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Voltage-Controlled Diffraction Modulation in Manganese-Doped Potassium Sodium Tantalate Niobate Single Crystals

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Paraelectric potassium sodium tantalate niobate (KNTN) crystals have been grown with two different Mn concentrations, namely, 0.25 and 0.5 mol %. High voltage-controlled diffraction efficiencies of 79 and 74%, and large two-wave mixing gains of 36 and 32 cm^{-1} were achieved for the 1.0 mm thick 0.25 and 0.5 mol % Mn-doped KNTN samples, respectively. The amplitude modulation of the diffracted beam by an AC external field at a frequency of 100 Hz was demonstrated. The results show that Mn-doped KNTN is a promising material for voltage-controlled diffractive applications.

Electroholography (EH) is a high-speed wavelength-selective beam steering method based on governing the reconstruction of volume holograms by an externally applied electric field.[1)] The implementation of EH based devices (EHDs) necessitated the development of a special photorefractive crystal, that is, a paraelectric voltage controlled photorefractive (PR) crystal. A potassium lithium tantalate niobate $K_{1-y}Li_yTa_{1-x}Nb_xO_3$ (KLTN) single crystal exhibits the unique properties to be the heart of EHDs.[2–4)] Unfortunately, some defects, such as bubbles, nebulous inclusions, and even hollows, usually appear in KLTN single crystals.[5)] Substituting Na for Li will effectively suppress the appearance of defects; thus, high optical quality $K_{1-y}Na_yTa_{1-x}Nb_xO_3$ (KNTN) single crystals can relatively easily to be obtained.[6)] However, detailed information on the voltage controlled diffractive properties of doped KNTN single crystals, which is essential in the design and optimization of EHDs was seldom reported.

In this letter, we report on the crystal growth and voltage-controlled diffraction modulation in manganese (Mn) doped KNTN (Mn:KNTN) crystals. A high diffraction efficiency and a large two-wave mixing gain coefficient were achieved. Furthermore, the amplitude modulation of the diffracted beam by an AC external field at a frequency of 100 Hz was examined.

KNTN single crystals doped with 0.25 and 0.5 mol % manganese were grown by the top-seeded solution growth method. The crystal growth condition of pure KNTN was adopted to grow Mn:KNTN. After the crystals were grown, they were cut to cubes along the crystallographic ⟨001⟩ axis with dimensions of 2.0 mm (width) × 5.0 mm (height) × 1.0 mm (thickness). The surfaces for light propagation were optically polished. Phase-transition temperatures (T_c) of the samples were determined by measuring the temperature dependence of dielectric coefficient with an LCR meter (Agilent E4980A). The refractive indices were measured by a spectroscopic ellipsometer (α-SE™, J. A. Woollam Co.). The quadratic electro-optic coefficients were determined by a Mach–Zehnder interferometer. Silver electrodes were coated on 5.0 × 1.0 mm^2 faces for voltage-controlled diffractive properties measurements. A two-wave mixing setup was used to measure the diffractive properties of samples. The measurement setup and configuration were described in detail in ref. 7. The measurements were made at 25 °C which the crystals were in the paraelectric phase. The diffraction efficiency η is defined by

$$\eta = I_d/I_t, \tag{1}$$

where I_t and I_d are the intensities of the transmitted beams before and after diffraction, respectively. The two-wave mixing gain coefficient Γ is determined as

$$\Gamma = \frac{1}{d}\ln\frac{I'_s}{I_s}, \tag{2}$$

where I_s and I'_s are the signal intensities before and after amplification, respectively, and d is the interaction length.

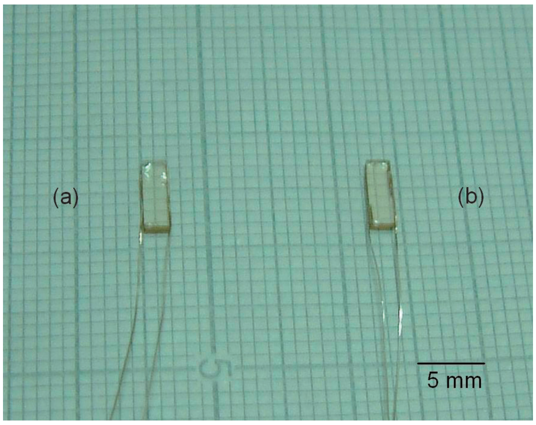

Fig. 1. Samples of Mn:KNTN crystals. (a) $Mn_{0.25}$:KNTN and (b) $Mn_{0.5}$:KNTN.

