Singly resonant proustite parametric oscillator tuned from 1.22 to 8.5 μm

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A singly resonant parametric oscillator based on proustite has been operated for the first time. The device was pumped by a Q-switched neodymium (1.065 μm) laser in a noncollinear phase-matching geometry. The output has been tuned over the wavelength range 1.22—8.5 μm. The idler power was typically ~100 W in a bandwidth of ~1 cm^{-1}.

Parametric oscillation with proustite has previously been observed[1,2] only in the doubly resonant oscillator configuration (DRO), i.e., with both the signal and idler waves resonant. Such a configuration generally leads to unacceptably large variations of output frequency.[3] A singly resonant oscillator configuration (SRO), in which one wave only is resonant, avoids this problem, and the well-behaved spectral characteristics of a SRO have been amply demonstrated.[4,5] We report here the first operation of a proustite SRO and its tuning over the range 1.22—8.5 μm. This tuning range encompasses a region of the infrared which cannot be covered by other infrared parametric oscillators based on $LiNbO_3$,[6] $LiIO_3$,[7] or CdSe.[8,9] Typical performance figures at a wavelength of 4.5 μm for the nonresonant idler are ~100 W idler power and a bandwidth of ~1 cm^{-1}. The above performance makes the proustite SRO strongly competitive with the pulsed spin-flip Raman laser[10] for a variety of applications.

A schematic diagram of the oscillator is shown in Fig. 1. The use of a noncollinear phase-matching scheme[11] eliminates the possibility of any feedback of the nonresonant idler within the oscillator. It also isolates the laser from the oscillator since pump reflections are not returned to the laser. Tuning has been achieved by rotating the crystal, keeping the oscillator mirrors fixed. The advantages of this scheme have been pointed out by Basu and Steier.[12] The choice of various parameters such as pump and signal confocal parameters, crystal length, mirror curvatures, and separation are a compromise between a number of conflicting requirements. The major factors which have determined the choice have been (i) the necessity to keep beam intensities below the proustite damage threshold,[13] (ii) the need for a short resonator to allow a large number of round trips during the short pump pulse (25 nsec FWHM), and (iii) the requirement for a long enough crystal to give sufficient gain to overcome the various losses.

Calculations of pump threshold have been made by taking into account (a) the finite pump pulse duration and (b) the incomplete overlap of beams due to both double refraction and noncollinearity. These calculations have been based on the work of Pearson *et al.*[14] and Basu and Steier,[12] respectively, and will be reported in more detail in the future. Since all threshold calculations necessarily make use of a number of quantities which are not accurately known, such as crystal losses and the effective crystal length as determined by inhomogeneities, there is clearly room for further optimization of the oscillator design parameters.

Three different mirror combinations have been used to span the tuning range of the signal (the higher of the two generated frequencies). Each was a standard $\lambda/4$ multilayer stack appropriately pitched. For idler wavelengths beyond 3.5 μm, the output mirror used a CaF_2 substrate. The concave mirror had either $\frac{1}{2}$- or 1-m radius of curvature, a reflectivity of > 99% over the appropriate signal range, and > 80% transmission for the pump. The plane output mirrors had reflectivities of > 96% for the signal and transmissions of > 90% for the idler. Work is in progress to produce a single stagger-stack coating to replace the three coatings used here.

The proustite crystal was a 10-mm cube cut for type I phase-matching with the optic axis at 28° to the face normal. The crystal orientation was such that the contributions from d_{15} and d_{22} were additive. As with $LiNbO_3$,[15] use of the correct quadrant is most important. For proustite the threshold for a crystal in the additive quadrant is 3—4 times smaller than for the subtractive quadrant. In fact the crystal used in the DRO reported in Ref. 2 has since been found to have been cut wrongly and the new crystal with correct orientation has been vital to the achievement of singly resonant oscillation.

Three-layer antireflection[16] coatings on the crystal provided low reflectivity for wavelengths from 1.0 to 4.0 μm. The transmission losses of the coated crystal were measured in a spectrophotometer to be less than 5% over the wavelength range from 1.1 to 3 μm. This method of measurement does not, however, allow any estimate to be made for the small-angle scatter loss.

