

**Single-ion spectroscopy of the $^2S_{1/2} - ^2D_{5/2}$
clock transition in Ba^+ ions and
laser cooling technique for their odd isotopes**

Hiroto Fujisaki

Single-ion spectroscopy of the $^2S_{1/2}$ - $^2D_{5/2}$
clock transition in Ba^+ ions and
laser cooling technique for their odd isotopes

Hiroto Fujisaki

Electronic Science and Engineering
Kyoto University

Abstract

The present research was initiated with the development of a laser capable of spectrally resolving clock transitions (i.e. clock laser). Given that clock transitions exhibit a narrow spectral width, clock lasers typically employ high-speed feedback control to narrow the oscillation line width, thereby achieving resonance within a high-finesse optical cavity. The infrared region is where the clock transition of Ba^+ is located, which poses a significant challenge in the procurement of both the laser itself and the high-reflectivity mirrors required for the high-finesse optical cavity. In the clock laser that excites the $1.76\text{ }\mu\text{m}$ $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition, the laser with a wavelength of 881 nm was first narrowed in linewidth to the resonance of a high-finesse optical cavity. The generation of the second harmonic of the $1.76\text{ }\mu\text{m}$ laser, followed by the detection of the beat with the narrowed linewidth 881 nm laser and phase-synchronisation, resulted in the creation of the clock laser. The linewidth of the 881 nm laser was evaluated at 200 Hz, in comparison to a laser with a linewidth narrowed by resonance in a different optical cavity. Given that the wavelength of the laser under comparison was 822 nm, the optical frequency comb (optical comb) was phase-locked to this, narrowing the linewidth of each mode of the optical comb. This was then used as an intermediary for comparison. The linewidth of the $1.76\text{ }\mu\text{m}$ laser is estimated to be 100 Hz, half of that.(this thesis, Chapter 3)

The $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ clock transition, which is necessary to achieve the final goal, has a wavelength of $2.05\text{ }\mu\text{m}$, and it is difficult to obtain a frequency-tunable laser. Therefore, we decided to use the method of generating a narrow-linewidth laser beam by phase-synchronising two lasers with the aforementioned line-narrowed optical comb and then mixing their difference frequencies. In this study, the difference frequency was set to $1.76\text{ }\mu\text{m}$, and the linewidth was evaluated in comparison to the aforementioned clock laser. The spectral width of the beat signal of the two lasers was 100 Hz, demonstrating the effectiveness of this method. It is possible to achieve a narrow linewidth laser beam with a wavelength of $2.05\text{ }\mu\text{m}$ by changing the wavelength of the

laser that generates the difference frequency. The observed linewidth is more than one order of magnitude narrower than the oscillation linewidth previously reported for difference frequency generation using a similar optical comb (Chapter 4).

Subsequently, a technique was established for the capture and cooling of a single Ba^+ ion in a linear RF trap. Given that the ultimate objective is the utilisation of multiple ions, a linear RF trap that can cool multiple ions along the trap axis was adopted. However, the primary objective of this study was to establish a technique to cool a single trapped ion to the Lamb-Dicke bound condition, where the range of ion motion is less than the wavelength of the clock transition, so that a carrier spectrum without Doppler shift can be clearly observed. In addition, Ba reacts violently in the air and is difficult to process; therefore, a getter was used as the source of Ba atoms. This represents a novel application of a getter in this context, and the study has demonstrated the efficacy of this approach for trapping a single Ba^+ ion. The generation of Ba atoms from the getter, their subsequent trapping through photoionisation within the trap, and their cooling through laser cooling via the 493 nm $^2\text{S}_{1/2} - ^2\text{P}_{1/2}$ transition, with repumping through the 650 nm $^2\text{D}_{3/2} - ^2\text{P}_{1/2}$ transition to complete the cooling cycle, were all achieved. The light sources employed were all semiconductor lasers or their second harmonic. The generation of Ba atoms from the getter was found to be distributed over a wide area, and the evaporation rate was found to be difficult to control. This made it difficult to introduce ions one by one into the trap. Therefore, a method was established for easily measuring the number of ions introduced by irradiating them with an LED. In real traps, unintended electrostatic fields usually occur, and the ions are not captured at the centre of the trap. If they are displaced from the centre of the trap, the RF electric field used in the trap becomes stronger, and the ions are excessively excited (excessive micromotion). The detection of this excessive micromotion was first achieved through the utilisation of a cooling laser irradiated from two directions, at an angle of 3 degrees from the axis, and then from the axis. The subsequent minimisation of this motion was accomplished through the superimposition of a DC voltage on the electrodes, thus demonstrating the efficacy of the method. Subsequently, a clock laser with a narrowed line width was utilised to perform single-ion spectroscopy of the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ clock transition. This process enabled the observation of vibrational sidebands, arising from the permanent motion of the ions, through their separation from the carrier spectrum. Furthermore, a spectrum was detected in which the carrier exhibited a significant magnitude superior to the

sidebands. With regard to the trap axis direction, the temperature reached by the permanent motion was evaluated to be 3.5 mK, thereby confirming the achievement of the Lamb-Dicke bound condition (Chapter 6). Concurrently, we successfully trapped and laser-cooled a single odd-mass isotope, $^{137}\text{Ba}^+$. The odd-mass isotopes $^{135}\text{Ba}^+$, $^{137}\text{Ba}^+$ possess hyperfine structures, necessitating the utilisation of two cooling lasers and three repumping lasers with distinct frequencies to excite all of the hyperfine structures. We configured independent lasers and phase-synchronised them as required to stabilise the frequencies. It was determined that the optical ionisation method employed in this study is incapable of separating $^{135}\text{Ba}^+$ and $^{137}\text{Ba}^+$. However, it was ascertained that if the ions are single, they can be introduced into the trap and laser cooled for specific isotopes (Chapter 7).

Acknowledgement

This study was conducted at quantum optical engineering laboratory in Kyoto University. I was able to complete this thesis under the long-term research guidance in Kyoto University.

First of all, I would like to express my sincere gratitude to Professor Yoichi Kawakami for giving me the opportunity to complete this research for my doctoral thesis.

I would also like to show my sincere gratitude to Professor Susumu Noda for taking time out of his busy schedule to give me various advices and suggestions on my research.

I would also like to acknowledge with appreciation the crucial role of Associate Professor Kazuhiko Sugiyama for his direct guidance, technical advice, and support in the laboratory.

A special gratitude I give to Professor Masao Kitano for giving me the opportunity to do interesting research in the laboratory.

I am grateful to Lecturer Toshihiro Nakanishi for the various technical in our discussions.

I am particularly grateful to Dr. Yasutaka Imai for helping me with my experiments until late at night and for his strong emotional support as a Ph.D. student.

I would like to express my gratitude to Dr. Masatoshi Mitaki, Mr. Yuya Nobuoka, Mr. Yoshikazu Hara, Mr. Yoshikazu Ideta, Mr. Keisuke Nishida, Mr. Shinnya Kawada and Mr. Shinnosuke Yukawa for helping me with my experiments in the laboratory.

I would like to thank Mr. Dai Osada, Mr. Ryuji Moriya, Mr. Ryohei Shibata and Mr. Kengo Takeshita for helping me complete this research as a co-researcher.

I would like to thank , Mr. Shohei Higashitani, Dr. Shuhei Hatanaka and Mr. Ken Hamada for various support at quantum optical engineering laboratory.

We are grateful to Professor Shinji Urabe of Osaka University for showing

us his linear-RF trap and Dr. Kazuhiro Hayasaka of the National Institute of Information and Communications Technology (NICT) for his support when designing the trap electrodes.

We are grateful to Dr. Jun Ishikawa of the National Institute of Advanced Industrial Science and Technology/National Metrology Institute of Japan (AIST/NMIJ) for fabricating the iodine cell.

I would like to thank Anritsu Corporation for understanding that I would be obtaining a degree after joining the company.

Finally, I would like to express my gratitude to my family for their financial and emotional support.

This study was partially supported by Grants-in-Aid for Scientific Research 21104006, 26287092, 21H01013, and JST-CREST.

Contents

1	Introduction	1
2	Principle	5
2.1	Barium ion	5
2.2	Paul trap	7
2.3	laser cooling	10
2.4	Laser linewidth reduction	11
2.4.1	FM sideband technique	11
2.4.2	Phase locked loop	17
3	Clock laser for the $^2S_{1/2}$ - $^2D_{5/2}$ clock transition at $1.76 \mu\text{m}$	23
4	Clock laser using difference frequency generation and optical frequency comb	31
4.1	Experimental setup	31
4.2	Results	33
4.3	Conclusion	35
5	Trapping and laser cooling of single $^{138}\text{Ba}^+$ in linear-RF trap	37
5.1	Light sources	40
5.1.1	Cooling laser	40
5.1.2	Repumping laser	42
5.1.3	Photoionization lasers	42
5.1.4	LEDs	44
5.2	Ion trap	44
5.2.1	Linear-RF trap	45
5.2.2	Beam steering	47
5.3	Results	47
5.3.1	Loading of trapped single $^{138}\text{Ba}^+$ ions	48
5.3.2	Micromotion minimization	50

6	Single-ion spectroscopy of the $^2S_{1/2}$ - $^2D_{5/2}$ clock transition	57
6.1	Procedure	57
6.2	Results	58
6.3	Conclusions	60
7	Trapping and laser cooling of odd isotopes of Ba^+ ions	63
7.1	Laser setup	63
7.2	Results	63
8	Conclusion	71
8.1	Prospects	72
8.1.1	optical clock using $^{138}Ba^+$ ions	73
8.1.2	multi-ion clock using odd isotopes	73

Chapter 1

Introduction

Singly ionized barium (Ba^+) has been attracted for use in various experiments using ion trap technique, such as optical frequency standards or simply called optical clocks [1], tests of the fundamental physics [2], and quantum information processing [3, 4], since the pioneering work of laser cooling [5] and observation of single ions [6] have been demonstrated using Ba^+ ions confined in radiofrequency (RF) traps. Regarding to the application to optical clocks, Ba^+ has clock transitions of narrow natural widths suitable for the reference frequencies. It has also cooling transitions of short lifetime for laser cooling, which are simultaneously used for state detection by detecting the fluorescence emitted from the upper state. As for the clock transitions, the lifetimes of the $^2\text{D}_{3/2}$ and $^2\text{D}_{5/2}$ states are $79.8(4.6)\text{ s}$ [7] and $31.2(0.9)\text{ s}$ [8], respectively. These are fairly long in the ion species used with ion trap technique. Long lifetime is essential for the clock transition referenced in the single-ion optical clock to make use of an advantage in ion trap technique of long interaction time. Therefore, the $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition at 20 nm and the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition at 1762 nm have potentials for the references used in optical clocks.

In addition, the $^2\text{D}_{3/2}$ state in the odd isotopes $^{135,137}\text{Ba}^+$ has the $F = 0$ hyperfine state, owing to the nuclear spin $I = 3/2$. Therefore, the $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition in $^{135,137}\text{Ba}^+$ is predicted to be insensitive to electric-field gradient owing to a very small electric quadrupole moment. [1] This property enables us to increase the number of trapped ions without degrading the uncertainty in the reference frequency. The $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition, of which the frequency was measured using single $^{138}\text{Ba}^+$ ions with an uncertainty of 10^{-15} level, [9] is also proposed to be applied to the multi-ion clock when dynamic decoupling techniques are applied. [10, 11] The $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$

transition is also useful in the quantum information processing for operation of an optical qubit [12,13] or application to state detection, [12] and sideband cooling. [14] In this work, we develop a system of a single-ion spectroscopy of the $^2S_{1/2} - ^2D_{5/2}$ transition in $^{138}\text{Ba}^+$ using diode-laser-based light sources towards realization of an optical clock.

For driving the $^2S_{1/2} - ^2D_{5/2}$ transition at 1762 nm, a color-center laser was first used. [15,16] Then, thulium-doped fiber lasers have been applied as more convenient light sources. [12,14] Diode lasers around 1.76 μm have been available several years ago and this allows us to easily build up tunable lasers as we compose extended-cavity diode lasers (ECDLs). [17] A few works using diode lasers at 1.76 μm have been reported so far. [9,13] To linewidth and frequency stabilize the clock laser, the high-finesse cavities are used as a reference to be feedback controlled. High reflection coatings for the high-finesse cavity are available in 1.76 μm region. [13,14] As an alternative approach, phase locking to a mode of an optical frequency comb (OFC) phase locked to a linewidth-narrowed laser at a different wavelength was used. [9] In this work, we phase lock an ECDL at 1.76 μm to a high-finesse-cavity-stabilized ECDL at 881 nm via second-harmonic generation (SHG). We show that this is also useful for another alternative scheme of the clock laser. The linewidth transfer via SHG has a potential for a better scheme than that via OFC. The power of the second harmonics could be larger than that of one mode of OFC. The linewidth at the fundamental laser is reduced by half from that of the high-finesse-cavity-stabilized laser.

We have one more reason why we adopt this scheme of clock laser. We have a plan of frequency comparison to the clock transitions in singly-ionized ytterbium ions (Yb^+). This enables us to evaluate the uncertainty and reproducibility. Moreover, the $^2S_{1/2} - ^2F_{7/2}$ transition in Yb^+ at 467 nm has the largest sensitivity to the variation of the fine structure constant α among the known clock transitions in atoms and singly-ionized ions. [18] The $^2S_{1/2} - ^2D_{3/2,5/2}$ transitions in Ba^+ have modest sensitivities to the α variation, which are opposite sign to that of the $^2S_{1/2} - ^2F_{7/2}$ transition in Yb^+ . [18] The comparison between the Ba^+ and Yb^+ transitions enhances the sensitivity by % from the comparison between the $^2S_{1/2} - ^2D_{3/2}$ and the $^2S_{1/2} - ^2F_{7/2}$ transitions in only Yb^+ . [19,20] Therefore, we develop a clock laser system by sharing one high-finesse cavity for the doubled frequency of the $^2S_{1/2} - ^2D_{5/2}$ transition in Ba^+ at 881 nm and the half frequency of the $^2S_{1/2} - ^2F_{7/2}$ transition at 934 nm. It could be possible to cancel the drift of the resonances of the high-finesse cavity when we conduct the future plan of the comparison.

Moreover, we have conducted two research subjects as a next step towards realization of a multi-ion optical clock using the $^2S_{1/2} - ^2D_{3/2}$ transition in $^{135}\text{Ba}^+$, $^{137}\text{Ba}^+$ ions. One is the development of the clock laser described in Chapter 3 and the other the isotope-selective photoionization loading of single $^{137}\text{Ba}^+$ ions.

the $F = 0$ hyperfine state in the $^2D_{3/2}$ in the odd isotopes $^{135}\text{Ba}^+$ and $^{137}\text{Ba}^+$ has very small electric quadrupole moment owing to the nuclear spin $I = 3/2$. [1] Therefore, the $^2S_{1/2} - ^2D_{3/2}$ clock transition to the $F = 0$ in the $^2D_{3/2}$ state is insensitive to electric field gradient, and this allows to increase the number of trapped ions without degrading the uncertainty in the transition frequency. The wavelength of the $^2S_{1/2} - ^2D_{3/2}$ clock transition is 2052 nm. Up to the present, a clock laser system based on a fiber laser has been developed [21]. The sophisticated techniques are required for realizing frequency tunability with narrow-line single-frequency oscillation in the fiber laser.

The phase locking of the two fundamental lasers to a line-narrowed optical frequency comb (OFC) is a potential scheme to realize a line-narrowed difference frequency radiation. We need one high-finesse cavity, which is used for line-narrowing a master laser to which the OFC is phase locked. The spectrum of the master laser is transferred to all modes of the OFC [22, 23]. This scheme of narrow-line difference frequency has been realized using an OFC based on a mode-locked fiber laser and was applied to molecular spectroscopy, where the linewidth was measured to be 3.5 kHz [24].

To line-narrow the difference frequency, we adopt difference frequency generation (DFG) to obtain coherent radiation at 2052 nm. To realize a narrow-line clock laser, fast feedback control to a resonance of a high-finesse cavity as shown in Chapter 3 is performed. When a laser is not available at the wavelength of the clock transition, nonlinear frequency conversion such as second-harmonic generation and sum or difference frequency generation is performed. When a sufficient power is generated in the sum or difference frequency generation, the narrow-line radiation is realized by controlling only one of the two fundamental lasers, as we analyze the frequency fluctuation using the generated radiation. [25, 26] In many cases, however, the generated radiation does not have a sufficiently large power. Two fundamental lasers are required to be independently line-narrowed before the generation. It is elaborating to develop two systems involved high-finesse cavities because considerable efforts are required to isolate the high-finesse cavity from vibrational and acoustic fluctuations.

The phase locking of the two fundamental lasers to a line-narrowed optical

frequency comb (OFC) is a potential scheme to realize a line-narrowed difference frequency radiation. We need one high-finesse cavity, which is used for line-narrowing a master laser to which the OFC is phase locked. The spectrum of the master laser is transferred to all modes of the OFC [22, 23]. This scheme of narrow-line difference frequency has been realized using an OFC based on a mode-locked fiber laser and was applied to molecular spectroscopy, where the linewidth was measured to be 3.5 kHz [24].

In this thesis, we investigate the potential of the scheme for realizing narrow-line difference frequency radiation. We generate the difference frequency radiation at 1762 nm. This is because we have independently developed a line-narrowed ECDL at 1762 nm for driving the $^2S_{1/2}$ – $^2D_{5/2}$ transition in Ba^+ as described in Chapter 3. We evaluate the absolute linewidth by comparing to the line-narrowed ECDL.

As mentioned above, because the odd isotope Ba ion has a nuclear spin of 3/2, two cooling laser beams and three repumping laser beams with different frequencies are required. We prepared independent lasers and phase-synchronised them as necessary to stabilise the frequency. Although the photoionisation method used in this study cannot separate $^{135}Ba^+$ and $^{137}Ba^+$, it was found that it is still possible to introduce single ions into the trap and laser cool specific isotopes.

The structure of this paper is as follows. First, Chapter 2 describes the important principles of this research. Chapter 3 describes the clock laser based on ECDL, and Chapter 4 describes the clock laser based on difference frequency generation. Chapter 5 describes ion trap technology and laser cooling technology. Chapter 6 describes clock transition spectroscopy using the clock laser described in Chapter 3. Chapter 7 describes laser cooling technology for odd isotope Ba^+ ions. Finally, in Chapter 8, we summarise the research and discuss future prospects.

