Control of photorefractive space-charge field and its deflection application in Mn-doped KTa$_{1-x}$Nb$_x$O$_3$ crystal

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Abstract

The photorefractive effect is an effective way of achieving control of a beam by inducing a space-charge field and controlling the distribution of the refractive index. Herein, the one-dimensional gradient distribution of a space-charge field is structured, and the deflection of the transmitted beam is achieved. The method of regulation and control of the space-charge field was studied by digital holographic microscopy based on a Mn-doped KTN single crystal. The realized deflector has a maximum deflection angle of 0.3° and maintains good frequency-response characteristics up to 50 kHz. This work provides guidance for optical functional devices based on engineering distribution of a space-charge field.

1. Introduction

The light-induced refractive-index change effect (that is, the photorefractive effect), as a nonlinear-optical characteristic of an electro-optical crystal, is an effective way of regulating and controlling distribution of the refractive index [1–3]. The change of refractive index in materials, controlled by irradiation, has been used in the optical memory field, self-pumping phase conjugation, and spatial light modulators [4–7]. The charge carriers excited upon irradiation are transited through drift, diffusion, and the photovoltaic field, forming the distribution of space charges. The space-charge field, originating from the distribution of space charges, yields the distribution of the refractive index [8–11]. Therefore, the space-charge field could be established through programmable space distribution of irradiation and to further realize engineering of optical devices. Optical deflectors, as an important type of optical device that changes the direction of a beam of light, are widely used in scanning, imaging, and integrated optical systems [12–14]. Materials with a gradient refractive index can achieve deflection of transmitted beams, which is used in the field of optical deflectors. The gradient distribution of the refractive index could be induced by a space-charge field by injecting space charge from electrodes to achieve beam deflection [15, 16]. Similarly, the gradient distribution of the refractive index could also be realized by engineering the composition distribution in the process of growing single crystals [17–19]. As mentioned above, the gradient refractive index could be achieved by building the one-dimensional space-charge field under the photorefractive effect. Hence, the method based on the photorefractive effect, which is contactless and does not require precise control of composition distribution, exhibits immense potential in building a space-charge field and realizing optical deflectors. Meanwhile, the distribution of a space-charge field induced by a programmable irradiation field is beneficial to the design and manufacture of new optical functional devices [20, 21].
Mn-doped KTN was grown using the top-seeded solution growth method [33]. The crystal was cut into a 2. Results and discussion

under external electric field while maintaining a good frequency response of up to 50 kHz. The methods of deflectors provide guidance for the design of optical devices.

walls. According to the previous researches, the dynamic polar nano-regions (that is PNRs) mainly enhance of the gradient distribution of the refractive index through a large quadratic electro-optical nanoscale and correlative characters of PNRs promote the quadratic electro-optical effect and keep the high contribute to the large quadratic electro-optical effect at the temperature slightly over the Curie point. The effect, but also maintains the quality of the transmitted beam, avoiding the scattering based on the domain optical quality [34].

The experimental setup is shown in figure 2. The results show that the achieved \( E_{sc} \) and the refractive index was designed by irradiation. Digital holographic microscopy (DHM) was used to monitor the distribution of the refractive index [30–32]. The results show that the achieved \( E_{sc} \) obeys a one-dimensional gradient distribution and a maximum up to 208 V mm\(^{-1}\), which is comparable to an applied electric field of 242 V mm\(^{-1}\). The change of the gradient refractive index under different applied electric fields leads to different deflection angles. Based on this, an electrically controlled optical deflector was achieved. The deflective angle reaches 0.3\(^{\circ}\) under external electric field while maintaining a good frequency response of up to 50 kHz. The methods of engineering the space-charge field distribution by applying spatial irradiation and achieving optical deflectors provide guidance for the design of optical devices.

2. Results and discussion

Mn-doped KTN was grown using the top-seeded solution growth method [33]. The crystal was cut into a cube of size of 4.13 \( \times \) 2.63 \( \times \) 1.57\(^{\circ}\) mm\(^3\)((001)_{C} \times [010]_{C} \times [100]_{C})\), and both (100) surfaces were polished. The gold electrodes were deposited on the \( yz \) faces. The dielectric permittivity was measured using an LCR meter (E4980A, Agilent Technologies, USA), applying a probing voltage (1 V). The temperature dependence of the relative dielectric constant \( \varepsilon_r \) is shown in figure 1(a). The results show that the Curie temperature of Mn-doped KTN single crystal is 24 \(^{\circ}\)C. The results also show that the value of \( \varepsilon_r \) reaches up to \( \sim 31 \) 000 over the range from 100 Hz to 100 kHz, revealing the high quality of Mn-doped KTN used in the experiments. To maintain the quality of the excited space-charge field, the experimental temperature was set as 26 \(^{\circ}\)C. A condition of slightly more than the Curie temperature not only benefits the enhancement of the gradient distribution of the refractive index through a large quadratic electro-optical effect, but also maintains the quality of the transmitted beam, avoiding the scattering based on the domain walls. According to the previous researches, the dynamic polar nano-regions (that is PNRs) mainly contribute to the large quadratic electro-optical effect at the temperature slightly over the Curie point. The nanoscale and correlative characters of PNRs promote the quadratic electro-optical effect and keep the high optical quality [34].

