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ROYAL SIGNALS & RADAR ESTABLISHMENT

CO_2 LASER FREQUENCY DOUBLING USING $AgGaSe_2$

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Defence Research Agency Electronics Division

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Date: September 1991

Abstract

$AgGaSe_2$ samples are examined using a CO_2 laser for the frequency doubling application. The results are compared with a coupled wave equation model which includes absorption. Good agreement between the results and the model is obtained. Best energy conversion efficiency obtained under our experimental conditions was 7% with an anti-reflection coated 2cm long sample.

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INTRODUCTION

$AgGaSe_2$ is a optical material which demonstrates a high figure of merit for nonlinear optics in the infrared. It transmits between 0.7 μm and 19 μm has a refractive index around 2.6, a band gap of ~ 1.82 eV and is a negative birefringent material of crystal class $\bar{4}2m$ [1]. This material can be phase matched for frequency doubling of CO_2 laser radiation and had shown up to 60% conversion efficiency[2] under best conditions. The material may also be phase matched to generate 3-5 μm and 8-14 μm tunable radiation in an OPO configuration pumped by HF or Er:YAG wavelengths in the 2.7-2.9 μm region. Phase matching for 3-5 μm radiation using Nd:YAG laser pumps in an OPO configuration is also possible[3].

We report a CO_2 laser frequency doubling study of $AgGaSe_2$ samples grown during the crystal growth programme in the UK and of one US grown sample. We examine the doubling efficiencies of the samples and compare our results with a theoretical model which takes into account absorption at the 10 μm and 5 μm wavelengths, beam shape and pulse profile. We also examine the angular acceptance of the crystal samples and relate these results to the single crystal nature of the samples. The first section of the report indicates the main theoretical ideas necessary for the understanding of the experimental results described and discussed in the second section.

THEORY

In order to examine our crystal samples to obtain the most information about crystal quality and the factors which influence the efficiency of the frequency doubling process it is necessary to begin with the coupled wave equation for the frequency doubling process. Yariv[4] gives for the solution of the electromagnetic wave equations in the case where three wave mixing can occur as

$$\frac{dE_{1i}}{dz} = -\frac{\sigma_1}{2}\sqrt{\frac{\mu_o}{\varepsilon_1}}\,E_{1i} - \frac{i\omega_1}{2}\sqrt{\frac{\mu_o}{\varepsilon_1}}\,d_{ijk}\,E_{3j}\,E^*_{2k}\,e^{-i(k_3-k_2-k_1)z}$$

$$\frac{dE^*_{2k}}{dz} = -\frac{\sigma_2}{2}\sqrt{\frac{\mu_o}{\varepsilon_2}}\,E^*_{2k} - \frac{i\omega_2}{2}\sqrt{\frac{\mu_o}{\varepsilon_2}}\,d_{kij}\,E_{1i}\,E^*_{3j}\,e^{-i(k_1-k_3+k_2)z} \qquad (1)$$

$$\frac{dE_{3j}}{dz} = -\frac{\sigma_3}{2}\sqrt{\frac{\mu_o}{\varepsilon_3}}\,E_{3j} - \frac{i\omega_3}{2}\sqrt{\frac{\mu_o}{\varepsilon_3}}\,d_{jik}\,E_{1i}\,E_{2k}\,e^{-i(k_1+k_2-k_3)z}$$

where the symbols have their usual meaning and d_{ijk} is the effective nonlinear coupling tensor.

The first term in these equations represents the absorption loss of the waves as they propagate through the crystal in the z direction (not the crystal z direction) while the second term represents the nonlinear coupling of the waves. It is required that for the i'th component of the nonlinear polarization which gives rise to the nonlinear term in the first equation of equation 1 that

$$\omega_1 = \omega_3 - \omega_2$$

We can define

$$A_1 = \sqrt{\frac{n_1}{\omega_1}}\,E_1 \qquad (2)$$

so that the wave intensities I_1 are given by:

$$I_1 = \frac{1}{2}\sqrt{\frac{\varepsilon_o}{\mu_o}}\, n_1 |E_1|^2 = \frac{1}{2}\sqrt{\frac{\varepsilon_o}{\mu_o}}\, \omega_1 |A_1|^2 \qquad (3)$$

and equation (1) reduces to:

$$\frac{dA_1}{dz} = -\frac{1}{2}\alpha_1 A_1 - \frac{i}{2} K A_2^* A_3 e^{-i\Delta kz}$$

$$\frac{dA_2^*}{dz} = -\frac{1}{2}\alpha_2 A_2^* + \frac{i}{2} K A_1 A_3^* e^{i\Delta kz} \qquad (4)$$

$$\frac{dA_3}{dz} = -\frac{1}{2}\alpha_3 A_3 - \frac{i}{2} K A_1 A_2 e^{i\Delta kz}$$

where

$$\Delta k = k_3 - (k_1 + k_2)$$

$$K = d_{eff}\sqrt{\left(\frac{\mu_o}{\varepsilon_o}\right)\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}}$$

and $\alpha_1 = \sigma_1\sqrt{\frac{\mu_o}{\varepsilon_o}}$

and 1, 2, 3 represent the polarization directions of the E fields.

For frequency doubling $\omega_1 + \omega_2 = \omega_3$ or

$$2\omega_1 = \omega_3 \quad \text{and} \quad A_1 = A_2$$

These equations can be solved numerically using an iterative technique on a small desk top computer in order to obtain the dependence of the doubling efficiency when crystal samples display absorption. The equations demonstrate that their is an optimum crystal length when absorption is present and can thus be used to specify minimum required absorption and best crystal length given the constraints imposed by the crystal growth process.

