Nonvolatile holographic storage in iron-doped lithium tantalate with continuous-wave laser light

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Holograms have been recorded in congruent $LiTaO_3$:Fe with continuous-wave laser light by use of a two-step process. Blue gating light ($\lambda = 488$ nm) sensitizes the crystals for holographic recording with red light ($\lambda = 660$ nm) of a diode laser. Refractive-index changes of as much as 1.0×10^{-5} are achieved for intensities of the red light of 1 W/cm^2. The saturation values are proportional to the intensity of the writing light. Nondestructive readout with red light is possible, and the holograms remain erasable for blue light. © 1999 Optical Society of America

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Volume holography is attractive for data storage because of its large storage densities and short access times. Photorefractive crystals such as iron-doped lithium niobate ($LiNbO_3$) and lithium tantalate ($LiTaO_3$) yield extremely large dark storage times of as much as 10 years.[1] In these crystals the iron ions occur in two valence states (Fe^{2+} and Fe^{3+}). To record a hologram one illuminates the crystal with a light-interference pattern of two intersecting laser beams, called signal and reference beams. The light can excite electrons from Fe^{2+} into the conduction band, where the electrons migrate before they are finally trapped at Fe^{3+}. This charge redistribution leads to an inhomogeneous space-charge field that modulates the refractive index through the electro-optic effect. To read the stored information, one illuminates the crystal with the reference beam and reconstructs the signal beam. One crucial problem with this process is the volatility of stored information, because during readout electrons are redistributed homogeneously, which leads to erasure of the recorded hologram. Thermal and electrical fixing techniques have been developed to overcome this problem, but the desired optical erasure is not possible.

As an alternative procedure, two-color recording has been proposed. During writing of a hologram with light of one wavelength, the crystal is illuminated homogeneously with light of a different wavelength. This gating light must not be coherent. In the absence of the gating light, the crystal is insensitive to light of the writing wavelength, and reading with one writing beam is nondestructive. Besides the nondestructive readout, this pure optical fixing process offers further advantages. As the crystal is not photorefractive for light of the writing wavelength, no undesired holographic scattering effects can occur during readout. Furthermore, multiplexing of many holograms is improved, because the crystals show no absorption at the writing wavelength and therefore thick samples can be used.

In earlier experiments, two-color recording was performed by use of pulsed lasers with light intensities of the order of 1×10^{11} W/m^2, and writing occurred through virtual or real intermediate states.[2–4] However, for compact low-cost data-storage applications pulsed lasers are not practical. Recently, approaches with near-stoichiometric reduced $LiNbO_3$ have been tried.[5,6] In $LiNbO_3$ the congruently melting composition is 48.6-mol. % Li_2O. The lack of Li is partially compensated for by Nb ions on Li sites (Nb_{Li}^{5+}). If $LiNbO_3$ is illuminated with blue light, electrons are excited from deep centers and the intrinsic Nb_{Li}^{5+} defects can capture electrons, forming small polarons (Nb_{Li}^{4+}). The energy level of these small polarons is close to the conduction band. Thus it becomes possible to record holograms with near-infrared light by excitation of electrons from Nb_{Li}^{4+} polarons with subsequent redistribution. The recording process becomes more effective with increasing polaron lifetime, and in $LiNbO_3$ the polaron lifetime increases with increasing Li content. Unfortunately, stoichiometric $LiNbO_3$ has a minor optical homogeneity compared with that of the congruent material.

In this Letter we present experimental results of a two-color writing process in congruent iron-doped lithium tantalate ($LiTaO_3$:Fe). Advantages of $LiTaO_3$ in comparison with $LiNbO_3$ are longer storage times and excellent optical homogeneity.[1] Holograms are recorded with a 660-nm diode laser, and gating is performed with either an Ar^+ laser at 488 nm or a 150-W Xe lamp. The dependences of holographic sensitivity and dynamic range on the intensities of the writing and the gating light are examined.

