aged (10.6  $\mu$ m) in the front focal plane of lens No. 1 as before. Kodak spectroscopic film Type I-M was placed at the back focal plane of lens No. 2 ( $f_2 = 500$  mm). The typical exposure time used was between 100 to 150 sec. Reduced intensity at the edge of images (Figs. 4 and 5) is partly due to phase mismatch³ and partly due to the intensity taper of 10.6- $\mu$ m illuminating beam. The recorded image at the sum frequency clearly indicates resolution down to the 3.56-line-pair/mm target element. The measured magnification of the upconverted image was found to agree reasonably well with the theory. 6-9

The experimental work described above supports the theoretical work on parametric image upconversion. It also demonstrates that a practical parametric image upconversion can be operated nearly diffraction limited.

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## Optically Erasable and Rewritable Solid-State Holograms\*

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Optical holographic storage in single-crystal LiNbO $_3$  is described which can be optically erased at room temperature and then rewritten with no degradation in efficiency or writing rate. The diffraction efficiencies associated with this process are about  $10^{-4}$ . Some variations from previously reported results include a lack of a threshold power density for writing, very-long-term persistence of the stored hologram, and a lack of a dependence of the diffracted intensity on the polarization of the readout beam.

Optically induced refractive-index changes in LiNbO<sub>3</sub> have recently been used as a mechanism for storing information holographically. Exposure of LiNbO<sub>3</sub> (as well as certain other ferroelectric crystals) to interfering light beams of the appropriate wavelength produces a volume hologram directly. Some additional results on lithium niobate solid-state storage are reported here including sequentially optically erasing and rewriting holograms in a sample with complete reproducibility and no loss of efficiency on successive writings. Therefore, in this case, there is no necessity to thermally erase the stored holograms by heating the crystal to 170 °C. This feature may be a very important consideration in achieving rapidly changeable high-density optical information storage.

A nominally pure poled-single-domain single crystal of  ${\rm LiNbO_3}$  was studied for optically induced refractive-index changes with the experimental configuration shown in Fig. 1. The two beams intersect in the crystal and form interference fringes in the geometry of vertical planes. This sinusoidal variation in light intensity across the crystal becomes written into the crystal in the form of index-of-refraction changes thus forming a thick diffraction grating. Holograms of an alphabetic character were also written by inserting a transparency in the object beam. The c axis of the crystal was oriented perpendicularly to the bisector of the writing beams and in the plane of the object and reference beams. The polarization of the writing beams for these experiments was in the plane of the beams, but as pre-

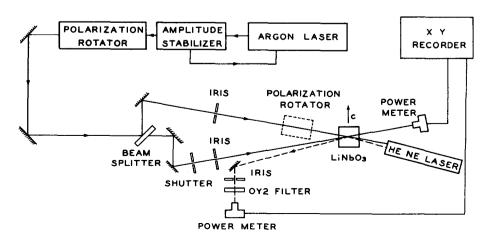


FIG. 1. Experimental configuration.

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<sup>&</sup>lt;sup>†</sup>Some of this work has been submitted to the faculty of Polytechnic Institute of Brooklyn as part of a dissertation in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Electrophysics.

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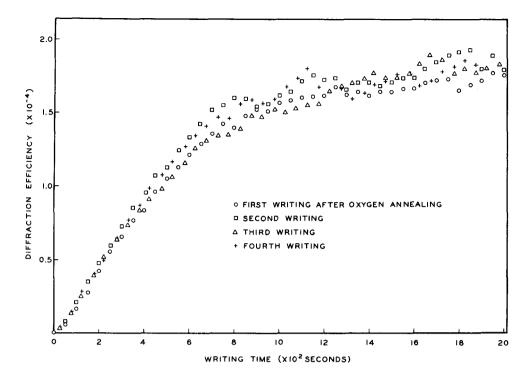


FIG. 2. Writing and rewriting characteristics of LiNbO<sub>3</sub> at a wavelength of 5145 Å using a total power density of 1.09 W/cm<sup>2</sup>.

viously reported, 1,2 changing the polarization of the writing beams did not change the storage process.

An argon-ion laser operating at 5145 Å was used throughout these experiments for writing and erasing. Two methods were utilized to measure the diffraction efficiency: (a) shuttering the object beam and reading the diffracted power in the direction of the object beam, and (b) continuously monitoring the diffracted power of a helium-neon laser operating at 6328 Å and aligned to its corresponding Bragg angle.

The maximum diffraction efficiency  $\eta_{\text{max}}$  in these experiments was about 0.03%. This corresponds to a change

in index of refraction,  $\Delta n$ , of  $6\times 10^{-7}$  for this experimental geometry and wavelength. The writing rate  $\Delta\eta/\Delta t$ , as given by the initial slope of the  $\eta$ -vs-t curve, increased linearly with total power density over the range from 0.023 to 4.5 W/cm² with a writing rate per unit power density  $\Delta\eta/\Delta t/P$  of  $3\times 10^{-7}/\text{sec/W/cm²}$ . The total energy required to achieve maximum diffracted power was about 850 J/cm². When the 6328-Å laser was substituted and used for writing, the writing rate per unit power density was  $5\times 10^{-9}/\text{sec/W/cm²}$  and the maximum observed diffraction efficiency was  $2\times 10^{-6}$ , much smaller values than for the 5145-Å case. No threshold for writing was observed at 5145-Å for a total power density down to 0.023 W/cm².

