Time-Resolved Study of Thermal and Electronic Nonlinearities in Nd^{+3} and Cr^{+3} Doped Solids

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In ion doped solids there is a nonlinear refractive index n_2 which is due to the polarizability difference $\Delta\alpha$ between excited and ground states. Thermal Lens effects are also very significant. This work shows that using time resolved Z-scan technique we can distinguish electronic and thermal nonlinearities. We present the first measurements of the $\Delta\alpha$ of Nd^{+3} ion in fluoride glasses and Cr^{+3} doped fluoride crystals. For SrAlF5:Cr^{+3} we measured $n_2 = (6.6 + 1.7i) \times 10^{-11}\text{cm}^2$ and calculated $\Delta\alpha = 3.1 \times 10^{-26}$ cm^3 and $\Delta\sigma = 1.7 \times 10^{-20}$ cm^2. In InSBZnGdN was observed $\Delta\alpha = 1.6 \times 10^{-26}$ cm^3. For ZBLAN we could only estimate $\Delta\alpha < 2 \times 10^{-27}$ cm^3.

1. Introduction

The study of nonlinear properties is particularly important in laser active media because standing waves in laser cavities produce self-focusing, temporal and spatial self-phase modulation and light-induced gratings that cause effects of hole-burning [1]. Thermal effects, like the thermal variation of the refractive index, thermal expansion and thermally in induced stress are also very important in solid-state laser design [2]. The nonlinear properties of different ion doped solids have been studied using several techniques. Like in others materials, the Z-scan technique have been shown to be the best technique for the study of ion doped solids where the nonlinearity is usually slow ($> 10{-}4$ sec.) allowing the use of time-resolved methods [3-4]. In ion doped solids the nonlinearity originates from the population of dopante ion metastable state, which has a complex susceptibility different from that of ground state. The real part of nonlinear refractive index is proportional to the polarizability difference $\Delta\alpha$ between excited and ground states and the imaginary part is proportional to the absorption cross section difference $\Delta\sigma$ between excited and ground states. This is the Population Lens effect (PL). Most of these solids present nonlinearity whose real part is one order of magnitude greater than the imaginary one.

Usually, part of the excited state decay is nonradiative, so the laser heats the sample and an optical path change is established owing to the temperature coefficient of the optical path, ds/dT, which causes the so-called Thermal Lens (TL) [5]. In fluoride glasses, usually $\Delta\alpha$ is very small and it was observed that PL is smaller than TL effect [7]. In this work, we show that it is possible to temporally distinguish these two effects in Nd^{+3} and Cr^{+3} doped solids.

When the ion doped solid is pumped in resonance with an absorption line, both thermal and electronic nonlinearities are proportional to the ion excited state population $N_{ex}(t)$ which time evolution can be calculated using rate equations [6]:

$$N_{ex}(t) = N_0 \frac{(1-e^{-t/\tau})I/I_s}{(1+I/I_s)} \quad (1)$$

with

$$\tau^{-1} = \tau_0^{-1}(1+I_0/I_s) \quad (2)$$

where I is the laser intensity, N_0 is the total ion concentration, $I_s = \hbar\omega/\sigma\tau_0$ is the saturation intensity, $\hbar\omega$ is the pump photon energy, σ is the absorption cross section and to the lifetime. The complex refractive index can be written as $n(t) = n_0 + \Delta n(t)$, where Δn is the laser induced variation due to PL or TL effects. The intensity dependence of the sample trough an aperture in the far field is proportional to the phase shift $\Delta\phi = (2\pi/\lambda)\Delta n L$, where L is the sample thickness.

First we consider the TL effect, where the phase shift is given $\Delta\phi_{th}$:

$$\Delta\phi_{th} = \frac{PAL}{K\lambda_p}\varphi\frac{ds}{dT} \quad (3)$$

where P is the excitation laser power, A is the absorption coefficient, K is the thermal conductivity, λ_p is the probe beam wavelength, L is the sample length and φ is the fraction of absorbed energy converted into heat per photon. If the sample fluorescence quantum efficiency is η, λ_{ex} is the excitation bean wavelength, λ_{em} is the average emission wavelength, then $\varphi = (1-\eta\lambda_{em}/\lambda_{ex})$. In the case of the Nd^{+3} ion, $\lambda_{ex} \sim 0.5$ μm and $\lambda_{em} \sim 1.06$ μm and h $\eta \sim 1$ so $\varphi \sim 0.5$ [8-9]. TL signal response time is given by:

$$t_c = w^2/4D \quad (4)$$

where $D = K/\rho C$ is the thermal diffusivity, ρ is the density, C is the specific heat and w is the excitation beam radius.

