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# The influence of spatial frequency in partial spatial erasure of holographic diffraction gratings within LiNbO<sub>3</sub>:Fe

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#### ABSTRACT

The effect of spatial frequency on partial spatial erasure of a hologram within a photorefractive crystal, lightly doped LiNbO3, was studied by capturing its reconstructed image on a digital camera at times varying from 40 s to 420 min after storage. The image's constituent spatial frequencies (6.2–62.3/cm) at these times were studied. Using a capacitor discharge model, lower spatial frequencies (6.2–41.6/cm) discharge exponentially after 180 min, the maximum space-charge field was found to occur for a median spatial frequency of 34.6/cm. Higher spatial frequencies charge continuously. Features oscillating at  $\sim\!2\,\mu\text{m/s}$  were superimposed on all frequencies  $>\!27.7/\text{cm}$ , evolving quadratically to a maximum amplitude at the spatial frequency of 48.5/cm before decaying. It is thought that the refractive-index field stored within the photorefractive crystal caused focussing of the read-out beam leading to the generation of a spatial soliton.

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#### 1. Introduction

#### 1.1. Holographic recording in photorefractive crystals

Interest in volume holography is not limited to the realisation of holographic memories [1] but also advances in a massive range of other applications such as interconnections, neural networks [2–4] and information processing systems. Volume holography technology has the potential to be the technological basis for alloptical devices capable of operating with wavelength division multiplexing networks such as optical attenuators and wavelength filters [5].

The principle of optical storage of data is based on a discovery in the crystal lithium niobate (LiNbO $_3$ ) in 1966, the photorefractive effect [6]. Here non-homogeneous illumination of LiNbO $_3$  causes spatial fringe patterns and charge re-distribution due to the migration of electrons from bright regions to dark regions. Charge distribution is no longer homogeneous; internal spacecharge fields are set up, which change the refractive index in sympathy with local illumination due to the linear electro-optic effect. In the simplest case the resulting interference pattern given by interfering signal and reference plane waves  $E_s$  and  $E_p$  respectively, is [4]

$$I(r) = I_0[1 + m\cos(Kr)] \tag{1}$$

where K is the spatial frequency of the resulting interference fringes; the depth of modulation  $m = 2(|E_1||E_2|)/(|E_1|^2|E_2|^2)$  and  $I_0$  is the mean intensity.

Many other photorefractive materials have since been discovered, and there is a wealth of information in the literature about the effect of light on photorefractive materials. In contrast, there is little scientific work in the literature concerning the effect photorefractive materials have on the incident light itself. This work describes such an effect caused when a 45° cut iron doped LiNbO<sub>3</sub> (LiNbO<sub>3</sub>:Fe) crystal was illuminated by a vertically polarised argon (Ar)-ion laser at 514 nm.

For data storage, the illumination incident on LiNbO<sub>3</sub>:Fe is the interference pattern of an object beam (from an object/information to be stored) with its reference wave. Since the object/information is complex, the illumination is a complex light pattern consisting of many spatial frequencies. This is stored in permanent form as a space-charge field hologram within the crystal. When the stored object is reconstructed (read) using a homogeneous reference beam the hologram acts as a diffraction grating with different spatial frequencies superimposed, stored as a refractive-index grating, to reproduce an image of the original object.

The amplitude of the refractive-index grating for ordinary polarised light is

$$\Delta n_{\rm s} = -\frac{1}{2} n_{\rm o}^3 r_{13} E_{\rm sc} \tag{2}$$

where  $n_0$  is the refractive index,  $r_{13}$  is one of the electro-optic coefficients and  $E_{\rm sc}$  the space-charge field set up by the inhomogeneous charge distribution.

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The spatial frequencies, K, are related to their grating spacing,  $\Lambda$ , by

$$K = 1/\Lambda \tag{3}$$

The reconstruction beam, being homogeneous, acts to return the charge distribution to uniformity again, eventually erasing the hologram. Thus, partial spatial erasure (volatile storage) is the main constraint in developing photorefractive materials into mass storage media.

Some techniques have been put forward for 'fixing' holograms, including doubly doped crystals, ferroelectric, dual-wavelength method, electrical, thermal fixing [7–11] and more recently triply doped crystals [12]. The charges which are re-distributed come from impurities or traps within the crystal lattice called photorefractive centres. These are located in the energy band gap between valence and conduction bands. In this paper LiNbO<sub>3</sub>:Fe at a dopant concentration of 0.03% was studied using Ar-ion laser beams of power density 400 mW/cm<sup>2</sup>.

#### 1.2. Charge transport model

LiNbO<sub>3</sub> has been experimentally shown [13–17] to behave as a one-centre charge transport model [18] at low light intensities.

Iron is a deep centre; hence thermal excitations are negligible compared to light, which excites electrons from the  $Fe^{2+}$  into the conduction band, where they migrate, driven mainly by the photovoltaic effect, in a preferred direction. They are trapped elsewhere by  $Fe^{3+}$  sites. This process can be considered to cause the formation of a capacitor, where electrons are charged from the photorefractive centres to the conduction band, eventually returning to the impurity levels with a characteristic time constant,  $\tau_e$ .

