Holographic Optical Elements for Visible Light Applications in Photo-thermo-refractive Glass

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HOLOGRAPHIC OPTICAL ELEMENTS FOR VISIBLE LIGHT APPLICATIONS IN PHOTO-THERMO-REFRACTIVE GLASS

by

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ABSTRACT

This dissertation reports on design and fabrication of various optical elements in Photo-thermo-refractive (PTR) glass. An ability to produce complex holographic optical elements (HOEs) for the visible spectral region appears very beneficial for variety of applications, however, it is limited due to photosensitivity of the glass confined within the UV region. First two parts of this dissertation present two independent approaches to the problem of holographic recording using visible radiation. The first method involves modification of the original PTR glass rendering it photosensitive to radiation in the visible spectral region and, thus, making possible the recording of holograms in PTR glass with visible radiation. The mechanism of photoionization in this case is based on an excited state absorption upconversion process in the glass when doped with Tb\(^{3+}\). By contrast, the second approach uses the original Ce\(^{3+}\) doped PTR glass and introduces a new modified technique for hologram formation that allows for holographic recording with visible light. Complex HOEs including holographic lenses and holographic curved mirrors were fabricated in PTR glass with visible light using both techniques. The third part of the dissertation takes a step in a different direction and discusses the development of the methods for fabrication of phase masks in PTR glass. A method for relatively straightforward and inexpensive fabrication of phase masks with the aid of a Digital Micromirror Device is presented. This method enabled to produce phase masks containing complex greyscale phase distributions for generation of vortex (helical) beams. A phase mask can be holographically encoded into a transmission Bragg grating where a holographic phase mask (HPM) is formed. HPM has an advantage over a regular phase mask of being capable of multi-wavelength operation. All optical elements recorded in PTR glass
preserve the advantages peculiar to VBGs recorded in PTR glass such as stability to heating and illumination with high-power laser beams.
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CHAPTER 1: INTRODUCTION

Optical elements such as mirrors or lenses have found their way into different areas of human life from science and technology to entertainment and household. Optical elements make it possible to harness light allowing for control of its dynamics and making use of light for a variety of applications. An optical function performed by an optical element may include reflection, diffraction, attenuation of light beams etc. Well-known optical elements such as lenses are used in eyewear for vision correction as well as in microscopes, telescopes, cameras and projectors. Such essential accessory as a smartphone contains a system of several aspheric lenses which enable using its CMOS camera to capture high quality photographs. As light-emitting diodes and other light sources become increasingly abundant with development of consumer electronics, the demand for optical elements is expectedly increasing. It has to be noted that optical elements are nearly always understood as mechanical elements made of traditional optical materials such as glass or polymer with its shape processed to fulfil a desired function and finalized by applying an appropriate coating. It appears, however, that the optical elements mentioned above can be also produced by holographic recording. The diffractive structures are fabricated by means of holographic imaging by transforming an interference pattern, formed by two intersecting beams, into a holographic optical element (HOE). This work is intended to familiarize the reader with general principles of operation and fabrication methods of such elements. The characteristic features of HOEs are presented, and a comparative analysis of their performance with respect to traditional mechanical optical elements is given. The main differences between HOEs and traditional mechanical optical elements are shown, and the advantages and drawbacks of both are discussed. Furthermore, the paper brings up a question of whether traditional optical elements can be gradually substituted for their holographic counterparts, at least in certain applications. An
attempt has been made to identify the shortcomings of the currently available holographic structures and design the new state-of-the-art holograms that meet the present day requirements. As a first step, however, the general overview of an HOE is presented.

1.1. Historical note

A principle of wavefront reconstruction was invented by a hungarian physicist Dennis Gabor in 1947 who was later awarded a Nobel prize “for the invention and development of the holographic method” [1, 2]. However, it wasn’t until the invention of the laser in 1960 [3] when the first hologram was possible to fabricate [4]. A first holographic mirror was demonstrated by Denisiuk as early as in 1962 [5]. That first success was then promptly built on with the first off-axis hologram created shortly by Leith and Upatnieks [6]. Later development of ray tracing techniques and the introduction of efficient holographic materials rendered it possible to fabricate practical HOEs of 3D objects [7]. The original concept described by Gabor is still being used as of now without undergoing any drastic transformation.

1.2. Introduction

An HOE is essentially a hologram of a point. Such element can be created by illumination of a photosensitive material with two or more beams from the same coherent light source. When the two beams intersect within a volume of the material, an interference pattern produced by the beams will be recorded [8]. The simplest case corresponds to the interference pattern of the two collimated beams with planar wavefronts in which case the pattern will represent a sinusoidal modulation throughout the length of the holographic plate. If one of the beams is scattered from an object, it thus carries information about the object which is then encoded into the hologram as demonstrated in the Fig. 1a. Once a hologram is formed, it can be used to produce an image of the object via
reconstruction. The hologram must be illuminated with the original reference beam for
reconstruction. When reference beam is incident on the hologram, a beam identical to the original
object beam is generated (Fig. 1b). As the latter was originally scattered from an object, the image
of the object will be produced. It is thus understood that the hologram performs the optical function
of the object. For instance, hologram of a lens will focus the beam just as if it possessed optical
power [9]. Similarly, the optical function of mirror, pinhole, beam splitter etc can be encoded into
a photosensitive material by means of holographic imaging. HOE can be later used in an optical
system in the same fashion as the original object.

Holograms are normally classified into thin and thick ones, amplitude and phase ones, and finally
into transmission and reflection holograms [8]. The hologram is considered thin if its thickness is
on the order of the interference planes separation. Those holograms exhibit several diffraction
orders producing several image waves. If the hologram thickness is significantly larger compared
to the wavelength of light, the interaction length of the beams in that case will be long, and that
type of hologram is referred to as a thick (volume) hologram. In that case only one diffraction
order that experiences constructive interference will be present while other orders will be largely

Figure 1  Formation and reconstruction of hologram.
attenuated. The diffraction efficiency achievable in the volume gratings is drastically higher than that of surface gratings, and can theoretically be as high as 100%.

Illumination of a hologram recording material with light might result in a change in the absorption \( \Delta \alpha \) of or in its refractive index \( \Delta n \) or both. Depending on the material type, one or the other effect can be more prominent. To distinguish the two, the HOEs are typically divided into amplitude and phase types [10]; an amplitude hologram is mostly based on the modulation of the absorption coefficient; by contrast a hologram where refractive index change (RIC) is prevalent (\( \Delta n \ll \Delta \alpha \)) is referred to as a phase HOE. In this work the focus is maintained on phase holograms owing to their useful properties.

Finally, transmission and reflection HOEs are discerned depending on the recording geometry. If incident and diffracted beams are located on the same side of the recording material, the hologram is known as reflective. When the incident and diffracted beams are found on opposite sides of the hologram, then the hologram is a transmissive one. Whether transmissive or reflective, an HOE is fabricated by recording an interference pattern in a plate of holographic material.

1.3. Advantages of holographic optical elements

The operation of a HOE thus relies on the modification of the optical wavefront by a two dimensional structural pattern rather than on variable physical thickness as it happens in conventional optical elements. Therein lies an important advantage of HOEs over the traditional mechanical optical elements. Here, conventional optical elements are produced by molding or grinding of a piece of an optical material to an appropriate three dimensional shape followed by high precision polishing to ensure the desired properties. By contrast, fabrication of an HOE does not require any material processing which reduces the fabrication costs and drastically simplifies the reproduction process. Besides a 2D pattern created in a plate of a holographic material would
be usually more compact than a corresponding 3D object which allows for more compact and lightweight solutions. Therefore, an optical system where an HOE is employed is likely to be more cost-efficient. This is just one of several advantages that HOEs possess over conventional mechanical optical elements. There are also other useful properties of HOEs worth mentioning. A single volume of a holographic material can be used to fabricate several holograms resulting in multiplexed optical structures [11]. Hence a single element can perform several different functions. Besides, HOEs can be designed and fabricated to have a diffraction efficiency from 0 to 100%. That means that an element can work as a beam splitter. Besides, it bears mentioning that an HOEs can be used for correction of certain aberrations adherent to traditional optical elements [12].

Meanwhile, the wavelength selectivity, a feature peculiar to volume holograms only, can be perhaps cited as the most important property of an HOE. The feature is attributed to a property of volume holograms that are designed to perform their function for the light at a certain wavelength only [12]. Practical advantage is that a volume HOE will combine the functions of an optical element and a wavelength filter. As an example, a holographic lens will only focus light which falls within a narrow spectral bandwidth while the rest of the spectrum will pass through without changing direction. Filtering property appears to be of particular importance since it can be particularly useful for certain applications some of which are discussed below. The advantages of HOEs discussed above are the reason why increasing number of applications substitute conventional optical elements with their holographic counterparts.

1.4. Applications of holographic optical elements

Below several applications are considered that can substantially benefit from using the filtering properties offered by HOEs. Atmospheric LIDAR, a remote sensing system used for study of the atmosphere, is an example of such application. Water vapor and its contents in the atmosphere are
important factors that have influence on weather, climate, and pollution processes. Scientists have been using LIDAR systems to perform comprehensive measurements of water vapor in the atmosphere. A typical LIDAR features a transmitter and a receiver as depicted in the Fig. 2. The transmitter comprises a pulsed laser source coupled to a set of mirrors followed by a beam expander that produce a collimated beam generally referred to as a probe beam which is directed upwards towards an atmosphere. The probe beam is scattered by molecules in the atmosphere such as aerosol particles, water vapor, and ice crystals. Some of the light travels back towards earth and is collected by a receiver. The backscattered is first collected by a telescope which directs it towards the curved mirror. The mirror in turn sends the signal towards the optical analyzer, and then, based on the amount of backscatter collected, an information about the density of the vapor phase in the atmosphere can be obtained. Detection of the backscatter, however, presents severe difficulties as a substantial amount of noise is collected along with the desired signal. This is due to a limited reflection bandwidth of the large tunable scanning mirror used in a LIDAR to collect the maximum possible amount of light incident from telescope at different angles. The problem can be addressed by substituting a traditional mirror with an HOE, whose filtering property allows to substantially alleviate the noise issue.
NASA has developed a scanning telescope based on a 400 mm reflecting HOE [13]. The HOE rotates in its plane thus replacing a large and costly laser sensor rotation system and allows to significantly reduce the number of parts contained in the receiver. Most importantly, however, the HOE provides filtering of the collected light to separate the desired backscatter signal from the noise. This technique allows for significant improvement of the signal-to-noise ratio of the system. As a result, attainable scanning range of the LIDAR system will be improved as well. The HOE was developed and fabricated by NASA and subsequently implemented in a Prototype atmospheric scanner system for environment remote sensing (PHASERS) LIDAR system operating at the wavelength of 532 nm [14]. The success of the system was followed by construction of the similar Holographic airborne rotating LIDAR instrument experiment (HARLIE) system for operation in the NIR at 1064 nm [15].

The filtering property and compact design were the factors that contributed to the success of the LIDAR systems from NASA. The same qualities were proved to be useful in air-to-air communication systems as well [16]. Air-to-air communication systems provide the airborne
vehicles with the means of communication with each other. Efficient communication requires constant tracking between the transmitter located on one aircraft with the receiver on the other. For that reason the tracking system has to feature a wide field of view (FOV). In the same time a receiver with a narrow field of view is needed to improve tracking accuracy. The compromise can be found by implementing a wide angle holographic lens as a laser receiver. A holographic lens allows for precise imaging of a signal onto a wide area detector, where the location of the transmitting source is determined from the position of the focused beam on the detector. Similarly to the LIDAR application, the communication receiver will largely benefit from fine spectral filtering that significantly improves the signal-to-noise ratio. That holds even for a system where a broadband transmitter source is used. Compact design, smaller weight of the HOE are of no less importance in the tight environment of the aircraft where any weight increase results in additional cost of the system.

So far HOE have been proved to allow for collection of large amounts of light into a wide FOV receiver. Meanwhile, it appears that holographic elements can be used for even larger angles such as pitch angles of antennas in concentrated solar power systems [17]. Those system produce electrical energy from sunlight by turning it into heat through a steaming process. Every concentrating solar power system comprises a solar concentrator to focus a large amount of sunlight onto a small area of a steam turbine. Commonly there are four main types of solar concentrators including a parabolic trough, compact linear Fresnel reflector, power tower or dish engine. Each of the mechanism employs a mechanical sun tracking system which is used to locate the position of the sun which changes due to rotation of the planet. Orientation angle of the concentrator is constantly steered to face the sun in the interval between 10 am and 2 pm corresponding to the most amount of collected sunlight. Again, use of an HOE renders possible to
avoid the bulk of mechanical tracking system and drastically reduces the number of elements in the system.

HOE have been already employed in solar concentrator system. A solar concentrator based on a multiplexed transmissive HOE is reported by [18]. An angular multiplexing property allows to concentrate sunlight coming from different angles onto a small area. In that case different holographic lenses multiplexed into a single element will focus light onto the same point from different positions of the sun at different hours. Meanwhile, wavelength selectivity of the hologram proves to be useful to filter the light at the desired wavelength while cutting out the unusable part of the spectrum such as UV. The reported system showed a reasonable efficiency of the total sunlight energy conversion of 60% where efficiency of the sunlight collection alone was measured to be 80%. Thus, solar concentration system can be an illustrative example of how the multiplexing property of an HOE can benefit the optical system.

There are also some applications that can use several advantageous properties of HOEs for their benefit. For example, spectral and wavelength selectivities as well as the multiplexing property of HOEs can be useful for applications in head-mounted display (HMD). HMDs are display devices that produce virtual and augmented reality. A typical HMD has two microdisplays that deliver two parallax images separately for each of the eyes creating a perception of a three-dimensional image. The display occupies the entire field of view of the user. Two types of HMDs discerned are a curved mirror and a waveguide HMD which employs an HOE. The systems of the first type embody a half-transparent curved mirror that is located in front of the user’s eye [19]. Curved mirror systems possess a number of bulky electronics which is needed to track the movement of the user’s eye. Besides, they are prone to distortion which has to be compensated electronically.
On the other hand, the waveguide HMDs as name suggest uses an optical waveguide for image projection avoiding the extra weight and space required by side electronics.

Holographic waveguide HMDs are now becoming more abundant due to low cost, compactness, and spectral and angular selectivity of an HOE. Reference [20] demonstrated a full color HMD whose principle of operation is shown in the Fig. 3. The holographic waveguide consists of a couple-in part, a waveguide itself, and the couple-out part. The image projected by a microdisplay is collimated by a lens as it enters the waveguide. Here the beam is incident on a reflecting HOE that directs beam down the waveguide using total internal reflection. The light is then reflected at a couple-out HOE and directed towards the eye of the user. Full-color HOE can be implemented by incorporating several different color reflective holograms into a multiplexed structure. The same result can be also achieved by stacking three single-color HOEs in a laminated structure. Both solutions yield three wavelengths of light being refracted simultaneously at the couple-in and couple-out HOEs resulting in an image projected as parallel rays towards the observer. HOE thus allows for projection of full color images onto an HMD without aberrations.

![Figure 3 Holographic optical display of waveguide type containing holographic optical element [20].](image-url)
There are two main considerations when designing an HOE-based HMD. The first consideration is the diffraction efficiency which determines how much of input power is refracted into the waveguide and then coupled out of it to reach the observer. It thus determines how much power has to be provided by the light source to yield necessary visibility of the image. This in turn directly affects energy efficiency of the device. Diffraction efficiency is mostly determined by a holographic material where the HOE is fabricated. Photopolymer is one of the currently used holographic materials that demonstrates relatively high values of diffraction efficiency. A full color HMD reported by [21] featured of total output efficiency of over 90%.

The other challenge encountered by HMDs is however more fundamental. Specifically, projected images in a traditional stereoscopic HMDs are two-dimensional and are formed at the conjugate plane of the microdisplay. Meanwhile the eyes are aimed at a different point in space which is determined by the image parallax. As the result, the eyes are focused at distance different from that of the imaginary three-dimensional image. This effect is known as accommodation-vergence conflict, and it is what causes the eye fatigue of the observer and as a consequence – discomfort during usage of the HMD. Authors in the reference [22] proposed and designed an HMD that projects real three-dimensional images using a spatial light modulator. The images are not confined to the conjugate plane of the display but are formed at a real distance. An extra challenge related to aberrations due to asymmetric refraction on the HOE is addressed by using pre-distorted hologram for aberration compensation. As the result, the system allows for projection of real aberration free three-dimensional images.

Described above are the three examples of successful incorporation of a holographic technology into an optical system. However the list does not end there. HOEs are widely used in optoelectronics for collimation of the laser diodes [23]. Laser diodes are known for large beam
divergence which can be compensated by a focusing HOE such as a holographic lens. Then, HOEs are often employed for free-space optical interconnects for coupling and decoupling light from fiber facet [24]. Those components can operate as diffractive or reflective structures as single fan-out elements as well as multifacet HOE arrays.

In conclusion to this general overview it is worthwhile to again mention the few characteristic features of HOEs that make it a valuable substitution for its mechanical counterpart? Among others it is its two-dimensional form factor as most of the HOEs use a plate substrate as opposed to the bulk of a mechanical element. As a consequence of the form factor, HOEs are usually lightweight compared to the similar mechanical optical elements. Thus use of HOE gives an edge over traditional elements in the mobile systems, space, aircraft, and other applications where extra weight comes at a price. On the other hand, wavelength and spatial multiplexing properties allow for several different functions being performed by a single element. The latter is unachievable with traditional optics. This property often allows to reduce the amount of components in the system or avoid the movement of the components. Then it is important to remember that reflective holograms combines the functions of an optical element and a fine wavelength filter. Indeed, wavelength filtering is inherent and unparalleled property of any HOE of the reflected type. As a result, bandwidth sensitive applications, such as sensing, benefit greatly from incorporation of such elements. Besides everything already mentioned, HOEs are often less expensive to produce, and their mass production is noticeably simplified. Cost efficiency is, in fact, often a determining factor when choice is made between HOEs and their mechanical counterparts.

All things considered the question then becomes why incorporation of HOE-based solutions take place in a relatively slow pace. The main drawback of HOEs lies in the imperfect reproduction of object beams upon reconstruction. Indeed, an HOE is not a complete replica of a traditional
mechanical element, rather it is a structure that is built to perform the same function. It is whether a mechanical optical element can be replaced with its holographic counterpart is what determines the success of the latter. In fact, often holograms demonstrate inferior performance compared to traditional optics. Essentially there are several properties that are peculiar to HOEs such as diffraction efficiency, resolution, spectral response etc. that determine how close performance of a hologram duplicates the performance of the mechanical optics. Later in this work the shortcomings of modern holograms are reviewed and possible methods for improvement are discussed. A brief overview of the basics of holography given below is meant to aid in full understanding of those properties.

1.5. Formation of a hologram

According to the definition holography is a method of recording of a light wave in a photosensitive medium with a capability for consequent reconstruction of the wave. The holographic principle is similar to the photography, however, with a crucial difference. Whereas photograph represents a distribution of light intensity captured on a photographic film using a lens, a hologram contains information about both amplitude and phase of light. In fact, hologram can be recorded with different types of waves such as X-rays, microwaves, acoustic waves, etc. as long as the waves are coherent, however it is optical holography that proved to be of use for many applications and thus appears to be of utmost interest [10].

The capacity of storing both phase and amplitude information in the hologram can be accounted for the principle of hologram formation. Here a hologram is formed by recording of the interference pattern of the two waves. The basic technique of hologram formation works as it is demonstrated in the Fig. 4. A beam from a coherent light source is split into two components thus creating two mutually coherent beams. While one of the beams propagates undeviated, another
beam is incident at an object. The second beam is then reflected from the object and directed towards the photosensitive medium. This beam is known as the signal beam. The undeviated beam is referred to as the reference beam, and quite often, just as it is in the case under consideration, it has a plane wavefront. The signal and the reference beams eventually intersect, and since the light is considered perfectly coherent, an interference pattern will be created at the intersection of the two beams. The photosensitive medium is then placed at the intersection point of the two beams, whereupon the medium interacts with light, leading to the light induced transformations in the material [25]. That transformation typically leads to local change of either the refractive index of the medium or there its absorption coefficient. For example, when silver halide emulsions is illuminated with light of an appropriate spectrum, the silver halides convert to silver atoms resulting in increased absorption in a material. The local distribution of refractive index or absorption coefficient in the photosensitive material is what constitutes a hologram. The hologram can be later bleached, and then silver atoms will be substituted with a transparent substance which does not absorb light but instead possesses a different refractive index. These two mechanisms result in formation of two different types of hologram, namely absorption and phase holograms that are going to be a subject of a detailed discussion later in the chapter.

Regardless of the nature of the effect, the amount of change in the material will be proportional to the amount of optical energy absorbed by the medium over a period of time significantly longer than the wave period [8]. Optical energy is in turn governed by the intensity of light which is described by

\[ I = 2\langle u \cdot u \rangle \]  \hspace{1cm} (1.1)

and characterizes the flow of optical energy through a unit area per unit time. The total amount of energy absorbed by a medium during an exposure time \( \tau \) is known as exposure \( E \) given by
The properties of a resulting hologram are largely dependent on the exposure. Now when process of hologram formation became more apparent, an attention should be given to the principle of the hologram operation.

\[ E = I\tau \] (1.2).

A hologram is essentially an interference pattern of the two waves mentioned above, the signal and the reference waves. The interference pattern creates a spatial distribution of fringes in space which is constant for a coherent light where relative phases of the two beams are constant. The intensity distribution is described by

\[ I = I_S + I_R + 2u_Su_S \cos(\varphi_S - \varphi_R) \] (1.3),

where the third term on the right hand side of the equation is known as interference term, and it is the one that contains the information about the relative phase. It can be inferred that the interference pattern will be formed only for two waves that have components of their electric fields parallel to each other. Meanwhile the third term will be naught for the two waves polarized orthogonally to each other, meaning that those waves will not interfere, and the total intensity will be a simple sum of the intensities of the two beams. By contrast, two perfectly coherent waves
with equal intensities of $I$ will create an interference pattern with the maximum intensity as high as $4I$ and the minimum intensity as low as naught.

**1.6. Hologram formed by interference of two plane waves**

The particular interference pattern depends on the amplitude and phase variation of the interfering waves, and can get rather complex. Hence it is worthwhile to begin by considering the most straightforward case of interference of two plane waves. The Fig. 5 demonstrates formation of the interference pattern in case when two waves with planar wavefronts intersect at an angle of 20° [26]. The lines labeled $F_1$ and $F_2$ denote the planes of maximum amplitudes of the first wave and the second wave respectively; those lines are known as wave crests. As two waves propagate in space, the intersection of their wave crests will form a line where light intensity is at a maximum. Hence the interference pattern will contain a set of bright fringes separated by the dark areas corresponding to the minimum intensity. Separation of the fringes defines the period $d$ of the hologram. The period is dependent on the intersection angle of the two waves and can be found from the equations describing the ratio of the sides of a triangle depicted in the Fig. 5. Now if a plate of holographic material is placed in the middle of the interference pattern, the intensity distribution will be converted into the change of refractive index to create a phase hologram (or in absorption for amplitude hologram). Changing the thickness of the photosensitive plate $T$ and its orientation with respect to the beam can result in different types of holograms created, such as transmission or reflection, planar or volume hologram. This distinction can be better understood on a different example when the interference pattern is obtained by intersection of a planar and a spherical waves.
1.7. Hologram formation geometries

Different hologram geometries are shown in the Fig. 6 where a spherical wave emitted from a point source forms an interference pattern with a planar wave [10]. Point source is considered here to be the object hence the spherical wave is referred to as the signal wave. Similarly the plane wave is regarded as the reference wave. Looking ahead it has to be noted that this geometry presents a
fundamental example of hologram formation technique since generally every complex hologram can be considered as a superposition of point sources. Thus an object wave will consist of a set of spherical waves that are emitted from the appropriate point source and will be interacting with the reference wave. Therefore, formation of a complex interference pattern follows the same principle as described below.

![Figure 6](image.png)

Figure 6 Various hologram formation geometries demonstrated by example of interference of plane and spherical waves.

Formation of an interference pattern of a spherical and a plane waves can be understood from the Fig. 6 where the lines represent the contours of maximum intensity. It can be seen that density of the fringes changes significantly across the diagram. In fact, placing a holographic plate in different locations and in different orientations will result in different types of a hologram recorded. The most indicative examples are considered here. For instance, if a plate is placed in the location 1 in the figure, the hologram will be formed by the interference pattern of two collinear
waves. This type of hologram is referred to as an inline hologram [27]. An in-line geometry implies the minimum difference in optical paths of the two waves. That geometry imposes the least requirements on coherence of the two waves, which is why it was possible to fabricate with a low coherence thermal light source. For that reason the first ever hologram, recorded by Gabor in 1948 before the invention of a laser, was indeed of that type. Besides low coherence requirements, the large separation of interference fringes in this area allows to use holographic materials with low resolution which was beneficial in the early days of holography.

The difference of the recording configuration 2 (Fig. 6) is in the angle introduced between the intersecting waves, in this case waves are no longer collinear. A hologram produced with this configuration is known as an off-axis hologram [28]. As opposed to location 1, there is a noticeable path difference between the two waves, and that places higher demands on the temporal coherence of light. Thermal sources do not provide enough coherence for this type of hologram to be formed. On the other hand, Leith and Upatnieks used an advantage of a coherent laser radiation which allowed to overcome the path difference issue. The hologram was recorded in a plate with thickness smaller than the fringe separation. It will be shown later that upon reconstruction of the hologram the beam interacts with one fringe only, and hence the hologram is of a planar type.

As angle between the wave normals increases so does the density of the interference fringes. If the holographic medium is thick enough so its sickness exceeds the fringe separation similarly to the manner as it is made in the configuration 3, a volume hologram will be formed. Holograms of this type exhibit selective properties in accordance with Bragg law which will be discussed later. First hologram of that type was shown by Pennington and Lin [29].

Lastly the interference pattern in the location 4 is formed by two counter-propagating waves. This case will demonstrate the highest density of fringes with fringe separation approaching $\lambda/2$. 

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When the hologram is formed, and a beam is incident at the hologram, it will encounter a sequence of alternating layers of high and low refractive index which act as a resonant reflective filter. The hologram was first designed by Denisiuk and is known as a reflection hologram [5]. Reflection holograms possess pronounced wavelength selective properties and thus are often used as spectral filters. The four cases described above are the most distinctive recording geometries.

1.8. General case of hologram formation

In general case, however, the hologram might be obtained by interference of any two or more waves as long as their coherence is maintained, and the reference wave does not necessarily have to be planar. In general case light from a single source can be split and used for illumination of two different objects. A hologram can be fabricated by placing a photosensitive plate in the region where the light diffracted by the two objects intersects and creates an interference pattern. Hologram pattern is determined by the exposure which is proportional to the intensity distribution which in this case takes the form

\[ I = (a_1 + a_2)(a_1 + a_2)^* \]  

where \( a_1 = a_1 \exp(i\phi_1) \) is a complex amplitude of the first wave, \( a_1 \) and \( \phi_1 \) are, respectively, amplitude and phase of the first wave that are both functions of spatial coordinates [8]. \( a_2, a_1, \) and \( \phi_1 \) are the corresponding values for the second wave. Symbol * designates a complex conjugate, and bold type in \( a_1 \) specifies a complex quantity. Although wave functions are actually real quantities, the complex quantities are introduced for the purpose of facilitation of the calculations. With that in mind, the intensity of the wave can then be written as \( I = a\cdot a^* \), then (1.4) can be put in the following form

\[ I = I_1 + I_2 + a_1 a_2^* + a_1^* a_2 \]  

(1.5).
This expression describes the spatial intensity distribution in the case of the hologram formation by two arbitrary wavefronts. Interference of two arbitrary waves is the most general case, and it finalizes the review of formation techniques. The principle of reconstruction of light waves with the aid of holograms is discussed next.

1.9. Reconstruction of hologram. Surface and volume gratings

Reconstruction of waves with hologram is based on a principle known as diffraction which describes bending of wave normal upon encountering an obstacle with dimensions on the order of the wavelength. When hologram is illuminated by one of the original beams, parts of the beam are diffracted at the fringes into the direction of the other beam, thus the latter will be reconstructed. Typically, a hologram is illuminated with the reference beam to reconstruct the signal beam (see Fig. 7a). The object beam emerges from the plate at the same angle as the original recording beam. In such manner an observer would perceive the reconstructed beam as being originated form a virtual object. It is quite interesting that the hologram can be read using a conjugate beam which is a beam completely retracing the reference beam in the opposite direction (Fig. 7b). In that case the reconstructed beam is directed towards the original object and forms a real image exactly at the original object location. That image can be then captured by a device such as CCD camera.

Similarly to hologram formation, reconstruction of a hologram of essentially any type can be simplified to one of the few approximation. Different hologram geometries were mentioned above depending on relative positions of the wavefront normal. The mention was taken of the fact that in a qualitative manner a hologram formed by arbitrary wavefront can be analyzed as a superposition of spherical or planar wavefronts interfering with the reference wave. The same approach can be employed for the reconstruction process where any hologram can be considered as an aggregate of plane gratings [10]. In that case interaction of the reference beam with the hologram can be
analyzed as consecutive interaction with a set of planar gratings. Then, all holograms can be divided in two types: surface and volume gratings. Surface grating has small thickness and an incident beam only interacts with one fringe contour. When beam is incident on such surface grating, it will be diffracted into multiple orders. The orders will emerge at angles that can be found from the condition of a constructive interference

\[ d (\sin \theta_m - \sin \theta_i) = m \lambda \]  \hspace{1cm} (1.6),

where \( d \) – is a period of the grating, \( \theta_i \) is incident angle, \( \theta_m \) diffracted angle of the m-th order, and \( \lambda \) is the wavelength of light. The condition (1.6) indicates that beams diffracted from all points on the grating will be in-phase. In that case intensity of the diffracted beam will have the maximum value.

On the other hand, if a recording medium is thick, the fringes straddle the entire thickness of the plate. The principle of consecutive superposition of light diffracted by the successive fringes still applies for the beam reconstruction here, however, the interaction distance in this case is longer and only one particular order will experience constructive interference given by

\[ \lambda = 2d \sin \theta \]  \hspace{1cm} (1.7)
The condition (1.7) is known as Bragg condition named after William Lawrence Bragg who formulated it for diffraction of X-rays [8]. This condition applies for all volume holograms. The difference between diffraction on a plane grating and on a volume grating is that the former can take place for various incident angles whereas Bragg condition is more deterministic, and only works for one particular angle for a given wavelength of light. Hence follow the selective properties of volume gratings.

1.10. Considerations for coherence of light

A Formation of holograms discussed above was based on the assumption of perfectly coherent light such as that originating from a monochromatic source. Coherence is, in fact, a very important requirement for the source which is intended to be used for holographic recording. In simple terms, the recording source has to emit highly coherent radiation in order to obtain a good quality hologram. Laser sources can have considerable coherence properties and for that reason are traditionally used for hologram recording. Use of sources with poor coherence properties such as thermal sources might result in inferior performance of the hologram. In the extreme case no hologram will be formed. Coherence of light is determined by a parameter known as coherence length which can be understood when an infinite monochromatic wave train is considered as it propagates in space [30]. Relative phase can be measured in two arbitrary points on the wave train. If the points are reasonably close to each other in space, the relative phase will be a constant value in time. On the other hand, as distance between the points increases, there will be a situation when the measured phases are no longer constant. Here the maximum distance at which the two phases are still correlated is referred to as the coherence length of light. Practical consideration implies that the path difference of the two recording beams were shorter than the coherence length of light. In the opposite case the performance of the hologram might degrade, sometimes to the extent of
complete destruction of the hologram. Generally, the relations stated above regarding the intensity
distribution in the interference pattern would still hold, however, the visibility of that hologram
will be affected. Visibility of a hologram is determined by the contrast between dark and bright
fringes in the interference pattern and is given by

\[ V = \frac{l_{\text{max}} - l_m}{l_{\text{max}} + l_m} \]  

(1.8).

