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ABSTRACT

A reliable continuously tunable source of 16μm radiation with ~0.5 kW peak power and repetition rates up to 20 pps is described. This consists of an Nd:YAG driven lithium niobate return pump parametric oscillator, with non-linear mixing of the signal and idler wavelengths in cadmium selenide. Pulse energies of 5 μJ and linewidths of 2.6 cm^{-1} at 16μm have been observed with the grating tuned OPO. Potential improvements in output energy and bandwidth (spectrum narrowing) are discussed.

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INTRODUCTION

The prospect of laser isotope separation has stimulated great
interest in the production of tunable sources at $16 \mu m$. A number of
sources for the production of such radiation and a variety of approaches
to this problem have been reported. A CdSe optical parametric
oscillator pumped by an HF oscillator-amplifier has produced 2.5 mJ
with bandwidths ranging from 9 cm^{-1} to 1.8 cm^{-1} and pulse lengths of $\sim$100
ns [1]. An optically pumped CO_2 laser has produced 300 μJ in 200 ns [2].
Laser action at $16 \mu m$ has also been reported from a gas dynamic CO_2 laser
[3], where energies in excess of 10 μJ with pulse widths of 150 ns have
been obtained.

Parametric processes using non-linear crystals have proved to be a
versatile method for tuning into the infra-red [4,5] Hanna et al [6] have
reported difference mixing in CdSe using the signal and idler of a
proustite (Ag_3AsS_3) parametric oscillator with output powers of a few
watts tunable from 10-22 μm. The emergence of an Nd:YAG pumped $LiNbO_3$
parametric oscillator capable of producing high energy outputs [7] and
its subsequent operation using a grating and a dichroic mirror [8,9] have
indicated the possibility of obtaining high energies with narrow
linewidths in the infra-red by mixing its signal and idler outputs in
CdSe.

On this basis we have set out to produce a reliable, high repetition
rate source generating narrow band tunable radiation at 16 μm by mixing
the signal and idler of a $LiNbO_3$ parametric oscillator in a CdSe crystal.

Initial efforts were concentrated on the reproducibility of the system,
which can now deliver over 5 µJ of tunable radiation in the 15-17 µm region
with a calculated power of over $\frac{1}{2}$ kW and a bandwidth of 2.6 cm^{-1}, at repet-
ition rates of up to 20 pps. There appears to be no problem in expanding the
system to deliver pulses with energies in excess of 100 µJ and much narrower
bandwidths. These possibilities, together with the more immediate aims
are discussed in the conclusions.

THE Nd:YAG SYSTEM AND FOCUSING

The experimental arrangement is shown in Fig. 1. A commercially available
Nd:YAG oscillator-amplifier system - model 2000 from J K Lasers - was used
in the experiments. The oscillator and amplifier heads contain 3" x $\frac{1}{4}$" rods
pumped by linear flashlamps and cooled with a recirculated liquid whose
temperature was maintained constant through a thermostat and a heat exchanger.
The system is capable of repetition rates of up to 40 pps with Q-switched
output energies of over 100 mJ. It is well known that the reliable operation
of parametric oscillators is very critically dependent on the quality of the
transverse mode. The construction of a completely reliable Nd:YAG oscill-
ator with good spatial and temporal stability is therefore a necessity.
Figure 2 shows the reproducibility of the Nd:YAG oscillator at 10pps. The
oscillograms show 10 superimposed pulses in (a) time taken using a fast
photodiode with rise time 0.4 ns, and (b) space taken with a beam scanning
array. The oscillator delivered 12 mJ in 20 ns and with a spot size at the
exit mirror (plane) of 0.77 µm. No attempt was made to narrow the spectrum which
was approximately 1 cm^{-1}. A 2:1 Galilean telescope placed at 45 cm from the
exit mirror was used to expand the beam before entering the amplifier. It is
important that for completely reliable operation the parametric oscillator
is placed at the beam waist of the expanded Gaussian beam. Thus, with the
parameters of the oscillator beam measured, the position of spot size and the
Rayleigh range of the expanded beam waist had to be calculated.

