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Laser-Microwave Double-Resonance Spectroscopy of Laser-Cooled ${}^9\text{Be}^+$ Ions in a Weak Magnetic Field for Studying Unstable Be Isotopes

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Laser-microwave double-resonance spectroscopy of laser-cooled ${}^9\text{Be}^+$ ions in a linear Paul trap has been performed under a weak magnetic-field condition for the first time. From the results of the double-resonance experiment, we could determine the magnetic dipole hyperfine constant (A) with an accuracy of $\Delta A/A = 4 \times 10^{-7}$. This method can be applied to measurements of the hyperfine constant of unstable isotopes (${}^{7,11}\text{Be}^+$) for studying the hyperfine anomaly.

KEYWORDS: beryllium, double-resonance spectroscopy, hyperfine structure, linear ion trap, laser cooling, optical pumping

§1. Introduction

A systematic measurement of the hyperfine anomaly (the Bohr-Weisskopf effect¹⁾) of chains of isotopes enables us to determine the nuclear magnetization distribution, and tests the present nuclear models.²⁾ A few systematic studies concerning the hyperfine anomaly of unstable nuclei have been performed so far.²⁾ Recently, a radioactive beam facility with an on-line isotope separator has come into wide use in the world. The development of such instruments has made it possible to perform a systematic study of hyperfine anomaly, including unstable nuclei.

We have started an experimental project to study the hyperfine anomaly of unstable nuclear ions trapped in an ion trap as one possible application of the radioactive beam facility at the Japan Hadron Facility.³⁾ Our first objects are unstable beryllium isotopes of ${}^7\text{Be}$ ($T_{1/2} = 53.3$ d) and ${}^{11}\text{Be}$ ($T_{1/2} = 13.8$ s). We will measure the hyperfine coupling constants and the nuclear magnetic moments of ${}^{7,11}\text{Be}^+$ with high precision using an ion-trap technique.

${}^{11}\text{Be}$ is known to be one of the neutron halo nuclei.⁴⁾ The valence neutron of ${}^{11}\text{Be}$ extends around the core nucleons. Since the hyperfine field at the extended nucleon is weak, a large hyperfine anomaly is expected. The valence neutron-density distribution of ${}^{11}\text{Be}$ can be studied through the hyperfine anomaly.

${}^7\text{Be}$ is the mirror nucleus of ${}^7\text{Li}$. Since the ${}^7\text{Li}$ - ${}^7\text{Be}$ pair is an isospin doublet, the two nuclei are connected through a mirror β -decay transition. Thus, we can determine the expectation values of both the isoscalar and

isovector spin and orbital angular momenta of nucleons separately based on the nuclear magnetic moments and the ft -value of the β -decay. Systematic measurements of the magnetic moments of mirror-pair nuclei in light elements have been performed and analyzed, except for the ${}^7\text{Li}$ - ${}^7\text{Be}$ pair.⁵⁾

In spite of the importance of the nuclear magnetic moments and hyperfine anomalies of ${}^{7,11}\text{Be}$, they have not been measured yet due to experimental difficulties. No β -rays are emitted from ${}^7\text{Be}$ because the decay mode is totally electron capture. Although the daughter nucleus, ${}^7\text{Li}$, in the excited state emits γ -rays, the spatial distribution of the intensity is uniform because the spin of the initial state is $1/2$. Thus, we cannot apply the ordinary β - or γ -NMR method for ${}^7\text{Be}$. In the case of ${}^{11}\text{Be}$, β -rays are emitted in the decay process. It is, however, hard to produce and maintain sufficient nuclear polarization in a catcher material.⁶⁾ Recently, a collinear laser spectroscopy experiment for ${}^7\text{Be}^+$ was performed at ISOLDE, CERN by using a newly developed laser ion source.⁷⁾ However, the accuracy must be insufficient for studying the hyperfine anomaly due to limitations of the natural line width of the optical transition and the instability of the beam velocity.

There are no such difficulties in laser-microwave double-resonance spectroscopy combined with the ion-trap technique. We can perform high-precision measurements using an ion trap, since Doppler-free spectroscopy can be easily realized for microwave spectroscopy⁸⁾ and a long interaction time between ions and microwave radiation can be obtained. We are also free from any consideration of the complex hyperfine field in the material. Moreover, ultra-high sensitive spectroscopy, even for a single ion, can be realized thanks to the laser-cooling technique. It is, in particular, useful for experiments of

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rare isotopes, such as radioactive nuclei.

