Atomic Pseudospin Resonance

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A new type of resonance is discovered in Rydberg atoms placed in a constant magnetic field $\vec{B}$ and a transient electric field that rotates at the constant frequency $\vec{\omega}$ in a plane perpendicular to $\vec{B}$. The dynamics is explained in terms of two pseudoparticles with spin $\frac{1}{2}$ in two generalized magnetic fields. The resonance frequency is predicted and found at $\vec{\omega} = (e/2m)\vec{B}$, where $-e/m$ is the electron's charge-to-(reduced)mass ratio. We discuss the applicability of the resonance to accurate magnetic field measurements and the prospects for determining e/m with improved precision.

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A resonance phenomenon analogous to nuclear magnetic resonance (NMR) [1] and electron spin resonance (ESR) [2] has been discovered in Rydberg atoms. The phenomenon can be understood in terms of the intrashell dynamics of hydrogenlike Rydberg atoms in weak, time-dependent electric and magnetic fields. It was recently recognized that this dynamics is exactly reducible to two independent pseudospin-$\frac{1}{2}$ problems [3]. These not only explain the new phenomenon, atomic pseudospin resonance (ApSR), but also expose clearly its similarity with NMR and ESR. We present the first theoretical analysis and experimental implementation of the ApSR and argue that it can be developed into a precise and sensitive technique for the measurement of magnetic fields.

The intrashell dynamics of hydrogenic Rydberg atoms in weak fields was studied already some time ago within the framework of distant collisions [4]. The problem was revisited recently [5–7] and extended to arbitrary field configurations and time dependencies [3,8,9]. The two spin-$\frac{1}{2}$ states were treated separately (spin-orbit coupling lifted), and in the analysis full advantage was taken of the O(4) symmetry of the Coulomb potential [10]. The angular momentum and Runge-Lenz-Pauli operators, $\vec{l}$ and $\vec{a}$, respectively, commute with the Hamiltonian [11]. This reflects the spherical and the additional "hidden" symmetries of the Coulomb potential. The n^2 eigenstates of a shell with principal quantum number n are described fully in terms of the two operators [11] $\vec{j}_{1,2} = \frac{1}{2}(\vec{l} \pm \vec{a})$, which commute, obey angular-momentum commutation rules, and take integer or half-integer values given by $\vec{j}_1^{\,2} = \vec{j}_2^{\,2} = j(j+1)$ with $j = \frac{1}{2}(n-1)$ [12]. In the presence of external electric, $\vec{E}$, and magnetic, $\vec{B}$, fields the Hamiltonian is $H = H_0 + \vec{j}_1 \cdot \vec{\omega}_1 + \vec{j}_2 \cdot \vec{\omega}_2$, where $\vec{\omega}_{1,2} = \vec{\omega}_L \pm \vec{\omega}_S$ (with $\vec{\omega}_L = \frac{1}{2}\vec{B}$ and $\vec{\omega}_S = \frac{3}{2}n\vec{E}$) are generalized magnetic fields. The atomic pseudospins $\vec{j}_1$ and $\vec{j}_2$ thus develop independently of each other like magnetic dipoles in external magnetic fields, i.e., $\vec{j}_1$ precesses about $\vec{\omega}_1$ and $\vec{j}_2$ about $\vec{\omega}_2$. The Majorana theorem allows each of the two pseudospin-j problems to be reduced to the product of $2j$ identical and independent pseudospin-$\frac{1}{2}$ problems [13]. This reduction holds for arbitrary time dependencies of $\vec{\omega}_{1,2}$. The intrashell dynamics is thus described fully by combinations of only two spin-flip probabilities p_1 and p_2, one for each of the pseudospin-$\frac{1}{2}$ problems.

In a more general context, the entire wealth of results obtained for the spin-$\frac{1}{2}$ problem can be applied to atomic pseudospins, for instance, concerning the quantum control issue [14]. It should be emphasized, however, that although full control could in principle be realized for a spin-$\frac{1}{2}$ particle, this is not so for higher values of the spin j due to the limited number of dynamical variables involved [8]. Even though it is possible to populate selectively some final states from some initial states, it is generally not possible to tailor external electric and magnetic fields so as to ensure complete population transfer between arbitrarily selected initial and final states. The number of dynamical variables is increased when the atomic potential deviates from a pure Coulomb potential in a core region near the origin. This leaves more room for quantum control, but in Rydberg atoms of high n the vast majority of states that do not overlap the ionic core are still hydrogenic, so the above conclusion holds in general.

