Parametric study of photorefractive beam coupling in $BaTiO_3$ at multiple wavelengths

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Abstract. Photorefractive two-beam coupling and energy transfer in $BaTiO_3$ are studied at multiple wavelengths using He-Cd, He-Ne, and diode lasers. The photorefractive signal beam gain is measured as a function of the pump beam intensity, beam ratio intensity, spatial frequency of the grating, and angle between the grating vector and *c* axis of the crystal. The exponential gain coefficient is calculated from signal beam gain, and its dependence on the same parameters is studied. The dependence of the signal beam gain and exponential gain coefficient on spatial frequency are also evaluated theoretically and found to be in good agreement with the experimental data. Also, the signal beam gain is studied as a function of wavelength, and a very high value is obtained at 441.6 nm. Figure-of-merit parameters such as the maximum change in the refractive index, the space charge field at saturation, the trap density of the charge carriers, and the photorefractive sensitivity of the crystal are calculated from the experimental data. Since the absorption in $BaTiO_3$ varies strongly with wavelength, its influence has been included in the calculations of the figure-of-merit parameters. The results are discussed in the light of the present understanding of photorefractive phenomena in crystals. [S0091-3286(00)01302-7]

Subject terms: photorefractive effect; two-beam coupling; signal beam gain; exponential gain coefficient; figure-of-merit parameters; $BaTiO_3$.

Paper 980373 received Sep. 26, 1998; revised manuscript received Mar. 29, 1999; accepted for publication July 30, 1999.

1 Introduction

Photorefractive (PR) materials are being studied with considerable interest in view of their potential for a variety of applications in areas such as optical information storage and data processing, phase conjugation, image processing, coherent image amplification, optical logic operations, dynamic holography, optical interconnects, and neural networks.[1–6] $BaTiO_3$ is one of the most efficient PR materials known, and several studies have been made on the basic and applied aspects of the photorefractive response of this crystal.[7–10] However, a complete picture is yet to emerge on the physical processes and mechanisms underlying its PR behavior, so that further efforts in this direction are called for.

Though two-beam coupling in $BaTiO_3$ has been studied at Ar^+-laser wavelength, such systematic studies at other laser wavelengths are sparse. Also, not much work has been reported on experimental estimation of the figure-of-merit parameters in this crystal and comparison with those evaluated on the basis of Kukhtarev's model.[11,12] A study of beam coupling at multiple wavelengths is expected to yield additional insight into the physical processes underlying the PR behavior of this crystal and also to probe its efficiency in applications at various regions of the optical spectrum. $BaTiO_3$ has been studied at 442 nm by Klein and Schwartz[13] to find the sign of the dominant charge carriers.

Here, we present a systematic study on the characterization and comparative study of two-beam coupling in $BaTiO_3$ at 441.6, 632.8, and 780 nm using He-Cd, He-Ne, and semiconductor diode lasers, respectively, by measuring the signal gain as a function of spatial frequency, crystal orientation, pump beam intensity, and beam intensity ratio. The exponential gain coefficient is obtained from these values and also independently evaluated theoretically as a function of spatial frequency. The signal beam gain is calculated theoretically, incorporating the effect of optical absorption. The results are found to be in agreement with the experimentally obtained values. The figure-of-merit parameters for PR response—namely, the change in steady-state refractive index (δn_{ss}), the space charge field at saturation (E_{sc}), the trap density N_A, and the photorefractive sensitivity S—are evaluated using the experimental data at the three wavelengths. The effect of optical absorption at each wavelength of operation is incorporated in the calculations. Such a study at multiple wavelengths addressing all these aspects is new to the best of our knowledge.

