

Optically Inhibited Collisional Dephasing

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We have performed a photon-echo experiment on the 555.6-nm intercombination line of atomic Yb vapor which demonstrates that an intense laser field acts to inhibit the dephasing resulting from velocity-changing collisions. This finding implies that the optical Bloch equations, which assume that relaxation is independent of the strength of the driving field, cannot correctly describe the behavior of gas-phase samples in all regimes.

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The Bloch equations¹ have provided an important and relatively simple means of calculating the response of a relaxing material to electromagnetic excitation. However, it was demonstrated by Redfield² and recently by DeVoe and Brewer³ that in certain cases the Bloch equations fail to predict the observed responses. The reason for this failure, as originally discussed by Redfield,² lies in the fact that the transverse relaxation time, T_2 , is in general a function of excitation field strength rather than a constant as assumed in the Bloch equations. Prior experimental work on this subject has been restricted to condensed-phase systems. Theoretical analyses^{2,4-8} have shown that the weak, continuous perturbations brought on by the fluctuating local fields characteristic of solids are essential to the existence of excitation-field-dependent relaxation rates.

The present work is concerned not with a solid, but with a gas-phase system. In dilute gases, local fields are strong and exist only briefly during collisions. The phase and state changes induced by these abrupt, intracollisional perturbations are the best known collisional relaxation mechanisms. Since these do not depend on excitation field strength, gas-phase systems have been overlooked in studies of the breakdown of the optical Bloch equations.

In this Letter, we describe an experiment which shows that field-dependent relaxation does, indeed, occur in gases. To understand the effect, recall that

gas-phase relaxation also proceeds via the effect of collisionally induced velocity changes.⁹ The velocity changes introduce weak, random Doppler frequency shifts which act between collisions and mimic local-field-induced frequency shifts in solids. We find that an intense excitation field inhibits velocity-change-induced relaxation.

We employed a modified, two-excitation-pulse photon-echo¹⁰ experiment (see Fig. 1 for nomenclature) to study the relaxation of the 555.6 nm $(6s^2)^1S_0-(6s6p)^3P_1$ transition of atomic ^{174}Yb vapor collisionally perturbed by argon. Holding the time $t_{e0} = 1200$ nsec ($t_{ij} = t_i - t_j$) fixed throughout, we measured the decay of the echo intensity, I_e , as a function of argon pressure, P_a , for several different values of t_{21} . We observed a significant drop in the decay rate as t_{21} approached t_{e0} , suggesting that the field of pulse 2 acted to inhibit the collisional relaxation.

In analyzing our experimental results we assume that the field of pulse 2 (1) does not affect the rate at which collisions occur, (2) does not affect the dephasing introduced by the phase- and state-changing aspect of collisions, and (3) while present completely suspends dephasing introduced by the velocity-changing aspect of collisions. These assumptions, together with the presumed statistical independence of the collisions, allow us to write the total, collisionally induced, echo-time phase error of the j th atom, $\Delta\phi_j$, as

$$\Delta\phi_j = \Delta\phi_j^p + k \left\{ \int_{t_0}^{t_1} \Delta v_j(t) dt - \int_{t_2}^{t_e} \Delta v_j'(t) dt \right\} - k \Delta v_j^{(12)} t_{e3}. \quad (1)$$

Here $\Delta\phi_j^p$ is the net phase change introduced by phase-changing collisions, $k = 2\pi/\lambda$, where λ is the laser wavelength, $\Delta v_j(t)$ is the total accumulated change in the atom's velocity since t_0 , $\Delta v_j^{(12)}$ is the net velocity change accumulated during pulse 2, and $\Delta v_j'(t) = \Delta v_j(t) - \Delta v_j^{(12)}$. The intensity of the echo, I_e , varies as $\langle \exp(i\Delta\phi_j) \rangle^2$, where the angular brackets denote an ensemble average. Ensemble averages corresponding to each of the terms in Eq. (1) have been calculated in analyses of two- and three-excitation-pulse echo collision studies.^{9,11} Using these results, we write

