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Results of experiments on the dynamics of scattering in barium titanate are presented. Asymmetric scattering and centrosymmetric scattering are characterized as a function of incident-beam size, and photographs are presented showing the development of the beam as light is asymmetrically scattered. Results are compared with existing theories of photorefractive scattering and are found to be in agreement with a near-forward-scattering theory. We propose that scattering in barium titanate begins with photorefractive inhomogeneities within the crystal, and experimental support is presented.

1. INTRODUCTION

The scattering of light (not specifically involving optical phase conjugation) in $BaTiO_3$ has been investigated for many years.[1-4] Here we report on the scattering of light within $BaTiO_3$ before the onset of optical phase conjugation. We present experimental results showing the dependence of scattering on incident-beam size. We address both asymmetric scattering in the plane created by the incident beam and the *c* axis of the crystal (sometimes referred to as asymmetric self-defocusing or beam fanning) and centrosymmetric scattering, which is observed orthogonal to the plane in which the *c* axis and the input beam lie. We also investigate the distribution of light scattered out of the incident beam during the scattering process and present some observations that lend insight into the process of asymmetric self-defocusing and self-pumped phase conjugation within the crystal. We then discuss how these results apply to existing theories of scattering and asymmetric self-defocusing in $BaTiO_3$.

2. EXPERIMENTAL PROCEDURE

To investigate scattering in $BaTiO_3$ an experiment was designed to measure the influence of a change in the incident-beam diameter on the scattering process. The experimental apparatus used for this work is shown schematically in Fig. 1. The laser was an Ar^+ variable-power (20-mW maximum) multimode laser operated at 514.5, 488.0, 476.5, and 457.9 nm. After leaving the laser, the beam was directed into a polarization rotator to rotate the beam from the ordinary to the extraordinary polarization for the crystal, and then it was directed through a Glan–Thompson polarizer. A beam splitter behind the polarizer was used to reflect any phase-conjugate beam. A variable aperture of the type common in photographic lenses restricted the beam size and provided a sharp-edged beam. The beam was then focused onto the crystal by uncoated lenses of either 34- or 16-cm focal length.

The crystal was one of four single crystals of $BaTiO_3$ manufactured by Sanders Associates and poled into a single domain. Two of the crystals measured approximately 5 mm × 5 mm × 5 mm and were transparent, with a slight yellow tint; two of the crystals measured approximately 5 mm × 5 mm × 2.5 mm and had no noticeable tint to them. The crystal under observation was mounted upon a rotatable platform capable of 12-cm translation along the axis of the optical bench, upon which all the components were mounted. After the crystal, and connected to the mounting table, was a knife edge that acted as a beam stop for the beam transmitted through the crystal. The knife edge was covered with black tape to suppress the formation of a resonating cavity with the crystal. Behind the knife edge, and also connected to the translation table, was a 1-cm^2 silicon photodiode connected to a Keithly 617 electrometer measuring the short-circuit current. The photodiode was placed such that only light scattered out of the incident beam in the direction of beam fanning was recorded. Light not scattered out of the incident beam was intercepted by the knife edge.

To measure the amount of light scattered into the plane of the *c* axis and the incident beam (i.e., asymmetric self-defocusing), the knife edge was aligned in such a way that the beam transmitted through the crystal was blocked from the detector for all observed beam diameters. The incident beam was then blocked, and the crystal was flooded with light from a 75-W incandescent lamp located approximately 12 cm away. After approximately 1 min, the light was turned off, and the laser beam was unblocked. The intensity of the light reaching the detector was then monitored, and the peak and steady-state intensities were recorded. Once the intensity had reached a steady-state value, the beam was again blocked, the crystal was translated (changing the incident-beam diameter) and flooded with white light, and the procedure was repeated.

