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Precision spectroscopy of the Zeeman splittings of the $^9Be^+2^2S_{1/2}$ hyperfine structure for nuclear structure studies

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Abstract

A precision measurement for the hyperfine Zeeman splittings of $^9Be^+$ ions, trapped and laser cooled in a linear combined ion trap was performed by using laser–rf double and triple resonance methods. The accuracies of 10^{-9} for the nuclear spin flip transitions and of 10^{-8} for the electron spin flip transitions were achieved at an arbitrary value of strong magnetic field of $\sim$0.47 T. This enables us to determine the hyperfine constant A and the ratio of nuclear g-factor to electronic g-factor with accuracies of 10^{-9} and 10^{-7}, respectively.

To show the wider applicability of our method, the particular strength of the magnetic field where the nuclear spin flip transition is independent of the magnetic field strength was intensionally not used. The confining fields of the linear combined trap have the advantage of simple and efficient concentration of the ions into the potential minimum at the center of the trap, when laser cooling of the stored ions is performed. The feasibility of single ion spectroscopy was also demonstrated and, when fully implemented will facilitate the on-line study of short lived isotopes, which can only be produced in minute numbers. © 2002 Published by Elsevier Science B.V.

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1. Introduction

In the past, precision measurements of Zeeman splittings of the ground state hyperfine structure (HFS) of trapped ions have been performed mainly by aiming towards the development of frequency standards. In contrast we have proposed to apply

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this technique to studying the nuclear structure of the various Be isotopes [1]. The hyperfine constant A shows a small but finite isotope dependence. The main part of this *hyperfine anomaly* stems from the finite distribution of the magnetism over the extended nucleus and is known as the Bohr–Weisskopf effect [2]. We aim at the investigation of the neutron halo structure of the ^{11}Be nucleus through the measurement of the Bohr–Weisskopf effect, which is sensitive to the radial distribution of the loosely bound valence neutron. A recent theoretical estimate supports the importance of the investigation of the Bohr–Weisskopf effect in connection with the nuclear structure of ^{11}Be [3].

So far we have been working towards the development of exact experimental techniques for this purpose [4–6]. To investigate the Bohr–Weisskopf effect of ^{11}Be, we must determine the hyperfine constant A and the nuclear g-factor g_I for all Be isotopes with an accuracy of 10^{-6} or higher. It should be noted that as long as we compare only the g_I-factors for the different isotopes, it is acceptable to determine the ratios of nuclear and electronic g-factors (g_I/g_J), since the isotopic dependence of the g_J-factor is negligible. However if one wants to know the absolute value of the nuclear g-factor, then the absolute strength of the magnetic field must also be measured. The Mainz group has used the cyclotron frequency of the electron for this purpose in their g_I-factor measurement of the ^{135}Ba isotope [7].

The NIST group has demonstrated an ultimate accuracy of 10^{-11} in the determination of the A constant of $^9Be^+$ and of 10^{-9} in the g_I/g_J ratio [8,9]. These measurements were carried out with a laser–microwave multiple resonance method [10] on laser cooled ions in a Penning trap. In this measurement, the particular strength of the magnetic field, where the nuclear spin flip transition is independent of the magnetic field strength in first order, was used. Such a transition is called a *clock* transition. For their application in a frequency standard, such an ultra highly accurate transition at that particular condition of a certain atom is needed. However, for our purpose, in the future various isotopic ions of several elements including short-lived isotopes are the objects to be studied. In this case there are not always such adequate *clock* transitions available. For instance, for the ^{11}Be ion, which at the present time is our most important nucleus to be studied next, there is no *clock* transition, since the nuclear spin is $I = 1/2$. On the more practical side and when studying unstable isotopes, it is also not simple to vary the strength of a precision superconducting magnet within the limited period of accelerator machine time.

For these reasons we have restricted ourselves to use an arbitrary fixed strength of the magnetic field, and to employ relatively short periods of time for each measurement cycle, so as to make our method also applicable to the study of unstable isotopes.

