

Hologram Multiplexing Using Two-Step Recording

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ABSTRACT

We present a comparison of different two-step holographic recording schemes based on their multiplexing performance using a unified framework. The difference between the different schemes is mainly due to the energy levels and sources of the traps. The influence of effective trap density and recording and sensitizing intensities are addressed. It is shown that for low light intensity operation, the performance of the doubly-doped crystals is superior due to the lack of dark depopulation of the shallow traps. Usage of such crystals also results in the suppression of the intensity threshold that exists for two-step recording in singly-doped crystals. The promising potentials for improvements in the two-step holographic recording in doubly-doped materials is also addressed.

Keywords: Holographic storage, photorefractive materials, Two-step recording, Two-center recording, Hologram multiplexing

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 electrooptic 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. Thermal fixing,⁷ electrical fixing,⁸ two-step recording,⁹ frequency-difference holograms¹⁰ and readout with wavevector spectra¹¹ are known techniques to overcome the problem of volatility. From all these techniques, two-step processes appear to be the most promising ones. They require no heating, no external electric fields and they may enable recording with a high dynamic range, i.e. multiplexing of many holograms with high efficiency.

Two-step holographic recording can be realized by using materials having an intermediate level. The material is first sensitized by a short wavelength sensitizing beam resulting in the promotion of the electrons from the deep traps to the shallow ones. Recording is then performed from the shallow traps with light of longer wavelength that promotes the electrons to the conduction band. These electrons are finally transferred to the deep traps and record the hologram there. Read-out is performed with the light of longer wavelength and therefore it is non-volatile.

Multiphoton photorefractive storage has been discovered in lithium niobate crystals (LiNbO_3),⁹ and the first non-volatile storage experiments utilizing two-step excitations were performed with LiTaO_3 , which is isomorphic to LiNbO_3 .¹² Picosecond light pulses (wavelengths 1064 and 532 nm) were used in these early investigations. Larger refractive index changes and better sensitivities were achieved using LiNbO_3 and nanosecond light pulses of the same wavelength.¹³⁻¹⁵ The crystals were doped with iron or with copper as the photorefractive centers. Iron and copper occur in LiNbO_3 and LiTaO_3 in the valence states $\text{Fe}^{2+}/3+$ and $\text{Cu}^{+}/2+$.¹⁶ Charge transport via the conduction band dominates, i.e. Fe^{2+} and Cu^{+} are the filled impurities. Two-step recording with cw light with lower intensities

in both Praseodymium-doped and nominally undoped stoichiometric LiNbO_3 have been reported.^{17–19} Recently, we proposed and demonstrated the two-step recording method in doubly-doped lithium niobate.²⁰

In this article, we explain the differences between the different two-step recording schemes. We use mainly the performance parameters regarding multiplexing many holograms as the measure for this comparison. Different two-step recording schemes are introduced in section 2. The theoretical model and the results of the comparison of these schemes are discussed in section 3. Section 4 is a brief comparison of the different schemes based on the published experimental results. Sections 3 and 4 provide an insight into the different electron transfer mechanisms involved in two-step recording schemes, and the consequences are discussed in section 5. Finally, conclusions are made in section 6.

2. TWO-STEP HOLOGRAPHIC RECORDING

Two-step holographic recording is performed in materials with two trap levels. Different two-step recording schemes are different mainly due to the position of these traps and how they are introduced into the material. Most of the previous work on two-step holographic recording has been performed in singly-doped crystals. The dopants, which are mainly either iron or copper in LiNbO_3 , result in the deep traps, and the shallow traps are believed to be due to small polarons. Congruent lithium niobate has a substantial lack of lithium ions. The lithium concentration is only 48.3 mol %.²¹ At least 1 % of the intrinsic defect niobium on lithium site (Nb_{Li}) is present.^{22,23} The valence state of this center is 5+, but one electron can be trapped, reducing the valence state to 4+ and creating a small polaron. All photorefractive properties of $\text{LiNbO}_3\text{:Fe}$ for illumination with continuous wave visible light and with high intensity green light pulses are quantitatively explained by a two-center charge transport model considering the photorefractive sites $\text{Fe}^{2+/3+}$ and $\text{Nb}_{\text{Li}}^{4+/5+}$.²⁴ Two-step transitions of electrons from Fe^{2+} via the small polaron into the conduction band are possible, because the concentration of small polarons is large and each Fe^{2+} ions has some empty Nb_{Li} close to its location. Effects like light-induced absorption changes, intensity dependent saturation values of the refractive index changes and photoconductivities, which are superlinear in the light intensity are successfully explained within this model.²⁴

