A novel method for persistent holographic recording in doubly–doped lithium niobate

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ABSTRACT

We present a novel method for recording persistent holograms in doubly–doped $LiNbO_3$. Simultaneous sensitization of the crystal by an inhomogeneous UV beam can result to 32 % diffraction efficiency recorded by red light in a 0.85 mm thick sample, while read-out with only red light is non-destructive. Rapid optical erasure by ultraviolet light is possible and light scattering is efficiently prevented.

Keywords: Holographic storage, photorefractive materials, Two-center recording, Persistent Holographic recording

1. INTRODUCTION

Volume holographic memories are promising for high density digital or analog data storage.[1] Excellent light sources, spatial light modulators and camera systems are available due to the applications of optics in communication and imaging. The critical issue for holographic storage is still the recording material. Volume holographic storage systems are tried and tested with photorefractive crystals as the recording medium.[2–4] Inhomogeneous illumination with an interference pattern of reference and signal beams excites charge carriers from impurity levels into conduction or valence band, the charge carriers migrate and they are trapped by empty impurity levels elsewhere. A space–charge field builds up and modulates the refractive index via the electro–optic effect. Different photorefractive centers can interact and the performance depends strongly on the host material, intrinsic and extrinsic defects, and experimental conditions.[5,6]

The photorefractive effect is reversible, i.e. homogeneous illumination redistributes the electrons back and new recording is possible. Thus read/write memories can be implemented. However, the major obstacle is that readout also requires homogeneous illumination which erases the stored information. One approach to obtain persistent (non–destructive read–out) storage is to copy the space–charge pattern into a pattern of immobile ions (thermal fixing).[7] Ions become mobile if the crystals are heated, they migrate to compensate the space–charge field and after cooling they become immobile again. The resulting ion pattern cannot be erased by light. Alternatively, the recorded space–charge field can be copied into ferroelectric domains (electrical fixing).[8] This is accomplished through application of external electric fields, which switch ferroelectric domains in the regions where the external and internal fields add up. The domain pattern is also stable against further illumination. Unfortunately heating or application of large external fields is not practical and rapid refreshing of the memory is not possible. Two-step recording[9] is an all-optical approach for non-volatile storage: Recording light of low photon energy and sensitizing light of high photon energy excite electrons via virtual or real intermediate levels to the conduction band. For read-out only light of low photon energy is used which cannot by itself generate free electrons, and thus the stored charge pattern is stable. The requirement of high light intensities and expensive light sources is the drawback of this approach. Two–step holographic recording experiments using both high-intensity pulses[10,11] and cw light[12,13] have been reported. Despite impressive recent progress in this field,[14] the dynamic range and the sensitivity are still not satisfactory. Recording with light of one wavelength and just reading with light of a shorter one causes substantial information losses due to the Bragg condition of volume diffraction.[15] Even though all these techniques work,[16–19] the outlined disadvantages have made holographic data storage impractical thus far.

In this article we present a novel holographic recording method (two–center method) that can lead to the realization of a practical persistent holographic memory system. In a 0.85 mm thick $LiNbO_3$:Fe:Mn crystal up to 32 % diffraction efficiency and non-destructive read-out are achieved for gratings recorded with red light, if during

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recording incoherent ultraviolet light is present. Rapid optical erasure by ultraviolet light is possible and light scattering is efficiently prevented. Two-center holographic recording method is introduced in section 2, and experimental demonstration of the method is presented in section 3. Performance characteristics along with possible ways for improvement are discussed in section 4. Finally, conclusions are made in section 5.

2. TWO-CENTER HOLOGRAPHIC RECORDING

Build-up of space–charge fields in photorefractive materials requires redistribution of charge. Transition metal ions can occur in inorganic crystals in different valence states, e.g. iron is present in $LiNbO_3$ as Fe^{2+} and Fe^{3+}.[20] Figure 1 (a) shows the energy band diagram for a $LiNbO_3$:Fe crystal. Inhomogeneous illumination excites electrons from Fe^{2+} to the conduction band, and they move due to the photovoltaic effect, diffusion and drift. The electrons are trapped by Fe^{3+} elsewhere and a space–charge field builds up, which modulates the refractive index via the electro–optic effect. Data can be erased by homogeneous illumination. However, readout also requires homogeneous exposure, which causes undesired erasure of the stored information. This is a general problem of all reversible storage media.

