Observation of Spontaneous Spin Polarization in an Optically Pumped Cesium Vapor

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(Received 20 November 1995)

We have observed spontaneous atomic spin polarization of a Cs vapor undergoing rapid spin exchange in a cell when illuminated with plane-polarized 894 nm ($D1$) light. The spin polarization shows marked hysteresis in switching between two stable orientations, fully aligned parallel or antiparallel to an external magnetic field, as the circular polarization of the pump light is varied about zero. We show that our observations agree with theoretical expectations and point out possible applications of spontaneous polarization.

PACS numbers: 42.65.Pc, 32.80.Bx

We have made the first observation of spontaneous atomic spin polarization in an alkali metal vapor optically pumped with plane-polarized light. Such polarization has been predicted to occur when optical pumping and electronic spin exchange take place faster than spin relaxation [1]. In our experiment, Cs vapor contained in a cell along with a nitrogen buffer gas becomes fully spin polarized when illuminated with linearly polarized 894 nm ($D1$) light tuned to excite atoms out of only the $F = 3$ ground state hyperfine level. For certain orientations of an external magnetic field, $\mathbf{B}_0$, the state of zero spin orientation is unstable; the slightest bias grows exponentially until the atomic spins are all lined up in the $F = 4$ state, either parallel or antiparallel to the field. The full atomic spin orientation remains indefinitely in either stable state in the presence of linearly polarized light and can be induced to switch from one stable state to the other by briefly introducing some circular polarization to the pump light. A striking feature of the spontaneous polarization, taken as unambiguous evidence that such has occurred, is hysteresis in the observed spin orientation when the circular polarization of the pump light is varied about zero, analogous to the hysteresis observed in ferromagnetism.

In this paper we describe the observations that establish the existence of this phenomenon and show that the main results are well explained by a generalization of the original analysis of Ref. [1]. Many properties of spontaneous polarization remain to be explored, such as the behavior in weak magnetic fields, in dense vapors, and at higher buffer gas pressures. It may also be possible to generate spontaneous polarization in coated cells [2] without buffer gas, in dense optically cooled vapors [3], or in optically pumped crystals. Spontaneous polarization may prove useful in precise measurements of spin precession, as in the search for a permanent electric dipole moment [4–6], and in spin exchange pumping of noble gas nuclei requiring a dense, fully polarized alkali vapor [7,8].

In a cesium atom, with nuclear spin $I = \frac{7}{2}$, the $6S_{1/2}$ ground state and the $6P_{1/2}$ excited state are each split into two hyperfine levels of total angular momentum $F = I \pm \frac{1}{2} = 4$ and 3. In our experiment, optical pumping takes place on the resolved $F = 3 \rightarrow F = 4$ hyperfine component of the $D1$ transition between $6S_{1/2}$ and $6P_{1/2}$. For this hyperfine component, spontaneous spin polarization appears when the polarization vector of the linearly polarized pump light is oriented perpendicular to $\mathbf{B}_0$. Absorption of a photon by an atom will then cause m, the magnetic quantum number along $\mathbf{B}_0$, to change by ± 1, with an increase in $|m|$ more likely than a decrease; the average change due to excitation from a given ground state sublevel m is found from Clebsch-Gordan coefficients to be $\Delta m = +9m/(m^2 + 20)$. (A ground state Cs atom in the $|F, m\rangle = |3, +2\rangle$ sublevel, for example, will make transitions to the $|4, +3\rangle$ and $|4, +1\rangle$ excited state sublevels with relative probabilities $\frac{7}{8}$ and $\frac{1}{8}$, respectively.) Thus any ensemble-averaged bias, $\mathcal{M} \equiv \langle m \rangle$, to the extent that it appears in the $F = 3$ ground state, will be reinforced in transitions to the excited state by an increment $\Delta\mathcal{M} = \langle \Delta m \rangle$, with the extra angular momentum being extracted from the light. In general only a fraction of $\Delta\mathcal{M}$ will be lost in the return to the ground state, resulting in a net gain in spin angular momentum over a complete optical pumping cycle.

By itself, optical pumping would soon depopulate the $F = 3$ state before adding appreciable angular momentum to the system. The crucial role of electronic spin exchange between atoms is to repopulate the optically pumped hyperfine level while conserving $\mathcal{M}$, thus allowing optical pumping to continue adding spin angular momentum as long as atoms are returned to the $F = 3$ state. In the ideal case of no spin relaxation, the atoms would end up in the unpumped $F = 4$ hyperfine level, all aligned with the field in the $|4, +4\rangle$ sublevel or all in the $|4, -4\rangle$ sublevel, and would no longer be affected by spin exchange. With a finite spin relaxation rate that is much smaller than the spin exchange and optical pumping rates, the atoms will be driven into a stable state of high spin polarization approaching the ideal case. Further discussion follows the description of the experiment.

