Refractive-index change and sensitivity improvement in holographic recording in $LiNbO_3$:Ce:Cu crystals with green light

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Nonvolatile holographic recording is performed with green light in $LiNbO_3$:Ce:Cu crystals. The refractive-index change and the recording sensitivity are times better than those obtained by recording with red light, and higher optical fixing efficiency is obtained. Correspondingly, theoretical investigations are given.

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Two-center holographic recording has become a topic of interest since it was proposed[1] to solve the problem of volatility. But small refractive-index change and low recording sensitivity are main problems with this method. Therefore, in recent years, comprehensive investigations of optimization for the holographic recording in $LiNbO_3$(LN):Fe:Mn crystals have been performed and significant improvement of the photorefractive performance has been found[2−5]. Other materials such as LN:Ce:Cu[6], LN:Fe:Cu[7], and LN:Ce:Mn[8] were also proposed and investigated. Among them, LN:Ce:Cu crystal is found to be a better material for nonvolatile holographic recording because it can provide high, persistent refractive-index change with a high recording sensitivity and weak light-induced scattering. Furthermore, holographic recording in LN:Ce:Cu can also be optimized by using theoretical analyses of the dopant densities and the oxidation-reduction processing for LN:Fe:Mn crystals[2−4], which were successfully used before. In this letter, we demonstrate the improvement of the recording performance in LN:Ce:Cu crystal by using a short wavelength recording light (green light). The refractive-index change and the recording sensitivity are increased, and higher optical fixing efficiency is also obtained.

Experiments of holographic recording with green light are performed comparing with the typical red-light recording in a congruent 2-mm-thick LN crystal doped with 0.085 wt.-% Ce_2O_3 and 0.011 wt.-% CuO, grown in air by the Czochralski method and annealed at 900 °C in air for 6 h. The experimental setup is illustrated schematically in Fig. 1. Ultraviolet (UV) light for sensitization (365 nm, 200 W/m^2) is produced by focusing of a filtered 75-W Hg lamp. Two ordinarily polarized recording beams (632.8 and 514.5 nm, respectively) with equal intensity 2500 W/m^2 are incident upon the crystal symmetrically at incidence angles of 15°. The grating wave vector is aligned parallel to the c axis of the crystal. In the experiments, the holograms are recorded by the two interfering beams, together with the UV light, until quasi saturation is reached. Then, the holograms are read with only one recording beam. During recording, one of the beams is blocked from time to time, and correspondingly, another beam is diffracted from the recording grating, allowing us to evaluate the diffraction efficiency which is defined as the ratio of the intensities of the diffracted and the total of diffracted and transmitted light. During reading, only the beam used for diffraction observation is still used for both readout and measurement.

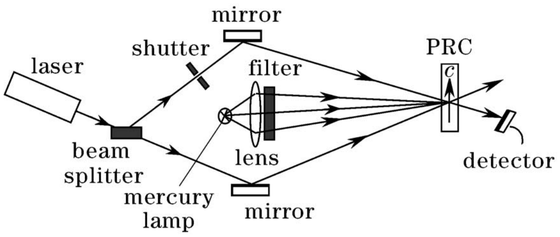

Fig. 1. Experimental setup for holographic recording experiments. PRC: photorefractive crystal.

Figures 2(a) and (b) show the experimental results

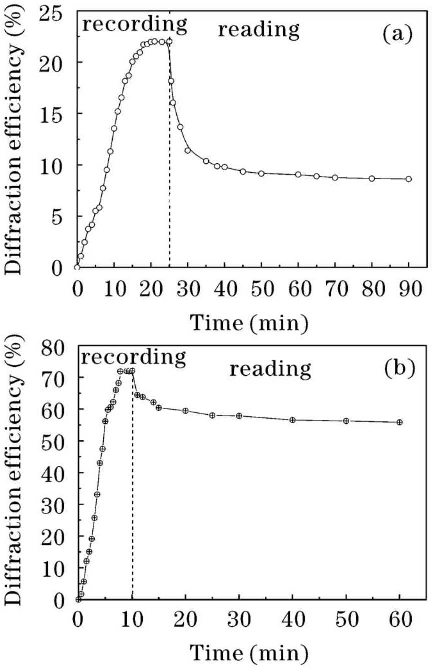

Fig. 2. Diffraction efficiency versus time for recording with (a) two red and (b) two green beams respectively, with simultaneous presence of UV light in the oxidized Ce:Cu:$LiNbO_3$ doped with 0.085 wt.-% Ce_2O_3 and 0.011 wt.-% CuO.

of holographic recording in LN:Ce:Cu crystal with red and green light, respectively. We can see that nonvolatile holographic recording can be successfully performed with green light besides that typically using red beams. And the saturation diffraction efficiency of the hologram recorded by green light is about 72%, which is more than three times of that obtained in red-light recording, witnessing larger refractive-index change in the crystal. For measuring of recording speed, the recording sensitivity S is defined as

$$S = \frac{1}{I_{\mathrm{rec}} d} \frac{\partial \sqrt{\eta}}{\partial t}\bigg|_{t=0}, \qquad (1)$$

where I_{rec}, d, t, and η are total recording intensity, depth of the LN:Ce:Cu crystal, time, and the diffraction efficiency, respectively. As measured from Figs. 2(a) and (b), the recording sensitivity is increased up to be more than two times by recording with green light. Moreover, in the one-hour nonvolatile holographic recording with green light, the optical fixing efficiency (which is defined as the ratio between the final and the saturation diffraction efficiency) is enhanced up to be 78%, which is almost two times of that in the red-light recording system.

