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Forced Raman scattering in air by a two-frequency laser beam

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Abstract. Stimulated Raman scattering in atmospheric nitrogen has been observed using two copropagating high power laser beams at 1.064 μm and 1.053 μm from a neodynium glass laser. This is due to the near coincidence of the beat frequency at 98.2 cm^{-1} with a rotational line of nitrogen (transition between $J=11$ and $J=13$) at 99 cm^{-1}. The beams were of total intensity 6×10^9 W cm^{-2} and had a common air path of $\approx$30 m. Large amplitude Stokes and anti-Stokes sidebands up to one half the pump beam intensity were generated. The observations are compared with a theoretical treatment of the interaction.

1. Introduction

The non-linear interaction of one or more intense laser beams with a medium generates many other frequencies such as harmonics, sum and difference frequencies and multiples thereof. Four-wave mixing and various forms of stimulated Raman scattering (CARS, CSRS, RIKES etc.) are particular examples of this and are used in spectroscopy and to generate coherent radiation (Reintjes 1985, Druet and Taran 1981, Shen 1984, Eckbreth 1988 and references therein). In a plasma the atomic and molecular transitions are replaced by the plasma resonances. This can be used to generate a relativistic plasma wave by the beating of two laser beams with frequency difference close to the plasma frequency (Rosenbluth and Liu 1972). This is of interest for particle acceleration (Tajima and Dawson 1979) and current drive (Cohen 1984). Indeed, the work presented in this paper was obtained during an experiment to generate a relativistic plasma wave by this process.

In experiments using the Nova laser (Henesian *et al* 1985) a high intensity beam of 10^{11} W cm^{-2} propagated over a long air path of 100 m. A number of sidebands, amplified from spontaneous background, were observed due to rotational transitions in air (Gross *et al* 1980). In this paper we report on the generation of Stokes and anti-Stokes sidebands in nitrogen using two high power laser beams of much lower intensity propagating over a shorter air path. This differs from the Nova experiment since here two laser beams are initially present and the forced oscillation at the beat frequency dominates over the growth from thermal fluctuations. The aim of our experiment was to generate a large amplitude plasma wave by the beat-wave process

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and to detect the plasma wave from the sidebands formed by plasmon-photon scattering. However, the sidebands due to stimulated scattering in atmospheric nitrogen were found to dominate. Both processes are very similar, and whilst extensive theoretical work has been done directly on SRS, in this paper we have used the theoretical description of beat wave plasmon generation (Karttunen and Salomaa 1986, 1987a, b) to the observed stimulated Raman scattering in molecular nitrogen.

In § 2 of this paper we briefly review the theory. The experiment is described in § 3 and a comparison between theory and experiment is presented in § 4. A summary and discussion are given in § 5.

2. Theory

Stimulated Raman scattering has been studied by using a semiclassical treatment where the molecular excitation is calculated quantum mechanically and the electromagnetic fields classically (Raymer *et al* 1979). The electromagnetic field is assumed to consist of many copropagating waves of the same polarisation

$$\boldsymbol{E} = \tfrac{1}{2}\left(\sum_m E_m(z, t)\boldsymbol{u}_x \exp(\mathrm{i}k_m z - \mathrm{i}\omega_m t) + \mathrm{cc}\right) \tag{1}$$

where E_m is the complex amplitude of the mth mode with frequency ω_m and wavenumber k_m and $\boldsymbol{u}_x$ is a unit vector perpendicular to the direction of propagation, z. The frequency difference between successive modes is taken to be close to the transition frequency ω_t so that

$$\omega_{m+1} - \omega_m = \omega_t + \Delta\omega \tag{2}$$

where $\Delta\omega$ is the frequency detuning from exact resonance. The equations for the electromagnetic waves and the complex amplitude Q of the excited molecular wave are (Raymer *et al* 1979, Penzkofer *et al* 1979)

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right) E_m = \mathrm{i}c(\omega_m/\omega_0)\kappa_1[Q^* E_{m+1} \exp(\mathrm{i}\Delta k_m z) + Q E_{m-1} \exp(-\mathrm{i}\Delta k_{m-1} z)] \tag{3}$$

$$\left(\frac{\partial}{\partial t} + \Gamma - \mathrm{i}\Delta\omega\right) Q = \mathrm{i}\kappa_2 \sum_m E_m^* E_{m+1} \exp(\mathrm{i}\Delta k_m z) \tag{4}$$

where

$$\Delta k_m = k_{m+1} - k_m - k_1 + k_0. \tag{5}$$

The coupling constants κ_1 and κ_2 are proportional to the dipole matrix elements between the initial and intermediate states and the intermediate and final states. The damping constant Γ is introduced phenomenologically and is proportional to the halfwidth at half maximum of the spontaneous Raman scattering line. The occupancy of the final state is assumed to be small.

