

A HIGH POWER, WIDELY TUNABLE INFRARED SOURCE BASED ON STIMULATED ELECTRONIC RAMAN SCATTERING IN CAESIUM VAPOUR

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Dye lasers pumped by the second harmonic of a ruby laser have been used to produce tunable stimulated electronic Raman scattering in caesium vapour. Using three different Raman transitions, the infrared tuning ranges were 2.5–4.75 μm , 5.67–8.65 μm and 11.7–15 μm with powers of up to 25 kW, 7 kW and 2 kW respectively.

Generation of long-wavelength infrared radiation by stimulated electronic Raman scattering (SERS) in caesium vapour has been reported by Sorokin and Lankard [1]. The pump radiation was in the form of ultrashort pulses at 347 nm (second harmonic of a mode-locked ruby laser) and therefore the Raman wavelength could not be tuned. A subsequent attempt to generate tunable infrared radiation using a low power, dye laser beam in potassium vapour gave only a very small range of Raman tuning [2]. More recently however, near-infrared stimulated Raman radiation ($\sim 3 \mu\text{m}$) with a useful tuning range has been obtained both in potassium vapour [3] and barium vapour [4]. In this letter we report a considerable increase in infrared tuning range, by using three Raman transitions in caesium vapour and using dye lasers pumped by the second harmonic of a high power ruby laser. In this way SERS has been tuned over the ranges 2.5–4.75 μm , 5.67–8.65 μm and 11.7–15 μm , with infrared powers up to 25 kW.

Fig. 1 shows the relevant energy levels of atomic caesium. For Raman frequencies ω_{S1} , ω_{S2} , ω_{S3} corresponding to infrared wavelengths in the ranges 2–5 μm , 5–10 μm and 9–20 μm respectively, the required dye laser wavelengths are 425–487 nm, 380–395 nm and 357–365 nm. This complete range can be covered using four or five different dyes.

A frequency doubled ruby laser giving 5–10 MW of 347 nm radiation in a 20 ns pulse was used to transversely pump a dye laser oscillator–amplifier combination. The oscillator, of similar design to that described

in [5], was pumped by $\sim 20\%$ of the 347 nm output, the remainder being used to pump a 2 cm amplifier cell. The performance of this laser system is summarised in table 1. The dye laser linewidth was typically $< 0.1 \text{ cm}^{-1}$ for the coumarin dye and $0.15\text{--}0.2 \text{ cm}^{-1}$ for the

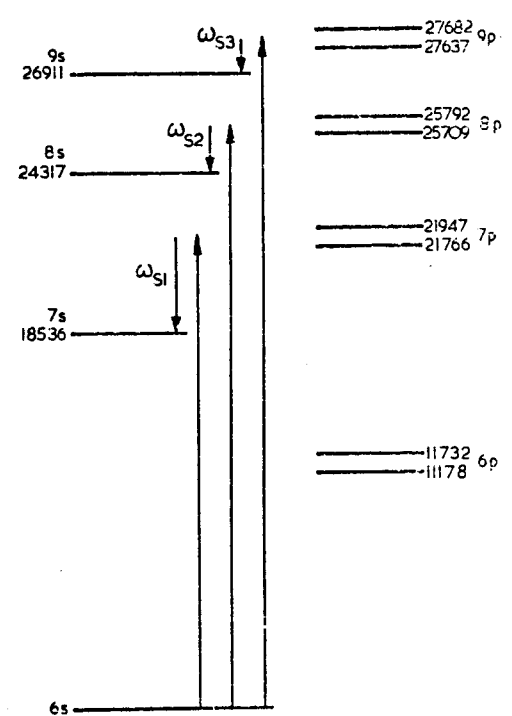


Fig. 1. Energy level diagram of atomic caesium.

Table 1
Summary of dye laser performance. (The wavelength limits given do not represent the full tuning ranges of the dyes.)

Wavelengths (nm)	Dye	Solvent	Output power (kW)	Pulse length (ns)
358–365	Butyl-PBD	Toluene	200–500	4– 6
380–390	PBD	Toluene	350–750	4– 8
388–395	PBBO	Cyclohexane	500–700	8–10
440–485	Coumarin 1	Methanol	500–800	10–13

shorter wavelength dyes, this difference in linewidth being due to the higher angular dispersion of the holographic grating at longer wavelengths. The output from the dye oscillator–amplifier was focussed into a 35 cm long, heat pipe oven containing caesium vapour. The output window was a CsI flat. Vapour pressures between 3 and 30 torr were used. The generated SERS radiation was detected after semiconductor filters and a grating monochromator. This was used to discriminate against any fixed frequency atomic emissions and also to measure the Raman wavelength. Several different detectors have been used (InSb, HgCdTe, Ge:Cu and a TGS pyroelectric). The minimum observable signal corresponded to powers of 100 μW – 1 mW incident on the detector. Using a 50 cm focussing lens, outputs were obtained over the following ranges: 2.5–4.75 μm , 5.67–8.65 μm , 11.65–15 μm for Raman transitions to the 7s, 8s and 9s levels respectively. The peak Raman output powers for these ranges were 25 kW, 7 kW and 2 kW, measured on a calibrated pyroelectric detector at the exit window of the heat pipe. Powers detected after the monochromator were greater than 1 W over $\sim 80\%$ of the total tuning range and could be easily monitored with the pyroelectric detector. Pulse widths for $\sim 7 \mu\text{m}$ and $\sim 13 \mu\text{m}$ SERS output were 4–7 ns and 1.5–3 ns respectively, measured using the Ge:Cu detector and a 500 MHz oscilloscope. Considerable substructure was frequently seen in the Raman pulse.

