Efficient second-harmonic generation in KTP crystals

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Conversion efficiencies in excess of 50% have been measured in frequency-doubling experiments in KTP using divergent fundamental 1.064-μm beams. Various doubling schemes have been investigated to avoid reconversion effects observed in 9-mm-long crystals. The phase-matching angular acceptance bandwidth, damage-threshold observations, and a measurement of effective susceptibility relative to KD*P are also reported.

INTRODUCTION

Many high-average-power (>20-W) solid-state laser systems currently being developed operate more efficiently with multitransverse-mode beam outputs as compared with single-transverse-mode operation. Among these systems are new tunable solid-state materials such as alexandrite,[1] GSGG,[2] and Ti:Al_2O_3.[3] Slab geometry lasers,[4] while showing promise for improved beam quality at high average powers, are likely to generate somewhat divergent outputs. Although applications can often tolerate beam divergences in excess of several times the diffraction limit, there are difficulties encountered in achieving efficient and reliable frequency conversion of multimode fundamental beams into the visible—a region of prime interest in many practical cases. Standard materials such as KD*P do not perform so efficiently as frequency converters of divergent fundamental beams because of their small acceptance angles.

Among various nonlinear crystals with large-angular-bandwidth properties, KTP is highly promising because of its large nonlinear coefficient, low absorption between 0.5 and 1.4 μm, and unusually large temperature bandwidth.[5] The last-named property is highly advantageous for maintaining pulsed energy stability of the converted beam. Recent experiments[6,7] have also shown KTP to be a long-lived nonlinear material not susceptible to statistical bulk damage, as in MgO:$LiNbO_3$, for example, even at relatively high-average-power levels. Experiments utilizing KTP as an intracavity doubler for a quasi-cw Nd:YAG laser have demonstrated that power outputs in excess of 25 W could be generated without damage for 3 mm × 3 mm × 5 mm KTP crystals.[7] Extracavity doubling experiments with KTP have also been encouraging. As much as 60% conversion efficiency of a TEM_{00} mode fundamental has been reported,[8] and greater than 50% conversion of a multimode beam has been demonstrated.[9]

In this paper we report measurements of the phase-matching angular bandwidth and conversion efficiency of multiple-transverse-mode and near-Gaussian diffraction-limited beams in hydrothermally grown KTP. Since we have seen evidence of reconversion of the second harmonic in long crystals with multimode beams, several double-pass schemes were investigated in order to improve frequency-conversion efficiency. We also report on preliminary observations of damage thresholds with single-mode and multimode beams and a measurement of the effective nonlinear susceptibility referenced to KD*P.

EXPERIMENT

The laser source used for all the experiments was a Molectron MY-32 Nd:YAG laser, which has a near-Gaussian transverse-mode intensity distribution and a 0.3-mrad beam divergence. The laser can be operated in single axial mode with a frequency bandwidth of 0.005 cm^{-1} or in conventional Q-switched mode with approximately 0.05-cm^{-1} bandwidth (10 axial modes). The pulse duration can be adjusted by changing the outcoupling ratio and pumping level. The energies of the fundamental and second harmonic were measured using calibrated beam splitters and Laser Precision pyroelectric probes. An external half-wave plate and polarizer permitted variation of the fundamental energy without affecting the laser pulse parameters. A schematic of the experimental setup is shown in Fig. 1.