We now use these equations to study the growth of additional modes by Raman scattering of a laser pump beam having one ($m = 1$) or two ($m = 0, 1$) frequency components.

2.1. *Single-frequency pump beam*

We begin by considering the generation of the first Stokes component, i.e. the $m = 0$ mode. The interaction is assumed to be weak so that there is no significant depletion of the pump beam. We also assume that the scattering medium is so dispersive that the wavevectors of the higher order modes are not matched, that is

$$\Delta k_m \gg L^{-1} \qquad \text{for } m \neq 0 \tag{6}$$

where L is the interaction length. The Stokes component grows from noise at exact resonance where the gain is the highest so that $\Delta\omega = 0$.

Equations (3) and (4) are made simpler in the retarded frame

$$z' = z \qquad t' = t - z/c. \tag{7}$$

The time derivative in equation (4) can be neglected if the laser pulse is smooth and its duration much longer than the inverse of the damping constant Γ. Solving for Q and substituting in equation (3) gives

$$\frac{\partial}{\partial z'} E_0 = \frac{\kappa_1 \kappa_2}{\Gamma} |E_1|^2 E_0. \tag{8}$$

In terms of intensities the solution is

$$I_0(z') = I_0(0) \exp(\gamma I_1 z') \tag{9}$$

where

$$\gamma = \frac{4\kappa_1 \kappa_2}{\varepsilon_0 c \Gamma}. \tag{10}$$

The quantity $I_0(0)$ represents the noise intensity at the Stokes frequency which is necessary to initiate the process. A gain $\gamma I_1 L = 25$ is usually required for the first Stokes frequency to grow from noise to a level comparable with the pump beam (Partanen and Shaw 1986).

The requirement that the higher order modes are not matched, equation (6), is a prerequisite for the unhindered growth of the first Stokes sideband. If the wavevectors are matched the first anti-Stokes component would prevent the growth of the excitation wave and consequently of the Stokes component. A more accurate condition than equation (6) for the amount of mismatch required is (Shen and Bloembergen 1965)

$$|\Delta k_2| > (\gamma I_1)^{-1}. \tag{11}$$

In the experiments using the Nova laser a large number of sidebands were observed. The simple plane wave phase matching arguments above are not applicable as the phase front of the beam is far from planar.

2.2. Two frequency pump beam

We now consider the growth of sidebands in a beam which initially contains two frequency components. We assume matching of the wavevectors, $\Delta k_m = 0$, which is a reasonable approximation for plane waves with small frequency differences in a medium with low dispersion. The frequency detuning $\Delta\omega$ is retained so that the two initial frequency components need not be exactly separated by the frequency of the molecular transition. We first consider the situation where the initial fields are not significantly affected by the interaction and remain much stronger than the sidebands. We then consider the case where the interaction is strong and many sidebands are produced, that is cascading occurs.

2.2.1. Weak interaction. Here the driving term in equation (4) can be approximated so that it contains only the two initial components $m=0, 1$. Thus in the steady state case we have

$$Q=\frac{\mathrm{i}\kappa_2}{\Gamma-\mathrm{i}\Delta\omega}E_1E_0^*. \tag{12}$$

Thus for the Stokes sideband we get

$$\frac{\partial}{\partial z'}E_{-1}=\frac{\omega_{-1}}{\omega_0}\frac{\kappa_1\kappa_2}{\Gamma+\mathrm{i}\Delta\omega}E_0^2E_1^*. \tag{13}$$

The solution of this equation expressed in terms of intensities is

$$I_{-1}(z')=\left(\frac{\omega_{-1}}{\omega_0}\right)^2(\tfrac{1}{2}\gamma' I_0 z')^2 I_1 \tag{14}$$

where the gain coefficient γ' is

$$\gamma'=\frac{\gamma\Gamma}{(\Gamma^2+\Delta\omega^2)^{1/2}} \tag{15}$$

where γ is the gain at resonance defined by equation (10). The solution for the anti-Stokes sideband is similar but with I_0 interchanged with I_1 and ω_{-1} replaced by ω_2.