Generally, unstable isotopes are supplied to an experiment as an energetic ion beam. Therefore, we must develop a new instrument to trap ions in an energetic beam into an ion trap. For this purpose, we have tested an ion-guide system consisting of a helium-gas chamber for cooling energetic ion beams to thermal energy. The sextupole radio frequency (rf) ion-beam guide (SPIG) as well as a novel linear trap have been developed at IPNS, KEK,^{3,9)} and tested in an on-line experiment.¹⁰⁾ In our scenario, the produced unstable ions are bunched in the SPIG system and transferred to a second linear Paul trap placed in a weak magnetic field where we shine laser light on the stored ions for laser cooling and optical pumping in order to produce nuclear polarization. We then transfer the ions to the last linear ion trap placed in a strong magnetic field of up to 1 T, where we perform laser-microwave double-resonance spectroscopy for measurements of the nuclear spin-flip transitions to determine the nuclear g_I factor. Development of the last part is under progress at ILS, UEC.³⁾

In this paper we report on the developments in the first part, i.e., laser cooling, optical pumping and laser-microwave double-resonance spectroscopy for determining the magnetic dipole hyperfine structure (HFS) constant (A) in a weak magnetic field.

Usually, the HFS constant (A) can be precisely determined in a measurement under a very weak field, where a first-order field-independent transition, i.e., the hyperfine clock transition, is measured by simple double-resonance spectroscopy for buffer-gas-cooled ions stored in a Paul ion trap.^{11–16)} The method, however, does not work for Be isotopes, since the HFS splitting is as small as about 1 GHz, which is much smaller than the Doppler width of the optical transition of the stored ions without laser cooling.

We need to achieve both laser cooling, which is necessary in order to discriminate the HFS states of Be^+ ion, and optical pumping, which is necessary to perform microwave spectroscopy. The optical pumped levels must be the maximum $|m_F|$ levels in order to satisfy these conditions. Unfortunately, they are field-dependent levels. Although the expected accuracy of the HFS constant (A) obtained from the field-dependent transition frequencies is not very high, we have tried to make measurements as accurately as possible under the present condition. Furthermore, a sensitive detection of the microwave resonance and high-resolution detection are different matters, though both are important for our experiment. We discuss our development results for these directions.

§2. Apparatus and Method

2.1 Ion trapping

For trapping Be^+ ions, we used a linear Paul trap comprising four separated cylindrical rods with a diameter of 6 mm. Each electrode was divided into three sections. The distance between the diagonal electrodes was 5.3 mm. The axis of the linear trap was tilted by 7 degrees to the direction of the laser beam for efficient laser cooling of ions moving in the radial direction. The lin-

ear Paul trap was enclosed in a vacuum chamber, which was evacuated to be lower than 1.3×10^{-7} Pa by a turbo-molecular pump (300 l/s) backed by a rotary pump.

A radio-frequency voltage generated by a synthesizer was amplified by an rf amplifier and a transformer. Then, this high rf voltage was applied to the trap electrodes. The frequency and amplitude were set to 4.24 MHz and 50 V, respectively. The maximum pseudopotential was about 1.3 eV in the radial direction. The two end sections have different DC offsets superimposed on the rf voltage to control ions in the axial direction.

There are some advantages of the linear Paul trap to our experimental project. The linear Paul trap can be easily connected with the SPIG system, by which unstable nuclear ions can be accumulated in a bunch. We can also perform mass selective injection of unstable ions in order to remove the impurities in the radioactive beam by using the end section as a quadrupole mass filter. Moreover, there is a one-dimensional field-free area where a lower rf heating effect is expected.¹⁷⁾

Stable $^9\text{Be}^+$ ions were produced by focusing a pulsed laser beam (14 mJ/pulse, 6 ns width) from a compact Nd:YAG laser on a metallic beryllium target. The peak power of the beam on the target was high enough to produce many $^9\text{Be}^+$ ions. The target was placed on an edge of the end section so as to introduce ions along the trap axis. A fraction of the created $^9\text{Be}^+$ ions were trapped by switching the DC potential of the end sections after a 12 μs delay from YAG-laser irradiation. The trapped ions were laser cooled to low temperature (< 10 K). The initial number of trapped ions was estimated to be on the order of 10^4 based on a measurement of an ion signal detected by a micro-channel plate device, which was placed at the down-stream of the linear trap.