$$I_e = I_0 \exp[-\beta(t_{21})P_a] \equiv I_0 \exp\{-[\beta_p + \beta_{\text{vcc}}^{(1)}(t_{21}) + \beta_{\text{vcc}}^{(2)}(t_{21})]P_a\}, \quad (2)$$

where the three terms in the exponential correspond in the same order to the terms in Eq. (1),

$$\beta_p = \gamma_{e0} \sigma_p, \tag{3a}$$

$$\beta_{\text{vcc}}^{(1)} = 2\gamma_{10} \sigma_{\text{vcc}} [1 - (t_{10})^{-1} \int_0^{t_{10}} dt \int_{-\infty}^{\infty} e^{ik\Delta v t} f(\Delta v) d(\Delta v)], \tag{3b}$$

$$\beta_{\text{vcc}}^{(2)} = \gamma_{21} \sigma_{\text{vcc}} [1 - \int_{-\infty}^{\infty} \exp(ik\Delta v t_{10}) f(\Delta v) d(\Delta v)], \tag{3c}$$

$\gamma_{ij} = 2(n_0/P_0)v_r t_{ij}$, n_0 is the perturber gas density at the pressure P_0 , $v_r = (8k_B T/\pi\mu)^{1/2}$ is the mean radiator-perturber relative speed, k_B is Boltzmann's constant, T is the absolute temperature, μ is the radiator-perturber reduced mass, σ_p (σ_{vcc}) is the total phase-changing or broadening (velocity-changing) cross section, Δv is the velocity change experienced in a single collision, and $f(\Delta v)$ is a one-dimensional coherence-type collision kernel⁹ whose magnitude gives the relative probability of the velocity change Δv . State-changing collisions play a small role in our system, and their effect is here included in β_p .

Equation (2) predicts and we observe a simple exponential decay of echo intensity versus perturber pressure (see Fig. 2). Equations (3) predict that the decay slope β varies with t_{10} [or equivalently t_{21} since $t_{10} = (t_{e0} - t_{21})/2$ and t_{e0} is fixed]. Using Eqs. (3) with $f(\Delta v) = (\delta\pi^{1/2})^{-1} \exp[-(\Delta \times v)^2/\delta^2]$, and inserting the previously measured¹² values of $\delta = 57$ cm/sec, $\sigma_p = 307$ Å², and $\sigma_{\text{vcc}} = 396$ Å², we obtain the values for $\beta(t_{21})$ represented by the solid line in Fig. 3. Our experimental values of $\beta(t_{21})$ (also shown in Fig. 3) are in excellent agreement with those calculated with the model described above. This fact provides strong support for our contention that the collisional relaxation in the Yb-Ar system is modified by the presence of a strong laser field and in particular that dephasing associated with collisional velocity changes is suspended by pulse 2. As pulse 2 gets longer velocity-change-induced dephasing is suspended for a longer period and $\beta(t_{21})$ gets smaller.

T_2 -type relaxation occurs as a result of frequency changes which in the course of time introduce phase changes. The laser does not stop collisions and their concomitant frequency changes. Rather

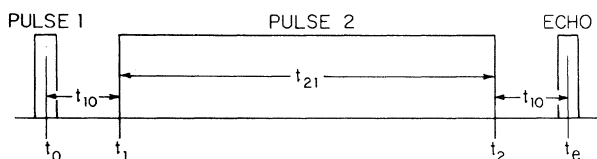


FIG. 1. Modified photon-echo pulse sequence.

