Nonvolatile Holographic Storage with Two-Step Recording in Lithium Niobate using cw Lasers

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We have demonstrated nonvolatile holographic storage with two-photon, two-step recording in nominally pure lithium niobate. An energy level slightly below the conduction band with lifetime 10–100 ms was found to be the intermediate state in the two-step electron ionization. The long lifetime allowed the use of cw lasers with moderate intensity at near infrared wavelengths. Nonvolatile readout of volume holographic images stored at near infrared wavelength was demonstrated. [S0031-9007(97)02927-X]

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Holographic data storage holds forth the promise of very high storage densities and parallel access capabilities. In a holographic recording system, a coherent light source is split into a reference beam and an object beam that is spatially modulated by a two-dimensional grid (or data source) containing the information to be recorded. These two beams are then directed onto a single region of a holographic recording medium where they interact to generate an interference pattern. The recording medium is made from a light sensitive material that records the resulting interference pattern, and thereby stores the information provided on the grid. A virtual image of the grid can be constructed at a later time from the recorded interference pattern by illuminating the medium with a read beam that has the same characteristics as the reference beam. Because of the restriction of the Bragg condition, many images can be independently recorded at slightly different wavelengths and crossing angles between the object and reference beams, which are commonly referred to as wavelength multiplexing and angle multiplexing. High storage densities are thus achieved.

Photorefractive materials, especially single domain ferroelectric crystals such as lithium niobate, have been investigated extensively as potential holographic recording media [1]. In such materials, incident optical radiation excites electrons from some partially filled, multivalent "deep traps" near the middle of the band gap to the conduction band where they migrate from the illuminated areas to the dark areas (for simplicity, we ignore the effects from holes). The electrons are then captured by the empty traps, forming a spatial density distribution that mimics the optical pattern. After the optical pattern is removed, the electrons remain trapped in the same locations that they assumed during the illumination. Thus the photorefractive material can record an optical interference pattern in the form of a spatial distribution of electrons. Through the linear electro-optical effect this electron distribution causes a corresponding spatial distribution in the refractive index that can diffract the reference beam to construct the virtual image.

One of the most attractive features of the ferroelectrics as storage media is their recording reversibility; i.e., they can be erased and recorded over and over again. On the other hand, this reversibility also makes the media volatile—they can be easily erased during the process of reading the recorded interference pattern. During the read out, the read beam causes the electrons in the recording media to reenter the conduction band where they diffuse to a uniform distribution, thereby erasing the recorded interference pattern.

In an effort to overcome this problem, it has been proposed to use a "two-photon," or "two-step," recording procedure, so named because it requires two photons to excite an electron to the conduction band [2]. It has been shown that two-photon recording can be accomplished by illuminating the photorefractive material with a "gating" beam at one wavelength and writing beams at a second wavelength (Fig. 1). The two wavelengths are chosen such that photons at the writing wavelength have insufficient energy by themselves to promote electrons to the conduction band. However, the sum of the photon energies for the two wavelengths is sufficient to promote electrons to the conduction band. In application, a first photon excites an electron to an intermediate state close to the conduction band. Then while the electron is temporarily residing in the intermediate state, a second photon (typically of a different wavelength) promotes it to the conduction band where it migrates and becomes trapped

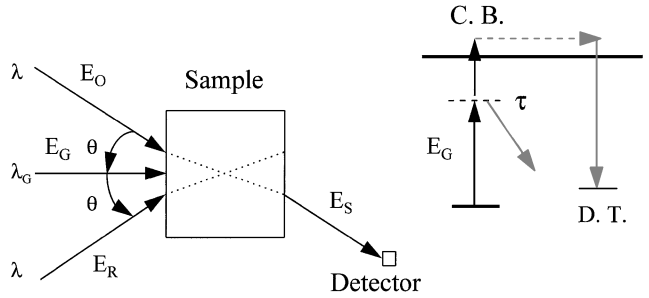

FIG. 1. Two-step holographic recording. C.B. and D.T. denote conduction band and deep trap. E_O, E_R, and E_G denote object, reference, and gating beams, respectively. In reading the hologram, only the reference beam is on. E_S denotes the diffracted beam.

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to record the interference pattern as described above. The recorded information can be read by a single read beam, the same as that in the conventional "one-photon" case. Because the photons from the read beam have insufficient energy to promote electrons to the conduction band, the reading process causes very little erasure. However, the recording reversibility remains because the media can still be erased using the gating beam (or gating beam plus reference beam) when it is desired.