2.2 Laser-Microwave Double-Resonance Spectroscopy for Laser-Cooled Be^+ Ions

In order to perform laser-microwave double-resonance spectroscopy for Be^+ ions, we applied optical pumping, a double-resonance method combined with a laser-cooling technique, as explained in the following.

A partial energy level diagram of $^9\text{Be}^+$ is shown in Fig. 1. In a weak magnetic field, the hyperfine states split into magnetic sublevels, denoted by (F, m_F) . Under this condition, we irradiated circularly polarized UV laser light, such as σ^- , onto the stored ions along the direction of the applied magnetic field. The laser was tuned to be slightly lower in frequency than the $^2\text{S}_{1/2} (2, -2) \rightarrow ^2\text{P}_{3/2} (3, -3)$ transition for laser-cooling. The $^2\text{S}_{1/2} (F, m_F) \rightarrow ^2\text{P}_{3/2} (F', m_F - 1)$ transitions were excited according to the E1 selection rule, $\Delta m_F = -1$.

The level population in the ground state was quickly shifted to lower m_F levels. By successive excitations, the population of ions was accumulated in the $^2\text{S}_{1/2} (2, -2)$ level. The optically pumped ions were excited only to the $^2\text{P}_{3/2} (3, -3)$ level, from which ions could decay to the original $^2\text{S}_{1/2} (2, -2)$ level. Thus, we realized the optical-pumping and laser-cooling cycle at the same time. Once ions are optically pumped and cooled to a low kinetic-energy state, each ion radiates strong fluores-

Fig. 1. Partial energy-level diagram of the $^9\text{Be}^+$ ion (not scaled). In a weak magnetic field, the hyperfine F states split into m_F sublevels (right hand side).

cence on the order of 10^6 photons per second. If we introduce microwave radiation with a resonant frequency of the $(2, -2) \leftrightarrow (1, -1)$ transition in the ground-state HFS, a part of the population in the $(2, -2)$ level transfers to the $(1, -1)$ level. Therefore, the strong fluorescence yield decreases due to the destruction of optical pumping. We observe a minimum in the fluorescence intensity spectrum at the microwave resonance.

If we change the laser polarization to σ^+ , we can pump all ions into the maximum m_F level of $(2, +2)$. Thus, we can also measure the resonant frequency of the $(2, +2) \leftrightarrow (1, +1)$ transition.

Since the resonant frequencies of the $(2, +2) \leftrightarrow (1, +1)$ and $(2, -2) \leftrightarrow (1, -1)$ transitions in the ground-state HFS are given as a closed form of the Breit-Rabi formula, the magnetic HFS constant can be obtained by fitting the measured transition frequencies to the formula. In our case, it is sufficient to take two independent parameters, i.e., the magnetic dipole HFS constant (A) and the applied magnetic field (B), since the effect of the g_f factor in the formula can be ignored in a weak magnetic field.

A schematic diagram of the experimental apparatus is shown in Fig. 2. In order to laser cool and detect the fluorescence from trapped ions, we used 313 nm UV laser light obtained by intracavity second-harmonic generation using an angle-tuned LiIO_3 crystal placed in a single-mode tunable ring dye laser (Coherent 899-21). The pumping laser for the dye laser was an argon ion laser (Coherent Innova 400, 20 W). The line width of the UV laser was smaller than 1 MHz without an extra stabilization system. The laser frequency could be scanned up to 15 GHz (30 GHz for UV).

Circular polarization of the UV laser light was produced by a Gran-Taylor prism and a $\lambda/4$ wave plate. The rotation angle of the wave plate was remote controlled by a stepping motor, which enabled us to change the σ^+ or the σ^- polarization quickly. The beam radius of the UV laser at the trap center was estimated to be about 170 μm .

Fig. 2. Schematic diagram of the experimental apparatus.

For tuning the UV laser frequency to the $2S_{1/2} \rightarrow 2P_{3/2}$ transition, we monitored the fundamental laser wavelength (626 nm) using a commercial wavemeter (Advantest, TQ8325). The resolution of the wavemeter was 1 pm and the accuracy 5 ppm. The molecular iodine absorption spectra were also used to calibrate the laser frequency.

