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# Effect of the parameters on diffraction efficiency after thermal fixing for transmission geometry hologram storage in $LiNbO_3$:Fe

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**Abstact**

For transmission geometry hologram storage in $LiNbO_3$:Fe, we have shown that the diffraction efficiency increases with doping level and thickness of storage material monotonically. When the angle between reference and object beam is large enough for getting a relatively small Bragg angle that is needed for angle multiplexing, smaller angle does good to diffraction efficiency after thermal fixing. And for absorption coefficient there is an appropriate value corresponding to optimal diffraction efficiency after thermal fixing and we develop a theoretical model that predicts achievable diffraction efficiency after thermal fixing as a function of crystal thickness, doping level, angle between reference and object beam and absorption coefficient. We compare this model with experimental results and get a good agreement.


*Keywords:* Absorption coefficient; Doping level; $LiNbO_3$:Fe

Volume holographic memory using photorefractive materials has become an important research focus that is pursued vigorously by many investigators for the potential in high-capacity storage and fast parallel readout. But for the practical realization, a reliable method for nondestructive readout of digital data must be demonstrated. A promising technique is thermal fixing [1] by compensation of mobile ion in photorefractive media and many investigations [2,3] have been done. In this paper, for multiplexed hologram storage in $LiNbO_3$:Fe, we have studied the influence of all parameters on the final diffraction efficiency after thermal fixing for transmission geometry.

As shown in Fig. 1, $\theta$ is the angle between reference beam or object beam and $z$-axis in the recording material and $c$-axis is perpendicular to $z$ axis. During hologram recording with extraordinary polarized light, the initial evolution of the local space-charge field can be obtained from the Kuktarev equation [4]. According to the analysis of Ref.[5], the first-order space-charge field could be expressed as

$$E_1(z) = m(z)E_q\{1-\exp[-t/\tau_I(z)]\exp[-jw_I(z)t]\} \times \left(E_{ph}^2+E_D^2\right)^{1/2}/\left(\left[(N_A/N_D)E_{ph}\right]^2+\left(E_D+E_q\right)^2\right)^{1/2},$$

where

$$\tau_I(z) = \tau_{di}(z)\frac{1+(E_D/E_\mu)}{1+(E_D/E_q)},$$

$$w_I(z) = \frac{1}{\tau_{di}(z)}\frac{N_A E_{ph}}{N_D E_q}\frac{1}{1+(E_D/E_\mu)},$$

and the parameters $N_D$ is total Fe doping, $N_A$ is initial $Fe^{3+}$ concentration, dielectric relaxation time $\tau_{di}(z) = (\varepsilon/q\mu)\left[\gamma_R N_A/sI_0(z)(N_D-N_A)\right]$, diffusion field $E_D = k_R TK/q$, saturation space-charge field $E_q = qN_A(N_D-N_A)/\varepsilon K N_D$, drift field $E_\mu = \gamma_R N_A/\mu K$ and photovoltaic field $E_{ph} = p\gamma_R N_A/q\mu s$. $S_z$ and $W_z$ are the signal and reference amplitudes at $z$, respectively, $I_0(z)$ is the local intensity, and $\alpha$ is the intensity absorption coefficient. The intensity ratio of incident beams $m(z) = 2S_Z W_Z/I_0(z)$ could be simplified to be a constant $m = 2S_0 W_0 \cos 2\theta/(S_0^2+W_0^2)$. The change of oxidation

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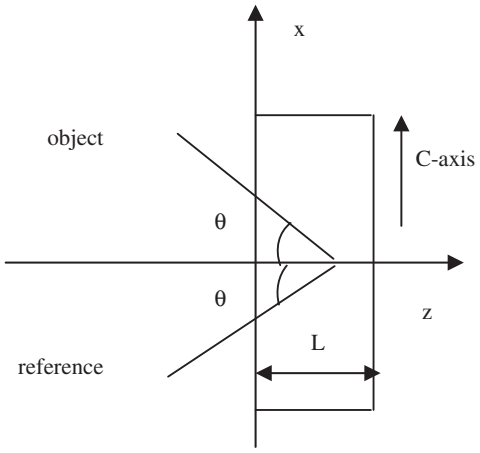


Fig. 1. Transmission geometry.

state affects terms $N_A$ and the absorption coefficient $\alpha$ (proportional to $N_D$–$N_A$ ). The total Fe doping $N_D$ remains unchanged during annealing.