In considering magnetic resonance phenomena, a particularly important field configuration consists of a constant field, $\vec{B}$, and a transient one, $\vec{B}_\omega$, that rotates at constant frequency, $\vec{\omega}$, in a plane perpendicular to $\vec{B}$. A particle with gyromagnetic ratio γ resonates with the fields when $\vec{\omega} = -\gamma\vec{B}$. Based on the pseudospin-$\frac{1}{2}$ representation of Rydberg states it is clear that a similar resonance phenomenon should be seen for Rydberg atoms in the same field configuration or in a novel configuration not used before in which the rotating *magnetic* field is replaced by a rotating *electric* field.

The new field configuration consists of a constant $\vec{B}$ along the z axis and a transient $\vec{E}$ rotating at constant frequency, $\vec{\omega}$, in the xy plane, $E_x(t) = E_0(t)\cos\omega t$ and $E_y(t) = E_0(t)\sin\omega t$. The switching function E_0 is positive only for a short period and zero otherwise. In a corotating frame, the electron experiences a fictitious, purely paramagnetic Coriolis field given by $-(2m/e)\vec{\omega}$ [4,15]. Thus, apart from the Coulomb potential, the electron sees a constant magnetic field $\vec{B}_{\text{eff}} = \vec{B} - (2m/e)\vec{\omega}$ and a

nonrotating, perpendicular electric field, $E_0(t)$. When $\vec{B}_{\rm eff}$ is tuned through resonance at

$$\vec{\omega} = e/(2m)\vec{B} = \gamma_e \vec{B}, \tag{1}$$

the dynamics becomes nonadiabatic. A selected initial state is then depleted and the population transferred to a large number of other states belonging to the n manifold. The γ value in (1) originates from the electronic orbital motion. For comparison, γ is twice as large for free electrons. The value of $\gamma_e/(2\pi)$ is 1.3996 MHz/G.

A resonance of this nature was not observed before. Apart from exposing yet another fascinating aspect of the Coulomb potential, ApSR may have practical applications as a tool for the precise, absolute measurement of magnetic fields. As compared to proton NMR magnetometers, the ApSR method has the distinct advantage of a much larger gyromagnetic ratio resulting in a significantly larger resonance frequency for a given field size (the gain is $m_p/2mg_p = 329$, where m_p is the mass and g_p is the g factor of the proton). This makes ApSR useful at fields as weak as Earth's magnetic field ($\simeq 0.5$ G), whereas NMR requires somewhat stronger fields ($\simeq 0.5$ kG or larger). The two methods are thus complementary. It might also be possible to refine the ApSR method sufficiently that the charge-to-mass quotient of the electron, $-e/m$, can be determined with improved accuracy. The relative uncertainty is presently 4.0×10^{-8}.

If $E_0(t)$ is a hyperbolic secant pulse

$$E_0^{\rm RZ}(t) = \frac{\mathcal{E}_m}{\cosh \alpha t}, \tag{2}$$

where $\mathcal{E}_m$ and α are constants, then the nonadiabatic probabilities pertaining to the spin-$\frac{1}{2}$ problems are given in an analytical form by the Rosen-Zener model [16],

$$p_{1,2} = \frac{\sin^2[3\pi n \mathcal{E}_m/(4\alpha)]}{\cosh^2[\pi(B_{\rm eff} \pm 3nE_\parallel)/(4\alpha)]}. \tag{3}$$

We have allowed for an electric field component parallel to the rotation axis, $E_z = E_\parallel$, to show explicitly that the two independent spin-$\frac{1}{2}$ problems may have different solutions. If the electron is initially in a circular Zeeman state with spherical quantum numbers $l = m_l = n - 1$, then the probability of its survival (i.e., adiabatic development) [3]

$$\mathcal{P}_{\rm ad} = [(1 - p_1)(1 - p_2)]^{n-1} \tag{4}$$

exhibits two distinct resonance dips at $B_{\rm eff} = \mp 3nE_\parallel$, one for each of the two pseudospins. This convincingly testifies that the dynamics is indeed described by *two* independent atomic pseudospins. A specific feature is the strong amplification of the resonance effect by the large power in (4) for Rydberg states. It can be understood as a collective effect within the Majorana reduction [13].