The fluorescence of the stored ions was focused by a lens system, and detected by a photon-counting image system (Hamamatsu, PIAS-TI). The lens system was designed so as to obtain a high total magnification (approximately $\times 20$) in order to discriminate each single ion in an ion crystal. To reduce a background signal, an interference filter for 313 nm was inserted in the optical path. The transmittance was about 14%. The total detection efficiency of the optical system was estimated to be 1×10^{-4} by a numerical simulation.

The image of the fluorescence yield was analyzed by a position analyzer, from which the position and time data were recorded event by event. This list-mode data-acquisition system was essential for the double-resonance experiment in the pulsed-microwave method (See Sec.3.3).

For laser cooling and optical pumping of $^9\text{Be}^+$ ions, we applied a weak static magnetic field to the ions using a Helmholtz coil, which could generate a magnetic field of up to 1 mT at the center of a linear trap. The direction of the applied magnetic field was parallel to the laser-beam propagation. To eliminate the stray magnetic field, we also placed two Helmholtz coils.

A microwave signal was generated in a precise microwave synthesizer (Hewlett-Packard, HP8665B) and amplified up to 4 W by a solid-state amplifier (R&K, A250HP-SMA); it was then irradiated onto the stored ions through one of the trap electrodes.

§3. Results and Discussions

3.1 Laser Cooling of $^9\text{Be}^+$ Ions

A typical frequency spectrum from laser-cooled $^9\text{Be}^+$ ions is shown in Fig. 3. The spectrum was obtained by scanning the UV laser frequency from the lower side of the resonance frequency. Since the trapped ions were sufficiently laser cooled, crystallization of the trapped

ions occurred (Fig. 3 (a)). The achievement of optical pumping into the maximum (or minimum) m_F state was also shown in observing this phenomenon.

In the case of Fig. 3, the residual Doppler width was about 60 MHz, which was estimated by fitting a part of the spectrum to a numerically evaluated Voigt profile. The line width was sufficiently narrow to discriminate the HFS splitting of $^9\text{Be}^+$, while that of the buffer-gas-cooled ions was as wide as about 16 GHz (Fig. 4).

Fig. 3. Laser-cooling effect on $^9\text{Be}^+$ ions. (a) Image of crystallized 11 $^9\text{Be}^+$ ions. The fluorescence signals were integrated for 44 s. (b) Typical frequency spectrum of laser-cooled $^9\text{Be}^+$ ions. The polarization of the laser was set to σ^+ . The sudden decrease of the fluorescence in the figure indicates that the trapped ions were crystallized. The ions disappeared at the higher side of the line center due to laser heating.

3.2 Laser-Microwave Double-Resonance Spectroscopy of $^9\text{Be}^+$ Ions

In double-resonance experiments, we fixed the UV laser frequency to approximately -300 MHz lower than the $^2\text{S}_{1/2} (2, -2) \rightarrow ^2\text{P}_{3/2} (3, -3)$ transition frequency. We then introduced microwave radiation.

The double-resonance spectrum of the $(2, -2) \rightarrow (1, -1)$ transition in the ground-state HFS at about 0.7 mT is shown in Fig. 5. The microwave frequency was swept under continuous irradiation of the UV laser light. Under the present condition, the Doppler width was relatively wide compared with Fig. 3, because the number of ions was larger (10^2 - 10^3). The resonant frequency in the spectrum was obtained to be 1264.757(2) MHz, which corresponds to a precision of 2×10^{-6} . The line width (FWHM) of the spectrum was 144(6) kHz.

Fig. 4. Fluorescence-intensity spectrum from buffer-gas-cooled ions as a function of the UV laser frequency. A Doppler width of 16.1(1) GHz was obtained by fitting the data to a Gaussian distribution function. We can not discriminate the ground HFS states under the buffer-gas-cooling condition, since the HFS splitting is about 1 GHz. The buffer gas pressure was 5.4×10^{-4} Pa.

Fig. 5. Fluorescence-intensity spectrum as a function of the microwave frequency. The $(F, m_F) = (2, -2) \rightarrow (1, -1)$ transition in the $^2\text{S}_{1/2}$ state was induced at about 0.7 mT. The UV laser and microwave radiation were continuously applied to the stored ions. At the resonance, a clear hollow was observed with a high signal-to-background ratio and high statistics.

