GENERATION OF TUNABLE MEDIUM INFRARED RADIATION BY OPTICAL MIXING IN PROUSTITE

D. C. HANNA, R. C. SMITH and C. R. STANLEY
Department of Electronics, University of Southampton, Southampton SO9 5NH, UK

Received 10 November 1971

The outputs of a ruby laser and a tunable dye laser (pumped by the ruby laser) have been frequency-mixed in a proustite crystal. Difference wavelengths have been generated around 5 and 11 μm; tuning being achieved for the latter covering the range 10.1 to 12.7 μm. Extrapolations from these preliminary results indicate that currently available lasers and nonlinear crystals should allow continuous tuning from 2.5 to 12.5 μm, with bandwidths substantially smaller than 0.1 cm^{-1} and at pulsed powers of the order of a kilowatt.

Recently considerable advances have been made in the operation of laser devices producing tunable medium infrared (IR) radiation. Of these the spin-flip Raman laser [1] and the various semiconductor laser diodes [2] suffer from restricted tuning ranges in practical devices. The proustite parametric oscillator [3, 4] has in principle a much wider tuning range although at the time of writing, tuning of this device has only been attempted in the near IR (from 1.82 to 2.56 μm) [4]. All these devices are relatively complex. Infrared generation by optical mixing is much simpler and, in addition, offers a very wide tuning range.

Martin and Thomas [5] first demonstrated medium IR mixing using an Nd laser and its Raman shifted output. In this way they produced a few fixed wavelengths around 10 μm by non-phase-matched mixing in CdS and CdSe. Extension of this work to produce higher powers and tunability of the medium IR had to await two advances. These were the availability of phase-matchable nonlinear crystals transparent from the visible to medium IR and the development of sources of high power tunable visible radiation for one of the mixing fields. Dewey and Hocker [6] have recently demonstrated the fruitful marriage of these two advances, by mixing a ruby laser and a tunable dye laser (pumped by the same ruby laser) in a crystal of $LiNbO_3$. The longest wavelength generated, 3.5 μm, was limited by IR absorption in the $LiNbO_3$. In this letter we report some preliminary experiments which extend the dye laser mixing technique further into the medium IR by using proustite as the mixing material. The

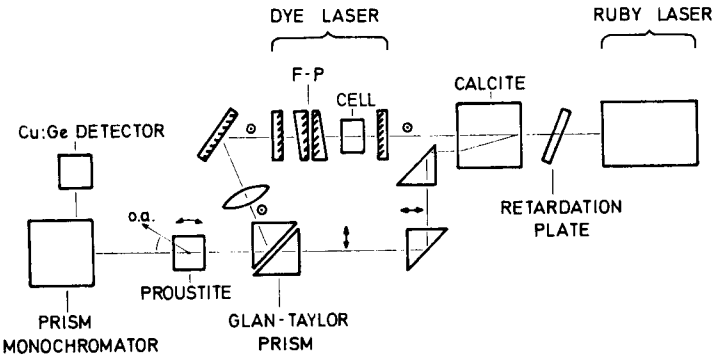

Fig. 1. Experimental arrangement.

attractions of proustite are its large non-linearity, its transmission from ≈ 0.6 to ≈ 13 μm and the fact that phase-matching is possible for any three-wave collinear process within its transmission band [7]. The material has a rather low damage threshold [8], particularly for ruby laser radiation (a few MW/cm^2) as the wavelength is close to the band edge. However, with care, damage can be avoided and the results of our preliminary experiments indicate that medium IR powers of the order of 1 kW are feasible with present-day laser and crystal technology.

The experimental arrangement is shown in fig. 1. The 75 mm × 6 mm ruby was Q-switched by a solution of vanadium phthalocyanine in toluene and oscillated in the TEM_{00} mode with a peak output power of 290 kW and a pulse duration (fwhm) of 12 nsec (see fig. 2b). The linearly-polarised ruby laser beam was split into two beams with orthogonal polarisations by means of a crystal quartz retardation plate followed by double refraction walk-off in a block of calcite. Tilting the retardation plate was a convenient way of adjusting the relative powers in the two beams without changing their parallelism or overlap at the mixing crystal. The longitudinally pumped dye laser [9] consisted of a 10 mm cell of 10^{-4} M cryptocyanine in glycerol [10] placed between plane mirrors 80 mm apart. The resonator also contained a plane Fabry-Pérot (FP) interferometer (free spectral range ≈ 250 cm^{-1} and finesse ≈ 20) for spectral narrowing and wavelength tuning by varying its angle [11]. The dye laser output (of the same polarisation as the ruby beam pumping it) was recombined collinearly with the other (orthogonally polarised) ruby beam by means of a Glan-Taylor calcite prism. A lens inserted in the dye beam provided approximate matching of spot-size (≈ 2 mm diameter) at the proustite crystal. Type I phase-matching has been used (ruby beam extraordinary, dye and IR ordinary). The crystal was 5 mm long and cut with its optic axis at $22^{\circ} \pm \frac{1}{2}^{\circ}$ to the face normal.