The energy band diagram along with the possible electron transitions in two-step recording is depicted in Figure 1. The main mechanisms that we should explain are sensitization, recording, and the dark depopulation of the shallow traps (electron transfer from the shallow traps to the deep ones without light intervention).

The main goal in sensitization is electron transfer from the deep traps to the shallow ones. This can be done either directly (transition 1) or via the conduction band (transitions 2 and 7) as shown in Figure 1. The relative strengths of these sensitization paths depend on the concentrations of the deep and shallow traps, their relative absorption cross sections and electron recombination coefficients and sensitization light intensity. For congruent LiNbO_3 doped with a fair amount of iron, the direct pathway is more important, while for nominally undoped stoichiometric LiNbO_3 , the indirect pathway is dominant due to smaller polaron concentrations.

Recording is performed from the shallow traps through electron transfer to the conduction band induced by the recording light. These electrons then move in the conduction band and they are trapped by shallow and deep traps (transitions 4, 8, and to some degree 7 in Fig. 1). The relative strengths of the holograms recorded in the deep and shallow traps depend on the properties of the traps as mentioned for the sensitization processes. The portion of the hologram recorded in the shallow traps is finally transferred to the deep traps and combined with the hologram already recorded there. This can be assisted by a homogeneous beam with recording wavelength specially for the cases where dark transitions are not effective.

Finally, we should consider the dark mechanisms (those that are present regardless of the presence of light). These result in the depopulation of the shallow traps in time. The lifetime of the polaron states could be from milliseconds to seconds and even 10's of seconds depending on the material growth and temperature. These dark mechanisms could also be divided into direct (transition 6 in Fig. 1) and indirect via the conduction band due to thermal excitations (transitions 5 and 8 in Fig. 1). The light-assisted depopulation of the shallow traps caused by the sensitizing light and the dc part of the recording light is also present (transitions 3, 4, and 8 in Fig. 1). Again, the relative strength of the different mechanisms depends on the concentrations and the properties of the shallow and deep traps and involved light intensities. Temperature is also important for thermal excitation mechanism (transition 8 in Fig. 1). For example, direct mechanism is the major cause for the dark depopulation of the shallow traps in congruent iron-doped LiNbO_3 , while the thermal dark depopulation mechanism is the dominant one for nominally undoped stoichiometric LiNbO_3 at room temperature.

