Nonlinear-optical interactions in fluorescein-doped boric acid glass

Mark A. Kramer, Wayne R. Tompkin, and Robert W. Boyd

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

Fluorescein-doped boric acid glass is a material characterized by an extremely low saturation intensity of ~ 15 mW cm^{-2} and a nonlinear susceptibility $\chi^{(3)}$ as large as ~ 1 esu. The saturated absorption of this material is shown both theoretically and experimentally to depend on the state of polarization of the saturating beam, even though the unsaturated absorption is polarization insensitive. Phase-conjugate reflectivities as large as 0.6% have been obtained through use of degenerate four-wave mixing in this material. These measured reflectivities are in good agreement with the predictions of a theory that includes the effects of excited-state absorption and grating washout. In addition, two-beam coupling due to the nonlinearity of saturable absorption has been demonstrated in this material. The magnitude of the coupling is maximized by inducing a frequency shift between the two beams of ~ 0.1 Hz.

INTRODUCTION

There is great need for nonlinear-optical materials that can be used with low-intensity light sources for applications such as phase conjugation, image processing, and optical switching. Large nonlinear-optical susceptibilities resulting from the nonlinear response of organic molecules have been reported recently for the case of single crystals of organic molecules,[1] for organic molecules in liquid solution,[2] and for organic molecules doped into various solids.[3–5] Some of the largest optical nonlinearities are those due to saturable absorption in materials with a small value of the saturation intensity. Fluorescein-doped boric acid glass has a very small saturation intensity of ~ 15 mW cm^{-2} and consequently the nonlinear $\chi^{(3)}$ susceptibility can be as large as 1 esu for samples of appropriate fluorescein concentration.

The origin of the extremely small saturation intensity of fluorescein-doped boric acid glass is illustrated in Fig.
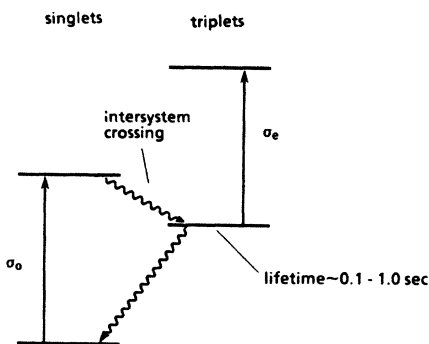

FIG. 1. Energy-level diagram showing the relevant optical interactions in fluorescein-doped boric acid glass. Due to its long luminescent lifetime, the lowest-lying triplet state acts as a trap level. σ_0 and σ_e denote the absorption cross sections of the ground and excited states, respectively.

1. The singlet ground-state absorption cross section is denoted as σ_0 and has a value[6] of $\sim 3 \times 10^{-16}$ cm^2 at a wavelength of 4579 Å; the absorption cross section of the excited triplet state is denoted as σ_e and has a value of $\sim 1.5 \times 10^{-16}$ cm^2. Following optical excitation from the singlet ground state to the singlet excited state, population can make an intersystem crossing into the lowest-lying triplet state. In fluorescein-doped boric acid glass, this level has an extremely long lifetime because many of the quenching mechanisms that are present in liquid solutions do not exist in a solid environment. At room temperature, the principal relaxation route out of the triplet state is thermally activated delayed fluorescence, which leads to nonexponential decay of the triplet-state population in a time of the order of 0.1–1.0 sec.[6–8] The response time of the nonlinearity resulting from the saturated absorption thus does not have a single well-defined value, although our results, and the results of several previous workers,[3,9] are consistent with an effective response time of $\tau = 0.1$ sec. A saturable absorber such as that shown in Fig. 1 can be characterized by a saturation intensity given by $I_s = \hbar\omega / \sigma_0 \tau$, which has a value of approximately 15 mW cm^{-2} for the fluorescein in a boric acid glass system.

In this paper, we report the results of several experimental and theoretical investigations of the nonlinear optical behavior of fluorescein-doped boric acid glass. In particular, we have studied polarization-dependent saturation, optical phase conjugation by degenerate four-wave mixing, and two-beam coupling in this material.