Equation (14) shows that the growth of the Stokes sideband is quadratic from an existing high level whereas for the case of a single frequency pump beam equation (9) shows that the growth is exponential from noise. The gain required for the Stokes sideband to have significant intensity (about 1%) in the case of a single frequency beam is about 25 (equation (11)) whereas with two initial frequency components it is only about 0.1.

2.2.2. Cascading. Analytical solutions can be obtained for the case where the transition energy is small compared to the photon energy and the number of sidebands generated is not excessive, so that $\omega_m \simeq \omega_0$. Using the coordinate transformation equation (7) and introducing the new variable

$$q=-\mathrm{i}\kappa_1 Q \tag{16}$$

equations (3) and (4) take the form

$$\frac{\partial}{\partial z'}E_m=q^*E_{m+1}-qE_{m-1} \tag{17}$$

$$\left(\frac{\partial}{\partial t'}+\Gamma-\mathrm{i}\Delta\omega\right)q=\kappa_1\kappa_2\sum_m E_{m+1}E_m^*. \tag{18}$$

The scaling in equation (16) is useful because the equations are now expressed in terms of the product $\kappa_1\kappa_2$ which is experimentally measurable. It should be noted that equations (17) and (18) are identical to the equations describing the interaction of electromagnetic fields with a plasmon (Karttunen and Salomaa 1986, 1987a, b).

It can be shown that the driving term on the right hand side of equation (18) is a constant (Karttunen and Salomaa 1986), that is

$$\frac{\partial}{\partial z'}\sum_m E_{m+1}E_m^*=0. \tag{19}$$

Equation (18) is therefore independent of z' and has the solution

$$q(t') = \kappa_1\kappa_2 \int_{-\infty}^{t'} \exp[-(\Gamma - i\Delta\omega)(t'-t'')] \sum_m E_{m+1}E_m^* \, dt''. \tag{20}$$

For pump beams with Gaussian pulse shapes

$$E_{0,1}(0, t') = F_{0,1} \exp(-t'^2/2t_p^2) \tag{21}$$

where $F_{0,1}$ are the peak electric fields at $z' = 0$, the excitation wave amplitude is

$$q(t') = \tfrac{1}{2}\sqrt{\pi}\, \kappa_1\kappa_2 F_0^* F_1 t_p \exp(-t'^2/t_p^2)\, W(-\Delta\omega t_p/2 + i\Gamma t_p/2 - it'/t_p) \tag{22}$$

where $W(\zeta) = \exp(-\zeta^2)\,\mathrm{erfc}(-i\zeta)$ is related to the complex error function. Equation (17) can then be solved giving for the intensity of the sideband with mode number m

$$I_m = \tfrac{1}{2}\varepsilon_0 c \exp(-t'^2/t_p^2)\{|F_1|^2 J_{m-1}(r)^2 + |F_0|^2 J_m(r)^2 - 2\,\mathrm{Re}[F_0F_1^* \exp(i\Phi)] J_m(r) J_{m-1}(r)\} \tag{23}$$

where $J_{m-n}(r)$ is the Bessel function of the first kind of order $m - n$ and the argument r is given by $r \exp(i\Phi) = 2qz'$.

3. The two frequency experiment

The experiment was performed using the Vulcan neodymium glass laser at the Rutherford Appleton Laboratory (Dangor *et al* 1986). A dual wavelength oscillator produced the wavelengths 1.064 μm (YAG) and 1.053 μm (YLF) which were then amplified in a common amplifier chain to give a total laser energy $\approx$100 J in 200 ps. The final beam diameter was $\approx$10 cm giving a peak intensity of $\approx 6 \times 10^9$ W cm^{-2}.

The sidebands formed on the pump light were monitored using a grating spectrometer coupled to an Hadland Imacon 675 streak camera fitted with an S1 photocathode and intensifier. The pumps passed through an air path of $\approx$30 m before being attenuated by a factor of about 10^{-4} by reflection from three glass wedges and then focused into the spectrometer.

Strong Raman scattering was observed as shown in figure 1. Both Stokes and anti-Stokes lines are clearly present together with the original pumps. The sidebands were not observed at the output of the laser or when the pumps were desynchronised.

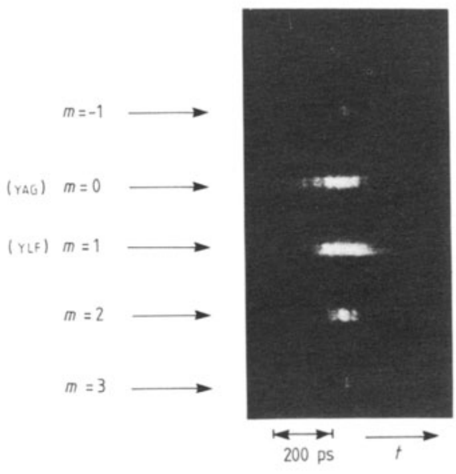

Figure 1. Streak camera record of the pump beams and sidebands (air path $\approx$30 m).

