Observation of a Magnetic-Field–Dependent g-Factor Ratio

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A new type of magnetic shift has been observed in high-precision measurements on the hyperfine structure of ground-state rubidium atoms. In the range of magnetic fields used, 4.6–7.8 T, the measurements in both ^{85}Rb and ^{87}Rb are consistent with a fractional shift of $-1.68(15)\times10^{-9}$ T^{-2} in the nuclear-to-electronic g-factor ratio, g_I/g_J. This effect is much larger than predicted by a second-order perturbation-theory analysis which gives other magnetic shifts in agreement with the measurements.

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The study of atomic systems in magnetic fields—a fundamental problem in physics—continues to provide new information. As has been exhaustively shown experimentally, the effect of a magnetic field on the hyperfine sublevels of a ^2S atomic ground state is given to high precision by the Breit-Rabi formula.[1] In this expression the nuclear-to-electronic g-factor ratio, g_I/g_J, and the hyperfine coupling appear as constant parameters. With increasing magnetic field or increasing resolution, deviations of the atomic levels from the predictions of the formula might be expected, because of magnetically induced changes in the structure of the atom and even the nucleus. Such changes may be expressed in terms of a magnetic field dependence of existing or possibly new parameters. Recently, we reported the results of precision measurements of the hyperfine structure of ground-state atomic rubidium in magnetic fields of up to 7.8 T, which revealed a magnetically induced change in the dipole hyperfine coupling and an induced quadrupole shift.[2] Analysis of the measurements also indicates a magnetic shift in the g-factor ratio, g_I/g_J, which is larger fractionally than the dipole hyperfine shift. Such a large g-factor shift has not been anticipated and remains unexplained since, unlike the hyperfine couplings, the nuclear and electronic g factors are, to first order, independent of the atomic environment. Thus one might expect a fractional shift which is smaller, not larger, than the fractional dipole hyperfine shift. While g-factor shifts due to recoupling of fine structure[3] and to buffer-gas collisions[4] have been observed in atomic resonance experiments in the past, a magnetic shift in the g-factor ratio of the more fundamental type reported here has not been previously observed. Our observation prompts basic questions about the origin of such a shift and may have important implications for high-field nuclear-magnetic-resonance (NMR) measurements, where g factors are assumed to be constant.

A strong external magnetic field causes the initially spherically symmetric electronic charge distribution of the ^2S ground state to shrink asymmetrically. This results in the observed increase in contact (dipole) hyperfine structure, and the induced quadrupole hyperfine interaction.[2] Associated changes in the atomic shielding of the electronic and nuclear moments,[5] and shifts due to the magnetic polarizability of the nucleus[6] have been anticipated. However, the observed shift in the g-factor ratio is much larger than would be expected from such effects based on the perturbation calculation described below, while the calculations for the dipole and quadrupole shifts are in reasonable agreement with the experimental results.

Since the experiment has been described previously,[2] only a very brief outline will be given here. Precision at the level of a few times 10^{-2} Hz was obtained in the roughly 10^9-Hz transition frequencies measured with a laser-optical pumping technique at magnetic fields of 4.6, 6.2, and 7.8 T. Microwave resonances, driven while the laser beam was blocked, were subsequently detected by use of a frequency- and intensity-stabilized laser probe beam, the circular polarization of which was photoelastically modulated to allow phase-sensitive amplification of the absorption signal. Natural-isotopic-abundance rubidium vapor was confined in four evacuated, wax-coated cylindrical glass sample cells which ranged from 1 to 2 cm^3 in volume and from 0.67 to 2.4 in length-to-diameter ratio. At each of the three values of the applied magnetic field all sixteen of the $\Delta m_J=0$, $\Delta m_I=\pm1$ transitions (ten in ^{85}Rb and six in ^{87}Rb) were measured in order to distinguish among possible shifts of different spin and isotope dependence.

The measured transition frequencies were fitted to the Breit-Rabi formula at each of the applied fields. These fits show a departure from the predictions of the formula in the form of a quadrupole ($\nu_Q m_I^2$) shift. Thus, the formula used for the hyperfine sublevels at each field is

$$\nu=\nu_c+m_I\nu_N+m_J\Delta\nu[X^2+4X(m_I+m_J)/(2I+1)+1]^{1/2}+\nu_Q m_I^2, \quad (1)$$

where $X=(g_J-g_I)\mu_0B/h\Delta\nu$, $\nu_N=g_I\mu_0B/h$ is the nuclear Zeeman frequency, $\Delta\nu=a(2I+1)/2$ is the hyperfine frequency, and ν_c is a constant with respect to m_I. When fits at the three different fields are compared, they reveal a magnetic-field dependence of the g-factor ratio as well as the dipole and quadrupole shifts. The

