Observation of Collision-Induced Subnatural Zeeman-Coherence Linewidths in the Doppler Limit

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We report the observation of the subnatural linewidth of Zeeman coherences in the presence of buffer gases by use of polarization-nearly-degenerate four-wave mixing. The origin of the ultranarrow linewidth is attributed to the nonlinear response of the ground state of the resonant system.

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Investigations of four-wave mixing processes in gaseous media have provided a rich variety of new spectroscopic results, especially in the presence of buffer gases. Examples are the experimental verification of pressure-induced extra resonances,[1,2] observation of collisional narrowing of spectral line features associated with the population relaxation rate,[3] and the appearance of a coherent dip in homogeneously broadened transitions.[4,5] These features can be interpreted as arising from the noncancellation of quantum-mechanical amplitudes mediated by collisions with ground-state perturbers.[1,6,7]

We report the first observation of an optical resonance having a subnatural linewidth generated by the nonlinear excitation of ground-state Zeeman coherence through the process of polarization-nearly-degenerate four-wave mixing (PNDFWM). Our results are explained by a simple physical model and analyzed by use of a statistical tensor formalism. We show that an upper bound of 10^{-18} cm^2 exists for the spin-depolarization cross section of Na($3S_{1/2}[F=2]$) in the presence of Ne buffer gas as inferred from the observed pressure dependence of the linewidth.

The technique of PNDFWM spectroscopy involves the nonlinear interaction of three input fields denoted $\mathbf{E}_f$, $\mathbf{E}_b$, and $\mathbf{E}_p$ oscillating at frequencies ω, ω, and $\omega+\delta$, respectively, with a set of moving two-level atoms having transition frequency ω_0. $\mathbf{E}_f$ and $\mathbf{E}_b$ are counterpropagating pump fields and $\mathbf{E}_p$ is the probe field which is nearly collinear with $\mathbf{E}_f$. We shall assume that the polarization states of $\mathbf{E}_f$, $\mathbf{E}_b$, and $\mathbf{E}_p$ are $\hat{\mathbf{x}}$, $\hat{\mathbf{x}}$, and $\hat{\mathbf{y}}$, respectively. With the x direction as the quantization axis, $\mathbf{E}_f$ and $\mathbf{E}_b$ can induce electric-dipole–allowed π transitions while $\mathbf{E}_p$ induces σ transitions. Each energy level of the resonant atom is described by a principal quantum number α and a total angular momentum J_α. The physical process giving rise to a four-wave mixing signal in such a system can be described as follows. The interference between $\mathbf{E}_f$ and $\mathbf{E}_p$ generates Zeeman coherences or electric quadrupoles in both the ground and excited levels. Because of the frequency difference between $\mathbf{E}_f$ and $\mathbf{E}_p$, the Zeeman coherence corresponds to a moving grating from which the field $\mathbf{E}_b$ scatters, thus generating a signal field $\mathbf{E}_s$ propagating in the opposite direction to $\mathbf{E}_p$. Conservation of energy imposes the condition that the frequency of the signal field is given by $\omega-\delta$. As a result of the motion of the atoms, two resonances are observed. The first occurs at $\delta=0$ when $\mathbf{E}_f$ and $\mathbf{E}_p$ are resonant with a specific velocity group and leads to the generation of a Zeeman-coherence signal. The width of this resonance is determined by the electric quadrupole dephasing time. The second resonance occurs at $\delta=-2(\omega_0-\omega)$ when $\mathbf{E}_f$ and $\mathbf{E}_s$ are resonant with the same velocity group, and its width is determined by the electric dipole dephasing time. Because of the distinct nature of collisional processes operative on electric dipoles and on quadrupoles, PNDFWM, in principle, permits a simultaneous and independent measurement of phase-interrupting and spin-depolarization collisional cross sections.

