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PARTICLE MANIPULATION VIA OPTICAL FORCES AND ENGINEERING SOFT-MATTER SYSTEMS WITH TUNABLE NONLINEARITIES

by

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Summer Term
2014

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ABSTRACT

One of the most intriguing properties of light-matter interaction is the ability of an electromagnetic field to exert mechanical forces on polarizable objects. This phenomenon is a direct consequence of the fact that light carries momentum, which in turn can be transferred to matter. Mediated by scattering, this interaction usually manifests itself as a “pushing force” in the direction of beam propagation. However, it is possible to judiciously engineer these optical forces, either by tailoring particle polarizability, and/or by structuring the incident light field. As a simple example, a tightly focused laser beam demonstrates strong gradient forces, which may attract and even trap particles with positive polarizability in the focal volume. The opposite occurs in the regime of negative polarizability, where particles are expelled from the regions of highest intensity. Based on this fundamental principle, one can actively shape the beam using spatial light modulators to manipulate individual objects as well as ensembles of particles suspended in a liquid. In the latter case, a modulation of the local particle concentration is associated with changes of the effective refractive index. The result is an artificial nonlinear medium, whose Kerr-type response can be readily tuned by the parameters of its constituent particles.

In the course of this work, we introduce a new class of synthetic colloidal suspensions exhibiting negative polarizabilities, and observe for the first time robust propagation and enhanced transmission of self-trapped light over long distances. Such light penetration in strongly scattering environments is enabled by the interplay between optical forces and self-activated transparency effects. We explore various approaches to the design of negative-polarizability
arrangements, including purely dielectric as well as metallic and hybrid nanoparticles. In particular, we find that plasmonic resonances allow for extremely high and spectrally tunable polarizabilities, leading to unique nonlinear light-matter interactions. Here, for the first time we were able to observe plasmonic resonant solitons over more than 25 diffraction lengths, in colloidal nanosuspensions.
ACKNOWLEDGMENTS

I would like to express my gratitude here to all the people who helped and supported me during my studies, above all my parents for their tremendous role in my educational path.

I would like to thank my advisor Professor Christodoulides for guiding me through my research and being patient with me, it was truly a great honor to work with him. Not only he is a creative scientist and caring advisor but also a great mentor for all his students. I am very grateful for everything he taught me.

I thank all the members of my committee for their time and stimulating discussions.

Special thanks to Professors: Zhigang Chen, Hagan, Van Stryland and Richardson for giving me a great opportunity to participate in their research and learn many things along the way.

I would like to thank all the CREOL staff, in particular Rachel who has been like a sister to me, Amy for her support, Gail, Richard, Deon, and Maria for their invaluable help and kindness.

Many thanks to my colleagues and friends, especially Dr. Samadi, Dr. Zhang, Dr. Heinrich, Dr. El-Ganainy, and Mr. Matthew Mills.

Finally, I would like to express my gratitude to my family; my dear Alessandro, wonderful in-laws, my brother Makan and sister Anya. They supported me with all their love and care. And since I know very well how annoying I can be when I am stressed, I thank them for being patient with me.
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**LIST OF ABBREVIATIONS**

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>1D</td>
<td>One Dimensional</td>
</tr>
<tr>
<td>2D</td>
<td>Two Dimensional</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full Width Half Maximum</td>
</tr>
<tr>
<td>SLM</td>
<td>Spatial Light Modulator</td>
</tr>
<tr>
<td>BPM</td>
<td>Beam Propagation Method</td>
</tr>
<tr>
<td>CW</td>
<td>Continuous Wave</td>
</tr>
<tr>
<td>NP</td>
<td>Negative Polarizability</td>
</tr>
<tr>
<td>PP</td>
<td>Positive Polarizability</td>
</tr>
<tr>
<td>MP</td>
<td>Mixed Polarizability</td>
</tr>
<tr>
<td>MNP</td>
<td>Metallic nano-particles</td>
</tr>
<tr>
<td>SPR</td>
<td>Surface Plasmon Resonance</td>
</tr>
<tr>
<td>QPD</td>
<td>Quadrant Photo-diode</td>
</tr>
<tr>
<td>SM</td>
<td>Steering Mirror</td>
</tr>
<tr>
<td>OT</td>
<td>Optical Trapping</td>
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CHAPTER ONE: INTRODUCTION

Since the invention of laser in 1960, this technology has influenced many different fields of science and technology due to its numerous properties. One of lasers many applications is the ability to manipulate matter via radiation forces, which nowadays plays a crucial role in several areas of physics, chemistry, and biology [1]. Optical trapping and manipulation of particles using lasers was pioneered by Ashkin three decades ago [2-6]. Since then this method has been used for transporting particles in a verity of sizes; from tens of nanometers to several micrometers, in many branches of science and has evolved significantly. The ability to manipulate matter optically, has been used widely in different branches of science. A few examples are; allowing scientists to control kinetic processes, study physical and mechanical properties of certain biological structures and how they function, the ability to sort mixed particles [7] micro-fabricate and study colloidal hydrodynamics and the possible nonlinear effects [5-7]. In chapter 2 we will discuss the principals of light-matter interactions that lead to optical forces which are the essence of optical manipulation. Briefly, there are two important optical forces that make manipulation with light, possible: One is the gradient forces which result from the intensity gradient near a focused laser beam and pull (repel) the particles to (from) maximum beam intensity, depending on particle polarizability sign. Another type is the scattering forces, which push particles along the direction of light propagation. In chapter 3, we first discus the principles of an optical tweezers setup, then examples of optical tweezers applications are demonstrated and a new method for simultaneously trapping and rotating particles and cells (Moire-based optical tweezers) will be experimentally presented.
Lasers have been used to study different types of light-matter interactions. The results of these studies have given in depth information, both macroscopically and microscopically about material properties, allowing scientists to further engineer them for novel applications. Synthesizing artificial material systems exhibiting unique optical properties not found in nature is nowadays one of the major scientific endeavors. This challenge becomes particularly acute in soft matter environments where light scattering loss has always been a formidable hurdle. In the nonlinear optics regime, this issue becomes even further complicated due to enhanced scattering, causing a dramatic drop in transmission. Clearly of importance will be to devise approaches to overcome these limitations. In chapter 4 through 6, we demonstrate that the nonlinear response of certain soft-matter systems can be tailored at will by appropriately engineering their optical polarizability. In particular in chapter 4, we observe synthesized colloidal nanosuspensions with negative polarizabilities, and observe for the first time robust propagation and enhanced transmission of self-trapped light that would have been otherwise impossible in nanosuspensions with positive polarizabilities. What greatly facilitates this behavior is an induced saturable nonlinear response accompanied by a substantial reduction in scattering via self-activated transparency effects. The close relation between the thermodynamic properties of these colloidal mixtures and the observed optical dynamics is also highlighted. These results may lead to new opportunities in developing novel versatile soft matter systems with tunable optical nonlinearities. In chapter 5 we demonstrate optical interactions between stable self-trapped optical beams in the aforementioned engineered soft-matter systems (with saturable self-focusing optical nonlinearities). Our experiments show that optical beam interactions can vary from attractive to repulsive, or can display an energy exchange depending on the initial relative
phases. The corresponding observations are in good agreement with theoretical predictions and behavior of solitons in photorefractive material. In Chapter 6, robust propagation of self-trapped light over distances exceeding 25 diffraction lengths has been demonstrated for the first time known, in plasmonic nanosuspensions. This phenomenon results from the interplay between optical forces and enhanced polarizability of the plasmonic particles that would have been otherwise impossible inside dielectric dispersions. Plasmonic nanostructures such as core-shell particles, nanorods and spheres are shown to display tunable polarizabilities depending on their size, shape, composition, as well as the wavelength of illumination. Here we demonstrate unique nonlinear light-matter dynamics in plasmonic nanosuspensions, which in turn allows for deep penetration of long needles of light through dissipative colloidal media.

In chapter 2 we demonstrate a general formulation for the optical forces acting on Mie particles. This general formulation is used in chapter 7 to theoretically analyze the optical forces exerted on a dielectric spherical particle illuminated by two non-interfering plane waves. We show both theoretically and experimentally how these beams of specific polarization are able to generate a transverse Poynting vector flow by demonstrating the particle's scattering pattern and it's motion.
CHAPTER TWO: LIGHT-MATTER INTERACTION AND OPTICAL FORCES

2.1. Light-matter Interaction Concepts

In this chapter, the different processes occurring in light-matter interaction will be discussed. In general, matter can react to an electromagnetic radiation in 4 ways. In Fig.1, this interaction and the following effects has been shown briefly. These effects are: 1. Reflection and Refraction, 2. Absorption and 3. Scattering. Among these losses [8], depending on the type of material and the wavelength, one dominates. The wavelength of radiation is an important factor; it determines the index of refraction and the loss coefficients. In many applications, knowing the absorption and scattering properties of our material is very important to fully understand the interaction.

![Figure 1 Geometry of reflection, refraction, absorption, and scattering.](image)

2.1.1. Absorption

While passing through a material, the intensity of an electro-magnetic wave is attenuated. The photon energy is converted into different electronic excitations or vibrations and rotations of the
absorbing molecules leading to heat. The absorption of a medium depends on several factors, most importantly is the atomic and molecular constitution of the atoms and molecules and the density of these absorbers, the wavelength of the light and thickness of the sample. The same material can be transparent in a certain range of wavelength, while opaque in another. As an example the cornea of the eye, though it seems transparent, it absorbs in the IR. There are two equation expressing absorption, each using different factors of the absorbing sample of study:

\[ I(z) = I_0 \ 10^{-az} = I_0 \ 10^{-\varepsilon cz} \]  

(2.1)

and

\[ I(z) = I_0 \exp(-\sigma Nz) \]  

(2.2)

Where \( I_0 \) and \( I \) are the intensity of the incident light and the transmitted light, respectively, \( \alpha \) is the absorption coefficient, \( z \) is the distance of propagation. The absorption coefficient can, in turn, be written as a product of the extinction coefficient of the absorber, \( \varepsilon \), and the concentration \( c \) of absorbing species in the material. \( \sigma \), is the absorption cross section and \( N \) is the density of absorbers. These equations are known as Beer-Lambert law.

In biological tissues, absorption is mainly caused by water absorbing in the IR, proteins and/or pigments absorbing in the UV and visible range [9]. In fig.2, the absorption of important cellular constituents and tissue fluorophores are demonstrated.
2.1.2. Scattering

When an electromagnetic wave is incident on an obstacle a redirection of the electromagnetic (EM) wave takes place, this phenomenon is called scattering. In more details; as the wave interacts with a particle, the electron cloud within the particle starts oscillating with the same frequency of the incident wave. The oscillation leads to induced oscillating dipole moment which would radiate EM waves. This radiation is known as scattered light. Most of the incoming light scattered by the particle is emitted at the same frequency (elastic scattering). In this process the particle size and shape and the frequency of the incoming light are important factors.

Overall the interaction of light with matter can reveal important information about the structure and dynamics of the material being examined. For instance structural repetition in the scattering medium will cause interference in the spectrum of scattered light. This fact helps to characterize
and extract information about the structure, spatial configuration, or morphology of the scattering medium.

![Figure 3: Scattering phenomena of an EM wave by a particle.](image)

Scattering is categorized based on; 1. size of particles and 2. whether the scattering is elastic or not.

2.1.2.a. Elastic Scattering

A general approach in finding the scattered field would be Mie theory for spherical particle of any type of material [11]. In this approach, the scattering intensity is sensitive to the particle size. If the particle is much smaller than the wavelength of incoming light, Rayleigh scattering would be the easier approach, which is strongly dependent to the wavelength of the light:

\[
I = I_0 \frac{1+\cos^2 \theta}{2R^2} \left( \frac{2\pi}{\lambda} \right)^4 \left( \frac{n^2-1}{n^2+1} \right)^2 \left( \frac{d}{\lambda} \right)^6
\]  

(2.3)

Here I is the intensity of scattered light, R is the distance to the particle, \( \theta \) is the scattering angle, \( n \) is the refractive index of the particle and \( d \) is the diameter of the particle. If a large particle is
non-spherical, there are several methods to solve the scattering problem. One method is the
discrete dipole approximation which the particle is considered as many Rayleigh particles [12].

2.1.2. b. Inelastic Scattering

If the wavelength of the scattered beam is the same as the incident, it would be elastic scattering
(Mie and Rayleigh). But in the case of Raman and Brillouin scattering the scattered beam would
show a shift in wavelength (inelastic). Brillouin scattering occurs from the interaction of photons
with acoustic phonons (vibrational quanta of lattice vibrations) in solids or with elastic waves in
liquids. The shift in frequency of the scattered light is very small (less than 100cm\(^{-1}\)). On the
other hand, in the Raman scattering the light interacts with optical phonons, which are
vibrational modes of the molecules with energies larger than acoustic phonons, leading to larger
shifts in frequency (100 to 4000cm\(^{-1}\)) [13].

2.2. Optical Forces

The fact that an electromagnetic field can exert mechanical forces upon polarizable objects was
known since the inception of Maxwell’s theory [14] and is a striking manifestation of the
principle of momentum conservation. In particular the momentum delivered by an
electromagnetic wave to an object in vacuum can be, at least in principle, exactly calculated by
evaluating the flux of the Maxwell’s stress tensor through a surface enclosing the scatterer [15].
Fundamentally more complex is the case of objects embedded in dielectric media, where the
very definition of electromagnetic momentum is not free of ambiguities. Nevertheless force
calculations based on the Maxwell’s stress tensor, albeit approximate, can be applied to a high
degree of accuracy also to objects embedded in incompressible dielectric media like fluids [16,17].

The most popular scattering term used so far is for the case of Rayleigh particles [18-21], where the scattering is described by the leading electric dipole contribution. Analytical expressions have recently been reported concerning the force contributions stemming from the first few higher order multipoles [22]. Here we will briefly observe the condition where these multipoles can play a role in the optical forces.

### 2.2.1. Optical Forces on Spherical Scatterers (Mie Regime)

In general, the average force experienced by a scattering and/or absorbing body immersed in a monochromatic field can be computed by evaluating the flux of the Maxwell stress tensor $\mathbf{T}$ through any virtual surface $S$ enclosing the object:

$$
\langle \mathbf{F} \rangle = \frac{1}{2} \text{Re} \left[ \int_S \mathbf{\hat{n}} \cdot \mathbf{T} \cdot dS \right]
$$

(2.4)

The Maxwell stress tensor is given by: 

$$
\mathbf{T} = \varepsilon_0 \varepsilon_r \mathbf{EE}^* + \mu_0 \mathbf{HH}^* - \frac{1}{2} \left( \varepsilon_0 \varepsilon_r \mathbf{E} \cdot \mathbf{E}^* + \mu_0 \mathbf{H} \cdot \mathbf{H}^* \right) \mathbf{I}
$$

Considering $\mathbf{E} = \mathbf{E}_t + \mathbf{E}_s$, $\mathbf{H} = \mathbf{H}_t + \mathbf{H}_s$ we can obtain the following expression for the Maxwell stress tensor:
\[ T = \left\{ \varepsilon_0 \varepsilon_\nu \mathbf{E}_i \cdot \mathbf{E}_i^* + \mu_0 \mathbf{H}_i \cdot \mathbf{H}_i^* - \frac{1}{2} \mathbf{I} \left[ \varepsilon_0 \varepsilon_\nu \mathbf{E}_s \cdot \mathbf{E}_s^* + \mu_0 \mathbf{H}_s \cdot \mathbf{H}_s^* \right] \right\} + \]
\[ + \left\{ \varepsilon_0 \varepsilon_\nu \mathbf{E}_i \cdot \mathbf{E}_s^* + \mu_0 \mathbf{H}_i \cdot \mathbf{H}_s^* - \frac{1}{2} \mathbf{I} \left[ \varepsilon_0 \varepsilon_\nu \mathbf{E}_s \cdot \mathbf{E}_i^* + \mu_0 \mathbf{H}_s \cdot \mathbf{H}_i^* \right] \right\} + \]
\[ + \left\{ \varepsilon_0 \varepsilon_\nu \mathbf{E}_s \cdot \mathbf{E}_s^* + \mu_0 \mathbf{H}_s \cdot \mathbf{H}_i^* - \frac{1}{2} \mathbf{I} \left[ \varepsilon_0 \varepsilon_\nu \mathbf{E}_i \cdot \mathbf{E}_s^* + \mu_0 \mathbf{H}_i \cdot \mathbf{H}_i^* \right] \right\} + \]
\[ + \left\{ \varepsilon_0 \varepsilon_\nu \mathbf{E}_s \cdot \mathbf{E}_s^* + \mu_0 \mathbf{H}_s \cdot \mathbf{H}_s^* - \frac{1}{2} \mathbf{I} \left[ \varepsilon_0 \varepsilon_\nu \mathbf{E}_i \cdot \mathbf{E}_i^* + \mu_0 \mathbf{H}_i \cdot \mathbf{H}_i^* \right] \right\} \]  

