Optical Hanle effect of Doppler-broadened transitions

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The results obtained in a previous article [Phys. Rev. A 23, 1365 (1981)] for the optical Hanle effect of an atomic beam (no Doppler broadening) are extended to the case of a dilute gas with Doppler width greater than the collision or natural width. It is shown that the optical Hanle effect is independent of the Doppler width for the case where the strong-field detuning is larger than the Rabi frequency and the weak-field excitation detuning. Analytical results are obtained for a $J=0\rightarrow J=1$ transition.

In previous articles we have discussed the optical Hanle effect which is the analog of the Hanle effect in magnetic or electric fields, except that these external fields are replaced by an intense laser radiation polarized either circularly[1] or linearly.[2] This radiation, nonresonant, produces a radiative shift which plays the role of an external field. These theoretical treatments did not include Doppler effects and thus described experiments on an atomic beam of Ba in which the fluorescence was observed in a direction perpendicular to both the beam and the propagation direction of the two lasers.[3,4]

Recently, the theory of the optical Hanle effect has been extended[5] to include finite bandwidth and arbitrary intensity of the driving lasers. In addition, various directions of observation and polarization of the emitting and driving lasers has been studied.[6] In the limiting cases of monochromatic and broadband source, Refs. 1 and 5 are in agreement. However, the Doppler effect was again not included.

In this article, we present the first treatment of Doppler effects on the optical Hanle effect. A two-level atomic or molecular system will be studied. It is assumed that the inhomogeneous broadening due to the Doppler effect is much larger than the homogeneous broadening (either natural or pressure) of the transition. The example to be studied will be a two level $c\rightarrow b$ ($J=0\rightarrow J=1$) transition where a simple physical interpretation of the results is possible.

It has been shown[4] that the important quantity to be calculated and which is accessible to experimental measurement is $\rho_{b_+b_-}$, the matrix element of the density matrix which describes the coherence between the $m=+1$ and -1 sublevels of the $b(J=1)$ level. We assume that this coherence is created by a monochromatic laser, linearly polarized of frequency ω_1, near the transition frequency ω_{bc}. By definition we call the quantity $\delta_1=\omega_1-\omega_{bc}$, the weak-field excitation detuning. We suppose that $\delta_1 \ll$ Doppler width. We further assume that the intensity is weak enough so that saturation effects can be neglected. A second strong saturating monochromatic laser of frequency ω_2 which is nonresonant and circularly polarized couples the b_- sublevel to the ground state $c(J=0,m=0)$. The two laser beams propagate along the same axis, either in the same ($\epsilon=+1$) or opposite ($\epsilon=-1$) direction. As stated above, the Doppler width ω_D is taken to be much larger than the homogeneous width Γ_{bc} of the transition. The fluorescence is observed in a direction perpendicular to the propagation axis of the lasers. By using the rotating polarization technique, it is possible to measure separately the absorption part, $S_A=-2\mathcal{R}e\rho_{b_+b_-}$, and the dispersion part, $S_D=2\mathcal{I}m\rho_{b_+b_-}$, of the coherent contribution to the fluorescence signal. This is separated from the incoherent contribution, $S_I=\rho_{b_+b_+}+\rho_{b_-b_-}$.

The intensity of the fluorescence, $L_F(\alpha)$, emitted in the $0\vec{y}$ direction in the presence of an excitation propagating along the $0\vec{z}$ axis with linear polarization of angle α with the $0\vec{x}$ direction is

$$L_F(\alpha)\sim S_I+S_A\cos2\alpha+S_D\sin2\alpha \quad . \tag{1}$$

The calculation of the matrix elements of ρ which are involved in $L_F(\alpha)$ is performed by summing over the velocities of the emitting atom which we assume are distributed according to Maxwell-Boltzmann statistics. The expression for $\rho_{b_+b_-}$ which was obtained in Eq. (1) of Ref. 4 now contains Green's functions $G_{ij}(n_1,n_2)$ which depend on $\vec{v}$, the center-of-mass velocity of the emittor as follows:

$$G_{ij}(n_1,n_2)=[n_1(\omega_1-\vec{k}_1\cdot\vec{v})+n_2(\omega_2-\vec{k}_2\cdot\vec{v})-\omega_{ij}+i\Gamma_{ij}]^{-1} \quad , \tag{2}$$

where n_1 and n_2 are the harmonics numbers with values 0, ± 1, or ± 2.

