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Nonvolatile holographic recording in Ti, Fe Co-doped $LiTaO_3$ crystal

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ABSTRACT

Ti, Fe Co-doped $LiTaO_3$ (LT) crystals have been grown by the Czochralski method from the congruent melts. The absorption spectra of crystal were measured before and after ultraviolet illumination. Holograms have been recorded in doubly doped crystals with continuous-wave laser light by use of two-color method. The maximum value of refractive-index changes 7×10^{-5} is achieved.

1. Introduction

$LiTaO_3$ (LT) crystals are widely used in many areas because of their excellent piezoelectric, electro-optic, and nonlinear optic properties. Because of its excellent photorefractive properties, LT crystals can be applied in many areas, such as piezoelectric, electro-optic, surface acoustic wave, waveguide, and nonlinear optical devices [1–5]. LT crystals also are promising materials for applications of holographic data storage. It is similar to $LiNbO_3$ (LN), but the photorefractive properties of doped LT are less studied. The reason for this might be the higher melting point of LT and the band gap is larger than that of LN [6]. The larger band gap made the spectral sensitivity of doped LT shift to the ultraviolet region. The most sensitive Fe:LN is around 480 nm, and Fe:LT is around 400 nm. The advantage of Fe:LT is the larger dark storage time than that of Fe:LN [6].

Volume holography is attractive for data storage because of its large storage densities. One crucial problem with photorefractive crystals is the volatility of stored information, because the electrons are redistributed during readout process. Two-color holography is an effective solution for the volatile problem [7]. The crystals have two different traps (shallow and deep), the energy levels of traps between the valence and conduction bands, and crystals are sensitive to the recording light only if additional gating light is present simultaneously. Without gating light, the readout light is nondestructive. In Fe:LT crystals, two-color holography was performed with continuous-wave laser light as reported in [8]. Holograms were recorded with a 660 nm diode laser, and gating light is an Ar laser at 488 nm or a 150 W Xe lamp. The two-color holography performed in the Fe:LT crystals relies on the existence of two sets of traps, $Fe^{2+/3+}$ is deep center and intrinsic $Ta_{Li}^{4+/5+}$ ions is shallow center. The problem of nonvolatile holographic storage in congruent Fe-doped LT is the diffraction efficiency is small and the recording light intensity is high. Because of the similarity between LN and LT with respect to the charge transport, the methods developed for LN also fit the LT crystals. Some efforts have been dedicated to lower the light intensity by use of doubly doped LN crystals and stoichiometric LN crystals in two-color holography. Two-color holographic recording used near-stoichiometric LT have done for improved diffraction efficiency [9].

In this letter, we have grown Ti:Fe:LT crystals using the Czochralski method. We also investigate two-color holographic in Ti:Fe:LT crystals. It shows that this material exhibits good two-color holographic characteristics.

2. Crystal growth and sample preparation

Ti:Fe:$LiTaO_3$ were grown from congruent melts by the conventional Czochralski (CZ) method using an intermediate frequency(IF) furnace. The raw materials used to grow the crystals were Li_2CO_3, Ta_2O_5, Fe_2O_3 (0.06 wt%), and TiO_2 (1 mol%) with the purity of 99.99%. The crystals were grown along the *b*-axis from the polycrystalline material in diameter-controlled equipment. And they were rotated at 10–20 rpm and growth rate was 0.5 mm/h. The axial temperature gradient of the IF furnace was 40–50 °C/cm. About 60% of the melts were crystallized. After growth, the crystals were annealed to room temperature at the

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speed of 60 °C/h. The diameter of crystals was 15 mm with a length of 20 mm; it appeared to be transparent, crack free, and inclusion free. The crystals were then polarized in another resistive furnace in which the temperature gradient was almost close to zero using an applied DC electric density of 10–15 mA/cm^2 at 700 °C. After 15 min, the crystal was cooled down to room temperature at the rate of 30 °C/h. The samples were cut into wafers with 2 mm thickness and polished to optical grade.

3. The photorefractive properties of Ti:Fe:LiTaO$_3$ crystal

The absorption spectra of the Ti:Fe:LT were recorded using CARYIE style UV–visible spectrophotometer with a wavelength range 200–700 nm.