In this measurement, the stored ions were always laser cooled due to continuous laser radiation. Thus, the Doppler width was kept smaller than the HFS splitting, and we could apply high-power microwave radiation to saturate the transition (the output power of the microwave radiation was 0.4 W). The maximum signal-to-background ratio as well as high statistics were easily achieved.

In the case of experiments for unstable $^{7,11}\text{Be}^+$ ions, the ground-state hyperfine constants are unknown. We must sweep the microwave frequency over a wide range in order to find the resonance frequencies. Thus, the present method, which was first tested with $^9\text{Be}^+$, will be useful for searching the microwave resonance frequencies of $^{7,11}\text{Be}^+$ during the first stage of each experiment.

The line width, on the other hand, was not narrow.

This is because a high excitation rate limits the coherent interaction time. Moreover, the intense laser field could cause a light shift in the HFS states.^{18,19} Thus, for more precise measurements, the laser light should be turned off when the microwave radiation is irradiated onto the ions, though we may lose simplicity and sensitivity.

3.3 Pulsed Microwave Method

We have applied the pulsed-microwave method while aiming at a precise measurement. The timing sequence of the method is shown in Fig. 6. Initially, trapped ions were laser cooled and optically pumped to the maximum or minimum m_F level during the period T_L . We then turned off the UV laser light and applied microwave radiation to the trapped ions for a duration of τ . After that, we turned the laser light on and counted the fluorescent photons for a certain period (t_{int}). The fluorescence intensity then obtained a steady state because optical pumping was achieved again. The integrated fluorescence intensity reflects any population difference between the $(1, +1(-1))$ and $(2, +2(-2))$ levels. We could thus observe the microwave resonance.

Fig. 6. Timing sequence of the pulsed-microwave method. Laser and microwave radiation were alternatively applied. The fluorescence yield was integrated from right after turning off the microwaves for a certain period, t_{int} (hatched area).

In the present experiment, appropriate power and duration of the microwave radiation should be given by the Rabi single-pulse method. The optical-pumped level $(2, +2(-2))$ and the corresponding level $(1, +1(-1))$ could be considered to be a two-level system when the laser light is turned off and the microwave radiation is turned on. We denote these levels as 1 and 2, respectively. If we define the interaction time (τ) between the applied microwave pulse and the stored ions, the population for each level after microwave-pulse irradiation can be written as

$$\begin{aligned}\rho_{11}(\tau) &= \frac{1}{2} + [\rho_{11}(0) - \rho_{22}(0)] \left[\frac{b^2}{2\Omega^2} (1 - \cos \Omega\tau) \right], \\ \rho_{22}(\tau) &= \frac{1}{2} - [\rho_{11}(0) - \rho_{22}(0)] \left[\frac{b^2}{2\Omega^2} (1 - \cos \Omega\tau) \right],\end{aligned}\quad (3.1)$$

$$\Omega = \sqrt{b^2 + \Delta\omega_m^2},$$

where $\Delta\omega_m$ and b are the detuning from the resonance

angular frequency and the amplitude of the applied perturbation, respectively. The maximum of the population difference at the resonance can be obtained when $b\tau = (2n+1)\pi$ (n :integer) is satisfied. Under the π -pulse condition of $b\tau = \pi$, the spectrum has one deep minimum.

Under the actual experimental conditions, there is a problem to perform pulsed measurements. When we turn off the laser light, ions are not laser cooled and rf heating occurs; then, the Doppler width increases. The intensity of the double-resonance spectrum in the pulsed method strongly depends on the Doppler width, the laser parameters and the integration time of the fluorescence yield (t_{int}). It thus becomes relatively hard to observe a double-resonance spectrum compared when using the previous method.

3.4 Simulation of the Pulsed-Microwave Method

In order to search for the dependences of the intensity of the double-resonance signal on the Doppler width as well as the laser parameters in the pulsed-microwave method, we performed numerical simulations.

