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1979

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ABSTRACT

A grating tuned $LiNbO_3$ parametric oscillator-amplifier system has been constructed and operated at a repetition rate of 10 pps using an unstable Nd:YAG oscillator-amplifier system as the pump. Peak power gains of the order of 10 have been obtained from a 5 cm $LiNbO_3$ crystal, with more than 20% energy conversion efficiency from the pump into parametric wavelengths. Frequency mixing of the signal and idler in CdSe has produced 50 μJ of tunable radiation at 16 μm with peak powers of up to 7 kW.

(Accepted for publication in Optics Communications)

June 1978

## Introduction

The grating tuned $LiNbO_3$ optical parametric oscillator (OPO) has recently become an important device for tuning in the near infrared [1, 2, 3]. Frequency mixing of the signal and idler in CdSe extends the tunability range further into the infrared [3]. Parametric devices using nonlinear crystals have hitherto been limited to working as oscillators due to the lack of long, good quality high-gain crystals. The $LiNbO_3$ crystals now available, with typical dimensions up to 15 mm diameter and 50 mm long, can provide sufficient parametric gains to suggest the feasibility of optical parametric amplifier (OPA) operation. An optical parametric amplifier pumped with the annular beam from an unstable Nd:YAG laser, has very recently been reported at a conference in the USA [4].

Straightforward calculations indicate that the small signal gain of a 5 cm $LiNbO_3$ crystal is of the order of $\sim 10^4$ for a $10^{12}$ watts/$m^2$ pump beam. On this basis we have set out to produce a parametric oscillator-amplifier system with subsequent mixing of the signal and idler in CdSe to extend the tunability range further into the infrared. The pump beams for the OPO and OPA originated from an Nd:YAG oscillator and were both brought to the far field before entering the crystals. A comparison between stable and unstable Nd:YAG oscillator beams for pumping parametric oscillators has been made. The important factors in the operation of the OPA are pointed out and qualitative measurements of the peak power and energy gains have been made. Line narrowing problems are discussed, together with calculations for the operation of bigger systems and frequency mixing in CdSe for the production of 16 $\mu$m radiation.

## The experimental arrangement

A commercially available Nd:YAG oscillator - model 2000 from J K Lasers - was turned into an unstable resonator according to the design described in [5] and [6]. The output beam from the unstable oscillator was propagated to the far field by sending the beam across the laboratory and using beam expanders to bring it to the required spot size. The beam was then apodized with more than 90 mJ available in the central peak to pump the parametric oscillator. The back mirror of the unstable Nd:YAG oscillator was partially transmitting, and the emerging beam was propagated to the far field using two 3:1 beam expanders, before entering a 3 x $\frac{1}{4}$" Nd:YAG amplifier. Up to 100 mJ were available from the latter to pump the OPA, and this beam was synchronized with the output from the Nd:YAG oscillator by using a long optical delay.

Fig. 1 shows the experimental arrangement of the parametric oscillator-amplifier system with frequency mixing in CdSe. The parametric oscillator was similar to that described in [3] utilizing a 50 mm long and 10 mm diameter $LiNbO_3$ crystal which was temperature controlled in a copper oven. The OPO cavity consisted of a 600 lines/mm replica grating blazed at 1.85 μm, a dichroic mirror transmitting the 1.064 μm and reflecting the 2.0 μm radiation at $45^o$ and a 56% output mirror (broadband around 1.9 μm) with a radius of 25 m. A 1 mm thick etalon could be inserted between the grating and the dichroic mirror. The pump beam was reflected back through the system to provide gain in the backward direction as well. The parametric amplifier was a similar crystal mounted on Burleigh mounts supported by four invar bars for stability. The OPO output and the pump beam for the OPA were made collinear using a dichroic mirror which had 98% T at 1.06 μm and 100% R at 2 μm at $45^o$.

Frequency mixing in CdSe was achieved using the crystal in the manner described in ref. [3]. Energy measurements were made using pyroelectric detectors and the pulses were recorded using InSb and Ge detectors.

