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Precise g_J- and g_I-factor measurements of Ba^+ isotopes

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Abstract. Laser-microwave double and triple resonance experiments were performed on clouds of Ba^+ ions confined in a Penning ion trap to induce and detect electronic and nuclear spin flip transitions. Collisions with buffer gas molecules in the trap was used to reduce the lifetime of a long lived metastable state of the ions, in which population trapping might occur, and to cool the ions to the ambient temperature. Loss of ions from the trap by collisions were prevented by coupling the magnetron and reduced cyclotron motions by an additional r.f. field at the sum frequency of the two motions. Electronic Zeeman transitions in $^{138}Ba^+$ and $^{135}Ba^+$ were observed at a full width of about 3 kHz at a transition frequency of 80 GHz. The uncertainty of the line center was 3×10^{-9}. From the magnetic field calibration by the cyclotron resonance of electrons stored in the same trap the g_J-factor for both isotopes could be determined to 2×10^{-8}. From radiofrequency induced $\Delta m_I = 1$ transitions of $^{135}Ba^+$ the nuclear g-factor could be determined 5×10^{-6}. Both measurements improve earlier results by about one order of magnitude.

PACS. 21.10.Ky Electromagnetic moments – 32.60.+i Zeeman and Stark effects

1 Introduction

The determination of g-factors has always been an important subject in atomic physics. Electronic g_J-factors serve as sensitive test of electronic wave functions, particularly for relatively simple systems such as the $S_{1/2}$ ground states of alkali atoms or isoelectronic ions. Progress in computational methods has made it possible to calculate the deviation of the g-factor from the value of the free electron, which is of completely relativistic origin, with great precision. g_I factors are basic properties of atomic nuclei and their knowledge is required for the interpretation of measured hyperfine splittings of atomic energy levels.

Classical methods of Zeeman spectroscopy, from which g_J- and g_I-factors are obtained, are optical pumping in gas cells or Rabi-type atomic beam resonance experiments. For neutral alkali atoms such measurements are reviewed by Arimondo *et al.* [1]. The development of the ion storage technique has allowed in recent years to obtain similar data also for the ions. The storage of ions in Penning traps greatly reduces the requirements on the homogeneity of the magnetic field, in a volume of about a few cm^3, and thus allow high precision measurements. Values of the g_J- and g_I-factors obtained in recent years by this method are listed in reference [2].

In this contribution we report about a new measurement of the ground state g_J-factor of Ba^+ ions in a Penning trap. We also present a precise value of the nuclear g_I-factor in $^{137}Ba^+$ obtained by direct nuclear Zeeman transitions. Ba^+ is of particular interest since accurate wave functions are required for the investigation of parity violation effects in these ions. An experiment to search for such an effect is presently under way [3]. Calculations of the $6S_{1/2}$ g_J-factor have been performed by Lindroth and Ynnerman [4] using accurate relativistic wave functions obtained in the Coupled-Cluster Singles and Doubles (CCSD) approximation, including correlations due to the Coulomb as well as the Breit interaction. In a previous experiment [5] we obtained a value for the g-factor in agreement with the calculated value within an accuracy of 0.5 ppm. As shown below, we have now succeeded to improve the accuracy by more than one order of magnitude.

The motivation for a direct determination of nuclear g-values arises from the fact that the high detection sensitivity of the ion storage technique allows spectroscopic experiments on rare species, particularly on radioactive isotopes. As shown in previous experiments on unstable Ba^+ and Eu^+ isotopes using Paul traps [6,7] hyperfine splittings of ionic ground states can be measured with great precision. The determination of the g_I-values of the same isotopes would allow a determination of the hyperfine anomaly, which is a measure of the distribution of the nuclear magnetization over an extended nuclear volume [8,9]. A systematic measurement in a chain of isotopes might improve our understanding of that effect [10]. We consider the determination of the g_I-factor of the stable isotope $^{137}Ba^+$, which we present in this paper, as a feasibility test for such systematic measurements.