In the following, a simple model of the relaxation will be used, chosen so as to illustrate the method and allow a comparison with the earlier results describing atomic beam experiments.

For this purpose, we asssume that the homogeneous width used in Eq. (2) is the rate of spontaneous emission,

$$\Gamma_{bc}=\Gamma_b=\Gamma_{b_\pm b_\pm}=\Gamma_{b_\pm b_\mp};\quad \Gamma_c=0 \ .$$

This assumption does not, of course, limit the results to be obtained.

The calculation follows the same lines as in Ref. 4 and includes the saturating field strength E_2 to all orders (nonperturbative treatment) and the weak field E_1 to lowest order (second-order perturbation in ρ). The strong field is detuned from the resonance frequency ω_{bc} by an amount $\delta_2=\omega_2-\omega_{bc}$, which is assumed to satisfy the condition

$$|\delta_2| >> \beta_2, |\delta_1| \ , \tag{3}$$

where $\delta_1=\omega_1-\omega_{bc}$ is the weak-field detuning, $\beta_2=d_{bc}E_2/2\hbar$ is the Rabi frequency, associated with the strong field and the transition with dipole matrix element d_{bc}. The condition, Eq. (3), is essential for the optical Hanle effect since it permits the neglect of radiative broadening with respect to the radiative shift,

$$\Delta=\frac{-\beta_2^2\delta_2}{\delta_2^2+\frac{1}{4}\Gamma_b^2}\sim-\frac{\beta_2^2}{\delta_2} \ , \tag{4}$$

where $\beta_2 >> \Gamma_b$ has been assumed.

Neglecting the velocity in the Green's function of Eq. (2), we obtain the usual expression

$$\rho_{b_+b_-}=\frac{\beta_1^2\rho_c^0}{\Delta+i\Gamma_b}\left[\frac{1}{\delta_1-\Delta+i\frac{1}{2}\Gamma_b}-\frac{1}{\delta_1-2\Delta-i\frac{1}{2}\Gamma_b}\right] \ , \tag{5}$$

where β_1 is the Rabi frequency associated with the weak field and ρ_c^0 is the c level equilibrium function, assumed only populated. If the velocity is retained, Eq. (5) becomes

$$\rho_{b_+b_-}(V)=\frac{\beta_1^2\rho_c^0}{\tilde\Delta+i\Gamma_b}\left[\frac{1}{\delta_1-V-\tilde\Delta'+i\frac{1}{2}\Gamma_b}-\frac{1}{\delta_1-V-2\tilde\Delta'-i\frac{1}{2}\Gamma_b}\right] \ , \tag{6}$$

where

$$\tilde\Delta=-\frac{\beta_2^2(\delta_2-\epsilon V)}{(\delta_2-\epsilon V)^2+\frac{1}{4}\Gamma_b^2},\quad \tilde\Delta'=\frac{-\beta_2^2[\delta_2+(1-\epsilon)V]}{[\delta_2+(1-\epsilon)V]^2+\frac{1}{4}\Gamma_b^2} \ , \tag{7}$$

and

$$V=k_1v_Z\approx k_2v_Z \ .$$

The $\tilde\Delta$ quantities are now velocity dependent, but still retain their interpretation as radiative shifts due to the strong pump field. However, now a distinction must be made between $\tilde\Delta$, the shift of the b_+ and b_- levels, and $\tilde\Delta'$, the relative shift of the b_- and c levels. $\tilde\Delta$ results from the saturation term and $\tilde\Delta'$ results from the interference term.[7] These terms are also referred to as speed-dependent dynamic Stark shifts.

