Saturated absorption and degenerate four-wave mixing in Nd^{3+} beta″ alumina

Robert W. Boyd, Mark T. Gruneisen, Paul Narum, and David J. Simkin*

Institute of Optics, University of Rochester, Rochester, New York 14627

B. Dunn and D. L. Yang

Department of Materials Science, University of California at Los Angeles, Los Angeles, California 90024

Received November 8, 1985; accepted December 30, 1985

Nd^{3+} beta″ alumina is one of a family of potentially useful nonlinear-optical materials based on rare-earth ions substituted into a beta″ alumina host. The saturation intensity of Nd^{3+} beta″ alumina has been measured to be in the range 16 to 35 kW cm^{-2}. Phase conjugation by degenerate four-wave mixing has been demonstrated, and the phase-conjugate reflectivity has been measured as a function of both laser wavelength and intensity.

There is currently great interest in the development of new nonlinear-optical materials for use in phase conjugation by degenerate four-wave mixing (DFWM) and in optical bistability. An important class of solid-state nonlinear-optical materials is those for which the nonlinearity is due to saturated absorption.[1–5] Desirable characteristics of such materials are that they possess a large unsaturated absorption coefficient, a small saturation intensity, and a short response time. A material that possesses these characteristics is Nd^{3+}-substituted sodium beta″ alumina. In their recent study of laser action in this material, Jansen *et al.*[6] observed that it exhibits an anomalously large absorption coefficient in the 580-nm spectral region, presumably due to the $^4I_{9/2} \rightarrow {}^4G_{5/2}$ crystal field transition of the Nd^{3+} ion.[7] The oscillator strength of this transition is 6×10^{-5}, compared with the more typical value of 7×10^{-6}, which occurs in Nd:YAG. Population placed into the $^4G_{5/2}$ level decays rapidly to the $^4F_{3/2}$ level, which acts as a trapping level, as illustrated in the inset of Fig. 1. In Nd:YAG, the lifetime of the trapping level decreases rapidly with increasing Nd^{3+} concentration, leading to an increased value of the saturation intensity. However, beta″ alumina can accept a large Nd^{3+} concentration without suffering concentration quenching. In fact, with a Nd^{3+} concentration of 1.5×10^{21} cm^{-3}, the fluorescent lifetime of the $^4F_{3/2}$ level has the reasonably large value of 350 μsec.[8] These observations suggest that beta″ alumina with a large Nd^{3+} concentration possesses a large absorption coefficient, a low saturation intensity, and sufficient speed for many applications.

The structure of sodium beta″ alumina ($Na_{1+x}Mg_xAl_{11-x}O_{17}$) consists of spinel blocks of closely packed Al^{3+} and O^{2-}, separated by loosely packed planes containing Na^+ and O^{2-}. Farrington and Dunn[8,9] have demonstrated that many divalent and even trivalent cations can be exchanged for the Na^+ ions, even to the extent of complete replacement for certain ions such as Nd^{3+}. The material used in the work reported here was prepared using an ion-exchange procedure[9] in which single crystals of sodium beta″ alumina were placed in a molten salt mixture at 600°C consisting of 55% NaCl and 45% $NdCl_3$. The extent of the Nd^{3+} exchange was determined gravimetrically. The crystal used in this study had a concentration of 8.5×10^{20} cm^{-3}, a thickness of 230 μm, and an absorption path length ($\alpha_0 L$) of 2.8. The crystals grow in the form of platelets, and selected samples are of high optical quality.

The saturation behavior was studied by focusing into the sample the output of a cw dye laser (Coherent 699-21 pumped by a 5-W argon-ion laser) tuned to the peak of the absorption band. The results giving the internal transmission as a function of the incident laser intensity (corrected for the 8% Fresnel reflection

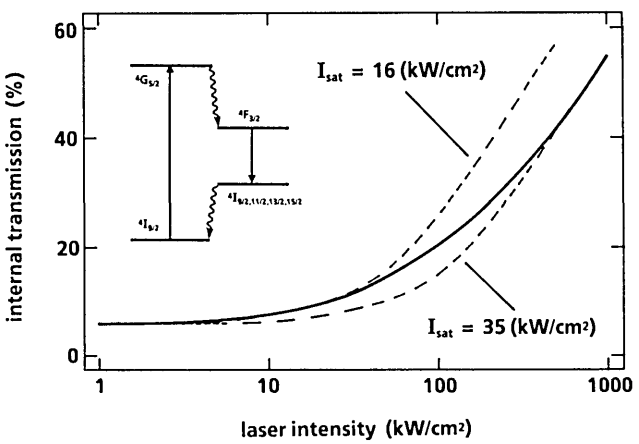

Fig. 1. Internal transmission of Nd^{3+} beta″ alumina versus laser intensity measured at a wavelength of 577 nm (solid curve). The dashed curves are theoretical predictions for an inhomogeneously broadened saturable absorber. The inset shows the relevant energy levels of Nd^{3+}.