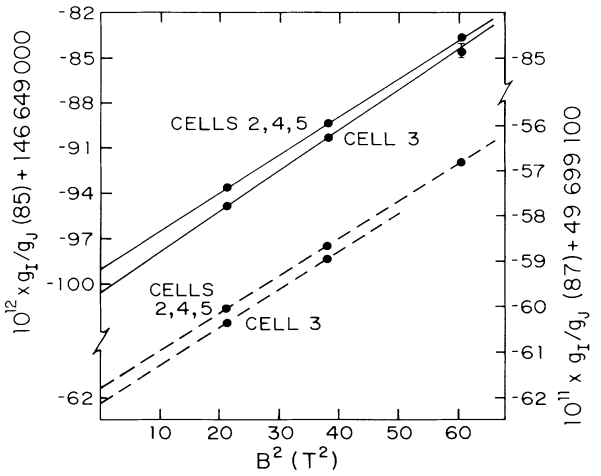

FIG. 1. Rubidium nuclear-to-electronic g-factor ratio, g_I/g_J, measured in each of four storage cells, as a function of the square of the magnetic field. The ^{85}Rb data and fits (solid lines) and the ^{87}Rb data and fits (dashed lines) are plotted with separate vertical scales which are in the ratio of the respective g_I factors. Data from cells 2, 4, and 5 are indistinguishable on the scales shown. The offset in the data in cell 3 may be due to a difference in the material used to coat the wall.

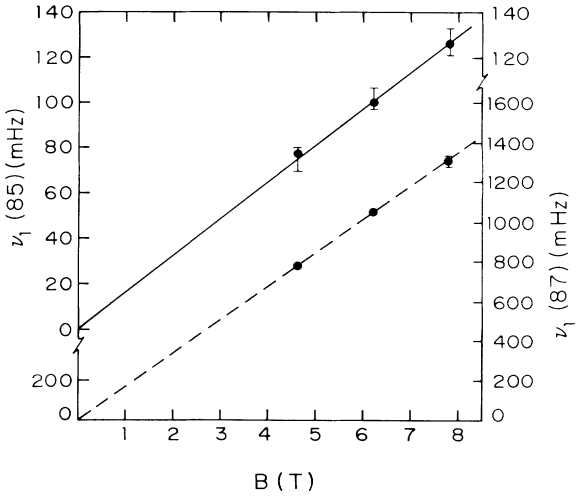

FIG. 2. Alternate representation of the rubidium g-factor ratio shift as a frequency shift ν_1 ($m_f^2 m_J$ dependent) vs magnetic field. The ^{85}Rb data and fit (solid line) and the ^{87}Rb data and fit (dashed line) are plotted with vertical scales which are in the ratio of the square of the respective g_I factors. The error bars indicate the range of values among all four cells. For each isotope the fit has been constrained to include the origin.

value of the g-factor ratio obtained from these fits is plotted as a function of B^2 in Fig. 1; the quadratic field dependence is readily apparent and may be parametrized by our setting $g_I/g_J = R(1+SB^2)$. The measured fractional shift, S, is the same in both isotopes; combining the results gives a value of $S = -1.68(15) \times 10^{-9}\ \mathrm{T}^{-2}$. The values for g_I/g_J extrapolated to zero field are $R = -1.46649099(2) \times 10^{-4}$ in ^{85}Rb and $R = -4.94991618(7) \times 10^{-4}$ in ^{87}Rb. The errors are chosen to include the differences among the cells discussed below.

Possible origins of the observed shift fall into two categories: (1) $m_I B^3$ or $m_J B^3$ energies corresponding to quadratic g-factor shifts, and (2) energy contributions proportional to $m_f^2 m_J B$. At sufficiently high fields, the two categories produce the same type of shift in the transition frequencies. A quadratic shift in g_J can have the same effect as an energy proportional to $g_f^2 m_f^2 m_J B$ because changes in the transition frequencies arising from changes in g_J are weighted by a factor of $1/B$ because of the decoupling of I and J. Thus, while in exact $m_I m_J$ coupling, the measured $\Delta m_I = \pm 1$ transition frequencies would be independent of g_I/g_J, at the fields used in these measurements the major effect of a quadratic g-factor shift is the same as a term proportional to $g_f^2 m_f^2 m_J B$. Terms in the first category must scale as g_I/g_J and terms in the second category must scale as g_f^2 for the two isotopes in order to be consistent with the observations.