In the absence of foreign perturbers, the ground and excited levels are coupled together by the vacuum radiation field. The spontaneous decay rate γ is a measure of the strength of this coupling. Because of it, both quantum levels evolve as a single entity even in the presence of applied radiation fields, and this entity has a unique spectral response, i.e., the bandwidth is determined by γ. In the presence of foreign perturbers, the ground and excited levels experience different collisional interactions which effectively decouples them even in the presence of the vacuum radiation field. In this case, the responses of the Zeeman coherence in the ground and excited levels to the interference of two distinct radiation fields are determined by different decay times. The responses have an additional but common factor due to the absorption and reemission of photons. This factor consists of the sum of two complex Lorentzians, whose widths are determined by the dipole dephasing time. Hence, the PNDFWM signal has contributions arising from both the ground and excited states separately. The spectral responses of the individual contributions are characterized by their respective collisional cross sections. For the signal arising from the ground level, $|J_1\rangle$, the bandwidth is determined by $\gamma_\tau+\Gamma_1$, while that of the excited level, $|J_2\rangle$, is determined by $\gamma+\gamma_\tau+\Gamma_2$. Here Γ_α is the effective collisional depolarization rate for level α and γ_τ is the reciprocal of the transit time for an atom through the laser beam. One notes that for low pressures, $\gamma_\tau+\Gamma_1 << \gamma+\gamma_\tau+\Gamma_2$. Hence, the contribution to the signal from the ground level will dominate over that from the excited level. The bandwidth is determined by $\gamma_\tau+\Gamma_1$ and is much narrower than the bandwidth associated with the rate γ. A measurement of the bandwidth at the resonance line $\delta=0$ will provide a direct measurement of the ground-level depolarization rate Γ_1 provided that the transit time $(1/\gamma_\tau)$ is known accurately.

A theoretical description of this physical phenomenon is accomplished by the solving of the density-matrix equation in the statistical tensor formulation.[8] This procedure is required to account for the degeneracy of the energy levels. The collisional rates are denoted by $\Gamma_{\alpha\beta}^{(k)}$, where k denotes either population $(k=0)$, orientation $(k=1)$, or alignment $(k=2)$. $\alpha\beta$ refers to the ground and/or excited levels. In writing such a decay rate, we assume that there is no coupling between distinct multipoles.[9] The solution of the set of statistical tensor equations in third-order perturbation theory is given by

$$\mathbf{P}^{(3)}(\mathbf{r},t)=\sum_{\alpha=1,2}\sum_{\bar{q}=0,\pm1}\sum_{k}\sum_{-k<q<k}\sum_{\substack{Q,\lambda,\lambda''\\=0,\pm1}}\frac{|\langle J_1||\mu||J_2\rangle|^4}{(ih)^3}\begin{Bmatrix}0&1&1\\J_2&J_1&J_1\end{Bmatrix}\langle T(J_1J_1)_{00}^{+}\rangle\hat{\epsilon}^*_{-q}$$
$$\times E_b^{(\lambda'')}E_f^{(\lambda)}E_p^{(Q)*}\exp[i(\mathbf{k}_f+\mathbf{k}_b-\mathbf{k}_p)\cdot\mathbf{r}-i(\omega_f+\omega_b-\omega_p)t](-1)^{\alpha+1}$$
$$\times M_{q\bar{q}\lambda''}^{k11}(\alpha)M_{qQ\lambda}^{k11}(\alpha)\int_{-\infty}^{\infty}d^3v\,W(\mathbf{v})L_{12}^{(1)}(\omega_f+\omega_b-\omega_p)$$
$$\times[L_{12}^{(1)}(\omega_p)+L_{12}^{(1)*}(\omega_f)]L_\alpha^{(k)}(\omega_f-\omega_p)[B_{qQ\lambda}^{k11}(\omega_f-\omega_p)]^{\beta}, \tag{1}$$

where $\beta=[1+(-1)^{\alpha+1}]/2$ and

$$B_{qQ\lambda}^{k11}(\omega_f-\omega_p)=(-1)^k\tilde{\gamma}^{(k)}\frac{M_{qQ\lambda}^{k11}(2)}{M_{qQ\lambda}^{k11}(1)}L_2^{(k)}(\omega_f-\omega_p)-1 \tag{2}$$

represents the contribution due to spontaneous emission. $\tilde{\gamma}^{(k)}$ is the spontaneous emission rate of the kth pole. $M_{qQ\lambda}^{k11}$ are just geometrical factors, given in terms of the Wigner's $3j$ and $6j$ symbols. $\langle T(J_1J_1)_{00}^{+}\rangle$ is proportional to the initial population of the ground level. The complex Lorentzians are defined by

