Phase cross correlation in the coherent Raman process

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The coherent anti-Stokes Raman scattering (CARS) process with two different-color phase-fluctuating fields was studied experimentally and theoretically. It was found that the efficiency of this process depends strongly on the cross correlation between the phases of the driving fields. Depending on the detuning from Raman resonance, the CARS signal can be either enhanced or suppressed with respect to the case of uncorrelated fields.

There have been several experimental and theoretical studies of the influence of optical-field fluctuations in the coherent Raman process, which is an example of a four-wave mixing process with a two-photon resonant intermediate state. It has been shown in these studies that the efficiency of various Raman four-wave mixing processes depends critically on the temporal correlations between the driving fields.[1–4] It should be noted that in all the previous investigations of the coherent Raman process, the two correlated fields were derived from a single laser by use of a beam splitter; thus the fields were exact replicas of each other, having the same center frequency and fluctuation behavior.

In this Letter we show experimentally and theoretically how cross correlation between optical fields with widely different center frequencies (i.e., different colors) affects the coherent anti-Stokes Raman scattering (CARS) process. To obtain such cross-correlated fields, we use stimulated Raman scattering (SRS). The presence of strong cross correlation between the pump laser field and the Stokes field generated by SRS has been proved theoretically and experimentally.[5–11] It has been shown that the complex amplitude of the generated Stokes field temporally follows that of the pump laser field in the limit that the laser bandwidth is much larger than the Raman linewidth. We describe how this cross correlation influences CARS as a function of the Raman linewidth and detuning from the Raman resonance. We clarify the role that pure phase fluctuations play in the CARS process in the absence of any fluctuations of the field amplitude. To achieve this experimentally, we have used broadband, "amplitude-stabilized, chaotic light"[12] as a pump field. This is the light from a cavityless pulsed dye laser that has been amplitude stabilized by passage through saturated amplifiers. A Stokes field that is phase correlated to the pump laser field was generated in a H_2 Raman cell pumped by the amplitude-stabilized chaotic light pulses. These two fields were used as driving fields in the CARS process. By time delaying or advancing the laser beam with respect to the Stokes beam, different degrees of correlation between them were achieved, and the resulting effects on the CARS process were measured.

The experimental setup shown in Fig. 1 was used to study the effects of phase cross correlation between different-color fields on the CARS process. Pulses of amplitude-stabilized chaotic light generated by the dye-laser system[12] were 4 nsec in duration and had a bandwidth of ~10 GHz, a pulse energy of 3 mJ, and a wavelength of 564 nm. The dye-laser beam was divided into two parts by the beam splitter. The stronger beam (about 80% of total energy) was weakly focused with lens L1 down to 0.65 mm inside the Raman generator cell and filled with H_2, producing Stokes light at 737 nm by vibrational SRS. A dichroic mirror and a colored-glass filter were used to block the pump beam that copropagated with the generated Stokes beam. The 20% of the dye laser beam that was deflected by the beam splitter was sent through a variable optical delay. Both this beam and the Stokes beam were collimated and then focused by lens L4 into a second H_2 cell, Cell(2). The phase-matching condition ($2\mathbf{k}_L = \mathbf{k}_S + \mathbf{k}_{AS}$) for generating anti-Stokes light in this H_2 cell was achieved by translating mirror M1 in the direction shown in Fig. 1. To prevent saturation in the CARS process, a neutral-density attenuator was in-

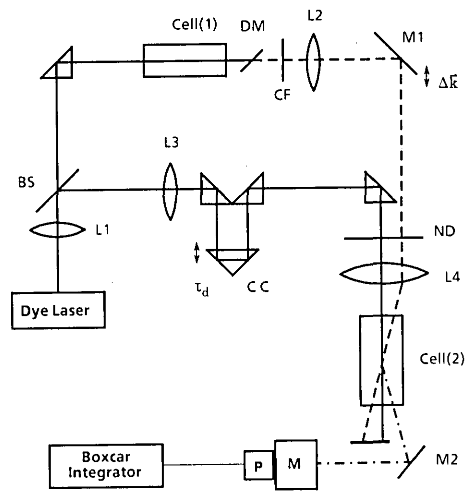

Fig. 1. Experimental setup. BS, beam splitter; DM, dichroic mirror; CF, colored-glass filter; L1–L4, lenses; ND, neutral-density filter; Cell (1), Raman generator cell; Cell (2), CARS cell; M1, M2, mirrors; M, monochromator; P, photodiode; CC, corner cube.

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serted in front of lens L4. The anti-Stokes signal from Cell(2) was filtered with a monochromator, detected with a photodiode, integrated, and then averaged with a microcomputer. The averaged anti-Stokes intensity was measured as a function of delay time τ_d of the dye-laser beam with respect to the generated Stokes beam. A delay time τ_d of zero corresponds to zero optical path difference between the two beam paths that leave the beam splitter and arrive at the four-wave mixing interaction region.

