Refractive index changes in solid state laser materials

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A few years after the invention of the ruby laser it became clear that laser-induced refractive index changes (RIC) have a major impact in laser behavior. In fact, all the main laser features (monochromaticity, directionality, coherence and brightness) are affected by RIC [1,2]. The propagation of intense optical beams through dielectric media induces RIC which cause self-focusing, beam breakup and damage in high power pulsed laser systems. For a laser beam of intensity, I, the fast host nonlinearity (crystal or glass) is important in high power short pulsed lasers, causing RIC with $\Delta n = \gamma I$ and $\gamma \sim 10^{-16}$ cm^{2}W^{-1}. Besides this effect, very important slow ($\sim 10^{-4}$sec) thermal and electronic RIC appears due to the pumping of the upper laser level which, for Solid-State Lasers (**SSL**) is usually the metastable state of a doping ion . For instance, in ruby and glass lasers it was soon realized that optical distortions caused by heat deposition could virtually preclude diffraction-limited laser operation. Moreover, athermal RIC associated with the excited state population of dopand ions (Cr^{3+}, Ti^{3+}, Nd^{3+}, Yb^{3+}, Tb^{3+}, etc.) usually have magnitude comparable to the thermal ones. This electronic contribution to the RIC arises from the fact the polarizability of the ion in its excited state (α_{pex}) is different from its value in the ground state (α_{pg}), so this RIC is proportional to $\Delta\alpha_p = (\alpha_{pex} - \alpha_{pg})$. The importance of $\Delta\alpha_p$ also appeared to explain frequency drifts as well as transverse or lensing effects observed in many SSL lasers. In fact, this lens like behavior determined the names Population Lens (PL) and Thermal Lens (**TL**) related to electronic (athermal) and thermal effects, respectively.

The refractive index change associated with the PL effect of an ion doped material is given by:

$$\Delta n_{PL} = \frac{2\pi}{n} N_{ex} f_L^2 \Delta\alpha_p \qquad \text{Eqn. 1}$$

where $f_L = (n_o^2 + 2)/3$ is the Lorenz local field correction factor, N_{ex} is the excited state population. We assumed that the system has only one excited metastable state, so the total ion concentration is given by $N_t \sim N_{ex} + N_g$, N_g being the ground state population. Let´s consider the case of ruby (Al_2O_3:Cr^{3+}) where $\Delta\alpha_p = 1.8\ 10^{-25}$ cm^{-3} with typical Cr^{3+} concentration $N_t \sim 1.6\ 10^{19}$ cm^{-3}. According to eq.(1) the maximum RIC can be estimated considering $N_{ex} \sim N_t$ so $\Delta n_{max} \sim 3\ 10^{-5}$. Analogously for a typical Nd:YAG crystal $N_t \sim 1.4\ 10^{20}$ cm^{-3}, $\Delta\alpha_p = 5\ 10^{-25}$ cm^{-3} and $\Delta n_{max} \sim 9\ 10^{-5}$. Although these Δn_{max} are not large, they are comparable to RIC due to TL effect or Δn of Fiber Braag Gratings, consequently they have important consequences in laser behavior.

For rare earth (RE) and transition metals doped materials, $\Delta\alpha_p$ is mainly accounted by transitions in the UV. Although far from resonance for visible light, these UV transitions have high oscillator strength,

typically 4 orders of magnitude greater than the forbidden transitions in the visible. In the case of rare earth doped materials, it is generally accepted that the 4f → 5d dipole allowed UV transitions in the should give the main contribution to the ion polarizability since their oscillator strength are typically 3 - 4 orders of magnitude higher than the 4f → 4f. Moreover, it has been observed in Cr^{3+}, Nd^{3+}, Yb^{3+} and Tb^{3+} doped materials that $\Delta\alpha_p$ varies strongly with the host character (~1 order of magnitude): fluorides < phosphates < silicates < oxides. In the case of RE ions, this observation is in qualitative agreement with higher energy position of the 5d levels of fluorides compared to other hosts. Fig.1.(a) show some data of Cr^{3+} doped oxide crystals where $\Delta\alpha_p$ decreases monotonically with the parameter ΔE, the energy difference between $^4T_2$ and ^2E levels of Cr^{3+} [3]. Not shown in Fig.1(a) is the case of the fluoride crystal $\Delta\alpha_p = 0.31\ 10^{-25}$ cm^3. Fig.1.(b) shows $\Delta\alpha_p$ of Nd doped crystals using the value of Nd:YAG as a reference ($\Delta\alpha_{pYAG}$). This procedure was adopted in to minimize experimental discrepancies between several experimental $\Delta\alpha_p$ data , determined by different groups and using different techniques. It appears that $\Delta\alpha_p$ decreases with band gap of the crystal host and this behavior is also related to the energy position of the 5d levels. Although we do not have the energy position of Nd^{3+} 5d level for all hosts, in general materials with high E_g values (like fluorides) present high energy 5d levels.

