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## A Study on Batch Method of Thermal Fixing for Multiplexed Holographic Recordings

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### ABSTRACT

We have investigated the batch method of thermal fixing for multiplexed holograms. The batch fixing process is pictured and analyzed, for the first time to our knowledge. The batch procedure of thermal fixing includes optical erasure of electronic gratings by subsequent recordings both in the same batch and in different batches, ionic compensation during thermal fixing in one batch, smoothing of ionic gratings during thermal fixing of subsequent batches, and revealing of ionic gratings. The inter-batch optical erasure time constants of  $\text{LiNbO}_3\text{:Fe:Ce}$  crystals are measured in experiment.

### INTRODUCTION

Photorefractive crystals are commonly used as a recording medium in volume holographic storage. According to the charge excitation and transport mechanism, the electronic charges constitute holograms in photorefractive crystals to store some desired optical information. However, the above mentioned mechanism similarly causes optical erasure of holograms in further illumination process. Therefore, the recorded holographic gratings, i.e., the electronic gratings, are not permanently maintained in photorefractive crystals. In order to overcome such optical erasure to holograms on readout, thermal-fixing technique has been developed [1-4]. Photorefractive holograms can be stabilized against readout by the thermal fixing process of heating the crystal during or after the writing stage to higher temperature. Most investigations on thermal fixing consist of a single process of heating. Since holographic storage in photorefractive crystals is aimed at achieving high-capacity storage, it is more important to investigate the thermal fixing of multiplex holograms. An et al. recently have performed the thermal fixing of 10000 holograms in  $\text{Fe:LiNbO}_3$  [5]. However, the procedure of such thermal fixing has not been described well, and the method of thermal fixing for multiplex holograms is still to be further improved for getting higher after-fixing diffraction efficiency.

In this paper, the batch fixing procedure is described in detail by analyzing respective behaviors of both electronic and ionic gratings at every stage. The mechanism of slow optical erasure of electronic gratings by subsequent recordings in different batches and smoothing of ionic gratings during thermal fixing of subsequent batches for batch fixing multiplex holograms is, for the first time to our knowledge, further presented. Moreover, according to the diffraction efficiency measured in every stage of all the batches, the inter-batch optical erasure time constant and the dark decay time constant of electronic gratings at elevated temperature for co-doped lithium niobate crystals have been fitted out.

### THEORY

#### Schemes of single thermal fixing

The procedure of thermal fixing consists of three steps: recording, fixing and revealing. Firstly, electronic gratings, which replicate the exposing light patterns, are recorded in a crystal

via illumination by spatially varying intensity patterns. In a fixing process when ionic conductivity dominates electronic conductivity at elevated temperature, the ions in the crystal move to compensate the light-induced space-charge distribution and form an ionic grating replicating the electronic grating. At room temperature the ionic conductivity is much smaller than the photoconductivity of the electrons. Therefore, in a revealing process at room temperature, the electronic gratings are partially erased by noncoherent uniform light, and then a net space-charge field dominated by the ionic gratings is left. Since the ionic gratings are much more stable against further optical erasure at room temperature than the relevant electronic gratings, the fixed holograms can be preserved for longer time.

At present, there are commonly two kinds of schemes available for thermal fixing of photorefractive holograms, called post-compensation and simultaneous storage and fixing at elevated temperature, respectively. For multiple holograms, the optical erasure time constant of light-induced gratings is a finite value. The diffraction efficiency of each hologram in a large scale of multiplexing holographic storage is commonly very small, since subsequent writing processes can partially erase the existing holograms stored in a photorefractive crystal. The diffraction efficiency of the hologram after fixing is usually much smaller than that of the original hologram. Therefore, if multiple holograms are fixed by using a post-compensation scheme, that is, the crystal is heated only after recording all the desired holograms, their diffraction efficiency will become rather small. If fixed by using a simultaneous-compensation scheme, electronic gratings recorded are almost compensated by ions thermally activated simultaneously, and the effect of ionic pattern's shielding against electronic gratings weakens the optical erasure. Thus, in comparison with a post-compensation scheme, a simultaneous-compensation scheme is of benefit to higher diffraction efficiency after fixing. However, since the recording and revealing processes are carried out at different temperatures respectively, the fixed holograms suffer from distortion caused by thermal contraction and change of the refraction index of the material. Consequently, the variation in the Bragg angles of the fixed holograms, resulting from the above reasons, will bring the difficulty in reconstructing data pages completely for a practical multiplexing storage system. In multiplexed holographic storage, recording and reconstruction of fixed holograms should be performed at the same temperature, commonly at room temperature. Therefore, a batch-scheme of thermal fixing, combining the advantages of both above schemes, will be a good choice.