The PL refractive index variation is given by $\Delta n_p = n_2 I$, then the PL phase shift can written as:

$$\Delta\phi_{pop} = (4\pi^2 f_L^2/\lambda n_0)\Delta\alpha L N_0 I_0/I_s \quad (5)$$

where $f_L = (n_0^2+2)/3$ is the Lorenz local field correction factor, n_0 is the real part of the linear refractive index, $\Delta\alpha$ is the polarizability difference between excited and ground states of the dopant ion, $I_0 = 2P/\pi w_0^2$, is the on axis intensity of the gaussian TEM_{00} profile and w_0 is the beam radius at focus.

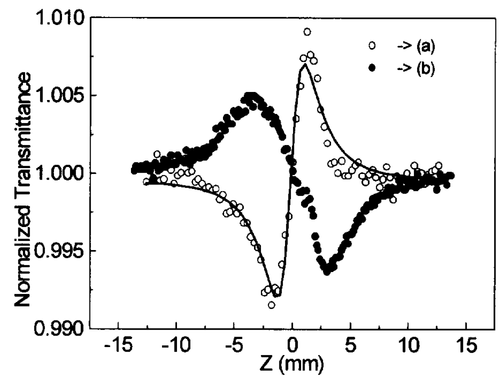

Figure 1. Time-resolved Z-scan of Nd^{+3} doped fluorindate glass where curve (a) shows data for chopper frequency f = 840 Hz and laser power P= 0.187 W and curve (b) shows data for f = 90Hz, P = 0.23W.

The Fig.1 shows our time-resolved Z- scan data performed at two different chopper frequencies in order to discriminate PL and TL effects in Nd^{+3} doped InSbZnGdN fluorindate glasses [7]. In this experiment the PL effect is faster than TL, so at high chopper frequency the characteristic Z-scan curve for positive nonlinearity was observed and attributed to PL effect. The PL curve shown is already normalized by the open aperture Z-scan (S_1=100%) as usually done in this technique. The open aperture data (S_1=100%) indicates a small absorption, so the complex n_2 is (1.1- 0.05i)10^{-10} cm^2/W. At lower chopper frequency the curve is inverted indicating $\Delta n < 0$ and consequently $ds/dT < 0$, as usually observed in fluoride glasses. The distance between peak and valley is doubled ($\Delta T_{pv} \sim 3.4 z_0$) as expected for a TL Z-scan[7]. From our PL data, we calculated $\Delta\alpha \sim 1.6^{-26}$ cm^3.

For ZBLAN and in both chopper frequencies we always observed $\Delta n < 0$. This indicates that either $n_2 < 0$ or the PL effect is too small compared to the TL one. Therefore we could only estimate $\Delta\alpha < 2 \times 10^{-27} cm^3$. In the sample YAG we obtained $n_2 = 1.4 \times 10^{-10}$ cm^2/W and $\Delta\alpha = 4.1 \times 10^{-26}$ cm^3.

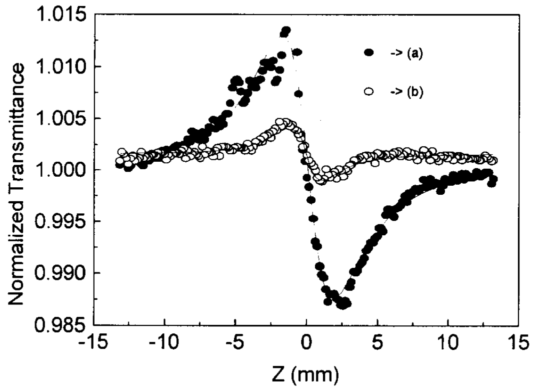

Figure 2. Normalized Transmittance obtained in ZBLAN sample. The curve (a) shows data for f = 186Hz and curve (b) shows data for f = 824Hz. Both curves shows $n_2 < 0$.

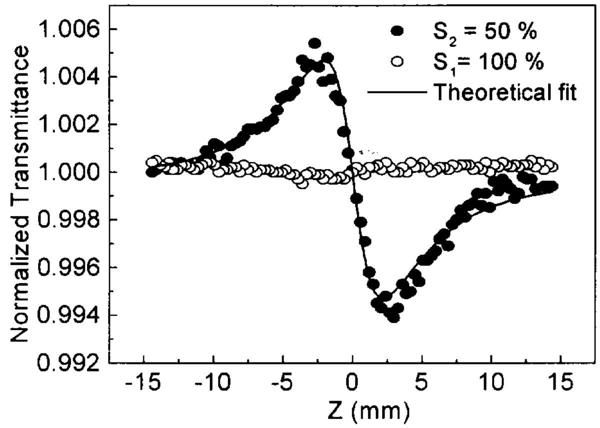

Figure 4. Normalized Transmittance obtained in $SrAlF_5Cr^{+3}$ sample with P = 0.175W and f = 186 Hz.