0146-9592/86/030162-03$2.00/0

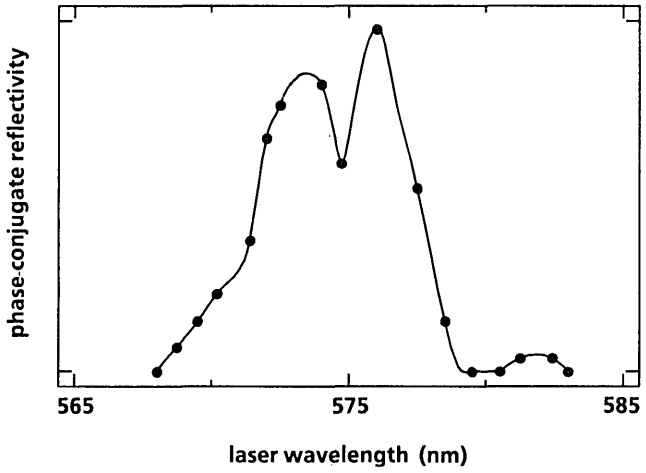

Fig. 2. Phase-conjugate reflectivity in arbitrary units versus laser wavelength at a pump intensity of 10 kW cm^{-2} in each beam.

loss on the entering surface) are shown in Fig. 1. Even at our largest laser intensities, we did not observe any thermal lensing effects. It is believed that crystal field transitions in Nd^{3+} beta″ alumina are inhomogenously broadened,[6] and thus we have attempted to fit the data by modeling the system as a four-level, inhomogeneously broadened saturable absorber. Under this assumption, the spatial evolution of the laser intensity $I(z)$ is described by

$$\frac{dI(z)}{dz} = -\frac{\alpha_0 I(z)}{[1 + I(z)/I_s]^{1/2}}, \quad (1)$$

where α_0 is the low-intensity absorption coefficient and I_s is the saturation intensity. The theoretical curves shown in Fig. 1 are obtained by solving this equation with $\alpha_0 L = 2.8$ and I_s equal to 16 and 35 kW cm^{-2}. The measured saturation curve fits between these two theoretical curves, with the low-intensity part lying close to the 16 kW cm^{-2} curve and the high-intensity part lying close to the 35 kW cm^{-2} curve. This increase in effective saturation intensity with increasing laser intensity could be due to thermal effects or to a decrease in the effective lifetime of the trapping level due either to excited-state absorption or to amplified spontaneous emission. We have also attempted to fit the data under the assumption that the transition is homogeneously broadened and have found the agreement with the experimental results to be far worse.

In order to demonstrate the potential of this material in nonlinear optics, we have performed a degenerate four-wave mixing experiment using the standard geometry of phase conjugation. The pump and probe beams were focused into the sample using 10× microscope objectives, yielding a spot size of 6 μm and a confocal parameter of 330 μm. The crossing angle between the pump and probe beams measured inside the crystal was 6°, and hence the beams did not completely overlap within the sample. The intensities of the two counterpropagating pump beams were approximately equal and were much larger than that of the probe beam.

The maximum phase-conjugate reflectivity that we observed was 0.3% at a laser wavelength of 576 nm and at our maximum laser power, which corresponds to an intensity of 30 kW cm^{-2} in each pump beam. The theoretically predicted[10] reflectivity taking pump absorption into account but assuming homogeneous broadening and infinite plane waves is 2%. We believe that the origin of this discrepancy is the use in our experiment of beams with a Gaussian transverse profile, the incomplete overlap of the beams, and the fact that for our medium the absorption saturates less rapidly (thus yielding a smaller nonlinearity) than for a homogeneously broadened medium.

