Linear and nonlinear waveguiding in Rhodamine-doped epoxy films

B. Rossi, H. J. Byrne, and W. Blau

Department of Pure and Applied Physics, Trinity College, Dublin 2, Ireland

G. Pratesi and S. Sottini

Istituto di Ricerca sulle Onde Elettromagnetiche del Consiglio Nazionale della Ricerche, Firenze, Italy

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Planar waveguiding of 350-ps pulses, at a wavelength in the region of the long-wavelength tail of the absorption, is observed by coupling by prism into spun films of Rhodamine-B-doped epoxy films. In a 13-μm-thick film, $\approx$20 modes are supported at 633 nm. The angle of optimum coupling is seen to be intensity dependent; the angle increases with increasing intensity, indicating a positive real component of the material nonlinearity at all wavelengths investigated. The throughput of the guide is also seen to be strongly intensity dependent, indicating a strong intensity dependence of the transmission of the film. The observed behavior is compared with the intensity dependence of the unguided transmission, and the intensity-coupling efficiencies are estimated. As we know the coupling efficiency, we estimate the real component of the nonlinearity to be $7.0 \pm 0.5 \times 10^{-20}$ m^2 V^{-2} at 616 nm and $5.0 \pm 0.5 \times 10^{-20}$ m^2 V^{-2} at 635 nm from the intensity dependence of the coupling angle.

INTRODUCTION

With the installation of high-performance optical communication networks in the past few years and the planned expansion of the network systems, the interest in optical technologies and their potential has increased tremendously. Exploitation of the inherent speed and high bit rate achievable with optical data transmission may only be fully realized through the development of high-speed switching and processing technology. Although the electronics industry has strained to meet the demands, the future of integrated all-optical switching appears secure.

The critical stage in the development of such an all-optical technology is the development and optimization of suitable nonlinear-optical materials. Although many functions and processes 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, which would allow for switching of low light intensities and for small device dimensions, and a fast response and decay time, which would enable high-bandwidth operation. Other material properties such as quality of film formation, mechanical strength, and chemical, thermal, and radiation stability are also critical. Of course, cost and availability of materials and ease of processing must also be considered.

Among the materials currently under investigation, organic materials have attracted particular interest. Their ease and low cost of synthesis, as well as their ease of processing and the ability of their optical and electronic properties to be tailored to specific applications, are obviously favorable factors. Organic conjugated polymers have been shown to exhibit large nonlinear susceptibilities with ultrafast response times[3,4] and appear to be the most promising organic materials for nonlinear optics. However, studies of the chain-length dependence of the polymer nonlinearity indicate that the effective conjugation length of the polymer is limited to approximately ten repeat units.[5] Although the origins of this behavior are unclear, a move toward less cumbersome, short-chain organic molecules seems reasonable.

In this study, the waveguiding properties, both linear and nonlinear, 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, on the long-wavelength side. The dye/epoxy system should serve as a good model for the study of the variety of processes observable in organic waveguides.

MATERIAL AND LINEAR OPTICAL PROPERTIES

The film was spun from a solution of one part MV757 commercial epoxy resin, one part epoxy hardener, and one part 2-g/L Rhodamine B in ethanol solution. A glass microscope slide was used as the substrate. A spinning rate of 500 rpm was employed, which produced a film of thickness 13.0 ± 0.5 μm.

Figure 1 shows the absorption and uncorrected fluorescence spectra of the film. Both spectra are singly peaked, the absorption spectrum at 545 nm and the fluorescence spectrum at 590 nm. The absorption spectrum compares well with that of Rhodamine B in solution,[6] indicating that the dye/epoxy film is a monodisperse quasi-solution, with no indication of aggregation. Assuming a value of $\epsilon(545) = 1.06 \times 10^5$ L mol^{-1} 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

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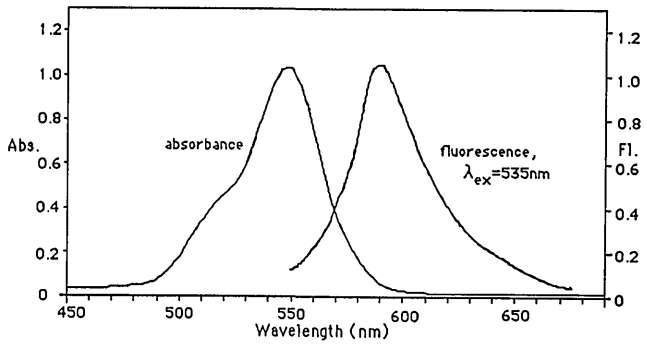

Fig. 1. Absorption and uncorrected fluorescence spectra of Rhodamine-doped epoxy film.