When coherence of light deteriorates, so does the fringe contrast and, as a consequence, the
diffraction efficiency of the hologram would be decreased.

Meanwhile measuring the relative phase between two points separated in space is equivalent to
a measurement of phase in a single point in space in two different moments in time separated by
the time interval \( \Delta t \). That time interval will be related to the coherence length by means of the
formula \( l = c \cdot \Delta t \). It can be shown [31] that \( \Delta t \) is inversely proportional to the emission bandwidth
of the light source hence so too is the coherence length which according to

\[ l = \frac{c}{\Delta f} \]  

(1.9).

It can be inferred that a narrow bandwidth sources such as lasers are advantageous for
holographic recording as they provide sufficient coherence length to accommodate for the path
difference between the beams. This is particularly important for three dimensional holograms since
they possess substantial physical depth which requires larger path difference between the recording
beams.

Two different aspects regarding the coherence property of light that are spatial and temporal
coherence. Temporal coherence is related to the relative phase of the wave in the single point in
space measured in two different moments separated by the time interval \( \Delta t \). On the other hand
spatial coherence determines whether there is a correlation between the phases measured
simultaneously in two different points on the wavefront. While essentially being representations
of the same effect, the two parameters have to be distinguished as they place different limitations on the holographic recording. For instance, a light source can exhibit sufficient temporal coherence but lack spatial coherence and vice versa.

In the above paragraphs the property of coherence was introduced, and its effect on holographic recording was outlined. However, just a qualitative explanation of the coherence property was provided. In order to analyze the effect quantitatively, a complex degree of coherence \( \gamma \) is introduced. The complex degree of coherence is a quantitative merit of coherence which determines the exact amount by which visibility will be decreased. Fringe visibility in a hologram can be then expressed in terms of \( \gamma \). This can be shown by considering the visibility of an interference pattern of light from an extended source [32]. The light from the source illuminates an opaque screen with two pinholes \( P_1 \) and \( P_2 \) in it as shown in the Fig. 8. Two light waves thus emerge from the pinholes and their interference is monitored at the point A located on another screen. The intensity pattern measured at A can be described by

\[
I = I_1 + I_2 + 2 \text{Re} \langle u_1 u_2^* \rangle \tag{1.10},
\]

where complex amplitudes of the waves are replaced with the appropriate complex electric field vectors \( u_1 \) and \( u_2 \). It is known that the average of the interference term in (1.10) will depend on the correlation between the phases of light in the two pinholes which, in turn, is determined by the coherence of light. Born and Wolf [30] define the complex degree of coherence according to

\[
\gamma_{12}(\tau) = \frac{\lim_{T \to \infty} \frac{1}{T} \int_{-T}^{T} u_{P_1}(t + \tau) u_{P_2}^*(t) dt}{\sqrt{\left( \lim_{T \to \infty} \frac{1}{T} \int_{-T}^{T} u_{P_1}(t) u_{P_1}^*(t) dt \right) \left( \lim_{T \to \infty} \frac{1}{T} \int_{-T}^{T} u_{P_2}(t) u_{P_2}^*(t) dt \right)}} \tag{1.11}.
\]
Given (1.11), the time average term in (1.10) can be expressed in terms of the complex degree of coherence using the following relation

$$2\text{Re}(u_1 u_2^*) = 2\sqrt{I_1 I_2} |\gamma_{12}(\tau)| \cos \beta_{12}(\tau)$$  \hspace{1cm} (1.12),

where $\beta_{12}$ is the phase of the complex interference term. Knowledge of the interference term allows to obtain the values for the minimum and the maximum intensities in the interference pattern which in turn yields the expression for the fringe visibility

$$V = \frac{2|\gamma_{12}|}{\sqrt{I_1/I_2 + I_2/I_1}}$$  \hspace{1cm} (1.13).

The expression (1.13) relates fringe visibility to the coherence of light expressed in terms of $\gamma$. Both spatial and temporal coherence are accounted for in the complex degree of coherence. Indeed, the expression appears to be useful for characterization of real-world light sources in terms of their applicability for holographic recording. Meanwhile, as parameter $\tau$ in (1.13) approaches zero, the expression describes solely the spatial coherence of the source. In the case under consideration the spatial coherence means that the interference patterns of light at the point $Q$ originated from different points on the extended source are correlated.

Figure 8  Double pinhole interference.
1.11. Coherence requirements on the recording source

Coherence property of recording sources is an important consideration for a practical holography. As it was mentioned before, the method of holography was invented before the development of the laser. For that reason, first holograms were created using thermal sources such as discharge lamps whose coherence properties were relatively poor. That meant that the size of the emitting source had to be decreased in order to provide sufficient coherence conditions for the recording. Size reduction prompted loss of the beam power which in turn resulted in extended recording durations. It is going to be shown later in the paper that prolonged exposure duration can be detrimental for hologram performance. Meanwhile, the invention of the laser which was designed to emit coherent high power radiation allowed for significant alleviation of the problem. Nevertheless, even with adequate coherence characteristics, there are still few considerations for using a laser for hologram recording [10]. In fact, radiation of a laser source appears to provide sufficient coherence only under the conditions of a single transverse and longitudinal modes. It has to be noted that the number of transverse mode will influence the spatial coherence of the laser whereas its temporal coherence will be determined by how many longitudinal modes are oscillating. Here the former of the two conditions is relatively unhindered since most gas lasers can emit in a single transverse mode or can be adjusted for single transverse mode operation by reducing the laser aperture. By contrast, it might take more effort to satisfy the latter condition as the number of oscillating longitudinal modes is dependent on such factors as emission bandwidth of the laser and its free spectral range. The latter is a direct function of the cavity length in obedience to

\[ FSR = \frac{c}{2l} \]  

(1.14).
An estimation of coherence length depending on the number of longitudinal modes is given in [33]. Using the expression (1.11) the authors determine the coherence length at which the complex degree of coherence is above reasonable for practical holography value of $1/\sqrt{2}$ to be defined by the formula

$$\Delta L_H = \frac{0.11c}{\Delta f} \quad (1.15),$$

where $\Delta f$ is the full Doppler width of the mode under consideration. Using a typical spectral width of 100 kHz a coherence length of 1 km can be obtained which satisfies requirements for any practical holographic applications by wide margins. On the other hand, the same authors show that the coherence length decreases if one more longitudinal mode is present, and, indeed, calculate the estimated coherence length to be 0.5 m in the case of two oscillating modes. This drastic decrease in coherence property implies that a laser with a single longitudinal mode is highly desirable for holographic recording.

Thus the basics of holographic methods have been reviewed, and the typical configurations of hologram recording and reconstruction were shown. A very important parameter of coherence was introduced and its effect on hologram performance was explained both qualitatively and quantitatively. It can be inferred that the choice of an appropriate light source is vital for successful implementation of any holographic methods. However, it is not just the appropriate light source that is crucial for achieving a high quality hologram. In fact, the number one concern in holography is not the source rather the recording material used, as it is the photosensitive materials and their properties that define whether all the features of the intensity pattern will be successfully transferred into the hologram and whether an object beam can be subsequently precisely reconstructed. In the next chapter the most significant parameters of hologram recording materials
will be discussed, and the main requirements will be listed. Comparison of some of the most widely used material up to date will be then given.

### 1.12. Introduction to holographic materials. Amplitude and phase holograms

Hologram recording materials are the materials that interact with light producing an induced structural change that results in formation of a hologram in the material. Properties of holographic materials are characteristics that determine whether a high quality hologram can be recorded with the appropriate light source as discussed in the previous chapter. In fact, while nowadays there is no lack of lasers that meet all the requirements for holography, the photosensitive material is often what presents a challenge for creation of a high-quality hologram. The main goal of a hologram recording material is to accurately transfer an interference pattern into a corresponding distribution of refractive index (or absorption coefficient). If a plate of a photosensitive material is considered, it has generally three parameters that will affect the intensity distribution of the transmitting beam, those being the refractive index of the plate material, its absorption coefficient as well as the thickness of the plate.

As was mentioned above, the holograms are normally discerned into amplitude and phase holograms depending on which of the three parameter experiences the most tangible change as a result of the exposure [8]. Holograms created by local variation of the absorption coefficient of the plate are referred to as the amplitude holograms. Contours of maximum induced absorption correspond to the planes of maximum intensity in the interference pattern. Hologram of that type operates by spatially modulating the intensity of the transmitted beam. Materials that yield amplitude holograms are classified as absorption materials. By contrast, phase holograms are created in the phase materials.
1.13. Diffraction efficiency of holograms

Phase holograms typically do not affect the amplitude of the beam, instead they produce a change of its phase. The change is described by

\[ \Delta \varphi = k_0 nt \]  

(1.16),

where \( k_0 \) is the wavenumber of the incident wave, \( n \) – the refractive index of the recording medium, and \( t \) – its thickness. As the beam propagating through the hologram (grating) acquires the phase, it might be fully or partially diffracted by the hologram [10]. Diffraction efficiency is a parameter that governs how much power of the incident beam is transferred into the diffracted beam and is given by

\[ \eta = \frac{I_d}{I_i} \]  

(1.17),

where \( I_i \) and \( I_d \) describe the intensities of respectively the incident and diffracted beams. The maximum diffraction efficiency of 1 takes place when the incident beam experiences the phase change of \( \pi \) radians. In that case, all the power in the reference beam will be transferred into the diffracted beam. Since acquired phase is a function of the refractive index of the medium as well as its thickness, then the spatial variation of at least one of those parameters will yield a phase grating. Although both parameters might be altered, typically the change in one of them is more significant.

Surface gratings are most often obtained by variation of the thickness of the photosensitive plate since it will produce more effect than the change of its refractive index. Typical variation of thickness is on the order of 0.5 μm which is on the order of average wavelength of light. Then, given the refractive index of 1.5 we shall obtain the maximum diffraction efficiency of approximately 40% achievable in the surface grating [25]. Higher values of diffraction efficiency can be obtained in volume gratings. Here due to larger thickness, a change in refractive index is
typically employed to achieve phase variation which can reach $\pi$ for few millimeter thick slabs and a refractive index change above $10^{-4}$. Efficiency of such volume gratings can be as high as 100%. It is owing to their diffractive properties, the phase gratings, both surface and volume ones, find application in a vast variety of areas, and are generally, more prevalent compared to absorption holograms. For that reason, the phase gratings are going to be the main topic of the discussion in the present paper.

Since phase holograms were chosen as a priority for the current paper, the analysis of hologram recording materials will be conducted based on their applicability for recording of phase holograms. The properties that are important for production of a high-quality hologram include photosensitivity, resolution, and recording noise. Besides, optical and mechanical properties of the material such as absorption and thermal expansion play an important role since they affect the operation of the recorded hologram. On the other hand, costs often have more influence on the applicability and industrial availability of the material than the properties above. Effect of each of the parameter above on the performance of hologram will be discussed next.

1.14. Figures of merit for hologram recording materials

Photosensitivity determines how much exposure is needed to obtain a necessary phase change in the material, for instance, $\pi$ radians. This depends on the absorption of the material as well as how efficient exposure is converted to the RIC. Here, a material with higher photosensitivity needs smaller exposure in order to provide the same diffraction efficiency which in turn results in shorter recording duration. On one hand, low sensitivity could be compensated by appropriately increased duration of recording. On the other hand, that is not desirable as long recording duration might result in deterioration of the hologram quality due to instabilities present during recording. Later the effect of instabilities will be discussed in greater detail, meanwhile, at this point general
estimation of 300 seconds can be considered as an upper limit for a recording duration. It can be inferred that given the upper limit of the recording duration, the photosensitivity will determine the maximum attainable diffraction efficiency in the hologram.

While diffraction efficiency is governed by the photosensitivity of the material, quality of a hologram is mostly dependent on the recording resolution [34]. As it has been mentioned before, a hologram is essentially a record of an interference pattern represented by spatial variation of refractive index. In fact, the spatial frequency of the refractive index modulation can be higher or lower. Indeed, it is the density of the contours of the maximum refractive index or simply fringe separation that determines the resolution of the hologram. The fringe separation is determined by the angle between recording beams as well as the fringe slant angle which in transmission geometry is expressed by

\[ d = \frac{\lambda}{\sin \theta_S - \sin \theta_R \cos \varphi} \]  

(1.18),

where \( \theta_S \) and \( \theta_R \) are angles that signal and reference beams make with respect to the grating normal, (angles are measured inside the medium). Considering a wavelength of 0.5 \( \mu \text{m} \) as an example, it can be found that the spatial frequency of such grating will be in the range \([0; 4000]\) lines per millimeter. In comparison, reflection gratings are created by interference of counter-propagating waves where angle \( \theta \) can reach \( \pi/2 \) radians. With the aid of Snell’s law, a fringe separation can be obtained as

\[ d = \frac{\lambda}{2\sqrt{n^2 - \sin^2 \theta}} \]  

(1.19)

Here assuming the same wavelength of 0.5 \( \mu \text{m} \), and a typical refractive index of recording medium of 1.5, it can be determined that the spatial frequencies in the reflective hologram will be in the range \([4500; 6000]\) lines per millimeter. It can be inferred that reflective gratings possess significantly higher spatial frequencies than transmission holograms.
A holographic material might be able or not able to properly resolve the small details in the hologram. In the latter case, the fringe contrast will be reduced for the features that are below the minimum distinguishable size. An ideal holographic material is supposed to have a flat frequency response which implies that the fringe contrast is independent of the line density in the hologram. From the comparison of the two geometries one can conclude that reflective gratings present greater challenge for maximum achievable resolution than transmission holograms. It has to be noted that the smaller the detail in the hologram, the more it is prone to degrading as the result of instabilities as will be shown later. For that reason, holograms with high spatial frequencies often impose additional requirements for reduced recording duration.

Another parameter of concern for evaluation of a holographic material is the scattering noise. Scattering might be introduced during recording of a hologram as well as during its reconstruction. Scattering is typically caused by microstructure of the material such as granule size as well as inhomogeneity and defects in the material [35]. Scattered light causes distortion of the reconstructed wavefronts which results in the introduction of noise and aberrations which can be devastating should the hologram be used for imaging applications. Beam shaping application will be compromised as well since diffraction efficiency of a hologram will be also reduced due to scattering. Therefore, a hologram with the highest quality and the best performance can be obtained in the recording material with high photosensitivity, high resolution, and the least amount of scattering. However, in practice the choice of a hologram material is more complicated since, generally, there is no material as of today that would possess all of the required properties. It appears that each of the materials presents a trade-off between advantages and drawbacks. The materials that are most commonly used for holographic recording include silver halide emulsions, dichromated gelatin, photoresists as well as photopolymers.
1.15. Hologram recording materials

1.15.1 Silver halide emulsions

When a holographic material is selected, first the benefits of each material are reviewed, and then decision on the material of choice is made based upon the primary requirements. Most often, recording resolution is given the highest priority since it determines if small details in the hologram can be at all resolved. Hence, photosensitivity and diffraction efficiency are considered of secondary importance. When resolution is the highest priority, attention should be paid to silver halide materials [36, 37]. Silver halide emulsion is a type of material where the first holograms were made. It still remains a common solution for holographic recording due to its high sensitivity and high resolution. High photosensitivity provides conveniently short exposure times that allow to significantly alleviate the requirements on light source and environmental stability. High resolution renders possible to record both transmissive and, most importantly, reflective holograms since latter are already known to possess features as small as 0.15 μm. Silver halide materials allow for formation of both amplitude and phase holograms [38]. As a result of light exposure and successive wet development, silver halides are converted to silver atoms that absorb light. The absorption is proportional to the density of silver grains which, in turn, is determined by the intensity of the interference pattern. This method results in formation of an absorption hologram. If necessary, this hologram can be bleached where silver atoms are converted into a transparent compound. This compound does not significantly absorb light, but its refractive index is different from the surrounding areas. Therefore, a phase hologram is obtained. Silver halide emulsion is a commercially available material, and several types of it can be purchase from companies such as Agfa and Kodak. However, while silver halide emulsions are distinguished by their high photosensitivity and resolution, they fall short in diffraction efficiency and scattering. The latter is attributed to a relatively large grain size of around 50 nm.
1.15.2 Dichromated gelatin

If diffraction efficiency of a hologram is of highest priority, a dichromated gelatin (DCG) can be suggested as a holographic material. That is one of the most common materials which features a fairly straightforward procedure of hologram fabrication [39, 40]. Optical element formation begins by coating a glass plate with a solution of gelatin and dichromate in water. The coated plate then undergoes a long-term exposure due to low sensitivity of DCG as compared to silver halide materials. Successive treatment, which includes chemical fixing, washing, and drying, finalizes the hologram formation. Sensitivity response of original DCG is limited to wavelengths in the blue-green region, however the range can be extended into the red by adding methylene dye sensitizers to the material [41]. Dichromated gelatin was found to possess a number of properties giving it an advantage over other holographic materials such as photoemulsions. High diffraction efficiency resulting from large refractive index modulation (RIM) values along with low scattering noise can be sited as an example of such properties. In reality, the listed advantages are strongly dependent on development such as thermal treatment, bias hardening, and dehydration rate [42]. Failure to perform the above mentioned procedures might introduce haziness as well as result in degrading of hologram quality due to its high sensitivity to humidity. Shortcomings of DCG include lower recording resolution as compared to silver halide emulsions and lower photosensitivity which results in extended recording durations.

1.15.3 Photoresists

Photoresists are materials that are used to make surface relief holograms. Surface relief is produced by exposure followed by dry treatment, and can be consecutively transferred to a plastic mold [43]. Most photoresists have reasonable photosensitivity to radiation in the blue and UV regions but can be modified to provide sensitivity up to 1 μm [44]. Shipley photoresists are among
the most commonly used. Simplified mass production can be named as a distinctive advantage of these materials whereas main disadvantage is in a compromised shelf life of the hologram.

1.15.4 Photopolymers

Lastly, another type of material used for holographic recording is photopolymer. Photopolymers are polymers that are incompletely polymerized. Light exposure activates a photosensitizer which in turn triggers the chain of reactions resulting in ultimate polymerization of the material [45]. The areas that underwent exposure to light have higher refractive index therefore a phase hologram is readily available. The chain of transformations is only triggered by light exposure and takes several minutes. Thus, hologram in photopolymer is readily available and does not require post-processing as opposed to other materials [46]. Although a dry processing can be carried out in order to enhance hologram properties [47]. Common examples include DMP-128 polymer from Polaroid and DuPont photopolymer.

1.16. Persistence and durability of holograms

The examples provided here support the statements made earlier regarding a trade-off of choosing one material over the other. Thus, the choice should be made based on the critical parameters for example depending on whether resolution or diffraction efficiency is of highest priority. The properties discussed above, however, mostly describe the imaging performance of a hologram. Imaging performance is known to describe how accurately the object beam can be reconstructed. However, no less important are the mechanical properties of the holographic material, and persistence of a hologram in particular. It could be expected that the properties of the hologram will be the same after a certain period of time as they were directly after the recording. This, however, is not necessarily the case since a hologram can fade which happens due to thermal relaxation of the photosensitive agents inside the material [35]. Fading can be caused by the
illuminating radiation and environmental effects as well as temperature. Fading in turn results in
deterioration of the holographic performance, degrading of its optical properties, and, finally, can
even lead to destruction of the hologram. In early days of holography extremely short lifetimes of
holograms were a common problem indicative to all recording materials. Nevertheless, overtime
the materials were gradually improved, and most materials nowadays can provide a hologram that
is stable for at least the duration of its reconstruction. Some elements can be even reused in case if
the reconstructing beam does not affect the properties of the hologram [8].

1.17. Considerations for high power applications

Holographic application in general can be divided into imaging and non-imaging application. So
far the emphasis was made on holographic imaging which is widely used for art, data storage etc.
Apart from that, however, HOEs are often employed for non-imaging applications such as beam
shaping as they are designed to substitute conventional mechanical optical elements. These
applications present new difficulties for a holographer. Optical elements are quite often used for
high power applications which means that if an HOE is intended to substitute a high power
mechanical optic, it too has to possess tolerance to high power laser beams. High power stability
implies that hologram persistence properties described above should be retained even for high
power levels. The above mentioned properties, however, do not necessarily apply for high power
applications. There two considerations for the high power capability of an HOE. First is attributed
to destruction or degradation of the hologram as a result of the high power beam illumination.
Second, the material where hologram is recorded might be damaged by high power laser beams
which inevitably leads to destruction of the hologram as well. All the materials listed above
generally are not meant to be used in high power applications [48]. Although the attempts have
been made to enhance the high power tolerance of such materials as DCG, at the present moment
they cannot fully replace the conventional mechanical optical elements. For that reason, when HOEs are designed with high power applications in mind it is worth considering another type of holographic material – photosensitive glasses.

1.18. Holography in photosensitive glasses

The first attempts to produce holograms in photosensitive inorganic glasses were made over 50 years ago. The method that was employed most often at that time was by bleaching of intrinsic color centers [49]. Color centers in glass were created by UV radiation and then bleached by visible light resulting in the formation of an amplitude hologram. This type of hologram only allows for low diffraction efficiencies and for that reason is of little interest for the present work. Later it was discovered that photoionization can cause RIC as well allowing for fabrication of phase holograms [50]. That RIC, however, was found to be on the order of 10^{-6} which is insufficient for any practical holography applications. One mechanism that indeed provided high diffraction efficiency was creation of surface relief gratings by photoexcitation and consecutive etching [51]. Yet those gratings had a different shortcoming: although created in glass and while they were found to be not able to tolerate high power illumination. As for the holographic structures recorded in the volume of glass, those suffered from insufficient diffraction efficiency due to low photosensitivity of inorganic glasses resulting in little RIC. An attempt was made to address the problem by combining the inorganic glass with organic polymer insets that allowed for significant increase in photosensitivity [52]. This approach, however, introduced a problem of additional absorption of polymer compounds which was ultimately incompatible with high power beams. The fundamental problem of low photosensitivity in the materials such as photopolymers is addressed by development process. Glasses, on the other hand, possess high densities leading to low diffusion coefficients which encumbers wet processing [53]. Thermal processing can be used instead as a
hologram development mechanism in inorganic glasses which results in improved photosensitivity.

1.19. Photo-thermo-refractive process and photo-thermo-refractive glass

Thermal processing of a previously UV exposed glass improves the photosensitivity of the glass and allows for larger RIC values that are sufficient for recording of phase volume holograms. This two-step process which includes exposure and thermal treatment is known as a photo-thermo-refractive process, and it is what provides the basis for operation of a commercially-viable photo-thermo-refractive (PTR) glass. PTR glass is a sodium-zinc-aluminum-silicate glass doped with cerium, silver, tin and antimony (full formula Na$_2$O-ZnO-Al$_2$O$_3$-SiO$_2$) [54]. The glass is widely used for fabrication of volume Bragg gratings (VBGs) [55] and phase masks [56] that, in turn, find their application in lasers beam control systems [57, 58]. The process of hologram fabrication in PTR glass comprises two main stages, namely, exposure to UV radiation and thermal development. On the first stage a plate of PTR glass is exposed to UV radiation from a He-Cd continuous wave (CW) laser at the wavelength of 325 nm. The UV beam is split at a beam splitter and the derived beams are interfered at an appropriate angle which is chosen to provide a desired grating period. Thus recording configuration for fabrication of a VBG is identical to the formation of a trivial transmission grating as it was demonstrated in the Fig 1.6 in the previous chapter. Large thickness of the glass plate yields a volume hologram while the period of the grating is determined by the intersection angle of the two beams. The difference in the case of PTR glass is that UV exposure alone actually creates no grating in the glass [59]. In fact, no RIC is present in the glass at that stage, rather exposure to the interfering UV beams creates what is known as a latent image in the glass. This latent image is converted to a real phase hologram upon thermal development.
Photosensitivity of PTR glass is determined by presence of the following dopants: silver ion acting as a photosensitive agent and trivalent cerium ion as a sensitizer. Photoionization of the glass can be carried out by exposure to UV radiation with wavelength falling within the range between 280 nm and 350 nm – the absorption band of the Ce$^{3+}$ ion [60]. Absorption of a UV photon with the energies corresponding to the indicated range results in excitation of the trivalent cerium ion (Ce$^{3+}$) from the ground state to the 5d$4f^7$ band which is located above the electron mobility threshold in PTR glass [61]. As a consequence, Ce$^{3+}$ ion is converted to a Ce$^{4+}$ ion, and a mobile electron is created. A mobile electron migrates from cerium ion and can be captured on traps such as Ag$^+$, Sb$^{5+}$ as well as impurities and defects in the glass. An electron captured in the trap is known as a color center. The electron can also return to the Ce$^{4+}$ ion resulting in recombination and restoring of the original Ce$^{3+}$ state. Meanwhile, color center formation concludes the chemical transformations that accompany UV exposure stage. As shall be seen next, those color centers play a crucial role in the photosensitivity process as they contribute to formation of the RIC. As mentioned above, UV exposed sample then undergoes a thermal development in order to produce a RIC in glass.

Thermal treatment or thermal development constitutes a second stage of hologram fabrication. Thermal development itself also comprises two stages, namely nucleation and crystal precipitation. During nucleation the glass is heated to the temperature of 485°C which is above the transition temperature of 470°C in PTR glass. Some color centers are unstable at high temperatures. Heat provides thermal energy that drives the electron out of the trap dismantling those color centers. Those electrons are captured by the remaining silver ions resulting in creation of neutral silver atoms and their consequent clustering [62]. Silver containing clusters serve as nucleation centers for consequent crystal growth which happens at the second stage of thermal development.
Meanwhile, as silver clusters are formed, glass is then cooled down to quench the nucleation process. After that no more silver clusters will be formed. Cooling is followed by the second stage of thermal treatment carried out at 515 °C. At these temperatures sodium fluoride crystals grow at the nucleation centers formed at the first development stage. As crystals are grown, a negative RIC takes place in the areas of the glass sample previously exposed to UV radiation. A maximum RIC value of over $10^{-3}$ (1000 ppm) can be achieved in PTR glass. The resulting RIC obtained during the two step procedure is dependent on the amount of the crystalline phase formed in the glass. The latter is a function of two parameters: the amount of the nucleation centers and the size of the crystals. Here, UV exposure is what determines the density of the nucleation centers created in the glass. On the other hand, the size of the NaF crystals is determined by the duration of thermal development and development temperature. Generally, larger RIC in the glass can be obtained by increasing both UV exposure and duration of thermal development. However typically the exposure is adjusted in order to obtain a certain desirable RIC. Meanwhile, duration of thermal development is kept at constant value corresponding to the optimum development conditions. Certain temperatures and durations of both development stages have to be maintained in order to achieve a desirable amount of crystalline phase. However, development at longer durations or higher temperatures will result in excessive crystalline phase that can introduce scattering in the glass. While larger refractive index values are often desirable for high diffraction efficiencies, scattering will decrease the diffracted output and hence will degrade the hologram performance. For that reason, an optimum duration and temperature of thermal treatment have to be maintained at the optimum values to provide the highest quality VBG in PTR glass. Those values had been determined to be 80 and 60 minutes for nucleation and crystal growth stages respectively for the standard recording procedure in PTR glass.
1.20. Photo-thermo-refractive glass as a hologram recording material for high power applications

Photo-thermo-refractive process allows for production of permanent RIC in PTR glass by UV exposure and consecutive thermal development. This implies that the glass can be used for fabrication of volume phase holograms. As a hologram recording material PTR glass is of particular interest to compare to the other materials discussed above. Here comparison of the maximum attainable RIC shows a value of $10^{-3}$ that can be achieved in the PTR glass which is lower than the maximum RIC of $10^{-2}$ in dichromated gelatin and some of the photopolymers [63]. However, RIC of $10^{-3}$ is sufficient to obtain volume holograms with diffraction efficiency as high as 99.9% in a 2 mm thick glass plate. Meanwhile, as an optical glass, PTR glass possesses immensely high resolution which by far surpasses other materials and allows to record structures with spatial frequencies as high as 10,000 lines per mm. Then PTR glass possesses low noise as scattering can be minimized by providing appropriate development parameters. Based on the comparative analysis of the parameters above, it can be summarized that the glass demonstrates superior parameters that place it in the same category with the most modern hologram recording materials. On the other hand, persistence and durability is where PTR glass has an edge over its counterparts. The persistence of holograms recorded in PTR glass is due to several factors. First factor, the environmental stability, is accounted for properties of the crystalline phase in the volume of the glass. The crystals inside the volume of the glass are not affected by the presence of mechanical or environmental disturbances. For example, possible scratches on the surface of the glass or other surface effects such as oxidation can be removed by additional polishing of the sample. For that reason, no fading of the hologram occurs overtime or during operation, and as a consequence the shelf life of the element is practically unrestricted.
Perhaps the main advantage of the PTR is its tolerance to high power laser radiation. The glass has extremely high laser damage threshold which goes as high as 40 kW/cm² for irradiance of CW sources and 40 J/cm² for pulsed beams with duration of pulse of 10 ns [64]. High damage threshold is accounted for very low absorption which is on the order of no more then 10⁻⁴ /cm across the entire window of transparency of PTR glass. This transparency window straddles from 350 nm in the UV region which denotes the residual absorption of the cerium and up to 2.7 μm in the NIR where absorption is introduced by hydroxyl groups introduced into the glass as contaminations at the fabrication stage. As a result, holograms recorded in PTR glass are designed to operate with high power beams from any lasers in the visible and NIR spectral regions falling within the designated bandwidth without causing any damage to the glass matrix itself or the crystalline structure forming the hologram. Besides, holographic elements recorded in PTR glass exhibit high stability to heat. In fact, the structures can be subjected to heating to temperatures of up to 400 °C without any noticeable deterioration of the holographic performance. Low thermal variation of refractive index is also important to mention as it implies stability of the resonant condition of the grating at different temperatures. Summarizing the properties listed here it can be inferred that PTR glass is a substrate that allows for fabrication of high efficiency holograms that are robust and can be used with high power laser radiation. The particular types of holograms produced in PTR glass are discussed below.

1.21. Volume Bragg gratings in Photo-thermo-refractive glass

Holograms recorded in PTR glass are planar transmitting or reflecting VBGs. VBG is a periodic perturbation in refractive index taking place throughout the volume of the glass plate. When a beam incident on the structure satisfies the Bragg condition as in (1.7) it will be diffracted on the periodic structure. VBGs are typically divided into transmission Bragg gratings (TBGs) and
reflection Bragg gratings (RBGs). The grating is referred to as a TBG if incident and diffracted beam are located at the opposite sides of the glass plate; in these structures fringes are often located perpendicular to the glass surface (see Fig. 9a). In the case of an RBG incident and diffracted beams are found on the same side of the glass plate. These gratings have fringes located parallel to the glass surface thus forming alternating layers of different refractive index, and hence they operate as resonant reflective filters for incident beams (Fig. 9b). However, a grating can be fabricated with slanted fringes in order to change the incident beam angle that satisfies the Bragg condition for a given wavelength of light. Slanted TBGs and RBGs are shown in the Fig. 9c and Fig. 9d respectively.

