Laser Induced Damage, Nonlinear Absorption, and Doubling Efficiency of $LiIO_3$

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Abstract—Laser-induced damage thresholds of single crystal $LiIO_3$ have been studied using picosecond pulses at 1.06 μm and 0.53 μm. These thresholds depend on wavelength, on crystal orientation, and on the number of times the sample has been irradiated. In addition, the doubling efficiency at high irradiance levels was observed to be a decreasing function of irradiance beyond a critical value. We present evidence to show that this results from the onset of optical parametric down conversion. In separate nonlinear transmission studies, reversible nonlinear transmission of 1.06 μm light was measured, and in self-diffraction experiments, both reversible and irreversible optically-induced complex index of refraction changes at 0.53 μm were observed.

I. Introduction

IT IS well known that the large nonlinear coefficient of lithium iodate ($LiIO_3$) makes it an attractive candidate for applications where second harmonic generation is required [1]. Consequently, we have begun a study of the laser-induced damage (LID) thresholds of this material and of the mechanisms that limit its second harmonic conversion efficiency. Here, we report the results of five separate but related experiments in this area. In the first of these (Section II), we measure the laser-induced damage thresholds of single crystal $LiIO_3$ using picosecond pulses at 1.06 μm and 0.53 μm. We find that the LID thresholds vary with wavelength, pulsewidth, crystal orientation, and the number of times that the sample is irradiated. Specifically, we observe that the sample is more easily damaged when green (0.53 μm) light is present and that the damage is initiated by some nonlinear absorption process, which is more efficient at 0.53 μm. The sample also damages more easily with repeated irradiations at both wavelengths. We thus obtain a single shot and a multishot threshold. Next (Section III), we determine the dependence of the second-harmonic conversion efficiency on incident 1.06 μm irradiance. We find that the efficiency initially increases with excitation level to a maximum of approximately 50 percent and then decreases. This decrease is consistent with any of three mechanisms: (a) any absorption of the second harmonic or nonlinear absorption of the fundamental, (b) a nonlinear refractive index change that destroys the exact phase-matching conditions, or (c) the onset of parametric down conversion. In an effort to identify the mechanism limiting the conversion efficiency and to identify the nonlinear mechanism responsible for the onset of damage, we perform three related studies. We first measure (Section IV) the nonlinear transmission of $LiIO_3$ at both 0.53 μm and 1.06 μm (under nonphase-matched conditions). We observe multiphoton absorption of the 1.06 μm radiation (the order appears to be greater than four) at irradiances well above the multishot threshold for damage. No nonlinear absorption at 0.53 μm is resolved up to the multishot damage threshold for green light, also the limit of our laser output. This threshold is considerably lower in irradiance than the 1.06 μm threshold. The onset of the observed nonlinear absorption of the fundamental occurs at too high an irradiance to account for the observed decrease in the conversion efficiency. Although no nonlinear absorption or index changes are observed at 0.53 μm in the transmission studies just described, both reversible and irreversible changes in the complex refractive index are observed at 0.53 μm by using a more sensitive background-free two-pulse self-diffraction technique (Section V). Finally, (Section VI), we measure the dependence of the spatial beam profile of the second harmonic on the fundamental irradiance. From the distortion in the 0.53 μm beam profile for large irradiances we conclude that optical parametric down conversion is responsible for the decrease in conversion efficiency with increasing irradiance.

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II. Damage Thresholds

The initial experimental arrangement to be used in damage and conversion efficiency experiments is shown in Fig. 1. The laser source was a passively mode-locked 1.06 μm Nd : YAG laser that produced Gaussian spatial mode pulses of temporal width externally variable between 40 and 200 ps (FWHM). Details of the experimental apparatus are given in [2]. The laser beam traversed the $LiIO_3$ with a uniform beam radius of 440 μm (all spot sizes are quoted as the half width at the e^{-2} point in irradiance). When 0.53 μm light was required, a second doubling crystal (KD * P) was inserted prior to the sample. The collimated 0.53 μm beam had a spatial width of 310 μm in the $LiIO_3$. In this case, residual 1.06 μm light was removed with polarizers and 1.06 μm blocking filters.