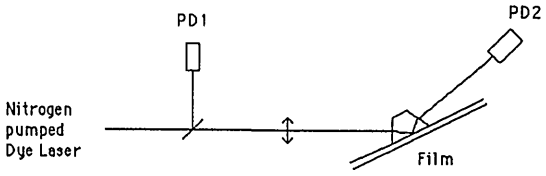

Fig. 2. Schematic diagram of waveguiding setup.

7.7×10^{-3} mol L^{-1}. The fluorescence spectrum is slightly red shifted from that of the solution.

LINEAR AND NONLINEAR WAVEGUIDING

Waveguiding experiments were conducted using a nitrogen-pumped dye laser. The laser dye employed for all experiments was Rhodamine B, which provided pulses of 350 ± 50 ps tunable in the range 595–640 nm. Typical pulse energies were in the range of 40–80 μJ per pulse, depending on the wavelength. Coupling to the waveguide was achieved by the prism-coupling method. A schematic of the experimental setup is shown in Fig. 2. The input beam was focused into the coupling region by a 30-cm lens. The sample was mounted on an Aerotech ARS-301 rotation stage, which enabled control of the coupling angle with 0.01° resolution. A silicon photodiode (PD2) was used to measure the throughput of the waveguide, and another silicon photodiode (PD1) was used to monitor the input-pulse energies. The photodiodes were calibrated using a Gentec ED-100A pyroelectric energy meter. The coupling prism employed was an SF6 double prism of base angle $\epsilon = 60°$ and refractive index at 633 nm of 1.806.

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 dark m lines on the uncoupled beam from the exit face of the prism, as well as a visible fluorescence streak in the film. Approximately 20 modes were observed, and the zeroth-order mode was seen to have a coupling angle to the input face of the prism of $\alpha = 1.05 \pm 0.01°$ with respect to normal. The coupling angle of the highest-order mode was found to be $\alpha = -5.81 \pm 0.01°$. Values of the effective indices (β/k) of the modes of the guide may be calculated from the equation[7]

$$\beta/k = \sin\alpha \cos\epsilon + (n_p^2 - \sin^2\alpha)^{1/2} \sin\epsilon, \qquad (1)$$

where n_p is the refractive index of the prism. The effective indices of the modes are found to lie in the 1.573–1.511 range. Since we know the effective indices of the modes and the thickness of the guide, we may establish the refractive index of the film by numerically solving the equation[8]

$$\tan b_1 d = \frac{b_1(p_0\eta_{sf} + p_2\eta_{cf})}{b_1^2\eta_{sf}\eta_{cf} - p_0 p_2}, \qquad (2)$$

where

$$b_1 = (k^2 n_f^2 - \beta^2)^{1/2},$$

$$p_0 = (\beta^2 - k^2 n_c^2)^{1/2},$$

$$p_2 = (\beta^2 - k^2 n_s^2)^{1/2},$$

$$\eta_{ij} = (n_i/n_j)^2 \quad \text{for TM modes and 1 for TE modes,}$$

n_f is the refractive index of the film, n_s is the refractive index of the substrate, and n_c is the refractive index of the cladding (air). At 633 nm, the refractive index of the film is calculated to be 1.5743 ± 0.0001. The total number of modes that may be supported by the guide may be calculated from the relation[8]

$$m \le \frac{1}{\pi}\left\{k(n_f^2 - n_s^2)^{1/2} - a \tan\left[\frac{(n_s^2 - n_c^2)^{1/2}}{n_{sf}(n_f^2 - n_s^2)^{1/2}}\right]\right\} \qquad (3)$$

to be 18 for both TE and TM modes. Comparison of this figure with the approximate number of modes observable implies that no distinction between TE and TM modes is observable, which indicates the absence of film birefringence.

Using neutral-density filters to achieve a variation in intensity, the intensity dependence of the throughput of

