Ultrabroad Bandwidth Multiline Stimulated Raman Scattering

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Introduction

Multiple high Stokes and antiStokes orders are frequently observed in stimulated Raman scattering (SRS). In the vast majority of cases the higher orders are the result of parametric 4-wave processes occuring under phase matched conditions. This phase matching requires that the various orders are generated at widely differing angles and the resulting multifrequency beam is not focussable to a single spot. The much more interesting case is where higher orders are generated with a phase mismatch but are collinear. Such a beam may have application to laser fusion [1, 2]. The rotational transitions in gases have been extensively used in Raman amplification and, recently, multiple components of comparable amplitude spanning a broad bandwidth have been generated using both H_2 and N_2 gas [3, 4]. Previously, it has been thought that it would not be possible to switch on bandwidth on a timescale much shorter than the T_2 time of the medium and that this delay would severely limit the usefulness of the generated spectrum [1, 2]. Experiment has tended to support this conclusion [4].

Multiseed SRS with identical pump and 1st Stokes pulse envelopes (symmetric bichromatic pumping) has not, to our knowledge, been investigated in the transient regime where turn on effects are important. It is precisely in this regime that we report the generation of more than 40 Stokes orders having intensity greater than 10% of the peak [5]. In addition to this, we demonstrate that, with an appropriate choice of experimental parameters, ultrabroad bandwidths may be turned on in a time much less than the T_2 time of the Raman medium [5]. The central motivation for this research is the optimisation of collisional absorption in high energy laser-target coupling experiments. Specifically, we are investigating powerful high bandwidth sources which may effectively raise thresholds for the competing parametric backscattering processes [1]. The benefits of increased bandwidth are multifold and may improve the efficiency of the laser fusion process and significantly increase the number of alternative experimental configurations [4]. Since the underlying mechanisms involved in this work are fundamental, there may be a much wider range of applications.

Model and Results

To model SRS we have expanded the total electric field in terms of constituent plane waves (the pump and Raman sidebands). Each wave has a complex envelope varying in both longitudinal space and time and has a carrier frequency which is given by the pump frequency plus an integer times the Stokes frequency. This integer may be positive or negative and is referred to here as the component number. To study the essential physics and scaling laws of the multiline generation process, we have recast the standard equations which describe multiwave interactions [6] in terms of dimensionless variables [5]. The amplitude of each line has been normalised to the peak value of the the pump pulse and the time scale has been scaled to its width, t_p. Propagation distances, z, have been scaled so that one deals directly with the gain-length product of the interaction, $Z = gIz$, where I is the peak input intensity of the pump pulse. We consider symmetric bichromatic pumping as the initial condition at $Z = 0$. In this case the input pulses are synchronous, collinear, coherent and have equal amplitude and pulse length. All other components are set initially to zero.

Results are presented for stimulated rotational Raman scattering in H_2 gas where the Stokes frequency is 587/cm. The pump beam considered is a second harmonic of Nd:YAG which has frequency 18900/cm. We choose the steady-state Raman gain coefficient to be unity, $gI = 1$/cm (g=0.5cm/GW and I=2GW/cm^2, for example), so that one can interpret Z as simply the cell length in units of cm.

It is instructive to firstly consider the multiline generation process without transient effects. Results for this case, valid for relatively long input pulses, are shown in figure 1. In part (a) the output spectrum generated from two input beams (pump and 1st Stokes – component numbers 0 an 1) is shown as a function of the gain-length product. For higher Z the multiline interaction becomes saturated and the spectrum remains unchanged. In figure 1(b) the effect of high gas pressure is demonstrated. Here, approximately the same output bandwidth is generated but it is shifted down to lower frequencies.

In steady-state calculations, such as thise shown in figure 1, cw or quasi-cw variables are assumed. However, when the durations of the input pump and Stokes pulses approach the dephasing time of the polarisation grating, T_2, transient effects can no longer be neglected. We now report on more general considerations involving multiline generation with pulses of input light and allowing for medium dynamics by solving a dynamic equation for the polarisation wave.

Figure 2 shows the output generated after propagation through a gas cell of length

$z = 200$cm. The intensity of the output pulse at each Raman frequency has been integrated over time to show the generated power power. To demonstrate the magnitude of this bandwidth one can calculate that the visible section of the spectrum only spans around 18 Stokes shifts.

