

## HIGH EFFICIENCY AND HIGH BRIGHTNESS RAMAN CONVERSION OF DYE LASER RADIATION

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A technique for efficient Raman conversion of an excimer pumped dye laser is described, based on the use of a Raman oscillator/amplifier combination. A photon conversion efficiency of 90% to first Stokes is demonstrated, with diffraction-limited beam quality.

Stimulated Raman scattering provides a simple means for extending the tuning range of dye laser radiation into regions where dye laser operation is inefficient or difficult to achieve. The simplest, and most widely used arrangement involves tight focussing of the pump beam into the Raman medium, with the Stokes radiation generated on a single pass through the medium. In practice however this arrangement gives poor control over the various processes which can occur, such as multiple Raman shifts, backward Raman scattering and generation of higher order Stokes and anti-Stokes radiation through four wave mixing. The results of such an uncontrolled mixture of processes can be poor conversion efficiency and a large angular spread of the generated radiation. By contrast, it has been shown that by arranging the Raman medium as an oscillator-amplifier combination [1-5] with a small fraction of the pump used for the oscillator and the main part of the pump used to drive the amplifier, good control can be obtained and high conversion efficiency to a selected Stokes order can be achieved. The main disadvantage of the oscillator-amplifier scheme is the extra complexity introduced and in particular the exacting requirement on spatial overlap of the pump beam and injected Stokes beam in the amplifier stage. Stapperts et al. [6] have also shown that where a broadband pump is used the gain in the amplifier is reduced unless the injected Stokes beam and pump beam at the amplifier are adjusted to travel over equal paths from the pump

laser. This however introduces a further complexity (fig. 1 shows a schematic set up). In this letter we describe a scheme for stimulated Raman scattering from a dye laser in which the required spatial and temporal overlap of the pump and Stokes beams at the Raman amplifier are automatically achieved. The technique can be extended to multiple Raman shifting but we report here the results obtained where conversion to the 1st Stokes was optimised. With a XeCl excimer-pumped dye laser of  $\sim 7$  mJ output, using  $H_2$  gas as the Raman medium, we obtain 90% photon conversion efficiency into a diffraction-limited 1st Stokes beam.

The experimental arrangement is shown in fig. 2. The essential feature is that Raman oscillator stage is before the final dye amplifier stage. The Stokes radiation and the residual dye laser radiation from the Raman oscillator are collinear and this collinearity is preserved after passing through the final dye amplifier cell. Further more in this scheme the final dye amplifier also maintains the temporal coincidence, first established in the Raman oscillator between the Stokes intensity fluctuations and the dye oscillator intensity fluctuations.

Our dye laser system consists of an oscillator and two amplifiers, pumped by a XeCl excimer laser with a 20 ns, 100 mJ output. The dye oscillator incorporated a four-prism beam expander and 2800 line/mm holographic grating to narrow the linewidth to  $0.3 \text{ cm}^{-1}$ . A 400  $\mu\text{m}$  diameter aperture was inserted in

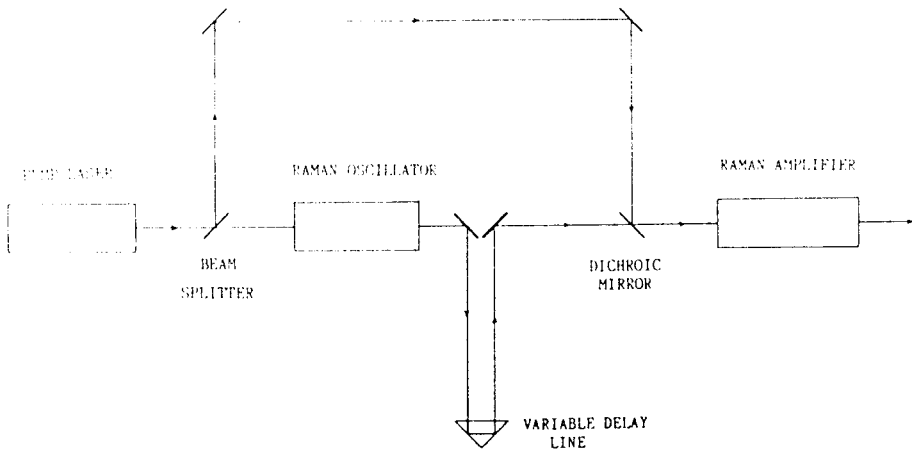


Fig. 1. Schematic of Raman oscillator/amplifier with provision for equalising paths from the pump laser to the Raman amplifier.