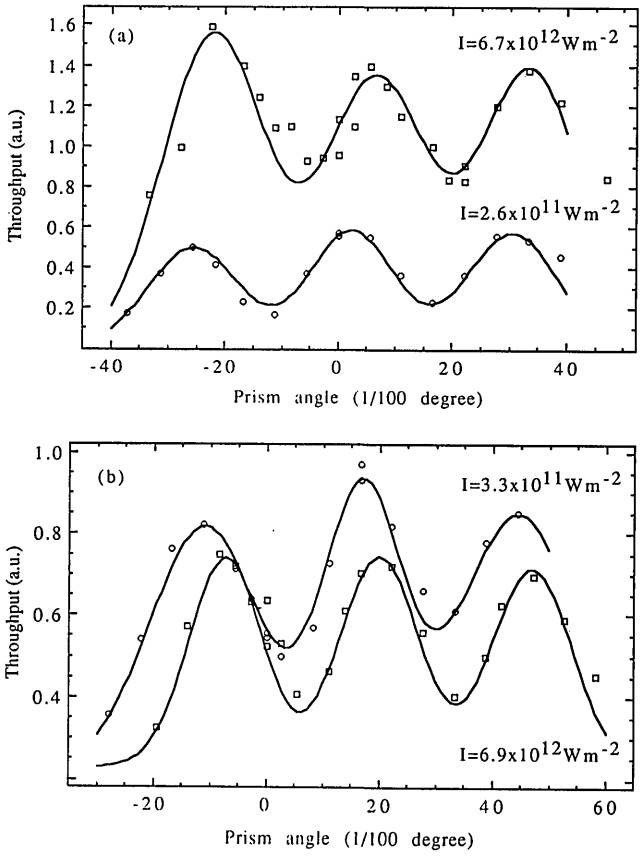

Fig. 3. Intensity dependence of coupling angle (a) 616 nm and (b) 635 nm. I is the intensity external to the guide.

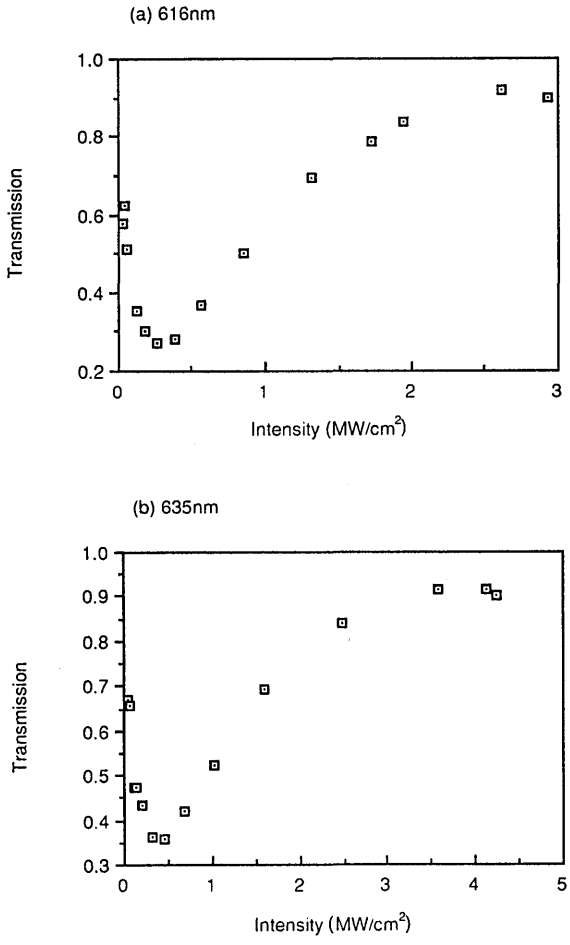

Fig. 4. Intensity-dependent absorption of Rhodamine-doped epoxy film at (a) 616 nm and (b) 635 nm.

the guide, as well as the intensity dependence of the coupling angle, was monitored at both 616 and 635 nm. The observed dependences are shown in Figs. 3(a) (616 nm) and 3(b) (635 nm) for a selection of modes of intermediate mode number. In both cases, there is a clear dependence of the coupling angle on intensity; the angle increases with increasing intensity at both wavelengths. This intensity dependence is indicative of an intensity dependence of the refractive index of the film. However, estimation of the degree of nonlinearity of the refractive index requires a knowledge of the intensity that is coupled into the film. As will be seen below, this may be acquired by considering the intensity dependence of the throughput of the guide.

At a wavelength of 616 nm, there is a dramatic increase in the throughput of the guide with increasing intensities, indicating the presence of an intensity-dependent loss process in the guide. Such behavior is characteristic of an absorption-bleaching process, by no means uncommon in organic-dye systems.[9] At 635 nm, however, the observed trend is the inverse. It has been shown that, in the presence of a strong nonlinearity, nonlinear coupling is accompanied by a significant loss of coupling efficiency.[10] In this case, however, the otherwise Gaussian profile of the angle dependence of the coupling efficiency becomes strongly asymmetric. In the present case, however, a reasonable fit of Gaussian profiles to the modes at all intensities can be performed; therefore it may be deduced that a behavior characteristic of an inverse saturable absorption is observed. Although such a process has also been observed in organic-dye systems,[11] the problem of understanding the reversal of trends within a small wavelength range must be addressed. With this in mind, the intensity dependence of the transmission of the film in an unguiding geometry was monitored at both wavelengths. Figures 4(a) (616 nm) and 4(b) (635 nm) show the observed dependences of the film transmission on intensity. A similar trend is observed at both wavelengths. The initial decrease of the transmission is indicative of an inverse saturation process, whereas at higher intensities a bleaching of the absorption is seen. The difference in the trends observable in the intensity dependence of the guided throughput at the two wavelengths may therefore be attributed to a difference in the intensity ranges studied at the two wavelengths (determined by the tuning curve of the lasing dye). The origins of the nonlinear behavior of the absorption of the Rhodamine-doped epoxy is as yet unclear, and further studies are under way to elucidate the relevant energy-level scheme.