Following the algebraic methods described in [10,11], expressions for
the transformation of the Gaussian beam parameters by a lens have been derived.
A Gaussian beam defined by

$$z_o = \frac{\pi w_o^2}{\lambda} \qquad w(z) = w_o \left[1 + \left(\frac{z}{z_o} \right)^2 \right]^{\frac{1}{2}} \qquad R(z) = \frac{1}{z} \left[z^2 + z_o^2 \right] \qquad ..(1)$$

where w_o is the spot size and z_o the Rayleigh range, is transformed by
a lens with focal length f to a Gaussian beam with new parameters w_1, z_1
obtained from

$$R_1 = \frac{fR_\ell}{f - R_\ell} \qquad\qquad z_1 = \frac{\frac{\pi w_\ell^2}{\lambda} R_1^2}{R_1^2 + \left(\frac{\pi w_\ell^2}{\lambda} \right)^2}$$

$$\ell_1 = \frac{\left(\frac{\pi w_\ell}{\lambda}\right)^2 R_1}{R_1^2 + \left(\frac{\pi w_\ell^2}{\lambda}\right)^2} = \frac{\pi w_\ell^2}{\lambda} \cdot \frac{z_1}{R_1} \qquad w_1 = \frac{\lambda}{\pi w_\ell} \sqrt{R_1 \ell_1} \qquad \qquad ..(2)$$

where

R_ℓ is the radius of curvature of the beam immediately before the lens

R_1 is the radius of curvature of the beam immediately after the lens

w_ℓ is the beam size at the lens

ℓ_1 is the distance of the new beam waist from the lens

and w_1 , z_1 are the new spot size and Rayleigh range respectively.

The 2:1 beam expander used, consisted of a plano-concave and a plano-convex lens with focal lengths of -7.5 cm and 15 cm respectively. The beam expander was placed at z = 45 cm from the exit mirror of the Nd:YAG oscillator. Applying relations (2) successively to the two lenses of the beam expander we find that for a lens separation of 7.7 cm a beam waist of 1.5 mm is formed at 260 cm from the exit lens of the beam expander with a Rayleigh range of 670 cm. The parametric oscillator was placed at 220 cm from this lens and the mixing crystal 35cm further along. No other lenses were used. For the present experiment the lensing of the amplifier rod was not taken into account since at our normal repetition rate of 10 pps and the maximum pumping used the focal length was longer than 8 m. Measurements of the lensing in the amplifier rod showed that its focal length could be approximated linearly according to $f(m) = \frac{1.25}{P(kW)}$.

The beam spot size at the position of the parametric oscillator was measured with a scanning diode array to have a radius of 1.52 ± 0.02 mm at the $\frac{1}{e^2}$ point which is in very good agreement with the calculated spot size. This spot size did not change at all with the pumping of the amplifier. The pulses however became less sharp and a slight degree of saturation could be detected at the peak for the maximum pumping used (60 mJ output). The FWHM of the Gaussian beam increased by 0.2 mm from the unpumped amplifier to the maximum pump energy used in the experiments, due to the slight saturation occurring. The reproducibility of the pulses in time was as good as that of the Nd:YAG oscillator, as can be seen from the oscillograms of Fig. 3.

The Nd:YAG system was followed by a Faraday isolator to facilitate the use of a return pump parametric oscillator. Its extinction ratio was measured to be better than 10^{-4}. A 7.4 mm thick crystal quartz plate was placed after the Faraday isolator to return the plane of polarization to

its original orientation. Alignment of the whole system was done with a
He-Ne laser placed behind the Nd:YAG oscillator. A second He-Ne collinear
with the first and with the laser radiation was placed in the manner shown
in Fig. 1 to facilitate the alignment of the apparatus from the parametric
oscillator onwards.

THE PARAMETRIC OSCILLATOR

The parametric oscillator was constructed using Burleigh mounts for
the grating and the output mirror, supported by four invar bars. Since
our final aim is to achieve very narrow band radiation with good wavelength
stability, invar was used for all distances affecting the length of the para-
metric oscillator. The LiNbO$_3$ crystal obtained from Crystal Technology was
5 cm long and 1 cm in diameter, antireflection coated at 1.8 μm and its
properties are described in [7]. It was held in a copper oven, although
no temperature control was attempted in the present experiments. The para-
metric oscillator cavity consisted of a 600 lines/mm replica grating from
Bausch and Lomb blazed at 1.85 μm, a dichroic mirror transmitting the
1.064 μm reflecting the 1.9 μm radiation at 45° and a 56% output mirror
(broadband around 1.9 μm) with radius of 25 m. The total cavity length
from the grating to the mirror was 11 cm. The arrangement was similar to
that described in [8] apart from that a return pump beam was used providing
gain in the backward direction as well. The output mirror and the mirror
returning the pump pulse were made of water free silica to avoid any absor-
ption of the generated wavelengths. The parametric oscillator was
tuned continually from 1.55 μm (signal) to 3.5 μm (idler) with the lower
threshold occurring around 1.97 um due to the blaze of the grating and the
increasing reflectivity of the output mirror.

