gen the coherent-excitation contribution to the time-averaged signal would be quite small.

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Spin-Exchange Shift and Narrowing of Magnetic Resonance Lines in Optically Pumped Alkali Vapors*

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We observe very narrow (~200 Hz) magnetic resonance lines in alkali vapors with atomic densities much greater than usual and with cell dimensions much smaller than usual. The Zeeman resonance frequency is shifted downward by a large factor, but the spin-exchange broadening vanishes in the high-density limit. This new regime affords important new possibilities for investigating basic interactions in dense alkali vapors, and it also opens the way to miniaturized optically pumped devices.

Spin-exchange collisions[1] are known to broaden the magnetic resonance lines of an alkali vapor. The line broadening has always been found to be equal to the product of the spin-exchange rate and a numerical constant on the order of unity. Measurements of the broadening as a function of the alkali vapor density have been used to deduce thermally averaged spin-exchange cross sections.[2] Since the broadening has been found to be approximately equal to the spin-exchange rate in all experiments so far, one might conclude that sharp magnetic resonance lines could not be observed when the spin-exchange rate is comparable to or greater than the magnetic resonance frequency. However, in this paper we show that in sufficiently dense alkali vapors, the spin-exchange broadening of the Zeeman resonance line vanishes, and the sole effect of spin-exchange collisions is to diminish the apparent gyromagnetic ratio of the atoms by a large factor. Thus, there is an important new high-density regime for optically pumped alkali vapors. We can now see narrow (~200 Hz) magnetic resonance lines in cells with 1 mm thickness and atomic densities greater than 10^{14} cm^{-3}. The potential for miniaturization of optically pumped devices is obvious. It is also clear that the high-density regime affords completely new possibilities for the investigation of spin-relaxation phenomena in dense alkali vapors. Since the spin-exchange broadening vanishes, any self-broadening of the Zeeman resonance line must be due to other, as

yet unknown mechanisms.

For simplicity we shall discuss spin-exchange collisions between identical atoms of a weakly polarized alkali vapor. Our arguments can be extended to cover mixtures of different alkali atoms and strongly polarized vapors. Suppose that the atoms have a nuclear spin I and that they are located in a weak external magnetic field $\vec{H}$. Consider first the limiting case of very rapid spin exchange where the spin-exchange rate is much greater than the ground-state Larmor frequency ω_0,

$$\vec{\omega}_0 = 2\mu_0\vec{H}/(2I+1)\hbar, \tag{1}$$

of the alkali atoms. Under these conditions we expect the atomic angular momentum to hop rapidly back and forth between the states of total angular momentum $F = I + \frac{1}{2}$ and $F = I - \frac{1}{2}$. The Larmor frequencies for these states are very nearly $+\omega_0$ and $-\omega_0$, respectively. The angular momentum will precess only an infinitesimal amount during the periods between spin-exchange collisions, and the average atomic angular momentum $\langle\vec{F}\rangle$ will appear to precess steadily at the weighted average of the frequencies $+\omega_0$ and $-\omega_0$. Since the statistical weight of the level $F = I + \frac{1}{2}$ is greater than that of the level $F = I - \frac{1}{2}$, we expect that the average precession frequency ω will be positive but smaller than the precession frequency ω_0 of the upper Zeeman multiplet. The situation is analogous to motional narrowing in nuclear magnetic resonance[3] or to exchange narrowing in paramagnetic resonance.[4]

Let us now consider the problem from a slightly more quantitative point of view. For sufficiently rapid spin exchange, the alkali vapor will come to spin-exchange equilibrium and the atomic density matrix will be[5]

$$\rho = Ce^{\vec{\beta}\cdot\vec{F}}, \tag{2}$$

where C is a normalizing constant and β, the equilibrium parameter, is much less than unity for a weakly polarized vapor. The total angular-momentum operator of the atom, $\vec{F}$, is the sum of the spin angular-momentum operator $\vec{S}$ and the nuclear angular-momentum operator $\vec{I}$,

$$\vec{F} = \vec{S} + \vec{I}. \tag{3}$$

To lowest order in β, the mean spin angular momentum is

$$\langle\vec{S}\rangle = \tfrac{1}{4}\vec{\beta}, \tag{4}$$

and the mean nuclear angular momentum is

$$\langle\vec{I}\rangle = \tfrac{1}{3}I(I+1)\vec{\beta}. \tag{5}$$

The torque on the atoms from a small external magnetic field $\vec{H}$ is very nearly

$$(2\mu_0\vec{H}/\hbar)\times\langle\vec{S}\rangle = d\langle\vec{F}\rangle/dt, \tag{6}$$

and the torque is equal to the rate of change of the total angular momentum if we assume that the spin-exchange collisions conserve $\langle\vec{F}\rangle$. We can rewrite (6) using (1), (4), and (5) to obtain an equation of motion for the equilibrium parameter $\vec{\beta}$,