For exposure times much shorter than $\tau_I$ , including the absorption of the readout beam and scatting beam, the effective writing can be written as

$$\frac{d\sqrt{\eta(t)}}{dt}|_{t=0} = \frac{S_0 W_0 k_0 n_e^3 \left(r_{33} - \left(n_o^4/n_e^4\right) r_{13} \tan^2(\theta)\right) \cos(2\theta)}{2\alpha\tau_I(z) I_0(z)} \times \left[\exp\left(-\frac{\alpha L}{2\cos\theta}\right) - \exp\left(-\frac{3\alpha L}{2\cos\theta}\right)\right] \times \frac{E_q\sqrt{E_{ph}^2 + E_D^2}}{E_D + E_q}.$$

During the erasure, the evolution of diffraction efficiency is

$$\eta'(t) \propto \int_0^L \exp\left(-\frac{\alpha z}{\cos\theta}\right) \exp\left(-\frac{2t}{\tau_I(z)}\right) dz.$$

Thus, the $M/\#$ in transmission geometry may be denoted as

$$M/\# = \left(\frac{d\sqrt{\eta(t)}}{dt}|_{t=0}\right) \times \tau_e = 2\left(\frac{d\sqrt{\eta(t)}}{dt}|_{t=0}\right) \frac{\eta'|_{t=0}}{-d\eta'/dt|_{t=0}}, \quad (1)$$

where $\tau_e$ is the erasure time constants.

Once the recording has been done, thermal fixing is needed for nondestructive readout of digital data. According to Ref.[6] and assuming that the electronic gratings are fully compensated by the proton grating after heating, which may be realized when the density of ions is sufficiently large, the thermal fixing efficiency under the short-circuit condition is denoted as

$$\eta_{fixing} = \frac{\left(\left(N_D - N_A/N_D\right)E_{ph}\right)^2 + E_D^2}{\left(\left(N_D - N_A/N_D\right)E_{ph}\right)^2 + \left(E_D + E_q\right)^2}. \quad (2)$$

For a large number of holograms, $M$, the final equalized diffraction efficiency [7] is $\eta^* = \left(M/\#\right)^2/M^2$ and so the diffraction efficiency after thermal fixing can be expressed by

$$\eta = \eta^* \eta_{fixing}. \quad (3)$$

According to the definition of these parameters, $E_D = 0.163/\Lambda$ and $E_q = 9.6 \times 10^{-9}\Lambda\left(N_A/N_D\right)\left(N_D - N_A\right)$ with $T = 300\,\text{k}$ and $\Lambda = \lambda_0 \times 10^{-7}/(2n_e \sin\theta)$ can be obtained. According to Refs. [8,9], the value of $E_{ph}$ is a range from $10^{-15}N_A$ to $3 \times 10^{-14}N_A$ and here we take $10^{-14}N_A$. For absorption coefficient $\alpha$, research shows it is proportional to the density of filled traps [10]. By the study of Phillips and Staebler [11], to 0.001–0.1 mol% Fe doped $LiNiO_3$, the amount of Fe ions per $cm^3$ is $1.89x \times 10^{20}$ for xmol% doping and especially $N_D - N_A = 1.51 \times 10^{17}\alpha$ for extraordinary absorption at 450 nm. For the study latter in this paper, $1.51 \times 10^{17}$ is adopted for 488 nm approximately and other parameters used include $r_{33} = 30.8 \times 10^{-12}\text{m/V}$, $r_{13} = 8.6 \times 10^{-12}\text{m/V}$, $n_e = 2.2446$, $n_o = 2.20$ and $S_0^2 + W_0^2 = 110\,\text{mV/cm}^2$. Actually the highest practical doping level is about 0.06 mol% limited by dark decay [12,13] and the angle between reference and object beam is limited by not only geometrical light-path and Bragg angle (when $\theta$ is too small, the Bragg angle is rather large and it is not good for hologram multiplexing) which decides the minimum value in practical experiment but also the refractivity of $LiNbO_3$ which decides the maximum value. Practically we take $5^\circ \leqslant \theta \leqslant 27^\circ$ in this paper.

