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Role of pump duration on temperature and efficiency of up-conversion in fluoride crystals co-doped with ytterbium and thulium

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Abstract: Pump pulse duration is shown to determine the maximum efficiency of the up conversion process in ytterbium, thulium co-doped fluoride crystals through its role in determining the emitter temperature reached. We show that up-conversion efficiencies should only be measured when using the same pumping conditions as would be used in a proposed application and that thermal management of the up converting material is critical to optimized performance.

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References and links

1. Introduction

The concept of frequency up-conversion (UC) of infrared-to-visible light in rare-earth (RE) doped materials was first reported more than forty years ago [1]. The efficiency that was observed or expected for this process was low in singly doped media, but it was quickly noticed that the mechanism could be made one or two orders of magnitude more efficient by using ytterbium (Yb$^{3+}$) as a sensitizer ion in addition to the active ion: erbium (Er$^{3+}$), holmium (Ho$^{3+}$), or thulium (Tm$^{3+}$) [2-7]. Efficient UC materials were extensively investigated, as they could make possible such photonic applications as UC lasers (visible lasers that are pumped by infrared diode lasers) or displays. However, because no powerful source existed in the 980-nm region to excite those up-converters, no practical product came out of the research. With the development of powerful 980-nm diode lasers lead by the telecommunication industry, a technology that appeared unworkable in the past now has significant potential applications.

It was noticed in the past that pumping conditions caused heating of the material and that higher efficiencies were obtained with low duty cycle excitation [8, 9] but no systematic study of the effect was conducted. It was also reported that for the same average input power, higher irradiance and thus, higher efficiencies were achieved using pulsed excitation than when using continuous wave (cw) excitation due to the quadratic nature of the process [10]. The specific properties of the pumping conditions in display applications of UC must be understood, as several technologies might be used to form the image. The infrared source can either be scanned (vector-addressed or raster-scanned), or the image can be directly projected using Digital Micromirror Devices (MEMS) such as in the Texas Instrument Digital Light Processing (DLP$^{\text{TM}}$) technology. In the first case the materials would be undergoing pulse-excitation, whereas they would be quasi-continuously excited in the second case.

In this paper, we examine the role of temperature on the efficiency of blue light emitting doubly doped fluoride up conversion materials. The materials are studied in the powder form in which they would be used in an UC display application. We concentrate on the blue emitter because it is commonly the least efficient in a UC display. Many phonons participate in the blue emission process due to energy level mismatches and result in increased emitter temperature and a drop in efficiency. In particular, we investigate the effect of various pumping rates and duration on the performance of our best performing blue UC material: Yb, Tm doped yttrium lithium fluoride or Yb,Tm$^{3+}$:YLF.
2. Results

The principle of photonic displays can be found in previous publications [11-14]. In order to help the reader follow the subsequent discussions, the energy diagram of Yb$^{3+}$ and Tm$^{3+}$ in YLF is given Fig. 1, along with the possible energy transfers from Yb$^{3+}$ to Tm$^{3+}$ and cross-relaxations.

![Energy level diagram of Yb$^{3+}$ and Tm$^{3+}$ in YLF](image)

Fig. 1. Energy level diagram of Yb$^{3+}$ and Tm$^{3+}$ in YLF. Possible energy transfers from Yb to Tm are indicated in red thick solid lines, radiative decays are indicated in blue thin solid lines, and cross-relaxations are indicated black lines with hollow arrowheads. Energy back transfer from Tm to Yb is also possible along the same pathways.

Previously published work [15] showed that the performance of the blue up-conversion material was altered when pumping with a continuous source compared to short pulse excitation. Rate equation analyses illustrated that this behavior could be explained only when transfer and cross-relaxation rates were made temperature-dependent, to include the phonon-assisted fluorescence. Length limitations prevent our including a more extensive description of the numerical simulations. These will be reported in a separate paper, and we now concentrate on the experimental results. To better understand thermal effects, we used the fluorescence intensity ratio technique (FIR) as described in [16] for other rare-earth ions. In [16] thulium was excluded from the list of suitable candidates for thermal sensing. However in fluorides such as YLF, the sublevels of the blue emitting levels are well separated and emission peaks from thermally coupled levels can be resolved. Also the low phonon number in fluorides together with the strong Yb$^{3+}$ absorption of the pump power allow for high UC efficiencies and strong fluorescence signals.

We now describe how we used FIR to study the role of pumping conditions in the co-doped Yb-Tm system. We recorded the emission spectra obtained for a powder sample of Yb,Tm:YLF when pumped with various intensities and pulse durations. Figure 2 shows the results of such measurements in the case where the output power was the same for three pumping conditions. These were continuous (cw) pumping, and pulse pumping at 30 Hz with pulses having either 2 or 5 ms duration, at average powers of 155, 50 and 54 mW respectively.
Fig. 2. Emission spectra of 0.4% Tm, 25% Yb:YLF. Three conditions of excitation were used: the red bold line corresponds to continuous pumping at 155 mW average power, the green thin line represents 5 ms pulse at a repetition rate of 30 Hz at 54 mW average power, and the black dotted line represents 2 ms pulse at a repetition rate of 30 Hz at 50 mW average power. The output luminance in the blue was the same in the three excitation conditions (58 mL/m). The sample holder was made of acrylic; the powder was packed binder-free into the hole.