The layout of the experiment is shown in Fig. 1. A Coherent 899-21 Ti:S ring laser provides the pump beam,

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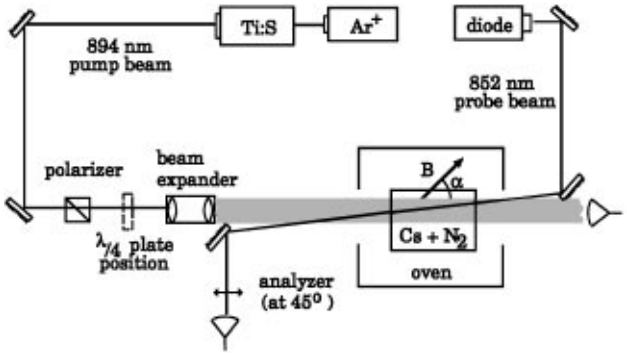

FIG. 1. The experimental arrangement. The analyzer measures the optical rotation of the probe beam due to the spin polarization of the cesium vapor.

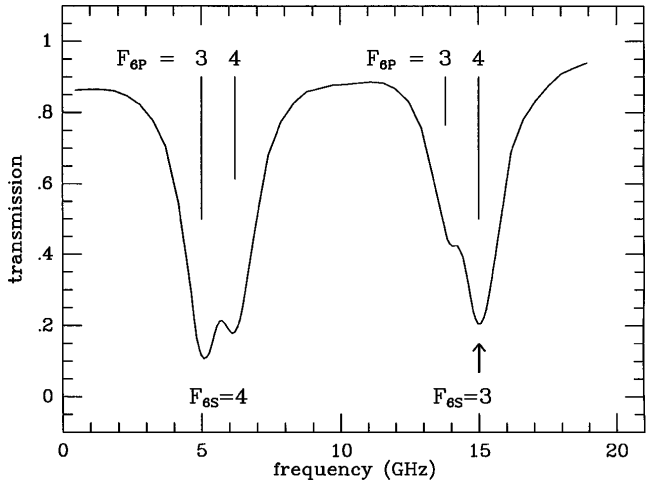

FIG. 2. The absorption profile of the $D1$ absorption line, showing the hyperfine splitting in the ground and excited states. The arrow marks the $3 \rightarrow 4$ hyperfine component used to pump spontaneous polarization.

which passes through a Glan-Thompson calcite plane polarizer and enters the cesium vapor cell after being expanded to a diameter of 1 cm. A static magnetic field, $\mathbf{B}_0$, adjusted in magnitude and direction by Helmholtz pairs, makes an angle α with the pump beam propagation direction and an angle β with the electric field vector of the pump light. A quarter-wave retardation plate [9] in a rotating mount can temporarily be placed after the polarizer to give some degree of circular polarization to the light when it is desired to cause existing spontaneous polarization along $\mathbf{B}_0$ to reverse sign. A weak plane-polarized probe beam from an external cavity diode laser is directed at 5° to the pump beam axis and overlaps the pump beam inside the Cs cell. The probe laser is tuned to the 852 nm $D2$ absorption line and is adjusted to be near resonance with the $F = 4$ ground state level. Optical rotation of the plane of polarization of the probe beam, measured by transmission through an analyzing polarizer at 45° to the input polarization, acts as a measure of the ground state spin polarization. An oven regulates the temperature of the cell, setting the Cs density.

The cells are 2.5 cm diameter pyrex cylinders ranging in length from 0.5 to 2.5 cm, with flat windows to pass the laser beams. The N_2 buffer gas serves both to prevent rapid diffusion of the Cs atoms to the walls, where spin relaxation takes place, and to quench the excited atoms to the ground state before they emit resonant spontaneous radiation that can depolarize atoms in the $F = 4$ ground state level. [N_2 has a particularly large collisional cross section ($\cong$86 Å^2) for quenching the $6P_{1/2}$ state compared with a much lower cross section ($\cong$5 Å) for changing $6P_{1/2}$ to the more readily depolarized $6P_{3/2}$ state [10].]

Figure 2 shows the $D1$ absorption profile with 46 torr buffer gas pressure as the pump laser frequency is swept across the hyperfine components of the line. The 9.2 GHz ground state hyperfine splitting is well resolved, and the 1.2 GHz splitting in the excited state, resolved with no buffer gas, is partially resolved at 46 torr.

