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PHOTOREFRACTIVE EFFECT OF Ce:Fe:LiNbO$_3$ CRYSTAL

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Ce:Fe:LiNbO$_3$ crystals were grown by Czochralski method, and was treated by oxidation or reduction. It was found that the exponential gain coefficient for the crystal with thickness of 0.5 mm was two times higher than that for one with the thickness of 2 mm. The effective charge density of the crystals with the reduction treatment was two times higher than that of the one with the oxidation treatment. The maximum phase conjugate reflectivity of crystals with the thickness of 2 mm was 75%.

Keywords: Ce:Fe:LiNbO$_3$ crystal; exponential gain coefficient; phase conjugating reflectivity.

1. Introduction

Single-crystal lithium niobate (LiNbO$_3$) is a useful material for various optical applications such as optical computing, image processing, optical modulators, volume holographic storage.[7,1] It is a well-known fact that the photorefractive properties of lithium niobate crystal can be enhanced significantly by doping the transition metal ions.[2,3] At present, Fe:LiNbO$_3$ becomes one of the most promising holographic storage material.[8] The main disadvantage of Fe:LiNbO$_3$ crystal as the holographic storage material is that its response rate is slow and the quality of output image or data is not satisfactory due to the scattering generated in Fe:LiNbO$_3$.[6] So it is needed to find the volume holographic storage materials whose light scattering resistance ability and response time are superior to those of Fe:LiNbO$_3$. Ce:Fe:LiNbO$_3$ crystal was first grown in our laboratory, and it was found that Ce:Fe:LiNbO$_3$ exhibits excellent photorefractive properties. This attractive feature makes it important for applications in holographic recording. In this paper, Ce:Fe:LiNbO$_3$ crystal was grown by Czochralski method, and the two-wave coupling exponential gain coefficient and the phase conjugate reflectivity was measured. It is found that for Ce:Fe:$LiNbO_3$, the thinner was thickness of the slice, the higher exponential gain coefficient was obtained. For example, that of the sample with the 0.5 mm thickness was up to its maximum value 91 cm^{-1} at the external angle of 34°, while that with the 2 mm thickness was up to its maximum value 28 cm^{-1} at the external angle of 36°.

2. Crystal Growth

Ce:Fe:$LiNbO_3$ crystal was grown by Czochralski method along the *c*-axis. The starting materials were Nb_2O_5, Li_2CO_3, CeO_2 and Fe_2O_3 with the purity of 99.99%. The melt composition was Li/Nb = 48.45/51.55 (atomic ratio). The weight concentration of CeO_2 and Fe_2O_3 was 0.1% and 0.03%, respectively. The temperature gradient was 35–40°C/cm, the pulling rate was 1–2 mm/h and the rotation rate was 15–25 rpm. The polarization current intensity was 5 mA/cm^2 at the polarization temperature of 1180°C. The crystal slice was treated by oxidation or reduction. Some crystal slices were reduced in the Li_2CO_3 powder at 500°C for 24 hours and others were oxidized in the Nb_2O_5 at 1100°C for 10 hours.

3. Exponential Gain Coefficient

It is a well-known fact that the two coupling exponential gain coefficient and phase conjugating reflectivity are the important technical parameters of the photorefractive crystal. If the external crossing angle 2θ for exponential gain coefficient of some photorefractive crystals is smaller than 10°, their applications are restricted greatly. The exponential gain coefficient Γ of the photorefractive crystal is defined as the ability that the energy of the pump light is transformed into the signal light during the coupling process. The measurement principle for Γ might be derived from the couple wave equation.[4] When the optical absorption and the reflections at the surface of the sample might be neglected, Γ was measured as

$$\Gamma = \frac{1}{d} \ln \left(\frac{I'_P I_S}{I_P I'_S} \right) \tag{1}$$

where I_S and I'_S were the transmitted intensities of signal with and without coupling, respectively, I_P and I'_P were the transmitted intensities of pump with and without coupling, respectively, and d was the interaction length in the crystal. Two writing beams for extraordinary polarization were derived from an argon ion laser, operating at 514.5 nm. The experiment setup for two-beam coupling is shown in Fig. 1.

The external crossing angle of two writing beams was designed as 2θ. The diameter of pump light was 5 mm. The diameter of signal was 1 mm with the intensity of 1.83 W/cm^2. The intensity ratio of the signal to pump light was 1844. Figure 2 showed the dependence of the exponential gain coefficient Γ on the 2θ. It showed that the exponential gain coefficient Γ of the sample with thickness of 2 mm was up to its maximum value 28 cm^{-1} at the external angle of 36° (a). That of the sample with the thickness of 1 mm was up to its maximum value 69 cm^{-1} at the

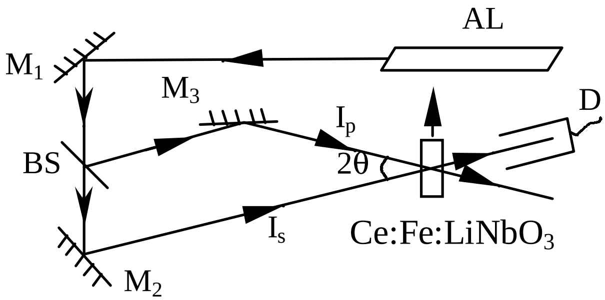

Fig. 1. Experimental schematic of two wave coupling: M_1, M_2 and M_3: mirrors; BS: beam splitter; AL: Ar^+-laser; D: detector; I_p and I_s: pump light and signal light.

