Stimulated Raman Scattering of Picosecond Light Pulses in Hydrogen, Deuterium, and Methane

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Abstract—**Experimental results are presented on stimulated Raman scattering of short pulses of approximately 100 ps duration in H_2, D_2, and CH_4, both in capillary waveguides and in a tight focusing geometry. Experimentally determined thresholds are in good agreement with calculation. Low thresholds (<20 μJ) are observed in CH_4 and preliminary results using a mode-locked dye laser as pump indicate a useful source of tunable short pulse radiation in the near infrared.**

Introduction

TUNABLE near-infrared sources of short pulse duration are being developed using a variety of different approaches. These include near-infrared dye lasers [1], color-center lasers [2], difference frequency generation [3], optical parametric generation [4], and stimulated Raman scattering (SRS) in various media, including optical fiber waveguides [5], caesium vapor [6], and molecular gases. The generation of widely tunable radiation via stimulated Raman scattering in H_2 gas has been extensively investigated using pulse durations of several nanoseconds [7], but relatively little work has been done with short pulses. Some recent work has reported picosecond UV pulse generation [8] and subpicosecond near-infrared generation [9]. In this paper, we report results of a study of SRS in H_2, D_2, and CH_4 using mode-locked pulses of approximately 100 ps duration at 1.06 and 0.53 μm using two different pumping geometries: tight focusing and waveguiding in a hollow capillary [7], [10]. The latter configuration permits a significant threshold reduction [11], [12] to less than 20 μJ for a 0.53 μm pump in CH_4. The capillary was particularly useful in the case of D_2 since it allowed a greater margin to be achieved between the SRS threshold and the gas breakdown. The observed thresholds, both guided and unguided, are found to agree with calculation to within a factor of two. Details of the calculation and the data used are presented. The threshold powers indicate that efficient SRS can be readily achieved with the power levels available from a pulsed mode-locked dye laser driven by a mode-locked Nd:YAG laser [13]. Our preliminary results on Raman scattering using such a dye laser have shown efficient multiple Stokes scattering to the near infrared.

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Threshold Calculation

Under steady-state conditions and for a plane wave pump of intensity I_p, the Stokes wave (angular frequency ω_s) has a power gain exp (G) over a length l of medium, given by $G = (g_R I_p l)$ where [14]

$$g_R = \frac{16\pi^2 c^2 \Delta N}{n_s^2 \hbar \omega_s^3 \Delta\omega_R} \left(\frac{d\sigma}{d\Omega}\right). \quad (1$$

ΔN is the difference in population between initial and final states and $\Delta\omega_R$ is the Raman linewidth (angular frequency, FWHM). The Raman scattering cross section $d\sigma/d\Omega$ is defined here in terms of the ratio of scattered and incident *intensities*. An alternative definition of cross section in terms of the ratio of scattered and incident *photon numbers* is also widely used [15], [16]; however, since most of the tabulated data on cross sections (e.g., [17]) are in terms of the intensity definition, we shall adhere to the latter. The relation between the two cross sections is

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{photons}} = \frac{\omega_p}{\omega_s} \left(\frac{d\sigma}{d\Omega}\right)_{\text{intensity}}.$$

In the infrared region, where $\omega_s/\omega_p \ll 1$, the distinction between these two definitions becomes important. The cross section data we have used for H_2 and CH_4 are based on values given by Schrötter and Klöckner [17]. Since their data are given for pump frequencies $\tilde{\nu}_p'$, other than those $\tilde{\nu}_p$ used in our experiments, we convert their data as follows:

$$\left(\frac{d\sigma}{d\Omega}\right)_{\tilde{\nu}_p} = 5.05 \times 10^{-52}\, \tilde{\nu}_s^4 \cdot \frac{[(\tilde{\nu}_{ig} - \tilde{\nu}_p)^{-1} + (\tilde{\nu}_{ig} + \tilde{\nu}_s)^{-1}]^2}{[(\tilde{\nu}_{ig} - \tilde{\nu}_p')^{-1} + (\tilde{\nu}_{ig} + \tilde{\nu}_s')^{-1}]^2} \sum_{j,\tilde{\nu}_p'} m^2/sr \quad (2$$

where $\Sigma_{j,\tilde{\nu}_p}$ is the "relative normalized differential Raman scattering cross section" for a pump $\tilde{\nu}_p$, as defined and tabulated by Shrötter and Klöckner. The $\tilde{\nu}$ are all expressed in cm^{-1}, and $\tilde{\nu}_s = \tilde{\nu}_p - \nu_R$ is the Stokes frequency corresponding to the pump $\tilde{\nu}_p$. The quantity in square brackets accounts for the dispersion of the Raman susceptibility, and assumes a single dominant intermediate leve