The signature of spontaneous polarization can be seen clearly in Fig. 3(a), which shows the observed changes in the spin polarization of the sample, $\mathcal{M}$ (as measured by the probe beam), as the degree of circular polarization of the pump light, σ^L, is varied by rotating the quarter-wave plate slowly and continuously. Note that $\mathcal{M}$ does not follow the variation in σ^L smoothly, but abruptly changes between two states of spin polarization, one parallel and the other antiparallel to $\mathbf{B}_0$, reversing polarity well after σ^L has passed through zero and acquired a sign tending to reverse $\mathcal{M}$ by direct optical pumping.

Figure 3(b) shows a plot of $\mathcal{M}$ vs σ^L from data taken as the quarter-wave plate is manually rotated in small increments from negative to positive σ^L and then rotated back. The plot exhibits striking hysteresis behavior similar to a saturated $\mathcal{M}$ vs $\mathcal{H}$ curve in single crystal ferromagnetism. We emphasize that, even when $\sigma^L = 0$, $\mathcal{M}$ automatically acquires a stable finite value which it keeps for as long as the pump light remains.

All data in Fig. 3 were taken with a field of magnitude $\mathbf{B}_0 = 55$ G and direction $\alpha = 80°$ relative to the pump beam. The linear polarization of the pump beam, before the quarter-wave plate, was set approximately perpendicular to $\mathbf{B}_0$ ($\beta \cong 90°$).

Spontaneous polarization appears above certain threshold values of pump light intensity and cesium density, determined by the cell geometry and buffer gas pressure (and within a range of values of α and β as discussed below). The data in Fig. 3 were taken with a cell 2.5 cm long containing 46 torr N_2, at a cell temperature of 82.5 °C with an experimentally determined electron spin exchange rate of $\Gamma_{se} = 2800$ s^{-1} and an approximate cesium density of 5×10^{12} atoms/cm^3. The threshold for spontaneous polarization in this cell occurs at a cell temperature between 57 and 64 °C, corresponding to a value of Γ_{se} between 430 and 730 s^{-1}. Cells with buffer gas pressures in the range of 10–160 torr were tested, and

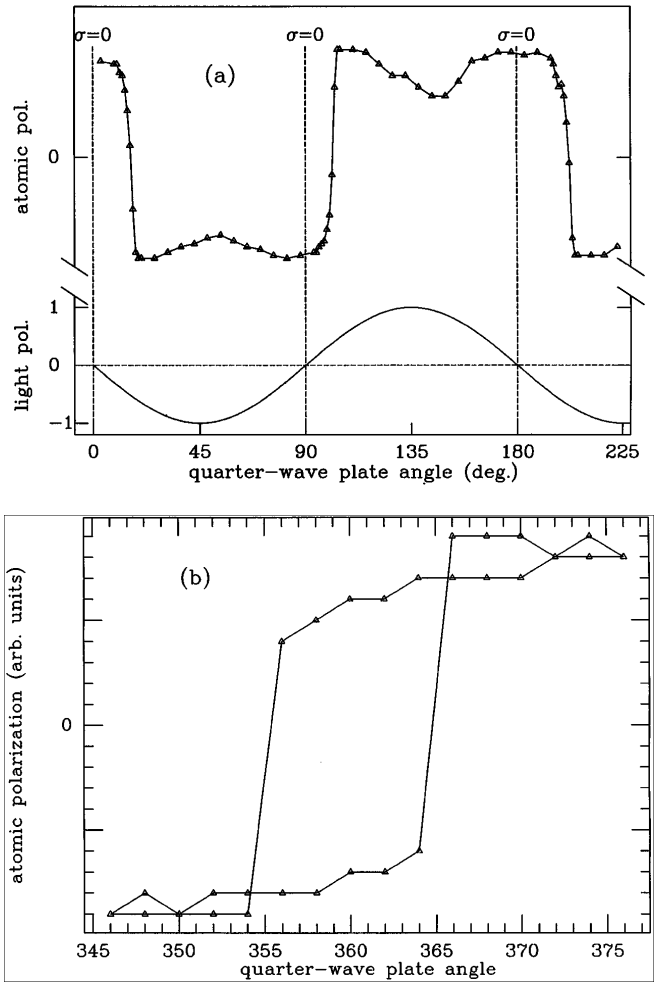

FIG. 3. Experimental data revealing the presence of spontaneous atomic spin polarization. (a) The variation of the atomic polarization, $\mathcal{M}$, with σ^L, the degree of circular polarization of the light, as the quarter-wave plate rotates, showing $\mathcal{M}$ switching between two stable, opposite polarity states well after σ^L has passed through zero and reversed sign. (b) A plot of $\mathcal{M}$ vs σ^L, showing the marked hysteresis. In both (a) and (b), the magnetic field points at an angle $\alpha = 80°$ away from the axis of the light beam, and only a small fraction of σ^L actually pumps $\mathcal{M}$ along $\mathbf{B}_0$. The approximate location of zero spin polarization has been indicated.

all exhibited spontaneous polarization up to the highest cell temperature tried, 95 °C, using typical values of 1–10 mW pump light and 1–10 μW probe light.

