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State-selective Rabi and Ramsey magnetic resonance line shapes

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We carry out state-selective Rabi and Ramsey magnetic-resonance experiments on ground-state ^{133}Cs($F=4$) atoms. Novel line shapes are obtained, which exhibit very sharp features with a width much smaller than the inverse duration of the magnetic-resonance pulse. The sensitivity of ordinary magnetic-resonance experiments with total spin F $>1/2$ is significantly less than the Heisenberg limit, which can be exactly realized only with maximally correlated spin states. We show that the state-selective resonances yield sensitivity very close to the Heisenberg limit, without any state preparation beyond ordinary optical pumping. [S1050-2947(99)51102-5]

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Atomic magnetic-resonance experiments are very important to precision measurement. For example, magnetic resonance is used to construct sensitive magnetometers [1] and to search for permanent atomic electric-dipole moments (EDMs) [2]. The experiments measure the torque on the magnetic-dipole moment $\boldsymbol{\mu}$ of an atom in a magnetic field $\mathbf{B}$. In principle, an electric-dipole moment and a field may also contribute to the torque [2]. The torque causes the atom's angular momentum $\mathbf{F}$ to precess at the Larmor frequency $\omega_0=\mu B/\hbar F$. A measurement of the Larmor frequency determines the magnetic field or moment, or possible EDM if an electric field is also present.

In this paper, we present results of state-selective Rabi and Ramsey magnetic-resonance experiments on ^{133}Cs atoms in their $6^2S_{1/2}$, $F=4$ ground-state hyperfine level. In contrast to conventional magnetic-resonance experiments, we do not prepare and detect a net polarization $\langle \mathbf{F} \rangle$ of the atomic spins. Rather, we prepare the atoms in a specific spin state $|F=4,\ M_F=0\rangle$ and, to a good approximation, we only detect the population in the state $|F=4,\ M_F=0\rangle$ following the Rabi or Ramsey pulses. The resulting line shapes show a complex structure that differs greatly from the much simpler line shapes observed in ordinary magnetic resonance. To our knowledge these line shapes have not been observed or calculated previously for any $F>1$. The case $F=1$ has been realized in an EDM search with atomic Th [3], where a much simpler line shape is observed due to the small number of Zeeman levels. Further, we find that these line shapes exhibit sharp features with a width much narrower than the inverse duration of the Rabi or Ramsey pulses. We show theoretically that these narrow line shapes lead to an uncertainty in a measurement of the Larmor frequency that is very close to the Heisenberg limit.

It is somewhat surprising that such a straightforward method can yield an uncertainty in the Larmor frequency close to the Heisenberg limit. For a measurement with a single atom, this limit is $\Delta\omega_{HL}=(2FT)^{-1}$, where T is the total measurement time [4]. For an atom with $F=1/2$, this limit is realized with a single Ramsey method interrogation of duration T, if the populations of the two spin states can be measured with unit efficiency [4,5]. However, for $F>1/2$, the Heisenberg limit can be realized only if the atoms are prepared in ''maximally correlated'' spin states $|\Psi_{max}\rangle = 2^{-1/2}(|F,M_F=+F\rangle+|F,M_F=-F\rangle)$ [4]. In this case $\langle \mathbf{F} \rangle=0$. In conventional magnetic-resonance experiments, a sample of atoms is given a net polarization $\langle \mathbf{F} \rangle$, and the precessing magnetization of the sample is detected by optical or radio-frequency methods; such measurements have an uncertainty significantly greater than $\Delta\omega_{HL}$. The use of maximally correlated spin states has previously been discussed in the context of a composite angular momentum $F>1/2$ made up of a collection of independent spin-1/2 atoms [4]; here we refer to the spin states within a single atom. For $F=1$, $|\Psi_{max}\rangle$ can be realized by subjecting an $|F=1,\ M_F=0\rangle$ state to a $\pi/2$ pulse. However, even within the spin states of a single atom, the realization of the state $|\Psi_{max}\rangle$ is not trivial for $F>1$. In fact, for $F>1$ the maximally correlated state cannot be accessed with a pure magetic-dipole interaction from any F_z eigenstate [6]. Such states can be prepared with ''quantum control'' techniques [6], but this requires a more complex arrangement of perturbing fields.

