# Simultaneous Observations of Stimulated Raman Scattering and Stimulated Collision-Induced Fluorescence

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We have observed collisional redistribution of laser light scattered by a three-level atom, thallium, in the presence of argon. The two observed spectral components correspond to Raman scattering and collision-induced fluorescence. We have seen the growth of both components from spontaneous to stimulated scattering as the laser intensity is increased. While the gain for the Raman component agrees with theory, the observed gain of the collision-induced fluorescence is over an order of magnitude lower than predicted.

Since the experimental study by Rousseau, Patterson, and Williams,[1] there has been considerable theoretical and experimental interest in the collisional redistribution of near-resonant scattered light.[2-6] For a two-level atom in the presence of collisions, the spectrum of the scattered light at low intensities consists of two components; elastic scattering at the frequency of the incident light, referred to as Rayleigh scattering, and inelastic scattering at the frequency separation of the two levels, which is called collision-induced fluorescence. For a folded three-level system, as shown in Fig. 1, there will also be two emission components near the 2–3 frequency: Raman Stokes scattering at $\omega_s$ as well as collision-induced fluorescence at $\omega_{23}$. Mollow[7] has predicted that both of these latter components can have gain. Therefore the possibility exists for simultaneous generation of stimulated emission at both frequencies, $\omega_s$ and $\omega_{23}$. In an experiment by Wynne and Sorokin,[8] there are indications that these two processes were occurring but the two components could not be well resolved and the population mechanism for the fluorescence was not determined.

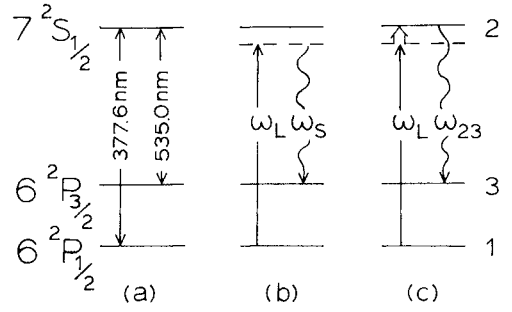


FIG. 1. (a) First three energy levels of thallium. With the laser tuned near the 377.6-nm resonance line, we observed scattered light near the 535.0-nm line. (b) Schematic representation of electron Raman scattering at the Stokes frequency $\omega_s$ from an incident laser at frequency $\omega_L$. (c) Schematic representation of collision-induced fluorescence. Collisions (which in our case were Tl-Ar collisions) transferred Tl atoms from the laser-induced virtual level to level 2, resulting in fluorescence at $\omega_{23}$. When level 3 is initially unpopulated, both of these components can become stimulated.

This Letter reports on the spectrally resolved observation of the simultaneous generation of stimulated Raman scattering (SRS) and stimulated collision-induced fluorescence (SCF). Using a dye laser tuned near the 377.6-nm ($6^1P_{1/2}$-$7^2S_{1/2}$) resonance line of Tl, we observed the growth of both of these components (near the 535.0-nm emission line) from the linear (with pump laser) regime, where the scattering is spontaneous, to the exponential regime, where the scattering becomes stimulated. In addition, we have studied the collisional dependence of the SCF and SRS. Recently SRS in vapors and gases has been used by a number of researchers as an efficient means of down-conversion.[9] This study is an effort to understand the effects of collisions on such stimulated scattering.

For a low-intensity monochromatic laser at frequency $\omega_L$, detuned far from resonance, the spontaneous Raman scattering at $\omega_s$ has a steady-state intensity (in photons $cm^{-3}$ $sec^{-1}$) given by[10]

$$I_R = \tfrac{1}{2} N \gamma_N^{23} (\Omega/\Delta)^2, \tag{1}$$

where $N$ is the number density of scattering atoms (in our case Tl atoms), $\gamma_N^{23}$ is the spontaneous decay rate from level 2 to level 3, $\Delta = \omega_{21} - \omega_L$ is the detuning, $\Omega = \mu E/\hbar$ is the Rabi frequency associated with the incident laser field $E$, and $\mu$ is the dipole matrix element between states 1 and 2. Similarly, the collision-induced fluorescence at $\omega_{23}$ has a steady-state intensity