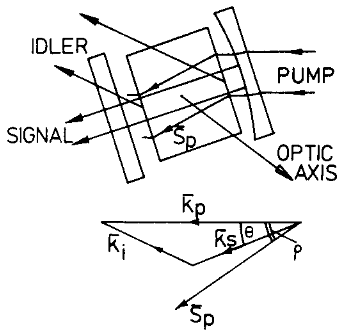

FIG. 1. Schematic diagram of noncollinear singly resonant oscillator configuration. For clarity the angles have been exaggerated; $\theta = 2$ mrad, $\rho = 78$ mrad. $\bar{S}_p$ is the Poynting vector of the pump.

The laser used was a flash-lamp-pumped Nd:$CaWO_4$ laser with a KD*P Pockels cell Q switch. The laser was operated in the TEM_{00} mode with the mirror curvatures and separation chosen to give the required spot size ($W = 0.6$ mm) at the parametric oscillator without the need for intervening mode-matching lenses. The output of the laser could be constrained to a single longitudinal mode by a method of switching the Pockels cell that has been previously reported.[17,18] Alternatively, by operating the Pockels cell as a conventional fast Q switch, the laser bandwidth of ~0.3 cm^{-1} would contain more than 30 longitudinal modes. For the DRO it was necessary always to operate the laser in a single longitudinal mode. For the SRO it has been found, as expected, that the threshold pump power is the same for single-mode or multimode pump.

For degenerate oscillation ($\lambda_s = \lambda_i = 2.13$ μm) the calculated steady-state threshold pump intensity (power/$\frac{1}{2}\pi W_p^2$, where W_p is the pump spot radius) for our oscillator is 1.2 MW/cm^2. This calculation makes use of the d coefficients quoted by Hulme *et al.*,[19] together with an estimated round-trip signal loss of 20% and a round-trip time of 0.25 nsec, corresponding to the 2-cm mirror separation actually used. By taking into account the 25-nsec duration of the pulse, the calculated peak pump intensity required to reach threshold (defined as the condition where output powers reach detectable levels) was 3.6 MW/cm^2. The observed near-degenerate threshold was ~5 MW/cm^2. Since the damage threshold of proustite under similar conditions of irradiation has been found[13] to be 20 MW/cm^2, this leaves a factor of 4 between the dynamic threshold of oscillation and pump-induced damage threshold.

The phase-matching angle for degenerate operation was found to be within the cutting tolerance ($\pm\frac{1}{2}°$). Signal wavelengths were measured using a 1-m grating monochromator (resolution 0.5 cm^{-1}) to an accuracy of a few cm^{-1} at a number of signal wavelengths between 1.42 and 1.26 μm (corresponding to an idler wavelength range of 4.2–6.8 μm). Idler wavelengths were also measured directly around 4 μm. The observed phase-matching angles for all these wavelengths were in excellent agreement with calculated values based on Hobden's refractive-index data.[20] The small degree of noncollinearity (< 2-mrad internal angle between pump and signal wave vectors) has a negligible effect on the computed phase-matching angle. Oscillation has been observed up to a phase-matching angle of 20°, which corresponds to signal and idler wavelengths of 1.22 and 8.5 μm, respectively. The entire tuning range is covered by a crystal rotation of 27°. In agreement with Basu and Steier's predictions,[12] a considerable portion of this tuning range was covered without any realignment of the oscillator cavity.

The signal and idler outputs were monitored with a number of different detectors: a room-temperature InSb detector, a cooled $Hg_xCd_{1-x}Te$ detector, and a TGS pyroelectric detector. Pump radiation was removed by a Ge plate. A measurement of idler output power made at 4.5 μm with a calibrated detector gave a value of 100 W peak. At the same wavelength the measured bandwidth between 10-dB power points was < 3 cm^{-1}. A calculation of the bandwidth[5] for the appropriate conditions gave a predicted value of ~1 cm^{-1}. It was found that over about 1 h the center frequency drifted by 2–3 cm^{-1}, an amount consistent with mechanical instabilities in the apparatus.