Our experimental arrangement is illustrated in Fig. 1(a). A vapor cell magneto-optical trap (MOT) apparatus [7] is housed in a three-layer magnetic shield. Cs atoms are trapped in the MOT by a combination of six 1-cm-diam, 3.5-

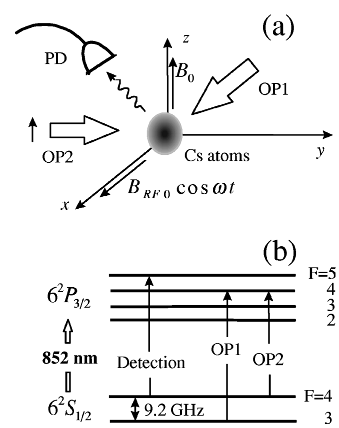

FIG. 1. (a) Schematic diagram of the experimental setup. The interaction region is in a Cs vapor cell housed in a magnetic shield not shown in the diagram. OP1 and OP2 are optical pumping beams. Detection beams (not shown) illuminate the Cs atoms from all three axes, and the flourescence signal is detected by the photodiode (PD). (b) Tuning of the optical pumping and detection beams.

mW/cm^2-intensity, -11-MHz detuning laser beams, a repumping laser beam, and a quadrupole magnetic field with an axial gradient of 10 G/cm. At the beginning of each experiment cycle, the MOT is loaded for 1 s. This produces a 1-mm-diam cloud of 10^7 atoms at a temperature between 40 and 100 μK. The MOT laser beams and the quadrupole magnetic field are then switched off, and the Cs atom cloud freely expands and falls under the influence of gravity. Two rectangular coils inside the chamber produce a uniform magnetic bias field $B_0\hat{\mathbf{z}}$, with $B_0 \approx 70$ mG, which defines a quantization axis $\hat{\mathbf{z}}$. This field is left on continuously, and with the quadrupole field off, it induces a Larmor precession of the spins at a frequency of $\omega_0/2\pi \approx 25$ kHz. Just after the atoms are released, two optical pumping beams OP1 and OP2 are pulsed on for 5 ms. OP1 has an intensity of 3 mW/cm^2, and is tuned to the $6\,^2S_{1/2}(F=3) \leftrightarrow 6\,^2P_{3/2}(F=4)$ transition. OP2 has an intensity of 5 μW/cm^2, is linearly polarized along $\hat{\mathbf{z}}$, and is tuned to the $6\,^2S_{1/2}(F=4) \leftrightarrow 6^2P_{3/2}(F=4)$ transition. The action of the two beams is to pump the atoms into the $|F=4,\ M_F=0\rangle$ state, because OP2 induces only $\Delta M_F=0$ transitions, and the $6\,^2S_{1/2}|F=4,\ M_F=0\rangle$ to $6\,^2P_{3/2}|F=4,\ M_F=0\rangle$ transition is forbidden.

After the cooling and state preparation of the atoms is completed, we drive magnetic-resonance transitions with a transverse radio-frequency magnetic field $\mathbf{B}_{RF}(t) = f(t)B_{RF0}\cos(\omega t)\hat{\mathbf{x}}$, produced by an additional set of coils. We study both Rabi and Ramsey resonances. For the Rabi resonance, the envelope function $f(t)$ is a unit amplitude square pulse of duration τ, with a pulse area $\Omega\tau=\pi$, where the Rabi frequency $\Omega=\mu B_{RF0}/2F\hbar$. For the Ramsey resonance, $f(t)$ consists of two unit amplitude square pulses of duration τ, separated by an interrogation time T where, in this case, the pulse area of each square pulse is chosen as $\pi/2$. After the radio-frequency pulse or pulses, we detect the atoms in a state-selective manner. We do this by first pulsing on OP2 for 2 ms. This removes the population from all $F=4$ Zeeman sublevels except for $M_F=0$. A calculation of this optical pumping process shows that while a few of these atoms are pumped into the $|F=4,\ M_F=0\rangle$ state, most are pumped into the lower hyperfine ($F=3$) state. Finally, we illuminate the atoms with a short pulse of laser light tuned to the $6\,^2S_{1/2}(F=4) \leftrightarrow 6\,^2P_{3/2}(F=5)$ cycling transition and we measure the induced fluorescence with a photodiode. In order to normalize these measurements, after each measurement cycle with the rf on, we repeat the same cycle, but with the rf amplitude set to zero. We plot the ratio of the signal level with the rf on to that with the rf off. To a good approximation, this yields the fraction of the atoms, which remain in the $|F=4,\ M_F=0\rangle$ state after the application of the rf pulse or pulses, having been prepared in that state initially.