given by[10]

$$I_F = \tfrac{1}{2} N \gamma_B(\Delta)(\Omega/\Delta)^2 \gamma_N^{23}/\gamma_N, \quad (2)$$

where $\gamma_B(\Delta)$ is the rate of quasielastic collisions (in our case Tl-Ar collisions) which make up the energy difference $\Delta$ needed to produce an atom in level 2 and $\gamma_N = \gamma_N^{21} + \gamma_N^{23}$. We have written the collision rate as a function of the detuning to account for the non-Lorentzian dependence outside the impact regime.[11] Effects of spatial degeneracy have not been included in Eqs. (1) and (2).

Mollow[7] has predicted that both $I_R$ and $I_F$ will have gain when level 3 is initially unpopulated. For laser intensities where $\Omega^2 < \Delta^2$, we expect the gain of the Raman scattering at $\omega_s$ to be

$$g_{SRS} = \frac{\pi}{2} N \frac{c^2}{\omega_{23}^2} \frac{\gamma_N^{23}}{\gamma_R} \frac{\Omega^2}{\Delta^2}, \quad (3)$$

where $\gamma_R$ is a measure of the Raman linewidth (to be discussed later). For the collision-induced fluorescence at $\omega_{23}$, the gain is expected to be

$$g_{SCF} = \frac{\pi}{2} N \frac{c^2}{\omega_{23}^2} \frac{\gamma_B(\Delta)}{[\gamma_D/\pi U]} \frac{\Omega^2}{\Delta^2}. \quad (4)$$

Here $\gamma_D$ is the Doppler linewidth and $U$ is the peak height of the normalized Voight profile,[12] both for the 2–3 transition. One can think of $\gamma_D/\pi U$ as an effective width for the Doppler-plus-collision–broadened transition, 2–3.

Expressions (1) and (2) for the spontaneous emission and Eqs. (3) and (4) for the gain can be used with simple photon propagation equations[13] to solve for the single-pass stimulated outputs (in photons $\sec^{-1}$) of the SRS and SCF collected by a solid angle $\alpha_2$:

$$I_{SRS} = (I_R A \alpha_1/4\pi g_{SRS})[\exp(g_{SRS} L) - 1] + I_R A(\alpha_2 - \alpha_1) L/4\pi \quad (5)$$

and

$$I_{SCF} = (I_F A \alpha_1/4\pi g_{SCF})[\exp(g_{SCF} L) - 1] + I_F A(\alpha_2 - \alpha_1) L/4\pi, \quad (6)$$

where $A$ and $L$ are the area and length of the excitation region, $\alpha_1$ is the solid angle formed by the excitation region, and it is assumed that $\alpha_2 \geq \alpha_1$. In both equations, the second term accounts for the spontaneous emission which exits from the sample outside of the gain region, but which still enters our collection angle $\alpha_2$. We note that Eqs. (5) and (6) predict that both the SRS and SCF will initially have a linear dependence on laser intensity when spontaneous scattering is dominant, but will eventually grow exponentially when the scattering becomes stimulated. Effects of saturation or population depletion have not been included.

The apparatus used in this experiment is similar to that described in detail by Carlsten, Szöke, and Raymer.[2] A tunable dye laser, pumped by an $N_2$ laser, was tuned near the $6^2P_{1/2}$-$7^2S_{1/2}$ resonance line of Tl at 377.6 nm. The dye laser had a pulse duration of 10 nsec, a spectral width of 0.03 nm, and an energy of 50 $\mu$J inside the excitation region. The beam was focused to 330 $\mu$m diam giving a power density of 2 MW/cm$^2$ inside an oven containing 0.1 Torr of Tl vapor and 5 to 80 Torr of Ar buffer gas. The oven input and output windows were put at an angle to avoid back-reflections, which would affect the growth of the stimulated emission. The length of the Tl vapor region was ~2.5 cm. The emission region was then imaged onto the slit of a monochromator of 0.06 nm resolution with an $f/10$ optical system that was capable of viewing the emission at right angles to the laser beam or along the laser beam direction.

