Ultraviolet laser-induced photovoltaic effects in miscut ferroelectric $LiNbO_3$ single crystals*

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This paper investigates the photovoltaic properties of miscut $LiNbO_3$ single crystal with different thicknesses under irradiation of a 248 nm ultraviolet laser pulse with 20 ns duration without an applied bias. Nanosecond photovoltaic response is observed and faster rise time is obtained in thinner samples. In accord with the 248 nm laser duration, the full width at half maximum of the photovoltaic signals keeps a constant of $\sim$ 20 ns. With decrease of the crystal thickness, the photovoltaic sensitivity was improved rapidly at first and then decreased, and the maximum photovoltage occurred at 0.38 mm-thick single crystal. The present results demonstrate that decreasing the $LiNbO_3$ single crystal thickness can obtain faster response time and improve the photovoltaic sensitivity.

Keywords: photovoltaic effect, $LiNbO_3$ single crystal, photodetector
PACC: 7847, 7240, 4280

1. Introduction

Lithium niobate $LiNbO_3$ (LNO), a ferroelectric crystal with a spontaneous polarisation caused by the displacement of Li^+ and Nb^{5+} cations along the crystallographic c axis, has been extensively researched for its excellent ferroelectric, piezoelectric, dielectric, pyroelectric electric-optical and nonlinear optical properties.[1−5] The LNO is widely used for holographic data storage, frequency doubling, waveguide structures, and even for acoustical memory.[6−8] Recently ultraviolet (UV) photodetectors have attracted a strong interest owing to their broad potential applications in the fields of automatization, short-range communications security, biological researches, and military services. The band gap of LNO single crystal is $\sim$4 eV,[9] and can be suggested as a promising material for UV photodetector since the present UV photodetectors based on various wide band gap semiconductors, such as III–V nitrides, silicon carbide, zinc oxide, and diamond, require a complicated fabrication process and high-cost manufacturing.[10−13]

In this paper, we focused on the thickness dependence of UV laser-induced photovoltaic properties of miscut pure congruent LNO single crystal. By decreasing the LNO crystal thickness, we not only observed the faster-response photovoltaic effect, but also found that the photovoltaic sensitivity was improved greatly.

2. Experimental details

In the experiment, a single polished commercial LNO single crystal with c-axis oriented is used for the photovoltaic studies. The sample geometry is 5×5 mm^2 with the thickness of 0.5 mm. The crystalline orientation is 10° miscut from the exact (001) orientation, which is characterized by x-ray diffraction with the usual $\theta - 2\theta$ scan using Cu Kα_1 and Kα_2 radiations. As shown in Fig. 1, the offset point is set as $\omega = \alpha$ or $45^\circ - \alpha$ to satisfy Braggs diffraction, where α is the miscut angle. The two peaks arise from the (006) and (0012) plane of LNO and are clearly apart for Cu Kα_1 and Kα_2 radiations. The inset of Fig. 1 shows the UV–visible absorption spectrum of LNO crystals as a function of the wavelength. The

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sharp absorption edge is ∼310 nm, corresponding to the optical band gap of LNO and indicating that UV light, e.g. 248 nm laser, can generate electron–hole carriers in LNO crystals.

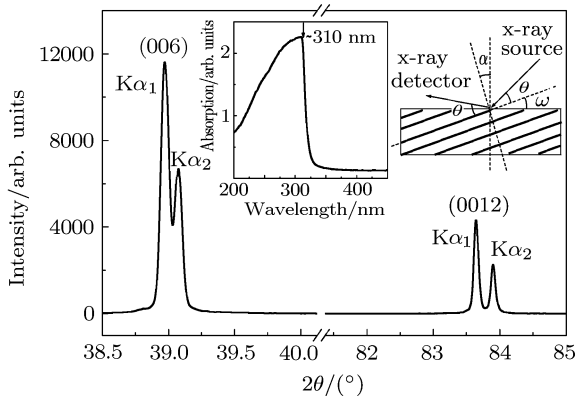


Fig. 1. The x-ray diffraction pattern of a miscut LNO single crystal. The left inset shows the absorption spectrum of LNO single crystal. The right inset shows the configuration of $\theta - 2\theta$ scan, where α is the miscut angle and ω the offset point.

