Volume holographic recording and storage in Fe-doped $LiNbO_3$ using optical pulses*

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(Received 4 June 1973)

Volume holographic recording and storage in Fe-doped $LiNbO_3$ using single 30–75-nsec duration optical pulses at 694.3 and 531 nm from Q-switched ruby and frequency-doubled Nd:glass lasers, respectively, is reported. The recording sensitivity for a pulsed writing source is found to be better than that estimated for a cw source. A sensitivity of 2 mJ/cm^2 at 476 nm and 2.5 mJ/cm^2 at 488 nm to record a hologram of 1% diffraction efficiency is the best sensitivity figure yet reported. The orders of magnitude of improvement in sensitivity is attributed to higher fractional concentration of Fe^{2+}.

Ferroelectric materials have recently become the subject of considerable interest on account of their potential use in optical storage and information processing. Such a ferroelectric storage medium must meet certain system requirements, which include high bit density storage capability, high sensitivity and speeds of recording, and high readout efficiency. In this work we discuss two of these parameters, speed and sensitivity of iron-doped $LiNbO_3$, which make it a most interesting storage medium in addition to strontium barium niobate (SBN). The results of the experiments conducted demonstrate the feasibility of volume holographic recording, storage, and retrieval of information by means of single optical pulses of nanosecond duration using the photoinduced index of refraction changes in lithium niobate. Here we report the best recording sensitivities yet published—an order-of-magnitude improvement over recent reported sensitivities of SBN[1] and almost two-orders-of-magnitude improvement in Fe-doped $LiNbO_3$.[2] These sensitivities are for the experiment conducted without any external electric field across the crystal. Further improvements in sensitivity are expected with external bias fields. Furthermore, our experiments indicate that the charge generation and transport processes occur with time constants considerably faster than 10^{-8} sec since no difficulty in recording and reconstructing was experienced using pulses from Q-switched lasers. The recording sensitivity for a pulsed source is slightly better than that estimated for a cw source.

Volume holograms in the form of interference fringes of two plane waves were recorded in two iron-doped $LiNbO_3$ crystals. Both these crystals were cut from the same boule grown by Crystal Technology, Inc. and contained 0.05 mole% concentration of iron. One of these crystals was annealed in pure oxygen atmosphere at 700 °C to increase the transmission in the shorter-wavelength region of the visible spectrum. The absorption spectra of both these crystals (1 mm thick) are shown in Fig. 1. In the same figure an absorption spectrum corrected for comparable crystal thickness from previously reported data[2] is plotted. The writing sources included single pulses from a Q-switched frequency-doubled Nd:glass laser capable of an output of 0.005 J at 531 nm with pulse duration of ~75 nsec, a ruby laser with 0.05 J energy at 694.3 nm with pulse duration of 30 nsec, and in addition a continuous wave argon laser using output at 476.5, 488, 496.5, and 514.5 nm and a He-Ne laser (632.8 nm). All these sources were made to operate in a single transverse mode. The recording of the hologram and the reconstruction was accomplished using an experimental setup shown in schematic form in Fig. 2. The recording beam is split into a reference and an object beam of approximately equal intensity using a beam splitter and is lightly focused using a 125-cm-focal-length lens. These beams intersect in the storage medium at an angle of 12°. The writing beams are polarized perpendicular to the c axis of the crystal and the plane of the writing beams. The diffraction efficiency and the writing curve are measured using the reference beam with the object beam shuttered intermittently during the recording process and also by a He-Ne laser polarized parallel to the c axis of the $LiNbO_3$ crystal. The diffraction efficiency in the pulsed mode is measured by simultaneously monitoring the transmitted and diffracted reference beams. Optimum reconstruction is observed when the reading beam is polarized parallel to the c axis which is in agreement with the previously reported observations.[3] The recording sensitivity defined in terms of the total required exposure in J/cm^2 to construct a hologram with 1% diffraction efficiency[4] is plotted in Fig. 3 for various recording wavelengths available from the various pulsed and cw sources for both untreated and annealed crystals. The results and conclusions obtained from the experimental data are as follows:

(i) Volume holographic information recording, stor-

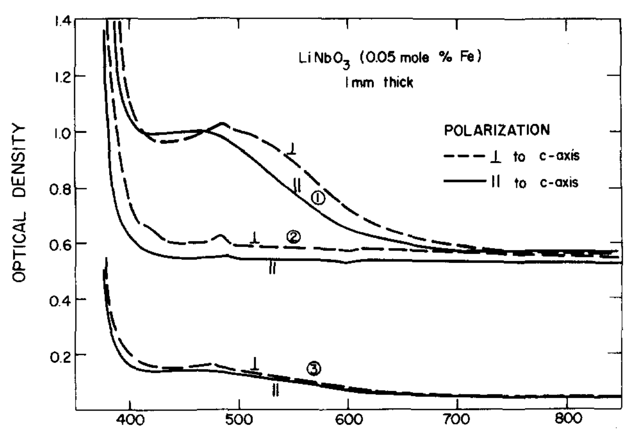

FIG. 1. Absorption spectra of 0.05 mole% iron-doped $LiNbO_3$ for (1) 1-mm-thick unannealed crystal and (2) 1-mm-thick annealed crystal. Curve (3) shows previously reported RCA spectral data corrected to a 1-mm-thick crystal for comparison.