FABRICATION

Although several methods for the preparation of fluorescein-doped boric acid glass have been presented in the literature,[9–11] we have found the most simple and trouble-free method to be that first described by Todorov *et al.*[9] A mixture of boric acid and fluorescein dye ($10^{-3}M$ concentration) is heated in a test tube to a liquid state by a Bunsen burner. A small amount of the fluorescein-doped boric acid glass is then sandwiched be-
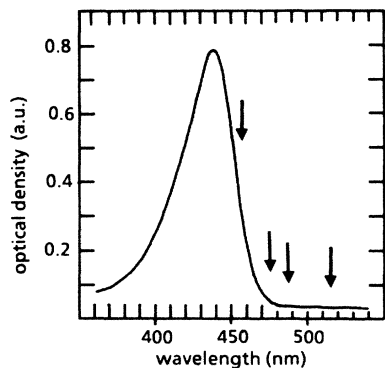
FIG. 2. Optical absorption in arbitrary units of fluorescein-doped boric acid glass. The arrows mark the wavelengths of prominent argon-ion laser lines.

tween microscope slides preheated to a temperature of $\sim 160\,^\circ$C and is pressed to a thickness of $\sim 100\ \mu$m. Samples thicker than $\sim 300\ \mu$m crack severely upon cooling. The small-intensity absorption length $(\alpha_0 l)$ of the samples can be controlled by varying either the thickness or the concentration of the sample. Figure 2 shows the absorption spectrum of boric acid glass doped with fluorescein at a concentration of $\sim 10^{18}$ molecules cm^{-3}. Superimposed upon the absorption spectrum are arrows denoting the wavelengths of prominent argon-ion laser lines.

POLARIZATION-DEPENDENT SATURATED ABSORPTION

Although the unsaturated absorption coefficient of fluorescein-doped boric acid glass is independent of the state of polarization of the applied radiation, the rate at which the absorption saturates is found to depend on the polarization state. In this section, we show that this effect is a consequence of the fact that the fluorescein molecule is highly anisotropic in its absorption properties and is held rigidly in the boric acid host. In addition, we present experimental results that illustrate this behavior and are in excellent agreement with theoretical predictions.

In order to establish notation, we first review the theory of saturable absorption for the more common case in which the saturation behavior is not polarization sensitive, such as in the case of atomic vapors or of molecules in a liquid solution. For the case of dye molecules in a liquid solution, the molecules can reorient in rotational relaxation times that are typically much shorter than the population relaxation times. As such, the initial orientation of the molecule with respect to the applied electric field vector plays no role in determining the extent to which the molecule is excited, and the absorption coefficient of a collection of such molecules is given simply by the well-known results

$$\alpha = N\sigma \ , \tag{1}$$

where N is the number of molecules per unit volume and

where σ is the saturated absorption cross-section given by[12]

$$\sigma = \frac{\sigma_0}{1+S} \ . \tag{2}$$

Here

$$\sigma_0 = \frac{4\pi\omega\mu_0^2 T_2}{3\hbar c(1+\delta^2)} \tag{3}$$

is the unsaturated absorption cross section,

$$S = \frac{S_0}{1+\delta^2} \tag{4a}$$

is the saturation parameter which characterizes the extent to which the absorption has been reduced, and S_0 is the saturation parameter for the case of exact resonance between the molecule and the exciting field. In terms of microscopic parameters, S_0 is given by

$$S_0 = \frac{4\mu_0^2 E_0^2 T_1 T_2}{3\hbar^2} \ , \tag{4b}$$

where μ_0 is the magnitude of the transition dipole moment, T_1 is the population relaxation time, T_2 is the dipole dephasing time, and $\delta = (\omega - \omega_0)T_2$ is the normalized detuning of the laser frequency ω from the resonance frequency ω_0. We have adopted the convention that the electric field experienced by a particular molecule is given by

$$\mathbf{E}(t) = \hat{\epsilon}E_0 e^{-i\omega t} + \text{c.c.} \ , \tag{5}$$

where $\hat{\epsilon}$ is the complex polarization unit vector.