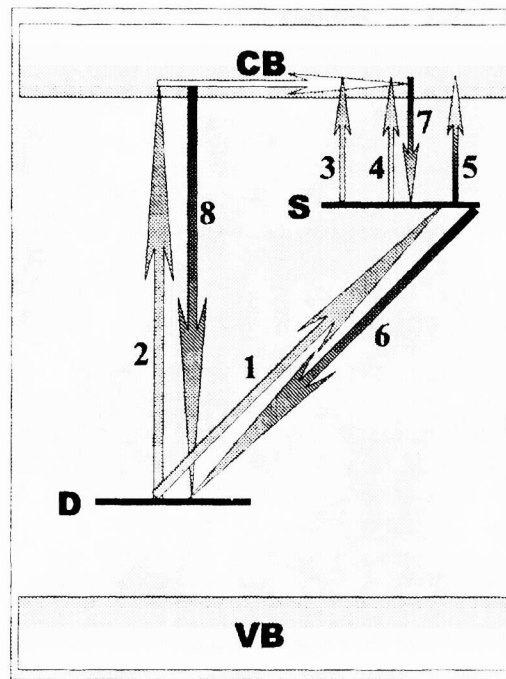


Figure 1. Energy band diagram and possible electron transitions for two-step holographic recording. Transitions 1, 2, and 3 are caused by sensitizing beam, and transition 4 by recording beams. Transition 5 is caused by thermal excitations, and all other transitions occur in dark without light assistance. VB, CB, D, and S stand for valence band, conduction band, deep trap, and shallow trap, respectively.

The dark depopulation of the shallow traps is the major problem in two-step recording in singly-doped or undoped crystals, resulting in the requirement of high light intensities in order to sensitize efficiently and use the electrons in the shallow traps for recording before they move back to the deep traps by the dark depopulating mechanisms. The shorter the lifetime of the polaron states (or the stronger the dark depopulation mechanisms), the higher the intensities that are needed for the same performance when everything else is the same. Recently, it has been shown¹⁹ that by using stoichiometric crystals, which have smaller polaron concentration, the lifetime of these shallow traps can be increased. Further increase of this lifetime can be achieved by using nominally undoped or very lightly doped stoichiometric crystals. These can be explained by noting that by reducing the concentration of the shallow traps, the direct depopulation mechanism becomes weak, and by reducing the deep trap concentration, re-trapping of the electrons from the conduction band by the shallow traps becomes more probable. As a result, polaron states will have longer lifetime, and holograms can be recorded with much lower intensities allowing recording with cw lasers. Although this scheme results in lower intensity requirements, it suffers from a major drawback. To record many strong holograms and have a large $M/\#$,²⁵ we need to have high concentration of the deep traps with optimized oxidation state. This by itself reduces the lifetime of the shallow traps, and therefore the sensitization efficiency and hologram strength. This dependence of the properties of the shallow traps on those of the deep traps makes the optimization of the material design difficult while the final performance is not very good despite the impressive research in the last few years.

The more recent idea, which we refer to as two-center method, is based on using crystals with two different dopants²⁰ that are deep enough to be safe from the thermal dark depopulation of the shallow traps. The practical concentrations of the traps are well below the minimum values necessary for direct transitions. Therefore, no dark depopulating effect is present. The only desirable mechanism that is lost is the direct sensitization process, but due to a much larger recombination coefficient of the shallow traps here, a good sensitization efficiency is still achievable. The hologram is recorded in both traps resulting in some initially partial erasure during read-out due to gradual transfer of the recorded charge pattern in the shallow traps to the deep ones, but the final hologram recorded in the

deep traps is effectively non-volatile. This method results in better performance than the other two-step scheme in all aspects (except at extremely high cw intensities).

3. MULTIPLEXING PERFORMANCE OF DIFFERENT TWO-STEP RECORDING SCHEMES

In this section we compare the theoretical predictions for multiplexing performance of the different two-step schemes. We consider mainly two different methods. The first one is the widely used two-step recording in singly-doped or nominally undoped crystals, where the shallow traps are due to polarons. We refer to this method as singly-doped method. The second one is the newly proposed two-center method which is performed in doubly-doped crystals. Our goal is to get insight into the qualitative differences between the two schemes rather than the precise quantitative differences. The later will be discussed in the next section where we compare the published experimental results. Since we are interested in multiplexing many holograms using these schemes, $M/\#$ would be a good measure for comparison. Using equal diffraction efficiency recording schedule²⁶ for multiplexing M holograms, the diffraction efficiency of each hologram is $\eta = \left(\frac{M/\#}{M}\right)^2$ where $M/\# = \frac{A_0\tau_e}{\tau_w}$. Here, τ_e and τ_w are erasure (with both recording and sensitizing beams present) and recording time constants, respectively and A_0 is linearly proportional to the saturation space charge field. In this paper, we concentrate on the saturation space charge fields of the different schemes, and we use it here as the comparison measure.

