

Experimental g_J factor in the metastable $5D_{3/2}$ level of Ba^+

K. H. Knöll, G. Marx, K. Hübner, F. Schweikert, S. Stahl, Ch. Weber, and G. Werth

Institut für Physik, Universität Mainz, D-55099 Mainz, Germany

(Received 9 February 1996)

The Zeeman splitting of the metastable $5D_{3/2}$ level of $^{138}Ba^+$ in a magnetic field of 6 T has been measured in a laser-microwave double resonance experiment in a Penning ion trap. The magnetic field at the ion's position is determined by the cyclotron frequency of electrons stored in the same trap. From the ratio of both transition frequencies we obtain a g_J value of 0.799 327 8(3). As a by-product, we confirmed earlier measurements on the $6S_{1/2}$ ground-state g_J factor to 2.002 492 2(10). The precision in both experiments is sufficient to test relativistic many-body calculations. [S1050-2947(96)04307-7]

PACS number(s): 32.60.+i, 32.80.Pj

I. INTRODUCTION

The study of the deviation of the magnetic moment of a bound electron from the value calculated by LS coupling provides an important source for the influence of relativistic and binding corrections. Sophisticated methods for many-body systems have been developed to treat even heavy atomic systems with great accuracy. Thus the comparison of those calculations to experimental value is a crucial test for such theories. These calculations can be particularly well performed on alkali-metal atoms or alkali-metal-like ions. While for neutral atoms accurate experimental values of g_J factors were obtained a long time ago by classical Rabi-type atomic-beam experiments [1], precise experimental results for ions have been available only in recent years by measurements in ion traps. The ground-state g_J factor of Be^+ , Mg^+ , Ba^+ , and Hg^+ has been measured with uncertainties below 10^{-6} , and generally the agreement with theory is good. A compilation of experimental and theoretical results on ions can be found in Ref. [2].

The ion trap technique seems particularly well suited for g factor measurements since the ions occupy a small volume in space of at most a few mm^3 . Magnetic fields with high homogeneity and temporal stability for such a small volume are readily available by superconducting coils. The high field strength of those magnets makes the Zeeman splitting of ionic energy levels large, which in general makes it easier to obtain a high precision. Effects of finite observation time can, of course, be completely neglected since ions can be stored and continuously observed for many hours.

Experiments so far are restricted to the ground state of atoms or ions and to our knowledge no experimental value of an excited state exists to date that matches the precision of ground-state experiments. The metastable $5D$ levels of Ba^+ , however, might be of particular importance in view of proposals and experiments, which attempt to use the $6S_{1/2}$ - $5D_{3/2}$ quadrupole transition on a single laser-cooled Ba^+ ion to measure parity-violating effects of the weak interaction [3]. The evaluation of the weak-coupling constants from experimental data requires a very good knowledge of the atomic wave functions. The g_J factors of the $6S_{1/2}$ and $5D_{3/2}$ levels are consequently a valuable test of corresponding calculations.

For the $6S_{1/2}$ ground level we have determined the g_J factor in a previous ion trap experiment and obtained $g_J=2.002\,490\,6(11)$ [4], in good agreement with a theoretical value of $g_J=2.002\,491\,1(30)$ [5] using relativistic wave functions obtained in the coupled-cluster single- and double-excitation scheme. For the $5D_{3/2}$ level, the most precise experimental value comes from a fast ion beam laser spectroscopy experiment and gives $g_J=0.800(9)$ [5], which agrees with $g_J=0.8$ from the LS coupling scheme but is not accurate enough to observe relativistic corrections. Since the radiative lifetime of the $5D_{3/2}$ level is 48.0 ± 5.9 s [6], it is very well suited for an ion trap experiment, where long observation times can be achieved and the precision compared to other methods can be substantially improved.

II. EXPERIMENT

We used the same apparatus as in our experiment on the $6S_{1/2}$ ground state of Ba^+ [4] with some minor modifications: A quadrupole ion trap with hyperbolic shaped electrodes made of oxygen-free copper (diameter of the ring electrode $r_0=13$ mm; distance between the endcap electrodes $2z_0=18.4$ mm) was placed in the center of a superconducting solenoid at $B=5.83$ T. Two holes of 4 mm diameter in the midplane of the ring electrode opposite one another served as an entrance and exit of laser beams. Laser-induced fluorescence can be observed through one end-cap electrode made from a mesh (transmission 72%) and a light pipe that guides the fluorescence quanta to a photomultiplier placed at 1 m distance from the trap outside the superconducting magnet. The total detection efficiency including finite solid angle, losses, and photo cathode sensitivity is 4.4%.