In particular, a high detection efficiency is an important issue when rare unstable isotopes are concerned. In this respect a linear combined trap is superior to an ordinary Penning trap. The presence of a strong focusing force due to the rf gradient field automatically and efficiently concentrates the ions in the center of trap during laser cooling [11]. This concentration can in a Penning trap only be achieved by additional sideband cooling and is then necessary due to the so called magnetron instability. Laser cooling in the combined trap enables single ion spectroscopy, which is the ultimate situation not only as far as the detection efficiency is concerned, but also for the homogeneity requirement of the magnetic field over the smallest possible measurement region confining the single trapped ion. The linear trap is in addition suitable for external injection. The linear structure matches ideally an upstream ion beam guide from which unstable nuclear ions can be introduced [5,12]. A larger phase–space acceptance of the combined trap is also helpful [13].

In this paper, we report the experimental procedure and results of precision spectroscopy of the Zeeman splittings of the HFS of ground state $^9Be^+$ ions using a linear combined trap. This is our first step towards future nuclear structure investigations of the other Be isotopes.

2. Experimental procedure

The HFS Zeeman levels of the $^2S_{1/2}$ state in a magnetic field B_0 are described by the exact Breit–

Rabi formula. We express the strength of the magnetic field B_0 in terms of the Larmor precession energy of the valence electron $b = g_J\mu_B B_0/h$, and the ratio of the nuclear g-factor to the g-factor of the valence electron as $\gamma = g'_I/g_J$. With these relations the energy splittings are described by

$$W_F(m_J, m_I, b) = -\frac{A}{4} - (m_J + m_I)\gamma b + m_J\sqrt{A^2\left(\frac{1}{2}+I\right)^2 + 2A(m_J+m_I)(\gamma-1)b + (\gamma-1)^2b^2}. \quad (1)$$

Here the energies are in frequency units, m_J, m_I are the magnetic quantum numbers of the electronic spin J and the nuclear spin I, and g'_I is the nuclear g-factor in units of the Bohr magneton μ_B. In order to find the three unknown parameters: A, γ and b, we must independently determine more than three orthogonal transition frequencies. The transition frequencies which we used are accessible by double resonance (ν_{e1}, ν_{e2}) and triple resonance (ν_{n1}, ν_{n2}) methods, and are indicated in Fig. 1.

Another prerequisite for the present study is to estimate the sensitivity of these measurable frequencies to the unknown physical quantities, A, γ and b at a given strength and homogeneity of the magnetic field. The deviation of the transition frequency ν, when these parameters are slightly varied, is

$$\frac{\Delta\nu}{\nu} = \left(\frac{\partial\nu}{\partial b}\frac{b}{\nu}\right)\frac{\Delta b}{b} + \left(\frac{\partial\nu}{\partial A}\frac{A}{\nu}\right)\frac{\Delta A}{A} + \left(\frac{\partial\nu}{\partial\gamma}\frac{\gamma}{\nu}\right)\frac{\Delta\gamma}{\gamma} = c_b\frac{\Delta b}{b} + c_A\frac{\Delta A}{A} + c_\gamma\frac{\Delta\gamma}{\gamma}. \quad (2)$$

The coefficients, c_b, c_A and c_γ, were derived from Eq. (1). They indicate the sensitivity to the values of the magnetic field, the hyperfine constant A, and nuclear g-factor, respectively. Assuming as an example that the strength of the magnetic field is 0.5 T, then the coefficients for the electron spin flip transitions (ν_e: $\Delta m_J = 1, \Delta m_I = 0$) are estimated to be $|c_b^e| \simeq 1$, $|c_A^e| \simeq 0.03/B_0$ (B_0 in T), $|c_\gamma^e| < 10^{-6}$ and for the nuclear spin flip transitions (ν_n: $\Delta m_J = 0, \Delta m_I = 1$) of $^9Be^+$ to be $|c_b^n| < 0.03$, $|c_A^n| \simeq 1$, $|c_\gamma^n| \simeq 0.02B_0$. The electron spin flip frequency ν_e depends dominantly on the magnetic field strength and has a contribution of only a few percent from A, and a negligible contribution from the nuclear g-factor. The nuclear spin flip fre-