The laser-induced damage (LID) thresholds for $LiIO_3$ were measured at 1.06 μm and 0.53 μm for various crystal orientations and pulsewidths. Both the single shot or 1 on 1 thresholds (i.e., each site irradiated only once) and multiple shot or N on 1 thresholds (i.e., each site irradiated many times with irradiance levels well below the single-shot damage threshold) were mea-

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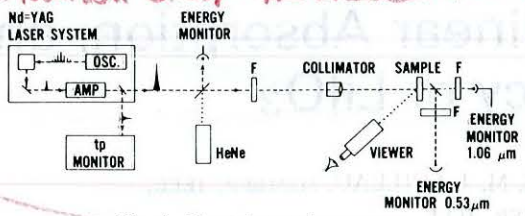


Fig. 1. Experimental apparatus.

sured. In all cases the light was collimated to determine practical limitations of use. In addition this configuration makes the calculation of irradiances trivial. The onset of damage was determined by observing both increased scattering of coaxial HeNe light and by observing other visible sample changes with a long working distance microscope. In $LiIO_3$, both damage signatures occurred simultaneously. The LID threshold is defined as that fluence or irradiance which produces visible damage with 50 percent probability as determined by the method of Porteus *et al.* [3]. The experimental uncertainties in the LID threshold measurements are indicated by the dotted lines in Tables I and II, and they include the relative uncertainity as determined in [3], absolute energy calibration error and the uncertainity in the spot size measurements.

The results of the damage measurements using 1.06 μm light are presented in Table I. The LID threshold was measured for 45 ps and 120 ps (FWHM) pulses. Both the fluence and the corresponding irradiance threshold are shown. The single shot (1 on 1) LID threshold fluence for front surface damage to the sample was approximately 1.3 J/cm^2 for both pulsewidths. For a beam radius that is constant throughout the sample, one would normally expect to see rear surface damage at lower fluence levels than front surface damage since the field at the exit surface should be approximately 30 percent greater than at the front surface for transparent samples. However, in this case (as we shall discuss in Section IV), depletion of the beam by nonlinear processes reduced the fluence at the rear surface by as much as a factor of 8 for input irradiance near the LID threshold levels. As expected, the surface damage thresholds were independent of crystal orientation (i.e, whether or not the crystal was phase-matched).

Because single shot damage first occurred on the sample front surface, the single shot bulk damage threshold could not be determined. However, approximately 20 percent of the shots at a fluence of 1.3 J/cm^2 resulted in bulk damage just below the front surface. From these measurements, we estimate a lower limit for the bulk, single shot, damage threshold to be 1.3 J/cm^2.

Table I also contains the results of the multiple shot or N on 1 measurements for 1.06 μm light for both phase-matched and nonphase-matched conditions. For the nonphase-matched (NPM in Table I) configuration, the $LiIO_3$ crystal was rotated about the laser beam propagation axis to an orientation 90° from the phase-matched orientation. In this configuration, no 0.53 μm light was visible. Each site was irradiated at levels far below the single shot threshold and the irradiance was slowly increased until damage was observed. Multiple shot damage was always initiated in the bulk, and the thresholds were determined to be substantially below the lower limit of 1.3 J/cm^2 found for the 1 on 1 experiments. Maximum lowering of the LID threshold was achieved after approximately 50 irradiations

TABLE I
DAMAGE THRESHOLDS FOR $LiIO_3$ AT 1.06 μm FOR SINGLE IRRADIATIONS (UPPER) AND MULTIPLE IRRADIATIONS (LOWER) GIVEN IN BOTH FLUENCE AND IRRADIANCE PM AND NPM INDICATE THAT THE SAMPLE WAS ORIENTED IN A PHASE MATCHED OR NONPHASE MATCHED GEOMETRY RESPECTIVELY