Chapter 2

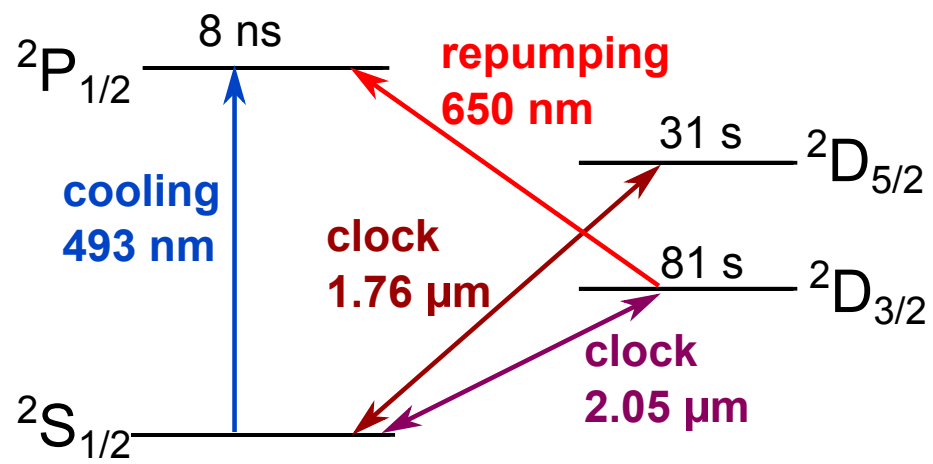
Principle

2.1 Barium ion

Barium (Ba) is an alkaline earth metal with an atomic number of 56. The pure element is highly reactive and difficult to handle, and there are seven isotopes of Ba. The isotope with the largest abundance is ^{138}Ba , which accounts for 71.7%. Furthermore, the abundance of odd-numbered isotopes is notably high, with ^{135}Ba and ^{137}Ba accounting for 6.59% and 11.2%, respectively. The transition structure of Ba^+ is illustrated in Figure 2.1. As illustrated in this figure, Ba^+ exhibits the $^2\text{S}_{1/2} - ^2\text{P}_{1/2}$ transition with a lifetime of 8 ns, which can be utilised for Doppler cooling, the $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition with a lifetime of 81 s, advantageous for clock transitions, and the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition with a lifetime of 31 s. Furthermore, the odd isotopes ^{135}Ba and ^{137}Ba possess a nuclear spin of $I = 3/2$, resulting in a hyperfine structure. Consequently, numerous lasers are required to cool them, yet the transition $(^2\text{S}_{1/2}, F = 2, m_F = 0) - (^2\text{D}_{3/2}, F = 0, m_F = 0)$ exhibits the following characteristics

- As this transition involves a change in m_F from 0 to 0, it does not undergo a first-order Zeeman shift.
- Furthermore, given that $^2\text{D}_{3/2}$ has $F = 0$, the $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition undergoes a minor frequency shift due to the quadrupole field.

Consequently, if the $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition of the odd isotopes $^{135}\text{Ba}^+$ or $^{137}\text{Ba}^+$ is adopted as the clock transition, a frequency standard with high frequency stability can be achieved without increasing the uncertainty by capturing and using multiple odd isotopes of Ba^+ .

Figure 2.1: level structure of Ba^+

2.2 Paul trap

Ion traps are technologies that use electromagnetic fields to trap ions in a ultra-high vacuum. There are two types of ion trap: the Penning trap, which uses an electrostatic field and a static magnetic field to trap ions, and the Paul trap, which uses an electrostatic field and a radio frequency (RF) field. The Paul trap is also known as an RF trap. Because the Penning trap uses a magnetic field to trap ions, the energy levels of the ions are split by the Zeeman effect. On the other hand, Paul traps do not use a large magnetic field to trap ions, and the energy level shift due to the Zeeman shift is relatively small. Therefore, in this study we used a linear RF trap, which is a type of RF trap. The outline of the structure is shown in Figure 2.2. Here the z axis is referred to as the axial direction of the trap and the x and y axes are referred to as the radial directions of the trap. In the linear RF trap used in this study, ions are trapped in the radial direction by the effective central force due to the RF voltage $V_0 \cos(\Omega t)$ and in the axial direction by the electrostatic field. As shown in Figure 2.2, the potential when an RF voltage $V_0 \cos(\Omega t)$ is applied to the electrode is given by

$$V(x, y, z, t) = \frac{V_0}{2} \left(1 + \frac{x^2 - y^2}{r_0^2} \right) \cos \Omega t. \quad (2.1)$$

Where r_0 is the distance between the trap axis and the electrode. The potential created by the electrostatic field is given by

$$U(x, y, z, t) = \frac{\kappa U_0}{z_0^2} \left(z^2 - \frac{x^2 + y^2}{2} \right). \quad (2.2)$$

Where κ is a factor determined by the structure of the electrode and z_0 is the distance between the flat electrodes. The potential formed in the trap region is the sum of equations (2.1) and (2.2). The electric field in the trap region is therefore given by

$$\begin{aligned} \mathbf{E}(x, y, z, t) &= -\nabla(V(x, y, z, t) + U(x, y, z, t)) \\ &= -V_0 \left(\frac{x\mathbf{e}_x - y\mathbf{e}_y}{r_0^2} \right) \cos \Omega t - \frac{\kappa U_0}{z_0^2} (-x\mathbf{e}_x - y\mathbf{e}_y + 2z\mathbf{e}_z) \end{aligned} \quad (2.3)$$

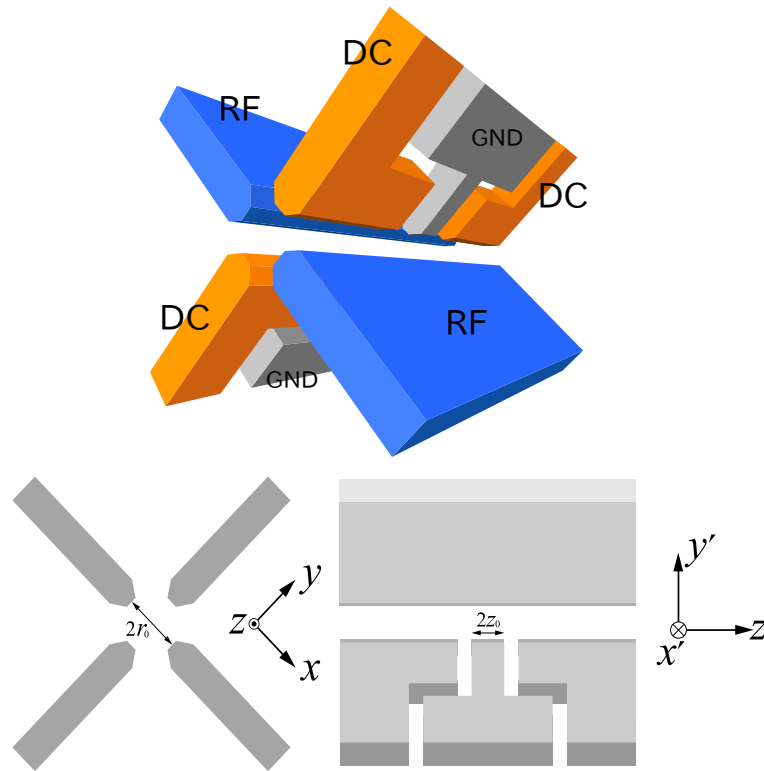


Figure 2.2: linear RF trap

Where $\mathbf{e}_x$, $\mathbf{e}_y$ and $\mathbf{e}_z$ are unit vectors in the x, y and z directions respectively. We consider the motion of the ions in the trap region. The equation of motion for a monovalent cation is given by

$$e\mathbf{E} = m \frac{d^2 \mathbf{u}}{dt^2} \quad (2.4)$$

Where $\mathbf{u}$ is the position of the ion. If we separate this equation into its x, y and z components, we get

$$\frac{d^2 u_i}{dt^2} + (a_i + 2q_i \cos \Omega t) \frac{\Omega^2}{4} u_i = 0 \quad (i = x, y, z). \quad (2.5)$$

Where

$$a_x = a_y = \frac{1}{2} a_z = -\frac{4e\kappa U_0}{mz_0^2 \Omega^2}, \quad (2.6)$$

$$q_x = -q_y = \frac{2eV_0}{mr_0^2 \Omega^2}, \quad q_z = 0 \quad (2.7)$$

The x and y components of equation (2.5) are known as the Mathieu equations. For stable ion trapping, the solutions of this differential equation must be such that a_i and q_i do not diverge. This gives the conditions for the RF voltage and DC voltage applied to the electrodes. The solution of equation (2.5) under the approximation $|a_i| \ll 1, |q_i| \ll 1$ ($i = x, y$) is

$$u_i(t) = u_{0i} \cos(\omega_i t) \left(1 + \frac{q_i}{2} \cos \Omega t \right). \quad (2.8)$$

Where

$$\omega_i = \frac{\Omega}{2} \sqrt{a_i + \frac{q_i^2}{2}}. \quad (2.9)$$

The oscillation of the frequency ω_i and Ω is referred to as secular motion and micromotion, respectively. Secular motion is due to the time average of the force exerted by the RF electric field.

2.3 laser cooling

When ions are loaded in a trap, they are at a high temperature. Furthermore the RF electric field applied to them heats them up. In order to keep the ions confined in the trapping region for a long time, it is necessary to cool them. In this study, we used Doppler cooling, a type of laser cooling, to cool the ions. Doppler cooling is a cooling method that makes use of the Doppler effect, which causes the frequency of the laser to change as seen from the ion. Consider the situation where an ion of velocity v and mass m interacts with a laser photon of frequency ω and wavelength k . If the direction of ion motion is opposite to the direction of laser irradiation, the frequency of the laser as seen by the ion is given by

$$\omega_b = \omega(1 + v/c) \quad (2.10)$$

It is assumed that v is sufficiently small compared to the speed of light, c . If the ion absorbs a photon in this case, the momentum of the ion is given by

$$mv' = mv - \hbar k \quad (2.11)$$

The ion is slowed down due to the law of conservation of momentum. Similarly, the frequency of the laser as seen by the ion, if the direction of the ion's motion and the direction of the laser beam are the same, is given by

$$\omega_f = \omega(1 - v/c) \quad (2.12)$$

If the ion absorbs a photon, the momentum of the ion is given by

$$mv' = mv + \hbar k \quad (2.13)$$

and the ion is accelerated. The ion that has absorbed a photon is excited to a level and returns to the ground state by spontaneous emission. The direction of the photon emitted is random, so the average speed of the ion does not change. If the frequency of the laser ω is detuned to a lower frequency than the absorption line of the ion, ω_f will be closer to the peak of the absorption line of the ion than ω_b . Therefore, the probability of interaction will increase if the direction of the laser beam is opposite to the ion's motion, and the ion

will be slowed down. To cool the ions to the lowest temperature, it is known that it is necessary to detune the cooling laser frequency by $\gamma/2$, where γ is the natural width. The limit of the Doppler cooling temperature is given by

$$T = \frac{\hbar\gamma}{2k_{\text{B}}} \quad (2.14)$$

We consider the laser cooling of the linear RF trap used in this study. Looking at Eq. (2.3), we can see that the electric field, which changes with time at a frequency Ω , becomes zero at $x = y = 0$. Therefore, on the trap axis, the RF electric field leads no force acting on it, and it can be cooled to low temperatures without being heated. Therefore, by using laser cooling in a linear RF trap, it is possible to line up several ions in a straight line and cool them to low temperatures.

2.4 Laser linewidth reduction

In this study, we first prepare a high-finesse optical cavity with a narrow-linewidth laser controlled at high speed, and use another laser with a narrowed linewidth as a clock laser by phase-locking it to the laser. In this section, we describe the FM sideband technique for linewidth narrowing the laser and the phase-locking technique for transferring the laser linewidth to another laser.

2.4.1 FM sideband technique

Since the natural width of the Ba⁺ ion clock transition is very narrow, at several mHz, a narrow-linewidth probe laser is required for the spectroscopic analysis of the clock transition. To obtain a narrow-linewidth laser, the frequency of the laser is controlled to resonate with the high-finesse optical cavity. However, the resonance signal of the optical cavity is symmetric with respect to the resonance point, so it cannot be used as an error signal as it is. Therefore, in this study we use the FM sideband technique to detect a signal that is antisymmetric with respect to the resonance point and to narrow the linewidth. In addition, the FM sideband technique uses the reflected light from the optical resonator. This has two advantages. One is that the response of the reflected light is faster than that of the transmitted light. The other is that it can be controlled without being affected by the intensity

fluctuations of the laser.

Resonance signal of optical resonator

The resonance signal of a Fabry-Perot resonator can be analysed as follows: when light with a complex amplitude of $E_{\text{in}} = E_0 e^{i\omega t}$ is incident on this optical resonator, if the complex amplitude of the reflected light from the optical resonator is E_{ref} , the amplitude reflectance of the electric field $r_{\text{FP}}(\omega)$ is given by

$$r_{\text{FP}}(\omega) = \frac{E_{\text{ref}}}{E_{\text{in}}} = -r_1 + t_1^2 r_2 e^{i\theta} \sum_{n=1}^{\infty} (r_1 r_2 e^{i\theta})^{n-1} \quad (2.15)$$

$$= -r_1 + \frac{t_1^2 r_2 e^{i\theta}}{1 - r_1 r_2 e^{i\theta}} \quad (2.16)$$

Here, r_i and t_i ($i = 1, 2$) represent the reflectance and transmittance of the reflectors at each end, respectively. Furthermore, if the free spectral range (FSR) of the optical resonator is Δf_{FSR} , the resonator length is L and the speed of light is c , then the formula is

$$\theta = \frac{2L\omega}{c} = \frac{\omega}{\Delta f_{\text{FSR}}} \quad (2.17)$$

In this study, it is assumed that the mirrors at both ends of the optical resonator are identical, i.e. $t_1 = t_2 = t$ and $r_1 = r_2 = r$, therefore we obtain

$$r_{\text{FSR}}(\omega) = \frac{-r(1 - e^{i\theta})}{1 - r^2 e^{i\theta}} \quad (2.18)$$

However, the energy conservation law $t^2 + r^2 = 1$ is employed. The frequency characteristics of the reflectance of the optical cavity, $|r_{\text{FP}}(\omega)|^2$, $\arg(r_{\text{FP}}(\omega))$, are demonstrated in Figures 2.3 and 2.4. In this study, the values of the optical cavity are substituted for each parameter: $r = 0.9999$, $\Delta f_{\text{FSR}} = 1.5$ GHz. A close analysis of these figures reveals that $|r_{\text{FP}}(\omega)|^2$ is symmetrical about the resonance point, thereby indicating that it is not possible to determine the direction of frequency shift based solely on the information regarding the optical intensity of the reflected light. However, given the antisymmetrical nature of $\arg(r_{\text{FP}})$ about the resonance point, it becomes possible to ascertain

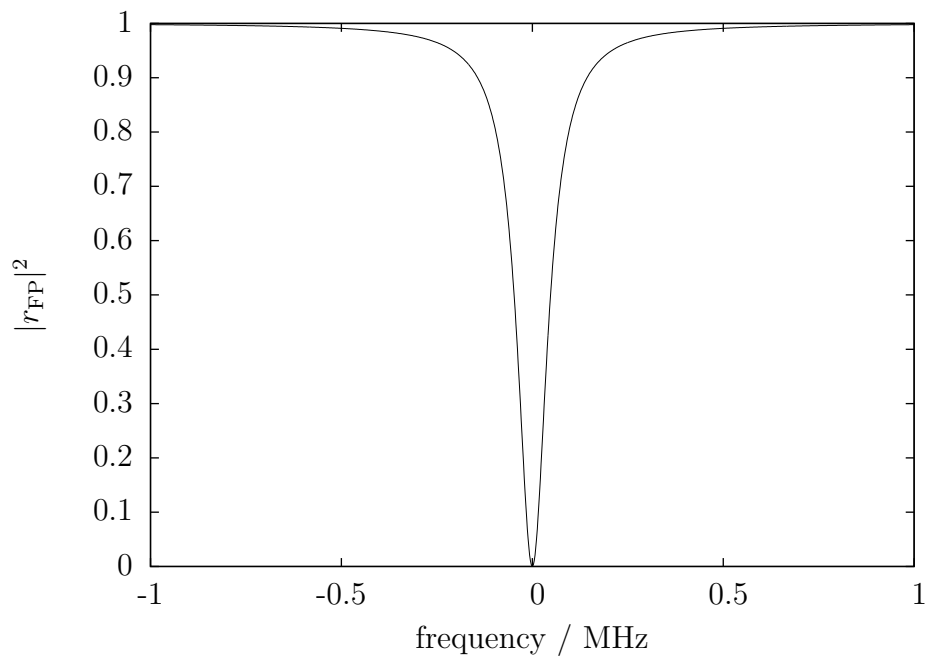


Figure 2.3: Frequency characteristics of the magnitude of the reflectance of a cavity

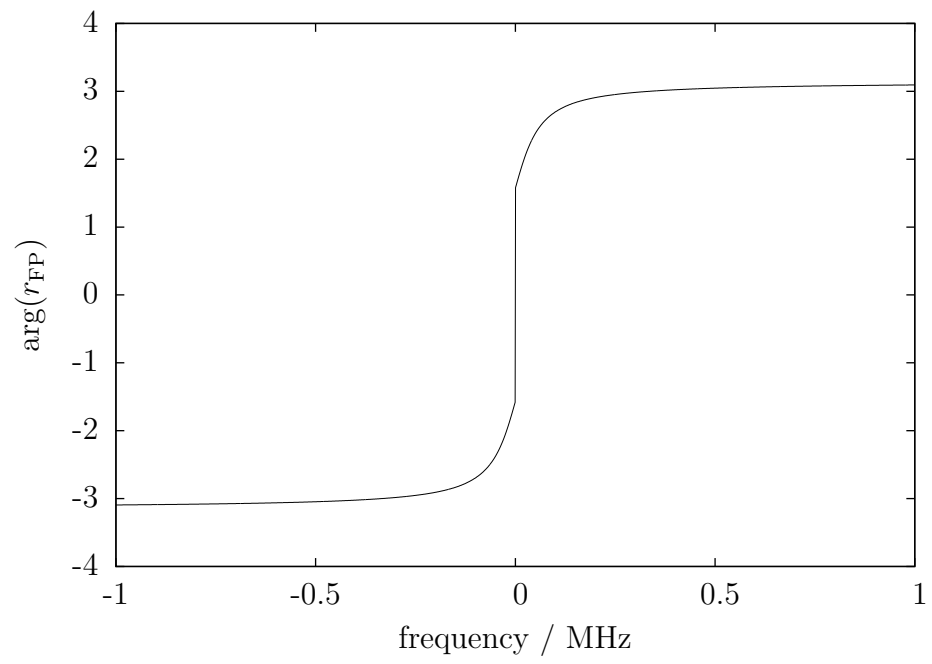


Figure 2.4: Frequency characteristics of the angle of the reflectance of a cavity

the direction of frequency shift, provided that the phase of the reflected light is taken into consideration.