Further insight into the doubling process may be obtained for the case when there is no absorption and crystal lengths are small so that little pump depletion occurs. In this case equation 1 reduces to

$$\frac{dE_{3j}}{dz} = -i\omega \sqrt{\frac{\mu_o}{\varepsilon_3}}\; d'_{ijk}\, E_{1i}\, E_{1k}\, e^{i\Delta kz} \tag{5}$$

Solving this equation and converting from electric field to power, P, one obtains for the conversion efficiency, ζ

$$\zeta = \frac{P^{2\omega}}{P^{\omega}} = 2\left(\frac{\mu_o}{\varepsilon_o}\right)^{3/2} \frac{\omega^2 (d'_{ijk})^2 L^2}{n^3} \left(\frac{P(\omega)}{\text{area}}\right) \frac{\text{Sin}^2\left(\frac{\Delta kL}{2}\right)}{\left(\frac{\Delta kL}{2}\right)^2} \tag{6}$$

Equation 6 contains the phase matching term $\mathrm{Sin}^2\left(\frac{\Delta kL}{2}\right) \Big/ \left(\frac{\Delta kL}{2}\right)^2$ which maximises when

$$\Delta k = 0 \quad \text{or} \quad \frac{2\pi n^{(2\omega)}}{\left(\frac{\lambda}{2}\right)} = 2 \cdot \frac{2\pi n^{(\omega)}}{\lambda}$$

Phase matching thus requires $n^{(2\omega)} = n^{(\omega)}$ (7)

Phase matching may be achieved by use of the birefrigent nature of the crystal. The extraordinary refractive index for a birefringent crystal depends on the angle, θ, of wave propagation from the optic (or z axis) of the crystal and is given by:

$$n_e(\theta) = \frac{n_e\, n_o}{\left(n_o^2 \mathrm{Sin}^2\theta + n_e^2 \mathrm{Cos}^2\theta\right)^{\frac{1}{2}}} \tag{8}$$

where n_o and n_e are the ordinary and extraordinary refractive indices. The requirement for phase matching determines the angle, θ, through the crystal that the waves must travel.

If the angle θ is varied about the optimum value the frequency doubling signal will vary according to the $(\mathrm{Sin}\ x/x)^2$ dependence in equation 6. The width of this curve depends on ΔkL and if refractive index data is available information on the length of crystal contributing to the nonlinear interaction can be obtained. Such measurements can therefore be used to determine the extent of the single crystal nature of a particular sample.

The nonlinear tensor term d'_{ijk} of equation 6 depends on the direction of propagation of the waves through the crystal. We have seen that the angle θ with respect to the optic axis is fixed. For a crystal of the $\bar{4}2m$ type and for Type 1 phase matching which are appropriate for $AgGaSe_2$ the nonlinear term is given by

$$d'_{ijk} = d_{eff} = -\, d_{14}\, \mathrm{Sin}\theta\, \mathrm{Sin}\, 2\phi \tag{9}$$

where ϕ is the azimuthal angle measured with respect to the x crystallographic axis and d_{14} is the relevant element of the nonlinear tensor appropriate for $\bar{4}2m$ class crystals. The optimum nonlinear coefficient will thus be obtained when $\phi = 45°$.

EXPERIMENTAL SET-UP AND RESULTS

UK grown crystals of $AgGaSe_2$ of 1 x 1 x 1 cm^3 were obtained which had been cut and polished for phase matching at 9.4 μm. θ was determined using the phase match condition 7 and equation 8 together with the Sellmeier equations[5]

$$n_o^2 = A + \frac{B}{1 - (D/\lambda)^2} + \frac{C}{1 - (E/\lambda)^2}$$

$$n_e^2 = F + \frac{G}{1 - (H/\lambda)^2} + \frac{K}{1 - (E/\lambda)^2} \qquad (10)$$

where $A = 3.9362$, $B = 2.9113$, $C = 1.7954$, $D = 0.38821$

$E = 40$, $F = 3.3132$, $G = 3.3616$, $H = 0.38201$

$K = 1.7677$ and λ is in μm.

These crystals were uncoated. In addition a US grown crystal 1 x 1 cm^2 x 2 cm long was obtained. This crystal was similarly cut for 9.4 μm use and had anti reflection coatings on both the input and output faces.

The laser used in these experiments is a hybrid TEA laser which operates on a single longitudinal mode and thus provides a very smooth pulse. This allows us to make more accurate measurements than would be the case if multiple longitudinal mode beating was superimposed on the pulse envelope. Figure 1 displays the CO_2 pump pul: en the cw section was operated below threshold. The laser was untuned and so operated on the P20 line at 10.59 μm. The laser typically emits 50 kW, 150 nsec, 10 mJ pulses.

Figure 2 shows the experimental set up. The output beam from the laser is divided by the beam splitter and monitored by the reference joulemeter. The remainder of the beam is focussed by a 50 cm lens into the crystal which is mounted on a tilt and rotate mount which allows adjustments to be made to optimise the beam propagation direction within the crystal. A sapphire filter is placed on the output side of the crystal to remove the remainder of the CO_2 beam and allow detection of the 5 μm signal using either a joulemeter or fast liquid N_2 cooled Au:Ge detector. Removable attenuators were placed at the output of the laser to allow the pump intensity to be varied.

Since the crystals were cut for phase matching at 9.4 μm a deliberate tilt from normal incidence was introduced so that phase matching could be achieved at the 10.59 μm laser wavelength. This tilt accorded with our calculations using the equations 7, 8 and 10. Crystal transmission and hence absorption was obtained using a Perkin-Elmer IR spectrometer. A typical transmission characteristic is shown in Figure 3. The decrease in transmission at shorter wavelengths is thought to be due to the presence of scattering centres within the crystal[6]. Absorption/loss coefficients for our crystals calculated from these measurements are given in Table 1. The absorption coefficients for the 2 cm coated sample are difficult to estimate since the transmission characteristic also includes the effect of the anti reflection coatings.