(2.5)

In Eq. (2.5) the first term depends only on the incident fields and clearly does not lead to any net force. The second term depends on the scattered fields only and due to the interaction between different multipoles of different order and degree of the particle, can lead to a force contribution. In other words the contribution of this second term to the optical force arises from the “cross-interaction” of different multipoles of scattered field. If the scattered field is described by only one single multipole, say the electric dipole as in the case of Rayleigh scatterers, the resulting force from this second term in the Maxwell stress tensor happens to be zero. The last two terms of Eq. (2.5) involve products of the incident and the scattered fields and describe the cross-interaction between incident and scattered multipoles. Hence the expression for the average optical force is due to the following contributions:  
\[ \langle \mathbf{F} \rangle = \langle \mathbf{F}_{ii} \rangle + \langle \mathbf{F}_{ii} \rangle + \langle \mathbf{F}_{ii} \rangle . \]

We can expand the incident and scattered fields in terms of spherical harmonics [11], as an example for the incident electric and magnetic fields:
\[ \mathbf{E}_i = \sum_{n=1}^{\infty} \sum_{m=0}^{n} \left( a_{enm} \mathbf{N}_{enm} + a_{omn} \mathbf{N}_{omn} + b_{enm} \mathbf{M}_{enm} + b_{omn} \mathbf{M}_{omn} \right) \]  

(2.6)
\( H_j = \frac{\sqrt{\varepsilon_b}}{i \eta_0} \sum_{n=1}^{n} \sum_{m=0}^{m} \left( a_{enm} M_{enm} + a_{emn} M_{emn} + b_{enm} N_{enm} + b_{emn} N_{emn} \right) \) \hspace{1cm} (2.7)

Where \( \varepsilon_b \) stands for the relative permittivity of the background dielectric medium in which the incident field propagates. The quantity \( \eta_0 \) is the vacuum characteristic impedance. Here the coefficients \( a_{nm} \) and \( b_{nm} \) represent the complex amplitudes of the various electric and magnetic multipoles, respectively. Similar expressions can be written for \( E_s \) and \( H_s \). Now replacing the field expressions in eq. (2.5) and after significant simplifications we will reach to the all the forces acting on the scattering particles. One of these equations showing the force contribution due to the interaction between different multipoles are illustrated bellow [22]:

\[
\begin{align*}
\langle F_{ii} \rangle &= \frac{1}{4} \text{Re} \sum_{n1=1}^{n1} \sum_{m1=0}^{m1} \sum_{n2=1}^{n2} \sum_{m2=0}^{m2} \varepsilon_b \varepsilon_b \\
&\quad \left( a_{enm}^* a_{enm2}^* + b_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} N_{enm1}^{(1)} N_{enm2}^{(1)}^* \hat{r} dS - \int_{S} \left( N_{enm1}^{(1)} \cdot N_{enm2}^{(1)}^* \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)}^* \right) \hat{r} dS \right] + \\
&\quad + \left( a_{enm}^* a_{enm2}^* + b_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} M_{enm1}^{(1)} N_{enm2}^{(1)}^* \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot N_{enm2}^{(1)}^* \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)}^* \right) \hat{r} dS \right] + \\
&\quad + \left( b_{enm}^* a_{enm2}^* + a_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} M_{enm1}^{(1)} N_{enm2}^{(1)}^* \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot N_{enm2}^{(1)}^* \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)}^* \right) \hat{r} dS \right] + \\
&\quad + \left( b_{enm}^* a_{enm2}^* + a_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} N_{enm1}^{(1)} N_{enm2}^{(1)}^* \hat{r} dS - \int_{S} \left( N_{enm1}^{(1)} \cdot N_{enm2}^{(1)}^* \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)}^* \right) \hat{r} dS \right] + \\
&\quad \times \\
&\quad + \left( a_{enm}^* a_{enm2}^* + b_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} N_{enm1}^{(1)} M_{enm2}^{(1)} \hat{r} dS - \int_{S} \left( N_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS \right] + \\
&\quad + \left( a_{enm}^* a_{enm2}^* + b_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} N_{enm1}^{(1)} M_{enm2}^{(1)} \hat{r} dS - \int_{S} \left( N_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS \right] + \\
&\quad + \left( b_{enm}^* a_{enm2}^* + a_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} M_{enm1}^{(1)} N_{enm2}^{(1)} \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot N_{enm2}^{(1)} \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS \right] + \\
&\quad + \left( b_{enm}^* a_{enm2}^* + a_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} M_{enm1}^{(1)} N_{enm2}^{(1)} \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot N_{enm2}^{(1)} \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS \right] + \\
&\quad + \left( b_{enm}^* a_{enm2}^* + a_{enm}^* b_{enm2}^* \right) \left[ 2 \int_{S} M_{enm1}^{(1)} N_{enm2}^{(1)} \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot N_{enm2}^{(1)} \right) \hat{r} dS - \int_{S} \left( M_{enm1}^{(1)} \cdot M_{enm2}^{(1)} \right) \hat{r} dS \right]
\end{align*}
\]
Similar expressions can be written for $F_{is}$ and $F_{si}$ [22]. Using these results as a starting point, it is then possible to infer the following set of general rules that apply to force calculations in terms of spherical multipoles. For the force along $x$, the non-vanishing contributions from the interaction are the following terms [22]:

1. Electric multipoles of degree $n$ and order $m$ with electric multipoles of degree $n\pm1$ and the order $m \pm 1$, and the same azimuthal parity.

2. Magnetic multipoles of degree $n$ and order $m$ with magnetic multipoles of degree $n\pm1$ and the same order $m\pm1$, and the same azimuthal parity.

3. Electric multipoles of degree $n$ and order $m$ with magnetic multipoles of the same degree $n$ and the order $m\pm1$, and opposite azimuthal parity.

For the force along $y$ we have nonvanishing contributions from the interaction of the following:

1. Electric multipoles of degree $n$ and order $m$ with electric multipoles of degree $n\pm1$ and the order $m\pm1$, and opposite azimuthal parity.

2. Magnetic multipoles of degree $n$ and order $m$ with magnetic multipoles of degree $n\pm1$ and the same order $m\pm1$, and opposite azimuthal parity.

3. Electric multipoles of degree $n$ and order $m$ with magnetic multipoles of the same degree $n$ and the order $m\pm1$, and the same azimuthal parity.

For the force along $z$ we have nonvanishing contributions from the interaction of the following:
1. Electric multipoles of degree $n$ and order $m$ with electric multipoles of degree $n \pm 1$ and the same order $m$, and the same azimuthal parity.

2. Magnetic multipoles of degree $n$ and order $m$ with magnetic multipoles of degree $n \pm 1$ and the same order $m$, and the same azimuthal parity.

3. Electric multipoles of degree $n$ and order $m$ with magnetic multipoles of the same degree $n$ and the same order $m$, and opposite azimuthal parity.

2.2. Rayleigh Regime

For very small particles (much smaller than the wavelength) they would be in the Rayleigh regime. We can assume without any loss of generality that the particle is located at the origin of our coordinate system. In this case the induced electric dipole moment is simply given by the product of the polarizability and the incident electric field at the particle location:

$$ \mathbf{p} = \alpha \varepsilon \mathbf{E} \bigg|_{r=0} $$

(2.9)

The resulting scattered fields from this dipole are then given by [15]:

$$ \mathbf{E}_s = \frac{1}{4\pi\varepsilon_0} \left\{ k_0^2 \left( \hat{r} \times \mathbf{p} \right) \times \hat{r} \frac{e^{ik_0r}}{r} + \left[ 3\hat{r} (\hat{r} \cdot \mathbf{p}) - \mathbf{p} \right] \left( \frac{1}{r^3} - \frac{ik_0}{r^2} \right) e^{ik_0r} \right\} $$

$$ \mathbf{H}_s = \frac{c k_0^2}{4\pi} \left( \hat{r} \times \mathbf{p} \right) \frac{e^{ik_0r}}{r} \left( 1 - \frac{1}{ik_0r} \right) $$

(2.10)
The scattered field can be described in terms of vector spherical Harmonics through the superposition electric dipole terms:

\[ E_s = a_e^{s0} N_e^{(1)} + a_{o1}^{s} N_{o1}^{(1)} + a_e^{s1} N_{e1}^{(1)} \]

\[ H_s = \frac{1}{i\eta_0} \left[ a_e^{s0} M_e^{(1)} + a_{o1}^{s} M_{o1}^{(1)} + a_e^{s1} M_{e1}^{(1)} \right] \]

(2.11)

It is straightforward to show that the spherical harmonics \( N_e^{s} \), \( N_{o1}^{s} \), \( N_{e10}^{s} \) are associated with the electric field scattered respectively by the Cartesian components \( p_x \), \( p_y \), \( p_z \) of the induced electric dipole moment \( p \). Imposing the equivalence between equations (2.10) and (2.11) one can relate the expansion coefficients with the particle polarizability in the following way:

\[ p = \alpha_E E_i |_{x=0} = -\frac{i4\pi \varepsilon_0}{k_0^3} \left[ \hat{x} a_{e11}^{s} + \hat{y} a_{o11}^{s} - \hat{z} a_{e10}^{s} \right] \]

(2.12)

The incident field in its most general form is given by the expression (2.6). When evaluated at the origin it reduces to: \( E_i |_{x=0} = \frac{2}{3} \left( \hat{x} a_{e11}^{s} + \hat{y} a_{o11}^{s} - \hat{z} a_{e10}^{s} \right) \), deducing from equation (2.12) the following expression that relates the scattered field and the incident field coefficients will be achieved as:

\[ a_{e11}^{s} = -\frac{i k_0^3 \alpha_E}{6\pi \varepsilon_0} a_{e11}, \quad a_{o11}^{s} = -\frac{i k_0^3 \alpha_E}{6\pi \varepsilon_0} a_{o11}, \quad a_{e10}^{s} = -\frac{i k_0^3 \alpha_E}{6\pi \varepsilon_0} a_{e10}. \]

At this point it is straightforward to apply the interaction rules given in the previous section in order to calculate the Cartesian components of the optical force of a Rayleigh particle under arbitrary field illumination. Since the scattered field is constituted only by electric dipole terms \((a_{(e/o)lm}^{s})\), the component \( \langle F_{ns} \rangle \) vanishes and the total force is given by \( \langle F_{ns} \rangle + \langle F_{ss} \rangle \). The only
components of the incident field that could lead to finite contributions to the total optical force (if
the scattering is electric dipolar) are the magnetic dipole terms \( b_{(e/o)lm} \) and the electric
quadrupole terms \( a_{(e/o)2m} \).

After some lengthy but straightforward manipulation, it is possible to show the well-known form
for optical forces on a dipolar scatterer is [16,22]:

\[
\langle F \rangle = \frac{1}{4} \alpha_i \nabla |E_i|_r^2 + \frac{k_0 \alpha_i}{\varepsilon_0} \left[ \frac{\text{Re} \left( E_i \times H_i^* \right)}{2c} + \frac{\varepsilon_0}{4k_0i} \nabla \times \left( E_i \times E_i^* \right) \right]_{r=0}
\]  

(2.13)

Notice that even though the scatterer is dipolar, the optical force itself is produced by the
interaction of higher-order multipoles of the incident field (electric quadrupole and magnetic
dipole) that do not directly participate in the scattering process.
CHAPTER THREE: OPTICAL MANIPULATION

It has been more than 20 years since Arthur Ashkin trapped micron-sized particles in both water and air, based on optical forces [23]. At the beginning, in order to create a stable 3D trap, he used two counter-propagating laser beams, which later was replaced with a tightly focused single beam optical trap [4], what is known today as “optical tweezers”. In the following years, optical tweezers became very popular for studies in biology [24-26] and nonlinear optical phenomena [27,28]. This device has been used to study an increasing number of biochemical and biophysical processes, from the basic mechanical properties of bio-particles to investigations on intracellular processes. The base of this technique has to do with the type of forces that were discussed in the previous section. Here we will discuss the physical principles behind an optical trap, its setup and how this device can be combined with different spectroscopy techniques to further extract information from trapped particles.

3.1. Theory of Optical Trapping

Optical tweezers (OT) work on the principle of pico-newton optical forces of highly-focused laser beams to trap and manipulate particles in the range of 10nm to 100um. This capability depends on the refractive index mismatch of the particle with respect to its media, the intensity of the laser and how tightly it can be focused. The main forces on a particle in the trap are the optical scattering and the gradient forces [29-31] (produced by the gradient of field intensity), as demonstrated in figure 4.
Scattering and gradient forces depend on the wavelength of the laser, and the particle size. In general, particles trapped with an OT can be divided into two main regimes: Mie regime [25,26] and Rayleigh regime [30], both discussed below.

In Mie regime, the particle is much larger than the wavelength of the trapping laser. In order to describe the forces on the particle in this regime, geometrical optics can be used, as demonstrated in figure 4. The principle of trapping is the conservation of the beam momentum while it passes through the particle [4,32]. Refraction leads to a light momentum change and consequently, depending on the values of np (refractive index of particle) and nm (refractive index of medium), the particle will either feel attracted towards or pushed away from the maximum intensity [33]. If np > nm, the particle will be attracted to the focus of the laser beam (maximum intensity), and the opposite will occur for np<nm (pushed to lower intensity region). For particles with high
reflection coefficient at the wavelength of incident, the same repulsive radiation force can be expected [34]. In both cases the force has two components; the axial and the lateral.

The spatial distribution of both intensity and radiation force potential of the focused laser are schematically shown in figure (5. b) shows the potential well produced by the focused laser beam when \( n_p > n_m \).

![Figure 5](image.png)

Figure 5 Schematic representation of the spatial distribution of a focused laser beam intensity (a) and the relevant potential of the radiation force (b-c).

For a particle with \( n_p < n_m \), there is no potential well and the particle is pushed out by the repulsive radiation force (figure 5.c).

In a Rayleigh regime, the particle is much smaller than the wavelength and thought to be an induced dipole that elastically scatters light. For particles which are substantially smaller than the wavelength of the beam, the direction of the force is independent of the particle shape (unlike the Mie approach) and its magnitude varies with the particle orientation. The electromagnetic field pulls the dipole particle towards the highest intensity of the beam. In this case, the scattering force is proportional to the optical intensity and points towards the propagation of the laser light.

The gradient force, also known as the Lorenz force, can be simply shown as:
\[ F_{\text{gradient}} = \pi \varepsilon_0 \alpha^3 n_m^2 \left( \frac{m^2 - 1}{m^2 + 2} \right) V E^2 \] (3.1)

While the scattering force can be expressed as:

\[ F_{\text{Scat}} = \frac{I_0}{c} 128 \pi^3 a^2 n_m \left( \frac{a}{\lambda_0} \right)^4 \left( \frac{m^2 - 1}{m^2 + 2} \right)^2 \] (3.2)

In these equations, \( r \) is the radius of the particle, \( \lambda \) is the wavelength of incident, \( m \) is the ratio of \( n_p \) and \( n_m \), \( I_0 \) is the intensity of the field, and \( c \) is the speed of light in a vacuum. The optical trap becomes unstable for very large particles because when \( r \) increases, the scattering force increases more than the gradient force. By increasing the intensity both fields will increase. However, in order for an optical trap to be feasible, the important factor is the gradient of the field, which can only be increased when the beam is tightly focused.

### 3.1.1. Optical Tweezers Stiffness Measurement

A trapped particle of mass \( m \) in an aqueous medium with damping \( \gamma_0 \) (at temperature \( T \)) can be considered as an example of a harmonic oscillator. The equation of motion of this particle can be described for Brownian motion [29] as:

\[ m \frac{\partial^2 x}{\partial t^2} + \gamma_0 \frac{\partial x}{\partial t} + kx = (2k_B T \gamma_0)^{1/2} \eta(t) \] (3.3)

Where \( k \) is the trap stiffness (linearly increases with laser power) and \( k_B \) is Boltzmann’s constant. The term on the right side of equation 3.3 accounts for all the Brownian forces on the trapped particles at temperature \( T \), with \( \eta(t) \) representing the random motion of the object as a function
of time. Due to the aqueous medium, the object is highly damped and thus the inertial and gravitational forces can be neglected [35].

In general two common methods are used to calculate the optical trap stiffness: equipartition theorem and the power spectrum [36].

![Methods for measuring trap stiffness](image)

Figure 6 Methods for measuring trap stiffness: (a) Position monitoring of the trapped particle (equipartition theorem), (b) A Fourier transform of the particle position yields a Lorentzian-shaped power density spectra [35].

1. Equipartition theorem is the simplest method for determining stiffness of the trap ($k_{\text{trap}}$). The equipartition theorem is mathematically expressed as:

$$\frac{1}{2} k \langle x^2 \rangle = \frac{1}{2} k_B T$$  \hspace{1cm} (3.4)

By monitoring the trapped particle movement (figure 6.a), using a quadrant photodiode and finding the mean-squared deviation in position ($\langle x^2 \rangle$) along one axis, one can calculate the $k_{\text{trap}}$.