Phase modulation

Assuming the complex amplitude of the electric field of the laser to be $E = E_0 e^{i\omega t}$, it can be demonstrated that if this laser is phase modulated with a modulation frequency Ω and a modulation index β , the electric field becomes

$$E_m = E_0 e^{i(\omega t + \beta \sin \Omega t)} \quad (2.19)$$

If this is expanded using the first kind Bessel function, it becomes

$$E_m = E_0 e^{i\omega t} \sum_{n=-\infty}^{\infty} J_n(\beta) e^{in\Omega t} \quad (2.20)$$

$$\simeq E_0 (J_0 e^{i\omega t} + J_1 e^{i(\omega+\Omega)t} - J_1 e^{i(\omega-\Omega)t}) \quad (2.21)$$

Here, we assume $\beta \ll 1$ and ignore the higher harmonics with $|n| \geq 2$. Assuming the vacuum impedance is Z_0 , the light intensity of the laser is given by $P_0 = |E_0|^2 / (2Z_0)$. Therefore, the light intensity of the carrier is $P_c = J_0^2 P_0$, and the light intensity of the sideband is $P_s = J_1^2 P_0$, as shown in

$$P_0 \simeq P_c + 2P_b \quad (2.22)$$

Subsequently, the consideration will be given to the case of the input of this modulated light into a Fabry-Perot resonator. The substitution of $E_{\text{ref}} = r_{\text{FP}} E_m$ into equation (2.21) and subsequent rearrangement will result in

$$E_{\text{ref}} = E_0 (r_{\text{FP}}(\omega) J_0 e^{i\omega t} + r_{\text{FP}}(\omega + \Omega) J_1 e^{i(\omega+\Omega)t} - r_{\text{FP}}(\omega - \Omega) J_1 e^{i(\omega-\Omega)t}) \quad (2.23)$$

The intensity of the reflected light $P_{\text{ref}} = |E_{\text{ref}}|^2 / (2Z_0)$ can be calculated from this, resulting in

$$P_{\text{ref}} = P_c |r_{\text{FP}}(\omega)|^2 + P_s (|r_{\text{FP}}(\omega + \Omega)|^2 + |r_{\text{FP}}(\omega - \Omega)|^2) + 2\sqrt{P_c P_s} (\text{Re}[R] \cos \Omega t + \text{Im}[R] \sin \Omega t) - P_s \{ r_{\text{FP}}(\omega + \Omega) r^*(\omega + \Omega) e^{i2\Omega t} + \text{c.c.} \} \quad (2.24)$$

However, this will be set as

$$R := r_{\text{FP}}(\omega)r_{\text{FP}}^*(\omega + \Omega) - r_{\text{FP}}^*(\omega)r_{\text{FP}}(\omega - \Omega) \quad (2.25)$$

In the FM sideband technique, the response to the resonance of the laser optical cavity is obtained by demodulating P_{ref} to extract R . Since R is contained in the coefficients of $\sin \Omega t$ and $\cos \Omega t$, it can be obtained by heterodyne detection at angular frequency Ω and then removing components above angular frequency Ω using a low-pass filter. The demodulation of the $\cos \Omega t$ coefficient is referred to as quadrature-phase demodulation, while the demodulation of the $\sin \Omega t$ coefficient is termed in-phase demodulation.

Error signal

The demodulated signal varies significantly with modulation frequency; consequently, an appropriate modulation frequency exists for utilisation of the demodulated signal as an error signal for control. Initially, the scenario will be examined in which the linewidth (full width at half maximum) of the resonator resonance is designated as $\Delta f_{1/2}$, and there exists a relationship of $\Omega \ll \Delta f_{1/2}$ with the modulation frequency. In this instance, Ω is considered to be sufficiently small, thus resulting in

$$\begin{aligned} R &\approx r_{\text{FP}} \left(r_{\text{FP}}^*(\omega) + \frac{dr_{\text{FP}}^*(\omega)}{d\omega} \Omega \right) - r_{\text{FP}}^*(\omega) \left(r_{\text{FP}}(\omega) - \frac{dr_{\text{FP}}(\omega)}{d\omega} \Omega \right) \\ &= \text{Re} \left[r_{\text{FP}}(\omega) \frac{d|r_{\text{FP}}(\omega)^*|}{d\omega} \right] \Omega = \frac{d|r_{\text{FP}}(\omega)|^2}{d\omega} \Omega \end{aligned} \quad (2.26)$$

Since this value is a real number, the demodulated signal in-phase is zero, and the demodulated signal in quadrature-phase is given by

$$e = 2\sqrt{P_c P_s} \frac{d|r_{\text{FP}}(\omega)|^2}{d\omega} \Omega \quad (2.27)$$

Subsequently, let us consider the case where $\Omega/2\pi \gg \Delta f_{1/2}$, i.e. where the optical resonator is resonating and Ω is near the resonance frequency. Given that the modulation frequency is sufficiently large, it can be assumed that the $\omega \pm \Omega$ component does not resonate. In other words, $r_{\text{FP}}(\omega \pm \Omega) = -1$.

Therefore, we have

$$R \simeq -i2 \operatorname{Im}[r_{\text{FP}}(\omega)]. \quad (2.28)$$

This is a pure imaginary number, resulting in a demodulated signal in quadrature-phase that is zero. Conversely, the demodulated signal in in-phase is given by

$$e = -4\sqrt{P_c P_s} \operatorname{Im}[r_{\text{FP}}(\omega)]. \quad (2.29)$$

The demodulated signal for $\Omega/2\pi \ll \Delta f_{1/2}$ is illustrated in Fig.2.5, and for $\Omega/2\pi \gg \Delta f_{1/2}$, the signal is shown in Fig.2.6. The parameters for these figures are identical to those in Fig. 2.3. The red line indicates quadrature-phase demodulation, while the blue line signifies in-phase demodulation. A comparison of these figures reveals that quadrature-phase demodulation is preferable when the modulation frequency is less than half the optical resonator's half-width, and in-phase demodulation is more suitable when the modulation frequency exceeds this value. A subsequent comparison of the demodulated signal in quadrature-phase in Figure 2.5 with the demodulated signal in in-phase in Figure 2.6. reveals that the former decays more rapidly than the latter when detuned from the optical resonator's resonance frequency. This observation indicates that the latter possesses a wider range of controllability in comparison to the former. In summary, it can be concluded that the demodulated signal in-phase with the modulation frequency Ω , which is larger than the half-width of the optical resonator, is more appropriate for use as the frequency control error signal.

2.4.2 Phase locked loop

The phase-locked loop (PLL) is a method of synchronising a target signal with a reference signal by extracting the phase difference between the reference signal and the target signal as an error signal. The electric fields $E_m(t)$ of the laser that serves as the reference signal (master laser) and $E_s(t)$ of the laser that functions as the target signal (slave laser) are expressed by

$$E_m(t) = E_{m0} \exp \{i(\omega_m t + \theta_m)\} + \text{c.c.} \quad (2.30)$$

$$E_s(t) = E_{s0} \exp \{i(\omega_s t + \theta_s)\} + \text{c.c.} \quad (2.31)$$

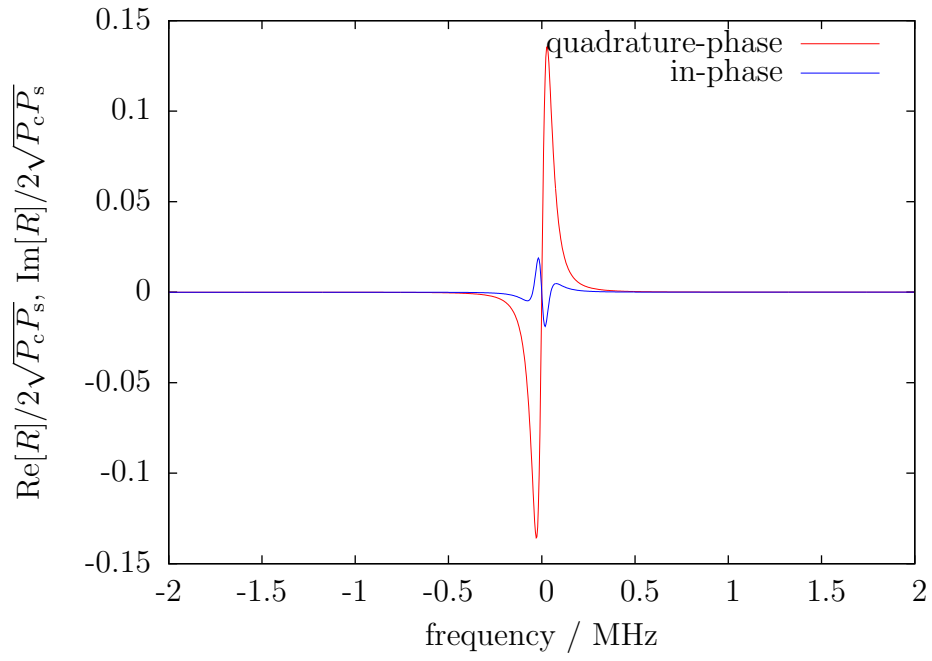
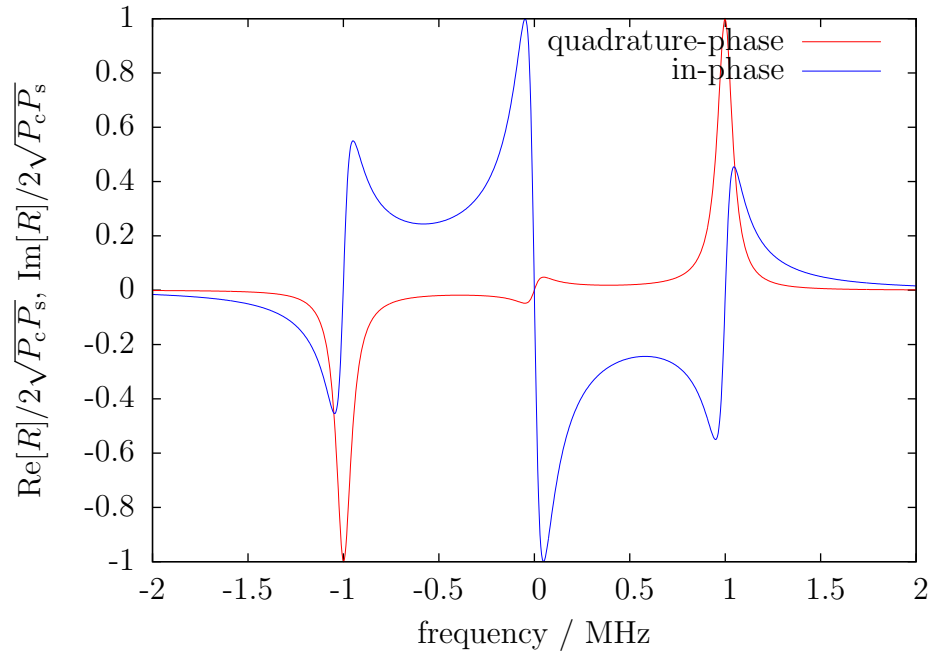


Figure 2.5: demodulated signal in condition of $\Omega/2\pi \ll \Delta f_{1/2}$

Figure 2.6: demodulated signal in condition of $\Omega/2\pi \gg \Delta f_{1/2}$

However, it is important to note that $\omega_m > \omega_s$. These lasers are then superimposed and incident on a photodiode (PD). The signal $v_{PD}(t)$ detected by the PD is proportional to the optical intensity $I(t)$, and is expressed by

$$\begin{aligned}
 I(t) &= \frac{(E_m(t) + E_s(t))^2}{Z_0} \\
 &= \frac{E_{m0}^2}{Z_0} + \frac{E_{s0}^2}{Z_0} + \left[\frac{E_{m0}^2}{Z_0} \exp\{i(2\omega_m t + 2\theta_m)\} + \frac{E_{s0}^2}{Z_0} \exp\{i(2\omega_s t + 2\theta_s)\} + \right. \\
 &\quad \left. \frac{E_{m0}E_{s0}}{Z_0} \exp\{i((\omega_m - \omega_s)t + \theta_m - \theta_s)\} + \frac{E_{m0}E_{s0}}{Z_0} \exp\{i((\omega_m + \omega_s)t + \theta_m + \theta_s)\} \right] + \text{c.c.}
 \end{aligned} \tag{2.32}$$

However, given the PD's insensitivity to the optical frequency, the observed signal is, in fact, expressed by

$$v_{PD}(t) = \frac{\alpha}{Z_0} [E_{m0}^2 + E_{s0}^2 E_m E_s \exp\{i((\omega_m - \omega_s)t + \theta_m - \theta_s)\}] \tag{2.34}$$

The coefficient α , which expresses the sensitivity of the PD, is therefore of significance. Should the DC component of $v_{PD}(t)$ be filtered out, the beat signal $v_{beat}(t) = (\alpha E_m E_s / Z_0) \exp\{i((\omega_m - \omega_s)t + \theta_m - \theta_s)\} + \text{c.c.}$ can be obtained, which contains information about the frequency difference $\omega_m - \omega_s$ and phase difference $\theta_m - \theta_s$ between the two lasers. Subsequently, by introducing the high-frequency voltage $v_1(t) = V_{l0} \exp\{i(\omega_1 t + \theta_1)\} + \text{c.c.}$, which possesses an angular frequency ω_1 , and $v_{beat}(t)$ into a multiplier (mixer) and performing a multiplication, the output will be

$$\begin{aligned}
 v_{mixer}(t) &= \frac{\alpha k E_m E_s}{Z_0} [\exp\{i((\omega_m - \omega_s - \omega_1)t + \theta_m - \theta_s - \theta_1)\} \\
 &\quad + \exp\{i((\omega_m - \omega_s + \omega_1)t + \theta_m - \theta_s + \theta_1)\}] + \text{c.c.}
 \end{aligned} \tag{2.35}$$

The conversion coefficient of the multiplier, denoted by k , is thus determined. Subsequent extraction of the previous term, facilitated by a low-pass filter, and utilisation of an oscillator with $\Omega = \omega_m - \omega_s$, results in the derivation of

$$v_e = \frac{\alpha k E_m E_s}{Z_0} \exp\{i(\theta_m - \theta_s - \theta_1)\} + \text{c.c.} \tag{2.36}$$

When setting $\Delta\theta = \theta_m - \theta_s - \theta_l - \pi/2$, it can be seen that

$$v_e = \frac{\alpha k E_m E_s}{Z_0} \sin(\Delta\theta) \simeq \frac{\alpha k E_m E_s}{Z_0} \Delta\theta \quad (2.37)$$

is obtained when $\Delta\theta$ is small. This is an antisymmetric signal that contains information about the phase difference signal. Therefore, if this signal is used as an error signal for control, it is possible to control $\Delta\theta$ to 0 within the range of $-\pi/2 < \Delta\theta < \pi/2$. Since the differential of the phase is the frequency, the phase-locked state is an integral control when viewed in terms of frequency. In the event that the phase fluctuation of the oscillator is sufficiently small, the phase of the slave laser will match that of the master laser, thus resulting in

$$\omega_s = \omega_m - \omega_l \quad (2.38)$$

Furthermore, the phase fluctuation of the slave laser will be almost equal to that of the master laser.

Chapter 3

Clock laser for the $^2S_{1/2} - ^2D_{5/2}$ clock transition at $1.76 \mu\text{m}$

We show a schematic of the light source for driving the $^2S_{1/2} - ^2D_{5/2}$ clock transition in Fig. 3.4. We linewidth an ECDL at 881 nm to a resonance of a high-finesse cavity using FM sideband technique. [27] Then we phase lock an ECDL at $1.76 \mu\text{m}$ to the linewidth radiation at 881 nm with an offset frequency. To realize this, we generate the SH of the ECDL at $1.76 \mu\text{m}$ and phase lock the beat between the two radiations at 881 nm to a reference frequency from a RF synthesizer. The frequency of the phase locked ECDL at $1.76 \mu\text{m}$ is precisely scanned by changing the reference frequency when we conduct single-ion spectroscopy.

We compose an ECDL at 881 nm of a commercially-available anti-reflection (AR)-coated DL chip (Eagleyard Photonics, EYP-RWE-0940-08000-0750-SOT01-0000) and a diffraction grating of a pitch of 1800 /mm and a braze wavelength of 400 nm. The first-order diffraction of 4.8% of the incident power is fed back to the DL chip, while 68% to the zeroth order is available as an output. The extended-cavity length is 45 mm. We obtain the wavelength tuning between 863 nm and 945 nm, and the maximum output power of 43 mW at 925 nm at the operation current I_{op} of 90 mA. The threshold current I_{th} is minimized to be 37 mA at 925 nm. At the operation wavelength λ_{op} of 881 nm, the output power P_{op} at I_{op} of 90 mA and I_{th} are 25 mW and 62 mA, respectively. We achieve a continuous frequency tuning range to be 6 GHz.

The finesse and the free spectral range of the high-finesse cavity, made of ultra-low-thermal expansion (ULE) glass, are approximately 29000 and 1.5 GHz, respectively. The cavity is placed in a temperature-controlled vac-

uum chamber, although the temperature is not adjusted at that of the zero thermal expansion coefficient. The cavity is vertically hold with respect to the optical table to decrease the sensitivity of the cavity length to the vertical vibration. [28] The fast component of the frequency fluctuation is reduced by proportion-integration-derivative (PID) control of the injection current of the DL. The servo bandwidth is achieved to be 1 MHz. The slow component is controlled by the cavity length of the ECDL using a PZT via an integrator. The linewidth relative to the resonance of the high-finesse cavity is estimated from the power spectral density (PSD) of the error signal voltage. It is possible to infer the error signal from the reflected light signal(Fig. 3.1) from the optical cavity, and it is possible to calculate the PSD from this error signal. In addition, by looking at this reflected light signal, it is possible to know the appropriate servo gain. The error signal voltage is converted to the frequency deviation using the discriminator slope of the cavity resonance signal. Then, we calculate the mean-square value of the frequency fluctuation $\Delta\nu^2$ by integrating the PSD from 50 Hz to 1 MHz, and we estimate the linewidth δf to be 29 Hz using the relation that $\delta f = \pi\Delta\nu^2/B$ as we approximate the frequency fluctuation to be a white noise, where B is the measurement bandwidth. [29]

We evaluate the absolute linewidth of the linenarrowed laser by comparing with another ECDL linenarrowed to a resonance of an independent high-finesse cavity. The wavelength of the compered ECDL is at 822 nm. To bridge the large frequency difference, we phase lock an OFC to the compared ECDL [30]. We measure the FWHM of the spectrum of the beat between the linenarrowed ECDL at 881 nm and a mode of the phase locked OFC to be 200 Hz.

The ECDL at 1.76 μm is composed of a commercially-avairable AR-coated DL chip (TOPTICA, LD-1700-0030-AR-1) and a diffraction grating of a pitch of 900 /mm and a blaze wavelength of 1200 nm. The first-order diffraction of 2% of the incident power is fed back to the DL chip, while 87% to the zeroth order is available. The cavity length of the ECDL is 40 mm. The maximum P_{op} is 22 mW at 1733 nm at I_{op} of 180 mA with I_{th} of 55 mA, while the oscillation wavelength range is between 1630 nm and 1778 nm. At λ_{op} of 1762 nm, P_{op} and I_{th} are 20 mW and 74 mA, respectively. A continuous frequency tuning range is 3 GHz.

