Effect and optimization of the diminution of the sensitizing intensity in two-center recording

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Received November 22, 2011; accepted December 12, 2011; posted online March 15, 2012

The effect of the diminution of the sensitizing intensity from initially high value on two-center recording is experimentally and theoretically investigated. A linear diminution of the sensitizing intensity is designed, and three different diminution rates are employed for the experimental investigation. The results show that the dynamic range can be improved by reducing the diminution rate of the sensitizing intensity. Compared with that of traditional two-center recording, the recording sensitivity obtained in the experiment is more improved when the maximum dynamic range is achieved. The effect of the diminution of the sensitizing intensity on grating uniformity and the effect of the initial sensitizing intensity on the recording sensitivity are theoretically investigated.

OCIS codes: 090.2900, 160.3730, 160.5320, 210.2860.

doi: 10.3788/COL201210.060901.

The two-center holographic recording method reported by Buse *et al.* has attracted attention because of its ability to record persistent holograms[1]. The advantage of this method from other grating–fixing methods is its all-optical, real-time process[2,3]. The two-center recording method uses two different dopants to produce shallower and deeper traps in photorefractive crystals. The key point of this technique is that, during the recording of an interference intensity pattern by two beams with longer wavelengths, the crystal is simultaneously sensitized by a beam with a shorter wavelength so that an inner-field grating can finally be built in the deep center, which is insensitive to the recording beam for further readout.

Much work has been done to improve the recording sensitivity and dynamic range in two-center recording. The effect of the dopant element and its concentration, annealing, and recording and sensitizing intensities and wavelengths on these parameters has also been investigated[4,5] based on stable recording and sensitizing intensity values. Researchers have observed that, for doubly doped $LiNbO_3$ crystals, the recording sensitivity monotonously increases as the intensity ratio (I_R/I_S) decreases. Meanwhile, the dynamic range increases to a maximum value at a certain intensity ratio and then monotonously decreases. The maximum dynamic range usually sacrifices the recording sensitivity[6−8]. However, a mass-storage system is necessary to improve the recording sensitivity.

In this letter, we utilize the diminution of the sensitizing intensity to optimize two-center recording. The recording sensitivity is well known to be directly proportional to the average electron concentration (N_{20}^-) in the shallow center. Similarly, the dynamic range is directly proportional to N_{20}^-/n_0, where n_0 is the average electron concentration in the conduction band[7]. The enhancement of N_{20}^- and n_0 depends on the improvement of the sensitizing intensity. If the sensitizing intensity is reduced from a high initial value to optimize two-center recording, a high recording sensitivity can be achieved at the start of the recording process. Then, the diminished sensitizing intensity results in the improvement of N_{20}^-/n_0, and a high dynamic range is obtained. However, if the sensitizing intensity becomes too weak, N_{20}^- diminishes and the recording light will partially delete the grating.

Hence, a novel two-center recording method is performed in this letter. In this method, the sensitizing intensity decreases with the recording time while keeping the recording intensity stable. The main achievement of this method is the improved recording sensitivity at the maximum dynamic range.

The congruent $LiNbO_3$ single-domain crystals used in the experiments were grown in air using the Czochralski method[9] and doped with 0.15 wt.-% Fe_2O_3 and 0.12 wt.-% RuO_2. The specimen was oxidized and then polished to a size of 10×12×1.5 (mm), with the *c*-axis parallel to the longest side. During the recording, two red beams of ordinary polarization were produced by a He–Ne laser. The beams each had a wavelength of 632.8 nm, beam diameter of 2 mm, and intensity of 250 mW/cm^2. The two recording beams were symmetrically incident on the crystal with an external intersection angle of 20°, and the holographic grating vector was positioned parallel to the crystal *c*-axis. A sensitizing light from a light-emitting diode (LED), which had a middle wavelength of 465 nm and intensity of approximately 66.8 mW/cm^2, was simultaneously radiated on the sample. In the experiments, the sensitizing intensity was altered by slowly adjusting the working voltage of the LED. The relation between the sensitizing intensity and recording time can be written as

$$I_S(t) = I_S(0) - at, \quad (1)$$

where $I_S(0)$ is the initial value and a is the diminution rate of the sensitizing intensity, whose unit is $mW/(cm^2{\cdot}s)$. If a is zero, the sensitizing intensity will remain stable.