2 Two-Beam Coupling and Energy Transfer

The photorefractive effect in $BaTiO_3$ has been studied using two-beam coupling. In this technique, two beams of the same frequency are superposed in the crystal, producing a spatially periodic irradiance pattern. As the resultant refractive index pattern is not in phase with the irradiance pat-

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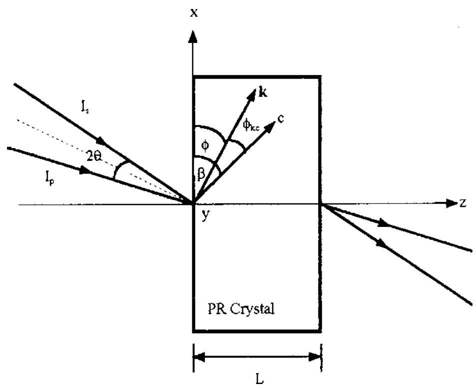

Fig. 1 Two-beam coupling configuration recording geometry, showing $I_p(0)$, $I_s(0)$, the incident angle 2θ, and the crystal orientation about the grating vector **k**.

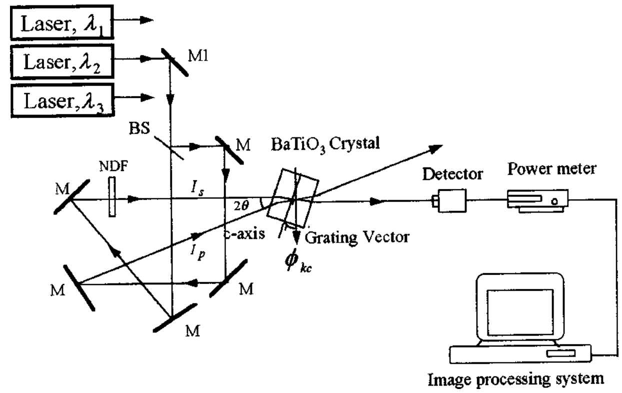

Fig. 2 Experimental setup for the two-beam coupling studies at multiple wavelengths.

tern, energy is transferred from one beam to the other. The theory for photorefractive two-beam coupling has been developed by Kukhtarev et al.[11,12] For the beam geometry and the crystal orientation shown in Fig. 1, the transmission of the signal beam is described by

$$\frac{I_s(L)}{I_s(0)}=\frac{I_p(0)\exp(\Gamma L)}{I_p(0)+I_s(0)\exp(\Gamma L)}, \tag{1}$$

where $I_s(0)$ and $I_s(L)$ denote the signal beam intensity before and after passing through the crystal, respectively, $I_p(0)$ is the pump beam intensity, Γ is the exponential gain coefficient, and L is the interaction length. For the case of negligible depletion of the pump wave $[I_s(0)\exp(\Gamma L) \ll I_p(0)]$, Eq. (1) reduces to

$$I_s(L)=I_s(0)\exp(\Gamma L). \tag{2}$$

When absorption is significant and assumed independent of intensity, Eq. (2) is rewritten as[14]

$$I_s(L)=I_s(0)\exp[(\Gamma-\alpha)L]. \tag{3}$$

The signal beam gain γ_0, when absorption is taken into account, is defined as

$$\gamma_0=\frac{I_s(L)\ \text{with pump beam}}{I_s(L)\ \text{without pump beam}}=\frac{(1+q)\exp[(\Gamma-\alpha)L]}{1+q\exp(\Gamma L)}, \tag{4}$$

where $q=I_s(0)/I_p(0)$ is the intensity ratio of the two incident beams before passing through the crystal. Equation (4) is valid when α is independent of intensity, which is the case in our experiments.[15] When depletion of the pump beam can be neglected but absorption is taken into account, $\gamma_0=\exp[(\Gamma-\alpha)L]$. In the absence of an applied electric field, the phase of the periodic space charge electric field is shifted from that of the irradiance distribution by $\pi/2$. For this case, the exponential gain coefficient is given by[16]

$$\Gamma=\frac{2\pi n_b^3 r_{\text{eff}} E_D E_Q}{\lambda(E_Q+E_D)}, \tag{5}$$

where E_D, E_Q are the diffusion field and saturation field, respectively, and are given by

$$E_D=\frac{2\pi k_B T}{\Lambda_g e},\quad E_Q=\frac{eN_A\Lambda_g}{2\pi\epsilon\epsilon_0}; \tag{6}$$