Interference effects for atomic pseudospins are also of interest. If the switching function E_0 has the form of two identical pulses separated by a time T, then the following expressions are to be substituted into formula (4):

$$p_{1,2} = 4p_{1,2}^0(1 - p_{1,2}^0) \times \cos^2[\tfrac{1}{4}(B_{\rm eff} \pm 3nE_\parallel)T - \chi_{1,2}], \tag{5}$$

where $p_{1,2}^0$ describe single-pulse transition probabilities and $\chi_{1,2}$ are small dynamic phases. For the pulse (2), $p_{1,2}^0$ are smooth functions given by (3). The rapidly varying interference phases in (5) lead to oscillations in $\mathcal{P}_{\rm ad}$ that could be referred to as Stueckelberg oscillations [17] or Ramsey fringes [18].

Returning to the case of a single pulse, the two dips merge when $E_\parallel = 0$ ($p_1 = p_2 = p$) and one finds $\mathcal{P}_{\rm ad} \simeq 1 - (2n - 2)p$ for small p. If ΔT is the full width at half maximum (FWHM) of the pulse (2) then the relative width (FWHM) of the resonance is independent of n and given by

$$\frac{\Delta\omega}{\omega_r} = \frac{\Delta B}{B_r} = \frac{8\ln(\sqrt{2} + 1)\ln(2 + \sqrt{3})}{\pi \Delta T \omega_r} \simeq \frac{\pi}{\Delta T \omega_r}, \tag{6}$$

where ω_r and B_r are values at resonance.

The essential parts of the experimental arrangement were described in detail previously [19]. Shortly, a vertical beam of thermal Li atoms was crossed by three pulsed laser beams. The lasers were on for about 5 ns at a repetition rate of 14 Hz. They excited the Li atoms to the uppermost Stark state of the $n = 25$ manifold in the presence of a strong, horizontal electric field and a weak, vertical magnetic field. The electric field was subsequently switched off adiabatically to produce horizontal, circular Zeeman states [20,21]. This was completed 4–5 μs after each laser shot and a few μs later an electric field rotating about a vertical axis at the frequency, $f = \omega/2\pi$, was switched on and off once or twice.

The Rydberg atoms were detected by selective field ionization (SFI) in a horizontal electric field shortly after the rotating field was turned off. The circular state, which is present when $\vec{B}_{\rm eff}$ is large, field ionizes at the classical limit $E_{\rm cl}$, but when $\vec{B}_{\rm eff}$ approaches $\vec{0}$ the rotating field populates a range of states most of which ionize only by tunneling at fields much larger than $E_{\rm cl}$. The SFI spectra therefore clearly revealed the transition from the adiabatic to the nonadiabatic regime as $\vec{B}_{\rm eff}$ was tuned through $\vec{0}$. The magnetic field was varied and f held constant to accomplish this. The field was formed by a vertical solenoid, and it was varied through resonance (1) by adjustments of the current.

Three typical resonances are shown in Fig. 1. The centroids fall at the same position, but the shapes depend on the amplitude and detailed form of the switching function E_0. For the pulse (2) the relative width of a shallow resonance is $\Delta I/I = \Delta B/B \simeq 1/(2\Delta T f)$. The narrow single resonance shown in Fig. 1 was measured with a single pulse closely resembling the form (2). The width of the resonance agrees fairly well with the estimate. The

