

# Theoretical study on lifetime of thermal-fixed volume holographic phase grating in photorefractive crystals

Yansheng Song (宋艳生)<sup>1\*</sup>, Jiarong Ji (季家榕)<sup>1</sup>, and Wenhua Dou (窦文华)<sup>2</sup>

<sup>1</sup>College of Opto-Electronic Science and Engineering, National University of Defense Technology, Changsha 410073, China

<sup>2</sup>College of Computer, National University of Defense Technology, Changsha 410073, China

\*E-mail: yansheng.song@gmail.com

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We review the dark decay of the electronic holographic phase grating before thermal fixing, and deduce the general analytic expression of the lifetime of thermal-fixed ionic holograms in the photorefractive crystal, by means of analogizing. Because the ions are optically inactive, the lifetime of thermal-fixed ionic holograms is only closely relate to the ionic decay rate which is determined by the conductivity of ionic species at a given temperature. We theoretically analyze and numerically simulate the influences on the lifetime of ionic grating from the crucial factors in the experiment and application. The results reveal that low temperature, low ion-concentration, and large grating spacing are advantages for extending the life of the thermal-fixed volume holographic phase grating in photorefractive crystal.

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Photorefractive crystals have been widely used for optical phase conjugation, optical image processing, volume holographic data storage (VHDS), etc.<sup>[1]</sup> By far, photorefractive crystals are the most efficient media for recording dynamic/static holograms<sup>[2]</sup>. In these media, information can be stored, erased, and retrieved by the illumination of light. In recent years, the researches on the applications of the photorefractive materials have been focused on VHDS. VHDS is becoming competitive due to its large storage capacity and fast access rate.

The index gratings and holograms can decay in dark and also can be erased by illumination with a plane wave. The ability to fix gratings with high efficiencies and long lifetimes is extraordinarily important for the applications of holographic data storage and for the fabrication of thick holographic optical elements. In 1971, Amodei *et al.* discovered that optically stored holograms in LiNbO<sub>3</sub> crystal could be stabilized against readout by heating the crystal to above 100 °C either during or after the storage process and illuminating it at room temperature subsequently<sup>[3]</sup>. That is called thermal fixing by which the information stored in the electronic space-charge distribution is compensated at elevated temperatures in a nonphotoactive ionic-charge distribution. Since then, the fixing and developing grating processes of doped LiNbO<sub>3</sub><sup>[4–10]</sup> and other photorefractive materials such as Sr<sub>0.75</sub>Ba<sub>0.25</sub>Nb<sub>2</sub>O<sub>6</sub><sup>[11]</sup>, BaTiO<sub>3</sub><sup>[12]</sup>, and KTa<sub>1-x</sub>Nb<sub>x</sub>O<sub>3</sub><sup>[13,14]</sup> have been extensively studied. The experimental results reveal that the lifetime of the holographic gratings stored in photorefractive crystals are prolonged by the thermal fixing<sup>[6,15]</sup>. However, there is no overall knowledge on the lifetime of the fixed ionic holograms. A general theoretical model and expression need to be established. On the analogy of electronic grating's decay, we show the analytic expression of the lifetime of thermal-fixed ionic holograms and discuss the factors that may have influences on it.

A typical thermal fixing is a three-step process with an ordinarily polarized recording light: recording at room

temperature, compensating at elevated temperature, and developing at room temperature. The evolution of a hologram from recording (phase 0) to having been fixed (phase IV) is sketched in Fig. 1<sup>[15]</sup>. What we are interested in is the phase IV that directly relates to the lifetime of the hologram in actual applications.

Electronic hologram decay contains two faces: the dark decay and the light erasing<sup>[16]</sup>. The decay rate can be expressed as

$$\Gamma_e = \Gamma_{e\text{-dark}} + \Gamma_{e\text{-light}}, \tag{1}$$

where  $\Gamma_{e\text{-dark}}$  is the electronic hologram decay rate;  $\Gamma_{e\text{-light}}$  is electron holographic grating decay rate induced by light erasing.

But the compensating-secondary carriers, i.e., ions, were confirmed to be optically inactive<sup>[14]</sup>. Yariv *et al.* concluded that ion transport is the mechanism that is responsible both for the fixed ionic hologram decay in the dark and for the erasure during the readout<sup>[15]</sup>. So the ionic hologram decay rate can be expressed as

$$\Gamma_i = \Gamma_{i\text{-dark}}, \tag{2}$$

where  $\Gamma_{i\text{-dark}}$  is the ionic hologram grating decay rate in dark.

