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Thermal lensing effects in Pr³⁺-doped ZnO–CdO–SiO₂ luminescent glass

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Abstract. Optical absorption, photoluminescence and fluorescence decay time of Pr³⁺ in a ZnO–CdO–SiO₂ glass were measured in the 400–800 nm range. Under continuous (CW) laser excitation the characteristics of thermal lensing (TL) were measured at room temperature, under resonant and non-resonant conditions. Our results are analysed based on a theoretical model for CW laser induced thermal lensing under a gaussian-beam approximation. The effects of the power input and the response time for TL are explained in terms of the thermal properties of the glass host, for non-resonant excitation, and in terms of the coupling of rare earth ions to the local vibrations for resonant excitations. We also report, for the first time, the observation of laser induced permanent changes in the refractive index of bulk rare earth doped oxide glasses, under non-resonant excitation conditions.

1. Introduction

Currently, the importance of glasses doped with rare earth ions as fluorescence standards and as medium for optical lasers has been stressed and aimed the efforts of physicist and engineers devoted to improve their optical and laser characteristics. For example, during lasing the active element of a laser heats up due to the absorption of pump radiation by the glass matrix. Furthermore, the temperature varies over the cross section of the element because of the finite thermal conductivity and the non-uniformity of the pump radiation. This change in the local temperature may cause a change in the refractive index known as thermal lensing (TL) [1]. Due to the importance of this effect during lasing operation TL has been studied in a large number of optical glasses [1–3]. In this paper we will discuss the results of a systematic study of the spectroscopic and thermo-optical characteristics of a Pr-doped ZnO–CdO–SiO₂ glass, and the effect of TL on the divergence of laser light.

2. Experimental procedure

The Pr-doped ZnO–CdO–SiO₂ glass studied in this work was formulated in the following composition: SiO₂ (40%), CdO (40%), ZnO (10%), Al₂O₃ (10%):Pr₂O₃ (3%).

The photoluminescence (PL) measurements were carried out using a Perkin–Elmer LS-5 fluorescence spectrophotometer fitted with a R-298 photomultiplier. This spectrophotometer