The variation of phase-conjugate reflectivity with laser wavelength is shown in Fig. 2 for a pump intensity of 10 kW cm^{-2} in each beam. Three resonances are evident in this tuning curve. These resonances occur approximately at the wavelengths of the peaks in the published absorption spectrum of Nd^{3+} beta″ alumina,[6] and in fact for our pump intensity and absorption path length the maximum reflectivity is expected to occur close to the center of the absorption line.[11]

The variation of phase-conjugate reflectivity with the pump intensity is shown in Fig. 3. For low pump intensities, the reflectivity scales as the square of the total pump intensity, as expected for a four-wave mixing process. At higher pump intensities, the reflectivity increases less rapidly, although for our maximum laser power we were not able to reach the regime where the reflectivity decreases with increasing laser intensity.

In conclusion, we have determined that the absorption of the 580-nm band of Nd^{3+} beta″ alumina saturates approximately in the manner expected of an inhomogeneously broadened medium with a saturation intensity in the range 16 to 35 kW cm^{-2}. We have performed a degenerate four-wave mixing experiment that produced a phase-conjugate reflectivity of 0.3%, limited by our laser intensity. It is possible to substitute any rare-earth ion[8] for the Nd^{3+} used in the pres-

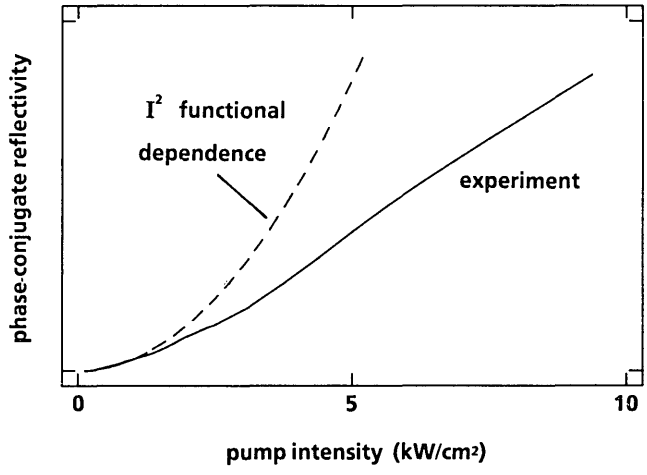

Fig. 3. Phase-conjugate reflectivity in arbitrary units versus pump intensity in each beam at a laser wavelength of 576 nm.

ent experiment, and thus it may be possible to develop a new class of solid-state, nonlinear-optical materials based on beta″ alumina with resonant transitions throughout the visible, ultraviolet, and near infrared.

We acknowledge useful discussions with A. L. Gaeta regarding the theory of DFWM. The portion of the work conducted at the University of Rochester was supported by National Science Foundation grant ECS-8408370 and the Joint Services Optics Program. The portion of the work conducted at the University of California at Los Angeles was supported by the U.S. Office of Naval Research.

* Permanent address, Department of Chemistry, McGill University, Montreal, Quebec, Canada.

References

1. R. L. Abrams and R. C. Lind, Opt. Lett. **2,** 94 (1978).
2. P. F. Liao and D. M. Bloom, Opt. Lett. **3,** 4 (1978).
3. D. S. Hamilton, D. Heiman, J. Feinberg, and R. W. Hellwarth, Opt. Lett. **4,** 124 (1979).
4. H. M. Gibbs, A. C. Gossard, S. L. McCall, A. Passner, W. Wiegmann, and T. N. C. Venkatesan, Solid State Commun. **30,** 271 (1979).
5. R. K. Ahrenkeil, J. F. Figueira, C. R. Phipps, Jr., D. J. Dunlavy, S. J. Thomas, and A. J. Sievers, Appl. Phys. Lett. **33,** 705 (1978).
6. M. Jansen, A. Alfrey, O. M. Stafsudd, B. Dunn, D. L. Yang, and G. C. Farrington, Opt. Lett. **9,** 119 (1984).
7. J. A. Koningstein and J. E. Geusic, Phys. Rev. **136,** A711 (1964).
8. B. Dunn and G. C. Farrington, Solid State Ionics **9/10,** 223 (1983).
9. G. C. Farrington and B. Dunn, Solid State Ionics **7,** 267 (1982).
10. M. T. Gruneisen, A. L. Gaeta, and R. W. Boyd, J. Opt. Soc. Am. B **2,** 1117 (1985).
11. A. L. Gaeta, M. T. Gruneisen, and R. W. Boyd, IEEE J. Quantum Electron. (to be published).