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TITLE: TRANSIENT EFFECTS AND PUMP DEPLETION IN STIMULATED RAMAN SCATTERING

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Transient effects and pump depletion in stimulated Raman scattering

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Abstract

Stimulated rotational Raman scattering in a 300-K multipass cell filled with para-H_2 with a single-mode CO_2-pumped laser is studied using a frequency-narrowed optical parametric oscillator (OPO) as a probe laser at the Stokes frequency for the $S_0(0)$ transition. Amplification and pump depletion are examined as a function of incident pump energy. The pump depletion shows clear evidence of transient behavior.

A theoretical treatment of transient stimulated Raman scattering, including effects of both pump depletion and medium saturation is presented. In a first approximation, diffraction effects are neglected, and only plane-wave interactions are considered. The theoretical results are compared to the experimental pulse shapes.

Introduction

Stimulated Raman scattering (SRS) can be a very efficient means of converting the frequency of laser radiation into desired spectral regions. Since its discovery and first comprehensive studies in the mid-1960's[1] various aspects of this nonlinear process have been studied experimentally and theoretically. In particular, there have been studies of pump depletion both in the steady-state regime ($\Gamma = \infty$)[2,3] and the hypertransient regime ($\Gamma = 0$)[4,5]. Also there have been studies of the transient regime (arbitrary Γ) where pump depletion has been neglected.[6,7] Nevertheless, it appears that to this time there has been no detailed comparison of a realistic theoretical model including pressure broadening to a set of experiments probing different degrees of pump depletion and medium saturation in the transient regime. Such comparison is important to demonstrate the validity of the semiclassical theory of SRS and its various approximate versions, and to assess their potential as analytical tools in the optimal design of Raman converters and oscillators.

The $S_0(0)$ rotational transition in para-H_2 provides an ideal system for studying stimulated rotational Raman scattering.[8,9,10] For scattering of a CO_2 laser near 10 μm, the usual complication of second Stokes generation is alleviated due to the large drop in gain with Stokes frequency. Competition from anti-Stokes generation is avoided by the use of circular polarization for the pump.[11] Superfluorescence, often a complication for stimulated Raman scattering in the visible, is easily avoided in the infrared since there is a large region over which one can measure substantial gains with a probe laser without competition from the growth of spontaneous Raman scattering. This is because the cross section for spontaneous Raman emission falls as ω_s^4 while the Raman gain falls only as ω_s.

In addition to avoiding many of the complications of Raman scattering seen on other systems and in other spectral regions, the molecular parameters for para-H_2 are also well known. The Raman linewidth in para-H_2 has been accurately measured[12] and the polarizability for the $S_0(0)$ transition in hydrogen has been accurately calculated.[13] Thus one has an ideal "two-level" molecule with which to study those effects of Raman scattering which are independent of the particular molecule, such as pump depletion in the nonsteady-state (transient) regime and the effects of laser bandwidth.

The main difficulty in obtaining stimulated rotational Raman scattering in H_2 in the infrared is the relatively low Raman gain in this region of the spectrum, despite the very narrow linewidth of the Raman transition. Since H_2 is a homonuclear molecule, there are no dipole-allowed rotational or vibrational transitions. Thus, the Raman susceptibility is determined solely by the electronic transitions, which lie in the region greater than 90,000 cm^{-1} above the ground state. It is therefore not surprising that the Raman susceptibility and hence the Raman gain is quite low for scattering of a CO_2 laser at ~1000 cm^{-1}.

However, the Raman gain can be increased in several ways. In particular for the J = 0 to J = 2 rotational Raman transition, the use of para-H_2, derived from the blow-off of liquid H_2,[14] eliminates the odd rotational levels. At room temperature this increases the gain by 4.0 over normal H_2 for the $S_0(0)$ transition. One can also cool the para-H_2 to liquid N_2 temperature[9] to decrease the J = 2 level population, increase the J = 0 population and thus further enhance the gain by 2.4. But an even larger increase in gain can come from the use of a multipass cell (MPC),[8,9,10,15] which by consecutive focusing can lead to increases in gain by 30 or more. These enhancements of the gain allow one easily to study the growth of the Stokes beam and the depletion of the pump beam with CO_2 lasers of modest energies (1 - 2 J).

