Spontaneous Spin Polarization and Bistability in Atomic Vapors by Optical Pumping with Unpolarized Light

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A vapor of paramagnetic atoms, under conditions in which electronic spin exchange takes place faster than collisional spin relaxation, should acquire a spontaneous spin polarization when illuminated by unpolarized light tuned to excite only certain ground-state atomic hyperfine levels. In magnetic fields above a certain size, the atomic polarization has two stable states, either parallel or antiparallel to the field. Experiments to demonstrate this effect and explore possible applications may be feasible by the use of alkali-metal vapors.

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Optical pumping with circularly polarized light is a standard method of obtaining high electronic- and nuclear-spin polarization in an atomic vapor.[1] Here we show that, with use of unpolarized light (and without optically resolving the Zeeman levels), it is still theoretically possible to induce spin polarization optically in a vapor. Any small spin polarization automatically grows under the combined action of the pumping light and electronic spin exchange between pairs of atoms, provided that the light acts only on those hyperfine levels in which the electronic and nuclear spins are antiparallel. In a large enough magnetic field, the polarization is forced to grow along the field, and can reach a stable state in either the parallel or antiparallel direction. If this spontaneous polarization is experimentally realizable, there should be interesting similarities to ferromagnetism, applications to the field of optical bistability,[2] and new phenomena to be explored. For example, the effect on the atoms of the optical polarization induced by the spontaneous spin polarization would lead, under some conditions, to instabilities and the possibility of flip-flop behavior.

In this paper we work out one specific mechanism for spontaneous polarization in alkali atoms, the most likely candidates for initial experiments, and take a brief look at experimental possibilities. A more general discussion will be presented elsewhere.[3]

An alkali atom in its $S_{1/2}$ ground electronic state with nuclear spin I has just two hyperfine levels with total angular momenta $F=I+\frac{1}{2}$ and $F'=I-\frac{1}{2}$. We assume that these two levels are resolved in optical transitions to the $P_{1/2}$ or $P_{3/2}$ excited states. To simplify the analysis, while still illustrating the salient features of spontaneous polarization, we also assume that the net effect of an optical transition to an excited state and subsequent return to the ground state is to randomize the electron-spin direction but to leave the nuclear-spin direction unchanged. At least in principle, both of these assumptions could be satisfied by use of a suitable buffer gas to disorient and quench the optically excited electronic states before the nuclear spin can be flipped by the hyperfine interaction in the excited state.[1,4]

Let incident monochromatic light be tuned to excite only the F' level, at a pump rate Γ_p'. Denote the electronic-spin exchange rate of an atom with other ground-state alkali atoms by Γ_{se}, and the rate of any additional ground-state spin relaxation by Γ_r. For convenience, Γ_r is taken to be due only to isotropic electronic-spin relaxation, as would be the case in ground-state collisions with buffer gas atoms. In general, the ground state will disorient much more slowly than the spatially asymmetric excited states. We require there to be a magnetic field B of such a size that the electron Larmor frequency is much larger than the Γ's (but much smaller than the hyperfine level splitting). The components of the angular momentum perpendicular to B will then average to zero, leaving only the parallel component of total angular momentum free to grow, while the hyperfine interaction still dominates the relative orientation of nuclear and electronic spin.

We first give a qualitative argument for the ideal case when $\Gamma_r=0$. Two possible stable conditions are for all atoms to be aligned with B in the $m_F=+F$ state, or all in the $m_F=-F$ state. In either case the atoms are unaffected by the pumping light, and since all of the atoms have the same electronic-spin direction, they are unaffected by spin exchange as well. Furthermore, when the ensemble of atoms is not in one of these two stable equilibria, the pumping light will drive the ensemble toward the equilibrium favored by any initial bias in M, the ensemble-averaged angular-momentum component along B. Here the key point is that the light acts only on the F' state where (for $I>\frac{1}{2}$) the average electron spin is antiparallel to M. Since by assumption the net effect of absorbing a photon is for the electron spin to be randomized, there must be a net gain in M. Spin exchange does not change M, but does keep returning atoms to the F' state for optical pumping until M becomes $+F$ or $-F$.

