

## Tunable Electronic Raman Laser at 16 µm

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Abstract. 6 s-9 s electronic Raman scattering in caesium vapour is used to generate tunable 16  $\mu$ m radiation. Output energies in excess of 8  $\mu$ J were obtained, with peak powers in the kilowatt range.

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Stimulated Electronic Raman Scattering (SERS) [1] is a well-known technique for frequency conversion of tunable dye laser radiation into the infrared spectral region. Little effort has been applied to extending the tunability of this method to produce outputs in the 16  $\mu$ m region, which is of interest for the laser separation of uranium isotopes. By using an improved superheated heat-pipe design, with caesium vapour as the SERS medium, we have obtained energies in excess of 8  $\mu$ J in the 16  $\mu$ m region, with a simple dye laser pump source.

A Lambda-Physik EMG 101 excimer laser was used with XeCl\* to pump a modified FL 2000 dye laser, which produced 10 ns pulses of up to 15 mJ at 3631 Å in a bandwitdth of ca.  $0.5 \text{ cm}^{-1}$ . The dye laser output then entered a caesium heatpipe oven through an optional telescope for optimisation of the focussing conditions. Most of the measurements, however, were carried out using the unfocussed beam from the dye laser; this beam was approximately circular in shape, and ca. 1.3 mm in diameter at the entrance window of the heatpipe oven. The heatpipe was of the split-wick type used in previous picosecond Raman experiments [2], with a central, superheated region 70 cm long. Superheating excess temperatures of 400 °C were obtained giving a reduction in caesium dimer density to 4%-10% of the saturated number density, at saturated caesium pressures between 15 and 100 mbar.

The 16  $\mu$ m radiation, generated by SERS from the 6s ground state to the 9s excited state of the caesium

atom, passed through the NaCl output window and Ge and InSb filters (cut-off wavelength: 12.5 µm), and was focussed onto a calibrated Mullard pyroelectric detector, either directly or through a 1/4 m monochromator. As the photodissociation of the caesium dimers remaining in the active region results in the production of excited caesium atoms, mainly in the  $9p_{1/2}$  state, for this excitation wavelength, an interfering atomic superfluorescent output is also present, on the  $9p_{1/2}-9s$ transition, at 13.76 µm [3]. This superfluorescence and the Raman emission were found to be the only sizable contributions to the pulse energy passed by the NaCl window and semiconductor filters. From their relative intensities, together with the total passed pulse energy, we obtained the absolute output energies per pulse, of the SERS and the competing superfluorescence. This is shown in Fig.1 as a function of caesium vapour pressure, for two levels of dye laser input, and without further focussing.

The dye laser pulse shapes at these input energies are shown in the inset. The faster rise time of the higher energy pulse results in a reduction of the fixed frequency laser output, probably due to a more rapid build-up of the SERS, allowing less time for competing ionisation and dissociation processes. A broad maximum for the 16  $\mu$ m energy, with peak energy of 8.5  $\mu$ J per pulse, occurs for pressures between 15 and 30 mbar, and the 13.76  $\mu$ m laser signal increases up to the highest pressures investigated.

The effect of varying the focussing of the dye laser beam into the heatpipe was investigated at a pressure of 20 mbar. It was found that a two-fold increase in beam diameter brought the Raman process close to threshold, and resulted in very weak unstable pulses,

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Fig. 1. Output energies of SERS at  $16 \,\mu\text{m}$  and of the atomic  $9p_{1/2} - 9s$  superfluorescence at  $13.76 \,\mu\text{m}$ , as a function of caesium saturated vapour pressure. The dashed and dotted lines refer to input energies of 15 and 7 mJ per pulse, respectively. The corresponding pulse shapes are shown as inset

while a two-fold beam reduction reduced the  $16 \,\mu\text{m}$  output to approximately half its original value. Still tighter focussing, using a 1 m lens in front of the heatpipe, to produce a confocal parameter for the dye laser beam of ~15 cm, resulted in complete quenching of the Raman signal, and bright yellow and red fluorescence could be seen from the focal zone.

At the optimum pressure of 25 mbar, and with the highest dye laser input, pulse-to-pulse amplitude stability of the 16  $\mu$ m signal was approximately half that of the pumping dye laser, with  $\pm 10\%$  peak-to-peak variation. Measurements of the spectral width of the generated radiation were not attempted here, although previous measurements with a subnanosecond dye laser system [4] had producded monochromator-limited linewidths below  $0.2 \text{ cm}^{-1}$ .

In conclusion, we have extended the range of electronic Raman laser action to the  $16 \,\mu\text{m}$  region, with peak pulse energies of  $8.5 \,\mu\text{J}$  at  $16 \,\mu\text{m}$ . Further improvements to the output energy are possible with higher dye laser energy, shorter pump pulses [2,4], and longer heatpipe ovens.

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