## Experimental Results

The unstable Nd:YAG oscillator beam was propagated to the far field and reduced to a spot size of 1.25 mm to pump the parametric oscillator. The pulses had a duration of 11 ns FWHM and energies up to 80 mJ, with energy outputs from the OPO up to 3.2 mJ. It is important to point out here that the OPO was neither as reliable nor as efficient as the one pumped with the equivalent stable Nd:YAG oscillator-amplifier configuration. In general we required nearly twice as much pump energy density to get the same output out of the OPO using the unstable Nd:YAG oscillator as compared with the stable configuration [3]. We attributed two reasons to this discrepancy. First the pump pulses are very short and there is not enough time after the build up for the signal to interact with the pump. From a number of experiments which we have carried out we have found that the optimum pump pulse duration for minimum energy density is approximately 20 ns. Secondly it was difficult to control the stability of the pump beam to a high degree of accuracy since it was propagated more than 15 m before reaching the OPO. However this arrangement enabled us to have the energy of the Nd:YAG amplifier available for pumping the parametric amplifier. The spot size of the pump beam at the OPA position was 1.6 mm and this beam was delayed by 3 ns with respect to the oscillator pump beam in order to be synchronized with the OPO pulse. Typical operating parameters of the system were:

| | |
|---|---|
| OPO pump pulse | 11 ns |
| OPO output pulse | 10 ns |
| OPO spot size | 1.2 mm |
| OPA pump spot size | 1.6 mm |

It is obvious that not all the available energy from the pump into the OPA was used during the parametric amplification due to the mismatch of the spot sizes.

The small signal gain of an OPA of length L is given by

$$G = \frac{I_s(L)}{I_s(0)} - 1 = \frac{\Gamma^2}{g^2} \sinh^2 gL \qquad (1)$$

where $$g^2 = \Gamma^2 - \left(\frac{\Delta k}{2}\right)^2$$

and $$\Gamma^2 L^2 = u_s u_i \omega_s \omega_i |d_{eff}|^2 |E_p|^2 L^2$$

$$= \frac{\omega_s \omega_i}{2 \eta_i \eta_s \eta_p \epsilon_o c^3} |d_{eff}|^2 \left(\frac{P_p}{\frac{1}{2}\pi w_p^2}\right) L^2 \qquad (2)$$

where $\omega_s$ , $\omega_i$ are the signal and idler frequencies respectively, $\eta$'s are the refractive indices, $P_p$ is the pump power and $w_p$ is the pump spot size. The effective nonlinear coefficient $d_{eff}$ is given by [1]

$$d_{eff} = d_{31} \sin(\theta_m + \rho) - d_{22} \cos(\theta_m + \rho) \sin 3\varphi \qquad (3)$$

where $d_{31}$ and $d_{22}$ have opposite signs [7] and $\theta_m$ is the phase matching angle and $\rho$ the double refraction angle ($\rho = 2.17^o$ and $\theta_m = 44.825^o$ at 2 μm). Since propagation takes place in the +Z -Y direction, $(\theta_m + \rho)$ is negative and $\varphi = 270^o$ and with $d_{31}$ and $d_{22}$ being of opposite sign eq. (3) becomes:

$$|d_{eff}| = |d_{31}| \, |\sin(\theta_m + \rho)| + |d_{22}| \, |\cos(\theta_m + \rho)| \qquad (4)$$

Substituting $d_{31} = 6.25 \times 10^{-12}$ m/V and $d_{22} = 2.9 \times 10^{-12}$ m/V , we find the effective nonlinear coefficient for $\lambda_s \simeq 2$ μm to be $|d_{eff}| = 6.55 \times 10^{-12}$ m/V Substituting this value in (2) together with $\epsilon_o = 8.85 \times 10^{-12}$ F/m we find that for $\lambda_s = 1.995$ μm ($\eta_s = 2.207$, $\eta_i = 2.201$ and $\eta_p = 2.165$)

$$\Gamma^2 L^2 = 0.664 \times 10^{-8} \left(\frac{P_p}{(\frac{1}{2}\pi w_p^2)}\right) L^2 \qquad (5)$$

where $P_p$ is in watts, $w_p$ is in metres, and L is in metres. For a pump beam with $w_p$ = 1.6 mm, $P_p \approx 8$ MW and a crystal of length 0.05 m we find $\Gamma^2 L^2 \simeq 36$ . From eq. (1) we find the small signal gain of the parametric amplifier to be $G = \frac{1}{4} e^{2\Gamma L} \approx 4 \times 10^4$ . These high gains (which is the gain of the signal when no idler is injected) suggest that if the signal and the pump only were injected into a parametric amplifier the parametric interaction should be easily achievable with the idler being generated at each position of the crystal according to the phase matching condition and the optimum phase angle.