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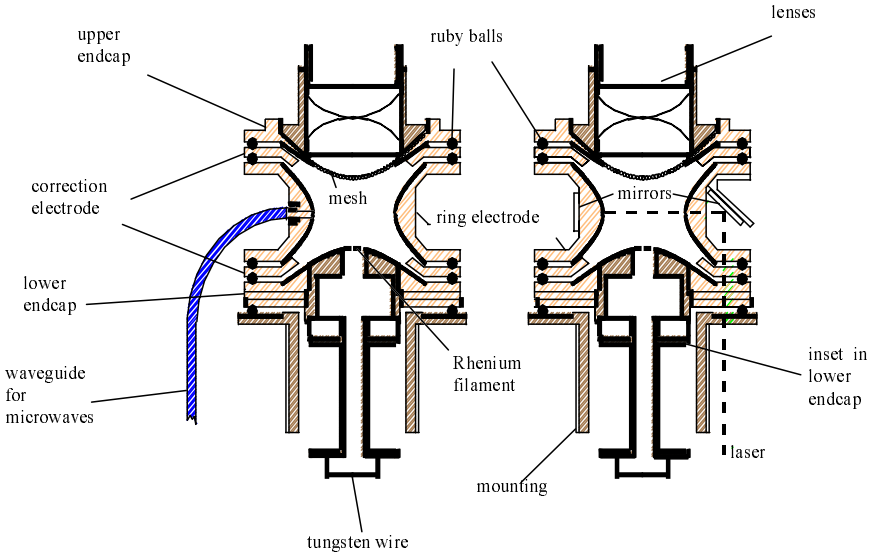

Fig. 1. Penning ion trap used in our experiment. The two sketches are at 90° to each other showing the laser beam direction and the microwave guide, respectively. The magnetic field is directed along the trap axis.

2 Experiment

2.1 Apparatus

Our experimental setup (Fig. 1) is described in reference [11]: a Penning ion trap of 13 mm radius of the hyperbolic shaped ring electrode was placed at the center of the horizontal room-temperature bore of a superconducting solenoid of 2.89 T magnetic field strength. Guard electrodes were placed between the ring and the endcap electrodes to compensate partially for possible trap imperfections. Ions were created by surface ionization of a sample of Ba isotopes on a Rhenium filament, placed in a slot in one of the endcap electrodes. The ions were excited at their $6S_{1/2}$–$6P_{1/2}$ resonance transition at 493.4 nm by a nitrogen pumped pulsed dye laser operated at 20 Hz repetition frequency. The laser beam was guided parallel to the solenoids axis, reflected into the trap through a hole in the ring electrode by a mirror and back reflected onto itself by a second mirror placed opposite to the entrance hole. Laser induced fluorescence light was collected by a two-lens optics into a light guide and detected by a photomultiplier tube outside the solenoid at a distance of about 1.5 m from the trap. The overall detection efficiency including solid angle, transmission losses and detector quantum efficiency was approximately 4×10^{-3}. Usually the decay of the excited $6P_{1/2}$ state into the metastable $5D_{3/2}$ level at 649.6 nm was detected and the laser stray light was blocked by an interference filter. Microwaves were guided into the trap by a waveguide through a hole in the ring electrode perpendicular to the laser beam direction to induce electronic Zeeman transitions at 80 GHz. To induce the nuclear Zeeman transitions we used the Ba-filament as antenna for the 2 GHz radio frequency field.

2.2 Buffer gas collisions

In our previous experiment to determine the g_J factor of Ba^+ [5] the laser induced fluorescence rate from the excited Ba^+ ion cloud was rather small, and long averaging times were required to collect sufficient data. This was due to the fact that the laser excited $6P_{1/2}$ level decays with 27% probability [11] into the metastable $5D_{3/2}$ state. The radiative decay time of this state is about 80 s [12]. Although the effective lifetime might be somewhat decreased due to collisions at the residual pressure of about 10^{-9} mbar, the ions remained most of their time in the metastable state and the average population of the ground state was small. In order to bring the ions back from the metastable state and to enhance the ground state population several lasers tuned to the $5D_{3/2}$–$6P_{1/2}$ transitions would be required, since the large Zeeman splitting of the $6P_{1/2}$ and $5D_{3/2}$ states of several tens of GHz at our magnetic field did not allow the use of a single repumping laser. Although this might have been feasible, our instrumentation did not allow such a scheme for signal enhancement.