TABLE I
STEADY-STATE RAMAN GAIN g_R AND DEPHASING TIME T_2, BOTH CALCULATED AT 30 atm PRESSURE

Raman Medium	Pump Wavelength	1st Stokes Wavelength	$dq/d\Omega$ m^2/sr	g_R(m/w)	T_2(ps)
H_2 Q(1) Shift 4155cm^{-1}	1.06μm (9,391cm^{-1})	1.91μm (5,236cm^{-1})	1.2x10^{-36}	0.97x10^{-11}	208
	0.53μm (18,782cm^{-1})	0.683μm (14,627cm^{-1})	7.9x10^{-35}	2.76x10^{-11}	
D_2 Q(2) Shift 2987cm^{-1}	1.06μm (9,391cm^{-1})	1.56μm (6,404cm^{-1})	2.0x10^{-36}*	3.7x10^{-12}	150
	0.53μm (18,782cm^{-1})	0.633μm (15,795cm^{-1})	8.0x10^{-36}*	1.0x10^{-11}	
CH_4 Q Shift 2917cm^{-1}	1.06μm (9,391cm^{-1})	1.54μm (6,474cm^{-1})	7.0x10^{-36}	3.3x10^{-12}	16
	0.53μm (18,782cm^{-1})	0.630μm (15,865cm^{-1})	2.7x10^{-34}	8.6x10^{-12}	

* Scaled from the measured value at 694nm reported by Devir [22]

of energy $h\nu_{ig}$ above the ground level. For H_2 and D_2, we take $\tilde{\nu}_{ig}$ to be 90 000 cm^{-1}, and for CH_4 to be 70 000 cm^{-1} [18].

The Raman linewidth $\Delta\omega_R$ is calculated from published data as follows. For CH_4, $\Delta\tilde{\nu}_R = 0.32 + 0.012p$ cm^{-1} where p is in atmospheres [19]; for H_2 and D_2 [$Q(1)$ and $Q(2)$ transitions, respectively] since we operate at pressures where the linewidth is proportional to pressure (> 10 atmospheres) we use the line broadening coefficients of 51 MHz/atm [16], and 70 MHz/atm [20], respectively. The value of ΔN in (1) is taken to be N_{tot} (the total molecular number density) in the case of CH_4 where the entire Q branch is unresolved at 30 atmospheres, 0.66 N_{tot} in the case of H_2 ($J = 1$ initial level), and 0.38 N_{tot} in the case of D_2 ($J = 2$, initial level) [21].

The calculated values of g_R corresponding to 30 atmospheres pressure for H_2, D_2, and CH_4 at pump wavelengths of 1.06 and 0.53 μm are given in Table I. Also included is the dephasing time T_2, given by $T_2 = 2/\Delta\omega_R$. Our calculated value of g_R for H_2 agrees with values calculated by Bischel and Black [16].

From the values of T_2 it can be seen that for our ~100 psec pump pulses the SRS process will clearly be transient in H_2 and D_2, and will even be showing some degree of transience in CH_4 (Typically, the pulse duration T_p must exceed ~20 T_2 if the steady value of gain is to be achieved). An analysis of transient Raman scattering for plane wave conditions has been given by a number of authors (see, e.g., [23]). The main result we wish to extract from this analysis is the factor F by which the peak pump intensity must be increased under transient conditions compared to steady-state conditions. This can be calculated, given the value of T_p/T_2 and the value of G which is the gain exponent needed to reach threshold. Following Laubereau and Kaiser [24], we take $G = 25$ as the appropriate condition, leading to a pump to Stokes' conversion efficiency of ~1 percent. Our experimental definition of threshold also corresponds to an observation of 1 percent conversion efficiency. Fig. 1 shows the calculated value of F versus (T_2/T_p) for a Gaussian-shaped pump pulse (T_p is