A new way to solve this problem is the usage of doubly–doped photochromic crystals.[21] In the recent work,[21] $LiNbO_3$ doped with manganese (Mn) and Fe is used. It is known that Fe/Mn doubly–doped $LiNbO_3$ is photochromic and that ultraviolet pre–illumination enhances the sensitivity for a few recording and erasure cycles with visible light.[22] The energy band diagram of such a crystal is shown in Figure 1 (b). Fe and Mn ions occur in the valence states $Mn^{2+/3+}$ and $Fe^{2+/3+}$,[23] and thermal depletion plays no role. Electrons can be excited by ultraviolet light either from Mn^{2+} or from Fe^{2+} into the conduction band while red light excites electrons only from the shallower Fe^{2+}, because the red light has a smaller photon energy. The conduction band electrons can recombine with both centers, and thus ultraviolet illumination populates the $Fe^{2+/3+}$ level partially while the red light empties the Fe sites. The filled Fe levels cause a broad–band absorption in the visible with a maximum at 477 nm light wavelength.[20] Thus ultraviolet light sensitizes the material while red light bleaches it. The basic idea of two–center holographic recording is to bring with the ultraviolet light electrons from Mn to Fe via the conduction band, use these electrons to record the hologram with red light, and eventually transfer the electrons from iron back to the manganese centers by red light. This results in a hologram stored in Mn centers that persists against further red illumination. One of the key material parameters in two–center holographic recording is the initial electron concentration in Mn and Fe traps. These concentrations can be varied by annealing (or so–called oxidation / reduction treatment).[24] For persistent holographic recording, it is necessary that the final hologram be stored in Mn centers to persist against further read–out by red light. Mn traps are deeper in the band gap than Fe traps. Therefore, electrons would fill the Mn traps before Fe traps. As a result, it is essential for persistent recording that all Fe traps be empty, and only a portion of the Mn traps be filled. This guarantees that the final hologram can be recorded in Mn traps after sufficient read–out.

3. EXPERIMENTAL RESULTS

We performed experiments with a 0.85 mm thick congruently melting x–cut $LiNbO_3$ crystal doped with 0.075 wt. % Fe_2O_3 and 0.01 wt. % MnO. The sample was oxidized enough to result in empty Fe traps and partially filled Mn traps. We used a 100 W mercury lamp as the ultraviolet light source (wavelengths 365 nm), and a 35 mW HeNe laser for generation of the coherent red light (wavelength 633 nm).

A proper ratio between the intensities of the red recording and the ultraviolet sensitizing light, I_{red}/I_{uv}, is essential to get good holographic recording performance. Too much ultraviolet light causes erasure while too much red light causes bleaching and low sensitivity. A convenient way to adjust the intensity ratio is to use sensitization and bleaching experiments. UV light causes electron transfer from Mn to Fe centers. This increases the absorption of the red light. On the other hand, red light causes electron transfer from Fe to Mn centers resulting in smaller absorption for red light. During sensitization experiment, the absorption of the crystal under UV illumination is monitored by measuring the transmitted intensity of a weak probe red beam (wavelength 633 nm, ordinary polarization). After sensitizing with UV light, the crystal is bleached by a strong red beam while the absorption of the crystal is monitored by measuring the transmitted intensity of the bleaching beam. Figure 2 shows typical results of the sensitization and bleaching experiments.

The time constants of sensitization and bleaching are measures for the rates of population and depopulation of the iron sites. They scale linearly with the light intensities, which allows tuning. The time constants should be of the same order of magnitude to achieve a strongly modulated Fe^{2+} concentration. Too strong UV light (compared

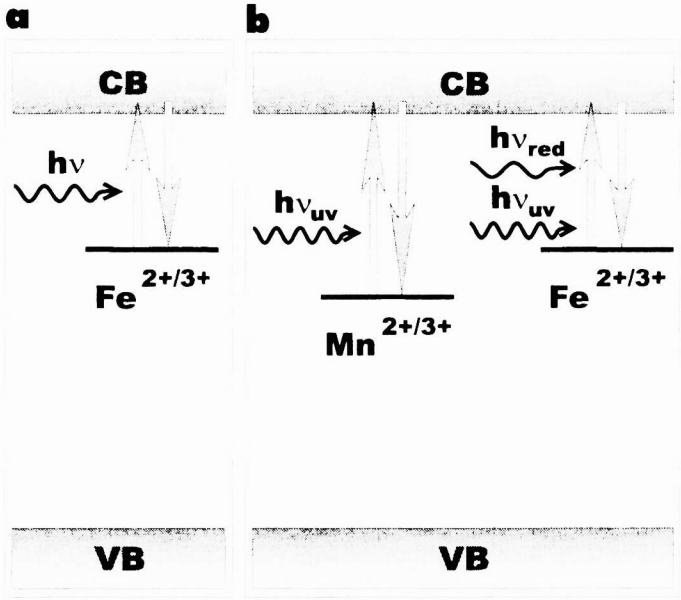

Figure 1. Energy band diagram for a typical $LiNbO_3$ crystal doped with **(a)** Fe, and **(b)** Fe and Mn. CB, and VB stand for conduction band and valance band, respectively.