Acceptance-Angle Measurements

Figure 2 depicts a contour plot of relative conversion efficiency versus external angle of incidence. For these measurements, a 3.4-mm-long uncoated KTP crystal was used, and the laser was operated in a near-Gaussian mode. The incident beam was focused with a 1-m focal-length lens to a spot diameter of 0.38 mm at the crystal. Polarization of the fundamental was set in the typical type-II orientation, and the angles ϕ and θ were referenced to the X axis in the $X-Y$ plane and the Z axis of the crystal, respectively.[10] The center coordinates of the phase-matching contours are θ = 90 deg and $\phi \cong$ 30 deg. Some distortion of the contours may have occurred owing to changes in Fresnel reflection at the air–crystal interface as a function of angle. The peak conversion efficiency was 20% for the results shown in the figure,

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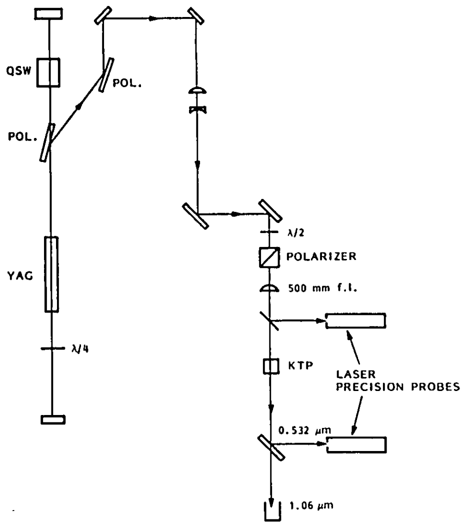

Fig. 1. Experimental setup. The main components of the Molectron laser are on the left and optics for frequency doubling on the right. f.l., focal length; QSW, Q switch; pol.'s; polarizers.

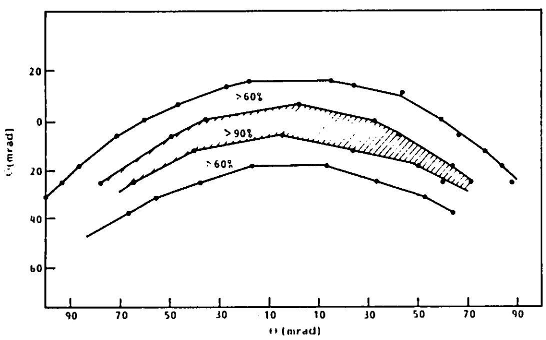

Fig. 2. Contour plot of conversion efficiency versus angle of incidence. For angular coordinates lying between the innermost two curves, a collimated beam is frequency doubled at 90% of the peak conversion efficiency.

and similar results are obtained, within the experimental error, at 4 and 10% peak conversion efficiency. Our results are in agreement with previously reported angular-bandwidth measurements.[8] From these measurements, we infer that a beam with a divergence as large as 12 mrad will be frequency converted at 90% of the level of a comparable collimated beam. The angular acceptance bandwidth at 80% of the peak conversion is 24 mrad in the ϕ direction and 100 mrad in the θ direction.

Multimode Doubling

The Molectron laser was altered to produce a multimode transverse intensity distribution by converting the usual apertured, quasi-stable resonator to a stable resonator configuration. The polarization outcoupling and pumping level were adjusted to optimize output energy, pulse duration, and mode pattern. The goal was to produce a laser pulse with many transverse modes, resulting in a more uniform energy distribution across the beam as well as a longer pulse duration (30 nsec). The latter feature was desired to simulate more closely the output properties of a Nd:glass or alexandrite laser at high average powers. The mode structure appears to be composed mainly of TEM_{00} and TEM_{02}, owing to the gain distribution in the YAG rod, the size of the limiting aperture, and the low pumping level. Divergence of the laser beam, determined by measuring the beam diameter at several distances from the output coupler, was 2.6 mrad for a waist diameter of 0.9 mm.

Some preliminary results are shown in Fig. 3 for 4-mm- and 9-mm-long KTP crystals. These crystals, as well as a 2.5-cm-long KD*P crystal, were compared under identical experimental conditions. All conversion efficiencies indicated in the figure were corrected for the measured losses at the KTP surfaces and the dichroic mirror. We note that there is an uncertainty of up to a factor of 2 in the absolute intensity scale shown in Fig. 3. This is due to the multimode nature of the spatial energy pattern and modulation of the pulse from mode beating. The intensity was calculated assuming a 30-nsec duration from the envelope of spikes in the laser pulse, the measured energy, and the measured beam diameter.