We report a study of stimulated rotational Raman scattering from para-H_2 at room temperature in a MPC, in which the 10R(12) line (10.3 μm, 970.55 cm^{-1}) of a single-mode CO_2 laser is downshifted to 616 cm^{-1} (see Fig. 1). The Stokes probe was a narrow-band (±50 MHz) CdSe OPO. Because this source could easily be observed after passage through the MPC in the absence of Raman gain, it was possible to study the temporal behavior of the Stokes field and the energy conversion from the small signal region to near-complete pump depletion.

Apparatus

The experimental layout is shown schematically in Fig. 2. The basic elements are a CO_2 pump laser, an OPO Stokes source, and a 3.47 m multipass Raman cell containing para-H_2. The system is described in detail in Ref. [16].

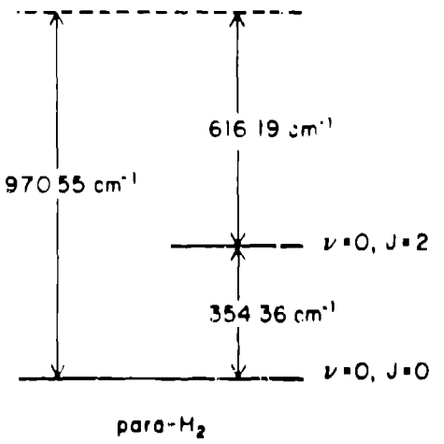

Figure 1. Energy level diagram for para-H_2 showing frequencies involved in Raman scattering studied.

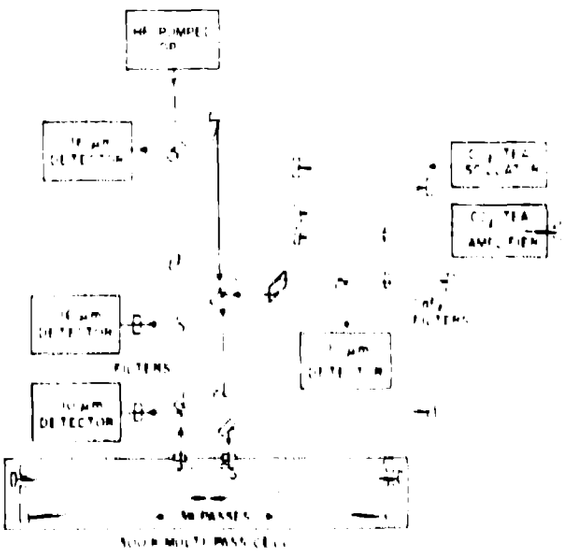

Figure 2. Schematic of experimental setup.

The single longitudinal mode pump is radially symmetric and approximately fundamental Gaussian. Up to 2 J in a temporally smooth pulse of ~25 ns full width at half maximum was available at the input to the MPC. By using calibrated attenuators the energy could be varied without changing the spatial or temporal characteristics.

The CdSe 16-μm source has previously been described.[17,18] For this experiment it was modified to provide better frequency control and more precise tuning to the Stokes frequency. The stability is estimated to be ±50 MHz from shot to shot with longer term drifts of ±150 MHz. The Stokes energy averaged about 9 μJ of the proper (circular) polarization at the input to the MPC.

The pump and Stokes beams were mode-matched[19] to the MPC confocal parameter of 1.36 m. The cell geometry was reentrant after 42 passes but the radiation was coupled out after 39 passes. The transmission of the pump through the cell indicated that the mirror reflectivities were 98.9% at 10 μm. The MPC was filled with 760 torr of para-H_2 from the boil-off of a L-N_2 dewar. Higher pressures of para-H_2 led to collisonal-induced absorption,[8,20] of the infrared beams. At 760 torr of para-H_2 at 300 K we estimate the collisional-induced absorption to be ~1%.