r_{eff} is the effective linear electro-optic coefficient; ϵ is the dielectric tensor, which can be calculated from experimental data[17]; λ is the laser wavelength; 2θ is the angle between the interfering beams outside the crystal; $\Lambda_g=\lambda/(2\sin\theta)=2\pi/k$ is the grating spacing; and **k** is the grating vector. In the case of zero applied electric field, the maximum value of exponential gain coefficient Γ occurs for the value of Λ_g for which $E_D=E_Q$. This value of Λ_g is referred to as the Debye length and is useful in determining N_A, the trap density in a crystal.[16]

The exponential gain coefficient Γ is related to the refractive index modulation as $\Gamma=4\pi\,\delta n_{ss}/\lambda$, where $\delta n_{ss}=\frac{1}{2}n_b^3 m r_{\text{eff}} E_{\text{sc}}$ is the maximum value of the index modulation due to the photorefractive beam coupling. The space charge field E_{sc} at saturation is given by $E_{\text{sc}}=E_Q E_D/(E_Q+E_D)$ in the case of zero applied field.

3 Experimental Setup

Figure 2 shows the experimental setup. A He-Cd laser (Omnichrome model 4074-40P) at 40 mW, a He-Ne laser (Spectra Physics model 127) at 35 mW, and a semiconductor diode laser (Spindler and Hoyer model DC 25C) at 25 mW are used as the sources for the present work. The actual laser source for a given experiment is selected by translation of mirror M_1 to the appropriate locations. A strong pump beam (I_p) and a weak signal beam (I_s) are obtained using a beamsplitter (BS) and a set of mirrors. The intensity I_s is varied with the help of a neutral-density filter (NDF). These beams are adjusted to have the same path length and are superposed in the $BaTiO_3$ crystal (Sanders Laboratory, USA) of dimensions 5 mm×5 mm×5 mm so as to form a refractive index grating. The crystal is kept at the center of a rotation stage. Intensities of the outcoming beams are measured using a digital power meter (Newport model 835) interfaced with a computer. The experimental geometry is so chosen that the c axis of the crystal is oriented at an

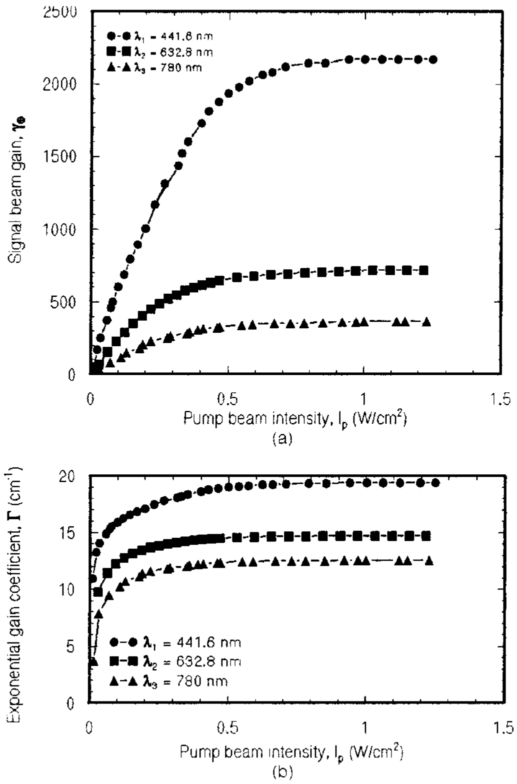

Fig. 3 Variation of (a) signal beam gain and (b) exponential gain coefficient as a function of pump intensity.