The low conversion efficiency obtained for KD*P is not surprising since the laser divergence is 2.5 times the angular acceptance bandwidth. By contrast, the two KTP crystals investigated are observed to be efficient for second-harmonic generation of a divergent fundamental beam. The conversion efficiency of the 9-mm-long crystal saturates at relatively low incident intensity and then decreases. This effect can be attributed to reconversion of the second harmonic back to the fundamental or to higher-order parametric processes, which produce light in the UV, which is absorbed in KTP. A two-photon absorption process[11] could also explain these results. A measurement of two-photon absorption using light at 532 nm was inconclusive. Two-photon absorption with one photon of 1.06 μm and one of 532 nm could also possibly contribute to the reduced conversion efficiency. By contrast, the data for the 4-mm-long crystal show no evidence of this reconversion effect at the intensities used.

Double-Pass Multimode Doubling

Several double-pass arrangements were investigated to improve on the single-pass conversion efficiency achievable with more easily available, short KTP crystals. In one arrangement (DP I) both the fundamental and the second harmonic generated on the first pass were reflected back through the crystal with a 300-mm radius-of-curvature dielectric mirror. In Fig. 4, the conversion efficiency of a 5.1-mm-long KTP crystal is plotted as a function of incident intensity for this two-pass configuration. The shape of the

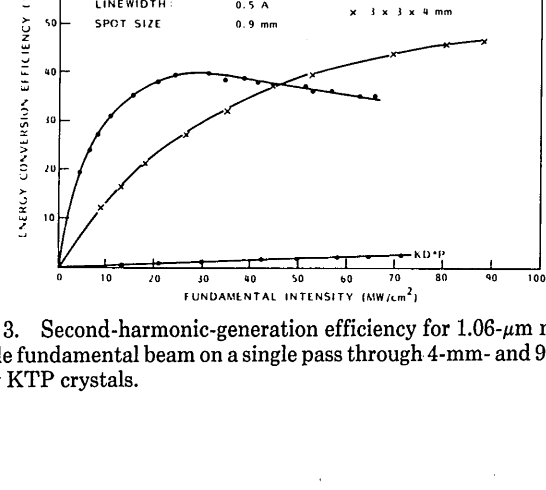

Fig. 3. Second-harmonic-generation efficiency for 1.06-μm multimode fundamental beam on a single pass through 4-mm- and 9-mm-long KTP crystals.

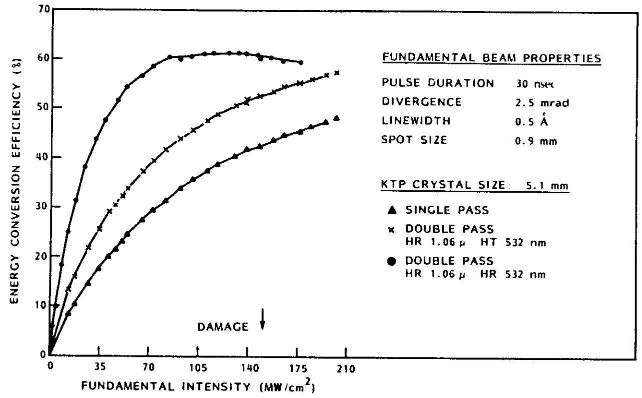

Fig. 4. Double-pass second-harmonic-generation efficiency for 1.06-μm multimode fundamental.

curve is similar to that obtained with the 9-mm-long crystal in single-pass geometry, with saturation setting in at low intensity and slight reconversion apparent at higher intensity. Although the data shown in Fig. 4 saturate at 60% conversion efficiency, other 5-mm-long crystals have reached 65% conversion efficiency. The conversion efficiency at saturation is higher for double passing the 5.1-mm-long crystal than for a single pass through the 9-mm-long crystal. This may be due either to better crystal quality or to a phase-shifting effect of the fundamental with respect to the second harmonic resulting from air dispersion.[12] This would reduce reconversion and higher-order processes, although the phases are somewhat random in a multimode beam.