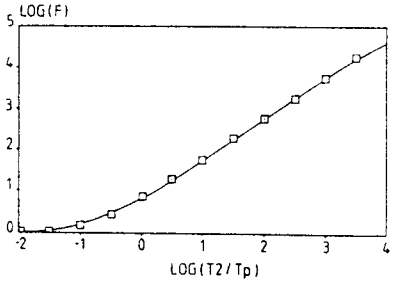

Fig. 1. Factor F by which the peak power is increased relative to the steady state threshold power. T_p is the FWHM of the Gaussian pump pulse. The threshold is defined by the requirement that the Stokes gain is exp 25.

the FWHM), for an assumed $G = 25$. This calculation was based on the analysis of [23]. Next we assume that the threshold power P_t for transient conditions with either the guided or unguided pump is simply the steady-state threshold power P_{ss} multiplied by the factor F.

The steady-state threshold powers are calculated as follows.

1) For the unguided case, assuming the pump beam to be Gaussian, with confocal parameter b, focused at the center of a Raman medium of length l, we have, from Cotter *et al.* [25]

$$P_{ss} = \frac{\lambda_s}{4g_R}\left[1 + \left(1 + \frac{G\lambda_p}{\lambda_s \tan^{-1}(l/b)}\right)^{1/2}\right]^2. \quad (3)$$

2) For the capillary waveguide, again assuming a net gain exp G is needed to reach threshold, we have

$$g_R \frac{P_{ss}}{A_{eff}} l_{eff} = G + \alpha_s l \quad (4)$$

l_{eff} is the effective length related to the actual capillary length l by [12].

$$l_{eff} = [1 - \exp(-\alpha_p l)]/\alpha_p \quad (5)$$

α_p being the pump attenuation coefficient, determined empirically from the measured pump transmission through the capillary. Since the Stokes' wave attenuation was not

measured, it is assumed to be given by the theoretical expression derived by Marcatili and Schmeltzer [26] which reduces in the case of fused silica capillary to [11]

$$\alpha_s = 0.43\ \lambda^2{}_s/a^3 \qquad (6)$$

where $2a$ is the capillary diameter.

The effective area A_{eff} of the pump beam is defined in the same way as for optical fiber waveguides [27], [28]. Assuming the pump beam to be in the form of a Gaussian beam of spot size w_0, the effective area is πw_0^2 [28], and this is the expression we use since it is only slightly different from the exact result obtained for the EH_{11} capillary mode [12]. The input beam to the capillary is focused to give $3w_0 = 2a$, for optimum launching [29], and the effective area is therefore $\frac{4}{9}\pi a^2$.

Experimental Arrangement and Results

The Nd:YAG laser used in these experiments consisted of an actively mode-locked and Q-switched Nd:YAG laser, having a long prelase operation [30], followed by a single pulse selector and an Nd:YAG amplifier stage (J. K. Lasers System 2000, AML-series). The TEM_{00} output gave a single pulse energy of ~ 8 mJ at 1.06 μm or ~ 4 mJ at 0.53 μm after doubling in KD*P. Measurement of the 1.06 μm pulse duration by a background-free autocorrelation technique gave typically 100 ps FWHM for an assumed Gaussian temporal shape. Somewhat different pulse durations were also used in some of the measurements. The 0.53 μm pulse duration was assumed to be $\sqrt{2}$ shorter than the 1.06 μm pulse, i.e., 70 ps corresponding to the 100 ps fundamental pulse. A measurement of the bandwidth of the 0.53 μm pulse, taken with the assumed 70 ps duration yielded a time bandwidth product of less than 0.6, confirming the essentially bandwidth-limited performance which results from the long prelase operation of oscillator [30].

Raman scattering measurements have been made with various cell lengths. Results given here are for 50 cm and 1 m lengths. Capillaries of two lengths have been used (40 and 78 cm) each of 200 μm internal diameter. The capillary was in the form of thin-walled tube of fused silica supported inside a thick-walled capillary. This was either inserted in the high-pressure cell (for the 40 cm length) or (for the 78 cm length) in a V-groove to maintain straightness [11], [12] with high-pressure attachments fitted to the ends.