One of the important aspects in the operation of the system were the
dichroic mirrors which were all made in our laboratory. The dichroic mirror
at 45° had a transmission of 98% at 1.06 μm and a reflectance from 1.4 μm to
2.1 μm of better than 99.5%. The output mirror had a broadband coating with
a reflectivity of 56% at 2 μm and 67% at 1.8 μm. Its transmission at 1.064
was better than 97%. The mirror reflecting the pump was better than 99.9%
whilst at the same time its transmission at 1.995 μm was 97% and at 2.28 μm
94% the two wavelengths of interest for the generation of 16 μm radiation.
The very high reflectivity of this mirror at 1.064 μm whilst at the same
time maintaining high transmission at the wavelengths of interest is very

important because of the low damage threshold of CdSe. We required no extra filters for the 1.064 µm radiation between this mirror and the CdSe crystal.

THE CdSe MIXER

Mixing of the signal and idler was achieved in a CdSe crystal 4.1 cm long and 1 cm^2 square cross section, cut at 70° to the optic axis. Only type IIa phase matching is allowed in this crystal, according to the relation

$$\eta_{ir}^{o}(\omega_{ir}) = \frac{\omega_s}{\omega_{ir}} \eta_s^{o}(\omega_s) - \frac{\omega_i}{\omega_{ir}} \eta_i^{e}(\omega_i) \qquad ..(3)$$

Since the signal and idler from the OPO are polarized in the same direction the CdSe crystal was set at 45° with respect to their polarization. This meant that only half of the available intensities of the signal and idler were used in the mixing process.

In order to reject the powerful radiation from the parametric oscillator special coatings were produced on Ge substrates. We were not able to make them broad enough to cover both wavelengths and a total transmission of 4% still remained. Work towards this goal is now in progress in our laboratory. An InAs filter was used to remove residual radiation from the parametric oscillator.   The combined rejection of the filter at 2 µm was better than 0.2×10^{-4} and had a transmission of 22% at 16 µm mainly due to the reflectivity of Ge. We are hoping to produce such filters with better than 90% at 16 µm by using antireflection coated Ge.

EXPERIMENTAL RESULTS

The LiNbO$_3$ parametric oscillator was tuned to the wavelengths 1.995 µm for the signal and 2.28 µm for the idler whose difference frequency corresponds to 16 µm. The CdSe crystal was set at 64.22° to its optic axis which is the angle for phase-matching these two wavelengths. Figure 3 shows oscillograms for each of the wavelengths involved in the production of 16 µm radiation. Fig. 3a shows 10 superimposed pulses of the 1.064 µm radiation (silicon detector) and 10 superimposed pulses of the 2.28 µm radiation (InAs detector with Ge filter) simultaneously recorded using a 7844 dual beam Tektronix oscilloscope. Fig. 3b shows 10 superimposed pulses of the 1.064 µm (silicon detector) and 10 superimposed pulses of the 1.995 µm radiation (Ge detector with Si filter) simultaneously recorded as in 3a. The time resolution of the Ge detector was not fast enough and the pulse width shown for the signal is not representative. The signal and idler pulses were 15 ns in duration and were shorter than the 1.064 µm pulses of duration ∼20 ns.

The system was 100% reliable and the reproducibility remained the same for repetition rates of 5, 7, 10 and 20 pps. The normal repetition rate of operation was 10 pps. Figure 3.c shows 10 superimposed pulses of the 16 μm radiation as recorded with a slow pyrolectric RKP 335 from Laser Precision. Other pyrolectic detectors (for example Plessey) have been used for the detection of the 16 μm radiation and they indicated the same reliability.

Fig. 4 shows the energy and power characteristics of the system. Figure 4a shows the percentage energy conversion efficiency of the 1.064 μm radiation into signal and idler wavelengths against the number of times above threshold. The threshold of the return pump parametric oscillator was 22 mJ. The maximum pump energy used of 60 mJ with a spot size of 1.53 mm corresponds to an energy density of 1.65 J/cm^2. This is well below the maximum allowable figure for the damage threshold of $LiNbO_3$, which has been reported to be above 2 J/cm^2 [8] but we did not want to risk the crystal until we had made damage measurements in our laboratory. Figure 4b shows the output energy at 16 μm plotted against the input energy (signal + idler) for the parametric oscillator. Energy measurements were taken using a Laser Precision pyroelectric energy meter RK 3232. All error lines in figs. 4a,b represent the maximum variation in the readings of the energy meter.

