Voltage-controlled photorefractive effect in paraelectric $KTa_{1-x}Nb_xO_3$:Cu,V

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Experimental results that demonstrate the formation of photorefractive gratings in KTN:Cu,V in the paraelectric phase are presented. These gratings are formed using the quadratic electro-optic effect, which allows amplitude modulation of the diffracted beam by an external electric field. High diffraction efficiencies of over 50% in a 3-mm-thick sample and amplitude modulation of the diffracted beam by an external field at frequencies of up to 20 kHz were observed.

The possible use of volume holography for performing various optical computing tasks has been considered in the past[1–3] and is currently arousing renewed interest because of its potential for realizing large-scale neural network models.[4]

Potassium tantalate niobate (KTN) was one of the first crystals in which photorefractive (PR) properties were identified.[5,6] However, although it is mentioned in most, if not all, review articles on PR materials as a promising photorefractive crystal, little has been done theoretically or experimentally to investigate its photorefractive properties.[7,8] There has been little research to explore the PR properties of KTN primarily because these crystals were considered hard to grow. We were able to grow large KTN crystals by carefully selecting the dopants. In what follows we describe a set of experimental data that demonstrate the storage of volume holograms with high diffraction efficiencies, and a controllable gain by an external electric field, in a KTN:Cu,V crystal in the paraelectric phase.

The chemical composition of KTN is $KTa_{1-x}Nb_xO_3$. It undergoes a ferroelectric phase transition at a temperature T_c that depends on the Nb concentration x (roughly 8.5 K/1% per mole Nb, such that for $x = 35\%$ $T_c \simeq 300$ K).[9] In the paraelectric region ($T > T_c$) KTN is cubic, and thus the electro-optic effect is quadratic and is given by[10]

$$\Delta n = \frac{1}{2} n_0^3 g P^2, \tag{1}$$

where Δn is the birefringence, n_0 is the refractive index, g is the appropriate electro-optic coefficient, and P is the static (low-frequency) polarization, given in the linear region by

$$P = \epsilon_0(\epsilon - 1)E. \tag{2}$$

Therefore the PR effect in paraelectric KTN can be described as follows[11]: When interference patterns are formed in the crystal by the illuminating beams, space-charge fields E_{sc} spatially correlated with these patterns are created, provided that some transport mechanism is present (either diffusion and/or drift in an external electric field). In the presence of an external electric field E_0 these space-charge gratings induce refractive-index gratings that are given by

$$\delta(\Delta n) = \Delta n(E_{sc} + E_0) - \Delta n(E_0)$$

$$= \frac{1}{2} n_0^3 g \epsilon_0^2 (\epsilon - 1)^2 (2E_0 E_{sc} + E_{sc}^2), \tag{3}$$

where it is assumed that the polarization is in the linear region. The last term is for a higher-order grating, which in a thick hologram will not diffract the writing beams, resulting in

$$\delta(\Delta n) = n_0^3 g \epsilon_0^2 (\epsilon - 1)^2 E_{sc} E_0. \tag{4}$$

Therefore the modulation depths of the diffraction gratings in KTN crystals in the paraelectric phase are expected to be linearly dependent on the applied electric field. This fact, together with the fact that the dielectric constant is high, gives rise to a possible use of KTN to implement amplitude-modulated phase conjugation and holographic memory devices. Electrical control of the holographic diffraction efficiency has been observed in lead zirconate titanate ceramic by Micheron *et al.*[12] and in strontium barium niobate (SBN) by Thaxter and Kestigian.[13]

The crystal used was a KTN crystal doped with V and Cu. The crystal was grown by us using the top-seeded-solution growth method. The chemical com-

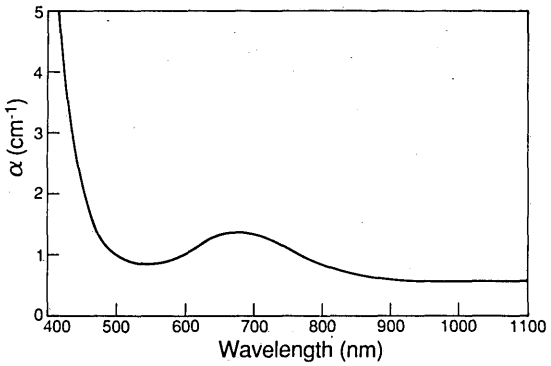

Fig. 1. Absorption spectrum of the KTN:Cu,V crystal.