Figure 2(a) shows a set of measurements of average anti-Stokes intensity versus time delay τ_d between the correlated laser and Stokes waves. The curves are normalized to unity for large time delay $|\tau_d|$. Each curve, which contains 600 points, was obtained by averaging 36,000 laser shots. The different curves correspond to different Raman detuning, with curve 1 having zero detuning and curve 6 having a detuning of 5.8 GHz. The laser linewidth (HWHM) was 5.0 ± 0.3 GHz and the Raman linewidth (HWHM) was 0.71 ± 0.03 GHz in the four-wave mixing cell [Cell(2)]. The Raman detuning, which is equal to the difference of the Raman frequencies in the two cells, was varied by changing the pressure of H_2 in the generator cell, while keeping the pressure in the four-wave mixing cell [Cell(2)] constant at 27 atm. Pressure line shift and linewidth data were taken from Ref. 13.

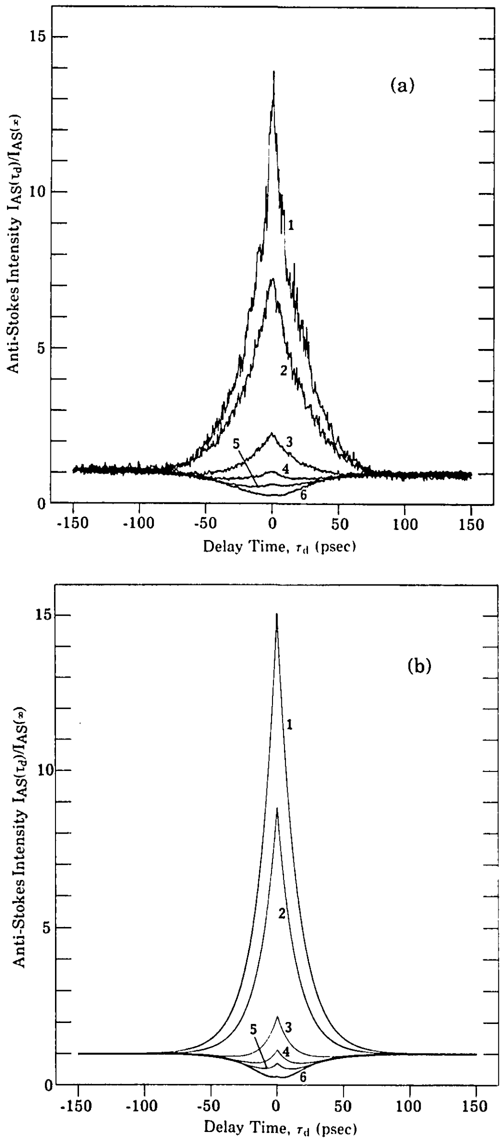

Fig. 2. (a) Measured average anti-Stokes intensity versus the delay time τ_d between laser and Stokes waves. Curves 1, 2, 3, 4, 5, and 6 correspond to the detuning from Raman resonance $\Delta = 0, 0.59, 1.7, 2.6, 3.4$, and 5.8 GHz, respectively. Raman linewidth $\Gamma = 0.71 \pm 0.03$ GHz, and laser bandwidth $\Gamma_L = 5.0 \pm 0.3$ GHz. All curves were normalized to unity for large $|\tau_d|$. (b) Theoretical dependence of anti-Stokes intensity versus the delay time τ_d for phase-diffusion laser light. Curves 1–6 were calculated with the parameters Γ, Γ_L, and Δ the same as the experimental parameters for corresponding curves in (a).

Note that for zero detuning a strong enhancement occurs when the pump and Stokes waves are correlated ($\tau_d = 0$). On the other hand, for large detuning the efficiency actually decreases when the two waves become correlated. For intermediate detuning, two minima in efficiency occur symmetrically around $\tau_d = 0$. It should be pointed out that zero delay time does not correspond to optimum overlap between laser and Stokes pulse envelopes, owing to the transient Raman effect in the generator. Rather, $\tau_d = 0$ corresponds to optimum correlation between the phases of laser and Stokes fields. Since the Stokes pulse was shorter and delayed with respect to the laser pulse, the CARS signal was slightly higher for negative τ_d than for positive τ_d.

The CARS process is described by the coupled equations of motion for the slowly varying complex amplitude of the anti-Stokes field $E_{AS}(t, z)$ and the slowly varying molecular vibration coordinate $Q(t)$;

$$\frac{\partial}{\partial z} E_{AS}(t, z) = -i\kappa_2 E_L(t) Q(t) \exp(-i\Delta\mathbf{k}\cdot\mathbf{z}), \quad (1)$$

$$\frac{\partial}{\partial t} Q(t) = (i\Delta - \Gamma)Q(t) - i\kappa_1{}^* E_L(t) E_S{}^*(t), \quad (2)$$