In experiments which we have carried out we have found that it was not necessary to inject the idler into the parametric amplifier in order to obtain parametric interaction. High gains were obtained by injecting the pump and the signal only into the amplifier crystal. We have also found that the idler beam was grossly divergent compared with the signal beam and that the idler divergence depended on the relative divergences of the signal and pump. We have concluded that as the pump and signal propagate through the amplifier crystal the idler is generated in such a way as to satisfy the phase matching condition and the optimum phase angle. The important factor in the operation of OPA is to have two matched parallel beams (long beam waists) for the pump and the signal within the crystal.

Fig. 2 shows the intrinsic peak power gain of the parametric amplifier as a function of pump energy for three different input energies from the parametric oscillator. It can be seen that the peak power gain is independent of the input energy from the parametric oscillator. The small difference in the slopes of the curves is attributed to the changes of the spot sizes and divergences of the beams for different pumping energies. We recall here that the spot size of the parametric oscillator beam (1.2 mm) did not completely overlap with the spot size of the pump beam (1.6 mm). It should therefore be possible to obtain much higher gains by proper beam waist matching.

Fig. 3a shows twenty superimposed pulses of the input and output pulses of the parametric amplifier. It can be seen that maximum gains occur at the peak powers. The synchronization of the parametric oscillator pulses with the pump pulses must be optimized for highest gains to be attained. Figure 3c shows the pump beam polaroid burn patterns at the position of the OPA, whilst Fig. 3d shows the parametric amplifier output beam polaroid burn patterns.

Finally line narrowing was achieved using etalons with finesse 6 in the parametric oscillator as well as in the unstable Nd:YAG oscillator. When an etalon was placed in either the Nd:YAG oscillator or the parametric oscillator no change in the output energy, efficiency or reproducibility in the performance of the system was observed. However when both were narrowed, the output of the parametric oscillator diminished considerably and the system was much less reliable. The reason for this is that the number of interacting mode combinations between pump and idler to give a single signal mode has diminished enormously, and since the pump pulses were short (~ 11 ns) compared to the cavity round trip period there was not enough time for the interaction of the pump with the signal after the latter grew from noise.

Frequency mixing for 16 μm radiation

The signal and idler beams from the parametric amplifier were mixed in a CdSe crystal 4.1 cm long to produce 16 μm radiation in the manner described in [3]. The parametric oscillator-amplifier system was tuned to the wavelengths 1.9948 μm and 2.28023 μm, which correspond to a frequency difference of 627.45 $cm^{-1}$ (15.9375 μm). The bandwidth of the generated radiation was approximately 2 $cm^{-1}$ and the peak of the spectrum was stable to better than 0.1 $cm^{-1}$ from shot to shot. Frequency narrowing of the beams using the intracavity etalons has produced narrow band radiation at 16 μm estimated at better than 0.1 $cm^{-1}$ (no better than 0.4 $cm^{-1}$ bandwidth could be measured with our spectrometer).

Figure 4 shows the output power at 16 μm plotted against the product of the input powers when no etalons were used. Pulses with energies of more than 50 μJ and powers of 7 kW were produced. These are much less than expected for high pumping energies (the dotted line represents the theoretically expected values). The curve follows closely the expected values for low pumping energies according $P_{16} = 1.2 \times 10^{-12}\ P_i\ F_s / 2\pi w^2$ and is a good continuation of the results described in [3]. Investigations have indicated that the idler beam was grossly divergent for high pumping powers due mainly to the change in the divergence of the pump beam with increasing pumping of the Nd:YAG amplifier rod. Since the beam had to travel 15 m before reaching the parametric amplifier and the thermal lensing of the rod for high pumping resulted in a focal length of less than 10 m, it was very sensitive to the pumping. We have found that at the position of the mixing crystal the idler beam cross-section for high pumping powers was nearly twice that of the signal, and this is in good agreement with the results of Fig. 4. Another interesting result is that 50 μJ at 16 μm corresponds to more than 1% energy conversion efficiency of the effective signal energy used in the mixing process.

