

Large thermal nonlinearities and spatial self-phase modulation in $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ and BaTiO_3 crystals

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We find strong optical nonlinearities in $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ ($x = 0.61$ and $x = 0.75$) and in BaTiO_3 crystals caused by thermally induced changes of the refractive indices. This gives strong spatial self-phase modulation and self-focusing of Gaussian light beams and light-induced birefringence. The light intensities are of the order of 10^2 – 10^3 mW/mm², and the time response of the effect is of the order of milliseconds.

$\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ (SBN) and BaTiO_3 crystals are probably the most intensively studied nonlinear photorefractive crystals.^{1,2} The standard photorefractive mechanism, based on space-charge fields that induce index of refraction changes, benefited from the large electro-optics coefficients of these crystals at room temperatures. The strongly enhanced dc susceptibility arises from being close to the phase transition temperature in the crystals.³ This is also the reason for the strong dependence on temperature of the susceptibility and the refractive indices of SBN^{3,4} and BaTiO_3 .⁵

Here we report on a strong nonlinear effect in SBN and BaTiO_3 crystals that has a thermal origin. It is shown to give strong spatial self-phase modulation and self-focusing with a Gaussian light beam traversing the crystals along a path of 2–5 mm with a power of the order of 10^2 – 10^3 mW/mm². This effect has not been reported yet in SBN and BaTiO_3 crystals, although its mechanism has a stronger capability of changing the refractive indices compared with the usual photorefractive effect and might be an important factor in wave-mixing experiments. Strong dependence of the refractive indices on temperature has been reported for these crystals.^{4,5} Thermal lensing has been observed recently⁶ in another crystal ($\text{Ba}_2\text{NaNb}_5\text{O}_{15}$), in liquid crystals,⁷ and in liquids.⁸ We also demonstrate light-induced birefringence that is due to polarization dependence of the thermal effect.

In the experiment we used several SBN ($x = 0.61$ and 0.75 and various nominal doping) and BaTiO_3 crystals. Most of them were strongly photorefractive, exhibiting large beam-coupling capability. A light beam from an argon-ion laser was shined directly or through a focusing lens onto the crystal. The beam direction was normal to the input surface, and the \bar{c} axis of the crystal was along one of these surface edges. The self-focusing effect, shown in Fig. 1, is obtained with SBN ($x = 0.61$, nominally doped with 0.05 wt. % Ce, and with an intensity absorption coefficient of 1.1 cm^{-1}). The effect is strong for an extraordinary polarization and weaker for an ordinary polarization.

We believe that the origin of the self-focusing effect is thermal. We rule out regular photorefractive,^{1,2} photovoltaic, or photoabsorptive⁹ origins for our self-focusing effect: the symmetrical rings of the diffraction in the present experimental geometry of the crystal and the beam cannot be explained by them. The fact that the self-focusing process is so strongly dependent on the input intensity, as discussed below (see Fig. 2), is also not in accordance with these mechanisms. Moreover we observed the same self-focusing effect even in samples that were nonphotorefractive. On the other hand, we know that SBN and BaTiO_3 crystals can give large changes of the index of refraction^{4,5} as they are heated or illuminated by a laser beam. We were also able to affect strongly the diffraction pattern by changing the thermal environment and boundary conditions. When we put a heat sink on the crystal surfaces we observed a decrease in the ring number and structure.

The thermal nonlinearity is not a local Kerr-like effect since it involves heat diffusion. For an exact analysis of the effect, we have to find the space- and time-dependent temperature distribution in the crystal driven by the heat source of the Gaussian light beam. This can be done by solving the heat diffusion equation. An explicit solution depends on the specific boundary conditions and can be complicated. An analysis of a simplified case is given in Ref. 8. In our experiment, the crystals were slabs with a width of 2–4 mm and surfaces with sides of 5–8 mm. The heat relaxation time is roughly given by^{6,8,10} $\tau \approx \rho Cl^2 / (4\kappa)$, where l is a characteristic dimension, ρ is the mass density, κ the thermal conductivity, and C the specific heat at constant pressure. Thus the time scale is composed from the flow in the z direction (along the light path), with the two surfaces (input and output) as quasi-isotherms, and in the plane that is transverse to the beam direction, with the relatively large crystal cross section compared with the small beam cross section. In fact, the nonperfect circular structure of the rings (seen in Fig. 1) is a result of the nonsymmetrical structure of the crystal (rectangular with one pair of