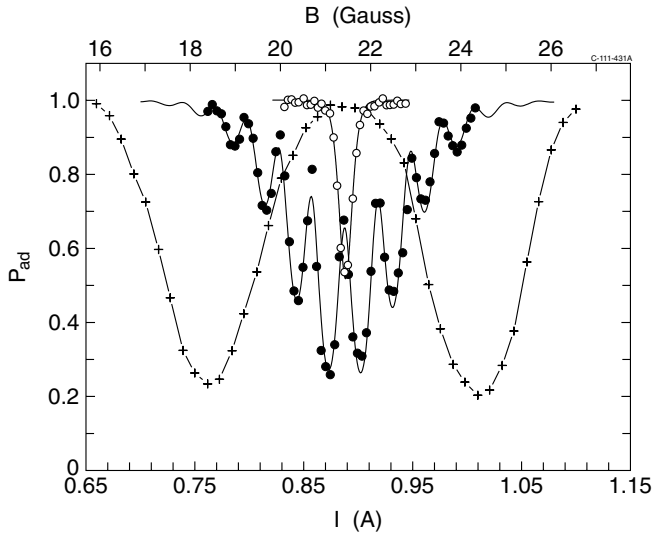

FIG. 1. Probability for adiabatic transformation, $\mathcal{P}_{\rm ad}$, vs current, I, and magnetic field, B, at $f = 30$ MHz. Open points: hyperbolic secant pulse (2) of duration $\Delta T \simeq 1.5$ μs; the smooth curve is a fit by (4) with (3); expected relative width $\Delta I/I_r \simeq 1/(2f\Delta T) \simeq 1.1\%$. Crosses: strong, sudden pulse and $E_\parallel \simeq 0.08$ V/cm leading to a wide, split resonance. Full points: two sudden pulses separated by 1 μs leading to Ramsey fringes; the smooth curve is a fit by (4) with (5) and (3).

resonance is split symmetrically about I_r when $E_\parallel \neq 0$. This is also illustrated in Fig. 1. In this particular example, the maximum electric field was strong and the pulse was switched on and off quite suddenly. This explains the larger widths and depths of the two components of the split resonance as compared to the unsplit one. The third curve in Fig. 1 shows a relatively broad resonance modulated by regular interference fringes. Two sudden pulses separated by $T = 1.0$ μs were used in this case. The period of the oscillations relative to the centroid of the resonance is $\Delta I/I_r = \Delta B/B_r = 1/(Tf) \simeq 3.3\%$. Even though the oscillating probabilities $1 - p_1$ and $1 - p_2$ each approach unity at regular intervals, the resulting probability $\mathcal{P}_{\rm ad}$ is less than unity throughout the resonance structure because the two oscillating probabilities have different phases (difference $= 0.31\pi$).

The sources of the rotating electric field were the charges (potentials) on eight vertical bars placed equidistantly on a circle to form a cylindrical cage [19] similar to the one described in [22]. The bars were driven by the harmonic voltage of a single generator modulated by the switching function. The voltage was fed to the bars through carefully adjusted delay lines to have a phase shift of exactly $\pi/4$ between consecutive bars at $f_0 = 30$ MHz. This results in a homogeneous field that rotates at the central frequency f_0 with a small frequency spread from the slowly varying switching function. If the generator is set at another frequency (delay lines unchange) the field is still quasiperiodic, and it can be perceived as consisting of two harmonic components. In complex notation, $E(t) = E_0(t)[A_+ \exp(i2\pi ft) + A_- \exp(-i2\pi ft)]$, where $A_\pm$ are constant in time but dependent on f/f_0. $A_- = 0$ at $f/f_0 = 1$ corresponding to a homogeneous, rotating field of positive helicity, but for $f \neq f_0$ there are two counterrotating fields. The ApSR can therefore be observed at any frequency, the atoms coming into resonance with one or the other rotating field, but at 30 MHz it is observable only if $\vec{B}$ and $-(2m/e)\vec{\omega}$ are antiparallel (as in the "rotating field approximation"). This is illustrated in the inset in Fig. 2.

The magnetic field strength on the axis of the solenoid was calculated by summing the contributions from the individual loops carrying a known current, and it was also measured carefully by a calibrated Hall element. The field is known to an accuracy of about 0.5%. The dependence of the resonance field B_r on f is shown in Fig. 2. The measurements imply an e/m value of $(1.7503 \pm 0.0003) \times 10^{11}$ C/kg, where we quote the statistical standard error. There is also a systematic error of 0.0088×10^{11} C/kg (0.5%). The derived e/m value thus agrees with the accepted value, $(1.7588\ldots) \times 10^{11}$ C/kg.

