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Efficient Second-Harmonic Generation in Tl_3AsSe_3 Using Focussed CO_2 Laser Radiation

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Abstract. The nonlinear material Tl_3AsSe_3 was used to convert pulsed 10.6 µm laser radiation into the second harmonic. The laser beam was tightly focussed, and an energy conversion efficiency of 57% was obtained, which is the highest reported to date in the mid-IR.

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A powerful and tunable mid-IR laser would be very useful for a number of applications, and one approach is to use a CO_2 laser and harmonic generation. This method has the advantage that CO_2 lasers are well developed, and the major uncertainty at present is the availability of suitable harmonic generation materials.

Tl_3AsSe_3 (TAS) can be grown at Westinghouse with high quality in lengths over 10 cm, and with an extremely low absorption coefficient of 0.0005 cm^{-1}. TAS also has a wide transmission range of 1.3–17 µm, a large nonlinear coefficient and a high damage threshold. These factors combine to make TAS an excellent harmonic generation material, and a second-harmonic generation (SHG) efficiency of 28% was previously reported using TAS [1]. The present results are considerably higher than previously reported, demonstrating that very efficient harmonic generation can be accomplished with TAS, which could lead to a useful laser source in the mid-IR.

1. Apparatus

A schematic diagram of the apparatus is shown in Fig. 1, which consists of a low-pressure oscillator and a high-pressure amplifier. The laser contained a grating, a ZnSe beam splitter with enhanced polarization coatings, a CdTe Pockels cell and a 4 m radius of curvature copper mirror. These elements were arranged in a cavity dumping configuration [2], which allows the cavity field to build up to its maximum value before being switched out of the laser cavity. The laser-pulse shape is determined by both the laser cavity length and the switching time of the Pockels cell. With infinitely fast switching, the pulse length would be equal to the laser cavity round-trip time.

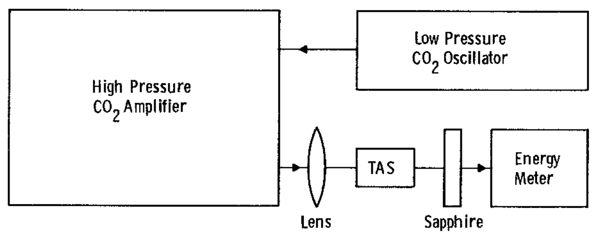

Fig. 1. Schematic diagram of SHG parametrization apparatus

The oscillator produced Gaussian TEM_{000} pulses on the $P(20)$ transition at 10.59 µm. The pulse shape was triangular with a FWHM of 20 ns, as shown in Fig. 2. The pulse energy was about 0.1 mJ, which was then increased in the high pressure amplifier to any level up to 30 mJ. This was accomplished with a double pass through the 150 cm long discharge medium.

The SHG efficiency was measured using a sensitive thermal energy meter, where a sapphire plate was used to block the 10.6 µm radiation while measuring the 5.3 µm energy. The various losses in the optics were also taken into account, to obtain the intensities inside the crystal.

The laser beam was determined to be nearly Gaussian by measuring the beam diameters at various points in the system, and comparing them with the predicted spot sizes of a Gaussian beam. The spot size ω is defined from the intensity relation

$$I = I_0 \exp(-2r^2/\omega^2), \quad (1)$$

where I_0 is the intensity at the center of the Gaussian, and r is the radial distance from the center. The spot sizes were experimentally determined by measuring the diameter which allowed half of the beam power to pass [3], which

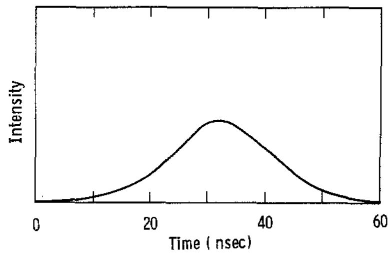

Fig. 2. Cavity dumped laser pulse illustrating triangular pulse shape

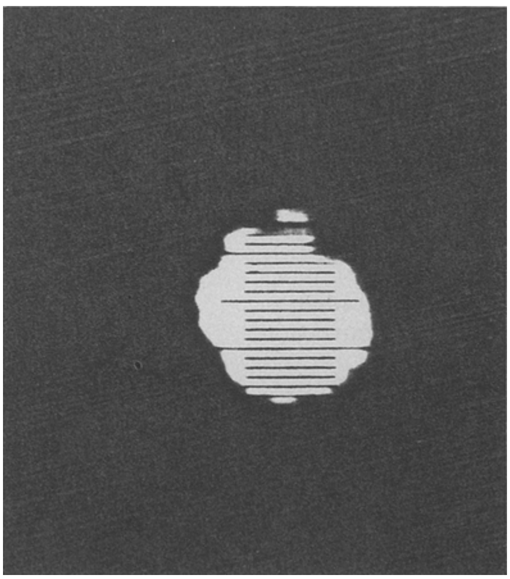

Fig. 3. Photograph of burn hole used to determine the focal spot size

occurs at a beam radius of 0.589ω. All of these measurements were performed with the amplifier off, so that any distortions produced by the amplifier were not included.