The input 16-μm Stokes beam was monitored with a liquid He-cooled Ge:Cu detector. The Stokes beam exiting from the MPC was separated from the 10-μm pump beam by a dichroic mirror and directed to a photon drag detector. The input and output CO_2 pulses were also observed on photon drag detectors.

Experiment

To study the CO_2 pump depletion, the temporal pulse shapes of the input CO_2 beam, the input Stokes beam, the output CO_2 beam, and the output Stokes beam were all monitored. The timing of the input Stokes was monitored on a liquid N_2 cooled Hg:Cd:Te detector. The other three beams were intense enough to be detected on Ge photon drag detectors. The pulse shapes were recorded on 400-MHz storage oscilloscopes and then digitized, with the resulting data being stored on magnetic disc to allow manipulation and plotting of the data. Figure 3 shows plots of this data for CO_2 pump energies of 1.06 J, 0.76 J, and 0.55 J. The input Stokes pulse shape was monitored prior to taking the data shown in Fig. 3 and a typical oscilloscope trace is shown in Fig. 4. However we did not have enough oscilloscopes to observe the input Stokes pulse while taking the data of Fig. 3.

All three photon drag detectors were calibrated by simultaneously monitoring a single shot of the CO_2 pulse in the absence of any depletion by blocking the 16-μm input to the multipass cell. In this way the relative amplitude and timing of all three pulses could be adjusted. By this technique the shot-to-shot amplitude accuracy was reduced to 10% and the shot-to-shot temporal accuracy to 2 ns. This calibration also allowed us to fold out all the optical losses not associated with the pump depletion so that in the absence of pump depletion the input and output pump pulse shapes are nearly identical.

At an incident energy of 0.55 J the pump depletion is just starting to occur and as the pump energy is increased to 0.76 J the pump depletion becomes more substantial. It is somewhat unexpected that at a pump energy of one Joule, the pump depletion should be so complete over the central 50 ns of the pump pulse. The pulse shapes of Fig. 3 were taken with the entire spatial distribution of the beam, attenuated by CaF_2 plates, focused onto the photon drag detector. Therefore the complete depletion indicates that the spatial wings of the pump, though of lower intensity, are also converting efficiently. The reason for this is not clear but it may be related to the multiple focus geometry. Studies of pump depletion for single focus experiments should help resolve this question.

Possible gain saturation due to depletion of the number of scattering molecules in the pumped volume of the scattering medium must be considered in analysis of the pulse shapes of Fig. 3. We assume that significant pump depletion will occur over 5-10 passes in the cell. We estimate that the number of molecules in the $J = 0$ level of p-H_2 at 1-atm pressure and room temperature in the focal volume of the CO_2 pump laser over 5 passes to be $\sim 10^{21}$ molecules. Since the number of photons involved in a 50% depletion of a 1.5-J CO_2 beam is $\sim 4 \times 10^{18}$, we find that depletion of the number of scattering molecules is not significant.

Theory

Pump depletion for interacting Gaussian beams in the steady state regime has been treated theoretically under the assumption that both the pump and Stokes beams maintain their Gaussian form after significant conversion.[2] Here we present a theoretical analysis of SRS in para hydrogen in the transient regime, which allows a detailed comparison to the corresponding experimental results.