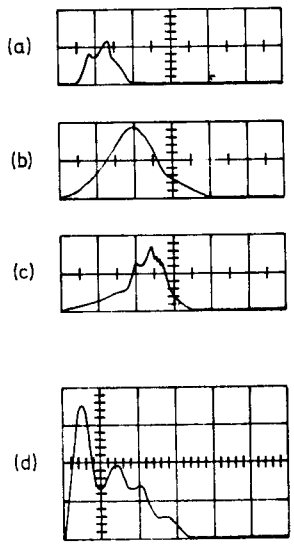

Fig. 2. (a) Dye laser output ($\approx$740 nm). (b) Ruby laser output. (c) Time-overlap of dye and ruby laser outputs. (d) Difference-frequency signal. Time bases: (a) - (c) 10 nsec/div; (d) 50 nsec/div.

The infrared radiation was monitored by a Cu-doped Ge detector (response time ≈ 50 nsec) after passing through a prism monochromator. The dye and ruby laser outputs (figs. 2a and 2b, respectively) were monitored by a planar vacuum photodiode (overall response time < 1 nsec). The good time-overlap of ruby and dye pulses is evident from fig. 2c.

We have observed, as reported by Stepanov et al. [12], that the cryptocyanine dye laser can oscillate simultaneously in two bands centred on 740 and 810 nm. The finesse of the FP was much reduced at the longer wavelength and tuning in this wavelength range was not attempted.

By varying the angle of the FP, tuning from 734 to 746 nm was achieved, this being limited by the free spectral range of the FP. When the powers at the crystal were 45 kW at 694 nm and 500 W at 743 nm the measured IR power was 100 mW, the wavelength being 10.6 μm. The IR wavelengths were tuned from 10.1 to 12.7 μm. The measured phase-matching angles agree well with those calculated from Hobden's [13] refractive index data. The bandwidth for phase-matching was calculated to be sufficiently wide to accommodate the frequency spread of the dye laser.

Mixing 250 W at ≈ 810 nm with 90 kW at 694 nm in the same crystal produced ≈ 6 W of IR power. In this case the monochromator was not used to measure the IR wavelength (expected to be 4.9 μm) but transmission of the radiation by filters indicated that it lay between 4 and 6 μm.

The difference power P_2 generated at frequency $\omega_2 = \omega_3 - \omega_1$ is given (in MKSA units)

$$P_2 = \frac{2^4\omega_2^2 d^2 \times 9 \times 10^9 P_3 P_1 l^2 T_1 T_2 T_3}{n_1 n_2 n_3 c^3 \ (W_1^2 + W_3^2)} \times \exp[-\tfrac{1}{2}\alpha_2 l] \ . \quad (1)$$

In deriving this expression, double refraction and diffraction effects have been ignored and depletion of P_3 assumed negligible. n_1, n_2 and n_3 are refractive indices at frequencies ω_1, ω_2 and ω_3 respectively; α_2 is the power absorption coefficient of the difference radiation; W_1 and W_3 are the spot sizes of the incident gaussian beams; l is the crystal length; and d is the effective nonlinear coefficient. T_3 and T_1 are the transmissions of the entrance face at frequencies ω_3 and ω_1, and T_2 is the transmission of the exit face at frequency ω_2. All the powers P_1, P_2, P_3 are external to the crystal.

Our crystal was not coated, hence $T_1 = T_2 = T_3 \approx 0.78$ due to Fresnel reflection. The crystal had been cut in such a way that the contributions of d_{31} and d_{22} were additive [7] thus giving an effective d coefficient of $\approx 3 \times 10^{-11}$m/V over the range of phase-matching angles used. With $W_1 = W_2 = 1$ mm, $P_3 = 90$ kW and $P_1 = 250$ W, eq. (1) predicts a power of 4.5 W at 4.9 μm (where α_2 can be neglected). For 10.6 μm generation with $P_3 = 45$ kW, $P_1 = 500$ W and $\alpha_2 = 1$ cm^{-1} the predicted IR power is 750 mW. The observed power at 4.9 μm was ≈ 6 W and at 10.6 μm was ≈ 100 mW. The transverse mode structure of the dye laser at 740 nm was noticeably less pure than at 810 nm probably contributed to the reduction in measured 10.6 μm power compared to that calculated.

Since the measured 4.9 μm power is in good agreement with that calculated, it is interesting to make predictions of the IR power which could be generated using the larger crystals and more powerful lasers available.

With large values of P_3 and P_1 (i.e., ≈ 1 MW) and for damage prone materials such as proustite and silver thiogallate [14] the spot sizes are determined by the need to keep power densities below damage threshold rather than by considerations of optimum focussing [15]. The values of W_1 and W_2 are then large enough for eq. (1) to be valid, i.e., diffraction and walk-off effects may be ignored. It is worth noting that the material may have significantly different damage thresholds for the two mixing wavelengths when these are close to the band edge.

