

# Nonlinear electronic line shape determination in Yb<sup>3+</sup>-doped phosphate glass

D. N. Messias and T. Catunda

*Instituto de Física de São Carlos, Universidade de São Paulo, USP, C.P. 369, CEP 13560-970, São Carlos, São Paulo, Brazil*

J. D. Myers and M. J. Myers

*Kigre Inc., 100 Marshland Road, Hilton Head Island, South Carolina 29926, USA*

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Nonlinear refraction spectroscopy has been performed in Yb<sup>3+</sup>-doped phosphate glass to determine the line shape of real and imaginary parts of  $n_2$  ( $n_2'$  and  $n_2''$ ). The  $n_2'$  spectrum presented an asymmetric feature due to the interference of resonant and nonresonant contributions, where the nonresonant term arises from the polarizability difference between excited and ground states ( $\Delta\alpha$ ). The measurements were performed in the transient regime to determine population dynamics and the pump saturation intensity at 975 nm (peak of the absorption spectrum). Because of the small quantum defect of Yb<sup>3+</sup>, we estimated that the magnitude of the thermal lens effect is  $\sim 20$  times smaller than the population lens effect, caused by  $n_2$ . © 2007 Optical Society of America

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With the advent of high-power diode lasers an increasing interest in Yb<sup>3+</sup>-doped systems has appeared. Since Yb<sup>3+</sup> has only one excited state in the visible–infrared range, a high doping concentration can be used without the usual care about concentration quenching. Yb<sup>3+</sup> is also attractive because of its relatively long lifetime ( $\sim 2$  ms). Moreover, the Yb<sup>3+</sup> emission wavelength is close to the pump diode wavelength, resulting in a thermal load lower than 10%. In addition to the thermal lens (TL) effect, it is well known that ion-doped solids present an electronic refractive index change due to the population of the excited metastable state, usually the upper laser level.<sup>1,2</sup> This effect is usually called a population lens (PL) and has been studied mostly in Nd<sup>3+</sup>- and Cr<sup>3+</sup>-doped materials.<sup>3,4</sup>

A complete picture of the refractive index change in Yb<sup>3+</sup>-doped materials is of primary importance to laser design. In this work we have performed Z-scan measurements in a Yb<sup>3+</sup>-doped phosphate glass, QX/Yb, to determine the line shape of the nonlinear refractive index,  $n_2$ . The experiments were performed with chopped cw lasers to investigate the transient behavior of the refractive index change in the milliseconds range.

In media with slow nonlinearity (response time  $\geq 100 \mu\text{s}$ ) the time-resolved Z-scan method is the most appropriate technique used for the study of population redistribution nonlinearity. It is very sensitive, simple, and allows the investigation of the temporal evolution of  $\Delta n$  and the elimination of spurious linear effects.<sup>5</sup> In this technique a cw laser is chopped to normalize the signal by making  $I(z, t)/I(z, 0)$  for each position of the Z-scan curve. As is usually done in the Z-scan method, after its passage through the sample the beam was split to allow the opened- (absorptive) and closed- (refractive) aper-

ture signals to be detected simultaneously, with aperture factors  $S_1=100\%$  and  $S_2=50\%$ , respectively.<sup>6</sup> In this work we studied a 2 mm thick,  $5.8 \times 10^{20} \text{ cm}^{-3}$  Yb<sup>3+</sup>-doped phosphate glass sample (QX/Yb) from Kigre Inc. The experiment was performed by using a cw Ti-sapphire laser pumped by an Ar<sup>+</sup> laser. First, the complex  $n_2$  (real and imaginary parts) spectra were obtained by repeating the measurement at several wavelengths, as shown in Fig. 1. Following the Z-scan recipe, the effect of nonlinear absorption was taken into account by the ratio  $S_2/S_1$  (more detail can be found in Ref. 4). A pronounced peak was observed at  $\lambda \sim 975$  nm, where  $n_2 = (2.5 + 2.3i) \times 10^{-8} \text{ cm}^2/\text{W}$  ( $n_2 = n_2' - in_2''$ ).