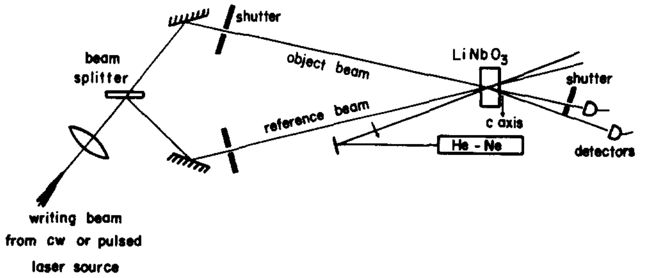

FIG. 2. Experimental arrangement for volume holographic storage. The writing beams used were from a cw argon laser (476.5, 488, 496.5, 514.5 nm), He-Ne laser (632.8 nm), and from a Q-switched frequency-doubled Nd:glass and ruby laser at 531 and 694.3 nm, respectively.

age, and retrieval has been accomplished in photorefractive crystals such as $LiNbO_3$ in times of 30 nsec, only limited by the duration of the optical source.

(ii) The recording sensitivities obtained for a pulsed writing source are slightly higher than the values estimated from sensitivity as a function of wavelength using cw sources at the other wavelengths.

(iii) The 1-mm-thick 0.05 mole% iron-doped $LiNbO_3$ crystal (not annealed) with a significant absorption band near the band edge had a recording sensitivity of 2.0 $mJ/cm^2/\%\eta$ at 476 nm and 2.5 $mJ/cm^2/\%\eta$ at 488 nm. These figures show that the crystal is at least 4 times more sensitive than the most sensitive strontium barium niobate crystal[1] and 60 times more sensitive than the previously reported most sensitive Fe-doped $LiNbO_3$.[2] In fact these sensitivities are even more significant if one considers the smaller interaction lengths of the crystal used in our experiments.

(iv) The storage with 10^{-8}-sec duration pulses indicates that the over-all time constant of recording process contributed by photoionization charge transport and retrapping is much shorter than the pulse duration times. Shorter pulses—such as picosecond duration—must be used to try to determine the dynamic response characteristics and ultimate speed limitations on the recording process.

(v) Increased sensitivity of the medium at longer wavelengths 632.8 nm and even at 694.3 nm is of considerable practical value since such storage material can be used with inexpensive low-power optical sources such as He-Ne lasers.

(vi) Comparison of the recording sensitivities and the absorption spectra of the two crystals cut from the same boule and heat treated after the growth conclusively indicates that the absorbing Fe^{+2} impurities play an important role in the recording sensitivity of the medium. The improvement in sensitivity for the unannealed crystal is by a factor of 100 over that of the annealed crystal characterized by the lower absorption. The unannealed crystal is also considerably more sensitive than the previously reported iron-doped $LiNbO_3$ of comparable doping concentration.[2] The improvement over the annealed crystal and the other reported

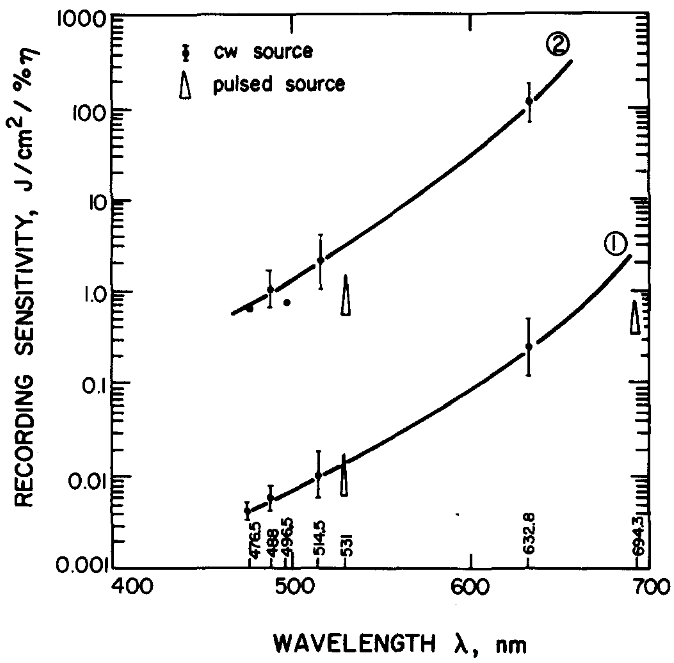

FIG. 3. Recording sensitivity measured in terms of energy required to write holograms capable of diffracting 1% of transmitted beam at 632.8 nm as a function of recording wavelength plotted for 0.05 mole% Fe-doped crystals; curve (1), 1-mm-thick unannealed and curve (2), 1-mm-thick but annealed. The required recording exposure (open triangles) for pulsed writing source is also plotted for these crystals.

results is significantly more than can be explained by only the increased absorption at writing wavelengths. Other processes in addition to photoionization, such as charge transport and retrapping, must play an additional role in determining the sensitivity. It is hypothesized that in addition to the above-mentioned effect the photoexcited carriers must be retrapped selectively to produce a space charge. This indicates that in addition to the absolute concentration of Fe^{+2} ions, the concentration of Fe^{+3} accepter ions and the relative concentration of the two ions must play a significant role in the improvement of the sensitivity. This point is also supported by the fact that the sensitivity data in Fig. 3. show that the ratio of the sensitivities of the two crystals seems relatively constant over the range of the studies.

*Work supported by the National Aeronautics and Space Administration.

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[4]The diffraction efficiency η is defined in terms of a fraction of transmitted light that is diffracted reconstructing the original information and is a function of interaction length L, change in index of refraction Δn, reading wavelength λ, and angle of incidence θ. All the diffraction efficiencies are either measured or corrected for a reading beam of wavelength $\lambda = 632.8$ nm using $\eta_\lambda = \sin^2(\pi \Delta n L / \lambda \cos\theta)$.