The two categories are only approximately equivalent, and measurements made in fields low enough, such that $\mathbf{I}$ and $\mathbf{J}$ are strongly coupled, should distinguish between them. The present data, however, are adequately described by either form. If the frequency given in Eq. (1) is considered to be shifted by an amount $\nu_1 m_f^2 (m_J/J) = \beta_1 m_f^2 (m_J/J) B$ and g_I/g_J is taken as fixed, the effect now appears linear in B with an obvious, nonzero slope β_1, as illustrated in Fig. 2. These fits are constrained by consideration of parity to include the origin, i.e., a field-independent $m_f^2 m_J$ term has not been included. The values obtained for β_1 are 0.0162(14) Hz/T in ^{85}Rb and 0.168(15) Hz/T in ^{87}Rb, which scale as g_f^2 for the two isotopes.

In order to interpret these results, we have attempted a consistent second-order evaluation of the hyperfine, Zeeman, and diamagnetic Hamiltonian, organized in terms $H(k_I, k_S)$ containing the rank-k_I nuclear-spin tensor and the rank-k_S electronic-spin tensor.[7] Both the diagonal and the nondiagonal matrix elements of this Hamiltonian are important. The Hamiltonian terms include

$$H(0,0) = (1 - C_0^{(2)}) e^2 B^2 r^2 / 12mc^2 + \mu_0 \mathbf{L} \cdot \mathbf{B}, \quad H(1,0) = -\boldsymbol{\mu}_I \cdot \mathbf{B} + \sqrt{10} (\boldsymbol{\mu}_I^{(1)} \boldsymbol{\sigma}(r)^{(2)})^{(1)} \cdot \mathbf{B} + 2\mu_0 \mathbf{L} \cdot \boldsymbol{\mu}_I / r^3,$$

$$H(0,1) = 2\mu_0 \mathbf{S} \cdot \mathbf{B} + c_{\mathrm{s.o.}} \mathbf{L} \cdot \mathbf{S}, \quad H(1,1) = (16\pi/3) \mu_0 \mathbf{S} \cdot \boldsymbol{\mu}_I \delta(r) + 2\mu_0 \sqrt{6} (\boldsymbol{\mu}_I^{(1)} \mathbf{S}^{(1)})^{(2)} \cdot \mathbf{C}^{(2)} / r^3,$$

$$H(2,0) = -e \mathbf{Q}^{(2)} \cdot \mathbf{C}^{(2)} / r^3, \quad H(2,1) = c_{ND} \{\mathbf{N}^{(2)} \mathbf{J}^{(1)}\}^{(1)} \cdot \mathbf{B}.$$

Enumerating the various interactions, we have in $H(0,0)$ diamagnetism (of orbital rank 0 and 2) and orbital Zeeman terms. The expression $H(1,0)$ contains the nuclear Zeeman term, shielding of the nuclear moment, and the orbital–magnetic-dipole term. Spin-Zeeman and spin-orbit terms occur in $H(0,1)$. $H(1,1)$ represents the contact and spin-dipole terms, and $H(2,0)$ is the electric quadrupole interaction. Finally, $H(2,1)$ displays a term due to the anisotropy of the nuclear magnetic susceptibility in the presence of electronic and externally applied magnetic fields. Here $\mathbf{N}^{(2)}$ denotes a second-rank tensor acting on the nuclear space. In high fields this last term directly reduces to an $m_I^2 m_J B$ dependence.

Using perturbation theory, we have looked for contributions to the observed dipole, quadrupole, and g-factor ratio shifts (both categories) in first, second, and then higher order. The only term in the Hamiltonian that may contribute to such shifts in first order is nuclear diamagnetism [$H(2,1)$], but the relevant nuclear susceptibility is believed to produce a shift (<1 mHz) below our level of resolution.[6,8,9] Moreover, despite the fact that the shift has an $m_I^2 m_J B$ dependence, it is not likely to scale as observed in the two isotopes.

The largest magnetic shift appearing in second order is a cross term involving the scalar part of the atomic diamagnetism in $H(0,0)$ and the contact hyperfine structure in $H(1,1)$. The magnetic-field–induced quadrupole hyperfine interaction results from a cross term involving the electric quadrupole interaction $H(2,0)$ and the anisotropic part of the atomic diamagnetism in $H(0,0)$. These two shifts vary as $m_I m_J B^2$ and $m_I^2 B^2$, respectively, at high field, and are therefore readily distinguishable from the shift in the g-factor ratio. The anisotropic diamagnetic component in $H(0,0)$ can also induce spin-dipole hyperfine structure in combination with $H(1,1)$. This term is largely masked by the contact shift described above, but can, in principle, be decorrelated by making measurements over a wide range of fields. Although in high fields this term produces a nonzero β_1, it is only a small part of the spin-dipole effect, and should not be significant in these experiments.