![Figure 9](image-url)

Figure 9 (a) Types of volume Bragg gratings a) unslanted transmission Bragg grating, b) unslanted reflection Bragg grating a) slanted transmission Bragg grating, b) slanted reflection Bragg grating.

In a general case of VBG with slanted fringes the Bragg condition in transmission and reflection geometries can be expressed by (20) and (21) respectively

\[
\lambda_T = 2\Delta \sin(\theta_i + \phi) \quad (1.20);
\]
\[ \lambda_R = 2\Lambda \cos(\theta_i + \phi) \] (1.21),

where \( \Lambda \) is the grating pitch, and \( \phi \) – slant angle. The performance of a VBG is evaluated by the three main parameters: its diffraction efficiency, its wavelength selectivity and angular selectivity. These parameters can be calculated using coupled wave theory developed by Kogelnik [65]. The theory describes propagation of a signal (S) and a reference (R) waves through a glass plate of refractive index \( n_0 \) and thickness \( d \) with an arbitrary grating of \( \Lambda \) pitch in it (Fig. 10). Grating fringes are slanted by an angle \( \phi \) with respect to the grating normal. This theory presents a straightforward and effective analysis of the grating performance, and it has been extensively used for simulation of performance of holograms mentioned later in this paper.

![Diffraction of a beam on a transmission Bragg grating.](image)

**Figure 10** Diffraction of a beam on a transmission Bragg grating.

### 1.22. Properties of volume Bragg gratings

Generally, Propagation of the wave through medium is described by the scalar wave equation that takes the following form

\[ \nabla E + \beta^2 E = 0 \] (1.22),

where \( E \) – is a complex amplitude of the wave, and \( \beta \) – its propagation constant

\[ \beta = \frac{2\pi n_0}{\lambda} \] (1.23).
In our case we have propagation of the S and the R waves coupled to each other with the total field equal to

\[ E(x, z) = R(z)e^{-jk_Rr} + S(z)e^{-jk_Sr} \]  

(1.24),

where the wave vectors \( k_S \) and \( k_R \) are related by the following equation

\[ k_S = k_R - K \]  

(1.25),

where \( K \) is the grating vector (\( K=2\pi/\Lambda \)). The vector relation can be depicted in a diagram form as shown in the Fig. 11, and it is what provides the satisfaction of the Bragg condition which takes the form

\[ \cos(\phi - \theta_R) = \frac{\lambda}{2n_0\Lambda} \]  

(1.26).

Meanwhile, coupling between the waves determines the energy transfer from the reference to the signal wave and is described by the coupling constant \( \kappa \)

\[ \kappa = \frac{\pi\Delta n}{\lambda} - i\frac{\Delta \alpha}{2} \]  

(1.27),

where \( \Delta n \) and \( \Delta \alpha \) designate the amplitude of the perturbation of the refractive index and the absorption coefficient respectively. Substituting the total field (24) along with (23) in the wave equation (22), a system of two independent equations can be obtained that describes the behavior of the R and S waves inside the grating

\[ \cos \theta_R \frac{dR}{dz} + \alpha R = -j\kappa S \]  

(1.28)

\[ \cos \theta_S \frac{dS}{dz} + (\alpha + j\Gamma)S = -j\kappa R \]  

(1.29),

where \( \Gamma \) is known as the mismatch parameter, and it determines how much the system is deviated from the Bragg condition in terms of wavelength (\( \Delta \lambda \)) or incident angle (\( \Delta \theta \))

\[ \Gamma = \Delta \theta K \sin(\phi - \theta_R) - \frac{\Delta \lambda K^2}{4\pi n_0} \]  

(1.30).
Solving the equation system (1.28) and (1.29) allows to obtain an expression for diffraction efficiency in the form of

\[ \eta = \frac{|\cos \theta_S|}{\cos \theta_R} SS^* \]  

(1.31)

VBGs recorded in PTR glass are essentially lossless and are purely phase gratings. Then in the case of transmission gratings (1.31) can be rewritten as

\[ \eta_T = \frac{\sin^2 \nu}{1 + \xi^2/\nu^2} \]  

(1.32).

\[ \xi = \frac{\pi f d}{\cos(\phi - \theta_S) - f \lambda/n_0 \cos \phi} \left( \Delta \theta \sin \theta_0 - \frac{f \Delta \lambda}{2n_0} \right) \]  

(1.36)

The diffraction efficiency is plotted as a function of the parameter \( \xi \) for several values of the parameter \( \nu \) as can be seen in the Fig. 12. It can be inferred from the graph that the parameter \( \xi \) describes the deviation from Bragg condition where efficiency goes to zero for a certain angle. Here the distance between the maximum and the first null is referred to as the angular selectivity. On the other hand, the parameter \( \nu \) defines depth of the phase modulation. It can be seen that the efficiency of the transmission grating will increase with thickness and refractive index modulation.
until it reaches the maximum of 100%. All incident power will be fully coupled into the diffracted wave when \( \nu = \pi / 2 \). If parameter \( \nu \) is increased beyond that point, direction of coupling will be changed, and the power will be then coupled back into the reference wave. It can be inferred that for a given thickness of the glass plate there is an optimum value \( RIM \) that yields 100% diffraction efficiency. If \( RIM \) is smaller or larger than the optimum value, the diffraction efficiency will be decreased. It can be also seen from the Fig. 12 that the grating with excessive refractive index change will have increased diffraction efficiency in the side lobes at the expense of the decreasing diffraction efficiency in the main lobe. Those gratings are referred to as overmodulated.

As far as reflective gratings are concerned, the expression (1.31) is typically represented in the following form

\[
\eta_R = \left[ 1 + \frac{1 - \xi^2 / \nu^2}{\sinh^2 \sqrt{\nu^2 - \xi^2}} \right]^{-1}
\]  

(1.37),

where parameters \( \nu \) and \( \xi \) are also different and are given by

![Figure 12: Diffraction efficiency curve for a transmission Bragg grating.](image-url)
\[ \nu = \frac{i\pi d\Delta n}{\lambda \cos \theta_R \cos \theta_S} \quad (1.38) \]

\[ \xi = -\frac{\Gamma d}{2 \cos \theta_S} \quad (1.39) \]

Similarly to the transmission case, the expression (36) in the case of unslanted grating can be simplified to

\[ \eta_T = \tanh^2 \left( \frac{\pi d\Delta n}{\lambda \cos \theta_S} \right) \quad (1.40) \]

Again, similarly to transmission gratings, angular and spectral selectivity of reflection gratings can be determined by writing the \( \xi \) parameter in terms of deviation of wavelength or incident angle

\[ \xi = \frac{\pi d\Delta n}{\cos(\phi - \theta_S) - f \lambda/n_0 \cos \phi} \left( \Delta \theta \sin \theta_0 + \frac{f \Delta \lambda}{2n_0} \right) \quad (1.41) \]

which replicates (1.36) with the exception of the change in the sign in front of the second term in brackets. In the case of reflection grating the dependence of the diffraction efficiency on the parameter \( \xi \) is demonstrated in the Fig. 13 where different curves again correspond to different values of the parameter \( \nu \). It can be seen that unlike transmission grating there is no optimum value of the \( \nu \) parameter, rather the diffraction efficiency is increasing with \( \nu \) approaching 100% in an asymptotic manner. Hence, refractive index modulation or thickness can be increased for improved diffraction efficiency in this case.
1.23. Applications of volume Bragg gratings in Photo-thermo-refractive glass

Two types of gratings exhibit very distinct properties when it comes to selectivity. As a matter of fact TBGs exhibit high angular selectivity whereas high wavelength selectivity is a property peculiar to RBGs. For example, angular selectivity FWHM of a TBG can be below 1 mrad which makes it a very efficient angular filter. RBGs are not as efficient to use as angular filters due to inferior angular selectivity. On the other hand, RBGs excel at their wavelength selectivity whose FWHM values can reach as low as 20 pm. Such high wavelength selectivity cannot be demonstrated in a TBG. This makes reflective gratings a suitable choice for use as an efficient wavelength filters. Due to their distinct properties of TBGs and RBGs, one or the other can be more suitable for different applications depending on whether wavelength or angular selectivity is required. For example, owing to their excellent angular filtering properties, TBG can be used to filter out higher order modes in a laser cavity [66]. This allows to maintain high beam quality even for high power outputs that require large mode area. In comparison, RBGs are typically used in
applications where fine spectral filtering is desirable. Such filtering was demonstrated in the beam combining experiment where two beams at wavelength separated by just several tens of picometers were spectrally combined to obtain a degenerate 2-channel laser cavity for further terahertz generation [67]. Both properties can be very beneficial for transverse and longitudinal mode selection of lasers. VBG are actively used for spectral stabilization of Raman lasers [58], they are used to produce notch filters designed for suppression of unwanted wavelengths useful for Stokes and Anti-stokes measurements. Besides, RBGs can be designed to have a grating pitch which varies continuously along the grating side. These gratings are known as chirped VBGs and they particular property is that the wavelength of the Bragg condition is varied along the grating’s side in accordance with the pitch. Such grating with a various pitch will stretch or compress an incident laser pulse depending on the direction of the chirp. For that reason chirped VBGs are widely used for stretching and compressing of the high power femtosecond pulses while preserving high diffraction efficiency. Lastly, TBGs can be used for beam steering applications which is possible as the angle of an incident beam can be multiplied up to 5 times upon diffraction on the grating.

All above said allows to conclude that PTR glass is a hologram recording material which is superior to the counterparts in several characteristics and allows for fabrication of robust high power stable holograms.

1.24. Challenge related to recording of complex holograms

The last sentence, however, is not completely true as the only HOEs that can be fabricated in PTR glass are planar VBGs. It appears impossible to produce complex HOEs in PTR glass using standard fabrication procedure. The reason for that is the sensitivity range of the original PTR glass which is limited to the 280-350 nm range in the UV. Hence only UV radiation falling within the designated range can be used for recording of holograms. Of high importance is the fact that these
are the wavelengths at which the hologram will be reconstructed. This can be better understood by going back to the technique of hologram formation which happens as the result of the wave interference where the intensity distribution is given by (1.5). When the hologram is reconstructed, it is illuminated with a reference beam which according to (1.5) has a complex amplitude of $a_1$. Intensity distribution just after the surface of the holographic plate can be then written as

$$w = a_1 I = a_1 (I_1 + I_2) + a_1 a_2^* + I_1 a_2$$

(1.42),

where the first term represents the intensities of the original beams, second is the interference term, and the last term is proportional to the original signal wave reconstructed by the hologram. This original beam can be reconstructed only by a reference beam with the same complex amplitude $a_2$ as the original recording beam. It can be inferred that a hologram can be fully reconstructed only if illuminated by the beam at the same wavelength and angle as in the recording configuration. In PTR glass the holograms are typically recorded using the UV radiation from a HeCd laser operating at the wavelength of 325 nm, and hence this is the wavelength at which the hologram can be reconstructed. Holograms for that spectral range, however, are not of the utmost interest since most applications for HOEs are found in the visible and IR spectral regions.

Meanwhile, VBGs that are also recorded by UV radiation, can operate in the whole transparency range of PTR glass. It follows that there is a profound difference in the production of VBGs and other planar structures and complex volume holograms. The difference is that a VBG can be recorded using indirect method which simply means that a hologram is reconstructed in the configuration which is different from the recording configuration. That is possible since a grating can be obtained by interference pattern of two plane waves. In terms of the expression (1.5) it means that both $a_1$ and $a_2$ are simple planar wavefronts. From (1.42) it follows that this hologram does not require exactly the same configuration and can be reconstructed with any planar wave.
that results in constructive interference which in the case of volume holograms is given by the Bragg condition. In practice it means that a grating can be designed to operate at different wavelengths by targeting an appropriate grating pitch. The latter, as is known to be determined by the intersection angle of the two beams. The technique can be applied to both TBGs and RBGs as can be easily seen from the Fig. 14 where an arbitrary grating is shown. Purple arrows in the picture represent the UV beams that the grating is recorded with, and changing the angle between the beams will result in different grating pitch. When the grating is fabricated, it can be used as either TBG or RBG depending on the direction of the reconstructing beam. Green arrows show the grating operation in transmission geometry (TBG) while orange arrows demonstrate how grating can be used as an RBG. Although in the real world a grating can rarely serve both purposes as grating pitch is much smaller for an RBG compared to a TBGs operating at similar wavelengths. Nevertheless, without getting into specifics it can be inferred that an indirect recording method allows to use one UV source to fabricate VBGs for operation at any desirable wavelength.

Figure 14 Recording and reconstruction geometry of arbitrary VBG.
The method, however, does not apply when at least one of the wavefronts is not planar. This is the reason why the standard recording procedure used in PTR glass cannot be used for fabrication of complex holograms for application in the visible and IR spectral regions. Complex holograms must be fabricated using the direct approach and thus have to be recorded using radiation at the exact wavelength as they will be later operate at. It can be inferred that the method has to be found to record HOEs in PTR glass using visible or IR radiation.

Development of a method for recording of complex holograms in PTR glass with long wavelength radiation has been the main goal in this work. As a more specific objective, a decision has been made to concentrate on the visible range situated around the wavelength of 532 nm as a target range for holographic recording. The choice was justified by the prevalence of laser sources at the wavelength of 532 nm (second harmonic of a Nd:YAG crystal resonance) and, as a consequence, a variety of application designed around that wavelength. Besides, the blue-green spectral region is in the vicinity of the UV sensitivity range of the original PTR glass, and the blue-green photons provide reasonable amount of energy that can be used for photoexcitation. Thus, the issue of recording using visible light has been addressed in the present work. The next chapters explain the methods of recording with visible light and show how the invented method can be employed for producing complex holograms in PTR glass. There have been two different approaches laid out for visible light recording. First approach involved modification of the original PTR glass with the purpose of its optimization for visible light recording. The holograms could then be recorded in a modified PTR glass by visible light. By contrast the second approach is based on the original PTR glass where, however, the recording procedure is modified accordingly. Ultimately, both methods were shown to allow for recording of holograms with visible light, including complex HOEs. Despite very different nature of the underlying effects and different
mechanisms, the holographic techniques used for fabrication of holograms with visible light were found to be quite similar and presented similar challenges for a holographer. These challenges as well as the specifics of visible light recording are discussed in greater detail in the succeeding chapters.
One approach undertaken to allow for visible light recording was to create a new glass which was photosensitive to the visible light. In this case the composition of the original PTR glass was modified in order to obtain the desired properties. This chapter describes the development of a modified PTR glass with justification of the conducted modifications, description of the fabrication process and characterization. To understand the concept of modification, the photosensitivity mechanism of the original PTR glass has to be first fully understood. The photosensitivity of PTR glass, as was discussed in the previous chapter, is based on its sensitizer, a trivalent cerium ion which has absorption ranging from 280 to 350 nm with a maximum at 305 nm. Illumination with the UV radiation with the wavelength in the region of 305 nm results in the excitation of the 4f-5d transition band of the rare earth dopant. The latter, in turn, results in so called interconfigurational transition of the Ce\(^{3+}\) ion where the latter is converted to the Ce\(^{4+}\) state and an electron is released. As already known from previous chapter, the mobile electron later triggers the chain of structural transformations ultimately resulting in the refractive index change. However, at that point the way the 5d band is excited is of higher concern. Excitation of the 5d band of a cerium ion requires a photon energy of around 4 eV. Reasonable assumption would be that choice of another rare-earth dopant with lower excitation energy would allow to use lower energy photons and thus shift the photosensitivity range to longer wavelengths. Yet, this does not appear possible since Ce\(^{3+}\) has the lowest 4f-5d transition energy of all the rare earth elements [68]. Hence there is no other dopant which could be photoexcited using a low energy photon. This, however, is only true for linear excitation by a single photon. Meanwhile, 4f-5d transition can be also excited with multiple photons with the use of intermediate levels. This process is known as upconversion, and in that
case energy of each of those photons can be drastically lower than of a single exciting photon. There are rare earth materials that allow for excitation of a 5d band using such multi-photon processes. The choice of a rare-earth material suitable for the task will be discussed later in the chapter. Yet, before that it is reasonable to understand the basics of upconversion and see how this process can be used to achieve visible light photosensitivity.

2.1. Upconversion theory

The term upconversion was first used by Bloembergen in 1959 where it was used in an IR photon counter [69]. The term is closely related to upconversion photoluminescence where the wavelength of emitted light is shorter than that of exciting light. In other words low energy photons are absorbed and then high energy photons are emitted as a result of an upconversion process. In such fashion upconversion is considered an Anti-Stokes process, and it is a non-linear process as well. There are several different mechanisms of excitation having a common property which is that more than one photon is absorbed per one photon emitted.

Excited state absorption (ESA) is the first and the simplest mechanism which is illustrated in the Fig. 15 [70]. The ion initially in the ground state 1 absorbs the first photon and gets excited to an excited state 2, which has to be a metastable level. Then the next absorption process takes place from the excited state, so the second absorbed photon excites to the upper level 3. Consequent emission takes place from the level 3 and has the higher energy than that of an absorbed photon. Often more steps and more intermediate levels are involved in the ESA. Sometimes the level the ion is excited to is not metastable. In that case the absorption process leads to an excited state from where the system decays to a metastable level. This type of ground state absorption is commonly referred to as non-resonant ground state absorption as opposed to resonant state absorption which is if a direct transition from level 1 to level 2 takes place.
It should be noted that ESA is a probability process and its efficiency is dependent on the lifetime of the particular metastable level [71]. If lifetime of the metastable level used in the upconversion is reduced so is the efficiency of the upconversion process. In that case the ion in the excited state is more probable to undergo a non-radiative decay before a second absorption process takes place. The processes that cause a metastable level lifetime to decrease are known as quenching processes and are often related to multi-phonon transitions [72]. The presence of additional energy levels located below the metastable level increases the probability of a multi-phonon transition to happen. In fact the probability of such transition between two levels depends on the energy gap between the levels and decreases exponentially as the gap increases according to the formula [73]

\[ R = R_0 e^{-\alpha \Delta E} \]  \hspace{1cm} (2.1)

Consequently if no levels are present between the metastable level and the ground state the probability of multi phonon transitions is low and ESA can be very efficient. However there are several other upconversion mechanisms as well that might have higher efficiency.

\[ \text{Figure 15 Upconversion processes [70].} \]
Besides sequential absorption there are several upconversion mechanisms involving energy transfer between nearby ions [70]. In such processes an ion which loses energy is normally called sensitizer, whereas the one who receives energy is an activator. Energy transfer upconversion (ETU) is one of the processes that requires an energy transfer. It is the most efficient upconversion process which often has higher probability compared to ESA [71]. The process is outlined in the Fig. 2.1b and affects three ions, one activator and two sensitizers. Each of the sensitizers first absorbs a photon and thereby gets excited to the intermediate metastable level 2, while the third ion stays in the ground state. Then the energy transfer happens between the sensitizers and the activator. Two ions in the excited state transfer their energy to the third ion in the ground state followed by excitation of the activator to the upper excited state 3. Activator then undergoes a radiative decay as it emits a high energy photon and returns to the ground state.

Cooperative sensitization (CS) also known as cooperative upconversion is an upconversion process similar to ETU [70]. The process is shown in the Fig. 2.1c and similarly to ETU involves an energy transfer between an activator and two sensitizers. However unlike ETU CS uses a quasi-virtual intermediate level instead of a real one. Consequently the energy transfer and excitation of the activator are considered to be concurrent processes in CS as opposed to ETU where they are consequent. Hence CS has significantly lower efficiency compared to ETU which employs resonant absorption [75].

ETU introduces an example of an advantageous use of energy transfer allowing for increase in upconversion efficiency [76]. Besides, energy transfer is associated with such process as cross relaxation [70]. The process where two nearby ions undergo non-radiative transitions in opposite directions as a result of energy transfer is called cross-relaxation (CR). A distinct upconversion mechanism can be based on CR as it is illustrated in the Fig. 2.1d. Two ions: one sensitizer and
one activator both excited to the metastable level 2 experience CR whereby the activator is excited into the upper state while the sensitizer returns to the ground state. The activator then undergoes a downward transition where it emits a high energy photon.

Often CR takes place within another upconversion mechanism such as photon avalanche (PA) upconversion which normally happens at high pumping intensities [70]. The diagram of the process is presented in the Fig. 2.1e [77]. The intermediate level 2 is first populated by non-resonant ground state absorption. ESA then takes place from the intermediate level and leads to some level 3’, from where the ion decays to level 3. Until this points PA closely resembles an ESA process described above. The core feature of the PA process is CR which happens thereupon between the ion at the level 3 and an ion in the ground state, resulting in both ions to be found at the level 2. As relaxation processes are much faster than other processes mentioned here, the population of the intermediate level will yet increase thus increasing consequent ESA events in an avalanche manner.

There are two nonlinear processes namely two photon absorption (2PA) and second harmonic generation (SHG) That might result in a high energy output. However those should be distinguished from the upconversion processes mentioned above as 2PA and SHG do not utilize an intermediate level [78]. The probability of those processes relies on the probability of two photons being simultaneously absorbed and thus 2PA and SHG have very low efficiency. Those processes should be only considered in the case of very high pump intensities. Summarizing all the mentioned upconversion schemes it bears repeating that ESA and ETU are among others the most efficient and thereby most commonly observed upconversion processes. Those were the processes encountered in the course of experimental work and will be described in greater details below.
Any upconversion process is a probability process and hence its rate is an important figure of
merit. Typically, upconversion rates are assessed by intensity of luminescence which succeeds the
multi-step absorption process. The parameter is known as the luminescence efficiency and it is
determined by the amount of luminescence light obtained from a certain amount of pump light
[79]. Ratios of intensities of emission to excitation are often quite small for upconversion
luminescence. Thus efficiency of the process is an important parameter to assess whether an
upconversion process is worthwhile for any practical applications. In fact luminescence efficiency
depends on pump power and the dependence is another important property of an upconversion
process. An n order process for n consecutive photon absorption steps that it takes to reach the
emitting state is normally found to have a $P^n$ type dependence for low pump power. However it is
worth noting that the slope of the dependence is generally not constant and decreases as pump
power goes up. It was shown in [80] that for an n order process the slope decreases from $P^n$ at low
pump intensity down to $P^1$ as it saturates at high pump intensities. The two limiting cases
correspond to infinitely low and infinitely high upconversion rates respectively. The decrease in
slope can be accounted for competition between nonradiative relaxation and upconversion
processes for depletion of an excited state. Specific type of upconversion process namely ESA or
ETU was also demonstrated to be possible to distinguish based on the dependence slope at low
upconversion rates. Hence the power dependence of upconversion luminescence can provide
important information on the process, the efficiency however is primarily depends on the medium
where the process is implemented. After everything mentioned about the upconversion
luminescence, it has to be kept in mind that luminescence would be a possible side effect if at all
observed whereas transition to a different valence state is a primary goal in the present work. Power
dependencies described above can be however used for estimation of the upconversion rates. In
the above paragraphs we became familiar with different upconversion schemes and mechanisms. Upconversion process is typically implemented using a rare-earth ion whose properties are favourable for higher upconversion rates. In the next section a review of the concept and main properties of rare-earth elements will be given, and different types of transitions that ions undergo will be discussed in relation to their application for upconversion.

2.2. Properties of Rare Earths

Rare earth elements or rare earth metals include 15 elements in the periodic table with atomic numbers from 57 to 71 that are known as lanthanides and two other elements: scandium and yttrium. All these elements belong to the rare-earths mostly for the reason that all of them are produced from the same ore deposits in the nature. However, it is the first 15 elements, the lanthanides, that are of particular interest since those elements possess very high similarity with each other in their physical and chemical properties [81]. Similarity begins with the electronic configuration which for neutral lanthanides is given by \(4f^{n+1}, 5s^2, 5p^6, 6s^2\). One \(4f\) electron and two \(6s\) electrons can be released resulting in a formation of a trivalent lanthanide ion. Trivalent state is prevalent in oxides since it is the most stable. Some rare earth oxides may however contain various valences of the same element as it is for example with terbium oxide \(\text{Tb}_4\text{O}_7\) which contains both \(\text{Tb}^{3+}\) and \(\text{Tb}^{4+}\) ions. Besides, some elements have similar energy of electrons at \(4f\) and \(5d\) subshells, in which case a \(4f\) electron might be promoted to the \(5d\) subshell resulting in a tetravalent rare-earth ion. The transition is known as interconfigurational, and it is the one that happens in \(\text{Ce}^{3+}\) under UV illumination with consecutive creation of a free electron.

Another important property of rare-earth elements is limited extension of the \(4f\) shells, and as a result, this shell cannot participate in the covalent interaction with nearby ions [82]. Hence the only types of bonding that occur for rare-earth ions are ionic or electrostatic bonding. \(4f\) shell is
also well shielded by the outer-lying shells from neighboring atoms such as those of the host lattice. Since most electron transitions in the rare-earths occurs within the 4f shell, those are well isolated from the influence of the host lattice. As a result, a rare-earth element will exhibit very similar properties across the wide range of host lattices. Properties include energies of the transitions undergone by rare-earth ions, and those are going to be reviewed next.

When a rare earth ion absorbs a photon, the energy is transferred to the electron, which can be transferred to a high energy orbital [82, 83]. Thus, the ion undergoes an upward transition effectively leading to an excited state. There are three types of transitions the lanthanide ion is involved in: sharp f-f transitions, broad interconfigurational 4f-5d, and charge transfer transitions [84]. Some types of transitions are permitted while the others are forbidden due to selection rules. Parity selection rule, for instance, implies that an electronic dipole (ED) transition cannot lead from one state to the other state which has the same parity [82]. Hence f-f ED transitions are forbidden, whereas magnetic dipole are allowed. However, the efficiency of the latter is low and so is that for allowed quadrupole transitions, hence, none of the two is normally observed.

Yet while the statements above hold for a free ion, they are not necessarily true if the ion is affected by a local environment. In some types of host lattices such as glass the presence of opposite parity states in the 4f band might be expected. In that case despite the selection rule ED transitions inside the 4f band become possible. Those are known as induced electric dipole transitions. However, the efficiencies of those transitions are fairly low and that is why absorption bands of f-f transitions are very narrow. Judd-Ofelt theory is used as a general rule for calculation of an exact transition probability value [85]. Other selection rules such as that requiring constant spin for the transition also apply. Meanwhile, in glasses those conditions are relaxed resulting in possibility of the transitions inconsistent with the selection rules. In such environments forbidden
transitions are commonly referred to as having low probability as opposed to high probability of
the allowed ones. Owing to the low efficiency of f-f transitions an ion excited to an upper state
will be found in that state for a long period of time before it decays. This explains long metastable
level lifetimes in lanthanides which is the fundamental reason for their use in upconversion.

Interconfigurational 4f-5d transitions are allowed according to the selection rules, and, hence,
their absorption bands are broader than those of f-f ones [82]. 4f-5d transition energies of
lanthanides will be discussed below, yet at this moment it is worth noting that Ce$^{3+}$, Pr$^{3+}$ and Tb$^{3+}$
are the only elements having the transition energy equal to or less than 5 eV [68]. These are the
transitions that are normally observed. The other rare earth elements have higher transition
energies which makes them unlikely to be experimentally observed. Cerium has the 4f-5d
transition with the lowest energy which provides advantages for its use as a sensitizer in PTR glass.

When electron is transferred from an orbit pertained to a lanthanide to another orbit situated
on another ion bonded to the lanthanide, that transition is called a charge transfer (CT) transition
and the ion is known as a ligand. Above said corresponds to Metal-to-Ligand Charge Transfer
transition (MLCT), whereas the reverse transition is called Ligand-to-Metal Charge Transfer
(LMCT) [82]. Similarly to 4f-5d transitions, CT transitions are allowed and have broad absorptions
bands. In the same fashion the transition energies are sufficiently high and therefore the only
transitions observed are Eu$^{3+}$ and Yb$^{3+}$ LMCT transitions. Of three types of transitions discussed
f-f and 4f-5d interconfigurational transitions will be mentioned in the present report as they both
contribute to the photosensitization process.

2.3. Choice of Rare Earth dopant

The above paragraphs explain why rare-earth ions are an optimal choice as a sensitizer for use
in a photosensitive glass. First of all, they have a 4f-5d transition which releases a free electron
and which can be excited with optical radiation. At the second, as a result of the low probability of the 4f-4f transitions the upper-state lifetimes can be substantially long. Hence, those upper-state levels can be used as an intermediate state for multi-step photoexcitation of the glass via upconversion. Meanwhile it appears that not all the lanthanides are suitable for upconversion photoexcitation. The choice of an appropriate rare-earth element requires an appropriate combination of 5d transition energy and the level structure. 5d-4f transition energy has to be low enough so it were equal or less then the combined energies of two visible photons involved in the process. Meanwhile, the energy diagram of the appropriate ion has to provide a convenient energy level for use as an intermediate state. Despite the wide selection of rare earth dopants available for use in upconversion processes, as stated above, only few ions were found to have suitable 5d-4f transition energies. Values of the lowest 5d energy states were predicted based on analyzing the absorption and emission spectra in [68] and [86]. Energies of 5d-4f transitions are given for each lanthanide element in the Fig. 16. It can be inferred that 5d band of Ce$^{3+}$ has the lowest energy of about 4 eV accounting for its use as a sensitizer in PTR glass. Successive lowest energies can be found for Pr$^{3+}$ and Tb$^{3+}$ both having a value of about 5 eV. Pm, Sm and Dy succeed with the closely spaced energy values residing above 6 eV. High 5d energy values require high energy pump and signal light to allow for photoionization by two step upconversion or involve three or more step upconversion processes being of less interest due to lower probabilities. Use of high energy light conflicts with the main idea of the work as to shift the photosensitivity region towards long wavelengths. For that reason only the rare earth elements with 5d energies below 6 eV specifically Ce$^{3+}$, Pr$^{3+}$ and Tb$^{3+}$ were taken into consideration.

The three ions were then compared in regard to their capacity of upcoversion photosensitization of 5d band. Upconversion capabilities can be deduced from the energy level structures of the ions
that are given in a parity plot known as the Dieke diagram presented in the Fig. 17 [87]. Thus it can be inferred from the diagram that upconversion in Ce$^{3+}$ is encumbered due to lack of long lifetime metastable levels. Upconversion luminescence as a result of three photon absorption process has been reported in Ce$^{3+}$ and was implemented with a femtosecond laser [88].