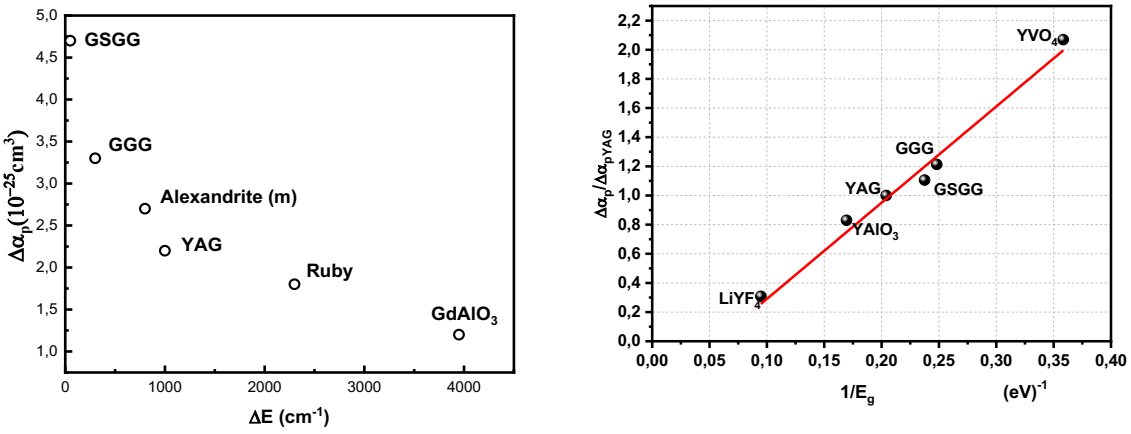

Fig. 1. The strong dependence of $\Delta\alpha_p$ of ion doped materials with the host character. (a) shows the dependence of $\Delta\alpha_p$ of Cr^{3+} doped crystals with the spectroscopic parameter ΔE, which is related to crystal field parameter Dq/B. (b) the dependence of $\Delta\alpha_p$ on Nd^{3+} doped crystal normalized by the value of Nd:YAG, showing that $\Delta\alpha_p$ increases with the inverse of the crystal bandgap (E_g).

The magnitude of the TL effect is related to the phase shift parameter θ_{PL}, which for solids is given by [1]:

$$\theta_{TL} = -\frac{P_{abs}}{\lambda K}\eta_h \frac{ds}{dT}. \qquad \text{Eqn. 2}$$

in which P_{abs} is the absorbed laser power, K is thermal conductivity, (ds/dT) is the temperature coefficient of the optical path length change [$\Delta s=\Delta(nL)/nL$]. The fraction of absorbed energy converted into heat is given by $\eta_h = 1-\Phi_f.\lambda_{exc}/\langle\lambda_{em}\rangle$, where Φ_f is the fluorescence quantum efficiency, λ_{exc} is the excitation wavelength, and $\langle\lambda_{em}\rangle$ is the average emission wavelength. For instance, for Nd^{3+} doped materials ($\langle\lambda_{em}\rangle$ ~1060nm) and excitation at λ_{exc} ~808nm, η_h~ *24%* for Φ_f ~1. One very important feature of Yb^{3+} doped materials is the low quantum defect with η_h~ *9%* (λ_{exc} ~ 941nm, $\langle\lambda_{em}\rangle$ ~1030nm and Φ_f ~1).

The TL response time is related to heat diffusion time, $t_c = w^2/4D$, where w is the waist of the pump beam and D is the heat diffusion (related to thermal conductivity by $K = \rho C_p D$, ρ is the density and C_p the specific heat).

The TL technique have been applied to the determination of important thermo-optical parameters like K and ds/dT, as well as the determination of small absorption coefficients of transparent solids. Moreover, the determination of the parameter η_h can be used in the determination of Φ_f as well as the study of energy transfer process. Other expressions for η_h can derived in order to consider other effects like laser action, amplified stimulated emission, energy transfer, upconversion, etc.

Most materials present simultaneously PL and TL so it is important to discriminate electronic and thermal effects. The magnitude of PL and TL phase-shifts can be compared considering the parameter θ_{PL}/θ_{TL}, where $\theta_{PL} = (2\pi/\lambda)\Delta n_{PL}L$. In the case of pulsed excitation, when heat diffusion can be neglected, the parameter $(\theta_{PL}/\theta_{TL})_{pul}$ is proportional to $\eta_h \rho C_p \Delta\alpha_p (ds/dT)^{-1}$. For cw excitation, $(\theta_{PL}/\theta_{TL})_{cw} = (\tau/t_c)\,(\theta_{PL}/\theta_{TL})_{pul}$, where τ is the PL response time (given by the ion excited metastable state lifetime).

In this talk we review the subject of RIC changes in ion doped SSL materials. The experimental techniques are based on the TL and PL effects, using the time-resolved Z-scan and mode-mismatched TL techniques. The measurements were performed with a single laser or pump-probe mode, using single frequency or tuneable lasers in order to study line shapes. Moreover, techniques to discriminate TL and PL effects will be discussed since in most cases they appear simultaneously. A comparison of the magnitude of thermal and electronic contributions to RIC in most relevant SSL lasers and amplifiers. Finally, perspectives and challenges will be discussed.

References

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