The photovoltaic properties are investigated under the illuminations of a KrF pulsed laser with the wavelength of 248 nm with 20 ns duration at a 10 Hz repetition. In order to study the influence of the thickness on the photovoltaic effect, the samples are polished mechanically into seven different thicknesses, which are 0.49, 0.45, 0.38, 0.28, 0.22, 0.17 and 0.09 mm, respectively. Before the photovoltaic measurements, the sample is cleaned by using an ultrasonic cleaner in alcohol and acetone under routine cleaning process. Two colloidal silver electrodes of about 1×5 mm^2 area, separated by 3 mm, are prepared on the mirror polished surface of the LNO single crystal and the opposite surface is exposed wholly to the laser irradiation, as shown in the inset of Fig. 2(a). The photovoltaic signals are recorded by using a sampling oscilloscope terminating into 50 Ω at room temperature. All the measurements are carried out in the room temperature without any applied bias.

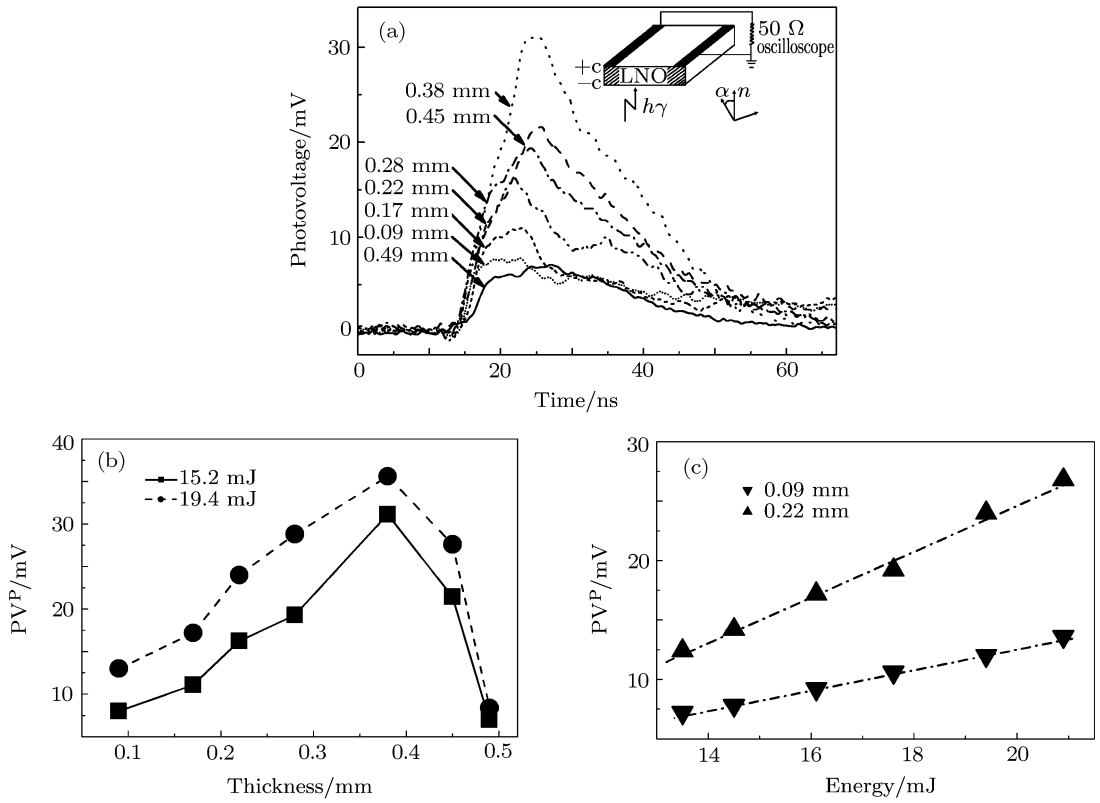


Fig. 2. (a) The photovoltaic pulses for miscut LNO single crystal under the illumination of a 248 nm laser for seven different thicknesses recorded by an oscilloscope with an input impedance of 50 Ω at room temperature without any applied bias. The on-sample energy was 15.2 mJ. The inset displays the schematic circuit of the measurement. (b) The peak signal PV^P dependence of the sample thickness. (c) The PV^P as a function of on-sample energy.