2. The Power spectrum method is a frequency-domain analysis based on the Fourier transform of the trapped particle position, which yields a Lorentzian-shaped power spectrum [36], demonstrated in figure 6.b, as:
\[ S_{xx}(f) = \frac{k_B T}{\pi^2 \beta (f_0^2 + f^2)} \]  

(3.5)

Where \( S_{xx}(f) \) is the power spectral density (PSD) in units of \((\text{nm}^2/\text{Hz})\), \( \beta \) is the Stokes drag on a spherical trapped particle of radius \( r \) (where \( \beta = 6\pi \eta r \)) and medium viscosity \( \eta \). \( f_0 \) is the roll-off frequency of the optical trap and for a sample in aqueous medium is well below 1 kHz, and \( f \) represents the frequency. \( f_0 \) can be found by fitting the PSD curve. The trap stiffness can then be found knowing:

\[ k_{\text{trap}} = 2\pi \beta f_0 \]  

(3.6)

For a 1\( \mu \)m sphere in water \( \beta = 10^{-8} \text{ Ns/m} \) and for typical biological applications, the stiffness has been calculated to be about 0.05pN/nm [35].

3.2. Optical Tweezers Setup

Figure 7 shows a version of a single beam OT setup. The laser source used for tweezing can be any type of CW laser with an output of a few mW. A beam that deviates significantly from a Gaussian profile, when focused will lead to a less efficient trap, so the best trap is created with the TEM\(_{00}\) mode. If the laser width is small, with a telescope system, the laser beam is expanded to match the back aperture of the microscope objective. A high-NA microscope objective (usually 100X) is used to create a tight focus. This objective is also used to image the trapped particles, on a CCD. With the use of a dichroic mirror (DCM) in the setup, we can separate the optical paths of trapping and imaging.
Conventional optical tweezers use a standard “single Gaussian laser beam”. But in the past few years researchers have used different types of beams and setups for trapping particles. Examples are; non-Gaussian beams like Bessel beams [37-41], two-beam interferometric optical tweezers [42-44] and creating multiple traps using three different methods [45-54] in order to extend the use of this device in particle manipulation and characterization. The main three different techniques used are: beam splitting and recombination techniques, scanning techniques and holographic techniques.

In the beam combination technique, a laser is usually split into two separate beams and later combined, so that two independent traps are formed and their position can be controlled (figure 8). This type of system is mainly used for force measurement research [29,35] and interaction of particles.
Figure 8 Schematic of the setup for a dual trap optical tweezers which is based on beam splitting [45].

In the scanning technique (figure 9.a) an acousto-optic deflector (AOD) is used to create a trap at different positions at different times, by time-sharing the light between trapping sites. So as long as the beam returns to the particle before it can diffuse away, the particle will remain trapped.

Figure 9 Schematic diagram of the setup used to create arrays of optical traps with the use of AODs for beam steering (a). Images of trapped 1.4 mm FITC–SiO₂ particles in a time-shared 2D array of 25 traps with square symmetry [46].
In the third method a 3D control of multiple trap sites is possible with the use of holograms which control the phase of the light beam. By reflecting the laser beam off the hologram (figure 10), the desired pattern of multiple traps is formed. This combination is referred to as holographic optical tweezers (HOT) [53].

Figure 10 Optical tweezers setup using spatial light modulator (SPM) (a). Detailed view of the spatial phase masks arrangement (b). Two examples of holograms used on SPM to produce: one (c) and four (d) trapping points respectively at the sample position. The resulting four traps at sample location, due to (d) is illustrated in (e) [53].

3.2.1. Laser Tweezers Applications in Biology

Since the invention of the optical tweezers, they have emerged as a powerful tool with a broad application in biology. Their capabilities have evolved from simple trapping and manipulation of cells and polymers to force/displacement measurement in the pico-newton and nanometer regime in order to understand the mechanical properties of bio-material [36,54]. Examples of
such studies are the stretching of the DNA molecule [55] and the mechanisms of molecular motors (made of proteins) in using chemical energy to perform mechanical tasks [56]. As demonstrated in figure 11, in these studies the molecules are attached to a suitable micron-sized dielectric bead, so instead of exerting the beam on the molecule, the dielectric bead is trapped and manipulated. This will prevent the sample from damage and better traps the macromolecules.

![Diagram](image)

Figure 11 Measuring DNA mechanical properties with an OT, by attaching a bead to one end of the DNA, while the other end is connected to the cover slip (a). Mechanical analogy of (a) showing the compliance of $k_{\text{DNA}}$ and the trap $k_{\text{trap}}$ (b). Interaction and binding of a single myosin (a motor protein in the muscle that creates external forces) with actin filament, causing muscle to shorten and the beads to move [55,35].

With the use of two counter-propagating diverging beams and based on the optical forces, cells and polymers have been trapped and their mechanical properties have been studied using the surface force (hundreds of pN [57]) that stretches the particle. This technique is known as optical
stretching and has been used to measure the elasticity of biological cells [57-59]. Based on these properties, diseased cells and cancer cells can be identified [54,60]. More recently scientists are using OT for cell sorting [61], as scissors or scalpels (to cut with submicron resolution) [62,63] and to build 3D biological structures with the use of multiple traps [64].

One great advantage of the OT system is that it can be integrated and combined with different spectroscopy [65,66], and spectroscopy based imaging systems [53,67-69]. This can simply be done by adding an optical path (using the right DCM and filter) to the OT setup to collect and transfer the signal to the spectrometer (and detector), as demonstrated in figure 12. In this case either the trapping beam can be the excitation source (for Raman or fluorescence) or a second laser beam is used.

![Figure 12 Typical setup of Raman Optical Tweezers. L: lens, M: mirror, DM: dichroic mirror, GM: galvo-mirror, NF: notch filter that only reflects Raman signal [70].](image)
The problem of optically trapped Raman imaging of cells is due to the fact that all living cells have to be kept in a suspension and consequently there is the Brownian motion of the cells, plus the inherent biological activity that continuously redistributes the organelles inside the cell. While the trapping laser captures the cell for hours, the cell can move or rotate in the trap [71] creating a very inaccurate map. In order to solve this issue and immobilizes the cell almost completely, instead of one trapping beam, multiple trapping beams can be used around the periphery of the cell. This will allow immobilization of even the larger or asymmetric cells and distributes the trapping power more evenly throughout the cell, minimizing the damage.

Figure 13 a) Single beam, traps the part of the cell with the highest refractive index, so the trapped cell can have different orientations inside the trap. b) Using two beams to trap the cell and stabilize it. c) Histograms of trapped cell movement recorded on a quadrant photo-detector (QPD), obtained with a single and double beam trap [72].

Figure (13.a) demonstrates a trapped cell with complex components having different optical properties. This will lead to different orientations of the cell inside the single trap. As shown in figure (13.b) by using two trapping beams, this undesired movement will reduce. To compare
how much adding a second beam, to trap the sample tighter and reduce the movements, the sample’s Brownian motion was recorded on a QPD for both cases of one and two traps. The result is shown in figure (13.c). Adding more beams for trapping can immobilize the cell to a great extent.

As discussed earlier, there are several ways to create multiple trapping beams that can be controlled separately. HOT and time-sharing tweezers have been successful in immobilizing larger biological samples with multiple traps and can rapidly scan the particle to create a Raman image of them: floating Jurkat cells (leukemia) 10-15 μm in diameter and RBC [53,69] 10-15 μm in diameter, are examples of two types of cells that has been imaged with this technique and studied.

Figure 14 Raman images taken by scanning the cell through the Raman probe, using four beams for trapping and scanning of: (a) Jurkat cell showing the distribution of proteins and lipids [53] (b) Healthy and thalassemic RBC showing the distribution of hemoglobin [69]. Each step, the cell was moved 3 μm for (a) and 0.5 μm (b) with 50ms integration time per pixel.
As demonstrated in figure (14.a) two separate Raman images of the Jurkat cell are achieved by plotting the intensity of the Raman bands assigned to two chemicals; proteins with 1005 cm\(^{-1}\) shift and lipids with 1300 cm\(^{-1}\) shift [73]. Though the resolution is not high (due to step size of 3µm) the different distributions of lipids and proteins inside the cell can be seen as expected. In figure (14.b) the distribution of hemoglobin (Hb) inside healthy and thalassemic RBCs have been imaged via Raman emission at 1547 cm\(^{-1}\). The Raman image for a normal RBC, presents the classical ring-shape distribution for Hb with no Hb in the center, while this distribution is modified in diseased cells and Hb can also be found in the center, as seen in figure 3.11.b for the thalassemic RBC.

In 2003, Alexander et al. demonstrated the first SERS Optical Tweezers [66]. In their experiment, they used a special SERS substrate (with 60nm gold nanoparticles attached) to measure the SERS of single trapped Bacterial Spores. They observed a 100-fold signal enhancement compared to normal Raman measurement, as demonstrated in figure 15.

![Figure 15 Schematic diagram for SERS measurement of bacterial spores (a). Comparison of normal Raman and SERS spectra of single Bacillus stearothermophilus spore (b) [66].](image-url)
Optical trapping is a powerful technique spanning numerous fields and operating in a large range from the single molecule to a large cell. By using holographic and time shared optical traps, the application of OT can be expanded, and the resolution of both particle manipulation and spectroscopy results will be much higher. More recently, due to the desire for high-resolution microscopy other spectroscopy techniques, like CARS has been added to the OT [74,75]. Optical tweezers application in Biology is growing fast and one of the main interests is finding ways to extract more biochemical information on a single cell level.

3.3. Optical Tweezers in Combination with Spectroscopy Techniques (Experimental Results)

A principal issue in laser spectroscopy of biological samples is the ability to keep them spatially confined over the duration of signal acquisition. Cellular samples are in constant motion due to inherent Brownian motion and suspension in aqueous solution. Since most spectroscopic techniques require integration times on the order of seconds, being able to keep the cell still without damage is crucial. Optical trapping has proven to be very useful in this effort [76]. By using a tightly focused laser beam, cells can not only be prevented from drifting in their surrounding medium, but they can even be manipulated and sorted [77]. Although biological cells are typically much larger than micrometers and composed mostly of water, various organelles within the cells are small enough to be trapped, thereby allowing the entire cell to be stabilized. It has been shown that eukaryotic cells are trapped by their densest structure, the nucleus [78], however it is possible to trap other organelles as well, such as mitochondria, which range in size from 0.5µm to 5µm [79]. Once the cell has been stabilized in the optical trap, various diagnostics can be performed with a higher signal-to-noise ratio (SNR). One of these diagnostic techniques is Raman spectroscopy, where it’s used commonly in situ biological
applications [80]. Raman spectroscopy is compatible with the optical tweezers systems because both techniques require an intense focused beam for optimal performance [81]. While the same trapping beam can be used to excite the vibrational levels measured in Raman spectroscopy, in some cases a secondary excitation beam is used, allowing for a stronger signal.

In general, acquiring a Raman signal from biological samples is not a trivial task for two main reasons: 1. Since these samples are in an aqueous solution they move all the time, and 2. Raman signal is inherently weak compared to the Rayleigh scattered signal, thus necessitating long acquisition times to accurately see the spectrum. The need for long acquisition times is obviated by stabilizing the suspended particles with optical tweezers [75].

Here we combine Fluorescence and Raman spectroscopy with an inverted optical tweezers system to study biological samples on a single cell level. We use a CW, 532nm laser source as both the trapping and excitation beam. It is possible to trap a cell while acquiring the Raman signature as well as fluorescence emission spectrum. Our experimental setup Figure 16 combines an inverted optical tweezers setup with an imaging system and with Raman and fluorescence signal acquisition elements. After expending the trapping beam more than 3 times via a telescope system, a dichroic mirror is used to selectively pass or reflect certain wavelengths, both for imaging and spectroscopy. Note that by forming a 4f system in a way that the steering mirror (BSM) and the back aperture plane (of the focusing microscope objective) become conjugate planes, the trap position in the sample plane could be easily moved without degrading trap performance. This conjugation is achieved by placing the BSM at the focal point of L1 and the
microscope objective (MO) at the focal point of L2. This MO is a 100x with a high numerical aperture (NA=1.25) in order to tightly focus the trapping beam.

The first sample used to calibrate the trap was 1 to 3μm polystyrene beads. Later we used 2μm dyed fluorescent polystyrene beads, and live yeast cells stained with Rhodamine B, hexyl-ester (suspended in a 10mM Hepes buffer with 5% glucose) were placed on 150μm glass coverslips and situated on a precise XYZ translation stage to provide 3-dimensional range of motion. Immersion oil was used to match the refractive index between the microscope objective and the glass coverslip for maximum focusing efficiency. A 10x microscope objective was placed above the sample, condensing a white light source that provided illumination for the CCD (Thorlabs USB 2.0 Digital Camera DCU224C-BG), which was placed on the back-reflected side of the dichroic mirror. Images were stored as 1280x1024 pixel JPEG files with a resolution of

Figure 16 Experimental setup for multispectral optical tweezers system [82].
98nm/pixel. A flipping mirror was used to allow the white light and backscattered light from the sample to be directed either to the spectrometer or CCD. The signal (either Raman or fluorescence emission) was collected after a 532nm long-pass edge filter (Semrock LP03-532RS-25), and focused into a fiber leading to an Ocean Optics HR4000 Spectrometer. Figure 17 demonstrates the motion of a 1μm polystyrene bead as it is translated sequentially from point (a) to point (e) by changing the angle of reflection of the BSM.

Figure 17 Moving the trapped 1μm polystyrene bead from point (a) to point (e) by varying the rotation of the beam steering mirror. Stable trap control was demonstrated over a displacement of 45μm [82].

Using a 532nm excitation source and a silicon wafer, calibration of the multispectral optical tweezers system in the Raman mode was performed to confirm the strong known Raman signal from silicon at 522cm⁻¹. Since the Raman spectrum of a trapped object is known to differ from that of the same object untrapped [10], 3μm polystyrene beads were used to ensure trap stability as well as Raman spectrum accuracy. As expected, the Raman spectrum Figure 18 shows a strong shift at 1001cm⁻¹ due to breathing of the phenyl group in the polystyrene beads, confirming proper calibration of simultaneously tweezing and performing Raman spectroscopy.
3.3.1. Raman Spectrum of Single Yeast Cell

Once the Raman optical tweezers system was calibrated, the 532nm source was used as both the trapping and the excitation source for live yeast cells suspended in water. The Raman spectrum was recorded using an integration time of 8 seconds, and 5 acquisitions were averaged to reduce the effects of background noise on the inherently weak Raman spectrum. The CCD captured live images of the trapped yeast cell. The combined image and Raman spectrum can be seen in Figure 19. The distinct peak at 1101cm⁻¹ is characteristic of a symmetric di-oxy stretch of the phosphate group in the yeast’s RNA [83].
A 5mW, 532 nm laser diode was used as both the excitation and trapping source, and the Raman spectrum shown is an average of 5 acquisitions of 8s integration time. A strong peak at 1101 cm\(^{-1}\) is characteristic of a symmetric di-oxy stretch in the phosphate group of the yeast’s RNA.

### 3.3.2 Fluorescence Imaging of Yeast Mitochondria

First, due to the ease of trapping polystyrene beads, we tried fluorescence imaging using dyed 2μm polystyrene beads (with \(\lambda_{ex} \sim 575\) nm and \(\lambda_{em} \sim 610\) nm) to make sure the system works well. Exciting with the same 532nm source, the fluorescence signal of the beads was acquired using the spectrometer. Figure 20 shows the resulting emission spectrum of the beads and validates the correct performance of our optical system.
Figure 20 Fluorescence emission spectrum and live image of dyed 2μm polystyrene beads in the optical trap, excited by the trapping beam at 532nm. Similar to the Raman results in Figure 3, the sharp onset of signal around 540nm is due to an edge filter placed before the spectrometer and CCD.

We used the same optical system to perform fluorescence imaging of yeast cells. Once the cells were dyed, the 532nm source was used as both the trapping and excitation beam, and the fluorescence image was recorded using the CCD. As shown in the enhanced image in Figure 21, fluorescence imaging shows three distinct bright spots, corresponding to three mitochondria in a yeast cell.

Figure 21 Fluorescence imaging of a trapped stained yeast cell, excited at 532nm. The image has been enhanced to show three areas of higher intensity from the mitochondria in the yeast cell [82].
In summary, the ability to combine multiple diagnostic methods into a single optical setup is highly desired for rapid evaluation of samples. Here we demonstrated a simple version of such multispectral optical tweezers system, allowing for various diagnostics of single biological cells. The achieved fluorescence and Raman signal from a single yeast cell trapped at the focus, confirms the sensitivity of our combined optical tweezers system in detecting biochemical information about bio-samples up to a single cell level. Most importantly, we were able to obtain strong signals at low power due to stabilization provided by the optical trap, allowing for nondestructive diagnostics of single cells. Combining more spectroscopic diagnostics and more resolved imaging techniques into the multispectral optical tweezers will allow for even further characterization of a given sample.