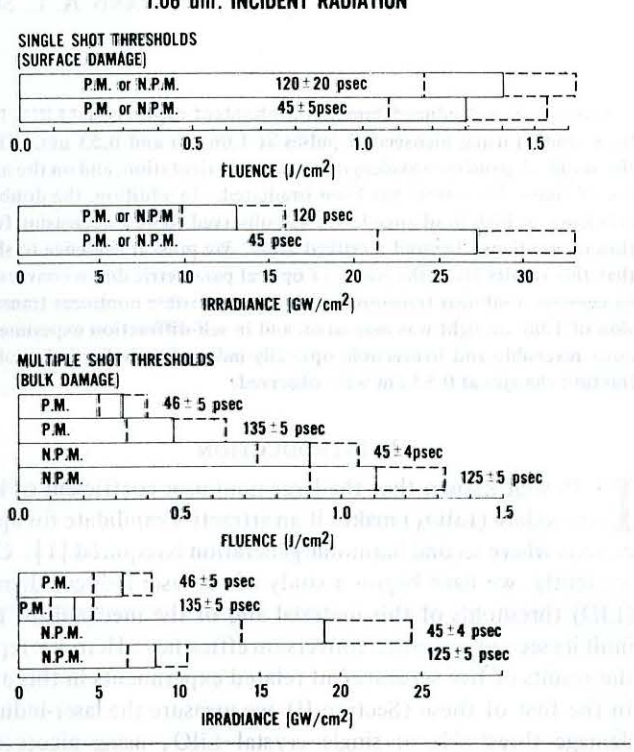
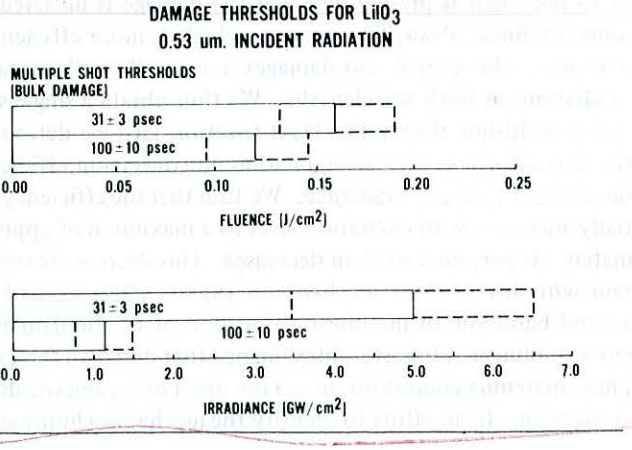


TABLE II
DAMAGE THRESHOLDS FOR $LiIO_3$ AT 0.53 μm FOR MULTIPLE IRRADIATION GIVEN IN BOTH FLUENCE AND IRRADIANCE



at 0.2 J/cm^2. Notice that the N on 1 thresholds are considerably lower for the crystal oriented to produce second harmonic light. Similar multiple shot damage threshold changes have been observed previously at 0.69 μm [4]. The 1.06 μm to 0.53 μm coversion efficiency (see Section III) was of the order of 50 percent for the input irradiance that produced breakdown. These results suggest that the green light may be responsible for damage under phase-matched conditions. To confirm this sug-

gestion, we measured the multiple-shot LID threshold for 0.53 μm radiation. For these measurements, a KD * P second-harmonic crystal was inserted following the Nd: YAG laser, and all residual 1.06 μm light was removed, as described above. Indeed, the results presented in Table II indicate that when 0.53 μm radiation is present, it is primarily responsible for initiating damage. Because of the role of the green light in determining the LID threshold when the crystal is phase-matched, we investigate the dependence of the second harmonic conversion efficiency on irradiance in the next section.

In addition, the lowered threshold for multishot irradiation is indicative of the formation of microscopic defects that eventually absorb enough energy to cause crystal fracture (what we observe as LID). This may be similar to the irreversible absorption changes seen in NaCl at 10.6 μm (Wu *et al.* [5]) or to a charge migration or photorefractive effect reported in other materials such as $BaTiO_3$ [6]. We conclude that these defects must be produced by a nonlinear process since no amount of irradiation at very low irradiance causes a lowering of the damage threshold. Also the defects appear to be more efficiently produced by 0.53 μm light as shown by the much lower multishot threshold at this wavelength. To investigate this supposition, we have also monitored the transmission of both 1.06 μm light and 0.53 μm light (no 1.06 μm light present) as a function of the incident irradiance (Section IV).