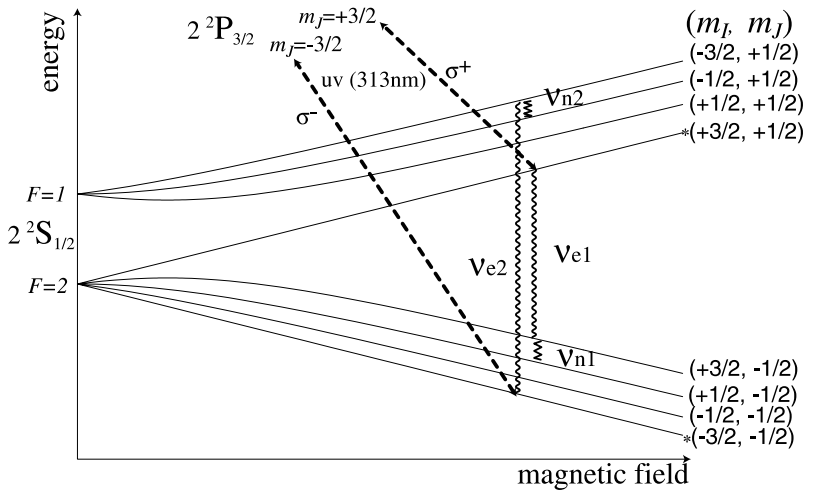

Fig. 1. Zeeman splitting of the ground state hyperfine structure of $^9Be^+$. The states $(m_I, m_J) = (+3/2, +1/2), (-3/2, -1/2)$ are optical pumped when circular polarized laser radiation (σ^+, σ^-) is used. The electron spin flip transitions, ν_{e1}, ν_{e2} are the transitions accessible by the laser–microwave double resonance method. The nuclear spin flip transitions, ν_{n1}, ν_{n2} can be measured by the laser–microwave–uhf triple resonance method.

quency ν_n, on the other hand, depends mainly on the A constant, and has a few percent contribution from the nuclear g-factor as well as from the magnetic field strength.

Next, we quantify how much accuracy for the determination of A and g_I'/g_J can be achieved from measurements of ν_e and ν_n under a given condition. For this purpose we define a figure-of-merit α as

$$\alpha = |c_\gamma|/|c_b| = \left|\frac{\partial \nu}{\partial \gamma}\gamma\right| \Big/ \left|\frac{\partial \nu}{\partial b}b\right| = \frac{|\gamma/\Delta\gamma|}{|b/\Delta b|} = \frac{|g_I/\Delta g_I|}{|B_0/\Delta B_0|}, \tag{3}$$

α is the ratio of the sensitivity of the nuclear g-factor to the sensitivity of the magnetic field. The figure-of-merit for the nuclear spin flip transitions ν_{n1} and ν_{n2} of $^9Be^+$ as well as of $^{11}Be^+$ versus the magnetic field are plotted in Fig. 2. We find singular points in the case of $^9Be^+$ at a particular strength of the magnetic field, where c_b^n becomes 0. This is the so called *clock* transition condition and it is the best condition for the determining g_I. It can be seen that no such conditions exist in the $^{11}Be^+$ case. If we use a fixed strength of the magnetic field of about 0.47 T and assume that the homogeneity of the magnet is 10^{-6} over the volume of the ion cloud, then the expected accuracy for nuclear g-factor is 10^{-6} ($\alpha \simeq 1$). In order to

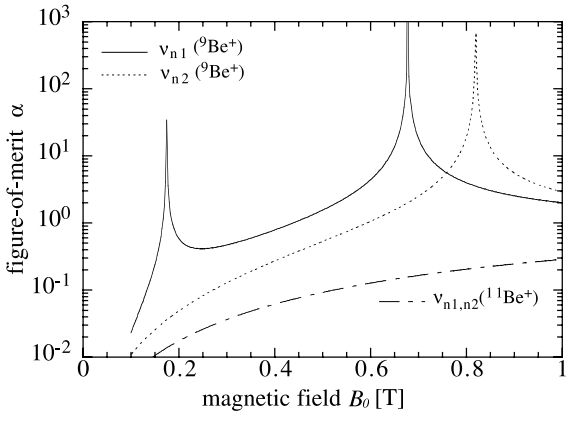

Fig. 2. Figure-of-merit $\alpha = |g_I/\Delta g_I|/|B_0/\Delta B_0|$ for the nuclear spin flip transitions of $^9Be^+(\nu_{n1}, \nu_{n2})$ and of $^{11}Be^+(\nu_{n1,2})$ as a function of the magnetic field. The singular points in $^9Be^+$ mark the *clock* transition conditions. There are no such points for $^{11}Be^+$.

achieve this accuracy, we need to measure ν_n with an accuracy of 2×10^{-8} corresponding to 6 Hz for a transition at 300 MHz, and ν_e with 10^{-6} corresponding to 10 kHz for a 12 GHz transition. For the hyperfine constant A, the expected accuracy under such conditions is then already $\sim 10^{-8}$.