The laser beam diameter was measured at the focus of the 50 cm lens using the scanning knife edge technique[7]. The results from a typical measurement are shown in Figure 4 together with the Gaussian fit to the data. The result gives a beam radius at $1/e^2$ of 0.625 mm so that a maximum energy density of $1J/cm^2$ and intensity of 4 MW/cm^2 could be obtained. Damage threshold of 1.1 J/cm^2 had previously been measured on trial slices so that care to avoid damage required the judicious use of attenuation.

The 50 cm lens provided a Rayleigh range of 30 cm which is greater than the crystal lengths so that our experiments were carried out under plane wave conditions. The more optimum confocal condition would have provided greater efficiency but would have required reduced pump power since the damage threshold would have been exceeded. Beam walk off was also avoided in these experiments since the beam diameter was sufficiently large.

A typical 5 μm pulse obtained using the Au:Ge detector is shown in Figure 1 together with a typical 10 μm pump pulse. As expected the 5 μm pulse is shorter than the pump pulse due to the expected $I^2_{(\omega)}$ dependence of the doubling process. More detailed measurements of the internal energy conversion efficiency obtained by measuring both input and output pulse energies are shown in Fig 5. These results have been corrected for the Fresnel losses associated with the uncoated $AgGaSe_2$ samples and for the losses associated with the sapphire filter. It can be seen from this data that internal conversion efficiencies range from 0.25% to 1% at the 8 mJ input level for the UK samples and up to 7% at 10 mJ for the larger US sample. The data also demonstrate a linear dependence on pump energy. This is to be expected when pump depletion does not occur and conversion efficiencies are low as is clearly the case in these measurements.

Figure 6 displays the intensity conversion efficiencies for the uncoated samples corrected for internal conversion. These are also linear as expected but display slightly larger conversion efficiencies varying from 0.6% to 1.6% at the 3 MW/cm^2 for the UK sample and up to 4% for the US sample at 3 MW/cm^2.

Examination of the absorption coefficients in Table 1 and comparison of the measured doubling efficiencies also indicated in the table show the correlation that the greater the crystal absorption the smaller the doubling efficiency as might be expected. The energy and power conversion efficiencies for the US grown sample are given in Figure 7. Due to the presence of anti-reflection coatings these have only been corrected for reflection losses associated with the sapphire filter.

These doubling efficiencies can be compared with the theoretical model based on equation 4 provided one recalls that this model deals with uniform plane waves and constant intensity. Since we have a Gaussian beam profile then the intensity conversion efficiency calculated using equation 4 will require to be reduced by a factor of 2. Similarly by considering the pulse shape of Figure 1 one can calculate that the energy conversion efficiency will be reduced from this intensity conversion efficiency by a further factor of 1.6. Figure 8 displays the theoretical results obtained in this way together with the experimental intensity and energy internal conversion efficiencies for crystal[5]. The upper theoretical curve is that obtained from solving equation 4 using the absorption coefficients for crystal[5]. This result is corrected for the Gaussian nature of the beam in the centre curve while the lower curve is again corrected for the pulse profile of Figure 1. It can be seen that the experimental results agree well, the intensity conversion efficiency fitting the centre theory curve well and the energy conversion efficiency fitting the lower theory curve as should be expected.

Results from the solution of equation 4 using appropriate absorption coefficients and corrected for Gaussian beam shape and pulse profile are given in Table 1 for comparison with the conversion efficiencies obtained experimentally. It can be seen that there is a good correlation between experiment and the theoretical model within experimental error. It is clear that absorption controls the obtainable conversion efficiency and that absorption on the pump frequency has a greater effect than absorption on the signal frequency. This might be expected since the doubled output depends on the pump input squared.

Figure 9 shows typical results obtained when a crystal is tuned through the optimum phase match angle θ. It is clear that the expected $(\text{Sin } x/x)^2$ dependence of equation 6 is obtained, the solid curve representing a $\text{Sin}\theta\left(\text{Sin}\frac{\Delta kL}{2}\Big/\frac{\Delta kL}{2}\right)^2$ fit to the experimental data using equations 6 and 9. The effect of changes in θ on d_{eff} through equation 9 is small over angles which the $(\text{sin } x/x)^2$ dependence is important. By measuring the width of these curves for each crystal it is found that the FWHH is typically 2.3° external to the crystal, for the 1 cm crystals and is 1.2° for the 2 cm long US sample. This compares well with an expected width of 2.3° and 1.2° for each length calculated using equations 8 and 10. Thus we can deduce that each sample is one single crystal throughout. The results obtained for each crystal are summarised in Table 1.

CONCLUSIONS

CO_2 laser frequency doubling experiments have been carried out on a number of 1 cm^3 UK grown samples of $AgGaSe_2$ and one US grown 2 cm long sample. Best results for the 1 cm long UK samples were 1% energy and 1.6% peak internal intensity conversion efficiency and 7% internal energy conversion efficiency for the US sample. The experimental results confirm the validity of the coupled wave model which includes absorption. Conversion efficiencies are

found to decrease with increasing absorption coefficient in accordance with the model when the effects of beam shape and pulse profile are taken into account. Angular tuning measurements provide the angular acceptance width for $AgGaSe_2$ of 2.3-2.4° cm as is expected from theory and refractive index data and thus confirm the single crystal nature of the samples.

It is clear from the results that absorption is the parameter which limits obtainable doubling efficiency and should therefore be of major concern for future crystal growth.