Fig. 4c shows the output power at 16 μm plotted against the product of the input powers. Since the CdSe crystal was set with its axis at 45° to the direction of polarization of the input beams only half the available powers were used and thus the product of the powers taking part in the interaction $P_s P_i$ was one quarter of the product of the total powers $P'_s P'_i$. The power of the 16 μm radiation plotted was calculated by assuming that the pulse length is 11 ns. i.e., approximately $\sqrt{2}$ shorter than that of the interacting Gaussian beams. The powers of the signal and idler were calculated by assuming that their energies were proportional to the quantum energies of their respective photons.

The intensity I_{16} of the generated beam is given by

$$I_{16} = I_i \frac{\omega_{16}}{\omega_i} \sinh^2 \Gamma L \qquad\qquad ..(4)$$

where L is the length of the crystal and $\Gamma^2 = \dfrac{\omega_{16}\omega_i}{4\eta_{16}\eta_i c^2} |d_{eff}|^2 |E_s|^2$,

d_{eff} is the effective nonlinear coefficient of CdSe, η's are the respective refractive indices $I_s = \frac{1}{2}\eta_s c \varepsilon_o |E_s|^2$ is the intensity of the signal. For small conversion efficiencies of less 40% $\sinh \Gamma L \approx \Gamma L$, and making these substitutions eqn. (4) becomes

$$I_{16} = \frac{\omega_{16}^2 |d_{eff}|^2}{2 \eta_{16} \eta_i \eta_s c^3 \varepsilon_o} I_i I_s L^2 \qquad ..(5)$$

from which it is seen that the generated intensity varies according to the product of the intensities of the other two beams. Substituting the numerical values of the quantities in (5)

$\omega_{16} = 1.178 \times 10^{14} \ sec^{-1}$, $d_{eff} = 19 \times 10^{-12} \ m/V$ [5], $c = 3 \times 10^8 \ m/s$

$\eta_{16} = 2.41 \quad \eta_i = 2.482 \quad \eta_s = 2.472$ [12], $\varepsilon_o = 8.85 \times 10^{-12} \ F/m$

and $L = 0.041 \ m$ we find

$$I_{16} = 1.2 \times 10^{-12} \ I_i I_s \qquad ..(6)$$

where the intensities are expressed in W/m^2. In terms of the power relation (6) can be written as

$$P_{16} = 1.2 \times 10^{-12} \frac{P_i P_s}{2 \pi w^2} \qquad ..(7)$$

where w is the parametric oscillator spot size and the factor of 2 is due to the approximation of the 16 µm radiation spot size by $w_{16} \approx \frac{w}{\sqrt{2}}$ as a consequence of the Gaussian beam interaction. For a parametric oscillator spot size of 0.0015 m and $P_i P_s = .45 \times 10^{10} \ W^2$, the expected power at 16 µm is $P_{16} = 0.38 \times 10^3 \ W$. This is in very good agreement with the results in graph 4c bearing in mind the assumptions made for the spot sizes of the parametric oscillator and the 16 µm radiation.

Fig. 5 shows the spectrum of the 16 µm radiation as taken with an Edinburgh Instruments 121S spectrometer(grating blazed at 16 µm) with a resolution of 0.4 cm^{-1}. The spectrum of the 16 µm radiation was found to be 2.6 cm^{-1} and it is much broader than that of the generating wavelengths. The spectra of the signal (~ 0.83 cm^{-1}) and idler (~ 1 cm^{-1}) were taken using the eighth and sixth order of their respective wavelengths, for which the resolution of the spectrometer was 0.7 cm^{-1}. Thus only the envelopes of the two spectra could be resolved. The idler bandwidth appears to be in good agreement with that of the pump (~ 1 cm^{-1}) as expected. The signal spectrum is slightly narrower than that reported in [8] but this may be due to the fact that a return pump parametric oscillator was used and the signal experienced gain in the backward direction as well. Finally the spectrum of the mixed wavelength (2.6 cm^{-1}) appears to be close to the convolution of the other two. The peak of the spectrum could be set with a reliability better than ± 0.1 cm^{-1}. Although the system is capable of being tuned from 10 µm to over 20 µm we only operated it in the 15-17 µm

region which was our area of interest.