The experimental ion trap setup has been reported in detail in a previous paper [11]. A linear rf trap made of four stainless steel rods (6 mm in diameter) and placed in a superconducting Helmholz magnet formed the linear combined trap. The homogeneity of the magnetic field was 10^{-6} at 0.5 T for a length of 1 mm at the center of magnet. In addition to the trap driving rf, two rf signals were introduced to the trap for spectroscopy. A variable 12–14 GHz microwave signal for inducing the electron spin flip transition was generated by a synthesizer (HP 83731B) and was amplified by a traveling wave tube amplifier (Hughes 8010H). The 300 MHz uhf signal for inducing the nuclear spin flip transition was generated by a synthesizer (HP 8643A) and amplified by a solid state amplifier (R&K 250-SMA). All synthesizers were locked to the 10 MHz reference signal of a GPS receiver (HP 58503B). The accuracy of the reference signal is almost 10^{-12} according to the specifications. These rf signals were transmitted to the chamber by coaxial cables and connected to one of the trap electrodes. UV laser radiation at $\lambda = 313$ nm was generated by intra-cavity frequency doubling of the light of a ring dye laser (Coherent 899-21). The circular polarization of the laser radiation was controlled by rotating a $\lambda/4$ plate. At resonance the laser induced fluorescence (LIF) signal was detected by spatially resolved photon counting with a two-dimensional camera (Hamamatsu PIAS-TI). Data acquisition was carried in both the simple MCS (multi channel scaler) mode and event-by-event recording mode (list mode). LabVIEW software controlled the entire sequence of the experiment by manipulating the mechanical shutter for chopping the laser radiation, the synthesizers for sweeping and chopping the rf signals, and the data acquisition system.

A typical timing sequence of the double (laser–microwave) and triple (laser–microwave–uhf) resonance method is depicted in Fig. 3. The scenario for the double resonance method is as follows.

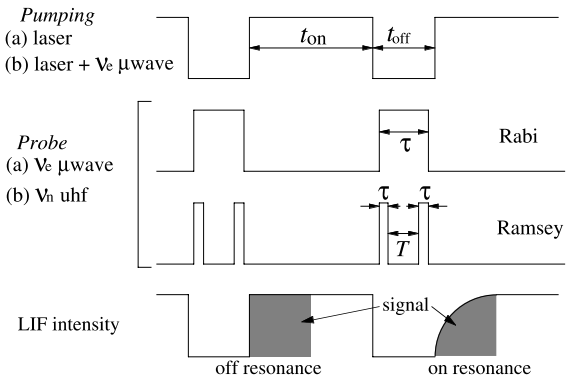

Fig. 3. Timing sequence of the double (a) and triple (b) resonance method. In the double resonance method, which is used for ν_e measurements, laser radiation was chopped and the probe microwave radiation was applied during the laser-off periods (t_{off}). In the triple resonance method, which was used for ν_n measurements, resonant microwave radiation for the corresponding ν_e transition was applied together with the laser radiation pulse. Probe uhf radiation was applied during the laser- and microwave-off periods in two different way. A single pulse with a width of τ was applied in the Rabi method, while two pulses of a width τ with a separation of T were applied in the Ramsey method. The LIF signal was accumulated during the initial part of the laser-on period and was reduced when Ramsey pulses were on resonance.

Optical pumping to the maximum m state, $(m_I, m_J) = (+3/2, +1/2)$, of the ground state is effected during the laser-on period, when the polarization of laser radiation is σ^+. In this situation strong LIF is detected. For a measurement of the electron spin flip transition, the 12 GHz microwave radiation is applied during the succeeding laser-off period. When the microwave frequency is on resonance with the electron spin-flip transition ν_{e1}, the integral LIF intensity decreases during the next laser-on period. For the nuclear spin flip transition measurement, on the other hand, the resonant microwave radiation (ν_{e1}) and laser radiation are irradiated simultaneously to realize a condition such that optical pumping into both (+3/2, +1/2) and (+3/2, -1/2) states is achieved and continuous LIF is observed. The 300 MHz uhf radiation is turned on during the succeeding laser- and microwave-off period. If the uhf frequency is on the resonance ν_{n1}, then the integrated LIF signal during the next laser- and microwave-on period decreases. This is the triple resonance method.