$$L_{12}^{(1)}(\omega_n)=[\Gamma_{12}^{(1)}+i(\omega_n-\omega_0-\mathbf{k}_n\cdot\mathbf{v})]^{-1}, \tag{3a}$$

$$L_\alpha^{(k)}(\omega_f-\omega_p)=\left\{\Gamma_\alpha^{(k)}+\left[\frac{1+(-1)^\alpha}{2}\gamma^{(k)}+i(\omega_f-\omega_p)-i(\mathbf{k}_f-\mathbf{k}_p)\cdot\mathbf{v}\right]\right\}^{-1}, \tag{3b}$$

and the integration is performed for a Maxwellian distribution $W(\mathbf{v})$. Expressions (1) through (3) are the main theoretical results of this paper and account for the arbitrary polarization state of the radiation fields. The integration has been carried out and the spectral response is plotted in the left-hand column of Fig. 1 for distinct values of neon buffer-gas pressure. Figure 1 corresponds to the case where the polarization states of the pump fields are linear and 90° cross polarized to that of the probe field. Figure 2 corresponds to the case where all input fields are linearly copolarized. The calculation performed for Fig. 2 includes the effect of velocity-changing collisions.[3] The data, in both figures, correspond to the case where the counterpropagating pump waves are on resonance with the two-level system, i.e., $\omega=\omega_0$. In this case, the two resonances overlap and in the absence of buffer gas their linewidths are determined by the spontaneous decay rate of the two-level system.

The experimental study of PNDFWM was made by use of the $3s\,^2S_{1/2}(F=2)$-$3p\,^2P_{3/2}(F=3)$ transition on the D_2 line of atomic sodium. This transition is not subject to ordinary hyperfine-structure optical pumping. The experimental setup was similar to that described earlier[3] where one frequency-stabilized, tunable, cw dye laser supplied the pump beams and the probe beam was supplied by a second cw frequency-stabilized laser. In the first set of experiments, the two pump beams were linearly s polarized and the probe was linearly p polarized. As studied earlier, the resultant signal is linearly p polarized and was detected with a photomultiplier tube and a p-oriented polarization analyzer while in the second set of experiments all beams were set to be s polarized. To avoid contributions from nonlinear Hanle effects, the Earth's magnetic field was cancelled by three orthogonal pairs of Helmholtz coils. The pump frequency was adjusted to be on or near the atomic resonance, and the signal was observed as a function of the probe frequency. The relative laser jitter was of the order of 1 MHz. The angle between the propagation directions of $\mathbf{E}_f$ and $\mathbf{E}_p$ was maintained at $<0.5°$. This geometry gives a residual Doppler width of the order of 3 MHz. In the ab-

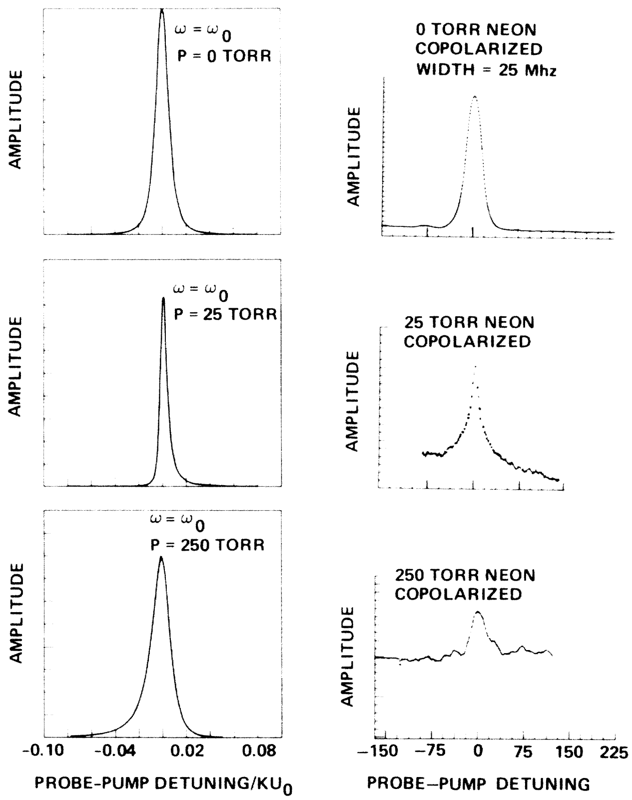

FIG. 1. PNDFWM spectral response as a function of neon gas pressure. The polarization states of $\mathbf{E}_f$, $\mathbf{E}_b$, and $\mathbf{E}_p$ are s, s, and p, respectively. Left-hand column corresponds to theory while right-hand column describes experiments.