When $\mathbf{B}_0$ is tilted closer to the light propagation direction, so that $\alpha < 60°$, spontaneous polarization is reduced or may not appear at all because as $\mathcal{M}$ is spontaneously generated in the front part of the cell it imparts a circular polarization to the light that tends to induce $\mathcal{M}$ of the opposite sign at the rear of the cell.

Preliminary tests were done at low magnetic field by placing the cell inside a set of magnetic shields. For magnetic fields below about 50 μG the feedback between $\mathcal{M}$ and the induced σ^L mentioned above creates an interesting steady oscillation in $\mathcal{M}$ that warrants further study.

To interpret our experiment quantitatively, the treatment of spontaneous polarization in Ref. [1] must be generalized to include the resolved hyperfine splitting in the excited state and arbitrary polarization of the pumping light. We quote results from a full analysis [11] that uses the complete optical polarization tensor to obtain the effect of optical pumping on the various multipoles [2] of the ground state spin density matrix. In that analysis, the collisional quenching of the excited $6P_{1/2}$ atoms by the N_2 buffer gas molecules is shown to produce the same relative populations among the ground state m sublevels as does spontaneous radiation, and the Larmor precession frequency about $\mathbf{B}_0$ is assumed large enough to average the off-axis spin components to zero. In the relevant special case of pumping with plane-polarized light on the $3 \rightarrow 4$ hyperfine component of the $D1$ line, and in the limit of large optical pumping rate compared to the spin exchange and spin relaxation rates (a condition that is well satisfied for most of our data), the rate of change of $\mathcal{M}$, the dipole polarization along $\mathbf{B}_0$, may be written as

$$\frac{d\mathcal{M}}{dt} = \Gamma \mathcal{M} , \qquad (1)$$

where a small contribution from the octupole distribution of the spin population has been ignored. When the gain coefficient, Γ, is positive, we expect spontaneous polarization to appear. In the strong pump regime assumed, the optical pumping rate drops out and Γ is given by

$$\Gamma = \frac{7(1 - 2\cos^2\beta)}{12(10 - \cos^2\beta)}(1 - \delta_{6P}) f_{se} \Gamma_{se} - \Gamma_{\mathrm{rel}} . \qquad (2)$$

The factor$1 - \delta_{6P}$ includes spin relaxation due to collisions between excited state atoms and the buffer gas, as well as the effect of pumping to the partially overlapping $F = 3$ excited level. Under the conditions of our experiment, $\delta_{6P} \ll 1$. Excited state losses are expected to be more severe in the $6P_{3/2}$ state and did show up clearly in some preliminary observations using $D2$ pumping light. The factor f_{se} gives the reduction in spin exchange rate when there is significant spin alignment among the atoms; $f_{se} \cong 1$ when $\mathcal{M}$ is small, but $f_{se} \rightarrow 0$ as $\mathcal{M} \rightarrow 4$ [1].

The last term in Eq. (2) is the total spin relaxation rate, Γ_{rel}, with contributions from a number of mechanisms: (1) spin relaxation in the ground state due to buffer gas collisions, (2) diffusion of Cs atoms to the cell walls, (3) optical pumping in the wings of the highly populated $F = 4$ hyperfine level, and (4) relaxation as a result of Cs_2 molecule formation [12]. The magnitude of each mechanism can be estimated with some reliability for any given conditions of cell geometry, Cs and N_2 density, and pumping light intensity from measurements of hyperfine pump and relaxation times, spin relaxation times, and pump beam absorption spectra. For the conditions under which the data of Fig. 3 were taken the main sources of

relaxation are from mechanisms (1), (2), and (3), and we estimate $\Gamma_{\rm rel}$ to be of order 40 s^{-1}.

Whenever $\Gamma > 0$, any arbitrarily small value of $\mathcal{M}$ will grow exponentially until reaching saturation, which occurs when the magnitude of $\mathcal{M}$ becomes so large that f_{se} begins to decrease, causing Γ to approach zero. At saturation, more atoms are in the $F = 4$ ground state level than when spin exchange is repopulating the $F = 3$ level at the full rate Γ_{se}, making the vapor more transparent to the pump light. This method for identifying a state of spontaneous polarization is particularly useful in low magnetic fields, where varying σ^L would result in light shift perturbations comparable to $\mathbf{B}_0$.