Ba^+ ions are created by surface ionization of a few milligrams of barium chloride placed on a rhenium filament near the inner surface at the center of one end-cap electrode. Heating the filament for several seconds produced sufficient ions to fill the trap until the space-charge limit of about 10^6 ions. Simultaneously with the desired Ba^+ ions, however, we produce and store many unwanted ions such as K^+ from impurities of the rhenium filament and $BaCl^+$ from the ion source. Because of the limited storage capacity of the trap these ions have to be removed in order to confine a sufficient number of Ba^+ . We do this by excitation of the K^+ and $BaCl^+$ cyclotron frequencies at 2.3 MHz and 520

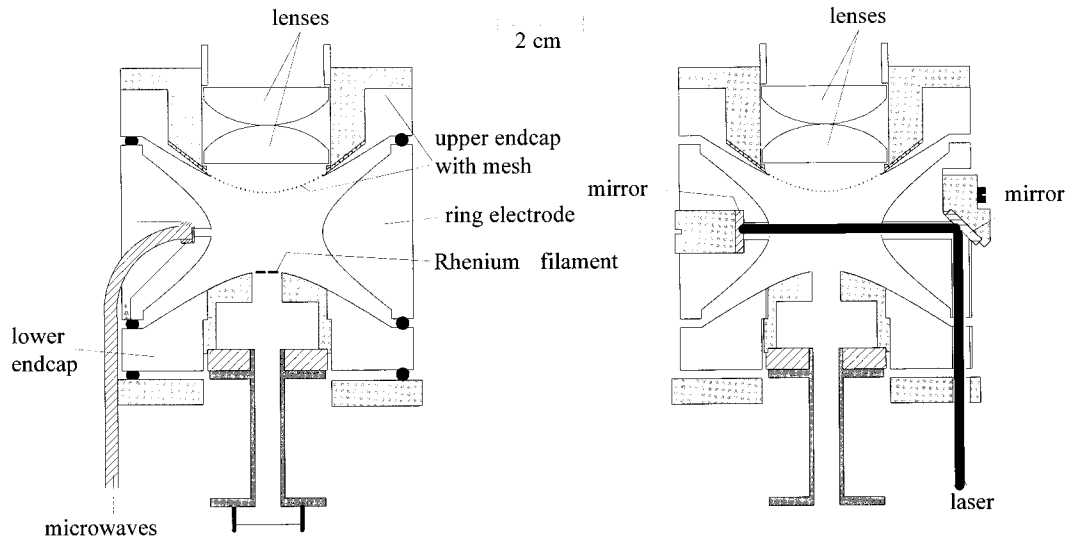


FIG. 1. Two cross sections of our ion trap ($r_0 = 13$ mm) showing the path of the laser beam and perpendicular to it the microwave guide.

kHz, respectively, with large amplitudes during ion creations. Excitation of the cyclotron orbits drives these ions out of the trap. At some distance below the endcap we mounted a tungsten wire that served as electron source for measurements of the magnetic-field strength as described below. At right angles to the laser entrance holes microwaves from the end of a waveguide were blown into the trap. Figure 1 shows a sketch of our trap.

To populate the metastable $5D_{3/2}$ state we used two lasers at 493.4 nm, which excited the two ground-state Zeeman components $m_J = \pm \frac{1}{2}$ to m_J states of the first excited $6P_{1/2}$ level (Fig. 2). At 5.8 T the splittings of the $6S_{1/2}$ ground level and the $6P_{1/2}$ excited level are 163 and 55 GHz, respectively, which far exceeds the Doppler width of the transition in the uncooled ion cloud (1.5 GHz) and the laser spectral bandwidth (6 GHz). By selection of the proper laser tunings we can excite either the $m_J = +\frac{1}{2}$ or $m_J = -\frac{1}{2}$ Zeeman level of the $6P_{1/2}$ state. As lasers, we used nitrogen pumped dye lasers with a repetition rate of 20 Hz. The metastable $5D_{3/2}$ level is then populated by radiative decay of the $6P_{1/2}$ state. The Zeeman substates of the $5D_{3/2}$ level become differently populated, depending on the laser-excited $6P_{1/2}$ Zeeman

