Reversible photomodification of LiNbO₃ and LiTaO₃ by femtosecond laser pulses

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ABSTRACT

We present experimental and theoretical study of refractive index modification induced by femtosecond laser pulses in photorefractive crystals. The single pulses with central wavelength of 800 nm, pulse duration of 150 fs, and energy in the range of 6-130 nJ, tightly focused into the bulk of Fe-doped LiNbO₃ and stoichiometric LiTaO₃ crystals induce refractive index change of up to about 10^{-3} within the volume of about $(2.0 \times 2.0 \times 8.0) \mu\text{m}^3$. The photomodification is independent of the polarization orientation with respect to the crystalline *c*-axis. The recorded region can be erased optically by a defocused low-intensity single pulse of the same laser. Recording and erasure can be repeated at the same position many times without loss of quality. These findings demonstrate the basic functionality of the ultrafast three-dimensional all-optical rewritable memory. Theoretically they are interpreted by taking into account electron photogeneration and recombination as well as formation of a space-charge field. The presented analysis indicates dominant role of photovoltaic effect for our experimental conditions, and suggests methods for controlling various parameters of the photomodified regions.

Keywords: Rewritable optical memory, LiNbO₃, LiTaO₃, modification by femtosecond pulses

1. INTRODUCTION

LiNbO₃ and LiTaO₃ crystals are widely used in linear and nonlinear optics. Recently they have attracted a new interest as materials suitable for implementation of 3D optical memory elements.¹⁻³ This capability stems from photo-refractive effect due to the formation of space charge field E_{sc} following optical irradiation by a spatially-varying optical field. The space charge field builds up due to carrier diffusion away from the impurity sites, and their subsequent re-trapping, which leads to the gradient in local charge density. The resulting electric field modifies refractive index of the material via electro-optical effect.⁴ Since carrier diffusion and re-trapping rates are relatively slow, these processes generally limit the speed of space charge buildup, and prolonged exposure may be necessary for reliable recording. Moreover, stability of the recorded structures is undermined by dark-current and photo-conductivity. In these circumstances, thermal fixing can be used to stabilize the recording,⁵ but at the same time all-optical character of the memory is lost, and its functionality becomes limited.

Here we show that exposure of LiNbO₃ and LiTaO₃ crystals to tightly-focused, powerful femtosecond laser pulses leads to the formation of localized spatial domains in which refractive index modulation of the order of $\sim 10^{-3}$ can be achieved. The photomodification sites are stable, can be erased optically, and survives multiple recording-erasure cycles. Physical mechanisms responsible for these observations are found to differ strongly from those encountered in the case of irradiation by weak continuous wave (cw) or pulsed lasers.^{6,7} Our estimates show that for the femtosecond pulses used in this work, the main contribution comes from photovoltaic effect

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Figure 1. (color online) Absorption spectra of the investigated samples. The wavelength used for optical recording, λ_{max} in the range of optical transparency in all materials.

2. EXPERIMENTAL DETAILS

2.1 Samples

Lithium niobate samples were Y-cut (parallel to the crystalline c -axis) single crystals of near-stoichiometric (Li/Nb = 49.85/50.15) Fe-doped (400 ppm) LiNbO₃. For reference, undoped LiNbO₃ samples were also used. Lithium tantalate samples were z -cut (perpendicular to the c -axis) near-stoichiometric LiTaO₃ single crystals.⁸ Figure 1 shows optical absorption spectra of the samples. For LiNbO₃, the effect of doping on the optical absorption can be seen clearly. All samples are nearly completely transparent for wavelengths longer than 750 nm. Hence, their linear absorption at the wavelength intended for recording, $\lambda_{laser} = 800$ nm is negligible, and nonlinear absorption is required for the photomodification.