III. Doubling Efficiency in $LiIO_3$

In the determination of the damage thresholds at 1.06 μm with the crystal in the angle phase-matched orientation, we also monitored both the transmission at 1.06 μm and the harmonic conversion efficiency (i.e., energy at 0.53 μm divided by incident 1.06 μm energy). Fig. 2 shows both the transmission and conversion efficiency as a function of input 1.06 μm irradiance for 40 ps (FWHM) pulses. Each data point is the average of five laser firings. The five data points at the highest irradiance were taken after damage was observed. Even after damage no large absorption is observed. The efficiency increases rapidly at low irradiance, reaches a maximum at ~3 GW/cm^2, corresponding to an efficiency of ~50 percent, and then decreases for higher incident irradiance levels (although the second harmonic energy continues to increase slowly). An identical experiment was performed using 140 ps (FWHM) pulses that reproduced the data of Fig. 2 up to ~3 GW/cm^2 where the sample damaged. Physical mechanisms that produce a theoretical fit to such a turnover in efficiency include nonlinear absorption of the second harmonic [7], [8], nonlinear refractive index changes that result in loss of phase matching at high irradiance levels [7], [8], and parametric down conversion of the 0.53 μm light [9], [10]. To distinguish the contributions of these separate mechanisms, we performed three related measurements described below.

IV. Nonlinear Transmission Measurements

The transmission of the $LiIO_3$ at 1.06 μm was measured with the sample oriented such that no second harmonic was produced. These data for 45 ps pulses are shown in Fig. 3 as a plot of the inverse third power of the transmission versus the cube of the incident irradiance. The sample was not AR coated such that the intercept of ~1.8 in Fig. 3 is indicative of the Fresnel

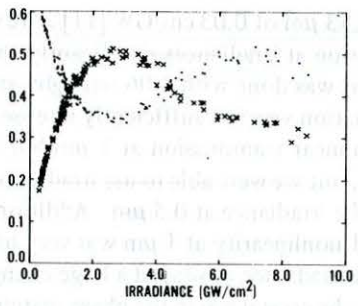


Fig. 2. Second harmonic conversion efficiency (x's) as a function of irradiance, and 1.06 μm transmission (o's) as a function of irradiance.

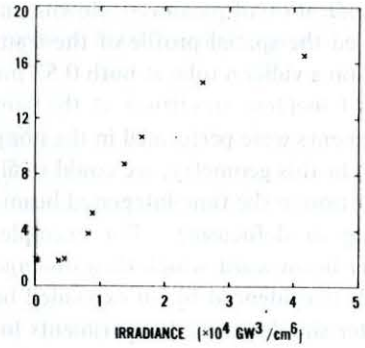


Fig. 3. Inverse cube of the 1.06 μm transmission versus the cube of the incident 1.06 μm irradiance.

losses of a material of index ~1.86. The data are plotted in this manner to investigate whether four-photon absorption might explain the results. Neglecting the Gaussian transverse structure of the beam, an exact solution for beam depletion due to four photon absorption should yield a straight line on such a graph, the slope of which would determine the four-photon absorption coefficient [2]. The line turns up rapidly indicating that the nonlinearity is of an order higher than four. We cannot account for the order of the nonlinearity even when the absorption caused by the subsequent photogenerated carriers is included. It is important that we emphasize that the data points are single laser firings and only a few shots were taken because the sample damaged more easily after each shot. The highest irradiance data point was taken first and the irradiance decreased with each subsequent shot. The data of Fig. 3 are for a single site. A final data point was taken at an increased irradiance to observe any possible hysteresis; none was observed for this small number of laser firings. Note also that several sites had to be irradiated before the data shown was obtained, since many sites damaged on the first shot. All the data shown were obtained at irradiance levels near to or above the multishot damage threshold. (The single shot surface threshold is ~27 GW/cm^2.) These transmission data were taken with a 1.06 μm spike filter in front of the detectors. The possibility of conversion of the 1.06 μm light to other frequencies is not excluded although no visible light was observed.