We consider the problem by dividing it to the following three steps: (i) Set the initial condition to $\rho_{11}(0)=1$ and $\rho_{22}(0)=0$ in Eqs. (3.1). In principle, we can achieve complete optical pumping into the $(F, m_F)=(2, +2(-2))$ levels by irradiating the circularly polarized laser light. (ii) Carry out calculations of the populations after irradiation of the microwave pulse, $\rho_{11}(\tau)$ and $\rho_{22}(\tau)$. (iii) Solve the rate equations numerically for all of the populations, including the excited levels under the initial condition calculated in step (ii). Then, the excited-state population is integrated for a certain period of t_{int} .

For simplifying the calculations in step (iii), we assumed that the velocity distribution of ions can be described by the Maxwell-Boltzmann distribution, that the Doppler width is much larger than the natural line width of the optical transition (~ 18 MHz),²¹ and that the intensity of the laser light is low enough to maintain a much smaller population of the excited state than that of the ground state. The last two assumptions always hold for Be^+ , even if ions are laser cooled to a few Kelvin and the laser intensity is up to a few hundred mW/cm².

Under these assumptions, one can write down the following rate equations:

$$\frac{dn_{g_j}}{dt} = \sum_i A_{e_i g_j} n_{e_i} + \sum_i W_{e_i g_j} n_{e_i} - \sum_i W_{g_j e_i} n_{g_j}, \quad (3.2)$$

$$\frac{dn_{e_k}}{dt} = \sum_i W_{g_i e_k} n_{g_i} - \sum_i W_{e_k g_i} n_{e_k} - \sum_i A_{e_k g_i} n_{e_k}, \quad (3.3)$$

$$\sum_j n_{g_j} + \sum_k n_{e_k} = 1, \quad (3.4)$$

where n_g and n_e indicate the ground and excited m_F level populations, respectively. N_e and N_g represent the number of excited- and ground-state sublevels, respectively. We needed a set of 24 equations to solve all level

populations. A_{eg} and W_{eg} are the spontaneous emission rate and the stimulated emission rate, which are given as²⁰⁾

$$A_{F_c m_c \rightarrow F_g m_g} = (a_{F_c m_c \rightarrow F_g m_g})^2 A_{J_c \rightarrow J_g}, \quad (3.5)$$

$$W_{F_c m_c \rightarrow F_g m_g} = \frac{3\pi^2 c^2 I_L}{\hbar \omega_L^3} (a_{F_c m_c \rightarrow F_g m_g})^2 A_{J_c \rightarrow J_g} \times \int_{-\infty}^{\infty} S_L(\Delta\nu_L, v_z) g(v_z) dv_z, \quad (3.6)$$

where c and $\hbar$ represent the speed of light in a vacuum and Planck's constant, respectively; $a_{F_c m_c \rightarrow F_g m_g}$ is the transfer coefficient, which determines the branching ratio of the spontaneous emission rate.²⁰⁾ I_L and ω_L indicate the intensity and the angular frequency, respectively. The laser detuning ($\Delta\nu_L$) was defined as the deviation from the resonant frequency of the $^2S_{1/2}$ ($2, +2(-2) \rightarrow ^2P_{3/2}$ ($3, +3(-3)$)) transition. $A_{J_c \rightarrow J_g}$ can be obtained from the lifetime of the $P_{3/2}$ state of $^9\text{Be}^+$, which is 8.77 ns.²¹⁾ The excitation rate ($W_{F_g m_g \rightarrow F_c m_c}$) is obtained in the same manner as $W_{F_c m_c \rightarrow F_g m_g}$. S_L is a Lorentzian function, which describes the frequency distribution of the light absorption for each ion; $g(v_z)$ represents the Maxwell-Boltzmann distribution, where we have only considered one component along the direction of laser propagation. Since the line width of the laser light was much smaller than the natural and the Doppler widths ($\Delta\nu_D$), it can be assumed to be monochromatic light.

We calculated the time dependence of the excited-state populations ($\sum n_{ek}(t)$) and integrated it from 0 to t_{int} . We defined the integrated populations as $I(t_{int})$. By calculating $I(t_{int})$ as a function of the microwave detuning, we obtained the calculated double-resonance spectrum in the pulsed-microwave method.

An example of the calculated spectrum is shown in Fig. 7. The integration time was taken to be 200 μs . The values of the other parameters are given in the figure caption. The ground-state Rabi resonance curve was directly reflected in the excited-state population.