The results of the threshold measurements are shown in Tables II and III, for the unguided and guided cases, respectively, with calculated values for comparison. All measurements were at 30 atm pressure. In calculating the threshold energy E_T, the relationship $E_T = 1.06\ T_p P_T$ was used since a Gaussian shape was assumed.

A comparison of measured and calculated thresholds shows agreement to within a factor of two, which, considering the number of uncertainties in both measurement and calculation, is reasonable. A feature of the results is that most of the measured energies at 1.06 μm are greater than

TABLE II
Threshold Energies Using Tight Focusing

	Experimental Threshold Energy		Calculated Threshold Energy	
	0.53μm	1.06μm	0.53μm	1.06μm
H_2	300μJ (f/b=20, f=1m, Tp = 80ps)	2.3mJ (f/b=20, f=1m, Tp = 120ps)	190μJ	1.21mJ
H_2	200μJ (f/b=8, f=0.5m, Tp = 70ps)	1.6mJ (f/b=4, f=0.5m, Tp = 100ps)	190μJ	1.25mJ
D_2	430μJ (f/b=8, f=0.5m, Tp = 70ps)	2.8mJ * (f/b=1.3, f=1m, Tp = 100ps)	390μJ	3.3mJ
CH_4	125μJ (f/b=20, f=1m, Tp = 80ps)	950μJ (f/b=20, f=1m, Tp = 120ps)	84μJ	540μJ
CH_4	61μJ (f/b=8, f=0.5m, Tp = 70ps)	490μJ (f/b=4, f=1m, Tp = 100ps)	80μJ	550μJ

* For a 0.5m cell, with up to 6.5mJ input energy the SRS threshold (predicted 2.4mJ) could not be reached due to the onset of gas breakdown. Gas breakdown was avoided by using a 1m cell and lower pump intensity.

predicted whereas the reverse is true at 0.53 μm. A comparison of Table II and III shows that the use of capillaries gives threshold reductions of up to an order of magnitude. An exception to the otherwise reasonable agreement is that in the case of D_2, with an unguided pump in a 50 cm cell at 1.06 μm, threshold could not be reached at the maximum input energy of 6.5 mJ. This discrepancy was due to the onset of gas breakdown. When a 1 m cell was used, the lower pump intensity allowed SRS threshold to be reached before breakdown and the observed SRS threshold of 2.8 mJ agrees well with calculation. A similar problem with gas breakdown occurred with a single longitudinal mode pump pulse of ~ 30 ns duration unguided in a 1 m cell. SRS threshold in D_2 was not reached even with an input energy of 140 mJ whereas the predicted threshold was 75 mJ. On the other hand, using a capillary waveguide (40 cm, $2a = 200$ μm) gave a threshold energy of 16 mJ, in good agreement with the predicted 11 mJ.

The variation of threshold with pressure has also been examined and agreement with calculation is again reasonable. Fig. 2 shows results obtained for D_2 in a 0.4 m capillary, with 80 percent transmission for the 0.53 μm pump. The threshold variation with pressure is due to the fact that T_2 is inversely proportional to pressure over the range of pressures covered, while the steady-state gain coefficient g_R remain constant. In addition to threshold measurements some measurements of conversion efficiency to 1st Stokes have been made for all three gases. Typically energy conversion efficiencies of around 10 percent are achieved at pump levels of twice the threshold value, with best results obtained from D_2 where conversion efficiencies of 17 and 27 percent were obtained with 1.06 and 0.53 μm, respectively. A detailed study of conversion efficiencies achieved using both guided and unguided geometries will be presented in a forthcoming paper with emphasis on high efficiency schemes such as oscillator/amplifier configurations.