In measuring the diffraction efficiency, one of the recording beams was occasionally blocked, and the diffracted power was detected by the photodetector. The diffraction efficiency η is defined as the ratio between the diffracted power and the sum of the transmitted and diffracted powers.

The recording sensitivity S and the dynamic range $M/\#$ are defined as[10]

$$S = \frac{\beta}{I_{\mathrm{r}} d} \frac{\partial \sqrt{\eta}}{\partial t}, \tag{2}$$

$$M/\# = \sqrt{\eta_{\mathrm{f}}} \frac{\tau_{\mathrm{r}}}{\tau_{\mathrm{e}}} \sim \sqrt{\eta_{\mathrm{f}}}, \tag{3}$$

where I_{r} is the total recording intensity, d is the sample thickness, η_{f} is the fixed diffraction efficiency, and τ_{r} and τ_{e} are the time constants of the recording and erasing processes, respectively. Furthermore, β is the ratio of the final hologram strength after the sufficient readout to the hologram strength immediately after the recording process. The sensitivity and dynamic range are calculated from the temporal profiles of the diffraction efficiency.

The proposed method primarily aims to confirm the of sensitizing intensity where the dynamic range increases as the sensitizing intensity decreases. First, the dependence of the recording sensitivity and dynamic range on the intensity ratio in the $LiNbO_3$:Fe:Ru crystal was experimentally investigated based on the traditional two-center recording method. The experimental results, which are shown in Fig. 1, are consistent with those reported in Ref. [10]. When the intensity ratio decreases within the range from 7.5 and 31, the recording sensitivity increases, but the dynamic range decreases. In these experiments, the maximum dynamic range and recording sensitivity are 0.51 and 0.041 cm/J, respectively. Pre-sensitizing was not performed before each experiment. Otherwise, a higher recording sensitivity would be achieved.

As shown in Fig. 1, an initial sensitizing intensity of 66.8 mW/cm^2 is required to achieve the maximum recording sensitivity. Then, the sensitizing intensity linearly decreases at a certain rate. Three typical diminution rates, namely, 0.07, 0.10, and 0.15 mW/(cm^2·s), were employed in the experiments. The temporal evolutions of diffraction efficiency with the decrease in sensitizing intensity were also obtained. The results are shown in Fig. 2. The dynamic ranges for the three different rates were 0.38, 0.49, and 0.58, respectively. As shown, the dynamic range increases as the diminution rate of the sensitizing intensity decreases. The effect of the diminution rate on the dynamic range can be explained

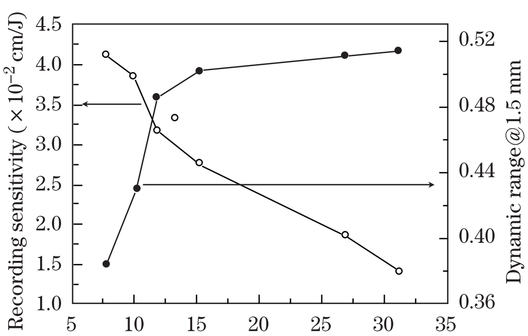

Fig. 1. Sensitivity and the dynamic range as a function of the intensity ratio of the recording to sensitizing intensity. The recording intensity is fixed at approximately 500 mW/cm^2.

