TUNABLE INFRARED DOWN-CONVERSION IN SILVER THIOGALLATE

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A ruby laser beam mixed in $AgGaS_2$ with the output of various ruby-pumped dye lasers has provided down-converted infrared radiation tuned from 4.6 to 12 μm. The measured phase-matching angles agree well with calculated values. Peak infrared powers of a few hundred milliwatts have been produced.

Chemla et al. [1] have drawn attention to the potential of the ternary chalcopyrite materials for infrared nonlinear optical devices. As an illustration of this, they demonstrated phase-matched second harmonic generation from 10.6 μm radiation in a crystal of silver thiogallate ($AgGaS_2$). This particular ternary has the attractive feature of a transmission range extending from 0.5 to 13 μm, thus allowing the possibility of infrared down-conversion tunable out to $\approx$ 13 μm by mixing the outputs of tunable dye lasers. A similar down-conversion experiment has previously been reported by us in proustite (Ag_3AsS_3) using a ruby laser and a ruby-pumped dye laser [2]. However, $AgGaS_2$ has advantages over proustite for this application. It has a wider band gap than proustite and is less liable to damage by visible laser radiation. In this letter we report the first tunable infrared generation by down-conversion in $AgGas_2$. Using the output of a ruby laser mixed with the outputs of ruby-pumped tunable dye lasers we have generated infrared radiation between 4.6 and 12 μm with peak powers of hundreds of milliwatts. The measured phase-matching angles are in excellent agreement with calculated values based on Sellmeier equations for the refractive indices [3].

In a very recent note by Bethea [4] it is briefly reported that fixed wavelength down-conversion in $AgGaS_2$ has been achieved at 5.52 μm by mixing two wavelengths emitted simultaneously by an Nd:YAG laser. No indication of the infrared power is given. His measured phase-matching angle agreed with calculations based on the data of Boyd et al. [5].

The Q-switched ruby laser used in our work gave a peak output power of 1 MW (TEM_{00} mode) with a pulse duration of 10 nsec (fwhm). Part of this power was separated by a beam splitter and was used to pump the dye laser. Quasi-longitudinal pumping was used, the ruby laser beam entering the dye cell nearly normal to the cell face. This same uncoated cell face served as the output reflector of the dye laser, the other reflector being a high reflectivity flat mirror. Frequency control of the dye laser was effected by means of a Fabry–Pérot etalon placed between the cell and the high reflectivity mirror. The etalon had a free spectral range of 70 cm^{-1} and a finesse of $\approx$ 10. Four dyes were used; DTTC in DMSO [6] for down-conversion in the region around 5 μm; 1,3,3,1′,3′,3′-hexamethyl-2,2′-indotricarbocyanine iodide (dye 17 of ref. [7]) for down-conversion around 7 μm; DDI in glycerol [8] for the 8.5 μm region; and cryptocyanine in glycerol for the 11.5 μm region.

The two laser beams were combined at a dichroic mirror, the dye beam polarised orthogonally to the ruby beam. Separate focussing of the ruby and dye laser beams ensured approximate equality of their spot sizes (diameter $\approx$ 2 mm) at the nonlinear crystal. Care was taken to ensure good overlap and colinearity of the two beams. Colinearity is most important as far as ease of detection is concerned, since in down-con-

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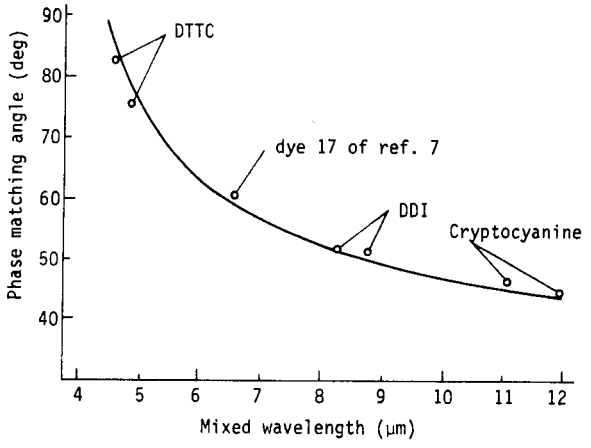

Fig. 1. Phase-matching angle versus infrared wavelength for difference mixing of ruby laser and ruby-pumped dye laser outputs in $AgGaS_2$. Calculated values are indicated by the solid curve and experimentally observed values by the circles.

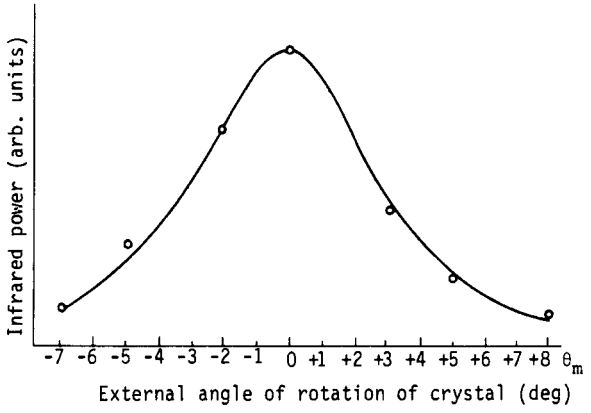

Fig. 2. Infrared power (arbitrary units) at 4.6 μm versus external angle of rotation of the 75° cut crystal (crystal B) with the dye laser wavelength kept fixed at 818 nm. The maximum power corresponds to a measured phase-matching angle of 82.7°.

version a small angle between the two high frequency beams gives rise to a large angular displacement of the infrared beam. This can make detection difficult. The infrared radiation was detected by either a cooled $Hg_xCd_{1-x}Te$ detector or an InSb detector. The ruby and dye laser beams were eliminated by a Ge filter in front of the detector.

