HIGHER-STOKES ORDER RAMAN CONVERSION TO THE NEAR INFRARED: HIGH EFFICIENCY AND BRIGHTNESS VIA A CAPILLARY WAVEGUIDE AMPLIFIER

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Efficient Raman conversion in H_2 gas from 0.53 μm to third Stokes (S_3) at 1.58 μm has been demonstrated using an oscillator/amplifier configuration. Seed radiation from the oscillator controls the divergence of the Stokes beams and a capillary amplifier configuration enhances the conversion. An overall 9% photon conversion efficiency to S_3 is achieved with the S_3 beam divergence twice the diffraction limit. Photon conversion efficiency within the amplifier is ~20%.

Stimulated Raman scattering of dye laser radiation in H_2 gas offers an attractively simple means of generating radiation with wide tunability. Multiple Stokes shifts can be produced simply by focussing the pump beam into the Raman medium. By using a capillary waveguide [1–3] the Raman gain can be considerably enhanced and this has allowed efficient conversion of dye laser radiation to third Stokes radiation in the infrared [2,4]. A problem generally encountered with such Raman generators, whether a guided or unguided geometry is used, is that the higher order Stokes radiation may be generated with a large angular spread. Potential uses of this Stokes radiation may be vitiated by this reduced spatial coherence and potential users are often unable to assess the potential utility since most reported results on such Raman generators do not include quantitative data on beam quality. Techniques for achieving efficient multiple Stokes generation with near diffraction-limited beam quality have been demonstrated [5–7]. The principle involves first generating the multiple Stokes radiation in a Raman oscillator and then injecting this radiation, after suitable spatial filtering, into a Raman amplifier where it acts as a seed from which each of the successive Stokes orders will grow. The generated Stokes radiation can then preserve the spatial coherence of the injected radiation. In their demonstration of this principle Komine and Stappaerts [5,6] used an ultraviolet pump laser, with 1st, 2nd and 3rd Stokes all in the visible region of the spectrum. In this paper we report an extension of this technique to infrared generation. Here the main difficulty to contend with is the reduced Raman gain at longer Stokes wavelengths. To overcome this problem we have used a capillary waveguide configuration to enhance the gain in the amplifier. We report the result obtained from this scheme. To summarize the experimental results, we have converted 0.53 μm pump radiation to 3rd Stokes radiation at 1.58 μm with a photon conversion efficiency of 9%. The measured divergence of the 3rd Stokes beam corresponds to twice the diffraction limited value, somewhat better than the beam quality (2.5×diffraction limit) of the pump laser itself. The conversion efficiency is better than that reported by Hartig and Schmidt [2] and comparable to that recently reported by Mannik and Brown [4] (both of whom used a capillary Raman generator alone) despite the fact that our pump power was around seven times smaller. This testifies to the advantage of the oscillator/amplifier configuration.

Since a dye laser of suitable power to act as the pump laser was not available for these measurements we used the second harmonic of a Q-switched Nd:YAG laser. The Nd:YAG laser was deliberately operated with a number of transverse modes, so that the second harmonic beam, with a measured divergence around 2.5 times the diffraction limit, simu-

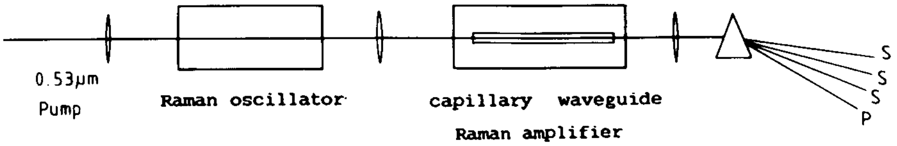

Fig. 1. Schematic arrangement of Raman oscillator/waveguide amplifier.

lated the beam quality of typical high power dye lasers, but the power (corresponding to a 20 mJ pulse of 22 ns duration) was somewhat lower.

The arrangement of Raman oscillator and amplifier is shown in fig. 1. This scheme is simple to use since the Stokes radiation generated in the oscillator is automatically aligned and temporally overlapped with the pump radiation as it enters the amplifier stage. A similar scheme has been found to give very efficient operation where performance was optimised for 1st Stokes generation [8]. For the scheme in fig. 1, the requirements are that 1st, 2nd and 3rd Stokes radiation be generated in the oscillator but without causing too great a depletion of the pump since this needs to be preserved for the amplifier stage. Suitable conditions for this to be achieved were found with a 10 atmosphere gas pressure and the pump beam focussed to a spot radius of ~ 240 μm at the centre of the 50 cm oscillator cell. The low gas pressure enhanced the production of higher order Stokes via four-wave mixing. With these conditions the generated energies from the oscillator were 2.2 mJ (S_1), 0.45 mJ (S_2), 45 μJ (S_3) with ~ 9 mJ of pump radiation still remaining.