The electronic hologram (before thermal-fixed) dark

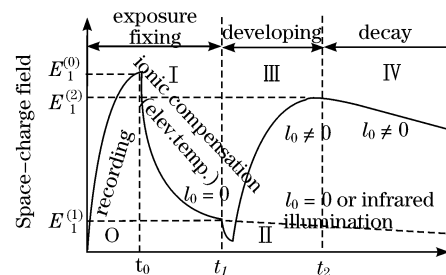


Fig. 1. Typical life evolution of a hologram in a photorefractive material. The Roman numerals are the phase numbers<sup>[14]</sup>.  $I_0$  is the intensity of illuminating light.

decay rate in BaTiO<sub>3</sub> is given as<sup>[16–17]</sup>

$$\Gamma_{e\text{-dark}} \propto f(K_g) \exp(-T_0/T), \quad (3)$$

where  $f(K_g) = 1 + \frac{K_g^2}{K_0^2}$ ,  $K_0$  is the characteristic vector and  $K_0^2 = \frac{n_e e^2}{\varepsilon \varepsilon_0 k_B T}$ ,  $K_g$  is the magnitude of the grating wave vector,  $n_e$  is the concentration of electrons,  $\varepsilon_0$  is the permittivity of free space,  $\varepsilon$  is the direct current (DC) dielectric constant along the direction of the grating wave vector, and  $T_0 \sim 10^4$  in the BaTiO<sub>3</sub>. The factor  $f(K_g)$  mainly describes the grating state and the exponential factor expresses the temperature dependence. Expression (3) was derived by Feinberg *et al.*<sup>[15,16]</sup> from experiments in BaTiO<sub>3</sub>, but it reveals the essence of electronic dark decay resulting from the electronic dark conductivity. The phenomena of photorefractive effect and electronic dark decay do not only exist in BaTiO<sub>3</sub>. There are just some factors such as  $n_e$ ,  $\varepsilon$ , and  $T_0$  depending on the crystal.

The essence of ionic conductivity-induced decay to the ionic grating is the same as the electronic conductivity-induced dark decay to the electronic grating. The dark conductivity is the reason for the dark decay. Similarly, the ionic hologram decay rate is determined by the conductivity of ionic species at a given temperature. We speculate that the expression of ionic grating dark decay rate should have a form as same as the electronic grating decay rate. So we deduce the expression of the ionic hologram dark decay rate as

$$\Gamma_{i\text{-dark}} = C f(K_g) \frac{\sigma_{\text{ion}}}{\varepsilon \varepsilon_0}, \quad (4)$$

where  $C$  is a constant. Ionic conductivity is

$$\sigma_{\text{ion}} = n_i e \mu_i, \quad (5)$$

where  $n_i$  is the concentration of ions,  $e$  is electronic charge, and  $\mu_i$  is the ionic mobility. Ionic charges migrate by means of hopping between adjacent sites, and the mobility is<sup>[18]</sup>

$$\mu_i = \frac{e}{k_B T} v_0 d^2 \exp\left(-\frac{E_a}{k_B T}\right), \quad (6)$$

where  $k_B$  is the Boltzmann constant,  $T$  is the temperature,  $v_0$  is vibration frequency,  $d$  is the distance of each step when ion is moving, and  $E_a$  is the activation energy. From Eqs. (2), (4), (5), and (6), we can get the decay time (lifetime) of the ionic gratings that exhibit Arrhenius behavior:

$$\tau = \frac{1}{\Gamma_i} = \frac{C_1}{v_0 d^2} \frac{1}{n_i e^2} \frac{1}{\varepsilon \varepsilon_0 k_B T + K_g^2} \exp\left(\frac{E_a}{k_B T}\right), \quad (7)$$

where  $C_1$  is a constant, the factors  $v_0$ ,  $d$ ,  $E_a$ , and  $n_i$  are the instinct characteristic factors of the photorefractive crystal, which have different values in different crystals; while  $T$  and  $K_g$  are external applied ones. We assign  $v_0 = 10^{12}$  Hz<sup>[18]</sup>,  $d = 3 \times 10^{-10}$  m,  $K_g = 2 \times 10^7$  m<sup>-1</sup>,  $E_a = 1.2$  eV<sup>[15]</sup> theoretically and have the curves of lifetime versus absolute temperature for a thermal-fixed hologram in

photorefractive crystal containing different ion concentrations. The three different lines in Fig. 2 correspond to three different ion concentration values, and those in Fig. 3 correspond to three different temperature values. The lifetime decreases quickly along with the increasing temperature. The dependence of the lifetime on the temperature is found to be closely related to the ion concentration. The phenomenon takes place because of the ionic conductivity increasing evidently at elevated temperature and being determined directly by the ion concentration. These results are consistent with the published ones<sup>[15,19]</sup>.