Absorption measurements were performed before and after 355 nm ultraviolet illumination for 30 min with 20 mW/cm^2, as illustrated in Fig. 1. Each dopant has an influence on the absorption spectra. The unexposed crystals having the absorption at 400 nm is due to an excitation of electrons from Fe^{2+} to the conduction. The absorption band edge of the Ti:Fe:LT crystals shifted to a longer wavelength than that of Fe doped. The other absorption at 550 nm is may be due to the Ti dopant. Absorption spectrum of the exposed sample exhibits increase in the absorption compared to that of the unexposed one.

The diffraction efficiency is defined as the ratio of diffracted beam intensities and total beams intensities. The diffraction efficiency is detected with a 780 nm laser and its beam is extraordinarily polarized (Fig. 2). Gating intensity dependence of saturation diffraction efficiency with a constant recording intensity of 5 W/cm^2 is shown in Fig. 3.

In LN and LT crystals, the most important shallow levels have been identified; they are Nb_{Li}^{5+} and Ta_{Li}^{5+} antisite defects. In Ref. [8], they assumed that intrinsic acts as a shallow center, and

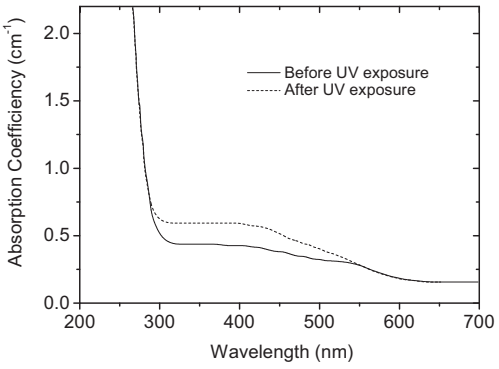

Fig. 1. Absorption spectra of Ti:Fe:LiTaO$_3$ crystals with and without ultraviolet illumination.

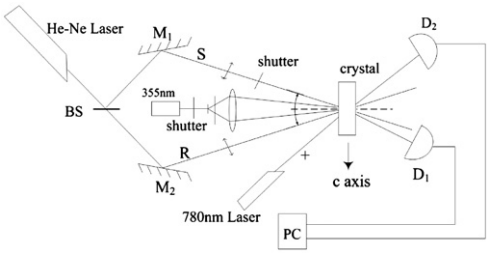

Fig. 2. Experimental setup of detects diffraction efficiency. BS: beam splitter; M1, M2: mirrors; D1, D2: detector; S: signal beam; *R*: reference beam; PC: computer.

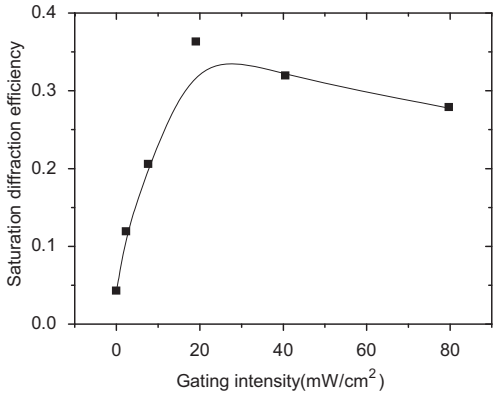

Fig. 3. Dependence of saturated diffraction on gating intensity at 365 nm with recording intensities of 5 W/cm^2 at 633 nm.

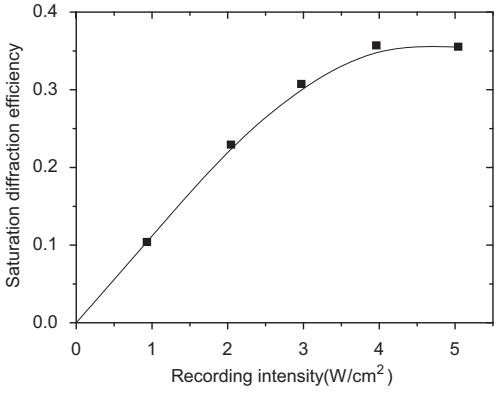

Fig. 4. Dependence of saturation diffraction efficiency on writing intensity at 633 nm with gating intensity of 20 mW/cm^2.