To study the  $n_2$  line shape, the measurements were then performed in a spectroscopic way.<sup>4</sup> The sample was fixed at the peak position of the Z-scan curve and the laser was continuously tuned from 967 to 983 nm across the absorption peak. As expected,  $n_2''$  is positive, which indicates a saturable absorption nonlinearity. The  $n_2'$  line shape is similar to the  $n_2''$ , but with a small blueshift. This behavior indicates that the po-

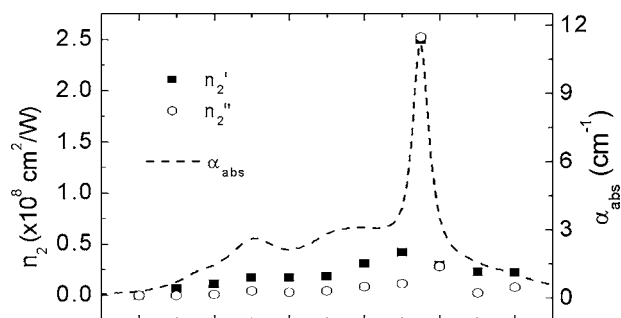


Fig. 1. Wavelength dependence of the complex nonlinear refractive index,  $n_2 = n_2' - in_2''$ , in the absorption range of Yb<sup>3+</sup>-doped glass. For comparison, the linear absorption spectrum is also shown.

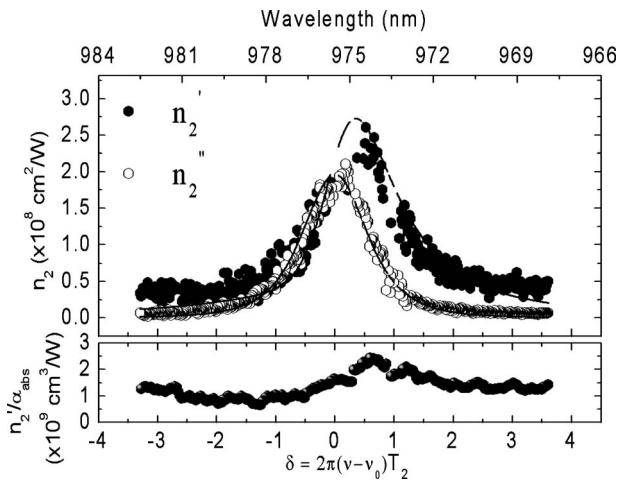


Fig. 2. Nonlinear refractive index line shape around the absorption peak. The filled and empty circles denote the real and imaginary wavelength behavior of  $n_2$ , respectively. The continuous lines represent the theoretical fit by Eq. (1). The dispersive character of  $n_2'/\alpha_{\text{abs}}$  is shown at the bottom.

larizability difference between excited and ground states,  $\Delta\alpha$ , gives the main contribution to  $n_2'$ . In fact, the  $n_2'$  spectrum shown in Fig. 2 is similar to the one observed in  $\text{Cr}^{3+}$ -doped crystals, ruby and alexandrite.<sup>4</sup>

The main features observed in this experiment can be explained by considering that the susceptibility is given by the effect of  $\Delta\alpha$  and the resonant interaction as shown in Ref. 4. In that work an expression for  $n_2$  was developed, so that

$$n_2 = \frac{\alpha_{\text{abs}}(0) i + \delta + A(1 + \delta^2)}{2kI_s (1 + \delta^2)^2}, \quad (1)$$

where  $\alpha_{\text{abs}}(0)$  is the line-center intensity absorption coefficient,  $\delta = 2\pi(\nu - \nu_0)T_2$  is the detuning parameter,  $I_s$  is the saturation intensity,  $k = 2\pi(\nu_0/c)$ ,  $A = 4\pi^2 f_L^2 \Delta\alpha / \lambda n_0 \sigma_g$ ,  $f_L = (n_0^2 + 2)/3$  is the Lorentz local-field correction factor, and  $n_0$  is the linear refractive index. The refractive index change associated with the population redistribution between excited and ground states is given by the factor  $A$  in Eq. (1). Since the polarizabilities of these levels are caused by the nonresonant interaction with all other high-lying levels of the system,  $A$  represents a nonresonant contribution to  $n_2$ . When  $A = 0$ , the standard expression for the resonant two-level system nonlinearity is recovered. Actually, Eq. (1) can be obtained from a more general expression derived by Butylkin *et al.*,<sup>7</sup> considering that the excited state lifetime  $\tau \gg T_2$ . This limit is well satisfied for ion-doped solids where, at room temperature,  $\tau \sim \text{ms}$  and  $T_2 \sim \text{ps}$ .