2.2 Optical setup and experimental techniques

Optical setup for the recording of 3D optical memory is shown in Fig. 2. The light source for recording is a femtosecond Hurricane X system (Spectra-Physics) with $\tau_{pulse} = 130$ fs, $\lambda_{laser} = 800$ nm, repetition frequency adjustable within the 1-1000 Hz range, and diameter of a Gaussian beam of about 4 mm (FWHM). The beam intensity is attenuated by a variable attenuator, and its slight divergency compensated by an optical telescope consisting of a confocal pair of lenses having the focal distances of 100 and 120 mm (not shown). After the telescope, a slightly expanded beam is obtained and coupled into an inverted optical microscope (Olympus IX71) which houses the sample and most of the optical, electrical and mechanical components required for the recording. The beam is reflected by the dielectric mirror D highly reflective at λ_{laser} and focused into the sample by a microscope objective lens. The microscope was equipped with a variety of air and oil-immersion lenses having various numerical apertures (NA) and magnification factors. For the recording, a lens with NA=0.5 and average working distance of 5 mm was used. Aberration-free focusing of a plane wave would correspond to the beam radius at waist of $w_0 = 0.61\lambda/NA \simeq 0.88$ μ m estimated at a $1/e^2$ -intensity level.

Although the lens with NA=0.5 was sufficient for *in situ* observation of the recording process and examination of the recorded memory bits, in some cases lenses having higher NA and magnification factors (such as oil-immersion lens with NA=1.35 and $\times 100$ magnification) were occasionally employed. To facilitate recording of 3D structures, the samples were attached to a high-precision piezoelectric transducer (PZT) controlled 3D translation stage mounted on the microscope and consisting of paired $x - y$ and z stages (Physik Instrumente P-517.2CL and P-518.ZCL) which provide a maximum positioning range of up to 100 μ m and accuracy of several nanometers. The samples are illuminated by a halogen lamp and a condenser C , and can be monitored in transmission within the spectral transparency range of the dielectric mirror D (visible wavelengths) using a video camera VC and monitor VM . For a properly chosen laser beam intensity, single-photon absorption is negligible, and laser pulses can be delivered into the bulk of the samples without absorptive losses. At the focal region,

Figure 2. (color online) Optical setup for femtosecond recording of three-dimensional memories. See the main text for details.

where local intensity is highest, nonlinear absorption is induced locally, and refractive index becomes modified. The index change monitored *in situ* is usually seen as a small bright/dark area. The threshold for recognizable modification at the depth of $50\ \mu\text{m}$ under the samples' surface was $3.8 \pm 0.5\ \text{nJ}$ in $\text{LiNbO}_3\text{:Fe}$ and $5.2 \pm 0.5\ \text{nJ}$ in undoped LiNbO_3 . The threshold for LiTaO_3 was $32 \pm 5\ \text{nJ}$ at the depth of $5\ \mu\text{m}$.

During the recording, the translation stage is controlled by a PC, which also controls a mechanical beam shutter (from Uniblitz). Such experimental system ensures that each laser pulse arrives to a pre-defined location in the sample.

For the monitoring of optically-induced refractive index changes and their relaxation, a holographic recording and readout setup was built. Such setup is shown schematically in Fig. 3. For recording in linear regime, a femtosecond optical parametric amplifier (OPA) pumped by the Hurricane system and emitting 130 fs pulses at the wavelength of 532 nm was used. The choice of wavelength was based on its common use for holographic recording at irradiance levels of the order of a few GW/cm^2 . At this wavelength absorption by Fe-band in Fe:LiNbO_3 is dominant. A diffractive beam splitter (DBS) was used to split the recording beam into several diffracted beams. The key advantage of the DBS use is that it ensures perfect temporal coincidence between the diffracted components.^{9–12} The collimated beams are passed to a selector plate with two apertures, which selects two beams needed for the grating recording. These beams are focused in the sample by another lens with a typical focal distance of 100 mm, and their interference field records a simple volume grating in the sample. The probing cw beam of a HeNe laser, despite the different wavelengths, pass through the same path as the recording beam, and intensity of its component, diffracted by the recorded grating, is monitored using a photodiode and an oscilloscope. Owing to the use of DBS, the probe beam perfectly fulfills the Bragg condition, which otherwise would be difficult to achieve in thick gratings having strong refractive index modulation. Temporal decay of the grating can be monitored by registering the diffracted intensity as a function of time. Absorption of the monitoring beam at the 633 nm wavelength is negligible in our samples (see Fig. 1).