We generate SH of the ECDL at 1.76 μm using a 10-mm-long periodically-poled lithium niobate (PPLN) crystal. We achieve a quasi phase-matching (QPM) at the temperature of 170 °C. The power of 170 nW is generated in SH from the fundamental power of 18.5 mW. We detect the beat between the

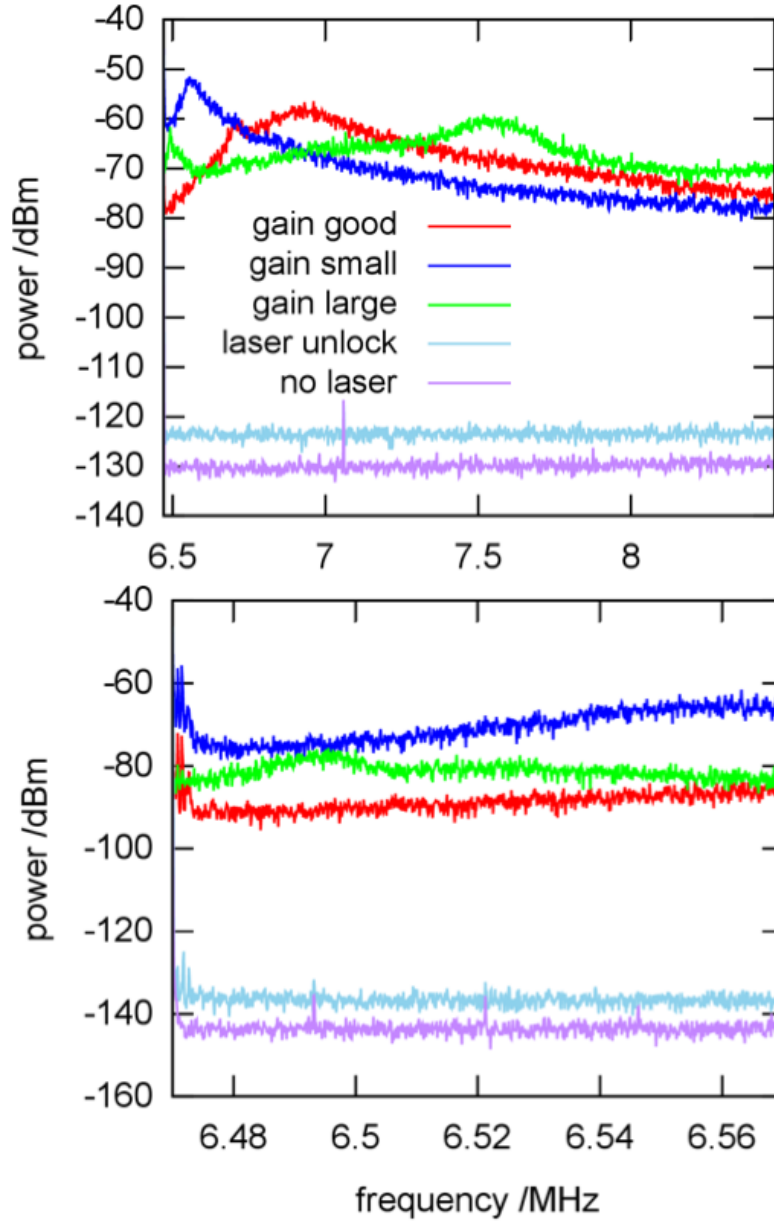


Figure 3.1: Frequency spectrum of reflected light from the optical cavity

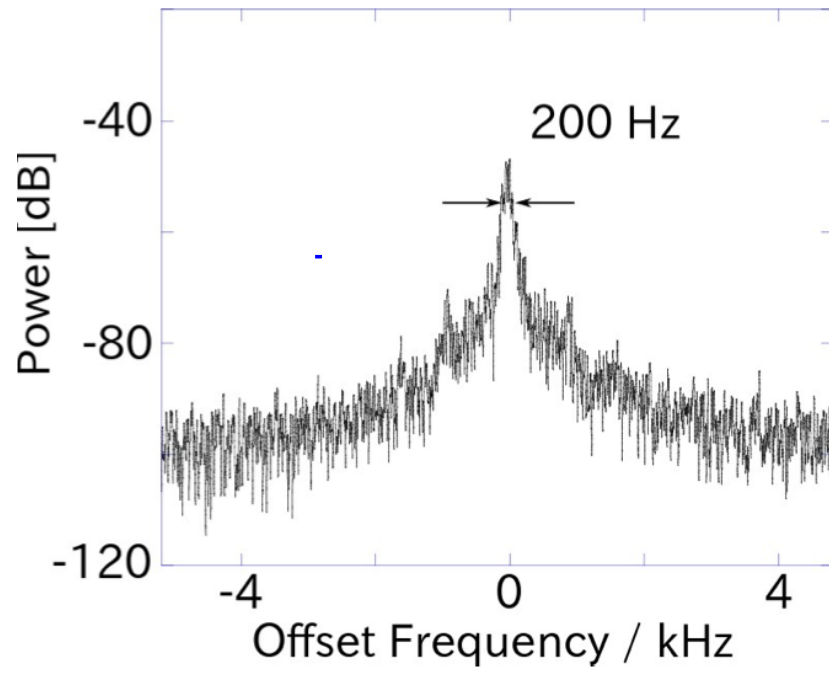


Figure 3.2: beat between 881-nm ECDL and OFC [30]

SH of the ECDL at $1.76\,\mu\text{m}$ and the linewidth-narrowed ECDL at $881\,\text{nm}$ with a signal-to-noise ratio (SNR) of 35 dB with a resolution bandwidth of 300 kHz. The FWHM of the beat spectrum is 450 kHz, which corresponds to the twice as wide as the FWHM linewidth of the free running ECDL at $1.76\,\mu\text{m}$. The phase deviation from the reference at $\sim -350\,\text{MHz}$ from the RF synthesizer is detected using a double-balanced mixer (DBM). The RF synthesizer is referenced to a GPS clock. The phase locking is performed by controlling the injection current of the DL chip at $1.76\,\mu\text{m}$ through a PID controller. The servo bandwidth is achieved to be 1.2 MHz. The locking is maintained in long term by controlling the extended-cavity length using a PZT through an integrator. Figure 3.3 shows the clock laser beat between $1.76\,\mu\text{m}$ SH and $881\,\text{nm}$ laser.

We evaluate the residual phase noise to be 0.79 rad using a signal analyzer (Anritsu 2840A-040 with option 010, 066), by integrating the PSD of the phase noise in the in-loop beat from 10 Hz to 10 MHz. Because the linewidth of the linewidth-narrowed ECDL at $881\,\text{nm}$ is successfully transferred, we estimate the linewidth of the ECDL at $1.76\,\mu\text{m}$ to be narrower than 100 Hz.

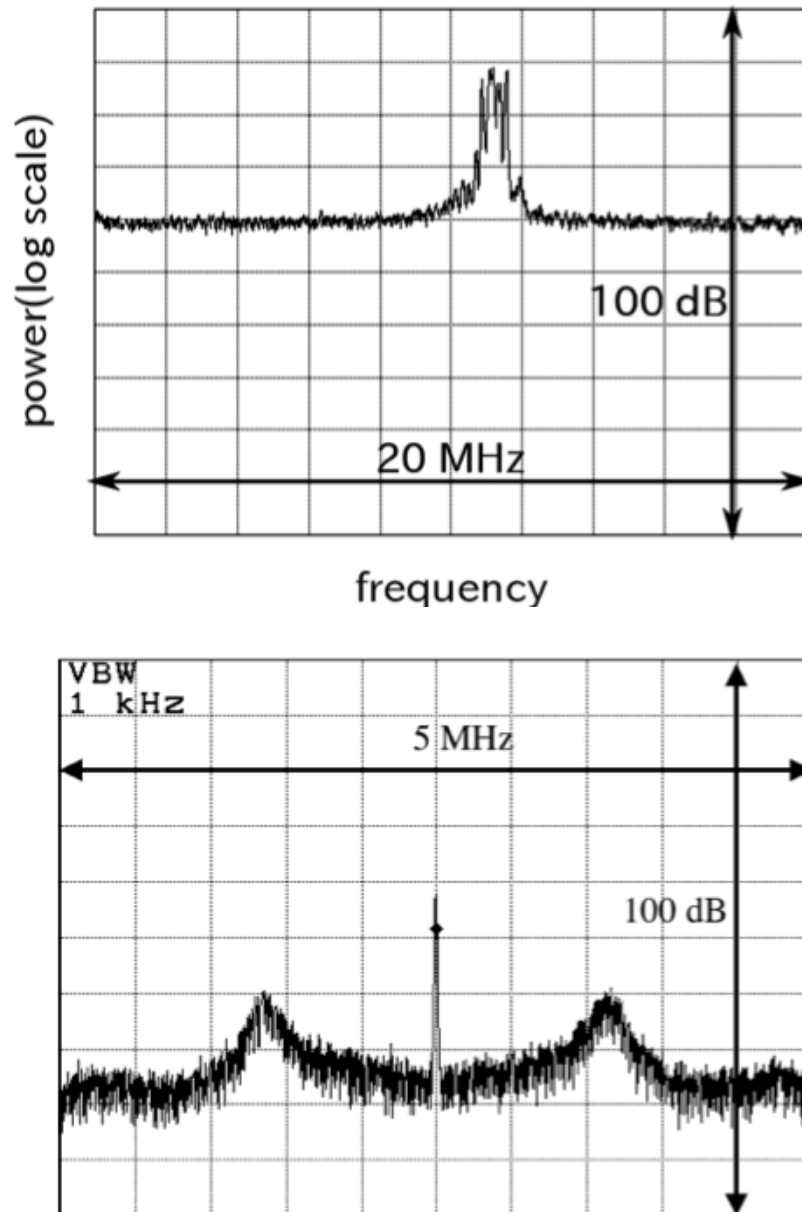


Figure 3.3: Beat between $1.76\ \mu\text{m}$ SH and 881 nm laser. The upper figure is when each laser is free-running. The lower figure is when they are phase-locked.

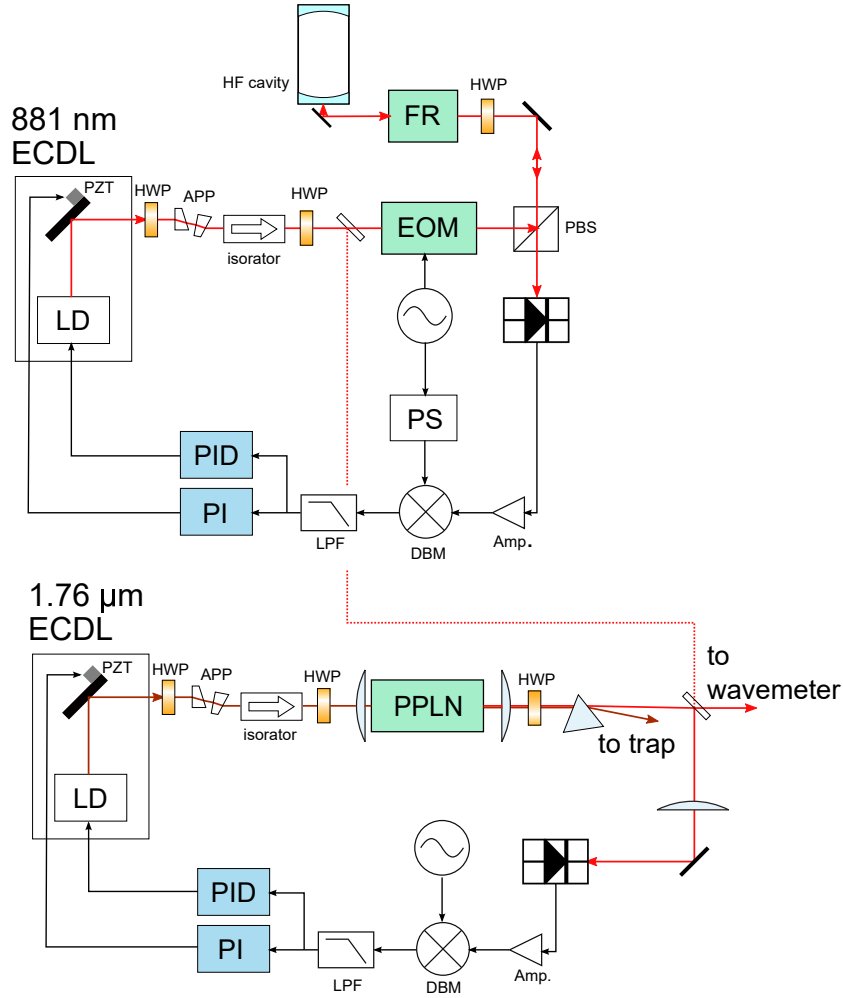


Figure 3.4: Schematic diagram of clock laser. DL, diode laser; ECDL, extended-cavity diode laser; PPLN, periodically-poled lithium niobate crystal; EOM, electrooptic modulator; PS, phase shifter; PZT, piezo-electric transducer; HWP, half-wave plate; APP, anamorphic-prism pair, PBS, polarizing beam splitter; FR, 45° Faraday rotator; HF cavity, high-finesse cavity; DBM, double-balanced mixer; AMP, amplifier; LPF, low-pass filter; PID, proportion-integration-derivative controller; PI, proportion-integration controller

Chapter 4

Clock laser using difference frequency generation and optical frequency comb

As an alternative approach, we adopt difference frequency generation (DFG) to obtain coherent radiation at 2052 nm. To realize a narrow-line clock laser, fast feedback control to a resonance of a high-finesse cavity as shown in Chapter 3 is performed. When a laser is not available at the wavelength of the clock transition, nonlinear frequency conversion such as second-harmonic generation and sum or difference frequency generation is performed. When a sufficient power is generated in the sum or difference frequency generation, the narrow-line radiation is realized by controlling only one of the two fundamental lasers, as we analyze the frequency fluctuation using the generated radiation. [25, 26] In many cases, however, the generated radiation does not have a sufficiently large power. Two fundamental lasers are required to be independently line-narrowed before the generation. It is elaborating to develop two systems involved high-finesse cavities because considerable efforts are required to isolate the high-finesse cavity from vibrational and acoustic fluctuations.

4.1 Experimental setup

To generate the radiation at 1762 nm, we perform DFG of radiation from a commercially-available Nd:YAG laser at 1064 nm and from a home-made ECDL at 664 nm using 45-mm-long 5mol%-magnesium-oxide-doped periodically-poled lithium niobate (PPMgLN) crystal of the poled pitch of 13.66 μm . The

two lasers are phase locked to neighbor modes of the line-narrowed f_{CEO} -stabilized octave-spanning OFC based on a homemade mode-locked Ti:Sa laser [23]. This is the same OFC used for the linewidth evaluation of the master laser at 881 nm in the clock laser system in Chapter 3.

The ECDL is constructed using a commercially available diode laser (DL) (Mitsubishi ML120G21)). The diverging output beam from the DL is collimated using a collimation lens having a numerical aperture of 0.55. We place a diffraction grating having a pitch of 1800 /mm and a blaze wavelength of 250 nm in Littrow configuration. The extended cavity length is 30 mm. The first order diffraction of 10% from the incident power is fed back to the DL. The output power is available as a zeroth reflection of 77%. We measure the wavelength tuning between 654.5 nm and 662.5 nm at room temperature. To generate the difference frequency at 1762 nm, we increase the temperature of the DL to approximately 55 °C. We obtain the output power of 49 mW at the operating current of 150 mA and the continuous tuning range of 4 GHz.

The Nd:YAG laser has a maximum output power of 1 W and the linewidth of 1 kHz in the specification (Innolight M1000NE).

We briefly describe the performances of the line-narrowed OFC. The details are shown in [23]. The homemade mode-locked Ti:Sa laser generating the OFC is dispersion-controlled by a pair of prisms. The pulse repetition frequency is ~ 100 MHz. The output power, spectral width and the pulse duration are 400 mW, 40 nm and 23 fs, respectively. The output from the mode-locked laser is passed through a ~ 5 -cm-long photonic-crystal fiber to broaden the output spectrum over one octave. Then, we measure the f_{CEO} using an f - $2f$ interferometer. The SNR of the beat at f_{CEO} is 25 – 35 dB with a RBW of 300 kHz. We phase lock f_{CEO} to a reference frequency from a RF synthesizer by controlling the pump-laser power using an AOM. We simultaneously control the angle of the end mirror after the pair of prisms to maintain the phase locking of f_{CEO} in long term. Then, we phase lock the f_{CEO} -stabilized octave-spanning OFC to an ECDL line-narrowed to a resonance of a high-finesse cavity (master ECDL) by phase locking the beat between one of the modes of the OFC and the master ECDL. The wavelength of the master ECDL is 822 nm and the SNR of the beat is typically 30 dB with RBW of 300 kHz. The phase-locking is achieved by controlling the cavity length of the mode-locked laser using a fast piezo-actuated mirror with a control bandwidth of 80 MHz. The residual phase noise is evaluated to be 0.35 rad from the integration of the PSD of the phase noise between 1 Hz to 1 MHz. We maintain the full phase locking over 40 minutes.

The red component of the output beam from the photonic-crystal fiber

was separated using a dichroic mirror and overlapped with the beam of the ECDL at 664 nm using a half mirror. The overlapped two beams were dispersed by a diffraction grating and were focused to an avalanche-photodiode (Hamamatsu S2381) to detect the beat between the neighbor modes of the OFC and the ECDL. We detected the phase deviation in the beat to the reference frequency from a RF synthesizer using a DBM. Then, we phase locked the beat to the reference by controlling the injection current of the DL. To maintain the phase locking, the slow component of the phase deviation was feedback controlled by the extended cavity length using a PZT.

The method of the phase locking of the Nd:YAG laser was similar to that of the ECDL. We took the beat using the beam after the second-harmonic generation in the $f-2f$ interferometer in the system of the OFC. We used a pin photodiode (Hamamatsu S5973) to detect the beat, and another RF synthesizer for the reference in the detection of the phase deviation. We externally place an AOM in the beam pass of the Nd:YAG laser. We feedback controlled the AOM as used for a phase shifter for the phase locking.

We evaluated the linewidth of the difference frequency by taking the beat with a line-narrowed ECDL at 1762 nm described in Chapter 3. The linewidth of the ECDL at 1762 nm was estimated to be 100 Hz in Chapter 3.

The beam of the line-narrowed ECDL at 1762 nm was transferred from the different optical table using a polarization-maintaining optical fiber. The two beams at 1762 nm are overlapped using a half mirror and are focused on to a photodiode (Hamamatsu G12181) having a response bandwidth of 80 MHz.

4.2 Results

We measured that the difference frequency power was proportional to each of the two fundamental laser powers available in this study. The maximum difference frequency power of $51 \mu\text{W}$ was generated from the fundamental power of 17 mW and 460 mW at 664 nm and 1064 nm, respectively. The quasi-phase matching was achieved at $\sim 80^\circ\text{C}$ with a temperature tolerance of

The SNRs of the beats with the neighbor modes of the line-narrowed f_{CEO} -stabilized octave spanning OFC were slightly higher than 20 dB and slightly lower than 30 dB for the ECDL and the Nd:YAG laser, respectively, with a RBW of 300 kHz. The servo bandwidth was achieved to be approximately 1 MHz and 130 kHz for the injection current of the ECDL and the external

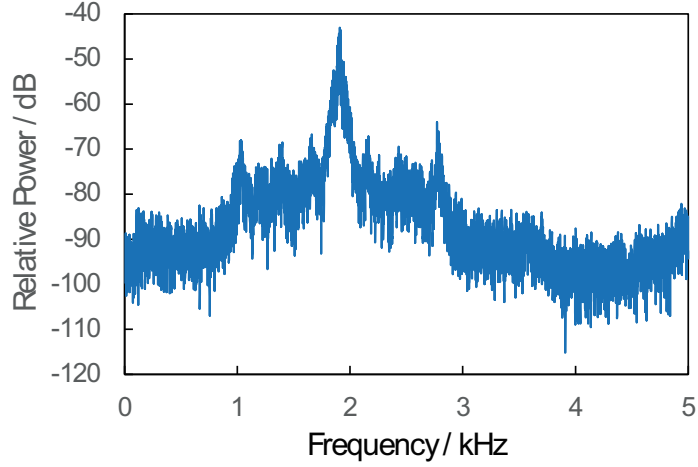


Figure 4.1: Beat spectrum between the line-narrowed difference frequency and the line-narrowed ECDL at 1762 nm. The beat frequency was 45.3 MHz. The span, resolution bandwidth, and video bandwidth are 5 kHz, 1 Hz, and 1 Hz, respectively.

