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Effect of UV light on multiplexing holograms in near-stoichiometric $LiNbO_3$:Ce:Fe

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Abstract

We performed recording–readout and bleaching experiment in lightly reduced near-stoichiometric $LiNbO_3$:Ce:Fe with 532 nm recording beams after UV pre-exposure to get the information of the effect of the UV light on erasure. 100 holograms of nearly equivalent strength were angle multiplexed in the crystal with the erasure time constant after UV pre-exposure. With multiplexing 100 holograms, an $M/^{\#}$ of 1.85 was obtained in a 3.5 mm thickness crystal.

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1. Introduction

Holographic storage in photorefractive crystal [1–3] has high storage density owing to three-dimensional storage and a high readout rate owing to parallel retrieval. However, in the process of multiplexing, the diffraction efficiency is gradually decreased with increasing numbers of stored holograms, because of bleaching of induced absorption by the recording light. In multiplexing many holograms, a reasonable recording schedule to equalize the diffraction efficiency of all holograms is needed. Several methods such as constant diffraction efficiency [4], constant recording time [5] and phase modulation for one of the recording beam [6] have been suggested.

Mok et al. [4] reported the $M/^{\#}$, here the dynamic range is expressed by a useful metric $M/^{\#}$, which determines how many holograms can be

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multiplexed in a single volume, depends on the physical characteristics of the photorefractive crystal, such as impurity doping level, photoconductivity, absorption coefficient, and oxidation state. For the photochromic materials, it is encouraging that the sensitivity can be improved after UV pre-exposure, without a sacrifice of the dark storage. Thus the $M/^{\#}$ could be improved in photochromic materials after UV pre-exposure. Multiplexing holograms by use of UV induced absorption provides an additional feature, i.e., a way of quickly erasing all stored holograms by UV illumination, which returns the crystal to the original colored state [7]. However, in multiplexing after UV pre-exposure, the induced absorption is gradually bleached and thus made the erasure time constant be no longer constant. Myeongkyu Lee et al [8] had observed a strong photochromic effect in near-stoichiometric $LiNbO_3$:Tb:Fe crystal and performed 50 holograms multiplexing of equal diffraction efficiency with determined the erasure-time constant τ_e by trial and error after UV pre-exposure. In the $LiNbO_3$:Tb:Fe crystal, it was believed the Tb ions play a role of stablizing Fe^{3+} valence state, which providing more empty deep traps, and the UV-absorption centers, which are just above the valance band, maybe the O^{2-} ions near lattice defect or another unknown impurities [9]. And an slightly improved $M/^{\#}$ of 1.73 obtained in $LiNbO_3$:Tb:Fe crystal of 3.3 mm with a cw green laser at $\lambda = 532$ nm (the total recording intensity was 315 mW/cm^2) after UV pre-exposure.

Recently, Yang et al. [10] report on Ce and Fe doubly doped $LiNbO_3$ suggested that these materials have a wide spectral response and show high sensitivity. And by changing the stoichiometry and the reduction state of photorefractive lithium niobate one can achieve a two order of magnitude increase in the two-photon response [11]. In this paper, we observed photochromic effect in near-stoichiometric LiNbO3:Ce:Fe crystal. This material exhibited a very long readout time at $\lambda = 532$ nm. By combing these features, we could achieve highly sensitivity and quasi-non-volatile holographic recording in near-stoichiometric LiNbO3:Ce:Fe crystals after UV pre-exposure at $\lambda = 532$ nm. In this paper, the effect of the UV pre-exposure on hologram erasure was also discussed and another approach for determination of the erasure time constant, which is used for calculation of recording time schedule for hologram multiplexing after UV pre-exposure, was suggested. Hologram multiplexing was performed in lightly reduced near-stoichiometric $LiNbO_3$:Ce:Fe with a second harmonic generation of Nd:YAG laser after UV pre-exposure. And with multiplexing 100 holograms, an $M/^{\#}$ of 1.85 was obtained in a 3.5 mm thickness near-stoichiometric LiNbO3:Ce:Fe crystals.