In the case that the polarization of the laser radiation is σ^-, the same methods can be performed, but now ν_{e2} and ν_{n2} (Fig. 1) are measured. The way of applying uhf radiation is in either case by a single square pulse modulation (Rabi method) or in a double pulse modulation (Ramsey method). The latter has higher resolution and facilitates the accurate determination of the resonance center, if the pulse conditions for producing coherent excitation are adequately chosen.

3. Results and discussion

Experimental results for the electron spin flip transition ν_{e1} measurements are shown in Fig. 4. The spectrum on the bottom was obtained when both laser radiation and microwave radiation were continuously applied (continuous method), while the upper spectrum was obtained by the Rabi method. The two measurements depicted were performed one after the other using the same ion

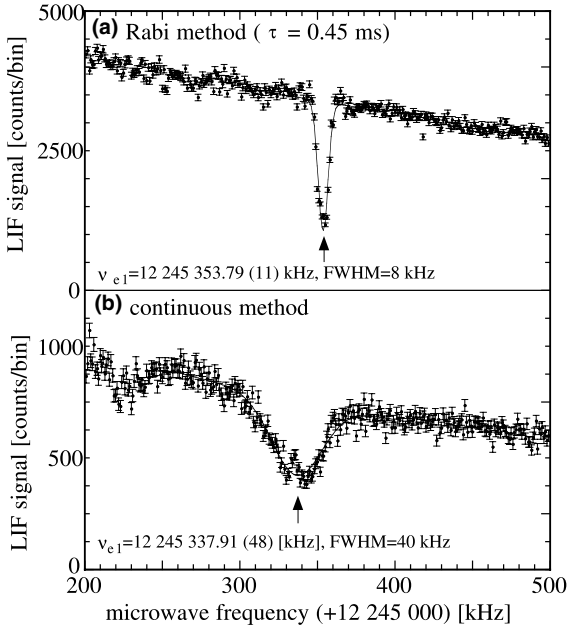

Fig. 4. Microwave resonance spectra for the electron spin flip transition ν_{e1} taken by the Rabi method (a) and the continuous method (b). The fluorescence signal was accumulated for a period of 28 pulses per data point (a), and for a continuous period of 500 ms per data point (b).

cloud to avoid any drift of the magnetic field strength. The comparison of the two spectra shows the existence of line broadening of 40 kHz, and a line shift of −16 kHz in the continuous method. These are the known power broadening and light shift effects due to the laser radiation [14,15]. The line width in the Rabi method spectrum was only 8 kHz FWHM which is consistent with the given inhomogeneity of the magnetic field, if we assume that the size of ion cloud is 1 mm.

Although the continuous method is not adequate for an accurate determination of the transition frequency, it is useful for finding the resonance at the start of the experiment. The electron spin flip transition frequencies ($\nu_{e1,e2}$) were obtained with an uncertainty of about 10^{-8} using the Rabi method. This is sufficient for our purpose as discussed in the previous section. Since the line width was not determined by the coherence time of the microwave radiation, we took the rather short pulse width of 0.45 ms, which was limited by the response of our mechanical shutter.

The nuclear spin flip transition frequency ν_{n1} was measured by the triple resonance method as shown in Fig. 5. After finding the corresponding electron spin flip transition frequency ν_{e1}, the microwave frequency was fixed to the resonance. The microwave radiation pulse was synchronized to the laser pulse. The probe uhf radiation was applied during laser- and microwave-off periods in two different time structures (Fig. 3). Fig. 5(a) is a typical spectrum taken with Rabi method. The coherence time τ was 45 ms. The LIF signal was accumulated for the first 55 ms in the succeeding laser-on period of 155 ms. Fig. 5(b) is a typical spectrum taken with the Ramsey method. The pulse parameters were: $T = 22$ ms, $\tau = 10$ ms. LIF signal were accumulated in the same way and plotted as a function of the uhf frequency. The Ramsey fringes were clearly observed when the proper intensity of the uhf radiation was applied. The uncertainty of both measurements for ν_{n1} was in the order of 0.5 Hz. We point out that these two measurements were carried out on different days illustrating the long term behavior of our measurements. The small shift in the observed resonance frequency is caused by the decrease of the magnetic field strength of the superconducting magnet.