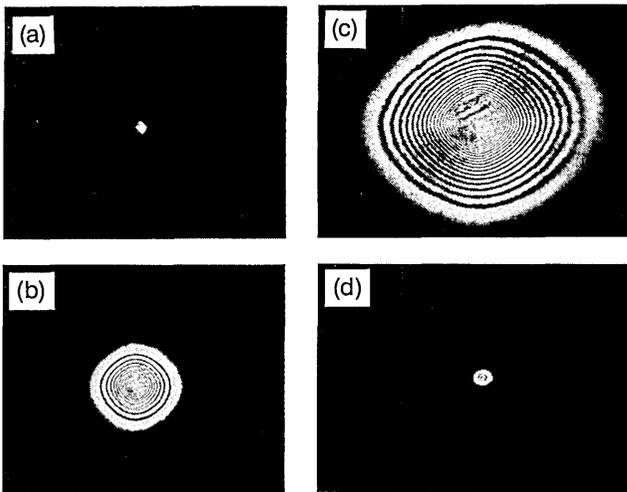


Fig. 1. Output beam from the SBN crystal for the following input-beam powers and conditions: (a) 3.5 mW and extraordinary polarization, which give a spot diameter of 7 mm at the detection screen; (b) 224 mW and extraordinary polarization, which give a spot diameter of 135 mm at the detection screen; (c) 318 mW and extraordinary polarization, which give a spot diameter of 360 and 290 mm (two axes of the elliptical shape); (d) 318 mW and ordinary polarization, which give a spot diameter of 35 mm at the detection screen. The input beam was focused onto the sample with a lens of 44-cm focal length, and the crystal location was 47.5 cm from the lens, which give a spot size of approximately 0.2 mm at the crystal. The detection screen, from where the pictures were taken, was 118 cm from the crystal.

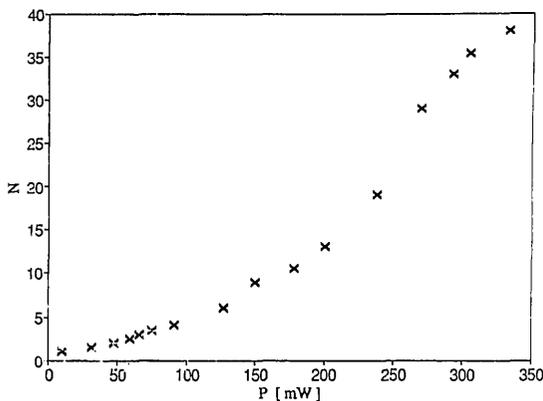


Fig. 2. Number of rings versus input-beam power (with the conditions of Fig. 1).

its edges coated with metallic electrodes that radiate heat and change the thermal boundary conditions). Another complication may arise from the expected change of κ owing to photoionization of carriers to the valence band.

The ring pattern is caused by spatial self-phase modulation owing to the nonlinear index change induced by the nonuniform Gaussian profile of the light beam. The ring pattern in the far field can be compared with the theory in Ref. 8, which results in a good agreement. A rough estimate of Ref. 7, for the change of the nonlinear index in the beam center by the number of rings N is given by $(\Delta n_e l) \approx N\lambda$. For the specific example of SBN shown in Figs. 1(c) and 1(d), we have for the extraordinary polarization

$N \approx 40$. Then with a wavelength $\lambda = 514.5$ nm and a crystal width of $l = 2$ mm, we obtain $\Delta n_e \approx 0.01$. For an ordinary polarization with the same conditions, we have $N = 3$ and $\Delta n_o \approx 0.0006$. These changes in the indices are similar to reported data⁴ for the temperature dependence in SBN. We observed the effect in several SBN crystals. The strongest self-focusing was achieved for SBN with $x = 0.61$, nominally doped with 0.1 wt. % of Ce,¹¹ but the pictures in Fig. 1 were taken for 0.05 wt. % of Ce.¹¹ In our BaTiO₃ samples the self-focusing effect was weaker. Typically we obtained $N = 2.5$ with direct illumination (without any lens) of ~ 1 W from an argon laser beam.

The number of rings was strongly dependent on the input-beam intensity, as shown in Figs. 1 and 2. The expected⁸ linear dependence on intensity is enhanced and becomes nonlinear when the intensity increases because the index of refraction changes faster⁴ as the crystal heats up and the temperature approaches the phase transition. When the intensity was raised beyond a certain value (of approximately 350 mW in this experiment), we saw a breakdown of the focusing. We attribute this behavior to an increase of the local temperature to values that are near (or above) the phase transition, such that the crystal partially loses its uniform, single-domain structure. The phase-transition temperature depends^{3,4} on x and on the doping and is in the range of 50–90°C. Sometimes, at the high-input-intensity regime, we saw a dramatic continuous increase in the rings number with an increasing self-focusing in the crystal (although the input was held constant), until the crystal was damaged and changed its uniformity. We also note that the self-focusing effect is stronger as the crystal doping and absorption were higher, as expected from the thermal origin of the effect.