With reference to Eq. (3), we plot in Fig. 2 the diffraction efficiency $\eta$, which refers to the diffraction after thermal fixing throughout this paper, as the function of thickness of recording material and the absorption coefficient with different doping level and $\theta$. One observation from these figures is that the diffraction efficiency is monotone increasing with the increase of thickness when absorption coefficient is within the range that relatively better diffraction efficiency could be gotten. This is because for the absorption coefficient corresponding to better diffraction efficiency and the increasing of dynamic range is much more than the loss caused by the absorption for thicker material. But for transmission geometry, reference beam is needed to cover object beam fully during the hologram recording processing and thus for high storage density, the thickness of recording material could not be too much. As to $m$, Eq. (3) tells us the maximum value is the perfect value. So $L = 0.5\,\text{cm}$ and $m = 1$ is adopted for the rest of our study.

To the influence of doping level on diffraction efficiency, our study gets the same result as that of thickness of $LiNbO^3$:Fe which is shown in Fig. 3. Theoretically, diffusion field $E_D$ is independent of doping level and saturation space-charge field $E_q$ has little relationship with it except when absorption coefficient is larger($\geqslant 0.2\,\text{cm}^{-1}$) and doping level is much low $\leqslant 0.005$ mol%. Thus only photovoltaic field $E_{ph}$ is normally proportional to doping level. So dynamic range is larger with high doping level. But thermal fixing efficiency has nothing with it because the influence of $E_{ph}$ to fixing efficiency is rather little.

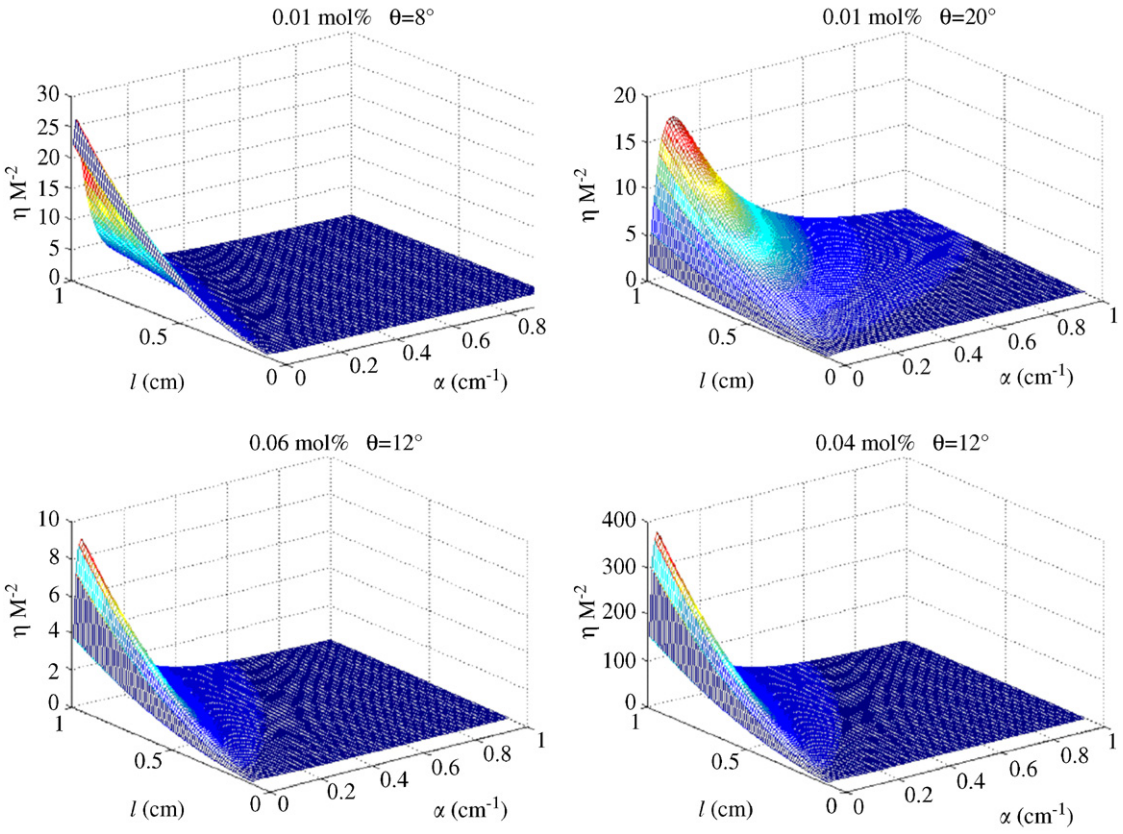


Fig. 2. Diffraction efficiency as a function of thickness of $LiNbO^3$:Fe under different conditions.