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Table 1 - Experimental / Theoretical Results

CRYSTAL	ABSORPTION COEFFICIENTS		ENERGY CONVERSION @ 10mJ / %		PEAK INTENSITY CONVERSION @ 3.0MW / cm² / %	
	$\alpha_{10.6}$ / m^{-1}	$\alpha_{6.3}$ / m^{-1}	EXPERIMENT	THEORY	EXPERIMENT	THEORY
1	28.8	45.9	0.79	0.64	0.85	0.84
2	52.0	104.7	0.33	0.35	0.36	0.50
3	12.1	22.1	1.45	0.81	1.58	1.12
4	49.1	81.4	0.42	0.39	0.54	0.58
5	12.3	28.4	0.81	0.75	1.02	1.08
6	18.1	29.1	0.69	0.74	0.86	1.02
7	23.2	44.2	0.72	0.69	0.79	0.90
US	8.7	12.1	4.52	3.43	3.76	4.06

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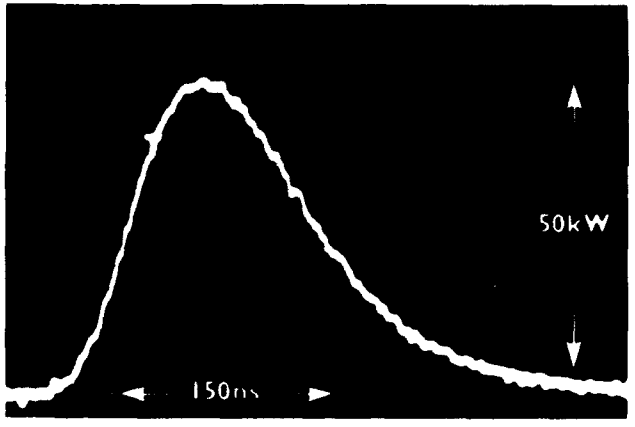
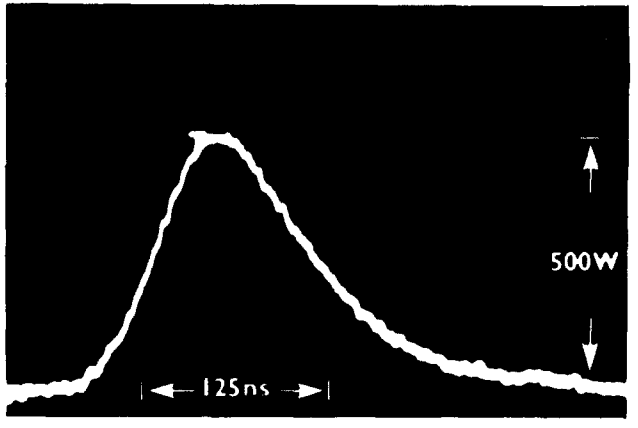
Typical Pulse profiles



a) Pump 10.6um pulse



b) Harmonic 5.3um pulse

FIG 1

Experimental set-up

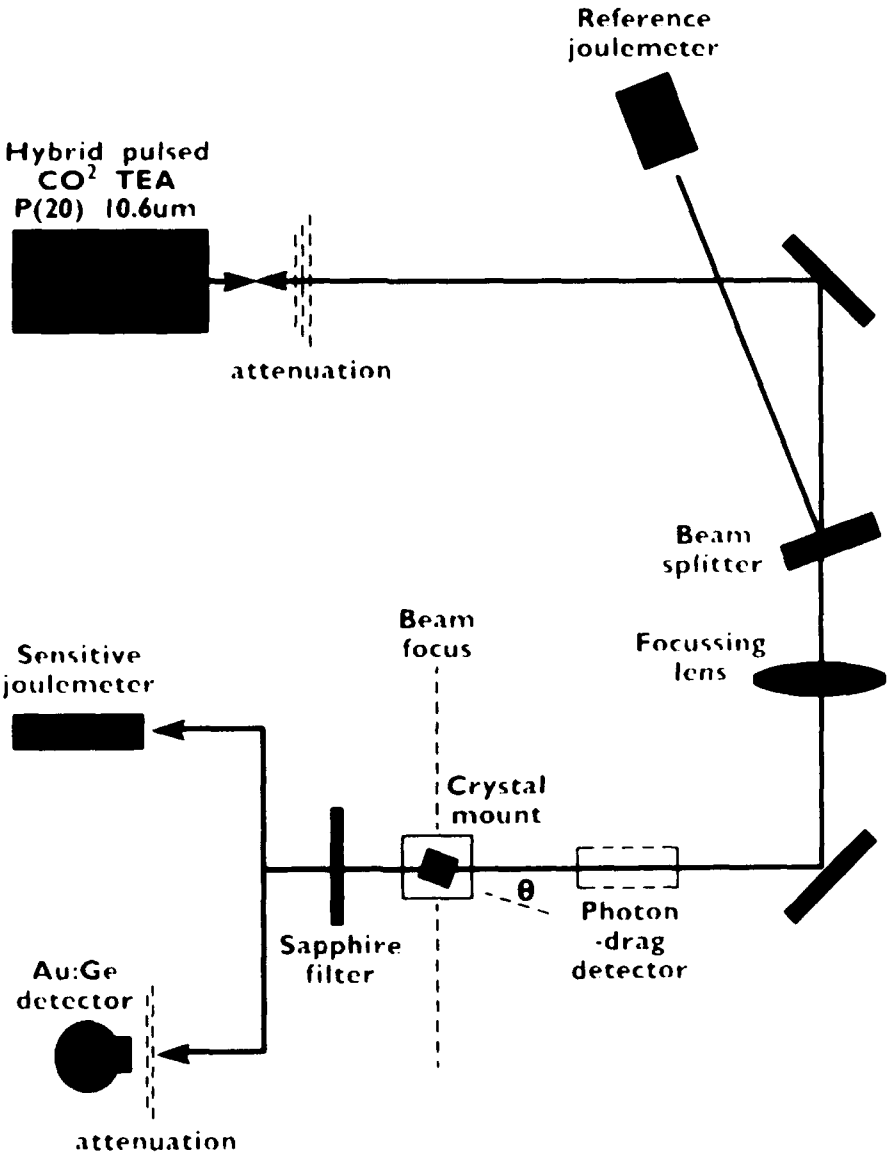


FIG 2

Transmission Characteristic — U.K. Crystal (3)