To explain our results theoretically, in this letter, we utilized the simulation model with the joint solution of two-center material equations and coupling-wave equations[4]. Referring to LN:Fe:Mn crystal[2−4], we estimate the electron recombination coefficients of Ce and Cu to be 1.65×10^{-14} and 2.4×10^{-5} m^3/s respectively. The photoexcitation coefficients of Cu for green and UV light were 1×10^{-8} and 3.6×10^{-5} m^2/J, respectively; and the photoexcitation coefficient of Ce for the red, green, and UV light were 3.3×10^{-6}, 1×10^{-5}, and 3.8×10^{-5} m^2/J, respectively. The bulk photovoltaic coefficient of Ce for the red, green, and UV light were 7×10^{-34}, 3.8×10^{-33}, and 1.4×10^{-32} m^3/V, respectively, and the bulk photovoltaic coefficients of Cu for the green and UV light were 5×10^{-36} and 1.1×10^{-32} m^3/V, respectively. The refractive-index of the LN:Ce:Cu at 632.8 nm is 2.286 and that at 514.5 nm is 2.33, and the absorption coefficient at 632.8 nm is 178 m^{-1} and that at 514.5 nm is 1000 m^{-1}.

As shown above, the photoexcitation coefficient of Ce traps at 514.5 nm is times larger than that at 632.8 nm and so does the bulk photovoltaic coefficient. Therefore, the recording sensitivity of holographic recording with green light is higher than that using red light. And in the normal nonvolatile holographic recording in LN:Ce:Cu crystals, red light can only excite electrons in the shallow centers, simultaneously with UV light moving electrons from the deep centers to the shallow ones to recorded gratings in both centers. The phase difference between deep and shallow gratings is nearly 180°, and the strength of the total grating (the superimposition of two gratings in both centers) in the crystal is low, as plotted in Figs. 3(a) and (b), respectively. However, in the green-light holographic recording, green light could slightly excite electrons from the deep centers besides that in the shallow ones, the phase difference between two gratings is reduced to be 160°, which is also plotted in Fig. 3(a). And the disadvantage of weakening the total grating caused by large phase difference between two gratings is alleviated. As also shown in Fig. 3(b), the refractive-index change is increased to be five times larger than that recorded with red light. In the reading phase, since green light excites electrons much faster than red light does and electrons move only a few nanometers on the average, most electrons are trapped in Cu centers within a much shorter distance shortly after they are excited from Ce centers, and the strength of the recorded total grating (the superimposition of the deep and shallow gratings) changes less. So, less partial of the total grating is erased by green light than that by red light and the optical fixing efficiency is correspondingly increased. However, with long time reading with green light, partial of the grating will be erased for green light slightly exciting electrons from the deep centers. But it is acceptable in practical applications.

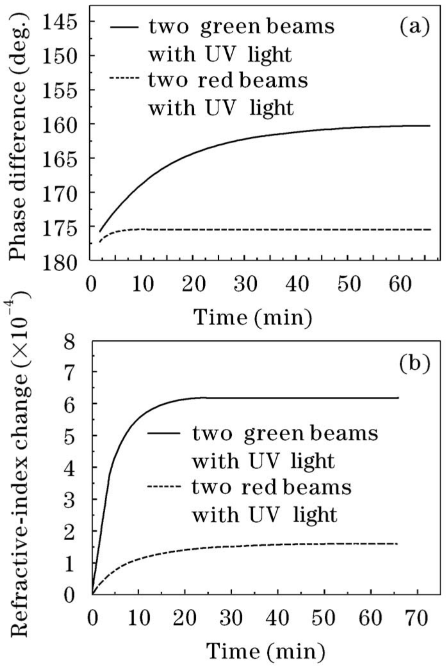

Fig. 3. (a) Phase difference between deep and shallow gratings, (b) refractive-index change versus time while recording with two red and green beams, respectively, and simultaneously with UV light.

In conclusion, nonvolatile photorefractive holograms have been recorded in LN:Ce:Cu crystals by recording with green light. Larger refractive-index change, higher recording sensitivity, and higher optical fixing efficiency can be obtained than that in normal nonvolatile holographic recording with red light, and the partial loss of the intensity of the grating in long time reading is acceptable in practical applications. Therefore, green light is superior to red light in nonvolatile holographic recording in LN:Ce:Cu crystals. Further optimization can be performed with the dopant densities and the oxidation-reduction processing of the LN:Ce:Cu crystals.

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