With our limited power we found it beneficial to enhance the gain in the amplifier stage by using a capillary waveguide. When an amplifier stage without waveguide was used, efficient generation of second Stokes (S_2) could be achieved (11% quantum efficiency) but the best efficiency for S_3 generation after testing various focusing arrangements and gas pressures in the amplifier, was $\sim 1.5\%$. This was increased by a factor of six when a capillary waveguide was used in the amplifier. The waveguide consisted of a fused silica capillary of 70 cm length and 350 μm bore diameter. Ideally a large bore diameter is desired to reduce four-wave mixing processes and thus allow domination by the cascade of Raman processes growing from the seed radiation [5,6]. A large bore in turn implies a long guide length to provide sufficient Raman gain. We have therefore used the maximum convenient length. However, the bore diameter had to be chosen small enough for the Raman gain to allow the cascade to proceed to the point where efficient S_3 generation was reached. A 350 μm diameter was necessary for this. To further suppress the four-wave processes in the amplifier the gas pressure was increased to 30 atm. We estimate, using the analysis below (Appendix) that this capillary was capable of enhancing the Raman gain for our multimode pump by a factor of ~ 4 compared with an unguided amplifier.

To launch the pump beam and S_1, S_2, S_3 beam into the capillary a lens of 20 cm focal length was used. The launch condition was adjusted to maximise the transmission of the pump light, resulting in $\sim 45\%$ transmission. This was significantly less than our estimate for maximum transmission of $\sim 85\%$ (Appendix). It is not clear whether this discrepancy stems from the inadequacy of our rough estimate, or whether, in view of the fact that an exhaustive optimisation was not carried out, a better transmission might be possible with greater care taken. The transmission of the capillary for the S_1 (683 nm), S_2 (954 nm) and S_3 (1.58 μm) inputs was also measured, without any H_2 gas in the capillary, giving respective values of 37%, 45% and $\sim 10\%$. Estimates of the Raman gain for S_1, S_2, S_3 under these pump conditions, using the Raman gain coefficient $G_R = 1.85 \times 10^{-11}/\lambda_s(\mu m)$ m/W [9] indicate that with these seed radiation intensities the cascade of Raman process should proceed to saturation for third Stokes generation. Fig. 2 shows the experimentally measured energies for S_1, S_2, S_3 emerging from the amplifier, as a function of gas pressure. At the maximum pressure of ~ 30 atm, the generated S_3 energy, 0.58 mJ, corresponds to a photon conversion of $\sim 20\%$ if one considers only the pump radiation input to the capillary. The overall photon conversion efficiency to S_3, from the 20 mJ of pump energy originally

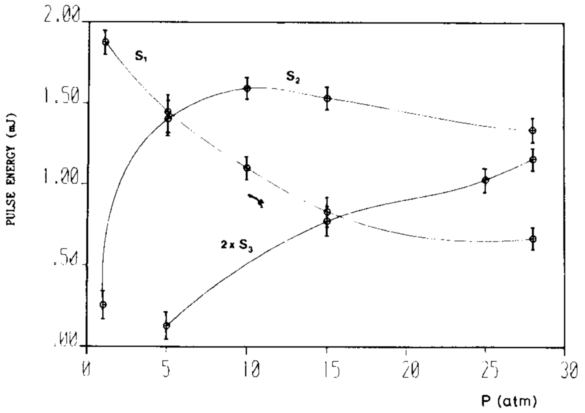

Fig. 2. Pulse energy for S_1 (683 nm), S_2 (954 nm), S_3 (1.58 μm) versus H_2 pressure in the Raman amplifier.

available was ~9%. The beam quality of the S_3 beam was checked by focussing the beam (after separation from S_1, S_2 by a prism), measuring the beam waist size and the divergence from this waist. A detector and scanning slit was used for these measurements. The result showed that the S_3 beam had a smooth profile with a central maximum and none of the ring structure characteristic of four wave mixing, and a beam divergence of 1.8 times the diffraction limit. This was despite the fact that the S_3 beam generated in the oscillator had its origin in a four-wave mixing process. In fact the process of launching into the capillary and of propagation of S_3 within the capillary both have a spatial filtering action which cleaned up the S_3 beam before it underwent Raman amplification

Conclusion. We have shown that the use of a Raman oscillator/amplifier configuration, with its ability to improve beam quality in multiple Stokes generation can be usefully extended to operation in the near infrared region. We have also shown that beam enhancement can be maintained when the amplifier incorporates a capillary waveguide. The gain enhancement provided by the waveguide has allowed us to demonstrate efficient performance with a laser of modest power. With typical commercial pulsed dye lasers, whose powers are greater than the pump used in these experiments, this approach should permit a significant extension of the infrared tuning range.

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Appendix

Capillary waveguide transmission and enhancement of Raman gain for a multitransverse mode pump.