Before introducing a general treatment, we consider the two limiting cases for $\Gamma_r \neq 0$, when either spin exchange or optical pumping dominates. When spin exchange dominates, Γ_{se} is much larger than both Γ'_p and Γ_r, and the ground-state atomic levels can be described by a spin temperature.[5] The general treatment given below then yields the following simple expression for the time evolution of M, valid while M is small:

$$\frac{dM}{dt} \cong \frac{3M[-\Gamma_r + \Gamma'_p(2F-1)(F-1)/6F]}{2(2F^2+1)} \quad (|M| \ll F;\ \Gamma'_p, \Gamma_r \ll \Gamma_{se}). \tag{1}$$

Note the difference in sign (for $F>1$) between the coefficients of Γ_r and Γ'_p in Eq. (1). If $\Gamma_r/\Gamma'_p < (2F-1)(F-1)/6F$ there is an exponentially increasing solution for M of either sign. As $|M|$ grows, the spin-exchange rate begins to reduce, until finally Eq. (1) no longer applies and instead $dM/dt \rightarrow 0$. When optical pumping dominates, Γ'_p is much larger than both Γ_{se} and Γ_r; most of the atoms are maintained in the F state by optical pumping, and the general treatment gives

$$\frac{dM}{dt} \cong \frac{2M(-\Gamma_r + C_{se}\Gamma_{se})}{2F^2+F+1} \quad (\Gamma_{se}, \Gamma_r \ll \Gamma'_p), \tag{2}$$

where $C_{se} \equiv (F-1)(1-\langle m_F^2 \rangle/F^2)/2$ is a positive number (for $F>1$) that depends upon the alignment of the atoms. $C_{se} = C_0 = (F-1)(2F-1)/6F$ when all m_F sublevels are equally populated, but reduces toward zero as the alignment increases. If $C_0\Gamma_{se} > \Gamma_r$ in Eq. (2), any small $|M|$ increases exponentially, leveling off as the alignment increases and reaching equilibrium when $C_{se}\Gamma_{se} = \Gamma_r$. The behavior in both limits suggests that, when optical pumping and spin exchange are sufficiently fast relative to Γ_r, the system has an unstable equilibrium at $M=0$, and has two stable equilibria at equal and opposite finite values of M.

We now begin a more general treatment by writing down the equations governing the density matrix of the ground-state atomic hyperfine sublevels. We then derive some useful expressions for the time rate of change of atomic angular momentum, present the general conditions for bistability, and plot numerical solutions of the complete density-matrix equations as functions of time to verify the spontaneous growth of the polarization to finite equilibrium values as sketched above.

The off-diagonal elements of the density matrix quickly average to zero in the assumed magnetic field. The density-matrix equations for the diagonal elements are[1,5]

$$\begin{aligned} d\rho_{F',m}/dt = (1/4F^2)\textstyle\sum_i \{ & (-3F^2+m^2-2mFP_i)\Gamma'_i\rho_{F',m} + (F^2-m^2)\Gamma_i\rho_{F,m} \\ & + [(F+m-1)\Gamma'_i\rho_{F',m-1} + (F-m+1)\Gamma_i\rho_{F,m-1}](F-m)(1+P_i)/2 \\ & + [(F-m-1)\Gamma'_i\rho_{F',m+1} + (F+m+1)\Gamma_i\rho_{F,m+1}](F+m)(1-P_i)/2\}, \\ d\rho_{F,m}/dt = (1/4F^2)\textstyle\sum_i \{ & (-3F^2+m^2+2mFP_i)\Gamma_i\rho_{F,m} + (F^2-m^2)\Gamma'_i\rho_{F',m} \\ & + [(F-m+1)\Gamma_i\rho_{F,m-1} + (F+m-1)\Gamma'_i\rho_{F',m-1}](F+m)(1+P_i)/2 \\ & + [(F+m+1)\Gamma_i\rho_{F,m+1} + (F-m-1)\Gamma'_i\rho_{F',m+1}](F-m)(1-P_i)/2\}, \end{aligned} \tag{3}$$

where we have incorporated the assumption discussed earlier that the net result of absorbing a photon is to randomize the electronic spin completely. The summation index i stands for optical pumping, spin exchange, or isotropic relaxation, and the P_i are defined as follows:

$$P_p = -P_l(J-1)^{-1}(2J+1)^{-1},$$

where J is the electronic angular momentum of the optically pumped excited state, and P_l is the circular polarization of the light $(-1 \leq P_l \leq 1)$;

$$P_{se} = 2\langle m_s \rangle = \textstyle\sum_m m(\rho_{F,m} - \rho_{F',m})/F; \quad P_r = 0.$$

We assume $\Gamma_{se} = \Gamma'_{se}, \Gamma_r = \Gamma'_r$, but usually $\Gamma'_p \gg \Gamma_p$.