From the earlier single-pass frequency-doubling measurements with multimode beams (see Fig. 3), it appears that a double-pass arrangement (DP II), in which only the fundamental retraversed the crystal, would avoid reconversion. Extrapolation of the results in Fig. 4 for higher intensities would appear to substantiate this assertion. However, the potential benefit of configuration DP II must be balanced against the need to operate at an intensity closer to the damage threshold and some overall efficiency loss incurred on combining the second-harmonic beams generated on the first and second passes.

Clearly the double-pass configuration DP I in short (5-mm) crystals, at intensities safely below the damage threshold, yields more efficient conversion of multimode beams than either a single pass through the 4-, 5-, and 9-mm-long crystals or a DP II configuration in a 5-mm-long crystal.

Gaussian Near-Diffraction-Limited Doubling

In contrast to the multimode doubling, conversion efficiency of a beam with near-diffraction-limited divergence and Gaussian transverse-mode structure saturates at a somewhat lower level, as shown in Fig. 5. This can be attributed to less severe intensity fluctuations from mode beating, an effect that has been shown to enhance the frequency-conversion efficiency.[13] We have observed only 35% conversion efficiency achieved at higher fundamental intensity by comparison with multimode doubling measurements in the same 5.1-mm-long crystal.

The same double-pass configurations, which have been described previously, were investigated for Gaussian fundamental beams. The comparison of DP I, DP II, and single-pass arrangements shows similar trends to those observed for multimode doubling, and most of the comments and observations made in that connection apply to the Gaussian-beam frequency doubling. The less severe mode-beating intensity fluctuations that led to reduced conversion efficiency at saturation apparently also result in reduced reconversion. Data on conversion efficiency as a function of fundamental intensity for a single pass through an 8-mm-long KTP crystal are plotted in Fig. 6. Compared with multimode doubling in long crystals (Fig. 3), only slight reconversion is seen for Gaussian beam doubling, even at very high fundamental intensity.

We have also measured the conversion efficiency of a 5.3-mm-long KTP crystal and compared the results with that for a 2.5-cm-long KD*P crystal under identical experimental conditions. From the slope of conversion efficiency versus incident fundamental intensity at low conversion efficiency, the effective susceptibility of KTP can be compared with that for KD*P. Provided that the fundamental is not depleted and there is no phase mismatch between fundamental and second harmonic, the expression for conversion efficiency is[14]

$$\frac{I(2\omega)}{I(\omega)} = \frac{128\pi^3}{n^3c^3}\,\omega^2{d_{\mathrm{eff}}}^2l^2I(\omega), \tag{1}$$

where n_ω is the refractive index at frequency ω, l denotes the crystal length, and d_{eff} is the effective gain coefficient.

Applying this expression to the data, we find

$$(d_{\mathrm{eff}})_{\mathrm{KTP}} = (17 \pm 1)(d_{\mathrm{eff}})_{\mathrm{KD^*P}}, \tag{2}$$

where we used 1.789 as the index of refraction for KTP and

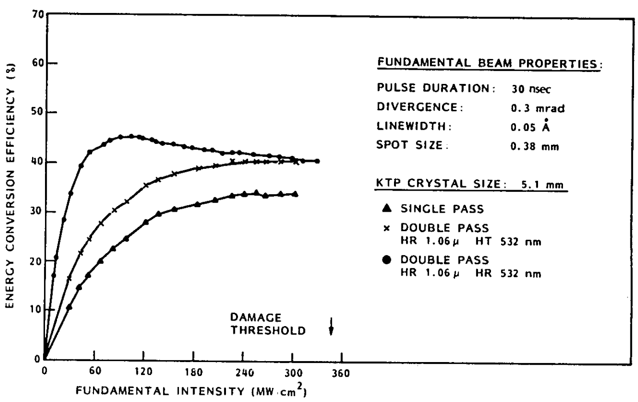

Fig. 5. Double-pass second-harmonic-generation efficiency for Gaussian 1.06-μm fundamental.

