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Quantitative characteristics of pressure-induced four-wave mixing signals observed with cw laser beams

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We report observation of a pressure-induced extra resonance in four-wave mixing (PIER4 signal) using single-mode continuous-wave dye lasers. The narrow laser linewidth permits for the first time a quantitative verification of the frequency, intensity, and pressure dependence of the PIER4 signal predicted by the theory of the nonlinear susceptibility $\chi^{(3)}$.

We recently reported the first observation[1] of a pressure-induced extra resonance in four-wave mixing (PIER4 signal) which exhibited a Raman-type resonance between two initially unpopulated excited states. Full verification of several theoretical aspects was not possible because of the large linewidths (about 10 GHz) of the pulsed dye lasers used. We have now observed this pressure-induced resonance with single-mode continuous-wave dye lasers. It is the purpose of this Communication to present new data which show the frequency, intensity, and pressure dependence of the PIER4 signal and permit a quantitative comparison with theory.

The experimental configuration has the "folded boxcars" three-dimensional geometry described earlier.[2,3] Three incident laser beams are brought to a common focus in a cell containing Na vapor at a density of less than 10^{15} atoms/cm^3, and helium gas with partial pressures p_{He} between 10 and 100 torr. The three beams travel in the near-forward direction and make (phase-matched) angles of about 4°–6° with each other. A Coherent model 599-03 dye laser, passively stabilized to a width of 50–100 MHz, provides two beams at a fixed frequency ω_1. The frequency ω_1 is chosen 15 GHz below the $3S$-$3P_{1/2}$ Na resonance transition. These two beams travel in the vertical plane and have mutually orthogonal linear polarizations, obtained by means of a beam splitter and a half-wave plate. A third input beam at frequency ω_2, propagating in the horizontal plane is provided by a Coherent model 599-21 dye laser, actively stabilized to a 1-MHz linewidth. This linearly polarized beam is chopped at 400 Hz, and its frequency is scanned continuously through a range of a few GHz, centered 15 GHz below the $3S$-$3P_{3/2}$ resonance line, so that $\omega_2 - \omega_1$ is about 17 cm^{-1}.

The intensity of the three beams at the common focal region is about 100 W/cm^2. This corresponds to a Rabi frequency of 1 GHz for the transition matrix element, much less than the detuning from resonance. A fourth beam is parametrically generated at a frequency $2\omega_1 - \omega_2$, with a wave vector $\vec{k}(\omega_1) + \vec{k}'(\omega_1) - \vec{k}(\omega_2)$ in the horizontal plane. The intensity of this signal beam is proportional to

$$I(2\omega_1 - \omega_2) = |\chi^{(3)}_{xyxy}|^2 I(\omega_1) I'(\omega_1) I(\omega_2) \quad . \tag{1}$$

This signal is detected by spatial (directional) and frequency filtering through a spectrometer with 5-cm^{-1} resolution. Further discrimination is achieved by its polarization perpendicular to that of the chopped beam at ω_2, and by the lock-in detection which eliminates scattered light from the beams at ω_1.

Figure 1 shows three experimental scans at three different partial pressures of helium p_{He}. The nonresonant background signal is the same in all three recordings and serves as a calibration for the strength of the resonant signal. The pressure-induced resonance has a width which varies linearly with p_{He}. Figure 2(b) shows that for $p_{\mathrm{He}} \gtrsim 35$ torr the slope is consistent with a value of 11 MHz/torr. The peak height at resonance approaches a constant value in the limit of high p_{He}, corresponding to four times the nonresonant four-wave-mixing (4WM) signal, as shown in Fig. 2(a). These two observations are consistent with a linear variation of the integrated intensity of the resonance signal with pressure, exhibited in Fig. 2(c). The nonzero intercepts for low values of the buffer gas pressure, $p_{\mathrm{He}} \to 0$, are ascribed to the effect of Na-Na collisions and to hyperfine splittings of the $3P$ states as discussed more fully below. The signal also shows the expected variation with the product of the intensities in each of the three incident beams. The data in Fig. 3 are in agreement with Eq. (1). If the 4WM signal were due to pumping of population into the excited $3P_{1/2,3/2}$ states, the expected signal would vary as a higher power of the intensities. Rayleigh scattering of the input beams at ω_2 and fluorescence produce the noise visible in the recordings of Fig. 1. The observed pressure dependence of the peak height, linewidth, and integrated intensity of the PIER4 signal agree well with the following theoretical expectations.

