Material optimization for low scattering noise during nonvolatile holographic recording in doubly doped LiNbO$_3$ crystals

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Scattering noises in four kinds of lithium niobate crystals with the same double doping system, which are LiNbO$_3$:Fe:Mn, LiNbO$_3$:Ce:Mn, LiNbO$_3$:Ce:Cu, and LiNbO$_3$:Fe:Cu, are observed and compared experimentally. The results show that nonvolatile holographic recording can effectively suppress scattering noise, which mainly depends on recombination coefficients of both the shallower centers and the deeper centers. The small recombination coefficients of the shallower centers and the large recombination coefficients of the deeper centers benefit the amplification of the signal gratings and the suppression of the noise gratings. In addition, the initial seed scattering also impacts the recorded scattering noise, and the little seed scattering results in low scattering noise. The theoretical simulations are performed for confirmation. Among the four kinds of doubly doped crystals, in LiNbO$_3$:Ce:Cu the performances of nonvolatile recording are the best with low scattering noise and high diffraction efficiency.

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Recently, an improved method to realize nonvolatile holographic storage by two-center recording in LiNbO$_3$(LN):Fe:Mn has been proposed[1]. It has advantages of high diffraction efficiency and all-optical process, and has been realized as the most promising holographic fixing technique in practical applications. However, large scattering noise during nonvolatile recording, such as in LN:Fe:Mn, is a great drawback in applications[2]. Up to present, there is no reports on material optimization for low scattering noise in doubly doped LN crystals during nonvolatile holographic recording. Therefore, it is significant to investigate the microcosmic amplification mechanism of scattering noise during nonvolatile holographic recording to optimize the materials for efficient recording with low scattering noise.

In principle, when a laser beam illuminates a crystal, there emerges an initial scattering of the incident beam from inhomogeneities within the crystal. The initially induced scattering beams are gradually amplified by photorefractive effect and build-up many scattering noise gratings. There are two necessary conditions for the buildup of scattering noise gratings: initial sources of scattering, so-called seed scattering, and a sufficient amplification mechanism. Seed scattering is intrinsic to the phases of crystal growth and processing, and it is inevitable. Therefore, the suppression of scattering noise can possibly be achieved by the reduction of the scattering amplification mechanism. Conventionally, there are two key methods to suppress the scattering noise. One is to suppress the buildup of scattering noise gratings during recording and/or to reduce the recorded scattering noise gratings during readout[3,4]. The other is that damage-resistant dopants are doped into the crystals and the scattering noise is partly prevented[5].

In fact, during photorefractive holographic recording, the signal gratings and the scattering noise gratings are simultaneously amplified and constructed on the basis of the same photorefractive effect. However, there is a dynamic nonlinear amplification competition between the signal gratings and the scattering noise gratings. If the relevant medium parameters are optimized to specially benefit amplification of the signal gratings, it can effectively suppress the scattering noise gratings and possibly provide a fundamental solution to the problem of scattering noise intrinsic to the recording media. In this paper, scattering noises in four possible LN crystals with the same double doping system are observed and compared in experiments. The dependence of the amplification competition between the signal gratings and the scattering noise gratings on the recombination coefficients of both the shallower centers and the deeper centers is investigated and deduced. Finally, the mathematical simulations of the jointed material equations are performed, in which the dynamic amplification competition between the signal gratings and the scattering noise gratings is considered.

In principle, in doubly doped LN crystals for nonvolatile holographic recording, there are alternatives of Cu and Mn in the deeper centers and that of Fe and Ce in the shallower centers[1]. So, there are four possible crystals: LN:Fe:Mn, LN:Ce:Mn, LN:Ce:Cu, and LN:Fe:Cu. The above four kinds of congruent LN crystals have been grown by the Czochralski method. Typically, in order to produce nonvolatile gratings in the deeper centers finally, the samples are properly oxidized[6]. All the specimens are then polished to 10×10×2.5 mm^3 size.