In contrast to the case just treated, we now show that anisotropic molecules that are randomly oriented but are not allowed to rotate freely, such as fluorescein in boric acid glass, have a saturated absorption coefficient that is polarization dependent. For this case, the absorption coefficient is calculated by performing an angular average of the absorption cross section σ_f for a molecule of fixed orientation:

$$\alpha = N\langle\sigma_f\rangle_{\theta,\phi} \ . \tag{6}$$

We next make the simplifying assumption, which is known to be highly accurate for linearly conjugated organic molecules,[6] that the molecule can absorb only that component of the incident radiation that is polarized along one particular molecular axis. We can therefore write the molecular transition dipole moment as $\mu = \mu_0\hat{\mu}$, where the unit vector $\hat{\mu}$ lies along this molecular axis. This unit vector can be expressed in the laboratory coordinate system $(\hat{x},\hat{y},\hat{z})$ as

$$\hat{\mu} = \hat{x}\sin\theta\cos\phi + \hat{y}\sin\theta\sin\phi + \hat{z}\cos\theta \ . \tag{7}$$

Since only the component of $\hat{\epsilon}$ parallel to $\hat{\mu}$ can induce an optical transition, the saturated absorption cross section of a molecule fixed in position is given by

$$\sigma_f = \frac{3\sigma_0 |\hat{\mu}\cdot\hat{\epsilon}|^2}{1+3|\hat{\mu}\cdot\hat{\epsilon}|^2 S} \ , \tag{8}$$

where σ_0 and S are given by Eqs. (3) and (4).

We now calculate the absorption coefficient as predicted by Eqs. (6)—(8) for the limiting cases of linearly and circularly polarized light. For linearly polarized light we choose the electric field polarization unit vector to be along the $\hat{z}$ direction. We then have $|\hat{\mu}\cdot\hat{\epsilon}|^2=\cos^2\theta$, and the absorption coefficient for linearly polarized light is given by

$$\alpha_{\text{linear}}=N\sigma_0\frac{3}{4\pi}\int_0^{2\pi}\int_0^{\pi}\frac{\cos^2\theta\sin\theta}{1+3S\cos^2\theta}d\theta\,d\phi$$

$$=\frac{N\sigma_0}{S}\left[1-\frac{1}{\sqrt{3S}}\tan^{-1}\sqrt{3S}\right].\qquad(9)$$

For circularly polarized light, the polarization unit vector is represented by $\hat{\epsilon}=(\hat{x}+i\hat{y})/\sqrt{2}$, and hence $|\hat{\mu}\cdot\hat{\epsilon}|^2=\frac{1}{2}\sin^2\theta$. The saturated absorption coefficient for circularly polarized light is then given by

$$\alpha_{\text{circular}}=3N\sigma_0\frac{1}{8\pi}\int_0^{2\pi}\int_0^{\pi}\frac{\sin^2\theta\sin\theta}{1+\frac{3S}{2}\sin^2\theta}d\theta\,d\phi$$

$$=\frac{N\sigma_0}{2S\sqrt{A}}\left[2\sqrt{A}+(A-1)\ln\left|\frac{1+\sqrt{A}}{1-\sqrt{A}}\right|\right],\qquad(10)$$

where $A=3S/(2+3S)$. We note that the saturated absorption coefficient for linearly polarized light given by Eq. (9) is different from that for circularly polarized light given by Eq. (10) and that both are different from that given by Eqs. (1) and (2) for light of arbitrary polarization for freely rotating molecules. We also note that for all three cases the absorption coefficient approaches the value $N\sigma_0$ as the intensity and hence S approaches zero, which demonstrates that in the linear regime the absorptive response is polarization insensitive.