The flooding of the crystal with white light effectively homogenized the charge distribution within the crystal and ensured that one reading was not affected by the previous one. Care was taken to ensure that all components were mounted upon the optical axis of the optical bench. The precision translation mechanism upon which the crystal ta-

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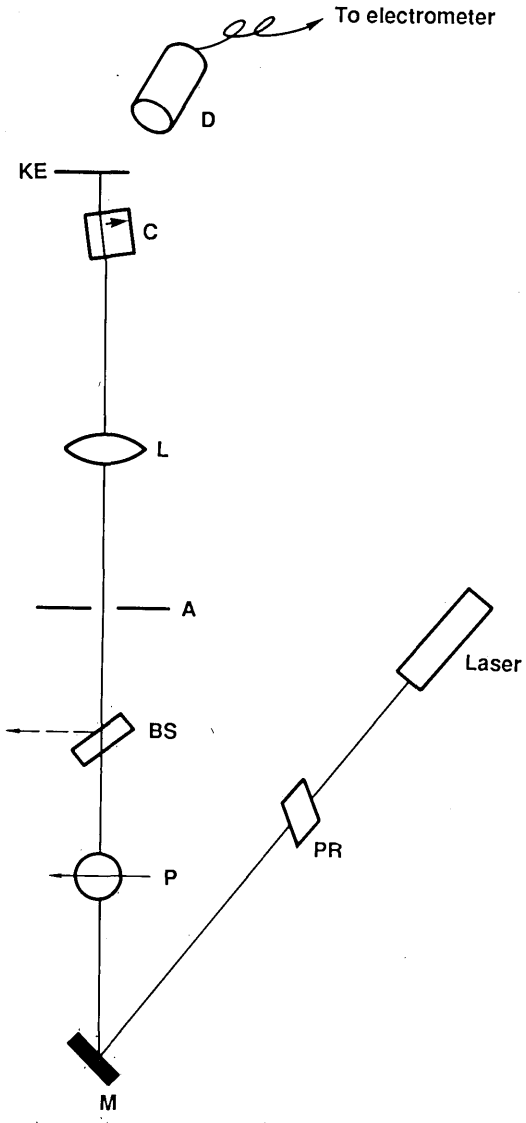

Fig. 1. Experimental arrangement using an Ar^+ laser: PR, polarization rotator; M, mirror; P, polarizer; BS, beam splitter; A, aperture; L, lens; C, $BaTiO_3$ crystal (the arrow indicates the direction of beam fanning); KE, knife edge; D, photodiode detector.

ble was mounted ensured that the beam was incident upon the crystal at the same spot for all measurements. These precautions effectively compensated for any surface inconsistencies on the crystal face, except at small laser-beam diameters ($\lesssim 50\ \mu m$).

Both peak and steady-state values of the scattered-beam intensity were recorded; however, only the peak values were used for final analysis. This decision was made because for large angles of incidence (>20°) the steady-state values, although correlated with the peak values, were inconsistent. One obvious problem with steady-state readings is the possibility of self-pumped phase conjugation that could reduce the amount of fanning recorded in the steady state. Also, it was possible (even likely) that the beam would fan across the detector, eventually coming to rest beyond the active area. Occasionally the scattered light was observed to leave the crystal face perpendicular to the face that the detector was monitoring. To reduce all these effects, the incident beam was originally permitted to enter at only small angles of the *c* axis (~2°–5°), and consistent results were obtained for various crystals, focal lengths of the lens, laser lines, and incident angles. Eventually, larger incident angles were used (~20°–40°) with similarly consistent results.

To ensure that the observed effects were not influenced by scattering from small (i.e., molecular-sized) crystal inhomogeneities, one configuration was chosen in which there was moderate fanning, and the fanned intensity was recorded for each of the available visible lines of the laser. The result was a mean ratio of incident intensity to fanned intensity that was constant to within 1% across all available wavelengths, thus indicating no resonances and no dependence of scattering on wavelength at the available laser lines.