Since the detuning from one-photon resonances is only 15 GHz, the impact approximation should be valid. In this case second-order perturbation theory gives a coherence density matrix element between

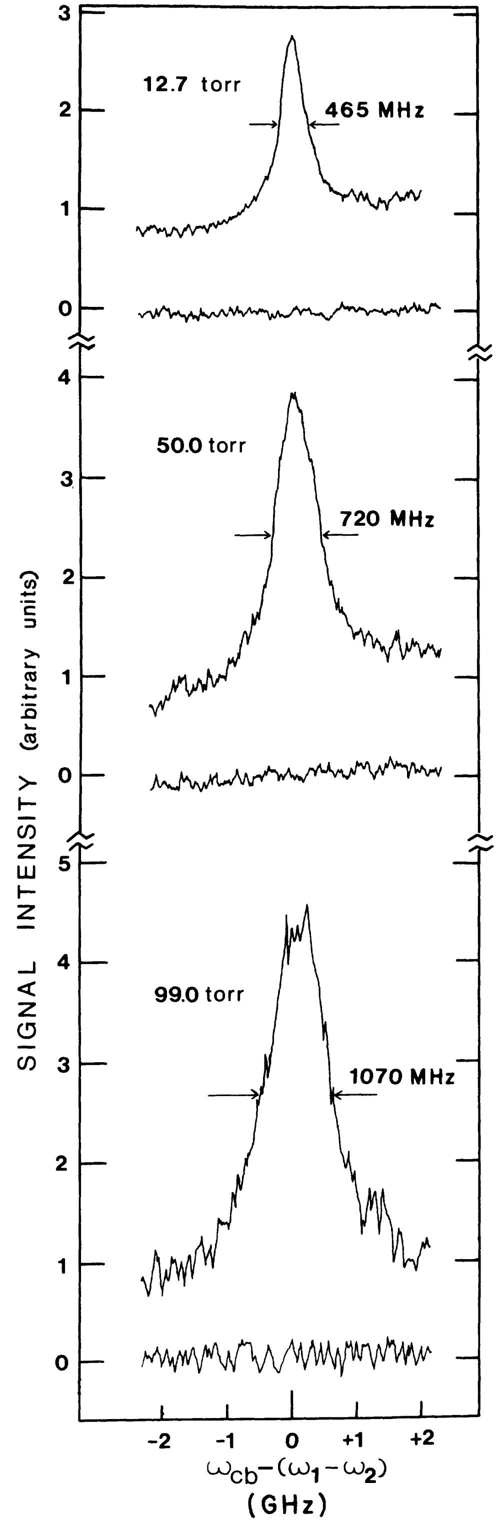

FIG. 1. Experimental scans of the intensity of the 4WM mixing signal at $2\omega_1-\omega_2$, for three buffer gas pressures. The lower scan at each pressure was taken with the input beams at ω_1 blocked. Note the nonresonant signal, independent of pressure, and the pressure-induced resonance.