The  $n_2'$  line shape is strongly determined by the  $A$  parameter, which represents the balance between nonresonant and resonant contributions. For  $A \ll 1$ ,  $n_2'(\delta)$  presents a dispersive character as observed in diluted atomic vapors. On the other hand, when  $A \gg 1$  the  $\Delta\alpha$  effect is dominant, so  $n_2'(\delta)$  roughly follows the Lorentzian absorption spectrum,  $\alpha_{\text{abs}}(\delta) = \alpha_{\text{abs}}(0)(1 + \delta^2)^{-1}$ . The  $A$  parameter can be easily ob-

tained from the experimental data by noticing that  $A = n_2'(0)/n_2''(0)$ . Therefore, for the  $n_2$  value at 975 nm,  $A \approx 1.1$  was obtained, which is in good agreement with  $A \approx 0.8$  by the fit of the  $n_2'(\delta)$  line shown in Fig. 2. Notice the small blueshift of the  $n_2'(\delta)$  spectrum relative to the  $n_2''(\delta)$ . This behavior is predicted by Eq. (1) for  $A \sim 1-5$  and was also observed in the alexandrite  $R_{1m}$  line, where  $A \sim 1.7$ .<sup>4</sup> Figure 2 also shows that the curve of  $n_2'(\delta)$  normalized by the absorption spectrum  $\alpha_{\text{abs}}(\delta)$  has a dispersive character. This behavior is in accordance to Eq. (1), from which  $n_2'/\alpha_{\text{abs}} \propto [A + \delta/(1 + \delta^2)]$ . Since at low intensity  $N_{\text{ex}} \propto I/I_s \propto \alpha_{\text{abs}}(\delta)$ , then the parameter  $n_2'/\alpha_{\text{abs}}$  is proportional to  $\Delta n/N_{\text{ex}}$  or the polarizability difference  $\Delta\alpha$ .

In the theoretical model that resulted in Eq. (1), the resonant interaction was represented by a two-level system. The same expression also represents the three-level system case, where the excited state decays to a metastable level. In fact,  $\text{Yb}^{3+}$  has two  $4f$  states ( ${}^2F_{7/2}$  and  ${}^2F_{5/2}$ ) that are split by the crystal-line field. Moreover, this structure is masked by the inhomogeneous broadening, which is comparable with the Stark splitting. The ground state is a quartet, while the excited state is a triplet, and these sub-levels are separated typically by  $\sim 200 \text{ cm}^{-1}$ , so they are thermally coupled.<sup>8</sup> The absorption spectrum has a pedestal shape with a peak at 975 nm, which corresponds to the transition from the lowest sublevel of the ground state manifold ( ${}^2F_{7/2}$ ) to lowest sublevel of the upper state manifold ( ${}^2F_{5/2}$ ). Figures 1 and 2 show that  $n_2''$  vanishes far from this peak (for  $|\delta| > 1$ ), which justifies our assumption of single resonance as a first approximation to this problem.

We investigate the behavior of the absorption saturation to test whether the population dynamics is adequately described by the simplified theoretical model [Eq. (1)]. The inset in Fig. 3 shows a typical open-aperture transient signal ( $S_1 = 100\%$ ), with the sample at the focus position ( $z \sim 0$ ). From simple rate equation analysis, for  $I \ll I_s$ , the excited state popu-

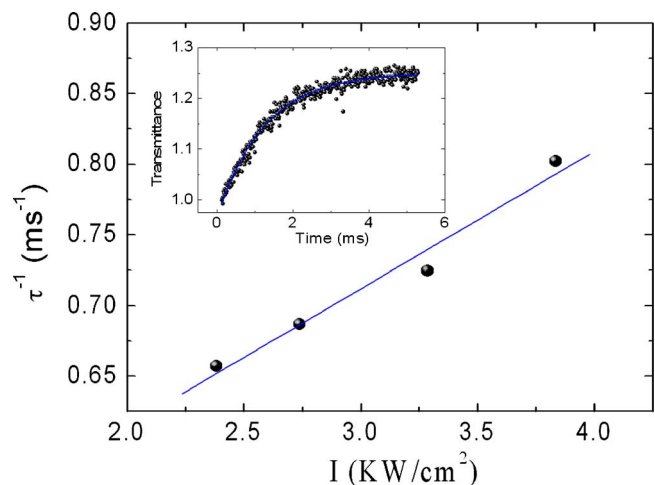


Fig. 3. (Color online) Intensity ( $I$ ) dependence of the nonlinear absorption response time ( $\tau$ ). The curve of  $\tau^{-1}$  versus  $I$  was fitted by a straight line. The inset shows a typical transient signal adjusted by an exponential to obtain  $\tau$ .