After the data collection, the beam diameter at the face of the crystal was measured with an Aeronca Electronics Laser Blade, and the direct laser intensity was recorded. Here, the beam diameter is defined as twice the distance from the center of the beam to the 10% intensity point.

Measurements of centrosymmetric scattering for various beam diameters were made by pointing the detector toward the unobstructed crystal face that is parallel to the plane created by the incident beam and the *c* axis of the crystal (i.e., placing the photodiode above the platform holding the crystal). In this configuration the amount of light scattered into the detector is proportional to the total amount of centrosymmetrically scattered light. Black tape shielded the front surface of the crystal from the detector to block any specular reflection off surface irregularities. Transmission measurements were also made by replacing the knife edge behind the crystal with an aperture, thus permitting only the transmitted portion of the incident beam to reach the detector.

It should be noted that the effects of asymmetric scattering could influence the measurement of centrosymmetric scattering. That is, significant amounts of asymmetrically scattered light may effectively increase the incident-beam size inside the crystal, thus giving erroneous and possibly inconsistent data. To ensure that this was not the case, centrosymmetric scattering data were taken with the incident beam entering the crystal parallel to the *c* axis of the crystal so that asymmetric scattering was minimized. It was discovered, however, that this effect was small (provided that there was no self-pumped phase conjugation) because, as will be shown in the photographs of Section 3, the onset of asymmetric scattering results in a reduction of the incident-beam size as the beam propagates through the crystal. This reduction in beam size helps to compensate for the added interaction area due to asymmetric scattering.

3. RESULTS

Typical results of the asymmetric scattering measurements are shown in Fig. 2. All intensity recordings have been normalized to the peak value for comparison purposes. Although any data presented are by necessity crystal specific, the *shape* of the curves is insensitive to the laser line used, beam angle, choice of $BaTiO_3$ crystal, and divergence or convergence of the incident beam (i.e., whether the crystal is placed in front of or behind the focal point of the focusing lens).

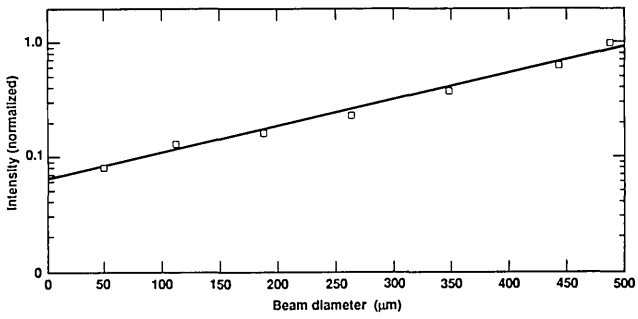

Fig. 2. Plot of normalized intensity of asymmetrically scattered light versus incident-beam diameter. The shape of the curve is insensitive to the choice of $BaTiO_3$ crystal, laser line, laser power (<20 mW), or convergence or divergence of the incident beam.