After 3 days of growth, yellow high quality Mn:KNTN single crystals were obtained. Figure 1 shows the cut and optically polished specimens of 0.25 mol % Mn-doped KNTN (designated as $Mn_{0.25}$:KNTN) and 0.5 mol % Mn-doped KNTN (designated as $Mn_{0.5}$:KNTN). The chemical compositions of the samples are $K_{0.95}Na_{0.05}Ta_{0.58}Nb_{0.42}O_3$. The T_c values of $Mn_{0.25}$:KNTN and $Mn_{0.5}$:KNTN are 21.5 and 23.5 °C, respectively. The large dielectric constants of the crystals near the phase-transition temperature will induce a large Kerr effect in the paraelectric phase.

In the paraelectric phase, the diffraction efficiency of plane-phase transmission holograms is given by[7)]

$$\eta = \sin^2\left(\frac{\pi d}{\lambda\cos\theta} n_0^3 g_{eff}\varepsilon_0^2\varepsilon_r^2 E_0 E_{sc}\right) = \sin^2\left(\frac{\pi d}{\lambda\cos\theta} n_0^3 s_{eff} E_0 E_{sc}\right), \tag{3}$$

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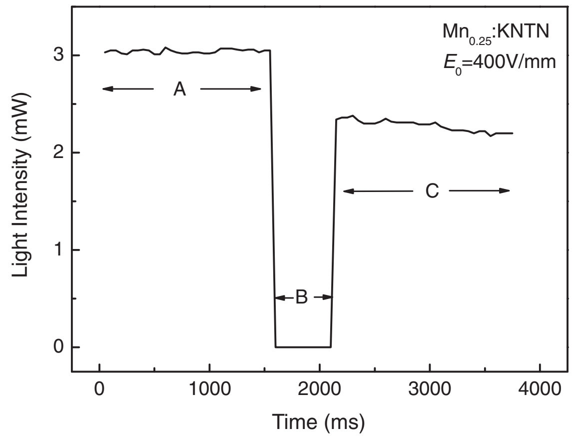

Fig. 2. Typical writing and reading processes for $Mn_{0.25}$:KNTN.

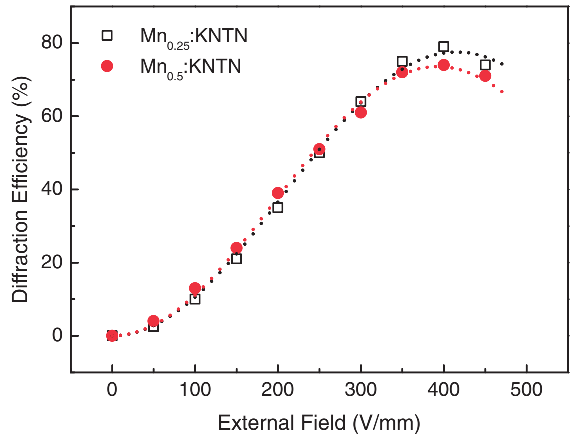

Fig. 3. Diffraction efficiency as a function of external field.

where λ is the optical wavelength, θ is the angle between the reading beams and the equiphase planes of the grating, n_0 is the refractive index, g_{eff} is the effective quadratic polarization-optic coefficient, s_{eff} is the effective quadratic electro-optic coefficient, ε_0 is the dielectric permittivity, and E_{sc} is the amplitude of the space-charge field. It is assumed in eq. (3) that the absorption can be neglected. Figure 2 illustrates the intensity of the transmitted beam in typical writing and reading processes for $Mn_{0.25}$:KNTN at $\theta = 5°$. During the writing process (A), two writing beams with the same intensity of 3 mW intersected inside the crystal and the PR grating was established. Then, both writing beams were turned off; thus, no light was detected (process B). For the reading process (C), a DC external electric field of 400 V/mm was applied to the crystal and one writing beam was turned on. A diffracted beam of 2.38 mW was detected; thus, the high diffraction efficiency (η) was calculated to be 79%. Because the PR grating was not fixed, it was erased by the reading beam in the reading process; thus, the diffraction efficiency was decreasing as time.

The diffraction efficiency as function of the external field for both crystals is shown in Fig. 3. In our experimental configuration, the values for the parameters in eq. (3) are $d = 1.0$ mm, $\lambda = 532$ nm, $\theta = 5°$, $n_0 = 2.27$, $s_{\mathrm{eff}} = 3.5 \times 10^{-15}$ m^2/V^2). As expected from eq. (3), η shows a quadratic dependence on the applied field and reaches 79 and 74% at a field of 400 V/mm. By fitting the experimental data with eq. (3) (the result was shown by the dashed curves in Fig. 3), the E_{sc} values of $Mn_{0.25}$:KNTN and $Mn_{0.5}$:KNTN were determined to be 15.6 and 17.5 V/mm, respectively. For comparison, the maximum diffraction efficiency of Cu:KLTN (2.9 mm thick) is 75% at 240 V/mm,[8] and for the maximum diffraction efficiency Cu, V:KTN (3.0 mm thick) is 60% at 300 V/mm.[9] Thus, Mn doped KNTN exhibits good voltage-controlled diffractive properties.