Since the intensity dependence of the sample absorption in an unguided mode has been investigated, the nonlinear throughput of the guide may be used to estimate the coupling efficiency into the guide. This may be achieved by fitting the rate of change of the throughput of the guide as a function of intensity to the nonlinear absorption curves. This procedure assumes that there is no significant loss of coupling efficiency as a result of nonlinear coupling. For the wavelengths of 616 and 635 nm, the intensity-coupling efficiencies are thus found to be 0.61 and 0.18, respectively. Given a measured spot size of 250-μm radius, these correspond to energy-coupling efficiencies of 0.020 and 0.008. It should be noted that the difference in coupling efficiencies at the two wavelengths is not the result of spectral differences in the material properties but rather the day-to-day variation in the coupling region.

Knowledge of the guided intensity enables an approximate calculation of the material nonlinearity from the intensity dependence of the mode number. When we assume that

$$\Delta\beta/k = \beta/k(I) - \beta/k(I_0), \tag{4}$$

the refractive index $n(I)$, and therefore Δn, may be found by solving Eq. (2) for $\beta/k(I)$, and hence $\chi^{(3)}$ may be calculated from the relationship

$$\chi^{(3)} = \frac{4\epsilon_0 cn^2 \Delta n}{3I} \tag{5}$$

to be $\chi^{(3)}$ (616 nm) = $1.5 \pm 0.5 \times 10^{-16}$ m^2 V^{-2} and $\chi^{(3)}$ (635 nm) = $1.0 \pm 0.5 \times 10^{-16}$ m^2 V^{-2}. We have much simplified this calculation to avoid numerical integration of nonlinear-coupling equations over the length of the coupling region. To allow for the fact that the nonlinear interaction occurs over this length rather than over the dimensions of the laser spot, a correction factor of 5×10^{-4} should be employed. The nonlinearity of the film at the two wavelengths is therefore estimated to be $\chi^{(3)}$ (616 nm) = $7.0 \pm 0.5 \times 10^{-20}$ m^2 V^{-2} and $\chi^{(3)}$ (635 nm) = $5.0 \pm 0.5 \times 10^{-20}$ m^2 V^{-2}. In a further study, degenerate four-wave mixing in the Rhodamine-doped epoxy film was performed in an unguided configura-

tion.[12] The values of the film nonlinearity are consistent with an extrapolation of the dispersion of the fully resonant nonlinearity to a slightly resonantly enhanced value. In the fully resonant case, temporally resolved measurements are consistent with saturation of the one-photon allowed transition as the primary nonlinear mechanism. Thermal contributions to the nonlinearity may be neglected owing to the efficiency of radiative relaxation processes in Rhodamine. These bulk nonlinearities may be converted to molecular hyperpolarizabilities, assuming a quasisolution of concentration 8.0×10^{-3} mol L^{-1}, yielding values of $\gamma = 2.0 \pm 0.5 \times 10^{-42}$ m^5 V^{-2} and $2.0 \pm 0.5 \times 10^{-42}$ m^5 V^{-2}. These values compare rather favorably with off-resonant nonlinearities of organic conjugated polymers.[13]

CONCLUSIONS

Spun films of a commercial epoxy, doped with a commercial laser dye, were employed to study nonlinear waveguiding phenomena at visible wavelengths. Nonlinear coupling and waveguiding of 350-ps pulses were observed. Although a strong intensity dependence of both the coupling angle and the throughput of the guide was observed, the Gaussian shape of the angle dependence of the coupling efficiency of the modes was preserved, indicating that there is no significant loss of coupling efficiency. Despite the low-energy coupling efficiencies, the high intensities afforded by the guided geometry produced strong nonlinear absorption effects. Coupling nonlinearities were estimated, and, significantly, the values of the nonlinearities were of reasonable size, although considerably resonantly enhanced, and they compare favorably with those of the more cumbersome organic conjugated polymers. This indicates the feasibility of a move toward less cumbersome, short-chain conjugated systems in the search for nonlinear-optical materials suitable for optical technologies.

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