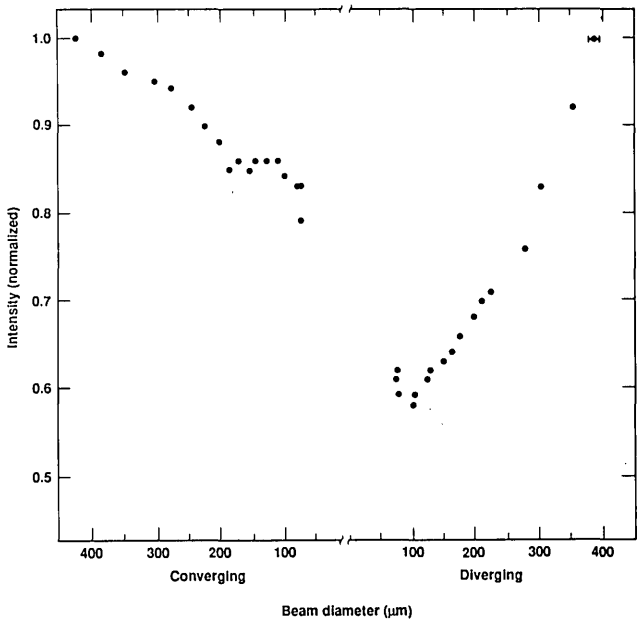

Fig. 3. Dependence of centrosymmetric scattering on incident-beam diameter as measured perpendicular to the plane created by the incident beam and the c axis of the crystal. Converging data were taken with the focal point of the lens behind the incident crystal face; diverging data were taken with the focal point in front of the incident crystal face. Except at very small beam diameters (<50 μm), the shape of the curve is insensitive to those factors mentioned in Fig. 2.

Figure 3 is a plot of the intensity observed perpendicular to the plane created by the incident beam and the c axis of the crystal for various beam diameters. Note that there is a much broader dynamic range in the intensity for diverging light (when the crystal face is placed past the focal point of the focusing lens) than for converging light (the crystal face is in front of the focal point of the focusing lens). Also of importance is that for constant laser-beam power the scattering is a minimum for the smallest beam diameters. Often, as the crystal passed through the depth of focus of the lens (beam diameter ~50 μm), there were drastic increases and decreases in the centrosymmetrically scattered intensity. This behavior was not consistent for all crystals and may have been due to surface inhomogeneities, dirt, or heating.

Removal of the knife edge and detector from behind the crystal permitted the observation of the image of the transmitted spot on a screen approximately 3 m away. Figure 4 shows the image of the near field projected directly onto photographic film at three different times after the beam was permitted to enter the crystal and before the onset of phase conjugation. Figure 4(a) is an image of the beam immediately after turn on; notice that the spot is generally undistorted. Figure 4(b) is a photograph between turn on and steady state, and Fig. 4(c) is a photograph just before the onset of self-pumped phase conjugation. It is apparent