The transmission of the $LiIO_3$ was also measured at 0.53 μm for both a phase-matched and nonphase-matched geometry. When great care was taken to eliminate all of the residual 1.06 μm light, no nonlinear transmission of the 0.53 μm light was observed up to the multishot damage threshold. These measurements determine an upper limit for the two-photon absorption

coefficient at 0.53 μm of 0.03 cm/GW [11]. We were unable to obtain transmission at irradiances significantly above the multishot threshold as was done with 1.06 μm light, since our source of 0.53 μm radiation was not sufficiently intense. The fact that we observe nonlinear transmission at 1 μm but not at 0.5 μm appears strange, but we were able to use irradiance levels at 1 μm about 6 times the irradiance at 0.5 μm. Additionally, the order of the observed nonlinearity at 1 μm was very high such that a small change in irradiance produced a large change in transmission. Whenever the crystal was in the phase-matched orientation and any residual 1.06 μm was allowed to strike the sample along with the 0.53 μm light, it was amplified depleting the 0.53 μm beam—a clear indication of parametric down conversion [9].

We also studied the spatial profile of the transmitted beams in the far field on a vidicon tube at both 0.53 μm and 1.06 μm as a function of incident irradiance at the same wavelength. These measurements were performed in the nonphase matched configuration. In this geometry, we could easily distinguish a half-wave distortion in the time-integrated beam profile caused by self-focusing or defocusing. For example, self-focusing causes a smaller beam waist which then undergoes greater diffraction, which is evidenced by an expanded beam in the far field. Computer simulation and experiments in materials that do show self-focusing indicate the stated sensitivity to a $\lambda/2$ beam distortion. For a 0.5 cm-thick sample, this means that we should be able to detect a change in index on the order of 10^{-4}. No detectable distortion was observed up to the multishot damage thresholds.

From these two types of nonlinear transmission measurements, we conclude that any induced change in either the absorption coefficient or index of refraction is far too small to account for the turnover in the doubling efficiency as displayed in Fig. 2.

V. Irradiance Dependent Complex Refractive Index Changes

The sensitivity of the nonlinear transmission measurements discussed in the previous section was limited by the large background signal present. That is, we were attempting to measure a very small change in a large signal. In this section, we describe the use of a more sensitive background-free self-diffraction technique to measure both transient and permanent optically-induced changes in the complex index of refraction at 0.53 μm. We emphasize that no such change was observed at this wavelength in the preceding experiments. In this technique, a single picosecond pulse at 0.53 μm was divided into two parts by a beamsplitter. These two pulses were then recombined so that they were temporally and spatially coincident in the $LiIO_3$ at an angle $\theta = 1.2°$. The interference of these two pump pulses spatially modulates the electric field which may cause a periodic change in the complex index of refraction of the sample. If such an irradiance dependence is present, each pump beam will be self-diffracted by this laser-induced grating into two first orders at $\pm\theta$. One first order for each pump beam will be scattered into the direction of the other pump, and one will be diffracted in a background-free direction (which we label $-\theta$). There is no signal at $-\theta$ unless a real or virtual grating is produced by the pump pulses.

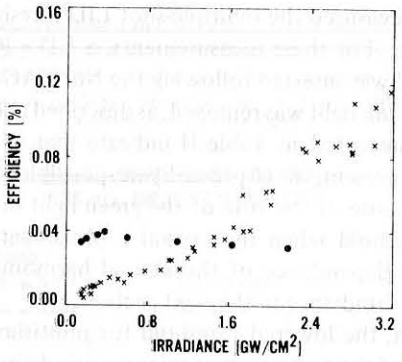


Fig. 4. Diffraction efficiency in a light-by-light scattering experiment versus the irradiance of the 0.53 μm pump beam as explained in Section V. Crosses indicate data taken with irradiance increasing. Dots indicate one beam blocked and irradiance decreasing.

The self-diffraction efficiencies as a function of irradiance are shown in Fig. 4 by the crosses. The crystal was oriented so the 0.53 μm beams were incident near the phase-matched condition for down conversion. The irradiances recorded in Fig. 4 are for one of two equally intense pump beams. Following these measurements, we subsequently blocked one pump beam while continuing to measure the diffraction efficiency of the other pump. The results are the solid dots in Fig. 4. Clearly, a permanent component to the grating has been produced that continues to diffract light when the modulation of the intensity has been removed. As expected, the diffraction efficiency of the permanent grating is independent of pump irradiance. This grating was not erased by irradiating with a single beam as occurs with photorefractive materials such as $BaTiO_3$ [6].