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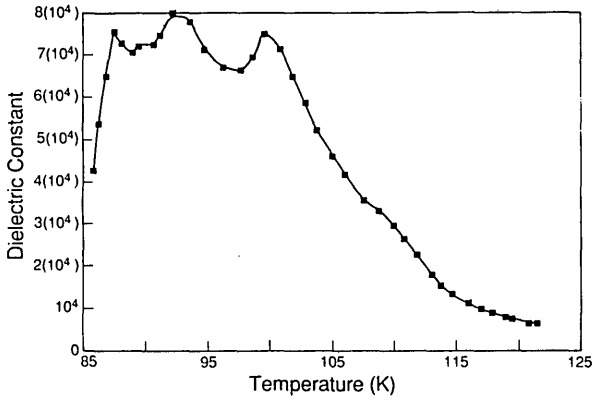

Fig. 2. Temperature dependence of the low-frequency dielectric constant. The curve through the data points is drawn as a guide.

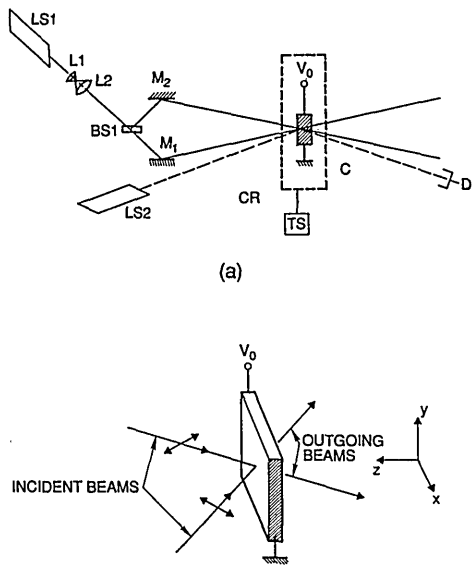

Fig. 3. (a) Experimental setup for diffraction measurements. LS1, Ar^+ laser with $\lambda = 514.5$ nm; LS2, He–Ne laser with $\lambda = 612.8$ nm; L1, L2, beam-expander lenses; BS1, beam splitter; M_1, M_2, mirrors; CR, cryostat; C, crystal; TS, temperature sensor; D, detector. (b) The sample position relative to the writing beams.

position of the sample was determined by electron-microprobe analysis and was found to be $KTa_{0.91}Nb_{0.09}O_3$. The dopants, although present in the flux, were found to be below the detection limit of the electron microprobe. A few samples were cut from the crystal and observed through crossed polarizers. A 3 mm × 3.5 mm × 5.5 mm sample that appeared to be homogeneous was finally selected and polished along the crystallographic [100] directions. The absorption spectrum of the sample is shown in Fig. 1.

The ferroelectric phase-transition temperature was determined by monitoring the temperature dependence of the low-frequency capacitance (Fig. 2). The cubic-to-tetragonal transition roughly corresponds to the highest temperature peak (at $T_c = 100$ K). The behavior of the dielectric constant below $T_c = 100$ K results from the tetragonal–orthorhombic and orthorhombic–rhombohedral transitions.[9]

A modified Sawyer–Tower circuit[14] was used to measure the dependence of the static polarization P on the electric field E. In accordance with the Devonshire theory of phase transitions in ferroelectrics, it was found that in the paraelectric region this dependence is described by

$$E = aP + bP^3, \tag{5}$$

where $a = 4.08 \times 10^8$ cm/F and $b = 4.15 \times 10^{19}$ cm^5/V^2 F^3.

Diffraction efficiencies were measured using the two-wave-mixing method. A schematic description of the experimental setup is given in Fig. 3. The sample position relative to the writing beams is shown in Fig. 3(a). The writing beams were generated by an Ar^+-ion laser operating at $\lambda = 514$ nm and were aligned so that their bisector is orthogonal to the sample (i.e., in the z direction); the beams were polarized horizontally [in the (x, z) plane]. All measurements were made at $T = 112$ K. The intensity of the writing beams was 32 mW/cm^2, and the angle between them was 11° (2.7-μm grating period). The exposure lasted 7.75 min under an applied electric field of $E_w = 900$ V/cm. The probe beam was a weak red He–Ne beam incident at the Bragg angle for the grating created by the writing beams [Fig. 3(b)]. It was verified that this beam was not capable of writing or erasing a photorefractive grating.

The dependence of the diffraction efficiency η on the electric field applied during reading is given in Fig. 4. As can be seen, η reaches approximately 60% and saturates at $E_R \simeq 3$ kV/cm. This saturation is discussed below.

In addition, in order to demonstrate the modulation capability of the voltage-controlled photorefractive effect, a 50-V/cm ac field at 20 kHz and a dc bias of 400 V/cm were applied to the crystal. The temporal behavior of η under this field is presented in Fig. 5.

Some diffraction was observed even for $E_0 = 0$. The diffraction efficiency here was small, $\eta \simeq 1\%$ (at this temperature), but it was not easily erased even when the polarity of the applied field was reversed.

