Nonlinear refractive index of erbium-doped $Y_2O_3$ integrated-optical waveguides

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Self-induced phase modulation in erbium-doped polycrystalline $Y_2O_3$ integrated-optic waveguides is studied by use of a Mach–Zehnder interferometric setup. We determined the wavelength dispersion of the nonlinear refractive index near the $4F_{5/2}$ absorption band and show that it corresponds qualitatively with a Kramers–Kronig analysis. The nonlinear coefficient at 670 nm was determined to be $1.4 \times 10^{-14}$ m$^2$/W and has an electronic origin with a slow relaxation time of 6 ms.

Materials that show a nonlinear intensity-dependent refractive index are of great importance for all-optical switching in integrated-optic circuits.\(^1\) Semiconductors and organics are examples of nonlinear materials frequently used in all-optical devices. Rare-earth-doped optical waveguides are well known for their amplifying and lasing characteristics but may be of interest for their intensity-dependent nonlinear index. These materials show sharp absorption peaks and therefore exhibit a relatively broad dispersive refractive-index change. The peak absorptions are relatively low so the waveguides can be operated close to resonance without a drastic increase of power absorption. A consequence, however, of operating close to resonance is that real transitions with long relaxation times are involved. Measurements of the nonlinear refractive index of erbium-doped glass fibers are reported, and attractive nonlinear optical properties are observed.\(^3\) However, rare-earth-doped glasses show inhomogeneous broadening of the absorption transitions, reducing the nonlinear effect. This broadening is expected to be less prevalent in rare-earth-doped polycrystalline $Y_2O_3$ materials because the rare-earth ion can occupy only two sites in the cubic Bixbyite lattice.\(^3\) Rare-earth ions enter the lattice by isomorphic substitution of the yttrium ions on these sites, so the material is stoichiometric and therefore high rare-earth concentrations are possible; this is in contrast to glassy hosts, in which rare-earth ions are dispersed impurities. Another advantage of $Y_2O_3$ is the low dominant phonon energy of 400 cm$^{-1}$,\(^5\) which leads to high radiative quantum efficiencies and therefore small thermal effects. In this Letter we present what is to our knowledge the first demonstration of the self-induced intensity-dependent refractive index of erbium-doped polycrystalline $Y_2O_3$ optical waveguides. We measure the dispersion of the nonlinear coefficient and show that the wavelength dependence agrees qualitatively with a Kramers–Kronig analysis. Therefore we conclude that the measured nonlinear coefficient is of electronic origin. The nonlinearities obtained may also be of importance for the modeling of rare-earth-doped integrated-optic amplifiers and lasers.

The physical mechanism of an intensity-dependent refractive index for a two-level system originates from the relation between the occupation of the levels and the refractive index.\(^6\) The ground state can be depleted by applying light with intensity $I$, which excites the ions to a higher-energy state. In the case of the three-level erbium system this excited state relaxes by a fast transition to the long-lived metastable $4I_{15/2}$ state, as shown in the inset of Fig. 1 for three possible excitation wavelengths. We assume that the $4I_{15/2} \rightarrow 4I_{15/2}$ transition does not contribute to the refractive-index change at visible wavelengths. The density of erbium ion in the ground state depends on the applied intensity according to $N_I = N_{Er}/(1 + I/I_0)$, where $I_0 = h\nu/\lambda\sigma$, $N_{Er}$ is the erbium-ion density, and $\sigma$ is the lifetime of the level involved.\(^7\) The ground-state population density is related to the absorption coefficient $\alpha$ by $\alpha(I, \lambda) = \alpha_{background} + \alpha_{Er}(I, \lambda)$, where $\alpha_{Er}(I, \lambda) = \sigma_{a}(\lambda)N_{I}(I)$ with $\sigma_{a}$ the absorption cross section. So a change in intensity results in a change in absorption $\Delta \alpha(I, \lambda)$. The accompanying refractive-index change $\Delta n(I, \lambda)$ can be calculated by using a Kramers–Kronig formalism\(^6\)^,\(^8\):

\[
\Delta n(I, \omega) = \frac{c}{\pi} \mathrm{P.V.} \int_0^\infty \frac{\Delta \alpha(I, \omega')}{\omega'^2 - \omega^2} \ d\omega',
\]

Fig. 1. Measured erbium absorption spectrum $\alpha_{Er}(\lambda)$ and the calculated maximum refractive-index change of an Er$Y_2O_3$ (0.34 at. % Er) waveguide.
A similar type of population redistribution, waveguide with an estimated efficiency of 20%. The rotation angle is 0. A cw DCM dye laser was used for intensity modulation at a certain position with a phase in the other branch by rotating a phase retarder (a glass slide) and detecting the resulting intensities in the waveguide are kept well below the saturating intensity of $I_s = 3.4$ GW/m$^2$. The dye laser is tunable in the wavelength range near 657 nm, so phase shifts can be measured at various wavelengths around the $4F_{9/2}$ absorption band. At each wavelength the nonlinear coefficient of the waveguide can be calculated from the phase difference between two fringe patterns measured at different intensities. These two fringe patterns were recorded together at one cycle of the phase retarder to reduce the influence of the temporal drift of the interferometer. A representative example of measured fringes is shown in Fig. 3.