We performed similar experiments at other orientations with various results. For example, with the crystal rotated by an angle $\alpha = 78°$ about the bisector of the angle between the two pump beams, away from the phase matched orientation, the diffraction efficiency is a maximum and no permanent grating is observed. In addition, the efficiency is nearly linear with irradiance. The dependence of the diffraction efficiency on the orientation angle α is shown in Fig. 5.

Subsequent examination of the $LiIO_3$ sample with a Nomarski microscope revealed that permanent gratings were formed both on the front surface and the rear surface of the crystal by the 0.53 μm pulses. The gratings were faint even under Nomarski illumination although those on the rear surface were notably sharper than those on the front surface due to the field enhancement at the rear surface. Permanent gratings in the bulk of the material were observed only in association with bulk damage sites. The surface gratings, by themselves, cannot explain the observed orientational dependent permanent diffraction since gratings of comparable visibility were observed on the surface corresponding to *both* crystal orientations. These gratings were also visible under bright field illumination, but not under dark field illumination.

For this sample thickness (~5 mm) and this grating spacing (~25 μm), we are in the Bragg grating regime. That is, the gratings produced here cannot be considered thin, and the measured self-diffracted signal at $-\theta$ violates the Bragg condition. This makes quantitative analysis of the results of Fig. 4 and Fig. 5 difficult. Although not all features of these data are understood

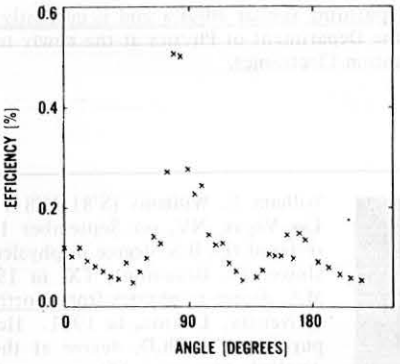


Fig. 5. Diffraction efficiency versus the angle about the beam direction as described in Section V. Both 0.53 μm pump beams were equally intense for this measurement.

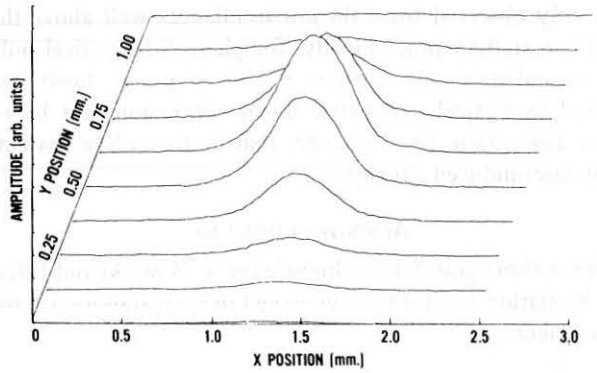


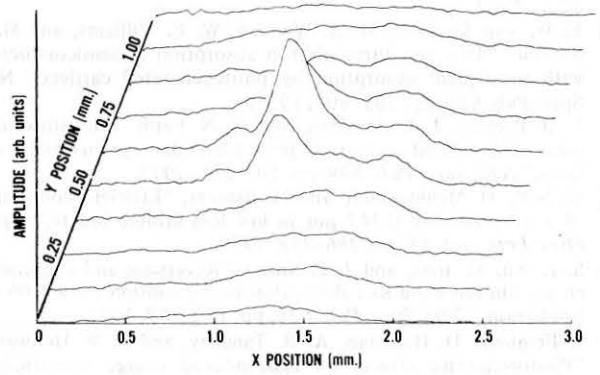
Fig. 6. Two-dimensional spatial beam profile of the second harmonic produced in $LiIO_3$ at low irradiance.

by the authors at this time, it is clear that we have observed permanent and transient index (or absorption) changes in the sample at 0.53 μm. We emphasize once again that these changes are too small to destroy phase match and account for the saturation and turn down in the doubling efficiency as shown in Fig. 2.