Figure 3. (color online) Schematic explanation for holographic recording of refractive index gratings and monitoring of their index modulation.

Figure 4. Optical transmission images of memory bits recorded by single pulses in LiNbO₃:Fe at the 10 μm depth by 7 nJ energy pulses. The recording sequence is explained in the text, (a) bits after a single recording, (b) bits after repetitive recording of 20 times. Polarization of the recording beam was perpendicular to c -axis, the white circles emphasize the positions of recorded bits.

3. RESULTS AND DISCUSSION

3.1 Single-pulse recording and erasure of memory bits

At first we will examine recording of memory bits by single pulses at high irradiance levels reaching TW/cm^2 level, but not exceeding the optical breakdown threshold. Under these circumstances, recording in Fe-doped LiNbO₃ can be accomplished most straightforwardly.¹³

Figure 4 shows optical images of Fe-doped LiNbO₃ areas subjected to a single (a) and multiple (b) recording cycles. Focusing depth under the surface of the sample during the experiments was 10 μm . The recorded bits can be seen as brighter or darker spots in the images; high brightness generally corresponds to a higher refractive index and vice versa.¹⁴ At pulse energies higher than 11-12 nJ, dielectric breakdown resulted for irradiation of spatially non-overlapping regions, causing strong, irreversible modulation of the refractive index. For irradiation of overlapping regions, optical damage resulted at even lower intensities (see Fig. 5).

The diameters of bits recorded with NA=0.5 lens was found to be approximately 1.1 μm (FWHM) from observation of the recorded structures using a microscope lens with higher NA and magnification. The axial diameter of the focal area was estimated from the expression $z_0 = (\pi w_0^2 n / \lambda) \simeq 6.8 \mu\text{m}$, where $n = 2.2$ is the real part of refractive index in LiNbO₃.

The recorded patterns consist of four horizontal lines (*i-iv*), recorded from left to right: line (*i*) consists of bits recorded with a small interval of 0.5 μm , and therefore merged together, line (*ii*) is the same as (*i*), but it

Figure 5. (color online) (a) Memory bits recorded by single pulses in LiNbO₃:Fe at 50 μm depth with a variable separation along the c -axis (or x -axis) direction, and (b) image intensity along the selected lines. The recording beam was polarized along the crystalline c -axis direction. Separation between the bits is given by numbers.

passes over a previously recorded bit at the middle in order to demonstrate the erasure, line (iii) has a single bit post-recorded at the middle in order to demonstrate over-writing, and line (iv) contains a single bit only as a reference. First and last bits of the lines are denoted as 1 and N , respectively. Single bits recorded after the lines are denoted as $N+1$. Comparison between the images in Fig. 4(a) and (b) clearly shows that after 20 re-writing cycles the pattern remains essentially the same.

Figure 5 shows optical image of the structure consisting of parallel lines with different intervals between the recorded bits. As can be seen from the Figure, the lines of bits drawn at intermediate y -axis positions become invisible (except for their extreme points) as a consequence of substantial overlap between the neighboring bits when bit-to-bit spacing becomes smaller than approximately 0.6 μm . We have verified in separate experiments that photomodification in these regions is reversible, i.e., multiple recording and over-writing similar to that demonstrated in the previous Figure is possible. Further decrease of the bit spacing beyond approximately 0.3 μm results in the appearance of dark lines manifesting an irreversible optical damage due to the excessive irradiation dose resulting from stronger overlap between the neighboring spots. The pattern in Fig. 5(a), which was recorded by a laser beam polarized along the c -axis has a higher optical contrast than the patterns in Fig. 4, which were recorded using a laser beam polarized perpendicularly to the c -axis. This difference is consistent with the tentative assumption that under excitation polarized conditions the space charge field evolves mainly due to photovoltaic effect.^{14,15}