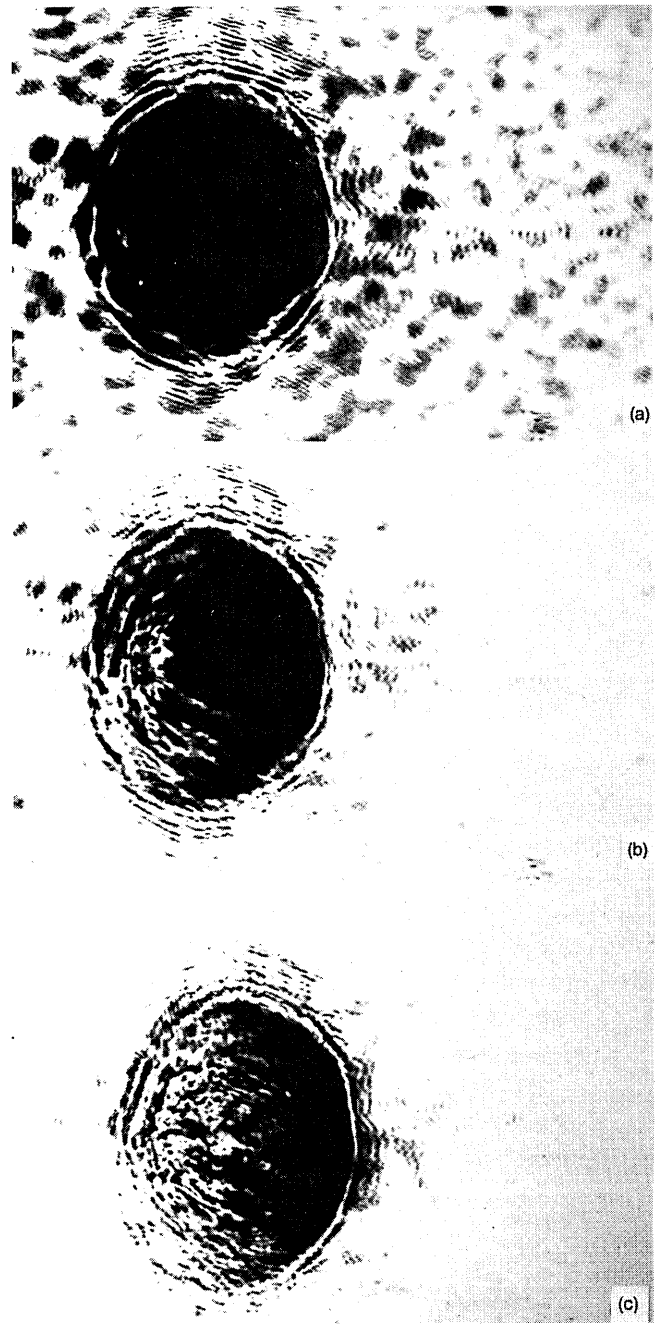

Fig. 4. Photographs of the near-field beam transmitted through the crystal at various times: (a) immediately after beam turn on, (b) during the process of beam fanning, and (c) just before the onset of self-pumped phase conjugation. Note that the scattering apparently begins at the periphery of the incident beam.

from these images that the energy of the beam was first extracted from the outside edge of the beam.

4. DISCUSSION

Asymmetric self-defocusing, or beam fanning, in $BaTiO_3$ has been attributed to an asymmetric change in the index of refraction across the incident-beam path that is due to the photorefractive effect,[1] to a nonlinear index of refraction due to an intensity-dependent frequency shift in a phase-conjugate field,[2] and to near-forward-stimulated photorefractive scattering.[4] Our data presented here and numerical simulations[5] do not support the first of these theories. The change in index of refraction along the beam path is given by

$$\Delta n = -(1/2)n^3 r_{\text{eff}} E(\mathbf{x}), \tag{1}$$

where n is the index of refraction, r_{eff} is the effective Pockels coefficient, and $E(\mathbf{x})$ is the space-charge field. The space-charge field is proportional to the gradient of the incident intensity.[1] For a Gaussian beam the intensity gradient is given by

$$\nabla I = \frac{-8P}{\pi\omega^4}(x\hat{x} + y\hat{y})\exp[-2(x^2 + y^2)/\omega^2], \tag{2}$$

where P is the total power of the beam and ω is the incident spot size.

Because, from Eq. (1), the change in index of refraction is directly dependent on the space-charge field and the space-charge field is inversely dependent on the incident-beam size, the magnitude of asymmetric self-defocusing for a given beam power should increase with a decrease in incident-beam size. However, we always observe an increase in asymmetric scattering with an increase in beam size for a constant beam power.

The exponential increase with beam diameter exhibited in Fig. 2 does support the theory of asymmetric self-defocusing by near-forward-stimulated scattering.[4] The total gain for any given noise wave in stimulated scattering is given by

$$G = \exp(gl), \tag{3}$$

where g is the intensity gain coefficient and l is the interaction length. Therefore an increase in beam diameter, which results in a linear increase in l for internally scattered waves, creates an exponential increase in the total gain G. This results in an exponential dependence of asymmetrically scattered intensity on beam diameter, as shown in Fig. 2.

The photographs of Fig. 4 also support a stimulated scattering theory. Whereas a change in the index of refraction within the entire beam path would cause beam fanning to begin at the center of the beam, where the intensity is largest, stimulated scattering may occur throughout the beam. The appearance of initial peripheral scattering is attributable to the fact that the gain paths of all stimulated beams must traverse the portion of the incident beam that lies between the initial scattering center and the beam periphery in the direction of gain (the direction of beam fanning). Because all scattered beams that experience gain take power from the incident beam along the entire path where they intersect, the amount of power lost at any point within the beam is the sum of the power taken by all the beams scattered through that point. As scattered beams compete for the power available in the incident beam, those with the longest lengths of interaction (for a given angle) win over those with shorter interaction lengths. Thus the power in the periphery of a beam is depleted first.