We measured the buildup behavior of the self-focusing by plotting the intensity of the central part of the far-field diffraction of the output beam (it passed through an aperture). This is shown in Fig. 3. The time constant is of the order of milliseconds, and it depends approximately on the input power P as $\tau \propto P^{-2.3}$. We observed, however, a much slower change in the ring pattern in the time scale of seconds to minutes. This is in accordance with the formula for τ given above. This is also seen and discussed in the next experiment, in which we examine light-induced birefringence. We therefore

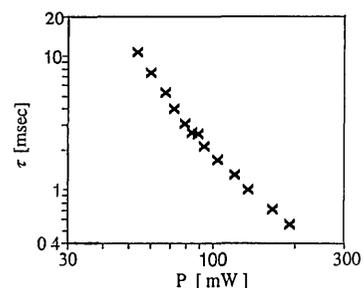


Fig. 3. Dependence of the time response on the input beam power (with the conditions of Fig. 1 with an aperture in front of the detector).

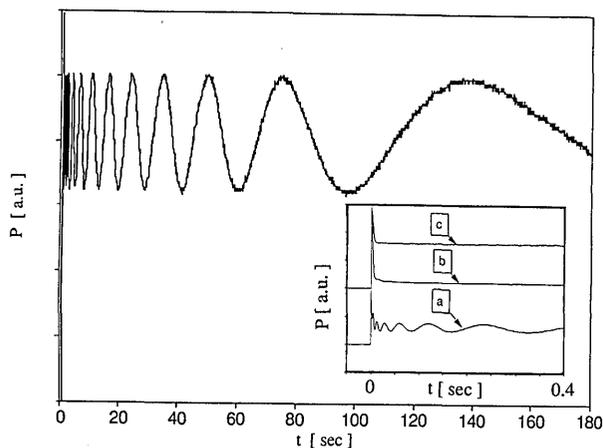


Fig. 4. Time dependence of the output intensity after the beam passes through an analyzer (the input beam was 45° to the crystal axes). The inset shows the initial response in Fig. 4 (curve a), which is the time dependence of the output intensity after the beam passes through an aperture and an analyzer, the response without the output polarizer (curve b), and the response without the output polarizer with extraordinary input polarization (curve c).

think that there are two time scales in the buildup of the temperature and indices changes. A part of the index change occurs in the short period of several milliseconds, and we can view it as local heating by the light beam. Then a heat flow with a much slower time scale causes a further change. This part strongly depends on the bulk geometry.

In another experiment, we examined the difference in the nonlinearity of the ordinary and extraordinary polarizations with refractive indices n_o and n_e , respectively. This gives a light-induced birefringence through a thermal effect. The input-beam polarization was at 45° with the c axis. The intensity-dependent retardation caused a change of the polarization state. We were able easily to observe retardations of several periods (multiples of 2π). This can be nicely shown in the buildup of the retardation for a Gaussian beam when we measure the time dependence of the output beam after it passed an analyzer (a polarizer with a perpendicular axis with respect to the input polarization) and an aperture (to detect only the central part of the diffracted beam). We found an expected oscillatory behavior, shown in Fig. 4, as a result of the rotation of polarization (in time) of the beam that passes the crystal. The input power was 135 mW. The oscillation period increased in time, corresponding to a slowing down in the buildup of the retardation. The oscillations can be compared to the changes of the ring pattern in the former experiment. This reveals that after the initial quick buildup of the self-focusing pattern there is a further slow change in the ring pattern that is related to the oscillations in Fig. 4. The initial rapid response, shown by curve a in the inset of Fig. 4, corresponds to the formation of the ring pattern of the former experiments. The fast decrease in the intensity is a result of the aperture that blocks diffracted light that is due to the self-focusing. In curves b and c of Fig. 4, we see the corresponding fast response in which the output

polarizer was removed and when the input had an extraordinary polarization, respectively. In these traces we again see the two time scales discussed above. A time dependence of the refractive index and also the oscillations in the reflectivity of a self-pumped phase-conjugate mirror that resembles the above slow response was recently observed in BaTiO_3 .⁵ There was no report on any self-focusing effect, and only the slow part of the index change was recorded. The reason why the intensity in the oscillation in Fig. 4 does not reach zero, as expected from a continuous change of the retardation, is the nonuniformity of the retardation in the beam cross section owing to its Gaussian profile. By taking the average value, we obtained the dependence of Fig. 4.

We have also performed a first study of induced gratings and beam coupling by the thermal effect. Except for the usefulness of this process in wave mixing, its dynamics can provide information about crystal parameters, the heat flow, and more. However, this process has been found to be inefficient because of diffusion and grating washout.

In conclusion, we have demonstrated a strong nonlinear behavior in SBN and BaTiO_3 crystals that originates from thermally induced index changes. This effect can be used as a tool to investigate crystal parameters and heat diffusion processes. The function of the light is twofold: to induce the thermal energy and to probe and visualize the changes in real time, for the whole medium in a parallel way.

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