2. Samples preparation

The near-stoichiometric $LiNbO_3$:Ce:Fe crystals were grown by Czochralski technique, containing 0.02wt% of Ce and 0.1wt% of Fe. The ratio of Li to Nb, which was estimated from the Curie temperature measurement, was higher than 49.8/50.2. Experimental studies on $LiNbO_3$:Mn:Fe crystals with different O/R states have showed that there is an optimal O/R state for optimal recording [12]. So we prepared near-stoichiometric $LiNbO_3$:Ce:Fe crystal with different O/R states for holographic recording. Some samples were buried in Nb_2O_5 powders at 1100 °C to be oxidized for 24 h (LN1), oxidized for 12 h (LN2), and some were buried in Li_2CO_3 power to be reduced at 550 °C for 4 h (LN3), reduced for 24 h (LN4). The crystals were cut into wafers with the dimension of $10 \times 10 \times 3.5$mm^3.

Some previous reports have shown that Cu and Ce ions occur in the valence states $Cu^{+/2+}$ and $Ce^{3+/4+}$, respectively [13,14], in $LiNbO_3$:Cu:Ce crystal. In this paper we hypothetically regarded Ce^{3+}/Ce^{4+} traps as the deep centers and this could be confirmed by the following absorption spectra measured at room temperature before and after UV exposure.

Fig. 1 shows the optical absorption spectra measured at room temperature before and after UV exposure. The UV light at 365 nm was obtained by filtering an Hg–Xe lamp which has several characteristic peaks in the range of 300–500 nm with $I = 0.4$ W/cm^2. From Fig. 1(a), small absorption was found above 400 nm in LN1, for almost all traps being empty. The absorption below this wavelength might come from the band-

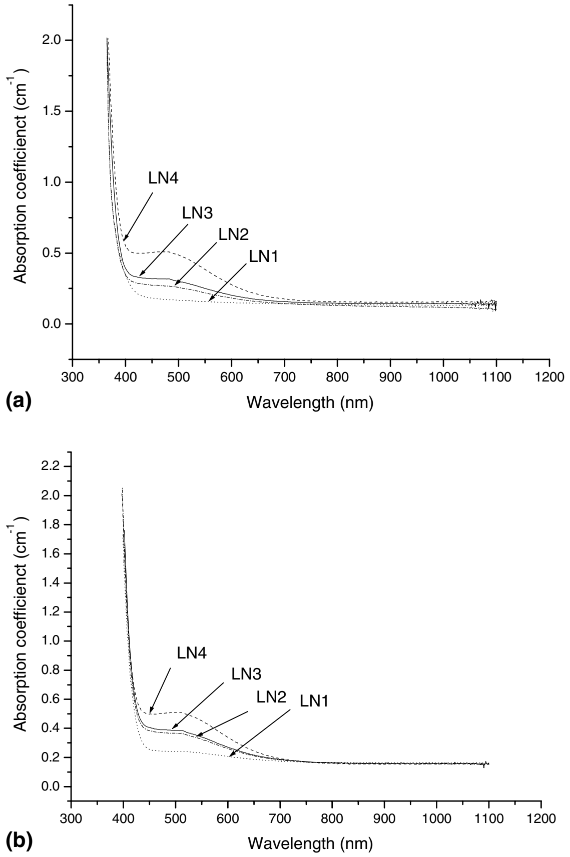

Fig. 1. Absorption spectra of near-stoichiometric $LiNbO_3$:Ce:Fe crystal without (a) and with (b) UV light.

to-band absorption of $LiNbO_3$, electrons transfer from the valence band to Fe traps (hole generation) [15], or the remaining electrons in Ce traps. We found larger absorption in LN2, which means more Ce traps were occupied by electrons. As we reduced the oxidized sample, some Fe traps begin to be occupied by the electrons for all the Ce traps being occupied and this causes an absorption band to appear around 500 nm, which means that the Fe traps began to fill the electrons [16,17], as shown in LN3 and LN4.

From Fig. 1(b), we found in LN2 the UV-induced absorption band was similar to the typical Fe^{2+} band in $LiNbO_3$, which is known to have a peak absorption around 500 nm. It implies that the UV-induced absorption is due to the valence change from the Fe^{3+} to Fe^{2+}, which possible caused by the photo-induced charge transfer from the Ce traps via the conduction band, as shown in Fig. 2. In LN1 a weak peak of absorption around 500 nm was also found which maybe because the remaining electrons in Ce traps transfer to Fe traps via conduction band.

