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Tunable Medium Infrared Generation in Silver Thiogallate (AgGaS₂) by Down-Conversion of Flash-Pumped Dye-Laser Radiation

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Abstract—The outputs of flash-lamp pumped rhodamine 6G and rhodamine B lasers have been mixed in a $AgGaS_2$ crystal. The generated difference frequency radiation has been tuned from 8.7 to 11.6 μm. Measurements of laser-induced damage of $AgGaS_2$ have been made in the region of 600 nm which show a rapid fall in threshold with decreasing wavelength.

Silver thiogallate ($AgGaS_2$) is an interesting nonlinear optical material [1]–[3]. Its transmission range extends from 500 nm to 13 μm, and its birefringence is large enough to allow phase matching through a useful part of this range. Tunable down-conversion by mixing ruby-laser and ruby-pumped dye-laser outputs in $AgGaS_2$ has recently been demonstrated [4]. In an earlier paper [5] we suggested that down-conversion of rhodamine dye lasers in $AgGaS_2$ could be an attractive scheme for generating tunable infrared radiation. This would exploit the 500-nm transmission limit of $AgGaS_2$ and the use of rhodamine dyes can offer the important advantage of higher repetition rates than dyes pumped by a ruby laser. In this correspondence we report the down-conversion of radiation from two rhodamine lasers and we believe this is the first published report of any down-conversion device using flash-pumped dye lasers. A tuning range from 8.7 to 11.6 μm has been covered.

The two dye lasers were pumped by a single flash lamp lying along the common focus of a double elliptical cylinder. In this way good simultaneity of the dye-laser pulses was ensured. The dye solutions were flowed through capillary tubes located at the other two foci of the double ellipse. These solutions were a 2 × 10^{-4}-M solution of rhodamine 6G in methanol (laser 1) and a 10^{-4}-M solution of rhodamine B in methanol (laser 2). Coarse frequency tuning of both lasers was effected by Brewster-angle prisms [6], and finer tuning and spectral narrowing was achieved by Fabry–Perot filters. The spectral linewidths were ~5 cm⁻¹ for laser 1 and ~8 cm⁻¹ for laser 2. TEM₀₀ mode operation was achieved by means of a circular aperture placed close to the output mirror of each laser. The angular divergence of the beams was ~2 mrad.

The output powers were ~1 kW in pulses of ~0.5-μs duration. Both lasers gave a horizontally polarized output and after rotating the plane of polarization of laser 1 through 90° by a z-cut quartz plate, the two beams were combined at a dichroic mirror. By means of angular adjustments and lateral displacements of the various mirrors it was possible to ensure good spatial overlap and, more importantly [4], a good collinearity of the two beams. The beams were focused into the $AgGaS_2$ crystal
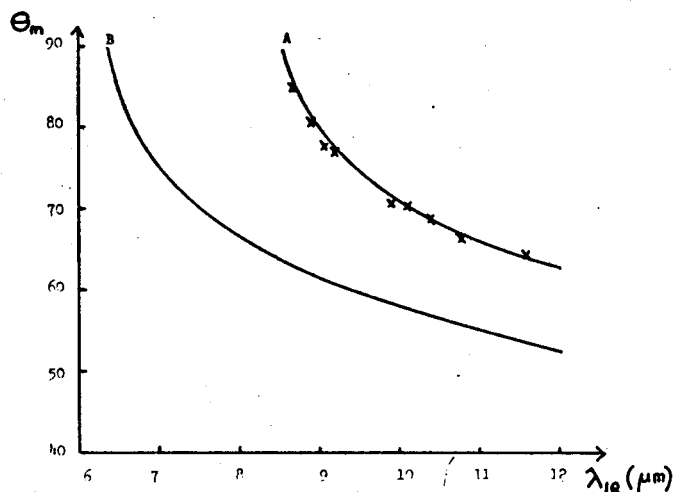
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Fig. 1. Tuning curves for $AgGaS_2$ down-converter. The crosses show experimentally observed phase-matching angles. The solid curves are calculated from refractive index data. Curve B shows the tuning behavior that would be obtained with one laser (rhodamine B) fixed at 633 nm, the other laser (cresyl violet and rhodamine 6G) tuned from 668 to 702 nm.

by a 50-cm lens and this produced an infrared power of ~100 μW at 10 μm. The predicted power for dye-laser spot sizes (W_0) of 1 mm in the 1.5-mm-long crystal is ~3 mW. The reason for the discrepancy between predicted and observed powers is not yet clear.

The $AgGaS_2$ crystal was one that had been used in a down-conversion experiment with ruby-laser pumped dyes (crystal B of [4]). It was in the form of a plate 1.5 mm thick with faces of 5 × 8 mm, cut for type I phase matching with the optic axis at 75° to the face normal. Phase matching was obtained by angle tuning and the crosses of curve A in Fig. 1 show the observed phase-matching angles as the infrared wavelength was tuned from 8.7 to 11.6 μm. This range was achieved by tuning the rhodamine B laser from 617 to 628 nm while keeping the rhodamine 6G laser fixed at 586 nm. The solid curve in the figure shows the corresponding predicted [7] phase-matching angles. With the particular dye frequencies used in this experiment, the rather small birefringence of $AgGaS_2$ limited the shortest infrared wavelength to 8.7 μm. The long wavelength limit for which infrared could be detected was determined by the falling sensitivity of the detection system rather than by the onset of two-phonon lattice absorption which sets in at about 13 μm. The dye lasers used in our experiment were of a rather modest power compared to typical commercially available lasers. To make a reliable estimate of the infrared power that can be expected when using these more powerful lasers it is necessary to know the threshold for laser-induced damage of $AgGaS_2$. Previously, measurements of damage in $AgGaS_2$ had been made only at 1.06 μm and 10.6 μm [8]. Using essentially the same techniques as in [8] we have now extended these measurements to the region between 580 and 625 nm using the lasers described above. The results are shown in Table I. The small difference between the values for extraordinary and ordinary radiation correlate with the observed polarization dependence of the absorption for the crystal used.