The nuclear shielding term in $H(1,0)$ may also contribute to observed shifts in combination with other interactions. A diamagnetic shift in shielding, proposed by Ramsey,[5] is an $m_I B^3$ energy proportional to the g factor. The same shielding term combined with the hyperfine interaction $H(1,1)$ (both scalar and tensor) gives rise to an $m_I^2 m_J B$ effect and scales as g_I^2. Thus either term is potentially the source of the observed shift in the g-factor ratio if either is sufficiently large.

In order to estimate the size of such second-order energies, the radial portions of the perturbations have been evaluated. The inhomogeneous Schrödinger equation has been solved, with use of modified Hartree-Fock-Slater wave functions,[10] for the perturbations r^{-3}, r^{-1}, and r^2. The shift in contact hyperfine structure is calculated by the evaluating of the fractional change in the electron wave function at the nucleus due to the diamagnetic term. The approximate result is $\delta a/a = 6.5\times10^{-10}$ T^{-2}, in reasonable agreement with the experimental value, $\delta(\Delta\nu)/\Delta\nu = 5.2(2)\times10^{-10}$ T^{-2}, obtained by fitting the data with Eq. (1).[2] If the effect of the core is taken into account in evaluating the induced quadrupole interaction, the calculation yields a value of 6.8×10^{-4} Hz T^{-2}, in agreement with the experimentally determined slope of ν_Q, $7.0(6)\times10^{-4}$ Hz T^{-2}, in Eq. (1).[2] The success in the calculation of the dipole and quadrupole shifts demonstrates the validity of the technique and verifies the presumed physical origin of each of the shifts. Despite this success with the hyperfine shifts, numerical evaluation of each of the suggested contributions to g-factor shifts results in a value orders of magnitude smaller than the observed shift.

Collisions of the atoms with the sample-cell walls induce other effects, including mainly the well-known wall shift in contact hyperfine structure.[11] A nonspherical cell may be expected to induce spin-dipole and quadrupole hyperfine structure as well. In addition to a contact dipole wall shift, there is also a suggestion of wall-induced (cell-shape–related) quadrupole shifts in the experimental data. These effects appear as cell-dependent, field-independent shifts. Another such shift is the offset in the value of g_I/g_J in cell 3, which was coated with a different form of wax from that used in the other cells. This offset might be related to the g-factor shift previously observed in buffer-gas collisions.[4] A small difference in slope also occurs in cell 3. If this difference is attributed to a wall interaction proportional to the dipole wall shift, a clear wall-independent g-factor ratio shift remains, with an increased error in the fractional shift of the g-factor ratio of $\pm0.31\times10^{-9}$ T^{-2}.

It may also be possible to have apparent shifts in g factors due to paramagnetic sites on the wall, but such shifts would be likely to be cell, temperature, and time dependent.[12] The dipole–hyperfine-frequency wall shift was observed to drift with time; correcting the data for this drift improved the consistency of the dipole hyperfine results. However, no systematic effect associated with temperature or time was observed in the case of the g-factor ratio shift.

Third- and higher-order perturbations can involve interactions, such as spin orbit, which can normally be ignored for a ^2S atomic ground state. An example is the core exchange spin-orbit g-factor shift,[13] which is related to anomalous $M1$ transition moments.[14] Analogous shifts involving magnetic interactions are likely to cause magnetic shifts in the g-factor ratio at some level, as suggested by Bender.[15] If this effect is included by the assumption of a diamagnetic shift due to the $5S$-$6S$ $M1$ moment, the result is much smaller than the present experimental resolution. Finally, while nonrelativistic wave functions were used in this analysis, a g-factor shift due to the change of mass of the electron in a magnetic field has also been predicted,[16] but this shift is several orders

of magnitude smaller than the observed shift.

In summary, a field-dependent g-factor ratio or closely related shift is clearly exhibited in the data. The perturbation expansion of the Hamiltonian suggests mechanisms which have appropriate character but which numerically evaluate to be much smaller than the observed shift. Future experiments, most likely on trapped ions,[17] may have the resolution to distinguish among the possible origins of the shift, if carried out over a large range in field (from $X < 1$ to $X \gg 1$) and in more than one isotope. We would like to thank N. F. Ramsey, W. M. Itano, and S. L. Gilbert for useful discussions relating to this work. The research was supported in part by the National Science Foundation.

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