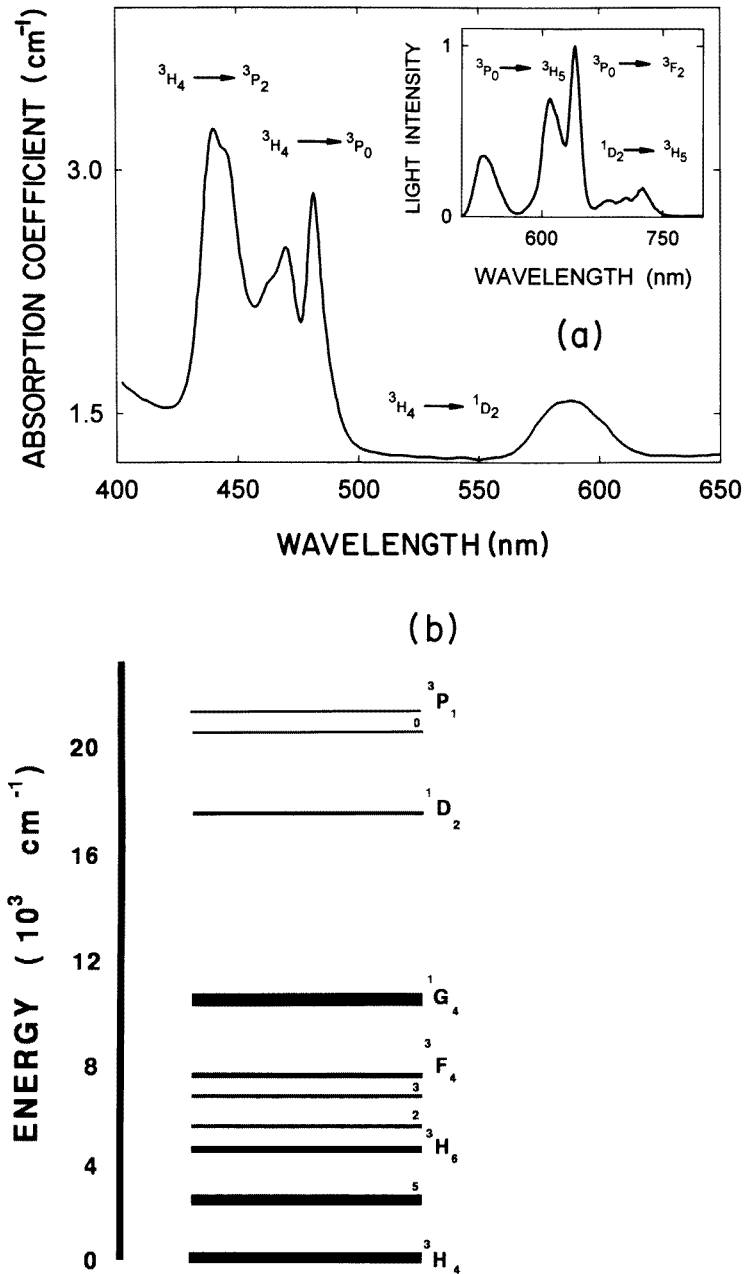


Figure 1. (a) Room temperature absorption spectrum of the Pr^{3+} -doped SiO_2 - CdO - ZnO glass sample. The inset shows the emission spectrum under continuous excitation at 488 nm. (b) Partial energy-level diagram for Pr^{3+} ions in oxide glasses.

provides corrected emission and excitation spectra. The light source was a 10 W pulsed xenon lamp with a width at half peak intensity of less than 0.01 ms. Lifetime measurements were obtained using 10 ns pulses from a EGG 2100 tunable DYE laser at 488 nm as

the excitation source. The sample fluorescence was monitored by a 0.45 m Czerni–Turner monochromator, detected by a cooled Hamamatsu R943-03 photomultiplier tube, and processed by a EGG/PAR 162 boxcar averager.

Thermal lensing experiments were performed following an experimental procedure similar to the one described by Durville and Powell [4]. The excitation source was a 5W-CW argon laser model Laser Ionics 1400. The different output lines of the laser allowed experiments to be performed in resonance or out of resonance with an electronic transition of the Pr³⁺ in our sample. A 10 mW He–Ne laser was used as a probe to detect changes in the index of refraction, induced by the excitation beam. The weak probe beam at this wavelength did not cause a perturbation of the refractive index.

The excitation and the probe beam laser beams were collinear and focused inside the sample with a 10 cm focal length lens. The thickness of the sample was 0.211 cm. The transmitted excitation beam was filtered out and the intensity, $I(t)$, at the centre of the probe beam was monitored using a pinhole and a Hamamatsu R943-03 photomultiplier tube. The signal was processed by using an EGG/PAR boxcar averager and readout on a strip-chart recorder.

3. Results and discussion

All measurements were carried out at room temperature (RT). In the visible range, the optical absorption and emission spectra of Pr in our glass sample were measured and are presented in figure 1. They arise from intraconfigurational transitions within the 4f² configuration.

The emission intensity at 527 nm (³P₀ → ³H₅) was monitored as a function of the time after the laser excitation, with 10 ns pulse width pulses at 488 nm. Lifetime measurements revealed that the decay scheme of the ³P₀ fluorescence consists of a pure exponential decay with an associated lifetime value of 1.27 μs. It is observed that this value is shorter than those previously reported for solids doped with low concentrations of Pr ions [5, 6]. However, it is quite similar to those measured for the praseodymium emission in heavily doped systems [6, 7]. Such results have been interpreted in terms of a resonant cross-relaxation process between Pr ions, and revealed the presence of a fast energy diffusion mechanism [7]. The effects of the experimental findings described above on the thermo-optical properties of our glass sample will be discussed below.

