Photorefractive effect in a $BaTiO_3$ crystal at the 1.5-μm wavelength regime by two-photon absorption

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We demonstrate the extension of the photorefractive effect in $BaTiO_3$ crystals to the 1.5-μm wavelength regime, which is obtained by two-photon absorption induced by intense femtosecond light pulses. The strong dependence of the effect on the light's intensity permits nondestructive readout of the photorefractively induced pattern by low-intensity light. One can thus use the photorefractive effect in the 1.5-μm regime to form optical components for communication applications. © 1996 Optical Society of America

The photorefractive effect permits the induction, in real time, of complex refractive-index changes in crystals by light illumination. Several applications of the effect for communication systems have been proposed. For example, the double-phase-conjugate mirror can be used to implement all-optical interconnection.[1] However, such applications were limited in the important communication wavelength regime near 1.5 μm because of the lack of suitable photorefractive materials.

$BaTiO_3$ is one of the most important photorefractive materials because of its large electro-optic coefficient. Attempts have been made to extend its spectral response. For example, specially doped $BaTiO_3$ crystals were grown to increase the maximum operating wavelength.[2] However, as far as we know, this crystal was never used in the important communication wavelength regimes of 1.3 and 1.5 μm. The photorefractive effect in the 1.5-μm regime was demonstrated in semiconductors (e.g., GaAs, InP, and CdTe; Ref. 3) but not in ferroelectric materials. The standard photorefractive effect is based on the absorption of photons by impurity levels. However, when the light intensities are strong enough, effects that are caused by two-photon absorption (TPA) can become important.[4,5] The TPA coefficient of $BaTiO_3$ crystal was measured in the visible region previously.[6]

In this Letter we extend, for the first time to our knowledge, the photorefractive effect in $BaTiO_3$ crystals to the 1.5-μm regime by TPA. The TPA can be enhanced by the existence of intermediate energy levels inside the forbidden bandgap (because of impurities).[4] The effect was obtained with high-peak-power, ultrashort pulses, which were focused to a small spot in the crystal. No significant photorefractive effect could be obtained without focusing.

The strong dependence of the effect on the light intensity and on the intensity threshold of the photorefractive effect (because of thermal excitation) can help to prevent the erasure of the photorefractively induced pattern: The writing process can be done with high intensities, whereas the readout is done with low intensities, such as those that are typical of optical communication systems, and hence the stored refractive-index pattern can be read without significant erasure. One can change or erase the photoinduced pattern by illuminating the crystal with high light intensities or by another light source with a lower wavelength that is absorbed in the crystal by a one-photon process. The photorefractive effect with ultrashort light pulses based on one-photon absorption was demonstrated in the visible wavelength regime.[7,8] The effect of TPA on the photorefractive recording was investigated in KTN and in $LiNbO_3$ crystals.[9,10] However, no significant photorefractive recording was obtained, unless beams with lower wavelength (which may permit a one-photon process) were used during the writing.

In our experiments we used two standard $BaTiO_3$ crystals (grown by Sanders, Inc). One of the crystals had dimensions of 5 mm $\times$ 5 mm $\times$ 5 mm, cut with its c axis at 45° relative to one of the sides. The other had dimensions of 5 mm $\times$ 6 mm $\times$ 3 mm and its c axis along the 6-mm side. The effect was observed in both crystals; however, the results that are given here were obtained with the first crystal. The crystal was illuminated by light from a synchronously pumped optical parametric oscillator with a 130-fs pulse duration, an 80-MHz repetition rate, and an average power of 250 mW operating at a wavelength of 1.5 μm. To obtain high intensities we focused the writing beams down with a strong lens with a 40–50-mm focal length.