Let E be the total electric field with a slowly varying envelope E_r, $r = p, s$ at the two frequencies and ρ the reduced density matrix of the molecule for the Raman transition (ground state 1, excited state 2)

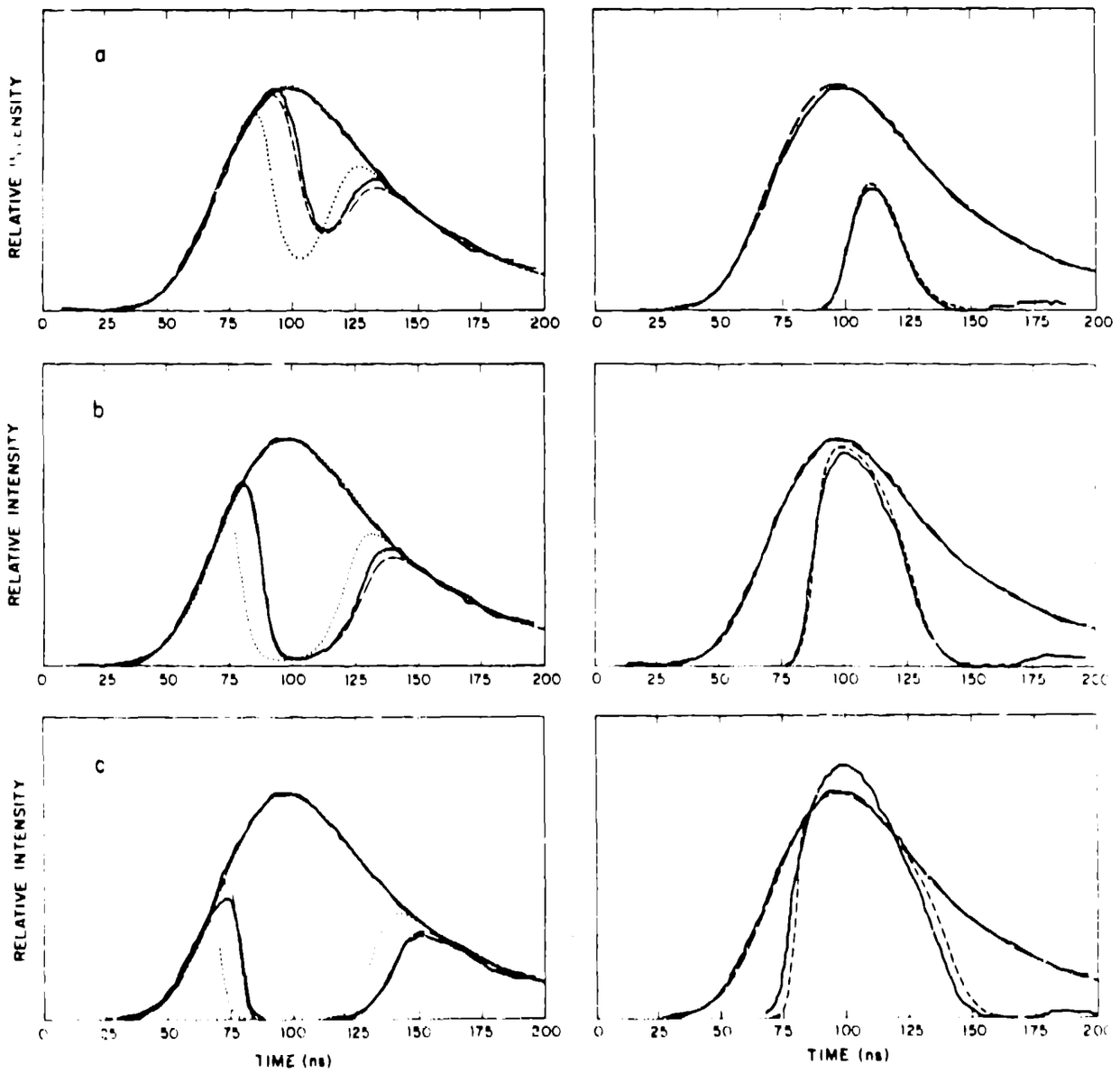

Figure 3. Experimental data compared with theory at input pump energies of (a) 0.55 J, (b) 0.76 J and (c) 1.06 J. In both columns the upper solid lines are the undepleted pump pulses observed experimentally and the associated dashed and dotted lines are analytical expressions for the input pump pulses used in the calculations. On the left the lower solid lines are the experimentally observed depleted pump pulses. The dotted lines are from calculations using steady state theory and the dashed lines correspond to transient theory. On the right the lower solid lines are the experimentally observed output Stokes pulses. The associated dashed lines result from the transient theory.