level: Excitation of $6P_{1/2}(m_J = -\frac{1}{2})$ does not allow the decay into the $5D_{3/2}(m_J = +\frac{3}{2})$ level and excitation of the $6P_{1/2}(m_J = +\frac{1}{2})$ leaves the $5D_{3/2}(m_J = -\frac{3}{2})$ level empty. The different population numbers of he $4D_{3/2}$ Zeeman substates are used to perform a double-resonance experiment on this state: A third laser at 649.6 nm (spectral width 1.2 GHz) tuned to the $5D_{3/2}-6P_{1/2}$ E1 transition probes the population of the $5D_{3/2}$ sublevels, which are separated by 65 GHz in our 5.8 T field. As a signal, we observe the fluorescence from the spontaneous decay of the $6P_{1/2}$ level into the ground state. Using a pulsed dye laser for the $5D_{3/2}-6P_{1/2}$ transition and placing the laser pulses in time between the pulses for the $6S_{1/2}-5P_{1/2}$ lasers we can observe the fluorescence free of laser stray background by gating the output of the photomultiplier on for a time on the order of a few nanoseconds during the $5D_{3/2}-6P_{1/2}$ laser pulse. The output of the photomultiplier is digitized by an analog-to-digital converter and fed to a personal computer for further data handling. Figure 3 shows the laser-induced fluorescence when we scan the entire range of the $5D_{3/2}-6P_{1/2}$ Zeeman structure. In this picture we have excited both $m_J = \pm \frac{1}{2}$ Zeeman levels of the

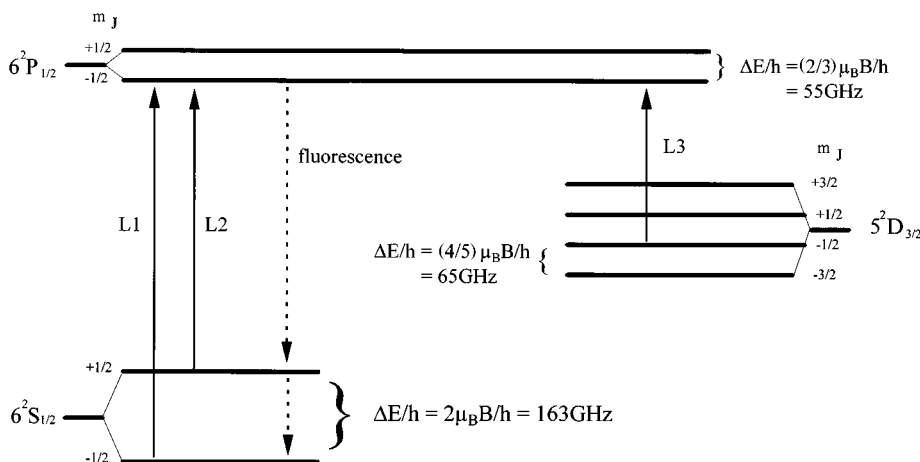


FIG. 2. Partial level diagram of Ba^+ in a magnetic field. $L1$, $L2$, and $L3$ are three lasers used to populate and prove the $D_{3/2}$ -state Zeeman levels.

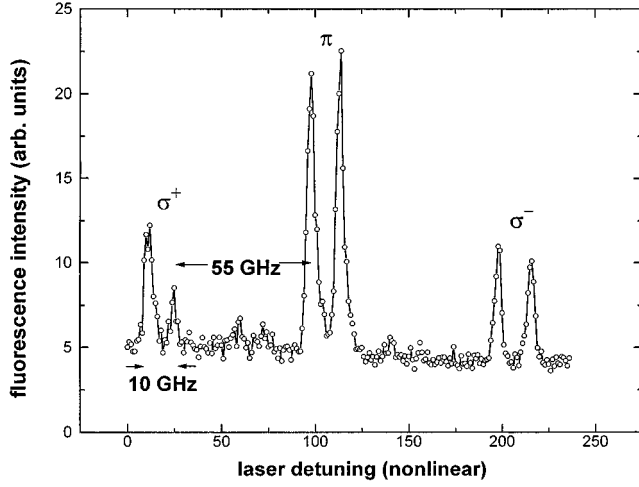


FIG. 3. Observed fluorescence spectra when we tune laser $L3$ (649 nm) across the $5D_{3/2}$ - $6P_{1/2}$ resonance. Lasers $L1$ and $L2$ are tuned to excite both m_J levels of the $6P_{1/2}$ states.

$6P_{1/2}$ state, which leads to a population of all Zeeman levels in the $5D_{3/2}$ state.

The microwaves to induce transitions between two sub-levels of the $5D_{3/2}$ state were produced in a 65-GHz-Gunn diode, which was phase locked to the fifth harmonic of a 13-GHz synthesizer. The spectral bandwidth was measured to less than 100 Hz and the frequency stability was controlled by a rubidium atomic standard. A Q -band waveguide was used to bring the microwave field into the trap. We used the same setup to guide microwaves at the cyclotron frequency of stored electrons at 164 GHz into the trap. This field was produced by frequency doubling of a 82-GHz klystron. The available output power was some $50 \mu\text{W}$, which is more than adequate to induce cyclotron transitions on the electrons.