In our simulations, we use the two-center model²⁴ which is based on the different electron transfer mechanisms depicted in Fig. 1 and basic electromagnetic laws. The parameters we use in the model are found either from the literature or by fitting the experimental data. The details will be published elsewhere. For the singly-doped method, we assume that the direct sensitization and dark depopulation mechanisms are dominant (although similar results could be obtained by assuming the dominance of the indirect mechanisms using appropriate values for the parameters of the model). The crystal is assumed to be congruent LiNbO₃ doped with 0.01 wt. % Fe₂O₃ with polaron concentration equal to 10^{26} m^{-3} unless otherwise specified. This is compatible with the above assumption regarding the dominance of the direct mechanisms. The sensitization and recording wavelengths are assumed to be 532 nm and 1064 nm, respectively. For the two-center method, we assume that direct transition between the traps is prohibited, and thermal excitation of the electrons from the shallower traps to the conduction band is negligible. The crystal is assumed to be LiNbO₃ doped with 0.15 wt. % Fe₂O₃ and 0.01 wt. % MnO unless otherwise specified. Here iron and manganese levels act as the shallow and deep traps, respectively. sensitization and recording wavelengths are assumed to be 365 nm and 633 nm, respectively. The predictions of the model with the parameters chosen are in fair agreement with the experimental results. In the calculation of the space charge field, we assume no absorption for both the recording and sensitizing beams in all cases unless otherwise specified. Furthermore, the recording beams are assumed to be cw plane waves with ordinary polarization and the sensitizing beam is assumed to be cw and homogeneous.

Figure 2 represents typical recording and read-out curves for the two cases. Recording is performed for 100 seconds with sensitizing and recording beams on. Then the grating is read-out for 200 seconds with one of the recording beams while all other beams are blocked. The intensity of the sensitizing beam is $I_s = 1 \text{ W/cm}^2$ and the intensity of each recording beam is $I_r = 2 \text{ W/cm}^2$. The iron concentration here is 0.075 wt. % (smaller than what we use in future simulations) to relatively enlarge the recording curve of the singly-doped method. The non-volatility in both cases along with partial erasure due to the transfer of the space charge from the shallow traps to the deep ones in two-center method is evident from Figure 2. It can also be seen that the two-center method can result into much higher space charge field resulting in better $M/\#$ and multiplexing performance. This is mainly due to the lack of dark depopulation of the shallow traps in the two-center method.

To see the effect of the deep trap concentration (which can be easily varied in both cases), we assume that it is varied in a long but practical range while all other parameters are kept constant, and calculate the final (non-volatile) saturation space charge field for both cases. We also assume that for each deep trap concentration, half of the deep traps are ionized. The results are depicted in Figure 3. Due to large concentration of polarons assumed (10^{26} m^{-3}), and the strong dark depopulation effects, saturation field in singly-doped method increases by increasing the concentration of the deep traps getting close to a saturation at very high doping concentrations. Therefore, adding more deep traps means adding more electrons to work resulting is a stronger grating until we get to saturation (although it might occur at non-practical doping levels) as increasing the concentration of the deep traps increases the dark depopulation rate, too. The situation in two-center method is different. The main

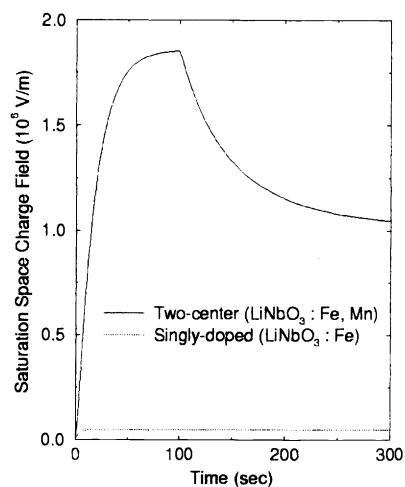


Figure 2. Recording and read-out curves for two-center and singly-doped methods. Recording is performed for the first 100 seconds where sensitizing and recording beams are all present. For the next 200 seconds, the sensitizing beam and one of the recording beams are blocked and read-out is performed with the other recording beam.