Unlike cerium, Pr$^{3+}$ has an energy level structure which is suitable for upconvertion, and the latter has been extensively studied in regard to the use of praseodymium in upconversion lasers. Specifically blue and orange upconversion emission has been reported in Pr$^{3+}$ doped systems under IR-excitation [89]. As can be seen from the diagram energy level, scheme of Pr$^{3+}$ is suitable for upconversion photoexcitation of the 5d band. In this respect of interest is the $^{1}\text{D}_2$ level which has energy of about 2.1 eV which corresponds to the 588 nm yellow light. If system is pumped by yellow light of the indicated wavelength then consequent ESA might take place from the $^{1}\text{D}_2$ level and a signal wavelength of about 450 nm is needed for 5d band excitation. However despite convenient level structure there are technical difficulties preventing from implementation of the scheme. High absorption at wavelength corresponding to the signal light is one of the obstructions.
Absorption spectrum of Pr$^{3+}$ is given in the Fig. 18 [90] and it can be concluded from the spectrum that use of blue light at 450 nm as signal light is encumbered due to large absorption in that region. Besides, $^1D_2$ level has been reported to have lifetime of about 100 μs which might be insufficient for the above-noted upconversion scheme. Short level lifetime can be accounted for dense spacing between the $^1D_2$ and underlying levels. As for energy levels located above $^1D_2$ state in the energy diagram they have even shorter lifetimes and consequently the upconversion process will be less efficient. For the reasons above Pr$^{3+}$ and Ce$^{3+}$ did not meet the requirements for efficient upconversion and therefore could not be used as active ions.

Figure 17 The Dieke diagram [87].

Tb$^{3+}$ is the most promising material for upconversion photosensitization of PTR glass for several reasons. First, as stated above its 5d energy band has sufficiently low energy of 5 eV. Besides, the long lifetimes of metastable levels are considered the fundamental advantage of the
ion over other RE materials. Trivalent terbium has two main metastable energy levels, namely $^5\text{D}_3$ and $^5\text{D}_4$, that are of interest for upconversion (see Fig. 17). $^5\text{D}_4$ level is separated from the $^7\text{F}$ ground state multiplet by a large energy gap of about 1.75 eV. Hence probability of multi phonon transitions from the level is sufficiently low so that long level lifetime can be expected. In fact, lifetime values of several milliseconds have been reported for both $^5\text{D}_3$ and $^5\text{D}_4$ states in various host materials [87]. Long lifetimes, in turn, allow for efficient level population resulting in higher ESA probabilities. $^5\text{D}_3$ state has energy of about 3.3 eV corresponding to 380 nm wavelength, while $^5\text{D}_4$ level can be excited with 480 nm radiation leading to a 2.6 eV transition from the ground state. Both levels can be used for upconversion photosensitization. ESA from $^5\text{D}_3$ and $^5\text{D}_4$ levels can be performed by optical radiation of the red to IR and the blue-green spectral regions respectively. Here another important advantage of Tb$^{3+}$ as an active ion has to be taken into account. The advantage lies in low absorption in the region corresponding to prospective signal light. Tb$^{3+}$ does not introduce any significant absorption in the 0.5-1.5 μm range into the glass which allows for use of high power signal sources [91]. As upconversion process is power dependent, the use of high power sources gives an opportunity to obtain photosensitization even under the conditions of low-probability upconversion transitions. Therefore trivalent terbium appears to be a single option for use as an active ion and for that reason it was chosen as a dopant for fabrication of a new PTR-type glass with extended photosensitivity region.
2.4. Novel Tb$^{3+}$ doped photo-thermo-refractive glass

A novel Tb$^{3+}$ doped PTR glass was fabricated, characterized and experimentally investigated in order to determine how far it was possible to extend the photosensitivity range of the original PTR glass. The fabrication of glass was carried out by following the instructions explained in details in the reference [60]. Two types of glass containing different amount of Tb$^{3+}$ dopant were fabricated to study the photosensitivity by means of upconversion. An atomic concentration of 0.7% of the dopant was used in the glass of the first type, while the other type contained less dopant at 0.08 at%. The indicated concentrations were chosen for the reason of distinct optical properties exhibited by the glass at two different Tb$^{3+}$ concentrations. For convenience the two types of glass will be hereafter referred to as Tb0.08 (0.6w%) and Tb0.7 (6w%) glasses. The terbium concentration effects on optical properties will be discussed in greater details below.

A detailed analysis of the glass composition is beyond the scope of this report. Nevertheless it is worthy of note that the Tb-doped PTR glass had the composition similar to that of a regular PTR glass with one important remark. No Ce$^{3+}$ doping was used in the new glass as the residual absorption of cerium overlaps with the absorption of Tb$^{3+}$ in the region of 380 nm. Hence excitation of the glass will result in refractive
index change (RIC) due to both sensitizers. Refractive index due to excitation of cerium is produced by UV radiation and is of no interest for the present work. Furthermore, refractive index change due to UV radiation will present an unwanted background reducing the effective amplitude of the refractive index change due to visible light. Thus all the photosensitivity in the glass is due to terbium doping and can be characterized by the refractive index modulation obtained as a result of the exposure. Both Tb0.08 and Tb0.7 glasses were characterized before photosensitivity experiments in order to study their structural, thermal and optical properties. The knowledge of glass properties was required for competent experiment design.

2.5. Characterization of Tb$^{3+}$ doped PTR glass

2.5.1 Absorption spectrum of Tb$^{3+}$ doped photo-thermo-refractive glass

UV-VIS absorption spectrum of the Tb$^{3+}$ doped PTR glass was measured using a Perkin Elmer Lambda 950 spectrometer. The measured spectrum of the Tb0.7 is presented in the Fig. 19. The UV-VIS spectrum was found to contain several absorption lines corresponding to the 4f-4f transitions with the peaks at the following wavelengths: 485, 378, 369, 352, 338 and 317 nm. While some peaks correspond to single transitions, the other contain two or more transitions that are overlapped due to inhomogeneous broadening [91, 92]. According to [93], 485 nm absorption peak is assigned to a transition from the $^7F_6$ ground state to the $^5D_4$ state. In the meantime the 378 nm peak corresponds to the transitions from the ground state to $^5D_3$ and $^5G_6$ energy levels. Two transitions cannot be discerned as a result of inhomogeneous line broadening in glass which was found to be larger than the 26 meV energy gap between the levels. The peaks at the shorter wavelengths are related to the upward transitions to the following levels or level groups: 369 nm - $^5L_{10}$; 352 nm - two states $^5L_9$ and $^5G_4$; 338 nm - two states $^5G_2$ and $^5L_6$; 317 nm - three states $^5H_7$, $^5D_1$ and $^5D_0$. 

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The IR absorption spectrum was measured by means of a Perkin Elmer Optica 100 FTIR spectrometer where several absorption peaks were detected. The peak with the highest energy in that spectral region was observed at 1903 nm and was found to have a spectral width of 248 nm. The peak corresponds to the transitions within the ground state multiplet: from $^7F_6$ to $^7F_0$, $^7F_1$ and $^7F_2$ respectively [91]. Thus no additional absorption introduced by terbium was found in the glass in the wavelength range from 0.5 to 1.5 μm.

The absorption spectrum of Tb$^{3+}$ doped PTR glass also contains 4f-5d transition lines located in the UV. As noted in [92, 94] the transitions correspond to the wavelength range from 220 to 260 nm. However, the lines at those wavelengths are not easily distinguishable as intense absorption peaks due to other dopants are also present in the area. For instance, silver as a photosensitive agent in the glass introduces absorption around 240 nm, while absorption at 270 nm is due to presence of antimony. For that reason an absorption spectrum was measured in the PTR glass matrix containing only one dopant - 2w% of Tb$^{3+}$. That glass was expected to have an absorption edge at around 220 nm due to glass matrix and no additional absorption peaks caused by the dopants. Another concern was related to the high absorption values in the region of interest. In

Figure 19 Absorption spectrum of PTR glass doped with 0.7a.%Tb.
fact, optical density of a regular 2 mm glass plate exceeds the upper limit of the spectrometer operating range in the given part of the spectrum. To resolve the problem a 1 mm thick glass plate was ground down to 300 μm in order to decrease its optical density and then the measurements were performed. The measured spectrum is shown in the Fig. 20. The spectrum comprises an absorption peak located at a wavelength of 228 nm and having a spectral width of 20 nm. The peak is related to the transition from the $^7F_6$-ground state to the 5d band in Tb$^{3+}$. Light at a wavelength of 228 nm can be used for photoexcitation of the 5d band in Tb$^{3+}$ doped PTR glass. Thus effect of various dopants on the UV-VIS absorption spectra was investigated and the experimental data was interpreted according to the available information on glass composition. That knowledge was required for assessment of the expected efficiency of a hologram recording process.

![Absorption spectrum of PTR glass doped with 2w.%Tb. (logarithmic scale).](image)

Figure 20  Absorption spectrum of PTR glass doped with 2w.%Tb. (logarithmic scale).
2.5.2 Luminescence of Tb$^{3+}$ doped photo-thermo-refractive glass

The luminescence properties of the new glass were investigated in order to estimate the efficiency of different absorption and recombination processes taking place in the glass. Both Tb0.08 and Tb0.7 glass samples were studied to estimate the effect of the dopant concentration on the luminescence properties of the glass. The luminescence spectrum was measured using the Ocean Optics S2000 spectrometer coupled to a fiber for light collection. The setup used for the measurements is shown in the Fig. 21. A beam from a 375 nm LED delivering 450 mW of optical power was focused into a 2mm spot inside the sample of Tb-doped PTR glass. UV light incident on the sample produced fluorescent light propagating in all directions. The collecting fiber was placed off the transmitted beam path so as to collect only the fluorescent light.

Luminescence spectra of both types of glass are presented in the Fig. 22 so that a comparison can be made. The luminescence spectrum of the Tb-doped PTR glass was found to be consistent with the spectra of the other Tb-doped silicate glasses [91, 93].

Figure 21 Experimental setup for measurements of luminescence spectrum of Tb-doped PTR glass.
The scheme of the luminescence process can be demonstrated using the energy level diagram of the Tb\(^{3+}\) ion shown in the Fig. 23. The energy level diagram of Tb\(^{3+}\) contains the following levels and groups of levels. The \(^7\)F\(_1\) ground state multiplet comprising 7 closely spaced energy levels is located on the bottom of the diagram with the highest \(^7\)F\(_0\) level having an energy below 1 eV. The energy gap of about 2 eV separates the ground state multiplet from the further two isolated \(^5\)D\(_3\) and \(^5\)D\(_4\) levels. The latter appear to be metastable due to large energy separation and thus they are of a particular interest for use in upconversion scheme. The upper metastable \(^5\)D\(_3\) state has energy of 3.3 eV corresponding to the upward transition from the ground state induced by the light at 378 nm. It was mentioned before that the \(^5\)D\(_3\) level cannot be separated from an adjacent \(^5\)G\(_6\) level due to small energy gap between the levels. An upward transition to the lower \(^5\)D\(_4\) level can be induced by the photon with energy of 2.6 eV at 485nm. The upper group of closely spaced 4f energy levels is situated just above the \(^5\)D\(_3\) and continues up to 4 eV. Those levels are of less concern for the experiment design due to their short lifetimes and high energy. As stated above the 5d band of Tb\(^{3+}\) ion is located at about 5eV.

Figure 22  Luminescence spectra in PTR glass with 0.08a.%Tb and 0.7a.%Tb by 375 nm light excitation.
As a result of the 375 nm light excitation the electron population is eventually found in the upper metastable \( ^5D_3 \) state. Luminescence follows from the \( ^5D_3 \) to the \( ^7F_j \) ground state multiplet states. Besides, a portion of electrons non-radiatively decay to the lower \( ^5D_4 \) level leading to subsequent luminescence from \( ^5D_4 \) to \( ^7F_j \) levels. Hence the luminescence spectrum consists of two groups of lines corresponding to the transitions from the upper and the lower metastable levels. Blue emission can be observed due to transitions from \( ^5D_3 \) level that take place in the range from 380 to 480 nm with the most intense line at 398 nm. Downward transitions from \( ^5D_4 \) state result in
green luminescence in the wavelength range from 485 to 620 nm. The most efficient green luminescence takes place at 545 nm and is due to $^5\text{D}_4 \rightarrow ^7\text{F}_5$ transition in Tb$^{3+}$.

Relative intensities of blue and green luminescence are subject to dopant concentration [93, 95]. Intensities of the most efficient blue and green transitions at 398 and 545 nm respectively are plotted against Tb$^{3+}$ concentration in the Fig. 24. While blue emission prevails at low dopant concentrations, it is eventually attenuated due to concentration quenching. Green luminescence is then enhanced at the expense of the blue at higher dopant concentrations. Quenching of blue luminescence in Tb$^{3+}$ doped glasses can be accounted for the cross-relaxation process between adjacent terbium ions. The cross relaxation process comes into effect when Tb$^{3+}$ concentration reaches the value of 0.5w% which corresponds to the 20Å distance between the neighbouring ions. One terbium ion initially in the $^5\text{D}_3$ state experiences a nonradiative decay as a result of an energy transfer to another ion originally in a ground state which undergoes an upward transition within the ground state multiplet. Efficiency of the cross-relaxation process increases with the dopant concentration and consequently decreases the lifetime of the upper metastable $^5\text{D}_3$ level. Thus most of the population of the $^5\text{D}_3$ state non-radiatively decays to the $^5\text{D}_4$ state and thereupon the green luminescence takes place from the lower metastable level.

The measurement results appear to be consistent with the stated above regarding the relative intensities of blue and green luminescence in both cases. Here the ratio of intensities of the most intense green and blue transitions is more than ten times for the Tb0.7 glass. In comparison, the same peaks have intensity ratio of 1.6 in the Tb0.08 glass. It can be inferred that blue luminescence is quenched due to cross-relaxation process in glasses with higher terbium concentration. An important consequence is that the lifetime of the upper state is significantly reduced in the Tb0.7 glass, hence the possibility of using that state for upconversion in unlikely. Nevertheless, the
lifetimes of lower and upper metastable levels were measured in both types of glass in order to estimate the probability of the ESA from the levels.

Experimental data obtained from the luminescence spectra was verified and compared with luminescent kinetics characteristics. The luminescence decay technique was used to measure the lifetimes of $^5D_3$ and $^5D_4$ metastable levels. The measurement setup is shown in the Fig. 25. A 20ms UV pulse from the 375nm LED is focused onto a glass sample. As shown above, that type of excitation results in population of the $^5D_3$ level followed by luminescence from the upper and the lower metastable levels. Luminescent light was focused onto a Thorlabs PDA100 photodetector connected to the oscilloscope, and the signal decay time was detected. An LP400 longpass filter from Thorlabs was used in order to cut off the scattered UV light. The signal measured in such fashion contained combined blue and green luminescence. Besides, additional filters were used to

Figure 24  Variation of green and blue luminescence intensities with Tb$^{3+}$ concentration in calcium aluminosilicate [93] glasses [73].
detect luminescence of each type distinctly. Here an SP500 shortpass filter was employed to detect blue luminescence only by cutting off green light. In contrast an LP500 longpass filter allowed for detection of green luminescence only. The lifetime due to combined luminescence was measured to be 3.7 and 3.8 ms for Tb0.08 and Tb0.7 glasses respectively. The distinct measurements of the Tb0.08 glass gave the upper $^5D_3$ level lifetime of 2.9 ms and the lifetime of the lower $^5D_4$ level was found to be 4.6 ms. The distinct measurement were not possible to conduct for the Tb0.7 glass for the reason of negligibly low efficiency of the blue luminescence due to concentration quenching effects. It can be inferred from the measurement results that both $^5D_3$ and $^5D_4$ levels can be employed for upconversion in Tb0.08 glass. However, in Tb0.7 glass only the lower $^5D_4$ metastable level can be employed for upconversion owing to insufficiently long lifetime of the $^5D_3$ level. The upper level cannot be used as an intermediate state for ESA in Tb0.7 glass.

Figure 25 Experimental setup for measurement of metastable level lifetime in Tb-doped PTR glass.
2.5.3 Excitation spectrum of green light luminescence in Tb$^{3+}$ doped photo-thermo-refractive glass

Location of a 4f-5d transition can be also determined from the excitation spectrum of 545 nm green light emission in the glass. The PTR glass matrix containing 2 w% of Tb$^{3+}$ was again studied to avoid distortion of the spectrum by additional luminescence due to possible energy transfers from other dopants. A Perking Elmer fluorimeter was employed for measurements of the excitation spectrum. As the 5d band is excited by the UV light it eventually decays to one of the metastable levels followed by a transition to one of the levels of the ground state multiplet attended with emission of visible light. Efficiency of the most intense $^5$D$_4$-$^7$F$_5$ transition leading to green light luminescence was detected as the excitation wavelength was scanned from 200 to 280 nm. The resulting excitation spectrum is shown in the Fig. 26. A broad peak of about 50 nm width centered around 250 nm wavelength can be seen in the spectrum. The peak is believed to be present due to the luminescence after excitation of the 5d band in the glass.

![Figure 26 Luminescence spectrum of PTR glass matrix doped with 2w.%Tb$_2$O$_3$. Arrows indicate end point of the transition from $^5$D$_4$ state when exposed to light at 522 nm (1), 449 nm (2) and 375 nm (3).](image)

The location of the 5d band obtained from the excitation spectra was found to be inconsistent with that available from the absorption measurement, since the location of the same band is shifted
towards longer wavelengths in the excitation spectrum with respect to the absorption spectrum. The inconsistency is likely to be accounted for the effect of surface absorption. Here intensity of light coming from the fluorimeter lamp is very low and consequently the light is mostly absorbed in the surface region of the sample. Carriers excited as a result of such absorption in the surface region are likely to undergo a non-radiative relaxation due to surface recombination. Thus efficiency of consecutive luminescence from the surface is reduced owing to the recombination with surface defects. However light at longer wavelengths can penetrate deeper into the material. Such system will exhibit higher quantum efficiency since it is less affected by surface defects. As a result, the luminescence efficiency is higher when light is absorbed in the thicker layer. For that reason the peak corresponding to maximum absorption of the 5d band is shifted towards longer wavelengths in the excitation spectrum. Thus the band location obtained from the absorption spectrum is perceived to be the correct one and provides the wavelength range which can be used for photoexcitation. It follows that the glass can be photoexcited using UV light in the wavelength range from 200 to 250 nm while light at about 228 nm provides the most efficient absorption. It has to be noted that photoexcitation based on an upconversion process which uses multi-step absorption of low energy light is also possible and is demonstrated below in this report.

2.5.4 Structural characterization of Tb$^{3+}$ doped photo-thermo-refractive glass

Structural characterization of the novel Tb$^{3+}$ doped PTR glass was carried out in order to understand the specifics of the crystallization mechanism in the glass with modified composition. It has to be taken into account that Tb:PTRG is based on the regular PTR glass whose photosensitivity mechanism was thoroughly studied. The refractive index decrement in the regular PTR glass is produced by presence of NaF crystals with the size of around 20 nm that precipitate inside the glass medium after exposure and thermal treatment. The crystalline content in Tb:PTRG
was studied in comparison with the regular Ce$^{3+}$ doped PTR glass to determine whether Tb$^{3+}$ doping affects the mechanism of crystal growth or properties of the crystalline phase.

Such comparative analysis is complicated to perform due to drastically different optical and thermal properties of the two glasses. First, as it will be seen in the following paragraphs, the Tb:PTRG was measured to have a transition temperature of 480°C which is by 10 degrees higher than the regular PTR glass. As a consequence, the thermal development procedure used for the Tb$^{3+}$ doped glass is also different. It will be seen later than the photosensitivity magnitude of the glass is also significantly lower than that of a regular PTR glass. It follows that it was impossible to compare two glasses with similar exposure and development parameters. Instead, samples of different glasses with similar induced refractive index values were compared. PTR glass samples were first exposed to the uniform UV radiation at the wavelength of 325 nm with the energy densities of 172 mJ/cm$^2$ and 348 mJ/cm$^2$. The samples were than nucleated and developed for 60 minutes at the temperature of 515°C which resulted in the RIC values measured to be 204 and 408 ppm, respectively. The samples of Tb:PTRG were exposed to UV radiation from the 375 nm LED with energy densities of 8.5 kJ/cm$^2$ and 38 kJ/cm$^2$. Nucleation and the subsequent thermal development of the samples, performed for 120 minutes at 515°C, produced RIC values of 197 and 399 ppm, respectively.

The glass samples were then transformed into powder whose X-ray diffraction (XRD) was measured at room temperature using a Rigaku MiniFlex II with a CuKα source. The glass powder was compressed into a 2 mm deep holder whereupon XRD diffraction patterns were recorded as 2Θ was continuously scanned in the range from 20° to 80° with the sampling interval of 0.02° and a rate of 0.2 deg/min. Fig. 27a demonstrates comparative XRD curves of the Tb:PTRG and the regular PTR glass. The main diffraction peak of NaF detected around 38.55° for all samples is
plotted in the Fig. 27b as it was normalized to the amorphous glass peak. The observation can be made that the XRD peaks are similar across the two types of glass for the samples demonstrated similar values of RIC. It can be inferred that the size and the concentration of the crystals is similar in the two glasses and the scalability of RIC with the crystalline phase content can be also applied to Tb:PTRG. Therefore, while it is evident that change of composition causes drastic change in the photosensitivity, the crystallization mechanism itself remains unchanged and no affect from Tb$^{3+}$ was detected.

2.5.5 Thermal characterization of Tb$^{3+}$ doped photo-thermo-refractive glass

DSC is a common technique for investigation of the thermal properties of crystals, glasses and polymers. The property being of primary concern was the glass transition temperature, which is about 470 °C in regular PTR glass. As glass is heated up, there is a certain temperature where a sharp increase occurs in heat capacity of the glass. The additional heat is absorbed due to vibrational motions of glass molecules that arise at high temperatures. The temperature corresponding to the increase in heat capacity of a glass is called the transition temperature. The other thermal properties of a glass such as crystallization temperature are influenced by the glass
transition temperature. The crystallization temperature is believed to rise if the transition temperature of the glass increases and thus the thermal development parameters should be adjusted accordingly.

DSC measurements were performed using a TA instruments calorimeter and were intended to identify the glass transition temperature and other characteristic temperatures of Tb-doped PTR glass. The measurements were conducted for three samples of Tb-doped PTR glass: an unexposed glass sample, a sample exposed to UV light and subjected to thermal development and an unexposed sample after thermal development. Each of the three glass plates was cut so that a tiny sample with a weight from 20 to 30 mg was obtained. The prepared sample was mounted inside the calorimeter. In the course of measurement the sample was heated, in fact the calorimeter provided uniform heating rate throughout the measurements. The amount of heat transferred to the sample from the heater at each temperature was detected.

The DSC curves for the three samples are presented in the Fig. 28. Positive Y-axis corresponds to the heat received from the sample whereas the negative denotes heat transferred to the sample. Interpretation of the measurement data is presented for the curve corresponding to the unexposed glass sample [98]. The curve comprises a dip with a minimum at about 510 °C and FWHM of 25 °C. It has to be noted that the dip in fact comprises two distinct features resulted from two different processes. The left-hand side of the dip is formed by a downward shift which is accounted for the increased heat capacity of the glass. As temperature is increased above glass transition point, more heat is pulled by the glass due to increased heat capacity. It can be seen that the change in heat flow is gradual and covers a temperature range from 450 °C to 510 °C. As a result the exact transition temperature value can be ambiguously determined. In that case it is accepted to define the glass transition temperature as the middle of the temperature interval where the shift occurs.
Thus glass transition temperature of the Tb-doped PTR glass was determined to be 480 °C. That value is 10 degrees higher than that of regular PTR glass. Inference can be drawn that the NaF crystal growth takes place at higher temperatures, consequently a thermal development program should be adjusted for the new glass.

![DSC curves for PTR glass doped with 6w.%Tb; virgin glass (1), glass after irradiation (2), after irradiation and thermal development (3).](image)

Detailed description of the thermal development routine is presented in the next paragraph. Meanwhile the right-hand portion of the curve designates the onset of crystallization. The crystallization peak however is not observed and presumable occurs at higher temperatures. It can be seen that the DSC curve of the irradiated and the non-irradiated samples after thermal treatment exhibits a similar dip, however, glass transition temperature is lower and happens to be at about 477 °C. Of interest is a peak that can be found at a temperature of 573 °C in the DSC curve of the irradiated sample. That peak is related to the induced crystallization as no such feature is found in the curve of the non-irradiated sample. As crystals start to precipitate they give away the excess energy in the form of heat which is detected by the instrument. That process accounts for the rise of the heat flow at temperatures just below 573 °C. Once NaF crystals are formed, the heat flow returns to the initial state so that the heating process continues with the same rate as before. It can

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be inferred that the induced crystallization only occurs in the areas of the glass that were exposed to UV radiation. Thus the refractive index change takes place in the irradiated areas while the rest of the area remains intact. The values of the glass transition temperature and crystallization temperature obtained from the measurement curves were used to prepare a thermal development program for the new glass. Similar measurements were carried out for the Tb0.08 glass and the DSC curves were obtained. The glass with lower Tb concentration was found to exhibit lower transition and crystallization temperature as can be seen in the Fig. 29.

![Figure 29 DSC curves for PTR glass doped with 0.6w.%Tb; virgin glass (1), glass after irradiation (2), after irradiation and thermal development (3).](image)

### 2.5.6 Structural characterization of Tb$^{3+}$ doped photo-thermo-refractive glass

Thermal treatment constitutes one of the two steps for producing of refractive index modulation. It is vital that the parameters of the thermal treatment are chosen appropriately given the known glass transition temperature to provide efficient refractive index modulation while retaining the inherent optical properties. The requirements for thermal development have been provided in the section 1.19, and are the same for terbium doped PTR glass as they are for the standard PTR glass, of course, with the exception of higher development temperatures. Briefly, both temperature and duration of development have to be above a certain value to provide sufficient amount of crystal
phase, however, it is important that both the temperature and the duration do not exceed the optimum value to avoid unwanted side effects such as scattering in the glass.

Practical implementation of the thermal development procedure is fraught with difficulties as the temperature experienced by the glass sample over time does not always directly correspond to the desired development temperature. Meanwhile, providing exact development temperatures is important to ensure that the refractive index change, and hence other properties of holograms in the developed glass are as required. Thermal development is conducted in a vacuum oven that features a high precision PID temperature controller and a thermocouple that measures temperature in the furnace. That system is designed to ensure that the target development temperature setpoint (for example, 485°C for PTR glass) is reached precisely, and the development temperature is maintained steadily at that value for the required time period (80 min for PTR glass). The challenge is introduced as the glass sample is placed in an aluminium box filled with sand which is done to provide maximum uniformity of the temperature field around the sample. The temperature inside the box is measured but is not directly controlled. This introduces a problem that the control loop is no longer closed and hence, incorrect temperature regulation is likely due to overshooting (oscillations due to underdamping) or undershooting. To improve the thermal development, loop tuning was performed using manual adjustment of P, I, and D parameters using a simple “rule-of-thumb” algorithm. Pre-heating stage was introduced into the development procedure where sample is preheated to a temperature of 450°C before reaching the target temperature. As a result, the glass has to be only heated by a small temperature amplitude of 35 °C which significantly alleviates the setpoint instability. On the other hand, the pre-heating stage does not affect the development process as the temperature of 450 °C is below the transition temperature, and hence, no structural transformation occurs in the glass at that stage. According to the transition temperature of 480°C
measured in the glass previously, development of the Tb-doped PTR glass was carried out with the following parameters: nucleation at 495°C for 80 mins and crystallization at 515 °C for 120 min. The resulting temperature curve measured for the treatment of Tb0.7 glass is illustrated in the Fig. 30. It can be seen that the setpoint temperature is reached precisely and is maintained steadily for the proper time duration.

![Temperature curve corresponding to thermal development of Tb:PTRG.](image)

**Figure 30** Temperature curve corresponding to thermal development of Tb:PTRG.

### 2.6. Photoexcitation of Tb$^{3+}$ doped photo-thermo-refractive glass by excited state absorption upconversion

ESA upconversion process based on consecutive absorption of two photons was implemented in the Tb-doped PTR glass for the excitation of the 5d band situated at 5eV energy. The type of the upconversion process used in the experiments is commonly referred to as double-wavelength
ESA upconversion and it can be beneficial if energy separation between lower and intermediate states is different from that between the intermediate and upper state. In that case upconversion is carried out using two sources emitting at two different wavelengths known as pump and signal sources. The ion initially in the ground state is excited to the intermediate state by the pump photon with energy matching that of the transition. The signal beam does not cause any transition at that stage as its energy is typically lower than required. From the intermediate state the ion is excited further to the upper level as a result of ESA by a signal photon. Here the signal photon energy should match the energy difference between the upper and the intermediate levels. ESA by another pump photon might take place as well leading to above situated energy levels if the latter are present. However it is often desirable to minimize the efficiency of that process, and therefore signal beams typically have sufficiently higher intensities compared to pump beams.

Implementation of the double-wavelength ESA upconversion in Tb-doped PTR glass is shown in the Tb\(^{3+}\) energy diagram in the Fig. 2.17a. A UV light at the wavelength of 375 nm was used as a pump to excite the ion to the metastable \(^5\)D\(_3\) state in both glasses. However successive process advancement happened differently for Tb0.08 and Tb0.7 glasses. ESA in Tb0.08 glass took place from the upper \(^5\)D\(_3\) state as well as from the lower \(^5\)D\(_4\) state as some population is eventually found in the lower state due to non-radiative decay (Fig. 31a). The signal wavelength was chosen in such manner that energies of the two photons added up to about 5eV which corresponds to location of the 5d band. In fact, a range of signal wavelengths was used in the experiments due to ambiguity in the 5d band location. Light in the visible spectral region was used for ESA from the \(^5\)D\(_4\) level while the ESA from the \(^5\)D\(_3\) level was performed by light in the VI/IR region. In the Tb0.7 glass, on the other hand, ESA upconversion only originated from the lower metastable \(^5\)D\(_4\) level due to insufficient lifetime of the upper \(^5\)D\(_3\) state (Fig. 31b). ESA was performed by visible light in Tb0.7
glass. The process was used for photoexcitation of the Tb-doped PTR glass. Yet it is not the only process that causes refractive index modulation in the glass.

![Diagram of ESA in Tb0.08 (A) and Tb0.7 (B) glasses and ETU (C) in Tb-doped PTR glass.](image)

Figure 31 Diagram of ESA in Tb0.08 (A) and Tb0.7 (B) glasses and ETU (C) in Tb-doped PTR glass.

### 2.7. Competing upconversion processes in the glass

Besides double-wavelength ESA upconversion there are other upconversion processes that result in photoexcitation of the glass. As stated above an ion excited to the intermediate level by a pump photon experiences ESA by another pump photon. Generally the process is of low concern since the energy of the pump photon typically does not match that of the transition to the upper state. However consecutive absorption of two UV photons in PTR glass may lead to excitation to the levels corresponding to the glass matrix. Hence such single-wavelength ESA process can also cause photoionization of the glass leading to refractive index modulation.
There is another competing upconversion process of higher concern. ETU process takes place when two ions excited to the intermediate state undergo an energy transfer resulting in excitation of one of the ions to the high energy states corresponding to the glass matrix (Fig. 2.17c). The efficiency of the ETU process can be two orders of magnitude higher than that of the ESA [70]. In fact, the ETU efficiency increases as the energy transfer efficiency goes up with doping concentration. Similarly to single-wavelength ESA, ETU is an unwanted process and thus measures should be taken to minimize its efficiency.

2.8. Layout for glass exposure experiments

Experimental investigation of photosensitivity of the new glass was performed by direct light exposure followed by the thermal treatment. 2 mm thick glass samples of each type were prepared for the exposure experiments. Glass plates having dimensions of 25x25 mm were cut from the glass slab and ground down to 2mm. The resulting samples were then polished to provide 1/4 or better surface flatness. During the experiments the samples were exposed to different dosages of light and after the thermal development the properties of the samples were compared. Two types of irradiation patterns were created in the course of the experiments, namely stripe and square irradiation patterns. The stripe irradiation pattern represents a 1mm thick stripe of refractive index change having Gaussian distribution of the refractive index across the stripe width (see Fig. 32a).

