Time-resolved Z-scan measurements in Nd^{3+} doped YAG

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Abstract

In this work we used the time-resolved Z-scan technique to measure the nonlinear refractive index, n_2, in Nd^{3+}:YAG. We also studied the energy-transfer upconversion (ETU) or Auger upconversion (γ), by monitoring the behavior of the population excited state. It is known that this energy-transfer can be a detrimental effect on laser operation.

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. The Z-scan is nowadays the most popular technique to investigate the nonlinear refractive index n_2 of different materials. This technique have been shown to be the best technique for the study of ion doped solids where the nonlinearity is usually slow ($>10^{-4}$s) allowing the use of time-resolved methods [3, 4, 5, 6]. 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 non-radiative, 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) [7].

In this work, nonliear refractive index, n_2, measurement of Nd^{3+} doped YAG using the time-resolved Z-scan technique is presented. In previous works we showed that in some cases, the time-resolved Z-scan may be used to temporally discriminate the PL and TL non-linearities [8, 9], so that the value of $\Delta\alpha$ can be obtained.

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 [10]:

$$N_{ex}(t) = N_o \frac{(1-e^{-t/\tau_0})S}{(1+S)} \tag{1}$$

where $S=I/I_s$ is the saturation parameter, I the laser intensity, $I_s = h\nu/\sigma\tau_o$ the saturation intensity, $h\nu$ the pump photon energy, σ the absorption cross section, τ_o the excited state lifetime and N_o the total ion concentration. The complex refractive index can be written as $n(t) = n_o + \Delta n(t)$, where Δn is the laser induced variation due to PL or TL effects. The beam intensity dependence trough an aperture in the far field is proportional to the phase shift $\Delta\phi = (2\pi/\lambda)\Delta nL$, where L is the sample thickness.

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

$$\Delta\phi_{pop} = (4\pi^2 f_L^2/\lambda n_o)\,\Delta\alpha L N_o I_o/I_s \tag{2}$$

where $f_L = (n_o^2 + 2)/3$ 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, $I_o = 2P/\pi w_o^2$, is the on axis intensity of the gaussian TEM_{oo} profile and w_o is the beam radius at focus.

Results and Discussions

The experimental was performed in resonance with ${}^2G_{9/2} + {}^4G_{7/2}$ (λ = 514nm) and ${}^4F_{5/2}$ (~ 810 nm) Nd^{3+} absorption lines. However, at λ = 514 nm the absorption is relatively weak and might have a contribution due to impurities present at host crystal. This would lead to an overestimation of the ion absorption cross section σ that would change the values of I_s and $\Delta\sigma$. Therefore we decided to do measurements with a tunable Ti-sapphire laser at λ = 810nm in resonance with the $^4F_{5/2}$ Nd^{+3} level. Details of experimental set up can be found in [6, 10].

Figure 1 shows single beam time-resolved Z-scan measurements. The measurements were made with a aperture factor $S_1 = 0.5$ and $S_2 = 1.0$ as defined in [4]. The Z-scan experiment was performed at a higher chopper frequency (f=840 Hz) to resolve the PL from the TL effects in Nd^{3+} doped solids [8, 9]. At higher chopper frequency the characteristic Z-scan curve present distance between peak and valley $\Delta Z_{pv} \sim 1.7z_0$ [3]. Beside, at a lower chopper frequency the distance between peak and valley is $\Delta T_{pv} \sim 3.4z_0$, which is two times larger than the electronic non-linearity [4, 11]. This increase in ΔZ_{pv} is a consequence of the larger refractive index profile $\Delta n(r)$, in the TL due to heat diffusion. Ours results indicates that in this regime the PL contribution to the nonlinearity is the dominant effect.

From open aperture ($S_2 = 1.0$) we can obtain the imaginary part of the non-linear refractive index, n_2". The transmittance increase at the focus indicates that the absorption decreases when the excited state is populated ($\Delta\sigma < 0$), so the material is a saturable absorber as shown in Fig. 1(b). Since the magnitude of the nonlinear absorption was significant, we had to normalize the closed-aperture signal ($S_1 = 0.5$), dividing it by the S_2 data, as is typically done in the Z-scan technique [4]. Figure 1(c) shows this ratio curve that was fitted by the proper expression. In this measurement, the axial peak intensity is I_0 = 3.9 KW/cm^2, which is smaller than the saturation intensity I_s = 17 KW/cm^2, obtained by $I_s = h\nu/\sigma\tau_o$, so saturation effects could be neglected. By fitting the experiments data, we obtained $n_2 = (5.6+2.2i)\text{x}10^{-9}$ cm^2/W, $\Delta\alpha = (6.2\pm0.2)\text{x}10^{-26}$ cm^3 and $\Delta\sigma = -5.1\text{x}10^{-20}$ cm^2.