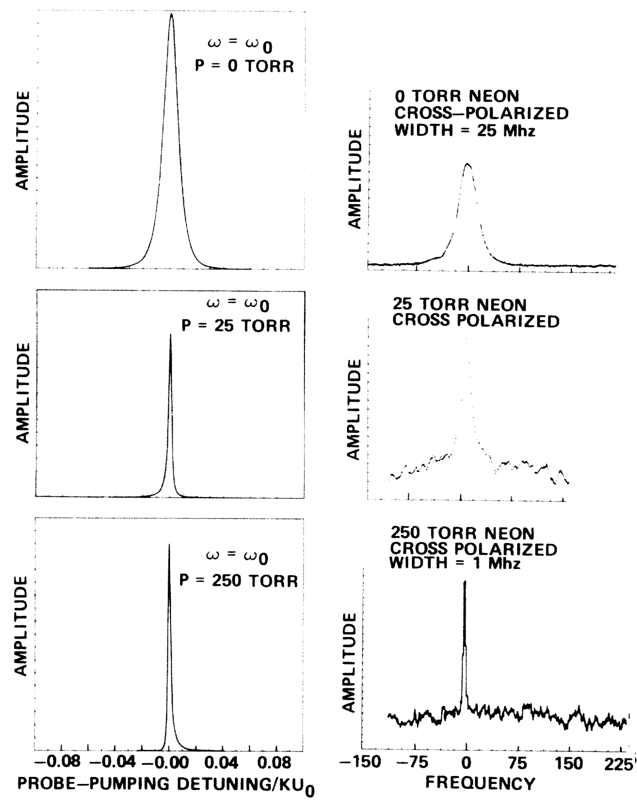

FIG. 2. NDFWM spectral response as a function of neon gas pressure. All radiation fields are linearly copolarized. Left-hand column corresponds to theory while right-hand column describes experiments.

sence of collisions, the measured linewidth of 25 MHz is given by the sum of the spontaneous emission rate, residual Doppler width, and laser jitter. The results of the experiment are illustrated in the right-hand columns of Figs. 1 and 2, and show excellent agreement with the theoretical results.

The distinct behavior of the two data sets can be traced to the origin of the collisional interaction for the population and electric quadrupole. In the presence of buffer gases, population relaxes according to velocity-changing collisions. The cross section for such a collisional interaction is approximately 10^{-15} cm^2 for the ground state of sodium atoms.[10] However, the ground-state electric quadrupole experiences spin-depolarizing collisions which have a cross section in the range of 10^{19}–10^{-26} cm^2 for the case of sodium vapor.[11] Since the collisional decay rate is proportional to the cross section, one expects that the decay rate for the population ($\Gamma_1^{(0)}$) is significantly larger than the one for the electric quadrupole ($\Gamma_1^{(2)}$) for the same pressure range. This physical argument explains the behavior of the linewidth as a function of buffer-gas pressure in Figs. 1 and 2. The results in Fig. 1 show that the subnatural linewidth arising from the ground-state electric quadrupole undergoes no additional changes when the buffer-gas pressure increases from 25 to 250 Torr. In contrast, the results in Fig. 2 show that the subnatural linewidth due to the ground-state population at 25 Torr experiences significant broadening when the buffer-gas pressure is raised to 250 Torr. Furthermore, the data in Fig. 1 allow a direct estimate of the magnitude of the spin-depolarizing cross section for the ground state of sodium. With the measured value of the linewidth at 250 Torr and the average value of the relative velocity of the perturbers with respect to the sodium atoms at 300 °C, the cross section for the spin-depolarizing collision is estimated to have an upper bound of 10^{-18} cm^2. This estimate is limited by the minimum linewidth capability of the laser system.

In conclusion, we used the technique of PNDFWM to study the effect of collisions on the population and electric quadrupole of the ground state of sodium vapor. Furthermore, we have elucidated the physical origin of the subnatural linewidth in the presence of buffer gases and attribute it to the production of ground-state population or electric quadrupole.[12]

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