Two $AgGaS_2$ crystals have been used, crystal A for wavelengths longer than ≈ 6 μm and crystal B for wavelengths shorter than ≈ 6 μm. Both crystals were correctly oriented for type I phase-matching (0 + 0 = E). Crystal A was in the form of a plate 1.7 mm thick with faces approximately 5 mm × 7 mm and cut so that the optic axis made an angle of 48° with the face normal. Crystal B was 1.5 mm thick with faces of ≈ 5 mm × 8 mm with the optic axis at 75° to the face normal. In both crystals the useful area was somewhat restricted by the presence of lamellar twins.

Fig. 1 shows the measured phase-matching angles, and the calculated angles (solid line) are shown for comparison. The Sellmeier equations used for the calculated angles have been obtained by incorporating new measurements [3] of the visible refractive indices of $AgGaS_2$ together with the infrared data of Boyd et al. [5]. It is seen that for both crystals the agreement between predicted and measured values is good.

Measurements of infrared power were made at two particular wavelengths. With 140 kW at 694 nm and 13 kW at 740 nm (cryptocyanine dye laser) incident on the crystal, the generated infrared power (at 11.1 μm) was measured to be ≈ 300 mW. The calculated power, using the recently remeasured [9] value of the nonlinear coefficient of $AgGaS_2$, is ≈ 3W. For the same ruby laser power and 10 kW at 808 nm (from the DTTC laser), the generated power at 4.9 μm was measured to be ≈ 200 mW. In this case the calculated power is ≈ 9W after taking into account increased crystal absorption in the region of 5 μm [3].

The small birefringence of $AgGaS_2$ means that for given mixing wavelengths the phase-matching condition is relatively insensitive to crystal orientation. Fig. 2 shows a plot of down-converted power (at 4.6 μm) versus crystal orientation with the dye laser wavelength fixed at 818 nm, a value leading to phase-matching near the non-critical condition. The measured width of the phase-matching curve agrees exactly with the calculated width and similar measurements throughout the entire tuning range have also given good agreement.

After several hundred pulses at the power levels used in this experiment (i.e., 140 kW of ruby power in a 10 nsec fwhm pulse with spot radius, $W = 1$ mm at the crystal) it was found that signs of surface damage began to be visible. This indicates a damage threshold of ≈ 10 MW/cm^2 under these conditions. A previous study of laser induced damage in $AgGaS_2$ has been made with 1.06 μm radiation [10].

In conclusion we have confirmed the values of phase-matching angles computed for down-conversion in $AgGaS_2$ and shown that a wide tuning range is possible. Measured infrared powers have been found to be one or two orders of magnitude smaller than those calculated. The reason for this discrepancy is thought to lie in multi-transverse mode behaviour of the dye lasers. Damage has not been found to be a serious limitation of $AgGaS_2$ for a down-conversion material and as larger crystals become available it should be possible to considerably increase the infrared power.

We are greatly indebted to Dr. G.C. Bhar for supplying the information on phase-matching angles and for many valuable discussions. Both crystals were grown at the Royal Radar Establishment where the orientation and polishing of one crystal was also carried out. The second crystal was oriented and polished at the Centre National d'Etudes des Télécommunications (C.N.E.T.) and constructive discussions with members of the nonlinear optics group at the C.N.E.T. are gratefully acknowledged. One of us (V.V. Rampal) wishes to thank the Association of Commonwealth Universities for the award of a Commonwealth Scholarship. The work has been supported by a grant from the Science Research Council.

References

[1] D.S. Chemla, P.J. Kupeck, D.S. Robertson and R.C. Smith, Opt. Commun. 3 (1971) 29.
[2] D.C. Hanna, R.C. Smith and C.R. Stanley, Opt. Commun. 4 (1971) 300.
[3] G.C. Bhar and R.C. Smith, unpublished.
[4] C.G. Bethea, IEEE J. Quantum Electron. QE-9 (1973) 254.
[5] G.D. Boyd, H. Kasper and J.H. McFee, IEEE J. Quantum Electron. QE-7 (1971) 563.
[6] P.P. Sorokin, J.R. Lankard, E.C. Hammond and V.L. Moruzzi, IBM J. Res. Develop. 11 (1967) 130.
[7] Y. Miyazoe and M. Maeda, Appl. Phys. Letters 12 (1968) 206.
[8] M.L. Spaeth and D.P. Bortfeld, Appl. Phys. Letters 9 (1966) 179.
[9] Ph.J. Kupecek, D.S. Chemla and C.A. Schwartz, unpublished.
[10] D.C. Hanna, B. Luther-Davies, H.N. Rutt, R.C. Smith and C.R. Stanley, IEEE J. Quantum Electron. QE-8 (1972) 317.