Sensitivity and Fatigue of LiTaO$_3$ for Holographic Recording*

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Abstract

The sensitivity of crystals of LiTaO$_3$ to hologram formation has been observed to vary with impurity concentration from a value comparable to the most sensitive doped LiNbO$_3$ for an impure crystal to a value more than five orders of magnitude smaller for a purer crystal. Fatigue effects were observed upon write-erase cycling. These effects were dependent upon writing and erasure polarization and power and could be virtually eliminated by proper choice of optical parameters.

Work by Chen$^1$ has indicated that LiTaO$_3$ is less susceptible than LiNbO$_3$ to laser induced refractive index changes. Other workers$^2$ have found some LiTaO$_3$ crystals to be insusceptible to laser damage. Our measurements indicate that, although a pure LiTaO$_3$ crystal is less sensitive to phase hologram formation than pure LiNbO$_3$,$^3$ impurities in LiTaO$_3$ can increase the sensitivity to a value comparable to that of the most sensitive doped LiNbO$_3$.$^4$ In LiTaO$_3$ the sensitivity to hologram formation was observed to decrease after repeated cycles of writing and optical erasure, i.e.
to fatigue. The sensitivity to hologram erasure was constant.

Proper choice of writing and erasure polarization and power results in virtual elimination of fatigue and is an important consideration in the use of LiTaO₃ as a medium for optical information storage.

Poled single crystals of LiTaO₃ were placed in the apparatus shown in Fig. 1. The argon ion laser was frequency stabilized by use of an internal etalon and was tuned to 514.5 nm., 488.0 nm., or 457.9 nm. The 632.8 nm. He-Ne read laser beam was incident on the crystal at the Bragg angle with its polarization such that the electric field is in the plane of the beam and the crystal c axis (denoted as $\vec{E} \parallel c$). The crystal c axis was in the plane of the two argon-laser write beams and normal to their bisector. Off-Bragg optical erasure was accomplished by moving a mirror until it intercepted the argon-laser beam and directed it onto the crystal at near normal incidence. The polarization rotators allowed the use of any polarization for both writing and erasure.

Two 0.5 cm. thick crystals of LiTaO₃ were used. One crystal had a higher impurity content than the other. A difference in the optical absorption of the two crystals was observed. The absorption edge was less sharply defined and was shifted toward the visible in the impure sample. This resulted in the impure crystal having a yellow color as contrasted to the purer crystal which was clear. Preliminary efforts to employ electron paramagnetic resonance to help identify the responsible defect(s) were inconclusive.
The sensitivity of a crystal to hologram formation was defined as the maximum slope of the diffraction efficiency vs. exposure curve and had units of %/joule/cm². The sensitivity of both crystals was measured for a writing wavelength of 488.0 nm. The less pure crystal had a sensitivity more than five orders of magnitude greater than the purer crystal whose sensitivity was $2 \times 10^{-5} \%$/joule/cm² and whose maximum diffraction efficiency was less than 0.02%. The purer crystal was not studied further because of its low sensitivity.

The initial sensitivity was defined as the sensitivity measured for the first writing exposure after the crystal had been thermally erased at 300°C for 2 hours. The initial sensitivity of the less pure crystal was found to be independent of the writing polarization. For a writing wavelength of 488.0 nm, the sensitivity increased with power from a value of $5.8 \pm 1.4 \%$/joule/cm² at 0.3 watts/cm² to a value of $10 \pm 1 \%$/joule/cm² at 7.5 watts/cm². At low writing power (0.3 watts/cm²) the initial sensitivities were $0.78 \pm 0.06$, $5.8 \pm 1.4$, and $8.9 \pm 0.9 \%$/joule/cm² for the writing wavelengths 514.5 nm, 488.0 nm, and 457.9 nm, respectively. The maximum diffraction efficiency was between 70 and 80 percent for all powers, polarizations, and wavelengths used.

The response of the crystal to repeated cycles of writing and optical erasure was measured at a wavelength of 488.0 nm.
For all combinations of writing and erasure power and polarization the qualitative response of the crystal was the same. The writing sensitivity measured after optical erasure was less than the initial writing sensitivity and this sensitivity continued to decrease through several write-erase cycles. After three or four write-erase cycles the writing sensitivity reached a stable value, called the fatigued sensitivity, which was constant through all subsequent write-erase cycles.

Quantitative measurements of the fatigued sensitivity were made. Variations of up to a factor of fifty in the fatigued sensitivity were produced by changes in writing and erasure polarization and power. There were no significant changes in the erasure sensitivity associated with the changes in writing sensitivity.

Figure 2 shows the dependence of the fatigued sensitivity on the writing power for a constant erasure power of 0.3 watts/cm$^2$. The writing beams were polarized $\vec{E} \perp c$. The two curves are for erasure polarizations $\vec{E} \perp c$ and $\vec{E} \parallel c$. The fatigued writing sensitivity is seen to be dependent upon the erasure polarization and to increase with increased writing power. Although not shown by Fig. 2, the fatigued sensitivity was observed to decrease with increased erasure power. When the writing and erasure powers were equal there was an increase in the fatigued sensitivity
with increased power as shown in Table I. The degradation of the sensitivity upon cycling can be virtually eliminated by choosing the writing polarization $\mathbf{E} \parallel c$, the erasure polarization $\mathbf{E} \perp c$, and by using higher powers. The fatigued sensitivity at 7.5 watts/cm$^2$ for this combination of polarizations was $8.2 \pm 0.3 \%$/joule/cm$^2$; the initial sensitivity was $10 \pm 1 \%$/joule/cm$^2$.

The sensitivity was found to be dependent only upon the writing and erasure conditions for the previous three or four cycles. Optical treatment prior to the last few cycles had no effect on the sensitivity.

No scatter of either the write or read beams due to large scale refractive index inhomogeneities was observed after a total exposure of 50,000 joules/cm$^2$.

Holograms in LiTaO$_3$ were observed to decay less than 1% per day at room temperature while exposed to a 1 milliwatt 632.8 nm laser beam.
References

*Work supported by the National Aeronautics and Space Administration.


5 Purchased from Union Carbide Corporation, Crystal Products Department, Cleveland, Ohio.

6 Purchased from Crystal Technology, Inc., Mountain View, California.
Table I. Fatigued sensitivity values for various powers and polarizations for a write and erase wavelength of 488.0 nm.

<table>
<thead>
<tr>
<th>Power Write &amp; Erase (watts/cm²)</th>
<th>Polarization Relative to Crystal c Axis</th>
<th>Fatigued Sensitivity (%/(joule/cm²))</th>
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Figure Captions

Figure 1. Experimental Apparatus.

Figure 2. Experimental writing power dependence of the fatigued sensitivity of LiTaO$_3$ for two erasure polarizations. Writing polarization: $\vec{E} \perp c$. Erasure power: 0.3 watts/cm$^2$. Write and erase wavelength: 488.0 nm.
Figure 1.
ERASURE POLARIZATION RELATIVE TO CRYSTAL C AXIS

Sensitivity (%/(Joule/cm²))

Writing Power (Watts/cm²)

Figure 2.