Altogether, the observations summarized in Figs. 4 and 5 demonstrate the possibility to record closely-spaced memory bits in LiNbO₃, and to erase them by overwriting with lines of closer-spaced bits. We would like to emphasize here, that although such kind of erasing leaves features recognizable as bits at the start and end of the erasing line, this circumstance can be incorporated into the protocol of the erasure, and should not limit functionality of 3D optical memory applications. One may conclude that erasure of *detectable bits* by overwriting does not necessarily manifest genuine erasure of their *index modulation profiles* i.e., overwriting induces uniform index modification along the erasure lines that are invisible under inspection by optical microscopy, which is most sensitive to rapid variations of the refractive index. However, it is well known that photo-refractive index modulation, recorded by a spatially varying optical field in LiNbO₃ and related materials, is completely erasable by exposure to a spatially uniform field.^{3,4} This functionality was demonstrated previously using continuous-wave or low-intensity pulsed irradiation. Here, essentially the same scenario is implemented using high-intensity pulsed irradiation, which allows rapid recording and erasure. Exposure to a uniform optical field required for the latter process is achieved by overwriting, which eventually creates nearly-uniform exposure, and suppresses formation of the space charge field along the overwriting lines. In these circumstances it is almost obvious that the overwriting leads to a genuine erasure of index modulation profiles of the memory bits. In order to verify this fact experimentally, a more sensitive imaging technique, such as phase-contrast optical microscopy may be used in the future.

Figure 6. Demonstration of a 3D recording in LiNbO₃:Fe sample. Letters Y, T, and A consisting of bits separated by 1 μm from each other were recorded on vertically stacked planes separated by 3 μm distance. The recording was done at the pulse energy of 6 nJ using oil-immersion microscope lens with NA=1.4.

Figure 7. (color online) Grating decay in LiNbO₃ following its recording by the interference field of two femtosecond pulses. The monitoring was done at first using a beam of cw HeNe laser (at 633 nm wavelength), and later, under additional spatially uniform irradiation by femtosecond pulses having different power levels I_0 and $2I_0$. The uniform illumination was done by closing all one of the interfering beams.

The data described above can serve as a demonstration of the functionality of all-optical 3D memory controllable by single femtosecond pulse irradiation. To further illustrate genuine 3D recording, Fig. 6 shows images of letters Y, T, and A, formed by bits separated from each other by 1 μm, recorded on three vertically stacked planes, separated by 3 μm intervals. To achieve a higher recording density, oil-immersion lens with numerical aperture of NA= 1.4 was used. The laser pulse energy was 6 nJ. After the recording, each of the planes shown in the Figure had to be imaged separately by adjusting the depth of focusing. At optimum focusing, no bits belonging to other planes could be seen in the images.

In order to test the temporal relaxation of photomodification and to determine the magnitude of index modulation, holographic recording experiments were performed using the setup described in Fig. 3.

For the recording, LiNbO₃:Fe sample was used due its stronger index modulation response to femtosecond laser pulses.¹³ First, a grating was recorded by the interference field of femtosecond pulses, till diffraction

Figure 8. (color online) Memory bit recorded by a single pulse with 14.5 nJ energy at the focus in LiTaO₃. Optical images after the recording and before it are compared on the top row. A difference image and the dependence of its brightness (integrated within the spatial range emphasized by the horizontal lines) on the position are given on the bottom row. The positions of recorded memory bit is emphasized by circles in the images. Crystalline *c*-axis of LiTaO₃ is perpendicular to the image plane.