The focussed spot size was measured by allowing the laser beam to burn through a piece of paper until half the beam power was projected through the hole. Such a burn hole is shown in Fig. 3, in which the hole diameter of 165 μm represents the half power point of the beam, and corresponds to a spot size of 140 μm. The paper thickness was 40 μm, which is thin compared to the hole diameter, so that diffraction effects from the edges of the paper were minimized, and the irregular burn pattern is due to the fiber structure of the paper.

2. SHG Formulation

The SHG process is typically formulated in terms of plane waves, and to apply the results to the present focussed and pulsed case, the plane wave results must be spatially and temporally integrated. The SHG energy efficiency for a pulse is then given by

$$\eta = \alpha \int\int I^2 da dt / \int\int I da dt , \qquad (2)$$

where the integrals extend over the pulse shape and profile, and α is the conversion efficiency coefficient for plane waves given by [4]

$$\alpha = (8\pi^2 \mu_0 v^2 / cn^3) d^2 L^2 \operatorname{sinc}^2(\beta \Delta\theta) , \qquad (3)$$

where μ_0 is the vacuum permeability, v is the fundamental frequency, c is the velocity of light, n is the index of refraction of the fundamental frequency, d is the nonlinear coefficient, and L is the interaction length. The conversion is also assumed to be uniform and in the small signal regime over the interaction length. The phase mismatch is taken into account by $\operatorname{sinc}^2(\beta\Delta\theta)$, where $\operatorname{sinc}^2(x)$ represents $\sin^2(x)/x^2$ and

$$\beta = (\pi v L/c) \sin(2\theta_m) [(n_e^{2v})^{-2} - (n_0^{2v})^{-2}] n^3 , \qquad (4)$$

where θ_m is the phase matched angle, n_e^{2v} is the extraordinary index of refraction at the harmonic frequency, n_0^{2v} is the ordinary index at the harmonic, and n refers to the ordinary index of the fundamental beam.

Performing the spatial integration over a Gaussian beam profile along with the temporal integration over the triangular pulse, the efficiency becomes

$$\eta = (1/3)\alpha I_0 , \qquad (5)$$

which applies to all Gaussian spatial distributions and any triangular pulse width, for either symmetrical or asymmetrical triangular pulse shapes. In terms of the fundamental pulse energy E_v, I_0 is given by

$$I_0 = 2E_v/(\pi\omega^2\tau) , \qquad (6)$$

where τ is the FWHM of the triangular pulse.

The focussed SHG formulation differs from the unfocussed case in that the variation of the beam in the propagation direction must be taken into account. Such an analysis was performed by Boyd and Kleinman for the confocal case with Gaussian beams [5], and they formulated an efficiency parameter h, which described the reduction in efficiency due to the focussing, where the focussed efficiency η_f is related to the unfocussed efficiency by

$$\eta_f = \eta h/\xi , \qquad (7)$$

where $\xi = L/b$ is a focussing parameter, and $b = 2\pi n\omega^2 v/c$ is the length over which most of the SHG interaction will occur. Therefore ξ represents the fraction of the available interaction length within the crystal. With a large spot size, b will be much larger than the crystal length, and ξ will be small. For tight focussing, b will be smaller than the crystal length, and ξ will be large. At small values of ξ, $h = \xi$, and η_f reduces to the unfocussed efficiency η.

The h parameter is also a function of the walk-off angle due to birefringence, which is equal to 2.12 degrees inside TAS at 10.59 μm. Walk-off reduces the efficiency, since the effective interaction length is decreased. For a given crystal length, the appropriate value of h for TAS can be found from the curves of Boyd and Kleinman.

3. Measurements

The SHG efficiency was measured using a 4.57 cm long TAS crystal having diamond turned surfaces with AR coatings, and initially the unfocussed beam with a spot size of 5.7 mm was used. The crystal was rotated about the matching angle, and the normalized SHG efficiency as a function of the internal angle $\Delta\theta$ was measured. The result is shown in Fig. 4, and it is compared with a $\text{sinc}^2(\Delta\theta)$ function having a FWHM of 1.15 mrad, which corresponds to the FWHM of the measured points.

The tuning curve indicates that the crystal is operating at an effective length of 3.3 cm, which is 72% of the measured 4.57 cm crystal length. It is believed that this reduction in effective length is due to crystal quality problems, and is not due to beam spreading effects, since the same tuning curve was measured at various other spot sizes using collimating optics, and it was found that the FWHM was unchanged for spot sizes larger than 2 mm. It is also believed that uncertainties in the indices of refraction used in calculating the tuning curve are not responsible for the widening effect. This was investigated analytically by expanding the tuning curve relationship in terms of the uncertainties in the indices, and assessing the widening as a function of the uncertainty. The Sellmeier equations of Ewbank were used [6], which are considered to be the best available formulation, and are within about 0.03% of the measured values at 10.59 μm, and should widen the tuning curve by 0.5% at the most. We can therefore conclude that the uncertainties in the Sellmeier equations are not responsible for the additional tuning curve width.