In order to improve the signal strength we now use the method of collisional quenching of the metastable state at higher background pressures. A problem arises from the fact, that the ion motion in a Penning trap becomes unstable under the influence of collisions through an increase of the magnetron radius. This can be overcome by coupling of the magnetron motion at frequency

$$\omega_m = \omega_c/2 - [\omega_c^2/4 - \omega_z^2/2]^{1/2} \quad (1)$$

to the reduced cyclotron frequency

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

by an additional radio frequency field at their sum frequency [13]

$$\omega = \omega_m + \omega_c' = \omega_c \quad (3)$$

$\omega_c = (e/m)B$ is the free ions cyclotron frequency and ω_z is the axial oscillation frequency in the trap. The cooling effect of collisions on the cyclotron motion supercedes the increase in magnetron radius and as a result the ions

concentrate near the trap center. Details of this technique are published elsewhere [14]. We benefit in our experiment from this method in several ways: besides the increase in ground state population density by quenching of the metastable state, the concentration of the ions near the trap center increases the spatial overlap with the exciting laser and improves the fluorescence signal. The same holds for the spectral overlap, since the ions are cooled. The maximum benefit, however, comes from the fact that by cooling the amplitude of the ion oscillation is reduced to a value which is smaller than the wavelength of the microwave radiation for the Zeeman transition. We then operate in the Dicke regime where the microwave spectrum consist of a central narrow line, unbroadened by first order Doppler effect, and sidebands at the ion oscillation frequency. As shown below, this reduces the spectral linewidth in the electronic Zeeman transitions by about two orders of magnitude compared to our previous experiment [6]. Finally, another advantage of the collisional cooling of the ions is the extended storage time from previously 20 min under UHV conditions to several hours at 10^{-5} mbar of N_2 buffer gas.

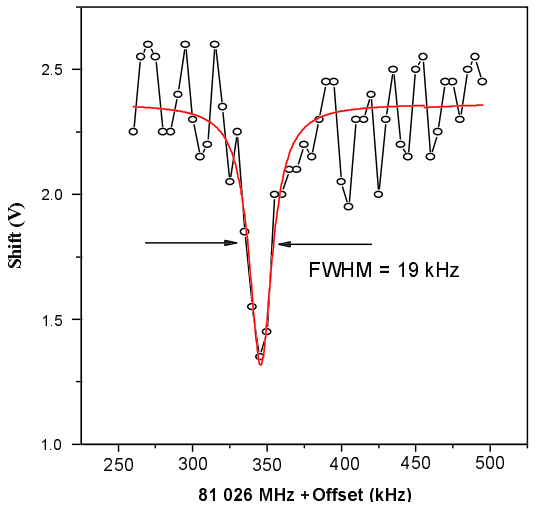

Fig. 2. Cyclotron resonance of stored electrons for magnetic field calibration. The voltage at which the axial resonance appears is plotted versus the applied excitation frequency. At resonant excitation the space charge in the stored electron cloud is reduced, which leads to a shift in the position of the resonance.

2.3 Magnetic field calibration

The magnetic field at the ions position was measured by the cyclotron frequency of electrons stored in the same trap after inversion of the trapping potential. Excitation of the motional eigenfrequencies of stored electrons by a radio frequency field increases the oscillation amplitude. This, in our experiment, is detected in the following way: the trap endcaps are connected by an outer inductance to form a tank circuit, which is weakly excited at its resonance frequency. The axial ion oscillation frequency of a stored particle of charge e and mass m in a trap of radius r_0

$$\omega_z^2 = (eV)/(mr_0^2) \tag{4}$$

depends on the square root of the potential V seen by the particles.This potential is the sum of the applied trapping voltage and the space charge potential of the electron cloud. When we sweep the trapping potential by a linear ramp voltage, ω_z coincides at a certain instant with the resonance frequency of the tank circuit. Then the ions absorb energy and dampen the circuit, which can be detected by a sensitive amplifier. Upon excitation of the electron cloud the space charge potential decreases and consequently the axial resonance appears at a different value of the applied trapping potential. We plot the applied voltage at which the axial electron resonance appears and obtain a resonance curve as in Figure 2. It fits well to a Lorentzian lineshape and we determine the center frequency with an uncertainty of 3×10^{-8}.

A problem may arise from the fact that the electron cloud may be stored at a slightly different position in the magnetic field, when contact potentials on the trap electrodes distort the trapping potential differently for both signs of the trapping voltage, as required for positive ions and electrons. This would lead to a wrong value of the magnetic field for the ions, when the field is inhomogeneous. To check for such a possibility we moved the trap by several mm along the magnet and measured the electron cyclotron frequency. We did not observe any change, more than the statistical fluctuations, for a shift in position of 2 mm which is much more than one can reasonably expect from contact potentials.