Figure 9. (color online) Temporal relaxation of memory bits recorded at two different pulse energies in LiTaO₃ monitored via optical transmission changes. The two dependencies are offset for clarity; the steady-state background transmission levels are indicated by dashed horizontal lines for each dependency. Temporal width and amplitude of transmission modulation during the laser pulse action are indicated by the rectangular boxes. The inset shows the image of linear trace, recorded by smooth translation of the focal spot at a constant velocity, taken immediately after the recording. The transmission data was acquired from the region indicated by the box.

efficiency of the grating saturated. For further analysis it is useful to define the diffraction efficiency η , which is the ratio between the intensities of the diffracted and incident radiation. For sinusoidal phase grating η can be expressed as

$$\eta = \frac{I_{df}}{I_{inc}} = \sin^2 \left(\frac{\pi \Delta n t}{\lambda \cos(\theta_i)} \right) \approx \left(\frac{\pi \Delta n d}{\lambda} \right)^2, \quad (1)$$

where Δn is the amplitude of refractive index modulation, λ is the wavelength, d is the grating thickness, and θ_i is the incidence angle. Thus, Δn can be extracted from the measurements of diffraction efficiency.

During the recording, as soon as the diffraction efficiency attained a constant value, the recording beams were blocked by the shutter, and intensity of the diffracted probe beam (HeNe laser beam with wavelength of 633 nm) was monitored versus time. In other experiments the grating was additionally irradiated by spatially uniform femtosecond pulses during the monitoring. This was achieved by opening one of the recording beams (i.e., no interference field was formed). The uniform femtosecond irradiation at two different power levels, denoted as I_0 and $2I_0$ was used. For I_0 the order-of-magnitude estimate is $I_0 \simeq 10 \text{ GW/cm}^2$.

The dependencies reflecting the temporal relaxation of diffraction efficiency are shown in Fig. 7. Without the uniform illumination, relaxation time of approximately 72 min can be inferred from the decay. Under uniform femtosecond illumination the initial magnitude of diffraction efficiency becomes lower, but the grating decay slows down to about 140 and 95 min at I_0 and $2I_0$, respectively. Relatively fast grating decay is most probably due to the electron tunneling between iron sites, characteristic for highly-doped $\text{LiNbO}_3\text{:Fe}$ crystals.¹⁶ This assumption is further supported by the observed decrease of the grating decay times during simultaneous probing and homogeneous photoexcitation of Fe^{2+} states. While the excess photocarriers in the conduction band screen the internal electric field, reducing diffraction efficiency of the grating, the $\text{Fe}^{2+}/\text{Fe}^{3+}$ ratio is also lowered, which leads to significantly lower electron tunneling rates.¹⁷ In order to achieve more detailed understanding of the observed dynamics, photoionization of deep Fe traps as well as recombination and capture of electrons should be modeled.

The maximum refractive index modulation in $\text{LiNbO}_3\text{:Fe}$ estimated according to (1) from the measured diffraction efficiency, $\simeq 1 \%$, was found to be $5 \cdot 10^{-4}$. In addition, diffraction grating with period of $\Lambda = 3 \text{ }\mu\text{m}$ and grating vector parallel to the c -axis was recorded using point-by-point scanning (the distance between the adjacent irradiated areas was $0.2 \text{ }\mu\text{m}$). This grating had slightly higher diffraction efficiency of $\eta \approx 2\%$, from which $\Delta n \approx 6 \times 10^{-3}$ was determined assuming $d = 6.8 \text{ }\mu\text{m}$, i.e., the grating thickness equal to the beam waist length. In both cases, the obtained index modulation can be regarded as high.