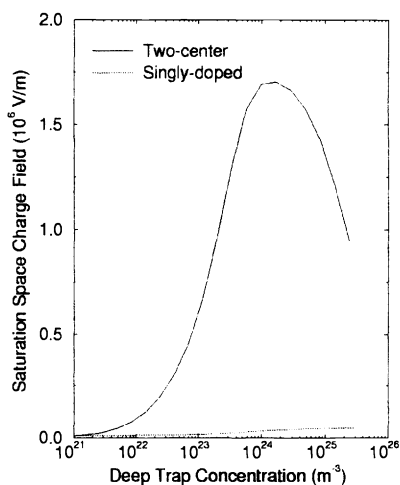


Figure 3. Variation of the non-volatile saturation space charge field with deep trap concentration for two-center and singly-doped methods.

competition here is between the two traps to capture the electrons from the conduction band. Each capture rate depends on the recombination coefficient of the traps involved (usually larger for the deeper traps), concentration of empty traps, and concentration of electrons in the conduction band. Although increasing the concentration of the deep trap results in the simultaneous increase of the sensitization rate by sending more electrons to the conduction band, it also increases the probability of electron capture by the deep traps (if we assume the same oxidation state for all doping concentrations) and therefore decreases the sensitization efficiency. Due to this trade off, an optimum is expected which occurs within practical doping levels here.

Figure 4 shows the variation of the final saturation space charge field with the recording and sensitizing light intensities. From Fig. 4(c) it becomes obvious that in the two-center method, the saturation field depends only on the intensity ratio and not on the absolute intensities if the absorption of the sensitizing beam can be neglected. This is due to the fact that too high recording intensities result in the bleaching of the shallow traps by the dc light while too high sensitizing beam intensities result in the strong erasure of any possible hologram. In other words, the electrons in the shallow traps should be used by the nonuniform part of the recording intensity to result in a hologram. There are three other mechanisms competing with this desirable mechanism, namely bleaching of the shallow traps by the dc part of the recording intensity pattern, depopulation of the shallow traps by the sensitizing beam, and erasure of the holograms in both traps by the sensitizing beam. The optimum intensity ratio results in the best balance between the desirable and undesirable mechanisms. The story is totally different for the singly-doped method. Due to the strong dark depopulation mechanisms, increasing recording and sensitizing beam intensities will both increase the saturation field as they both help earning more electrons for the desirable mechanism in competition with all undesirable mechanisms dominated by the dark depopulation processes. We can certainly get to a saturation and then decrease if we keep increasing the intensities, but that could happen only at impractically high intensities for the material assumed in these calculations.

Besides having much higher diffraction efficiencies and therefore the $M/\#$, two-center method has another major advantage over singly-doped method as suggested by Figure 4(c): there is no intensity threshold for the two-center method, but there is always one for the singly-doped method since the dark depopulation mechanisms are always there. It is now clear that the main problem of the singly-doped method is dark depopulation that can be rephrased as short lifetime of the shallow traps. A lot of efforts have been made to increase this lifetime to improve the performance, for example to decrease the intensity threshold. Improvement of this lifetime from milliseconds to seconds has been reported by using nominally pure stoichiometric crystals.¹⁹ Although this looks impressive, it suffers from the important drawback mentioned in the last section and evident from Figure 3. To record strong holograms and get large $M/\#$ we need a considerable concentration of the deep traps that by itself reduces the lifetime of the shallow traps dramatically. This dependence of the properties of the traps is a problem in optimization. In the two-center method, the concentration of the shallow and deep traps and the initial oxidation state of the material are all independent of each other and can be varied to improve the recording performance. This could be simply thought of as having more degrees of freedom in the two-center method which results in the logical conclusion of having better performance for the optimum case.