A microwave-induced transition between Zeeman levels of the $5D_{3/2}$ state is observed as a flop-in signal: We tune the red probing laser (649.6 nm) to the π components of the $5D_{3/2}$ - $6P_{1/2}$ line. These two lines, corresponding to the two $\Delta m_J = 0$ transitions, are separated by 10 GHz, the difference between the $5D_{3/2}$ and $6P_{1/2}$ Zeeman splitting. The spectral bandwidth of the laser operated without intracavity étalon was about 30 GHz and covered both transition wavelengths simultaneously. By optical pumping both $m_J = \pm \frac{1}{2}$ states of the $5D_{3/2}$ level are depleted after several laser pulses. Consequently the observed fluorescence drops to the scatter level. Increase of fluorescence occurs if we populate an empty m_J substate by induced $\Delta m_J = 1$ transitions. This allows us to determine the transition frequency $\Delta\nu = (g_J) \times (\mu_B)(B/h)$ for the $m_J = \frac{3}{2} \rightarrow m_J = \frac{1}{2}$ and $m_J = -\frac{3}{2} \rightarrow m_J = -\frac{1}{2}$ transition. It does not allow, however, the observation of the $m_J = +\frac{1}{2} \rightarrow m_J = -\frac{1}{2}$ transition. Attempts to deplete one of the $m_J = \pm \frac{1}{2}$ states separately by a narrow-band laser and to induce microwave transitions failed because the population in the $m_J = \pm \frac{1}{2}$ substates by spontaneous decay of the $6P_{1/2}$ level was too small to be observed in our experiment.

To derive the g_J factor from the measured transition frequencies we need to know the value of the magnetic field in the trap. This is accomplished by a measurement of the cyclotron frequency $\omega_c = (e/m)B$ of free electrons in the same

trap. We store electrons after reversing the sign of the trapping voltage and observe their induced voltage in an outer tank circuit, formed by the trap's endcaps as capacitance and a connecting inductance. The circuit is tuned to the axial oscillation frequency of the electrons. Exciting the cyclotron frequency of the electrons drives them out of the trap and thus reduces the observed absorption signal.

III. MEASUREMENTS

Figure 4 shows the microwave-induced transitions $m_J = +\frac{3}{2} \rightarrow m_J = +\frac{1}{2}$ and $m_J = -\frac{3}{2} \rightarrow m_J = -\frac{1}{2}$ in the $5D_{3/2}$ state observed as an increase in fluorescence intensity on the $6P_{1/2}$ - $6S_{1/2}$ transition. We fitted the lines by Gaussians and a linear background due to ion loss during the measurement. The linewidth of about 270 kHz is attributed to unresolved sidebands at the axial oscillation frequency of Ba^+ in our trap, which at a typical trapping voltage of 10 V was 145 kHz. By repeated scans and statistical averaging we reduced the uncertainty of the center frequencies. The measured values are

$$\nu(\frac{3}{2} \rightarrow \frac{1}{2}) = 65\,239\,515.2(3.3) \text{ kHz},$$

$$\nu(-\frac{3}{2} \rightarrow -\frac{1}{2}) = 65\,191\,914.6(5.9) \text{ kHz}.$$

For the measurement of the magnetic field we applied a negative voltage to the trap's end caps and stored free electrons. Excitation of their motional eigenfrequency

$$\omega'_c = \frac{\omega_c}{2} + \left[\frac{\omega_c^2}{2} - \frac{\omega_z^2}{4} \right]^{1/2} \quad (1)$$

gives a value for B . Here $\omega_c = (e/m)B$ is the free-electron cyclotron frequency, $\omega_z = [eU/mr_0^2]^{1/2}$ the axial oscillation frequency in a trap of radius r_0 and an applied trapping voltage U , and ω'_c the slightly perturbed cyclotron frequency of the stored electron. Figure 5 shows an example of a measurement where we monitor the number of stored electrons. They are driven out of the trap when we excite their eigenfrequency ω'_c . We obtain a value of $\omega'_c/2\pi = 163\,176\,280(66)$ kHz. The correction of the measured frequency ω'_c to obtain ω_c from Eq. (1) is only 2×10^{-9} and can be neglected. The quoted frequency contains corrections from a temporal drift of the magnetic field, which was assumed as linear and measured to $(4.4 \pm 3.4) \times 10^{-10}$ h. The contribution to the frequency uncertainty is 35 kHz. We also applied a relativistic mass shift to the electron's cyclotron frequency, assuming that the mean kinetic energy is $\frac{1}{2}$ of the maximum value allowed by the trap's potential depth. Very conservatively we assigned an uncertainty of 100% to this correction, which contributes 56 kHz to the total error.