it eliminates frequency-change-induced phase changes. We suggest a simple vector model¹³ picture of this process. Consider two atoms, one assumed (for convenience) resonant with a driving field and the other detuned by some amount Δ . Bloch vectors $\vec{p}(0)$ and $\vec{p}(\Delta)$, respectively (see Fig. 4), represent these atoms. At low field intensities, the effective fields $\vec{\Omega}(0)$ and $\vec{\Omega}(\Delta)$ will, as shown in Fig. 4, have significantly different orientations [$\epsilon = \tan^{-1}(\Delta/\chi)$] and magnitudes, $|\vec{\Omega}(\Delta)| = (\chi^2 + \Delta^2)^{1/2}$. Here $\chi = 2d\mathcal{E}/\hbar$, and d and \mathcal{E} , respectively, represent the transition matrix element and the electric field of pulse 2. Consequently, $\vec{p}(0)$ and $\vec{p}(\Delta)$ precess at different rates about different directions, and their relative phase evolves in time. As the driving field intensity increases relative to Δ , however, both the directions and magnitudes of $\vec{\Omega}(0)$ and $\vec{\Omega}(\Delta)$ become ever more nearly the same, and increasingly $\vec{p}(0)$ and $\vec{p}(\Delta)$ precess at the same rate about the same direction. If $\chi \gg \Delta$, the difference in precession angle, $\Delta\theta$, after an interval τ_I is $\Delta\theta = [\Omega(\Delta) - \Omega(0)]\tau_I \cong \Delta^2\tau_I/2\chi$. If $\Delta\theta \ll \pi$, which is true when

$$\chi \gg \Delta^2\tau_I/2\pi, \tag{4}$$

dephasing has in effect been suspended¹⁴ over the bandwidth Δ for the time τ_I . When Eq. (4) is satisfied, Bloch vectors representing atoms within the bandwidth and initially distributed arbitrarily about

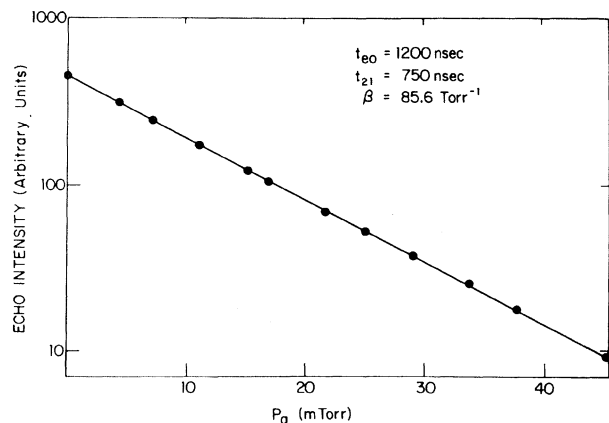


FIG. 2. Echo intensity vs Ar pressure.

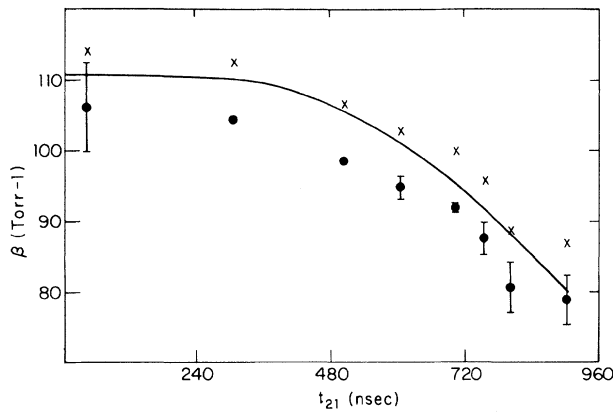


FIG. 3. Variation of β with t_{21} . Solid line, calculated under the assumption that velocity-changing-collision-induced dephasing is suspended for the duration of pulse 2. Solid circles, raw measured β 's. Crosses, measured β 's corrected for Ar-induced variations in Yb density. Error bars represent the standard deviation of several measurements of β (see Fig. 2) for each t_{21} .

the unit sphere can be viewed (over intervals of τ_l) as rotating as a rigid sphere about the driving field. In our experiment, velocity changes lead to frequency shifts of typical magnitude $\Delta_{vcc} \approx k\delta$. Since, for our pulse 2, $\chi \approx 10^8 \text{ rad/sec} \gg \Delta_{vcc}^2 \times t_{21}/2\pi \approx 6 \times 10^6 \text{ rad/sec}$ (using $t_{21} = 900 \text{ nsec}$), Eq. (4) predicts, as we earlier assumed, that velocity-change-induced dephasing will be suspended during pulse 2. Intracollisional frequency shifts, however, are so large ($\approx 10^{12} \text{ rad/sec}$) that their contribution to dephasing is unaffected by our excitation field.