$$d\vec{\beta}/dt = \vec{\omega}_\infty\times\vec{\beta}, \tag{7}$$

where

$$\vec{\omega}_\infty = \frac{3(2I+1)}{3+4I(I+1)}\vec{\omega}_0. \tag{8}$$

The high-density Larmor frequency ω_∞ is equal to the normal Larmor frequency ω_0 if the nuclear spin I is 0 or $\frac{1}{2}$, but if I is greater than $\frac{1}{2}$, ω_∞ is less than ω_0. A frequency shift was also predicted in earlier theoretical papers on spin exchange.[6,7] However, that shift was proportional to the product of the spin-exchange rate T^{-1}, the electronic spin polarization (i.e., β), and a parameter κ which could be positive, negative, or zero. The shift discussed in this paper is independent of the parameters T, β, and κ for sufficiently small T and β, and the shift is always toward smaller frequencies.

Having deduced ω_∞, the high-density magnetic resonance frequency for a weakly polarized alkali vapor, we now consider the detailed dependence of the magnetic resonance frequency and linewidth on the spin-exchange rate. From the work of Grossetête[7] or Gibbs[8] one can deduce that the transverse electronic and nuclear angular momenta obey the coupled differential equations

$$\begin{aligned} d\langle S_+\rangle/dt &= -\{A - i\omega_0/[I]\}\langle S_+\rangle \\ &\quad + \{B + i\omega_0/[I]\}\langle I_+\rangle + O(\beta^3), \\ d\langle I_+\rangle/dt &= \{A + 4i\omega_0 I(I+1)/[I]\}\langle S_+\rangle \\ &\quad - \{B + i\omega_0/[I]\}\langle I_+\rangle + O(\beta^3), \end{aligned} \tag{9}$$

where

$$A \equiv 8I(I+1)/3T[I]^2, \tag{10}$$

$$B \equiv 2/T[I]^2, \tag{11}$$

$$[I] \equiv (2I+1). \tag{12}$$

If we ignore the terms of order β^3 in (9) ($\langle S_+\rangle$ and $\langle I_+\rangle$ are of order β), the resulting linear system

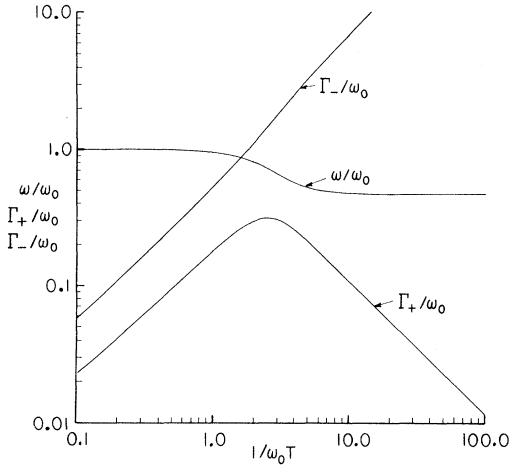

FIG. 1. Theoretical plot of the damping rate for the positive-frequency resonance, Γ_+, the damping rate for the negative-frequency resonance, Γ_-, and the observed Larmor frequency ω versus the spin-exchange rate T^{-1} for the case of alkali atoms with nuclear spin $I=\frac{5}{2}$. The above quantities have been normalized with respect to the Larmor frequency without spin exchange, ω_0.

has eigensolutions of the form

$$\langle S_+\rangle = ae^{\lambda t}, \quad \langle I_+\rangle = be^{\lambda t}, \tag{13}$$

with eigenvalues

$$\lambda = -\tfrac{1}{2}(A+B) \pm \tfrac{1}{2}\{(A+B)^2 + 4i\omega_0[I]B - 4\omega_0{}^2\}^{1/2} = -\Gamma_\pm \pm i\omega. \tag{14}$$

The eigenfrequency ω and the two damping rates Γ_+ and Γ_- are plotted in Fig. 1 as a function of the spin-exchange rate T^{-1} for an alkali atom with $I=\frac{5}{2}$.

For future reference we list approximate values of ω, Γ_+, and Γ_- for slow spin exchange when $T^{-1} \ll \omega_0$. From (14) one finds to lowest order in T^{-1}

$$\omega = \omega_0 - \frac{1}{18\omega_0 T^2}\left(1-\frac{1}{[I]^2}\right)\left(1-\frac{4}{[I]^2}\right), \tag{15}$$

$$\Gamma_+ = 2I(2I-1)/3T[I]^2, \tag{16}$$

$$\Gamma_- = 2(2I^2+5I+3)/3T[I]^2. \tag{17}$$

For slow spin exchanges there will be two magnetic resonance signals, corresponding to rf magnetic fields rotating clockwise and counterclockwise about the static magnetic field at a frequency ω. The linewidth Γ_+ of the positive-frequency resonance will be less than the width Γ_- of the negative-frequency resonance, but both widths