The experimental setup is shown in Fig. 1. A 20-mW He-Ne laser provides two ordinarily-polarized beams which intersect symmetrically inside the crystal with the intersection angle of 30°. A transparency of the letter "E" is put into one of the recording beams, which is called signal beam in this paper, for the convenience of observation of scattering noise. The other recording

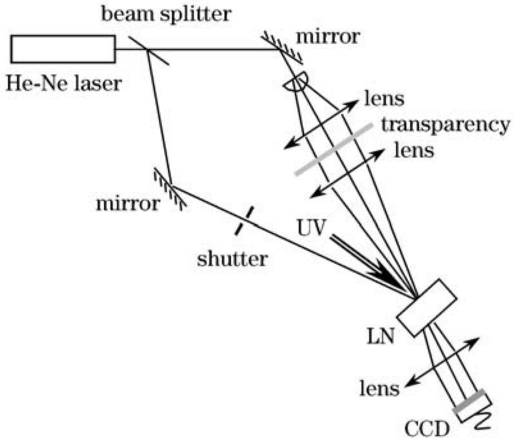

Fig. 1. Experimental setup for observation of scattering noise during nonvolatile holographic recording in doubly doped $LiNbO_3$ crystals.

beam is called reference beam accordingly. A filtered 75-W mercury lamp is used as the source of ultraviolet (UV) light. During recording, two red laser beams together with UV light are incident on the crystal to record a holographic grating. Note that using a single laser beam to radiate the crystal is a very effective technique for the observation of amplification mechanism of scattering noise in photorefractive crystals[7]. However, this method can not embody the competition between signal gratings and scattering noise gratings during holographic recording. In this paper, in order to comparably investigate the effect of nonvolatile holographic recording on the scattering noise, we perform observing the image of the letter "E" constructed along the signal beam both after the single signal beam exposure and after nonvolatile holographic recording. Therefore, in the latter case, after nonvolatile holographic recording it is the image of the letter "E" reconstructed along the signal beam, instead of the holographic image reconstructed by the reference beam, that is detected by charge-coupled device (CCD).

For comparison, three groups of systemic experiments are performed. Firstly, the CCD receives the images just at the beginning of which the signal beam is singly incident into the crystals. Secondly, the images are monitored when the crystals are radiated by the signal beam half an hour later. Thirdly, the same images are detected after half an hour of nonvolatile holographic recording in other positions of the crystals.

Firstly, while the signal beam is incident into the samples of four kinds of LNcrystals, the radiation time is too short to amplify the seed scattering and the initially detected scattering noise is very low, as shown in Figs. 2(b)−(e). However, it is important to be noted that the initially scattering noise is different among the four kinds of crystals: scattering in LN:Fe:Mn and LN:Fe:Cu is larger than that in LN:Ce:Mn and LN:Ce:Cu. In fact, little initially scattering in LN:Ce:Mn and LN:Ce:Cu is due to the fact that the Ce ions benefit better optical quality than other dopants[8] and result in little seed scattering. Secondly, after the crystals have been exposed to the signal beam for 30 minutes, the monitored images along the signal beam are shown in Figs. 2(f)−(i). Obviously, the amplified scattering noise is larger in LN:Fe:Mn and LN:Fe:Cu in which the initial scattering noise is larger. Accordingly, it is less in LN:Ce:Cu in which the initial scattering is less. This kind of discrepancies in amplified scattering noise when the single signal beam singly radiates the crystals are mainly due to the difference of the initial seed scattering.