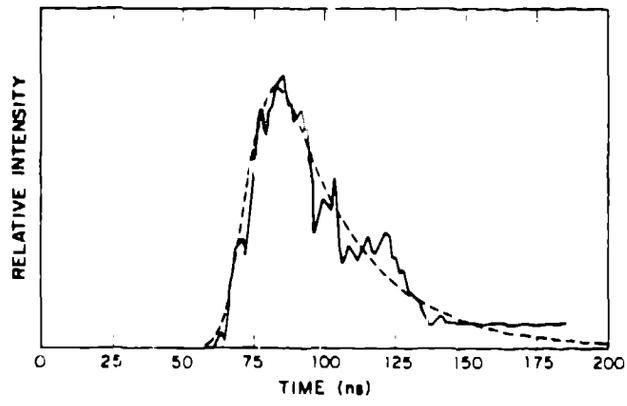

Figure 4. Solid line: a typical input Stokes pulsed observed experimentally. Dashed line: the input Stokes pulse shape used in the calculations.

$$\vec{E} = \sum_{r=p,s} E_r e^{i\Phi_r} \qquad \Phi_r = k_r z - \omega_r t$$

$$\rho = (\rho_{ab}) \qquad a,b = 1,2$$

$$A = ie^{i\Phi_p - i\Phi_s}\rho_{12}$$

$$B = \rho_{11} - \rho_{22}$$

The coupled equations of motion for the density matrix and Maxwell equations for the fields are:[5,21]

$$\frac{\delta}{\delta t} A = -\Gamma A + 2i\Delta (|E_p|^2 + |E_s|^2)A + \kappa E_p^* E_s B \qquad (1)$$

$$\frac{\delta}{\delta t} B = -4 \text{ Re } (\kappa^* E_p E_s^* A) \qquad (2)$$

$$\left(\frac{\delta}{\delta z} + \frac{1}{c}\frac{\delta}{\delta t}\right) E_s = 2\pi k_s N (i\Delta B E_s + \kappa A E_p) \qquad (3)$$

$$\left(\frac{\delta}{\delta z} + \frac{1}{c}\frac{\delta}{\delta t}\right) E_p = 2\pi k_p N (i\Delta B E_p - \kappa^* A^* E_s) \qquad (4)$$

The first term in Eq. (1) represents collisional decay of coherence (homogeneous broadening) with Γ equal to the pressured broadened linewidth (HWHM Lorentzian). The second term in Eq. (1) represents the two-photon Stark shift. κ is the Raman scattering tensor (which reduces to a scalar in our case), and 2Δ is the difference in polarizability between ground state and excited state. For the $S_0(0)$ transition in hydrogen we have[21]

$$\kappa = \sqrt{\frac{2}{15}}\,\gamma, \quad 2\Delta = -\frac{2}{21}\,\gamma \qquad (5)$$

where γ is the anisotropy of the polarizability. At the intensities typical for these experiments the Stark shift term is about two orders of magnitude smaller in absolute value than the broadening term, and will be neglected.

Also the temporal change of the Raman inversion B is at least one order of magnitude smaller than its initial value. Hence the first term in Eqs. (3) and (4) can be eliminated by assuming that the index of refraction is constant and redefining the velocity of light c in the medium. N is the number density of molecules in the Raman cell.