For thermal lensing studies, different output lines were chosen from the excitation source. Specifically, the 488 nm line was used to excite resonantly the ³H₄ → ³P₀ transition while the 514.5 nm line was used for off-resonance excitation of the glass. With the sample placed just before the beam-waist of the probe beam we observed an expansion of the transmitted probe beam profile when the excitation beam was turned on. This indicates that the excitation beam causes the sample to act as a positive lens [4].

Typical results for the time evolution of the intensity at the centre of the transmitted probe beam after turning on the excitation beam are shown in figure 2. At low input powers the decay curves for $I(t)$ show two well defined stages under on- and off-resonance excitation. In figure 2, the first stage (ST1) lasts a few seconds while the second stage (ST2) lasts several minutes before it reaches equilibrium. If the excitation beam is turned off after the equilibrium point, the signal does not return to its original level. This suggests the creation of a permanent change in the refractive index. It should be pointed out that, to our knowledge, in figure 2 the observation of permanent refractive index changes in bulk rare earth doped oxide glasses induced under off resonance laser excitation conditions is reported for the first time. On the other hand, using a 500 mW laser excitation at 488 nm the intensity $I(t)$ reaches equilibrium after a few seconds, and it returns to its original

value once the excitation beam is turned off (figure 2, inset). In this case, no evidence of a permanent change in the refractive index was observed.

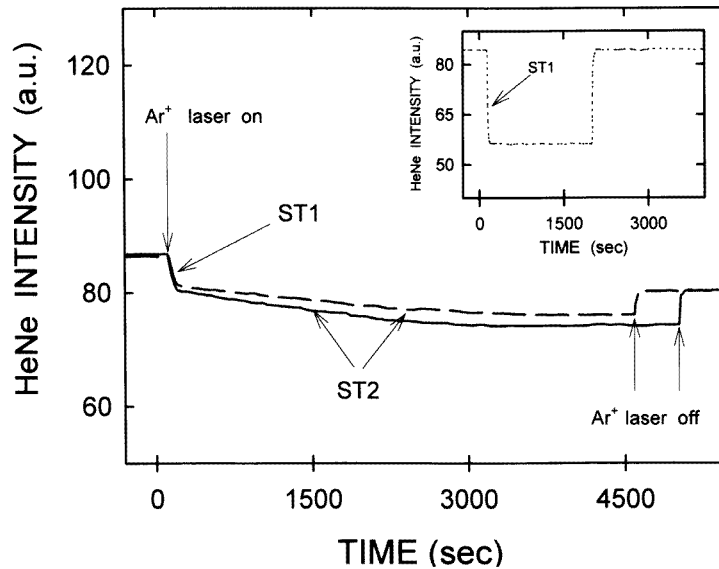


Figure 2. Time evolution of the intensity $I(t)$ at the centre of the transmitted probe beam under 50 mW-CW resonant laser excitation at 488 nm (—) and 500 mW-CW non-resonant laser excitation at 514 nm (- - -). The inset shows the time evolution of the intensity at the centre of the transmitted probe beam under 500 mW resonant laser excitation. In this case, no permanent refractive index changes are observed.

On the ST1 region the general characteristics and the response time observed in our experiments indicate that thermal lensing is the mechanism dominating the laser induced refractive index changes observed in our sample, at medium power levels (10^2 – 10^3 W cm $^{-2}$) [4]. Also, for our experimental configuration, the relative change in the intensity at the centre of the transmitted probe beam can be determined from ray analysis by using transfer matrices, under a gaussian-beam approximation [4, 8]. According to this model the ratio $I_0/I(t)$ is given by:

$$\frac{I_0}{I(t)} = 1 + \theta[1 + \tau_c/2t]^{-1} + \theta^2\{4[1 + \tau_c/2t]^2\}^{-1} \quad (1)$$

where $\tau_c = \omega^2/4D$, ω is the excitation beam radius inside the sample, and D is the thermal diffusivity, in units of cm 2 sec $^{-1}$. The parameter θ is a dimensionless collection of variables and can be viewed as characterizing the strength of the thermal lens [4]. A computer fit to the experimental data using equation (1) was obtained by treating θ and τ_c as adjustable parameters. Figure 3 shows a typical result of such a fitting. Good agreement is obtained between the theory and the experimental results; the values obtained for the adjustable parameters are listed in table 1.