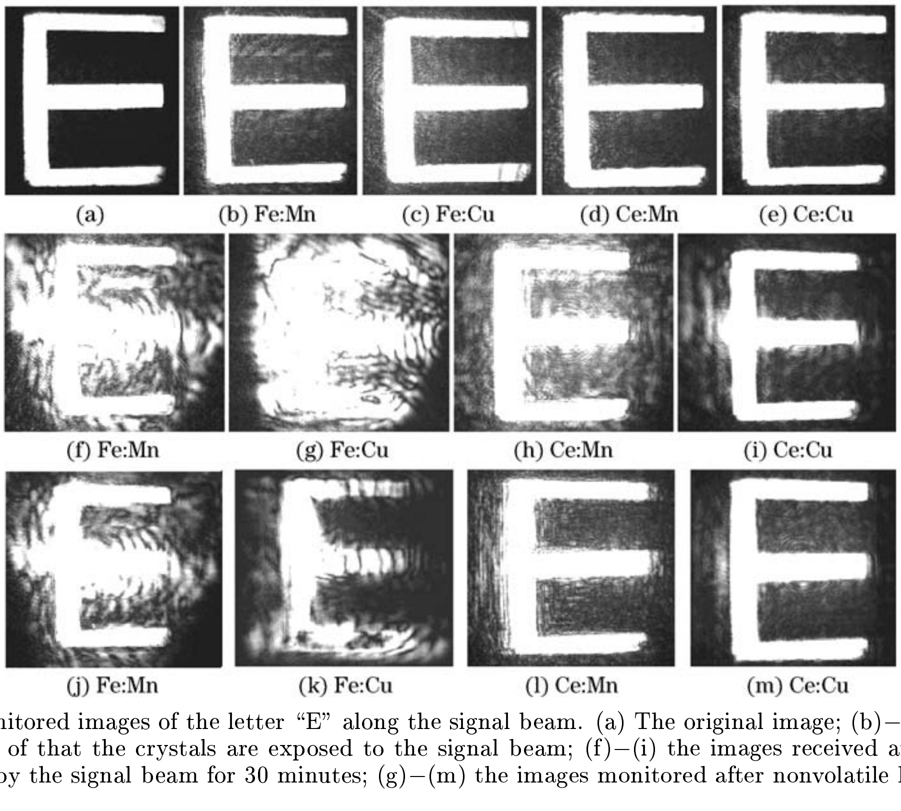
(a) (b) Fe:Mn (c) Fe:Cu (d) Ce:Mn (e) Ce:Cu
(f) Fe:Mn (g) Fe:Cu (h) Ce:Mn (i) Ce:Cu
(j) Fe:Mn (k) Fe:Cu (l) Ce:Mn (m) Ce:Cu

Fig. 2. The CCD monitored images of the letter "E" along the signal beam. (a) The original image; (b)−(e) the images gotten just at the beginning of that the crystals are exposed to the signal beam; (f)−(i) the images received after the crystals have been singly radiated by the signal beam for 30 minutes; (g)−(m) the images monitored after nonvolatile holographic recording for 30 minutes.

Thirdly, in order to observe the effect of nonvolatile holographic recording on the amplification of scattering noise, the crystals are moved to another position and nonvolatile holograms have been recorded for 30 minutes. After recording, both the reference beam and UV light are shut off and the images reconstructed along the signal beam are monitored, as shown in Figs. 2(g)−(m). Compared with the former results in the case of radiating only by the signal beam, the detected scattering noise in the letter "E" after nonvolatile recording is greatly reduced in the four crystals, respectively. The kind of decrease in scattering noise after holographic recording is due to the fact that there is a dynamic amplification competition between the signal gratings and the scattering noise gratings during holographic recording[9]. This kind of competition limits the amplification of the noise gratings, and thus nonvolatile holographic recording can suppress scattering noise at a certain degree.

During nonvolatile holographic recording, this kind of dynamic competition mainly depends on the microcosmic mechanism of the photorefractive effect in the medium, especially the charge transport mechanism during recording. Because the photovolatic effect is the predominant charge transport mechanism in doped LN crystals[10], the photovolatic fields have important effect on the amplification competition. In doubly doped LN crystals, the recombination coefficients of the shallower and the deeper centers are proportional to the photovolatic fields[11] and thus determine the amplification competition between the signal gratings and the scattering noise gratings greatly. In the four kinds of doubly doped LN crystals, it is known that the recombination coefficient of the shallower centers Fe is larger than that of Ce[11,12], and the recombination coefficient of the deeper centers Cu is larger than that of Mn[11]. Through the comparison of the observed scattering noise after nonvolatile recording as shown in Figs. 2(e)−(g), it is thought that the effect of nonvolatile recording on the suppression of scattering noise in LN:Ce:Cu is the best among the four kinds of crystals and the diffraction efficiency is also high[13,14]. And there comes a phenomenological conclusion that the combination of small recombination coefficients of the shallower centers and large recombination coefficients of the deeper centers benefits low scattering-noise and high diffraction efficiency during nonvolatile holographic recording.