3. Experiment preparation

To investigate more deeply the inherent mechanism and possible optimal conditions for oxidation/reduction states, we performed recording–readout experiments with the four near-stoichiometric $LiNbO_3$:Ce:Fe crystals before and after UV pre-exposure.

In these experiments, a cw green laser (λ = 532 nm) was split into two beams of equal intensity, with the total recording intensity being fixed at 400 mW/cm^2 and these two beams of extraordinary polarization were made to intersect symmetrically inside the crystals, with an external crossing angle ~90°. The recording–readout experiments were performed in these crystals with and without UV pre-exposed (intensity, 0.4 W/cm^2) for 30 min. In the process of recording, one of the recording beams was blocked by a shutter from time to time, so that we could use the other recording beam to monitor the grating buildup process and to measure the diffraction efficiency η. Here η was defined as $I_d/(I_d + I_t)$, where I_d and I_t were the diffracted and transmitted intensities of the readout beam, respectively.

Fig. 3 compares the recording and readout characteristics measured before and after UV pre-exposure. From Fig. 3(a), we found that

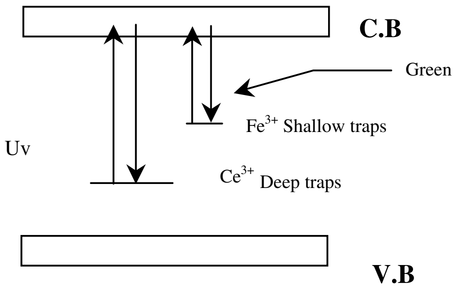

Fig. 2. Possible charge transfer mechanism.

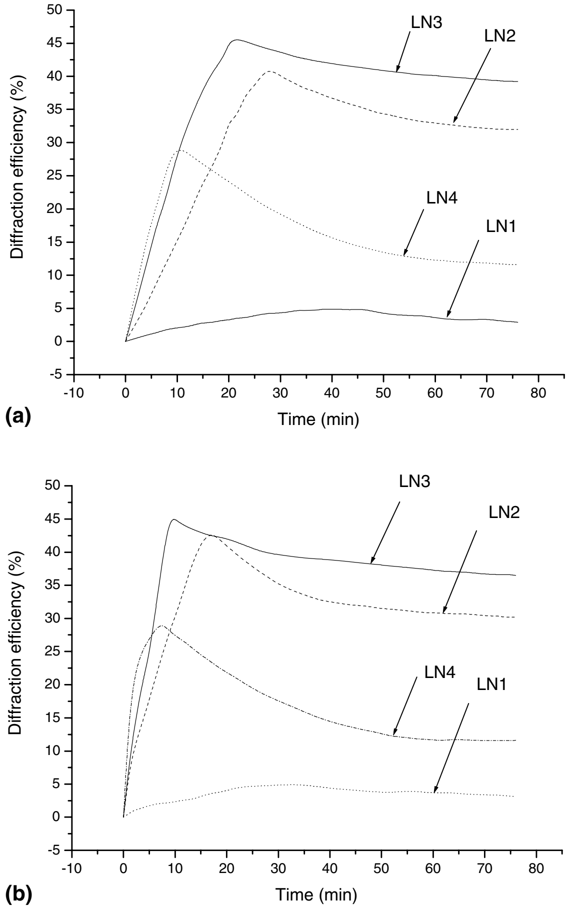

Fig. 3. Recording and readout characteristics measured before (a) and after (b) UV pre-exposure. Total recording intensity, 400 mW/cm^2 at 532 nm; UV pre-exposure time, 30 min at the intensity of 400 mW/cm^2.

LN1 and LN4 were not candidates for quasi-non-volatile holographic recording. This is because in LN1 there are few electrons available in Ce traps while all the Fe traps are completely empty. In LN4, all Ce traps and most of the Fe traps are occupied by the electrons, thus there are no enough empty traps to store a strong space-charge pattern. From Fig. 3(a), we found the crystals of LN2 and LN3 exhibited long readout time and we were able to achieve quasi-non-volatile holographic recording at $\lambda = 532$ nm. However, from Fig. 3(a), we found that LN3 has the larger quasi-non-volatile diffraction efficiency than LN2. It is well known that in $LiNbO_3$ both the sensitivity and the maximum diffraction efficiency depend on the photovoltaic constant [18]. From Fig. 1(a) we know that in LN2 the holograms exist in the Ce traps and in LN3 they exist in Fe traps. So the recording and readout curves for both LN2 and LN3 could be explained with the much smaller photovoltaic constant [19] in Ce traps.