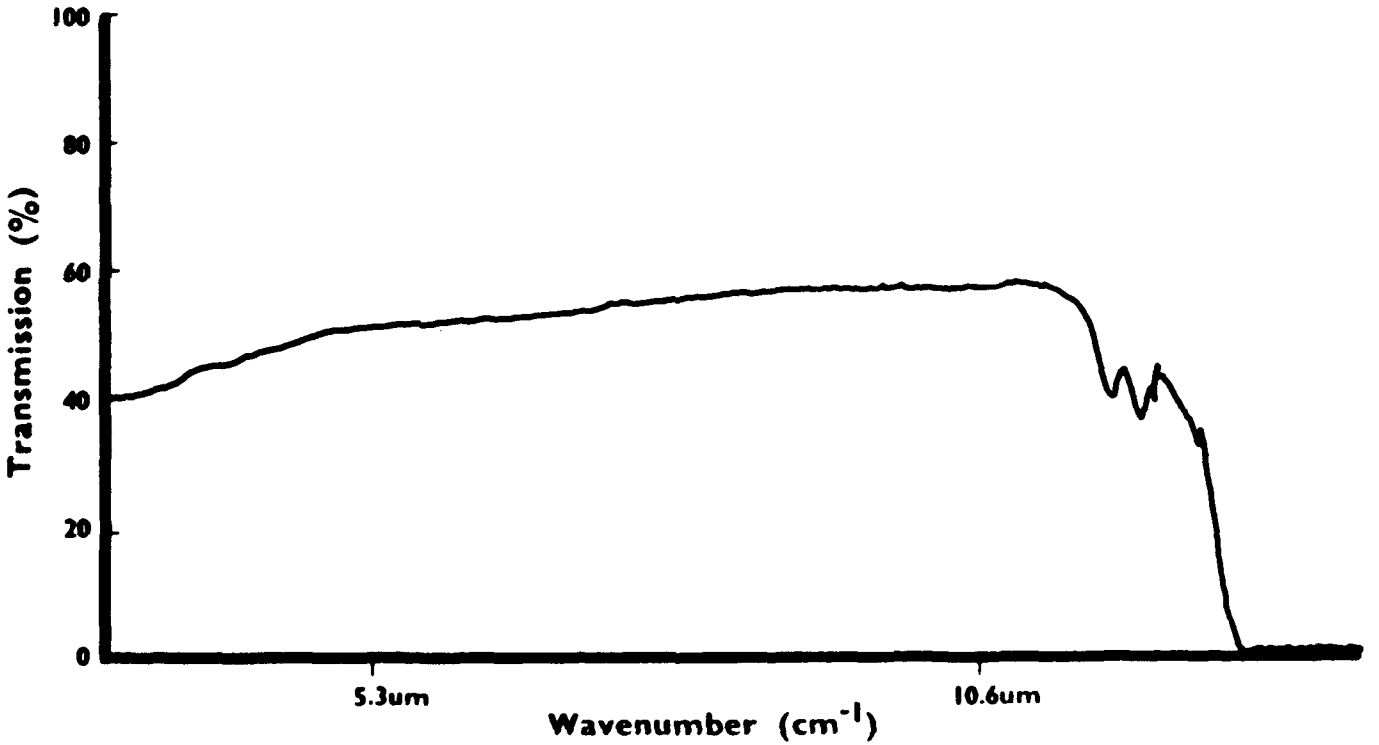


FIG 3

$1/e^2$ Beam Radius Measurement

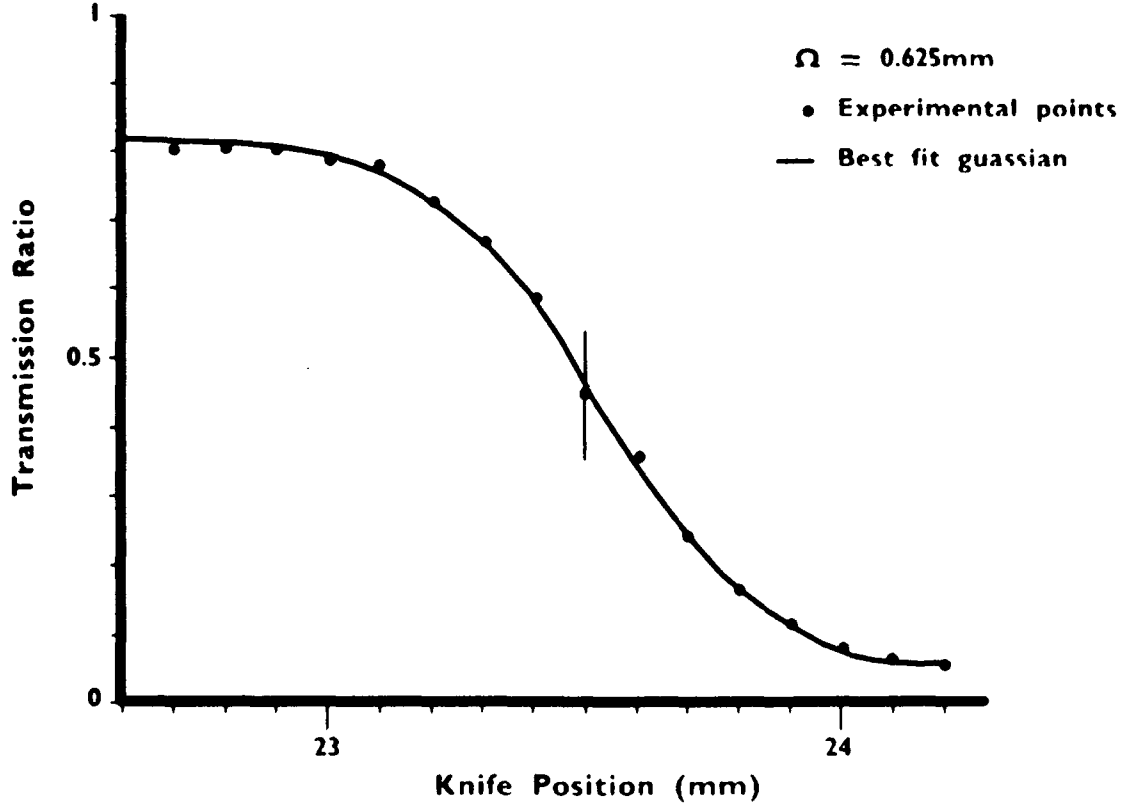


FIG 4

Internal Energy Conversion Efficiencies (U.K.)