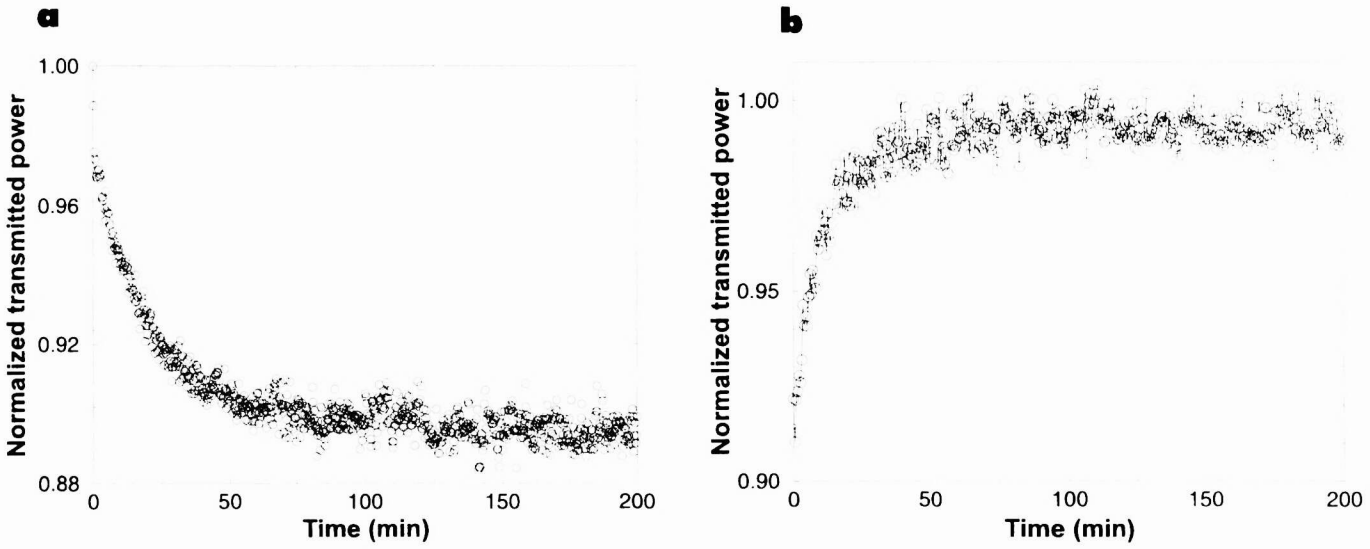

Figure 2. Normalized transmitted red intensity in a 0.85 mm $LiNbO_3$:Fe:Mn crystal. (a) Sensitization experiment: The crystal is sensitized with a homogeneous UV beam (wavelength 365 nm, intensity 20 mW/cm^2) while monitored by a weak red beam (wavelength 633 nm, intensity 0.6 mW/cm^2, ordinary polarization), (b) Bleaching experiment: The sensitized crystal is bleached with a strong red beam (wavelength 633 nm, intensity 300 mW/cm^2, ordinary polarization)

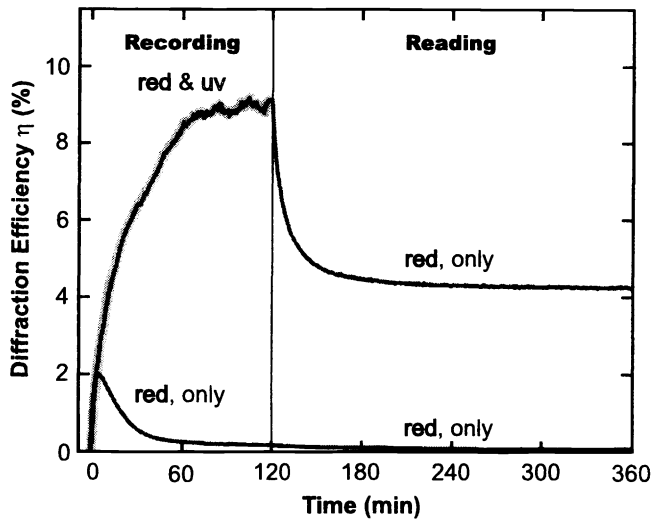

Figure 3. Diffraction efficiency η versus time for recording without and with simultaneous presence of ultraviolet light, and for subsequent reading in a $LiNbO_3$:Fe:Mn crystal.