In figure 3 the results from a large number of simulations are summarised to outline the role of dispersion. For each value of t_p/T_2 considered, a suitably large gain-length product is specified to minimise z-dependent variation in the output bandwidth. In part (a) the variation of the 10% bandwidth of the time-integrated spectra is shown. This definition of bandwidth reflects the width of the output spectrum within which the integrated-intensities of the lines are greater than 10% of the peak value. For $t_p/T_2 = 1, 8$ and low dispersion the bandwidths are slightly lower than their maximum values. It is possible that this effect is related to the gain suppression of phase-matched interactions which has been quantified previously in models of three-wave coupling [6]. Also for low dispersion, one may observe an enhancement of the output bandwidth due to transient effects. This may be attributed to a longer interaction length which results from the longevity of the polarisation grating. For high gas pressures dispersion tends to inhibit the generation of ultrabroad bandwidths when transient effects are strong. Thus, for fixed steady-state gain, there exists an optimal gas pressure for maximising the output bandwidth. As one may expect, instantaneous bandwidths (at particular values of local time) may exceed those of the time-integrated spectrum and can be greater that 50 Stokes shifts.

In figure 3(b) the points in time for maximum (instantaneous) bandwidth are shown. While changes in the (global) parameter of time-integrated bandwidth tend to saturate at large z, the switch on time is more local and remains sensitive to z. The overall trends and level of sensitivity can be interpreted from correlating the results of such a large number of simulations. Generally, with decreasing t_p/T_2 the switch on time is seen to increase. However, the delays shown are still less than the input pulse width, t_p. For $t_p/T_2 = 0.125$

increasing the gas pressure tends to halt the generation of ultrabroad bandwidth. It is thus not surprising that switch on times for these smaller bandwidths do not greatly increase with gas pressure and, instead, stay of the same order. When T_2 is less that t_p ultrabroad bandwidth may be switched on essentially instantaneously or even during the local rise time of the input pulses.

In figure 4 the variation of bandwidth with t_p/T_2 is shown for fixed gas pressure and gain-length product. In the direction of the extreme transient limit, $t_p \ll T_2$, the suppression of ultrabroad bandwidth, that was indicated in Fig. 3(a), is seen to continue while towards the steady-state regime, $t_p \gg T_2$, bandwidth can be maximal. It is also in this latter regime where switch on time can be minimal minimal [5]. Even while t_p/T_2 varies over two orders of magnitude, symmetric bichromatic pumping leads to significant bandwidth which coincides (temporally) with the pumps to within a pulse width.

Summary

In summary, we have demonstrated that in stimulated rotational Raman scattering in H_2 gas, output bandwidths greater that the pump frequency may be generated when symmetric bichromatic pumping is implemented. Previous research has shown that long transients may occur before bandwidths of just several components can switch on and that such transients may limit the usefulness of the generated spectrum. Here we have demonstrated that, with the correct choice of experimental parameters, *ultrabroad* bandwidths may be generated essentially instantaneously. It is expected that the principles underlying these results for linearly polarised light will have consequences in other polarisation geometries. Work is in progress to generalise these results through the systematic inclusion of asymmetries in the bichromatic pumping scheme. Studies with more complex models to determine limiting factors that could suppress the generation of such ultrabroad bandwidth need to be undertaken. The experimental realisation of just a fraction of the predicted bandwidth would be a very significant result.

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Figure Captions

1. Variation of the steady-state multiline spectrum as a function of the gain-length product (Z=0-60). Each Raman line is separated by integer multiples of the Stokes frequency (the component number). (a) Neglecting the effects of dispersion. (b) A significant level of dispersion (approximately 3.6 atmospheres of H_2).

2. Time-integrated intensity of the generated Raman lines ($\log_1 0$ scale). Parameters are $t_p/T_2 = 1$, gI=1/cm ,z=200cm and the gas pressure is 0.66 atmospheres H_2.

3. The characteristics of the time-integrated output as a function of gas pressure for 3 ratios of pulse width to polarisation dephasing time: t_p/T_2 =0.125,1 and 8. (a) 10% bandwidths (in units of Stokes shift). (b) Points in the local time frame for maximum bandwidth (values of t_p/T_2 are as indicated in (a)).

4. Variation of the generated bandwidth with t_p/T_2 for z = 500/cm and 0.66 atm. H_2. 10% bandwidths and magnitude of the pump frequency in units of the Stokes shift.