In the optical Hanle effect, the emitted fluorescence is detected (in the case discussed here, only S_A and S_D are studied) with a fixed frequency ω_1 (usually $\delta_1=0$) and ω_2 [nonresonant by hypothesis, Eq. (3)]. The fluorescence is then studied as a function of the intensity of the strong field. This intensity variation is equivalent to a variation of the radiative shift and causes zero-field level crossings to occur in the same manner as would be the case with external fields and Stark or Zeeman shifts. However, when the radiative shift depends on the velocity of the emitting atom, it is no longer clear that this Hanle effect analogy still holds. To examine this situation we consider two cases which are common in two laser field atomic spectroscopy experiments:

Case I. $|\delta_2| >> \omega_D >> \beta_2, \Gamma_b$. The Doppler width is much larger than the Rabi frequency of the strong field, but much smaller than the strong-field detuning. In this case, the radiative shift becomes essentially velocity independent; $\tilde\Delta=\tilde\Delta'=\Delta=-\beta_2^2/\delta_2$, and $\rho_{b_+b_-}$ becomes (for $\delta_1=0$)

$$\rho_{b_+b_-}=-2\pi\rho_c^0\beta_1^2\frac{\Gamma_b+i\Delta}{\Delta^2+\Gamma_b^2} \ .$$

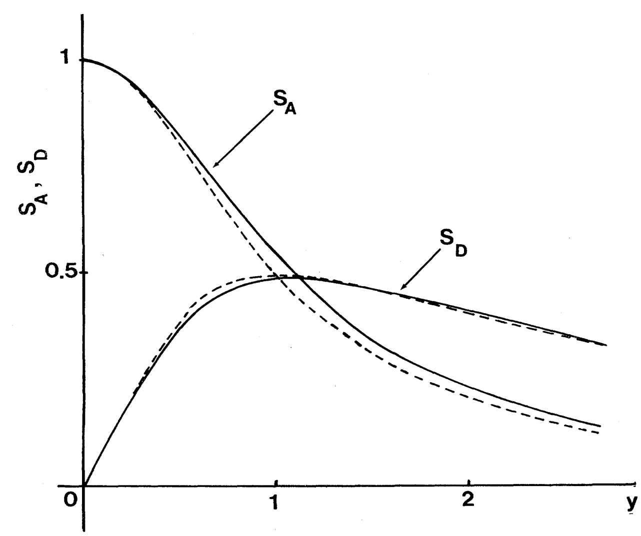

FIG. 1. S_A and S_D resonance curves vs the reduced variable $y=-\Delta/\Gamma_b$ in the two level case. Full line: narrowband excitation with Doppler effect ($\omega_D=400\Gamma_b$; $\delta_2=200\Gamma_b$; $\delta_1=0$; and $\epsilon=+1$). Dashed line: broadband excitation without Doppler effect ($\delta_2=200\Gamma_b$).

This is the same expression as that found for the optical Hanle effect when broadband pumping is used and the Doppler effect neglected. This results from the fact that the case I condition written above implies that although the radiative shift is velocity independent, the weak-field frequency, ω_1, is shifted by the Doppler effect to $\omega_1 - V$ and because of the large Doppler width is equivalent to excitation at a broadband of frequencies. This result is independent, of course, of ϵ, the relative direction of propagation of the two laser beams.

Case II. $\omega_D >> |\delta_2| >> \beta_2, \Gamma_b$. The Doppler width is much greater than the strong-field detuning and Rabi frequency. In this case, the ϵV or $(1-\epsilon)V$ terms cannot be neglected with respect to the detuning δ_2 as in case I. Nevertheless, whenever $\delta_1 = 0$, that is, for resonant excitation of the ω_{bc} transition by the weak field E_1, the range of speeds which contribute is of the order of several Γ_b. Therefore, under these conditions, the terms ϵV or $(1-\epsilon)V$ can be again neglected with respect to δ_2 and the radiative shifts $\tilde{\Delta}$ and $\tilde{\Delta}'$ are essentially velocity independent as in case I. Numerical calculations in two cases; $\delta_2 = 200\Gamma_b$, $\omega_D = 100\Gamma_b$, $\epsilon = \pm 1$, $\delta_1 = 0$, and the same values except $\omega_D = 400\Gamma_b$, give essentially the same result (Fig. 1).

In conclusion, the optical Hanle effect observed on a two-level transition, $J=0 \rightarrow J=1$, with two monochromatic lasers incident on a dilute gas contained in a cell (i.e., with a Doppler width much larger than the collision or natural width), becomes essentially identical to the optical Hanle effect observed on an atomic beam (no Doppler effect) with broadband excitation. However, this result is quite different from the monochromatic excitation without Doppler effect.[4]

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