The effect of an optical pump on the absorption coefficient of magnesium-doped near-stoichiometric lithium niobate in terahertz range*

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The absorption coefficient of magnesium-doped near-stoichiometric lithium niobate crystal is measured by terahertz time-domain spectroscopy in a frequency range of 0.2 THz–0.9 THz at room temperature. The absorption coefficient is modulated by external optical pump fields. Experimental results show that the absorption coefficient of near-SLN:Mg crystal is approximately in a range of 22 cm^{-1}–35 cm^{-1} in a frequency range of 0.2 THz–0.9 THz and tunable up to nearly 15%. Further theoretical analysis reveals that the variation of absorption coefficient is related to the number of light-induced carriers, domain reversal process, and OH^- absorption in this crystal.

Keywords: near-stoichiometric $LiNbO_3$:Mg, terahertz, absorption coefficient, domain reversal

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1. Introduction

Lithium niobate ($LiNbO_3$, LN) exhibits attractive material properties for quasi-phase matching frequency conversion.[1] In the past several decades the research on the generation of terahertz (THz) wave in periodically poled lithium niobate (PPLN) has drawn much attention.[2–4] However, the high power application of PPLN-SHG devices is limited by the photorefractive damage and the green light-induced infrared absorption.[5] Furukawa *et al.*[6] observed that the optical damage of LN may be reduced by Mg doping. Pálfalvi *et al.*[7] reported that doping small amount of MgO could reduce the light-induced absorption of LN. Since then magnesium-doped LN (LN:Mg) has been regarded as a potential material for quasi-phase-matching of nonlinear-optic and electro–optic devices because of its high resistance to optical damage and its low coercive field for ferroelectric domain reversal.[8]

It is noted that compared with congruent $LiNbO_3$ (CLN) crystal, near-stoichiometric $LiNbO_3$ (near-SLN) crystal has some improved optical properties, such as short response time,[9] high exponential gain coefficient,[10] high sensitivity and dynamic range,[11] strong photodamage resistance ability,[2] large electro–ferroelectric effects,[12] and short absorption edge.[13] Due to the excellent optical performance of near-SLN, many properties of this crystal have been experimentally and theoretically studied.[14]

However, the effect of optical pump on the absorption coefficient of near-SLN crystal in the THz range has rarely been studied so far. In this work, the transmission spectrum of magnesium-doped near-stoichiometric $LiNbO_3$ (near-SLN:Mg) in the THz range under an external optical pump field is measured. Experimental results show that the absorption coefficient of near-SLN:Mg crystal in a frequency range of 0.2 THz–0.9 THz is approximately between 22 cm^{-1} and 35 cm^{-1} and tunable up to nearly 15%. Further theoretical analysis reveals that the variation of absorption coefficient is related to the number of light-induced carriers, domain reversal process, and OH^- absorption in this crystal. This work may be helpful for the applications of SLN crystal in the THz field.

2. Experiment

A near-stoichiometric $LiNbO_3$:Mg crystal containing 1 mol% Mg was grown using the top-seeded-solution-growth method.[15,16] The ratio of Li to Nb, estimated from the Curie temperature measurement, was higher than 49.8/50.2. The crystal was cut and polished to form a plate with its polar face perpendicular to the z axis and dimensions of 10 mm × 10 mm × 0.41 mm.

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A THz time-domain spectroscopy (THz-TDS) system[17] was used to measure the transmission spectrum at room temperature, which is shown in Fig. 1. In this THz-TDS system, a fiber femtosecond laser beam (780-nm central wavelength, 50-MHz repetition rate, and 90-fs pulse width) was divided into two beams by a polarized beam splitter 1 (PBS1), namely, pump beam and probe beam. A half-wave plate was placed to change the ratio between the pump beam and probe beam. The pump beam was directed through retroreflector 1 (RR1), quarter wave plate 1 (QWP1), and PBS1 to lens 1 (L1) and focused on the GaAs emitter to generate THz radiation. The probe beam was directed to PBS2 by mirrors of M1 and M2. The reflected beam passed through QWP2 and was reflected by RR2 and mirror M3. RR1 and RR2 were attached together on a linear step stage to form the time delay line.