Equations (3) can be used to derive the time evolution of the average angular momentum in each hyperfine level. With $M_{F'} = \sum_m m\rho_{F',m}$, $M_F = \sum_m m\rho_{F,m}$, and $M = M_{F'} + M_F$, we have

$$\begin{aligned} \frac{dM_{F'}}{dt} &= \frac{-M_{F'}}{4F^2}[(\Gamma'_p+\Gamma_r)(2F^2+F+1) + \Gamma_{se}(F+1-A)] + \frac{M_F}{4F^2}[\Gamma_r(2F^2-3F+1) + \Gamma_{se}(F-1-A)], \\ \frac{dM_F}{dt} &= \frac{M_{F'}}{4F^2}[(\Gamma'_p+\Gamma_r)(2F^2+3F+1) + \Gamma_{se}(F+1-A)] - \frac{M_F}{4F^2}[\Gamma_r(2F^2-F+1) + \Gamma_{se}(F-1-A)], \\ \frac{dM}{dt} &= \frac{M_{F'}\Gamma'_p}{2F} + \frac{(M_{F'}-M_F)\Gamma_r}{2F}, \end{aligned} \tag{4}$$

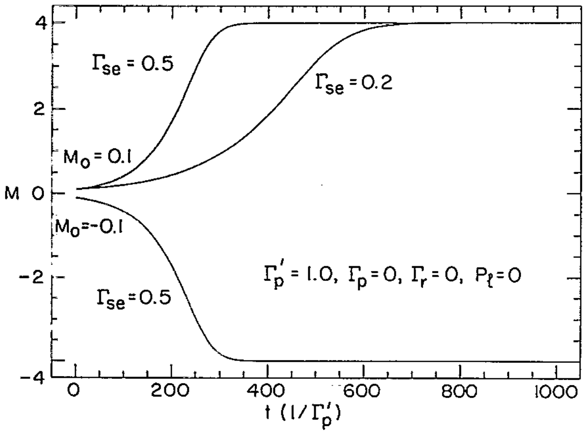

FIG. 1. Time evolution of M, the angular-momentum component along an external magnetic field, of an ensemble of Cs atoms when unpolarized light is resonant with the $F'=I-\frac{1}{2}=3$ atoms alone, and there is no spin relaxation. The sign of the initial value for $M=M_0\pm 0.1$ determines the sign for the growth of M.

where

$$A\equiv\sum_m m^2\rho_{F,m}(F-1)/F^2+\sum_m m^2\rho_{F',m}(F+1)/F^2,$$

and we have assumed that $P_l=0$ and $\Gamma_p=0$. When all the m_F and $m_{F'}$ levels are equally populated, or more generally when there is zero alignment, $A=A_0=(F^2-1)/3F$. Equations (4) lead to Eqs. (1) and (2) in the appropriate limits.

We can proceed as we did with Eqs. (1) and (2) to obtain general conditions for the existence of bistability. We are interested in exponentially increasing solutions of Eqs. (4) when the alignment is small ($A\cong A_0$). Such solutions exist when the Γ's satisfy the inequality

$$\Gamma_p'\Gamma_{se}(F-1-A)>2\Gamma_r(\Gamma_p'+\Gamma_{se}+\Gamma_r). \tag{5}$$

This equation reduces to the conditions already discussed in the limit of large Γ_{se} or large Γ_p'.

We have numerically solved the complete density-matrix equations [Eqs. (3)] as a function of time for ^{133}Cs ($I=\frac{7}{2},F=4$) for various values of the Γ's. Some results are shown in Figs. 1 and 2. These curves clearly show that, for $P_l=0$ and values of the Γ's satisfying Eqs. (4), an initially small M automatically rises to some finite equilibrium value. (Because $M_0=0$ is a state of unstable equilibrium, in Figs. 1 and 2 a small amount of angular momentum is given to the otherwise thermally distributed initial hyperfine level densities.) Figure 3 shows the effect of a light beam with a small degree of circular polarization ($P_l=0.04$), which drives an initially unpolarized atomic ensemble into one of the bistable states.

It may be feasible to observe this spontaneous spin polarization in properly designed experiments. For example, measurements on Cs vapor[1,4] indicate that in 200 Torr of N_2 buffer gas, $\Gamma_r=200$ sec^{-1}, while at a Cs den-