We consider two examples. First a ruby-pumped dye laser of 0.5 MW power, bandwidth 0.02 cm^{-1}, and a ruby laser of 0.5 MW power, bandwidth $\ll 0.02$ cm^{-1}, both operating in the TEM_{00} mode. The possibility of achieving such performance figures has already been demonstrated [16, 17]. In addition Miyazoe and Maeda [18] have shown that a range of ruby pumped dye lasers is available which allows tuning of the IR wavelength from 2.5 μm up to the long wavelength transmission limit of proustite (≈ 13 μm). In a 10 mm cube of proustite, with spot sizes $W_1 = W_2 = 5$ mm and hence with beam centre density below the measured damage threshold (3 MW/cm^2) at 694 μm [8], the above laser powers would give tunable IR power ranging from 5.5 kW at 2.5 μm to 150 W at 12.5 μm with a bandwidth of ≈ 0.02 cm^{-1}.

The second example considers two rhodamine 6G dye lasers each having TEM_{00} powers of 50 kW, mixed in silver thiogallate [13]. We assume a cube of only 3 mm side for this material since this is the maximum size currently available. The measured damage threshold is > 6 MW/cm^2 for wavelengths longer than 590 nm [19]. Thus using the two rhodamine 6G lasers, one at the fixed wavelength of 590 nm and the other tunable from 619 nm to 633 nm, phase-matched IR difference mixing is possible from 8.7 μm to 12.6 μm. With spot sizes $W_1 = W_2 \approx 1$ mm (hence power density ≈ 6 MW/cm^2) and $\alpha_2 \approx 1$ cm^{-1}, the IR power would range from ≈ 300 W at 8.7 μm to ≈ 100 W at 12.6 μm. Dye laser bandwidths of the order of 0.02 cm^{-1} have been achieved (see [20], for example) allowing the possibility of an IR bandwidth of 0.04 cm^{-1}. As yet rhodamine 6G lasers having simultaneously the performance figures of 50 kW TEM_{00} power and 0.02 cm^{-1} bandwidth have not been reported but the rapid progress of dye lasers [21] gives good expectation of their achievement in the very near future. The good chemical stability of the rhodamines compared to the cyanine dyes and the higher repetition rate operation possible with rhodamine lasers compared to ruby lasers probably make this the preferred scheme for tunable IR generation through difference mixing.

In conclusion we have reported the first demonstration of tunable difference mixing for wavelengths well into the medium IR. Proposals are made for generation of up to one kilowatt of peak power and for continuous tuning over the whole range 2.5 to 12.5 μm. The comparative simplicity of this approach suggests applications in fields such as atmospheric pollution measurement.

We wish to thank Mr. G. C. Bhar for the various phase-matching computations required. The proustite crystal was obtained from the Electronic Materials Unit of the Royal Radar Establishment. This work was partially supported by a grant from the Science Research Council.

REFERENCES

[1] C. K. N. Patel and E. D. Shaw, Phys. Rev. B3 (1971) 1279.
[2] K. W. Nill, F. A. Blum, A. R. Calawa and T. C. Harman, Appl. Phys. Letters 19 (1971) 79.
[3] E. O. Amman and J. M. Yarborough, Appl. Phys. Letters 17 (1970) 233.
[4] D. C. Hanna, B. Luther-Davies, H. N. Rutt and R. C. Smith, Appl. Phys. Letters, to be published.
[5] M. D. Martin and E. L. Thomas, IEEE J. Quantum Electron. QE-2 (1966) 196.
[6] C. F. Dewey and L. O. Hocker, Appl. Phys. Letters 18 (1971) 58.
[7] K. F. Hulme, O. Jones, P. H. Davies and M. V. Hobden, Appl. Phys. Letters 10 (1967) 133.
[8] D. C. Hanna, B. Luther-Davies, H. N. Rutt, R. C. Smith and C. R. Stanley, IEEE J. Quantum Electron., to be published.
[9] P. P. Sorokin, W. H. Culver, E. C. Hammond and J. R. Lankard, IBM J.Res. Develop. 10 (1966) 401.
[10] M. L. Spaeth and D. P. Bortfeld, Appl. Phys. Letters 9 (1966) 179.
[11] D. J. Bradley, G. M. Gale, M. Moore and P. D. Smith, Phys. Letters 26A (1968) 378.
[12] B. I. Stepanov, A. N. Rubinov and V. A. Mostovnikov, JETP Letters 5 (1967) 117.
[13] M. V. Hobden, Opto-Electron. 1 (1969) 159.
[14] D. S. Chemla, P. J. Kupecek, D. S. Robertson and R. C. Smith, Opt. Commun. 3 (1971) 29.
[15] G. D. Boyd and D. A. Kleinman, J. Appl. Phys. 39 (1968) 3597.
[16] D. J. Bradley, A. J. F. Durrant, G. M. Gale, M. Moore and P. D. Smith, IEEE J. Quantum Electron. QE-4 (1968) 707.
[17] V. Daneu, C. A. Sacchi and O. Svelto, IEEE J. Quantum Electron. QE-2 (1966) 290.
[18] Y. Miyazoe and M. Maeda, Appl. Phys. Letters 12 (1968) 206.
[19] V. V. Rampal, private communication.
[20] H. Walther and J. L. Hall, Appl. Phys. Letters 17 (1970) 239.
[21] I. Itzkan and F. W. Cunningham, Paper 1.7 - Conference of laser engineering and applications, Washington D. C. (1971).