Typical results, showing the normalized fluorescence signal as a function of the rf drive frequency ω, are plotted with a solid line in Fig. 2 for the Rabi method and in Fig. 3 for the Ramsey method. If we were to prepare and detect a net polarization $\langle\mathbf{F}\rangle$ of the atoms, these would yield the familiar Rabi and Ramsey resonance line shapes [8]. However, with the state-selective preparation and detection, a much more complex resonance line shape is observed. For the Rabi resonance, we used a π pulse of duration $\tau=3.0$ ms. The line shape consists of an envelope that is much wider than $1/\tau$, in contrast to the ordinary Rabi line shape whose width is $0.80/\tau=267$ Hz. Further, within the envelope, the line shape exhibits a series of sharp maxima and minima. The transition probability out of the $|F=4,\ M_F=0\rangle$ state is zero rather than 1 in the exact center of the line. A remarkable feature is the narrow 56-Hz width of the central fringe, which is much less than the inverse pulse duration. The same phe-

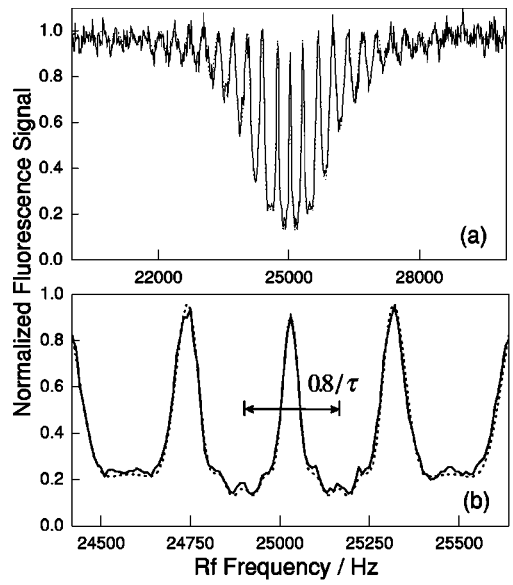

FIG. 2. (a) Rabi resonance line shape with a 3-ms π pulse. Solid line, experimental results; dotted line, theoretical line shape with an inhomogeneous broadening $\Delta\omega_B/2\pi=27$ Hz, and a Larmor frequency $\omega_0/2\pi=25\,040$ Hz. (b) Magnified view of the central fringes. The full width at half maximum (FWHM) of the central fringe is 56 Hz, and is much narrower than the width of $0.8/\tau$ for an ordinary Rabi resonance line shape. Also, the envelope of the fringes is much wider than $0.8/\tau$.

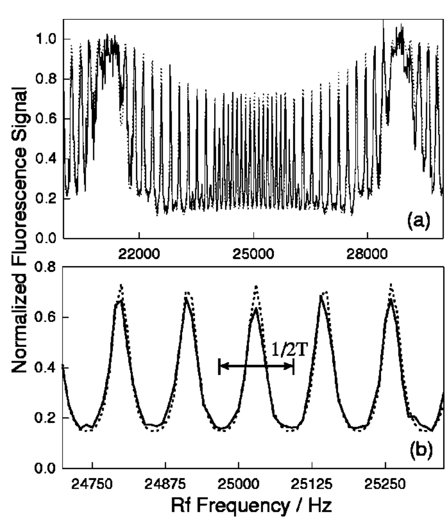

FIG. 3. (a) Ramsey resonance line shape. Data is obtained by applying two $\pi/2$ pulses of the same duration $\tau=0.25$ ms with an interrogation time $T=4$ ms between them. Solid line, experimental results; dotted line, theoretical line shape, with an inhomogeneous broadening $\Delta\omega_B/2\pi=27$ Hz and a Larmor frequency $\omega_0/2\pi=25\,040$ Hz. (b) Magnified view of the central fringes. The FWHM of the central fringe is 36 Hz, and is much narrower than $1/2T$.