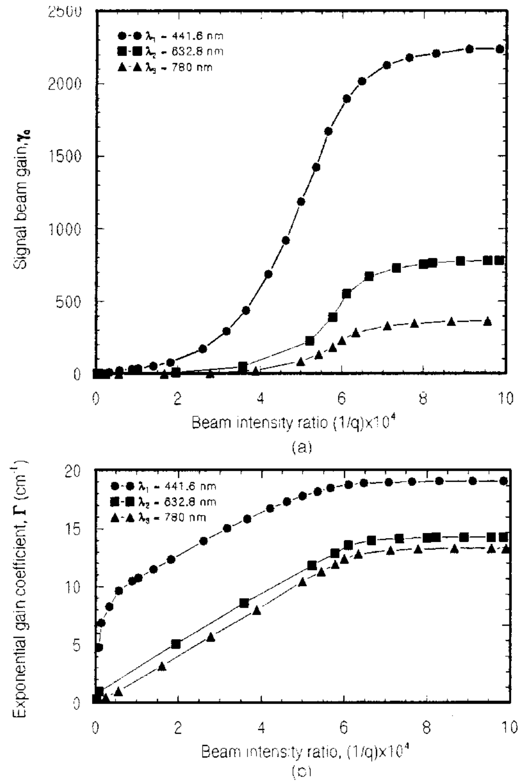

Fig. 4 Variation of (a) signal beam gain and (b) exponential gain coefficient as a function of input beam intensity ratio *q*.

angle around 19 deg with respect to the grating vector. This angle is referred to as ϕ_{kc}, and the dependence of signal beam gain on it is studied separately. The angle 2θ between the two beams outside the crystal is chosen so as to provide maximum gain at each wavelength; this angle is around 30 deg, corresponding to fringe spacings $\Lambda_g = 0.9$, 1.22, and 1.41 μm for wavelength values 441.6, 632.8, and 780 nm, respectively. The maximum value of pump intensity used in the present experiments is 0.85 W/cm^2, and the input beam intensity ratio q is less than 10^{-4} in most cases.

The optical absorption spectra are recorded on a Hitachi spectrophotometer (model U3400), and the measured values of the optical absorption coefficient are 3.9, 1.6, and 0.8 cm^{-1} at 441.6, 632.8, and 780 nm, respectively. The crystal from Sanders Laboratory is expected to have Fe impurities in the range of 100 to 150 ppm.[15] The value of the absorption coefficient of a crystal with Fe content of 150 ppm, obtained from the measured coefficient of 441.6 nm for various crystals with different Fe contents, is lower than 3.9 cm^{-1}. Assuming that the values of α obtained in Ref. 13 are correct, our crystal may have Fe impurities more than 180 ppm.

4 Results and Discussion

The signal beam gain γ_0 has been measured as a function of (1) pump beam intensity, (2) beam intensity ratio, (3) spatial frequency of the refractive index grating, and (4) angle between the grating vector and the c axis of the crystal. The exponential gain coefficient Γ is calculated using Eq. (4) in each case.

4.1 *Characterization*

Figures 3(a) and 3(b) show the variation of the signal beam gain γ_0 and the exponential gain coefficient Γ respectively as functions of pump intensity I_p at the three wavelengths studied, for a beam intensity ratio of less than 10^{-4}. Both γ_0 and Γ increase initially and then saturate in all cases. Such gain saturation behavior was observed earlier at Ar^+ laser wavelength and explained on the basis of the photoconduction model.[18] The saturation values γ_s of the signal beam gain are 2200, 700, and 365 at 441.6, 632.8, and 780 nm, respectively, and the corresponding values of Γ are 19.33, 14.72, and 12.56 cm^{-1}, respectively, when absorption is taken into account. The gain at 441.6 nm is much larger than that at the other wavelengths. Data from these experiments are used to evaluate the figure-of-merit parameters discussed in the next section.

Figures 4(a) and 4(b) show the dependence of γ_0 as a function of the inverse beam intensity ratio q. Initially the variation of γ_0 with q is slow, followed by a sharp rise and then a saturation for all wavelengths; the saturation value is much larger at 441.6 nm than at other wavelengths. This plot helps us to estimate the ideal beam ratio for maximum signal gain.