the dye oscillator to ensure a diffraction-limited output. With  $\sim 3$  mJ pump energy into the dye oscillator, its output was  $40 \mu\text{J}$  in a 12 ns pulse and this was amplified to  $\sim 700 \mu\text{J}$  in the first dye amplifier cell, pumped by  $\sim 10$  mJ from the excimer laser. The Raman oscillator cell, filled with  $\text{H}_2$  to a pressure of 25 atm., contained a fused silica capillary waveguide of  $200 \mu\text{m}$  bore diameter and 72 cm length. The use of the capillary offered two main advantages. The lower SRS threshold [7] meant that more pump

energy was available for the second dye amplifier, hence improving the overall conversion efficiency. Also the output beams from the capillary (both the Stokes beam and the residual dye beam) were constrained to be diffraction-limited by the spatial filtering action of the guide. The measured transmission of the capillary with the dye laser beam optimally focussed into the capillary was 70%, somewhat lower than the calculated value [8] of 90% for a dye laser wavelength of 565 nm. With  $680 \mu\text{J}$  of dye laser input to the

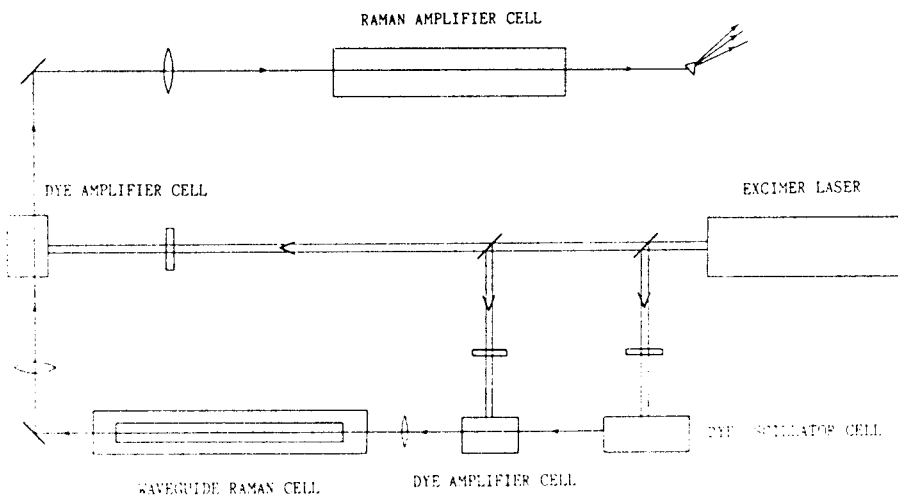


Fig. 2. Experimental configuration of the dye-laser-pumped Raman oscillator-amplifier.

Raman oscillator (i.e. about a factor of two above threshold), the 1st Stokes (738 nm) output energy was 60  $\mu\text{J}$ . In this experiment we deliberately kept the pump energy low enough to prevent significant second Stokes generation since our aim was to achieve maximum conversion to first Stokes in the Raman amplifier. The first Stokes and remaining dye laser beam from the capillary exit then pass through the second dye amplifier, a 2 cm long cell pumped by 70 mJ of excimer laser energy. The first Stokes beam suffered a small attenuation, from 60  $\mu\text{J}$  input to 40  $\mu\text{J}$  output, in the dye medium (Rhodamine 6G in methanol) whereas the dye laser beam was amplified to an energy of 7 mJ. The collinear dye and Stokes beams were then loosely focussed by a lens of 50 cm focal length, into the Raman amplifier cell. The cell length  $l$  was

35 cm and the dye beam waist, formed at the centre of the cell, had a spot size radius  $w_0$  of 350  $\mu\text{m}$ . Under these conditions the power gain  $e^G$  in the amplifier cell was insufficient for Stokes generation to occur without the injected Stokes beam. A rough calculation of  $G$ , showed that with the maximum dye input (5.5 mJ in 12 ns), the peak, on-axis Raman gain  $G$  was only approximately 20 confirming that self oscillation would not be expected. Typically around 30  $\mu\text{J}$  of Stokes radiation could be injected into the Raman amplifier cell and fig. 3 shows the measured energy conversion efficiency to 1st Stokes as a function of dye laser input energy. Energy measurements were made with a pyroelectric energy meter. The maximum energy efficiency was 70%, corresponding to a

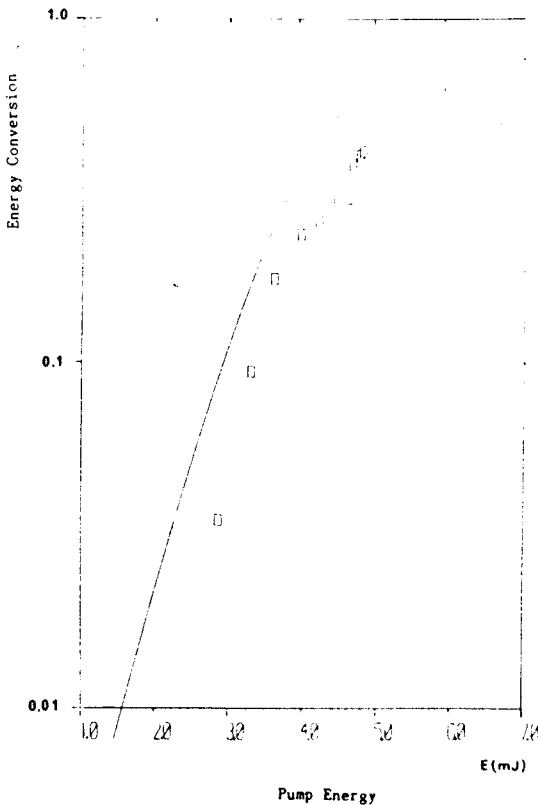


Fig. 3. Raman energy conversion efficiency into the first Stokes as a function of the input dye laser energy. The squares indicate the experimental results, the solid line corresponds to the calculation.