Fe acts as a deep center. Ti ions occupied Ta sites. Its absorption is close to trapped polaron centers. Electron transfer from the Ti^{3+} center to lattice Ta results in Ta^{4+} trapped polarons. The small continuous wave laser intensities can be used, because then the concentration of polarons is high in doped crystals. The doped crystals almost have two different types of energy levels: metastable shallow electron traps (antisite defects and Ta^{4+} trapped polarons) and Fe deep electron traps.

The saturation diffraction efficiency increases with increase in gating intensity first, and then decreases slowly. This is because the strong gating light reduced electrons from the recording center (shallow electron traps) and erased the grating. Weaker ultraviolet light produced less population in the recording center.

Fig. 4 shows that the saturation diffraction efficiency can reach a maximum with a suitable recording intensity. The nonvolatile holographic recording experiment chose the gating intensity as 20 mW/cm^2 and the recording intensity as 5 W/cm^2 at 633 nm.

4. Nonvolatile holographic recording in Ti:Fe:LiTaO$_3$ crystal

Two-color recording has been carried out at 633 nm with 365 nm gating light (Fig. 2). A diode laser was split into two beams of equal intensity, with 2.5 W/cm^2. These two beams of extraordinary polarization were made to intersect symmetrically inside the crystal, and the diameters of two beams are 2 mm, overlapped by a 6 mm diameter gating light. The grating vector is parallel to the *c*-axis. The period of elementary holographic gratings is about 3 μm. The writing intensity is adjusted with a half wave plate and a polarizer. And the gating light can be

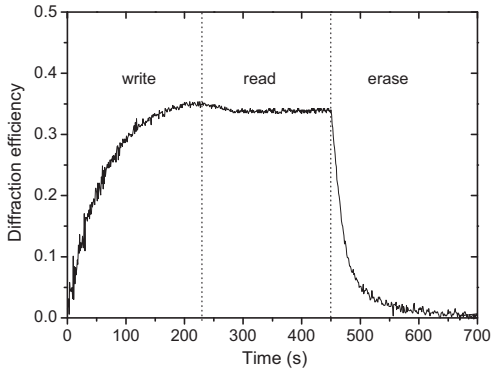

Fig. 5. Evolution of diffraction efficiency during a typical write–read–erase process in Ti:Fe:$LiTaO_3$ crystal.

changed with neutral density filters. The diffraction efficiency is detected with a 780 nm laser and its beam is extraordinarily polarized.

Fig. 5 illustrates the evolution of diffraction efficiency during recording and readout. The diffraction efficiency decreases at the initial readout process and then remains constant. The Ti:Fe:LT is insensitive to recording with red writing light alone. But it is possible to record hologram for illuminated one with additional gating light. We assume that in Ti:Fe:LT crystals, intrinsic defects and Ta^{4+} trapped polarons act as a shallow center, and $Fe^{2+/3+}$ acts as a deep center. So the recording process can be understood with a two-center charge-transport model. Homogeneous illumination with the gating light excites electrons from Fe^{2+} and Ti^{3+} to Ta_{Li}^{5+}, forming small polarons (Ta_{Li}^{4+}). Simultaneous illuminations with red light transfers the electrons to the conduction band, and migrate until trapped by Fe^{3+} ions. The photon energy of red light is not sufficient to excite electrons from Fe^{2+}, but it can remove the electrons from the shallow traps until all of them are trapped in deep traps and then the diffraction efficiency remains constant, so reading with red light is nondestructive.

Using Kogelnik's formula

$$\eta = \sin^2\left(\frac{\pi \Delta n d}{\lambda \cos\theta}\right) \quad (1)$$

where d denotes the crystal thickness, λ the vacuum wavelength, and θ half of the angle between the recordings beams inside the crystal. We can calculate the refractive-index change Δn. The maximum refractive-index change is 7×10^{-5}. It is a little small than that of nondoped near-stoichiometric LT [9]. But it is larger than the maximum value of 1×10^{-5} obtained before [8].

5. Conclusion

We have grown Ti, Fe-doped $LiTaO_3$ (LT) crystal by the Czochralski method, and demonstrated nonvolatile two-color holographic recording on it. The maximum saturation diffraction efficiency could be obtained by fixing the gating and recording intensities in the Ti:Fe:LT crystal. The maximum refractive-index change is 7×10^{-5}.

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