There are two basic techniques for fabricating a Gaussian stripe in a glass sample. In the first technique the glass sample mounted on a translation stage is swept across a laser beam of about 1 mm diameter (Fig. 32b). The translation speed is adjusted so that the sample is irradiated with a certain exposure of optical radiation, therefore, slower movement means larger exposure. As exposure is defined as beam power times exposure duration, the resulting density of absorbed radiation has Gaussian distribution across the stripe width. As a result of a sweep technique, a
Gaussian stripe was recorded in the glass blank. The refractive index modulation is uniform along the stripe, and transverse distribution is close to Gaussian.

![Figure 32](image)

Figure 32  A. Gaussian refractive index profile B. Stripe irradiation technique by sweeping beam C. Stripe irradiation using a photomask.

The second recording technique involves exposing a sample to a large-area beam through a photomask (Fig. 2.18c). The photomask is created on the glass slide using e-beam lithography technique where a stripe shaped pattern is created on the glass surface. The pattern comprises horizontal dashes that are randomly distributed across the stripe length. Transverse distribution of the dashes density is inverse Gaussian, hence the dashes are densely spaced at the stripe edge. A thin layer of metal is then deposited onto those areas of the stripe pattern where dashes are present. Unpatterned surface of the glass is also coated with metal. When a large area beam having uniform spatial distribution is incident on the photomask, the output of the photomask is a stripe-shaped beam which has a transverse intensity profile close to Gaussian. Placing the glass sample against the photomask allows for recording of the Gaussian stripe similar to that created with a sweep technique.
Square pattern is created by exposing a large glass surface area to light with uniform spatial intensity profile. As a result, a large square of uniform refractive index modulation is recorded in the glass sample. Such uniform refractive index pattern is used for the measurements of the induced absorption in the glass. The stripe irradiation pattern on the other hand was used for measurements of induced refractive index in the glass.

2.9. Technique for measurement of induced refractive index

Refractive index modulation in the stripe pattern can be measured using a Shearing interferometer with a liquid cell [99]. Liquid cell contains a matching liquid filling a cavity between its front and a rear windows. When a glass sample is inserted inside the cell, its refractive index matches that of the surrounding liquid. That allows for separation of the phase change produced by refractive index difference in the glass bulk from that caused by surface profile of the sample.

The measurement setup is shown in the Fig. 33, where a beam from a 633nm He-Ne laser transmitted through an attenuator is incident on the cell. The beam is partially reflected from the front surface of the front window while the other part is transmitted through the sample and is then reflected from the back window. The reflected beams produce an interference pattern which is recorded by a CCD camera. The pattern comprises vertical fringes such as those illustrated in the Fig. 34a. As refractive index change takes place in the glass sample, a shift of the interference fringes can be observed (see Fig. 34b). Here the recorded interferogram is analyzed by a software where the corresponding refractive index change is calculated from the detected fringe shift.
The measurement technique described above was used in the experiments owing to its several advantages over other methods for refractive index modulation measurement. One of the main advantages is the use of the matching liquid for refractive index matching as mentioned above. If sample resides in air, a notable part of fringe shift will be accounted for sample surface irregularity. In fact, a sample thickness variation of 1 μm will lead to the fringe shift comparable to that due to bulk refractive index change on the order of 1000 ppm which is on the same order of magnitude as the induced refractive index in the glass. Immersing the sample into matching liquid allows for dramatic decrease of the surface variance contribution (from 50 to 500 times). A measurement precision of 1 ppm was achieved in the measurement setup described above. Additional advantage is the wedge shaped front cell window that prevents parasitic fringes due to interference of the signal beam with the beam reflected from the backside of the front window.

Figure 33  A. Gaussian refractive index profile B. Stripe irradiation technique by sweeping beam C. Stripe irradiation using a photomask.
2.10. Photosensitivity due to single-wavelength excited state absorption

Series of exposures were carried out to experimentally investigate the photosensitivity of Tb-doped PTR glass. In the first experiments the glass was exposed to a single UV source in order to estimate the single-wavelength photosensitivity magnitude. A 375 nm LED delivering 450 mW of optical power was used as the irradiation source. A beam from the LED was pointed at the glass sample such that the beam area was of the square form with a 7.5 mm side. Spatial intensity profile was sufficiently close to uniform so that the beam was considered flat top in experimental design. Thus the square irradiation pattern was readily available by pointing the beam at the sample. The beam was also passed through the photomask for stripe recording. The time of exposure was chosen to be 80 min giving a maximum total exposure dosage density of 3.5 kJ/cm². The irradiation was followed by thermal development procedure. After that, the liquid cell Shearing interferometer was used to measure the obtained refractive index modulation in the glass. As a result, a change of refractive index of up to 220 ppm was found in the Tb0.7 glass after the single source irradiation. Nevertheless, the presence of refractive index change alone cannot serve as a compelling evidence of an upconversion process happening in the glass.
2.11. Experimental evidence of upconversion process

Presence of Tb$^{4+}$ ions can be considered a reliable proof of the upconversion process resulting in photoionization of Tb$^{3+}$ ions in the glass. While the primary glass components only contain the trivalent terbium ions, Tb$^{4+}$ ions can be created as a consequence of photosensitivity process accompanied by reduction of silver. Presence of Tb$^{4+}$ ions can be determined from the absorption spectrum of the irradiated PTR glass sample before thermal development. A difference of such a spectrum and a spectrum of an unexposed Tb-doped PTR glass sample is presented in the Fig. 35. A broad line that can be observed in the spectrum at the wavelength of about 350 nm is related to Tb$^{4+}$ ions as can be inferred from the Ref. [100]. Similar peaks were observed in Tb$^{3+}$ doped calcium aluminum glasses [101] and other Tb-containing materials after UV exposure. Absorption magnitude measured in the Tb-doped PTR glass after UV exposure was also found to be similar to that measured in the calcium aluminum glasses. Therefore the peak indicated presence of Tb$^{4+}$ ions in the glass which in the case at hand is indicative of the upconversion process taking place.

Figure 35  Induced absorption in Tb-doped PTR glass after exposure to 375 nm UV light.
2.12. Optimization of dynamic range of induced refractive index due to visible light

It has been shown that exposing the Tb-doped PTR glass to UV light results in refractive index change due to single-wavelength upconversion process. The process referred to is believed to be the single-wavelength ETU since it is likely to have higher efficiency compared to the single-wavelength ESA leading to the same result. However the refractive index due to single-wavelength upconversion is not of particular interest for the purpose of the present project and is only an intermediate step before implementation of the double-wavelength upconversion, where UV pump light is used alongside the visible signal. In fact the refractive index change caused by ETU is an unwanted effect in this report as it creates an undesirable background for the refractive index change obtained using visible light. The maximum induced refractive index change possible to obtain in glass has a saturation value which is yet unknown for the Tb-doped PTR glass. If a parasitic background from ETU is present, the total refractive index change can have a value between that of the background and the saturation value. Hence the dynamic range of the possible refractive index change due to visible light is decreased.

To resolve the problem of the dynamic range decrease, the measures were taken to reduce the contribution of the ETU process to the induced refractive index change. It is important to realize that the magnitude of the refractive index modulation is proportional to the square of beam irradiance for the ETU, since 2 photons are involved in the process. By contrast double-wavelength ESA only involves one visible photon contributing to the refractive index change and hence the refractive index modulation has linear dependence on the light intensity in that case. Therefore reducing the UV pump intensity is beneficial for the UV background attenuation. In the present report the intensity of the 375nm LED beam was reduced to 0.84W/cm² and the exposure time was chosen to be 120 min resulting in the refractive index modulation of 50 ppm in Tb0.7 glass. That value was experimentally chosen since it can be clearly detected yet being far from the saturation
conditions. Thus dynamic range for the refractive index change due to visible light was increased. Here the UV light dosage was kept constant throughout the experiments with Tb0.7 glass, so that the visible light contribution to the induced refractive index change could be easily determined.

In Tb0.08 glass, the absorption magnitude is about 10 times lower owing to the reduced concentration of terbium. As a result, a larger dosages of incident optical radiation are required to provide the amount of absorbed radiation equal to that in Tb0.7 glass. Thus exposure times were increased to 10h to provide the same 50 ppm refractive index modulation after UV exposure and thermal treatment.

2.13. Photosensitivity due to double-wavelength excited state absorption

Double-wavelength ESA was implemented by simultaneously exposing a glass sample to the UV pump light and the high intensity signal light. Multimode broad area LDs at different wavelengths in the visible and the IR spectral regions were used as signal sources in the experiment. Signal beam was focused into a 5 mm horizontal stripe with uniform longitudinal refractive index distribution and a Gaussian lateral profile. The glass sample was placed at the intersection of the pump and the signal beams. Beam alignment was carried out in such manner that a stripe-shaped signal beam was imposed onto the center of a square shaped pump beam (Fig. 36a). Such arrangement allowed for convenient measurement of induced refractive index change in the stripe caused by visible light on the uniform UV exposed background using the Shearing interferometer described above. The signal wavelengths were chosen on the basis of the optimum transition energy. Here the best result is expected if the transition energy matches the maximum of 5d density of states distribution given by the 5d absorption line discussed above. Therefore, a blue and green visible LDs emitting light at 449nm and 522nm respectively were employed as the signal sources for exposure of the Tb0.7 glass samples. Photons at those wavelengths provide
optimal overlap with the 5d band if the ESA takes place from the lower $^5\text{D}_4$ state which is the case for Tb0.7 glass. Green LD delivered an optical power of 1W providing $39\text{W/cm}^2$ maximum light intensity on the stripe axis. Optical power of 1.5W was obtained from the blue LD resulting in the maximum intensity of $59\text{W/cm}^2$.

Figure 36  A. Gaussian refractive index profile B. Stripe irradiation technique by sweeping beam C. Stripe irradiation using a photomask.

As a result, a refractive index modulation of 223 ppm was obtained in the stripe in the glass sample after 120 minutes of concurrent exposure to the UV pump light and the green signal light and consequent thermal development (Fig. 2.22c). A 240-minute long exposure to the blue light alongside UV pump resulted in the maximum of 190ppm induced refractive index change after the thermal treatment.

Detailed consideration of the experimental results revealed an inconsistency between the obtained data and the expectations based on the glass characterization results. In fact, the glass was found to have higher photosensitivity to green light than that to blue light. Normalizing the
obtained refractive index change to the exposure dosage at both wavelengths the final value for 522 nm green light was calculated to be 3.5 times higher than that obtained for 449 nm blue light. Generally, the expected relative photosensitivity magnitudes at different wavelengths could be assessed on the grounds of Tb$^{3+}$ 5d band location, which can be deduced from absorption or luminescence excitation spectra. Here the position of the peak and the shoulder of the band on the energy scale can be matched with resulting energy of an upconversion transition. In the case under consideration the electron at $^5$D$_4$ level is excited to a position corresponding to the energy of 4.93 eV as it absorbs a “green” photon at 522 nm. In the same time a “blue” photon at 449 nm elevates the electron from 2.6 eV at $^5$D$_4$ state up to 5.31 eV. It can be seen from the Fig.2.12 that energy value of 4.93 eV corresponds to the point in the vicinity of the peak of the 5d band in the excitation spectrum. On the other hand the same value appears to fall within the shoulder area of the band in the absorption spectrum as it can be seen in the Fig. 2.6. Often a priority for interpretation of the measurement data should be given to the absorption spectrum, and in that case an inconsistency becomes apparent. However, the experimental results could be possibly explained by presence of yet unknown mechanism taking place at the position of 4.93 eV. As carriers are excited to the position of 4.93 eV, the mechanism prevents both carrier relaxation to the lower states with subsequent luminescence and carrier transfer to the glass matrix. Hence, the intensity of both luminescence and photosensitivity would have a maximum at that spectral position where the above mentioned mechanism is less apparent. According to the experimental results, the maximum values of both photosensitivity and luminescence was observed at the position corresponding to 250 nm wavelength or 5.31 eV of energy. The character of the mechanism mentioned above is yet not completely understood and will be investigated in the future. Nevertheless it can be inferred
that the double-wavelength ESA upconversion provides induced refractive index change in Tb0.7 glass after exposure to green and blue light followed by thermal treatment.

Unlike Tb0.7 glass the low concentration Tb0.08 glass allows for use of the upper metastable $^5D_3$ level where the photon with lower energy is required. For that reason, infrared LDs operating at 808 nm and 975 nm were used as signal sources for Tb0.08 glass exposures. As mentioned before Tb0.08 glass exhibits sufficiently lower absorption magnitudes compared to Tb0.7. Hence significantly higher intensities of the signal beam were required as well. Owing to the limited maximum output power of the LDs, the beam area was decreased to provide higher intensities in the Tb0.08 exposure experiments. Here the signal beam was focused into a circular dot with 1mm diameter. The beam alignment was performed in such fashion as it is demonstrated in the Fig. 2.22b. Such dot pattern does not allow for reliable measurement of the induced refractive index using the Shearing interferometer such as stripe pattern, however the ballpark estimate of the refractive index change is possible.

After 10 hours of the simultaneous exposure to the UV light and the IR light at 808 nm a dot shaped refractive index modulation such as that shown in the Fig. 2.22d was observed in the glass sample. The induced refractive index change was estimated to be on the order of 300 ppm based upon the number of the fringes in the interferogram. The similar exposure involving the IR light at 975 nm resulted in the refractive index modulation of less than 150ppm.

It can be inferred that much higher intensities of the signal beam are required to obtain the induced refractive index in Tb0.08 glass comparable to that in Tb0.7 due to lower absorption magnitudes in the former. On the other hand, the ESA from the upper $^5D_3$ metastable level allows for lower energy signal photons resulting in extension of the glass sensitivity range into the IR spectral region.
2.14. Experimental study of holographic capabilities of Tb\textsuperscript{3+} doped photo-thermo-refractive glass

Refractive index modulation obtained in Tb\textsuperscript{3+} doped PTR glass by concurrent exposure to UV and visible light enables an opportunity for hologram recording in the glass using visible light. In fact induced refractive index of 200 ppm allows for recording of VBG with the diffraction efficiency of 98\% or above in regular 2mm PTR glass plate. Thus a 300 ppm refractive index change obtained in Tb\textsuperscript{3+} doped glass by upconversion involving green light renders possible to record complex holograms in glass plates thicker than 2mm. Since visible light holography in PTR glass is of interest as a prospective application, an attempt was made to record a complex hologram in the Tb0.7 glass using the ESA upconversion described above.

Unfortunately the available signal sources did not meet the requirements for spatial and spectral coherence imposed by the typical complex hologram recording process. For that reason a special type of complex hologram applying relaxed constrains at the recording process was chosen to demonstrate the capacity for hologram fabrication in Tb\textsuperscript{3+} doped PTR glass [102]. The beam requirements are relieved since the hologram recording setup contains only one beam i.e. no reference beam is present. A metal grid with 100 μm pitch was chosen as an object. The recording procedure was conducted in the following manner (see Fig. 37). The sample was placed against the backside of the grid. UV beam from the 375 nm LED, with the 7.5 mm square shape was incident on the backside of the sample at an angle of about 20°. UV light was used as a pump for upconversion process providing excitation of the ion to the intermediate metastable state for consequent ESA. The latter was carried out by a signal beam from the green LD normally incident on the front surface of the sample through the grid. The green light thus served as a signal beam in the hologram recording setup. Incident beam was diffracted on the grid so that the beams diffracted on different elements of the grid produced an interference pattern inside the glass plate.
The intensity pattern inside the glass resulted in the volume hologram fabrication after subsequent thermal treatment. For the reconstruction procedure the grid was removed and a sample was irradiated with the green beam alone. The image of the hologram was observed on the screen positioned about 3m away from the sample.

![Diagram](image)

**Figure 37** Hologram of a metal grid recorded in blank of Tb-doped PTR glass.

The image of the diffraction pattern from the hologram of the grid is shown in the Fig. 38 alongside the image of the green beam diffracted on the grid. Close resemblance of the two images can be clearly seen. A similar hologram was fabricated using blue light as a signal in the same recording setup as described above. It was thus demonstrated that the new Tb$^{3+}$ doped PTR glass is capable of holographic recording of optical structures despite its lower photosensitivity compared to the regular PTR glass. Hence, the possibility to record complex volume holograms in the Tb$^{3+}$ doped PTR glass can be safely suggested if high performance laser sources are available. It can be inferred that the new glass allows for recording of complex holograms in Tb$^{3+}$ doped PTR glass for use in visible light.
2.15. Issue of low photosensitivity of Tb$^{3+}$ doped photo-thermo-refractive glass and possible ways of optimization

Tb:PTRG is a type of PTR glass, where Ce$^{3+}$ dopant is replaced with another rare earth element – Tb$^{3+}$. Photoionization here is performed in a similar fashion as in regular cerium doped PTR glass – by excitation of a 5d band, placed above an electron mobility threshold in PTR glass. Trivalent terbium has a 5d band with energy higher than that of cerium, consequently direct excitation of Tb:PTRG requires beams with very high photon energies corresponding to the far UV region [68, 86]. Yet Tb$^{3+}$ allows for excitation of the 5d band using two photon excited state absorption (ESA) process [70].

PTR glass doped with 0.7 at% terbium was chosen for the purpose of recording of a HOE with visible light using ESA upconversion excitation process. The type of ESA implemented in the experiments, employs two photons with different energies for two two types of transitions: ground state absorption and ESA. This type of ESA is known as double-wavelength ESA. Here the photon that provides excitation from a ground state $^7F_6$ to the intermediate metastable state $^5D_4$ is called a pump photon, whereas the photon responsible for the excitation from the intermediate state is known as a signal photon. In the experimental environment double-wavelength ESA was

![Figure 38: Diffraction on a metal grid (A) and complex hologram (B).](image-url)
performed by simultaneously exposing a glass sample to two different sources emitting at two different wavelengths.

Refractive index change due to visible light was obtained in Tb:PTRG following its simultaneous exposure to UV and visible sources. However, the amount of refractive index change which is possible to obtain with that technique has to be recognized to be significantly lower than what can be achieved by hologram recording materials mentioned in the previous chapter. In turn, insufficient refractive index change is known to directly affect the diffraction efficiency of recorded holograms. Furthermore, long exposures on the order of several hours were needed to achieve even the small values of RIC shown in the previous chapter. It is understood that those types of exposure are not compatible with any practical holographic applications. It followed that if the glass had to be used for recording of real-world holograms, the recording duration had to be drastically reduced. There are two main approaches that may lead to reduction of recording duration. The first and obvious one is the photosensitivity of the glass. This however is a complicated subject as there are many parameters of glass composition and fabrication that affect its photosensitivity. Here the only straightforward option is to increase the concentration of Tb$^{3+}$ sensitizer in order to increase the upconversion rate. However, increasing the concentration of Tb$^{3+}$ beyond 0.7at% was shown to have a negative effect on homogeneity of the resultant glass phase. For that reason, it was decided to keep the glass composition unchanged and pursue other methods of recording time reduction.

The second method is no less obvious, and it involves the use of recording sources that may supply the necessary exposure to provide sufficient amount of RIC in a hologram. In case of Tb:PTRG the visible light source had to supply sufficient amount of energy to compensate for low photosensitivity. There were several considerations for choice of the light source. First of all,
sources that deliver higher power outputs are preferable as they allow to decrease the amount of exposure time. It is also important to keep in mind that since upconversion rate is dependant on the peak power, choice of pulsed sources over CW ones would be of a great benefit as pulsed sources might have peak power by several orders of magnitude higher than the CW sources. Besides, as it follows from 2.5.3, spectral dependence of the ASE had to be taken into account in order to ensure that the wavelength of the chosen source provides the maximum efficiency of the photoexcitation. The other consideration is the pump exposure and intensity. Unlike the signal beam which has to provide maximum power possible, pump parameters are more complex as too much pump irradiance will benefit an unwanted competing ETU process whereas excessive exposure will result in a parasitic UV induced RIC background. Lastly, thermal development also has its effect however that had been already chosen before and its duration was determined by the maximum RIC obtainable before the onset of scattering in glass.

Optimization of all the above mentioned parameters was an important step towards recording of real hologram and to proceed from a proof of concept to applications. Spectral optimization of the sources was relatively straightforward. Here the pump and signal photons had to be chosen so that the sum of their photon energies corresponded to the position of the center of the 5d band on the energy diagram. That would result in maximum ESA efficiency that is attainable with the given source intensity. It was determined that if pumping is provided by a UV source operating between 365 and 375 nm, maximum ESA can be carried out with a signal source at the wavelength of approximately 540 nm. The choice is accounted for an LED pump source which provides resonance excitation of a $^5\text{D}_3$ state with maximum efficiency. Besides, the glass sensitivity is, thus, situated exactly in the region of interest with the target wavelength of 532 nm just 10 nm away from the ESA maximum.
2.16. Light sources for photosensitivity measurement and holographic recording

The choice of the pump and signal sources was made within the scope of target photon energy and also on the basis on the availability of high quality and high power light sources in that spectral region. The sources that were used in the photosensitivity experiments and later for the hologram recording were the following ones. An LED from Thorlabs mounted on the high-thermal conductivity metal-core printed circuit board mounted onto a heat sink was used as a pump source. The LED was designed for operation at the central wavelength of 365 nm, and its emission spectrum was found to have a good overlap with absorption band of Tb:PTRG corresponding to the 380 nm peak. The LED delivered 980 mW of CW power and could be focused to achieve maximum irradiance of 7.76 W/cm². Spectral analysis of LEDs operating at 365nm and 375 nm demonstrated that 375 nm LED provides more efficient pumping. Nevertheless, the 365 nm LED was opted for as it offered over two times more power as its counterpart.

As far as the signal is concerned, the benefit of high peak power was provided by a pulsed Nd:YAG Powerlite PL8020 laser manufactured by Continuum to provide high-energy pulses and high beam quality. Visible output at 532 nm was produced by second harmonic generation from a fundamental Nd:YAG line at 1064 nm. The laser delivered 10 nanosecond pulses with the repetition rate of 20 HZ and with the maximum energy of 200 mJ. From there the peak power was found to be 20 MW, and it is the peak power that that determines the rate of the ESA process. Meanwhile the average power which governs the total exposure was 4W. High spatial and temporal coherence becomes of high importance as the holographic capabilities of the laser source are concerned. The laser met all the requirements in that respect as it featured injection seeding technology. A seed beam from a single frequency fiber laser is injected into the oscillator cavity, which results in close to transform limited linewidth of the pulses. In fact, the particular laser model was designed for holographic purposes and featured a custom built seeder which provided
The output laser emission was specified by the manufacturer to have a linewidth below 100 MHz resulting in over 1 m of coherence length. Thus, the laser provided sufficient peak power to provide high efficiency upconversion process, and owing to its coherence properties, it was also later used for holographic recording. Next step was to determine the requirements for irradiances of the both sources which however was more complicated.

2.17. Upconversion transition kinetics in Tb$^{3+}$ doped photo-thermo-refractive glass

Upconversion photoexcitation is a probability process which involves several different transitions happening simultaneously. In this case efficiency of the process depends not only on the rates of the constituting transitions but also on the rates of competing processes as well. Kinetics of the upconversion is ultimately governed by the evolution of the carrier population on the lower excited $^5D_4$ state which was initially populated by non-resonant absorption. The main mechanisms are demonstrated in the Fig. 39 where absorption of a pump photon at the wavelength of 375 nm leads to the excitation to the upper excited state $^5D_3$ whose lifetime is insufficient for use as a metastable level, and hence the population decays non-radiatively to the lower excited state. That allowed to make a reasonable assumption that the decay rate from the upper level is significantly higher than any competing processes. Hence, for simplicity, it was assumed that the lower excited state is populated by non-resonant absorption of UV photons. Here there are at least four different mechanisms of depopulating the $^5D_4$ level. First mechanism is the target ESA by a visible photon which provides an upward transition to the 5d band with consecutive photoionization of the glass. The second mechanism is again ESA, however, carried out by another UV photon. Absorption of another UV photon by an ion in the $^5D_4$ state leads to the upward transition to bands situated above the 5d band and whose physical nature is yet unknown. It is understood, however, that the ion undergoes a non-radiative relaxation down to the 5d band where
photoionization happens in the similar fashion. Although ESA can be carried out by both UV and visible photons, it has to be noted here that the ESA rates are known to depend on the absorption cross section as well as on the peak power of the signal beams. Since photons in the green part of the spectrum experience significantly larger cross sections, and since that signal beam is pulsed and its power is significantly higher than that of the UV pump beam, it is possible to neglect the ESA due to UV photons without any loss of generality. Then, there is another upconversion process, namely ETU, which is also caused by another pump photon, however, that cannot be neglected as ETU tends to have efficiency approximately two orders of magnitude higher than ESA. ETU is also dependent on the doping concentration and hence is higher in the medium where distance between adjacent Tb$^{3+}$ ions is smaller. Lastly, the carrier population is depleted by a set of radiative downward transitions from the metastable $^5\text{D}_4$ state to the different states of the $^7\text{F}_j$ ground state multiplet from where the carriers undergo a nonradioactive decay to the ground state. The most efficient transition at 545 nm from $^5\text{D}_4$ to $^7\text{F}_5$ state becomes dominant at high pump powers and hence it can be considered as a main mechanism for luminescent depopulation of the metastable state.

By taking account of all the three remaining mechanisms, the evolution of carrier population at the lower metastable $^5\text{D}_3$ state can be represented as

$$\frac{dN}{dt} = \sigma_{\text{GSA}}\phi_{\text{pump}}N_{\text{GS}} - \sigma_{\text{ESA}}\phi_{\text{sig}}N - \sigma_{\text{ETU}}\phi_{\text{pump}}N - \frac{N}{\tau}$$

(2.2),

where the first term determines population of the level by ground state absorption, second and third terms correspond to the ESA and ETU mechanisms, respectively, while the fourth term represents spontaneous emission to the ground state multiplet. In the equation above $\sigma_{\text{GSA}}$ is defined by ground state absorption (GSA) of Tb:PTRG and \(\tau\) is the level lifetime at 3.2 ms. Photon
flux is defined as $\varphi = I/\nu$, and hence depends on beam irradiance; these were the variable parameters in order to determine the unknown quantity – the level population $N$. ...

![Diagram of energy level transitions](image)

**Figure 39** Mechanisms of population and depopulation of energy level $5D_4$.

The task is made more complex by the fact that the cross sections of the ESA and ETU processes are also unknown. Those cross section values are available in the literature [103, 104] for other Tb$^{3+}$ doped media. The ESA cross section in Tb$^{3+}$ doped silicate glasses tends to be situated around the value of $7 \cdot 10^{-11}$, whereas ETU expectedly shows higher probability with the cross sections on the order of $3 \cdot 10^{-9}$. It is understood that the actual cross section is also dependent on the host medium and hence can be different in PTR glass. For that reason cross section of the ESA was experimentally measured for Tb07 glass using a lock-in amplifier.
2.18. Measurement of Excited state absorption

ESA was the main process responsible for photoionization and producing of the RIC in Tb:PTRG using visible radiation. ESA is the process that follows excitation to a metastable level resulting from GSA of a UV photon. While GSA was studied in detail and it is discussed in the preceding sections of this work, only spectral dependence of ESA on the wavelength of the visible signal light was mentioned. It was determined that radiation originating from the spectral region corresponding to green light results in the highest efficiency of the upconversion process. However, no quantitative description of the ESA was given before. On the other hand, the ESA cross section $\sigma_{\text{GSA}}$ is an important parameter for modeling of the kinetics of the upconversion process. $\sigma_{\text{GSA}}$ was experimentally measured using a setup shown in the Fig. 40.

![Experimental setup for measurements of Excited state absorption.](image)

The performed experiment was of a pump-probe type. A CW frequency-doubled Nd:YAG laser operating at 532 nm radiation served as a probe beam as it passed through the pencil of Tb07glass
entering and exiting through the polished ends of the sample. Pumping of the sample was collinear and was carried out by a 375 nm UV LED which was modulated by a diode driver to emit 5 ms rectangular pulses with a frequency of 100 Hz which was used as a reference frequency for the lock-in amplifier. UV radiation was focused from the end of the sample into its center for increased intensity leading to larger number of GSA events. Collinear alignment gave advantage of better overlap between pump and probe beams potentially increasing the obtainable signal. Collinear alignment was possible as a probe beam was injected from the side using an RBG and then the 532 nm beam was extracted from the UV beam path by another RBG where the signal was collected by a photodetector. UV pump radiation was filtered out by means of an aperture as well as a longpass dichroic filter. Additionally, the signal was detected as a difference with the direct output of the laser which was made for the purpose of mitigation of beating effects that were present possibly due to presence of several longitudinal modes in the laser emission. In the end, the ESA coefficient $\alpha_{\text{ESA}}$ was measured to be $2.058 \times 10^{-6} /\text{cm}$ which was consistent with the $\sigma_{\text{GSA}}$ values measured in other vitreous media doped with Tb$^{3+}$ ions.

2.19. Optimum exposure and irradiance parameters for the recording sources

Efficient ESA in Tb:PTRG takes place if the requirements, imposed onto intensities and dosages of radiation from pump and signal sources, are fulfilled. Requirements for signal source are quite straightforward, and claim that signal beam intensity should be as high as possible for efficient ESA. The refractive index change in that case will be proportional to the dosage of visible (signal) light. Constraints imposed on the pump source require that a certain ratio of UV and visible beam intensities is maintained. Intensity of the UV source should be high enough, and its dosage should be large enough to provide efficient pumping to the metastable level. It was determined from (2.2) that ESA requires the minimum UV dosage of 150 J/cm² when pumping is carried out by the 365
nm UV LED at full power. At the same time, excessively large dosages of pump radiation are unwanted, as they would result in refractive index change due to UV radiation. RIC due to UV radiation results from ETU process, hence UV light intensity should be kept significantly lower than that of the visible light, and the UV dosage has to be maintained below a certain value in order to keep the UV induced RIC reasonably low. The UV dosage of 2 kJ/cm² was found to result in RIC of 50 ppm due to UV radiation after 120 min of thermal development. Here, any refractive index change caused by UV radiation will decrease the dynamic range of RIC due to visible light, consequently the performance of resulting holographic elements is expected to deteriorate. For the reasons described above, the parameters of UV and visible sources were chosen so that to provide the most efficient ESA upconversion possible.

2.20. Photosensitivity of Tb:PTRG using improved exposing technique

The efficiency of ESA in Tb:PTRG can be determined from the photosensitivity of the glass to visible light, which in turn can be assessed by the amount of refractive index change caused by exposure to visible light. Photosensitivity of Tb:PTRG to visible radiation at 532 nm from the high power laser mentioned above was characterized by the induced refractive index. Samples of polished Tb:PTRG were irradiated with the stripe having a Gaussian lateral intensity profile, which allows for precise measurement of RIC. Exposure geometry is presented in Fig. 41a. A collimated beam from the green laser was focused onto the sample surface using a cylindrical lens, so that the illuminated spot was a stripe with a close to Gaussian lateral profile in vertical direction. However, the longitudinal beam profile obtained in such geometry is inhomogeneous due to nonuniform intensity profile of the collimated beam in horizontal direction. The longitudinal inhomogeneity problem was fixed by sliding the sample back and forth along the stripe axis (Fig. 41b). This pattern at 532 nm was produced by the signal beam, whereas the 375 nm UV LED, mentioned
above, provided the pumping. UV beam was focused into a square with a side of 7.5 mm with uniform intensity across the beam area. Beam alignment was performed so that a stripe-shaped signal beam was imposed upon the center of a square-shaped LED beam. A set of Tb:PTRG samples was irradiated with different dosages of visible radiation, followed by thermal development. RIC in stripes was measured using a technique based on the shearing interferometer, described in details in the section 2.9.