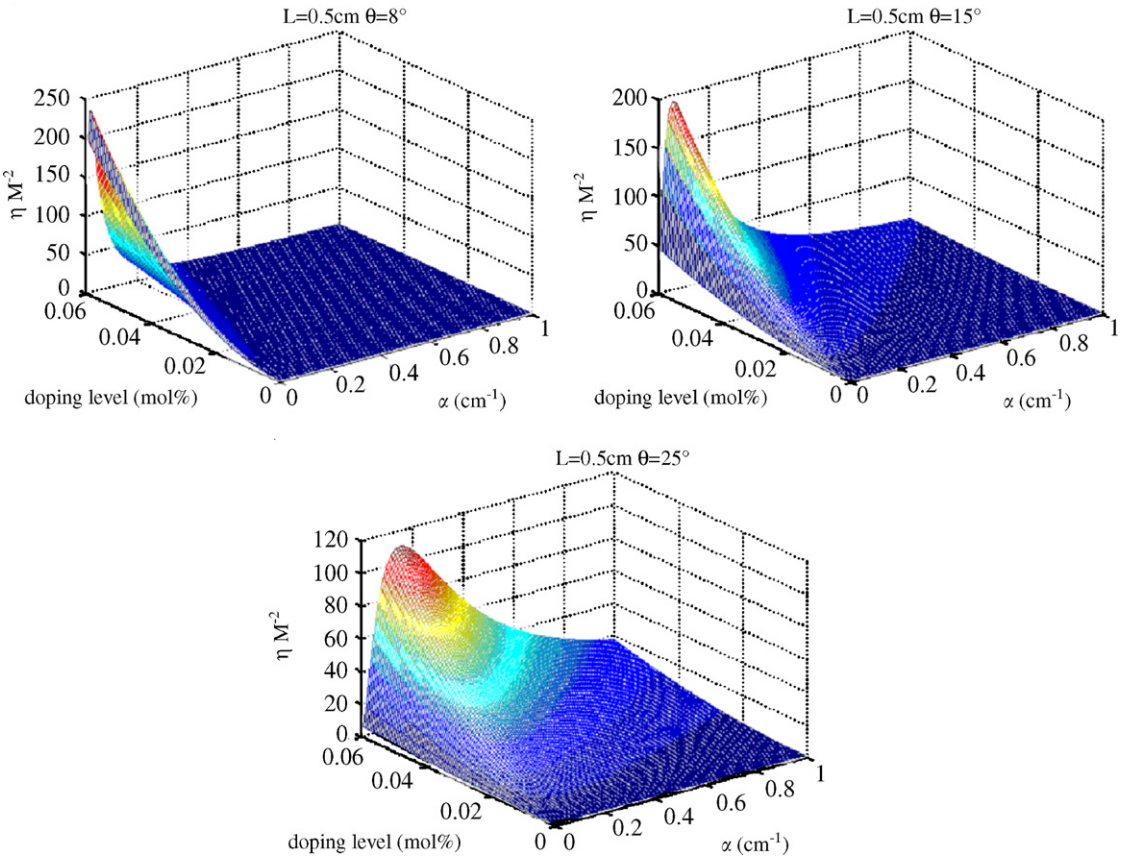


Fig. 3. Diffraction efficiency as a function of doping level with different $\theta$ and absorption coefficients.

Experimentally for the preparation of material, high doping level does harm the optical quality of $LiNbO^3$:Fe. Generally moderate doping level, 0.01∼0.03 mol% is favorable for hologram recording.

Without consideration of the minimum limitation of $\theta$, it could be found the diffraction efficiency has a maximum value within 0° and 27° as shown in Fig. 4. This is because with $\theta$, the thermal fixing efficiency rises up but the dynamic range falls down. Thus these two factor lead to the formation of the maximum value. When the absorption coefficient is less than 0.01 $cm^{-1}$, the maximum values of diffraction efficiency change very little. In fact, it is difficult to realize very small absorption coefficient exactly in the preparation of $LiNbO_3$:Fe. So our calculation takes $\alpha \geqslant 0.01\,cm^{-1}$.