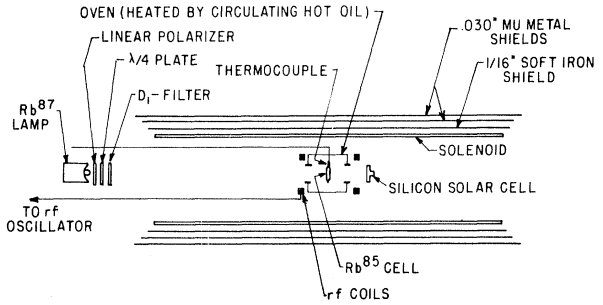

FIG. 2. Experimental apparatus.

will be proportional to the spin-exchange rate T^{-1}. The Larmor frequency is shifted downward by an amount proportional to the square of the spin-exchange rate.

For very rapid spin exchange when $T^{-1} \gg \omega_0$, we see from Fig. 1 that only the positive-frequency resonance is observable, since the negative-frequency linewidth greatly exceeds the resonance frequency. The approximate resonance frequency and linewidth for rapid spin exchange can be obtained from (14) and are, to lowest order in T,

$$\omega = \omega_\infty + 2\omega_\infty{}^3 T^2 \frac{I(2I-1)(2I+1)^2(2I^2+5I+3)}{(4I^2+4I+3)^2},$$

$$\Gamma_+ = \tfrac{2}{3}\omega_\infty{}^2 T \frac{I(2I-1)(2I^2+5I+3)}{(4I^2+4I+3)}.$$

The Larmor frequency approaches its high-density limit, ω_∞, as T^2, and the linewidth Γ_+ approaches zero as T.

We have observed the predicted narrowing and frequency shift in two alkali vapors, Rb^{85} and Cs^{133}. Our apparatus is sketched in Fig. 2. Two concentric, high-permeability magnetic shields allow us to take data in very weak magnetic fields. We used very thin (~1 mm) cells with high buffer gas pressure (~2000 Torr) to minimize the optical thickness of the cell. Magnetic resonance transitions were detected by transmission monitoring of the spin polarization of the optically pumped alkali vapors. A comparison of our results with the theoretical predictions of Eq. (14) is shown in Fig. 3. The residual linewidth at low and high temperatures is probably due to rf power broadening, diffusional and buffer-gas broadening. Although there are some small systematic discrepancies between theory and experiment, the overall agreement is excellent, especially considering the uncertainties in the atomic vapor pressure and the spin-exchange rate constants.

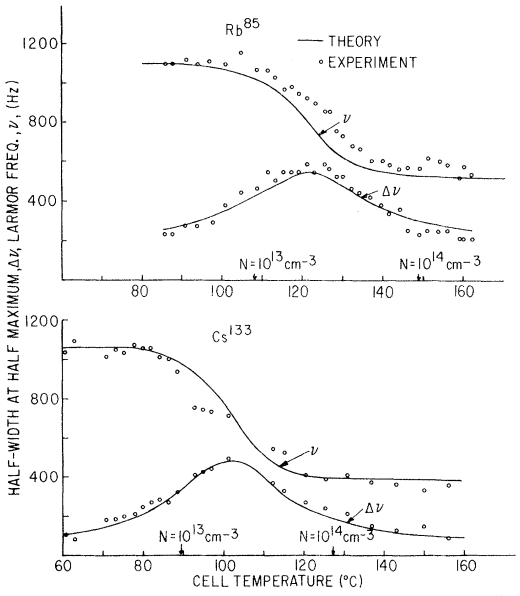

FIG. 3. Magnetic resonance frequency ν and the half width at half-maximum, $\Delta\nu$, of the magnetic resonance line in Rb^{85} and Cs^{133} versus the resonance cell temperature. Theoretical curves, calculated using the spin-exchange constant (Ref. 8), data for rubidium vapor pressure versus temperature (Ref. 9), and data for cesium vapor pressure versus temperature (Ref. 10).

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Time Evolution of a Two-Dimensional Classical Lattice System

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We exhibit a dynamical model of particles in a two-dimensional lattice. We study a particular class of equilibrium states, and we investigate the simplest time correlation functions for these states. As a consequence, the propagation of sound waves with velocity $1/\sqrt{2}$ can be proved rigorously. A transport coefficient analogous to a viscosity is given by a Kubo-type integral formula. The convergence of the integral is discussed. Some preliminary computer results are reported.

We consider an infinite two-dimensional square lattice. On each lattice site there are at most four particles. Each particle may have four velocities, all equal in absolute value, but whose directions are one of the four numbers $\{1, 2, 3, 4\}$ corresponding respectively to the four unit vectors $\{(1, 0), (0, 1), (-1, 0), (0, -1)\}$. Configurations are excluded where there are at least two particles with the same velocity on the same lattice site. We denote by X an allowed configuration and by K the set of all X. It is