Measurements are performed with different single-domain $LiTaO_3$ crystals containing 0.002- to 0.02-wt. % Fe, as determined earlier by mass spectroscopic and x-ray fluorescence analysis.[1] It is possible to record nonvolatile holograms in all these crystals, but the present results have been obtained with a 0.65-mm-thick sample doped with 0.02-wt. % Fe. We reduce this sample by annealing it in vacuum (10^{-2} mbars) at a temperature of ~1000 °C for 2 h, increasing the absorption of Fe^{2+} near 400 nm (3.1 eV). The number of Fe^{2+} ions can be derived from the absorption spectrum, and we find that $c_{Fe^{2+}} = 105 \times 10^{23}$ m^{-3}.[1] $LiTaO_3$ is isomorphous to $LiNbO_3$,

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and crystals that were grown from the congruently melting composition have a Li deficit and contain a large amount of Ta antisite defects (Ta_{Li}^{5+}). The Curie temperature of our crystal is 618 °C, which corresponds to a Li_2O content of 48.21%.[7]

For hologram recording, two interfering red beams ($\lambda = 660$ nm) of a 30-mW diode laser write elementary holographic gratings with a period $\Lambda = 3.2$ μm. The writing beams have equal intensity and are extraordinarily polarized. They have a beam diameter of ~1.5 mm and are overlapped by a 5-mm-wide beam from either an Ar^+ laser or a 150-W Xe lamp. The writing intensity is adjusted with a half-wave plate and a polarizer. The intensity of the gating light of the Ar^+ laser can be changed with neutral-density filters. The diffraction efficiency η is monitored with cw light from a 785-nm diode laser that is extraordinarily polarized and Bragg matched. Here η is defined by the ratio $I_d/(I_d + I_t)$, where I_d and I_t are the intensities of the diffracted and the transmitted light, respectively. The saturation value Δn_S of the refractive-index modulation can be calculated from Kogelnik's formula.[8]

$LiTaO_3$:Fe is insensitive to holographic recording with red writing light alone. Nevertheless, recording becomes possible if the crystal is illuminated with additional gating light. Figure 1 shows the amplitude of the refractive-index change for a typical write–read–erase cycle. Here white light of a 150-W Xe lamp is used for gating. The recording process can be understood with a two-center charge-transport model (Fig. 2). We assume that, in addition to the deep $Fe^{2+/3+}$ center, intrinsic $Ta_{Li}^{4+/5+}$ ions act as a shallow center. Holographic recording is realized by a two-step process, in which homogeneous illumination with the gating light excites electrons from Fe^{2+} directly to Ta_{Li}^{5+}, forming small polarons (Ta_{Li}^{4+}). Simultaneous illumination with an interference pattern of red light transfers the electrons from the shallow center to the conduction band, where they migrate before they are trapped by Fe^{3+} ions. Reading with red light is nondestructive, because the photon energy of the red light is not sufficient to excite electrons from Fe^{2+} ions. The Ta_{Li}^{4+} polarons induce a broad light absorption with a maximum at 570 nm.[9] In our crystal we cannot detect any light-induced absorption for the gating intensities because the sample is too thin. Nevertheless, light-induced absorption changes can be measured by use of light ($\lambda = 355$ nm) from a Q-switched Nd:YAG pulse laser for pumping.[4] These measurements yield a polaron lifetime of a few milliseconds, which strongly depends on the amount of Fe^{3+} ions. The lifetime increases with decreasing Fe^{3+} concentration. This result is due to direct transitions between deep and shallow centers (Fig. 2). Thus, reduced $LiTaO_3$:Fe crystals show a relatively long lifetime of excited electrons, and two-step recording with small light intensities becomes possible.