Considering the limitation mentioned above, the relationship between diffraction efficiency and absorption coefficient and $\theta$ is figured as Fig. 5 within the range of $\alpha \geqslant 0.01\,cm^{-1}$ and $5^\circ \leqslant \theta \leqslant 27^\circ$. It shows that with the diminution of $\theta$, the value of maximum diffraction efficiency increases as the corresponding absorption efficient decreases. The role of absorption coefficient is also divided into two aspects. With respect thermal fixing, fixing efficiency increases with the decrease in thermal fixing, but to dynamic range, its influence is a little sophisticated: a peak value of dynamic range is formed at a rather large absorption coefficient. This can be explained through Eq. (2): for dynamic range the two dominant terms are $E_q$, which increases with absorption, and $\exp(-\alpha L/2\cos\theta)$, which decreases with absorption. Intuitively, dynamic range is small for low absorption because the number of photogenerated electrons is small. But for higher absorption coefficient, the losses due to bulk absorption rapidly dominate, reducing the dynamic range. So the competing effect of bulk absorption and photorefractive dynamics leads to an absorption coefficient corresponding to the maximum dynamic range. It is the same for the diffraction efficiency after thermal fixing according to Eq. (3) that the value of absorption coefficient could be optimized for the best diffraction efficiency as the result of the completing between fixing factor and dynamic range factor.

We used the experimental setup as shown in Fig. 6 to experiment hologram storage with angle multiplexing. The fixed object beam is simplified to be a parallel beam and 500 parallel reference beams are in the plane containing reference and object beams with 0.03° (outside the $LiNbO_3$:Fe) separation around the reference beam of $\theta = 13^\circ$. Sequential exposure was adopted for equivalent diffraction efficiency. Then the storage medium, flaky