As shown in Fig. 3(b), the recording sensitivity of LN2 and LN3 are greatly enhanced by the UV pre-exposure. This maybe because that when pre-exposed with the UV light, the electrons are excited from the Ce traps and then trapped by the Fe traps via the conduction band, which increase the absorption and photoconductivity in the Fe traps. Thus increase the sensitivity of the recording procedure.

From the above discussion, we found LN3 is the best candidate for performing hologram multiplexing after UV pre-exposure. In order to investigate the erasure mechanism of LN3 after the UV pre-exposure, another experiment was performed in LN3 crystal with UV light for 30 min. The photochromic effect [20,21], which was due to the electrons excited from Ce^{3+} centers to Fe^{3+} centers via the conduction band, was found in LN3 crystal. The measurement of bleaching was performed with one green beam, which was fixed at 200 mW/cm^2 too. Due to the green light excites, the electrons, which were excited from Ce^{3+} centers to Fe^{3+} centers after UV pre-exposure, were thermally unstable and transferred from the Fe^{2+} centers to the Ce^{4+} centers via the conduction band. Fig. 4 shows the bleaching process with one green beam.

From Fig. 3(b), we found in LN3 the erasure curve first decayed non-exponentially and then decayed in a near-exponential way. This maybe due to the bleaching process was much faster than erasure process and thus makes the erasure curve first decayed in a non-exponential way. The non-exponential erasure characteristic made it difficult to calculate the erasure-time constant τ_e, which was defined as the time taken to $1/e^2$ value of the original diffraction efficiency, with the one-center erasure mechanism [22] and two-centers erasure mechanism [23]. Thus, a modified recording schedule should be required. According to the consistency of the function curve, we could calculate the

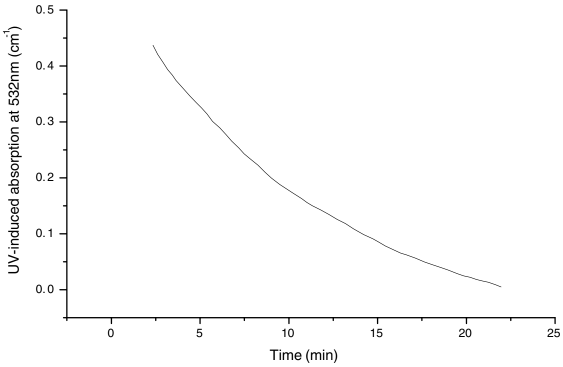

Fig. 4. The bleaching process with one green beam (intensity, 200 mW/cm^2) at $\lambda = 532$ nm.

erasure time constant with the erasure curve of after bleaching by using the least squares method, which is a standard procedure when calculating linear coefficient of function interpolated through measured values. Thus the conventional recording schedule [4] could be expressed as

$$t_{m-1} = t_m \exp\left(\frac{t_m}{\tau_m}\right), \tag{1}$$

where τ_m is the erasure time constant of the completely bleached state.

4. Multiplexed hologram storage in LiNbO$_3$:Ce:Fe crystal

In the multiplexing procedure, a cw green laser ($\lambda = 532$ nm), which was fixed at 400 mW/cm^2, was equally split into two green beams of extraordinary polarization. These two beams were made to intersect symmetrically inside the crystal, with an external crossing angle ~90°. One of the recording beams could be rotated with a mirror mounted on a rotation stage, which followed by a 4-f Fourier-transform system. Thus 100 holograms could be recorded in the crystal with angle-multiplexing method. In the process of readout, we could use the recording beam, which could be rotated with the mirror, to measure the diffraction efficiencies of the recorded holograms.

By the least squares method, the erasure time constants of the crystal before UV pre-exposure and after UV-pre-exposure could be calculated from Fig. 3 as around 2200 and 1600 s, respectively. When we multiplexed 100 plane-wave holograms with an identical exposure time ($t_1 = 70$ s, $t_{100} = 17$ s), which were calculated by Eq. (1) with $\tau_e = 2200$ s before UV pre-exposure, we found the diffraction efficiency of the stored holograms was gradually increased, as shown in Fig. 5.