Important parameters in characterizing holographic storage materials are sensitivity and dynamic range. The dynamic range depends on the saturation value Δn_S of the refractive-index change. From Fig. 3 it can be seen that Δn_S increases linearly with increasing writing intensity. The diffraction efficiency η in the 0.65-mm-thin sample with $\Delta n_S \approx 10^{-5}$ is of the order of 10^{-4}. However, for a one-photon writing process Δn_S does not depend on the writing intensity. The intensity dependence can be explained if we assume that the red writing light generates a photovoltaic current in the conduction band. Such a current is proportional to the writing intensity as well as to the concentration of filled shallow levels. The photoconductivity, however, is mainly determined by the intensity of the gating light. There is also an influence of the gating light on the dynamic range: A larger gating intensity leads to a smaller saturation

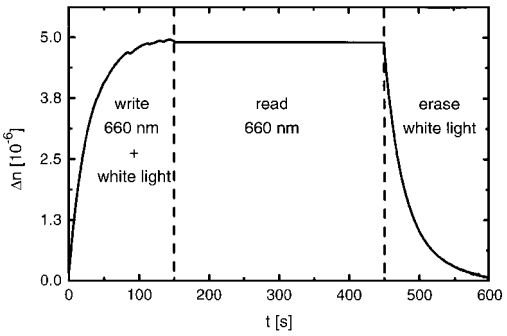

Fig. 1. Refractive-index change for a typical write–read–erase cycle. The hologram is recorded with a 660-nm diode laser ($I_{writing} = 0.65$ W/cm^2) and white light from a Xe lamp for gating. Nondestructive readout is performed with one of the writing beams, and hologram erasure is possible with the gating light.

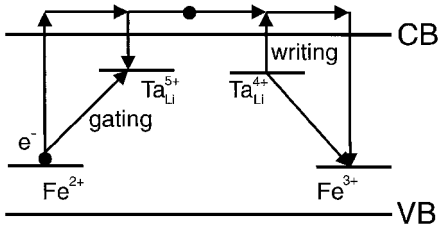

Fig. 2. Two-center charge-transport model in $LiTaO_3$:Fe. Deep center, $Fe^{2+/3+}$; shallow center, $Ta_{Li}^{4+/5+}$; VB, valence band; CB, conduction band.

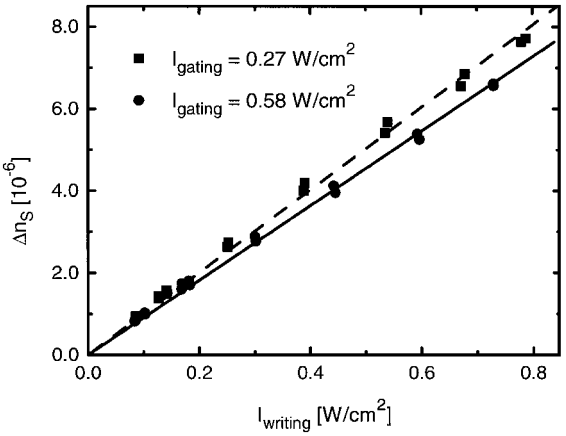

Fig. 3. Saturation values Δn_S of refractive-index changes as a function of the writing intensity $I_{writing}$ for two different gating intensities. The lines are linear fits to the measured values.

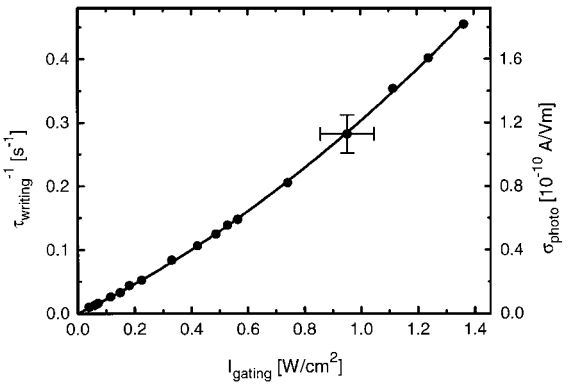

Fig. 4. Inverse time constant $1/\tau_{\text{writing}}$ for hologram writing and photoconductivity σ_{photo} versus gating intensity at 488 nm. The curve is a fit according to $\sigma_{\text{photo}} = aI_{\text{gating}} + bI_{\text{gating}}^2$, where a and b represent fit parameters.