CONCLUSIONS AND DISCUSSION

A reliable, tunable source of 16 μm radiation using parametric processes in non-linear crystals has been described. Pulses with energies of 5 μJ and bandwidth of 2.6 cm^{-1} with an estimated power of $\frac{1}{2}$ kW have been produced at repetition rates of up to 20 pps. Controlled focussing of the pump radiation was the most important factor in the reproducibility of the system and expressions for the focussing of the Gaussian pump beam have been derived. The pulse widths, the energies and spectra of the interacting modes of radiation have been measured. The experimental results are in close agreement with theoretical predictions for the growth of the power of the difference wavelength. The main advantage of this method of production of 16 μm compared to other methods is the short duration of the pulses ∼10 ns.

In the present experiments only half the available power of the signal and idler were effective in the mixing process owing to the coincidence of their polarization ($LiNbO_3$ is type I phase-matchable). The power of the 16 μm radiation can be quadrupled by rotating the plane of polarization of one of the beams by 90°, so that phase-matching using the total powers of the input beams would be feasible, as described by eq. (3). This is presently being attempted in our laboratory. The main difficulty in achieving this lies in the closeness of the two wavelengths.

The spectrum of the parametric oscillator signal has been shown to narrow by a big factor with the introduction of thin, high finesse etalons [8], to the order of less than 0.1 cm^{-1}. Attempts are also being made to produce a reliable single longitudinal mode Nd:YAG laser. Already reliable operation of Nd:YAG with 3 to 4 longitudinal modes is feasible and bandwidths of 0.02cm^{-1} can be obtained. 16 μm tunable radiation with a bandwidth of less than 0.1cm^{-1} should then be possible.

It is important to note that until now we have used peak powers of less than 10 MW/cm^2 incident on the CdSe crystal. These are considerably lower than the reported damage threshold of >50 MW/cm^2 [5]. It appears from the above discussion that by rotating one of the polarizations of the input beams to its correct orientation and pumping the parametric oscillator harder, energies of between 50 - 100 μJ with a bandwidth of less than 0.1 cm^{-1} and pulse width of ∼10 ns could be achieved. The use of unstable Nd:YAG oscillators should further increase the efficiency.

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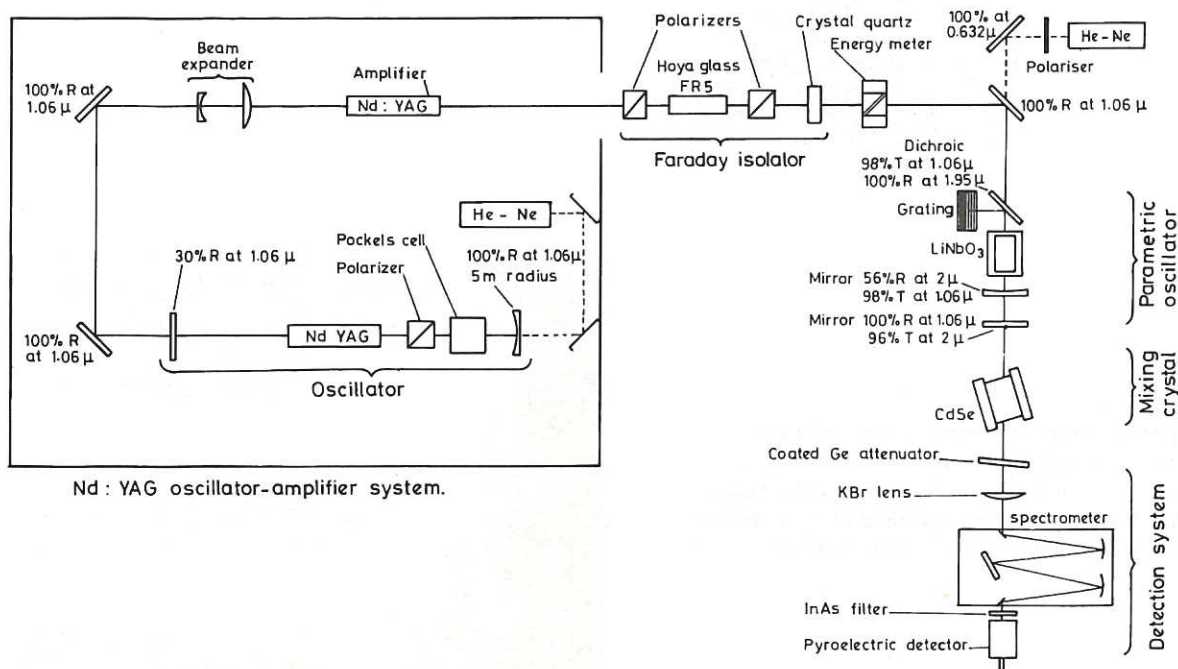
Fig.1 Experimental arrangement.