where $E_L(t)$ is the complex amplitude of the laser field, which is a random process, and $E_S(t)$ is the complex amplitude of the Stokes field from the generator. Since depletion of the laser field and amplification of the Stokes field are negligible, and the angle between the two beams is small, they depend only on the local time variable $t = t_{1ab} - z/c$.[10] Γ is the collisional dephasing rate, and $\Delta = \omega_L - \omega_S - \omega_{21}$ is the Raman detuning, i.e., the mismatch between the difference of pump laser and Stokes frequencies and the molecular vibrational frequency ω_{21} in Cell(2). κ_1 and κ_2 are coupling constants, and the phase mismatch, $\Delta\mathbf{k} = 2\mathbf{k}_L - \mathbf{k}_S - \mathbf{k}_{AS}$, will be taken equal to zero. The solution of Eqs. (1) and (2) is

$$E_{AS}(t, z) = -\kappa_1{}^*\kappa_2 z \int_0^t dt' \exp[(i\Delta - \Gamma)(t - t')] \times E_L(t) E_L(t') E_S{}^*(t'). \quad (3)$$

The average anti-Stokes intensity is given by the ensemble average with respect to the fluctuations of the driving fields, $I_{AS} = \langle E_{AS}(t) E_{AS}{}^*(t)\rangle$. This intensity depends on the correlation function

$$C(t, t', t'') = \langle |E_L(t)|^2 E_L(t') E_S^*(t') E_L^*(t'') E_S(t'') \rangle. \quad (4)$$

As was stated at the beginning of this Letter, the generated Stokes field amplitude $E_S(t)$ approximately follows the laser field amplitude $E_L(t)$ in the case of a laser linewidth much larger than a Raman linewidth. The quantum theory of the Stokes generator shows that the Stokes field entering Cell(2) is related to the laser field in the high-gain limit by

$$E_S(t) = \xi(t - \tau_d) E_L(t - \tau_d), \quad (5)$$

where τ_d is the variable optical delay time and $\xi(t)$ is an operator.[5] It can be shown that, in the high-gain limit, $\xi(t)$ can be treated as a classical random process, with a correlation time approximately equal to the collisional dephasing time in the generator cell [Cell(1)].[14] Thus, during time intervals of length $1/\Gamma_L$, $\xi(t)$ acts approximately as a constant. To evaluate $C(t, t', t'')$, and thereby I_{AS}, we used the phase-diffusion model, which assumes that the laser field amplitude $|E_L(t)|$ is constant and the phase undergoes a diffusionlike process.[15] Although the amplitude-stabilized chaotic light is not strictly speaking a phase-diffusion process, the two processes are similar in the sense that both have constant amplitude and fluctuating phase with uniform distribution.[12] By using the phase-diffusion model, I_{AS} can be evaluated in steady state to give

$$I_{AS}(\tau_d) = |\kappa_1{}^* \kappa_2 z|^2 \frac{I_L{}^2 I_S \gamma}{\Gamma(\Delta^2 + \gamma^2)} \left\{ 1 + \frac{2\Gamma_L}{\gamma(\Delta^2 + \Gamma^2)} \times [(\Gamma\gamma - \Delta^2)\cos \Delta|\tau_d| - 2\Delta(\Gamma + \Gamma_L) \times \sin \Delta|\tau_d|] \exp(-\gamma|\tau_d|) \right\}, \quad (6)$$

where $\gamma = 2\Gamma_L + \Gamma$, Γ_L is the laser linewidth (HWHM in radians per second), and the laser and Stokes intensity are $I_i = |E_i(t)|^2$. It can be shown that when $\tau_d = 0$, the laser linewidth Γ_L completely drops out of Eq. (6). This is because the Stokes field frequency fluctuations compensate for those of the laser field. The result is to drive the molecules with a nearly monochromatic effective field.[16]

By using the experimental values of Γ, Γ_L, and Δ, a set of theoretical curves corresponding to experimental curves 1–6 in Fig. 2(a) was calculated according to the formula given in Eq. (6), and the results are shown in Fig. 2(b). The overall behavior of these curves is in excellent agreement with the data. The slight quantitative discrepancies can be attributed to our using the phase-diffusion model to represent the laser field.

In conclusion, we have shown quantitatively to what extent the CARS process is sensitive to phase cross correlation between the laser and Stokes waves in the absence of intensity fluctuations. Depending on the magnitude of the detuning, either enhancement or suppression of anti-Stokes generation is observed for strongly correlated pump and Stokes waves. According to Eq. (6), and measurements not shown here, the enhancement for zero detuning increases with decreasing ratio of Raman linewidth to laser linewidth, Γ/Γ_L. This is because when the laser and Stokes waves are uncorrelated the fraction of the effective field spectrum that overlaps the Raman line is roughly $\Gamma/2\Gamma_L$, while when they are strongly cross correlated the linewidth of the effective field spectrum is near zero.

The results of this study can also be viewed as an indirect proof that the phase of the Stokes field produced in the generator cell is strongly correlated to the phase of the pump field when $\Gamma_L > \Gamma$. This has been demonstrated in steady state in the absence of any significant intensity fluctuations, a condition not previously accessible experimentally.

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