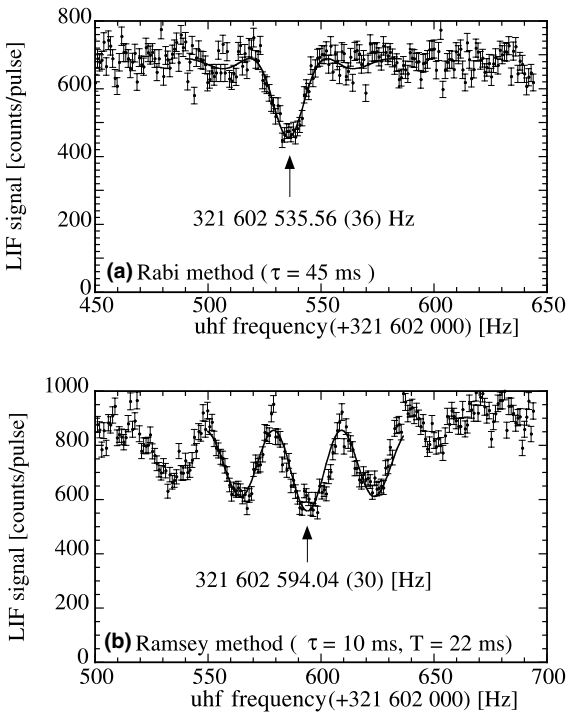

Fig. 5. uhf Resonance spectra for the nuclear spin flip transition ν_{n1} taken by the laser–microwave–uhf triple resonance method. The upper curve was obtained by the Rabi method while the bottom curve was measured with the Ramsey method. Each data point was an accumulation of the LIF signal during one pulse cycle. The total acquisition period was 40 s (200 ms × 200 ch). Note that these two measurements were carried out on different days. The drift of the magnetic field strength causes the shifted frequency.

When we repeated cycles of alternative measurement for ν_e and ν_n, we found a drift of the effective strength of the magnetic field which is considerably large for a precision measurement. The trend of the drift consisted of a long term constant decay component and short term fluctuations. The latter ones were considered to be due to the variation of the position of the ion cloud. When we loaded Be ions into the trap, the pulse of a YAG laser irradiated a metallic Be target which was located close to the trap electrode. Large amounts of ion–electron pairs were produced by the laser ablation, and when some of them stick to the trap parts they may cause shifts in the trapping potential.

We took care of the effect of the short term fluctuation by sequentially measuring a set of transitions, ν_e–ν_n–ν_e, without reloading the ion species. In the data analysis, we took the average of the two ν_e measurements as the values for ν_e with a large uncertainty which covered the two measurements, and gave different magnetic field parameters b_i for each set of the measurement. Results for the two different polarization experiments are summarized in Fig. 6. From these experimental results, we obtained that the hyperfine constant $A = -625,008,835.23(75)$ Hz and the relative nuclear g-factor $g'_I/g_J = 2.13478033(28) \times 10^{-4}$. These values are slightly inconsistent with the NIST results by about 2σ [8]. A possible reason of this disagreement could be a quadratic dependence of the hyperfine constant A and the g-factor ratio γ to the strength of the external magnetic field [16,17]. We plan to confirm this fact in further experiment of the magnetic field dependence with higher accuracy. Fig. 6 also indicates that there is still some room to improve the accuracy by increasing the accuracy especially for ν_{n1}, even if we stay with the present homogeneity of the magnet. This improvement can be done, for example, by an increase of the coherence time as well as the statistics of the measurement.