AOM for the control of the Nd:YAG laser, respectively.

We evaluated the residual phase noise from the error signal voltage at the output of the DBM. We measured the PSD of the error signal voltage using a fast Fourier transform (FFT) analyzer. We converted it to the PSD of the phase noise using a discriminator slope of the phase deviation to the voltage generated at the DBM. We obtained the residual phase noise to be 0.9 rad and 0.2 rad for the ECDL and the Nd:YAG laser, respectively, from integrating the phase noise PSD between 1 Hz and 10 MHz.

We detected the beat between the line-narrowed difference frequency and the line-narrowed ECDL at 1762 nm with a SNR of 30 dB with a RBW of 300 kHz. Figure 4.1 shows the beat spectrum with a RBW of 1 Hz when we phase locked the two fundamental lasers used for the DFG to the line-narrowed OFC. The FWHM of the beat spectrum was narrower than 200 Hz. Because the estimated linewidth of the ECDL at 1762 nm was 100 Hz, we estimated the linewidth of the line-narrowed difference frequency to be 100 Hz.

We continuously phase locked the entire system during 15 minutes. This is limited by the duration of which the phase locking of the Nd:YAG laser maintained. This can be improved by additional control of the length of the laser cavity via a PZT and/or the temperature of the Nd:YAG crystal.

4.3 Conclusion

We have developed a system of the narrow-line difference frequency generation by phase locking of two fundamental lasers to a line-narrowed f_{CEO} -stabilized octave-spanning OFC. We evaluated the absolute linewidth of the line-narrowed difference frequency to be 100 Hz. In this system, all phase locking were successfully transferred the spectra and linewidths. We can improve the absolute linewidth by improving the linewidth of the maser laser used as a reference for phase locking of the OFC.

This scheme of generating narrow-line difference frequency radiation requires only one high-finesse cavity and widely known techniques. Narrow-line radiation can be realized in wide wavelength range by exchanging the ECDL to other wavelengths using an appropriate nonlinear crystal. To realize the clock laser for driving the $^2\text{S}_{1/2}$ - $^2\text{D}_{3/2}$ transition at 2052 nm, the wavelength of the ECDL is 701 nm.

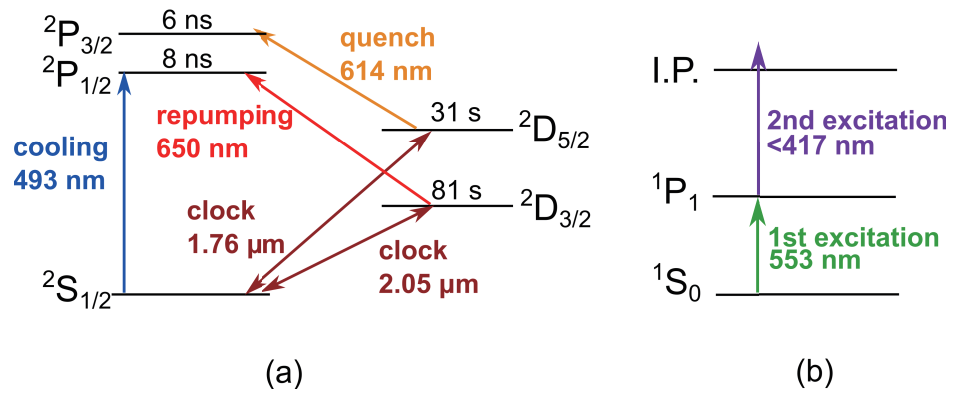
Chapter 5

Trapping and laser cooling of single $^{138}\text{Ba}^+$ in linear-RF trap

A partial-term scheme for Ba^+ related to this study is shown in Fig.5.1. The $^2\text{S}_{1/2} - ^2\text{P}_{1/2}$ transition is driven by laser cooling at 493 nm. In the $^2\text{P}_{1/2}$ state, Ba^+ decays to the $^2\text{D}_{3/2}$ metastable state with a branching ratio of approximately 1/3. We drive the $^2\text{D}_{3/2} - ^2\text{P}_{1/2}$ transition at 650 nm to close the cooling cycle as a repumping transition. We observe the fluorescence emitted by spontaneous emission from the $^2\text{P}_{1/2}$ to the $^2\text{S}_{1/2}$ state for the detection of single Ba^+ ions. We conducted single-ion spectroscopy of the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition at 1762 nm. The excitation to the $^2\text{D}_{5/2}$ state is detected through fluorescence disappearance by electron shelving in the $^2\text{D}_{5/2}$ state. To obtain the spectrum of the clock transition in a single ion, we determine the excitation probability by repeating the excitation of the clock transition. The lifetime of 31 s of the $^2\text{D}_{5/2}$ state in Ba^+ prevents the determination of the excitation probability within a short time. To repeat the excitation faster, we drive the $^2\text{D}_{5/2} - ^2\text{P}_{3/2}$ transition at 614 nm after state detection. In addition, to determine the number of trapped Ba^+ ions, we drive the $^2\text{S}_{1/2} - ^2\text{P}_{3/2}$ transition at 455 nm. We determine the number from the step-wise decrease and recovery in the fluorescence by the decay to the $^2\text{D}_{5/2}$ state and subsequent decay to the $^2\text{S}_{1/2}$ state, respectively.

Ba^+ ions are generated inside the trap by the two-step photoionization of Ba atoms. For this purpose, we drive the $^1\text{S}_0 - ^1\text{P}_1$ transition at 553 nm as the first excitation of the photoionization. [31] We photoionize Ba atoms in the $^1\text{P}_1$ state by irradiation with a wavelength shorter than 417 nm.

A schematic of the experimental setup is shown in Fig. 5.2. The details are as follows.

Figure 5.1: Partial term schemes for (a) Ba^+ and (b) Ba in this study.

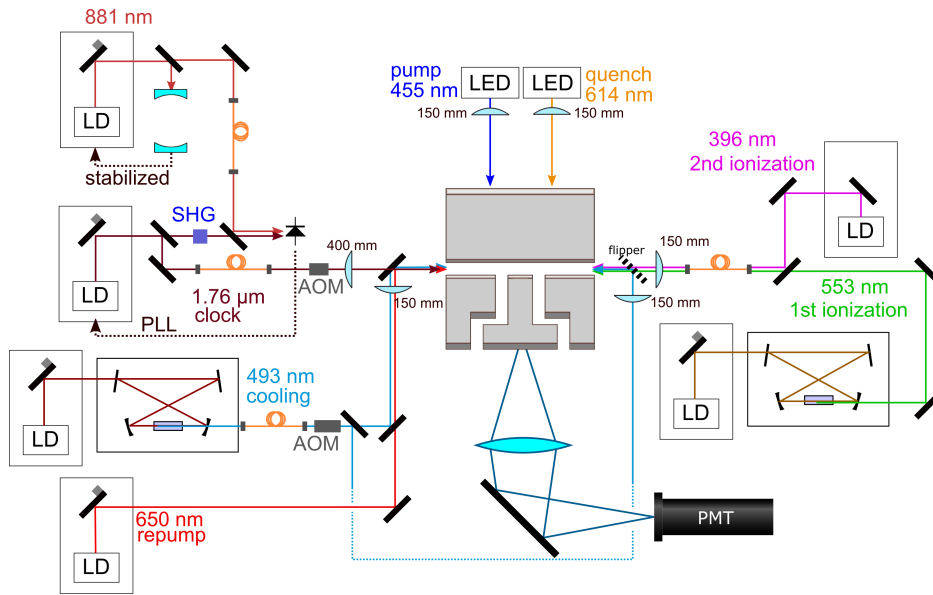


Figure 5.2: Schematic of clock laser. DL, diode laser; ECDL, extended cavity diode laser; PPLN, periodically poled lithium niobate crystal; EOM, electrooptic modulator; PS, phase shifter; PZT, piezo-electric transducer; HWP, half-wave plate; APP, anamorphic-prism pair, PBS, polarizing beam splitter; FR, 45° Faraday rotator; HF cavity, high-finesse cavity; DBM, double-balanced mixer; AMP, amplifier; LPF, low-pass filter; PID, proportion-integration-derivative controller; PI, proportion-integration controller

5.1 Light sources

We used DLs as we constructed the ECDLs to drive the transitions described above, except for three. For the second excitation of the photoionization, we used a DL as it is sufficient to excite Yb atoms above the ionization potential. We used light-emitting diodes (LEDs) to drive the $^2D_{5/2} - ^2P_{3/2}$ transition at 614 nm and the $^2S_{1/2} - ^2P_{3/2}$ transition at 455 nm. [32] The former is for simplicity and the latter is suitable for measuring the number of trapped $^{138}\text{Ba}^+$ ions.

We constructed the ECDLs as per MacAdam *et al.* [17] We collimated a diverging output beam from a DL using a collimation lense, and placed a diffraction grating in a Littrow configuration. We preferred the Littrow configuration over other Littmann configurations, because the output power is higher and an extremely large continuous frequency tuning range is not required. Adjusting the extended cavity length and diffraction efficiency of the grating is essential. As we increase the extended cavity length, the linewidth of the ECDL is narrower; however, the ECDL tends to oscillate in multiple modes or decrease the continuous frequency tuning range. During construction, we began with an extended cavity length of approximately 100 mm. We then shortened the cavity length until single-frequency oscillation and a continuous frequency-tuning range of more than 3 GHz were achieved. A solution of 3 GHz was derived from our experience for easy tuning to the transition. Considering the diffraction efficiency, a small efficiency in the first-order diffraction as feedback indicates that a large output power is available as the 0-th order reflection. However, an overly small feedback ratio narrows the oscillation wavelength range, preventing oscillations at the intended wavelength. We selected a grating to obtain a first-order diffraction efficiency of 10–20 %. To realize such small diffraction efficiency, the polarization of the DL beam was set parallel to the grating grooves and the blaze wavelength was shorter than the oscillation wavelength. When we increased the diffraction efficiency, the blaze wavelength was rendered closer and/or the polarization was perpendicular.

5.1.1 Cooling laser

Figure 5.3 shows a schematic of the cooling laser, that is, the radiation source used to drive the cooling transition at 493 nm. This was realized by SHG of radiation from an ECDL at 987 nm. SHG was performed using a KNbO_3 crystal with a length of 5 mm placed in an external cavity with a coupling

mirror of a reflectivity of 91%. The cavity resonance was stabilized to the frequency of the ECDL using the signal detected with the polarization technique. [33] Phase matching was achieved at 34 °C. We obtained a SH power of 3.1 mW from a fundamental power of 42 mW at the cavity input and the fundamental enhancement factor was determined to be 33. It should be noted that SH power has been gradually decreasing over years to 300 μ W.

The ECDL frequency was stabilized at the resonance of the transfer cavity. The transfer cavity was stabilized to the ECDL frequency at 822 nm, which was offset-locked to the frequency of the laser line-narrowed to a resonance of a temperature-stabilized high-finesse cavity made of ULE glass (master laser). The frequency drift of the master laser was smaller than 50 Hz/s, although we did not adjust the temperature of the high-finesse cavity to realize zero thermal expansion coefficient. We generated the offset frequency as the zero-crossing frequency of a RF interferometer [34] launched by the beat between the master laser and transfer laser at 822 nm. The RF interferometer comprised the followings. The beat signal was amplified, splitted to two arms by a power splitter. Then, the signals through the two arms were mixed using the DBM. In one of the arms, we inserted a 1-m-long coaxial cable to introduce a differential delay between the two arms, and a phase shifter (Mini Circuit ZXPHS-431-S+) to tune the zero-crossing frequency [35]. By tuning the zero-crossing frequency, we adjusted the detuning of the cooling laser through the offset frequency.

We measured the stability of the interferometer to be ± 30 kHz/h as we input a RF from the RF synthesizer referenced through the GPS clock. On offset locking the transfer laser, the in-loop stability of the offset frequency was measured to be ± 50 kHz/h and its root Allan variance was evaluated to be $< 1 \times 10^{-12}$ at an averaging time of 1000 s. The frequency stability, including that of the master laser, was sufficient for the cooling laser, because the natural linewidth of the cooling transition is 20.26(3) MHz. [36]. The tuning range of the zero-crossing frequency by the phase shifter was measured to be 180 MHz, that is, 360 MHz at 493 nm. This tuning range was covered for compensating the frequency drift of the cooling laser of approximately 1 MHz and for easily loading Ba^+ ions into the trap at the beginning of the experiments. In our experience, the cooling laser should be detuned by several tens MHz on the low-frequency side for easy loading. However, on using this system, we mixed a fixed RF from a RF synthesizer to the beat between the master and transfer lasers, because the offset frequency required for the laser cooling was unknown. Once the experimental parameters of laser cooling is established, the offset frequency could be fixed. The mixed

RF could be removed by selecting a phase shifter that operates within an appropriate frequency range.

5.1.2 Repumping laser

To drive the $^2D_{3/2} - ^2P_{1/2}$ repumping transition at 650 nm, we composed an ECDL. To frequency-stabilize the repumping ECDL, we constructed another ECDL at 650 nm, which is frequency-stabilized to a spectrum of a hyperfine transition in the R(25) line in the 6–5 band in iodine molecules detected using modulation transfer spectroscopy [37, 38]. Then, we phase locked the repumping laser to the iodine-stabilized laser with an offset frequency.

We performed the modulation transfer spectroscopy of the iodine molecule in a 100-mm-long glass cell using the latter ECDL. The two beams were counter propagated through the cell. One of the beams, known as the pump beam, was frequency modulated using an electro-optic modulator. We detected the signal at the modulation frequency generated in the other beam called the probe beam. The modulation frequency and the modulation index are 1.4 MHz and 6, respectively. The SNR of the spectra was 4 through a low-pass filter with a cutoff frequency of 50 Hz. We frequency-stabilized the ECDL to the hyperfine component at the second lowest frequency in the line. We controlled the extended cavity length using a PZT through an integrator. Then, we phase locked the repumping ECDL to the iodine-stabilized ECDL with an offset frequency of approximately -420 MHz. The phase fluctuation in the beat between the two ECDLs to the reference from a RF synthesizer at the offset frequency was detected using a DBM. Then, phase locking of the repumping ECDL was achieved by fast feedback control of the injection current, whereas slow component was controlled by the extended cavity length using a PZT to maintain phase locking. The servo bandwidth was achieved as 2.2 MHz. The residual phase noise integrated from 10 Hz to 10 MHz was 0.44 rad.

5.1.3 Photoionization lasers

Radiation at 553 nm for the first excitation of the photoionization of Ba atoms is realized by SHG of radiation from an ECDL at 1107 nm. We compose the ECDL using a commercially available AR-coated DL chip (Toptica LD-1120-0300-1) and a diffraction grating having a pitch of 1200/mm and a blaze wavelength of 500 nm. The first-order diffraction of 11 % of the incident power is fed back to the DL chip, while 85 % to the zeroth order is used for

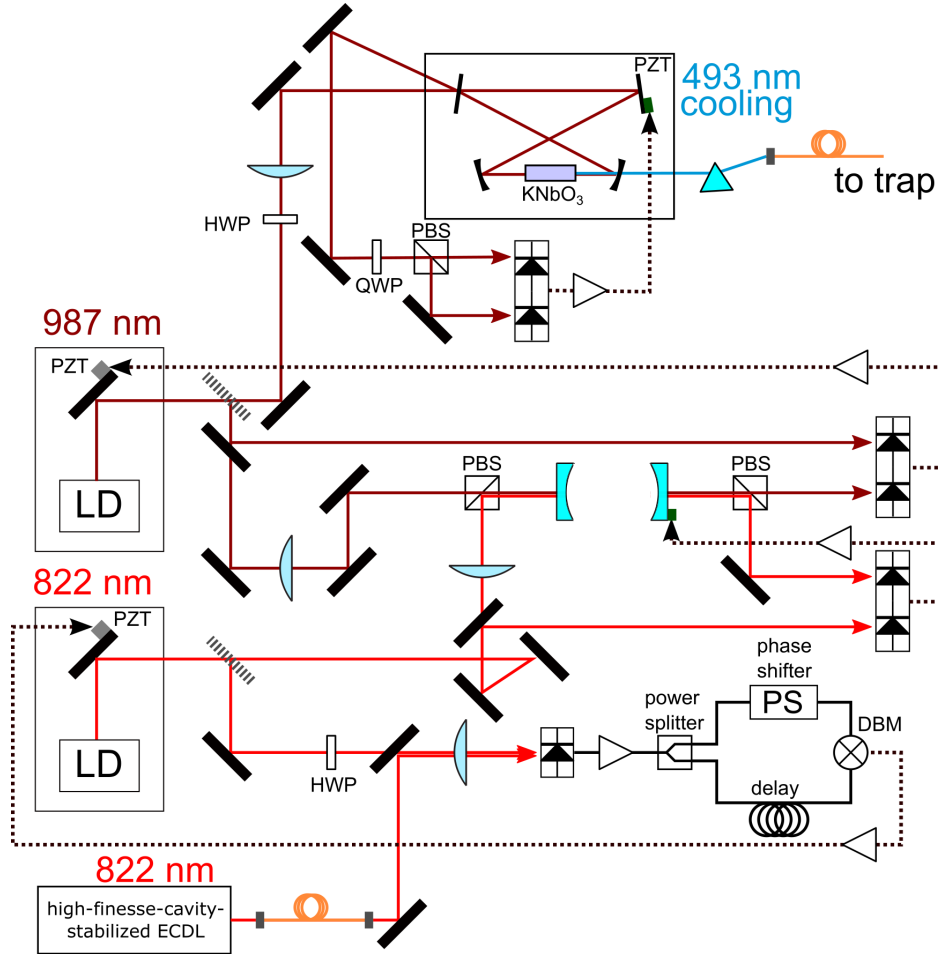


Figure 5.3: Schematic of the cooling laser. DL, diode laser; PZT, piezo-electric transducer; HWP, half-wave plate; QWP, quarter-wave plate; PBS, polarizing beam splitter; PS, phase shifter; DBM, double-balanced mixer.

SHG. The external cavity length is 100 mm. The single-frequency oscillation is realized between 1100 nm and 1125 nm. The threshold current is minimized to be 42 mA at 1116 nm, while it increases to be 59 mA in the region between 1100 and 1107 nm. We obtain the output power of 131 mW at the transition wavelength of 1107.404 nm with a operation current of 400 mA.