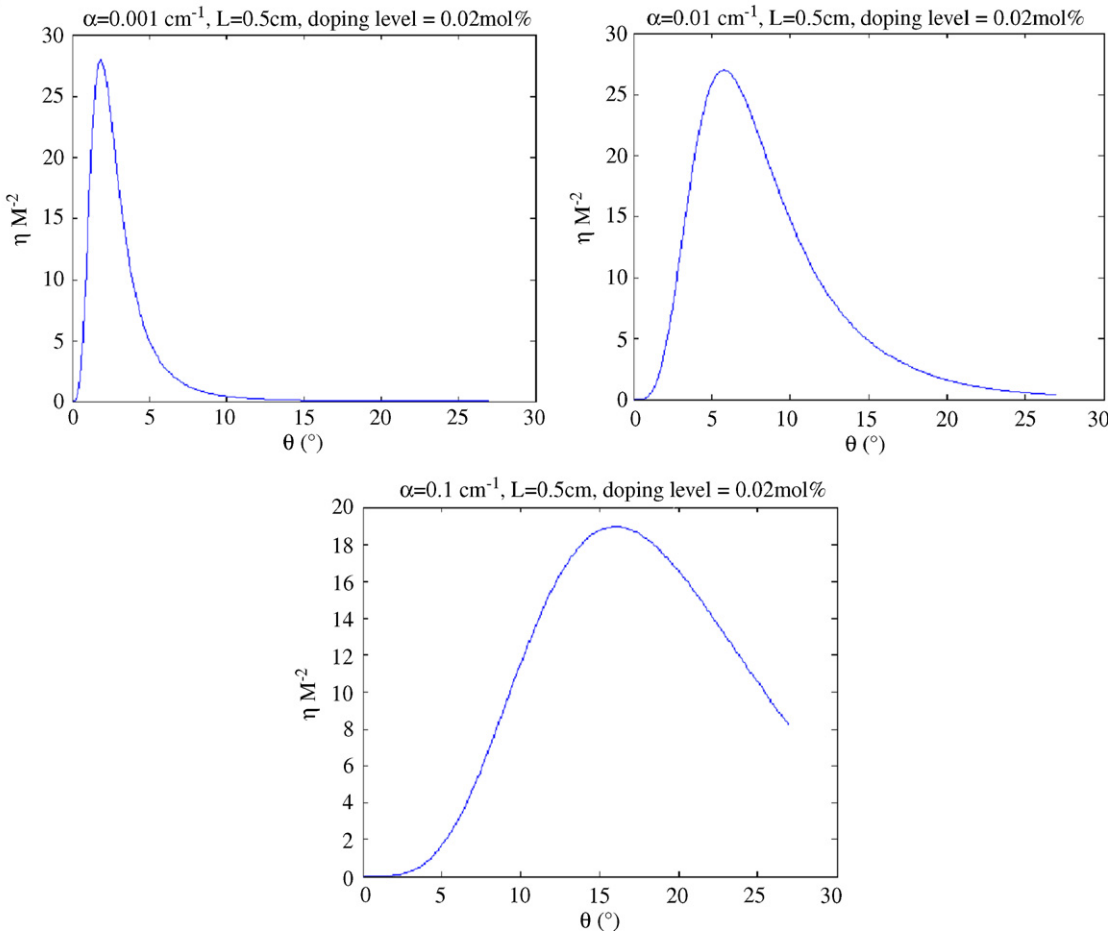


Fig. 4. Diffraction efficiency as a function of $\theta$ with different absorption coefficient at 0.02 mol% doping

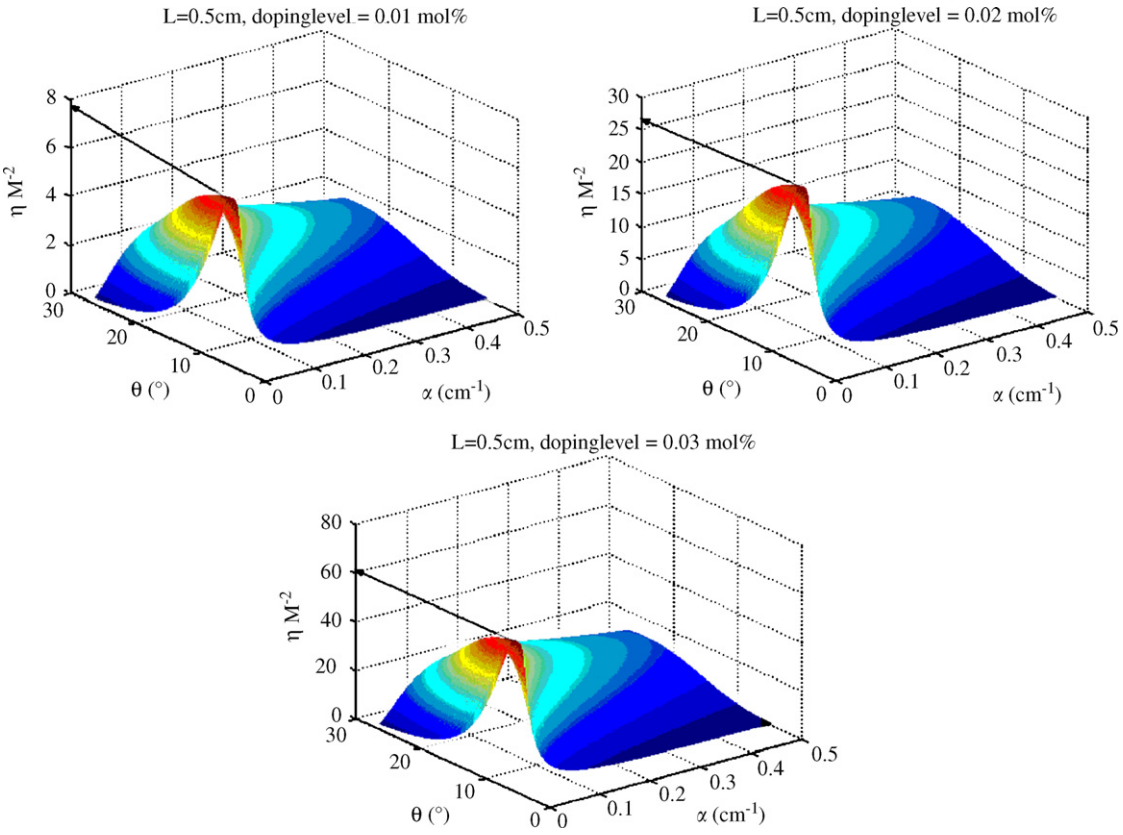


Fig. 5. Diffraction efficiency as a function of $\theta$ and absorption coefficient with different doping levels.