The pump-induced damage threshold was found to be identical to our earlier value for proustite.[13] It is possible to avoid this damage by careful control of intensity. There was some evidence of signal-induced damage at the highest output powers. Useful output powers could, however, be obtained without having to change from a resonant signal to a resonant idler configuration.[9]

In conclusion, we have demonstrated singly resonant parametric oscillation in proustite tuning over a substantial portion of the infrared. Much of this range cannot be covered by other parametric oscillators. An output power of ~100 W in a bandwidth of ~1 cm^{-1} at 4.5 μm is a typical performance figure obtained to date. However, further optimization of a number of parameters is possible which should extend the range of 1.2–9.5 μm, together with a substantial increase in power. The present bandwidths may be considerably reduced by frequency selectors which restrict oscillation to a single mode.[5,21] By down-conversion[22] of the output of our oscillator, it is now possible to generate tunable coherent radiation over the whole range 1.2–25 μm. The large tuning range, high power, and narrow bandwidth should lead to wide use in infrared spectroscopic applications.

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[1] E.O. Ammann and J.M. Yarborough, Appl. Phys. Lett. 17, 233 (1970).

[2] D.C. Hanna, B. Luther-Davies, H.N. Rutt, and R.C. Smith, Appl. Phys. Lett. 20, 34 (1972).

[3] J.A. Giordmaine and R.C. Miller, in *Physics of Quantum Electronics*, edited by P.L. Kelley, B. Lax, and P.E. Tannenwald (McGraw-Hill, New York, 1966), pp. 31–42.

[4] J.E. Bjorkholm, Appl. Phys. Lett. 13, 399 (1968).

[5] L.B. Kreuzer, Appl. Phys. Lett. 15, 263 (1969).

[6] R.W. Wallace, Appl. Phys. Lett. 17, 497 (1970).

[7] L.S. Goldberg, Appl. Phys. Lett. 17, 489 (1970).

[8] A.A. Davydov, L.A. Kulevskii, A.M. Prokhorov, A.D. Savel'ev, and V.V. Smirnov, JETP Lett. 15, 513 (1972).

[9] R.L. Herbst and R.L. Byer, Appl. Phys. Lett. 21, 189 (1972).

[10] R.B. Dennis, C.R. Pidgeon, S.D. Smith, B.S. Wherrett, and R.A. Wood, Proc. Roy. Soc. Lond. A 331, 203 (1972).

[11] J. Falk and J.E. Murray, Appl. Phys. Lett. 14, 245 (1969).

[12] R. Basu and W.H. Steier, IEEE J. Quantum Electron. 8, 693 (1972).

[13] D.C. Hanna, B. Luther-Davies, H.N. Rutt, R.C. Smith, and C.R. Stanley, IEEE J. Quantum Electron. 8, 317 (1972).

[14] J.E. Pearson, U. Ganiel, and A. Yariv, IEEE J. Quantum Electron. 8, 433 (1972).

[15] J.E. Bjorkholm, Appl. Phys. Lett. 13, 36 (1968).

[16] R. Henderson and H. N. Rutt (unpublished).

[17] D. C. Hanna, B. Luther-Davies, and R. C. Smith, Electron. Lett. 8, 369 (1972).

[18] D. C. Hanna, B. Luther-Davies, and R. C. Smith, Opto-Electron. 4, 249 (1972).

[19] K. F. Hulme, O. Jones, P. H. Davies, and M. V. Hobden, Appl. Phys. Lett. 10, 133 (1967). We have taken d_{31} for $LiNbO_3$ to be 1.5×10^{-8} esu.

[20] M. V. Hobden, Opto-Electron. 1, 159 (1969).

[21] J. Pinard and J. F. Young, Opt. Commun. 4, 425 (1972).

[22] G. C. Bhar, D. C. Hanna, B. Luther-Davies, and R. C. Smith, Opt. Commun. 6, 323 (1972).