Figure 8 shows image of a bit recorded in undoped LiTaO_3 , observed along the c -axis direction. In this material the recorded bits were short-lived and disappeared after few tens of milliseconds. For pulse energies above 17 nJ modification became permanent when exposure to multiple pulses with repetition rate of 1 kHz was applied. Although contrast of the transient bit is somewhat lower than in $\text{LiNbO}_3\text{:Fe}$, we have verified that within its life span the bit is erasable and rewritable by single-pulses, i.e. essentially the same functionality as in $\text{LiNbO}_3\text{:Fe}$ is achieved. To enhance the visibility of recording, background subtraction was performed on the image as illustrated in Fig. 8. The difference image exhibits modulation of optical transmission of about $\sim 2.3\%$. Small magnitude and short lifetime of the photomodification can be attributed to purely refractive index modification. The maximum contrast of modulation is related to the defect density. For intrinsic LiNbO_3 , typical $\text{Nb}_{\text{Li}}^{4+/5+}$ antisite defect density is about 1%. This amount limits the maximum refractive index modulation via photovoltaic effect, or buildup of the space charge field and index modification via Pockels effect. Judging from the optical transmission modulation in LiNbO_3 and LiTaO_3 , the maximum index modulation induced by a single pulse irradiation is approximately 5×10^{-4} for y -plane in LiNbO_3 and 2.5×10^{-4} the for c -plane in LiTaO_3 .^{13, 18}

Temporal relaxation of bit modulation in LiTaO_3 was monitored by recording a line of bits at a constant velocity of $0.1 \text{ }\mu\text{m/ms}$ at 20 Hz laser pulse repetition, and taking a sequence of optical images at the video rate of 1/30 s. Under these conditions, the interval between neighboring irradiation sites was nearly equal to the spot diameter. Recording of the line at a constant speed, followed by a snapshot optical image of the line immediately afterwards, allows one to extract the dependency of bit transmission versus the time elapsed after the recording. Such dependency is shown in Fig. 9. At both intensities of the laser pulses used, a full relaxation of the bit modulation occurs within about 300 ms time. The fast recovery can be tentatively explained as a result of diffusion and recombination of photogenerated e-h pairs (the probability of two-photon absorption at 800 nm in undoped LiTaO_3 is negligible).

3.2 Analysis of the refractive index modulation mechanism

In this Section we analyze the possible physical mechanisms behind the reversible photorefractive modification by femtosecond pulses in LiNbO₃ and LiTaO₃. In these estimates we will assume size of the irradiated region typical for the case of focusing by a lens with NA=0.5, irradiation by single pulses having 150 fs duration. At first we will estimate free carrier electron generation rates via avalanche and multiphoton ionization (MPI) processes.

Joule heating of electrons oscillating in the electromagnetic field of laser beam can be approximated as:

$$\frac{d\varepsilon_e}{dt} \approx 2\varepsilon_{osc} \frac{\omega^2 \nu_{e-ph}}{\nu_{e-ph}^2 + \omega^2} \approx 2\varepsilon_{osc} \nu_{e-ph}, \quad (2)$$

where ν_{e-ph} is the electron-phonon collision rate, ε_{osc} the electron quiver energy, ω is the frequency. The rate of electron-phonon collisions can be estimated from the known value of the heat diffusion coefficient $D = 1.5 \times 10^{-2} \text{ cm}^2\text{s}^{-1}$ by using the relationship $D \approx v_s^2 / (3\nu_{e-ph})$, where $v_s = 4 \text{ km/s}$ is the sound velocity. Thus, one obtains $\nu_{e-ph} = 3.5 \text{ THz}$. As an alternative estimate, the approximate range of ν_{e-ph} can be determined by dividing the sound velocity by the range of interatomic distances of 5.148-13.865 Å. This method yields $\nu_{e-ph} = (2.9 - 8.0) \text{ THz}$. In the following an average value of $\nu_{e-ph} = 5 \text{ THz}$ will be used.