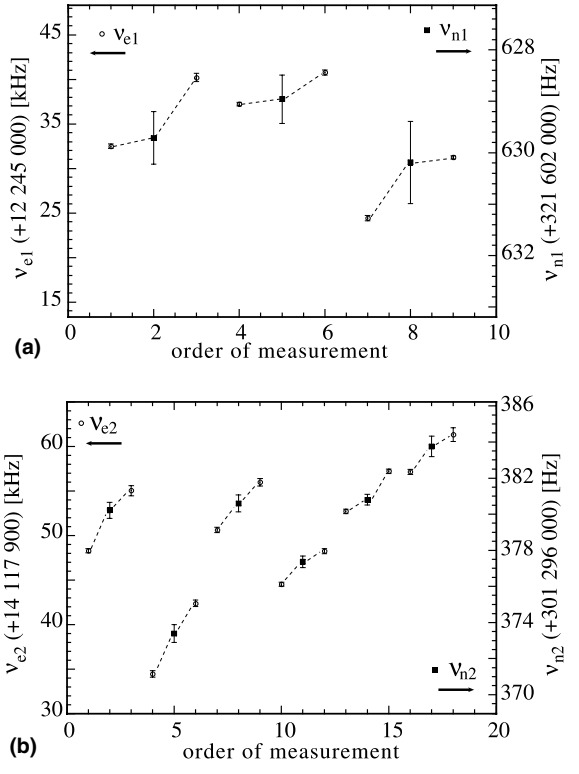

Fig. 6. Data fluctuation in a series of measurement for ν_{e1} (circle), ν_{n1} (box) (a), and ν_{e2} (circle), ν_{n2} (box) (b). The dotted lines connect each set of measurement. The range of the vertical scale corresponds to the deviation of field strength of $\Delta B/B = 2.66 \times 10^{-6}$ for both ν_e and ν_n. It is equivalent to those frequencies: of $\Delta\nu_e = 35$ kHz, $\Delta\nu_{n1} = -6.1$ Hz, and $\Delta\nu_{n2} = 17.3$ Hz. Since ν_{n1} is more insensitive to the deviation of the magnetic field ($\alpha_{n1} > \alpha_{n2}$), the statistical uncertainty of ν_{n1} was enhanced.

As a prospect for future work, we have also tested single ion spectroscopy for the electron spin flip transition. This is the best possible situation not only as far as efficiency is concerned, but also for eliminating any spatial inhomogeneities of the field. For this measurement the detuning of the laser radiation was only −100 MHz, while it was −900 MHz in the previous experiments. Fig. 7 shows the signals for an ion crystal consisting of a few ions and for a single ion. In each case we observed the resonance of the electron spin flip transition, but on the other hand for the single crystallized ion, a fake resonance due to a motional instability was also recorded. In order to completely realize spectroscopy for a single ion, a higher stability in the crystallized state [18] and the consideration of acquiring data under the conditions of such a quantum jump are required. To register a resonance signal, the number of quantum jumps in a suitably chosen period must be counted as a function of the uhf frequency.

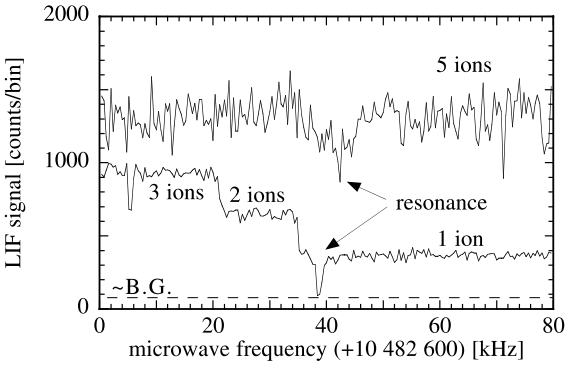

Fig. 7. Test result for crystallized ions made with a few ions (upper trace) and a single ion (lower trace).

4. Conclusion

We have developed a novel technique using a linear combined ion trap to measure on-line the hyperfine constant A as well as the ratio of the nuclear g-factor to the electronic g-factor with an accuracy of 10^{-9} and 10^{-7}, respectively. Even though we did not use a particularly high homogeneous magnet (the homogeneity was about 10^{-6} for 1 mm), nor the optimum value of the magnetic field (*clock* condition), the obtained accuracy is sufficient for studying the Bohr–Weisskopf effect. This result is of importance, when in the future this technique will be extended to unstable isotopes. The number of trapped ions was around one hundred in typical experiments. However we have also tested possible single ion spectroscopy. The sensitivity of the double and triple resonance method when coupled with the laser cooling technique is extremely high. This is a decisive advantage when applying the method to ions of unstable isotopes, which are usually available only in limited amounts. Another difficulty in unstable nuclear isotope research is the fact that the isotopes decay according to their nuclear life time. We have taken only about 40 s for getting a spectrum in the present experiments. This time is sufficient for studying many isotopes, but not yet short enough for a shorter lived one, such as ^{11}Be ($T_{1/2} = 13.8$ s). However, it should be noted that in one cycle of measurements with either the Rabi or the Ramsey methods, the total time period for the cooling and repumping phase and the rf resonance phase, was as short as 200 ms. Therefore, in principle, we can reload new unstable nuclear ions during each measurement cycle. This will allow us to also apply our method towards such short lived nuclei which have a half-life of the order of one second.

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