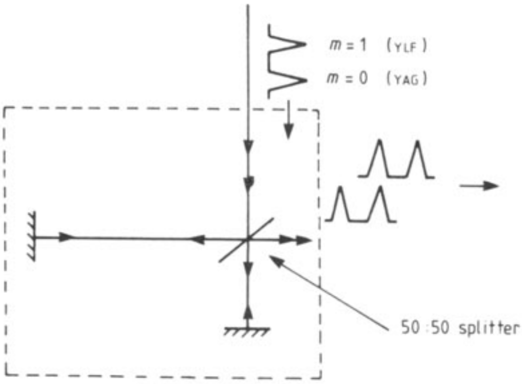

Figure 2. Michelson arrangement used to synchronise the pump beams.

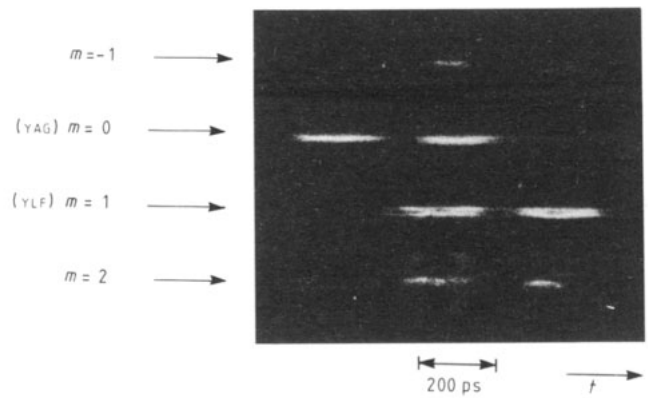

Figure 3. Streak camera record obtained with Michelson arrangement (air path ≃5 cm).

An attempt to reduce the effects of scattering by atmospheric nitrogen was made by desynchronising the beams at the oscillator and recombining them with the Michelson arrangement shown in figure 2. The path difference between the two arms was set equal to the relative delay of the pumps. This reduced the common air path to ≃5 cm. To assist the detection of the weak sidebands the pump light was attenuated by 100 using an ND strip placed over the central region of the entrance slit of the streak camera. An example of the data obtained is shown in figure 3. This shows that the anti-Stokes line also appears with the second YLF pulse and suggests that the low frequency rotational wave has not fully decayed.

4. Modelling

The strong Raman scattering is a result of the near coincidence of the beat frequency $\omega_1 - \omega_0$ at 98.2 cm^{-1} with the rotational transition between $J = 11$ and $J = 13$ in molecular nitrogen ω_r at 99 cm^{-1} (Henesian *et al* 1985). The rotational lines of oxygen are too far from the beat frequency to give any contribution to the sideband generation (Henesian *et al* 1985). The theory presented earlier is applicable as $\omega_r/\omega_0 \simeq 10^{-2}$ and the wave vector mismatch $\Delta k_2 \simeq 10^{-4}$ cm^{-1} is negligible (Eimerl *et al* 1980). The product of the coupling constants $\kappa_2\kappa_2$ needed for the analysis depends on the damping constant and the gain coefficient for a single frequency pump laser beam (equation (10)). This coefficient has been measured in beam propagation experiments with the Nova laser (Henesian *et al* 1985) to be $\gamma = 2.5 \times 10^{-12}$ cm W^{-1} for the rotational transition from $J = 10$ to $J = 12$ at 91 cm^{-1}. The gain coefficient at 99 cm^{-1} is reduced by a factor of 2.2 because of differences in the populations and degeneracies of the different energy levels (Averbakh *et al* 1978). The damping constant is evaluated using $\Gamma = \pi\Delta v$ where $\Delta v = 2.4$ GHz is the measured width of the spontaneous Raman scattering line (Jammu *et al* 1966).

The frequencies of the pump laser beams are known with an accuracy of about 100 GHz. This is much greater than the spontaneous Raman scattering linewidth and so it is impossible *a priori* to define the detuning from exact resonance $\Delta\omega$ and is therefore used as a parameter in the analysis. However, it can be assumed that $\Delta\omega < 20\Gamma$ otherwise the interaction is too weak. The intensities of each of the pump laser beams were not measured but were known approximately from the measured total intensity.