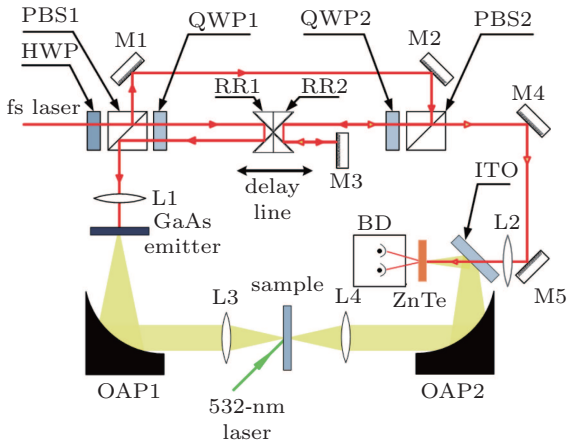


Fig. 1. (color online) Installation diagram of TDS. A CW green laser is obliquely incident upon the surface of the sample at an angle of 45° with the polar axis.

After passing through PBS2, the probe beam was directed to a ZnTe crystal and a balance detector (BD) through two mirrors (M4 and M5), L2, and an ITO crystal. The generated THz radiation was collimated by an off-axis parabolic mirror 1 (OAP1) and then passed through two confocal polyethylene lenses, L3 and L4. The sample was placed at the focus of L3 and L4. Finally, the transmitted THz radiation was focused by OAP2 and reflected by ITO crystal to ZnTe crystal and BD for coherent detection. The detectable frequency ranged from 0.2 THz to 0.9 THz (6.7 cm^{-1} to 30 cm^{-1}), with a dynamic range of more than 60 dB. The measured frequency resolution was 10 GHz, and the spot size at the focus was 3 mm. An all-solid-state continuous wave (CW) green laser (central wavelength of 532 nm) was used to provide an external optical pump field. The light was obliquely incident on the surface of the samples at an angle of 45°, and the spot size had a diameter of 5 mm.

3. Results and discussion

The time-domain waveforms are obtained experimentally. As shown in Fig. 2(a), the time lag between the reference (air) and the sample is approximately 5.52 ps. The second and third waveforms at 33.18 ps and 46.96 ps, respectively, are induced by the multiple internal reflections of the sample. With the intensity of optical pump increasing from 0 mW to 260 mW, time shifts are observed in the time domain. For example, the transmission waveform shifts approximately 0.16 ps when the external optical pump intensity is 260 mW compared with that without optical pumping as shown in Fig. 2(b). The intensity dependence of time shift is shown in the inset of Fig. 2(b).

We calculate the transmission spectrum and the intrinsic phase shift of the sample by the Fourier-transform method.

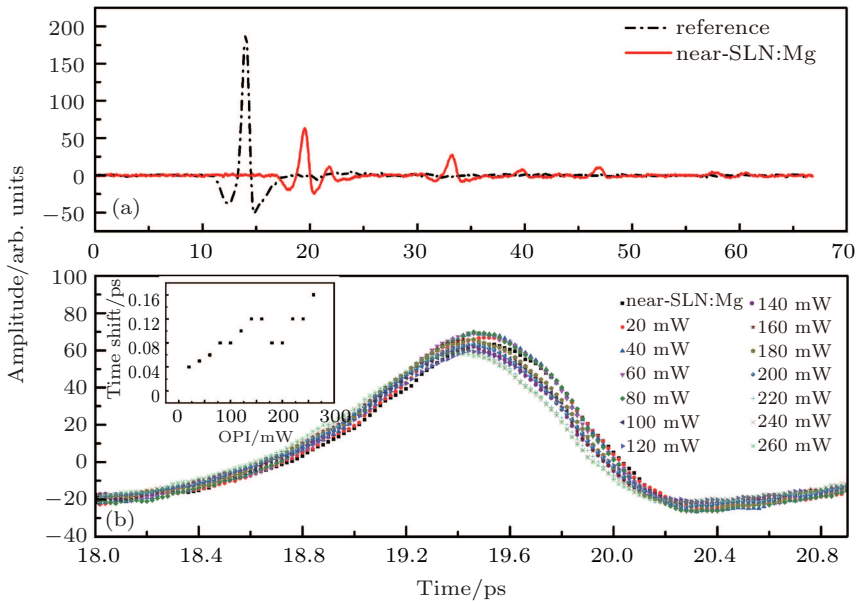


Fig. 2. (color online) (a) Time-domain reference and transmission waveforms of near-SLN:Mg crystal and (b) time shifts under different external optical pump fields at room temperature. The inset shows the time shift (relative to the curve belonging to zero intensity) versus external optical pump intensity (OPI).