The Fig.3 shows our results open (S_1) and closed-aperture (S_2) data, to $SrAlF_5$:Cr^{+3} at high frequency. The open-aperture data present a decrease in transmittance near the focus due to a excited state absorption. The division curve indicates a positive nonlinearity with $n_2 = 6.6 \times 10^{-11}$ cm^2/W and using Eq. (5) we calculated $\Delta\alpha = 3.1 \times 10^{-26}$ cm^3. The Fig.4 shows our Z-scan data lower frequency. In this case, the closed aperture curve indicate a negative nonlinearity, which was attributed to TL effect. This curve can be fit with theoretical thermal lens model [5] where we obtained tc which is used to calculate the thermal diffusivity $D = 6.5 \times 10^{-3}$ cm^2/s.

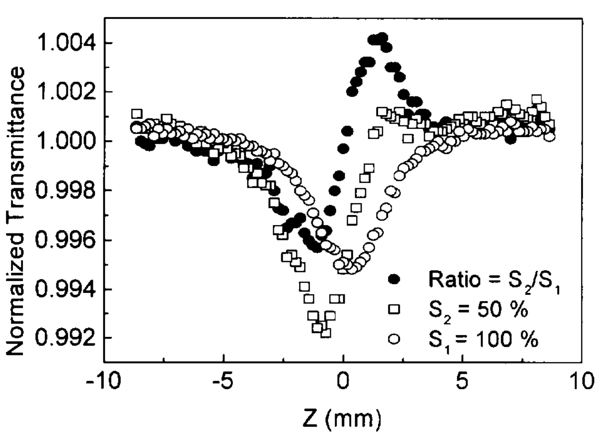

Figure 3. Normalized Transmittance obtained in $SrAlF_5$:Cr^{+3} sample for laser power P = 0.09W and frequency f = 822 Hz.

In this work, we have shows that even when $\Delta\phi_{th} > \Delta\phi_{pop}$ these two effects can be distinguished temporally by measuring with appropriate chopper frequencies. The same procedure has been used before to distinguish the nonlinear effect of the two different sites of Cr^{+3} in alexandirte [6].

Acknowledgments

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References

1. H. J. Eichler, P. Gunter, and D. W. Pohl, *Laser-Induced Dynamic Gratings*, vol. 50 of Spring Series in Optical Sciences. Spring-Verlag, Berlin, (1986).
2. W. Koechner, "Solid-State Laser Engineering". Spring-Verlag, New York, (1988).
3. M. Sheik-Bahae, A. A. Said and E. W. Van Stryland, "High sensitivity, single beam n2 measurements", Opt. Lett. **14**, 955 (1989).
4. L. C. Oliveira and S. C. Zilio, "Single-beam time-resolved Z-scan measurements of slow absorbers" Appl. Phys. Lett. **65**, 1 (1994); L. C. Oliveira, T. Catunda and S. C. Zilio, "Saturation Effects in Z-scan Measurements". Jpn. J. Appl. Phys. **35**, 2649 (1996).
5. M. L. Baesso, J. Shen and R. D. Snook, "Mode Mismatched Thermal Lens Determination of Temperature Coefficient of optical path Length in Soda Lime Glass at Different Wavelenghts". J. Appl. Phys. **75**, 3732 (1994).

6. V. Pilla, P. R. Impinnisi and T. Catunda, "Measurement of saturation intensities in ion doped solids by transient nonlinear refraction". Appl. Phys. Lett, **70**, 817 (1997).
7. T. Catunda; M. L. Baesso, M. L. Y. Messaddeq; M. Aegerter, "Time-resolved Z-scan and thermal lens measurements in Er^{+3} and Nd^{+3} doped glasses", J. Non-Cryst. Solids, **213&214**, 225 (1977). A. A. Andrade, T. Catunda, R. Lebullenger, A.C. Hernandes and M. L. Baesso, "Time-resolved study of thermal and electronic nonlinearities in Nd^{+3} doped fluoride glasses glasses". Eletronics Letter, **38**, 117 (1998).
8. M. L. Baesso, A. C. Bento, A. A. Andrade, J. A. Sampaio, E. Pecoraro, L. A. O. Nunes and T. Catunda, "Absolute thermal lens method to determine fluorescence quantum efficiency and concentration quenching of solids". **57**, 10545 (1998).
9. M. L. Baesso, A. C. Bento, A. A. Andrade, T. Catunda, J. A. Sampaio and S. Gama, "Neodymium concentration dependence of thermo-optical properties in low silica calcium aliminate glasses". **219**, 165 (1997).