The two-wave mixing gain coefficient Γ was also measured at $\theta = 5°$ with different external fields. Γ is related to the PR index grating, as shown by

$$\Gamma = 4\pi n_0^3 g_{\mathrm{eff}} \varepsilon_0^2 \varepsilon_r^2 E_0 E_{\mathrm{sc}} \sin\frac{\Phi}{m\lambda\cos\theta} = 4\pi n_0^3 s_{\mathrm{eff}} E_0 E_{\mathrm{sc}} \sin\frac{\Phi}{m\lambda\cos\theta}, \quad (4)$$

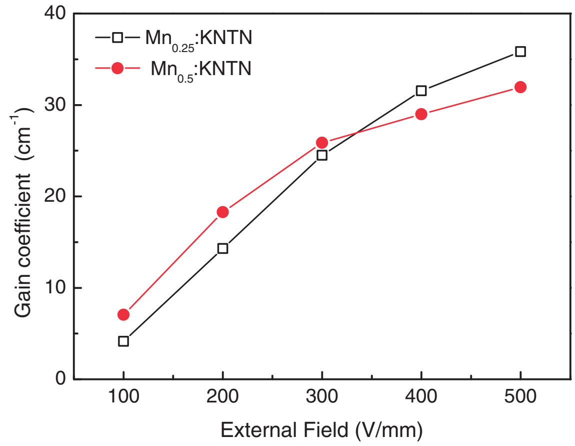

Fig. 4. Two-wave mixing gain coefficient versus external field. Square and circle dots indicate the experiment data. The lines serve only as visual guides.

where Φ is the phase shift between the optical interference pattern and the PR index grating, $m = 0.06$ is the modulation depth of the incident optical interference pattern, $\lambda = 532$ nm, $\theta = 5°$, $n_0 = 2.27$, $s_{\mathrm{eff}} = 3.5 \times 10^{-15}$ m^2/V^2. The phase shift Φ is $\sim \pi/2$ when the PR process is dominated by diffusion. Figure 4 shows Γ as a function of the external field at the pump beam intensity of 3 mW. The gain increases with external field and reaches large values of 36 and 32 cm^{-1} at $E_0 = 500$ V/mm for $Mn_{0.25}$:KNTN and $Mn_{0.5}$:KNTN, respectively. By applying a higher external field, the gain will reach a larger value.

In order to demonstrate the modulation capability of the voltage-controlled diffractive effect, a 75 V/mm (max value) AC field at 100 Hz and a DC bias of 75 V/mm were applied to the crystal. The temporal behavior of diffraction efficiency under this field is presented in Fig. 5. The diffraction efficiency varies with external field as shown by eq. (3) and indeed has a frequency of 100 Hz. The frequency is limited by our experimental instruments. As well known, the response time of KLTN series crystals is less than 10 ns;[1] thus, the potential of modulation frequency can reach a value as high as 10^8 Hz.

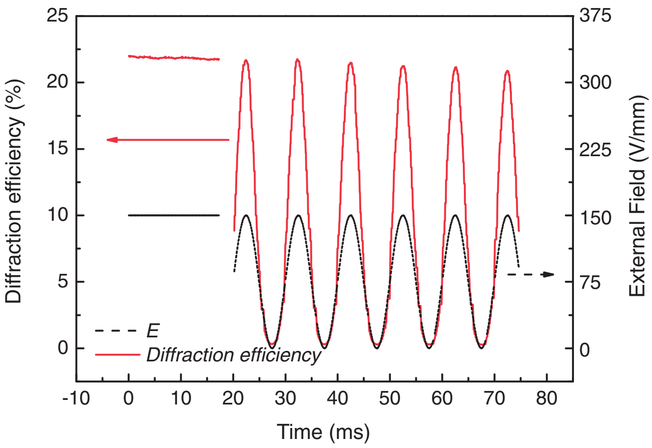

Fig. 5. Modulation of the diffractive beam.

In conclusion, high voltage-controlled diffractive performance was observed in Mn doped KNTN single crystals. High diffraction efficiencies of 79 and 74%, and large two-wave mixing gains of 36 and 32 cm^{-1} were achieved for $Mn_{0.25}$:KNTN and $Mn_{0.5}$:KNTN, respectively. A modulation of the diffractive efficiency under AC field was demonstrated. Our results indicate that Mn-doped KNTN crystals seem very promising materials for EH applications.

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