(a) Ten superimposed pulses in time, obtained with a fast photodiode having a rise time of 0.4 ns. Time scale 10 ns/div.
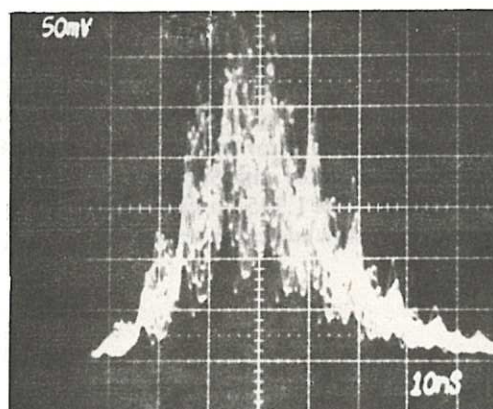
(b) Ten superimposed pulses showing the TEM$_{00}$ mode with a scanning diode array placed at 110 cm from the output mirror of the oscillator. Time scale 20 μs/div. The space correspondence is 0.066 mm/μs showing a beam waist of 0.88 mm at the 1/e^2 point. This corresponds to 0.77 mm at the exit mirror of the Nd:YAG oscillator.
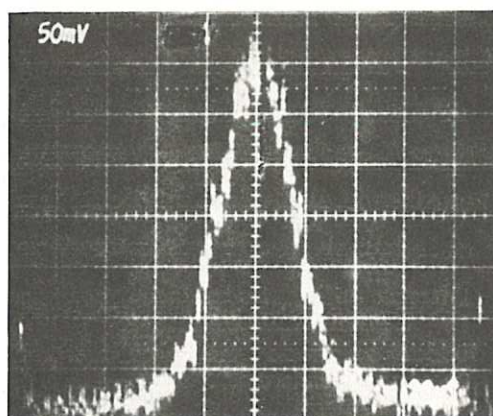
(c) Beam burn patterns from the Nd:YAG oscillator just before entering the beam expander.
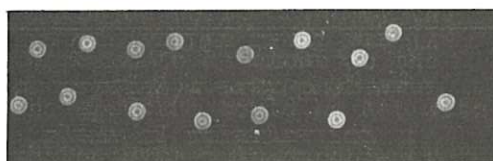
Fig.2 Nd:YAG oscillator characteristics.

Fig.3

(a) The top trace shows 10 superimposed pulses of the
1.064 μm radiation, and the bottom of the idler (2.28 μm),
simultaneously recorded using a dual beam 7844 Tektronix
oscilloscope. A silicon detector was used at $\lambda = 1.064\,\mu$m
and an InAs detector with a Ge filter was used at $\lambda = 2.28\,\mu$m.
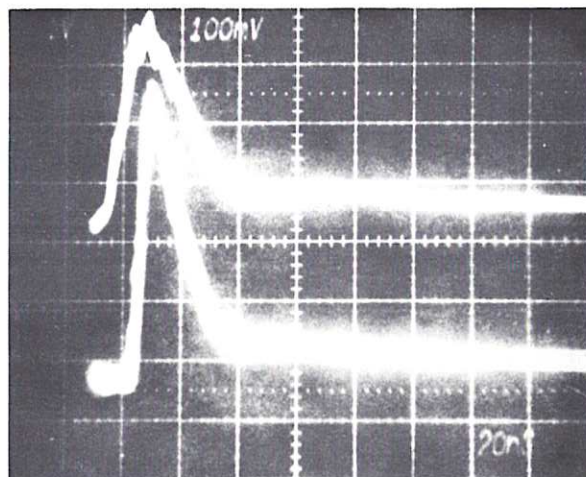
(b) The top trace shows 10 superimposed pulses of the
1.064 μm radiation, and the bottom of the signal (1.995 μm),
simultaneously recorded using a dual beam 7844 Tektronix
oscilloscope. A silicon detector was used for the 1.064 μm
radiation and a Ge detector with Si filter was used for the
1.995 μm radiation. The 1.995 μm signal duration is
determined by the (slow) detector response.
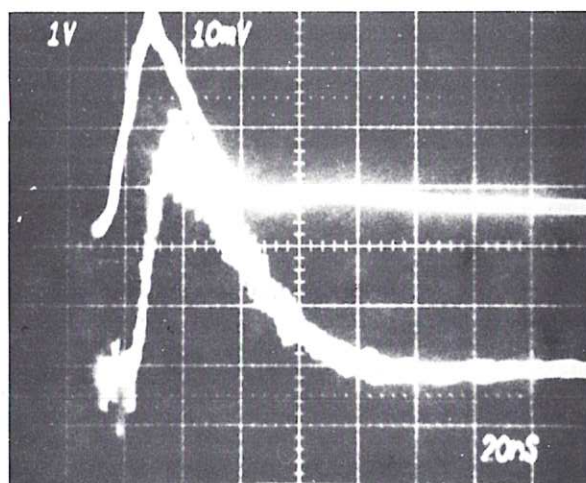
(c) Ten superimposed pulses of the 16 μm radiation recorded
using an RKp 335 Laser Precision pyroelectric detector.