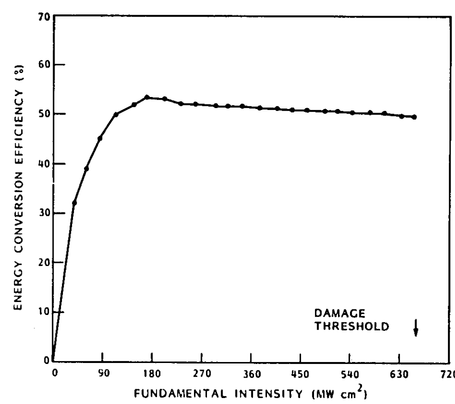

Fig. 6. Second-harmonic-generation efficiency for Gaussian fundamental; single pass through 8-mm-long KTP crystal.

1.493 for KD*P. The calculated value[15] using the results of Zumsteg *et al.*[5] for the susceptibility tensor elements, $d_{eff} = 17.7 \times 10^{-9}$ esu, agrees quite well with these experimental results.

Taking the slope of the conversion-efficiency curve in Ref. 8, however, we calculate a value of 4.8×10^{-9} esu for KTP. A similar result obtained from the slope of the curve in Fig. 5 gives $d_{eff} = 4.1 \times 10^{-9}$ esu. The fact that these methods give a somewhat lower value for d_{eff} than measured relative to KD*P is attributed to averaging the conversion efficiency for spatial and temporal variations in intensity.

Damage-Threshold Observations

With the small clear aperture currently attainable for KTP, the damage threshold plays an important role in applications of this crystal. We have shown that the second-harmonic conversion efficiency can reach its saturation value well below the damage threshold either in long crystals or in a double-pass arrangement. The maximum energy at the second harmonic, with the highest conversion efficiency, depends on the maximum currently available clear aperture, which is 5 mm × 5 mm.

There are several damage mechanisms that have been observed in the seven KTP crystals examined. One type of bulk damage appeared to occur along a plane of the crystal at 45 deg to the entrance face of the 8-mm-long crystal. The crystal cleaved at an intensity of 650 MW/cm^2 after several repeated hits on the damage site. No damage was seen in the crystal for intensities below this level while taking the conversion-efficiency data of Fig. 6. Additional damage sites were already present on the same plane in different areas of the crystal. Other crystals that suffered bulk damage at the intensities indicated in Figs. 3–5 did not suffer the same peculiar cleavage.

Another damage mechanism, which appeared in one crystal, darkens the interior of the crystal at an intensity of 150 MW/cm^2 without causing an inclusion. Degraded conversion-efficiency results following this effect can probably be attributed to absorption of 0.532- and/or 1.06-μm light. Since the average power levels were always below 200 mW for all data taken, the damage mechanisms would appear to be related to peak power.

CONCLUSION

We have demonstrated efficient second-harmonic conversion of divergent, multimode fundamental beams in KTP. Reconversion in long (9-mm) crystals limits the maximum conversion efficiency to 40%, but a double-pass geometry with short (5-mm) crystals avoids this effect. Reconversion of the second harmonic in long (8-mm) crystals appears to be less severe for Gaussian fundamental beams. Effective modeling of second-harmonic generation in KTP including reconversion, one- and two-photon-absorption effects, and integration over spatial and temporal intensity variations is beyond the scope of this paper. This work is currently in progress and will be reported at a later date. We have also presented preliminary observations of damage thresholds for Gaussian and multimode beams in several KTP crystals; however, inconsistent crystal quality from impurities and defects affects these observations. We have also measured the phase-matching angular bandwidth and the effective susceptibility relative to KD*P. In the future we plan to investigate sum-frequency mixing efficiency and damage thresholds for wavelengths close to the absorption band edge.

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