The second-harmonics is generated from a 10-mm-long PPLN crystal placed in a power build-up cavity. The quasi-phase matching is achieved at 122 degree C. The fundamental enhancement factor of 34 is realized using a coupling mirror of the reflectivity of 92 %. We obtain the second-harmonic power of 61 mW at 553 nm from a fundamental power of 97 mW. This is the region where the depletion of the fundamental power is significant. [39]

A part of the second-harmonic beam is introduced into a commercially available hollow-cathode lamp (Hamamatsu L2783-56NE-BA) to tune its frequency.

For the second excitation, we use a DL having a wavelength of 392 nm.

5.1.4 LEDs

We used two high-power LEDs as described above. One emitted radiation at approximately 455 nm (Luxeon, LXHL) and the other emitted radiation at approximately 614 nm (Cree LED, XRCRD0-L1-000-00M01). Each radiation source was passed through a band-pass filter and focused on the trap region using a lens. The DLs that oscillated at 614 nm were not commercially available during this research. Alternatively, the SHG of radiation from the DL at 1228 nm was used. [14]

5.2 Ion trap

Ba⁺ ions were confined in a linear RF trap. We adopted the linear RF trap as a future plan for multi-ion optical clocks. Herein, we describe the trap design and beam steering for cooling and single-ion spectroscopy. The Ba atoms, photoionized inside the trap, were generated from the wire of a getter (Saes Getters, ST14/FD Wire) by increasing its temperature as current passed through it. We selected the getter as the source of Ba atoms, because Ba metal is hard and has high reactivity in air, and its products after the reaction in air are toxic. The trap electrodes and getter wire were placed in a vacuum chamber and evacuated using an ion pump at an evacuation rate of 20 L/s. The part of the vacuum chamber surrounding the trap is made of five plates

of Pyrex glass. The fluorescence was collected by a combination of lenses placed perpendicular to the trap axis through one of the Pyrex plates, and was detected using a photomultiplier with a photon-counting method. We placed three pairs of coils close to the configuration of the Helmholtz coil to eliminate the environmental magnetic field and apply a magnetic field of appropriate direction and strength.

5.2.1 Linear-RF trap

Figure 5.4 shows a schematic of the linear RF trap. The voltages applied to the trap electrodes are shown in the figure. The linear RF trap comprises of four plates distributed at an equal angle of 90° in the radial direction. We applied a RF voltage required for the radial confinement to a pair of plates facing each other, and the other pair at the ground potential about the RF voltage. One of the ground plates was divided into three groups. DC voltages for axial confinement were applied to the two side plates, while the center plate was fixed to the ground potential.

To minimize the radial micromotion in two dimensions, a DC voltage was superimposed via a bias-T circuit onto one of the two RF electrodes, and the other DC voltage was applied to the undivided RF-ground electrode. To minimize the axial micromotion, the DC voltages applied to the two side plates for axial confinement were adjusted independently.

We designed the shape of the electrodes using a computer simulation of the electric field (solution of Maxwell's equation by the Finite Integration Algorithm MAFIA). The edge of each plate faced to the trap region has a cross section of trapezoid. The lengths of the upper and lower sides, equal to the plate thickness and height were 0.2, 0.8, and 0.2 mm, respectively. The minimum distance from the trap center to the upper side r_0 , that is, half of the radial trap region, is 0.4 mm. In the cross section, the gap angle between two neighboring plates to the trap axis was 23° . We designed this cross section to reduce the high-order potential. Expressing the radial potential as $V = a\xi^4 + b\xi^2 + c$, where ξ is the radial distance from the trap axis normalized to r_0 , the ratio of the octupole and quadrupole potential a/b is 3.6×10^{-3} . In the axial design, increasing of the gap between the center and side plates increases the unwanted axial RF field when the distance between the two side plates is fixed. We decided the gap to be 0.15 mm by considering manufacturing ease, while the distance between the two side plates, that is, the axial trap region, was 1 mm (the center plate had a length of 0.7 mm along the trap axis).

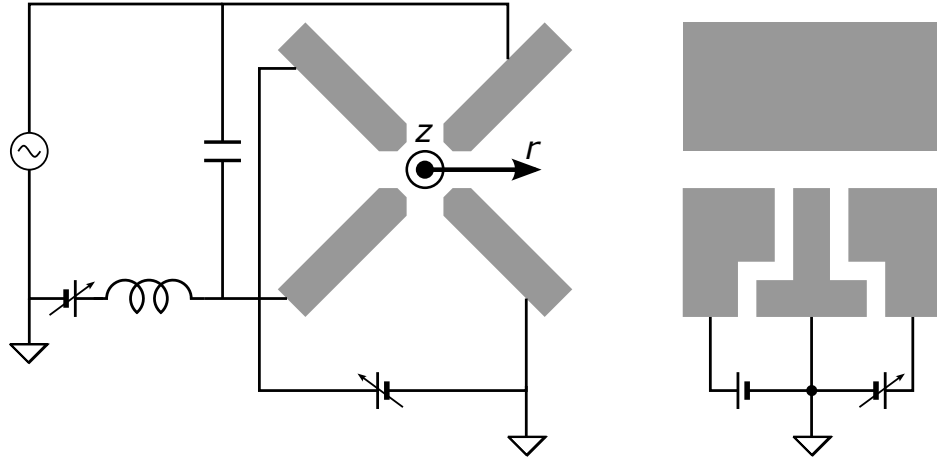


Figure 5.4: Electrodes of the linear RF trap. The RF and DC voltage circuits for trapping and micromotion minimization are shown in Figure. Variable DC voltages show the compensation voltages for the excess micromotion minimization (the variable voltage to the one of the electrodes for the axial confinement includes the confinement voltage.).

We apply a RF voltage to the electrodes and superimpose DC voltages in order to minimize an excess micromotion.

5.2.2 Beam steering

The cooling beam was transported using a polarization-maintaining optical fiber to the table where the linear RF trap system was located. After the output beam from the fiber is collimated, it passes through an acoustooptic modulator (AOM) for switching between the introduction and blocking of the beam to the trap. Then, the cooling beam was split into two beams to be introduced into the trap at two different radial angles. This is aimed at three-dimensional cooling of secular motion, even in the case of the degeneration of two radial secular motions. The two beams were introduced in a counter-propagating manner into the trap. One beam, cooling beam #1, was tilted vertically at an angle of 3° with respect to the trap axis. The other, cooling beam #2, was first introduced along the trap axis to measure excess micromotion in the axial direction. After the minimization, the beam was tilted horizontally at an angle of 3° . The repump laser was placed on the table on which the trap was located. The beam was transported without using an optical fiber and overlapped with cooling beam #1. The clock beam was transported from a different table using a polarization-maintaining fiber. Then, the collimated beam from the fiber was passed through an AOM for switching and was overlapped with cooling beam #1. The beams of the first and second excitation lasers for photoionization were introduced using the same optical fiber. Although these lasers were placed on the table on which the trap was located, we used a fiber to ensure overlap of the two photoionizing beams. Then, the collimated output from the fiber was overlapped with cooling laser #2 when introduced along the trap axis.

Each beam was focused using a lens, whose focal length is as shown in Fig. 2, when introduced into the trap. The power of the beams immediately before the vacuum chamber was $1\ \mu\text{W}$, $40\ \mu\text{W}$, $40\ \mu\text{W}$, $1.2\ \text{mW}$, $200\ \mu\text{W}$, and $200\ \mu\text{W}$ for the clock, cooling #1, cooling #2, repumping, first ionization, and second ionization beams, respectively.

5.3 Results

To conduct single-ion spectroscopy, we established methods for loading single $^{138}\text{Ba}^+$ ions and minimizing the excess micromotion in three dimensions.

After describing them, we show the results of single-ion spectroscopy of the $^2S_{1/2} - ^2D_{5/2}$ transition.

As a common experimental parameter, we applied a magnetic field of approximately $300 \mu\text{T}$ perpendicular to the propagation and polarization directions of the repumping and clock laser beams. This is intended to avoid generation of the dark states in the repumping transition and increase the fluorescence intensity, [40] and also to allow to driving only $\Delta m_F = \pm 2$ Zeeman components in the clock transition. Before applying the desired magnetic field, the magnetic field was eliminated. To conduct this, we appropriately set the polarization direction of the repumping laser and measure the current of a pair of coils that minimize the fluorescence intensity from a single $^{138}\text{Ba}^+$ ion owing to pumping to the dark state in the repumping transition. We repeated this measurement for three pairs of coils to eliminate the environmental magnetic field in three dimensions.

5.3.1 Loading of trapped single $^{138}\text{Ba}^+$ ions

When we load single $^{138}\text{Ba}^+$ ions using photoionization, we tune the first excitation laser for the photoionization to the peak of the absorption signal of the $^1S_0 - ^1P_1$ transition in the Ba atoms in the hollow-cathode lamp. Upon loading, we introduce the cooling and the repumping beam simultaneously, and we observe the fluorescence from the Ba^+ ions trapped and laser cooled during the loading.

A stepwise increase can be observed in the fluorescence accompanying one-by-one ion loading, as we apply the minimum current to the getter required for the loading. However, multiple Ba^+ ions could be easily loaded simultaneously. This is because the evaporation rate of Ba atoms from the getter increased sharply as the temperature increased, and the Ba vapor from the getter spread widely. Therefore, we determined the number of Ba^+ ions after loading. After observing the fluorescence, we immediately blocked the first excitation laser for the photoionization to stop loading. The LED at 455 nm was then irradiated to observe the electron shelving in the $^2D_{5/2}$ state, as shown in Fig. 5.5. We repeated the loading process until we conclude that a single Ba^+ ion was trapped. It was also possible to conduct a one-by-one release of Ba^+ from the trap by blocking the cooling beam. However, this method has poor reproducibility.

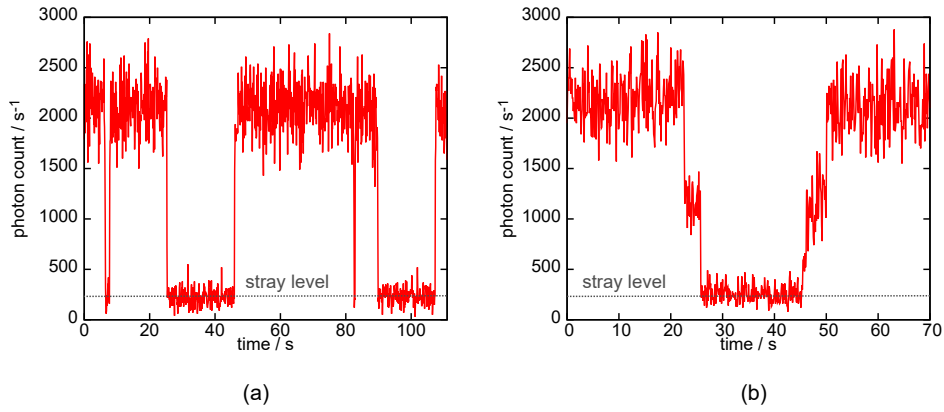


Figure 5.5: Stepwise decreases and increases in fluorescence intensity from (a) single and (b) two trapped $^{138}\text{Ba}^+$ ions by electron shelving to and deexcitation from $^2\text{D}_{5/2}$ state, respectively. Electron shelving occurs by driving the $^2\text{S}_{1/2} - ^2\text{P}_{3/2}$ transition at 455 nm with the LED and subsequent spontaneous decay from the $^2\text{P}_{3/2}$ to the $^2\text{D}_{5/2}$ state. Deexcitation was caused by the quenching of the $^2\text{D}_{5/2}$ state. This measurement is always performed to determine the number of trapped $^{138}\text{Ba}^+$ ions.

5.3.2 Micromotion minimization

After loading a single $^{138}\text{Ba}^+$, the micromotion was three-dimensionally minimized. For this purpose, we detected micromotion using RF-photon cross-correlation. [41, 42] In this measurement, we detected the modulation in the excitation probability by the Doppler shift caused by micromotion. To obtain the correlation, we repeated the measurement of the time interval from the detection of a photon emitted from the trapped single $^{138}\text{Ba}^+$ ion to the upward zero-crossing of the driving RF voltage and drew a histogram as the measured time interval was accumulated in each time bin.

First, excess micromotion was detected in the axial direction when the cooling beam #2 was introduced along the trap axis. We minimized excess micromotion by adjusting one of the DC voltages for the axial confinement. Once the axial micromotion was minimized, we tilted cooling beam #2 in the horizontal plane. Then, we detected the radial micromotion in the vertical and horizontal plane using cooling beam #1 and #2, respectively. We adjusted the two compensation voltages applied for the radial micromotion minimization to simultaneously minimize the correlations detected using the two cooling beams.

Specifically, we did the following. Once the axial micromotion was minimized, the radial micromotion in the vertical plane was minimized detecting the correlation using cooling beam #1. (Fig. 5.6) We adjusted one of the two radial compensation voltages V_1 as the initial value V_a . (Fig. 5.7)

Subsequently, we tilted cooling beam #2 in the horizontal plain to detect the radial micromotion in the horizontal plain. To minimize the micromotion in the horizontal plane, we changed V_1 to $V_a - V_b$ and adjusted the other compensation voltage V_2 to $V_a + V_b$ to minimize the influence of the vertical minimization (Fig. 5.8). We finally checked, or finely adjusted the two compensation voltages if required, to simultaneously minimize the correlations detected using the two cooling beams. (Fig. 5.9)

Figure 5.10 shows the voltage of the DC electrode that minimises axial micromotion. By finding this line, it is possible to adjust the axial potential depth while maintaining the micromotion minimised condition.

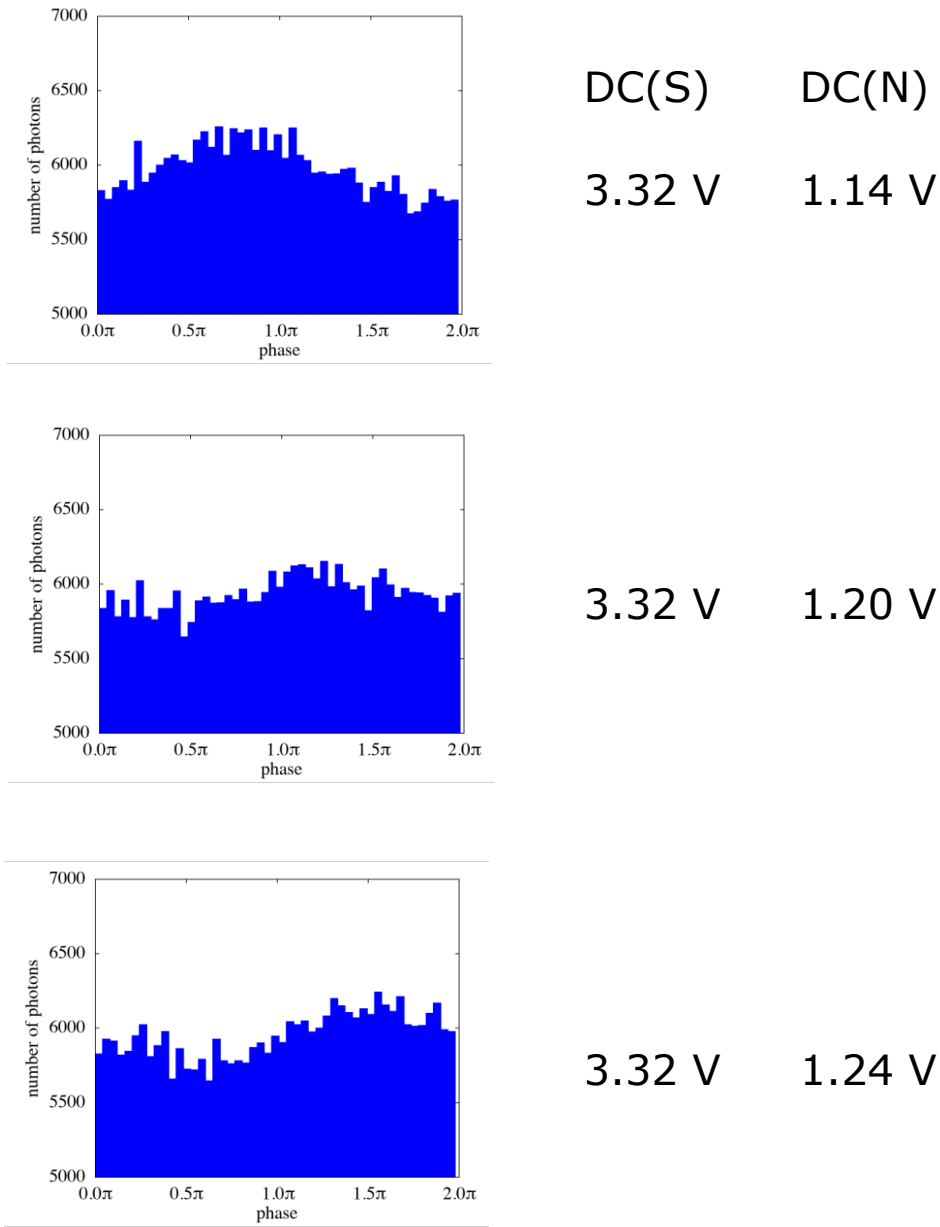


Figure 5.6: RF-photon cross correlation signals in component of axial direction.

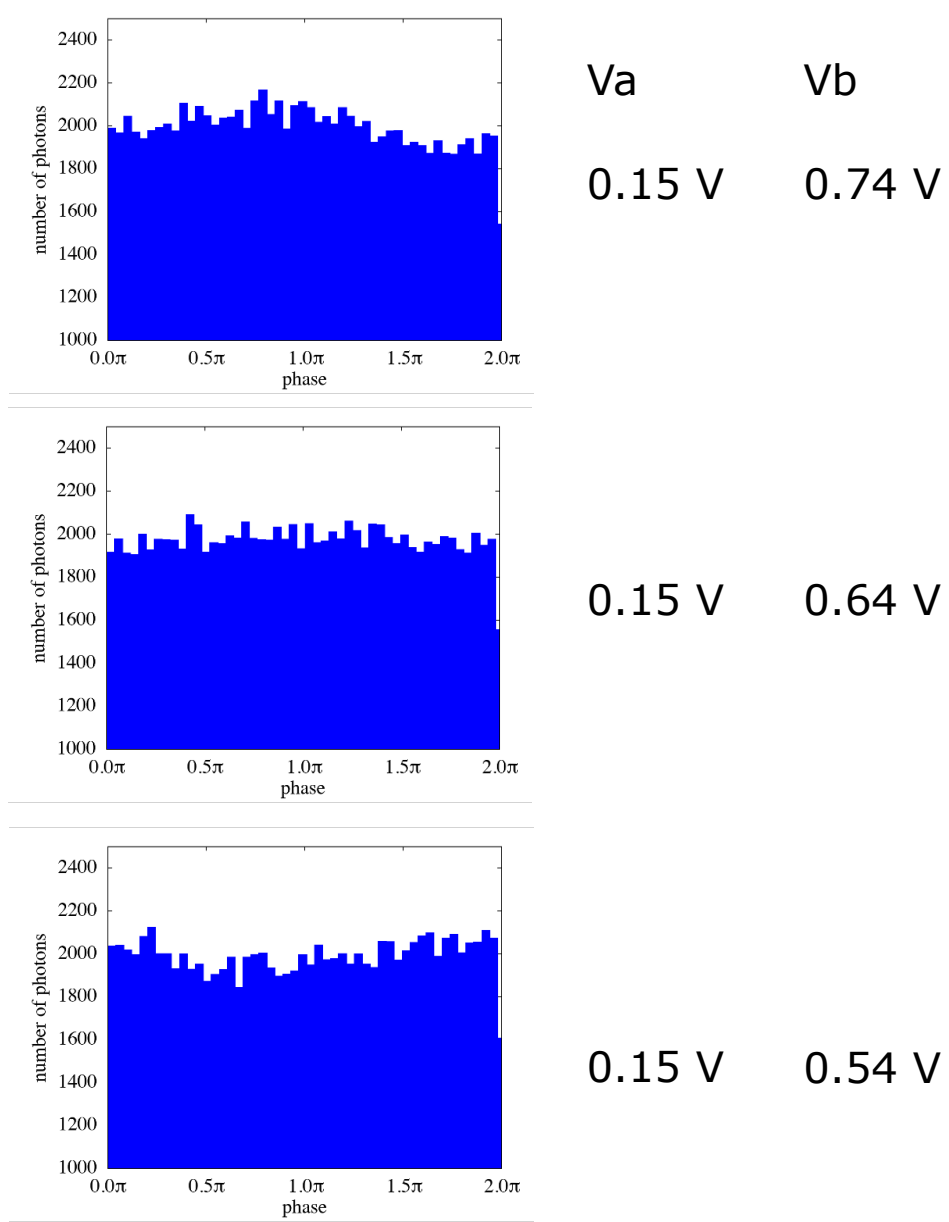


Figure 5.7: RF-photon cross correlation signals in vertical component of radial direction.