The most striking feature of the damage measurements is the rapid fall in threshold with decreasing wavelength in this spectral region. Obviously, there is then an advantage to be gained from using dye lasers operating further towards the red. This is also an advantage for another reason, since the birefringence increases for longer wavelengths, permitting a wider tuning range for the difference frequency. Curve B in the figure shows a calculated tuning curve using one dye laser fixed at 633 nm while the other laser tunes from 668 to 702 nm. These wavelengths can be obtained from flash-pumped laser systems using rhodamine B and

TABLE I

DAMAGE MEASUREMENT ON AgGaS₂ USING FLASH-LAMP-PUMPED DYE LASERS

| | Damage Threshold Intensity MW/cm² for Laser Pulses of 0.5-μs Duration | | | | | |
| | Extraordinary Wave | | | Ordinary Wave | | |
Wavelength	1 shot	3 shots	10 shots	1 shot	3 shots	10 shots
580 nm			1.2			0.8
590 nm	>4	3.0	2.0	3.8	2.4	1.8
625 nm	5.0	4.5	3.6	4.0	3.6	2.5

cresyl violet [9] with efficiencies comparable to rhodamine 6G [10]. It therefore seems realistic to consider the infrared power that could be obtained by mixing 10 kW of power from two such dye lasers in a 3-mm-long crystal of AgGaS₂. Keeping the total power density a factor of 2 below the measured multiple-shot damage threshold (this would require crystal faces of 2 mm × 2 mm), the calculated infrared power at 10 μm is 0.5 W. As seen from Fig. 1., this would be tunable from 6.5 to 13 μm. A more useful figure to quote is the energy/pulse which, with typical duration of 0.5 μs would be 0.25 μJ. This energy would give a S/N ratio of better than 10⁵ with typical pyroelectric detectors for any one pulse. Operation of these flash-pumped dye lasers at 50 Hz [11] would give an average power of 10 μW. Linewidths of flash-pumped dye lasers under 10⁻³ cm⁻¹ have been reported [12] and therefore comparable infrared linewidths would be produced by down-conversion. It is clear that the practicality of a down-converter device based on AgGaS₂ and using flash-pumped dye lasers is restricted by the rather short life of the flash lamps. . Even bearing this in mind we would suggest that such a device might merit serious consideration for a number of applications in the infrared.

ACKNOWLEDGMENT

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Electronic Tuning of a Dye Laser with Simultaneous Multiple-Wavelength Output

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Abstract—Rapid simultaneous multiple-wavelength tuning of a dye laser has been achieved electronically by inserting an acoustooptic beam deflector inside the laser cavity. A beam deflector and diffraction grating combination serve as the tuning element. By applying multiple-RF signals to the acoustooptic deflector, multiple-wavelength operation is readily obtained.

It is possible to tune a laser electronically with simultaneous multiple-wavelength operation. Two-wavelength operation has previously been achieved by different methods [1], [2]. A scheme in which the two wavelengths are decoupled by forcing the dye laser to produce them with mutually orthogonal polarization [3] has been demonstrated recently. Also, simultaneous laser oscillation at multiple-wavelengths was achieved by simply using more than one diffraction grating [4]. The gratings were oriented such that the Bragg condition of each of the gratings allowed different laser frequencies to oscillate. This correspondence deals with an electronic method of producing simultaneous multiple-wavelength laser tuning.

Single-frequency electronic tuning has been demonstrated by inserting an acoustooptic beam deflector in the laser cavity [5]. The intracavity optical beam was deflected onto a diffraction grating, which in turn produced a narrow-band high-Q condition. Sweeping the RF signal, which is applied to the acoustooptic crystal, caused the optical beam to scan across the diffraction grating, thus providing for an electronically tunable laser. The acoustooptic deflector and diffraction grating combination yields the following dispersion equation [6]:

$$\frac{\Delta\lambda}{\Delta f} = \frac{\lambda}{\dfrac{mv}{2d\cos\theta} - \dfrac{2v}{\lambda}\sin\left(\dfrac{\theta - \beta + \phi}{2}\right)} \quad (1)$$

where λ is the optical wavelength, m is the diffraction grating order, v is the acoustic velocity, d is the grating spacing, $\theta = \sin^{-1}(m\lambda/2d)$, β is the angle between the diffraction grating and the beam deflector, and ϕ is the Bragg angle of the acoustooptic crystal at the center of the bandwidth of the acoustic signal and center optical wavelength of the laser. The Bragg angle is selected in the above manner because the acoustooptic beam deflector cannot be rotated at the speed required to keep up with the speed of tuning. Thus the deflector is kept fixed and the Bragg condition is not satisfied exactly for all frequencies.

With the development of a new acoustooptic material (TeO₂) multiple-wavelength operation using the previously described transverse geometry has become feasible. The anomalously low sound velocity of TeO₂ (0.617×10^5 cm·s⁻¹) provides for an acoustooptic beam deflector with a very large scan angle. This