4. EXPERIMENTAL RESULTS

In this section, we present results from the experiments we did using two-center method and compare the performance parameters with those of the singly-doped method reported to date. Experiments were performed with a 0.85 mm thick LiNbO₃ crystal doped with 0.075 wt. % Fe₂O₃ and 0.01 wt. % MnO. We used a 100 W mercury lamp as the ultraviolet light source and a 35 mW HeNe laser for holographic recording. The unpolarized ultraviolet light illuminates the crystal homogeneously (wavelength 365 nm, intensity 20 mW/cm²) and the HeNe laser light is split into two plane waves which interfere at the crystal (wavelength 633 nm, $1/e^2$ beam diameter 2.0 mm, intensity of each wave averaged over the $1/e^2$ area 300 mW/cm², transmission geometry). The grating vector of the interference pattern is aligned along the c axis of the sample. 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. We first pre-expose the crystal to uv for 3 hours. Then the recording is performed by the two red beams with simultaneous uv exposure. For read-out, the uv beam and one of the red beams are blocked and the diffraction efficiency is measured by the other red beam.

The experimental result is depicted in Figure 5. The shape of the curve is exactly what we would expect theoretically from the two center model discussed in the last section. The obtained performance is exciting: We reach a non-volatile diffraction efficiency of 4 % for ordinarily polarized light and, due to a larger electrooptic coefficient, 32 % for extraordinary polarization. The square root of the saturation efficiency yields the $M/\#$ which is 0.2 for ordinary and 0.6 for extraordinary polarization which is very good for a 0.85 mm thick crystal. It should be noted here that the absorption coefficient of the crystal for ultraviolet light was close to 90 cm⁻¹ resulting in a lower effective thickness. Recently, Guenther et al. reported some nice experimental results for non-volatile singly-doped two-step method at low intensities. They reported $M/\# = 0.1$ for a 8 – 10 mm thick crystal at sensitizing intensity of 1 W/cm² total recording intensity of 600 mW/cm² with extraordinary polarization for the recording beams. This value can be raised to $M/\# = 1$ by increasing the total recording intensity to 10 W/cm². This is due to the almost