TABLE III
THRESHOLD ENERGIES USING A CAPILLARY WAVEGUIDE

	Experimental Threshold Energy		Calculated Threshold Energy	
	0.53μm	1.06μm	0.53μm	1.06μm
H_2	33μJ ℓ=0.78m; ℓ_{eff}=0.48m Tp=53ps	140μJ ℓ=0.78m; ℓ_{eff}=0.42m Tp=110ps	36μJ	132μJ
H_2	28μJ ℓ=0.4m; ℓ_{eff}=0.35m Tp=70ps	290μJ ℓ=0.4m; ℓ_{eff}=0.26m Tp=100ps	47μJ	186μJ
D_2	91μJ ℓ=0.4m; ℓ_{eff}=0.35m Tp=70ps	350μJ ℓ=0.4m; ℓ_{eff}=0.34m Tp=100ps	99μJ	490μJ
CH_4	14μJ ℓ=0.78m; ℓ_{eff}=0.57m Tp=67ps	93μJ ℓ=0.78m; ℓ_{eff}=0.38m Tp=110ps	13μJ	66μJ
CH_4	12μJ ℓ=0.4m; ℓ_{eff}=0.3m Tp=70ps	160μJ ℓ=0.4m; ℓ_{eff}=0.26m Tp=100ps	24μJ	86μJ

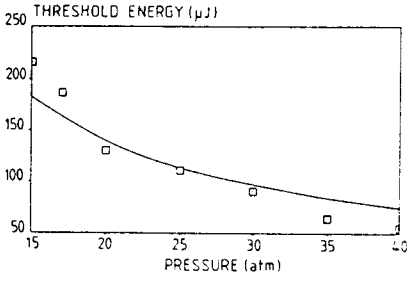

Fig. 2. Threshold energy versus pressure for D_2 in a capillary waveguide (40 cm, 200 μm core) using 0.53 μm pump pulses of 70 ps duration. The curve is calculated from theory.

The low threshold energy requirement, particularly in methane suggest that SRS using a mode-locked dye laser as pump could provide a widely tunable source of short pulses in the near-infrared, and that this could prove a more convenient route to infrared generation than some of the other approaches being pursued. Mode-locked dye lasers with pulse energies in the millijoule region have been reported [13], which as our results show, is sufficient to exceed SRS threshold by at least an order of magnitude, or two orders if a capillary guide is used. Under these conditions, efficient multiple Stokes generation can be anticipated, as obtained in the steady-state regime (see, e.g., [7]). We have made some preliminary measurements of SRS performance using a synchronously pumped mode-locked dye laser similar to that described by Wokaun *et al.* [13]. The dye laser was pumped by the frequency doubled train of pulses from an actively mode-locked and *Q*-switched Nd:YAG oscillator. A single pulse from the Nd:YAG oscillator was selected, amplified in a Nd:YAG amplifier, frequency doubled (~4 mJ at 0.53 μm) and then used to pump a dye amplifier. In this way a single dye laser pulse was produced and under the conditions of our experiments using Rhodamine 6G dye had the following characteristics—pulse duration 50–70 ps, pulse energy up to 700 μJ at the peak of the gain curve (~560 nm), with bandwidth-limited performance. (This laser system was kindly made available to us by JK Lasers, and the assistance of K. A. Ure in using this laser is gratefully acknowledged).

The dye laser beam was tightly focused ($l/b \sim 10$) into a high pressure cell of 35 cm length containing CH_4. The observed threshold energies were 120 μJ at 30 atm, 170 μJ at 20 atm, and 200 μJ at 15 atm. These results are again in good agreement with calculated values. At 30 atm pressure it is found that at the maximum input energy of 640 μJ, i.e., about five times above threshold, the energy conversion efficiencies to first Stokes (~670 nm), second Stokes (~830 nm) and third Stokes (~1.1 μm) were respectively 10, 10, and 1 percent.

CONCLUSIONS

Measurements of stimulated Raman scattering thresholds in H_2, D_2, and CH_4, using mode-locked pulses of 50–100 ps duration have been made. The results agree well with calculations, despite the fact that the calculation involves approximations in extending the analysis of transient Raman scattering to the nonplanewave situation. The low threshold energy requirements, and thus the ability to pump at intensities well above threshold have led to good conversion efficiencies even to third Stokes using a mode-locked dye laser as pump. This approach can now provide a convenient source of tunable short pulses in the near-infrared.

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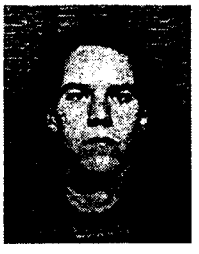

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