RIC dependence on dosage of visible radiation for Tb:PTRG is presented in the Fig. 42. The glass underwent two series of exposures with beams of 0.5 and 1 J/cm² pulse energy densities. The magnitude of refractive index change depends not only on the parameters of the visible beam, but on the additional conditions as well, including UV light dosage, development time and temperature. The results presented in the figure are the highest values of RIC that were possible to

Figure 41  Experimental setup for simultaneous exposure of Tb:PTRG sample to a Gaussian stripe at 532 nm and square patter at 375 nm. A – geometry of beam combining, B. geometry of exposure patterns.
obtain throughout the experiments. It can be seen from the graph that the maximum achievable refractive index change approaches 400 ppm in Tb:PTRG. This value allows for recording of VBGs with diffraction efficiency of 99% and better. It can be then inferred from the obtained results that the Tb:PTRG demonstrates photosensitivity sufficient for holographic recording in the glass.

![Graph showing photosensitivity to visible radiation after exposure to 532 nm beam with different pulse energy densities (ED) for Tb:PTRG.](image)

Figure 42  Photosensitivity to visible radiation after exposure to 532 nm beam with different pulse energy densities (ED) for Tb:PTRG.

### 2.21. Holographic recording in Tb:PTRG

HOEs in (Tb:PTRG) were fabricated by concurrent exposure of the glass plate to UV radiation at 365 nm and visible radiation at 532 nm. Again a hologram is fabricated in the glass upon completion of the thermal development, which takes place at higher temperatures in the terbium doped PTR glass compared to the regular PTR glass. A transmitting VBG (TBG) was recorded in
the glass since the efficiency of such optical structure can be easily compared to the VBGs produced using standard recording procedure with UV exposure of PTR glass. An optical setup shown in Fig. 43 was devised for recording of a TBG. A collimated beam at 532 nm was split by means of a beam splitter, and then the two beams were combined at the full incidence angle of around 6°. A sample of Tb:PTRG was placed at the intersection of the beams, so that the interference pattern can be recorded in the glass plate. Pumping was carried out by the 375 nm LED beam focused from the backside onto the sample in a 7.5x7.5 mm² square. The sample was exposed to visible radiation with the total dosage of 32 kJ/cm², while the UV dosage was measured to be 1 kJ/cm². The TBG was formed in the glass sample after the exposure and subsequent thermal development. The period of the grating was measured to be approximately 5 μm, and its diffraction efficiency was 50%. A reflective VBG (RBG) was recorded in Tb:PTRG as well. In that case the beams were combined in a way as it is shown in Fig. 44, so that the converging angle could be easily adjusted.

![Figure 43 Setup for TBG recording in Tb:PTRG.](image-url)
Performance of a TBG was upfront simulated using the technique described in 1.22 to calculate the required visible radiation dosage for highest diffraction efficiency. The TBGs recorded in the experiment were designed to target 95% of diffraction efficiency. Unfortunately the results did not quite meet the expectation as only 50% diffraction efficiency was measured in the transmitting grating while efficiency of the RBG was yet drastically lower below 1%. The reasons behind poor performance of the HOEs recorded with visible light will be described in more detail below. Meanwhile the main challenge lies in the drastically lower photosensitivity of Tb:PTRG to visible light compared to the photosensitivity of the regular PTR glass to UV radiation. As a result, maximum attainable values of RIC in the glass are lower and the exposures durations are longer leading to deterioration of the hologram performance. Discussion of the figures of merit of different hologram recording materials in 1.14 mentioned its photosensitivity as one of the main criteria for the choice of the material. A material with high photosensitivity is highly desirable as it allows to maintain the exposure durations relatively short. When photosensitivity of the holographic recording material is low, exposure durations are longer and this introduces the
problem of recording stability. Quality of a hologram can degrade if recording fringes displace with respect to the material during recording. In this work attempts were made to overcome the problem of fringe stability and provide stable recording environment suitable even for recording in the low photosensitivity material such as Tb:PTRG.

2.22. Recording stability in relation to hologram performance

High quality grating can be achieved if two main conditions are met, high contrast refractive index modulation (RIM) and a high-visibility fringe pattern. That would provide a stable beam resonance condition resulting in the high diffraction efficiency. If one of the conditions is violated the performance of the grating is going to deteriorate. The first condition is addressed by targeting the appropriate RIM value by controlling the duration of the holographic recording or adjusting the intensity of the recording beam. It has to be noted that visible recording procedure has lower RIM saturation values and lower photosensitivity resulting in longer durations of recording. High fringe visibility can be achieved by providing stable environment during holographic recording. The primary factor that aggravates the fringe contrast is the shift of the fringes in the process of recording. This shift can be caused by different instabilities such as mechanical vibrations, air movement, temperature etc. The primary effect of the fringe shift is smearing of the interference planes and subsequent decrease in the depth of modulation. HOEs produced using visible light can have spatial frequencies as high as 6 million lines per mm. Hence displacement by a fraction of a wavelength will result in smearing of the interference pattern. It can be inferred that two main ways of improving the performance of the HOEs would be by increasing the photosensitivity of the glass while maintaining a stable recording environment. Stability of the recording stage means fulfillment of two requirements: stability of a holographic plate with respect to the stage and stability of the fringe pattern with respect to the plate. While the former simply means fastening
the sample securely to the stage and prevent any possible movement of the sample, it is more complicated to ensure a perfect fringe stability.

Fringe stabilization is a complex issue, however for simplicity all methods can be divided in the two main groups: passive phase stabilization and active phase stabilization systems. The former methods aim to solve the instability issue by neutralizing the sources of instabilities. If no instabilities are present, the fringe stability will be readily provided. Meanwhile the second group of methods are known as active phase stabilization systems. Active systems constantly detect the relative phases of the recording beams, and provide compensation should a fringe shift occur. The advantage of the active methods is that fringe stability is supposed to be maintained even in the presence of instabilities. Each of the two approaches by itself, however, might not be sufficient. Here it is not always possible to completely mitigate all instabilities in the system. Hence, active phase stabilization might be required. On the other hand the active phase stabilization might not provide compensation with enough accuracy if the amplitude of the instabilities is excessively large. The two groups of methods hence should be combined for best results.

2.23. Vibration isolation

In this paper a hologram recording system is presented that takes advantage of both approaches. First, a passive phase stabilization was carried out. First, the sources of the instabilities had to be properly identified in order to be able to develop an isolation strategy. Visual estimation can be quite challenging here since the effect takes place on the scale of the fraction of a micron. Reference [105] presents an approach for identifying the existing source of aberrations by recombining the two recording beams and analyzing the behavior of the resulting interference pattern. Exact analysis of the interference effects is rather complicated and is beyond the scope of this paper. However, it is relatively easy to get a grasp of the practical concept of the pattern
behavior as seen from [106]. Then all the instabilities can be generally divided into three main groups including mechanical vibrations, air turbulence, and temperatures effects. Each type of instabilities will have a distinct effect on how interference pattern evolves overtime. For instance, mechanical vibration present in the recording setup will cause jitter of the recording fringes. By contrast, air movement causes medium lifetime fluctuations of the recording fringes without a discernable log-term displacement. Finally, thermal fluctuations can be detected by a constant short or long-term drift of the interference fringes. Once the effects of the instabilities are clear, an appropriate perturbation is simulated to confirm the observed effect on the interference fringes. The method was used, and as a result, air turbulence and mechanical vibrations were found to be the two major concerns while thermal instability was found to be off less importance which is explained by the use of a pulsed laser radiation as opposed to a CW source.

Once instability sources were determined, measures were taken for their mitigation. Vibration isolation methods are fairly well studied and straightforward. The recording setup was mounted on a vibration-isolating table which allows to prevent vibrations of the building to be transmitted into the movement of the delivery optics. A vibration isolating table consists of the tabletop which is supported by the isolators. Isolators contain two air chambers with a piston that moves therebetweeen. While piston movement is caused by direct disturbances, air encumbers the motion of the piston thus resulting in damping of the vibrations. The resonance frequency of the damper is much lower than that of typical vibrations in a building. This system works well for both small and large amplitude displacements. Table top placed on top of the isolators provides further vibration isolation due to its honeycomb construction which provides stiffness of the structure. Isolating optical table can prevent detrimental effects from ground vibrations such as those of seismic origin, acoustic vibrations, and direct disturbances. Obviously any movement of the optic
with respect to the table should be prevented as well. This is provided by placing all the optics on thick, vibration-proof posts and mounts.

Acoustic noise can be also coupled to the recording stage by air. Air turbulence produces disturbance in the refractive index of air resulting in the change of the optical path. Turbulent air flows might result from air conditioning units, air ducts, fans, heaters and other sources. The recording stage thus was isolated from surrounding air by confining all the delivering optics and a sample holder assembly within a 5 sided acrylic box. A recording laser beam was introduced into the setup through a 20 mm round opening. Before each recording the setup was conditioned for 30 minutes to ensure all the air movement inside the box is settled down and stable recording environment is provided. It has to be noted that air flows inside the confinement box can be produced by heat caused by a laser beam (such as heating of a recording sample), however no such effects were detected. Presence of heating of the sample was evaluated by tactile contact where no temperature raise by over 1 °C was detected.

2.24. **Transmissive holographic optical elements**

As changes to the stability of the recording stage had been implemented, the attempts to record a hologram were resumed. As before, transmitting gratings were chosen first as those gratings allow for convenient characterization and comparison with standard UV gratings. The layout of the recording setup was identical to that used before and shown in the Figure 41. The TBGs recording procedure targeted 99% diffraction efficiency which corresponded to the RIM of 150 ppm in a 2 mm sample. After recording and thermal development a relative diffraction efficiency of 98% was measured in the transmitting grating which was obtained as a result of exposure to 0.7 KJ/cm² of UV radiation and 22 kJ/cm² of visible light. Absolute diffraction efficiency which
includes losses and scattering in the glass was found to be 85%. Efficiency curve of the 85% efficient TBG recorded by means of the visible radiation is demonstrated in the Fig. 45.

Although planar Bragg gratings allow for fair estimation of the holographic capabilities of a material, they were not of most concern for the present work, since those structures can be recorded in the regular PTR glass with UV radiation. A hologram of a positive convex lens with a focal length of 200 mm was recorded in a sample of Tb:PTRG as an example of a complex HOE utilizing the same laser beam at 532 nm for both recording and reconstruction. The recording setup is shown in the Fig. 46 and resembles that used for a regular TBG recording except for one modification. The lens system was placed in one of the arms of the setup so that the appropriate beam was focused, and then defocused after reaching its minimum size. The distance between the lens and the sample was chosen in such manner that the focused beam returned back to its initial size as it reached the recording beams intersection point. Therefore the two beams had the same size at the intersection point, which enabled recording a hologram with the largest possible diameter. Such a hologram is basically a two-dimensionally chirped TBG where surfaces of constant refractive index are not planar and grating period is varied along the side of the hologram. As the beam is diffracted by such a hologram, it experiences convergence in a similar way as it would after passing through a lens, or divergence depending on the direction of the incident beam. Operation of the holographic lens in the focusing mode is demonstrated in Fig. 47. The diffracted beam starts to converge upon emerging from the grating (Fig. 47a), and reaches its minimum size as it goes through the focal point. After that, the beam begins to diverge, so its size eventually larger than that of the transmitted beam (Fig. 47b).
Figure 45  Diffraction efficiency versus angle of incidence for a TBG recorded in PTR glass using visible light.

Figure 46  Setup for recording of holographic lens in Tb:PTRG.
An interesting property peculiar to the holographic lens that bears mentioning is that unlike the conventional mechanical lens, whose focusing ability is independent on the direction of the incident beam (front or back), a holographic lens exhibits distinct behavior depending on the orientation of the element. When reconstruction is carried out with the original reference beam in the recording configuration, the lens will operate in focusing mode as described above. On the other hand, should it be illuminated with a beam which is conjugate to the reference beam, the lens will reconstruct the original object beam directed backwards. Hence, defocusing mode is obtained when the opposite orientation of the lens is used. As far as the diffraction efficiency of the HOE is concerned, that was measured to be 96% in the holographic lens. Absolute diffraction efficiency was measured to be 85%.

Figure 47 Holographic lens operation, focusing of a diffracted beam a) transmitted and diffracted beams 30 mm from the lens; b) transmitted and diffracted beams 80 mm from the lens. Photos are with different magnification – diameter of the transmitted beam is the same at both photos.
2.25. Conclusion on holographic recording in Tb$^{3+}$ doped PTR glass

In summary, the modified version of PTR glass doped with Tb$^{3+}$ ions was designed and fabricated for the purpose of holographic recording by means of visible light. Full characterization of Tb:PTRG was performed, and the glass was proven capable of holographic recording. Photoexcitation of this glass is carried out using an ESA upconversion process involving a UV photon that provides initial excitation to an intermediate state and a visible photon responsible for the photoionization of the glass. Hologram can be recorded in Tb:PTRG by simultaneous exposure to UV radiation and high-irradiance visible beam. It is, in fact, important to maintain the highest irradiance levels of the visible beam possible since the efficiency of the upconversion process is proportional to the peak power of the visible beam. The recording durations are substantially longer for recording in Tb:PTRG with visible light in comparison with the standard recording procedure in PTR glass. Long recording durations present a challenge of the fringe stability which can ultimately affect the performance of the hologram. Stable environment was provided during recording by providing vibration isolation. The means included suspended optical table, vibration-proof optical mounts, air confinement, etc. Vibration isolation technique was proven to yield favourable results as efficiency of the transmission gratings recorded with visible light was significantly improved approaching that in the VBGs created with UV radiation in PTR glass.

On the other hand, vibration isolation was found to have little effect on the performance of the reflection holograms. RBGs recorded using visible light still demonstrated very low diffraction efficiencies on the order of a percent. It appears that despite the rewarding effects, vibration isolation provides insufficient fringe stability for recording of reflective HOEs. Recording of reflective structures is more sensitive to any disturbances, and hence additional measures have to be taken to provide required fringe stability. The problem was addressed and the proposed solution will be described in the following chapter. Despite the stability issues, it can be concluded that
Tb:PTRG was demonstrated to be a worthy alternative to current hologram recording materials as it allows for fabrication of durable, high-power and heat stable HOEs for visible applications.

Thus the approach of creating a hologram recording material sensitive to visible light on the basis of the original PTR glass by modification of the glass was overall successful. There are however few drawbacks associated with this approach. Here, Tb:PTRG is a new type of glass as opposed to the original PTR glass that is a commercial glass which has been optimized and developed over a long period of time. As a consequence, the new glass is prone to several issues related to its optical properties. First, choice of an appropriate vendor of Tb$_4$O$_7$ component is important as the properties of the compound tend to differ depending on the vendor. In particular, that means that two glass batches fabricated using Tb$_4$O$_7$ supplied by different vendors might have different ratio of trivalent and tetravalent terbium concentrations. As it is only the Tb$^{3+}$ that provides the photosensitivity of the glass, photosensitivity properties of the two glasses will be different. Depending on the amount of Tb$^{4+}$ present, the glass might even lose the photosensitivity completely. The second problem consists in the increased viscosity of the glass which results in inferior homogeneity of the glass compared to the standard PTR glass. Lastly, there is a disadvantage of the recording procedure itself which is related to the spatially non-uniform pumping intensity. Here the emitting surface of a pump LED is not uniform but rather the electrons are shaped in a geometrical pattern. Although the PTR plate is placed out of the focus plane, the intensity profile is still non-uniform. Non-uniform intensity pattern results in non-uniform exposure and hence parasitic modulation of the refractive index of the grating leading to degrading of the grating performance. On the other hand, availability of pumping sources with sufficient optical power in that spectral range is severely restricted, therefore the choice of the pump sources is limited to the above stated LEDs. All the above said raises the question of whether hologram
can be recorded with visible light in the original non-modified PTR glass with Ce\(^{3+}\) as a sensitizer. It appears that recording with visible light is possible in the original PTR glass as well, in fact, the next chapter is dedicated to exactly that. It will be demonstrated that it is indeed possible to produce holograms with visible light similar to the ones described in this chapter in the original glass. The two methods will be also compared to reveal the advantages and disadvantages of each.

### 2.26. Appendix. Photosensitivity to blue light

As up to the present moment photosensitivity to visible radiation implied the possibility of photoexcitation by green light at the wavelength of 532 nm. Despite its high applicability as a common laser line, there is an interest of using other parts of the spectrum. In fact, the Tb:PTRG offers possibility of extending the photosensitivity range into the blue part of the visible spectrum, in particular for use at 488 nm which corresponds to the wavelength of an Argon laser. The photosensitivity mechanism in that case is based on a consecutive absorption of two photons with the energy of about 2.54 eV (488 nm) (see Fig. 48). This energy is fairly close to the resonant absorption band corresponding to the lower metastable level \(^5D_4\) which is located 2.6 eV above the ground state in Tb:PTRG. Stated differently the mechanism involves a single wavelength ESA upconversion process. Plausibility of the method was demonstrated by irradiating a sample of Tb:PTRG by radiation from a pulsed 10 ns laser providing 10 mJ/cm\(^2\) at 488 nm. After exposure and consecutive thermal treatment a refractive index of 120 ppm was measured in the sample. A big advantage over the mechanism presented above in the paper is that single wavelength ESA prevents any possibility of the issues related to non-uniform pumping which were reported for 532 nm recording. That in turn means potential for higher quality HOEs in comparison with those recorded using green light. On the downside while the wavelength of 532 nm corresponds to the maximum efficiency of the ESA process, the blue line falls on the shoulder of the efficiency curve,
and hence smaller maximum achievable RIM is expected. However an estimation showed that 4 W CW power at 488 nm is sufficient for holographic applications. No HOEs were recorded using blue light due to absence of the suitable laser sources available.

Figure 48  Energy level diagram of Tb$^{3+}$ with single wavelength excitation scheme using 488 nm radiation.
It appears that a volume hologram can be recorded in the original PTR glass by bleaching of color centers. The idea of using the color center bleaching method for hologram recording is not new; the method has been widely used for fabrication of amplitude holograms in inorganic glasses. Actually, this was one of the first approaches undertaken for holographic recording in vitreous media [49]. Color centers can be created in glass when electrons and holes are excited across the bandgap by means of ionizing radiation whereupon the electrons or holes are trapped on the impurities and various glass defects. Such trapped carrier, in fact, creates a color center in the glass. Those color centers can be identified by the additional absorption of the glass that spans from the UV region to the IR region and is known as induced absorption [60]. Color centers can be intrinsic if they belong to the glass matrix or extrinsic if they are related to impurities in glass. In all cases the color centers can be bleached by optical radiation with subsequent recombination of electrons and holes. Thus, the spatial absorption profile can be modified, and hence this effect can be used for fabrication of an amplitude hologram [107].

First holograms produced by this method were fabricated by bleaching of intrinsic color centers using radiation in the visible spectral region [108]. The main disadvantage of those holograms were low diffraction efficiency which was on the order of 1% as well as poor persistence which was caused by low stability of the intrinsic centers at room temperatures. Overtime the technique was improved to provide stability of holograms by bleaching of extrinsic color centers. Such holograms can be recorded in the CaF$_2$ crystals where stability is provided by thermal development of the hologram [109]. Yet such holograms still belonged to an amplitude type which means that only fairly low diffraction efficiency was attainable using that method. The maximum values
barely reached 15% which is still significantly lower than most up-to-date hologram recording materials. On the other hand, a possibility of using color center bleaching method for fabrication of phase holograms would be of primary interest. This implies that PTR glass with its extremely low absorption and stable holograms owing to the thermal development can be considered as a suitable candidate for the objective. This chapter describes an attempt to implement the color centers bleaching approach for the PTR glass to enable visible light recording.

3.1. Photosensitivity of photo-thermo-refractive glass to visible light

A method for fabrication of HOEs using visible radiation in PTR glass was presented a while ago, where a hologram could be fabricated by consecutive exposure to UV and visible radiation [110]. It has to be noted that the type of glass used in the procedure was the original Ce$^{3+}$ doped PTR glass. Recording procedure in that case involved two consecutive exposures of a glass sample, first to a spatially uniform UV radiation and then to high irradiance beam from a nanosecond pulsed laser operating at 532 nm. The hologram was created by a patterned visible beam on a uniform background provided by UV radiation.

When Ce doped PTR glass is exposed to spatially uniform UV radiation, the Ce$^{3+}$ ion undergoes an upward transition to its excited state which is situated above the electron mobility threshold in PTR glass. As a consequence, Ce$^{3+}$ ion is converted to a Ce$^{4+}$ ion thus releasing an electron. A mobile electron migrates from cerium ion and is trapped on defects in the glass known as color centers. Color center formation concludes the chemical transformations that accompany UV exposure stage. It should be recalled that the UV exposure is followed by thermal development in the standard procedure of a UV hologram fabrication in PTR glass. As glass is heated to elevated temperatures at the onset of thermal development, electrons are released from the defect states where they are captured by silver ions converting the latter to neutral silver atoms. Heating also
causes the silver atoms to diffuse and create silver containing clusters. Finally, the clusters serve as nucleation centers for the growth of NaF crystals at the second stage of thermal development carried out at temperatures above 500 °C. As crystals are grown, a negative RIC takes place in the UV exposed areas of the glass sample. It can be inferred that color centers created during the UV exposure stage supply electrons for creation of silver containing clusters during thermal development, and hence they directly affect the RIC produced in the glass.

The color centers can be also bleached before thermal development procedure begins whereupon electron is released from the trap and is recombined with the Ce⁴⁺ ion. While the latter undergoes a reverse transformation thus restoring the Ce³⁺ state, no contribution to the creation of silver cluster takes place. In [110] the color centers were bleached using high irradiance pulsed radiation from a 532 nm laser. The areas illuminated with high irradiance visible radiation were found to obtain smaller RIC compared to the surrounding areas exposed to UV radiation alone. Hence, the areas exposed to visible radiation exhibited effective positive change of refractive index with respect to the UV exposed background. Thus, if visible beam is structured with information, a hologram can be fabricated in PTR glass using this technique, in fact, such holograms produced with visible light were demonstrated in [110]. However, it has to be noted that the magnitude of RIC obtained in [110] was significantly smaller compared to one produced by standard UV exposure in PTR glass. Small RIC values such as those demonstrated are essentially insufficient for creation of an HOE with high efficiency.

3.2. Optimization of the recording procedure for high-efficiency holograms

The present paper employed the approach described above for the fabrication of complex HOEs for visible light application. It showed also that bleaching with higher efficiency can be done resulting in larger values of RIC. It was demonstrated that an optimized recording procedure allows
for recording of transmitting holograms including complex HOEs with efficiency approaching that in the holograms created in the original PTR glass created with UV radiation. The highlights of the method are discussed and advantages and disadvantages of both methods are compared.

3.2.1 Effect of cerium concentration

First, different parameters which affect the efficiency of the bleaching process were studied. Efficiency here is determined by the maximum attainable positive RIC due to visible radiation. Factors such as cerium concentration, UV excitation wavelength, visible beam irradiance, and development time were found to influence the efficiency. It was identified that the RIC induced by visible radiation tends to increase for glasses with increased Ce$^{3+}$ concentration (Fig. 49). In fact, the glass with the cerium concentration decreased by 3 times compared to the standard value was found to be lower than in standard glass. Similarly the glass with ten times the standard cerium concentration was measured to have larger RIC compared to the standard glass. Influence of cerium concentration comes from the probabilistic nature of bleaching process. Here the electrons released from the trap by optical bleaching have a possibility of being captured by a Ce$^{4+}$ ion, returning back to the original defect state, or being captured by another trap [111]. Consequently, the higher the cerium concentration – the higher the probability of the electron being captured by a cerium ion resulting in restoration of the Ce$^{3+}$ state. That, in turn, leads to a larger number of color centers bleached and eventually in larger positive effective RIC. Still it has to be noted that the glass with 20 times the standard Ce$^{3+}$ concentration demonstrated a decrease in the efficiency of the bleaching process. It is fair to assume that a competing process exists whose physical nature however remains yet unknown.
3.2.2 Effect of the UV excitation wavelength

The UV excitation wavelength was found to be the second parameter to affect the efficiency of the bleaching process. Bleaching of color centers was carried out in the PTR glass samples previously exposed to UV radiation from the two sources emitting within the range of Ce$^{3+}$ absorption band. The sources included a He-Cd laser operating at 325 nm, used in the standard recording procedure in PTR glass, as well as pulsed laser emitting the third harmonic of Nd:YAG. The dosages of UV radiation were normalized according to the magnitude of the Ce$^{3+}$ absorption to ensure that the same amount of UV radiation was absorbed for all wavelengths. All glass samples were exposed afterwards to the same dosage of high irradiance visible radiation at 532 nm from the second harmonic of the Nd:YAG laser. The effective positive RIC measured in the samples was found to be larger when UV exposure was carried out at 355 nm compared to that when 325 nm radiation was used for exposure of the PTR glass. The difference in RIC between
the two cases amounted to approximately 3 times for the glass with 10 times standard Ce$^{3+}$
concentration. Meanwhile the corresponding values were only found to differ by less than 1.5
times when glass with the standard Ce$^{3+}$ concentration was investigated. The difference was even
smaller for the glass with lower Ce$^{3+}$ concentration.

3.2.3 Proposed explanation for UV wavelength dependence

In support of the experimental data it should be assumed that color centers of not one but several
(at least two) types are formed as the result of the UV exposure which differ in their activation
energies. In fact, one type of color center appears to be bleached more readily than the other.
However, that assumption alone does not explain the effect of the UV excitation wavelength. The
wavelength effect comes from different UV photon energies used for excitation. Different photon
energies result in favorable conditions for formation of one or the other color centers [112, 113].
In the case under consideration it appears that low photon energy, corresponding to 355 nm
exposure, results in creation of a larger amount of color centers that are susceptible to bleaching.
By contrast, standard UV excitation with 325 nm laser leads to smaller amount of bleachable color
centers. As a result, more color centers are going to be bleached and, consequently, larger effective
positive RIC will be obtained in the glass subjected to UV exposure at 355 nm. The analysis of the
UV excitation performed in the paper is far from comprehensive as the number of available sources
was limited. The analysis is planned to be continued by an optical parametric oscillation device
whose output tunable across the entire range of Ce$^{3+}$ absorption band can be used for more in-
depth study.

3.2.4 Dependence of photosensitivity on exposure dosages

The effect of optical bleaching wavelength on the efficiency of the process was not studied due
to lack of available high power and high irradiance light sources other than the mentioned 532 nm
in the range of interest. The effect of the exposure dosages however was investigated for both precursory UV exposure and the visible light exposure. It was found that the effective positive RIC increases with UV dosage until saturation. The change in refractive index was measured to increase until the dosage per square cm of 355 nm radiation reached 100 J/cm² whereupon the growths of the RIC ceases. Growth cessation here can be explained by the saturation of color center formation, and the consequent UV illumination results in de-excitation of Ce⁴⁺ ions which is a competing process in that case. The dependence of the RIC on the dosage of visible light was found to be similar to the UV dosage dependence. The RIC was discovered to increase with the dosage of visible light until saturation occurs. The maximum value of RIC was found at the dosage of visible light of 32 kJ/cm² after which the increase of dosage did not seem to affect the RIC. Here the magnitude of the maximum effective positive RIC constituted less than a third of the refractive index decrement due to UV exposure. It follows that a fairly large portion of the color centers are insusceptible to bleaching which appears to be consistent with the assumption of several color center types that was made above.

3.2.5 Effect of thermal development time

Thermal development duration was determined to be another important parameter to influence the effective positive RIC. The RIC dependence on the development time was discovered to exhibit a maximum, thus an optimum duration of thermal development was found to exist (Fig. 50). This dependence is drastically different from that of the UV induced RIC which is known to continuously increase with thermal development time until saturation. The effect can be accounted for by the different rate of crystal growth in the areas exposed to the UV radiation only (type A) and those that underwent consecutive UV and visible exposures (Type B). Since certain amount of color centers was bleached in the type B areas, that can be approached as those areas were
simply exposed to a smaller dosage of UV radiation. Hence, the amount of crystal phase in the A
type areas will experience faster growth compared to that in the type B areas [114]. Hence RIC
will grow faster in the type A areas than in the type B areas but the saturation will occur sooner
for the type A areas while RIC in the areas of the type B will be still increasing until saturated at
longer development times. It follows that the dependence of the effective positive RIC on the
duration of the thermal development should have a maximum. An optimum development time
(corresponding to the maximum RIM was indeed determined in the course of the experiments.
Glass samples developed for the time periods shorter or longer than referred optimum time were
measured to have smaller effective refractive index increment.
Finally, the efficiency of bleaching was studied as a function of intensity/irradiance of both UV and visible sources. Intensity/irradiance of the UV sources did not demonstrate any effect on the bleaching procedure. Here the samples exposed to UV radiation showed the same initial refractive index decrement due to UV exposure and effective positive RIC due to visible light regardless of the intensity/irradiance of the UV radiation given the same total absorbed dosage of UV radiation. This is consistent with the current understanding of the photosensitivity process in PTR glass where RIC scales linearly with UV dosage [54]. By contrast, irradiance of the visible 532 nm beam was found to have a profound effect on the bleaching efficiency. Effective positive RIC was measured to increase quadratically with the beam irradiance while no RIC due to visible light was

3.2.6 Effect of irradiance of the light sources

Finally, the efficiency of bleaching was studied as a function of intensity/irradiance of both UV and visible sources. Intensity/irradiance of the UV sources did not demonstrate any effect on the bleaching procedure. Here the samples exposed to UV radiation showed the same initial refractive index decrement due to UV exposure and effective positive RIC due to visible light regardless of the intensity/irradiance of the UV radiation given the same total absorbed dosage of UV radiation. This is consistent with the current understanding of the photosensitivity process in PTR glass where RIC scales linearly with UV dosage [54]. By contrast, irradiance of the visible 532 nm beam was found to have a profound effect on the bleaching efficiency. Effective positive RIC was measured to increase quadratically with the beam irradiance while no RIC due to visible light was
found in the glass illuminated with low irradiance beams. This shows that the bleaching is in fact a non-linear process which appears to be consistent with the results shown in [110].