As a by-product we repeated our earlier measurements on the ground-state Zeeman splitting. When we use only one laser on the $6S_{1/2}$ - $6P_{1/2}$ transition, we obtain a population difference between the ground Zeeman levels by optical pumping, which is reduced if we induce a transition between the two m_J states. It is indicated by an increase in laser

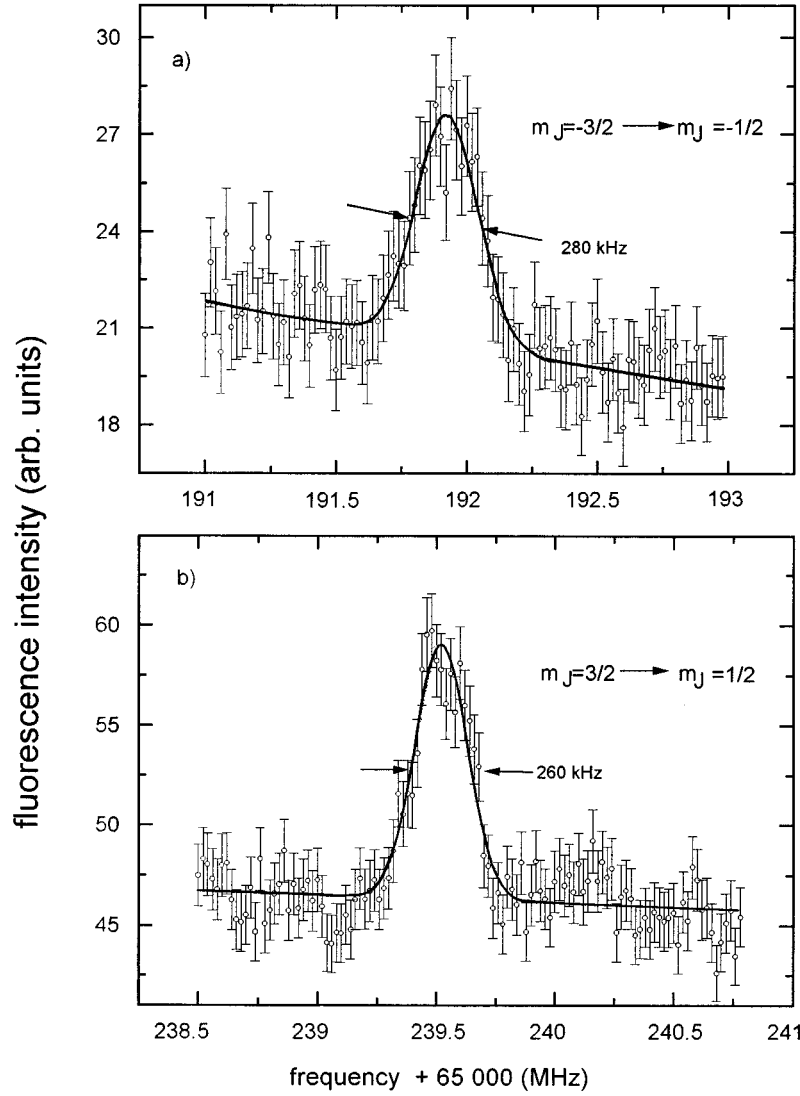


FIG. 4. Microwave-induced Zeeman transitions between the $m_J = \frac{3}{2} \rightarrow m_J = \frac{1}{2}$ levels (a) and the $m_J = -\frac{3}{2} \rightarrow m_J = -\frac{1}{2}$ states level (b) of the $5D_{3/2}$ state.

induced fluorescence. The frequency is close to the free electrons cyclotron frequency. Figure 6 shows an example. The measured transition frequency after averaging over several sweeps is

$$\nu = 163\,379\,633(14) \text{ kHz.}$$

IV. DISCUSSION AND RESULTS

The frequencies of the $m_J = -\frac{3}{2} \rightarrow m_J = -\frac{1}{2}$ and $m_J = \frac{3}{2} \rightarrow m_J = \frac{1}{2}$ transitions differ by about 47.6 MHz. This difference is caused by a quadratic B -field dependence of the energy levels. It arises (a) from diamagnetic corrections and (b) from a mixture of finestructure levels $5D_{3/2}$ and $5D_{5/2}$ in the external magnetic field by incomplete LS coupling.

