A HIGH POWER, SHORT PULSE STIMULATED RAMAN SOURCE AT 1.54 μm

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An actively modelocked Q-switched Nd:YAG laser produces efficient stimulated Raman scattering in high pressure methane, using a Raman oscillator/amplifier configuration. First Stokes output at 1.54 μm is generated up to 30 MW in a single pulse of ~40 ps duration. Good temporal and spatial coherence is maintained with the same pulse energy stability, ±6%, as the Nd:YAG output.

With the increasing availability of dye lasers and Nd:YAG lasers [1–3] which can produce high power modelocked pulses there is a growing need for efficient nonlinear frequency conversion techniques to extend the spectral range of such sources. Stimulated Raman scatterings (SRS) is one such proven scheme with the attractions of simplicity, cheapness and availability of suitable Raman media. With the power levels available in modelocked pulses (typically ~1 mJ in 100 ps ie. tens of megawatts [1,2]) the threshold for SRS in gases (eg. CH_4, H_2 and D_2) can be exceeded by at least an order of magnitude [4]. This suggests that high conversion efficiency should be possible. However, in practice unless precautions are taken, competing processes (eg. four-wave mixing) can decrease the efficiency and degrade the spatial coherence of the generated radiation. Komine and Stapperts [5] have shown, using pulses of several nanoseconds duration, that four-wave mixing can be suppressed by the use of a Raman oscillator/amplifier configuration thus allowing the generation of high brightness Stokes radiation (see also refs. [6–8]). Here we report the succesful application of a Raman oscillator/amplifier with much shorter pulses (~100 ps). To demonstrate this technique we have used a Nd:YAG laser (1.06 μm) with high pressure CH_4 gas ($\tilde{\nu}=2917$ cm^{-1}) as the Raman medium. This particular combination provides first Stokes radiation at 1.54 μm, a wavelength of interest in a number of current laser applications. With the Raman oscillator/amplifier arrangement we have generated first Stokes output with an overall energy efficiency of 20% (30% quantum efficiency), and peak powers of 30 MW in pulses of ~40 ps duration. Good temporal and spatial coherence is maintained in the Raman process with the same amplitude stability (±6%) as the pump since the amplifier is driven into saturation.

The pump laser was a pulse-pumped (10 Hz repetition rate) actively modelocked and actively Q-switched Nd:YAG laser (JK Lasers System 2000 AML Series). The TEM_{00} mode Nd:YAG oscillator produced a train of modelocked pulses from which one was selected and amplified in a single stage amplifier to give a pulse energy of 6.9 mJ. The pulse duration, measured by background-free second harmonic autocorrelation, and assuming a gaussian temporal shape, was found to be 100 ps, giving a time–bandwidth product of 0.6.

Initial measurements of SRS performance were made with the 1.06 μm output focussed into a single cell of CH_4 at 30 atm pressure. The pump beam was focussed at the centre of the cell ($L=50$ cm) with a confocal parameter, $b=12.5$ cm ie. $L/b=4$. Fig. 1 shows the results for energy conversion efficiency to first Stokes. The observed threshold energy of 490 μJ is in good agreement with calculation [4]. The peak energy conversion efficiency of 8.5% is reached at 2.5 times the threshold power and at this point, second Stokes and first anti-Stokes were observed. The Raman gain for second Stokes driven solely by the

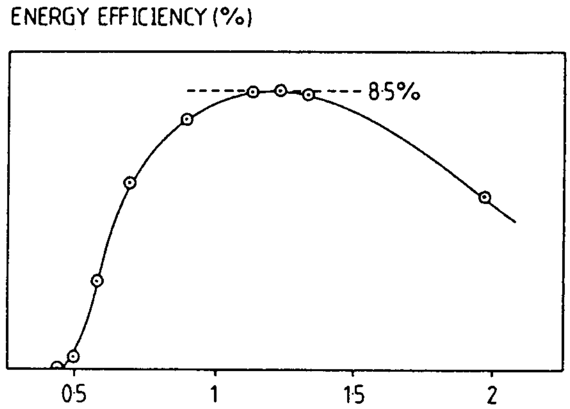

Fig. 1. First Stokes (1.54 μm) efficiency versus pump (1.06 μm) energy from tight focussing arrangement in CH_4 (30 atm).

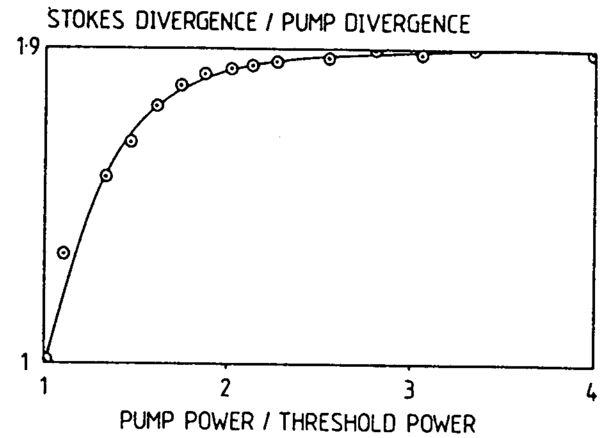

Fig. 2. Stokes divergence versus pump power from a tight focussing arrangement in CH_4 (30 atm).