to red light) results in too rapid sensitization and therefore too rapid erasure of the hologram. Therefore, strong holograms can not be recorded. On the other hand, too strong red light (compared to UV light) results in too fast bleaching of the Fe traps, and lack of enough electrons in Fe traps for efficient holographic recording. Therefore, it is important to optimize the ratio between UV and red intensities. We find from the sensitization / bleaching experiments an optimum intensity ratio of $I_{\mathrm{red}}/I_{\mathrm{uv}} \approx 30$.

To get information about the holographic performance, plane–wave gratings are recorded and reconstructed. The unpolarized ultraviolet light illuminates the sample homogeneously; the HeNe laser light is split into two plane waves which interfere at the crystal ($1/e^2$ beam diameter 2.0 mm, transmission geometry, period length of the grating 0.9 μm). The grating vector is aligned parallel to the c–axis of the sample. The crystal is pre–exposed to UV light for at least 3 hours before recording. During recording, one of the HeNe beams is blocked from time to time and the second beam is diffracted from the written grating to obtain the diffraction efficiency η as the ratio between diffracted and total incident light powers. Figure 3 shows the results. The diffraction efficiency raises quickly and drops afterwards almost to zero with no ultraviolet light present during the hologram formation. After some reading, the grating finally disappears completely. With assistance of ultraviolet light during recording, much higher efficiencies are obtained. Subsequent reading erases first the grating partially, but the remaining grating persists despite further red illumination.

As shown in Fig. 3, the presence of ultraviolet light during hologram formation is crucial for obtaining large diffraction efficiencies. This is due to the fact that the grating can not be recorded in the Mn traps in the absence of UV during recording. This can be understood by the following argument: The electron recombination rates of Mn and Fe centers have the same order of magnitude. Therefore, the probabilities of trapping a conduction band electron at Mn and Fe centers are comparable. As a result, when an electron is excited from Fe centers to the conduction band, it will end up in Mn centers after at most a few retrapping at Fe centers (for example, if the trapping probability at each center is 1/2, the average number of retrapping at Fe centers before being trapped at Mn centers is 2). An electron moves only a few nanometers in the conduction band before getting retrapped at either centers due to small mobility of $LiNbO_3$. Therefore, if there is no UV illumination during recording, an electron moves only a few nanometers on the average which is much less than the grating period (usually around 1 μm). This is due to the fact that red light is not able to excite electrons from Mn centers. When an electron is trapped at these centers, it can not be used for holographic recording any more. Having simultaneous UV illumination during recording makes the Mn electrons available for recording and increases the average distance an electron can move through multiple cycles of excitation. This results in a successful recording to large saturation diffraction efficiencies.

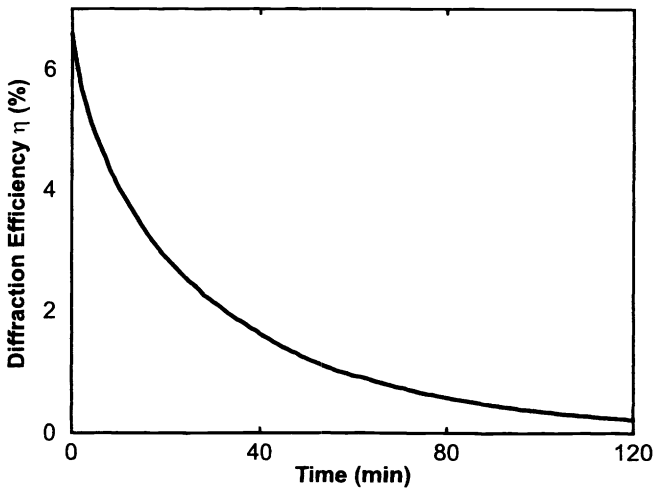

Figure 4. Diffraction efficiency η versus time for erasure with simultaneous presence of UV and one of the red recording beams. The hologram was recorded by simultaneous presence of UV and two red recording beams to an arbitrary diffraction efficiency of close to 7 %.