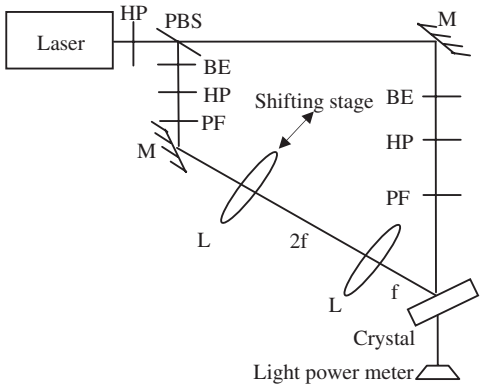


Fig. 6. Experimental setup for thermal fixing. HP, half-wave plate; PBS, polarizing beam splitter; BE, beam extender lens; PF, polarizing film; M, mirror.

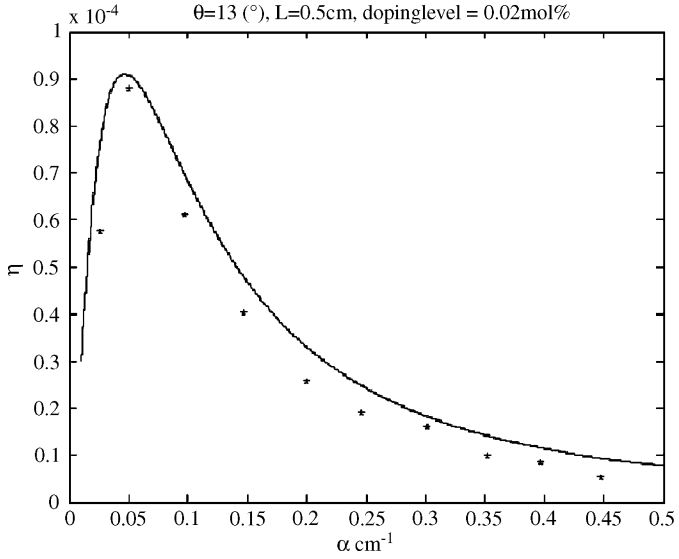


Fig. 7. Comparison of diffraction efficiency between experimental and theoretical predictions as a function of absorption coefficient.

$LiNbO_3$:Fe with $L = 0.5$ cm, $m = 1$, $\theta = 13^\circ$ (corresponding to $30^\circ$ outside $LiNbO_3$:Fe) at 0.02 mol% doping level, may be heated to 150 °C for thermal fixing. The crystal was allowed to cool and illuminated with UV light to reveal the fixed holograms. After each diffraction efficiency measurement, the crystal was annealed at 980 °C in an argon–oxygen mixture. A combination of oxygen partial pressure and time was used as the control variable for changing the absorption coefficient measured at 488 nm extraordinary polarized light. No spatial variations in absorption coefficient were observed. Taking the diffraction efficiency corresponding to the reference beam of $\theta = 13^\circ$, Fig. 7 shows the comparison between experimental results and theoretical predictions. Except that the experimental data are a little less than the prediction as a whole, a good agreement between the theoretical prediction and the experimental data points. All the deviation could be explained as follows: (1) to the thermal fixing, the

electronic gratings could be seen are not fully compensated by the proton grating after heating; (2) to angle multiplexing, the angle of references which is a range around $\theta = 13^\circ$ is different from our model taking $\theta$ as $13^\circ$ exactly; and (3) the coefficient of $\alpha$ to $N_D - N_A$ is not exactly for 488 nm.

In conclusion, from our study we know higher doping and thicker $LiNbO_3$:Fe are beneficial for diffraction efficiency. Also the angle between reference and object beam is better as small as possible except that too small angle would lead to a big Bragg angle which limits the angle multiplexing. For moderate angle between reference and object beam, an appropriate absorption coefficient is needed for optimal diffraction efficiency after thermal fixing and our model may predict this value effectively. This has a practical value for hologram storage including thermal fixing in $LiNbO_3$:Fe.

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