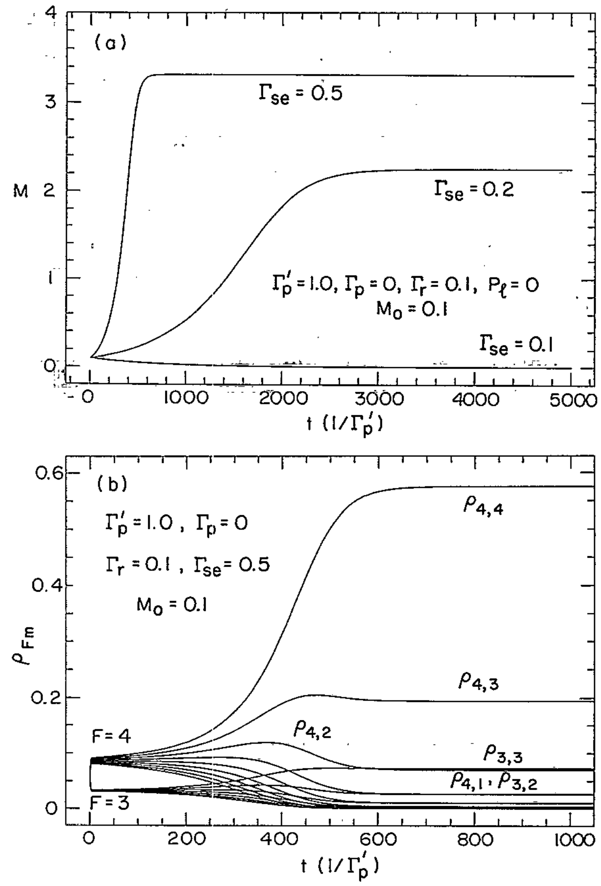

FIG. 2. (a) Time evolution of M when unpolarized light is resonant with $F'=3$ Cs atoms having $M_0=0.1$ and there is isotropic electronic-spin relaxation ($\Gamma_r=0.1$). For Γ_{se} too small, there is no growth of M. (b) Time evolution of the Cs-atom ground-state hyperfine level densities for the case in (a) when $\Gamma_{se}=0.5$. Optical pumping makes the populations of the two hyperfine multiplets different after a few pump times.

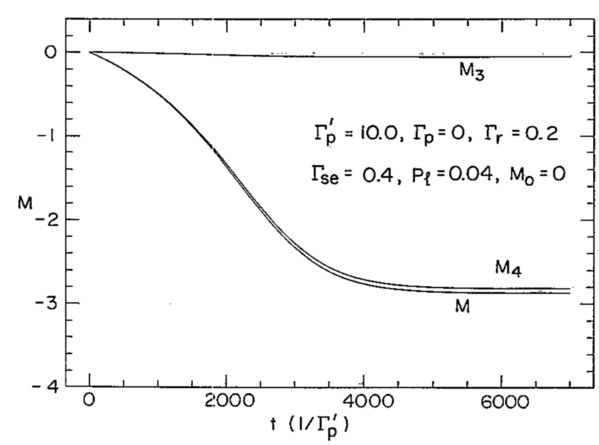

FIG. 3. Time evolution of M_3, M_4, and $M=M_3+M_4$ for an initially unpolarized atomic Cs ensemble ($M_0=0$) when light with 4% circular polarization is resonant with the $F'=3$ atoms alone.

sity of 1.5×10^{11} atoms/cm^3, $\Gamma_{se}=1000$ sec^{-1}. A laser beam tuned for excitation of the $F'=3$ state to either the first or the second P state can readily produce the needed pump rate, Γ_p', while the observed collisional width of the absorption lines leads one to expect a small Γ_p ($\cong\Gamma_p'/16$) because of the small overlap with the $F=4$ line. The key experimental issue is how well the nuclear spin is actually preserved in an optical pumping cycle at this buffer-gas pressure. If higher pressures are needed to avoid loss of nuclear angular momentum in the excited state, then Γ_p/Γ_p' may become too large. Other issues are the optical thickness at high alkali-metal vapor densities, and the loss of angular momentum by diffusion to the cell walls. The spontaneous spin polarization could be detected by optical rotation of the pumping light. This polarization should show hysteresis as a function of the ellipticity of the light.

Spontaneous polarization should also be possible with alkali-metal vapors in cells without a buffer gas, but with suitable wall coatings to inhibit spin relaxation. In this case, the calculations must include the effect of resolved hyperfine coupling in the excited state[3]; the results are similar to those presented here, although the exact conditions for spontaneous polarization [analogous to Eq. (5)] depend upon which hyperfine level is used in the excited state. Experimentally, the relaxation of the F (unpumped) ground state by absorption of resonance reradiation must be avoided by proper cell geometry or by tuning of the incident laser beam to the wings of a line where the resonance light is only weakly absorbed.

In summary, we have shown that it should be possible to exploit the angular-momentum–conserving properties of electronic-spin exchange to produce spontaneous polarization and spin bistability in atomic vapors illuminated by unpolarized light. Experiments are in progress to search for these effects.

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