The change of refractive index along the \( x \) axis is related to the quadratic electro-optic coefficient \( s_{11} \) and applied electric field \( E \), obeying the following equation:

\[
\Delta n_x = -\frac{1}{2} n_o^2 s_{11} E^2,
\]

(1)

where \( n_o \) is the intrinsic refractive index of Mn-doped KTN. The value of \( s_{11} \) is important to follow-up research related to the distribution of \( \Delta n_x \). Thus, distribution of \( s_{11} \) in the crystal was shown in figure 1(b). The experimental setup is shown in figure 2. The results show that \( s_{11} \) is uniform in the crystal, because \( s_{11} \) and \( n_o \) are constants at a certain temperature. \( \Delta n_x \) mainly depends on the \( E \), indicating that the distribution of \( \Delta n_x \) can be designed by employing the space-charge electric field. While, the form of irradiation induces the different \( E \). Mn doping is an effective way of improving the photorefractive properties of KTN [3]. According to the band transport model, the charge carriers are excited upon irradiation in Mn-doped KTN. Then, the electrons or holes move from the acceptor energy level to the conduction or valence band. The movements of charge carriers are controlled by diffusion, drift, and the photovoltaic effect. After iterated captures and excitations, the stable distribution of space charge is built, which provides \( E_{sc} \) as follows [35]:

\[
E_{sc} = E_{\infty} + \frac{I_{0} + I_{\infty}}{I + I_{d}} \frac{k_b T}{e} \frac{1}{I + I_{d}} \frac{\partial I}{\partial x} - E_{B} \frac{I - I_{\infty}}{I + I_{d}},
\]

(2)

where \( E \) is the applied electric field; \( I_{0}, I_{\infty} \), and \( I \) are the dark irradiation intensity, light intensity at the location of infinity on the \( x \) axis, and irradiation intensity, respectively; and \( k_b, T, \) and \( e \) are the Boltzmann constant, environmental temperature, and electron charge, respectively. The distribution of \( I \) is the function of \( x \). The first, second, and third items show the drift, diffusion, and photovoltaic effect, respectively. The photovoltaic effect in Mn-doped KTN mainly originates from the polarization of ferroelectric domains,
Figure 1. (a) Temperature dependences of relative dielectric constants $\varepsilon_r$ for Mn-doped KTN single crystal. (b) Quadratic electro-optic coefficient $s_{11}$ of Mn-doped KTN crystal. White dotted rectangle shows the crystal region.

Figure 2. Experimental setup for constructing and controlling refractive index gradient. $\lambda/2$, half-wave plate; SF, spatial optical filter; L1, lens; L2, cylindrical lens; M, mirror; BS, beam splitter; CCD, charge-coupled device.

Figure 3. (a) Distribution of $\Delta n$ after irradiation under a 1000 V applied voltage. (b) $E_{sc}$ corresponding to different locations in the sample.

which could be ignored in this research because the sample is in the cubic phase. Equation (1) shows $E_{sc}$ mainly related to the intensity and distribution of irradiation, providing an approach to control the distribution of $E_{sc}$ based on the designed distribution of irradiation.

To excite charge carriers, a 491 nm laser was used to irradiate the sample at 26 °C. The form of irradiation is shown in figure 2. The 491 nm laser was modulated by a cylindrical lens and slit to provide a gradient distribution of intensity in the $x$ direction in the sample. The He–Ne laser (632.8 nm), which
cannot excite the charge carriers because of the small photon energy, carries the information of the distribution of the refractive index in the sample. The UV–vis DRS was measured to make sure the irrelevant of He–Ne laser in exciting of space-charge, as shown in figure S1 (supplementary material (https://stacks.iop.org/NJP/23/013014/mmedia)). The results shows the bandgap of Mn-doped KTN is 2.5 eV according to the Tauc formula. The polarization of the He–Ne laser is parallel to the x axis and is employed to monitor the signal controlled by $s_{11}$. $\Delta n$ upon irradiation is shown in figure 3(a), demonstrating the one-dimensional distribution of $\Delta n$ along the x axis. Since $\Delta n$ is controlled by $E_{sc}$, the distribution of $E_{sc}$ can be calculated using equation (1). The gradient distribution of the space-charge field in different locations along the x axis is shown in figure 3(b). The red region is the bright area of irradiation, which is mainly the region of charge-carrier excitation. $E_{sc}$ has a nearly linear relation with $x$ in the range of −0.6 to 0.7 mm in the x direction, while the difference along the y axis is small. The maximum of $E_{sc}$ reaches up to 208 V mm$^{-1}$, which is comparable to an applied electric field of 242 V mm$^{-1}$.