(a) *Diamagnetic correction.* If we describe the interaction of the Ba^+ ion with the vector potential \vec{A} of the magnetic field by a single-particle wave function in the spherical potential $\phi(r)$ of the Ba^{2+} core, we have the Hamiltonian from the Dirac equation

$$\begin{aligned} H = & mc^2 e \phi(r) + \frac{1}{2m} (\vec{p} + e\vec{A})^2 + \frac{e}{m} \vec{S} \cdot \vec{B} \\ & + \xi(r) [\vec{r} \times (\vec{p} + \vec{A})] \cdot \vec{s} + \frac{\hbar^2 e}{8m^2 c^2} \Delta \phi(r) \\ & - \frac{1}{8m^3 c^2} [(\vec{p} + e\vec{A})^2 + 2e\vec{s} \cdot \vec{B}]^2. \end{aligned} \quad (2)$$

The first two terms are spin- and angular-independent rest and potential energies. The kinetic-energy term $\frac{1}{2}(\vec{p} + e\vec{A})^2$ can be expanded:

$$\frac{1}{2m} (\vec{p} + e\vec{A})^2 = \frac{p^2}{2m} - \vec{\mu}_L \cdot \vec{B} + \frac{e^2}{2m} A^2(r), \quad (3)$$

$$\vec{\mu}_L = -g_1 \mu_B \vec{L} / \hbar.$$

The third term describes the interaction of the spin's magnetic moment with magnetic field. The fourth term gives the spin-orbit interaction $\xi(r) \vec{L} \cdot \vec{S}$, $\xi(r) = (e/2m^2 c^2)(1/r)(d\phi/dr)$, and a linear term $\xi(r) (\vec{r} \times e\vec{A}) \cdot \vec{S}$, called the Breit-

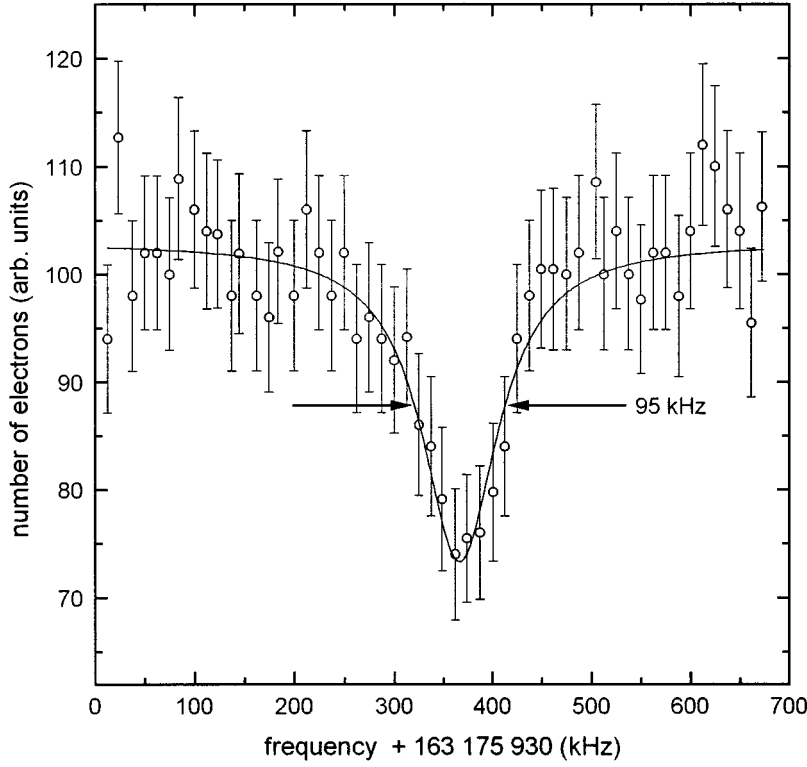


FIG. 5. Cyclotron resonance of stored electrons.

Margenau correction. The Darwin term $(\hbar^2 e/8m^2 c^2)\Delta\phi(r)$ is spin and angular independent. The leading part in the last expression in Eq. (2) is a correction $-p^4/8m^3 c^2$ to the kinetic energy, independent of spin and angular coordinates.

The quadratic energy dependence of the energy levels in the $5D$ states arising from the diamagnetic Hamiltonian $H_D = (e^2/2m)A^2(r)$ [Eq. (3)] is obtained as we set

$$\vec{A} = \frac{1}{2}(\vec{B} \times \vec{r}). \quad (4)$$

We then have

$$H_D = \frac{e^2}{8m} B^2 r^2 \sin^2 \theta = \frac{e^2}{8m} B^2 r^2 \left\{ \frac{2}{3} - \frac{4\sqrt{\pi}}{3\sqrt{5}} Y_{20}(\theta, \varphi) \right\}, \quad (5)$$

where Y_{20} is the Legendre polynomial for $L=2$, $m=0$. Calculating $\langle J, m_J | Y_{20} | J, m_J \rangle$ and setting $\bar{r}^2 = \int_0^\infty r^2 dr |\Psi(r)|^2$, we obtain