Figure 3 shows the transmission spectra of near-SLN:Mg crystal under different external optical pump intensities at room temperature. It could be found that the transmittance is dependent on the external optical pump intensity.

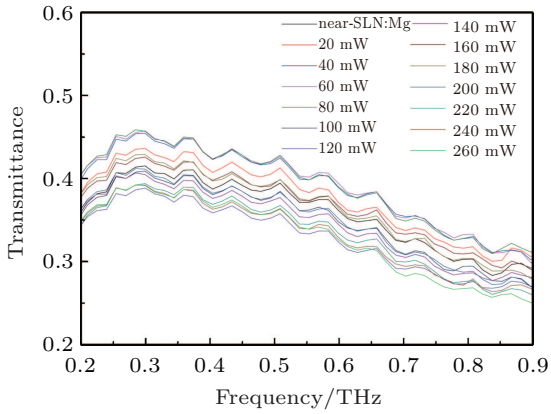


Fig. 3. (color online) Transmission spectra of near-SLN:Mg crystal under different external optical pump intensities at room temperature.

In order to investigate the influence of an optical pump on the absorbance of near-SLN:Mg crystal, we calculate the frequency dependence of absorption coefficient as shown in Fig. 4(a). The absorption coefficient of near-SLN:Mg crystal can be expressed as $\alpha = -\ln(T)/d_0$, where T is the transmittance and d_0 is the thickness. In a frequency range from 0.2 THz to 0.9 THz, the absorption coefficient of near-SLN:Mg crystal is approximately in a range from 22 cm^{-1} to 35 cm^{-1}. This result is in good agreement with the previous discussion on SLN and CLN.[18] Under different external optical pump intensities at room temperature, the absorption coefficient is tunable up to nearly 15%. The variations $\Delta\alpha$ of absorption coefficient with the intensity of optical pump are plotted in Fig. 4(b). $\Delta\alpha$ is the difference in absorption coefficient between the conditions with and without an optical pump. The previous theory indicated that as the number of light-induced carriers increases with optical pump intensity increasing, the conduction loss will continue to increase, resulting in the increase of absorption coefficient.[19,20] However, in our experiments, when the external optical pump intensity increases from 0 mW to 260 mW, the absorption coefficients does not keep increasing, but decreases in regions from 0 mW to 60 mW and from 140 mW to 180 mW.

For the purpose of studying what causes the absorption coefficient to decrease in the regions of 0 mW–60 mW and 140 mW–180 mW, we study the micromechanisms of the effect of external optical pump on the absorption coefficient of near-SLN crystal.

In the region of 0 mW–60 mW, we think that the decrease of absorption coefficient is attributed to the absorption of OH^- in crystal. The OH^- stretching vibration is sensitive to the change of the ion environment and it has been reported that the infrared absorption band is related to the OH^- stretching vibration in LN.[21–23] There are two reasons leading to the decrease of the absorption coefficient with external light intensity increasing.[24,25] Firstly, the transition dipole moment is diminished due to some charge redistribution in the near-SLN:Mg during light illumination and hence the OH bond is IR inactive. Secondly, the OH bond is completely broken. With the optical pump further increasing to 140 mW, the vibrational mode and the number of OH^- would not change any more because of the limited concentration. The conduction loss plays a major role. So the variation of absorption coefficient increases with external optical pump intensity increasing.