3.4. Moire-based Optical Propelling Traps

So far we demonstrated the principles of optical tweezers and how they can be used to trap particles. In addition to linear translations in three dimensions, rotation of trapped particles offers another important degree of freedom for optical manipulation. Over the years, a variety of techniques have been proposed for rotating trapped particles, including the alteration of optical angular momentum [84,85] the holographic optical tweezers (HOT) [50,86] specially designed electric or magneto-optic manipulators [87,88] or the more popular, rotating, non-symmetric trapping beams (as obtained with higher-order transverse laser modes, cylindrical lenses, rectangular apertures, revolving interference patterns, and so on) [89–96]. Most of these techniques rely on either mechanical rotation of beam-shaping components or phase-sensitive
interference, thus deterring stabilized control of the tweezers against ambient vibrations or perturbations.

Recently, a novel approach to generate rotating intensity blades (optical propelling beams) was proposed, using the moiré technique [97]. These optical propelling beams can be generated with controllable rotation speed and direction without the need for any mechanical movement or phase-sensitive interference. Here we demonstrate how these optical propelling beams can be utilized to trap particles with the advantage of dynamic control of their rotation. With intelligent beam design, we obtain a well-resolved single/multi-blade intensity structure, even when the whole beam is tightly focused, and demonstrate stable two-dimensional trapping and controlled rotation of polystyrene micro-beads and Escherichia coli bacteria. We show that in principle our technique for rotating tweezers can be implemented with incoherent light sources, as multi-blade white-light propelling beams are readily demonstrated in our experiment. These computer-controlled, rotating multi-traps that do not require time-share may open new avenues for optical trapping and manipulation.

3.4.1. Generation of Optical Propelling Beams Experimental Setup

These multi-blade intensity patterns are created by overlapping a moving straight-line grating with a fork-type grating [97]. The dynamic phase mask used is the same as the pattern resulting from the interference of a plane wave with a vortex beam. The number of intensity blades is determined by the topological charge of the vortex, while the direction of beam rotation depends on the direction of the grating movement and the rotation speed is proportional to the speed of the grating motion. The important fact in generating the rotating multi-blade intensity patterns
via moiré-based rotating optical tweezers is to design the dynamic mask in a way that each blade acts as an optical gradient trap well separated from the other blades.

The main advantage of this technique is that the number of blades, their angular velocity and direction of rotation can be changed very simply by a computer-controlled spatial light modulator (SLM). More importantly, because no phase-sensitive interference is involved, the resulting pattern is remarkably stable during rotation regardless of environmental vibrations.

![Experimental setup for moiré-based rotating optical tweezers. SLM: spatial light modulator; BS: beam splitter; L: lens; O: objective lens; WLS: white light source; CCD: charge-coupled device [98].](image)

The experimental setup used is sketched in Fig. 22. A collimated Gaussian beam, with a wavelength of 532 nm is reflected from a SLM with a very small angle (less than 10 degrees). With appropriate spatial filtering, the designed moire patterns can be retrieved. By simply setting the straight-line grating into linear motion, the propelling beams are generated, and then sent into a setting typically used for optical tweezers. To form a multi-trap rotating tweezers with our propelling beams, we use an objective lens (60X, NA = 0.85) to match the size of individual traps with the typical size of the particles used in our experiments. The power of the trapping
beam before the sample is about 20 mW. Using this setup, first 2um polystyrene beads were trapped. Later the sample used contained E. coli cells (with an average length of 2um) suspended in aqueous solution and sandwiched between two thin cover slides (150um thin). The sample is illuminated with a white-light source from the opposite direction and imaged on a CCD camera. Examples of masks used for the SLM, to achieve this rotation are illustrated in figure 23. To achieve dynamic masks 2 different gratings have to overlap and move in the opposite direction with a certain speed. For instance to generate the 3-blade rotating beam shown in figure (24.a) a straight-line grating (24.b) and a fork-type vortex grating (3.21c) of topological charge $m = 3$, were used.

![Figure 23](image)

Figure 23 From left to right: the Moiré pattern used on the SLM for generating 2, 3 and 4 blade rotating beams

Another important matter is to separate the focal plane of the Gaussian beam and vortex. If they both share the same focal point, at tightly focused conditions essential for trapping, the 3 blades will no longer be distinguished from one another due to the presence of the Gaussian focus. This is demonstrated in figure (24.d) for point 2.
Figure 24 (a) Moiré pattern used for generating 3-blade rotating beams by overlapping (b) a straight-line grating and (c) a fork-type vortex grating of topological charge $m = 3$. (d) Illustration of beam propagation and focusing of the Gaussian (solid) and vortex (dashed) components. (1-3) experimental transverse intensity patterns taken at different longitudinal positions marked in (d) [98].

To overcome this problem, a quadratic phase is added to the vortex grating at the position of the SLM, resulting in a spiral-type moiré fringe as shown in Figure (25.a). This time, by sending a Gaussian beam to reflect off the SLM, the output vortex component has a different wave-front convergence as compared to that of the Gaussian component, leading to different focal points demonstrated in figure (25.b,c). In Image 25: (a) shows the new moiré pattern used and (b) illustrates the propagation and focusing of the Gaussian beam (solid) and the vortex beam (dashed). The 3-blade pattern at the focal point of the vortex beam can now be clearly distinguished and 3 traps are formed (25.c.2). Similarly, other numbers of blades can be readily reconfigured by setting the curved vortex grating with different numbers of singularities.
Figure 25 (a) Moiré mask used for generating 3-blade rotating beams with a added quadratic phase. (b) Illustration of beam focusing for the vortex (dashed) and Gaussian (solid) beams at different spots marked with 2 and 4 respectively. (c,1-5) Experimentally retrieved moiré patterns at different longitudinal positions Marked in (b) [98].

3.4.2. Experimental Results of Trapping and Rotation

To demonstrate optical trapping and manipulation with the propelling beams generated above, we employ an objective lens to tightly focus the beams down to several microns as in a typical setting for optical tweezers. To prove the feasibility of our rotating traps, a sample of 2-μm polystyrene beads suspended in water is used. The motion of beads due to the rotating traps is monitored with the use of white light source focused on the sample plane, and collected from the other side by the same microscope objective that created the traps, towards the CCD camera.
Figure 26 Top panel: Snap shots of trapping and rotation of 2-μm polystyrene beads driven by the rotating tweezers. Bottom panel: snapshots of the E. coli cell in different orientations during rotation. The bacterium is about 2 μm in length and 1 μm in width. The white dashed line serves as a visual guide for the orientation of the bacterium, and the white arrow shows the direction of rotation [98].

As expected, a rotating beam with 3 blades can trap and rotate the same number of particles by the optical gradient force, as shown in Fig. (26.top row). By reversing the rotation of the propelling beam (achieved by reversing either the sign of vortex topological charge or the translation direction of straight-line grating), the rotation direction can be reversed. The polystyrene beads follow the propelling beam at the same frequency (about 0.1Hz). We emphasize that, since no interference or mechanical motion is involved in this technique, the dynamic multi-trap established here is very stable, not susceptible to ambient perturbations. Next, we demonstrate stable trapping and rotation of biological specimens using the above setup. One of the samples used was live E. coli bacteria suspended in de-ionized water. The experimental results are shown in Fig. (26. bottom row) where several snapshots of a rotating bacterium are displayed in different panels. Similar to the case for polystyrene beads, the trapped bacterium can rotate either in a clockwise or counter-clockwise with a controlled speed which is
the same as that of the input rotating beam, with a period of roughly 40 seconds for a full 360° turn. Since the *E. coli* cell is somewhat rod-like, its shape does not appear the same in every picture as the bacterium moves in and out of the imaging plane. Trapping and rotating other bacteria of different shapes with our dynamic tweezers is currently underway, and issues such as the optimal beam configurations (2-blade, 3-blade, etc.) for trapping a specific bacterium or a particular cell shape will be studied. These studies may lead to biological applications for our moiré-based rotating tweezers, such as sorting and separation of bacterial cells.
CHAPTER FOUR: SELF–TRAPPED BEAMS IN ENGINEERED NONLINEAR SOFT- MATTER SYSTEMS

In general, optical beams tend to spatially diffract as they go through a medium. Optical spatial solitons are beams of light that propagate in some type of nonlinear media where the nonlinearity balances diffraction leading to a constant beam cross section during propagation [99]. In order to balance optical diffraction, the beam-media interaction has to induce a self-focusing nonlinear effect that is adequate to cancel diffraction but not too large to create strong self-lensing that leads to catastrophic collapse and breakup of the beam. We can also think of solitons as beams that induce a waveguide and guide itself in the nonlinear medium by locally modifying its refractive index. A side-view image shown in figure 27 represents both beam diffraction at the linear regime and a self-trapped beam at the nonlinear regime in the same medium [99].

There are various types of optical spatial solitons [100]. Askaryan was the first to propose the idea of optical beam wave-guiding in nonlinear media at 1962 [101]. The first experimental demonstration of optical beam self-focusing was studied in materials with Kerr nonlinearities [102]. Kerr nonlinearities are defined by local, instantaneous refractive index change of the media where the index change is proportional to the beam intensity ($\Delta n=n_2 I$). All media show optical Kerr nonlinearities but it is a weak effect since it occurs at frequencies far from resonance. Typical values measured for $\Delta n$ are in the range of $10^{-4}$ or smaller [99]. In 1985, the first Kerr-type 1D spatial soliton was observed in a slab waveguide filled with liquid CS$_2$ by Barthelemy et al [103]. After that, 1D solitons have been demonstrated in a variety of materials displaying Kerr-type nonlinearity, including glass, semiconductors and polymers [99,104]. However bright Kerr-type solitons are only stable in planar 1D systems and the 2D solitons
undergo catastrophic self-focusing [105]. Also 1D soliton-like beams in a bulk media are very unstable and due to instabilities they tend to break apart [99,106].

Figure 27 Left panel: Schematic showing the spatial beam profiles (solid line) and phase fronts (dashed line) for (A) beam self-focusing, (B) natural beam diffraction, and (C) soliton propagation [99]. (D): Side views of experimental demonstration of an optical spatial soliton propagating through a 5mm long media. Top: Beam diffraction under linear condition, bottom: same beam under nonlinear condition of the media [107]. (E) Propagation of a plane wave in nonlinear Kerr medium is shown here for clear demonstration of the phenomena [107].

In order to achieve stable solitons in bulk medium, the light- matter interaction that changes the material’s optical properties should saturate (saturable nonlinearity). In 1974 the first 2D spatial
soliton was observed in sodium vapor at higher beam powers, where the nonlinearity encountered was a saturable self-focusing type that exists near an electronic resonance in a two-level system [108]. A good example of saturable nonlinear media where solitons are stable both in slab waveguides and bulk media are the photorefractive materials [99]. In 1982 Ashkin demonstrated artificial Kerr-like behavior in dielectric nano-suspensions [2,5], which later was shown that the nonlinearity is more of a super-Kerr type [109] that can lead to catastrophic beam collapse. In these media, the gradient forces acting on particles change particle distribution leading to local refractive index change in the media allowing optical waveguiding [110]. Later, it was shown that depending on the polarizability of the nano particles, this type of artificial nonlinearity could be engineered to behave as a saturable type leading to a stable 1D and 2D solitons [109].

4.1. Engineering Nano-suspensions as Artificial Nonlinear Media

Controlling light transport in soft matter systems could be crucial in many and diverse fields of science and technology. In colloidal media, for example, this can be accomplished through optical radiation forces capable of manipulating particle concentration and molecular kinetics at the mesoscopic level [111-113]. In principle such optically induced processes can be exploited for initiating and regulating chemical reactions, for sorting different species of nanoparticles, and for influencing diffusion and osmotic pressure effects, to mention just a few. Approaches of this sort may have important ramifications in life sciences where for instance separation of cells and viruses is often necessary. So far however, meeting these goals has been challenging. Not surprisingly, what impeded progress in this area has much to do with the underlying physics of light propagation in optically non-uniform media. In random environments, light scattering
dominates thus, greatly diminishing transmission. Given that the polarizability of most stable colloids happens to be positive, then at higher intensities, particles tend to migrate into the high field regions of an optical beam making propagation conditions even worse because of enhanced scattering [110]. At the same time, this situation is further exasperated by optical catastrophic self-focusing collapse initiated by a non-saturable Kerr nonlinearity originating from such positive polarizability arrangements [2,5]. In view of this, the question naturally arises whether one could overcome these obstacles by judiciously crafting the nonlinear response of a colloidal system. If so what are the key physical variables in controlling these effects?

These issues can be best addressed by first considering the relation between optical nonlinearities purely activated by radiation pressure and particle polarizability. In general, a particle displays a positive polarizability (PP) whenever its refractive index exceeds that of the background medium, while in the converse case a negative polarizability (NP) arises. As indicated in several studies [5,110], under the action of optical gradient forces, the PP dielectric particles are attracted towards the center of a beam where intensity is higher whereas their NP counterparts are repelled (as illustrated in Fig. 28.a,b). In view of these dynamics, one can readily see that the refractive index of a colloid will in both cases increase along the path of the beam - always resulting into a self-focusing nonlinearity. Interestingly, for many years, the common belief has been that the PP arrangements exhibit a typical Kerr nonlinearity, in which case the nonlinear index change is expected to vary linearly with intensity. Yet, recent z-scan measurements have revealed that the nonlinearity in PP suspensions is in fact super-critical (rises faster than linear) because of the exponential Boltzmann distribution [109]. This super-critical nature of the nonlinearity explains why an optical beam becomes quickly highly unstable and collapses
catastrophically in experimental settings with PP suspensions. A possible way out of this problem was suggested in a subsequent theoretical study [110] where it was indicated that nonlinearities associated with NP suspensions should instead exhibit a saturable Kerr behavior. If indeed this is so the case, then one could expect stable optical self-trapping, entirely free of the aforementioned complications [101]. Furthermore, by appropriately mixing the PP and NP suspensions (as illustrated in Fig. 28.c), one might achieve tunable polarizabilities in such soft-matter systems that could lead to optimized nonlinear response and enhanced light transmission. Unfortunately however, apart from a specific micro-emulsion system operating at a critical phase-transition temperature [114] to our knowledge, no other NP colloidal suspension has been deliberately synthesized thus far to assess its nonlinear response. In this study, we experimentally demonstrate for the first time a new class of synthetic NP colloidal nanosuspensions, and observe robust propagation and enhanced transmission of an optical beam through such nanosuspensions over several diffraction lengths. Furthermore, we show that the nonlinear response of such a soft-matter system can be tailored at ease by targeting its optical polarizability. Specifically, by appropriately mixing the PP and NP colloidal suspensions, the resulting “mixed” polarizability (MP) can be fine-tuned, leading to controllable nonlinear optical response of the nanosuspensions. Comparing with conventional PP colloidal systems, the effects of self-activated transparency enable an increase in light transmission up to fourfold when the concentration of the NP nanoparticles is gradually increased. Our experimental observations are corroborated with theoretical analysis. These results bring about many possibilities for synthesizing colloidal materials with engineered optical nonlinearities, promising for applications where low-loss penetration through scattering media is desirable.
The key to understand light-particle interactions in colloidal suspension systems is the optical forces. Under dipole approximation, apart from radiation pressure and spin-density pressure, the optical gradient force plays a major role, as encountered in an optical tweezers [4,110]. The gradient force can be expressed as

\[ F = \nabla U = \alpha \nabla I/4 \]  \hspace{1cm} (4.1)

Where \( U \) is the potential energy and \( I \) is the optical intensity. \( \alpha \) is the particle polarizability defined as:

\[ \alpha = 3V \varepsilon_0 n_b^2 (m^2 - 1)/(m^2 + 2) \]  \hspace{1cm} (4.2)

\( V \) is the particle’s volume, \( n_b \) is the refractive index of the surrounding liquid background, and \( m = n_p/n_b \) is the refractive-index ratio between the particles and the background [4,110].