$$\langle J, m_J | H_D | J, m_J \rangle = c_D \left(\frac{15}{4} + m_J^2 \right) B^2, \quad (6)$$

$$c_D = \frac{\bar{r}^2 e}{60m}. \quad (7)$$

(b) *Mixing of fine-structure states.* In our strong magnetic field, LS coupling is not completely preserved. Second-order perturbation theory gives additional energy corrections to the Zeeman levels in the $5D_{3/2}$ states:

$$E_{J, m_J}^{(2)} = \sum_{j \neq j'} \frac{\langle j' m_{j'} | H_p | j m_j \rangle^2}{E_{j'} - E_j}. \quad (8)$$

The perturbation Hamiltonian H_p contains the sum of all spin- and angular-independent terms in Eq. (2). $E_{j'} - E_j$ can be

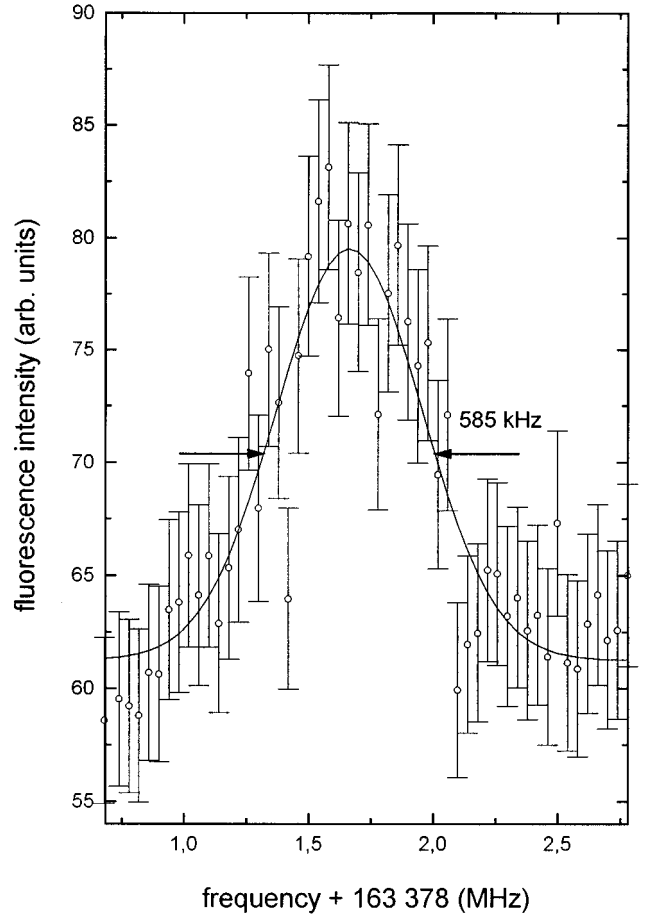


FIG. 6. Zeeman resonance in the $6S_{1/2}$ ground state of Ba^+ .

reduced to the energy difference of the fine-structure levels $5D_{3/2}$ and $5D_{5/2}$. Additional terms with different angular momenta L can be neglected here. If we reduce H_P to the Paschen-Back operator $H_{PB} = -(\vec{M}_L + \vec{M}_S) \cdot \vec{B}$, where M_L and M_S are the magnetic moments of orbital and spin angular momenta, we obtain

$$E_{3/2}^{(2)} = -c_{PB} \left(\frac{25}{4} - m_J^2 \right) B^2, \quad (9)$$

$$c_{PB} = \frac{2(g_S - g_L)^2 \mu_B^2}{125 \xi^2 \hbar^2}. \quad (10)$$

As the next order in the perturbation series we obtain a cubic dependence of the energy levels on the magnetic field:

$$E_{3/2}^{(3)} = c_3 \left(\frac{25}{12} m_J - \frac{1}{3} m_J^3 \right) B^3, \quad (11)$$

$$c_3 = \frac{24(g_S - g_L)^3 \mu_B^3}{3125 \xi^2 \hbar^4}. \quad (12)$$

Using the experimentally determined fine-structure splitting $h\xi = 9.604\,819\,326\,847\,2(160)$ THz [8], we obtain

$$c_3 = 0.23 \text{ kHz}/T^3. \quad (13)$$

Summarizing the contributions from (a) and (b) we obtain for the two observed transition frequencies:

$$\nu(m_J = -3/2 \rightarrow m_J = -1/2) = g_J \mu_B B / \hbar - 2(c_{PB} + c_D) B^2 + c_3 B^3, \quad (14)$$

$$\nu(m_J = +3/2 \rightarrow m_J = +1/2) = g_J \mu_B B / \hbar + 2(c_{PB} + c_D) B^2 + c_3 B^3. \quad (15)$$