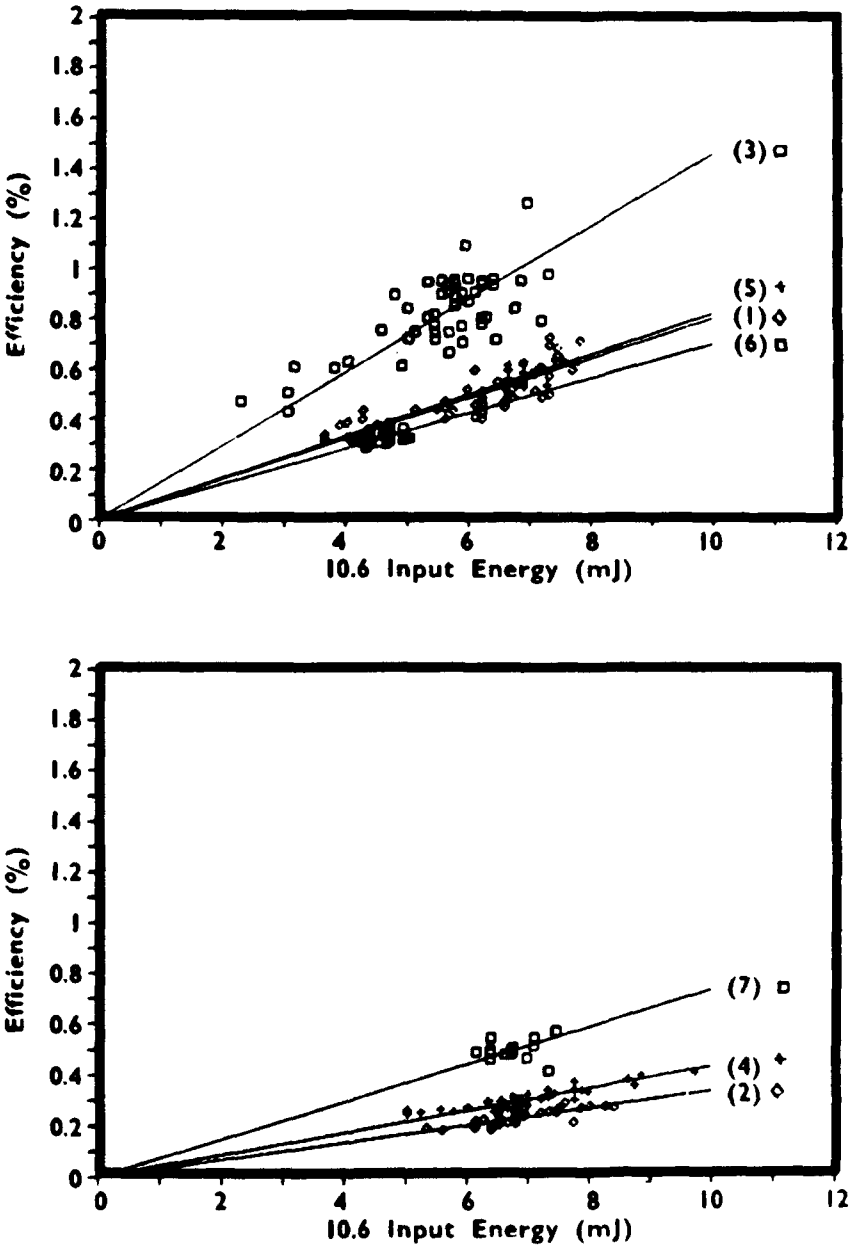
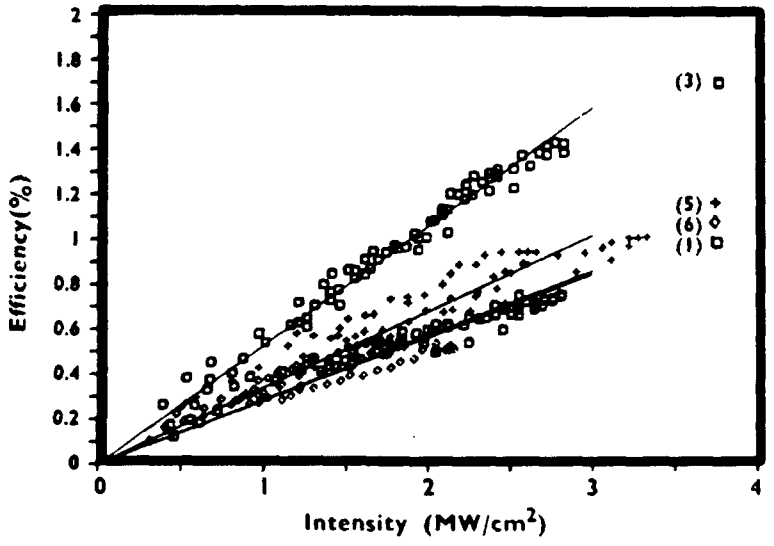


FIG 5

Internal Intensity Conversion Efficiencies (U.K.)