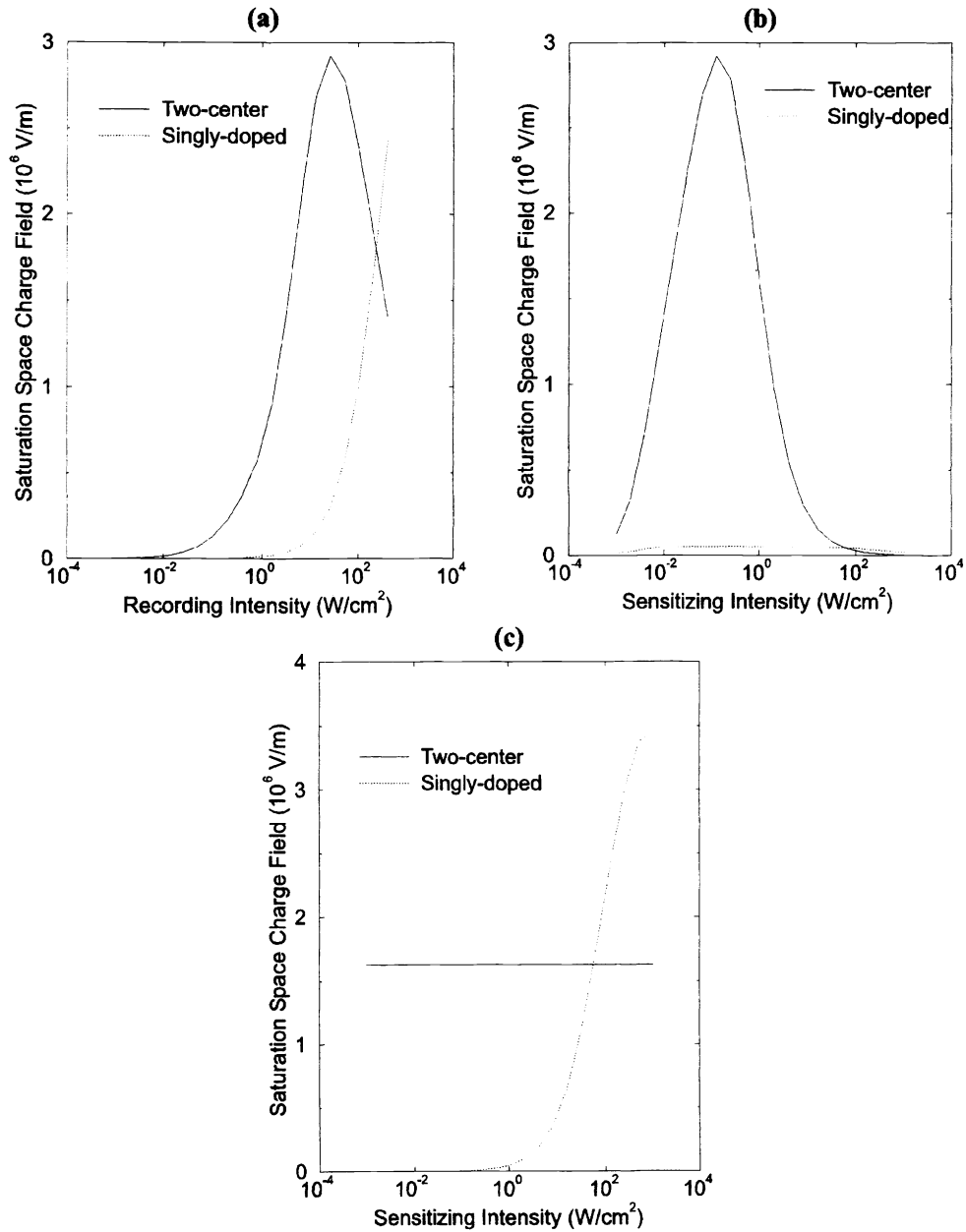


Figure 4. Effect of sensitizing and recording beam intensities. (a) Variation of the non-volatile saturation space charge field with total recording intensity when sensitizing beam has a fixed intensity of $I_s = 1 W/cm^2$, (b) variation of the non-volatile saturation space charge field with sensitizing intensity when each recording beam has a fixed intensity of $I_s = 2 W/cm^2$, and (c) variation of the non-volatile saturation space charge field with sensitizing intensity when the ratio of the total recording intensity to sensitizing intensity is fixed at 4.

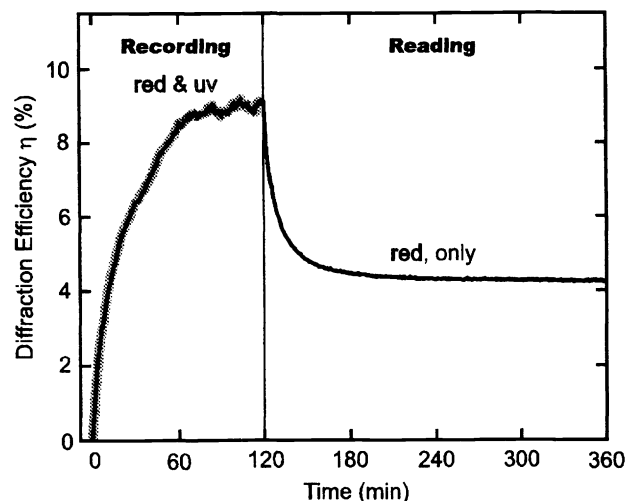


Figure 5. Holographic recording and read-out curves for two-center method. The diffraction efficiency η , i.e. the ratio between the intensity of the diffracted and of the total incident read-out light, is shown versus time. The crystal is pre-exposed with the ultraviolet light (wavelength 365 nm , intensity 20 mW/cm^2 , 120 minutes), then the hologram is recorded (wavelength 633 nm , ordinary polarization, intensity 600 mW/cm^2 , 120 minutes) and finally it is read by one of the recording beams (633 nm , intensity 300 mW/cm^2 , 240 minutes). The ultraviolet light is present during recording.