At the focal point of a lens the beam is so small that the effective interaction length is ~0; thus no detectable asymmetric scattering (fanning) occurs. However, the symmetric scattering does not go to, or even approach, zero. This suggests that there is another scattering mechanism within the crystal. The data presented in Fig. 3 indicate that the mechanism responsible for centrosymmetric scattering is not simply a stimulated scattering phenomenon because the magnitude of the intensity of the centrosymmetrically scattered light is not exponentially dependent on the incident-beam size. Similarly, centrosymmetric scattering is not solely scattering that is due to uniformly distributed static scattering centers within the crystal because the magnitude of the centrosymmetrically scattered light does not change as the square of the incident-beam radius (i.e., proportionally with the cross-sectional area of the incident beam). We propose that this other significant scattering mechanism is due to nonstimulated photorefractive scattering, that is, scattering due to index variations caused by the nonuniform distribution of charges within the crystal.

That the dominant scattering mechanism is due to a dynamic process may be shown by measuring the scattered intensity for a given configuration, uniformly illuminating the crystal with light, and then measuring the scattered intensity again, all without moving any part of the apparatus. The illumination of the crystal between measurements ensures that the second measurement is not affected by the first because any index variations due to the incident beam are erased and the internal charge is homogenized. For both centrosymmetric scattering and asymmetric scattering the results between the two measurements differ. This difference may be as much as almost 0.5% when the symmetric scattering is measured and a few percent when the asymmetric scattering is measured. The greater difference between two successive measurements seen when one is observing the asymmetric scattering is because the amount of light reaching the detector is strongly dependent on how and where the scattered light leaves the beam; whereas, if it is viewed orthogonally to the plane created by the incident beam and the c axis of the crystal, there may be little effect on intensity because the total amount of scattered energy remains almost the same.

We next discuss the observations of beam curvature of the scattered beams within the crystal, a phenomenon that cannot be explained by a stimulated scattering theory. A study of photographs and video tapes of many crystals during asymmetric self-defocusing taken by ourselves and others[6] has produced no instance in which beams in $BaTiO_3$ actually curve. The curved appearance of internal beams is due to one of two processes. In a majority of cases the curved appearance arises from a series of short discontinuous straight sections. These sections, usually two or three for any given internally scattered beam, are all straight and are connected by small regions of fuzz. It is assumed that a fuzzy section is where stimulated scattering occurs because a new beam leaves this area in a new direction. So the appear-

ance of curvature in these cases stems from multiple scattering along the beam path.

The second process that may give the appearance of beam curvature is the coupling of energy between different parts of the beam. That is, although the beam travels in a straight line within the crystal, one side of the beam is reduced in intensity as coupling occurs toward the c axis as the beam progresses down the crystal, producing a curved appearance. This effect, caused by beam coupling, is especially prevalent in cases when the incident beam is totally absorbed into the scattered beam, which finds the closed loop, by total internal reflection, to create self-pumped phase conjugation by degenerate four-wave mixing.[7]

Studying photographs has also shown that many scattered beams commonly leave the incident beam at different angles and at different points in straight lines. Additionally, others have photographed in real time the instantaneous appearance of auxiliary beams, which do not intersect other beams in the crystal (eliminating other gain mechanisms such as four-wave mixing), after the crystal has appeared to reach steady state.[6] Both of these observations are consistent with a stimulated scattering theory.

5. CONCLUSIONS

We have presented evidence that demonstrates that asymmetric self-defocusing in $BaTiO_3$ is not due to an asymmetric change in the index of refraction across the incident-beam path. We have also shown that the dependence of this self-defocusing on beam diameter is exponential and that scattering in $BaTiO_3$ begins with some dynamic scattering mechanism, which we propose is nonstimulated photorefractive scattering. All evidence presented supports a theory that the phenomenon of asymmetric self-defocusing is due to a stimulated scattering phenomenon, which begins because of photorefractive inhomogeneities within the crystal.

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