When a single pulse is used, the magnetic field is measured by the resonance frequency and the calibration factor, $B/f = 4\pi m/e$, is a fundamental constant. Considered as a principle for a magnetometer, the method thus connects a magnetic field with a frequency that can be determined with high precision, and it avoids system-dependent parameters. Therefore, the method does not need calibration, and it is potentially very stable and sensitive. The FWHM, Δf, of the resonance is inversely proportional to ΔT, which for a thermal beam is ≤ 25 μs, so Δf is typically larger than 20 kHz. If the centroid of the resonance curve can be determined with a precision of 1%, then the uncertainty of the resonance frequency is about 200 Hz. According to (6), this corresponds to a relative precision of 2×10^{-4} at $f = 1$ MHz or $B = 0.714$ G (Earth's

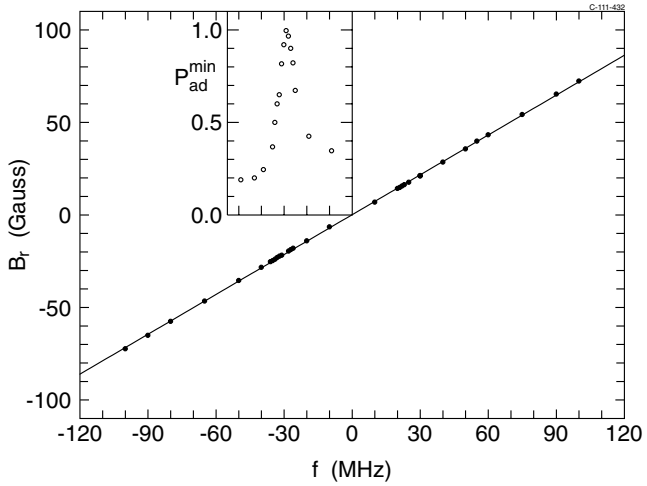

FIG. 2. Magnetic field at resonance vs frequency, $B_r(f)$, with $f < 0$ for $-\vec{B} \parallel \vec{\omega}$ and $f > 0$ for $\vec{B} \parallel \vec{\omega}$. The inset illustrates resonance suppression. It shows minimum values of probabilities for adiabatic transformation, $\mathcal{P}_{\rm ad}^{\rm min}(f)$, near $f = -30$ MHz.

magnetic field $\simeq$0.6 G). The relative precision is inversely proportional to f, reaching 2×10^{-7} at $f = 1$ GHz. The vector character of (1) makes the method quite sensitive to stray electric fields, $\vec{E}^s$, from contact potentials or bound charges on insulating surfaces. The parallel component of $\vec{E}^s$ relative to $\vec{B}$, $E^s_\parallel$, splits the resonance symmetrically and the perpendicular one, $E^s_\perp$, broadens it. Stray fields may be the limiting factor for the method at the highest level of precision.

A scalar technique, less sensitive to stray fields, is offered by the oscillations in (5), which to first order do not depend on $E^s_\perp$. The oscillations are thus governed by the two phases $[(\frac{1}{2}B \pm \frac{3}{2}nE^s_\parallel) - \frac{m}{e}2\pi f]T - \chi_{1,2}$. Measurements of the oscillations as a function of f for several values of T relates B (as well as $E^s_\parallel$, T, and $\chi_{1,2}$) to a frequency f. The precision is an increasing function of T. An atomic fountain (or a Zeeman slower) allows cold atoms to be launched into a region of a homogeneous magnetic field and to stay almost at rest within a small volume of space for as long as perhaps 25 ms. During this time circular Rydberg states can be prepared (in $\leq$10 μs) and then used as probes for the magnetic field. For $n = 50$ the lifetime is about 30 ms. This gives a theoretical precision of 2×10^{-9} at $B = 70$ G, so, if B is determined to the same precision (known coil geometry and current), it should be possible to improve the precision of e/m which presently is known to 4×10^{-8}.

In conclusion, a novel resonance in Rydberg atoms has been discovered and explained in terms of two independent pseudospins in two generalized magnetic fields. The experimental results give the most direct dynamical evidence of the reality of the *two* pseudospins, which mathematically stem from the O(4) = O(3) $\times$ O(3) relation between the group algebras.

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