To account for a temporal variation of the magnetic field we measured the electron cyclotron frequency before and after a Zeeman resonance measurements for the Ba^+ ions. The measured drift of the field was $\delta B/B = 1 \times 10^{-9}$ h^{-1}. By linear interpolation of the measured field strength from the electron cyclotron resonance, we determined the field strength at the time of the Zeeman resonance measurement.

2.4 g_J-factor measurements

We determined the g_J-factor of the two Ba^+ isotopes with mass 138 and 135 in laser-microwave double resonance experiments. After storage of about 10^5 ions from isotope enriched samples, cooling and confinement of the ions in the center of the trap as described above, one of the electronic ground state Zeeman sublevels was selectively excited by a broadband ($\delta\nu = 7$ GHz) nitrogen pumped pulsed dye laser operated at a repetition rate of 20 Hz. After a few seconds the observed fluorescence decreases to the scatter level by depletion of the pumped state. When the microwave frequency is swept across the $m_J = +1/2 - m_J = -1/2$ Zeeman resonance, we observe an increase in the fluorescence intensity. At higher microwave powers a central peak occurs alongwith strong sidebands at combinations of the ions axial and radial oscillation frequencies (Fig. 3). At lower microwave powers,

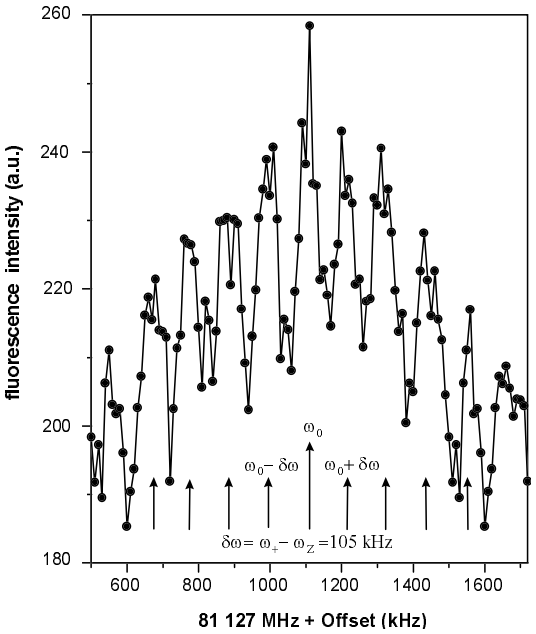

Fig. 3. Microwave induced Zeeman resonance for $^{138}Ba^{+}$ in a magnetic field of 2.89 T. At high microwave power a narrow central peak occurs and sidebands at combinations of the ions oscillation frequencies in the trap.

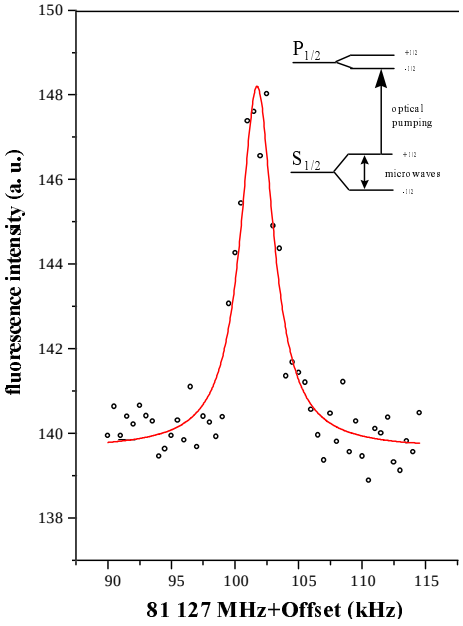

Fig. 4. High resolution scan of the central resonance of Figure 3. The experimental ponts are least-squares fitted by a Lorentzian Lineshape. The statistical uncertainty of the line center is 3×10^{-9}.

the amplitude of the sidebands decreases. When we scan with higher resolution over the central resonance, which is unaffected by first order Doppler effect, the resonance shows a Lorentzian lineshape. The minimum linewidth which we obtained was 3 kHz at a transition frequency of 81 GHz for the even isotope $^{138}Ba^{+}$ (Fig. 4). The line center could be determined by a least squares fit