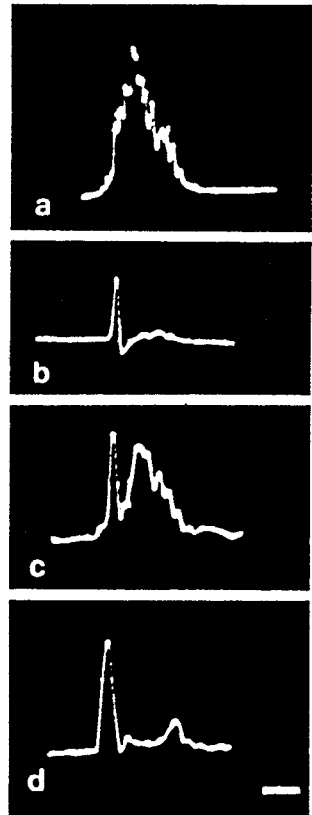


Fig. 4. The temporal profiles of the dye laser at various stages in the configuration shown in fig. 2: (a) at the input of the capillary, (b) at the output of the capillary, (c) after the second stage dye amplifier, (d) after the Raman amplifier. The bar in (d) represents 5 ns.

photon conversion efficiency of 90%. Also shown in fig. 3 is a calculation (solid line) of conversion efficiency, including the effect of pump depletion. The calculation is approximate since it makes the simplifying assumption that the pump beam of power  $P$  has a uniform transverse profile with intensity  $I_p$  given by  $I_p = P/\pi w_0^2$ . The Raman gain coefficient used in this calculation is  $g_R = 2.4 \times 10^{-11}$  m/W ( $g_R I_p l \equiv G$ ), corresponding to the more general expression

$$g_R = \frac{1.8 \times 10^{-11} \text{ m/W}}{\lambda_S (\mu\text{m})},$$

where  $\lambda_S$  is the Stokes wavelength [9].

Figs. 4 and 5 show the temporal behaviour of the dye laser pulse and first Stokes pulse at various stages through the system. In fig. 4b it is seen that the dye pulse emerging from the capillary is heavily depleted except for a spike left at the beginning of the pulse. Fig. 4c shows the dye pulse leaving the second dye amplifier and it can be seen that most of the energy now goes into the depleted portion of the dye pulse rather than the spike. This is an important feature for

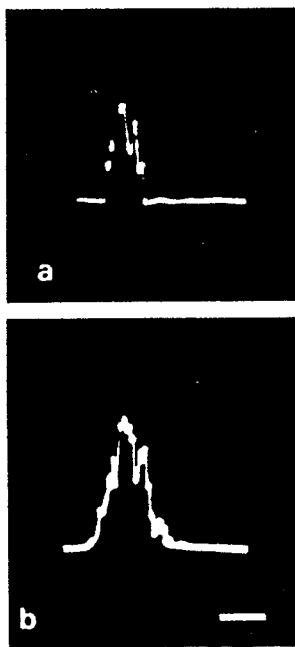


Fig. 5. The temporal profiles of the first Stokes radiation: (a) after the capillary waveguide, (b) after the Raman amplifier. The bar in (b) represents 5 ns.

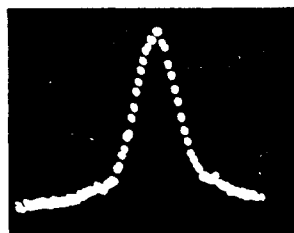


Fig. 6. Transverse intensity profile of the amplified first Stokes pulse, as monitored by a photodiode array.

efficient Stokes conversion in the Raman amplifier since the injected Stokes radiation coincides with the depleted portion of the dye pulse. Fig. 4d shows the transmitted dye pulse after the Raman amplifier (vertical scale of 4d ten times less sensitive than fig. 4c). Figs. 5a and b show the Stokes pulse before and after the Raman amplifier, respectively, with 5b having a vertical scale ten times less sensitive than in 5a.

The Stokes output beam quality was assessed by focussing the beam then measuring the waist-size and the beam divergence from this waist. This measurement showed that the beam was diffraction-limited and fig. 6, which shows the intensity profile recorded with a diode array confirmed the smooth profile that was noted visually.

**Conclusions.** We have demonstrated a stimulated Raman scattering arrangement involving a Raman oscillator/amplifier configuration which automatically ensures excellent spatial and temporal overlap between the pump beam from a dye laser and the injected Stokes beam. This results in a very high conversion efficiency and suggests that near infrared radiation may be generated more efficiently by this method rather than by direct pumping of near infrared dyes.

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