CLM-P 496
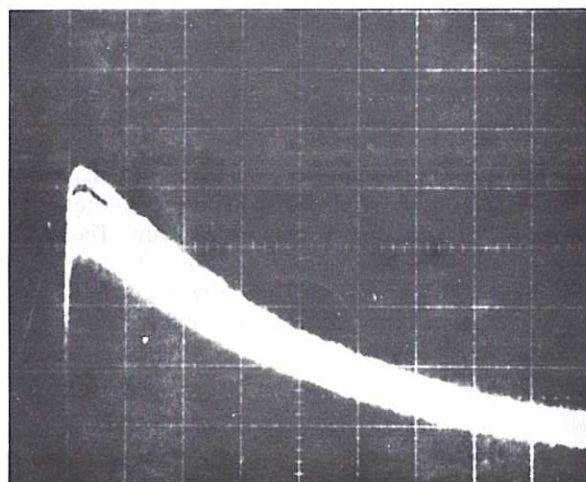
Fig.4

(a) The percentage energy conversion efficiency of the parametric oscillator vs. input (normalized to threshold energy).
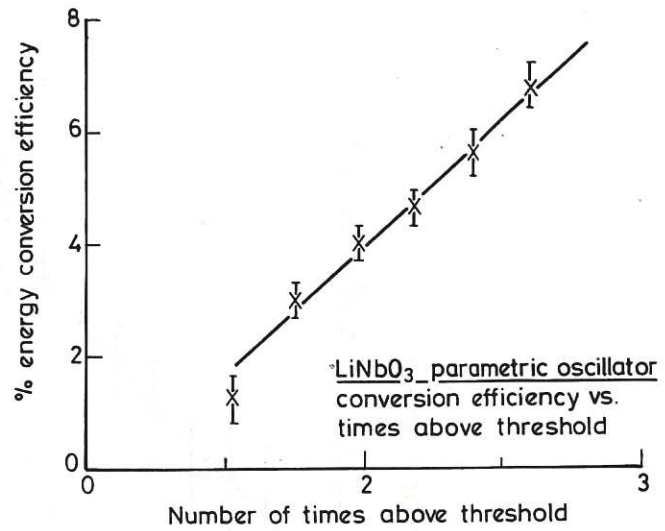
(b) The output energy at $16\,\mu m$ as a function of the input energy (signal + idler) in the CdSe crystal.
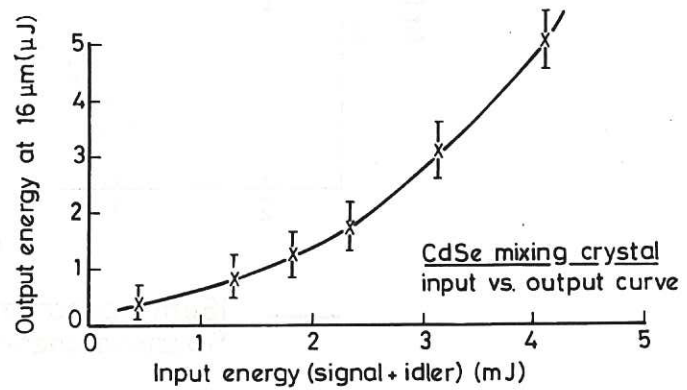
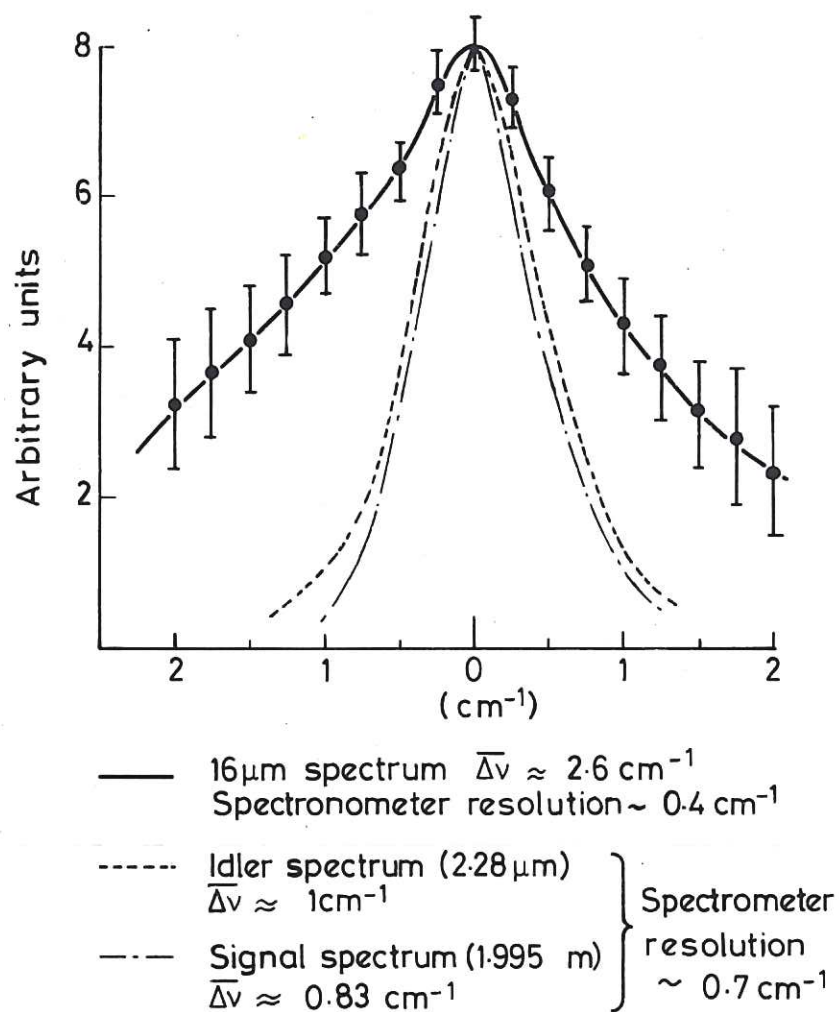(c) The output power at $16\,\mu m$ as a function of the product of the powers of the generating beams.