Here, we define the signal-to-background ratio ($R_{S/B}$) as

$$R_{S/B} = I_S(t_{int})/I_B(t_{int}). \quad (3.7)$$

The background intensity (I_B) means the integrated intensity at completely off-resonance of the microwave transition, while I_S indicates the intensity at the resonance (See Fig. 7). If we increase the integration time to observe the spectrum, $R_{S/B}$ quickly decreases, since the effect of the microwave resonance decreases.

In order to find suitable conditions to observe the double-resonance spectrum, we calculated the dependences of $R_{S/B}$ on the laser parameters and the Doppler width of the optical transition. In the following calculations the magnetic field strength was fixed to 0.7 mT and the π -pulse condition in the Rabi's method was assumed. The laser intensity was also assumed to be 30 mW/cm², which was a typical value in the present experiment.

Fig. 7. Calculated microwave spectrum in the pulsed-microwave method. The integrated population of the excited state as a function of the microwave detuning. The fluorescence yield is obtained by multiplying the spontaneous emission rate by the number of ions. In this simulation, the π -pulse condition was assumed and the following parameters were used: $\tau=400 \mu\text{s}$, $t_{int}=200 \mu\text{s}$, $\Delta\nu_D=600 \text{ MHz}$, $\Delta\nu_L=-200 \text{ MHz}$, $I_L=30 \text{ mW/cm}^2$.

Since the difference of $R_{S/B}$ with a change in the laser polarization is small under the weak magnetic field condition, we only show the result of a calculation for the case of σ^+ polarization.

Figure 8 shows the dependence of $R_{S/B}$ as a function of the integration time (t_{int}). Several cases for the Doppler width ($\Delta\nu_D$) were plotted. The laser detuning was set to -200 MHz. In the case of $\Delta\nu_D < 800 \text{ MHz}$, the magnitude of $R_{S/B}$ quickly decreases as t_{int} increases. We can measure the microwave resonance with a higher $R_{S/B}$ for a smaller Doppler width.

Fig. 8. Integration time (t_{int}) dependence of $R_{S/B}$ under a certain Doppler width. The detuning and intensity of the laser light were set to -200 MHz and 30 mW/cm², respectively. The duration of the microwave radiation was $\tau=400 \mu\text{s}$.

Laser detuning is also important. Figure 9 shows $R_{S/B}$ as a function of t_{int} for several cases of laser detuning. The Doppler width was assumed to be $\Delta\nu_D=600 \text{ MHz}$, which was a realistic value under our experimental conditions. If we tune the laser frequency very close to the $F=2$ state, e.g., $\Delta\nu_L=-60 \text{ MHz}$, we can observe microwave resonance with a high $R_{S/B}$. On the other hand,

Fig. 9. $R_{S/B}$ as a function of t_{int} under various laser detunings. The Doppler width and laser power were set to 600 MHz and 30 mW/cm², respectively. The duration of the microwave radiation was $\tau=400$ μ s.

if we set the laser frequency closer to the $F = 1$ state than to the $F = 2$ state, which corresponds to $\Delta\nu_L \leq -617$ MHz, $R_{S/B}$ has a negative value. This means that a peak spectrum is observed. In this condition, the excitation rate should be low and the cooling power of the laser is insufficient.

Obviously, the simulation results show that the small laser detuning and the small Doppler width are good conditions to observe the microwave resonance with a high $R_{S/B}$. However, these two conditions are somewhat contradictions. Very small laser detuning is inefficient for laser cooling. The proper laser detuning must be determined experimentally according to the guideline given by the simulation.

The simulation results also show that the very short integration time ($t_{int} \leq 1$ ms) yields a high $R_{S/B}$. In this mode, we have to repeat measurements many times in order to obtain sufficient statistics. This can be realized by using a rapid optical chopper.

In principle, the line width of the resonance becomes narrow as the duration of the microwave pulse increases under the π -pulse condition. However, the Doppler width also increases due to the rf heating effect during the laser-off period, as mentioned before, so that the signal-to-background ratio ($R_{S/B}$) quickly decreases. Moreover, the transition frequencies are very sensitive to any fluctuation of the magnetic field, because we measure the field-dependent transitions ($d\nu_{\pm}/dB \sim \pm 20$ kHz/ μ T at around 0.7 mT). The precision of the resonant frequency is already limited by the stability of the magnetic field. Thus, we set the duration of the microwave pulse to a realistic value of 400 μ s.