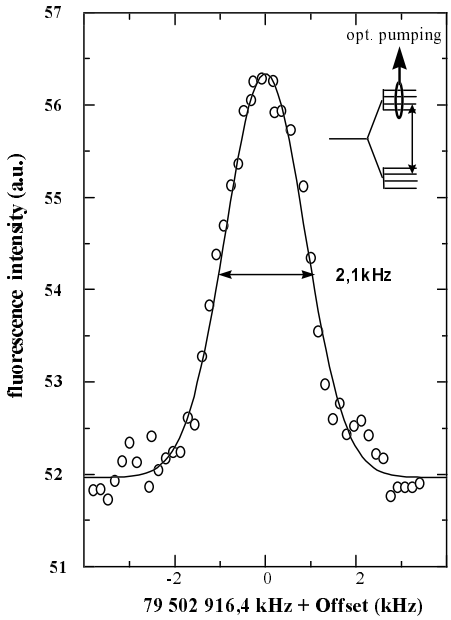

Fig. 5. Microwave induced Zeeman resonance for $^{137}Ba^{+}$ ($m_I = -1/2 \rightarrow m_I = -1/2$). Zeeman effect and hyperfine interaction shift the resonance by about 2 GHz compared to $^{138}Ba^{+}$.

of a Lorentzian to the data points with an uncertainty of 0.22 kHz, corresponding to a fractional uncertainty of 2.5×10^{-9}. Further reduction of the uncertainty was limited by the temporal stability of the magnetic field produced by the superconducting solenoid. Furthermore the uncertainty of the magnetic field calibration could be performed with a precision of 10^{-8} only and hence improvements on the Zeeman resonance would not improve our results.

In a similar way a $\Delta m_J = 1$, $\Delta m_I = 0$ transition was induced in the odd isotope $^{135}Ba^{+}$. Here the transition frequency is shifted to 79.5 GHz compared to $^{138}Ba^{+}$ by hyperfine interaction and nuclear Zeeman effect. The experimentally obtained linewidth was of the same order as for the even isotope (Fig. 5).

From the transition frequency in $^{138}Ba^{+}$ and the magnetic field strength, we drived the value for the g_J-factor in the $6S_{1/2}$ ground state of Ba^{+} as:

$$g_J = 2.002\,491\,92\,(3).$$

The standard deviation ($1\sigma = 1.5 \times 10^{-8}$) as quoted above is entirely due to the uncertainty in the calibration of the the magnetic field.

For the odd isotope $^{135}Ba^{+}$ we obtain a value for the g_J-factor using the Breit-Rabi formula. For the hyperfine structure coupling constant A we used the value $A = 3\,591\,670\,117.45\,(29)$ Hz [15] and for the nuclear g_I factor $g_I = 0.557\,67$ [16]. The g_J-value for $^{135}Ba^{+}$ agrees within the limits of error with that of $^{138}Ba^{+}$ and we can quote an upper limit for the isotope dependence of the g_J-factor:

$$g_J(^{135}Ba^{+}) - g_J(^{138}Ba^{+}) = 1(5) \times 10^{-8}.$$

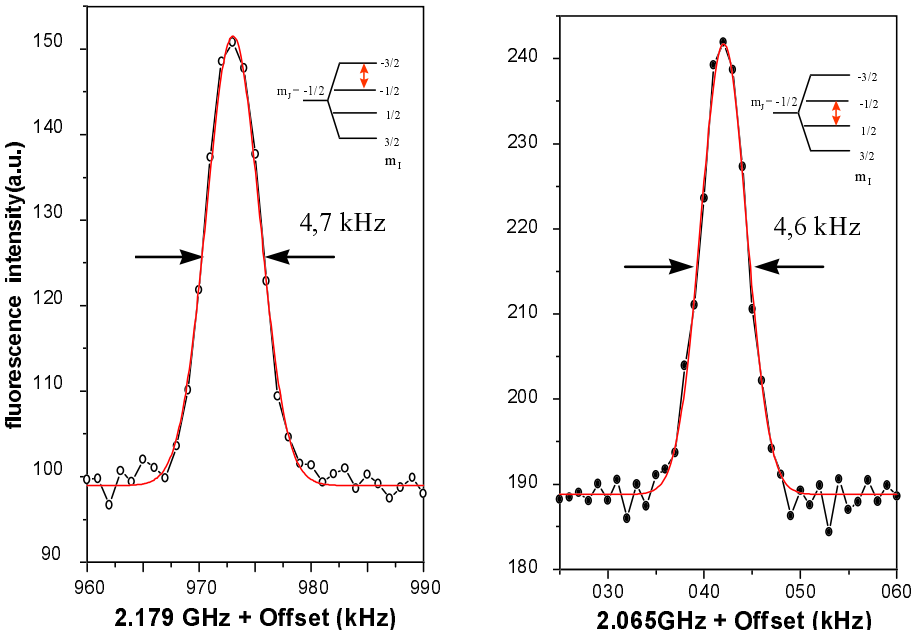

Fig. 6. Microwave induced nuclear Zeeman transitions in a triple resonance experiment on $^{135}Ba^+$. (a) $m_I = -1/2 \rightarrow m_I = -3/2$, (b) $m_I = -1/2 \rightarrow m_I = +1/2$. The experimental data are least squares fitted by a Lorentzian. The relative uncertainty of the line centers is 3×10^{-8}.