For focused Gaussian beams an effective intensity I_{eff} and effective cavity length ℓ_{eff} can be defined in terms of the power P, confocal parameter b and cavity length ℓ:[1,3]

$$I_{eff} = 4P/b(\lambda_p + \lambda_s)$$

$$\ell_{eff} = b \tan^{-1} (\ell/b) \quad .$$

For comparison with experiment (b) the following numerical values were used in for the model using an enhancement of 31.8 for ℓ_{eff} due to the multiple passes:[16]

$$\Gamma = 0.3204 \text{ rad/ns}$$

$$\ell_{eff} = 52.09 \text{ m}$$

$$P = 5.43 \text{ MW}$$

$$I_{eff} = 5.98 \times 10^7 \text{ W/cm}^2 \quad ,$$

where we note that the effective intensity I_{eff} is well below the typical geometrical mean intensity for Rabi flopping which is 1.46×10^9 W/cm^2.

If the Stark shift and the index of refraction terms in Eqs. (1)-(4) are neglected, it is a self-consistent condition to require that all quantities are real. This is a valid approximation provided the phases do not change too rapidly. The dephasing time is $\Gamma^{-1} = 3.12$ ns, while the pump and seed pulses have typical risetimes of about 50 ns. In this situation the off-diagonal matrix element A can be expected to follow the driving term in Eq. (1) closely with a time lag $\Delta t \leq 3$ ns. Furthermore the effect of detuning can be modeled to a good approximation by replacing Γ in Eq. (1) by $[\Gamma^2 + (\Delta\omega)^2]^{1/2}$ where $\Delta\omega$ is the average detuning.

Dimensionless forms of Eqs. (1), (2), (3) and (4) were solved numerically by a two-dimensional implementation of the trapezoidal rule with fixed step rise. In each of the calculations presented here 200 integration steps for each dimension were found to be sufficient. Calculations were repeated with double number of steps, and no significant improvement of results was found. A single run took about 12 sec of time on a CDC 7600. Input pump and Stokes seed pulses were fitted to an analytical expression. Figures 3 and 4 show experimental pulse shapes for pump and seed pulse and the analytical fits.

Figures 3(a), (b), and (c) show experimental and theoretical depletion profiles together with the corresponding steady state result and the analytical pump pulse profile. A constant detuning parameter $\Delta\omega = 0.33\ \Gamma$ gave best overall fit of the depletion profiles. Fig. 4 shows the timing for the input Stokes beam used in the calculation for experiment (c). For (a) and (b) the pulse was delayed by 25 ns and 10 ns, respectively. The agreement is surprisingly good, given the fact that our model neglects relative phases and treats detuning in a very approximate fashion. A discrepancy occurs in the trailing edge for experiments (a) and (b), where the large local detuning is probably not modelled well enough.

The results show a marked retardation of the depletion profile as compared to the steady state case. The thresholds for the beginning and end of the depletion process are shifted about 10 ns relative to their steady state values, which is about four times the effective dephasing time $(\Gamma + \Delta\omega)^{-1}$. This gives clear evidence of the importance of transient effects in SRS, even when the dephasing times are an order of magnitude smaller than the typical times of variation for the pulses, as in our case.

Using the digitized data, the height of the output Stokes pulse was adjusted to make its total area equal to that of the experimental pump depletion for each shot. In (a) and (b) good agreement is obtained with calculated pulse shapes. In (c) however, the observed Stokes pulse shape is narrower than the calculated shape and, as a result, has a higher peak power than the pump. We believe this is a result of the beam crossing inside the multipass cell seen previously.[9] These crossings occur between different temporal sections of the pulse allowing the generated Stokes pulse to interact with an earlier or later temporal section of the pump pulse. This can then lead to further pump depletion in regions other than that near the peak of the pump pulse.

Summary

We have formulated a theory of transient Raman scattering allowing for a finite decay time of the off-diagonal matrix element. A simplified version of the theory was used to analyze experiments on SRS from para-H. In this version the relative phase between pump and Stokes field was assumed to be zero and Stark shift effects were neglected. Effects of detuning were modelled by a plane wave gain formula with effective gain length and beam intensity. The model gave good agreement between the theoretical and experimental pulse shapes, which show marked deviations from the steady state results.

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