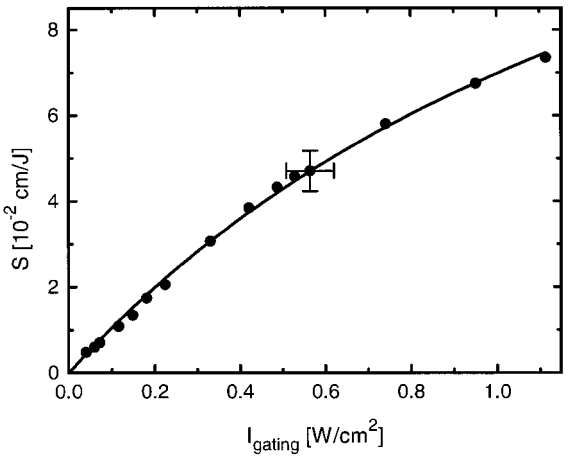

Fig. 5. Dependence of photorefractive sensitivity S on the gating intensity at 488 nm. The curve is a fit according to $S = aI_{\text{gating}}/(b + I_{\text{gating}})$.

value Δn_S (Fig. 3). As the gating light is able to empty filled shallow levels, hologram erasure during recording with the gating light increases with increasing gating intensity. The inverse time constant $1/\tau_{\text{writing}}$ for hologram writing shows a slightly superlinear dependence on the gating intensity (Fig. 4). The time constants for writing and erasure have the same values and do not depend on the intensity of the writing light. For a gating intensity of $1\,\text{W/cm}^2$ the writing time constant is ~ 4 s. The photoconductivity σ_{photo} is large because of a high $c_{Fe^{2+}}/c_{Fe^{3+}}$ ratio. Hologram readout with one of the writing beams ($I = 0.5\,\text{W/cm}^2$) also leads to small erasure effects with a time constant of ~ 4 days. But this value is more than 80,000 times larger than the writing time constant. An even higher ratio can be achieved with a longer writing wavelength, because in the near infrared the excitation of electrons from Fe^{2+} is much weaker. The dark storage time of our crystal is estimated to be approximately 90 days. In oxidized samples dark storage times of 10 years were measured.[1] The sensitivity, $S = 1/(\mathrm{d}I_{\text{writing}})\partial\sqrt{\eta}/\partial t|_{t=0}$, where d is the crystal thickness, increases with increasing gating intensity (Fig. 5), and for large intensities a saturation behavior is expected. We estimate a saturation value of the order of 0.18 cm/J. For an intensity of the gating light of $1\,\text{W/cm}^2$ the sensitivity is 7.0×10^{-2} cm/J. Writing with red light alone ($I_{\text{writing}} = 1\,\text{W/cm}^2$) yields a sensitivity that is 3 orders of magnitude smaller than for gated recording ($I_{\text{gating}} = 1\,\text{W/cm}^2$). The sensitivity of our iron-doped congruent $LiTaO_3$ crystal is already larger than the sensitivity in near-stoichiometric $LiNbO_3$ crystals.[5] Nevertheless, an improvement of sensitivity is expected with near-stoichiometric $LiTaO_3$:Fe crystals, as is an improvement of the dynamic range, which here is already comparable with that of near-stoichiometric $LiNbO_3$.[5]

In conclusion, we have demonstrated that holograms can be recorded by a two-step process in congruent reduced $LiTaO_3$:Fe by use of cw laser light intensities. Blue or white gating light excites electrons from Fe^{2+} to Ta_{Li}^{5+} antisite defects, forming small polarons (Ta_{Li}^{4+}). Red writing light redistributes electrons from these small polarons to Fe^{3+} ions. Erasure effects during readout of the hologram with red light are almost 5 orders of magnitude smaller than erasure effects with the gating light. The dynamic range can be increased by use of a larger writing intensity, and the sensitivity can be optimized in a wide range of the gating intensity. Further improvements in the recording performance are expected with near-stoichiometric $LiTaO_3$:Fe crystals.

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