Experiments with a stable Nd:YAG oscillator system

An experimental system similar to that described in [3] was constructed but where high finesse etalons were used to produce narrow band beams. We have introduced a 2 mm etalon with a finesse of 8 in the Nd:YAG oscillator cavity which operated at repetition rates of up to 20 pps and reproducibility of better than 98% from shot to shot. The output was beam expanded and amplified through an Nd:YAG rod to give 70 mJ in 20 ns with a spot size of 1.5 mm. An etalon with a finesse of 8 was used in the parametric oscillator and the output beams were mixed in CdSe to produce 5 μJ of narrow band radiation at 16 μm, whose reproducibility was better than 85%. From the finesse of the etalons and the gain bandwidths we estimated that the resulting radiation had a bandwidth of less than 0.1 $cm^{-1}$, although the best we could actually measure was 0.4 $cm^{-1}$ due to the low resolution of our spectrometer.

## Discussion and Conclusions

A $LiNbO_3$ optical parametric amplifier has been constructed and operated at repetition rates of 10 pps with peak power gains of 10 and more than 20% energy conversion efficiency from the pump into parametric wavelengths. The most important factor in the operation of parametric amplifiers is that the signal and idler beams are parallel (they form long beam waists) at the position of the crystal so that the generated idler has a low divergence. We see no problem in introducing a second parametric amplifier crystal after the first to provide further conversion of the pump into parametric wavelengths. The second OPA should be placed with its optic axis opposite to that of the first so that the walk-off angles are in opposite directions and the full length of the crystal is utilized effectively. The operation of parametric amplifiers opens up the field of tunable lasers in the infrared to energies in excess of 100 mJ.

A comparison of the performance of the $LiNbO_3$ parametric oscillator when pumped using a stable Nd:YAG oscillator-amplifier system and when using the far field of an unstable Nd:YAG oscillator has been made. Four important aspects are pointed out:

(i) The pulses produced by the Nd:YAG unstable oscillators ($\sim$ 11 ns) are too short for the efficient operation of parametric oscillators. No unstable Nd:YAG oscillator system at present has been reported producing longer pulses. In general we required more than twice the pump energy densities for pulses of 11 ns duration to produce the same output energy from the parametric oscillator as with pulses of 20 ns duration.

(ii) The long distances which the beams from the unstable Nd:YAG oscillator have to travel to come to the far field make the beam waists strongly dependent on the pumping of the Nd:YAG rods, and also the accuracy of the system becomes much lower. Attempts to bring the beam to the far field using 3:1 inverted beam expanders have failed as a result of damage to the negative lens caused by the high intensity of the reduced beam.

(iii) The beam waists of the Gaussian beams are easily calculable as described in [3] and the crystals can be placed at the optimum positions for best operation.

(iv) Frequency control of the pump beam using high finesse etalons is easier using a stable oscillator, and frequency control of the parametric oscillator using high finesse etalons is also made more reliable due to the longer duration of the pump pulses. In general the use of a stable Nd:YAG oscillator in conjunction with two amplifiers is preferable to the unstable resonator system.