linear dependence of the $M/\#$ to the recording intensity of the singly-doped method as depicted in Fig. 4(a) and explained in the last section. Note that for a singly-doped 0.85 mm thick crystal with the same parameters, the $M/\#$ would be smaller by a factor of 10 due to the linear variation of the $M/\#$ with crystal thickness. We should note, however, that the doubly-doped crystal described here can not be used with thicknesses at the cm range because of the large band-to-band absorption at 365 nm (the wavelength of the sensitizing beam).

5. DISCUSSION

The results of the sections 3 and 4 simply suggest that the two-center method has better performance characteristics than the singly-doped method. It should be noted that the present two-center results are really unoptimized. The crystal has a large absorption coefficient at 365 nm . This results in the waste of some part of the crystal. This large absorption coefficient is partly due to band-to-band transitions which can be prevented by using longer sensitizing wavelengths. One example would be 404 nm line of the mercury lamp. The oxidation state of the crystal (the percentage of the initially ionized deep traps) is another important parameter to optimize. Highly oxidized crystals are not good for recording as they do not have enough electrons in the deep traps to begin with. On the other hand, all the deep traps are initially filled with electrons in highly reduced samples. Although strong initial holograms can be recorded in these samples due to transfer of some of these electrons to the shallow traps, final diffraction efficiency is negligible as those electrons finally come back to the deep traps to erase the existing hologram. As an example for potential improvement, we got $M/\#$ close to 0.3 in a crystal with the same specifications as that mentioned in section 4 with a better oxidation state and using only 4 mW/cm^2 of sensitizing intensity at 404 nm . By increasing the iron concentration to the highest practical limit and choosing optimum manganese concentration, further improvement by at least a factor of 2 is possible.

Another important fact to note is that the choice of iron for recording in red is not a good one due to small absorption coefficient and photovoltaic constant for the iron traps at this wavelength. Cerium would be a much better choice. Simple approximate calculations of the $M/\#$ with typical parameters for cerium show the possibility of improvement in the $M/\#$ by a factor of 20 which can be raised to about 50 if the optimum doping levels are used. By using longer sensitizing wavelengths, we hope to improve the performance further. For example, we can use

copper as the deep trap and raise the sensitizing wavelength to 430 nm. All these facts suggest that the two-center method has a lot of potential for improvement.

It should be noted that a lot of optimization effort has been made for the singly-doped scheme. The best recording wavelength has been used, different doping levels and degrees of oxidation have been tried, and both congruent and stoichiometric samples have been investigated, but the performance measures are not better than those of the non-optimum two-center method. We think this is mainly due to the dark depopulation processes and the dependence of the properties of the shallow and deep traps. Note that by going to very high cw intensities with congruent crystals, singly-doped samples will have better performance parameters due to very large polaron concentration which can not be achieved in the two-center method for the shallow traps. Note that in this regime, the dark depopulation mechanisms are not important compared to the much stronger desirable transitions.

6. CONCLUSIONS

We presented a comparison of different two-step holographic recording schemes using one general picture. The difference between the different schemes is mainly due to the energy levels and sources of the traps. The major problem in the conventional two-step recording in singly-doped crystals is the dark depopulation of the shallow traps which is overcome by using doubly-doped crystals. This results in a better performance along with the suppression of the intensity threshold. The $M/\#$ for the two-center method depends on the ratio of recording and sensitizing intensities and not on the absolute intensities when the absorption of sensitizing and recording beams are not very large. We also showed that there is a lot of room for improving the two-center method. We think that among different non-volatile holographic recording schemes two-center method offers the most promising performance.

ACKNOWLEDGMENTS

This research was supported by JPL, funded by DARPA/ITO, and by grants from Rome laboratories.

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