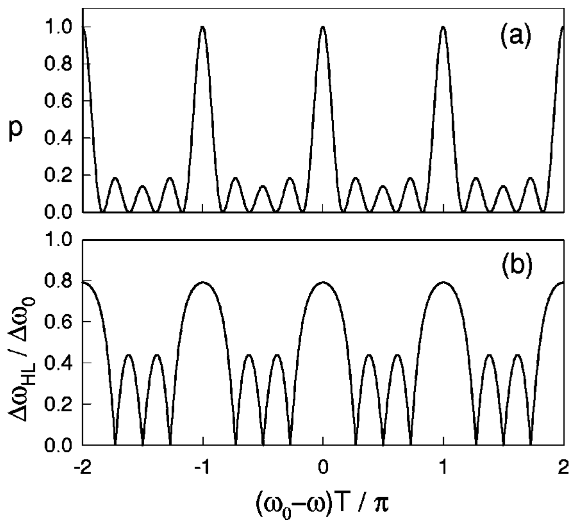

FIG. 4. (a) Theoretical line shape for the $F=4$, $M_F=0$ state-selective Ramsey fringe, for ideal detection, negligible broadening, and a vanishing $\pi/2$ pulse duration. (b) Inverse of the statistical uncertainty $\Delta\omega_0$ in a measurement of the Larmor frequency for the line shape shown in (a), in units of the Heisenberg limit $\Delta\omega_{HL}=(2FT)^{-1}$.

nomenon is seen in the Ramsey fringe, where we used two $\pi/2$ pulses of duration $\tau=0.25$ ms and an interrogation time $T=4$ ms. In the ordinary Ramsey fringe line shape, the width of the central fringe in Hz is $1/2T$, which corresponds to 125 Hz in our case, whereas our data show a central feature of width 36 Hz.

The results agree very well with calculated line shapes, shown as the dotted lines in Figs. 3 and 4. The Schrödinger equation describing our experiment is

$$i\hbar\frac{d}{dt}|\psi(t)\rangle=-\frac{\mu}{F}[B_0\hat{\mathbf{z}}+f(t)B_{RF0}\cos\omega t\hat{\mathbf{x}}]\cdot\hat{\mathbf{F}}|\psi(t)\rangle. \tag{1}$$

For $F=4$ ground-state Cs, μ is equal to the Bohr magneton. We then switch to a representation of the wave function in the corotating frame [8] by writing $|\psi(t)\rangle=e^{-i\omega t\hat{\mathbf{F}}_z/\hbar}|\varphi(t)\rangle$ and, making the rotating-wave approximation, find

$$i\hbar\frac{d}{dt}|\varphi(t)\rangle=(\Delta\hat{\mathbf{F}}_z+\Omega f(t)\hat{\mathbf{F}}_x)|\varphi(t)\rangle, \tag{2}$$

where $\Delta=\omega_0-\omega$ is the detuning of the rf frequency from resonance. The solution of Eq. (2) is

$$|\varphi_f\rangle=e^{-i(\Delta\hat{\mathbf{F}}_z+\Omega\hat{\mathbf{F}}_x)\tau/\hbar}|\varphi_i\rangle \tag{3}$$

for the Rabi method, and

$$|\varphi_f\rangle=e^{-i(\Delta\hat{\mathbf{F}}_z+\Omega\hat{\mathbf{F}}_x)\tau/\hbar}e^{-i\Delta\hat{\mathbf{F}}_zT/\hbar}e^{-i(\Delta\hat{\mathbf{F}}_z+\Omega\hat{\mathbf{F}}_x)\tau/\hbar}|\varphi_i\rangle \tag{4}$$

for the Ramsey method, where $|\varphi_i\rangle=|F=4,\ M_F=0\rangle$ is the initial state and $|\varphi_f\rangle$ is the atom state at the time of detection. For an ideal state-selective experiment we would measure $|\langle F=4,\ M_F=0|\varphi_f\rangle|^2$. We also consider two nonideal effects in our model. First, we include the small contribution to the signal from the atoms not in the $|F=4,\ M_F=0\rangle$ level by summing the occupation probability of all the Zeeman sublevels times their probability of decaying to the $|F=4,\ M_F=0\rangle$ level during the 2-ms OP2 pulse. Second, we assume that the resonance frequencies ω_0 have a Gaussian distribution of a half width $(1/e)$ $\Delta\omega_B$ to account for possible broadening due to magnetic-field inhomogenity, and compute the signal averaged over this distribution. In the model, the rf field amplitude and frequency are taken to be their experimentally measured values. The central value of ω_0 and its width $\Delta\omega_B$ are adjusted to produce a good fit to the data.