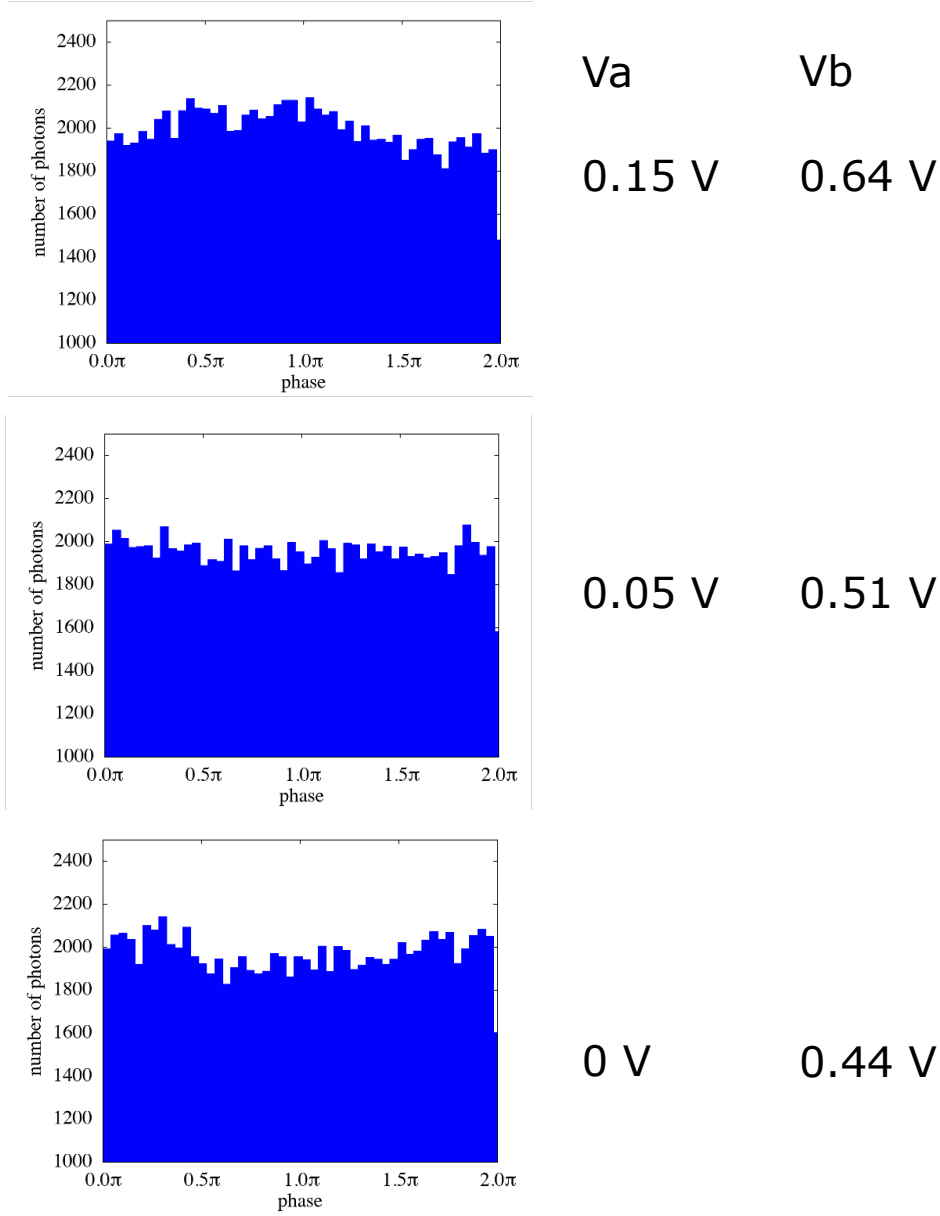


Figure 5.8: RF-photon cross correlation signals in horizontal component of radial direction.

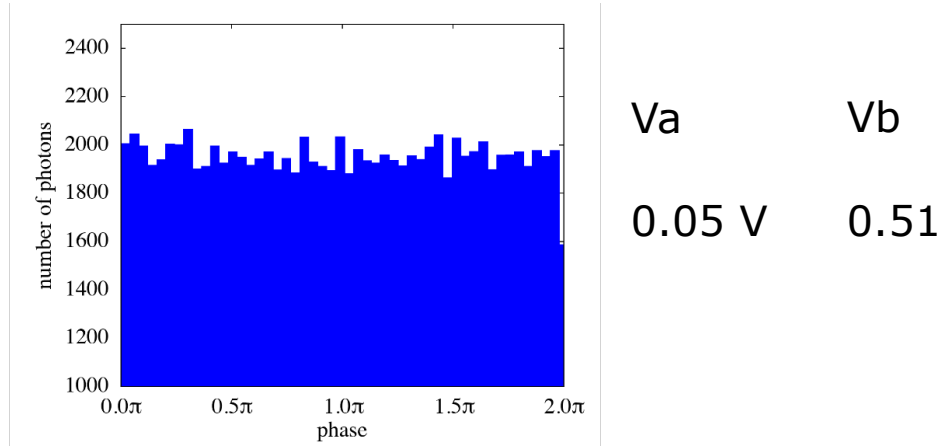


Figure 5.9: RF-photon cross correlation signals in vertical component of radial direction(final checking).

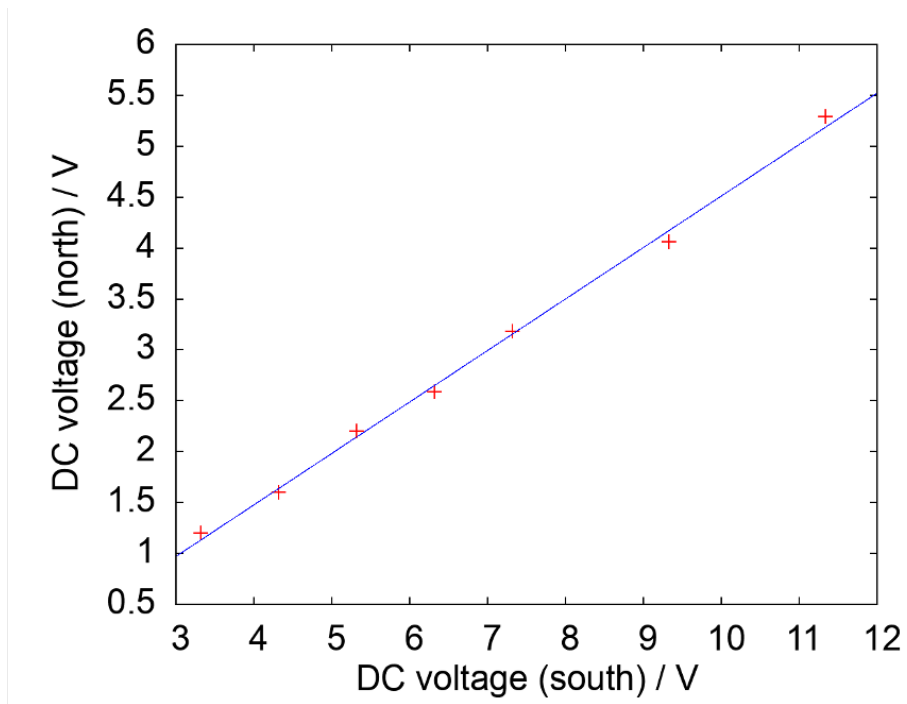


Figure 5.10: Voltage conditions for DC electrodes that minimise axial micro-motion

Chapter 6

Single-ion spectroscopy of the $^2S_{1/2}$ - $^2D_{5/2}$ clock transition

6.1 Procedure

After the excess micromotions are minimized three dimensionally, we conduct single-ion spectroscopy of the $^2S_{1/2}$ - $^2D_{5/2}$ transition. Figure 6.1 shows the time chart of the single-ion spectroscopy. We continuously irradiate with the repumping beam. The procedures are described as follows. (1) We laser cool a single $^{138}\text{Ba}^+$ ion for 100 ms. (2) We block the cooling beam by switching off the driving voltage applied to the AOM to avoid the light shift. This procedure prepares the $^{138}\text{Ba}^+$ ion in the $^2S_{1/2}$ state. (3) We introduce the clock beam pulse. The duration is varied 10–100 ms according to the interaction time intended. (4) We perform the state detection by irradiation with the cooling beam during 100 ms. (5-1) If we observe the fluorescence from the $^2P_{1/2}$ to $^2S_{1/2}$ states, this shows that the $^{138}\text{Ba}^+$ ion has not been excited to the $^2D_{5/2}$ state. We start the next interrogation as we go back to procedure (1). (5-2) If we observe no fluorescence, this shows that the $^{138}\text{Ba}^+$ ion has been excited to the $^2D_{5/2}$ state. Then, we turn on the LED at 614 nm to accelerate deexcitation from the $^2D_{5/2}$ state. The deexcitation rate was roughly measured to be shorter than 100 ms. We continue observing the fluorescence with a gate time of 100 ms until the fluorescence recovers. After the fluorescence recovers, we turn off the LED and we start the next interrogation as we go back to procedure (1). (6) By repeating the trial of procedures (1)–(5), we determine the excitation probability. We repeat the trial typically 50 times to determine the excitation probability. (7) We repeat the determination of the excitation probability as we step-wisely scan the

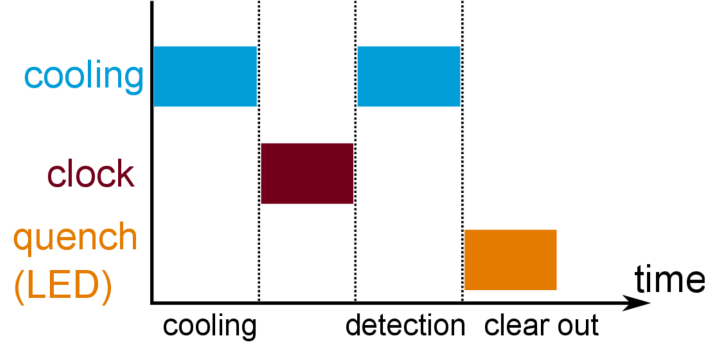


Figure 6.1: chart of single-ion spectroscopy of the $^2S_{1/2} - ^2D_{5/2}$ transition in $^{138}\text{Ba}^+$

frequency of the clock laser by changing the frequency of the RF synthesizer used for the offset locking of the slave laser in the clock laser system. Then, we obtain a spectrum of the transition.

6.2 Results

First, we conduct the single-ion spectroscopy in the range of approximately 30 MHz to identify the four Zeeman components. Then, we observe one of the two stronger components in the four in detail with a smaller frequency interval of the frequency scanning of the clock laser.

Figure 6.2 shows a spectrum of the $m_j = -1/2$ to $m'_j = -5/2$ component of the $^2S_{1/2} - ^2D_{5/2}$ transition. The actual offset frequency in the phase locking of the SH of the ECDL at $1.76\,\mu\text{m}$ to the linewidth-narrowed ECDL at 881 nm was $\sim -377\,\text{MHz}$. This spectrum was detected with a frequency interval of 2.5 kHz and an irradiation duration of the clock laser beam of 50 ms. We conducted the interrogation of the clock transition 50 times at each frequency to determine the excitation probability. The result shows that the carrier spectrum is clearly identified from the sidebands. We identified the sidebands generated from the axial and radial secular motions, and estimated the origin of the higher-order sidebands generated by mixing of the modulation by the axial and radial secular motions. The identifications are shown in the figure.

We observed the carrier and each sideband spectrum with a higher resolution of 500 Hz interval. The results are shown in Fig. 6.3 with a Lorentz profiles fitted to the spectra. The full-width at half maximum of the carrier spectrum was determined to be 1.8 kHz. This value was broader than the

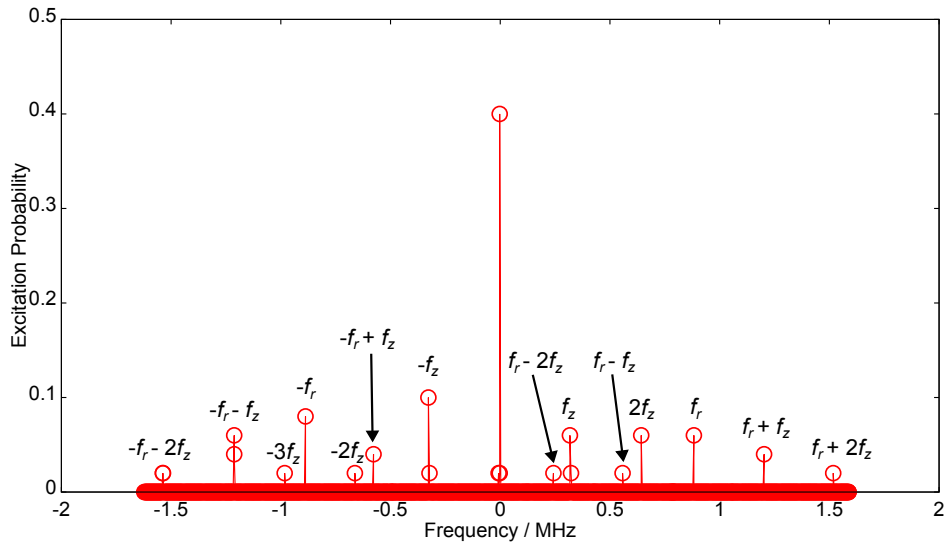


Figure 6.2: Spectrum of $m_j = -1/2$ to $m'_j = -5/2$ component of $^2S_{1/2} - ^2D_{5/2}$ transition in single $^{138}\text{Ba}^+$. The spectrum was detected with a frequency interval of 2.5 kHz and an irradiation duration of the clock laser beam of 50 ms. The excitation probability was determined from 50 trials of the clock transition excitation at each frequency.

clock laser linewidth of 100 Hz as described in chapter 3. The possible reason for the residual broadening is the magnetic field fluctuation.

We estimate the energy of the secular motion from the intensity ratio of the first sidebands to the carrier. The intensity of n -th order sideband and the carrier ($n = 0$) is proportional to a Bessel function of $J_n(kx_i)$, where kx_i is the Lamb-Dicke parameter with the wavenumber k of the clock laser and the amplitude x_i of a secular motion where $i = r, z$ which indicates the radial and axial direction, respectively. From the average of the ratio of the upper and lower first axial sidebands to the carrier, we estimate that $kx_z = 0.94(15)$. Because $kx_z \sim 1$, the single $^{138}\text{Ba}^+$ ion was almost confined in the Lamb-Dicke region of the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ clock transition in the axial direction. The temperature corresponding to the axial secular motion is deduced to be 4.8(1.6) mK. In the same manner, we can estimate the temperature of the radial secular motion to be 3(3) K. The large error is caused by the fact that the clock laser beam propagates with the small angle to the trap axis and the radial component of the beam is small. This prevents from precise analysis of the energy of the radial secular motion.

6.3 Conclusions

We developed a single-ion spectroscopy system for the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ clock transition in Ba^+ . Using this system, we successfully observed resolved-sideband spectra of the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition in single $^{138}\text{Ba}^+$ ions. For the axial direction, we almost confined single $^{138}\text{Ba}^+$ ions in the Lamb-Dicke region of the clock transition, and achieved the temperature of 4.8 mK. The results show that the clock laser phase locked via SHG is useful for the single-ion spectroscopy, and the getter is applicable for single $^{138}\text{Ba}^+$ trapping as a source of Ba atoms.