We demonstrated and analyzed the effect by performing two-wave mixing experiments. The power of the two beams was controlled by an attenuator, with the ratio between the beam intensities kept constant (1:6). The angles between the propagation directions of the strong (weak) beams and the c axis were approximately 50° (40°) outside the crystal. A small angle between the beams gives a better spatial overlap between the beams[7] but causes a weaker photorefractive effect if the diffusion process

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is the dominant effect in creating the photorefractive field. This reduction of efficiency is particularly important at the wavelength of 1.5 μm, where the grating period is larger by a factor of 2–3 compared with those of gratings formed by light sources in the visible regime. When the crystal was illuminated simultaneously by the two beams, the intensity of one of the beams (which had the smaller angle between its propagation direction and the c axis) increased, while the intensity of the second beam decreased, as expected for the photorefractive effect in this configuration. The direction of the power flow between the two beams was reversed when the crystal's c axis was inverted. The polarization of the two beams was extraordinary. No significant effect was observed when the polarization was ordinary or when one of the beams was delayed, so the pulses did not overlap in time inside the crystal.

Figure 1 shows the time-dependent buildup of the amplified beam intensity. We could partially erase the induced grating by illuminating the crystal with an unfocused beam at $\approx$0.8 μm from a pulsed Ti:sapphire laser with an average power of 300 mW and a spot size of ~0.5 cm, as shown in the figure. Amplifications of the order of ~50% were achieved. We believe that one could obtain higher amplifications by optimizing the time and the spatial overlap between the two beams and by optimizing the crystal and the beam directions. Note that the spot size of the beams in our experiment was ~20 μm and the spatial extent of the pulses along their propagation direction was ~14 μm. The relatively large spectral bandwidth of 20 nm associated with these short pulses also decreased the efficiency of the process.

Figure 2 shows the dependence of the estimated exponential time constant τ of the buildup process on the input power P. We can fit the results to $\tau \propto P^{-x}$, where $x = 1.37$, compared with the values of $x = 0.5–1.0$ obtained in the conventional (single-photon) photorefractive effect. In the past we measured a dependence of $\tau \propto P^{-0.7}$ for $\lambda = 0.514$ μm at low intensities.[11] Our measurements also show a second component of the buildup process with a much longer time constant, which has an intensity dependence of P^{-1}. This time constant may originate from a weak one-photon process.

Using a single focused beam, we measured the changes in a beam profile and its direction after it passes through the crystal. Figure 3 shows the profile of the beam after it has passed through the crystal. Also shown is the profile of a beam with ordinary polarization, for which the photorefractive effect is ineffective. We see that the extraordinarily polarized beam acquires a slightly asymmetric profile. By launching the beam along different directions, we verified that the beam profile changed according to the symmetry dictated by the photorefractive effect. As in the two-wave experiments, we could erase the effect by illuminating the crystal with the Ti:sapphire laser. The changes in the beam profile, which are caused by wave mixing between different spatial frequency components of the incident beam, were observed before in the visible regime.[12] We also note that no significant fanning could be observed because the beam was focused inside the crystal and therefore the interaction length was limited.[12]

The time constants in the one-beam experiments were more than 1 order of magnitude shorter than those measured in the two-wave mixing experiments. In single-beam experiments there is an optimal, self-obtained, spatial overlap between the interacting frequency components, which could explain the faster response time. This suggests that one could significantly reduce the time constant in the two-wave mixing experiments by improving the spatial overlap between the two beams.

With an autocorrelator we measured the temporal distortions of the laser pulses after they passed through the crystal. A significant broadening of the pulse by the $BaTiO_3$ crystal was measured in the visible region.[7] According to the Sellmeier relation, the dispersion of a $BaTiO_3$ crystal[3] should be much smaller at the 1.5-μm regime than that in the visible regime. Indeed, after that pulse had passed through the crystal its width had actually decreased from 130 to 120 fs,