As shown in Figs. 2 and 3, our model fits the data very well. In both cases, we set the resonance frequency $\omega_0/2\pi=25\,040$ Hz and $\Delta\omega_B/2\pi=27$ Hz. The main features of the resonance line shapes can be easily understood. As indicated by Eq. (3), the Rabi pulse effectively rotates the initial state by an angle $\varphi=(\sqrt{\Delta^2+\Omega^2})\tau$ around the axis $\hat{\mathbf{n}}=(\Delta\hat{\mathbf{z}}+\Omega\hat{\mathbf{x}})/\sqrt{\Delta^2+\Omega^2}$. The transition probability goes to zero whenever φ equals an integer multiple 2π. In addition, for an initial state $|F=4,\ M_F=0\rangle$ the transition probability is zero when $\Delta=0$ and $\Omega\tau=\pi$, because a π rotation about $\hat{\mathbf{x}}$ leaves the $|F=4,\ M_F=0\rangle$ state unchanged. Further, because there are many Zeeman levels, a rotation through only a small angle is required to cause a significant decrease in the projection of the state onto $|F=4,\ M_F=0\rangle$. This explains both the narrow width of the central fringes and their broad envelope. Alternatively, one can understand the narrow features as being analogous to a multiple slit interference fringe.

The narrow central fringes suggest that very good statistical uncertainty should be possible in a measurement of the Larmor frequency. At present, we are limited by technical noise rather than statistical noise, but it is interesting to calculate the theoretical uncertainty. In Fig. 4(a), we show the theoretical projection $p=p(\omega)=|\langle F=4,\ M_F=0|\varphi_f\rangle|^2$ for the case where the nonideal effects discussed above are absent, and τ is negligibly small compared to T. If we assume that the noise in the experiment is limited only by the "projection noise," during the final measurement of the atom populations, then the statistical uncertainty of resonance frequency can be written as [5]

$$\Delta\omega_0=\sqrt{p(1-p)}/|dp/d\omega|. \tag{5}$$

In Fig. 4(b), we plot the ratio $\Delta\omega_{HL}/\Delta\omega_0$ for the Ramsey fringe of Fig. 4(a). As has been noted previously [5], the uncertainty can be a minimum near $p=1$ or 0, even though $|dp/d\omega|=0$, since the variance in p is also zero at those points. In the presence of a small amount of technical noise it would be desirable to operate the experiment closer to the points of maximum $|dp/d\omega|$. It is somewhat surprising that near the center of the line, the theoretical sensitivity of the state-selective Ramsey fringe reaches 79% of the Heisenberg limit, even though we employ no methods beyond ordinary optical pumping and magnetic resonance.

A possible application of this method is the determination of the electric-dipole moment d_e of the electron using laser-cooled ^{133}Cs atoms [9]. Laser-cooled atoms have the advantage that the coherent interaction times can be much longer than in a beam or cell. However, one disadvantage of ^{133}Cs is its large nuclear spin of $I=7/2$. This reduces the Larmor frequency by a factor of $2I+1=8$, relative to the case for

$I=0$. This tends to reduce the sensitivity of the experiment with conventional methods, relative to $I=0$, since eight times less Larmor phase is accumulated for the same precession time. However, if the Heisenberg limit $\Delta\omega_{HL}$ can be realized, the statistical error Δd_e in a measurement of d_e is independent of I; our work shows that it is possible to nearly realize this limit with straightforward techniques, even for $I=7/2$.

In summary, we investigated Rabi and Ramsey magnetic-resonance line shapes with state-selective preparation and detection in an $F=4$ spin system. The line shapes show a novel structure that exhibits sharp minima and maxima, with a width much less than the inverse duration of the magnetic-resonance interrogation. Due to their sharp structure, we find that the statistical uncertainty in a measurement of the Larmor frequency with these line shapes can be very close to the Heisenberg limit. The result presented in this paper can be generalized to any spin system. Furthermore, the ^{133}Cs atom we study here is of fundamental interest because of its application to the search for an electron EDM. The large nuclear spin of ^{133}Cs does not necessarily significantly impair the statistical sensitivity of such an experiment.

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