

1991-04-01

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Recommended Citation

Rossi, B., Byrne, H. & Blau, W. (1991) Degenerate Four-Wave Mixing in Rhodamine Doped Epoxy Waveguides. *Applied Physics Letters*, Vol. 58, no.16, 22 April. doi:10.1063/1.105117

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Degenerate four-wave mixing in rhodamine doped epoxy waveguides

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(Received 19 October 1990; accepted for publication 29 January 1991)

Self-diffraction from laser-induced gratings is observed in a 13 μm -thick film of rhodamine B doped epoxy. The decay of the grating is measured to be 2.05 ns. Degenerate forward four-wave mixing in a guided geometry is observed by coupling out all beams after propagation over 1 mm. At 595 nm the third-order nonlinear susceptibility of the film is measured to be $1.5 \times 10^{-19} \text{ m}^2 \text{ V}^{-2}$.

The critical stage in the development of an all-optical technology is the development and optimization of suitable nonlinear optical materials. Although many functions have been demonstrated on a prototype level,^{1,2} exploitation has been limited by the lack of an optimized material. Inherent material requirements include a large nonlinear optical susceptibility, a fast response and decay time, and low inherent losses. In this study, the nonlinear optical properties of an epoxy film doped with a commercial laser dye, rhodamine B, are investigated. The wavelength range studied is in close proximity to the resonance of the dye.

The film was spun from a solution of one part MV757 commercial epoxy resin, one part epoxy hardener, and one part of 2 g/l rhodamine B in ethanol solution. A glass microscope slide was used as substrate. A spinning rate of 500 rpm was employed, producing a film of thickness $13.0 \pm 0.5 \mu\text{m}$. Both the absorption and uncorrected fluorescence spectra of the film spectra are singly peaked, at 545 and 590 nm, respectively. The absorption spectrum compares well with that of rhodamine B in solution,³ indicating that the dye/epoxy film is a monodisperse quasi-solution. Assuming a value of $\epsilon(545) = 1.06 \times 10^5 \text{ l mole}^{-1} \text{ cm}^{-1}$ for the molar extinction coefficient of rhodamine B at 545 nm, the concentration of dye in the film may be calculated to be $7.7 \times 10^{-3} \text{ mole l}^{-1}$.

The experimental method employed in these studies was that of forced light scattering from laser-induced gratings.⁴ The light source is a nitrogen-pumped dye laser emitting linearly polarized pulses of $350 \pm 50 \text{ ps}$ duration of energy 40–80 μJ per pulse. The dyes used were coumarin 485 and rhodamine B, giving tunability in the range 495–550 nm and 595–640 nm, respectively. The method, which is described in detail elsewhere,⁵ is based on the interference at the sample of two spatially and temporally overlapped beams, producing a spatial modulation of the intensity-dependent refractive index of the material. Under thin grating conditions,⁴ an expression relating the diffraction efficiency, η into the first order, to the third-order material nonlinearity may be derived:

$$|\chi^{(3)}| = \frac{4\epsilon_0 c n^2 \lambda \alpha \sqrt{\eta}}{3\pi I_0 (1 - T) \sqrt{T}}, \quad (1)$$

where c is the speed of light, ϵ_0 is the permittivity of free space, n is the refractive index of the sample, α is the absorption coefficient, T is the sample transmission, and I_0 is the input pulse intensity.

For the purpose of investigating the temporal decay of the grating, a third beam was employed to read the grating in a folded boxcar's geometry.⁶ Delay of this beam with respect to the arrival of the other two beams enables resolution of the recovery time of the material nonlinearity.

Waveguiding was achieved by the prism-coupling method. The sample was mounted on an Aerotech ARS-301 rotation stage which enabled control of the coupling angle with 0.01° resolution. The coupling prism employed was an SF6 prism of base angle $\epsilon = 45^\circ$, and refractive index at 633 nm of 1.806. For simplicity, a two-beam geometry was employed for the degenerate four-wave mixing (DFWM) in the guided mode. A second, identical prism was used to limit the interaction length within the guide by coupling out the light after 1 mm. This was necessary for operation under thin grating conditions.