The energy of electrons oscillating in the optical field of a linearly polarized laser pulse can be expressed in scaling form as $\varepsilon_{osc} = 9.3I\lambda^2 \text{ [eV]}$, where $I = 10^{14} \text{ W/cm}^2$ and λ is expressed in micrometers. Assuming $I = 6.7 \text{ TW/cm}^2$, $\varepsilon_{osc} = 0.4 \text{ eV}$ is obtained. Using the previously estimated value of $\nu_{e-ph} = 5 \text{ THz}$, the rate of electronic energy gain is $2 \times 10^{12} \text{ eV/s}$, while the avalanche ionization rate is $w_{av} \approx d/(\Delta_{gap}\varepsilon_e dt) \sim 0.5 \text{ THz}$. These estimates imply that $w_{av}t_p = 0.075$; hence the rate of electronic transitions from the valence to conduction bands is insignificant (however, electron generation from impurity levels located approximately 0.4 eV below the conduction band is still possible).

MPI rate per atom per second can be determined from the following expression:

$$w_{mpi} \approx \omega n_{ph}^{3/2} \left(\frac{\varepsilon_{osc}}{2\Delta_{gap}} \right)^{n_{ph}}, \quad (3)$$

where $n_{ph} = \Delta_{gap}/\hbar\omega \sim 4/1.55 \text{ eV} = 2.575$ is number of photons required for excitation of one electron across the band gap ($\sim 4 \text{ eV}$). Assuming $\omega = 2.356 \text{ fs}^{-1}$, and gap width of 4 eV, $w_{mpi} \approx 4.9 \times 10^{15} (I/10^{14})^{2.575} \text{ s}^{-1}$ is obtained. Assuming a Gaussian beam shape at the waist and a pulse duration of 150 fs, $w_{mpi} = 1.46 \text{ THz}$, and density of conductivity electrons generated by this process is $n_e = w_{mpi}n_a t_p = 2 \times 10^{22} \text{ cm}^{-3}$ (where $n_a = 9.45 \times 10^{22} \text{ cm}^{-3}$ is the atomic density). This value is larger than the critical density for plasma generation $n_{cr} = 1.76 \times 10^{21} \text{ cm}^{-3}$ at the wavelength of 800 nm. We assume here that critical density is achieved only at the end of the exciting pulse ($\omega_{pe} \sim \omega \gg \nu_{e-ph}$), and during the action of the laser pulse the real part of dielectric function is not modified. The imaginary part which describes absorption, $\varepsilon'' \approx \nu_{e-ph}/\omega$, can be expressed as $\kappa \approx \varepsilon''/2n_0 = \nu_{e-ph}/2n_0\omega = 5 \times 10^{-4}$. Assuming the refractive index $n_0 = 2.2$, the absorption length is $l_s = c/\kappa\omega \approx 250 \text{ } \mu\text{m}$. It can be therefore concluded that MPI is not important under our conditions.

Temperature of the electrons can be deduced from the the energy loss to the crystal lattice:

$$\frac{\partial C_e n_e T_e}{\partial t} = \frac{2A}{l_{abs}} I(r, z, t), \quad (4)$$

where A is the absorbance, l_{abs} is the absorption length, C_e is the specific heat capacity per electron, T_e the electron temperature, and n_e the electron density. The heat conduction is neglected. At the end of the laser pulse the temperature becomes

$$T_e \approx \frac{2AIt_p}{C_e n_e l_{abs}}. \quad (5)$$

The temperature of degenerate electron gas having heat capacity $C_e = (\pi^2 T_e)/(2\varepsilon_F)$, density $n_e \simeq n_{cr} \sim 10^{21} \text{ cm}^{-3}$, absorbance $A \sim 2z_0/l_s$, $l_s \sim 2.5 \times 10^{-2} \text{ cm}$, and Fermi energy $\varepsilon_F \sim 10 \text{ eV}$ can be expressed as $T_e^2 \approx \frac{8z_0 I t_p}{\pi^2 l_s^2 n_e} \varepsilon_F$. From this expression, increase in the the electron temperature comprises $T_e = 0.2 \text{ eV}$ (in energy