Frequency mixing in CdSe has hitherto produced 50 μJ with 7 kW and the extension of the system to the 1 mJ region is a matter of scaling up the system. It has been shown that a reliable narrow band system operating at 15.937 μm can be constructed with good frequency stability and bandwidth (better than 0.1 $cm^{-1}$). One of the interesting questions is the energy at 16 μm which such a system can provide with present day available crystals and equipment. Taking the maximum beam damage intensity for CdSe to be 60 $MW/cm^2$ [1] we find that for pump pulses of 15 μs duration, and a beam spot size of 1.7 mm the maximum energy which can be allowed to fall on the crystal is 40 mJ (signal = 21.3 mJ, idler = 18.7 mJ according to their respective quantum energies for 16 μm). These energies can now be easily obtained from parametric amplifiers, and the intensities are high enough to produce saturation effects in the mixing of the beams if the crystal is sufficiently long. It should be possible to use two CdSe mixing crystals, each 5 cm long, with their axes in opposite directions to eliminate beam walk-off effects, providing a total mixing length of 10 cm. It is also possible to propagate the beams at the correct polarizations in the CdSe so that the total powers are used in the mixing process [8]. Thus even if we allow for a modest 5% conversion efficiency (less than half the theoretical maximum) it is possible to produce more than 1 mJ at 16 μm with peak power ⩾ 100 kW (already 1% conversion efficiency has been achieved in the present work with a 4.1 cm long crystal). The above considerations were based on beam parameters for which parametric devices have actually been operated at. It should be possible to expand these systems to much higher outputs, although at the moment it does not look as if the efficiency of the machine can be improved to better than $0.5 \times 10^{-4}$.

Acknowledgements

The author would like to thank Mr K Fletcher of Culham Laboratory for the production of the specialized optical coatings for this experiment. He would also like to thank Drs I J Spalding and A C Selden for their interest shown in this work.

13 June 1978

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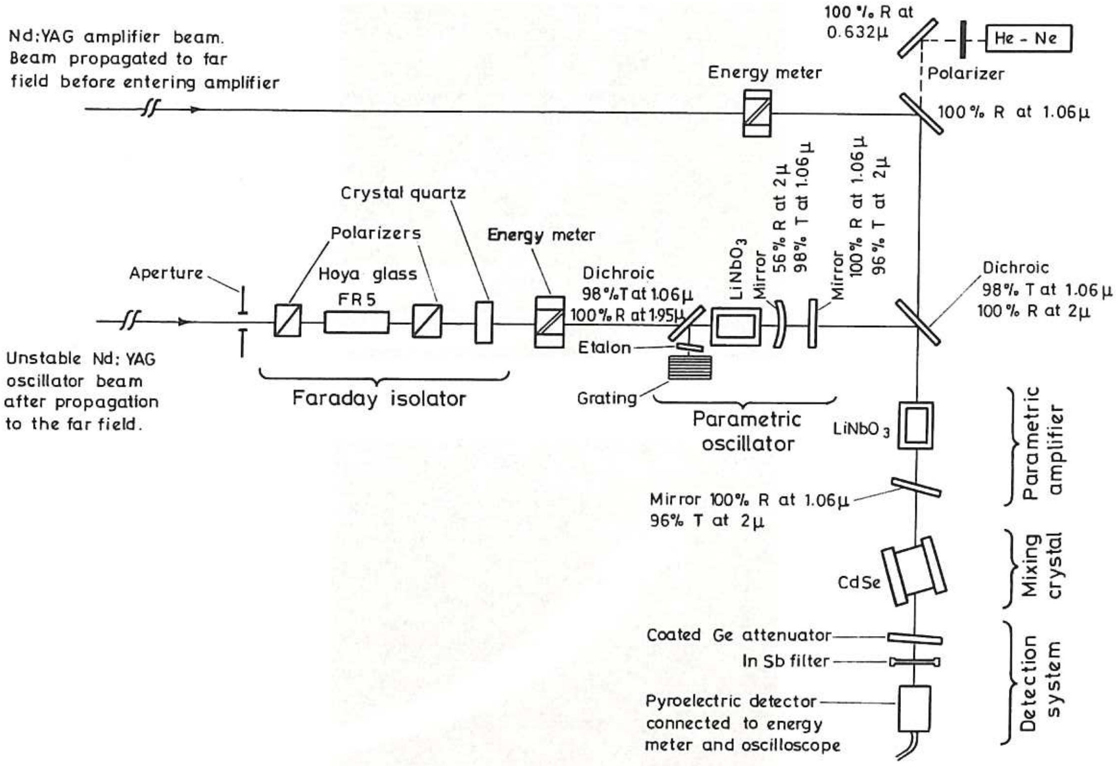