When the incident laser is tuned on resonance, only one spectral component, centered at $\omega_{23}$, is observed in the emission. We have studied this emission at 535 nm in both the side and forward directions when the laser was tuned to the 377.6-nm resonance line of Tl. At low laser intensities we observed that both the side and forward emissions were linear in laser intensity. Above ~10 kW/cm$^2$, the forward emission became stimulated and eventually saturated, allowing a maximum photon conversion efficiency of 60%.

In order to study the collisional effects upon this stimulated scattering, we tuned the laser 0.14 nm to the red side of resonance. We were then able to resolve spectrally the Raman emission at $\omega_s$ and the collision-induced fluorescence at $\omega_{23}$. The dependence of these two resolved components on laser intensity is shown in Fig. 2. The Ar pressure for these data was 20 Torr.

In the side direction [Fig. 2(b)] both the Raman scattering $I_R$ and collision-induced fluorescence $I_F$ were linear in laser intensity. By measuring the ratio $I_F/I_R$, we obtained an absolute measure of the collisional redistribution function. From Eqs. (1) and (2), this ratio is expected to be

$$I_F/I_R = \gamma_B(\Delta)/\gamma_N. \quad (7)$$

Using[14] $\gamma_N = 6.7 \times 10^7$ rad $\sec^{-1}$ and our measured value for $I_F/I_R$ we obtain $\gamma_B$(0.14 nm to red) = 7.4

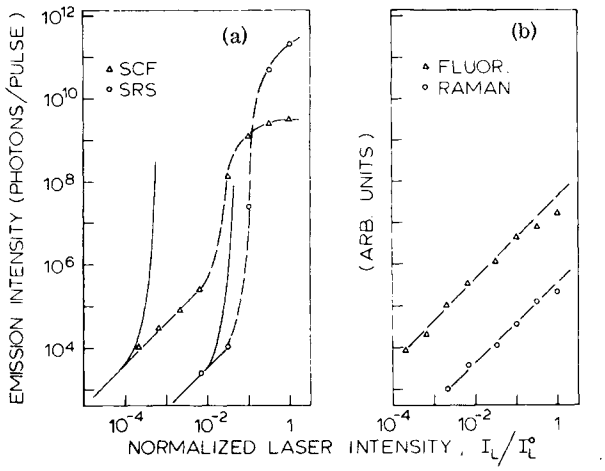


FIG. 2. Dependence of Raman scattering and collision-induced fluorescence on laser intensity when the laser was tuned off resonance (0.14 nm to the red side of the 377.6-nm resonance line). (a) Growth of the stimulated Raman scattering (SRS) and stimulated collision-induced fluorescence (SCF) from spontaneous, linear scattering in the forward direction. The solid curves are theoretical curves obtained from Eqs. (5) and (6) for the SRS and SCF, respectively. (b) Spontaneous, linear scattering in the side direction for comparison. The laser intensity $I_L^0$ was 2 MW/cm$^2$.

$\times 10^9$ rad sec$^{-1}$. This is a factor of 3 larger than the value determined by the emission line-shape measurements of Cheron, Scheps, and Gallagher.[15] According to recent results of Nienhuis and Schuller,[11] we expect the two experiments (collisional redistribution and collisional line broadening) to give the same value for $\gamma_B(\Delta)$ over the experimental range that we studied. The existing discrepancy may be due to the transient nature of our experiment as well as neglect of degeneracy in Eq. (7).

In the forward direction, as in the side direction, the emission is initially linear at low intensities. However, eventually both the Raman emission and the collision-induced fluorescence grow exponentially when the scattering becomes stimulated, and at still higher intensity saturation occurs. Using Eqs. (5) and (6) multiplied by the pulse duration we calculated the expected exponential growth for the Raman scattering and collision-induced fluorescence. The results are shown as the solid curves in Fig. 2(a). The vertical scale was considered a free parameter, but agreed to within a factor of 4 with an absolute-calibration estimate. The laser profile was taken to be uniform in intensity over the 300-$\mu$m diam of the excitation region. It is important to note that in calculating the SRS gain we have used the result of Akhmanov, D'yakov, and Pavlov[16] that, for forward SRS in a dispersionless medium, a broad-band laser has the same gain as a monochromatic laser. This is because the intensity variations of the Stokes emission follow those of the pump laser as the two pulses travel with the same velocity through the medium. Thus for $\gamma_R$ we have used the Raman linewidth (which is predominantly the Doppler width for the 1–3 separation). This results in an SRS gain which is 240 times larger than that calculated by taking $\gamma_R$ equal to the laser linewidth. We see that while the predicted initial exponential growth of the Raman scattering is quite close to our experimental results, the gain for the collision-induced fluorescence is more than an order of magnitude less than predicted by Eq. (6). We do not know the reason for the discrepancy, but problems may arise from use of the steady-state theory as well as from the assumption of a constant spatial profile.