The gain is also studied experimentally as a function of the spatial frequency k obtained by changing the angle 2θ between the beams outside the crystal. Figures 5(a) and 5(b), respectively, show the dependence of γ_0 and Γ on k. The experimental data shown by symbols are superposed on theoretically evaluated curves. The exponential gain Γ is also evaluated theoretically as a function of spatial fre-

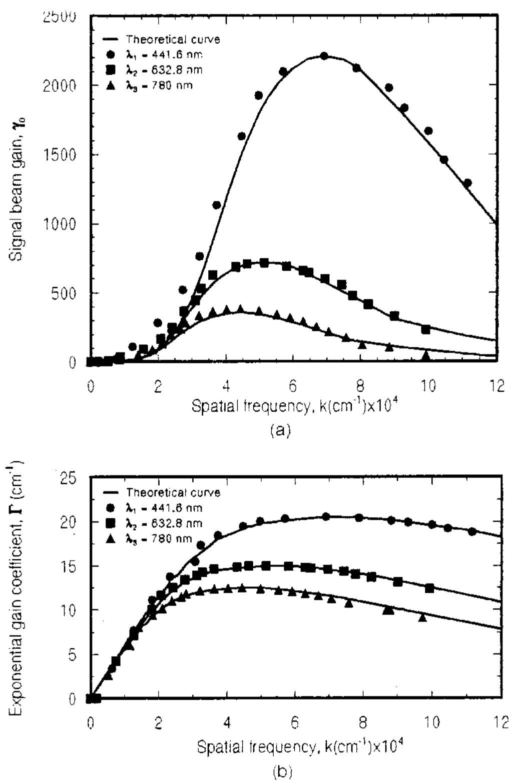

Fig. 5 Variation of (a) signal beam gain and (b) exponential gain coefficient as a function of spatial frequency *k*. The experimental data are denoted by symbols, and the theoretical evaluation is shown by a solid line.

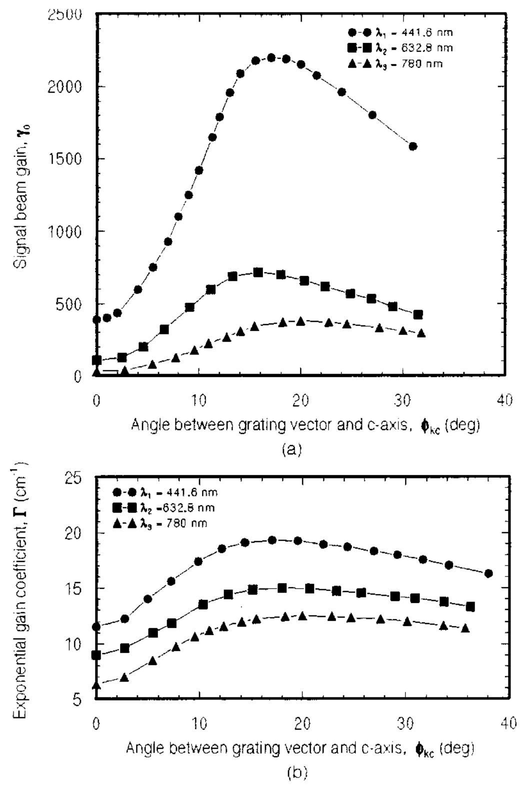

Fig. 6 Variation of (a) signal beam gain and (b) exponential gain coefficient as a function of the angle ϕ_{kc} between the grating vector **k** and the *c* axis of the crystal.

quency k using Eq. (5) and data available from literature.[19–21] The signal beam gain γ_0 is calculated from Γ using Eq. (4). The plots exhibit broad maxima. The experimental data are found to agree well with the theoretically evaluated plots, indicating the validity of Kukhtarev's model in understanding PR wave-mixing phenomena in this crystal at all the three wavelengths used.