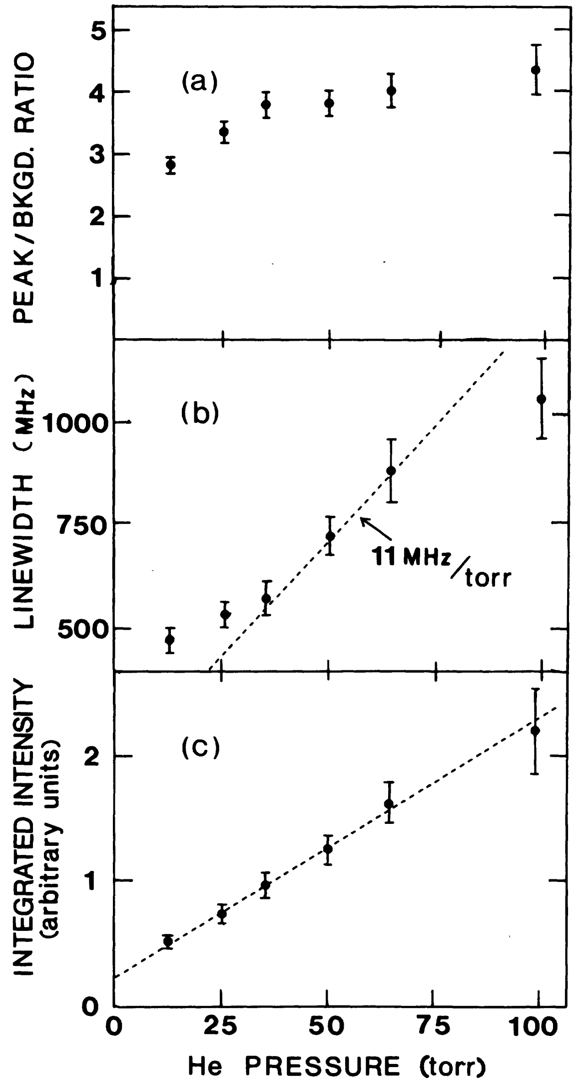

FIG. 2. PIER4 signal characteristics as a function of buffer gas pressure p_{He}. (a) Ratio of peak height to nonresonant signal, (b) linewidth (FWHM), and (c) integrated intensity of resonant signal.

the admixture of the two excited $3P_{1/2,3/2}$ states to the system of N_a atoms in the ground state $|a\rangle$. If the excited states are denoted by $|b\rangle$ and $|c\rangle$, respectively, one finds

$$\rho_{cb}(\omega_1-\omega_2)=\frac{\hbar^{-2}\mu_{ca}\mu_{ab}E_1E_2^*N_a}{(\omega_{ca}-\omega_1-i\Gamma_{ca})(\omega_{ba}-\omega_2+i\Gamma_{ab})}\times\left[1-\frac{i(\Gamma_{ca}+\Gamma_{ab}-\Gamma_{cb})}{\omega_{cb}-(\omega_1-\omega_2)-i\Gamma_{cb}}\right] . \qquad (2)$$

The nonlinear susceptibility $\chi^{(3)}$ is proportional to this expression, which was also derived by Grynberg[4] on the basis of dressed atom states. For spontaneous lifetime broadening, $\Gamma_{ca}+\Gamma_{ab}=\Gamma_{cb}$, and no reso-