Without UV, the recording light bleaches the Fe centers. This bleaching is faster in high light intensity regions. Therefore, at some point in time, the bulk photovoltaic current ($j_{\mathrm{ph}} = \kappa N_{\mathrm{Fe}^{2+}} I_{\mathrm{Red}}$) at the peaks of the interference pattern becomes weaker than the bulk photovoltaic current at positions away from the peaks. This reverses the prevailing charge transfer direction and causes erasure. The peak in the recording curve (with no UV) corresponds to this reversal of direction. Simultaneous UV illumination during read–out prevents the full bleaching of Fe centers by continuously transferring electrons from Mn centers to Fe centers. This maintains a steady–state electron concentration in Fe centers resulting in stronger photovoltaic current and stronger hologram.

Therefore, the key idea that allows non-volatile storage is to illuminate the crystal with ultraviolet and red light simultaneously, wait until saturation is reached and then switch the ultraviolet light off. The hologram is recorded in both Fe and Mn traps. During read–out by red light, electrons are transfered from Fe to Mn traps resulting in a partial erasure of the hologram. After all electrons are transfered to Mn centers, the remaining hologram persists against further red read–out. We can erase the hologram by UV light or by simultaneous UV and red illumination. Figure 4 shows the diffraction efficiency vs. time for a plane–wave hologram erased by UV and red beams simultaneously. Recording was performed by two red beams with simultaneous illumination with a UV beam, while erasure was performed by the UV beam and one of the red recording beams. The specifications of the beams are the same as those of the previous experiment.

4. DISCUSSION

As shown in section 3, two-center holographic recording is an efficient method for recording persistent holograms in $LiNbO_3$. The obtained performance is exciting: We reach a persistent diffraction efficiency of 4 % for ordinarily polarized light and, due to a larger electro–optic coefficient, 32 % for extraordinary polarization. The square root of the saturation efficiency yields approximately the M/#, a measure of how many holograms can be multiplexed.[25] We get a M/# of about 0.6 with the 0.85 mm thick crystal. Typical values for volatile recording with green light in iron-doped material using ordinary polarization are around M/# = 1 for approximately 1 cm thick crystals.[25] Since the M/# is proportional to the thickness of the recording material, a 1 cm thick crystal with the new technique should yield M/# = 7. However, the absorption coefficient of the sample at 365 nm is large resulting in smaller M/# and sensitivity. A major part of the absorption is caused by band-to-band absorption of $LiNbO_3$ itself, and not the sensitization mechanism. One way to solve this problem is to use longer wavelength for sensitization. We recently performed experiments with 404 nm sensitizing light, and obtained better performance due to smaller UV absorption. Illuminating the sample from both sides with two UV sources can also improve the performance. Another

way to solve the absorption problem is to use other dopants. For example, we can replace Mn by Cu, which has higher energy level in the band-gap, and use higher wavelength for sensitization. To obtain persistent recording, we need to replace Fe by Ce for example and use higher wavelength for recording, too.

The main advantages of our technique compared to other non-volatile storage methods are: no need of heating, of external electric fields or of light of high intensity. Furthermore, during recording the ultraviolet light prevents build-up of holographically amplified scattered light or of screening fields created by accumulation of charge at the boundaries of illuminated regions. During readout the crystal is insensitive and cross-talk build-up due to two-wave mixing effects does not occur. Thus the fidelity of the stored information is significantly improved and the error rate drops. Smooth and continuous recording curves like in Fig. 3 are hard to obtain in conventional experiments. It appears that we have discovered a recording method with which essentially all desirable features of the holographic memory are improved. The prospects for practical holographic storage devices using this technique, are excellent.

5. CONCLUSIONS

We presented here a novel method for persistent holographic recording in doubly–doped $LiNbO_3$ crystals. For persistent holographic recording, the shallower traps must be empty, and the deeper traps must be partially filled. After sufficient read–out, the hologram is stored in the deeper traps and persists against further read–out. The final diffraction efficiency depends on the ratio of the sensitizing and recording light intensities. Sensitization and bleaching experiments are very helpful in choosing the optimum beam intensities. We also showed that the simultaneous presence of the sensitizing beam during recording is essential for obtaining large persistent diffraction efficiencies. It turns out that recording in doubly–doped crystals with simultaneous presence of long–wavelength recording and short–wavelength sensitizing light is a promising approach for all–optical persistent holographic data storage, and the underlying processes are correctly understood

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