units). The electronic excess energy becomes transferred to the lattice within the time $t_{e-L} \approx \nu_{e-ph}^{-1} \sqrt{M/m^*}$. Assuming the average atomic mass $M = 29.6$ a.u., and free electron mass equal to their effective mass m^* , one finds $t_{e-L} = 4.6 \times 10^{-11}$ s. Total absorbed energy per volume is $E_{abs} \approx (2A \cdot I \cdot t_p)/l_{abs} \sim 1.85$ J/cm³. Thus, assuming the specific heat capacity $c_p = 628$ J/(kg·K), lattice heating by the amount of $\Delta T \simeq 0.6$ K can be obtained. This value is small, and implies negligible contribution of pyroelectric effect to the buildup of space charge field.

Contribution from diffusive charge redistribution is

$$E_{dif} \approx \frac{T_e}{e} \frac{\nabla n_e}{n_e}, \quad (6)$$

where e is the electron charge. Substituting the value of $T_e \sim 0.2$ eV, and approximating the density gradient by the ratio of density to the lateral size of the focal region, $\nabla n_e/n_e \propto w_0^{-1} \sim 2 \times 10^4$ cm⁻¹, a moderate field strength of $E_{dif} \sim 3.6$ kV/cm is obtained.

Finally photovoltaic contribution can be evaluated from the photovoltaic current, j_{ph} , non-centrosymmetric medium:

$$j_{ph} = l_{abs}^{-1} G I, \quad (7)$$

where G is Glass constant. For the beam incidence along the direction perpendicular to c -axis, photovoltaic current is excited in the c -axis direction by an electric field, estimated for quasi-steady conditions as $E_{ph} = j_{ph}/\sigma$. Real part of the conductivity during the laser pulse action is:

$$\sigma = \frac{e^2 \cdot n_e \cdot \nu_{e-ph}}{m_e^* (\omega^2 + \nu_{e-ph}^2)}. \quad (8)$$

After the laser pulse is gone, the conductivity becomes $\sigma \approx (e^2 n_e)/(m_e^* \nu_{e-ph})$. For $\omega_{pe} \sim \omega \gg \nu_{e-ph}$, one can rewrite (8) as $\sigma \approx \nu_{e-ph}/4\pi \sim 0.4$ THz. By taking $G_{33} = 3 \times 10^{-9}$ cm/V, and using (7), one obtains $E_{ph} = 1.62$ MV/cm.

According to the above estimates, photovoltaic effect is by far stronger than other effects considered here. Modulation of the refractive index by the internal electric field resulting from charge separation is¹⁴

$$\Delta n = -\frac{n^3 r}{2} E_{sc}. \quad (9)$$

In LiNbO₃, $(\lambda/n^3) \cdot r = U_\pi \approx 2$ kV at the wavelengths of $\lambda=630$ nm. At the wavelength of 800 nm, $n^3 \cdot r \approx 6.35 \times 10^{-8}$ cm/V, where r is the electro-optic coefficient. Therefore, the theoretical limit to the index modulation is $|\Delta n| = 5.14 \times 10^{-2}$. Most likely this value is overestimated, since the rate of electron-phonon interaction was obtained indirectly, from numerical estimates.

4. CONCLUSIONS

We have present experimental and theoretical studies of refractive index modification induced by femtosecond laser pulses in LiNbO₃ and LiTaO₃ photorefractive crystals. Refractive index change of up to about 10^{-3} was induced by tightly focused laser pulses within the volume of about $(2.0 \times 2.0 \times 8.0) \mu\text{m}^3$. The photomodification is optically erasable, and the recording-erasure cycle be repeated at the same position many times without loss of quality. Theoretical analysis of these observations has revealed a dominant role of photovoltaic effect in the buildup of photorefractive modification. Optical recording of memory bits demonstrated in this work exhibits the basic functionality of ultrafast three-dimensional all-optical rewritable memory.

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