3.5 Results of the Pulsed-Microwave Method

The observed microwave spectra of the $(2, +2(-2)) \rightarrow (1, +1(-1))$ transitions are shown in Fig. 10 (a) (b). In order to realize efficient laser cooling and quick repumping after the microwave transitions, the detuning was set to about -190 MHz. The power of the laser was set to a maximum value of about 90 μ W under the present condition. The microwave pulse was 400 μ s and the duty ratio of the pulse was set to about 30% of the total pe-

riod of the sequence. The output power of the microwave radiation after the amplifier was about 13 mW.

By fitting the spectra to a Gaussian profile, the resonant frequencies were obtained as

$$\nu_- = 1264.7353(1) \text{ MHz},$$

$$\nu_+ = 1235.4149(3) \text{ MHz},$$

where the uncertainties represent the statistical errors. The precisions were 8×10^{-8} and 2×10^{-7} , respectively.

Fig. 10. Observed double-resonance spectra of (a) the $(F, m_F)=(2, +2) \rightarrow (1, +1)$ transition (σ^- polarization case) and (b) the $(2, -2) \rightarrow (1, -1)$ transition (σ^+ polarization case) at approximately 0.7 mT. The integration time (t_{int}) was about 900 μ s. The line widths (FWHM) were obtained as (a) 5.9(7) kHz and (b) 7.0(3) kHz, respectively.

In the pulsed-microwave method, the origins of the line width come from: 1) the interaction time of microwave radiation with ions, which is determined by the duration and power of the microwave pulse; 2) the short-term instability of the magnetic field. The former was given by the duration of the microwave pulse, $\tau=400$ μ s, which corresponds to a width of 2 kHz under the π -pulse condition.

We measured the double-resonance spectra at various microwave powers in order to find a proper condition. When we gradually decreased the microwave power down to about 0.1 mW, the width did not become narrow, but the signal became small. At a high power of about 400 mW, we observed a wide spectrum, as shown in Fig. 11. On the other hand, we never observed Rabi resonance curves like the one in Fig. 7. These facts suggested that the dominant contribution to the line width was from the short-term fluctuation of the magnetic field, i.e., the AC magnetic field from the surroundings.

Fig. 11. Power broadening of the microwave spectrum. The laser polarization was σ^- . The resonant frequency and width (FWHM) were $\nu_0=1264.7204(2)$ MHz and $20.2(4)$ kHz, respectively. The condition of the magnetic field strength was slightly different from that in Fig. 10.

The frequency shifts caused by the long-term drift of the magnetic field strength were $\Delta\nu/\Delta t = \pm 83.3$ Hz/min. during the present measurement; meanwhile, it took 6 minutes to obtain the two spectra in Fig. 10. Thus, a frequency shift of ± 500 Hz could possibly occur. By adding these systematic errors to the statistical errors of the measured frequencies, we could derive the HFS constant from the measured transition frequencies.

By a least-square fitting of the measured frequencies in Fig. 10 to the Breit-Rabi formula, we obtained the ground-state magnetic dipole hyperfine constant of $^9\text{Be}^+$ as

$$A_9 = -625.00882(26) \text{ MHz.}$$

The accuracy was $\Delta A_9/A_9 = 4 \times 10^{-7}$. The magnetic field was also obtained as $6.97517(22) \times 10^{-4}$ T. The result was consistent with that measured at a strong magnetic field by the NIST group.²²⁾

§4. Conclusion

We have established both highly sensitive and precise laser-microwave double-resonance spectroscopy for laser-cooled Be ions in a weak magnetic field.

If we measure the magnetic HFS constants of unstable $^7,^{11}\text{Be}^+$ ions with the same accuracy of the present $^9\text{Be}^+$ experiment, it should be sufficient values to study the hyperfine anomaly.

We can also obtain the unknown nuclear g_I factor using the relation

$$g_{7,11} \approx g_9 A_{7,11}/A_9,$$

where the accuracy is limited by the differential hyperfine anomaly (Δ), which might be 10^{-5} for ^7Be , and 10^{-3} for ^{11}Be .²³⁾ These g_I factors are also important to set the proper magnetic field to measure the accurate g_I factors in the next step of our project.

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