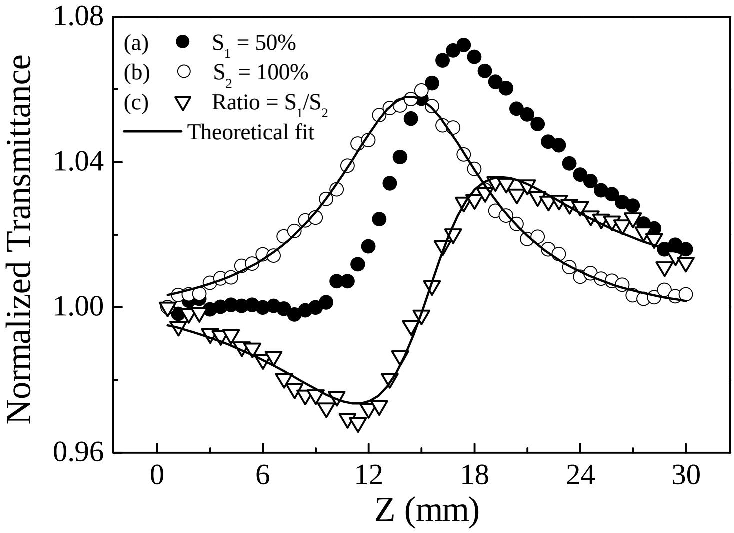

Figure1 – Z-scan data in YAG:Nd^{3+} at λ = 810 nm. $N_0 = 1.03 \text{ x } 10^{20}$ cm^{-3} and L =2 mm.

We also realized the measurement with open aperture (S_2) in different excitation beam power as is shown Fig. 2. It show the normalized transmittance variation, ΔT_{pv}, versus $S = I/I_s$. This curve possibility us to determine the Auger upconversion parameter (γ) or energy-transfer upconversion (ETU), which in solid materials can be a detrimental effect that depopulate the excited state. ETU or Auger upconversion, is an energy transfer process involving two excited (metastable) ions, where one ion is promoted to higher excited while the other ion is transferred to the ground state. In this paper, a theoretical model that considers the parameter γ was developed. In this case the normalized transmittance variation ΔT can be written as:

$$\Delta T = \left[\frac{-(1+S)+\sqrt{(1+S)^2+4\beta S}}{2\beta}\right]\Delta\sigma L N_0 \qquad (3)$$

where $\beta = N_0\gamma\tau_0$. By fitting the experimental date with Eq. (3) we obtained $\beta = (5.7\pm 1.1)$ so that $\gamma = (2.4\pm 0.5)\text{x}10^{-16}$ cm^3/s. This results is in good agreement with the value found in literature $\gamma = 2.8\text{x}10^{-16}$ cm^3/s [12]. The Fig. 2 shows also the simulation where we had consider $\gamma = 0$. We can observe that the γ contribute strongly for the depopulation of excited state.

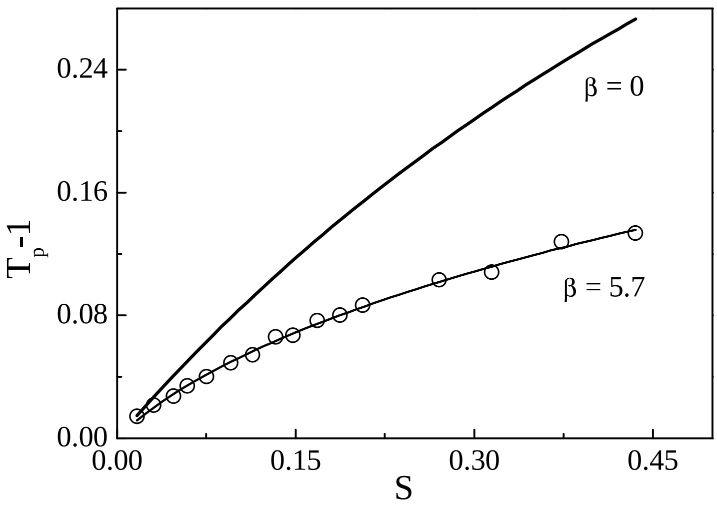

Figure2 – Amplitude of open aperture versus $S=I/I_s$.

Conclusions

In conclusion, we have determined the refractive index, n_2, in Nd^{3+}:YAG by the time-resolved Z-scan technique and we also demonstrated that the time-resolved Z-scan technique can be used to studied the ETU or Auger upconversion (γ), by monitoring the behavior of the population excited state. The results obtained are in good agreement with the results found in literature, for both $\Delta\alpha$ [13] and γ [12].

Acknowledgements

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