Equation (2) provides a good qualitative and quantitative description of the spontaneous polarization data. In the absence of relaxation, Γ would be positive and spontaneous polarization would occur when $|90° - \beta| < 45°$. Under the conditions of Fig. 3 spontaneous polarization was actually observed within the range $|90° - \beta| < 38°$, the difference being due to a finite value of $\Gamma_{\rm rel}$. This qualitative agreement with Eq. (2) can be made quantitative by setting the expression for Γ equal to zero at the maximum observed value of $\cos^2\beta$ (= 0.37). Using the experimentally determined rate $\Gamma_{se} = 2800$ s^{-1}, we find that $\Gamma_{\rm rel}$ must be $\cong 40$ s^{-1}, in agreement with the earlier estimate based on the known relaxation mechanisms. Using $\Gamma_{\rm rel} = 40$ s^{-1} in Eq. (2) at the optimum β of 90° we can estimate the threshold spin exchange rate in this cell, and find that $\Gamma_{se}(\text{threshold}) \cong 700$ s^{-1}, which lies within the observed threshold range already mentioned.

Let us turn now to explaining the shape of $\mathcal{M}$ vs σ^L in Fig. 3(a). When Eqs. (1) and (2) are modified for the general case of elliptically polarized pump light, a direct pumping term proportional to $\sigma^L \cos\alpha$ is added to the right side of Eq. (1), and in Eq. (2) $\cos^2\beta$ must be replaced by $\langle\cos^2\beta\rangle = \frac{1}{2}\sigma_L^2 \sin^2\alpha$. Note that $\sigma^L = \sin 2\phi$ [9], where ϕ is the quarter-wave plate angle measured relative to the initial plane of polarization of the light. When $\sigma^L = \pm 1$ the coefficient of Γ_{se} in Eq. (2) reduces to zero, which is the reason for the pronounced dip in the magnitude of $\mathcal{M}$ that occurs near $\sigma^L = \pm 1$ in Fig. 3(a). [The center of the dip is shifted slightly to the right of $\sigma^L = \pm 1$ in Fig. 3(a) because the axis of the linear polarizer is not exactly at 90° relative to $\mathbf{B}_0$.] Using a different plate with much smaller retardation at 894 nm causes the dip in $\mathcal{M}$ to disappear, as expected, since σ^L then remains small for all values of ϕ. All other features of the curves in Fig. 3, such as the differences between positive and negative $\mathcal{M}$ in Fig. 3(a), including the values of σ^L at which switching occurs, are well explained when optical imperfections and birefringence are taken into account.

With sufficiently small instrumental asymmetries, spontaneous polarization would grow directly from the $1/\sqrt{N}$ statistical fluctuations in $\mathcal{M}$, where N (typically $>10^{12}$) is the number of atoms that are mixed by diffusion during the spontaneous polarization rise time. Although a residual circular polarization of the pump light, $\Delta\sigma^L$, projected along $\mathbf{B}_0$, creates an instrumental bias in $\mathcal{M}$ of order $\Delta\sigma^L \cos\alpha$, this bias could be made $\ll 10^{-6}$ since σ^L and α can each readily be adjusted to better than 10^{-3}. At thermal equilibrium, the bias in $\mathcal{M}$ created by the Zeeman splitting is of order $\mu\mathbf{B}_0/kT$, which in fields below 100 mG will be negligible.

In other future work, pumping with isotropic unpolarized light [$\langle\cos^2\beta\rangle = 1/3$ in Eq. (2)] would allow spontaneous polarization along an arbitrary magnetic field direction. In a low magnetic field, with optical perturbations removed, the spin polarization might precess freely about $\mathbf{B}_0$ for exceptionally long times. In the case of exchange pumping of ^{3}He as applied to neutron spin polarizers [8], spontaneous polarization might be particularly useful because the pumping beam direction does not have to be aligned with the static magnetic field. Finally, in applications with no buffer gas, and hence no collisional quenching of the Cs excited state, it may be necessary to pump in the wings of a hyperfine component of the absorption line to avoid depolarizing the atoms with reabsorbed radiation.

We wish to thank Warren Nagourney for help with the pump laser system, Jason Pruet for work during the early stage of the experiment, and Blayne Heckel and Peter Bender for useful discussions. This work is supported by National Science Foundation Grant No. PHY-9506361.

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