The cascading produced by the two pump laser beams over the long air path of $\simeq 30$ m is described by the analytical solution for Gaussian pulses given by equation (23). The best fit to the experimental results is found for a detuning $\Delta\omega = 10\Gamma$ and is

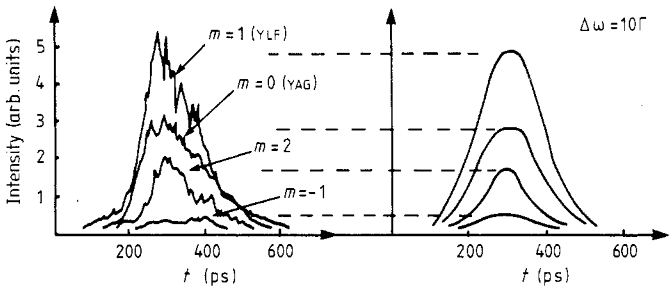

Figure 4. Measured and theoretical profiles of the pumps and sidebands. The total intensity is 6×10^9 W cm^{-2} over a path length of $\simeq 30$ m with the best fit obtained for an initial ratio $I_1/I_0 = 4$. Here $\Gamma = 7.5 \times 10^9$ s^{-1} and the detuning $\Delta\omega = 10\Gamma$.

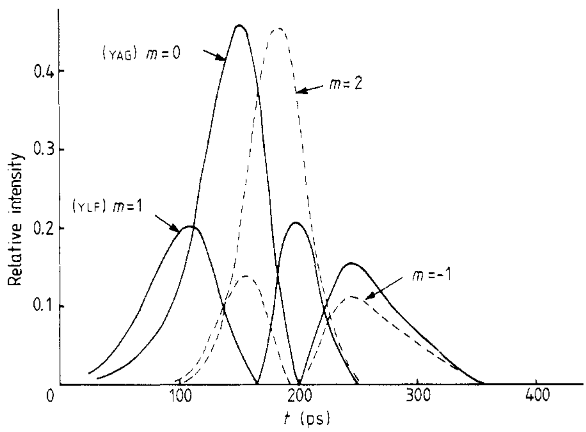

Figure 5. Theoretical profiles of the pumps and sidebands for exact resonance $\Delta\omega = 0$. The other parameters are as in figure 4. Note that the YLF intensity I_1 is reduced to below that of the YAG I_0 as a result of the strong interaction at resonance.

shown in figure 4. For comparison the case of exact resonance is given in figure 5 and shows a much stronger interaction with a much more complicated temporal structure of the modes.

The results obtained with the Michelson arrangement (figure 3) can be used to give an estimate of the damping constant. The ratio of rotational wave amplitude generated at time t_1, immediately after coincidence of the two pump laser pulses, to that at the time t_2, when the last laser pulse appears, is $\simeq \exp[\Gamma(t_2 - t_1)]$. Since the sidebands induced by the last laser pulse are small and so effect the amplitude of the rotational wave only slightly. Thus the relative amplitude of the anti-Stokes sideband is also $\simeq \exp[\Gamma(t_2 - t_1)]$. This gives a value for the damping constant corresponding to $\Delta v =$ 2.6 GHz. This is close to the value assumed in the analysis above of the long path experiment.

5. Summary and discussion

The difference between this experiment and those with the Nova laser (Henesian *et al* 1985) is that there are pump laser beams at two frequencies to drive the molecular transition. Thus the sidebands are driven harder than in the Nova experiments where they are spontaneously amplified from noise. Strong induced Raman scattering occurs because the difference frequency of the pumps is close to the frequency of a molecular transition in nitrogen. The results obtained are in good agreement with the theory presented which is based on the plasma theory model. It should be emphasised that the Raman scattering observations were obtained in an experiment to study the generation of plasma waves induced by the beat-wave process. Thus a measurement of the spectral widths of the incident laser beams and a systematic study of the scattering was not possible given the time available on the large Vulcan laser system.

Sideband generation by stimulated Raman scattering can be used to spectrally broaden the laser driver in fusion experiments. The effect of this is similar to induced spatial interference which has been found to be advantageous. Since the molecular excitation wave is similar to the plasma wave in a beat wave accelerator, a molecular gas medium can be used instead of a plasma. Thus important effects such as cascading and dephasing may be investigated in a simpler experiment not requiring a plasma with a density at resonance.

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