A schematic diagram of our apparatus is shown in Fig. 5. Excitation pulses were generated by an acousto-optic modulator (AOM1). Pulse 1 was generated only with alternate pulse 2's to provide a means of background subtraction. The experimental repetition rate was $\approx 5 \text{ kHz}$. The pulses were

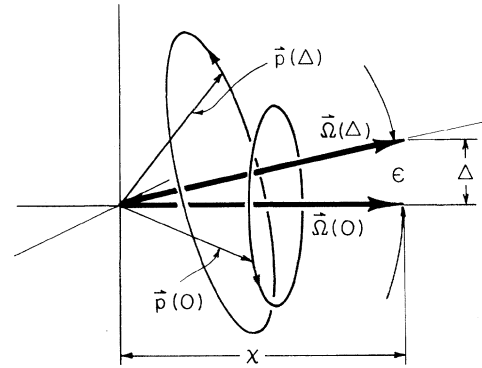


FIG. 4. Vector model picture of optically inhibited collisional dephasing.

optically combined, circularly polarized, collimated to about a 1.5 mm diameter, and resonant with the $^1S_0\text{-}^3P_1$ transition of ^{174}Yb . Pulse 1 (2) had a power of 8 mW (25 mW). A uniform magnetic field of $\approx 70 \text{ G}$ was applied throughout the Yb vapor region (maintained at $\approx 500^\circ\text{C}$) and oriented along the laser propagation direction. Since ^{174}Yb has no nuclear spin, and since we used circular polarized light, our transition closely resembles a two-level system with an upper-state radiative lifetime of 875 nsec. Pulse 1 (2) had a duration of 20 nsec (40 to 900 nsec).

Two problems were encountered in the experiment. First, a free-induction-decay (FID) signal generated by pulse 2 constituted an unwanted coherent background at the time of the echo. AOM2 and AOM3 were thus used to give successive excitation pulse pairs (and hence the FID and echo) a random relative phase so that, averaged over many shots, echo-FID coherence was unimportant. We then simply subtracted the FID intensity (generated by pulse 2 with pulse 1 turned off) from the FID-plus-echo intensity. To rule out the

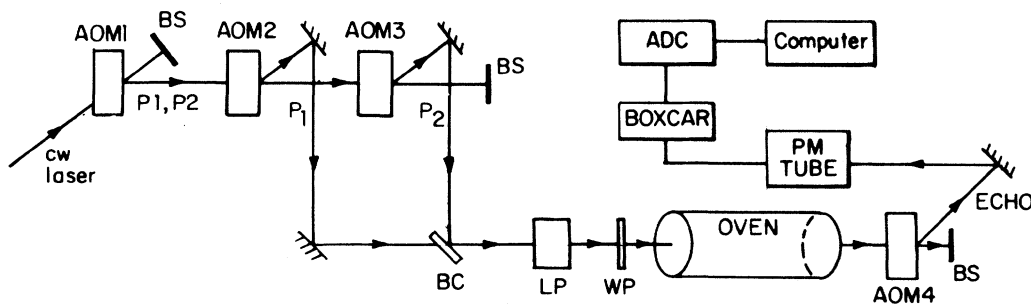


FIG. 5. Experimental schematic. BS, beamstop; BC, beam combiner; P1(2), path of pulse 1 (2); AOM, acousto-optic modulator; LP, linear polarizer; WP, $\lambda/4$ plate.

possibility that our observed variation of β with t_{21} is an artifact of our background subtraction method (the background is larger for long t_{21}), we repeated the experiment with $t_{e0} = 500$ nsec. A much smaller variation of β with t_{21} was observed as expected from Eq. (3). The second problem was a coupling of Yb and Ar densities. The weak-signal, line-center absorption of our sample increased from approximately 40% to 60% as the argon pressure was increased from zero to its maximum value. Throughout this range, increases in Yb density tend to increase I_e , partially offsetting the effect of collisions. A simple correction for this effect brings our β 's into closer agreement with calculated values (see Fig. 3). Since Ar pressure changes lead to a hundredfold collisional decrease in I_e while concomitant Yb density increases lead only to roughly a twofold increase, the correction to β is small.

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