Figure 28 Schematic illustrations (top) and experimentally observed snapshots (bottom) of light-particle interaction in colloidal suspensions. (a,d) Attraction of particles towards laser beam in a suspension of positive polarizability \((n_p>n_b)\); (b,e) Repulsion of particles away from laser beam in a suspension of negative polarizability \((n_p<n_b)\) and (c,f) Hybrid motion of particles in a suspension of mixed polarizability \((n_{p1}>n_b>n_{p2})\).
Clearly, apart from the intensity gradient $\nabla I$, the optical gradient force depends also linearly on the induced polarizability. As such, the PP dielectric particles ($n_p > n_b \Rightarrow \alpha > 0$) are attracted towards the high intensity regime of an optical beam, whereas the NP particles ($n_p < n_b \Rightarrow \alpha < 0$) are pushed away. To visualize these phenomena experimentally, we launch a focused laser beam into three samples of colloidal suspensions, which exhibit the PP, NP, and MP, respectively. Typical experimental results are shown in the bottom panels of Fig. 28. For these experiments, 2μm polystyrene beads ($n_{p1}=1.59$) were suspended in water ($n_b=1.33$) where these particles will demonstrate PP. For this suspension, after the focused laser is switched on, the particles are strongly attracted towards the beam center due to optical forces which dominate Brownian motion (fig. 28.a). This polystyrene-based PP arrangement has previously served as a test bench for studying nonlinear phenomena in colloidal suspensions including nonlinear wave mixing, self-focusing and self-trapping [2,115]. However, as mentioned earlier, an intense optical beam propagating through such a PP colloidal system would be afflicted with enhanced scattering as well as catastrophic self-focusing collapse. To illustrate the fundamental difference of light-particle interactions in NP colloidal suspensions with PP colloids, a sample containing glass bubbles with an average size of 7μm (with an average refractive index $n_{p2} < 1.33$) suspended in water is used, and a snapshot showing repulsion (rather than attraction) of glass bubbles by the laser beam is shown in the bottom panel of Fig. (28.b). In addition, we demonstrate clearly in Fig. (28.c) how the laser beam drives particles in a sample of MP suspensions ($n_{p1} > n_b > n_{p2}$), where simultaneous attraction of high-index polystyrene beads ($n_{p1}$) and repulsion of low-index glass bubbles ($n_{p2}$) are observed. The typical setup used for the experimental images
demonstrated in figure 28 and our further self-trapped beams studies in nano-suspension environment, is shown in figure 29.

![Experimental setup used to demonstrate light matter interaction in a colloidal suspension. Verdi: Laser, RT: Reversed telescope, L: lens, CCD: charge-coupled device which is used to take the snapshots at the sample position, and the side view camera records images of beam propagation along the sample.](image)

In Figure 29, the reverted telescope system is used to expand the beam so the first lens is be able to make a tight enough focus inside the suspension. The final lens is used as the imaging lens for the CCD, since moving this lens back and forth will allow taking different snapshots of the beam profile along beam propagation, as needed. A side-view camera zoomed on the beam is used at all time, in order to study beam propagation and to record the results.

In what follows, we shall show that only in those colloidal suspensions exhibiting overall NP can a laser beam undergo stable nonlinear self-trapping and enhanced transmission.
4.2. Beam Propagation through Positive and Negative Polarizability Nano-suspensions

So far we discussed the effects of optical forces and how a focused beam interacts with particles in a PP and NP colloidal suspensions, under the same experimental conditions. A schematic illustration and description of our experimental setup is shown in figure 29. Basically, a focused laser beam (λ=532 nm) is sent into 10-mm-long glass cuvettes filled with different colloidal suspensions, and its linear and nonlinear propagation through the sample is visualized from scattered light using a side-view camera. To synthesize a stable NP colloidal suspension, we chose Polytetrafluoroethylene (PTFE) particles with a refractive index $n_p=1.35$ [116], suspended in glycerin-water mixture (at 3:1 ratio) that has an average index $n_b=1.44$. The PTFE particles have an average diameter of 200 nm and a volume filling factor $f=0.3\%$. For a fair comparison, the PP colloidal suspension is made of polystyrene (PS) particles ($n_p=1.59$) of the same average size and same filling factor suspended in the same liquid background. With the same input laser beam, while no appreciable difference is noticed at low powers, dramatically different behavior of nonlinear beam propagation is observed at high powers as shown in figure 30. (a) and (b). Specifically, in the PP nanosuspension, the beam simply cannot penetrate through the sample, as it collapses after less than 5mm of propagation due to strong scattering loss accompanied by catastrophic self-focusing. In contrast, in the NP nanosuspension, the same beam propagates much longer distance throughout the 10-mm-long sample, as the particles are pushed away from the beam path. In fact, in the latter case, the beam forms a stable self-trapped channel, as the optically induced self-focusing nonlinearity is now saturable (rather than non-saturable as inherent to traditional PS-water-type Kerr nonlinear systems [5,110]). This represents the first
observation, to our knowledge, of robust propagation and self-induced transparency in any synthetic nanosuspensions.

Just to compare with linear propagation (which cannot be easily pictured from side-view of scattered light at low power), we show in Fig. (30.c) linear beam diffraction in a refractive-index-matched suspension (PTFE particles suspended in a dilute glycerin-water mix of $n_b=1.35$).

![Figure 30](image)

**Figure 30** Experimental observation (side-view images) of beam propagation in colloidal suspensions with different polarizabilities: a) For PP suspension, strong nonlinear self-focusing leads to unstable soliton that gradually fades due to strong scattering along the beam path. b) For NP suspension, a long, stable soliton is formed accompanied by enhanced transmission; c) For refractive-index matched (zero-polarizability) suspension, typical linear beam diffraction is observed. In all three cases, the experimental conditions (input beam, background solvents etc.) remain the same except for the refractive index mismatch.

In this index-matched sample, the beam diffracts to about 14 times of its original width after 10 mm of propagation, regardless of the increase of input power, indicating that no nonlinearity exists in such a zero-polarizability ($n_p=n_b \Rightarrow \alpha=0$) setting. From this result we can also conclude
that the beam self-trapping which took place when $\alpha \neq 0$, is purely due to optical forces and not a thermal phenomenon. Our experimental results agree well with earlier theoretical predictions [109,117].

As an example of employing the saturable nonlinearity in NP suspensions, we show nonlinear self-trapping of a light beam into an optical spatial soliton in our PTFE-based nanosuspensions. Although spatial solitons were attempted in previous experiments with PP suspensions [2], no stable soliton formation was achieved simply because the saturable nature of the nonlinearity is essential to support two-dimensional spatial solitons [101]. In Fig. 31, a circular spatial soliton (self-channeled beam) is established in the PTFE nanosuspension [synthesized with the same particle filling factor $f=0.3\%$ and the same background solution as in Fig.30 (a, b)], where snapshots of transverse intensity patterns and their corresponding intensity profiles are presented in (a-c), along with the direct side-view of the soliton beam from scattered light in (d). At a low input power (5 mW), the incident beam (focused to about 11 $\mu$m at input to the sample) diffracts naturally to about 32 $\mu$m after 2mm of linear propagation. At a high input power (3W), however, the beam self-traps to about its initial size (within the 1$\mu$m resolution limit of the camera). The side-view image of Fig. (31.d) demonstrates clearly the formation of a stable spatial soliton even after 5mm of propagation (corresponding to 5 diffraction lengths). We emphasize that the bright spatial soliton realized here based on the saturable nonlinearity in NP nanosuspensions, fundamentally different from all previously observed soliton-like phenomena in soft condensed matter systems including, for examples, those from modulation instability mediated by Kerr-type nonlinearity in PP colloidal systems [4,5,110], those from hydrodynamic supercavitation-like
phenomena in absorbing colloidal material near liquid-gel transition [114], and those “hot-particle” solitons resulting from thermophoresis effects [118].

Figure 31 Observation of stable optical self-trapping in an NP colloidal suspension with \( f_0 = 0.3\% \). (a) Input beam profile; (b) Linear output intensity after a propagation distance of 2 mm at a low input power of 0.1W; (c) Nonlinear output corresponding to a stable soliton at a high input power of 3.0W. The side-view photograph (d) shows the propagation of such a self-trapped beam over a distance of 5 mm.

In fact, our colloidal suspensions with **PTFE** nanoparticles suspended in glycerin-water mixture represent clearly a NP soft matter system, in which the nonlinear optical response is found to be strongly dependent on the refractive index contrast between the particles and hosting liquid mixture, the particle size and the particle filling factor as well. As such, it is important to point out that the nonlinear self-trapping and self-induced transparency phenomena observed in this work do not originate from the thermal effects (the nanoparticles are only weakly absorptive at the wavelength used), but rather from the optical gradient forces mediated by the material polarizabilities.
4.2.1. Numerical Model of Beam Propagation in NP Suspensions

To better understand the beam propagation through a NP suspension and the observed enhanced transmission compared to the PP sample, we modeled the dynamics of beam propagation in above PTFE-based nanosuspensions starting from the Helmholtz equation: \( \nabla^2 E + k_o^2 n_{\text{eff}}^2 E = 0 \), where the effective refractive index can be determined from \( n_{\text{eff}} = (1 - f)n_b + fn_p \). Considering \( E(x, y, z) = \varphi(x, y, z) \exp(ik_0 n_b z) \), the slowly varying field envelope satisfies the following nonlinear Schrödinger equation [110]:

\[
\frac{i}{\partial z} \varphi + \frac{1}{2k_0 n_b} \nabla^2 \varphi + k_0 \left( n_p - n_b \right) f \varphi + \frac{i}{2} \sigma \varphi \varphi = 0 \tag{4.3}
\]

Here \( k_0 = 2\pi / \lambda_0 \) is the wavenumber, \( \rho \) is the particle density and \( \sigma \) is the scattering cross-section

\[
\sigma = \frac{128\pi^3 a^7 n_b^4}{3} \left( \frac{a}{\lambda_0} \right)^4 \left( \frac{m^2 - 1}{m^2 + 2} \right)^2 .
\]

Note that the particle density thus the local filling factor \( f = \rho V \) is a function of the beam intensity along the beam path. In the PTFE suspensions, the NP dielectric particles are pushed away from the beam center by the optical gradient forces, resulting in a decrease of particle concentration and the filling factor. The third term in eq (4.3) represents saturable Kerr type nonlinearity in the case of NP suspensions or a super-critical (exceeding Kerr) nonlinearity in the case of PP [109,110].

For simplicity, we consider the exponential model for the induced change of particle concentration, since it has been shown that the exponential nonlinearity is very similar to the
Debye-Huckel model for the colloidal systems involving NP dielectric particles [109]. Hence, using

$$\rho(I) = \rho_0 \exp\left(\frac{\alpha}{4k_BT} I\right)$$  \hspace{1cm} (4.4)$$

In Eq. (4.4) $\rho_0$ is the initial unperturbed particle density and $\alpha$ is the particle polarizability as defined earlier. One can normalize Eq. (4.3) so that the nonlinear evolution equation of beam propagation through the nanosuspension would be easier solved numerically [110]. After doing so, the result of numerical simulation of beam propagation in experimental conditions of $n_p=1.35$, $n_b=1.44$, $f=0.7\%$, particle average size of 200nm and 11 micron beam (FWHM) in the high power (nonlinear) regime are presented in Fig. (32.a). The corresponding experimental result is demonstrated in Fig. (32.b). As observed, the beam is stable in a NP suspension where it propagates about 10 diffraction lengths (more than 6mm) without collapsing or noticeable beam divergence.

Figure 32 Beam propagation through NP nano-suspension of PTFE with $f=0.7\%$, showing soliton formation in this type of media for powers larger than 2W: a) numerical modeling and b) Experimental results.
4.3. Mixed Polarizability Nano-suspensions and Nonlinear Tunability

The successful demonstration of NP behavior now allows one to synthesize soft-matter systems with tailored optical nonlinearities by judiciously mixing both PP and NP particles within the same dispersant. In such a setting, the resulting self-focusing arises from the collective action of both species involved with PP particles being attracted and NP ones being repelled by the beam. Yet, despite the fact that both processes lead to an increase in the refractive index, the respective Boltzmann contribute to the nonlinearity in entirely different ways. As a result, a gradual inclusion of NP particles into an initially pure PP system can transform its super-critical response into a saturable one. This scenario is depicted in Fig. 33(a) along with the “Kerr plane” marking the transition between these two regimes. Experiments confirming these prospects were carried out in mixtures of PTFE and PS particles. As Fig. 33(b) shows, the beam FWHM tends to decrease as more PP particles are introduced into an NP suspension (0.7%) - indicating that while the net nonlinearity increases, self-focusing remains stable. Yet, once the polystyrene content exceeds 0.3%, the system enters the supercritical phase and hence the beam undergoes catastrophic collapse. We next investigate self-induced transparency effects in NP dispersions. In general, scattering losses in colloidal systems increase with concentration. Given that in NP arrangements, the filling factor decreases exponentially with intensity, the overall transmission nonlinearly increases. In other words, in an NP setting, an intense beam should be capable of clearing up the haze from its own path—a desirable feature in scattering environments. Along these lines, we experimentally observe a substantial enhancement in transmission (from 18% to 34%) when the power is increased to 1.6 W in a pure 0.7% PTFE NP dispersion (Fig. 33(c)). This is in stark contrast to what we measure in an equivalent polystyrene PP suspension, whose
initial filling fraction is chosen to be 0.22% so to have the same initial transmission of 18% at the lowest input power. The transmission through this PP sample drops by a factor of two at the same high power level. Self-induced transparency effects are clearly evident in the NP systems.

Figure 33 (a) Normalized nonlinear response of a mixed-polarizability suspension. The collective nonlinearity arises from a superposition of the two components for a fixed overall volume filling factor. The pure NP dispersion corresponds to a mixing ratio of \( c = 0 \), while a ratio of \( c = 1 \) indicates a pure PP system. The transition between saturable NP and supercritical PP behaviors is indicated by the Kerr plane (gray). (b) Measured output beam size after 2 mm of propagation as a function of the input power for mixed colloidal suspensions containing 0.7% of NP and varying amounts of PP particles. (The input beam width was 11 \( \mu \)m). (c) Measured intensity-dependent transmission for different suspensions (black: pure NP \( (f_0 = 0.7\%) \); red: pure PP \( (f_0 = 0.22\%) \); blue or green: mixed) after 2 mm of propagation. All mixtures were appropriately diluted to exhibit the same linear transmission of 18% at the lowest input power. In (b) and (c) the error ranges are indicated by the shaded regions surrounding the respective graphs.

In addition, mixtures of these two suspensions at different percentages are used for two MP samples. As seen in Fig. (33.c), at low input power, the transmission is about the same for all four samples. However, as the input power increases, the transmission ratio decreases in the pure PS-suspension for reasons discussed above, but increases dramatically for the pure PTFE-suspension. Moreover, as the input power is raised to about 1.25 W, nearly fourfold increase in
light transmission is achieved in the PTFE-suspension as compared with the PS-suspension, indicating the emergence of nonlinear self-induced transparency. As explained earlier, such enhanced transmission and self-induced transparency is a direct outcome of light-matter interaction in the NP colloidal systems, and was never demonstrated before. In the two samples of MP suspensions, due to co-existence of two kinds of particle motion driven by the laser beam, the total scattering loss and thus the transmission ratio falls in between the pure PP and NP cases. As expected, higher PS percentages would result in overall lower transmission ratios at high power levels. These experimental results clearly suggest the possibility of engineering such synthetic nanosuspensions for tunable optical nonlinearities.

It is important to emphasize that stable self-trapping supported by the saturable NP nonlinearities is fundamentally different from that discussed for pure PP arrangements, such as those due to electrostrictive nonlocal response, optically induced thermodiffusion and thermophoresis effects, or nonlinear effects in polymers and liquid crystals. In particular, the nonlinear phenomena explored earlier such as supercavitation and nematicons in soft-matter systems do not originate from negative-polarizability optical gradient forces such as the ones discussed in our work. In fact, NP suspensions represent a new territory of materials, yet they have hardly been studied due to the difficulty in making such stable suspensions. The ability to penetrate light through fluids and manipulate their local properties in strongly scattering suspensions is very important, as opaque suspensions of particles are abundant in our daily life. In this work, we show one endeavor to achieve such a goal – deep penetration of light up through scattering suspensions by tuning the nonlinearity with NP.
CHAPTER FIVE: INTERACTIONS BETWEEN SELF-CHANNELED OPTICAL BEAMS

In the previous chapter we studied laser-material interaction inside different types of nano-suspensions and how it can lead to beam self-guiding. In this chapter we investigate how these wave-guided beams interact with one another in a system of negative polarizability particles which demonstrate saturable type of nonlinearities.

The nonlinear interaction of a laser beam with colloidal suspensions has been studied for more than two decades [2-5,122]. Yet, very recently, nano.suspensions as artificial nonlinear materials have attracted increasing interest in the scientific community [109,110,117]. A few groups have attempted to understand the physics behind the light-matter interactions, beyond the simple Kerr model [109,123-125]. For these soft matter systems the relatively high optical nonlinearity demonstrated is mostly due to the optical gradient forces acting on the particles. These interactions can lead to the well-known nonlinear phenomena such as beam self-lensing, formation of self-channeled beams (spatial solitons) and four-wave mixing, inside the nanosuspension [2-6,110,123]. In a study by Smith [2] it was shown that a polystyrene nano-suspension can illustrate an effective nonlinear coefficient ($n_2$) of about $10^3$ times that of CS2.

Spatial solitons and their interactions have been studied in a verity of nonlinear materials [99] but there are only a few experimental studies [2-6,109,123] demonstrating soliton type propagation in these engineered soft-matter systems. In the previous chapter we showed that the nonlinearity demonstrated in a negative polarizability (NP) suspension is of a saturable type, which can support stable 2D self-channeled beams. In this chapter we demonstrate, theoretically
and experimentally, how these self-guided beams affect one another’s “waveguide” and interact in a NP suspension.