Mok et al. [4] had mentioned that the parameter $M/^{\#}$ can be measured from the recording and erasure of a single hologram, $M/^{\#} = (A_0/\tau_r)\tau_e$, A_0 is the saturation grating strength, τ_r is the recording time constant, τ_e is the erasure time constant. But for hologram recording in the LiNbO3:Ce:Fe crystal after UV pre-exposure, it is difficult to measure the ratio A_0/τ_r. This maybe because that at the beginning of recording, the recording speed was much faster for the excess electrons in the Fe traps, which were excited from Ce^{3+} centers to Fe^{3+} centers after UV pre-exposure. However, due to the recording light excites, these electrons were unstable and transferred from the Fe^{2+} centers to the Ce^{4+} centers via the conduction band. This makes the electrons concentration in the Fe traps decrease gradually and at the same time makes the recording speed decrease gradually, too. Thus make the recording slope deviate from exponential behavior as the hologram evolves.

However, for a large number of holograms M, the effect of the UV light become little and the final equalized diffraction efficiency is $\eta = [(A_0/\tau_r)\tau_e/$

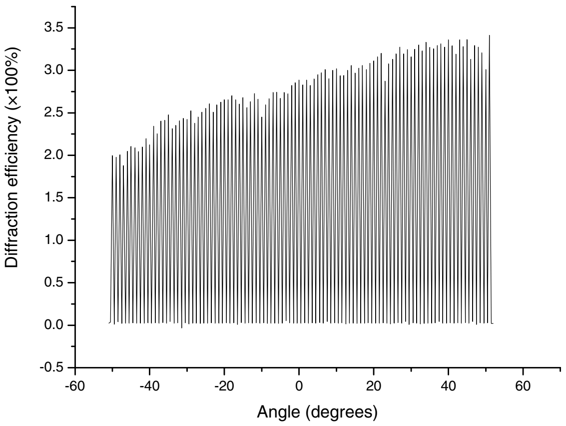

Fig. 5. Angle-multiplexing of 100 plane-wave holograms after UV pre-exposure, recorded with the conventional recording schedule with $\tau_e = 2200$ s ($t_1 = 70$ s, $t_{100} = 17$ s).

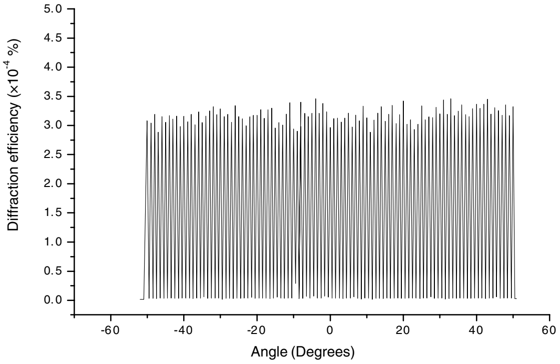

Fig. 6. Angle-multiplexing of 100 plane-wave holograms after UV pre-exposure, recorded with the conventional recording schedule with $\tau_m = 1600$ s ($t_1 = 80$ s, $t_{100} = 13.5$ s).

$M]^2 = (M/^{\#}/M)^2$ [24]. So we can calculate the $M/^{\#}$ with $\eta = (M/^{\#}/M)^2$. Fig. 6 shows the angle-multiplexed 100 plane-wave holograms stored with the erasure-time constant $\tau_m = 1600$ s after UV pre-exposure. A nearly equal efficiency of $\eta = 0.034\%$ was achieved. Substituting these values into $\eta = (M/^{\#}/M)^2$, we get the $M/^{\#} = 1.85$. From Fig. 5, the diffraction efficiency $\eta \sim 0.03\%$ was obtained, which gave an $M/^{\#}$ of 1.73. The increase of the $M/^{\#}$ may be because the sensitivity increases, which mainly from the increase of material absorption and photoconductivity at the recording wavelength after UV pre-exposure, and the erasure time τ_e decreases too.

5. Conclusions

In conclusion, we showed the effect of the UV light on multiplexing holograms and performed holograms multiplexing at $\lambda = 532$ nm in lightly reduced near-stoichiometric $LiNbO_3$:Ce:Fe. We got nearly equal efficiency with the recording schedule with the erasure-time constant after bleaching. And an $M/^{\#}$ of 1.85 was obtained in the 3.5 mm thickness crystal.

Acknowledgements

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