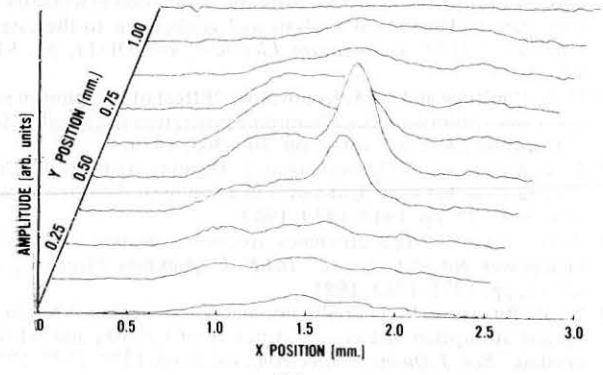
VI. Phase-Matched Second Harmonic Spatial Profiles

Having shown that laser-induced absorptive and index changes (both at 0.53 μm and 1.06 μm) are small, we suspect that the eventual decrease in the doubling efficiency with increasing 1.06 μm irradiance is caused by down conversion. In this section, we show this is indeed so.

In these experiments, we monitored the spatial profile of the second harmonic produced by phase matching the $LiIO_3$ crystal as a function of incident fundamental irradiance. At low incident 1.06 μm irradiance, the profile at 0.53 μm is a smooth Gaussian as shown in Fig. 6. As the 1.06 μm irradiance is increased past the efficiency maximum (as shown in Fig. 2), the profile is distorted as shown in Fig. 7(a). That is, the second harmonic is skewed to one side. This can be understood by recalling that $LiIO_3$ possesses a large walk-off angle (4°) between the fundamental and second harmonic when phase matched. For a fundamental beam radius of 0.44 mm (half width at the e^{-2} point in irradiance) and a crystal length of 5 mm, the two beams (fundamental and second harmonic) will be separated by 0.33 mm at the exit surface. This separation is of the order of the 1.06 μm beam radius. Down-conversion would be expected to be important only in regions where the fundamental and second harmonic beams overlap and where both irradiances are large, i.e., near the rear of the crystal. This would produce a lopsided spatial distribution of second harmonic light, as shown in Fig. 7(a). We would expect then that by rotating the crystal 180° about the incident beam direction that both the walk-off direction and the distortion would be inverted. That this is the case can be seen in Fig. 7(b). From these results, we conclude that parametric down conversion is primarily responsible for limiting the harmonic conversion efficiency [9], [10].



(a)



(b)

Fig. 7. (a) Two-dimensional spatial beam profile of the second harmonic produced in $LiIO_3$ at high irradiance. (b) Two-dimensional spatial beam profile of the second harmonic produced in $LiIO_3$ at high irradiance. The crystal has been rotated 180° about the beam axis from the position used in Fig. 7(a) as described in Section VI.

VII. Conclusions

Laser-induced damage thresholds in $LiIO_3$ have been determined for two pulsewidths (45 and 145 ps), for two wavelengths (0.53 and 1.06 μm), and for phase-matched and nonphase-matched crystal orientations. Multiple shot thresholds were lower than single-shot thresholds for all pulsewidths, crystal orientations and wavelengths studied. In addition, the multiple shot LID thresholds were lower at 0.53 μm than at 1.06 μm, indicating that the laser-induced threshold is lowered by cumulative defects produced by absorption of the 0.53 μm radiation. Self-diffraction experiments confirmed the presence of both reversible and irreversible changes in the material refractive index prior to damage even though no nonlinear absorption was resolvable at 0.53 μm for the maximum irradiances from our system. Higher order nonlinear absorption was, however, observed for 1.06 μm light. We emphasize that this absorption

was only observed for 1.06 μm irradiances well above those available at 0.53 μm. Finally, for picosecond optical pulses, the second harmonic conversion efficiency was shown to be limited by optical parametric down conversion, not by nonlinear absorption, index changes that destroy phase matching, or by laser-induced damage.

Acknowledgment

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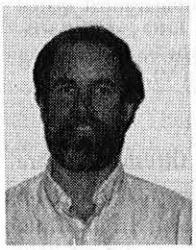

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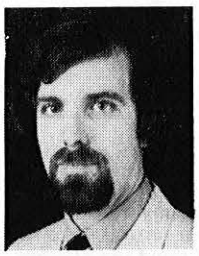

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