We have performed an experiment to verify the existence of polarization-dependent saturation in fluorescein-doped boric acid glass. The experiment was conducted using the 4579Å output of an argon-ion laser. This wavelength was chosen because it is close to the peak-absorption wavelength of our material and hence could most readily saturate the transition. The laser output was passed through a Pockels cell followed by a linear polarizer in order to vary the intensity of the laser beam. The beam was then passed through a second Pockels cell which controlled the polarization state of the beam. The transmission of a 200-μm-thick sample containing $\sim10^{18}$ molecules cm^{-3} (yielding an unsaturated absorption of $\alpha_0 l=4.6$) was measured as a function of the laser intensity for both linearly and circularly polarized light. The results are shown in Fig. 3. It is seen that the material is more readily saturated by linearly polarized than by circularly polarized light. This behavior occurs because, at a given intensity, linearly polarized light has a larger peak field strength than does circularly polarized light, and hence interacts more strongly with those molecules oriented along its polarization direction. The transmittance for elliptically polarized light lies between the values measured for linear and circular polarization. The solid lines shown in Fig. 3 represent the transmittance predicted
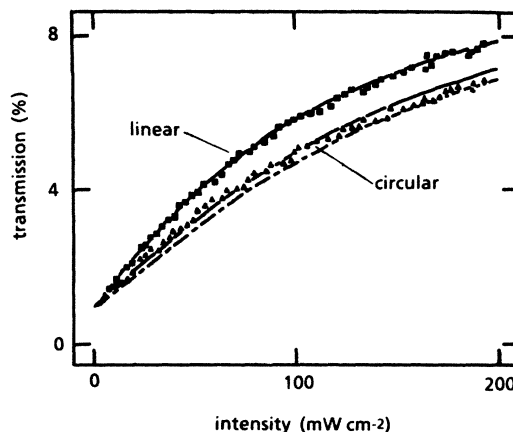

FIG. 3. Transmission of fluorescein-doped boric-acid glass plotted as a function of the laser intensity for linearly and circularly polarized light at a wavelength of 4579 Å. The solid lines are theoretical predictions for the following values of the parameters: $\alpha_0 l=4.76$, $\alpha_e l=2.26$, and $S=18$ at the maximum intensity of 200 mW cm^{-2}. The dashed line is the theoretical transmission for an ensemble of freely rotating molecules under the same set of conditions.

theoretically, including the effects of propagation and excited-state absorption. The predicted transmission is obtained by numerically integrating the differential equation for the intensity I:

$$\frac{dI}{dz}=-(\alpha+\alpha_e)I,\qquad(11)$$

where α is the ground-state absorption coefficient calculated above and $\alpha_e=N_e\sigma_e$ is the excited state absorption coefficient. Here σ_e is the excited-state absorption cross section and N_e is the excited-state population given by $N_e=N(1-\langle\sigma_f\rangle/\sigma_0)$. Very good agreement between theory and experiment is obtained through use of the values $\alpha_0 l=N\sigma_0 l=4.76$, $\alpha_e l=2.26$, and $S=1$ corresponding to an intensity of 11 mW cm^{-2}. Also shown in Fig. 3 for the purpose of comparison is the predicted transmission of a collection of freely rotating molecules for the same values of the parameters. We note that this predicted transmission saturates more slowly than for either case with fixed molecules. This behavior occurs because, for any incident polarization, the light interacts with the molecules by means of the rotationally averaged absorption cross section which is 3 times smaller than the maximum possible value that can occur when the molecules are fixed. This effect is qualitatively different from the anisotropic absorption saturation measured by Shank and Ippen for DODCI (3,3'-diethyloxadicarbocyanine iodide) in a liquid solution,[13] because in their experiment the molecular orientation relaxation time is of the order of picoseconds, whereas in our experiment the molecules were held rigidly.

The results of this experiment can be used to estimate the size of the nonlinearity of our material. The third-order nonlinear susceptibility can be expressed in terms of the unsaturated absorption coefficient α_0, the unsaturated refractive index n_0, and the saturation intensity I_s by

$$\chi^{(3)}=(n_0^2 c^2\alpha_0/24\pi^2\omega I_s)(\delta+i) \ ,$$

which, for $\alpha_0=238\,\text{cm}^{-1}$, $I_s=11$ mW/cm^2, and $\delta=1$ gives $\chi^{(3)}=2(1+i)$ esu or $|\chi^{(3)}|\sim 3$ esu.