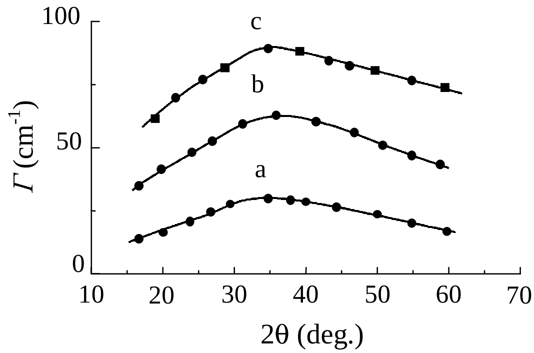

Fig. 2. Dependence of the exponential gain coefficient Γ on 2θ.

external angle of 34.5° (b) while that of the sample with the thickness of 0.5 mm was up to its maximum value 91 cm^{-1} at the external angle of 34° (c).

The relationship between the exponential gain coefficient and the external crossing angle can be expressed as follows[5,9]

$$\Gamma = \frac{A\sin\theta}{(1+B^{-2}\sin^2\theta)} \cdot \frac{\cos 2\theta_i}{\cos\theta_i} \tag{2}$$

where θ and θ_i are the half external and half internal beam crossing angles, respectively. A and B are the material constants. The effective charge density N_{eff} can be calculated by the following equation

$$N_{\text{eff}} = \left(\frac{4\pi}{e\lambda}\right)^2 \varepsilon_0\varepsilon k_B T \cdot \sin^2\theta_{\text{peak}}\,. \tag{3}$$

Here θ_{peak} was external crossing angle of two beams at maximum Γ value. The correlative parameters for doped $LiNbO_3$ were shown as follows: $e = 1.602 \times 10^{-19}$ C, $T = 300$ K, $\lambda = 488.0$ nm, $\varepsilon\varepsilon_0 = 2.832 \times 10^{-12} \text{C} \cdot \text{J}^{-1} \cdot \text{cm}^{-1}$. Table 1 shows the experimental results of two beam coupling of the crystals with 2 mm thickness.

Table 1. Results of two beam coupling experimental.

Crystal	Treatment	Γ (cm^{-1})	2θ (deg.)	N_{eff} (cm^{-3})
Ce:Fe:$LiNbO_3$	Reduction	28	36	2.9×10^{15}
Ce:Fe:$LiNbO_3$	As grown	22.2	28	1.4×10^{15}
Ce:Fe:$LiNbO_3$	Oxidation	9.8	21	1.0×10^{15}

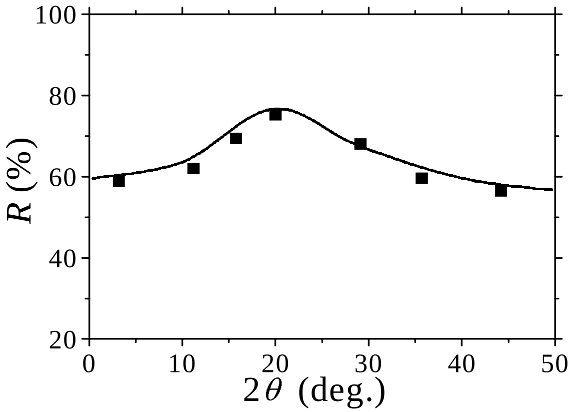

Fig. 3. Dependence of the phase conjugate reflectivity (R) on 2θ.

It is obvious that the exponential gain coefficient and the effective charge density were related to the reduction/oxidation treatment. The exponential gain coefficient and the concentration of effective charge density of Ce:Fe:$LiNbO_3$ with the reduction treatment were two times higher than those of the one with the oxidation treatment.

4. Phase Conjugate Reflectivity

The phase conjugate reflectivity of the crystals with 2 mm thickness was measured by the four-wave mixing experiment at room temperature. Ar^+ laser operating at 514.5 nm was taken as light source. The sample was reduced and the light was shoot from its y-face. The pump lights I_1 of 1.25 W/cm^2 and I_2 of 1.16 W/cm^2 were used. The expression of conjugate reflectivity R is given as follows:

$$R = \frac{I_3}{I_4} \times 100\% \tag{4}$$

where I_3 and I_4 is the phase conjugate light and the signal light, respectively. The maximum value of the phase conjugate reflectivity of 75% was obtained when the crossing angle between I_1 and I_4 was 20° (Fig. 3).

5. Discussion

In the two-wave coupling experiment, it is found that the weak signal light is amplified, while the strong photo induced scattering with large angle is observed from the sideface of the sample. When the intensity of the pump light is high and its diameter is more than the thickness of the crystal, the scattering lights with large angle can be reflected time after time. These scattering lights coupled with the sectional pump lights are enhanced. They can crawl out of the light-shooting area. The reason why the signal lights with low intensity are amplified is that the small sectional signal lights are coupled with the directly encountered pump lights, the crawling lights in cross area or the nearby scattering lights with the small angle to get their energy, which makes the exponential gain coefficient increase enormously. In addition, no matter how the indent angle varies, the coupling will occur so long as the diameter of the pump light is large enough because the crawling light derived from the scattering light generally exists in the crystal. So the range of the angle that exponential gain coefficient depends on is wide.

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