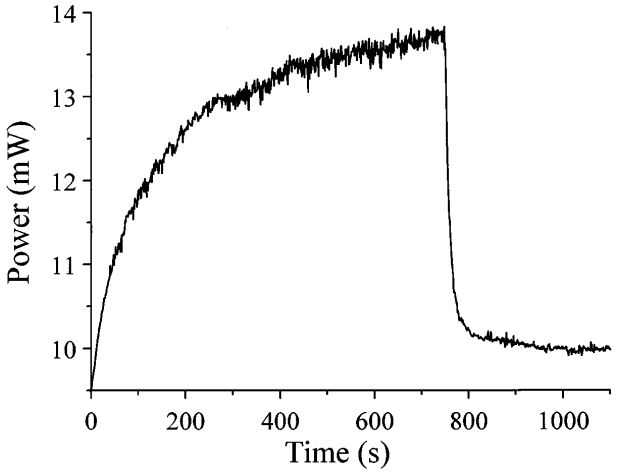

Fig. 1. Time dependence of the output signal intensity in a two-wave mixing experiment. Illumination starts at $t = 0$. At $t = 750$ s an erasing beam at $\lambda = 800$ nm is turned on.

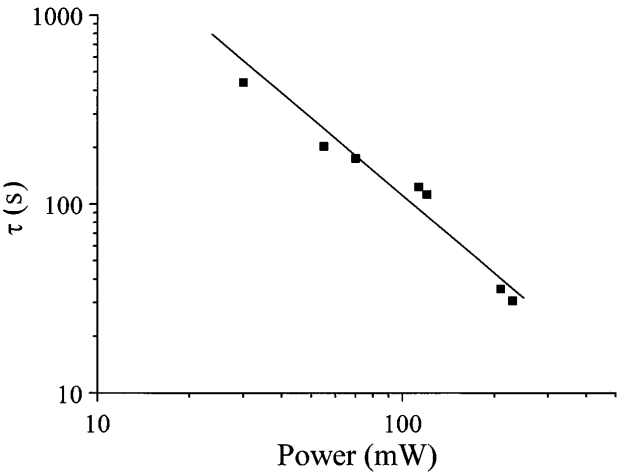

Fig. 2. Dependence of the estimated time constant (τ) of the buildup process on the input power (P). The solid line shows a fit to $\tau = \tau_0 P^{-1.37}$.

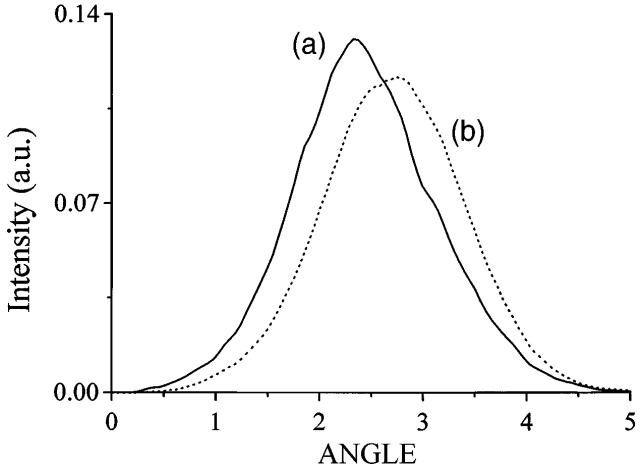

Fig. 3. Profiles of beams that have passed through the crystal for incident beams with (a) extraordinary light polarization and (b) ordinary polarization, where the photorefractive effect is ineffective.

probably because of the removal of the small frequency chirp of the pulses.

The TPA can be added to the conventional rate equations that are used to model two-wave mixing in photorefractive media by an absorption term that is proportional to the square of the intensity. The results indicate that the TPA will cause the formation of a strong second-order grating with a spatial period that equals half of that of the light interference pattern, in addition to the first-order grating, built in the conventional way.

In conclusion, we have extended the operating wavelength of the photorefractive effect in $BaTiO_3$ to the 1.5-μm regime, using two-photon absorption. This could enable the photorefractive effect to be used for forming complicated optical elements for optical communication systems. In this case the writing of refractive holograms will be induced by an intense laser, whereas the readout will be done with low intensities such as those typical of optical communication systems.

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