OPTICAL PHASE CONJUGATION

It has been shown by several groups that it is possible to observe phase conjugation by degenerate four-wave mixing using the nonlinearity of saturable absorption in solids doped with organic molecules.[4,9,14] Because the line-center saturation intensity of fluorescein-doped boric acid glass is only ~ 15 mW cm^{-2}, it is possible to observe four-wave mixing using even the unfocused output of a continuous-wave laser.[9,15] In order to make an independent measurement of the nonlinear susceptibility of our material, we have measured the phase-conjugate reflectivity using the standard geometry of degenerate four-wave mixing with counterpropagating pump beams of equal intensity at a wavelength of 4765 Å. This wavelength produced better phase-conjugate reflectivity than any other argon-ion laser wavelength, because it is close enough to the line center of the absorption band to allow significant resonant enhancement of the nonlinear susceptibility, yet is sufficiently detuned to avoid serious absorption losses. The angle between the forward pump and the probe was 7°. The measured phase-conjugate reflectivity is shown in Fig. 4 plotted as a function of the intensity of either pump beam for several samples of various unsaturated absorption lengths ($\alpha_0 l=0.30$, 0.50, and 0.66). The data are seen to be in good agreement with the solid curves which represent theoretical predictions that include the effects of unsaturable excited-state absorption and grating washout arising from mechanical instabilities. These theoretical predictions are obtained by modifying the nondepleted pump theory of Abrams and Lind[15] to include the effects of excited-state absorption by taking the total absorption coefficient to be[16]

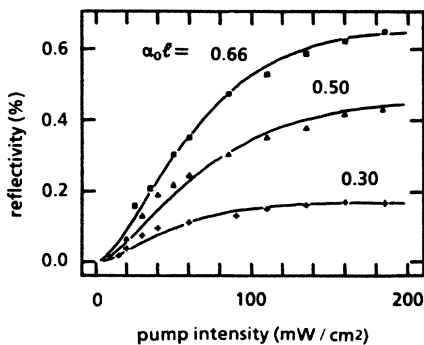$$\alpha_{\text{tot}}=\alpha_s+\alpha_u \ , \tag{12}$$

FIG. 4. Phase-conjugate reflectivity of fluorescein-doped boric acid glass at a wavelength of 4765 Å plotted as a function of the intensity of each input pump beam for three different samples. The solid lines are theoretical predictions based on a model that includes the effects of excited-state absorption and grating washout, for the following values of the parameters: $\delta=3.5$, $\alpha_e=0.23\alpha_0$, and $S=6.5$ at the maximum intensity of 200 mW cm^{-2}.

where α_s is the saturable part of the absorption coefficient given by

$$\alpha_s=\frac{N}{1+S}(\sigma_0-\sigma_e) \tag{13}$$

and α_u is the unsaturable part given by $N\sigma_e$. By performing a derivation analogous to that of Abrams and Lind,[15] we find that the phase-conjugate reflectivity is given by

$$R=K\frac{|\kappa\sin(wL)|^2}{|w\cos(wL)+(\alpha_\mu+\gamma)\sin(wL)|^2} \ , \tag{14}$$

where

$$w=[|\kappa|^2-(\alpha_u+\gamma)^2]^{1/2} \ , \tag{15a}$$

$$\gamma=N(\sigma_0-\sigma_e)\frac{1}{(1+\delta^2)}\frac{1+2S_0}{(1+4S_0)^{3/2}} \ , \tag{15b}$$

and

$$\kappa^*=iN(\sigma_0-\sigma_e)\frac{(1-i\delta)}{(1+\delta^2)}\frac{S_0}{(1+4S_0)^{3/2}} \ . \tag{15c}$$

Here L is the length of the nonlinear medium and K is a numerical factor lying in the range 0—1 which gives the effect of grating washout. When grating washout effects are not present, K has the value 1. Due to its long response time of 0.1 sec, fluorescein in boric acid glass is extremely sensitive to grating washout due to mechanical instabilities in the experimental setup. In general, two gratings of quite different spatial period contribute coherently to produce the phase-conjugation signal. Under our experimental conditions, only one of these gratings contributes effectively to the phase conjugation signal, and hence K is equal to 0.25. We have established that grating washout is in fact occurring by measuring the efficiency with which a helium-neon laser beam scatters off of the grating formed by the interference of two beams from the argon-ion laser. The grating period was varied by changing the angle between the two argon-ion laser beams. We found that the scattering efficiency fell nearly to zero for gratings of period less than 1.0 μm. The solid lines shown in Fig. 4 are theoretical curves obtained by evaluating Eq. (14) with the parameters $\delta=3.5$, $\sigma_e/\sigma_0=0.23$, $S=1$ corresponding to an intensity of 400 mW/cm^2, and $\alpha_0 l$ as given in the figure. The quality of the fit is seen to be very good. The saturation intensity at the wavelength of the experiment is thus 400 mW/cm^2, which implies a value of the saturation intensity at line center of $I_s/(1+\delta^2)=30$ mW/cm^2, which is in fair agreement with the value inferred from the polarization-dependent saturation experiment. The discrepancy probably results from our use of a single relaxation time in modeling these experiments. For the sample with $\alpha_0 l=0.66$, the nonlinear susceptibility is estimated using the values given above to be $\chi^{(3)}=0.05$ $(1+0.3i)$ esu or $|\chi^{(3)}|\sim 0.05$ esu. This value is smaller than that measured in the polarization-dependent saturation experiment both because the laser detuning was larger in the present case and because the fluorescein concentration was lower in the sample used in the present case.