A reasonable explanation for the widening of the tuning curve is crystal quality problems. This speculation is supported by noting that the measured curve of Fig. 4 does not attain zero, and that it is not symmetrical in the wings. These effects, and especially the asymmetry tend to support crystal quality problems, in which asymmetrical misorientations occur over the length of the crystal. It would be difficult to explain the asymmetry with instrumental effects. We must therefore conclude that the TAS crystal quality is not perfect, and that the effective length with regard to SHG is 3.3 cm.

Since the SHG process is not effectively interacting over the entire crystal length L, the measured d value will be less than the actual value. The measured d coefficient can be scaled more closely to the actual value by assuming the interaction length is the effective length L_{eff}, rather than L. This technique should give reasonably correct results when $L_{\text{eff}} \sim L$.

Using an effective length of 3.3 cm, and $d = 20$ pm/V, the focussed efficiency given by (7) was plotted in Fig. 5. The calculation does not include depletion of the fundamental beam, and the plot is linear. The experimental points are correctly predicted by the theory in the small signal region using these values of length and d, but deviate at higher efficiencies. Also shown is a $\tanh^2(\eta^{1/2})$ function, which describes depletion for the plane wave case, and it also appears to predict the effects of depletion with focussing.

The present d value is considerably less than the 41 pm/V measured by Feichtner and Roland [7], and the value of up to 29 pm/V given in [1]. A possible explanation is that longitudinal laser modes were contributing to the higher measured d values, where enhancement factors of two between single and multiple mode operation can exist [8]. The present result corresponds to single-longitudinal.mode operation, and is expected to be lower than those measured with a multiple-mode laser.

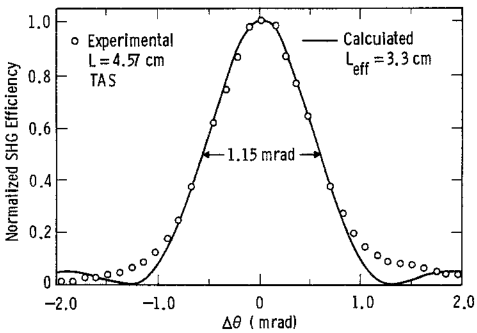

Fig. 4. Measured tuning curve compared to $\text{sinc}^2(\Delta\theta)$ prediction

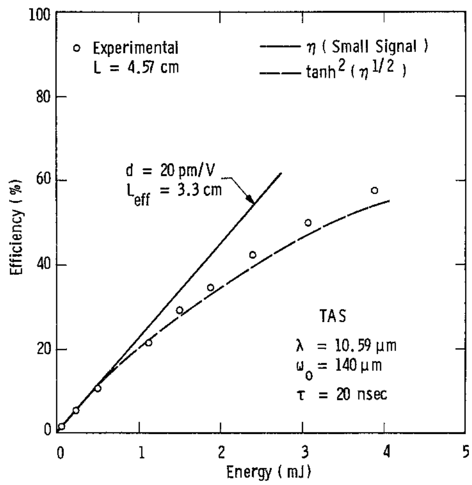

Fig. 5. Focussed SHG results showing high SHG efficiency for TAS

A maximum efficiency of 57% was obtained without damage at an internal energy of 3.9 mJ, giving an energy density of 12.7 J/cm^2 at the focus. With the 20 ns FWHM pulses used for these measurements, the power density was 630 MW/cm^2 at the focus. Assuming that the focus was situated at exactly the center of the crystal, as prescribed for the confocal case, beam expansion would have reduced the energy density to 5.3 J/cm^2 through the surfaces. Although this represents a very high surface energy density, it is less than recently measured damage levels for TAS of 10 J/cm^2 using 130 FWHM ns pulses at 10 μm [9].

4. Conclusions

TAS has been shown to be a very efficient harmonic generation material, and an efficiency of 57% was demonstrated using a pulsed CO_2 laser at 10.6 μm with focussing in the crystal. The data was correlated to the SHG theory for focussed Gaussian beams, which gave a nonlinear d coefficient of 20 pm/V. This value was obtained with a single longitudinal mode laser, so that enhancement effects from multiple modes were not present.

The high demonstrated efficiencies of this study indicate that TAS could be useful as a harmonic generation material for a mid-IR laser. With appropriate harmonic generation and mixing, tunability from quadrupled CO_2

laser radiation to doubled CO_2 wavelengths could be obtained with such a laser.

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