3.2.7 Results of photosensitivity optimization

The collected experimental data rendered possible to determine the important parameters of the recording procedure in order to obtain the maximum possible RIC induced by visible light. The maximum RIC was found in the glass with 10 times standard Ce³⁺ concentration exposed to 100 J of radiation at 355 nm and bleached with 32 kJ/cm² of the visible radiation from a 532 pulsed laser beam with the pulse energy per area of 0.6 J/cm² and was equal to 170 ppm. Significantly shorter duration of thermal development was used to achieve this RIC in comparison with the standard development time. It has to be noted that the maximum RIC is not limited by the value presented above, and larger values can be obtained if bleaching is performed by a beam with higher irradiance. For instance, a RIC of almost 300 ppm was demonstrated for 4 J/cm² pulse energy per area of the visible beam. The latter was however obtained by focusing the visible beam at full irradiance, and hence those values are inaccessible in the actual recording procedure. Effective RIM of 170 ppm is thus the maximum value that can be targeted in the hologram. Nevertheless, that value is almost by an order of magnitude larger than that demonstrated in [111] (25 ppm). Such increase in RIM is accounted for the use of the PTR glass with higher concentration of Ce³⁺, lower energy UV excitation, and shorter thermal development. Larger RIM in the hologram, in turn, indicates higher diffraction efficiency. HOEs were fabricated in PTR glass using the technique and parameters described above.
3.3. Transmitting holographic optical elements fabricated in photo-thermo-refractive glass by color center bleaching

Transmitting HOEs including planar transmitting VBGs (TBG) as well as complex holograms were recorded using the bleaching approach in Ce doped PTR glass. The vibration isolation technique outlined in a previous chapter was employed again in this case to ensure that the efficiency of the hologram remains high and is not deteriorated by any random or broadband noise present during recording. A TBG was the first type of hologram to be recorded by consecutive exposure to UV and visible radiation. The plate of PTR glass was first illuminated to a uniform UV beam at 355 nm resulting in the exposure energy density of 32 J/cm$^2$. The subsequent holographic recording was performed using the setup shown in the Fig. 51 which resembled the TBG recording setup in Tb:PTR glass with the exception that this time, of course, no UV pump beam was present. Total exposure to visible light amounted to 32 kJ/cm$^2$, and as a result, the relative diffraction efficiency of 99% was measured in the grating while absolute efficiency was found to be 91%. Similarly, a holographic lens was recorded in Ce10 glass using a setup depicted in the Fig. 52. It can be seen from the picture that the beam alignment in this case was identical to the one in Tb:PTRG with the exception of UV pump which was not used. The diffraction efficiency of the holographic lens was measured to be 97%.
3.3.1 Imaging performance of the holographic lens

The advantage of the color center bleaching method for holographic recording is in the absence of UV beams, hence only visible signal beams are employed. This eliminates the risk of non-uniformity issue and results in improved quality of the hologram as well as enhanced the imaging.
performance which is one of the most important properties of a complex HOE such as lens. As an HOE is considered as a substitution for a conventional mechanical optical element, it is of utmost concern that the performance of the original object is most closely reproduced by the hologram. Attention has to be paid to any possible additional aberrations introduced by a hologram into the imaging system. This is most conveniently determined by the beam spot size which will inevitably increase should any aberration be present. The minimum beam spot size was measured in the focal plane of both original lens system (Fig 53A) and the holographic lens (Fig. 53B). It can be inferred from the pictures that the imaging system comprising the holographic lens showed only 8% increase in the RMS spot size with respect to 250 μm in the original lens system. It may be deduced that based on the high diffraction efficiency and acceptable imaging performance, the holographic lens can be considered as a substitution for the original conventional lens system.

![Figure 53](image)

Figure 53  Dependence of photosensitivity (RIC) to visible radiation on duration of thermal development. Dependence holds for both UV wavelengths.

### 3.4. Transmitting holographic optical elements fabricated in photo-thermo-refractive glass by color center bleaching

The results above showed that transmitting HOEs can be recorded in PTR glass with visible radiation when fringe stability is maintained. Nevertheless, despite all efforts to mitigate the instabilities, there will be residual instabilities present during recording. Even those, residual
instabilities will be significant for recording of HOEs with high spatial frequencies that exceed 5 million lines per mm. Such structures include reflective HOEs such as reflective gratings with the pitch below 300 nm. It was in fact experimentally demonstrated that passive stabilization system had little effect on the performance of the reflective structures whose efficiencies were still well below 1%. Indeed, fringe stability had to be maintained as precise as \(\lambda/20-\lambda/30\) to obtain reasonable diffraction efficiency of 95%. The stringent conditions require the use of the active phase stabilization systems.

Phase stabilization systems have been widely used for holography and interferometric applications. An active phase stabilization system contains a phase detection system which detects the relative phase of the recording beam with respect to the reference beam, a feedback loop, and a phase-shifter which is used to provide compensation for the detected phase change. Typically, a piezo-transducer (PZT), mounted to one of the mirrors, is used as a phase shifting element as it changes the optical path in a corresponding arm of the recording system. What distinguishes the different stabilization systems is the phase detection system which can be designed in different ways. The most common approach for phase detection has the recording beam being passed through an auxiliary grating at Bragg condition [115]. Here intensity of the diffracted beam emerging from the auxiliary grating is detected. As phase of the recording beam is changed, the deviation from resonance condition occurs resulting in decrease in the efficiency of the detected diffracted beam. That approach demonstrated effective control of the fringe pattern however possessed a drawback attributed to the auxiliary grating. It appeared that a different auxiliary grating had to be fabricated every time the recording setup was modified. The necessity of an auxiliary grating can be avoided in the fringe stabilization system invented by [116] where phase detection is based on the beam recombination using a corner cube retroreflector. The stabilization
system was initially developed for recording gratings using UV radiation in PTR glass, and can be used for fabrication of gratings of any geometry and significantly simplifies the alignment of the system. The main mechanism presented in [116] was used for fringe stabilization for visible light recording described in the present paper.

The recording system used for visible light recording, however, presented a few challenges that had to be taken into account. First challenge is associated with the recording beam which is a high energy pulsed beam from a Q-switched laser. That beam has enough energy to cause damage to optics not compatible with high-energy radiation. Besides, significant amount of energy had to be extracted from the system so that to avoid dissipation of that energy inside the recording box causing severe heating problem. For that reason a beam-recombination and phase detection approach was taken from [117] which utilizes different geometry. Here the recording and reference beams are sampled upon passing through the recording plate and recombined using a beam splitter. The recombined beams are maintained codirectional by correcting for an appropriate interference pattern of the beams. Interference pattern of the beams is sent onto a photodetector. The intensity measured by the detector is given by

$$I = I_S + I_R + 2\sqrt{I_SI_R} \cos \varphi$$  

(3.1)

where,

$$\varphi = \psi + \psi_d \sin(\Omega D)$$  

(3.2)

is the phase between the wave fronts of the two beams. The first harmonic term of $\Omega$ is described by

$$I^\Omega = 4J_1(\psi_d) \sqrt{I_SI_R} \sin \psi \sin(\Omega t)$$  

(3.3)

This term is actually collected by the photodetector. Deviation of the phase will thus result in the change of the detector signal. The latter is used to compute the proportional gain which is supplied
to the PZT. The PZT moves the mirror so as to compensate for the phase change measured at the
detector. As can be seen from [117], the system possesses good sensitivity to the lateral shift as it
directly corresponds to the shift of the interference fringes measured by the photodetector.

All of the above however is true in the case of a CW recording beam such as that used in the
reference above. The current system however had to interact with the pulsed radiation. Detection
of the pulsed radiation presents a number of challenges. It is known that the system converts the
detected optical power into the phase information. Laser pulse energy, however, exhibits pulse-
to-pulse fluctuations that introduce a substantial amount of noise into the system.

The pulse energy can be averaged over time, however, the pulse rate of a Q-switched laser is
fairly low which means a fraction-of-wavelength displacement can be detected in several seconds
which is by several orders of magnitude longer than an average settling time of the vibration
isolation system in the optical table. Due to the challenges presented here, a different method based
on a probe beam was implemented in the current setup for fringe stabilization.

The current fringe stabilization system in application to RBG recording is presented in the Fig.
54. A probe beam at the wavelength of 543 nm is introduced into the setup using the same
beamsplitter as used to split the recording beam. After the beamsplitter the paths of the reference
beam and the transmitted probe beams are aligned in the horizontal plane, whereas the reflected
probe beam path follows the path of the signal beam. In the vertical plane the axis of the probe
beam is displaced above the axis of the appropriate recording beam by 10 mm. This allows for
convenient separation of the beams afterwards. Pinhole alignment is used to ensure that the paths
of the signal and the probe beam actually coincide. While the signal beam passes through the glass
sample the probe beam does not. This is not critical since neither the thickness of the plate nor its
refractive index changes during the recording as was shown before. On the other hand, this
drastically simplifies the alignment of the system. Upon passing the intersection point the beams are sampled and then directed towards the second beam splitter. Here the signal and the reference beams are blocked and the probe beams are recombined and sent onto the photodetector. Arrived at the photodetector is thus the interference pattern of the two parts of the probe beam. The intensity is measured at the detector whereupon the feedback system comes in operation as outlined above in the paper. PZT is used to move the turning mirror in the reference arm thus controlling the relative phase of the recording beam.

It is worthy of note that the described stabilization system is inferior to the one reported in [116] in terms of sensitivity since the beam does not return along its path and hence the phase deviations are not doubled. However, the new system allows for high-power beam alignment for which the system in [116] is unsuitable. Another drawback of the new system is the presence of an idle path where phase deviations are only introduced into the probe beam and not the recording beam. The effect of the this is however minimized by making the paths of the two probe beams equal. It is also counterintuitive to provide compensation for a beam at a different wavelength where optical paths can theoretically differ by 2% which can in theory introduce an error in the phase stabilization. Nevertheless, the equal arm lengths ensure that the difference of the phase deviation of the two wavelengths will be the same as the difference in the phases introduced by the system compensation. Concluding the analysis it can be seen that the system is supposed to provide adequate control of the fringes although it is less sensitive to the phase instabilities.
3.5. Fabrication of reflective holographic optical elements

The active phase stabilization system was employed for recording of the reflective HOEs that are known to possess much higher spatial frequencies than transmissive elements and thus could not solely rely on vibration isolation methods. Again, a planar reflective grating, an RBG, was chosen as a first element for recording. Those elements allow for simple comparison with its counterparts fabricated with UV radiation. For the recording procedure, the Fig. 3.6 should be again referred. It can be seen that unlike the UV indirect recording, recording here is performed using a direct method hence recording is carried out at the same geometry as the readout afterwards. That prevents any possible mismatch between the targeted and actual Bragg wavelengths. The recording arms are maintained close to equal for improved accuracy of the phase detection. Probe beam paths are maintained equal as well which is controlled by the visibility of the detected interference pattern.
RBGs were recorded by bleaching technique in 6 mm thick plates of Ce10 glass previously exposed to 32 J/cm$^2$ of UV radiation. Diffraction efficiency of 99% requires a RIM of 117 ppm which corresponds to 64 kJ/cm$^2$ dosage or two hours of recording. Even with phase stabilization in operation two hour duration of recording seemed unreasonable hence lower diffraction efficiencies were targeted. The recordings were carried out with the following target diffraction efficiencies: 90% at 50 ppm (32 kJ/cm$^2$ recording dosage), 71% at 35 ppm (19.2 kJ/cm$^2$ recording dosage) and 50% at 25 ppm (12.8 kJ/cm$^2$ recording dosage). The characterization of the resulting gratings have shown that the phase stabilization system is operational, and, indeed, improves the performance of the gratings, however the phase instabilities are not entirely eliminated and there are some concerns in the system operation yet to be addressed. Here it appeared that the phase stabilization system performed more effectively for shorter durations of the recording. The maximum diffraction efficiency of 40% was measured in the RBG recorded with 12.8 kJ/cm$^2$ dosage (24 min) where 50% efficiency was targeted. The efficiency curve for the grating is presented in the Fig. 55, it can be seen that the modulation depth is indeed decreased which indicates the presence of the hologram smearing. The RBGs recorded with larger dosages demonstrated lower efficiencies despite the expectations. It is unclear yet what exactly is the cause of the residual fringe shift during recording. An effect of gradual drift of the phase was observed with variable amplitude during recordings. In certain cases the drift was sufficiently strong so that the phase deviation exceeded the accepted limit of the phase stabilization system resulting in destruction of the hologram. It is believed that the effect has a cumulative nature such as that related to a thermal process which, however, has not been detected. Hence the detrimental effect is increased as recording duration increases. The source of the drift will be investigated in the
future using equipment such as thermal vison cameras to detect any possible heating with high
precision.

Meanwhile the visible light recording approach is mostly valuable owing to its ability for
fabrication of complex holograms which is not possible with UV radiation. An example of a
complex HOE in reflecting geometry is a curved mirror. An attempt was made to fabricate a
holographic curved mirror in Ce10 using high-power radiation at 532 nm. The recording setup for
fabrication of the curved mirror was based on the RBG recording setup and is demonstrated in the
Fig. 56. The focusing function was introduced by placing a lens system in the signal arm of the
recording stage. Here the signal beam was first passed through a negative lens resulting in
divergence of the beam. Diameter of the signal beam was thus increased. The beam was then

![Relative diffraction efficiency of RBG versus incident beam angle. RBG recorded using visible light.](image)

Figure 55  Relative diffraction efficiency of RBG versus incident beam angle. RBG recorded using visible light.

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recording stage. Here the signal beam was first passed through a negative lens resulting in
divergence of the beam. Diameter of the signal beam was thus increased. The beam was then
collected by a fast positive lens and thus ultimately focused emerging from the lens system. The position of the lens system was chosen so that by the time the collimated reference beam and the focused signal beam reach their intersection point, their diameters are equal. Equal beam sizes ensured the most efficient use of beam energy for recording.

Operation of the holographic curved mirror at 532 nm in the focusing and defocusing modes is shown in the Fig. 57. It can be seen that the size of the reflected beam can be increased or decreased depending on the orientation of the holographic plate. Size of the collimated transmitted beam is shown for comparison. The targeted diffraction efficiencies, dosage used, and hence time durations were the same as those for the RBGs. Similarly, the maximum diffraction efficiency of 38% was measured for a hologram fabricated with 12.8 kJ/cm² dosage (24 min exposure). The efficiency curve of the holographic mirror is presented in the fig. 58 and is drastically different from that of an RBG in the way that its angular acceptance appears to be significantly larger.

![Setup for recording of holographic curved mirror with visible light using hologram fringe stabilization system.](image_url)

Figure 56 Setup for recording of holographic curved mirror with visible light using hologram fringe stabilization system.
Figure 57  Operation of holographic curved mirror recorded in PTR glass.

Figure 58  Relative diffraction efficiency of holographic curved mirror versus incident beam angle. Mirror recorded in PTR glass using visible light.
3.6. Comparison with the method for holographic recording in Tb:PTRG

One might most certainly notice that fabrication of an HOE by means of visible light is what the described technique has in common with the technique based on recording in Tb:PTR glass discussed in the previous chapter. The technique described in the present paper involves consecutive exposure to UV and visible radiation of PTR glass whereas Tb:PTRG is simultaneously exposed to UV and visible beams. Resemblance can be noted in the photosensitivity levels as well as recording setups. Hence the interest is aroused in comparison of the two techniques in particular regarding their holographic capabilities. A comparative analysis showed that two techniques yield fairly similar results in terms of performance demonstrated by the HOEs. On one hand, Tb:PTRG, which was designed for visible light applications, has somewhat higher photosensitivity to visible light than the Ce doped PTR glass. In fact, Tb:PTRG demonstrated maximum RIC of over 450 ppm due to visible radiation whereas 300 ppm was obtained in the Ce doped PTR glass in the similar exposure conditions. RIC larger by 50% can be certainly considered as an advantage of Tb:PTRG and simultaneous exposure as it allows for reduction of exposure times which, in turn, results in less severe effects from the recording instabilities. On the other hand, the presence of both UV and visible beams during recording is a drawback of the technique. The problem resides in the UV beam which has to provide perfectly uniform illumination to ensure good quality of recorded holograms. Unfortunately, real world beams often exhibit non-uniform spatial intensity profile which results in parasitic modulation of the refractive index ultimately leading to decreased diffraction efficiency. By contrast, only visible signal beams are used in the recording technique described in the this chapter which eliminates the risk of non-uniformity issue resulting in performance deterioration. In evidence, the diffraction efficiency of all HOEs recording in the regular PTR glass by means of the visible radiation was measured to be on average by 10% higher than that in the elements recorded in Tb:PTRG.
3.7. **Conclusion for holographic recording using bleaching of color centers**

In conclusion, a method for fabricartion of HOEs in PTR glass with visible radiation by means of color center bleaching was demonstrated. The technique is implemented by consecutive exposure of the Ce doped PTR glass sample to spatially uniform UV radiation and patterned visible beam followed by the thermal development. Both planar volume Bragg gratings and nonplanar HOEs (holographic lens and mirror) operating in the visible spectral range were recorded using that technique. Transmission holograms fabricated by visible light demonstrated remarkable performance approaching that in the UV recorded VBGs. Implementation of an active phase stabilization system rendered possible to achieve better performance of the reflective HOEs. Diffraction efficiency of the RBGs was increased from below 1% to 40%.
CHAPTER 4: HOLOGRAPHIC PHASE MASKS FOR BEAM SHAPING APPLICATIONS IN PHOTO-THERMO-REFRACTIVE GLASS

It has been mentioned before that the PTR glass is a photosensitive material that was designed for fabrication of high quality VBGs. Furthermore, the glass exhibits notable optical and mechanical properties compared to other holographic materials which make it attractive for a wide range of holographic applications. Indeed, in the previous paragraphs the possibility to extend the capability of the glass for recording of complex holograms was demonstrated. Despite certain challenges related to fabrication of complex holograms, the subject appears to be of high interest because of the advantages that those elements possess when recorded in PTR glass. For example, PTR glass is a unique holographic material which allows for fabrication of complex HOEs with high power and heat stability. This cannot be achieved with other materials such as photopolymers that are intolerant to high power radiation and high temperatures. This is the reason why it is advantageous to develop complex holography application in PTR glass.

Yet the capability of a holographic material is not limited to holograms only. For instance, such material as photopolymer is used for fabrication of other types of structures as well including photonic crystals, waveguides, and phase masks with refractive index change. In particular, the latter are optical elements that introduce a phase incursion into the transmitted beam resulting in a change in the beam intensity profile in the far field. Thus, those elements can be employed for generation of beams with various spatial intensity profiles from the original laser output beam, typically that of a Gaussian intensity distribution. Hence, phase masks are tools for beam shaping alongside spatial light modulators, q-plates, and refractive optics. The advantage of phase masks over the competing methods is that the phase profile can be tailored for a particular application where production of a single optical element is less expensive compared, for instance, to the cost
of using a spatial light modulator. On the other hand, low durability and low tolerance to high power due to limitations of the photopolymer could be sited as disadvantages of the elements.

Stability of PTR glass to high power laser radiation is the reason why it is of interest as a material for production of phase masks. If phase masks can be fabricated in PTR glass, those optical elements will be tolerant to high power radiation. This would make possible to use the phase masks in PTR glass for shaping of high power beams which could be useful for material processing applications. Normally, shaping of high power beams can be carried out using phase masks of different type, those produced using surface profiling of a glass plate. On the other hand, UV exposure is significantly less expensive compared to etching and deposition techniques. If the same element can be created by refractive index change instead of surface profiling, that means significant reduction of the cost of beam shaping.

In fact, phase masks in PTR glass have been demonstrated before [118]. Those masks were shown to be able to perform mode conversion and generate several different spatial modes. The masks were created using a contact-copy method with the aid of an amplitude mask, the method which is typically employed for fabrication of phase masks in a photopolymer. Yet, the phase masks created using this method contained binary phase profiles whereas most beam shapes used in material processing can be generated with greyscale phase profiles. This chapter describes a novel technique for fabrication of phase masks in PTR glass where phase masks can be of either binary or greyscale type. The method is based on a digital micromirror device (DMD) and allows for production of masks for generation of beams used in material processing such as vortex (helical) beam. It renders possible to avoid surface processing and thus reduce the cost of phase masks with greyscale phase profile by fabricating them with refractive index change in PTR glass.
This together with high-power stability of PTR glass makes them an attractive solution for laser material processing.

The proposed method, however, does not eliminate a profound disadvantage of phase mask which, as a phase sensitive element, can only operate at a single wavelength. This means that several different optical elements have to be fabricated for operation at various wavelengths. This chapter elaborates on how the issue can be resolved by using a holographic phase mask instead of a regular phase mask. Holographic phase mask is based on a TBG and allows for multi-wavelength operation. Those elements have been demonstrated in PTR glass [56] and are produced by encoding a desired phase profile into a transmitting grating. The same technique was employed in order to encode greyscale phase profiles into TBGs so that such beam shapes as vortices can be generated at multiple wavelengths.

4.1. Beam shaping

Laser beam shaping is a technique of altering the spatial distribution of phase or intensity of a laser beam. Typically, the beam intensity profile determines the shape of the beam whereas its propagation characteristics are governed by the phase profile. Being able to control the beam shape appears to be very useful since numerous applications can greatly benefit from using a laser beam whose wavefront possesses a particular shape [119-120]. While most lasers emit radiation with Gaussian spatial intensity profile, in many instances other beam shapes are more suitable. There are a variety of various beam shapes intended for different applications. The beam shapes most commonly used for material processing are top-hat, ring, and multispot profiles. Top-hat and ring shaped beams are described in more detail below. It will be shown that, in fact, application of those two beam shapes stretch far beyond material processing and hence there is a high interest in capability of generation of those beams. Later, the novel technique for generation of such beam
shapes using phase masks created in PTR glass is described. However, first it is reasonable to familiarize with the beam shapes and their properties and applications.

### 4.2. Top-hat beams

Top-hat beam is a beam whose intensity remains constant over a certain spatial region. Those beams are widely used for laser material processing including drilling, ablation, welding, etc [121-123]. The uniform intensity spot provides homogeneous illumination of the working surface to ensure that all areas of the workpiece are processed evenly. The area of the beam possessing uniform intensity can itself be round, rectangular, square, etc. in shape. The edges of the area must be sharp to ensure that there is a clear boundary between treated and untreated areas. Mathematically, such shape would be described with a rectangular function \( \text{rect}(r) \). In the real world, however, top-hat beams tend to have smoother edges where the spatial profile can be approximated using a supergaussian formula

\[
I(r) = I_0 \exp\left(-2\left(\frac{r}{w}\right)^n\right) \tag{4.1}
\]

where \( w \) is the beam waist and \( n \) is the order of the supergaussian polynomial, and the edges are sharper for larger \( n \).

Laser material processing is not the only application of top-hat beams. Historically, semiconductor lithography has been the primary application where the beams were used to provide uniform illumination of the wafer while trying to prevent diffraction effects in order to achieve smaller feature size [124]. Nowadays, the beams with uniform intensity are widely employed for drilling of holes in integrated circuit assemblies and trimming of circuit components. Other applications include optical data storage, medical, laser printing, and imaging. The importance of the uniform illumination for imaging purposes will be evident as the process of fabrication of phase masks will be explained further in the text. Yet phase masks are not the only tools for generation
of top-hat beam profiles. Several different techniques have been developed over time to produce a uniform intensity pattern from a Gaussian beam.

4.3. Generation of top-hat beams

First technique for beam shaping involved cutting a Gaussian beam by means of apertures to obtain a modified spatial profile [119, 125]. The resulting profile was dependent on the positioning of the cutting aperture. The range of attainable beam profiles in this case was limited to parts of the Gaussian distribution while also the beam power was significantly reduced. Those issues can be avoided by using another beam shaping technique known as field mapping. This technique uses optics that bend the light rays in such fashion that the uniform intensity profile is obtained [126]. In its simplest case, the optical system consists of two elements with the shapes of the optics chosen so that the second element compensates for the phase introduced by the first element. As a result, a beam passed through both lenses will emerge having a uniform intensity distribution. Field mapping allows for better control over the resulting profile shape and renders possible to eliminate any power loss. However, the resulting profile is highly dependent on beam size and there is an additional issue related to diffraction effects that can result in deterioration of the targeted beam profile. For that reason, the applications that require higher accuracy of the intensity profile employ a different beam shaping method based on beam integrators. A beam integrator typically consists of two elements, a lens array and a focusing lens. A lens array splits the beam into multiple sections where each section obtains a particular phase change. The focusing lens collects all the beams and overlaps them again creating a uniform spatial profile [127]. This technique allows for the highest beam quality of the three methods and although it can suffer from various diffraction effects, their weight is negligible for relatively large beam sizes. The high uniformity, however, comes at a cost with the need of an expensive lens array. For that reason, the use of the other two techniques is
preferred for applications where high homogeneity is not critical. It has to be noted that such techniques as beam apertures can be only applied for generation of uniform intensity profiles. Generation of other beam shapes requires more sophisticated methods as it is in the case of a vortex beam.

4.4. Vortex beams and their applications

Another beam type often used for material processing purposes is the vortex beam, which is a beam with a wavefront twisted around the propagation axis. Its cross section has a ring shaped intensity profile with an optical vortex (zero intensity) in the middle. This type of beam is commonly employed for laser drilling, micromachining, and cutting where the presence of optical vortex facilitates heat and material removal from the processed area [128, 129]. Yet the application range of vortex beams stretches far beyond material processing. This can be accounted for a property of the vortex beams to exhibit a helical phase dependence of \( \exp(il\phi) \) and therefore possess optical angular momentum (OAM). The magnitude of the OAM is known as the topological charge and is equal to the number of times the wavefront is twisted around the axis within one wavelength [130, 131]. Thus, there is an infinite number of vortex beam variations having different intensity profiles and connected by the zero intensity property.

Vortex beams are also known as helical beams which corresponds to helical Ince-Gaussian (IG) modes since the transverse intensity of a vortex beam can be described as a superposition of even and odd IG polynomials in the form

\[
HIG_{p,m}^\pm(r) = IG_{p,m}^e(r) \pm IG_{p,m}^o(r)
\]

(4.2),

where even and odd IG polynomials are given by

\[
IG_{p,m}^e(r) = \frac{A\omega_0}{\omega(z)} C_p^m(\xi) C_p^m(\eta) \times \exp \left[ \left( \frac{-r^2}{\omega^2(z)} \right) + i \left( k z + \frac{k r^2}{2 R(z)} - (p + 1) \Phi_G(z) \right) \right]
\]

(4.3),
\[ IG_{p,m}^o(r) = \frac{B\omega_0}{\omega(x)} S_p^m(i\xi) S_p^m(\eta) \times \exp \left[ \left( -\frac{r^2}{\omega^2(x)} \right) + i \left( kz + \frac{kr^2}{2R(z)} - (p + 1)\Phi_G(z) \right) \right] \] (4.4),

where \( p \) and \( m \) are the order and the degree of the beam [132, 133]. Vortex beams of different order and degree are demonstrated in the Fig. 59. The top row depicts transverse intensity profiles of vortex beams given by

\[ |HIG_{p,m}^\pm(r)|^2 \] (4.5)

The lower row contains the phase distributions needed to generate the corresponding vortex beam. The required phase distribution can be described by

\[ \text{Im} \left( \log HIG_{p,m}^\pm(r) \right) \] (4.6)

Grey level in the picture indicates a phase shift which varies from 0 to 2\( \pi \). It can be inferred from the pictures that the area of the zero intensity increases with the mode order whereas the degree of the beam determines the number of gradient sections in the meridional direction [134]. The two types of helical phase profiles shown in the first two images on the left are known as vortex and double-vortex profiles, respectively. Those phase distributions were selected as patterns for greyscale phase masks in PTR glass as examples of beam shaping optics for material processing. Besides material processing, vortex beams can be applied in variety of areas that can benefit from their OAM property.
OAM beams have recently become a topic of an intensive research and found their way into a range of applications from micromanipulation to telecommunications. One of the most promising applications of vortex beams is optical tweezers where circular polarization allows for the rotation of the trapped particle around the beam axis [135, 136]. Spatial division multiplexing (SDM) is yet another application of OAM beams and it is addressed in the present paper. Dividing data streams into individual channels encoded by means of orbital angular momentum (OAM) is one way to achieve increased optical bandwidth of a communication link. Vortex beams of different helical modes are mutually orthogonal allowing for efficient multiplexing of channels without crosstalk [137-139]. This technology can be successfully employed to achieve increased capacity of fiber and free space optical communications, which is highly demanded due to the growing amount of data.

### 4.5. Generation of helical beams

Vortex beams possess spatial phase and intensity distributions which appear too complex to obtain by refracting techniques discussed above, hence, diffractive elements have to be used for
precise phase manipulation. Vortex beams are typically generated by means of special type of phase mask where phase distribution is defined by the thickness profile rather than the change of the refractive index. Those optical elements are also known as vortex lenses or spiral phase plates [140, 141]. Those elements are normally produced from the transparent plate of glass or polymer where spatial selective etching and deposition are employed to modify the surface profile in order for the desired phase distribution to be achieved. A photograph shown in the picture 60. clearly explains the idea behind the spiral phase plates. The drawback of using the phase mask of this type is sensitivity to environmental conditions such as humidity. It has to be noted that the elements made out of glass are more durable than their counterparts produced in a polymer but their fabrication is more expensive due to expenses for glass processing. In fact, high cost of surface processing makes those phase masks an expensive solution regardless of the material. Besides, high chromaticity of the elements have to be taken into account which is resulted from their phase dependence. For that reason, the use of spiral phase plates can be justified in the case where single source is used for beam shaping and only one certain beam shape is of interest. In the instances where operation at multiple wavelengths is desired, other methods of vortex beam generation should be considered.

![Figure 60 Spiral phase plate.](image-url)

Generation of helical beams at multiple wavelengths became possible with the emergence of spatial light modulators (SLMs). Since their advent in the eighties, SLMs worked their way from
bulky devices incorporating only a few sections/pixels to high precision devices with thousands of pixels. High resolution SLMs can produce virtually any phase pattern and, hence, can be used for generation of a wide variety of beams [142-144]. SLMs are, however, not compatible with high power laser radiation because of their liquid crystal nature and this encumbers their application for material processing. Besides, the high price of the SLM technology presents another challenge for its implementation particularly in the areas where only one single beam shape needs to be generated. The alternative methods for production of vortex beams include q-plates and diffractive axicons which have their advantages as well as limitations.

4.6. Phase masks in Photo-thermo-refractive glass

The motivation for creating phase masks for generation of helical beams is in high durability of the elements to the environmental factors such as temperature and humidity as well as lower fabrication costs since, instead of surface profiling, the elements are created by profiling of refractive index across the volume of the glass plate. In fact, binary phase elements have been demonstrated in PTR glass, and those were produced using contact-copy method which is also typically used for production of phase masks in photopolymers [145]. The contact-copy method employs an amplitude mask where certain areas are coated with chromium to block the light incident on the mask whereas light can freely transmit through its other areas. In order to produce a phase mask, a plate of PTR glass is placed in contact with the amplitude mask and is illuminated with UV radiation through the amplitude mask. The areas of the glass exposed to UV radiation will ultimately attain a change of the refractive index.

Binary phase masks, such as the four-sector phase mask illustrated in the Fig. 61a, were created using the indicated method [118]. This mask is used for generation of a free-space TEM$_{11}$ (LG$_{04}$) mode from a Gaussian beam as the latter is incident onto the center of the mask. For
characterization of the mask, the latter was illuminated by a collimated beam from a 633 nm He-Ne laser. As phase incursion was introduced into the beam upon transmission through the mask, Fourier transform was provided by a positive lens, and the far field intensity distribution was observed at the focal plane of the lens. As a result, a Gaussian beam was demonstrated to experience mode conversion to the \( \text{TEM}_{11} \) spatial mode as the beam was incident on the center of the mask as can be seen from the Fig. 61b. A \( \text{TEM}_{01} \) mode was possible to obtain as the beam was incident on the horizontal boundary area between the sections in the way that it is shown in the Fig 61c. All phase mask were designed for operation at a fixed wavelength of 633 nm.