LiNbO$_3$ parametric oscillator conversion efficiency vs. times above threshold

CdSe mixing crystal input vs. output curve

16μm radiation, 16 μm power vs product of generating powers

$P'_s P'_i / 4 = P_s P_i \times 10^{10}\ W^2$
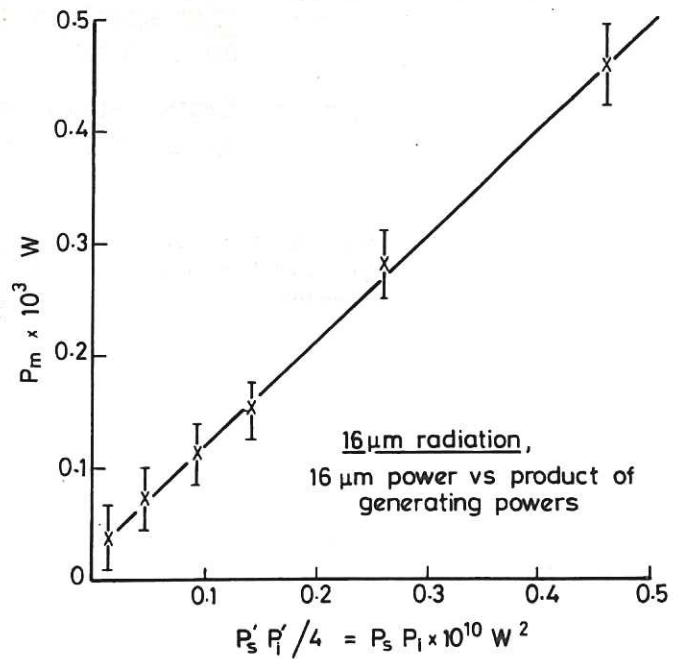
Fig.5 The spectrum of the 16 μm radiation and the envelopes of the spectra of the two generating beams (signal and idler of the parametric oscillator).