All the subsequent holograms are broken into several batches. Each batch is recorded at room temperature and then fixed at higher temperature, and finally all the batches are revealed in whole at room temperature. The batch scheme can avoid incomplete Bragg matching caused by thermal effect, because recording and readout both occur at room temperature. With many times heating, the diffraction efficiency of multiple holograms fixed in the batch scheme is able to increase effectively.

#### **Batch scheme of thermal fixing**

In the batch scheme of thermal fixing for multiplexed holograms, the multiplexing holograms to be recorded and fixed are divided into several sets, every set of holograms is subsequently recorded at room temperature and fixed at elevated temperature in order, respectively, up to having fixed all the sets of holograms. And then, after cooling down to room temperature, all the holograms of ionic gratings are revealed at the same time by illuminating them with noncoherent or uniform light.

All the  $N$  holograms are divided into  $S$  sets, each set including an equal number of holograms,  $N_h=N/S$ . Firstly, the  $N_h$  holograms in the first set are subsequently recorded in a

crystal, and then the crystal is heated in an oven so that all the  $N_h$  holograms are thermally fixed at the same time. In this process, the ions rapidly migrate at elevated temperature and form ionic gratings of compensating the first set of electronic gratings completely, which makes the net space-charge field become zero. After cooling down to the room temperature, the second set of  $N_h$  holograms are recorded in turn, which slowly erases the previous set of electronic gratings and thus partially reveals the respective ionic gratings. Since the migration rate of electrons is larger than that of ions ( $\mu_e \gg \mu_i$ ), the optical erasure of ionic gratings is negligible in this process. Next, after heating the crystal again for the same amount of time, we find that the revealed portion of the first set of ionic gratings has been smoothed out and the second set of electronic gratings compensated with migration of thermal activation ions. The procedure above-mentioned is repeated until all  $N$  holograms are recorded and fixed. Thus, in the recording process, each set of electronic gratings except the first set suffer optical erasure by subsequent recordings both in the same batch and in different batches, and then in the fixing process, ionic compensation to the same batch of electronic gratings and smoothing for previous different batches of ionic gratings revealed partially during thermal fixing in every batch are achieved. The electronic grating strength of set  $m$  ( $m=1,2,\dots,S$ ) can be expressed by its index modulation  $\Delta n_e^m$  as

$$\Delta n_e^m = \Delta n_0 e^{-\frac{\sum_{i=1}^{S-m} t_{m+i}}{\tau_f}} \quad (1)$$

where  $t_{m+i}$  is the total writing time of holograms in the batch  $m+i$ ,  $\tau_f$  is the optical erasure time constant of electronic gratings by recordings in subsequent batches, and  $\Delta n_0$  is the index modulation of electronic gratings in the first batch before thermal fixing.  $\Delta n_0$  can be taken the equal value for all the gratings in the first batch by using proper exposure procedure. In the respective process of optical erasure after ionic compensating, with partially revealing effect of writing beams in subsequent batches on ionic gratings, movable electrons light-excited can drift and diffuse under effect of ionic charge field, and then screen the ionic patterns. The screening effect of trapped electrons on ionic gratings can reduce optical erasing, in that  $\tau_f$  is larger than the optical erasure time constant  $\tau_E$  of holograms without fixing. The screening depth considerably depends on the concentration  $N_a$  of electrons trapped deeply, space-frequency  $K$  of holographic gratings and the strength of photovoltaic effect.