5.1. Theoretical and Experimental Study

To better understand how a beam would evolve inside a colloidal media with artificial nonlinearity mediated by the NP, we modeled the dynamics of beam propagation in PTFE-based nanosuspensions starting from the Helmholtz equation $\nabla^2 E + k_0^2 n_{eff}^2 E = 0$, where the effective refractive index can be determined by $n_{eff} = (1-f)n_b + fn_p$, where $f$ is the particle volume filling factor, $n_b$ and $n_p$ are the refractive index of media and particles respectively. Assuming $E(x, y, z) = \varphi(x, y, z)\exp(ik_0n_bz)$ with a slowly varying field envelope, we reach the following nonlinear Schrödinger equation as seen in chapter 4 as:

$$i \frac{\partial \phi}{\partial z} + \frac{1}{2k_0n_b} \nabla^2 \phi + k_0 (n_p - n_b) \phi \rho V + \frac{i\sigma\rho}{2} \phi = 0 \quad (5.1)$$

Here $f$ is not a constant value and for particles with NP it will decrease with beam intensity as:

$$f(I) = f_0 \exp \left( \frac{\alpha}{4k_b T} I \right) \quad (5.2)$$

In Eq (5.1) $k_0 = 2\pi / \lambda_0$ and $\sigma$ is the Rayleigh scattering cross section. The third term in eq. (5.1) represents saturable Kerr type nonlinearity in the case of NP suspensions while the forth term denotes the nonlinear loss.
First, using two identical but out of phase beams separated by 13µm (2W power each) as the input, we solve Eq (5.1) numerically in a two dimensional configuration for certain experimental parameters of: \( n_p=1.35 \), \( n_b=1.44 \), \( f_0 = 0.3\% \), particle average size of 200nm and 12 micron beam (FWHM). The results of this simulation for the beam propagation and profile at their input and output planes, are shown in figure 34.(a-c) respectively.

![Figure 34](image)

Figure 34 Repulsion of two out of phase self-trapped beams, 13µm apart, in a NP nano suspension of PTFE (a-c). Numerical simulation of 3.5mm-long beams interaction at 2W: (b) beam profile at the input and (c) output. (e,f) show the corresponding experimental snapshots taken under the same nonlinear conditions. In image (g) one beam is blocked before the cuvette so in the absence of interaction, the beam keeps a constant distance from the dashed line (d) Beam diffraction at low powers (linear regime).

The electromagnetic fields of the two out-of-phase beams interfere at their overlap, canceling the optical forces effect in the center point between them. This in turn, modulates a refractive index
gradient in the soft-matter system that increases away from the midpoint, causing the beams to diverge away from each other as demonstrated here in fig (34.a). The same repelling dynamics for out of phase beams have been demonstrated for Kerr [99,126] and photorefractive [99,127] media.

The experimental setup is illustrated schematically in fig. (35), where a 532nm (Verdi) CW laser is split in two equal-intensity beams directed and focused inside a 5mm-long NP sample, by a system of mirrors, beam-splitters and lens (L1). The distance between the parallel beams is adjusted by translating mirror M1 while the relative phase between them can be finely adjusted by driving the piezoelectric transducer which the second mirror (M-PZT) is mounted on.

![Experimental setup for two self-trapped beam interaction in nano-suspension systems. BS: beam splitter; L: lens; M: mirror, PZT: piezoelectric transducer; CCD: charge-coupled device.](image)

The CCD shows profile images of the beam at desired distances away from the input surface of the cuvette by moving lens 2 (L2). The experimental results for out of phase self-trapped beam interaction are presented in fig. 34.(d-g), where the input and output transverse intensity patterns

65
illustrated in (e) and (f) are in good agreement with the corresponding theoretical plots (b) and (c). If one of the beams is blocked, now in the absence of beam interaction, we expect the single beam to propagate straight through the suspension. The snapshot taken under this condition for the beam output is observed in fig. (34.g). For low input powers (5 mW), the optical forces are weak, consequently no significant index gradient will form in the nano-suspension and the natural beam diffraction dominates, as observed in fig. (34.d).

How these self-confined beams affect each other’s induced waveguide through the NP-PTFE suspension and consequently the final separation between them is critically sensitive to their initial phase difference. In the case of two in-phase beams as demonstrated in fig.36, their interaction results in attraction and the two almost coalesce after 3mm of propagation. For these data we used the same suspension as before while the initial beam separation was increased to 26µm, each with 2W power and 12µm (FWHM) width.

Figure 36 Attraction of in phase self-trapped beams, 26µm apart, in the NP-PTFE with 2W power. (a-c) Numerical simulation for two 3mm-long beams show attraction during propagation: (b) beam profile at the input and (c) output. (d,e) show the experimental beam profile images at the input and output respectively.
In our last study, using the exact same suspension, we change the initial relative phase between the two beams to $\pi/2$ and their distance to 20µm, at the input. The results are presented in fig.37. Here the beams affect each other in a more complex manner; Not only do they repel each other, but they also exchange energy through a four-wave mixing process as observed for 2D solitons in certain nonlinear materials [99].

![Image](image_url)

Figure 37 Interaction of $\pi/2$ out of phase, 3mm-long self-trapped beams, in the NP-PTFE suspension: (a-c) Numerical simulation showing power exchange between the beams with 20µm initial separation and 2W power. (d,e) Show the corresponding experimental results recorded at the same initial condition.

In conclusion, we have demonstrated self-trapped beam propagation through a suspension of PTFE nanoparticles. Now in the presence of a second beam, depending on their relative phase and how they affect the media we observed them repelling, attracting and transferring power between each other. It is worth noting that the possibility to create stable 2D self-guided beams
inside this (NP-PTFE colloid) and to study their interactions were due to the nature of optical forces in this engineered saturating nonlinear soft-matter system. This ability to synthesize pre-engineered saturating nonlinear soft-matter systems may now allow a systematic study of physical kinetic effects in the presence of optical forces exerted by self-channeled beams.
CHAPTER SIX: TUNABLE POLARIZABILITY AND SELF-TRAPPING OF LIGHT IN PLASMONIC NANOSUSPENSIONS

Developing engineered materials demonstrating extraordinary optical properties has become one of the major scientific researches, especially in the case of soft-matter media [128-134]. For these environments, light-matter interactions have always been dominated by strong scattering losses. The beam propagation becomes further dissipated in the nonlinear regime where optical beams tend to catastrophically collapse inside a typical positive polarizability system [110,135]. To this end, the ability to control these dynamics in order to extend beam propagation will have a significant impact on numerous applications, for example in opto-fluidics, biology and optical manipulation in scattering media.

Noble-metal nanoparticles (MNP) and their wide range of application [136-141], have attracted a great deal of attention in the past decades. Their distinctive optical properties are significantly affected by surface plasmon resonances (SPR) [11,142], which are a function of particle parameters [142–144]. By judiciously altering these parameters, one can tailor their optical properties, both in linear and nonlinear regime, to their advantage. Although the linear optical properties of these structures have been extensively studied, much remains to be explored and understood in terms of their nonlinear properties. In this letter we study how the MNP size, shape and composition affect nonlinear light-matter interactions, in order to overcome the aforesaid complications in colloidal media and allow deep beam penetration with suppressed diffraction.

As mentioned in previous chapters, during the propagation of a focused beam inside a colloidal suspension, depending on their polarizability sign, particles tend to be attracted or repelled from
the beam center. This is due to the gradient optical forces \( F_{\text{grad}} = \frac{\alpha_r}{4} \nabla |E|^2 \) present for focused beams, where \( E \) is the electric field and \( \alpha_r = 3\epsilon_0\pi(n^2 - 1)/(n^2 + 2) \) is the real part of the polarizability. So for samples with positive polarizability (PP) the particles are attracted towards the beam center, whereas their negative polarizability (NP) counterparts are repelled. Despite the difference in the dynamics, in both cases the result is a self-focusing nonlinearity, inducing a waveguide capable of trapping an optical mode. Optical wave propagation and the refractive index change in such photoresponsive system obey a nonlinear Schrödinger-like equation as mentioned in chapter 4.

Since the polarizability of most dielectric colloids tends to be positive and particles will drift into the maximum intensity region of the beam, this causes even worse propagation conditions due to enhanced scattering. At the same time, due to strong self-focusing the beam will eventually collapse [145]. In chapter 4, we discussed how to solve this issue by engineering the soft-matter system to obtain NP at the wavelength of desire. This results in particles moving away from the beam center and consequently reduced scattering, while at the same time leading to a saturable Kerr response. Consequently, stable low-loss propagation of needles of light inside dielectric suspensions could be achieved. However intrinsic NP suspensions are rare, difficult to produce and mostly composed of dielectric particles with fixed and relatively small polarizability values resulting in weak optical forces. Hence, one would need a high density of particles to create a large enough refractive index change capable of self-trapping the beam. This in turn limits the strength and depth of effective gradient forces, restricting the wave-guiding length to only a few millimeters, as demonstrated in a recent work [145]. Here we investigate aqueous colloids of
Plasmonic structures where their SPR, enhances polarizability values up to 2 orders of magnitude, compared to dielectric particles of same volume. This effect along with other impacts of SPR, allows not only the NP but also the PP Plasmonic suspensions to support self-channeling of light up to more than 5cm of propagation, as demonstrated here for the first time known. We discuss the different nonlinear mechanisms behind these phenomena which is the result of an overall positive $n_2$. The way light affects these media can be instrumental in controlling mesoscopic kinetic processes like diffusion, used for sorting particles or applied to certain spectroscopy techniques. A summary of the different nanoparticles used in our study and their polarizability type, at the wavelength of interest (532nm), is presented in figure 38:

![Figure 38](image)

Figure 38 Overview of the four exemplary plasmonic particle geometries investigated in this study.

**6.1. NP: Plasmonic Nanosuspensions**

The only reason for NP of dielectric particles in a colloid is that their refractive index is less than the background and this polarizability value depends mostly on particle volume. On the other hand due to SPR and its dependency on particle shape, size, composition, wavelength and $n_b$,
Plasmonic dispersions can be cleverly designed to exhibit artificial NP, with tunable values. In this section, we study the conditions where two different Plasmonic structures demonstrate NP in an aqueous suspension and how self-activated “needles of light”, can be achieved. These two particles are: gold nanorods with a 50nm width and 100nm of length (sample 1) and silica–gold core-shells with a 120nm diameter silica core and 15nm thick gold shell (sample 2).

As mentioned earlier, optical forces acting on particles depend on the real part of the polarizability ($\alpha_R$). In Fig 39 the dependency of SPR and consequently $\alpha_R$ of both samples 1 and 2, on particle size ratio and light wavelength have been presented. All calculated plots for nanorods consider its larger axis parallel to the linearly polarized E-field of our laser source for the reason that follows: A linearly polarized E-field of the laser induces transverse and longitudinal dipole resonances in a randomly oriented rod. A summation of these dipoles leads to an overall dipole moment $p$, indicated by a blue arrow in Fig 40.a. In general, the orientation of asymmetric particles in the presence of an electric field $E$ depends mostly on their size and shape. While larger particles (compared to the beam waist) tend to align along the direction of propagation, smaller particles align orthogonal to it and along the electric field [146-148]. Since the nanorods used in our experiments are much smaller compared to beam waist and the longitudinal polarizability is stronger than the transverse mode, due to the potential energy $U= -p.E$ it is energetically more favorable for the particles to orient their longer axis parallel to the field. In this new alignment, polarizability values of nanorods have considerably increased, enhancing the optical force effects which can result in a larger local refractive index change. Since we have only accounted for the nanorod’s longitudinal resonance, the measured extinction
spectrum that is taken under nonpolarized light has an extra peak compared to our calculated one in Fig 39(b).

Figure 39 Calculated values for the polarizability of (a) gold nanorods (sample 1) and (d) silica-gold core-shell particles (sample 2) when suspended in water. Here, the blue and red curves represent the real part $\alpha_R$ and imaginary part $\alpha_{im}$, respectively. Both types of particles exhibit NP behavior at the experimental wavelength of 532 nm (indicated by vertical lines). The calculated and measured normalized extinction cross sections for samples 1 and 2 are displayed in (b) and (e), respectively. (c) Illustrates the tunability of $\alpha_R$ for gold nanorods as a function of the rod length when the width is fixed at 50 nm. For reasons of visibility, all plots have been normalized. (f) Demonstrates the tuning of $\alpha_R$ for silica-gold core-shell particles similar to those in sample 2 as a function of the silica core size for a fixed shell thickness of 15 nm.

Depending only on $n_p$ and $n_b$, dielectric suspensions display either PP or NP for all wavelengths while for plasmonic structures, the polarizability sign and value change significantly with wavelength, observed in Fig 39 (a,d). As the wavelength increases, $\alpha_R$ changes from negative values on the short-wavelength side of the SPR, to zero exactly on resonance and becomes
positive on the longer wavelength side. This leads to repelling optical forces for laser wavelengths bellow resonance and attractive forces above it. Hence by adjusting the SPR, whose wavelength can be tuned throughout the visible and near-IR [11], the wavelength range and the strength of repelling forces can be modified at will. Fig 39(c) shows calculated $\alpha_R$ for gold nanorods of fixed width (50nm) and different lengths varying between 100nm and 260nm. As the rod’s length increases, the wavelength range of NP red-shifts. A similar effect occurs for core-shell structure observed in Fig 39(f), where the core to shell ratio increases. In both figures, in order to show the shift clearly, all plots have been normalized to their NP peak (-1).

Next we experimentally studied light propagation inside commercially available samples 1 and 2, at the wavelength of 532nm where the polarizability is negative in both cases. At all time, our experiments were performed under the same condition. We launched a focused laser beam (532nm, 15μm FWHM) into a 5cm-long glass cuvette containing the aqueous suspension. The respective volume filling fractions were chosen between $f_v = (2 \ldots 5) \cdot 10^{-6}$ to obtain samples of comparable optical density.

Typical light propagation inside sample 1, for low and high powers are captured by a side-view camera and shown in Fig 40(c,d), in hot-color presentation. For each image a different neutral density filter was placed in front of the camera or CCD, depending on the sample absorption and scattering. This was done in order to prevent saturation and maximize contrast. At low power levels ~10mW the beam considerably expands due to linear diffraction (Fig 40(c,e)). As we slowly increased the power, due to stronger gradient forces, more particles are pushed away, slowly increasing the self-focusing nonlinearity. As a result, the beam divergence reduces
gradually, evident from the output profiles in Fig 40(f,g) till finally at a certain power level ($P_{\text{threshold}}=250\text{mW}$) it self-traps, forming a 5cm long filament (Fig 40(d,h)). This is due to the anticipated saturable Kerr-type response of the NP media that finally balances diffraction.

Figure 40 (a) Orientation of gold nanorods along the electric field $\vec{E}$ of a linearly polarized beam. (b) Field distribution around the nanorod at the longitudinal plasmon resonance. (c) Linear diffraction of a low-power beam (10 mW) when injected into an aqueous suspension of gold nanorods (sample 1). (d) Formation of a stable self-trapped filament at 250 mW over 5 cm (25 diffraction lengths) mediated by the negative polarizability of the colloid. (e-h) Beam profiles observed after 5cm of propagation at different input power levels, showing the transition from diffractive broadening at 10 mW to self-trapping at 250 mW. For reasons of visibility, the output beam profiles have been normalized with respect to their individual peak intensities.
In the same experiment for sample 2, we observe similar dynamics as illustrated in Fig 41. This was expected, since both samples demonstrate NP at 532nm, but there is one major difference. At $P_{\text{threshold}}$ (300mW), the 5cm long self-trapped beam inside sample 2 was more than 3 times wider than in sample 1, which can be explained as the following; comparing the polarizability values of these samples at 532nm from Fig 39(a,d) we see that the core-shells have a fivefold larger $\alpha_R$ value compared to the rods. This consequently results in larger negative optical forces, expelling higher density of particles away from center, leading to a waveguide with a larger cross section.

Since the polarizability values of these Plasmonic structures are at least an order of magnitude more than the typical dielectric nanoparticles with NP, the gradient forces are significantly stronger and more effective, even at longer propagation distances for the case of MNP. This is why we observe such a long needle, propagating more than 25 diffraction lengths for samples 1 and 2 here, while for NP dielectric colloids only a few diffraction lengths (a few mm) was experimentally possible as demonstrated in chapter 4.
As mentioned earlier, close to the SPR of plasmonic structures, due to free electron oscillation of the particles they show strong polarizability values. This in turn results in stronger optical forces in the presence of a field gradient $\nabla E$, much stronger than the forces acting on dielectric beads.
As shown in chapter 4, PP dispersion of polystyrene particles cannot create a stable self-channeling and in the best case scenario the beam catastrophically collapses after a few millimeters of propagation, due to supercritical self-focusing effects. Now, in the case of PP Plasmonic colloid with much larger values of $\alpha_r$, one would anticipate that at higher power levels the beam will collapse much faster, due to the stronger gradient forces. Nevertheless as can be seen from Fig 43(c) for spherical gold nanosuspension with PP, the opposite occurs; the beam self-channels and can stably propagate for more than 10cm (50 diffraction lengths). Also the fact that many research groups, based on the closed aperture Z-scan have reported negative $n_2$ values for gold and silver nanosuspensions [149-151], is in stark contrast with the self-focusing type behavior we demonstrate here. Why is that?