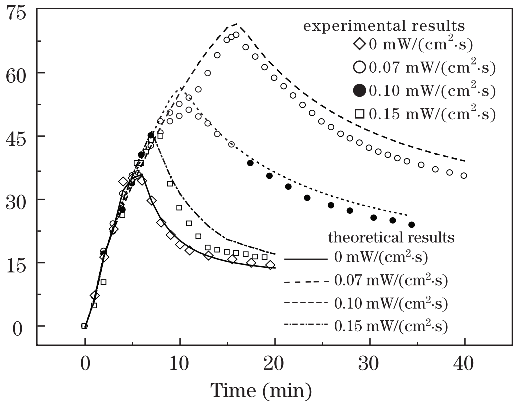

Fig. 2. Time evolution of diffraction efficiency in the recording and readout process with different diminution rates. The total recording intensity is 500 mW/cm^2, and the initial sensitizing intensity is 66.8 mW/cm^2. In fitting the theoretical curves to the experimental results, we assume that the OR degree is 0.78.

as follows. A low intensity ratio requires a long recording time. However, a high diminution rate causes the sensitizing intensity to weaken rapidly. Hence, the strong grating does not have enough time to build up.

For the diminution rates of 0.10 and 0.07 mW/(cm^2·s), the recording sensitivities are similar with those obtained using the traditional two-center recording method at $I_{\mathrm{R}}/I_{\mathrm{S}}$=7.5. Among the four recording schemes (Fig. 2), the lowest recording sensitivity (0.028 cm/J) was obtained with the rate of 0.15 mW/(cm^2·s). The decrease in recording sensitivity is highly attributed to the diminution of the sensitizing intensity at the beginning of the recording process.

In traditional two-center recording, the recording time needed for the grating to reach the saturated state is 25 min, when the maximum dynamic range of 0.51 is achieved. For the rate of 0.07 mW/(cm^2·s), the recording time is 15 min. Compared with traditional recording, the proposed method achieves an improved recording sensitivity when the maximum dynamic range is obtained.

The sensitizing intensity was not zero when the diffraction efficiency became saturated. For example, the end value was approximately 6.3 mW/cm^2 for the diminution rate of 0.07 mW/(cm^2·s). As aforementioned, the recording beams erase the grating when the sensitizing intensity becomes too weak. Thus, the saturated phenomenon can be observed before the sensitizing intensity reaches zero. This observation meets the theoretical expectation mentioned earlier in the letter.

To verify the effect of the diminution of the sensitizing intensity theoretically, the current study employs the simulation model with the joint solution of the material and wave-coupling equations[6]. In the simulations, the Fe and Ru trap densities were $N_{\mathrm{Fe}}=5.2\times10^{25}$ m^{-3} and $N_{\mathrm{Ru}}=2.6\times10^{25}$ m^{-3}, respectively. The critical and real oxidization-reduction (OR) degrees were $Xc=N_{\mathrm{Fe}}/(N_{\mathrm{Fe}}+N_{\mathrm{Ru}})=0.67$ and $x=1-N_{\mathrm{A}}/(N_{\mathrm{Fe}}+N_{\mathrm{Ru}})=0.78$, respectively. The total intensity of the red beams (632.8 nm) was 500 mW/cm^2 (each red beam had 250 mW/cm^2). The intensity of the sensitizing light (465 nm), with an initial value of 66.8 mW/cm^2, was assumed to decrease linearly, following Eq. (1).

In these simulations, the parameters of Ru are unknown. Hence, their values are estimated from previous Refs. [5,6,11,12]. The electron recombination coefficients of Fe and Ru are 1.65×10^{-14} and 24×10^{-14} m^3/s, respectively. The photo-excitation coefficients of Fe and Ru for 465-nm light are 2×10^{-5} and 1.7×10^{-5} m^2/J, respectively, and the photo-excitation coefficients of Fe and Ru for the red beam are 3.3×10^{-6} and 1×10^{-8} m^2/J, respectively. Moreover, the bulk photovoltaic coefficients of Fe and Ru for 465-nm light are -8×10^{-33} and -5×10^{-33} m^3/V, respectively, whereas those for 633-nm light are -7×10^{-34} and -4.45×10^{-36} m^3/V, respectively. The time evolutions of diffraction efficiency with different diminution rates are also plotted in Fig. 2, and the theoretical results can verify the experimental results. The difference between the theoretical and experimental curves may be caused by the deviation of the practical from the ideal parameters in the calculations, the optical quality of the specimen, the stability of the laser intensity and its mode, and the shock resistance of experimental equipment[13].