1.54 μm first Stokes acting as pump (i.e. in the absence of any four-wave mixing processes) is calculated to be ~0.2 cm/GW [4]. The observed power at 1.54 μm at the peak of the curve in fig. 1 would give a gain which is around an order of magnitude less than that required to reach threshold, and it is therefore clear that four-wave mixing processes are involved in the generation of second Stokes. The phase matching conditions for first anti-Stokes and second Stokes generation via four-wave mixing are respectively:

$$\kappa_A = 2\kappa_p - \kappa_{s1}, \quad \kappa_{s2} = 2\kappa_{s1} - \kappa_p .$$

As a consequence of the medium dispersion these phase matching conditions can only be satisfied in a noncollinear geometry and, provided both the pump and first Stokes diverge significantly less that the required phase matching angles, the four-wave processes are suppressed. This condition can apply close to SRS threshold where the Stokes divergence is comparable to the pump divergence. However, as threshold is exceeded, so the length of medium in which first Stokes reaches threshold is reduced, and Stokes radiation can then be generated with greater divergence. We have measured the increase of Stokes divergence for a tightly focussed pump beam ($L/b = 20$) as the pump was increased above threshold, and the results are shown in fig. 2.

This increase in Stokes divergence reduces the Stokes brightness directly, and also indirectly by allowing the generation of second Stokes via four-wave mixing, thus limiting the efficiency of first Stokes generation. To overcome the efficiency limitation and increase in Stokes divergence we have used the Raman oscillator/amplifier configuration shown in fig. 3. In choosing the focussing parameters for this configuration it is necessary to have regard for the breakdown limit of the gas and of the cell windows. Breakdown for the CH_4 gas (99.5% pure, 0.22 μm pore filter) was not observed for the highest input energies implying a breakdown threshold greater than 75 GW/cm^2, however, damage to the fused silica cell windows was observed at intensities as low as 20 GW/cm^2 (although in general it was significantly higher). These figures are taken as the maximum safe levels in designing the oscillator/amplifier. Mirror M1 reflects 8% of the pump energy into the oscillator (CH_4 at 40 atm, $L = 50$ cm focussed to $L/b = 2.6$) to generate sufficient seed radiation (30 μJ) at 1.54 μm. Lens 1 is placed before M1 to provide comparable spot sizes for both the Stokes seed and remaining

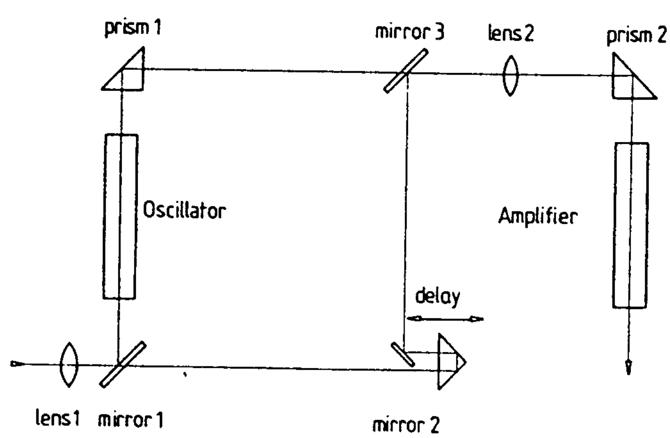

Fig. 3. Schematic layout of the Raman oscillator/amplifier.

pump at the location where they are recombined. In fact, the Stokes mode size, and hence confocal parameter b_s at threshold is roughly equal to that of the pump b_p, with $b_s=0.7\ b_p$ being the calculated result [9].