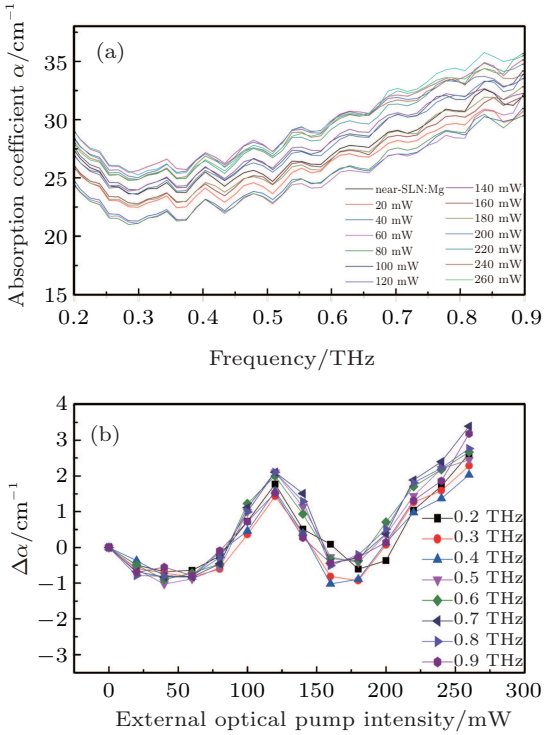


Fig. 4. (color online) Frequency dependences of (a) absorption coefficient and (b) variation values of absorption coefficient for near-SLN:Mg crystal under different intensities of light pump at room temperature.

In the region of 140 mW–180 mW, the decrease of absorption coefficient is due to the fact that the internal space charge field in the near-SLN:Mg is sufficiently enhanced when the external optical pump intensity increases to a value higher than 140 mW. Thus, light-induced domain reversal occurs in the crystal, resulting in the decrease of the spontaneous polarization.[8] This effect gives rise to a reduction in the area of hysteresis loop, i.e., the hysteresis loss,[26] resulting in the corresponding decrease of absorption coefficient. When the external optical pump intensity increases to a value higher than 180 mW, the domain reversal is saturated, and the increase of absorption coefficient is attributed to the increase of conduction loss again.

Note that the light-induced domain reversal in the near-SLN would lead to a nonlinear variation of refraction index.[27] As a result, the external optical pump intensity de-

pendence of variation of refraction index is calculated to confirm the existence of domain reversal in this crystal as shown in Fig. 5. When the optical pump intensity increases from 0 mW to 140 mW, due to the photorefractive effect,[28,29] the variation of refractive index linearly increases. When the intensity of external optical pump is higher than 140 mW, the refractive index first decreases and then increases. This result demonstrates that when the external optical pump intensity increases from 140 mW to 180 mW, the light-induced domain reversal process occurs, and the decrease of absorption coefficient is due to the domain reversal in the near-SLN:Mg crystal.

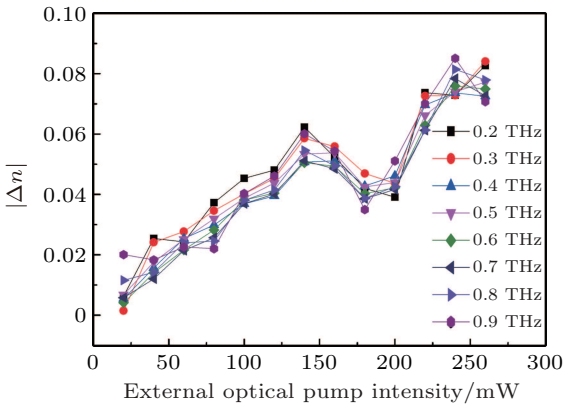


Fig. 5. (color online) Plot variation value of refractive index for the near-SLN:Mg crystal versus external optical pump intensity at different frequencies.

4. Conclusions

In this paper, we have both experimentally and theoretically investigated the effect of an external optical pump on the absorption coefficient of near-stoichiometric $LiNbO_3$:Mg crystal in the THz range at room temperature. An obvious modulation of absorption coefficient of near-SLN:Mg crystal is observed by applying different external optical fields. The two decreasing regions of absorption coefficient with optical pump intensity increasing are theoretically explained by the OH^- absorbance and domain reversal process respectively. Furthermore, we experimentally confirm the existence of domain reversal in the near-SLN:Mg crystal. It is concluded that the OH^- absorbance and domain reversal process occur in different pump intensity regions respectively, resulting in the observed modulation of the absorption coefficient in the near-SLN:Mg.

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