We have calculated the theoretical threshold and hence tuning range of SERS in caesium vapour subject to the following assumptions; (1) that the spontaneous Raman linewidth is the result of Doppler broadening (i.e. typically 0.03 cm^{-1}), (2) that the dye laser linewidth is less than the Doppler width, and (3) the dye

laser beam is assumed to be gaussian, focussed at the centre of the heat pipe with a confocal parameter equal to the length of the vapour column. For these calculations, we have taken the published values for the atomic energy levels [6] and the dipole radial matrix elements [7]. We have included the effects of spin–orbit splitting [8] and also the considerable diffraction loss of the generated Raman radiation [9]. Fig. 2 shows the calculated threshold for a caesium vapour pressure of 10 torr. It may be seen from these curves that a cancellation of the Raman susceptibility occurs for a pump wavelength between the doublet p levels. This cancellation was observed experimentally as a drop in Raman output (although not to zero) when the pump was tuned between the $9p_{3/2}$ and $9p_{1/2}$ levels and the observed minimum is within 0.5 cm^{-1} of the calculated cancellation point.

Fig. 2 also indicates that complete coverage of the 2–20 μm range should be possible using a dye laser power of only 10 kW provided the laser linewidth is less than 0.03 cm^{-1} . The observed thresholds are as much as two orders of magnitude higher than these predicted thresholds (even when allowance has been made for the finite dye laser linewidth).

A number of other observations are also inconsistent with the theoretical calculations. Thus, the tuning ranges were hardly affected by changing the dye laser linewidth from 0.1 to 1 cm^{-1} , or by changes in the spatial quality of the dye laser beam. Also it was found that changing the caesium pressure over the range 3–30 torr had only a little effect on tuning range whereas the theoretical calculation suggests that this tenfold increase of pressure should give approximately a threefold increase in tuning. The linewidth of the generated radiation in the 7 μm and 13 μm regions was measured to be between 0.3 cm^{-1} and 1.0 cm^{-1} . Little change in this linewidth was observed when the caesium pressure was changed from 5–30 torr. On the other hand preliminary measurements have shown that the linewidth decreased with decreasing dye laser intensity, produced either by attenuating the dye beam or by focussing it less tightly.

Many of these observations appear to be consistent with an intensity-dependent broadening mechanism which broadens the final level of the Raman transition to a value considerably in excess of the calculated Doppler width. One possible mechanism is two-photon ionisation with subsequent Stark broadening of the

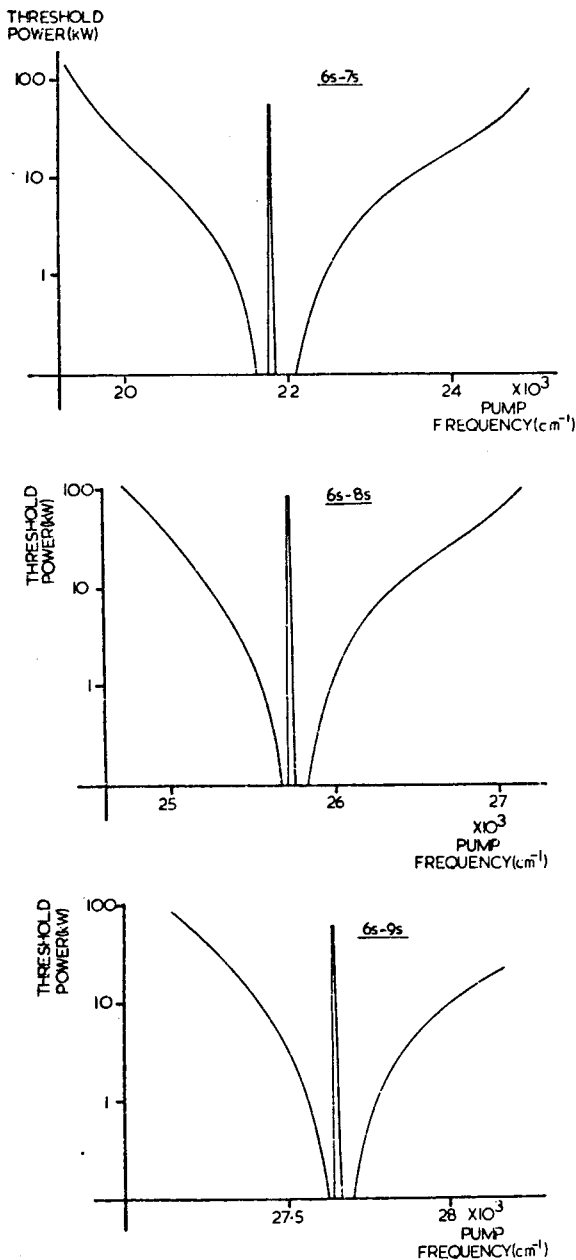


Fig. 2. Calculated thresholds for SERS using the three indicated Raman transitions in caesium vapour at 10 torr pressure.

atomic levels by the ions and electrons, although further measurements will be required to check this sug-

gestion. Without knowing the precise mechanism for the intensity-dependent broadening it is still possible to suggest an approach for increasing the observed tuning range. By expanding the dye laser beam (and hence reducing its intensity) the resulting reduction in the Raman gain per unit length would be more than compensated by the increased length over which this gain could be maintained, either by using a longer heat pipe oven or by using a Raman oscillator cavity [9].

In conclusion we have shown that stimulated electronic Raman scattering in caesium vapour provides a simple way of extending the tuning properties of dye lasers to a wide range of medium infrared wavelengths. Infrared linewidths of less than 1 cm^{-1} and powers of several kilowatts are readily obtained. The observed threshold and tuning are still far from agreeing with calculated values and the source of this disagreement has yet to be conclusively identified. Nevertheless it is to be expected that significant improvements in tuning range and linewidth will be achieved.

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