TWO-BEAM COUPLING

We have observed two-beam coupling resulting from the nonlinearity of saturable absorption in fluorescein-doped boric acid glass. Two-beam coupling is the transfer of energy from one beam to another through a nonlinear interaction.[17] This process has been studied extensively for the case of a nonlinearity due to the photorefractive effect in barium titanate, and it is found that a large fraction of the power in one laser beam can be transferred to the other in a single pass through a few millimeters of crystal.[18,19] In these exeriments, the two laser beams have the same optical frequency; the sense of energy flow is determined by the orientation of the two beams with respect to the positive c axis of the barium titanate crystal. The origin of two-beam coupling in a photorefractive crystal is that the two incident waves interfere to form a standing-wave intensity pattern which, due to the nature of the photorefractive effect, produces a refractive-index grating that is shifted in phase with respect to the intensity distribution. This phase shift is necessary to break the symmetry between the two beams and allow a net power transfer between them.

Two-beam coupling can also occur for the nonlinearity of saturable absorption, as long as some means is provided for shifting the nonlinear response grating relative to the intensity pattern that formed it. This shift can result either from a motion of the nonlinear material (due, for instance, to vibrations) with respect to a fixed intensity pattern or from a motion of the intensity pattern with respect to the material, as will occur if the two beams have slightly different frequencies. In our experiment, a frequency difference between the two beams of ~ 0.1 Hz was induced by reflecting one of the beams off a mirror that was translated at a constant velocity by a piezeoelectric transducer (PZT).

Two-beam coupling in a saturable absorber can be described analytically by an extension of the formalism of Silberberg and Bar-Joseph,[17] which applied to the case of a Kerr nonlinearity. In order to include the effects of absorption, we write the refractive index as a complex quantity

$$n = n' + in'' + (n_2' + in_2'')I \qquad (16)$$

which includes the effects of saturable absorption and dispersion. The nonlinear refractive index $n_2 = n_2' + in_2''$ is related to the nonlinear susceptibility introduced above and the linear refractive index $n_0 = n' + in''$ by $n_2 = 3\pi\chi^{(3)}/n_0$. By performing a derivation analogous to that of Silberberg and Bar-Joseph, we find that the intensity I_1 of one of the beams varies spatially as

$$\frac{dI_1}{dz} = -\alpha I_1 + 2n_2'' k (I_0 + I_1)I_1$$

$$+ 2n_2'' k \frac{I_0 I_1}{1+\Delta^2} + 2n_2' k \frac{\Delta}{1+\Delta^2} I_0 I_1 , \qquad (17)$$

where k is the propagation constant of the light and where $\Delta = (\omega_0 - \omega_1)\tau$, ω_0 and ω_1 being the frequencies of the two interacting beams and τ being the response time of the medium. The first term on the right-hand side of
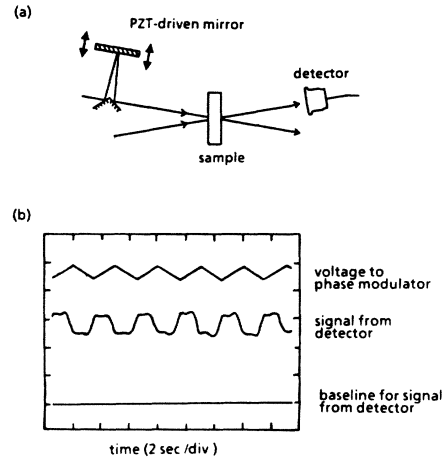

FIG. 5. (a) Experimental setup used to produce two-beam coupling. The moving mirror introduces a small frequency difference between the two beams that interfere within a sample of fluorescein-doped boric acid glass. (b) Oscilloscope trace showing the periodic variation of the intensity of one of the transmitted beams.