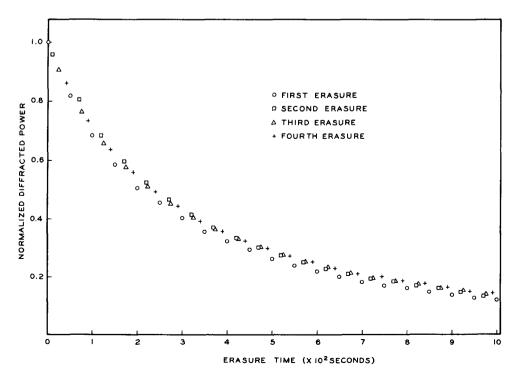


FIG. 3. Optical erasure and re-erasure characteristics of  $\text{LiNbO}_3$  at a wavelength of 5145 Å using a reference-beam power density of 0.59 W/cm<sup>2</sup>.

Appl. Phys. Lett., Vol. 20, No. 1, 1 January 1972

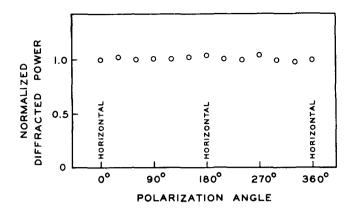


FIG. 4. Dependence of diffracted power on the polarization of the readout beam.

Following the results reported by Chen, LaMacchia, and Fraser, 1 it was found that annealing the crystal above 170 °C in an oxygen atmosphere completely erased all holographic storage. In addition, it was found that optical erasure, by allowing only the reference beam to illuminate the crystal, could be followed by rewriting with no loss of writing rate or efficiency, a process which has not been previously reported. Figures 2 and 3 show the writing and erasing characteristics obtained by sequentially optically writing-erasing-writing-erasing, etc., in the same region of the crystal. No trends in the small changes of writing rate, maximum diffraction efficiency, or erasure characteristics were observed. At each step in the process the experiment was allowed to continue until final saturation or complete erasure was obtained (longer time scales than shown in Figs. 2 and 3). At least 20 write-erase cycles over a range of power densities were obtained with no detectable fatigue.

The grating holograms written in this manner exhibited a strong angular selectivity due to their volume nature. An angular half-power width of 1.4 mrad was observed for this 0.5-cm-thick crystal in comparison to a theoretical value³ of  $\Delta\theta \approx 0.87L/t = 0.59$  mrad, where L is the grating spacing and t is the length of the interaction region.

The dependence of diffracted intensity on reference-beam polarization as reported in  ${\rm LiNbO_3}$  (Ref. 1) and  ${\rm Ba_2NaNb_5O_{15}}$  (Ref. 4) was not observed. A variation of about 5% as shown in Fig. 4 was measured using a smaller diameter reference beam for readout than was used for writing. This was done to reduce the problems of small-scale beam deflection associated with slightly nonparallel surfaces in the polarization rotator as it was rotated. This experiment was immediately followed by optical erasure at high intensity to verify that only diffracted light was being observed.

The optically induced holograms were observed to exhibit relatively long lifetimes. For the conditions of room temperature and storage in darkness, less than 10% degradation in diffraction efficiency was measured after 276 h. This is apparently longer than the previously observed persistence, though the conditions of the two experiments being compared were not the same. Exposure to 0.33 W/cm² of 6328-Å light caused the diffraction efficiency to decrease to 1/e of its original value in  $7.0 \times 10^3$  sec.

In summary, additional experimental results are reported on volume holographic storage in  $\text{LiNbO}_3$  via induced changes in the index of refraction. Many of the observed characteristics are similar to those previously reported with the notable exceptions of (a) repititive optical erasing and rewriting, (b) no apparent threshold writing power density, (c) a lack of dependence of diffracted power on readout polarization, and (d) long-term persistence.

## A New Ferroelectric: Semicarbazide Hydrochloride

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Dielectric measurements on a single crystal of semicarbazide hydrochloride show three maxima at temperatures of -230, 19, and 21 °C. Initial data for the highest peak clearly suggest a ferroelectric-paraelectric transition with a Curie temperature of 21 °C. In the paraelectric region the Curie law is well followed.

At about 20-21 °C, semicarbazide hydrochloride:  $H_2NCONHNH_2\cdot HCl$  crystallizes in the orthorhombic system, space group  $P2_12_12_1$ , and presents a ribbon-

like structure made up of protonated semicarbazide cations and of chloride anions. In addition to the electrostatic forces between these ions, a three-dimen-

<sup>\*</sup>Work supported by the National Aeronautics and Space Administration.

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