In every process of heating a crystal for fixing holograms of different batches, electrons and ions are thermally activated at elevated temperature, respectively. Electronic diffusion and drifting both occur to decline the grating strength. Meanwhile, since the ionic migration rate is bigger than the electronic one, ions rapidly move to compensate electronic gratings once again. As a result, the strengths of ionic gratings in previous batches further decrease with their respective electronic gratings. Electronic gratings will decay exponentially in dark case. The index modulation  $\Delta n_{ed}^m$  of the set  $m$  of electronic gratings, which experience the procedure of optical erasure and dark decay effect during  $(S-m)$  times, can be written as

$$\Delta n_{ed}^m = \Delta n_e^m e^{-\frac{(S-m)T}{\tau_r}} \quad (2)$$

where  $T$  is the time for heating a crystal to achieve thermal fixing in one batch,  $\tau_i$  is the dark decay time constant of electronic gratings at elevated temperature, relative to decay of grating strength in the dark case. The ionic grating strength is equal to that of electrons.

After recording and fixing according to the batch procedure, all the fixed holograms are revealed sufficiently. Finally, ionic holograms are obtained in batch fixing. Obviously, the last set of holograms experiences only one thermal fixing procedure of recording - compensation - revealing, but other sets also suffer the procedures of optical erasure and smoothing with different times. The time of optical erasure and smoothing for one batch of holograms depends on the number of batches recorded after it. Therefore, having accomplished the thermal fixing for  $S$  batches of holograms, the batch  $m$  experiences  $(S-m)$  procedures of optical erasure and smoothing and one procedure of thermal fixing with post-compensation.

We refer to a single thermal fixing as the procedure of only including recording, compensation and revealing steps, no matter how many multiplexed holograms to be recorded. In the revealing process of a single thermal fixing, ionic gratings screened in part by trapped electrons are unable to be fully revealed, even though they completely replicate electronic holograms in the fixing process. Therefore, the ionic holograms are partially readout after a sufficient revealing process. For a single thermal fixing, the thermal fixing efficiency  $\eta_F$  for multiplexed holograms is defined as

$$\eta_F = \frac{\eta_i}{\eta_e} \quad (3)$$

where  $\eta_i$  is the diffraction efficiency of revealed ionic gratings,  $\eta_e$  diffraction efficiency of electronic gratings before fixing. Although  $\eta_F$  is a complicated function of both the storage material and optical system [5], it can also be measured experimentally.

Since the index modulation is proportional to the square root of the diffraction efficiency in the small modulation case, according to equations (1)~(3), we can derive the index modulation  $\Delta n_i^m$  of revealed ionic holograms in the batch  $m$  as follow

$$\begin{aligned} \Delta n_i^m &= \sqrt{\eta_F} \Delta n_{el}^m \\ &= \sqrt{\eta_F} \Delta n_0 e^{-\left( \frac{\sum_{i=1}^{S-m} t_{m+i}}{\tau_i} + \frac{(S-m)T}{\tau_F} \right)} \end{aligned} \quad (4)$$

The time constants  $\tau_i$  and  $\tau_F$  are the complex functions of parameters of photorefractive material. Although time constants  $\tau_F$  can be calculated according to electronic decay in the dark phase theoretically [3], but it is not practical to calculate  $\tau_F$  for the respective crystal in theory. On other hands, the time constant  $\tau_F$  is an important parameter in calculating exposure schedule of multiplexed holograms to equalize their diffraction efficiencies. Therefore, it is necessary to measure time constants  $\tau_F$  for different crystal samples experimentally.