At mirror M3 where the beams are recombined, the Stokes spot size is calculated to be 1.5 times larger than the pump. This ensures efficient energy extraction from the entire pump beam in the amplifier and also provides some spatial filtering of the seed radiation (although in practice this was not found to be necessary as the seed was already diffraction limited). An adjustable optical delay was incorporated into mirror M2 to achieve synchronism between the pump and seed pulses; synchronism to within 20 ps was required for optimum conversion. Good spatial overlap was also necessary and this was achieved by first aligning the system with a HeNe laser collinear with the 1.06 μm beam. Particular care was taken in positioning focussing lens 2 so that pump/seed beams passed through its centre thus preventing walk off due to chromatic dispersion. The available pump energy entering the amplifier was measured to be 4.5 mJ. Lens 2, 50 cm focal length, was chosen to give a gently convergent pump beam forming a waist ~1 m away from lens 2 (calculated waist spot size $\omega_0=290$ μm, $b=0.5$ m). The amplifier cell (CH_4 at 30 atm, $L=45$ cm) was positioned to give a mean pump spot size of 650 μm, however, more or less gain could be easily achieved by moving the amplifier cell along the convergent pump beam. With a mean on-axis pump intensity of 7 GW/cm^2 and a Raman gain coefficient of 0.33 cm/GW [4] the calculated small-signal steady-state gain is $G=g_R Il$ exp(100). Taking into account the effect of transience [4], this gives a gain of exp(40), just sufficient for threshold to be reached from noise. In fact significant noise amplification was observed when no seed signal was injected, and in addition, owing to the loose focussing geometry which allows first Stokes generation at large angles to the axis, there was evidence of four-wave mixing, in the form of anti-Stokes generation. With seed injection, no evidence of four-wave mixing was found confirming that (a) the seed amplification was saturning the gain and so supressing noise amplification and (b) the amplified Stokes had low divergence. No second Stokes was observed and an energy conversion efficiency of 30% within the amplifier (≡ 1.35 mJ of Stokes) was measured, (see fig. 4). This corresponded to an overall energy efficiency (from the Nd:YAG output) of 20%. Saturation within the amplifier results in a Stokes stability (± 6.2%) comparable to the pump stability (±5.8%). Generation of second Stokes from noise by the first Stokes as pump would require a first Stokes intensity some three times greater than that generated in our experiment. To analyse the temporal evolution of the Stokes pulse we have made calculations based on the equations of Carman [10] for transient SRS, extending their analysis to include the effect of pump depletion. The treatment is restricted to a simple plane wave analysis. The predicted pulse profile (for a 100 ps pump pulse) corresponding to an energy conversion efficiency of 30% is shown in fig. 5. The Stokes pulse duration is 45 ps and an instantaneous photon

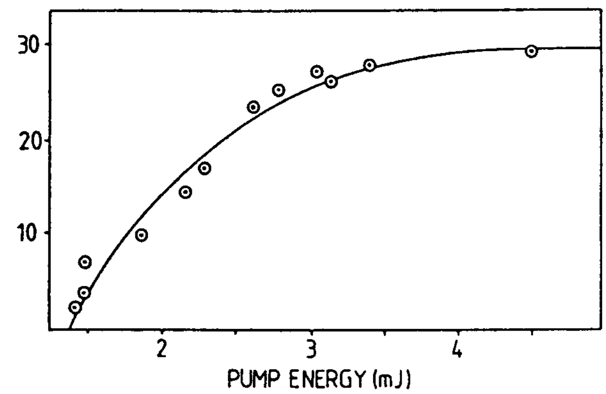

Fig. 4. Performance of the Raman amplifier: energy conversion to first Stokes versus pump energy into the amplifier.

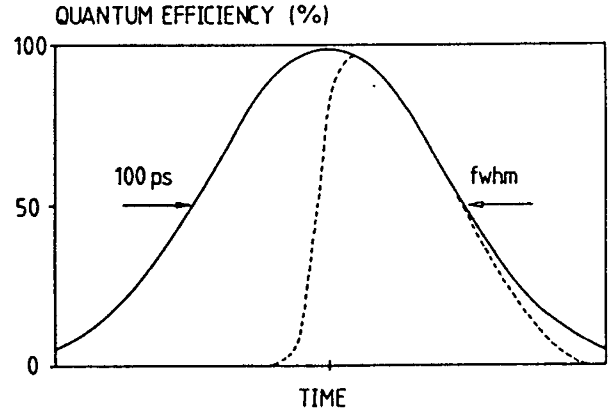

Fig. 5. Pump pulse shape (solid curve, assumed 100 ps fwhm gaussian) and calculated Stokes pulse shape (dashed curved) expressed as quantum efficiency. Conditions of the calculation correspond to 30% energy conversion efficiency.

conversion efficiency of ~90% is predicted. For CH_4 at 30 atm pressure, the ratio of pulse duration to dephasing time is ~5 however, characteristic transient effects are already apparent. To confirm the predicted pulse compression, we have measured the 1.54 μm pulse duration, using background-free second harmonic autocorrelation, to be typically 50 ps with a maximum Stokes energy of 1.35 mJ. This implies a peak power of ~30 MW. A measurement of spectral bandwidth at 1.54 μm has not been made, however, assuming it to be comparable to the spontaneous Raman linewidth, 0.6 cm^{-1} at 30 atm [4], this would imply a time-bandwidth product of ~0.8. In fact, the bandwidth is likely to be somewhat less than the spontaneous linewidth [11], suggesting that the pulses are near bandwidth-limited.

These results indicate that a properly designed Raman oscillator/amplifier can give very efficient conversion to Stokes pulses with well controlled temporal and spatial properties as well as good amplitude stability. In this work we have concentrated on optimization of first Stokes generation. For a mode-locked dye laser source the need arises for multiple Stokes generation in order to extend the tuning range into the near infrared and experiments are now in hand to test the efficiency of the oscillator/amplifier configuration in producing well controlled outputs under conditions of multiple Stokes scattering.

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