In the transmission mode, self-diffraction was carried out using Coumarin 485 as the source laser dye. When placed in the interference region, a clear diffraction pattern from the sample was observable over much of the tuning range. The diffraction efficiency into the first order was measured by using silicon photodiodes to monitor the input and diffracted energies. The intensity dependence of the diffraction efficiency was investigated by transmission of both input beams through a variable neutral density filter and Fig. 1 shows the dependence at 540 nm. The plot shows a cubic dependence of the diffracted signal on the input intensity at low intensities, characteristic of a third-order nonlinear process. At higher intensities, the dependence flattens to a linear behavior, characteristic of the onset of a saturation of the absorption band. From a two-level model, the saturation intensity of rhodamine B may be calculated to be $3 \times 10^9 \text{ W m}^{-2}$. This compares favorably to the intensity range in which the saturation is observed here.

The temporal response of the nonlinear process was investigated by the introduction of a third "read" beam whose arrival at the sample with respect to the two "writing" beams could be varied via a delay line. Figure 2 shows the decay of the grating at 540 nm. A good fit to a single exponential decay is found with a decay time $\tau/2 = 2.05 \text{ ns}$. This is consistent with the decay of a grating formed by a saturated absorption with a ground-state recovery time of $\tau = 4.1 \text{ ns}$.⁴ Such lifetimes are characteristic of fluorescence decay times of rhodamines.⁷

The dispersion of the nonlinear optical response was monitored by tuning over the wavelength range of the dye.

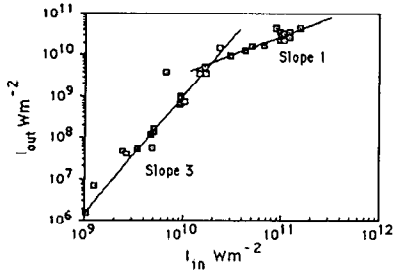


FIG. 1. Intensity dependence of transmission diffraction efficiency at 540 nm.

In Fig. 3 the dispersion of the nonlinearity is plotted as well as the absorption spectrum. The nonlinear response roughly follows the square of the linear absorption of the film, indicating that the diffraction process is due to saturable absorption.

Self-diffraction in the guided mode was performed using rhodamine B as the source laser dye. At low pulse energies (≈ 100 nJ per pulse), waveguiding was readily achievable in the rhodamine doped film across the range of the dye and was observable as a visible fluorescence streak in the film. Approximately 20 modes were observable and the zeroth order mode was seen to have a coupling angle $\alpha = 29.15 \pm 0.01^\circ$ with respect to the normal to the input face of the prism. The coupling angle of the highest order mode was found to be $\alpha = 21.44 \pm 0.01^\circ$. Values of the effective indices, β/k , of the modes of the guide may be calculated⁸ and are found to lie in the range 1.573–1.511. Knowing the effective indices of the modes and the thickness of the guide, the refractive index of the film may be established.⁹ At 633 nm, the refractive index of the film is calculated to be 1.5743 ± 0.0001 . The total number of modes which may be supported by the guide may be calculated from the equation⁸

$$m \leq \frac{1}{\pi} \left[kd \sqrt{n_f^2 - n_s^2} - a \tan \left(\frac{\sqrt{n_s^2 - n_c^2}}{\eta_{sf} \sqrt{n_f^2 - n_s^2}} \right) \right] \quad (2)$$

to be 18 for both TE and TM modes. n_f is the refractive index of the film, n_s the refractive index of the substrate, and n_c the refractive index of the cladding (air). Comparison of this figure with the approximate number of modes observable implies that no distinction between TE and TM modes is observable, indicating the absence of film birefringence.