Figures 6(a) and 6(b) show the variation of γ_0 and Γ on the angle ϕ_{kc} between the c axis and the grating vector inside the crystal, respectively. The maximum energy transfer is found to occur at 441.6 nm. The optimum of signal beam gain is obtained for ϕ_{kc} of 17, 18, and 22 deg at 441.6, 632.8, and 780 nm, respectively. Its value for the Ar^+ laser is reported to be around 20 deg.[18]

4.2 *Evaluation of Figure-of-Merit Parameters*

Some important parameters, such as the saturation change in refractive index (δn_{ss}), the space charge field in the crystal (E_{sc}), the trap density N_A, and the photorefractive sensitivity S, have been generally considered as figure-of-merit parameters describing PR response in the context of its potential use in optical data processing.[19] We have evaluated and compared these parameters for $BaTiO_3$ at the three wavelengths. Here δn_{ss} is calculated from the measured values of Γ. The effective electro-optic coefficient r_{eff} and hence the space charge field E_{sc} are calculated from the experimental data. The trap density N_A is calculated using the values of δn_{ss} and E_{sc}. The PR sensitivity S is defined as the change in index of refraction per unit absorbed photon energy per unit volume, and is given by $S = \Delta n / \alpha I_0 t$. It is calculated using the experimental data. The calculated figure-of-merit parameters are listed in Table 1.

The optical absorption coefficient α of the crystal is larger at 441.6 nm than at the other wavelengths. Correspondingly the value of δn_{ss} is smaller at this wavelength, which is the trend to be expected from diffraction efficiency considerations. Correspondingly, the value of E_{sc} is larger for shorter wavelengths. The value of N_A is found to be of the same order of magnitude at the three wavelengths, as expected, though the actual values are larger at shorter wavelengths. The values are of the same order of magnitude as obtained at Ar^+-laser wavelength.[18] The larger values of N_A at shorter wavelengths could probably be due to generation of additional carriers through photoexcitation processes. The values of the trap density help us to estimate the response time, which, under appropriate conditions

Table 1 Figure-of-merit parameters for $BaTiO_3$.

λ (nm)	δn_{ss}	E_{sc} (V cm^{-1})	N_A (m^{-3})	S (cm^3 kJ^{-1})
441.6	6.79×10^{-5}	798	3.0×10^{22}	17.10
632.8	7.41×10^{-5}	561	2.7×10^{22}	35.18
780	7.79×10^{-5}	552	2.0×10^{22}	50.16

(e.g., short recombination times), reduces to the dielectric relaxation time[19] and can be written as

$$\tau_{\mathrm{di}}=\left(\frac{\epsilon\gamma_R}{4\pi e\mu s}\right)\left(\frac{N_A}{N_D-N_A}\right)\frac{1}{I_0},$$

where γ_R is the recombination rate, μ is the mobility, I_0 is the average irradiance, and s is the photo-ionization cross section. This is useful parameter for applications in which the grating must be written or erased in a set time scale. The trap density is the largest at 441.6 nm, and hence we can expect a larger response time at this wavelength, which has been experimentally observed. The large values of α at higher quantum energies lead to a reduction in the PR sensitivity S. The sensitivity is found to be largest at 780 nm, where the PR gain is comparatively low. Thus, there appears to be a trade-off between PR gain and figure-of-merit considerations for optical data processing, emphasizing the need for a detailed PR characterization of materials at multiple wavelengths so as to facilitate a proper choice of wavelength and material suited for the requirements of any given application.

Acknowledgment

We are thankful to the referees, whose reviews have significantly improved the contents of the paper.

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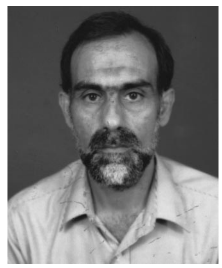

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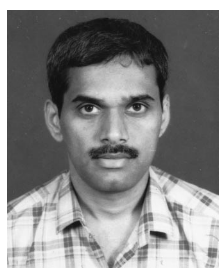

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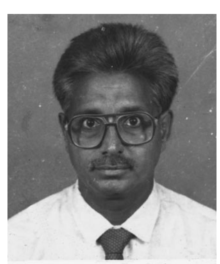

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