To simulate theoretically the function of nonvolatile holographic recording on the suppression of scattering noise, all the scattering noise beams are considered as a plane wave propagating along the bisector between the reference beam and the signal beam[15]. And for simplification, the situation that the intensity of the reference beam is much larger than that of the signal beam is supposed and only two kinds of beam coupling are considered: the coupling between the reference beam and the signal beam, and the coupling between the reference beam and the scattering beam. Based on the two-center material equations describing nonvolatile holographic recording in doubly doped LN crystals[6], the intensity modulation depth of the reference beam and the scattering beam is included into the modified model. The dependence of amplification competition between the signal gratings and the scattering noise gratings on the recombination coefficients of the shallower centers and the deeper centers is deduced. Figure 3 gives the dependence of the ratio of the saturation diffraction efficiency of the signal gratings to that of the scattering noise gratings after recording on the recombination coefficients for the shallower centers (γ_2) and the deeper centers (γ_1), respectively. The results show that the ratios of saturation diffraction efficiency of the signal gratings to that of the scattering noise gratings decrease with the increase of γ_2 while they become large with the increase of γ_1. It is obvious that the combination of the small γ_2 and the large γ_1 benefits the suppression of scattering noise, which is consistent with the observed results very well.

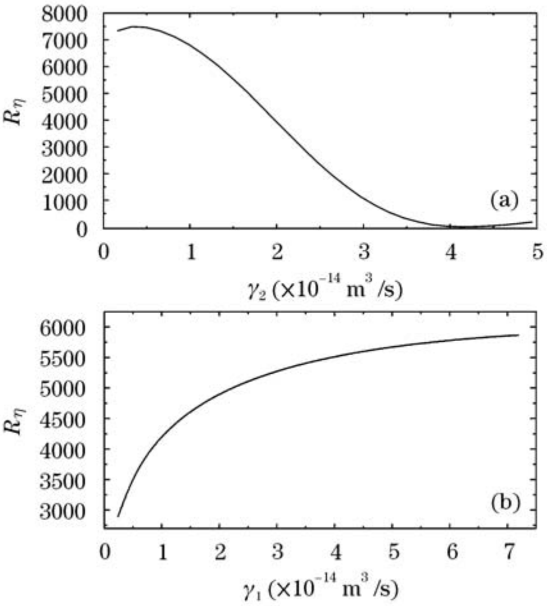

Fig. 3. Dependence of the ratio of saturation diffraction efficiency of the signal gratings to that of the scattering noise gratings (R_η) after recording on the recombination coefficients for the shallower centers γ_2 (a) and the deeper centers γ_1 (b).

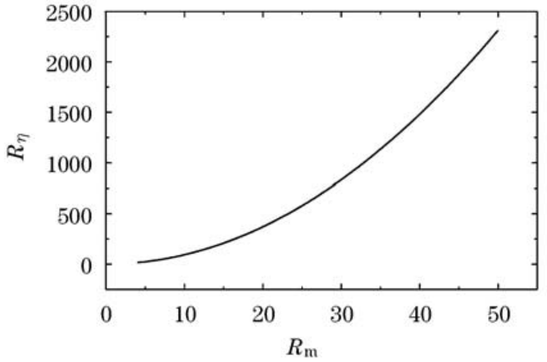

Fig. 4. Dependence of the ratio of saturation diffraction efficiency of the signal gratings to that of the scattering noise gratings (R_η) on the ratio of the intensity modulation depth for the signal gratings to that for the scattering noise gratings ($R_{\rm m}$).

At last, the effect of the initial seed scattering on the suppression of the scattering noise after nonvolatile recording is simulated with the fixed recombination coefficients of the shallower and the deeper centers. For evident physical profile here, the dependence of the ratio of the saturation diffraction efficiency of the signal gratings to that of the scattering noise gratings on the ratio of the intensity modulation depth for the signal gratings to that for the scattering noise gratings is investigated while the intensity modulation depth for the signal gratings remains fixed. The result in Fig. 4 shows that the larger the initial seed scattering, the higher the recorded scattering noise.

In conclusion, nonvolatile holographic recording in doubly doped LN crystals can suppress scattering noise, and small recombination coefficient of the shallower centers and large recombination coefficient of the deeper centers benefit the amplification of the signal gratings and the suppression of the noise gratings. In addition, little initial seed scattering results in low scattering noise.

Among the four kinds of crystals with the same double doping system for nonvolatile holographic recording, the performances of LN:Ce:Cu is the best. There are two reasons: the recombination coefficients of both Ce and Cu are favorite to the suppression of the scattering noise, and the dopant Ce has less effect on the development of intrinsic defects and benefits better optical quality of the crystal than other dopants.

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