In the early work [2,3] on the two-photon recording, the intermediate states used were either the "virtual states" formed by high intensity laser fields or the lower levels of transition metal dopants. Because of the short intermediate state lifetime, the resulting recording systems require expensive high intensity mode-locked or Q-switched lasers. More recently, two-step recording via some unidentified "shallow traps" using Q-switched pulsed lasers was also demonstrated [4]. In all of these cases, lasers with peak intensity >1 MW/cm^2 were used. The cost and the physical size of the high intensity lasers present a big obstacle to the realization of a commercially viable, nonvolatile read/write holographic recording system.

Like many other optical devices, it is of very high practical importance for a holographic recording system to incorporate inexpensive semiconductor diode lasers in its design. With currently available diode lasers, the obtainable intensity is about 1 to 10 W/cm^2 in the near infrared spectral region. Using the analysis in Ref. [2], one can estimate that the required intermediate state lifetime is $\sim$10 to 100 ms for efficient two-step recording in lithium niobate. However, dopants having such a long radiative lifetime are rarely known. Those that do may not have a large enough absorption cross section to produce a sufficient number of free electrons.

In the course of our research, we found that there are some energy levels with lifetimes $\sim$10 ms naturally existing just below the conduction band in many ferroelectrics, and that these levels can be used as the intermediate states for the two-photon recording using low intensity cw lasers. These levels can be accessed through the tail of the band edge absorption, and thus can provide sufficient absorption at properly selected gating wavelengths. In the second step excitation, efficient holograms can be written at red and near infrared wavelengths, which fall right in the diode laser wavelength range. Because the gating beam generally does not have to be from a coherent light source, a nonvolatile read/write holographic recording system based on low cost diode lasers and incoherent lamps are within reach.

We carried out two-step recording measurements in several single domain ferroelectrics, including lithium niobate, lithium tantalate, barium strontium niobate, and barium titanate. Since lithium niobate was found to give the best overall performance, the results from a pure (undoped) lithium niobate sample are presented.

A $5 \times 5 \times 5$ mm^3 nominally pure single domain lithium niobate crystal was used in the experiment. The direction of the macroscopic electric dipole is, by definition, denoted to be positive c axis. As is generally true in most ferroelectrics, light modulations along the c axis tend to give the largest change in the refractive index. A cw Ti:sapphire laser pumped with an argon ion laser was used for writing and reading the holograms. The Ti:sapphire laser was continuously tunable from 690 nm to 1 μm. In writing the holograms, the laser was split into two beams, an object beam and a reference beam, by a beam splitter ($R \cong 40\%$) and then focused to the crystal with a crossing angle of $\sim 5°$. The laser beams were polarized approximately along the c axis of the crystal. For two-color, two-step gated recording, a small portion of the argon ion laser at 515 nm was brought to the interaction region in the crystal. In reading the holograms, the object beam was blocked and the diffraction of the reference beam propagating along the object direction was detected. The diffraction efficiency η, which is defined to be the ratio between the diffracted beam intensity and the reference beam intensity, was thus measured. All the measurements were carried out at room temperature.

In the first set of measurements, the Ti:sapphire laser was tuned to various recording wavelengths from 690 to 995 nm. At each wavelength, holograms were recorded with and without gating beam. In the absence of gating beam, the diffraction efficiency was found to drop very rapidly with the increase of wavelength, and became practically nonmeasurable at $\lambda > 800$ nm. The intensity dependence was found to be characteristically that of a two-photon recording, $\eta \sim I^4$. In the presence of the 515 nm gating beam, the diffraction efficiency became orders of magnitude larger, and more importantly, decreased much slower with the recording wavelength. In Fig. 2, diffraction efficiencies are plotted as functions of recording wavelength. All data points were taken with a fixed writing intensity of 2.0 W/mm^2 and exposure time of 1.0 sec. For two-color writing, the 515 nm gating beam with an intensity of 0.8 W/mm^2 was simultaneously on during the exposure. Note that the vertical scales are different for the two sets of data. The gating efficiency, defined to be the ratio between the diffraction efficiencies obtained with and without the gating beam, is extrapolated to be on the order of 10^4 at 800 nm. On the short wavelength side, gating efficiency is extrapolated to be on the order of 10 at 600 nm, which sets a practical upper limit on the writing photon energy.