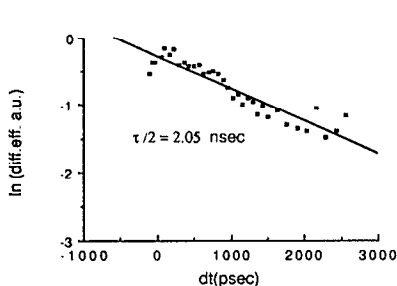


FIG. 2. Temporal decay of the diffraction efficiency at 540 nm.

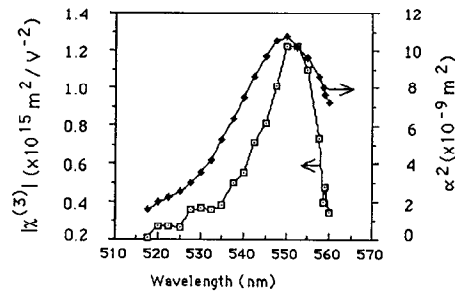


FIG. 3. Wavelength dependence of nonlinear susceptibility.

The prism-coupling apparatus was introduced into the interference region of the DFWM experiment and a second prism was placed on the sample, 1 mm from the input coupling region. The intensity dependence of the diffraction process at 595 nm was monitored by use of a variable neutral density filter. Figure 4 shows the measured curve. The input intensity has been corrected for the nonlinear throughput of the guide in the absence of self-diffraction, described in detail elsewhere.¹⁰ The behavior of the nonlinear throughput was compared to the measured intensity-dependent transmission of the sample. From the rate of change of the waveguide throughput with intensity, estimates can be made of the intensity within the guide and values of the coupling efficiency, calculated to be 0.61 for intensity and 0.20 for energy. The corrected intensity dependence shows the same behavior as the transmission mode (Fig. 1), consisting of an initial cubic dependence followed by a linear dependence at higher intensities. A diffraction efficiency of 0.25 at a guided intensity of $2.64 \times 10^{10} \text{ W m}^{-2}$ is observed. The material nonlinearity at 595 nm may be calculated from Eq. (1) to be $1.5 \times 10^{-19} \text{ m}^2 \text{ V}^{-2}$, which, assuming a quasisolution of concentration $7.7 \times 10^{-3} \text{ mole l}^{-1}$, corresponds to a molecular hyperpolarizability for rhodamine B of $6.0 \times 10^{-42} \text{ m}^5 \text{ V}^{-2}$. It is worth noting that this value, although considerably resonantly enhanced, compares rather favorably with those measured for organic conjugated polymers.¹¹

Spun films of a commercial epoxy, doped with a commercial laser dye, have been employed to study nonlinear phenomena at visible wavelengths in both transmission and waveguiding modes. Degenerate forward four-wave mixing is readily observable in the transmission mode and temporal measurements show the nonlinear mechanism to be

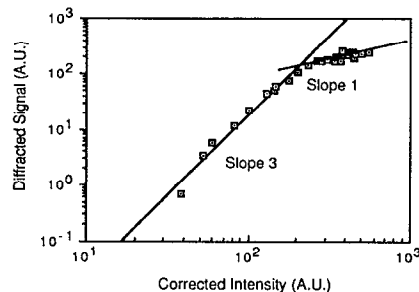


FIG. 4. Intensity dependence of guided diffraction efficiency at 595 nm.

largely absorptive in nature. In the guided mode, self-diffraction is observable in the long wavelength tail of the absorption. Despite the low-energy coupling efficiencies, the high intensities afforded by the guided geometry produce strong nonlinear absorption effects. Material nonlinearities have been calculated and, significantly, the values of the nonlinearities are of reasonable size. This indicates the feasibility of a move towards less cumbersome, short chain conjugated systems, in the search for nonlinear optical materials suitable for optical technologies.

This work has been completed and is presented in loving memory of Barbara Rossi, who died tragically on the 1st of March, 1990.

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