An estimate of the enhancement of Raman gain provided by a capillary waveguide for a TEM_{00} mode pump has been given previously [3]. Here we derive a more general expression to include also the important case of a pump laser which is not diffraction-limited. First we consider the gain enhancement for a TEM_{00} mode pump, where it is assumed that this mode couples perfectly to the EH_{11} mode of the guide when the waist size w_0 at the guide enetrance is equal to $2a/3$ where $2a$ is the guide diameter [10]. The Raman gain exponent G_g (i.e. where $\exp(G_g)$ is the power gain) for a pump power P in a guide of length L is then given by [9]

$$G_g = P g_R L_{eff} / A_{eff} , \quad (1)$$

where

$$L_{eff} = [1 - \exp(-\alpha L)]/\alpha , \quad (2)$$

and

$$\alpha = 0.43 \lambda_p^2 / a^3 , \quad (3)$$

The effective area A_{eff} is taken to be $\pi w_0^2 = 4\pi a^2/9$ [9].

If the effect of Stokes beam diffraction is ignored, then for the unguided TEM_{00} pump focussed to give a confocal parameter b, the Raman gain exponent G_u evaluated along the beam axis is [3],

$$G_u = (4 P g_R / \lambda_p) \tan^{-1}(L/b) . \quad (4)$$

To compare the values of G_g and G_u, i.e. to work out the gain enhancement factor due to the guide, $S \equiv G_g/G_u$, we assume the same spot size in the guided and unguided case. (Generally, since the value of w_0 is such that $L/b \gg 1$, hence $\tan^{-1}(L/b) \sim \pi/2$, this assumption is not restrictive). Thus S is given by

$$S = \frac{9\lambda_p[1 - \exp(-\alpha L)]}{16\pi a^2 \alpha \tan^{-1}(L/b)} . \quad (5)$$

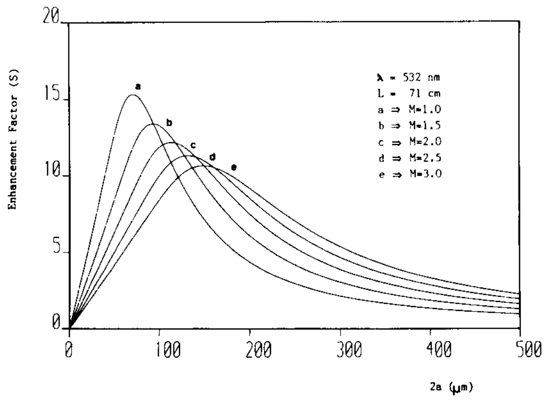

Fig. 3. Enhancement of Raman gain due to the waveguide, as a function of capillary diameter. The parameter M specifies the extent to which the pump beam divergence exceeds the diffraction limit.

For a multimode pump we characterise the beam quality by the factor M by which the pump beam divergence exceeds the diffraction limited value $\lambda/\pi w_0$. Here w_0 refers to the $1/e^2$ intensity radius of the multimode pump beam. The guide loss for such a multimode excitation can be estimated by a simple application of a ray analysis. Such an analysis has been shown [11] to give a remarkably good agreement with the loss calculated for individual guide modes by the full em wave treatment [12]. Here we model the behaviour of the multimode propagation by assuming the rays to travel at an angle to the guide axis given by $M\lambda/2a$ where this represents the divergence of a beam of diameter $2a$. Then the number of reflections per unit guide length and the Fresnel reflection at each bounce can be calculated, leading to an expression for the guide loss per unit length given by $M^2\lambda^2/a^3$. Apart from a numerical factor of the order of unity this agrees for the single mode case ($M=1$) with the exact wave analysis. The effect of the multimode beam is therefore essentially to replace λ by $M\lambda$. To give agreement with the single mode case the loss per unit length is taken to be $0.43M^2\lambda_p^2/a^3$. This simple model gives the guide transmission as $\exp(-M^2\alpha L)$. It is also seen that the value G_u taken by G_u for the unguided case is simply given by eq. (4) with λ_p replaced by $M\lambda_p$ and b ($=2\pi w_0^2/\lambda_p$) replaced by $b'=2\pi w_0^2/M\lambda_p=b/M$. With these changes the factor S' by which the guide enhances the Raman gain for a multiple pump is

$$S'=\frac{G'_g}{G'_u}=\frac{9M\lambda_p[1-\exp(-M^2\alpha L)]}{16\pi a^2 M^2\alpha \tan^{-1}(ML/b)}\,. \qquad (6)$$

Fig. 3 shows plots of S' versus guide diameter for a length $L=70$ cm and $\lambda_p=0.53$ μm, with M as a parameter. For the conditions of our experiment the enhancement is predicted to be ~ 4.

Further refinement of this model can be made by taking account of the Stokes wave diffraction loss for the unguided case. For a TEM_{00} beam eq. (4) becomes [13]

$$G'_u=[(\lambda_s/M\lambda_p)\tan^{-1}(ML/b)] \times [(4Pg_R/\lambda_s)^{1/2}((4Pg_R/\lambda_s)^{1/2}-2)]\,. \qquad (8)$$

and G'_u for an unguided multimode beam becomes

$$G'_u=[(\lambda_s/M\lambda_p]\tan^{-1}(ML/b)] \times [(4Pg_R/\lambda_s)^{1/2}((4Pg_R/\lambda_s)^{1/2}-2)]\,. \qquad (8)$$

For large powers, i.e. where $Pg_R/\lambda_s \gg 1$, eq. (8) reduces to the simpler expression (4).

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