In this section, we study the possible reasons for the long needle formation inside PP plasmonic dispersions and explain the disagreement with previous findings. The particles used are: gold spheres with D=40nm (sample 3) and silver spheres with D=100nm (sample 4), where their aqueous suspension at 532nm show PP. Also, as observed in Fig 42(a,d) this wavelength is close to the SPR of both samples, however silver particles present one order of magnitude larger values of $\alpha_r$ with respect to that of gold.
Figure 42 Calculated values for the polarizability of (a) gold spheres (diameter 40 nm, sample 3) and (d) silver spheres (diameter 100 nm, sample 4) when suspended in water. Here, the blue and red curves represent the real part \( \alpha_R \) and imaginary part \( \alpha_{im} \), respectively. Both types of particles exhibit PP behavior at the experimental wavelength of 532 nm indicated by vertical lines. The calculated and measured normalized extinction cross sections for samples 3 and 4 are displayed in (b) and (e), respectively. (c,f) Calculated absorption- and scattering cross-sections \( \sigma_A \) and \( \sigma_S \) of the gold and silver spheres, respectively. Note that for sample 3, absorption is the dominant effect (\( \sigma_A \gg \sigma_S \)), while in sample 4 scattering plays the major role (\( \sigma_A \ll \sigma_S \)) at 532 nm.

Here, we repeat the same light-matter interaction done for NP suspensions, under the same experimental conditions. As before both samples have similar OD at 532nm. Typical light propagations inside sample 3, captured by the side-view camera are shown in Fig 43 (b-d), in hot-color presentation. At low power levels \(~10\text{mW}\), diffraction occurs similar to the NP case. As we increased the power, the divergence reduced slowly, up to a point (\( P_{\text{threshold}} = 150\text{mW} \))
where self-trapping occurred, forming a long filament. Given that the dispersed gold nanoparticles of sample 3 exhibit a PP at 532nm, one may wonder, why at higher powers the beam did not collapse?

Intuitively, this might be explained as follows. As the power is increased, more particles are dragged towards the beam, increasing the particle concentration. Eventually, this density growth stalls because of the induced osmotic pressure caused by particle-particle interactions [109]. Nevertheless, even in this case, the self-focusing nonlinearity is still supercritical. On the other hand, unlike PP dielectric suspensions, sample 3 demonstrates significant absorption since 532nm is right at the peak of absorption curve (Fig 42(c)). This in turn leads to a thermal defocusing as a result of negative dn/dT effects in water [121]. The combined action of self-focusing (due to the gradient forces) and self-defocusing (due to thermal effects) results in a stable wave-guided beam, akin to that in a cubic-quintic saturable nonlinear medium. Indeed, as the power is further increased above $P_{\text{threshold}}$, nonlinear defocusing dominates and the beam diverges strongly (Fig 43(d)).
Figure 43 Field distributions at the plasmon resonances of (a) 40 nm gold spheres (sample 3) and (b) 100 nm silver spheres (sample 4), respectively. Sample 3: (b) Linear diffraction at 10 mW, (c) thermally mediated stable self-trapping under PP conditions at 150 mW, and (d) thermal nonlinear defocusing at 500 mW. Sample 4: (f) Catastrophic collapse in the PP suspension at 10 mW. (g) Around 2000 mW, thermal effects become strong enough to balance the supercritical PP nonlinearity and stabilize the beam. (h) Thermal defocusing eventually overcomes the self-focusing process. For each side view image a different neutral density filter was placed in front of the camera, depending on the sample absorption and scattering to prevent saturation and to maximize the contrast.

Mathematically this balance can be expressed as:

$$i \frac{\partial \phi}{\partial z} + \frac{1}{2k_0 n_b} \nabla^2 \phi + k_0 (n_p - n_b) \rho V \phi - |k_0 \Delta n_T| \phi + \frac{i \sigma \rho}{2} \phi = 0$$  \hspace{1cm} (6.1)$$

The third term in eq (6.1) presents a super-critical (exceeding Kerr) nonlinearity in the case of PP suspensions [109]. Here $\Delta n_T$ is the change in the refractive index of the solution due to temperature rise caused by the thermal effects discussed, approximated by:
\[
\frac{\partial n}{\partial T} = \left( \frac{\partial n_{\text{Solution}}}{\partial T} \right) (1 - V_f)
\]  

(6.2)

Where \( V_f \) is the local volume fraction of the particles and \( \left( \frac{\partial n_{\text{Solution}}}{\partial T} \right) \) is a negative value.

The third and fourth term in eq (6.1) describe the nonlinear behavior of the media and are self-focusing and defocussing effects respectively. While both terms increase as a function of beam intensity, eventually at a certain power level they will add up to balance diffraction which lies in the second term.

To further investigate our hypothesis, we repeat the experiment for sample 4. For this sample, at 532nm, while the polarizability value is 15 times that of sample 3, its absorption cross section is significantly less. As a result, the gradient forces rising from strongly polarized silver particles will be significantly larger while the thermal effects are minimized (scattering dominates as seen in fig 42.f). Consequently, the beam collapses even at lower power levels as observed in Fig 43(f). This effect can be demonstrated for laser powers < 1W. As we increased the power above 1W, the self-focusing term in eq (6.1) eventually saturates while the thermal defocusing term builds up, till about 2W (\( P_{\text{threshold}} \)) the defocusing term becomes strong enough to stabilize and self-trap the beam as seen in Fig 43 (g). Once we raised the power to 4W, self-defocusing becomes dominant and the beam diverges (Fig 43(h)).

The most common method to measure \( n_2 \) (nonlinear refractive index) of any material at a specific wavelength (\( \lambda_s \)), is the closed aperture Z-scan technique [152]. In this method, a thin sample of the media is moved along a focused laser beam of \( \lambda_s \), starting from tens of Rayleigh range before the focus to the same distance after the focus, while the transmitted light is recorded
at each sample position. Now in light of our findings in this paper, let's discuss the reasons why the Z-scan technique could show a negative $n_2$ (self-defocusing) in most cases for plasmonic nanosuspensions, as has been established [149-151]. As we demonstrated in the previous sections, for a strongly focused beam inside the colloid at certain power levels, regardless of the sign of $\alpha_r$ and due to different mechanisms, the plasmonic system behaved as a self-focusing media where the beam underwent self-trapping. Now, if we moved the sample till the position of focus was before or after the sample (as happens in the Z-scan), we would never observe self-channeling due to absent or significantly weaker gradient forces. Also depending on the power level, diffraction or thermal defocusing can dominate in these environments, which in both cases results in a negative $n_2$.

In Summary, we demonstrated that the optical polarizability of plasmonic soft-matter systems can be tailored at will. Depending on the polarizability sign and value, the optical forces can be greatly modified and strongly enhanced if close to a SPR. As a result here, for the first time we observed self-trapping and robust propagation of light over significantly long distances (up to 25 diffraction lengths) that would have been otherwise impossible. The longest self-channeled beam in dielectric nanosuspensions yet reported to our knowledge is only a few diffraction lengths. This is mainly due to the substantially larger $\alpha_r$ values of the Plasmonic structure, where along with their other tunable optical properties, mediate controllable nonlinear dynamics. Our findings here can open up new opportunities in developing novel, versatile soft-matter media with engineered optical nonlinearities. In addition to optical manipulation, these results can help improve certain spectroscopy techniques such as SERS in order to obtain a stronger signal.
CHAPTER SEVEN: ANOMALOUS OPTICAL FORCES ON MIE-PARTICLES BY CREATING A TRANSVERSE POYNTING VECTOR FLOW

One of the most intriguing properties of light-matter interaction is the ability of an electromagnetic field to exert mechanical forces upon polarizable objects. Such phenomenon is a direct consequence of light scattering and momentum conservation, which usually leads to a “pushing force” on an object in the direction of beam propagation. However, recent studies [153–158] have demonstrated that particles can be pulled by nonparaxial light beams towards the light source. So far, several theoretical contributions have dealt with the possibility of exerting reverse optical forces over dielectric objects either by exploiting structured illumination or complex environments [153-160].

Radiation pressure has fascinated scientists for centuries. In 1619, Johannes Kepler proposed the existence of radiation pressure, by considering light as composed of small particles able to transfer their momentum and push small particles in the direction of the photon flow. It is now well known that the spatial redistribution of electromagnetic waves upon scattering, causes the transfer of momentum to any polarizable obstacle by means of optical forces [157].

Since the very first experimental observations of opto-mechanical interactions by Lebedev [161] and by Nichols and Hull [162], the subject of optical forces has been explored in depth, both from the theoretical and the experimental point of view. Optical tweezers [23] are perhaps the best example of the ways in which optical forces can be harnessed and even turned into a powerful tool for the remote manipulation of microscopic objects. This tool has become a mainstay of research in biology, physical chemistry and soft condensed matter physics.
7.1. Pulling Forces

In the modern way of micromanipulation, structured beams of light are used to; push, trap and translate particles[4,29]. The fundamental dynamics of optical manipulation are rather simple for dipolar objects: particles much smaller than the wavelength of incident light develop an electric dipole moment in response to the light’s electric field. The induced dipole is then drawn by field intensity gradients, radiation pressure and spin force [11,22]. In free space, the radiation pressure is proportional to the Poynting vector, which determines the direction and magnitude of the momentum flow. The spin force is a non-conservative force arising in a light field of non-uniform ellipticity, proportional to the curl of the spin angular momentum of the light field [21]. It is possible to manipulate these micro-particles by appropriately designing the irradiating beam shape to control; local vortices, light spin patterns and complex Poynting vectors [157]. However, it was believed that there is no way to move a particle in any other direction other than the photon stream. It has been recently shown by Chen et al. [154] that for a larger particle it is possible to create a pulling force that acts against the optical power flow. In this case, the multipole radiation field due to the incident field, interferes in a way that the net scattering becomes strongly focused in the forward direction. This narrowing of the scattered filed can only result to a pulling force when the irradiating beam’s projection of the total photon momentum along the propagation direction is small. Thus due to the momentum conservation the particle will be pulled towards the source of the incident beam, shown in figure 44 In order to demonstrate this effect, theoretical studies have proposed different types of beams; interfering plane waves [156,157] or a single Bessel beam with a semi-apex angle close to 90° [154,155]. Nevertheless, the practical use of Bessel-based "pulling beam" is limited due to their short
working distances (about 10 μm). In another study, Lee and colleagues [153] used a solenoid beam to pull a micro-particle over a distance of 8 μm along a helical trajectory towards the light source, while Brzobohaty et al [158] interfered two wide Gaussian beams at a large angle theta to pull polystyrene particles more than 50 μm away and used this force to sort particles of different sizes. Nevertheless in all these studies, this so called "tractor beam" has been demonstrated to be limited to the plane of incidence. The question that naturally arises here is in what kind of arrangement, the particle can be accelerated perpendicularly to the plane of incidence?

![Figure 44 Scattering and optical forces acting on: (a) Interaction of a plane wave with a Rayleigh particle, which induces only an electric dipole (resulting in isotropic scattering) and a net force pushing the particle in the forward direction. (b) The same plane wave interacts with a larger particle, leading to interference between the multipolar fields of the scattered field. This strongly focuses the scattered beam in the forwards direction while reducing the total pushing force. (c) Two plane waves with a large enough angle between them interacting with the same particle as (b), results in a pulling force due to the conservation of photon momentum [157].](image-url)
7.2. A Transverse Optical force on a Mie-particle

In this chapter, we investigate a different and counterintuitive optical force effect associated with an anomalous Poynting vector. We demonstrate for the first time, how a superposition of non-interfering plane waves can generate a transverse Poynting vector [160]. Surprisingly enough, an isotropic scatterer immersed in such field distribution experiences a transverse optical force orthogonal to the incidence plane. The underlining physics here is the interaction of perpendicularly polarized beams mediated by the induced electric and magnetic dipoles. The relatively simple geometry of our study could add an additional degree of freedom to optical manipulation and provide a novel perspective in the understanding of light–matter interaction.

The momentum transferred by an electromagnetic field to a scattering and/or absorbing body is given by the flux of the Maxwell’s stress tensor through any arbitrary surface enclosing the object [15]. This in general requires the exact knowledge of the incident field as well as of the field scattered by the object. Such demanding task is somehow alleviated when analytical representations of the fields are available, as happens in the case of spherical objects. Structured illumination and polarization control offer important degrees of freedom in tailoring the behavior of optical forces [155,158,159] which results in new and interesting phenomenology. Here we would like to show that a combination of plane waves of appropriate polarization and coplanar propagation vectors can, quite surprisingly, lead to non-conservative optical forces which are orthogonal to the power flow associated with each plane wave component.
To this end, the superposition of two plane waves is considered, with propagation vectors lying on the XZ plane and forming an angle $2\theta$ of as shown in Figure 45(a). With respect to this plane of incidence, the first plane wave is assumed to have its electric field along $y$ (TE wave), while the other will have its magnetic field along $y$ (TM wave). Each beam by itself will contribute to a Poynting vector that has components only in the XZ plane:

$$S_{TE} = \left( |E_0| \hat{\eta} \right) \left[ \hat{x} \sin(\theta) + \hat{z} \cos(\theta) \right]$$  \hspace{1cm} (7.1)$$

$$S_{TM} = \left( |H_0| \hat{\eta} \right) \left[ -\hat{x} \sin(\theta) + \hat{z} \cos(\theta) \right]$$  \hspace{1cm} (7.2)$$
It is very well known that the superposition of TE and TM waves, due of their orthogonal polarization, does not lead to any interference when observed on a plane defined by a normal vector $\mathbf{n}$ contained in the XZ plane. The total power flow density is trivially given by the superposition of the Poynting vectors associated with the two waves. A net power flow along $z$ is therefore expected. Nevertheless such overly simple field distribution hides some rather counterintuitive characteristics which can be highlighted by considering the Poynting vector distribution of this system:

$$ S = \left( |E_0| / n_0 \right) \left( \hat{z} \cos(\theta) - \hat{y} \sin(2\theta) \cos \left[ 2k_0 \sqrt{\varepsilon_0} \sin(\theta) x \right] / 2 \right) $$

(7.3)

Figure 46 Showing electric and magnetic field components of the two interfering TE and TM plane waves. The different cross-interaction of $E_x \times H_y^*$ that can lead to a poynting vector in the y direction is marked with squares of the same color: the yellow squares which are the electric field of TM wave (in the z direction) and the magnetic field of the TE field (in the x direction) and the green squares show the electric field of TM wave (in the x direction) and the magnetic field of the TE field (in the z direction).
As clear from eq. (7.3) there is in fact a component of the Poynting vector directed along the transverse direction \( \hat{y} \) which arises from the cross-interaction of the TE and the TM waves, highlighted in green and yellow squares in figure 46. If on one hand the interpretation of the \( z \) component of the Poynting vector as power flow is immediate and unambiguous, the meaning of the \( y \) component is less intuitive because of its anomalous features. There is in fact no wave propagation or phase accumulation in the \( y \) direction. Moreover, as eq. (7.3) indicates, this transverse component of the Poynting vector is characterized by periodic reversals depending on the \( x \) coordinate shown as red and blue stripes in figure 45.b. The width of each channel (\( \Delta x \)), where the sign of the Poynting vector remains the same, is given by \( \Delta x = \frac{\lambda}{4n \sin \theta} \). Here \( \lambda \) is the wavelength of the monochromatic field used and \( n \) is the refractive index of the particle media. Notice that the same phenomenon occurs for finite size beams of appropriate polarization, as shown in Figure 45(c), for the case of two Gaussian beams. As the simulation clearly indicates the Poynting vector lines circulate in the transverse plane \( XY \).