3. Results and discussion

Typical voltage transients of the LNO single crystal of different thicknesses to a pulsed laser illumination are presented in Fig. 2(a). The peak photovoltage (PV^P) as a function of the sample thicknesses is shown in Fig. 2(b), and the energies on the sample are 15.2 and 19.4 mJ, respectively. With the decrease of the crystal thickness from 0.49 to 0.09 mm, the PV^P increases rapidly at first and then descendes. The maximum PV^P occurs at the thickness of 0.38 mm and reaches 35.6 and 31.1 mV for 19.4 and 15.2 mJ, with the corresponding photovoltaic sensitivities of 1.83 and 2.04 mV/mJ, which is several times larger than that at 0.49 and 0.09 mm. In addition, the PV^P is found to depend linearly on the on-sample energy from 13.5 to 20.9 mJ for selected crystal thicknesses of 0.22 and 0.09 mm, respectively, as shown in Fig. 2(c).

Figure 3 shows the 10%–90% rise time (RT) and full width at half maximum (FWHM) as functions of the sample thickness for the on sample-energy of 15.2 and 19.4 mJ, respectively. With the decrease of sample thickness, the carriers reaches the two colloidal electrodes faster for thinner sample so that faster photovoltaic response can be observed. The RT descends obviously from 11.83 ns at 0.49 mm to 3.946 ns at 0.09 mm, suggesting that decreasing the sample thickness is a useful way to obtain faster response detection. Since the miscut LNO single crystal exhibits an optical response time of $\sim$ ns, it can be applied to detect the present laser pulse, which is further confirmed in Fig. 3, where the FWHM is independent of crystal thickness and almost keeps a constant of $\sim$20 ns in accord with the 248 nm laser duration.

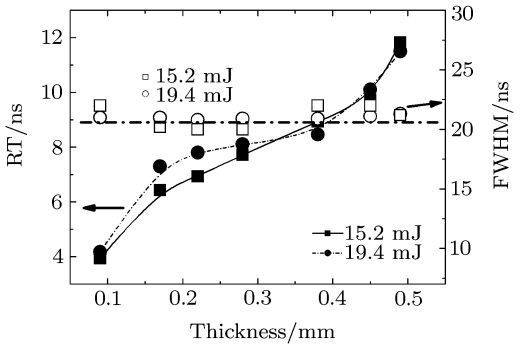


Fig. 3. The RT and FWHM as functions of the sample thickness.

It is well known that pure congruent LNO is a spontaneous polarisation crystal and there exists a spontaneous polarisation electric field (SPEF) along the c axis. In the experiment, the crystalline orientation is 10° miscut from the exact (001) orientation, so is SPEF as shown in Fig. 4. Under the 248 nm laser irradiation, photo-induced carriers are separated and assembled at the two electrodes by the SPEF. Only those carriers reaching to the electrodes in the back can be collected and give rise to photovoltaic signals. With decrease of crystal thickness the amount of carriers collected by electrodes in the back increases due to lower loss, which is resulted from the shorter transport distances as well as less traps and recombination in thinner samples. Thus the signals for thinner samples are much larger than that for thicker samples. On the other hand, the decrease of thickness also leads to decrease of autologous carriers in LNO single crystals, which limits the amount of carriers collected by electrodes. The competition between the two above factors results into a maximum photovoltaic signal at an optimum thickness at 0.38 mm.

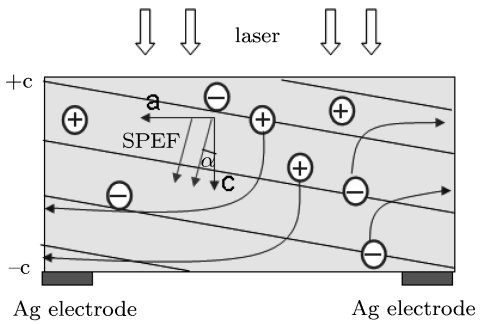


Fig. 4. The schematic diagram of the transporting photogenerated carriers, indicating that the carriers were separated and assembled at the two Ag electrodes by the SPEF.

4. Conclusion

In conclusion, we have investigated the thickness dependence of the photovoltaic effect in miscut LNO single crystal. With the decrease of the crystal thickness, the photovoltaic response time decreased monotonically, the photovoltaic sensitivity is improved rapidly at first, and then decreases. The experimental results show that decreasing the crystal thickness is an effective method for obtaining faster response time and improving the photovoltaic sensitivity in single crystals, suggesting potential applications in optoelectronic detection.

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