**Fig.1 Experimental arrangement of the parametric oscillator-amplifier system together with frequency mixing in CdSe.**

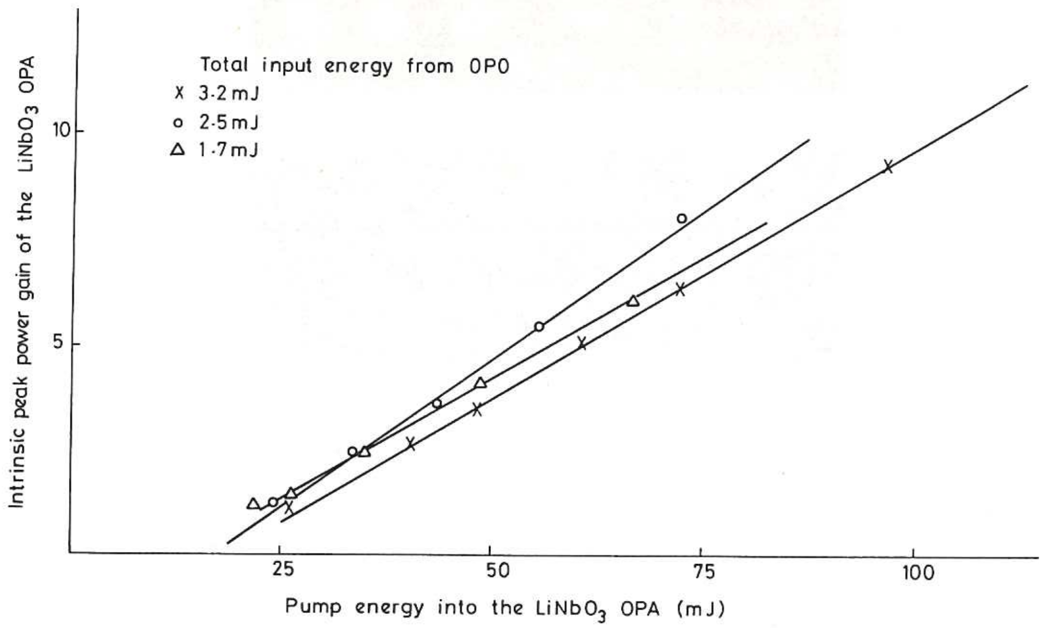
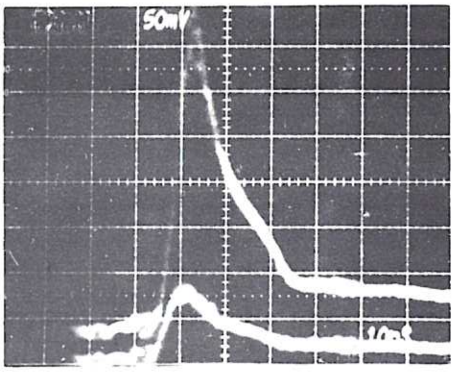



**Fig.2 The peak power gain of the $LiNbO_3$ parametric amplifier as a function of pump energy. Pump spot size 1·6 mm.**

(a)

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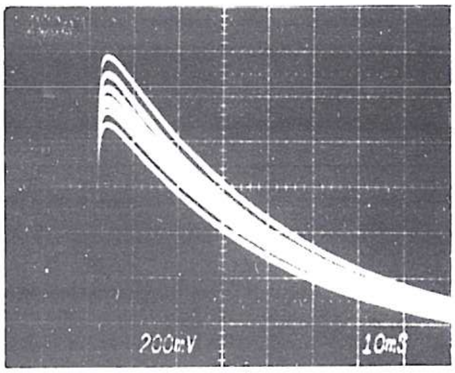


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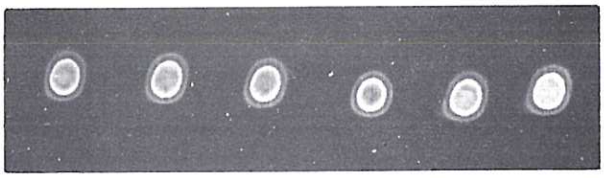

(d)