In traditional two-center recording, the nonuniformity of the grating is primarily caused by the variation of I_R/I_S along the transmission direction of the recording light, which is due to the absorption to the sensitizing light[14]. When the sensitizing intensity decreases with time, I_R/I_S correspondingly varies at each slice of the crystal. The effect of the diminution of the sensitizing intensity on grating uniformity was investigated, and the results are shown in Fig. 3. In traditional two-center recording, the maximum space charge field (SCF) can be found at z=0.4 mm when the grating reaches a saturated state. However, the SCF decreases with depth z, and the maximum SCF can be achieved at z=0 if the sensitizing intensity decreases with time. When the grating is read enough, the maximum values appear at z=0 for all four schemes.

Furthermore, in traditional two-center recording, the recording sensitivity increases as the sensitizing intensity increases. Similarly, in the proposed scheme, the recording sensitivity is improved by enhancing the initial sensitizing intensity. The effect of the initial sensitizing intensity on recording sensitivity is further theoretically investigated, and the results are shown in Fig. 4. In the simulations, the diminution rate is adjusted so that the sensitizing intensity decreased to zero after 16 min. The results show that the recording sensitivity increases when the initial sensitizing intensities increase from 5.0 to 140.0 mW/cm^2. In addition, a higher initial sensitizing intensity induces a higher dynamic range.

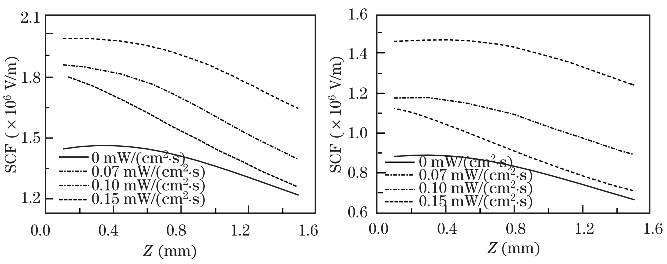

Fig. 3. Variation of (a) saturated SCF and (b) fixed SCF with thickness of crystal.

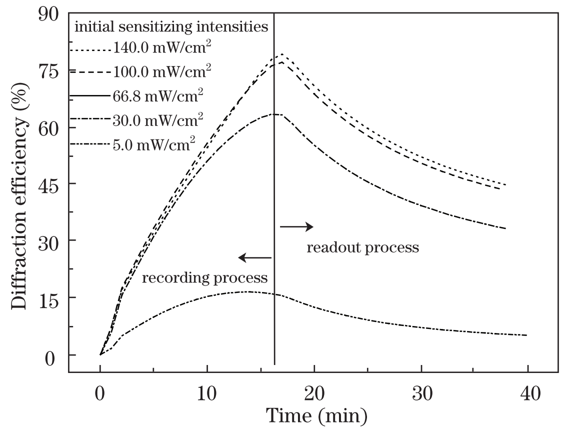

Fig. 4. Time evolution of diffraction efficiency in the recording and readout process with different initial values of sensitizing intensity. In simulations, we decrease the sensitizing intensity to zero at 16 min by adjusting the diminution rate.

In conclusion, the diminution of the sensitizing intensity from the initially high value is utilized to optimize the two-center recording process in $LiNbO_3$:Fe:Ru crystals. The results show that the recording sensitivity can be improved by reducing the sensitizing intensity for a mass-storage system.

This work was supported by the National Natural Science Foundation of China (No. 60908029) and the Natural Science Foundation of Shanghai (Nos. 09ZR1435300 and 09ZR1431300).

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