In Fig. 2, the blue output power was the same for the three pumping conditions. The peaks at 465.5 and 483 nm originate from the same energy level, the $^1G_4$ but different Stark split sub-levels. Those sub-levels are thermalized and the spectral distribution is therefore a signature of the temperature inside the sample. The emission band centered at 450 nm originates from the $^1D_2$ level of Tm, which is populated via a fourth UC step from the $^1G_4$ level and cross relaxations. However it lies 7000 cm$^{-1}$ above $^1G_4$, so these two levels are not thermally coupled, which was confirmed by lifetime measurements. Therefore, to correctly estimate the spectral distribution of the 480 nm band as a function of temperature, one has to first subtract the tail of the emission band at 450 nm. From the resulting spectrum, it is possible to say that when the ratio of the emission at 465.5 to that at 483 nm increases, it means that the temperature of the emitting powder is increasing. It can be seen from Fig. 2 that for the same output luminance, the temperature of the sample is much higher when continuous pumping is used than when pulsed excitation is used.

To get more quantitative measurements, we measured the spectral distribution around 480 nm after correction for the $^1D_2$ emission at various pre-set temperatures. A hot plate and a thermocouple were used to vary and record the temperature, and for each temperature the emission spectrum was measured after excitation with a low power laser pulse (no heating due to excitation). After subtracting the emission due to the $^1D_2$ level, the ratio of the 465.5 to the 483 nm emission was determined as a function of temperature. We used the results from this experiment to determine the temperature inside the sample for different pumping conditions while operating at room temperature. The incident power was varied as well as the pump duration. Figure 3 shows the estimated powder temperatures reached when cw pumped and when pumped with a 30 Hz, 5 ms pulse in either an acrylic or a copper powder holder. YLF, acrylic and copper have thermal conductivities of about 6, 0.2 and 400 W/(m.K), respectively [17-19]. The powder was composed of 50 µm size particles and was packed binder-free in a cylindrical hole in the holder that was 750 µm diameter and 500 µm deep. There are two conclusions that can be drawn from that plot: first, for the blue emitting Yb-Tm system at a given output power (or brightness), the temperature reached when pumping continuously is significantly higher than when a short duration excitation pulse is used.
Second, using a substrate that extracts the heat produced (e.g., has a high thermal diffusion coefficient) significantly reduces the heating of the powder.

Finally, we can measure the total output power as the temperature of the sample is raised for a fixed input power. The excitation was a 4 ns, 10 Hz pulsed OPO laser operating at 150 µJ and tuned to 959 nm (the peak excitation wavelength for Yb,Tm:YLF). The change in efficiency with temperature of the blue emitting Yb,Tm:YLF is shown in Fig. 4. Higher temperature means higher phonon density which increases the rates of phonon assisted energy transfers, cross-relaxation and energy back-transfer. Above room-temperature the net result of the increased rates of these processes in Yb,Tm:YLF is a decrease in the blue emission. Figure 4 shows that the up-conversion emitter performance diminishes significantly when operated at temperatures above room temperature while other experimental results show that temperatures above 100°C can be reached in this emitter (Fig. 3). Therefore, lowering the operating temperature is key to optimizing blue up-conversion materials performance. Preliminary simulations using FEMLAB™ and a simple heat-conduction model show that using a material with high heat-diffusion-coefficient as a substrate in a UC display (e.g., silver or aluminum coated copper for a reflective display or appropriately coated diamond prepared by chemical vapor deposition for reflective or transmissive displays), and reducing the pixel size will enable up-conversion materials to operate close to room-temperature. The data in Fig. 4 suggests that the efficiency of such a display will be between 2 and 4 times greater than possible if the display were designed with no attention paid to emitter thermal management. Such potential improvements are being investigated experimentally and results will be reported shortly.
3. Conclusion

Photonic displays based on up-conversion materials have numerous advantages that make the technology appealing: the phosphors emit very narrow lines which produce a very wide color gamut and saturated colors, high-brightness (several kCd/m²) can be achieved without damage to the phosphors, and no vacuum enclosure or high-voltage is required. However, improving the materials’ efficiencies is paramount to making this technology competitive with existing display technologies. In this work, a major step was identified that can lead to optimized performance of the up-conversion phosphors: using a thermally conductive substrate to conduct heat away from the blue emitting UC pixel will reduce the operating temperature of the powder and improve the efficiency of the display.

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