In order to study further the predictions of Eqs. (5) and (6), we measured the dependence of both the SRS and SCF as a function of Ar pressure. Profiles similar to those in Fig. 2(a) were taken at 5, 20, and 80 Torr of Ar buffer gas, at both $\pm 0.14$ nm detuning. While the SRS showed no pressure dependence, the SCF was highly pressure dependent, as expected from the theoretical discussion. In particular, the collision-induced fluorescence was linear in Ar pressure at low intensities, while the high-intensity behavior is described by noting that the SCF gain was roughly linear in Ar pressure. Thus, curves calculated from Eq. (6), but with the gain decreased by an overall factor of 30, were in good agreement with these data. These results show that the experimental dependence of the SRS and SCF on the collision rate was in accord with theory.

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# Optical Rotary Saturation

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The first observation of optical rotary saturation is reported. Bloch-like equations of motion for rotary saturation are solved by a perturbation calculation and also numerically on a computer. In addition to the rotary-saturation resonance, multiphoton transitions are predicted and observed. A "Debye resonance in the rotating frame" naturally emerges from the equations. The case of two traveling waves interacting with a two-level system is shown to be mathematically equivalent to the rotary-saturation equations.

We report the demonstration of the optical analog of NMR rotary saturation.[1] By means of the Stark switching technique of Brewer and Shoemaker[2] we investigate a double-resonance interaction between a two-level system and two fields, a laser traveling-wave field and an electric radio-frequency (rf) standing wave. Consider a two-level system (Fig. 1) acted upon by a long pulse

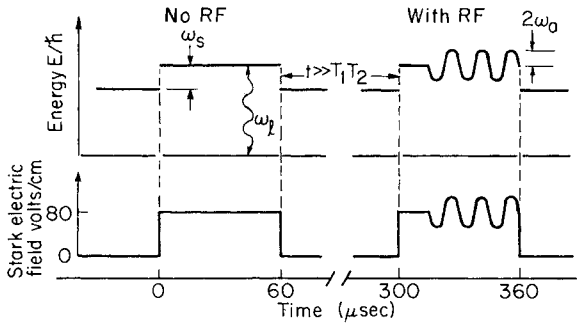


FIG. 1. Stark-voltage pulse sequence (below) and corresponding two-level energies (above). The 80-V/cm Stark pulse for all data (at $^{13}CH_3F$ pressure of 1 mTorr) correspond to $\omega_{s0}/2\pi = 3.7$ MHz.

of electric field. The first-order Stark effect tunes the transition into steady-state resonance equilibrium with the applied laser frequency $\omega_l$ after the pulse is applied, and free-induction decay (FID) occurs immediately after the pulse is switched off. When more than one of a closely spaced set of two-level transitions interacts with the laser, a series of interference FID signal[3] beats will appear which can be Fourier analyzed to yield spectral information from within inhomogeneously broadened lines.[4]

In order to achieve rotary saturation we add to the applied Stark electric field a parallel component oscillating at radio frequency $\Omega$, resulting in a corresponding oscillation of amplitude $2\hbar\omega_a$ in the energy level spacing. The total Hamiltonian can now be written as

$$\mathcal{H} = \hbar \begin{pmatrix} \omega_0/2 + \omega_a \cos\Omega t & -\kappa\mathcal{E}_l \\ -\kappa\mathcal{E}_l & -\omega_0/2 - \omega_a \cos\Omega t \end{pmatrix}, \quad (1)$$

where $\hbar\omega_0$ is the Stark-shifted level spacing in the absence of the rf field, $\mathcal{E}_l$ is the laser field