Eq. (17) represents linear absorption, the second and third term predict increased transmission at high intensities due to saturation effects, and the last term, which is due to dispersion, is responsible for two-beam coupling.

To measure two-beam coupling in fluorescein-doped boric acid glass, the experimental setup shown in Fig. 5(a) was used. Two beams of wavelength 4880 Å are formed by passing the output of an argon-ion laser through a beam splitter. The beams cross at an angle of 5° within the sample. The 4880-Å line was found to give better coupling than any other line of the argon-ion laser. As shown above, only the real part of n_2 leads to two-beam coupling, and hence it is desirable to work far in the wing of the absorption line. The samples used in our experiment had a value of n_2 given approximately by $n_2 = 0.23(1+0.2i)$ esu. By alternately moving the mirror in and out at constant velocity v, a periodic positive and negative frequency difference $\Delta\nu = \pm(2v/c)\nu\cos\theta$ is intro-
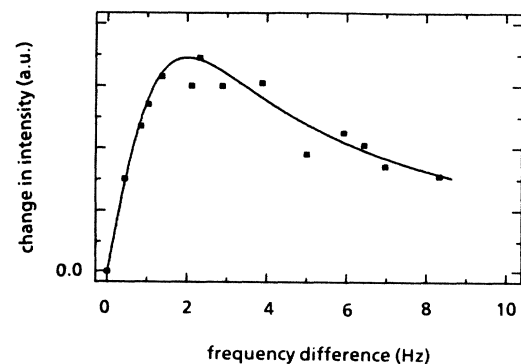

FIG. 6. Change in intensity of one of the transmitted beams due to two-beam coupling plotted in arbitrary units as a function of the frequency difference between the beams. The solid line shows the frequency dependence predicted by Eq. (17) for the value $\tau = 100$ ms.

duced between the two beams, where v is the optical frequency. Following transmission through the fluorescein-doped glass, the intensity of either beam is observed to alternatively increase and decrease by a constant amount. Figure 5(b) shows an oscilloscope trace of both the ramp voltage to the PZT and a detector output representing the intensity of one of the transmitted beams. Figure 6 shows a plot of the change in intensity of one of the beams due to two-beam coupling plotted as a function of the frequency difference between the two beams. The frequency difference was measured interferometrically. The solid line is a theoretical fit of the data to the dependence on the frequency difference predicted by the last term in Eq. (17). Good agreement is obtained through use of the value $\tau = 100$ ms.

DISCUSSION

In order to verify that the nonlinearity giving rise to the effects described in this paper is in fact saturated absorption and is not due to thermal effects, we have repeated the phase-conjugation and two-beams coupling experiments using samples of Rhodamin 6G in boric acid glass, for comparable values of $\alpha_0 l$. These samples thus possess thermal characteristics similar to that of fluorescein-doped boric acid glass, but do not display long-lived fluorescence. Hence the thermal contribution to $\chi^{(3)}$ should be the same for the two samples, although that due to saturated absorption should be much smaller for the samples containing Rhodamin 6G. Neither experiment produced a measurable output signal using the Rhodamin

6G samples. The output signal, if present, was less than 10^{-3} of that produced by the fluorescein-doped boric acid glass. We thereby conclude that thermal effects did not contribute appreciably to the nonlinear optical processes described in this paper.

In summary, we have shown that fluorescein-doped boric acid glass displays interesting and useful nonlinear optical properties. It is characterized by a $\chi^{(3)}$ susceptibility as large as 1 esu, as compared to $\sim 10^{-12}$ esu for the commonly used Kerr liquid CS_2. Since this material is isotropic on a macroscopic scale, its linear optical properties are polarization independent; however, its nonlinear optical properties have been shown to depend on the polarization state of the saturating field because the individual molecules are not free to reorient during the relevant interaction times. We have also shown that it is possible to perform phase conjugation and two-beam coupling using the nonlinear response of this material even for unfocused continuous-wave laser beams. The results of these experiments are in good agreement with theoretical predictions.

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