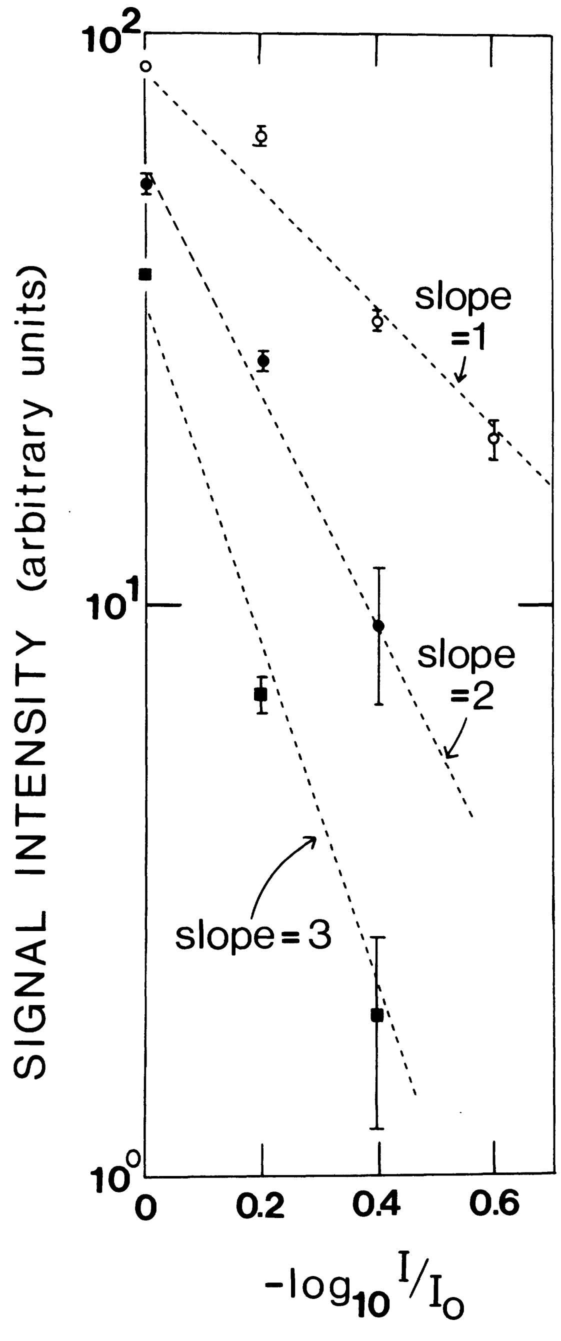

FIG. 3. PIER4 signal intensity as a function of the intensity of the input beams. Open circles: Beam at ω_2 is attenuated. Full circles: The two input beams at ω_1 are attenuated. Square boxes: All three beams are attenuated. The unattenuated points have been shifted arbitrarily in the vertical direction for clarity of presentation.

nance occurs at the energy separation ω_{cb} between the two unpopulated excited states. In the presence of collisions we may write $\Gamma_{ca} = \Gamma_{ca}^{SP} + \gamma_{ca} p_{\text{He}}$, etc. Mossberg *et al.*[5] have recently measured γ_{cb}, and have shown that $\gamma_{cb} \sim \gamma_{ca} \sim \gamma_{ab} = \gamma = 5.5$ MHz/torr [half width at half maximum (HWHM)] at 600 K. Substituting these values into Eq. (2), one finds that the 4WM signal is proportional to

$$|\chi^{(3)}|^2 \propto N_a^2 \left| 1 + \frac{3(\gamma p_{\text{He}})^2 + 2\gamma p_{\text{He}} \Gamma_{cb}^{SP}}{(\omega_{cb} - \omega_1 + \omega_2)^2 + (\Gamma_{cb}^{SP} + \gamma p_{\text{He}})^2} \right| . \tag{3}$$

This expression predicts a resonance width [full width at half maximum (FWHM)] increasing linearly at the rate of 11 MHz/torr; in agreement with the data in Fig. 2. The intercept for $p_{\text{He}} \to 0$ is mainly caused by the fact that levels $|c\rangle$ and $|b\rangle$ are not strictly single energy levels. The hyperfine interaction caused a splitting[6] of the $3p_{1/2}$ of 199 MHz between the $F = 1$ and 2 states. The overall splitting of the $F = 3, 2, 1, 0$ quartet of the $3p_{3/2}$ state is 112 MHz. In addition, the frequency jitter of the dye laser at ω_1 causes an instrumental broadening of about 100 MHz.

At high p_{He}, where the width of the resonance is largely determined by the pressure broadening $\gamma_{cb} p_{\text{He}}$, the peak height becomes independent of pressure and equal to four times the nonresonant background signal. At high pressures the width of the resonance and the integrated intensity are both proportional to p_{He}. These features of Eq. (3) are strikingly confirmed in Fig. 2. Deviations at lower pressure and the nonzero intercepts are due to the influence of Na-Na collisions and the hyperfine splitting of the $3p$ states. Further investigation of the low-pressure regime with better frequency stabilization of the laser at ω_1 would be of interest, and might resolve splitting of the excited states. Obvious extensions would also include the effect of external magnetic fields and other polarization combinations of the incident beams.

In conclusion, observation of PIER4 signals with cw laser beams has confirmed for the first time certain detailed characteristics of the damping terms in the nonlinear susceptibility $\chi^{(3)}$. In particular there is no ambiguity about the correct sign of these terms, contrary to suggestions in the literature.[7] In other situations PIER4 signals may be used to obtain additional information about line-broadening mechanisms and splittings of excited states.

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[7] See, for example, L. A. Carreira, L. P. Goss, and Th. B. Malloy, J. Chem. Phys. 69, 855 (1978), and references quoted therein.