At large light intensities (>200 W/cm²) [19,20] the one-centre model is insufficient for description of charge transport in LiNbO<sub>3</sub>:Fe, and it has been described by the two-centre model with direct transitions[13,21–24]. The additional photorefractive centre is the shallow centre identified as the intrinsic defect [21] Nb<sup>4+</sup> on the Li<sup>+</sup> site. At least 1 mol% of this intrinsic defect is present in congruent LiNbO<sub>3</sub> and direct excitations of electrons from Fe<sup>2+</sup> into Nb<sup>5+</sup>, forming Nb<sup>4+</sup>, are possible [25] and grating erasures occur with two distinct time constants [13]. The one-centre model applies in this work, since laser intensities used were low.

Reasons for volatile storage suggested in the literature are: (i) Inhomogeneity of illumination of the crystal during recording. Here the excitation of electrons from  $Fe^{2+}$ -populated centres to  $Fe^{3+}$ -unpopulated centres through the conduction band [9] can be considered to cause the formation of a capacitor. Such a 'capacitor' will decay quicker if it is charged by low-intensity light (partially charged) compared to higher-intensity light (fully charged). The effect of this is to cause different parts of the hologram to fade out in different time periods. (ii) Spatial inhomogeneity of the crystal doping. This is related to erasure time since its time constant,  $\tau_e$ , is inversely proportional to photoconductivity [26]:

$$\tau_{\rm e} \propto \frac{c_{\rm Fe^{3+}}}{c_{\rm Fe^{2+}}} \tag{4}$$

This is proportional to the reciprocal oxidation/reduction state of the crystal and may cause fluctuation in photorefractive centres throughout the crystal.

In previous work [27] the decay time of partial spatial erasure of holograms was found to be dependent on location within the stored image. In this work reconstructed images of objects stored in the crystal were filtered into 10 spatial frequency intervals using an annular shaped band-pass filter. The intensities of all pixels within each interval were integrated to obtain the total

contribution of each of 10 spatial frequency intervals, within the range 6.3–69.3/cm, to the Fourier transform. These intensities, *I*, are proportional to the number of charged 'capacitors' in the hologram.

#### 2. Theory

Although no absorption is caused by Fe<sup>3+</sup> ions, its concentration should be large enough so that any kind of trap limitation is avoided and dominant photovoltaic charge transport is ensured. This is described by [26]

$$c_{\mathsf{F}\mathsf{e}^{3+}} \gg c_{\mathsf{F}\mathsf{e}^{2+}} \tag{5}$$

The space-charge field can be simplified to

$$E_{\rm sc} = -\frac{e}{\varepsilon \varepsilon_{\rm o} K} c_{\rm Fe^{2+}} \tag{6}$$

Thus, from Eq. (2),

$$\Delta n_{\rm s} \propto \frac{e}{\varepsilon \varepsilon_{\rm o} K} c_{\rm Fe^{2+}} \tag{7}$$

where K is the spatial frequency and  $c_{\mathrm{Fe}^{2+}}$  is the density of donor sites or electrons available for re-distribution. Thus, the space-charge field is limited by  $c_{\mathrm{Fe}^{2+}}$ .

Despite Eq. (7), a study of the saturation values of refractive index changes of highly doped LiNbO<sub>3</sub> crystals [26] reported that  $\Delta n_s$  values are independent of K between 1.5 and  $14\,\mu\text{m}^{-1}$  at  $\lambda = 514\,\text{nm}$ , indicating that saturation of refractive index changes for  $c_{\text{Fe}^{3+}}$  at concentrations above 0.06 wt% Fe<sub>2</sub>O<sub>3</sub> cannot be attributed to trap limitations; Fe<sup>2+</sup> concentration is too large; so erasure takes place over prolonged time scales as indicated by Eq. (4).

The following relations are also valid for LiNbO<sub>3</sub>,

$$\Delta n_{\rm s} \propto c_{{\rm Fe}^{3+}}$$
 (8)

$$j_{\rm phv} \propto Ic_{\rm Fe^{2+}}$$
 (9)

$$\sigma_{\rm ph} \propto I(c_{\rm Fe^{2+}}/c_{\rm Fe^{3+}}) \tag{10}$$

where  $\Delta n_s$ ,  $j_{phv}$  and  $\sigma_{ph}$  are the refractive-index change, bulk photovoltaic current density and photoconductivity, respectively.

#### 3. Experimental

In this work the Ar-ion laser beam at 514.5 nm was split into two s-polarised beams, an object beam and a reference beam of power  ${\sim}400\,\text{mW/cm}^2.$  Holograms were stored on a 45° cut 0.03 mol% (0.032 wt%) iron (Fe<sub>2</sub>O<sub>3</sub>) doped LiNbO<sub>3</sub> crystal in a 90° two beam coupling configuration [27]. The experimental set-up is shown in Fig. 1.

The image of an extended shape (plus sign '+') was stored in the crystal. After the hologram was stored for 5 min, the object beam was blocked and the stored image was reconstructed by the reference beam on reflection from the hologram and recorded at fixed times on a digital camera.

The times at which photographs were taken ranged from 40 s to 420 min after storage. The data were transferred to a computer and stored as  $(2500 \times 1720 \times 3)$  true-colour unsigned integer (uint16) data files, which were then converted to  $(2500 \times 1720)$  grey-scale double precision data files.

The spatial frequencies were determined [28] to vary between 0 at the centre to  $\sim$ 69/cm at the outer edge of the diffraction pattern. The schematic diagram of Fig. 2 indicates the measurements made.

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