4.7. Holographic phase masks

Single wavelength operation is a fundamental disadvantage of phase mask as a phase sensitive element. This implies that a new optical element has to be fabricated each time a laser source of a different wavelength is used. On the other hand, the approach undertaken in the present paper uses a holographic phase mask (HPM) produced in the volume of PTR glass which lacks the above mentioned downsides such as single-wavelength operation or low durability. An HPM is a quasi-
achromatic optical element that is produced by encoding a desired phase distribution from a corresponding phase mask into a transmitting volume Bragg grating (TBG) [56]. The resulting element will resemble a transmitting grating in appearance, and the profile of the grating will be modified according to the encoded phase distribution. For example, the profile of a regular grating is shown in the Fig. 62a with proper periodicity of the hologram fringes. The same grating encoded with the four-sector distribution will possess a profile with fringes displaced from the regular interference pattern positions exhibiting a phase shift of \( \pi \) at the boundary between the sections (Fig. 62b).

Holographic grating with encoded phase information will diffract light just as a standard grating, in addition the diffracted beam will experience mode conversion according to the phase incursion encoded into the grating. Therefore, mode conversion properties of a phase mask and an HPM are identical. The main advantage of an HPM over a standard phase mask is that an HPM is capable of performing the desired mode conversion over a wide range of wavelengths. Wavelength tunability originates from the property of a TBG that allows for tuning of the Bragg condition by adjusting the incident angle. In the case of an HPM, adjustment of the angle of incidence results in the Bragg condition being met for different wavelengths whereas the phase information incurred into the beam remains unchanged. This is drastically different from a regular phase mask where imposed phase is directly dependent on the wavelength. This implies that several phase masks operating at different wavelengths can be effectively replaced with one holographic phase mask. Encoding of the phase profile into a grating is carried out by placing the phase mask into one of the arms of a two-beam interference system. In fact, this technique of HPM fabrication has been already presented. TBGs were encoded with the four-sector profiles discussed above whereupon generation of the free space TEM\(_{11} \) mode was demonstrated at different wavelengths [56]. The
same technique was employed for fabrication of HPMs encoded with other binary phase profiles (eight-sector etc.) from the corresponding master phase masks.

4.8. Novelty of this work

It can be inferred that the phase masks and HPMs fabricated in PTR glass are robust devices insensitive to environmental conditions that allow for shaping of high power beams. However, the variety of beam shapes possible to generate with that technique is fairly limited. Only beam shapes corresponding to binary phase profiles can be generated as those profiles can be produced in phase masks with contact-copy method. On the other hand, such beams as vortex beams that are very useful for a number of application including material processing, can be only generated using phase masks containing greyscale phase distribution. While fabrication of such elements is possible using the contact-copy method, very high cost of greyscale amplitude mask fabrication, even in comparison with spiral phase plates, render the devices economically unviable. If an alternative, less expensive method for fabrication of phase masks in PTR glass were found, that would significantly extend the range of producible optical elements. The novelty of the present

Figure 62  A. Profile of regular transmitting grating B. Profile of grating encoded with four-sector phase profile.
paper is indeed in the method of production of phase masks in PTR glass by refractive index profiling. The presented method is versatile and allows for relatively simple and inexpensive fabrication of phase masks with greyscale phase profiles. The phase masks discussed in this paper were created using a Digital micromirror device (DMD). Greyscale phase profiles, in turn, render possible to produce an HPM encoded with a vortex profiles that are capable of generation of helical beams as shown below. The fabrication method is cost effective in comparison with contact-copy and surface profile techniques while phase masks can be used for shaping of high power beams and are extremely robust as phase information is contained inside the volume of a glass plate.

4.9. Digital micromirror device

The phase masks presented in this paper were created with the aid of a DMD which is a special type of spatial light modulator similar to the one shown in the Fig. 63. The device contains an array of micron-sized mirrors each of which can be individually addressed to tilt to an angle of 12°. This angle corresponds to an “on” state when incident light is reflected by the mirror [146]. Applying voltage to individual mirrors will switch the mirrors between “off” and “on” positions, and in such manner the direction and amplitude of incident light can be modulated. The DMD allows for reflection of only the desired portions of the incident beam thus acting as an amplitude mask. If the amplitude modulated reflected beam is incident on a slab of photosensitive material such as PTR glass, radiation from the mirrors in the “on” position will be incident onto the glass surface and will result in refractive index change. Therefore, amplitude modulation produced by a DMD was converted into a phase modulation by volume refractive index profiling. In the experiments described in the present paper the DMD was used in conjunction with an imaging system to image the exact spatial pattern generated at the DMD plane into the slab of PTR glass. This was done in
order to avoid distortion of the spatial pattern due to diffraction while propagating from the DMD plane which could result in deterioration of the quality of phase mask.

Figure 63  Digital micromirror device.

The capability of producing binary phase patterns similar to ones obtained by contact-copy method was not of highest importance in this paper. The main advantage of imaging a DMD pattern was in the ability to reproduce greyscale patterns. Grayscale phase distribution can be produced by varying the effective (total) time of mirror opening during the exposure. Here if the mirrors are in “on” position for the entire duration of exposure, the areas on the sample would acquire a phase shift of $2\pi$. On the other hand, if some mirrors are in “on” position only for a certain fraction of exposure time, the areas collecting radiation from those mirrors will obtain smaller phase shift. By selecting the suitable effective opening time, it was possible to produce any phase shifts in the interval between 0 to $2\pi$. The DMD used in the experiments provided 256 grey levels which along with the high pixel resolution ($1080\times1920$) enabled successful implementation of the linear phase gradient in the vortex phase mask. It is understood that the recording of patterns with fine details can be only implemented in a photosensitive material with high resolution. In the case under consideration the phase masks as well as the holographic phase masks were fabricated in the volume of PTR glass which provided recording resolution as high as 10,000 lines/mm.
4.10. The phase mask recording system

The phase masks were produced by imaging the desired pattern from the DMD into the volume of 1-2 mm thick plates of PTR glass. The refractive index change Δn with respect to unexposed areas targeted in the PTR glass slab was dependent on the slab thickness t and the operating wavelength λ according to

\[ \Delta \varphi = \frac{2\pi}{\lambda} t \Delta n \]  \hspace{1cm} (4.7).

That was done by adjusting the UV exposure of the plate or duration of its thermal development according to the sensitivity curve of PTR glass for the corresponding UV source. The phase mask recording system with the DMD device, henceforth referred as the recording system, used in the present paper, is demonstrated in the Fig. 64. The DMD is illuminated by a spatially uniform UV beam coming from a UV light source. Over the course of the experimental work mainly two sources including a 325 nm He-Cd UV laser and a mercury lamp were used as light sources. The recording UV beam is patterned upon reflection from the DMD chip. Then, the reflected beam is passed through a 4f imaging system which includes a Fourier lens, an iris for spatial filtering located at the Fourier plane, and the second Fourier lens that produces the pattern identical to that projected by the DMD. Ratio of the focal lengths is maintained at 2:1 to provide larger dimension of phase masks which was found to improve the beam shaping performance. The spatial filter was used to remove the noise in the image which was possible due to dust or scattered light present in the system.
4.11. Concerns regarding illumination field uniformity and power stability

It was found extremely important that the light field illuminating the DMD plane has spatially uniform intensity. Uniform illumination ensured that all areas with constant target refractive index change indeed had exactly the same refractive index change leading to the uniform phase profile. Presence of non-uniformities causes irregularities in the phase profile of the fabricated phase mask, and this, in turn, will result in generation of a beam with distorted shape. In this regard it was extremely important to homogenize the intensity of radiation from the utilized sources. In the course of work measures were taken to convert the output of the mercury lamp and the He-Cd laser into a spatially uniform field.

Homogenization was performed differently for the two sources as demonstrated in the Fig. 65. Here the emission of the mercury lamp possessed a quasi-supergaussian profile with a relative flat top region. Owing to the high power of lamp emission (3 W), it was possible to simply expand the beam and then select the uniform region for the imaging purposes. The beam was then collimating by means of a lens whose aperture also served as a spatial filtering element. The resulting illumination was measured to have uniformity on the order of λ/10 which was comparable with the surface relief of PTR glass plates.
By contrast, the laser emission was of a Gaussian intensity profile of considerably high quality. Expansion of such beam will not eliminate phase variation in the way it was possible to achieve with the lamp besides significantly lower power of 40 mW made it impractical. For that reason a system of aspheric UV lenses was used to transform the beam shape whereupon it was expanded to match the size of the DMD chip. As a result, notably good uniformity of the illumination of no more than $\lambda/10$ was measured. This property was found to be particularly crucial for recording of phase masks with gradient phase profiles such as vortex masks as accurate targeting of refracting index change results in precise implementation of the desired linear phase gradient.

The alternative methods for obtaining uniform light field exist and include lens arrays, rotating diffusers, as well as modal interference in a multimode fiber [147]. Lens arrays were omitted due to high cost of such devices whereas rotating diffusors resulted in considerable loss of power. One
method, implemented along the ones discussed above, was the use of modal interference. Here the UV light from the He-Cd laser was coupled into a multimode fiber at an angle corresponding to favorable excitation of the higher order modes. The launching angle was then adjusted until a flat-top beam was obtained at the output of the fiber. Mechanical vibrations transmitted to the fiber via a speaker provided constant variations of modal interference in order to prevent speckle formation. The light field uniformity achieved with this method was found satisfactory however the method was ultimately abandoned due to large output power instabilities caused by the changes in modal interference.

Output power stability was another property which was found to be critical for fabrication of high quality phase masks. Whereas spatial uniformity was essential to produce homogeneous spatial distribution of the refractive index, power stability affected the accuracy of the exposure metering which, in turn, resulted in an error in the targeted RIC leading to an improper phase shift. In the course of experiments the exposure was targeted by adjusting the time necessary to obtain the desired RIC at the measured average power. Deviations of the source power from the average value resulted in decreased accuracy of produced RIC. In this regard the mercury lamp emission was preferable since it exhibited little instabilities in power of less than 0.1%. By contrast, the laser emission demonstrated larger fluctuations of power on the order of 5% which affected the accuracy of the induced refractive index and phase which was found to have an effect on the performance of the phase masks. Nevertheless, the issue in the case of the laser emission was ultimately not critical as power was found to fluctuate evenly around the average value thus the average value was sufficiently stable for long exposures.
4.12. Edge sharpness maintenance

The phase profiles of both binary and greyscale phase masks contain several regions with different phase increments and the edges that are formed by the boundaries between adjacent sections. Sharpness of the boundaries between regions has to be maintained and the edge profile has to resemble step with the highest possible accuracy to ensure that the desired beam shape will be obtained. Smearing of the step profile will affect the performance of the phase mask and will result in aberration introduced into the beam. Smearing of the edges, in turn, can happen due to natural diffraction of light as it propagates towards the sample as well as aberrations caused by the imaging system where chromatic aberration was found to have the strongest effect.

Diffraction effects were mitigated by means of an imaging system which projected the spatial pattern located at the DMD plane directly into a PTR glass plate. Here, several factors had to be taken into account as they affected the sharpness of the edges. Those were focusing accuracy, depth of focus, and thickness of the sample. The 4f-system presented in Fig. 4.6 was employed in order to increase the depth of focus of the imaging system and decrease the negative effect of smearing, and samples with small thicknesses were used for the same reason. Here, in the course of the experimental work it was found that phase masks recorded in a 1 mm thick slabs generally show better performance compared to those made in a 2 mm thick glass, and it was imperative that the slab of PTR glass was placed at the focal plane with the highest possible accuracy.

However, even if all the requirements stated above are met, smearing of the boundaries is possible due to chromatic aberration if the light source exhibits a broad emission spectrum, which was the case with the mercury lamp. This problem was addressed by using a special coating on the DMD window that only allowed transmission of radiation within the desired wavelength range onto the DMD chip while the rest of the spectrum was reflected away from the window. Additional correction for chromatic aberrations was attained by using UV achromats for the 345-370 nm
wavelength region as Fourier lenses. All the precautions allowed to significantly alleviate the problem of chromatic aberrations since only radiation within one spectral peak of the mercury lamp emission spectrum was incident on the sample. However, regardless of all the measures, it was impossible to eliminate the problem completely. In that regard, the He-Cd gas laser was found to be a preferable tool for fabrication of high quality phase masks. As a monochromatic source, the gas laser was devoid of chromatic aberration and, hence, demonstrated by far superior boundary sharpness. This advantage was particularly important in the instances where the projected pattern contained fine details as it was in the case of the Fresnel zone plate. On the downside of using the laser is the homogenization of its radiation which proved to be more cumbersome compared to the simple expansion method available with the mercury lamp.

4.13. Performance test with a binary phase mask

The challenges related with production of high quality phase masks using the recording system encouraged to perform a system test to ensure the expected performance of the recorded optical elements. This can be achieved by fabricating a test phase mask whose performance can be compared to a similar element fabricated in PTR glass using a contact-copy method. Since only binary phase distributions could be fabricated with contact-copy approach, a binary four-sector phase mask for operation at 633 nm was selected as a test phase mask. The mask was fabricated by the recording system whereupon its beam shaping performance was compared to that by a similar mask made using contact-copy method. Spatial phase distribution was measured by means of a Fizeau interferometer and is demonstrated in false color in the Fig. 66a. Beam shaping performance was evaluated by producing mode conversion of a Gaussian beam using both test mask and the etalon mask, made by means of photolithography, and comparing the far field intensity profiles after mode conversion. A Gaussian beam was converted to TEM\textsubscript{11} free-space
transverse mode as it passed through both masks: one produced by photolithography (Fig. 66b) as well as the one made with the recording system (Fig. 66c). It can be seen from the figure that both phase masks demonstrated almost identical conversions to $\text{TEM}_{11}$ mode. It was inferred that the phase masks produced by the recording system show performance on par with their counterparts fabricated by photolithography. Hence, the imaging performance of the system made possible to continue work on phase masks incorporating more complex phase profiles.

![Image](image.png)

Figure 66 Four-sector phase mask with actual phase profile (photograph of measurement screen) (A), mode conversion by reference mask (intensity profile) (C), mode conversion by test mask (photograph) (D).

### 4.14. Greyscale phase masks for generation of helical beams

Among many complex phase profiles, the ones containing vortices were of utmost interest in this paper which is accounted for open prospects for application of vortex beams combined with challenges of their generation. For that reason this work was focused on phase masks for generation of helical beams. In fact, as a proof of concept, two of the profiles were selected including vortex and double-vortex phase profiles for generation of helical beams of first and second order, respectively. The phase profile of the vortex mask produced by means of the recording system and measured using a Fizeau interferometer is shown in the Fig. 67a. The phase undergoes a linear change from 0 to $2\pi$ in a full circle around the mask where it experiences a step of $2\pi$. This phase mask can be used to convert a Gaussian beam into a helical Ince-Gaussian beam of the first order.
IG$_1$ with an optical vortex in the middle. Important quality parameters of the mask include radial phase which should be uniform, meridional phase profile which must experience a linear change from 0 to $\pi$, and, inevitably, the sharpness of the borderline between 0 and $\pi$. In reality, the linear phase gradient was substituted for 16 sections with consecutively increasing phase increments. This was done in order to facilitate the procedure of programming procedure of the DMD. It can be seen in the measured phase profile, indeed, that the phase is increasing in discrete steps as opposed to linear increase. Black areas that can be noticed in the vicinity of the phaseshift are not related to the mask performance. The areas are the result of limitation of the camera used in the interferometer which did not possess sufficient resolution to discern fairly sharp step profile. Black areas correspond to the regions where camera data was dropped due to excessively sharp phase shift. Another method based on a Shearing interferometer was employed for evaluation of the edge sharpness and, as a result, the mask was proved to be suitable for beam shaping. The phase mask shown in the figure was fabricated to operate with a laser beam at the wavelength of 633 nm. Beam shaping was performed by illuminating the phase mask with a collimated laser beam whose size was chosen so as to cover the major portion of the mask while not exceeding its dimensions. The output of the mask was transformed by means of a Fourier lens and the far field intensity distribution was analyzed in the focal plane of the lens. The vortex mask demonstrated conversion of the incident Gaussian beam into a helical beam of the first order IG$_1$. The resulting ring shape intensity profile is shown in Fig. 67b and closely resembles the profiles expected from numerical simulation results [134]. The minor discrepancies are believed to be related to the use of a discrete step phase gradient as opposed to the smooth linear gradient. Development of the advanced DMD patterns for phase mask fabrication is expected to provide improved beam shaping performance.
It has to be noted that while the presence of zero intensity is necessary condition for observation of a vortex beam, it is not sufficient. Also the beam has to possess OAM. To ensure that the generated beam in fact has OAM, the phase mask was placed into one of the arms of a Mach-Zender interferometer operating at the wavelength of 633 nm whereupon the phase mask output was interfered with a plane wave from a beam shaper. A fork interference profile observed upon the beam overlap and shown in the Fig. 67c demonstrates OAM property of the generated beam. Thus it was shown that the phase mask produced in PTR glass is capable of generation of vortex beams. Fig. 67d demonstrates an IG₁ mode generated from a Gaussian beam by means of a similar mask produced for operation at 1064 nm.

Figure 67 Vortex phase mask with actual phase profile (photograph of measurement screen) (A), mode conversion of beam at 633 nm (photograph) (B), interferogram with plane wave (C), mode conversion of beam at 1064 nm (photograph) (D).

IG₁ mode is one example of the great variety of helical beams of different shapes and properties. Another phase mask known as double-vortex phase mask, was fabricated in PTR glass for
generation of helical beam of the second order corresponding to IG\(_2\) mode. The phase mask has more complex phase profile as compared to the vortex mask discussed above with two consecutive 0 to 2\(\pi\) gradients around the full-circle. The phase profile measured in the mask by means of the Fizeau interferometer is demonstrated in the Fig. 4.10a. The same comments regarding the discrete step increment of phase and dropped data given for the vortex mask can be also applied for the double-vortex phase mask. Mode conversion produced by the element was consistent with the expectations based on the theoretical modelling [134] and is shown in the Fig. 4.10b. As predicted, the area of the optical vortex increased with the order of the helical mode whereas the number of intensity nulls in the meridional direction increased with the mode degree.

![Image](image.jpg)

Figure 68 Double-vortex phase mask with actual phase profile (photograph of measurement screen) (A) and mode.

### 4.15. Holographic phase masks for generation of vortex beams

A capability of generation of helical beams of different orders and degrees was demonstrated using phase masks produced in PTR glass. Phase masks, produced using the technique described above, can themselves be used for generation of vortex beams as long as they are designed to operate at the desired wavelength. However, of high interest is the possibility of using the elements in conjunction with the technique of phase encoding into a TBG. As described above in the text,
the phase profile from the mask can be holographically encoded into a TBG resulting in production of an HPM. Now that it has become possible to encode HPM with greyscale profiles such as vortices, the corresponding helical modes can be generated at a wide range of wavelengths.

The holographic setup for recording of an HPM is illustrated in the Fig. 69. It can be seen from the diagram that the setup, in fact, resembles that for recording of a regular TBG with a phase mask inserted in one of the arms. The phase masks have to be encoded with a desired phase profile and they have to be designed for operation at 325 nm, the emission wavelength of a He-CD laser. As the UV beam is split, portion of it is directed towards the phase mask where the corresponding phase incursion is introduced. 4f system was implemented in the recording setup for exact imaging of the phase profile into a TBG.

![Figure 69 Setup for recording of holographic phase mask.](image)

An HPM encoded with a vortex phase profile was fabricated using the setup described above. The profile of a vortex HPM is presented in the Fig. 70a and is essentially a fork profile since it is obtained by interference of a plane wave with a helical beam. The optical element demonstrated diffraction of an incident beam while the diffracted beam was simultaneously converted to a vortex
beam of the first order. The far field intensity profile of the diffracted beam is shown in the Fig. 70b-d and is consistent with that obtained using a regular phase mask. The crucial difference of an HPM is in the capability for multi-wavelength operation. In fact, the vortex HPM was tested at several wavelengths of the visible and IR spectral regions. The angle of incident beam was adjusted to satisfy the Bragg condition for each wavelength, and, as a result, similar mode conversion was achieved in all cases. Figures 70b-d demonstrate generation of a vortex beam at 450 nm, 633 nm, and 1064 nm with almost identical far field intensity profiles. The only parameter of the device that changed with wavelength was diffraction efficiency. Since an HPM is essentially a TBG with an addition of a phase incursion, similarly to a TBG, an HPM possesses a diffraction efficiency property. Relative diffraction efficiency can be as high as 100% in TBGs recorded in PTR glass and this also applies to HPMs. In the case under consideration the diffraction efficiency at the resonance wavelength of 1064 nm was measured to be 95%. As the wavelength was tuned away from the resonance, the efficiency will be inevitably decreased, however, the value can be maintained well over 50% in the range of 0.5 μm.

![Image](image_url)

Figure 70 (a) Degenerate clockwise and anti-clockwise modes in a single ring resonator (b) Degeneracy is lifted upon a perturbation by a nanoparticle.

Similarly to the HPM described above, another HPM was encoded with the double-vortex phase profile. The HPM was fabricated in the same fashion as explained above. As a result, the double-vortex HPM demonstrated simultaneous diffraction and generation of second order vortex beam.
Operation was tested at the resonance wavelength of 1064 nm as well as at the visible wavelength of 633 nm with very similar intensity distributions (Fig. 71). Wavelengths longer than 1064 have not been tested in the experiments due to absence of corresponding laser sources at the moment of the experiment. As far as diffraction efficiency is concerned, that was measured to be equal to 94% at the resonance.

![Figure 71 Generation of helical IG2 beam by double-vortex holographic phase mask at two different wavelengths.](image)

### 4.16. Multiplexed holographic phase mask for simultaneous generation of several vortex beams

It was already shown in the paragraphs above that HPMs produced in PTR glass can be used for generation of individual helical modes at a wide range of wavelengths. Besides, the functionality of the optical elements can be further extended using the multiplexing property of PTR glass. Owing to the two-stage process of fabrication of optical elements in PTR glass, several volume gratings can be consecutively recorded in a single volume of PTR glass. Since no RIC is present in the glass before thermal development, subsequent recording will not erase or alternate structures produced before. Similar to TBGs, several HPMs, encoded with different phase profiles, can be combined in the single slab of glass. Such device can be then used to produce several different vortex beams, one at a time. Alternatively, the elements can work simultaneously if several
gratings are designed for the same angle of incidence. In the present paper two HPMs encoded with the vortex and the double-vortex phase profiles, respectively, were multiplexed in the same plate of PTR glass in such manner that both HPMs were designed for normal incidence while having opposite symmetrical diffraction angles. Such element worked as a coherent beam splitter resulting in simultaneous generation of helical beams of the first and the second order. When the multiplexed HPM (MHPM) was illuminated with a Gaussian beam at normal incidence, the beam was split into two diffracted beams where each of the beams experienced mode conversion to the corresponding order vortex profile (See Fig. 72). The optical element was fabricated for operation at the wavelength of 1064 nm and provided a 50:50 power division between the two diffracted beams. The device is high-power stable, simple, and robust, and combines the functions of vortex beam generator and OAM mode multiplexer. MHPMs with larger number of fractions are projected to be fabricated in the volume of PTR glass in future using the presented recording system. It has to be noted that the number of individual HPMs composing the multiplexed element are only limited by the available maximum refractive index change ($10^{-3}$ in PTR glass). Such device allows for straightforward incorporation of spatial division multiplexing technology into existing FSO links, and is capable of use in high-power laser systems.
4.17. Beam homogenization using Fresnel lens array

While vortex beams certainly were of highest interest in the present paper, attention was also devoted to other beam shapes such as top-hat described above in the text. Its applications for material processing and photo-lithography have been already mentioned. However a special interest is that the top-hat intensity profile was required for the experiments described in the present paper. Thus the phase mask for generation of a top-hat profile would be beneficial for the phase mask recording system itself and can be placed into the recording system for producing uniform illumination of the DMD. The approach for homogenization undertaken in the present paper is based on the concept of a microlens array which is a common tool for homogenization of various light sources. Typically, lens array is incorporated into an optical system in the manner as it is shown in the Fig. 73. As a collimated beam is incident on the array, it is divided into multiple beamlets. The beamlets then pass through a spherical lens that overlaps them in the Fourier plane in such way that the total intensity is homogenized. In this way the principle of operation of a lens array is similar to beam integrator. Despite being very efficient homogenizers, lens arrays are used in limited number of applications due to their cost-intensive fabrication process. The idea of this
paper proposes to substitute an array of mechanical lenses with an array of Fresnel zone plates that could be recorded in a plate of PTR glass. Ultimately, the Fresnel zone plate array can be fabricated at a fraction of a cost of a microlens array and still achieve comparable performance acceptable for non-imaging applications.

First, a single Fresnel zone plate was fabricated in a plate of PTR glass using the recording system. The phase profile of the mask consists of radially symmetric rings known as zones where the phase shift between the adjacent zones alternates between 0 and $\pi$ radians. Radii of the zones are chosen in the way that light from even and odd zones interferes constructively in the focal point [44]. This type of phase profile is binary and contains details with high resolution, and for that reason it was fabricated using a He-Cd laser. Operation of the Fresnel zone plate for focusing of a laser beam at the wavelength of 633 nm is shown in the Fig. 74. Some astigmatism was detected in the lens which, however, is believed to be mostly caused by the shape of the original laser beam. The Fresnel zone plate had a focal length of 1000 mm and contained 17 distinguishable zones.
Lastly, following a successful demonstration of a Fresnel zone plate, an attempt was made to combine several elements on a single glass plate. A prototype of a Fresnel zone plate array was fabricated in PTR glass, and its performance was tested by observing multiple intense light spots at the focal plane of the device. It is understood that the device is not yet capable for operation as a homogenizer due to large separation of focusing elements. In the future, the distance between adjacent Fresnel zone plates is planned to be decreased while in the meantime increasing the efficiency of each focusing element. Then the device can be incorporated into the recording system to provide uniform illumination of the DMD while preserving power which is otherwise lost due to expansion.

4.18. Conclusion on holographic phase masks for beam shaping

In conclusion, a method has been demonstrated for relatively straightforward and inexpensive fabrication of phase masks with virtually any phase profile that can be used for generation of a wide range of optical beam shapes. For example, the phase masks can be encoded with vortex phase profiles that enable generation of helical beams. Helical beams of the first and the second order generated using the phase masks were demonstrated.
Phase masks are fabricated in PTR glass which gives an advantage of high-power stability. Besides the masks are not affected by high temperature or by other environmental effects, and possess considerably long shelf-life compared to their counterparts produced by photolithography in a photopolymer and also can contain greyscale profiles which are cumbersome to produce by photolithographic method. On the other hand, the fabrication of phase-masks in PTR glass is more cost-efficient compared to phase plates produced by surface processing. Wide range of beams, possible to generate with the phase mask, combined with the unique optical and mechanical properties of the material make phase masks in PTR glass an ideal solution for beam shaping for applications in material processing.

Despite all the advantages, phase masks have a fundamental limitation of single-wavelength operation. In this paper the novel technique of greyscale phase mask fabrication was combined with the method for encoding the phase profiles into a TBG resulting in fabrication of an HPM. Unlike the regular phase mask, an HPM is capable of multi-wavelength operation. For instance, one element was shown capable of generation of helical beams at a wide range of wavelengths. Finally, several HPMs can be multiplexed in a single volume of PTR glass which allows for generation of various beam shapes using the same element. MHPM was fabricated which splits an incident Gaussian beam into several diffraction orders where each diffraction order is converted to an individual helical mode.
The dissertation reports on a variety of optical elements fabricated in PTR glass for applications in the visible spectral region. PTR glass is a photosensitive silicate glass that allows for permanent refractive index change which is employed for fabrication of high-quality as volume Bragg gratings (VBGs). VBGs are planar structures, and for that reason they can be recorded using UV radiation for operation at different wavelengths such as those of the visible and IR spectral regions. This approach cannot be implemented in the case of complex holograms that can only be fully reconstructed at the wavelength at which they were recorded. Hence, it follows that complex holograms recorded in PTR glass can be only designed for operation in the UV spectral region. On the other hand, of interest are complex HOEs for the visible spectral region as those are of high interest and in demand for such as applications as LIDAR, virtual and augmented reality, free space optical communication etc. PTR glass is a photosensitive glass with outstanding optical and mechanical properties which is used for recording of high-quality optical elements such as VBGs. However the range of optical elements capable for production in the glass is limited to planar structures such as VBGs. This paper presents methods for extension of the glass capabilities for fabrication of complex holographic structures and demonstrates conceptually new optical elements in PTR glass. The research discussed in the paper was focused on fabrication of complex HOEs in PTR glass for applications in the visible spectral region. Production of such elements was inaccessible before due to photosensitivity range of the PTR glass limited to the UV region. This paper describes two different approaches for addressing the problem of the photosensitivity. First method involves modification of the PTR glass making it photosensitive to radiation in the visible spectral region and thus making possible to record holograms in PTR glass with visible radiation. By contrast, the second approach uses the original glass and introduces a new modified technique
for hologram formation that allows for holographic recording with visible light. However, the photosensitivity by both methods appeared to be significantly lower compared to the original glass resulting in longer exposure durations that, in turn, presented a challenge of stability during holographic recording. As a consequence of long exposure times, the quality of fabricated HOEs was compromised due to instabilities present during recording that caused smearing of holographic fringes. In the course of work the recording stability problem was addressed by means of both passive (vibration isolation) and active phase stabilization methods. Ultimately, both techniques demonstrated capability for holographic recording with visible light, and HOEs recorded with visible radiation were demonstrated. The presented elements offer new capabilities for laser beam control while preserving the unique properties peculiar to VBGs in PTR glass such as high-power and temperature stability.

The other types of optical elements presented were phase masks and HPMs. Phase masks are optical elements that introduce a spatial dependent phase change into a beam as the latter is transmitted through the mask. This leads to a change in the far field spatial intensity profile of the beam thus altering the shape of the beam. Indeed, phase masks can contain various phase distributions for generation of a great variety of optical beam shapes. This paper presented an innovative method that allows for fabrication of phase masks in PTR glass with virtually arbitrary phase profile including greyscale profiles. Here it is useful to highlight the phase masks produced in PTR glass that demonstrated generation of helical (vortex) beams. Those optical elements can be directly used for generation of helical beams in a single-wavelength system. However, the functionality of a phase mask can be extended as its phase profile can be also encoded into a TBG resulting in fabrication of an HPM. An HPM allows for beam shaping similarly to regular phase masks and on top of that possesses important advantages including multi-wavelength operation
and multiplexing ability. It has to be noted that both phase masks and HPM are robust devices that can be used for shaping of high power beams and whose operation is not affected by environmental conditions.

The optical elements produced in PTR glass possess exceptional properties that make them stand out from their counterparts produced in other holographic materials. Besides the scientific interest, those devices are of interest for a wide range of real-world applications. For instance, spatial and spectral selective properties of complex HOEs are beneficial in LIDARs, AR and VR systems, FSO communications etc. On the other hand, the phase masks and HPMs are in demand for material processing, free-space and fiber optical communications, imaging etc. Finally, phase masks and HPMs recorded in PTR glass facilitate generation of highly demanded vortex beams thus opening new prospects in various areas of scientific research.
REFERENCES