In the small diffraction limit, the diffraction efficiency is proportional to the square of the amplitude of electron density modulation, n_e. For the two-step recording, n_e can be approximately expressed as [5]

$$n_e = [\alpha(\lambda_g) I_g \tau][\sigma^*(\lambda_w) I_w t] \times \text{const}, \qquad (1)$$

where α is the absorption coefficient of the medium,

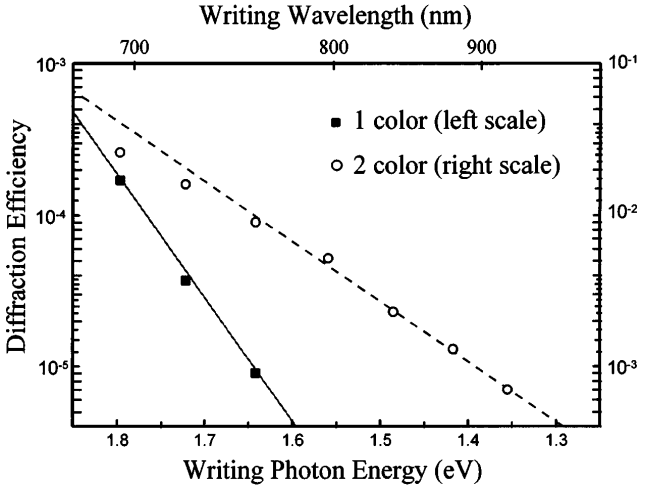

FIG. 2. Two-color (circles) and one-color (squares) photorefractive responses. The horizontal scale is the writing photon energy (wavelength). Note the difference between the two vertical scales.

λ_g (λ_w) is the gating (writing) wavelength, I_g (I_w) is the gating (writing) photon flux (intensity normalized by photon energy), τ is the intermediate state lifetime, σ^* is the absorption cross section for the excitation from the intermediate state to the conduction band, t is the exposure time, and $t \gg \tau$ is assumed. The terms in the first parentheses give the electron population in the intermediate state. The terms in the second parentheses describe the second step excitation. Note that depending on the perspective, subscripts g and w are interchangeable; i.e., I_g (and I_w) can function as both gating and writing beams. In particular, I_g is to be replaced by I_w in the case of one-color, two-photon writing. From Eq. (1), we can see that, when the gating beam and the writing beam are of comparable intensity, the gating efficiency is given by $[\alpha(\lambda_g)/\alpha(\lambda_w)]^2$. The large gating efficiency observed in our experiment is the result of extremely small absorption at the near infrared writing wavelength. One can expect the gating efficiency to further increase at blue to UV wavelengths where the absorption is larger (we have indeed observed much larger gating efficiency using a broadband UV filtered halogen lamp for given gating intensity).

The lifetime of the intermediate state was measured with time-delayed two-step recording. The writing wavelength was at 795 nm and the gating at 515 nm. The laser beams were chopped by acousto-optical modulators to produce pulses of 10 ms in duration. The writing pulses were delayed from the gating pulse. After being excited to the intermediate state by the gating pulse, the electron population in the state starts to decay according to $\exp(-t_D/\tau)$, assuming the population decay is exponential. The diffraction efficiency will decrease accordingly,

$$\eta = \eta_0 \exp(-2t_D/\tau), \qquad (2)$$

where t_D is the time delay between the writing pulses and the gating pulse. Thus lifetime of the intermediate state can be derived from the time-delayed recording measurements. In Fig. 3, the diffraction efficiency is plotted as a function of time delay between the writing pulses and the gating pulse. The intermediate state lifetime is found to be 39 ms. (Intermediate state lifetimes in lithium niobate crystals from different sources vary from 5 to 100 ms. Intermediate state lifetime in a lithium tantalate was measured to be 12 ms. Intermediate state lifetime in a barium titanate was measured to be 33 ms.)

To demonstrate nonvolatile holographic image storage, we expanded laser beams to 2 mm in diameter and recorded an image of the United States Air Force target chart using the two-step recording procedure. The results are shown in Fig. 4. The total writing intensity was 10 W/cm^2 at 810 nm, and the gating intensity was 10 W/cm^2 at 515 nm. Total exposure time was 13.5 sec. The diffraction efficiency is 2×10^{-5}. The hologram was stored up to 200 h and read continuously for 20 h. No reduction of diffraction efficiency was observed in the entire period.

In order to identify the migrating charge carriers, we performed a two-beam coupling measurement. In this experiment, the intensity of the object beam was measured at the two opposite crystal (z-axis) orientations. When the positive z axis and the object beam form an acute angle, the intensity became lower in the presence of the pump beam (coupling loss). In the opposite orientation, coupling gain was observed. According to the analysis in Ref. [6], this indicates that the charge of the free carriers is negative; i.e., the carriers are indeed electrons, which is consistent with our physical picture.