lation is expected to evolve as  $N_{\text{ex}}(t) \sim N_t [1 - \exp(-t/\tau)] I/I_S$  with  $\tau^{-1} = \tau_0^{-1} (1 + I/I_S)$ , where  $\tau_0$  is the exponential fluorescence decay time and  $N_t$  is the total ion concentration.<sup>1</sup> Several transient measurements were performed as a function of pump intensity, with the sample at a fixed position. All transient signals were well fitted by an exponential, and a linear behavior of  $\tau^{-1}$  versus  $I$  was observed as shown in Fig. 3. The values  $\tau_0 = 2.4$  ms and  $I_S = 4.3$  KW/cm<sup>2</sup> were obtained from the fit. This  $\tau_0$  value compares well with  $\tau_{\text{exp}} = 2.3$  ms, obtained from a direct fluorescence decay measurement. Alternatively,  $n_2''$  is related to absorption saturation by  $\alpha_{\text{abs}} = -2kn_2''I_S$ , since Yb<sup>3+</sup> has no excited state absorption in the infrared range. Therefore, from the  $n_2''$  value at 975 nm (line-center wavelength)  $I_S = 3.8$  KW/cm<sup>2</sup> is obtained. This result is in good agreement with the value 4.3 KW/cm<sup>2</sup> obtained from transient measurements (Fig. 3). These  $I_S$  values are about the double of the theoretical two-level system value  $I_S = h\nu/2\sigma_g\tau = 2.3$  KW/cm<sup>2</sup>. This discrepancy indicates that the system is better modeled by a four-level system, for which Eq. (1) is valid with  $I_S = h\nu/\sigma_g\tau$ . A similar behavior was previously observed in Cr<sup>3+</sup>-doped crystals.<sup>1</sup>

The microscopic parameter,  $\Delta\alpha = 2.3 \times 10^{-26}$  cm<sup>3</sup>, could be obtained assuming that at line center  $n_2' I = (2\pi/n_0) f_L^2 N_{\text{ex}} \Delta\alpha$  and  $N_{\text{ex}} \sim I/I_S N_t$  with  $I_S = h\nu/\sigma_g\tau$ . This  $\Delta\alpha$  value is comparable with 1.9, recently obtained for Yb<sup>3+</sup>:YAG,<sup>9</sup> and 2.8, obtained for a Yb<sup>3+</sup>-doped silica fiber<sup>10</sup> (all  $\Delta\alpha$  values in 10<sup>-26</sup> cm<sup>3</sup>). Similarly, nearly constant  $\Delta\alpha(\lambda)$  was obtained by using  $n_2'(\lambda)$  data, which corroborates the nonresonant character of  $\Delta\alpha$  expected by our model. The absorption cross-section difference between excited and ground states was found to be  $\Delta\sigma = -2.2 \times 10^{-20}$  cm<sup>2</sup>. As there is no excited state absorption, this value is close (in modulus) to the ground state absorption cross section,  $\sigma_g = 1.9 \times 10^{-20}$  cm<sup>2</sup>.

Finally, electronic and thermal contributions to the refractive index change,  $\Delta n$ , coexist and usually have comparable strengths.<sup>11</sup> To estimate the relative magnitude of these effects, one can take the ratio between its induced phase shifts,  $|\Delta\phi_{\text{pop}}/\Delta\phi_{\text{th}}| \propto \varphi\rho C_p \Delta\alpha$ , where  $\varphi$  is the fraction of absorbed energy converted into heat,  $\rho$  is the density, and  $C_p$  the specific heat.<sup>3</sup> For pulsed excitation (disregarding heat diffusion) we have obtained  $|\Delta\phi_{\text{pop}}/\Delta\phi_{\text{th}}| \sim 20$ . This value is  $\sim 40$  times larger than those calculated by Powell *et al.*<sup>3</sup> for Nd<sup>3+</sup>-doped flashlamp-pumped amplifiers, where  $\varphi \sim 0.5$ .<sup>3</sup> The main difference is due to

the very small quantum defect of Yb<sup>3+</sup> ions ( $\sim 3\%$ ). In the cw regime, it can be shown that  $|\Delta\phi_{\text{pop}}/\Delta\phi_{\text{th}}|_{\text{cw}} \sim (\tau/t_c) |\Delta\phi_{\text{pop}}/\Delta\phi_{\text{th}}|_{\text{pul}}$ , where  $t_c$  is the characteristic TL response time.<sup>11</sup> In this experiment  $t_c \sim 2$  ms  $\sim \tau$ , so the TL contribution to the refractive index change is negligible, as observed in the transient measurements, where the data was very well fitted by considering only the PL effect. TL measurements performed by using another experimental setup confirmed these estimations.<sup>12</sup>

In conclusion, using a variation of the Z-scan technique, we obtained the complex  $n_2$  spectrum of Yb<sup>3+</sup>-doped phosphate glass. We observed a population dynamics well described by a four-level system, with pump saturation intensity  $I_S \sim 4$  KW/cm<sup>2</sup>. Although the PL effect is usually neglected in laser engineering, we demonstrated that it makes the dominant contribution to the refractive index in Yb<sup>3+</sup> because of its very small thermal load.

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