The gradient refractive index can be used for optical deflection. As a light beam (polarized parallel to the electric field) propagates through the crystal, the deflection angle $\theta$ of the output light is expressed as [17]

$$\theta = l \left| \frac{d\Delta n(x)}{dx} \right| = n_{0}^{3} l s_{11}(E + E_{sc}(x)) \left| \frac{dE_{sc}(x)}{dx} \right|,$$

where $E$ is the external electric field, $\Delta n(x)$ is the distribution of the refractive index along the x axis, and $l$ is the thickness of the sample in the direction of light transmission. Equation (3) indicates that $\theta$ is proportional to $E$ because the distribution and intensity of $E_{sc}$ are invariable in the test period. Figure 4 indicates the distribution of $\Delta n$ under different $E$. The gradient distribution of $\Delta n$ along the x axis in the center of the sample is obviously altered under the effect of $E$. To further study the distribution features of $\Delta n$, we chose the $\Delta n$ at the position of $x = −1.7$ mm and $y = 0$ mm as the original point $\Delta n_0$. The differences between the $\Delta n$ on the line of $y = 0$ mm in the range of $−1.7$ to $1.8$ mm along the x axis and $\Delta n_0$ under different electric fields are shown in figure 4(c). The distribution of $\Delta n$ can be divided into three parts. The range from $x = −1.7$ mm to $−0.7$ mm shows a very small change of $\Delta n$, because only a few charge carriers can be excited in the absence of enough irradiation [36]. The uniform $\Delta n$ in this partition is mainly attributed to the quadratic electro-optical effect yield by applied electric field. The range from
Figure 5. (a) Theoretical and experimental results of deflection angle \(\theta\) under different electric fields. (b) Frequency response of deflection signal under ac voltage with an amplitude of 150 V and a frequency of 50 kHz.

\[ x = -0.7 \text{ to } 0.7 \text{ mm} \] indicates the large gradient of \(E_{sc}\). The range from \(x = 0.7\) to \(1.8\) mm matches the region upon high intensity irradiation, which exhibits a large \(E_{sc}\). \(\theta\) at different \(x\) locations under different voltages is calculated according to equation (3), as shown in figure 4(d). The time stability is necessary for the application. The additional time dependence of space-charge field was measured in supplementary material, as shown in figure S2. The results show that it has good time stability within 5 h with keeping 66% of original value. The similar research was studied in Mn:Fe:KTN co-doped crystal with over 18 days of maintaining stability, which reveals the potential for application [37].

The center of the beam is extracted to represent the average theoretical deflective angle, which is shown in figure 5(a). The red line is the theoretical deflection angle of the center of the beam spot, while the black line denotes the experimental results by measuring the change of spot location. Obviously, the theoretical results agree well with the experimental results, exhibiting a linear relation of \(\theta\) versus \(E\). Figure 5(b) shows the frequency response of designed deflector, manifesting the good frequency response of the device. Deflection performance is maintained at 50 kHz. The response time was also measured by applying an impulse voltage. The result shows that the response time of the deflector, defined as the time 90% of maximum signal, is \(\sim 19\ \mu\text{s}\), which is larger than the response time of the quadratic electro-optical effect [23]. One of the reasons for this is the drift of charge carriers for the photorefractive effect. The drift of charge carriers needs time, which leads to the longer response time. The other reason is that the sample is in the phase boundary between ferroelectric and paraelectric phases at experimental temperature. Previous research showed that KTN crystal has local correlated dipoles near Curie temperature. The reorientation dynamics under applied electric field of large-size correlated dipoles need more time to respond the electric field, which is also the important reason of extended response time [34, 38]. Nevertheless, the response time of the implemented deflector is still fast enough, which meets the requirement of response rate for current optical communication and imaging optics.

3. Conclusion

In summary, the photorefractive space-charge field was controlled upon setting the designed light irradiation in an Mn-doped KTN single crystal. The distribution of space-charge field was studied by DHM. The distributed space-charge field induces the gradient distribution of refractive index. Furthermore, the gradient distribution of refractive index was modulated under applied electric field through the quadratic electro-optical effect. The optical deflector was achieved based on the field-alterable refractive index gradient. The deflection angle reaches 0.3° and scanning frequency response maintains good characteristics at 50 kHz. The experimental results of deflection agree with the theoretical results, which proves the feasibility of the construction method of space-charge field and design of the deflective device proposed herein. The findings in this work can provide guidance for engineering space-charge fields and distributed refractive indexes based on the photorefractive effect, which is beneficial for designing and manufacturing highly functionalized optical devices.
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