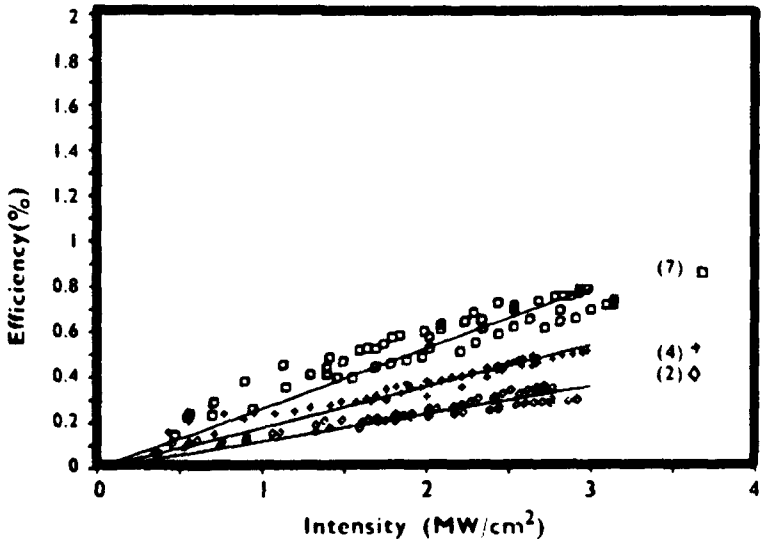
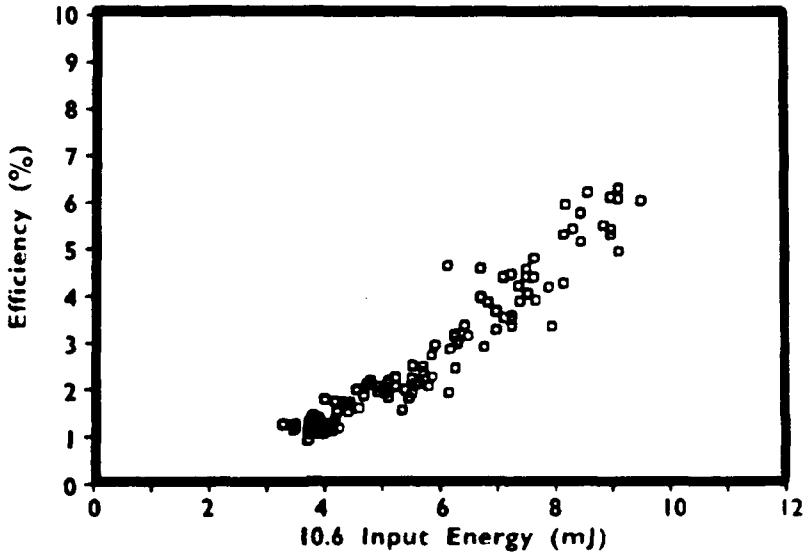


FIG 6

Internal Energy & Intensity Conversion Efficiencies (U.S.)

a) Energy:



b) Intensity:

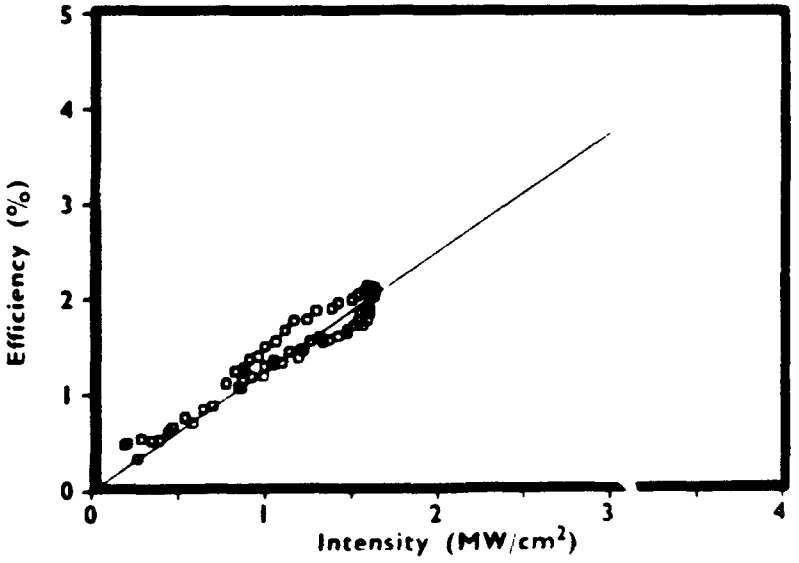


FIG 7

Theory/Experiment Comparison U.K. Crystal (5)