The sum of the two observed frequencies is independent of any quadratic contribution and we have

$$g_J \mu_B B + c_3 B^3 = 65\,215\,714.9(3.3) \text{ kHz}. \quad (16)$$

The third-order correction gives at our field of $B = 5.83$ T a contribution of 45.5 kHz and we have

$$g_J \mu_B B = 65\,215\,669.4(3.3) \text{ kHz}. \quad (17)$$

The value of the magnetic field is taken from the cyclotron frequency of stored electrons extrapolated to the time when we performed the Zeeman measurements. We obtain for the g_J factor of the $5D_{3/2}$ state

$$g_J = 0.799\,327\,8(3). \quad (18)$$

The quoted error is the quadratic sum of the statistical uncertainty (1σ) of the Zeeman transition frequencies (6.7 kHz) and the error of the electron cyclotron frequency (66 kHz). We add quadratically a possible systematic uncertainty of 50 kHz, which is one-half of the full linewidth of a typical electron cyclotron frequency to account for the fact that the electrons might not occupy the same trap volume as the Ba^+ ions and thus might react to a different magnetic field. We consider this estimate of a systematic uncertainty as rather conservative, since its relative value of 5×10^{-7} is

larger than the quoted magnetic-field inhomogeneity for a reasonable estimate for a different trapping volume for the two species.

From the difference of the two Zeeman transition frequencies (13) and (14) we obtain a value for the quadratic contribution to the splitting:

$$4(c_{PB} + c_D) B^2 = 47\,600.6(6.7) \text{ kHz}. \quad (19)$$

The correction $c_{PB} B^2$ arising from the fine-structure mixing of the two $5D$ states can be calculated from Eq. (10). We obtain for our field

$$4c_{PB} B^2 = 44\,076.9 \text{ kHz}. \quad (20)$$

The diamagnetic correction is the difference between this calculated value for $c_{PB} B^2$ and the measured sum of both parts:

$$c_D B^2 = 880.9(1.7) \text{ kHz}. \quad (21)$$

Using Eq. (8) we derive a value for the mean-square radius of Ba^+ $5D_{3/2}$ of $\bar{r}^2 = 3.672(7) \text{ \AA}^2$.

From the measurement of the ground-state Zeeman splitting and the electron cyclotron frequency we obtain a value for the $6S_{1/2} g_J$ factor

$$g_J(6S_{1/2}) = 2.002\,492\,2(10). \quad (22)$$

Again we have added one-half of the full linewidth at half maximum of the electron cyclotron resonance as a possible systematic uncertainty to the statistical error. Our value is somewhat higher than our previously obtained g_J factor of 2.002 490 6(12) [4], but agrees well with a theoretical calculation by Lindroth and Ynnerman, who obtained $g_J = 2.002\,491\,1(30)$ [5].

The ratio of the g_J factors on the $6S_{1/2}$ and $5D_{3/2}$ states is independent of the magnetic field value, which represents the otherwise largest uncertainty in our experiment. Only the statistical uncertainties contribute and we obtain

$$g_J(6S_{1/2}) / g_J(5D_{3/2}) = 2.505\,220(2)(8 \times 10^{-7}). \quad (23)$$

V. CONCLUSION

The experimentally obtained value of the g_J factors as well as the value of the mean-square radius of Ba^+ in the $5D_{3/2}$ state should be a sensitive test for relativistic wave functions. While for the $6S_{1/2}$ ground state such calculations have been performed [5] and agree well with our experiment, to our knowledge no such calculation exists for the $5D_{3/2}$ level except for a first-order estimate by Johnsson [9], who arrives at $g_J = 0.799\,46$ using Dirac-Hartree-Fock wave functions. Obviously a second-order calculation is required to match the experiment uncertainties.

We would like to note that a reduction of the experimental errors is possible if required by improved theoretical calculations. The experiment was performed using nitrogen-pumped pulsed dye lasers, which were technically obsolete, had frequent breakdowns, and made data taking difficult. Using state-of-the-art solid-state lasers, which are available for the required wavelength, would substantially improve the

signal-to-noise ratio. Electron cyclotron resonances can be measured up to a few times 10^{-8} by sensitive detection of their induced currents in the trap electrodes [10]. The largest systematic uncertainty from the ambiguity that the electrons might not occupy the same trap volume as the Ba^+ ions could be reduced by mapping the magnetic field inside the

trap, when we apply a bias voltage between the endcap electrodes and thus shift the center of the ion cloud.

ACKNOWLEDGMENT

Our experiment was supported by the Deutsche Forschungsgemeinschaft.

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