So far we have shown how the two interacting plane waves with TE and TM polarizations can lead to an unexpected Poynting vector, perpendicular to the plane of incidence of the beams. But would a particle of any size actually accelerate in the \( y \) direction? To answer this question we will first consider the simple case of a dipolar scatterer. The net optical force on a Rayleigh particle endowed with an isotropic electrical polarizability \( \alpha^E \) is given by [16,22]:

\[
\langle \mathbf{F} \rangle = \frac{1}{4} \alpha r \nabla |\mathbf{E}|^2 + \frac{k_0 \alpha}{\varepsilon_0 c} \left[ \frac{\langle \mathbf{S} \rangle}{c} + c \nabla \times \langle \mathbf{L}_S \rangle \right] \tag{7.4}
\]
The first term is the gradient force which in the case of plane waves is zero. The second term is the radiation pressure \( F_{RP} \) which as we shown in eq. (7.3) has contributions in both z and y directions:

\[
F_{RP} = \hat{z} \alpha_i E_i^2 k_0 \cos(\alpha) - \hat{y} \alpha_i \frac{E_i^2}{2} k_0 \sin(2\alpha) \cos[2k_0 \sin(\alpha) x]
\]  
\[
(7.5)
\]

The third term of eq.(7.4) is a result of Spin forces (Fs) and for a Rayleigh particle is calculated as:

\[
F_S = \hat{y} \alpha_i \frac{E_i^2}{2} k_0 \sin(2\theta) \cos[2k_0 \sin(\theta) x]
\]  
\[
(7.6)
\]

As seen from eq. (7.5) and eq. (7.6) the optical forces acting on a Rayleigh particle due to radiation pressure and spin have the same y component value but opposite sign, canceling each other’s effect. So this indicates that a Rayleigh particle with the sole electric dipole complex polarizability \( \alpha^E = \alpha^E_r + i \alpha^E_i \) cannot detect the transverse Poynting vector associated with the TE-TM plane wave combination. This is consistent with the symmetry with respect to y of the scattered radiation pattern shown in figure 47(a).

A sufficient condition to develop a transverse optical force is that the scatterer is endowed with a magnetic polarizability \( \alpha^M = \alpha^M_r + i \alpha^M_i \) in addition to the electric polarizability \( \alpha^E \). In this case, due to the additional magnetic dipole term, the forces acting on the particle cannot be described as simple as equation (7.4). In general, the average forces experienced by a scattering
object in a monochromatic field can be computed by evaluating the flux of the Maxwell stress tensor \( T \) through any virtual surface enclosing the particle [11]:

\[
< F > = \frac{1}{2} \text{Re} \left[ \int_{S} \hat{T} \cdot \hat{n} dS \right]
\]  

\[
(7.7)
\]

\[
\hat{T} = \varepsilon_0 \varepsilon_r \varepsilon \mathbf{E} \mathbf{E}^* + \mu_0 \mu_r \mathbf{H} \mathbf{H}^* - \frac{1}{2} \left( \varepsilon_0 \varepsilon_r \mathbf{E} \cdot \mathbf{E}^* + \mu_0 \mu_r \mathbf{H} \cdot \mathbf{H}^* \right) \hat{I}
\]  

\[
(7.8)
\]

In expression (7.7), \( \hat{n} \) is the unit vector normal to the enclosing surface \( S \) and pointing outward.

**Figure 47** Radiation pattern of a single particle in the presence of superimposed TE and TM waves which induce: (a) only an electric dipole which has a symmetric pattern and (b) both electric and magnetic dipoles which demonstrate an asymmetric scattering with respect to \( z \) and \( y \) axis.

In particular, in the presence of intersecting TE and TM fields with angle of \( 2\theta \), in the XZ plane we obtain the following force components [22,160]:

92
\[
F_x = \frac{\left(E_{TE}^2 - E_{TM}^2\right) k \left[6\pi \left(\alpha_i^E + \alpha_i^M \varepsilon_0 \mu_0\right) - k^3 \left(\alpha_i^E \alpha_i^M + \alpha_r^E \alpha_r^M\right) \mu_0\right] \sin \theta}{12\pi}
\]

(7.9)

\[
F_y = \frac{E_{TE} E_{TM} k^4 \left(\alpha_i^E \alpha_i^M + \alpha_r^E \alpha_r^M\right) \mu_0 \sin 2\theta}{12\pi}
\]

(7.10)

\[
F_z = \frac{\left(E_{TE}^2 + E_{TM}^2\right) k \left[6\pi \left(\alpha_i^E + \alpha_i^M \varepsilon_0 \mu_0\right) - k^3 \left(\alpha_i^E \alpha_i^M + \alpha_r^E \alpha_r^M\right) \mu_0\right] \cos \theta}{12\pi}
\]

(7.11)

Where \(\alpha_i^E\) and \(\alpha_i^M\) are the electric and magnetic polarizabilities respectively. If \(E_{TE} = E_{TM}\) then \(F_x\) in eq. (7.9) will be zero as expected. As we can see from eq (7.10), there is a non-zero force component perpendicular to the plane of incidence which was not expected. This proves the fact that as a consequence of complex interplay between spherical multipoles, a fully symmetric geometry can generate optical forces orthogonal to the plane of incidence.

To better understand the origin of this rather counterintuitive optical force component, we compare the radiation patterns of a dipolar scatterer with and without the magnetic polarizability. The illumination field is the superposition of a TE and a TM wave. As seen in figure 47(b), we observe a broken symmetry in the y direction of the radiation pattern of the particle with the extra magnetic polarizability. This asymmetric scattering is the source of the anomalous force in the transverse direction.
7.3. Experimental Demonstration of Symmetry-Broken Radiation Pattern

To verify the proposed anomalous transverse Poynting vector, we designed an experimental configuration that uses intersecting TE and TM laser beams with an angle of \(2\theta\) onto polystyrene particles stuck to a glass slide. The two perpendicularly polarized beams have the same power and are Gaussian beams rather than plane waves. For both our theoretical and experimental results we observed that the asymmetric scattering only occurred for certain particle diameter \(D<\Delta x\), so \(D\) is a function of beam wavelength \(\lambda\) and angle \(\theta\).

![Diagram of experimental setup](image)

Figure 48 (a) Schematic of the experimental setup for observation of 140nm particle scattering pattern in the presence of both TE and TM irradiation; DMO: Dark-field microscope objective for both irradiation and scattering collection; L: Lens; CCD: Charge-coupled device; PBS: Polarizing beam splitter; HWP: Half-wave plate; M: Mirror; S: Sample cover slide; PTS: Piezo translating stage. (b) Simulation showing particle asymmetric scattering patterns depending on the "channel" they are in (formed by the TE and TM beams).
For the experimental condition of $\lambda=532\text{nm}$, $n_m=1$ and $2\theta \approx 130^\circ$, $\Delta x=150\text{nm}$ which is the upper limit of the particle diameter. If the diameter is larger than this value the particle will not fit in a single channel (shown as red and blue stripes in figure 45.b) and the effect of the phenomena under study will be reduced due to the contribution of the neighboring channels. On the other hand the particle cannot be too small since it will not have a magnetic dipole contribution. Here we chose $D=140\text{nm}$ for a polystyrene bead ($n_p=1.59$) as the scattering object. Before doing the experiment we first simulated the scattering pattern of the chosen particle, for this specific size and material to make sure this particle will be large enough to demonstrate magnetic dipole in addition to the electric dipole, based on Mie theory [11]. The results were schemed as polar plots for better demonstration, shown in figure 49. For an electromagnetic field incident on a Rayleigh particle, the scattered field is only due to electric dipole radiation, so the resulting scattering pattern is symmetrical as seen in (a). For the case of 140nm polystyrene bead in air with an illuminating beam of $\lambda=532\text{nm}$ the scattering pattern is demonstrated in figure (b). The asymmetric pattern in this image is a clear indication of the presence of magnetic dipole.
Figure 49 Normalized calculated polar plots showing both parallel (blue) and perpendicular (red) scattering patterns of: (a) Rayleigh particle, (b) 140nm polystyrene bead in air, illuminated with a beam of $\lambda=532$nm and (c) 300nm polystyrene bead in water, illuminated with a beam of $\lambda=1064$nm. In all cases, light is incident from the top on a spherical particle located at the center of the polar plot.

In order to eliminate possible alignment issues of overlapping the TE and TM beams at the desired angle, which would compromise the clear identification of the anomalous transverse Poynting vector, we used a 150X dark field microscope objective (DMO) for both illumination and radiation pattern observation observed in 48 (a). The dark field objective used has NA of 0.9 and 0.7 for illumination and collection respectively. As demonstrated in figure (48.a), the TE and TM beams are sent through the back aperture of the objective and overlap on 140nm polystyrene particles on a glass slide. To avoid multiple scattering effects we have an extremely low density of particles on the slide so they are not too close to each other and can be almost isolated.

As mentioned earlier, due to the cross-interaction of perpendicularly polarized beams, at the position of the sample, there would be parallel channels (like hypothetical interference fringes shown in figure 48 (b)) where the Poynting vector of each has the opposite sign relative to its
closest neighbor. In order to check the radiation pattern of a particle, we first positioned it in one of the channels, using a piezo translating stage with 5nm step resolution. This could be only realized by imaging the Fourier plane of the objective on to the CCD in order to demonstrate at which position the particle demonstrates maximum asymmetric radiation pattern from the particle and at the same time recorded the results shown in figure (50.c,d).

Figure 50 Demonstration of radiation pattern of a Mie particle irradiated by intersecting TE and TM beams in phase (a,c) and Π out of phase (b,d). The top panel shows simulation results while the bottom panel illustrates our experimental results.
In Figure 50 (a) and (c), the TE and TM beams are in phase while (b) and (d) show the result when the TE (or TM) beam has accumulated a total of $\pi$ phase shift with respect to the TM (or TE) before reaching the sample.

### 7.4. Experimental Demonstration of Transverse Optical Forces on a Mie-particle and measuring the anomalous force value

In this section we design a second experiment in order to demonstrate particle acceleration in the transverse direction ($y$), in other words, perpendicular to the plane of incidence and measure the anomalous optical force in that direction ($F_y$). The experimental setup used is shown in figure 51. Here, in order to better study the optical force effect on the particle, minimize random particle motion and be able to measure particle movement systematically, we first trap the particle using an optical tweezers system. This was done using a 532nm laser beam tightly focused to 400nm in diameter by a 100X objective with a NA of 1.4 (where the beam power at the sample position is about 50mW). The particle is far from any surface so the corrections due to Stokes’ law can be neglected [31].
For a particle trapped inside an optical tweezers, in addition to the small Brownian motion, the particle can move due to external forces (up to tens of nanometers [29]) which can be measured experimentally using a quadrant photo-detector (QPD). The interaction of optical forces with a trapped particle, behaves like a microscopic version of Hookean spring where the force can be found as $\mathbf{F} = -k \times \Delta \mathbf{y}$, while $\Delta \mathbf{y}$ is particle displacement from trap center and $k$ is the trap stiffness. Now in order to create the transverse pointing vector flow mentioned before, we used two perpendicularly polarized 1064nm beams (TE and TM) irradiating the particle from the other side of the setup, shown as the 2 red beams in figure 51. The two beams are focused down...
to a width of 25μm (FWHM) and overlap with each other and the particle passed their focus, where their width is 30μm.

The first step in measuring anomalous transverse forces is to determine \( k \), which depends on laser power, particle size, temperature and the viscosity of surrounding medium. There are a few different methods to calibrate the trap and find \( k \). One of the most well-known approaches relies on the Power Spectrum (PS) analysis method. In the following section trap calibration is presented.

7.4.1. Calibration of Optical Tweezers and Finding Trap Stiffness

In this section we present the calibration method [29, 31,163-165] we used in order to find trap stiffness and measure particle displacement inside the trap. This is required in order to convert particle translation inside a trap to the force exerted on it. In our setup (figure 51) the backscattered laser trapping light [166], is used to detect lateral motion and consequently measure transverse force. This can be achieved by monitoring the backscattered light on a Quadrant Photodiode (QPD) which records the relative displacement between an object and the center of the trap with nanometer accuracy [31,165]. The fluctuations of a trapped bead due to Brownian motion can be expressed by the Langevin equation where the power spectral density of the particle position \( S_y(f) \) is given by the Lorentzian expression [29] :

\[
S_y(f) = \frac{k_b T}{\pi^2(f_c^2 + f^2)}
\]  

(7.12)

Where \( k_b \) and \( T \) are the Boltzmann’s constant and temperature respectively, \( f \) is the frequency, \( f_c \) is the cut-off frequency and \( \gamma \) is the drag coefficient.
Figure 52 Power spectrum density of a 300nm polystyrene bead in the 532nm laser trap. Green dots represent experimental data (Each dot is the average of 50 positional data) and solid black line shows the Lorentzian fit. Based on the power spectrum analysis the corner frequency has been found to be $f_c = 370 \pm 9.5 \text{ Hz}$ and the conversion factor $\beta = 2.67 \mu \text{m/Volt}$.

Fitting the experimental values of $S_y(f)$ (the green circles) to the Lorentzian model described in eq (7.12) (the black curve), shown in figure 52 we found the corner frequency $f_c = 370 \pm 9.5 \text{ Hz}$. Now, trap stiffness can be calculated using $k = 2\pi \gamma f_c$ and $\gamma = 6\pi \eta r$ where $\eta$ is the viscosity of the surrounding medium ($\eta_{\text{water}} \sim 10^{-3} \text{ Pa.s}$) and $r$ is the radius of trapped particle (150nm).

Using the given values we find $k = 6.57 \pm 0.17 \text{ fN/nm}$.

Note that the detector measures an uncalibrated power spectrum $S_u(f)$, which can be easily converted to the real power spectrum through $S_y(f) = \beta^2 S_u(f)$, where $\beta$ is the conversion factor.
representing the linear sensitivity of the detector [31] and can be found as : \( \beta = \left( \frac{k_B T}{S_u(f)\pi^2 Y} \right)^{1/2} \).

For our experiment we find \( \beta=2.67 \, \mu m/\text{Volt} \).

7.4.2. Measuring Particle Displacement and Calculating The Transverse Force

For this experiment the particle used was 300nm polystyrene beads inside water media, irradiated by 1064nm laser beams. Just as before, using Mie theory, we first checked the chosen particle scattering pattern inside water to make sure that the magnetic dipole exists. The result is observed in figure 49 (c). After trapping a 300nm polystyrene particle with the 532nm laser beam and calibrating the trap, the two perpendicularly polarized 1064nm beams are turned on to study the Anomalous force. In this case, the particle displacement due to this transverse force can be measured by detecting the backscattered 532nm laser beam (used for optical tweezers), which is collected by the same objective creating the trap and directed to the QPD where the motion of the particle is analyzed.

In order to move the trapped particle with a few nano-meter accuracy relative to the 1064nm laser beams (the channels) and to maximize the anomalous force effect, we added beam steering capability to the trapping section of the setup. This part is composed of a steering mirror (SM) and a 4f system shown in figure 51, where the surface of the SM and the back aperture of the trapping objective are conjugate planes [165]. Once the particle was trapped, we moved the particle using the SM system till particle displacement in the y direction was maximized. As indicated in figure 51, due to the specific design of the experiment, y is perpendicular to the table (same as the Poynting vector channels). Hence the trapped particle, due to the aforementioned
transverse anomalous force would move up or down, depending on the channel it's positioned at. The resulting particle displacement in $\hat{y}$, detected by the QPD is presented in figure 53.

![Graphs showing particle displacement](image)

Figure 53 Experimental results of the trapped particle displacement, due to the anomalous transverse optical forces in the $\hat{y}$ direction, detected by the QPD: (a) particle moves ($\Delta y = -28\text{nm}$) downwards when TE and TM beams are in phase and (b) particle moves ($\Delta y = 32\text{nm}$) upwards, once the TE and TM beams are $\pi$ out of phase, with respect to each other.

In figure 53 (a) the particle has displaced $\Delta y = -28\text{nm}$, when the two 1064nm (TE and TM) beams are in phase while in (b) the particle moves $\Delta y = +32\text{nm}$, once the TE and TM beams are $\pi$ out of phase, with respect to each other. The same result occurred once the two beams were in-phase and the particle was moved one channel, using the SM system. From these results we have found the average particle displacement in the presence of the two perpendicularly polarized beams to be $\bar{\Delta y} = 30\text{nm}$.
As mentioned earlier knowing the stiffness of the trap and the displacement, one can easily calculate the experimental transverse force value using Hooks law \((F = -k \times \Delta y)\) to be \(F_{y}^{\text{Exp}} = -197.1\ fN\). At the same time using eq (10) one can find it's theoretical value to be \(F_{y}^{\text{Theory}} = 154\ fN\). This difference of force values can be due to the fact that the particle gets slightly pushed along the z direction and in its new position the spring stiffness is slightly smaller than the one found before. This leads to larger particle displacements that was not accounted for in \(F_{y}^{\text{Exp}}\). Another reason could be due to possible thermal effects and change of viscosity of the surrounding media.

In this chapter, we have investigated a counterintuitive optical force effect associated with an anomalous Poynting vector. We demonstrated for the first time how superposition of two non-interfering plane (or Gaussian) waves can generate a transverse Poynting vector. This was realized by an isotropic scatterer immersed in such field distribution, where it experienced a transverse optical force orthogonal to the plane of incidence. The underlining physics here is the interaction of perpendicularly polarized beams (one TE and the other TM) with a scatterer of the right size, large enough to demonstrate both electric and magnetic dipoles. We observed this novel phenomenon in two different experimental setups; one studying the radiation pattern of the particle and the other by demonstrating particle motion in the transverse direction, which allowed us to experimentally find the anomalous force value. The relatively simple geometry of our study could add an additional degree of freedom to optical manipulation and provide an innovative perspective in the understanding of light–matter interaction.


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