2.5 g_I-factor measurement

We performed a test to demonstrate that the Penning trap technique is capable of producing accurate values for nuclear g-factors in a triple resonance experiment on $^{137}Ba^+$. The $m_j = 1/2$ manifold of the nuclear Zeeman structure was depleted by selective excitation with a broadband dye laser in a similar way as in the experiments described above on an even isotope. A second transition between the $m_J = 1/2$, $m_I = -1/2$ and $m_J = -1/2$, $m_I = -1/2$ levels depletes one of the nuclear Zeeman levels in the $m_J = -1/2$ manifold. Finally a radio frequency field between the depleted $m_I = -1/2$ and the adjacent $m_I = -3/2$ or $m_I = +1/2$ levels is detected by an increase in fluorescence intensity. Figure 6 shows the two resonances. The linewidth of 4.7 kHz at a transition frequency of about 2.1 GHz is limited by power broadening. Since the line intensity was rather weak, averaging times of about one hour were required to record the data. Since the precision obtained was more than adequate for the purpose of determination of the hyperfine anomaly, which requires uncertainties of the order of 10^{-5} in unfavorable cases, we did not continue to reduce the linewidth by using lower radio frequency power. This could have resulted in smaller linewidths of the same order as in the case of measurements of the g_J-factor discussed above.

From the two transition frequencies and a magnetic field calibration by a Zeeman resonance in $^{138}Ba^+$, we obtain two independent values for the g_I-factor, which agree with each other within the statistical error. The average value from both the measurements is

$$g_I' = 0.620\,235\,(3).$$

Table 1. Published values of g_J-factors in the $6S_{1/2}$ ground state of Ba^+.

	g_J-factor	Reference
experimental	2.002 491 92 (3)	this work
	2.002 490 6 (11)	5
	2.002 492 2 (10)	18
theoretical	2.002 491 1 (30)	4

Table 2. Nuclear g_I-factors of ^{137}Ba.

g_I-factor	Method	Reference
0.623 876 (3)	Penning ion trap	this work
0.623 82 (4)	Opt. Pumping in gas cell	16
0.623 88 (1)	Opt. Pumping in gas cell	19
0.623 8 (5)	Nuclear magnetic Resonance	20

3 Discussion and conclusion

Table 1 compares the measured g_J value with results from earlier experiments and with a theoretical calculation. All values agree within their quoted uncertainties. Compared to earlier experiments in Penning traps, we have improved the experimental value by a factor of 35. This was made possible by significant signal enhancement by collisional quenching of a long lived metastable state and ion cooling and confinement near the trap center. Further reduction of the uncertainty was limited by calibration of the magnetic field. The theoretical value is two orders of magnitude less accurate than the experimental value and considering the complexity of calculations in a multi-electron system like Ba^+ it may be a difficult task for the theory to match the experimental precision.

Table 2 lists available values for the g_I-factor of $^{135}Ba^+$. We have applied a diamagnetic correction $(1-\sigma)^{-1} = 1.005\,90$ [17] to our experimental value of $g'_I = 0.620\,235$. All the quoted values agree with each other. Compared to the most accurate previous measurements on neutral Ba atoms by optical pumping in a buffer gas cell, our measurements using Penning trap have one order of magnitude lower uncertainty. Further improvement, if required, is possible by reducing the power broadening, as mentioned above.

In conclusion, we have demonstrated that the Penning trap technique can yield very accurate values of electronic and nuclear g-factors. Since it has been shown earlier [6] that radioactive isotopes of minute quantities can be successfully investigated in ion traps, the extension of the method described above to unstable isotopes could result in improved values for nuclear magnetic moments which are not derived from hyperfine structure measurements, but measured by direct nuclear Zeeman transitions.

This would in combination with measured hyperfine coupling constants of the same isotopes, result in values of the hyperfine anomaly in unstable isotopes, which is desirable for a better understanding of the distribution of magnetization over extended nuclei.

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