We also attempted to identify the origin of these long lived intermediate states by having the sample composition analyzed with spark mass spectroscopy. It was found that all of the major transition metal impurities,

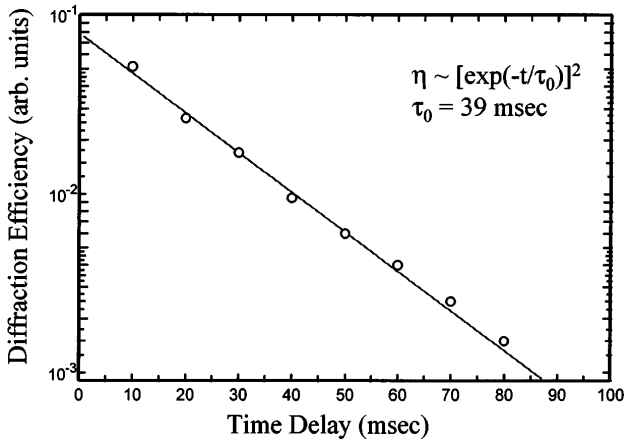

FIG. 3. Time-delayed two-step writing. The horizontal scale is the time delay between the writing pulses and the gating pulse.

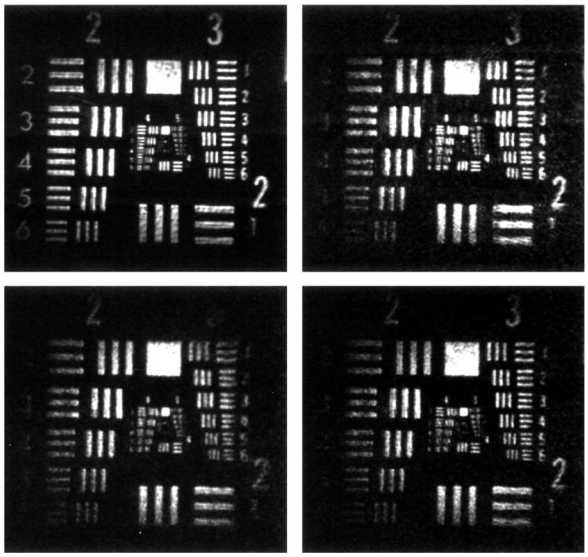

FIG. 4. Nonvolatile holographic image storage at 810 nm using two-step recording. Clockwise from top left: object image, virtual image from the hologram a few minutes after writing, after 100 h storage and 10 h continuous reading, and after 200 h storage and 20 h reading.

including Fe, Cr, Cu, and Mo, are below 1.3 ppm, and all of the rare-earth impurities are below 0.03 ppm. Thus it is clear that these states are intrinsic; i.e., they are associated with some defects in the host lattice.

In some previous low temperature light induced absorption measurements [7,8], it was found that the absorption profile of nominally pure lithium niobates could be drastically modified by intense light illumination at $T < 80$ K. In particular, prominent absorption appeared at the near infrared region after illumination. Accompanying this induced absorption, a signature of Nb^{4+} was observed in electron spin resonance measurements. The Nb^{4+} was believed to be in the form of a "small polaron," i.e., an electron in the conduction band causing distortion in lattice and being trapped by a Nb^{5+}, which is the natural valence state in lithium niobate. The binding energy of the small polaron was estimated to be 0.4 eV, while the peak of the induced absorption was around 1.6 eV [8]. Because of the complex nature of the ferroelectrics, induced absorption usually was not expected to produce the two-step photorefractive effect [9]. However, it does seem plausible to attribute the intermediate states observed in our experiment to these small polarons.

Therefore the two-step recording process can be described as follows. The electrons are first brought by the gating beam to the lower edge of the conduction band where they form small polarons. Although stable at low temperatures, these small polarons can thermally "break up" at room temperature, which results in the finite lifetime observed here. When exposed to the writing laser beams, the bound electrons are excited to higher orbits in the conduction band where with large mobility they migrate away from the excitation region, and then are captured by some deep traps in the dark region. A hologram is thus recorded. With this picture, it is clear that for the two-step effect of occur, the polaron states have to greatly outnumber the deep traps. In our experiment, we found from the intensity dependence that, in the visible region around the gating wavelength 515 nm, one-color recording is a mixture of one-photon and two-photon processes. The competition of these two is described by the "branching ratio"—the ratio between the relaxation rate to the polaron state and that to the deep trap states. These rates are proportional to the densities of states. This explains why the cw two-step effect was not observed in commonly used multivalent transition metal ions doped materials: Concentrations of the deep traps are simply too high. For example, in a 0.01% Fe doped lithium niobate commonly used for one-photon recording, more than 90% of the iron ions are in the Fe^{3+} states (deep traps). However, dopants such as rare earths that exist only in the single valence state and hence do not act as deep traps are expected to improve the two-step sensitivity, because they can increase the lattice distortion and the band tail absorption [10].

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