## EXPERIMENT

The holograms were recorded with two ordinarily polarized beams with equal-intensity in reflection geometry. In the experiment, all 25 holographic gratings are divided into 5 batches, each batch including 5 gratings. Firstly, one batch of gratings was recorded at room temperature by use of angular multiplexing. After recording, the writing beams were blocked and the sample with one batch of gratings was heated in the dark to  $\sim 150^\circ\text{C}$  to cause fast ionic transport, and soaked for about 20 min. to ensure that these electronic gratings were compensated by ions completely. Next, the sample was cooled to room temperature and the second batch of 5 holographic gratings was recorded in it. Then, the sample stored two batches of holographic gratings are heated to accomplish ionic compensation. Such procedure of recording and heating was repeated for all 5 batches of holographic gratings. Finally, ionic holograms were revealed with a non-coherent erasing beam.

In each recording stage, besides optical erasure inside one batch, recording of the subsequent batch of gratings slowly erased the previous gratings fixed, which results in a little revealing of ionic gratings. The smaller diffraction efficiency generated by the revealed portion of ionic gratings was observed in the experiment. We refer to this kind of optical erasure in different batches as inter-batch optical erasure, which have a longer erasure constant time than one in multiplexed recording without thermal fixing. During each heating process, in addition to compensating the present batch of electronic gratings, fast ionic transport also effaces the revealed portion of ionic gratings in previous batches. Therefore, there was no diffraction to be observed at the end of each heating stage of the experiment. As taken dark decay time constant  $\tau_T = 10^{14}\text{s}$ , the inter-batch optical erasure time constant  $\tau_F$  of electronic gratings was fitted out according to the diffraction efficiency measured in every stage of all the batches (see figure 1). The inter-batch erasure time constants  $\tau_F$  of batch thermal fixing for co-doped crystal samples, with different concentrations and treatments, were listed in Table I.

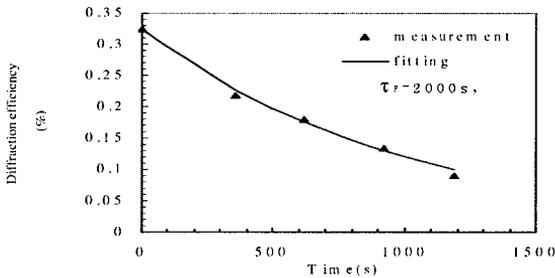


Figure 1. A fitted curve of the inter-batch optical erasure time constant  $\tau_F$  for a Fe:Ce:LiNbO<sub>3</sub> crystal.

**Table I.** Comparison of ordinary erasure time constant and inter-batch erasure time constant (the wavelength  $\lambda=0.633\mu\text{m}$ , the total intensity  $I_0=37\text{ mw/cm}^2$ , dark decay time constant  $\tau_T=10^{14}\text{s}$ ).

No.	Concentration (%)	O/R state	ordinary erasure time constant $\tau_E$ (s)	Inter-batch erasure time constant $\tau_F$ (s)
FC1	Fe:0.03 Ce:0.05	1050°C oxidation, 20 hours	3000	14000
D2B	Fe:0.03 Ce:0.05	As grown	1900	4300
FC2	Fe:0.07 Ce:0.20	1050°C oxidation, 20 hours	1500	4500
D3B	Fe:0.07 Ce:0.20	As grown	455	2000

The experimental results indicate that the inter-batch optical erasure time constant  $\tau_F$  is indeed much longer than the ordinary erasure time constant  $\tau_E$ . The result demonstrates that, since the migration of light-induced electrons is hampered by corresponding ionic gratings during inter-batch erasure, the optical erasure becomes much slower in the case of screening effect of ionic gratings on electronic ones.

## CONCLUSIONS

The work investigated the batch method of thermal fixing for multiplexed holograms, in which the behavior of both electronic and ionic gratings is discussed in detail. There is a good agreement between the experimental result on inter-batch optical erasure time constant and the theoretical prediction. The result shows that the hampering effect of ionic gratings on trapped electrons reduces optical erasing in batch procedures, and hence, enhances the diffraction efficiency of fixed multiplexed holograms. This optical erasure time constant is required for designing the exposure schedule for large scale multiplexed storage to achieve equal diffraction efficiency holograms.

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