From the data presented in this table it is observed that the difference in the mechanisms of generating the thermal effect for the resonant and non-resonant excitation is responsible for difference in the time constant τ_c and in the strength of the thermal lenses θ . For non-resonant excitation the observed thermal lensing is due to the absorption of light by the network of the glass host, and the time evolution is governed by the thermal properties of

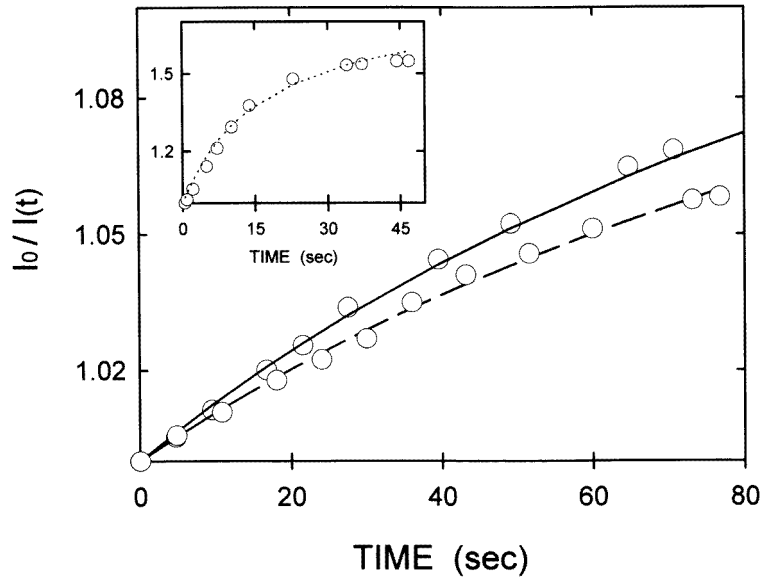


Figure 3. Theoretical fit of the time evolution of the signal in the Pr^{3+} doped glass sample: 50 mW input power at 488 nm (—), 500 mW input power at 514 nm (- - -) and 500 mW input power at 488 nm (· · · · ·) (inset). The circles are the experimental points.

Table 1.

λ_{ex} (nm)	P (mW)	α (cm^{-1})	τ_c (sec)	θ
488	50	1.5	292.7	0.2015
	500		28.75	0.6773
514	500	0.012	319.7	0.1815

the bulk sample. For the resonant excitation the temperature increases through radiationless relaxation processes that then transfer their energy to the network of the host. Under laser excitation at 488 nm, the fluorescence measurements show that the emission occurs mainly at 640 nm, from the ${}^3\text{P}_0 \rightarrow {}^3\text{F}_2$ transition (figure 1(a), inset). Non-radiative relaxation then occurs from the ${}^3\text{F}_2$ state to the ${}^3\text{H}_4$ ground state. In rare-earth (RE) doped glasses, radiationless relaxation transitions have been shown to be multiphonon emission processes, that generate several high-energy phonons [9, 10]. Since the thermal diffusion in oxide glasses is typically slow, the phonons generated by the RE remain localized creating a high level of vibrational energy around each RE ion. This can produce a high effective local temperature and high values for the strength of the TL effect, in good agreement with the data portrayed in table 1.

At variance with the above results, because of the permanent change in the refractive index observed in figure 2 requires more than 30 minutes to take place it is not a direct thermal effect, but is a combination between thermal expansion and the local temperature rise. However, it should be observed that at 500 mW input power at 488 nm (figure 2, inset) the redistribution of the input energy due to the energy migration observed between the Pr ions means that the strength of the thermal effect could be high enough to prevent any stress-optic permanent change in the refractive index. All these results must be combined

with previous observations on laser induced refractive index changes in RE doped oxide glasses [11–12], and considered by further investigations to develop an understanding on the active mechanism for laser induced effects in this kind of materials.

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