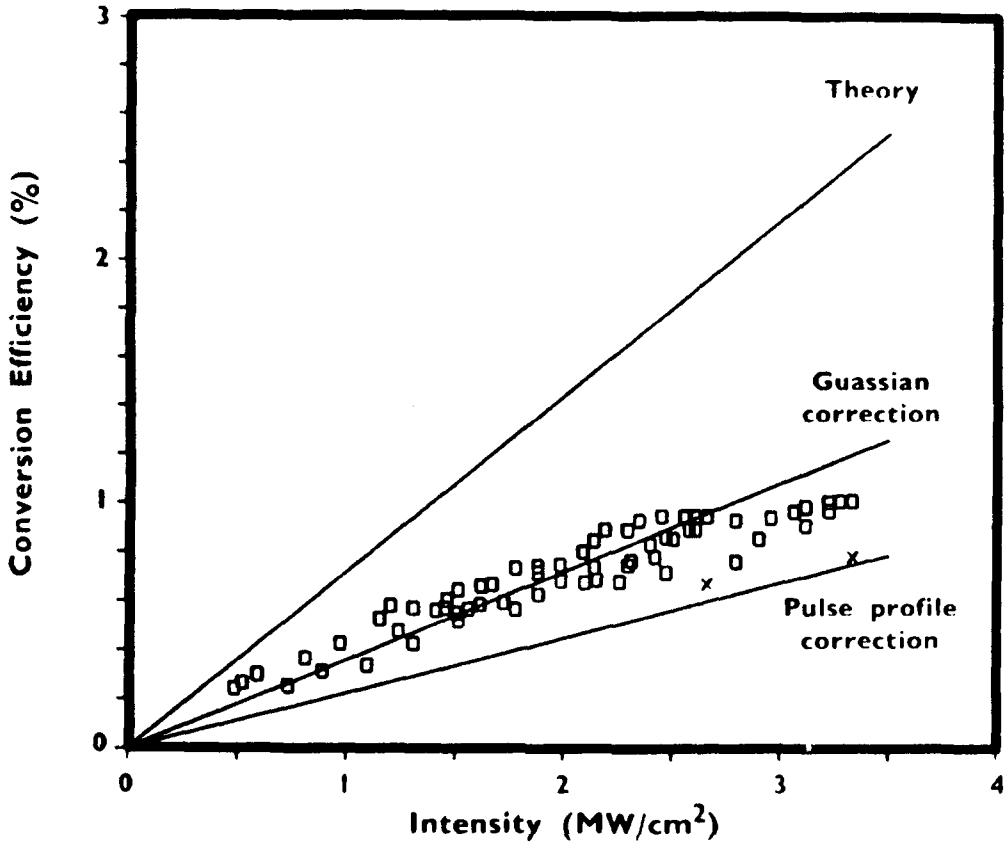
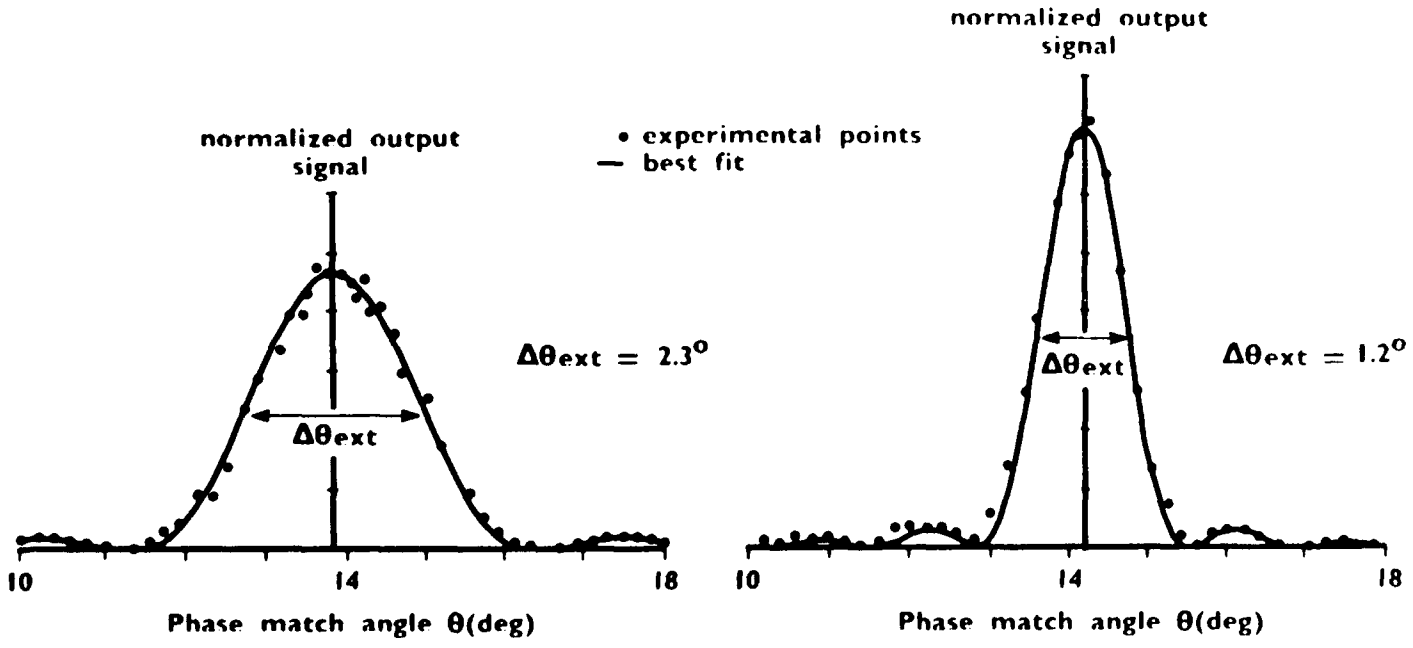


FIG 8

Tuning Curves



a) U.K. Crystal (3)

b) U.S. Crystal (2cm)

FIG 9

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