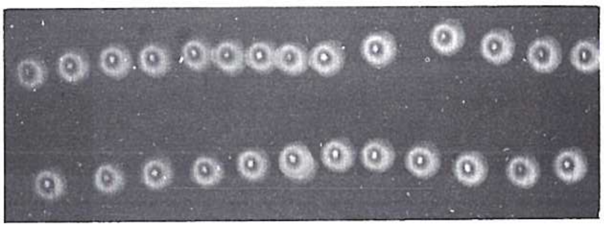

Fig.3 (a) Twenty superimposed pulses showing the input into the parametric amplifier (lower trace) and the output of the parametric amplifier (upper trace) showing a peak power gain of 5. (b) Twenty superimposed pulses at 15·937$\mu$m with an average energy of 50$\mu$J per pulse recorded using a pyroelectric detector. (c) Pump beam of the parametric amplifier with a spot size of ~ 1·6 mm. (d) Output beam of the parametric amplifier (signal and idler) with a spot size of ~ 1·2 mm.

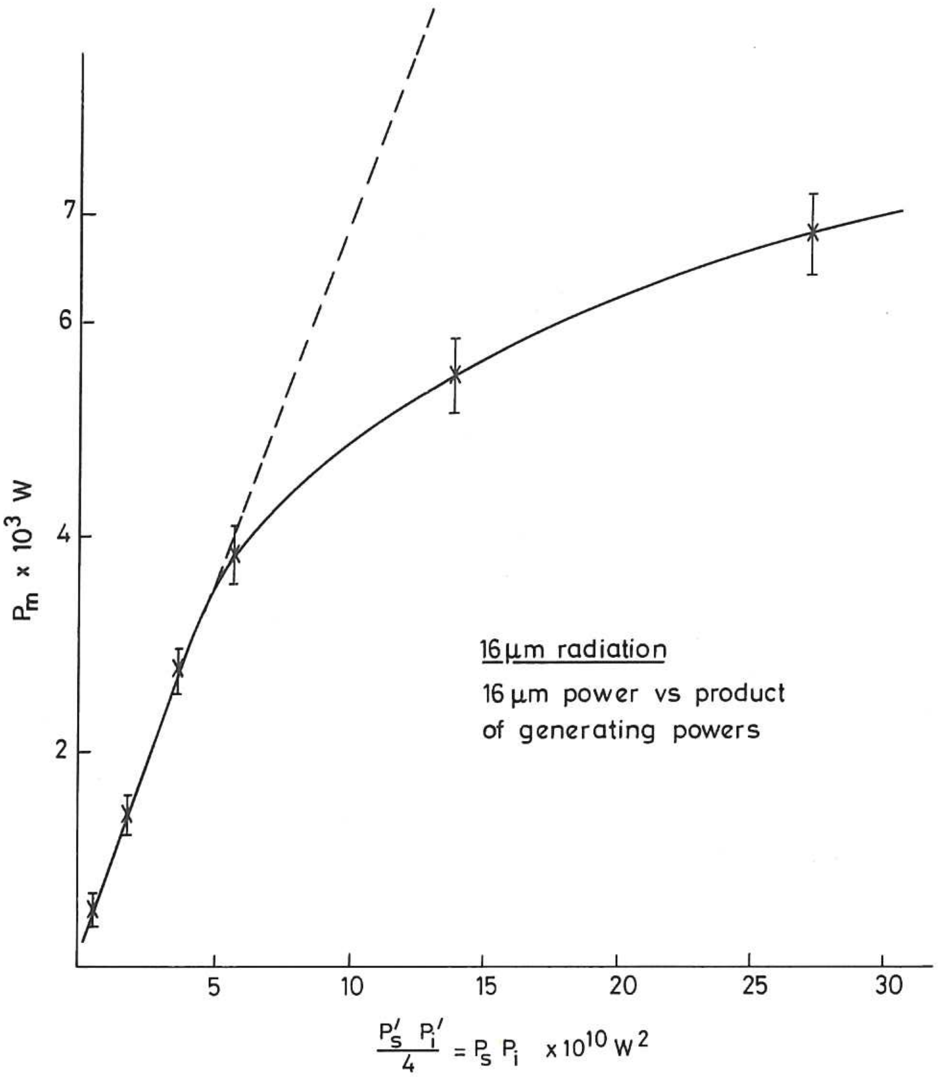


Fig. 4 The output power at 15·937 µm as a function of the product of the powers of the generating beams. The dotted line represents the theoretically expected curve.