Supposing the volume holographic grating spacing is  $\Lambda$  and the lifetime is  $\tau$ , we show the curves of  $\tau - \Lambda$  at different temperatures in Fig. 4 for  $K_g = \frac{2\pi}{\Lambda}$ . The speed of ionic conductivity induced decay is slower for larger grating spacing than for smaller one. These results are consistent with the published ones<sup>[20]</sup>. So we can enlarge holographic grating spacing to increase the storage lifetime. Larger grating spacing requires larger recording wavelength  $\lambda$  and smaller incidence angle  $\theta$  according to the Bragg condition (*i.e.*,  $\Lambda = \lambda / (2n \sin \theta)$ ), where  $n$  is the diffraction index of the crystal). However, smaller recording wavelength has the advantage of higher recording efficiency. Thus, we can reduce the crossing angle of writing beams but not change the wavelength to approach the aim of long storage time.

We have no consideration of the reading erasure effect, because the ions usually are optically inactive when the reading beam is of a low intensity. The lifetime of a thermal-fixed ionic grating is mainly determined by the conductivity induced decay. The case that the fixed grating is illuminated by high intensity light is out of our consideration.

From Eqs. (4) and (5) we can discover that, in the limit

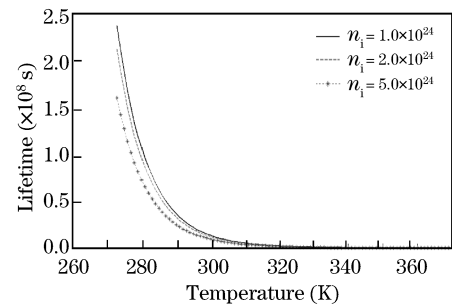


Fig. 2. Lifetime versus temperature for a thermal-fixed hologram in photorefractive crystal containing different ion concentrations.

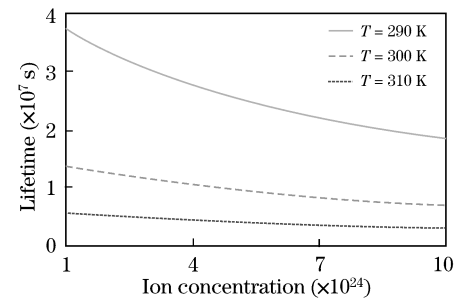


Fig. 3. Lifetime versus ion concentration for a thermal-fixed hologram in photorefractive crystal at different temperatures.

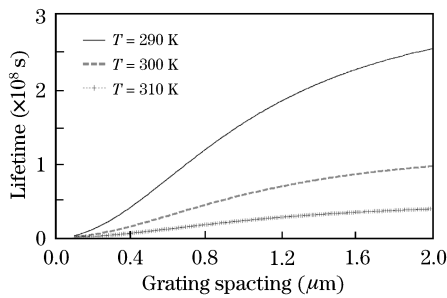


Fig. 4. Lifetime versus grating spacing for a thermal-fixed hologram in photorefractive crystal at different temperatures.

$K_g \rightarrow 0$ , the ionic decay rate  $\Gamma_i$  approaches the dielectric relaxation frequency for conducting ions:  $\omega_i = \frac{n_i \mu_i \epsilon}{\epsilon \epsilon_0}$ .

Ionic grating developed (or revealed; Fig. 1, phase I) by uniform incoherent and non-Bragg-matched erasing beam can avoid the decay during this phase. Uniform incoherent light with the single wavelength energy less than  $E_a$  cannot activate the ions. The non-Bragg-matched beam affects the crystal but has no influences on  $K_g$ , which means that the beam has no influence on the periods of the ionic grating.

Equation (7) reveals that a substantial reduction of ion density can increase the available storage time. While the maximum space-charge field  $E_{sc}$  that can be reached depends on the effective number of electron traps in the crystal. When ion density is reduced, as well as  $E_{sc}$ , the refractive index modulation induced by electro-optical effects such as Pockels effect and Kerr effect is reduced. Subsequently, the diffraction efficiency is reduced. We have to find a balanced point between the long storage lifetime and the high diffraction efficiency in application to deal with the contradiction.

In conclusion, efficient thermal fixing is based on the great disparity between the dark electronic conductivity at elevated temperatures and the ionic conductivity. At elevated temperatures the ionic conductivity is dominant, and ions readily compensate for the holographic electric field pattern created by photo-excited electrons by mimicking their spatial distribution. At low temperatures the ionic conductivity is low, permitting the quasi-permanent storage of the initial electronic-hologram ionic replica. Because ions are optically inactive, the residual ionic conductivity at low temperature determines the lifetime of the fixed hologram. Only under certain and restricted conditions can the decay time be acceptably

and available long.

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