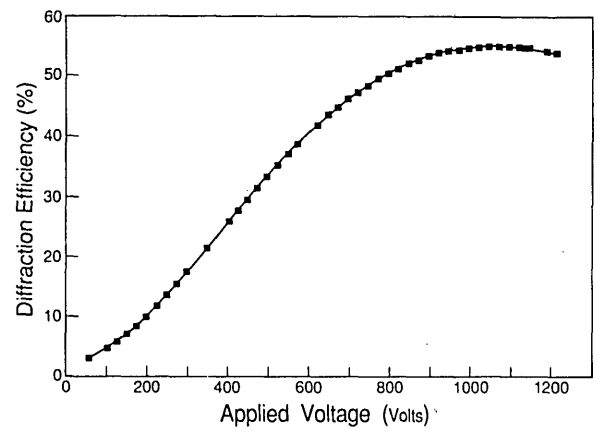

Fig. 4. Diffraction efficiency of the He–Ne beam as a function of the applied voltage.

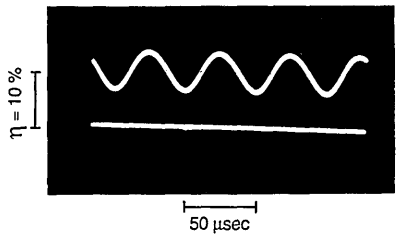

Fig. 5. Amplitude modulation of the diffracted beam. The modulating field is 50 V/cm rms, and the dc bias field is 400 V/cm.

The results described above clearly demonstrate the possibility of amplitude modulating the photorefractive gratings in paraelectric KTN crystals by an external electric field. The modulating frequency in our experiment was limited to 20 kHz by the bandwidth of our oscillator. The true upper limit determined by the performance of the crystal is in the range of 100–500 kHz owing to the critical slowing of the dielectric response ϵ near the phase-transition temperature.

It should be remembered, however, that the diffraction efficiency saturates at strong electric fields. This is attributed to two mechanisms: (a) The $\sin^2[\pi\delta(\Delta n)d/\lambda\cos(\theta/2)]$, where $\delta(\Delta n)$ is given by Eq. (4), d is the thickness of the sample, and θ is the crossing angle of the writing beams, and (b) the saturation of the static polarization P at high electric fields. To a first order of approximation P can be described as

$$P(x) = P_0 + \left.\frac{dP}{dE}\right|_{E=E_0} E_{sc}(x), \tag{6}$$

where $P_0 = P(E_0)$. Thus P^2 will contain a grating term given by

$$\delta P^2(x) = 2P_0 \left.\frac{dP}{dE}\right|_{E_0=E} E_{sc}. \tag{7}$$

Using Eq. (5) we get for the induced index grating

$$\delta(\Delta\eta) = \eta g \frac{P_0}{(a + 3bP_0^2)} E_{sc}(x), \tag{8}$$

from which we expect $\delta(\Delta n)$ to vary nonlinearly with the applied electric field.

The fixation effect that was observed at $E_0 = 0$ is similar to what was observed by Micheron and Bismuth[15] in SBN crystals and by us in a KTN:Cr,Fe (Ref. 11) crystal. Micheron and Bismuth attribute the fixing mechanism to domain reversal, while in our case the crystal is in the paraelectric phase. However, in doped crystals in the paraelectric phase, ionic movement in the proximity of the phase transition is common.[16] We therefore suggest that the fixing observed is a result of the formation of ionic gratings, which is possible owing to the softness of the crystal in the proximity of the phase transition. The effect is currently being investigated further.

As mentioned above, the phase transition of KTN is determined by the Nb concentration. The sample used here contained 9 mole % of Nb, and therefore the working point was low (at ≈112 K). The working point was selected to be close enough to the phase transition so that the diffraction is large ($\epsilon \gg 1$) but far enough from the transition so that the mean field approximation is in effect. The latter requirement is necessary to avoid the critical slowing of the dielectric response that occurs close to T_c. The growth of doped KTN crystals with large Nb concentrations will enable us to increase the working point to room temperature. We are currently growing such crystals and already have some samples with $T_c \simeq 250$ K. It should be borne in mind, however, that temperature stabilization of the sample will always be required owing to the fact that in the vicinity of T_c the dielectric constant and hence the PR properties are extremely temperature sensitive.

The PR sensitivity based on the above results is $S \simeq 10^{-5}$ cm^3/J. This is much lower than the usually quoted results for KTN ($S \simeq 0.1$ cm^3/J) for the two-photon excitation process described in Ref. 6. The reason for this loss of sensitivity is not completely clear at this time. It may be a result of a reduction in carrier mobility owing to high dopant concentration.

In conclusion, we have demonstrated a voltage-controlled photorefractive effect by making use of the quadratic electro-optic effect in KTN:Cu,V. High diffraction efficiencies, full amplitude modulation, and modulation frequencies of 20 kHz were observed. This effect can be of use in applications where the amplitude of a photorefractive grating needs to be controlled.

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