Phase differences were determined for various wavelengths, and the corresponding dispersion of the nonlinear coefficient is shown in Fig. 4. The expected dispersion of the refractive-index change resulting from a Kramers–Kronig analysis of the measured absorption spectrum of this waveguide is also shown in Fig. 4 and corresponds qualitatively with the measurements, indicating the measured nonlinear coefficient has an electronic origin. However, the refractive-index change is caused by real electron transitions, so the time constants involved are quite large. In the case of our three-level system, the longest relaxation time involved is that of the intermediate metastable $4I_{13/2}$ level, which has a decay time of 6 ms. However, a short turn-off pulse at 1535 nm may be used to stimulate the transition to the ground state and reset the refractive-index change.$^5$

The usefulness of these types of material in nonlinear optics is determined by the inducible phase change per unit absorption length.$^1$ This is expressed in the figure of merit, $W = \Delta n_{\text{sat}}/\alpha_{\text{Er}}$, which should be larger than 1. Whereas we measured slightly off resonance a refractive-index change of $10^{-5}$ for $I < I_s$, we have $\alpha_{\text{Er}} = 0.2$ cm$^{-1}$ and $\Delta n_{\text{sat}} \approx 10^{-6}$. This results in an acceptable value of $W \approx 0.8$.

Fig. 2. Mach–Zehnder interferometer setup for measuring self-induced phase changes of integrated-optical waveguides.

where P.V. denotes the Cauchy principal value. A similar type of population redistribution, resulting in a refractive-index change, is a common phenomenon with saturation of laser amplifiers.$^9$ The absorption of our three-level system also saturates at high intensities, where the corresponding maximum refractive-index change $\Delta n_{\text{sat}}$ can be computed from an absorption spectrum by using the Kramers–Kronig formalism and is shown in Fig. 1 for an Er:Y$_2$O$_3$ optical waveguide. Such an intensity-dependent refractive index is commonly written in terms of a nonlinear coefficient $n_2$ by $n(I) = n + \Delta n(I) = n + n_2 I$. High-quality polycrystalline Er:Y$_2$O$_3$ optical waveguides were deposited onto oxidized silicon substrates by a reactive sputtering technique.$^{10}$ Scanning electron microscope photographs and x-ray diffraction spectra indicate that the deposited layers consist of crystalline columns. The composition is determined to be stoichiometric through energy dispersive x-ray analysis and Rutherford backscattering. Ridge-type channel waveguides were formed by photolithography and ion-beam sputter etching. The realized waveguides have an erbium concentration of 0.34 at. %, a refractive index of 1.92 at 633 nm, a thickness of 0.8 $\mu$m, a length of 25 mm, a ridge height of 80 nm, and a width of 6 $\mu$m. For this configuration the mode confinement to the nonlinear waveguiding layer is close to unity. To determine the intensity-dependent refractive-index change in these waveguides a Mach–Zehnder interferometer scheme is used,$^{11}$ which is shown in Fig. 2.

The nonlinear coefficient $n_2$ (m$^2$/W) of the waveguide introduces a self-induced nonlinear phase shift $\Delta \phi(I) = (2\pi/\lambda)n_2 I\alpha^{-1}[1 - \exp(-\alpha L)]$ in one branch of the interferometer, where the absorption $\alpha$ is taken as being intensity independent, which is valid for $\alpha_{\text{background}} > \alpha_{\text{Er}}(I, \lambda)$. As a result of this phase shift the fringe pattern will move when the intensity is varied. The fringes are measured by shifting the phase in the other branch by rotating a phase retarder (a glass slide) and detecting the resulting intensity modulation at a certain position with a photodiode and a pinhole. The phase shift introduced by the phase retarder can be calculated from its rotation angle $\theta$. A cw DCM dye laser was used as light source, and its output was coupled into the waveguide with an estimated efficiency of 20%.

Fig. 3. Fringes for powers of 16 mW (top) and 36 mW (bottom) at 670 nm. Dotted curves, measured fringes; solid curves, calculated fringes.
In spite of the contribution of the background loss to the total loss, it is not included in the figure of merit because it does not saturate. However, the demand for lower background absorption still exists, and such an improvement may be achievable by improving the host material or by using longer wavelengths.

In conclusion, we observed that the Er:Y$_2$O$_3$ optical waveguides showed a self-induced phase shift as a result of an intensity-dependent refractive index. The nonlinear coefficient of Er:Y$_2$O$_3$ with an erbium concentration of 0.34 at. % was determined to be $1.4 \times 10^{-14}$ m$^2$/W at a 670-nm wavelength. The wavelength dependence of the nonlinear coefficient corresponds well to the behavior predicted by a Kramers–Krönig analysis, which indicates that the refractive-index change is of electronic origin. The optical nonlinearity that is due to the sharp absorptions, the availability of a wide range of transitions, and the possibility of high doping levels make rare-earth-doped optical waveguides promising for all-optical switching. A low background attenuation is necessary to make this type of material useful for application in devices.

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References