In this work, the small gaps between the electrodes in the radial direction prevent from introducing the cooling beam in a large angle to the trap axis owing to generation of a large scattering light. It seems be impossible for us to irradiate the trapped Ba^+ ions also with the clock laser beam without guide of the cooling laser beam colinearly overlapped. This disables us to precisely estimate the temperature of the radial secular motions. Moreover, there is a possibility that the small radial component of the cooling beams would not sufficiently laser cool the radial secular motions. For a further development, we need to design the trap electrodes which allows us to introduce the cooling beams with sufficiently large radial components, and more precisely estimate

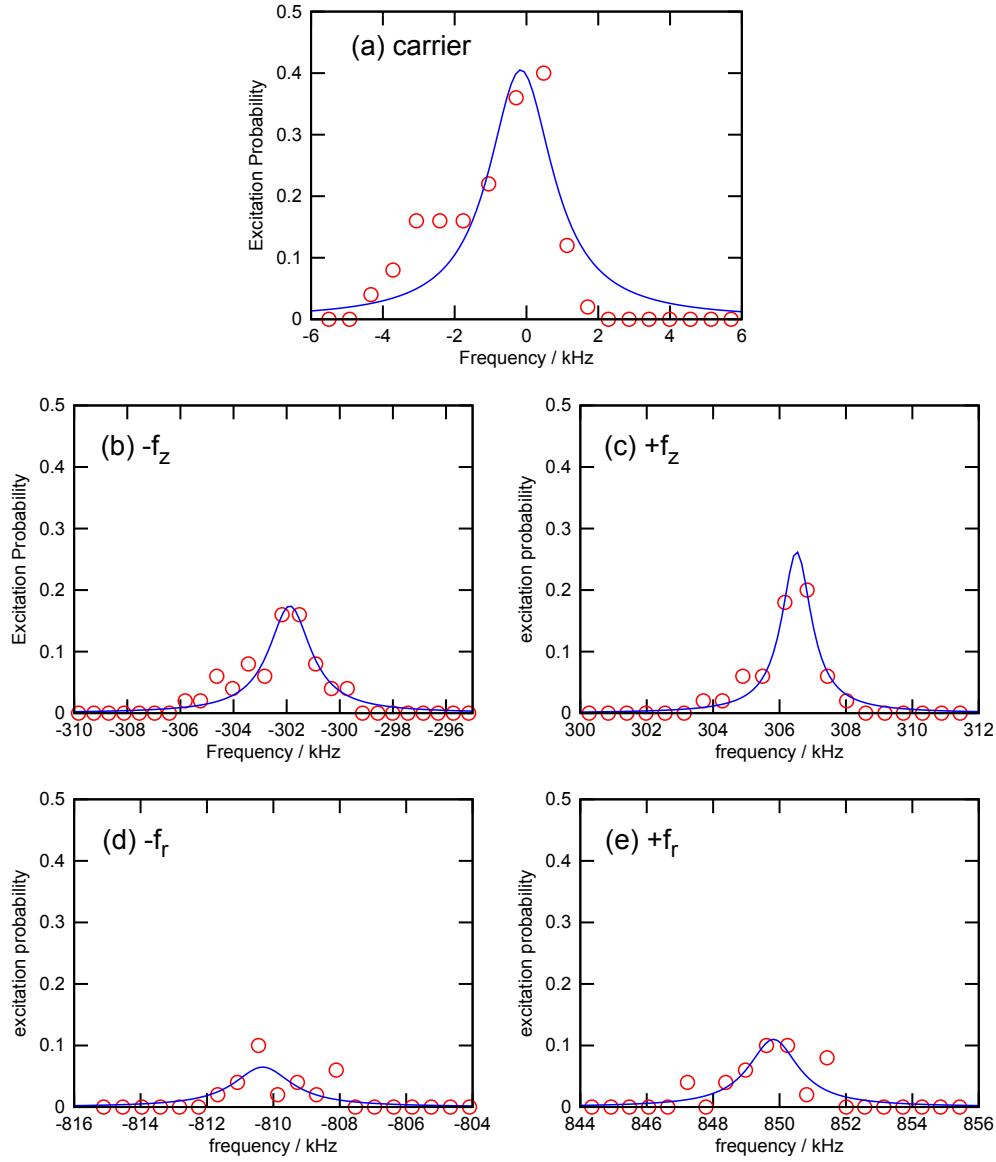


Figure 6.3: High-resolution frequency scanning of carrier and each sideband spectra of $m_j = -1/2$ to $m'_j = -5/2$ component of $^2S_{1/2} - ^2D_{5/2}$ transition. The frequency interval was 500 Hz. The Lorentz profiles fitted to the spectra are also shown.

the temperature of the radial secular motions.

Chapter 7

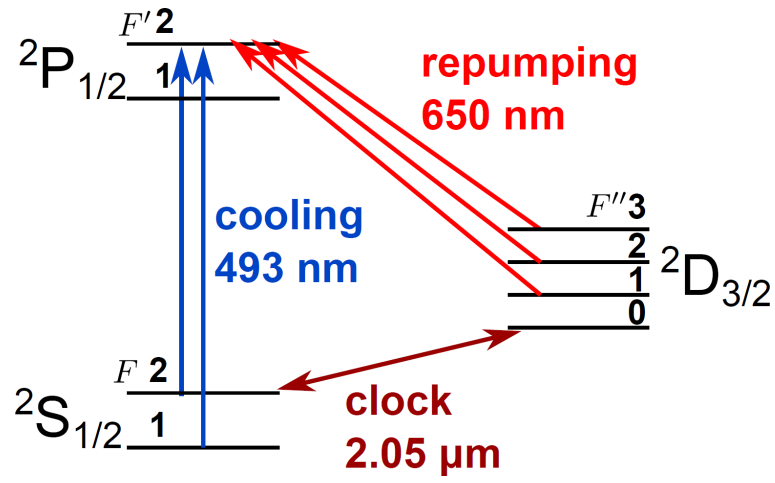
Trapping and laser cooling of odd isotopes of Ba^+ ions

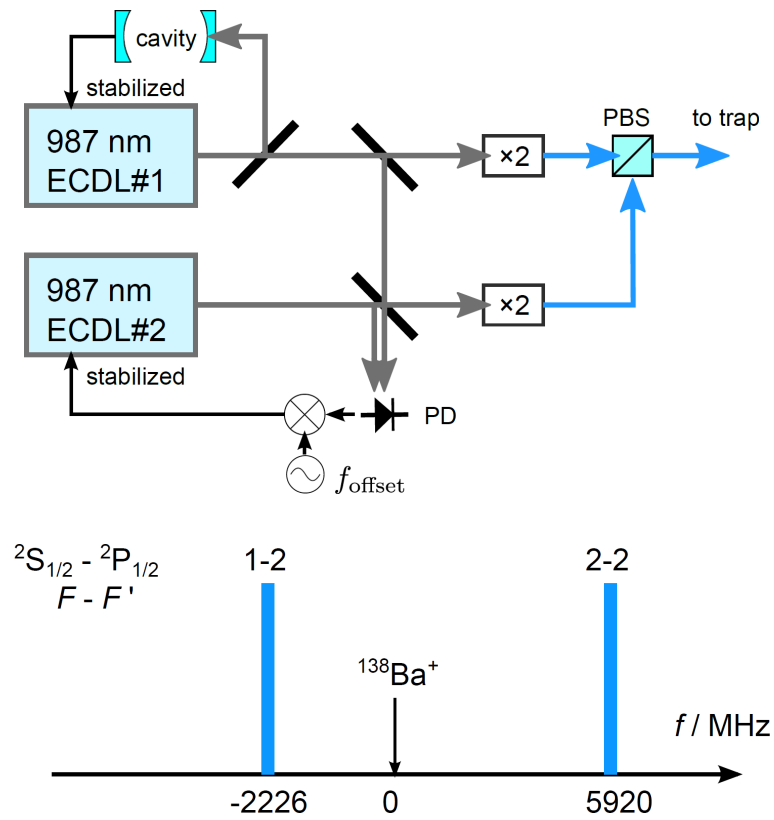
7.1 Laser setup

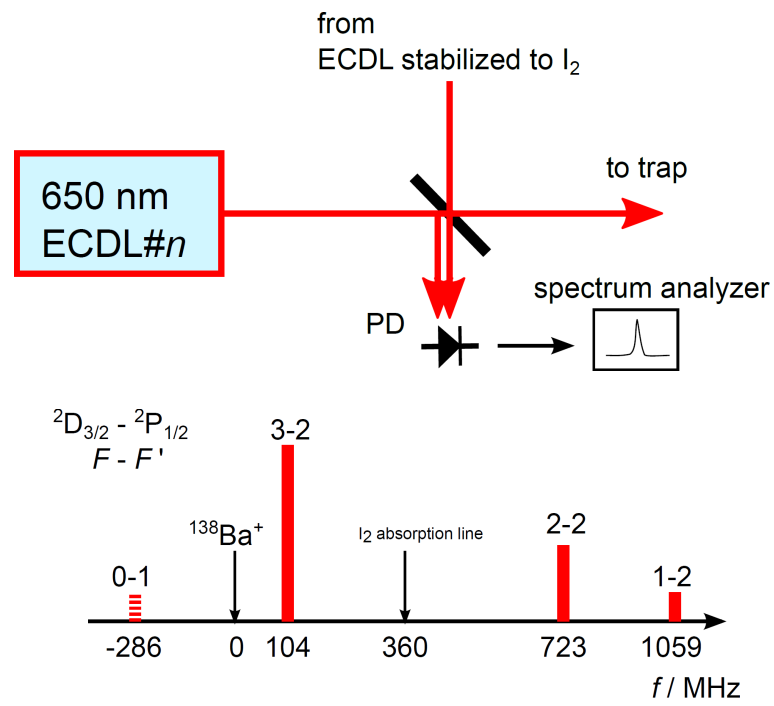
Figure 7.1 shows the level structure of the odd isotope Ba^+ . The odd-numbered isotope Ba^+ has a nuclear spin of $3/2$, so it has a hyperfine structure as shown in the figure. Therefore, in order to laser cool the odd isotope Ba^+ , two cooling lasers and three repumping lasers are required. Figure 7.2 shows the configuration of the cooling lasers. Cooling lasers #1 and #2 are phase-locked to each other and their frequencies are stabilized relative to each other. Cooling laser #1 is stabilized by frequency control of the resonance of a temperature-stabilized optical cavity. The second harmonic of each laser is generated, the beams are overlapped using a PBS, and the beams are introduced into the trap via optical fibers. Figure 7.3 shows the configuration of the repumping lasers. The beat between the repumping lasers #1 to #3 and the stabilised laser using an iodine cell is obtained. The repumping laser #1 is frequency-stabilised by phase-locking, and the frequencies of the repumping lasers #2 and #3 are adjusted manually by free-running. The beams of each repumping laser are overlapped by a half mirror and introduced into the trap.

7.2 Results

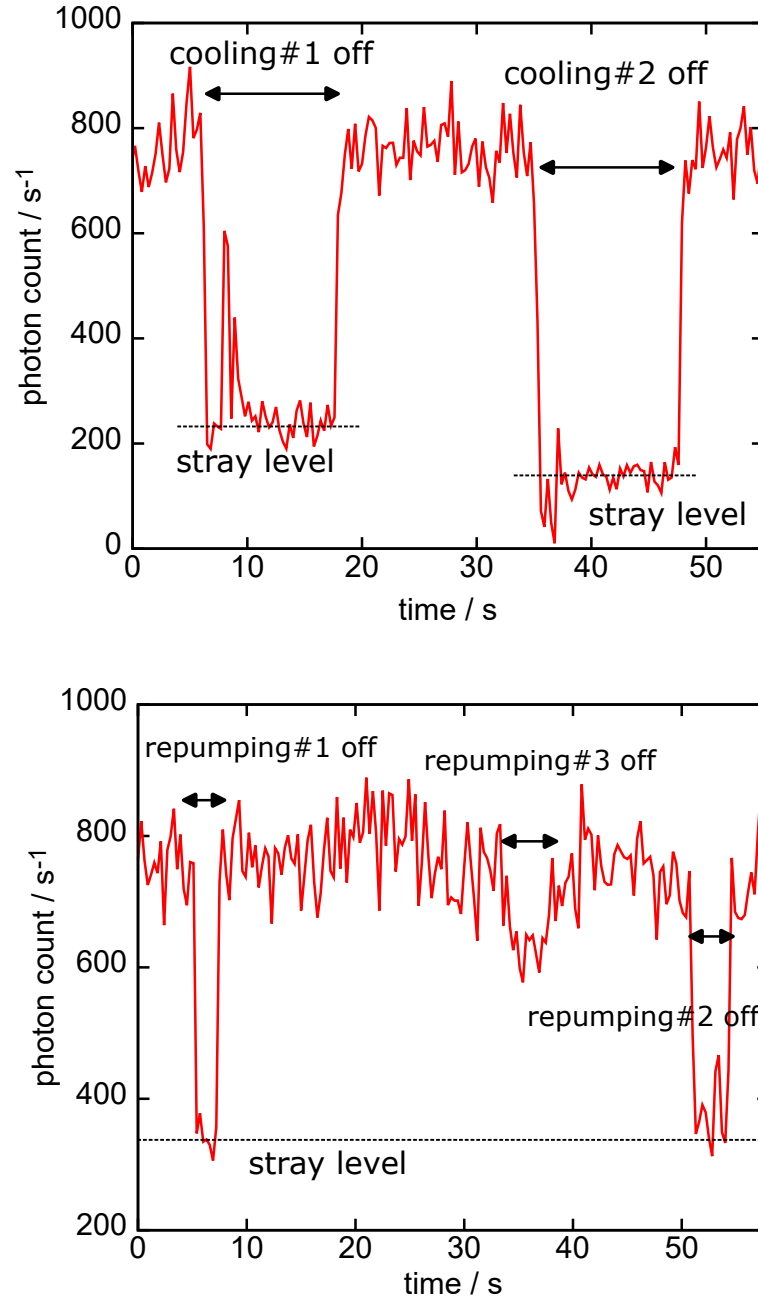
Initially, we trapped ions. Cooling fluorescence was observed through the application of a current of approximately 3.6 A to the barium getter, with

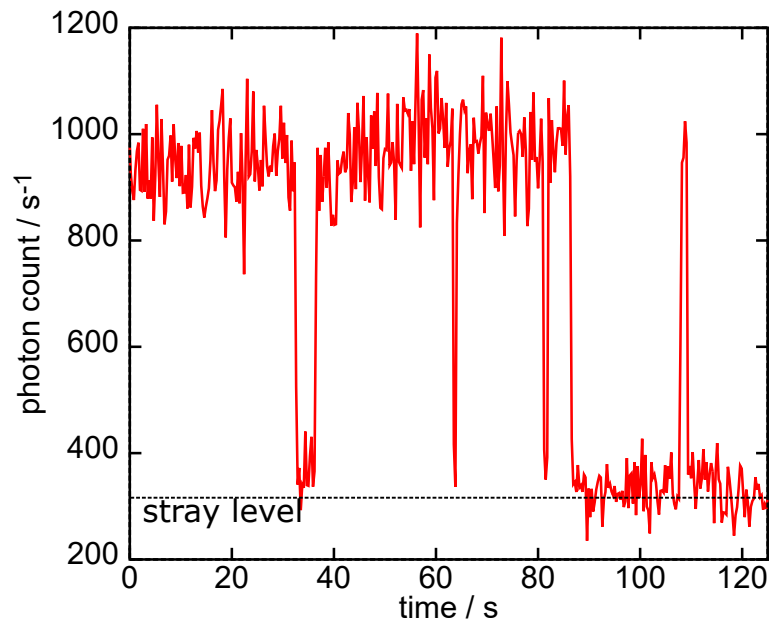
Figure 7.1: level structure of odd isotope Ba^+

Figure 7.2: cooling lasers for odd isotope Ba^+

Figure 7.3: repump lasers for odd isotope Ba⁺

the first ionisation laser being tuned to the resonance frequency of the odd isotope. It is possible to trap single ions by periodically deactivating the RF electric field of the ion trap. As illustrated in Figure 7.4, the fluorescence of the trapped $^{137}\text{Ba}^+$ is evident. It was observed that when each of the cooling and repumping lasers was deactivated, the fluorescence diminished, indicating that all lasers were contributing to the process of laser cooling. Figure 7.5 illustrates quantum jumps observed using an LED that excites the $^2\text{S}_{1/2} - ^2\text{P}_{3/2}$ transition. It is noteworthy that a single $^{137}\text{Ba}^+$ ion was found to generate 700 cps of fluorescence.

Figure 7.4: fluorescence of $^{137}\text{Ba}^+$ ions

Figure 7.5: quantum jumps of single $^{137}\text{Ba}^+$ ions

Chapter 8

Conclusion

In this study we have established a clock transition spectroscopy technique for $^{138}\text{Ba}^+$ ions and a laser cooling technique for odd isotope Ba^+ ions. The conclusions of each chapter are summarised below.

First, we developed a laser capable of spectroscopy of the $1.76\text{ }\mu\text{m}$ $^2\text{S}_{1/2}$ - $^2\text{D}_{5/2}$ clock transition (clock laser). By controlling the 881-nm laser to resonate with the optical cavity, we narrowed the line width. The clock laser was achieved by detecting and phase locking the beat between the 881 nm laser, which had been narrowed by generating the second harmonic of a $1.76\text{ }\mu\text{m}$ laser, and the 881 nm laser. The linewidth of the 881 nm laser was evaluated at 200 Hz compared to the linewidth of the laser linewidth narrowed by resonance in another optical cavity. As the wavelength of the laser being compared was 822 nm, the optical frequency comb was phase locked with it and the linewidth of each mode of the optical comb was linewidth narrowed and the beat between the comb and the 881 nm laser was observed. The linewidth of the laser at $1.76\text{ }\mu\text{m}$ wavelength is estimated to be half that at 100 Hz. (Chapter 3)

A clock laser was developed by generating a narrow linewidth laser beam by differential frequency mixing using two phase-locked lasers with a narrow linewidth as described above. In this study, the difference frequency was set to $1.76\text{ }\mu\text{m}$ and the linewidth was evaluated by comparison with the clock laser described above. The spectral width of the beat signal of the two lasers was 100 Hz, demonstrating the effectiveness of this method. By changing the wavelength of the laser that generates the difference frequency, it is possible to obtain a narrow linewidth laser beam with a wavelength of $2.05\text{ }\mu\text{m}$. (Chapter 4)

We then established a technique for trapping and cooling a single Ba^+ ion

in a linear RF trap. Since our ultimate goal is to use multiple ions, we adopted a linear RF trap that can cool multiple ions on the trap axis. However, in this study we aimed to establish a technique for cooling a single trapped ion to the Lamb-Dicke state, where the range of ion motion is less than the wavelength of the clock transition. In addition, a getter was used as a source of Ba atoms. Ba atoms were generated from the getter, trapped by photoionisation in the trap, laser cooled using the 493 nm $^2S_{1/2} - ^2P_{1/2}$ transition and repumped using the 650 nm $^2D_{3/2} - ^2P_{1/2}$ transition to complete the cooling cycle.

In real traps, unwanted electrostatic fields usually occur and ions are not trapped in the centre of the trap. If they are displaced from the centre of the trap, the RF electric field used for trapping becomes stronger and the ions are overexcited (excess micromotion). This excess micromotion was first detected and minimised in the axial direction. Next, it was detected and minimised in the radial direction using cooling lasers irradiated from two directions at an angle of 3 degrees from the axis. (Chapter 5)

We then performed single-ion spectroscopy of the $^2S_{1/2} - ^2D_{5/2}$ clock transition using a narrow linewidth clock laser. We observed the vibrational sidebands caused by the continuous motion of the ions by separating them from the carrier spectrum, and detected a spectrum in which the carrier was sufficiently larger than the sidebands. With respect to the direction of the trap axis, we estimated the temperature reached by the secular motion to be 3.5 mK, confirming that the Lamb-Dicke region had been reached. (Chapter 6)

In parallel, we succeeded in trapping and laser cooling a single odd isotope $^{137}\text{Ba}^+$. The levels of the odd isotope $^{135}\text{Ba}^+, ^{137}\text{Ba}^+$ each have hyperfine structures, and two cooling lasers and three repumping lasers are required to excite all the hyperfine structures. We prepared independent lasers and phase locked them as necessary to stabilise their frequencies. The photoionisation method used in this study cannot separate $^{135}\text{Ba}^+$ and $^{137}\text{Ba}^+$. However, it was found that single ions could be introduced into the trap and laser cooled for specific isotopes. (Chapter 7)

8.1 Prospects

In terms of future prospects, we discuss the realization of a single $^{138}\text{Ba}^+$ ion optical clock and a multi-ion clock using the odd isotope Ba^+ .

8.1.1 optical clock using $^{138}\text{Ba}^+$ ions

The largest frequency shift is the first-order Zeeman shift. We can remove this shift by measuring the magnetic field dependence on the clock transition frequency and extrapolate the value at the zero magnetic field. Then, we average the zero-magnetic-field frequency of each Zeeman component and find the center of gravity of the clock transition.

To measure this precisely, a narrow spectral width is essential. The main reason for the broad linewidth of the clock-transition spectra in single $^{138}\text{Ba}^+$ is the fluctuation of the ambient magnetic field mainly caused by the AC current of the electricity. This can be improved by detecting the clock-transition excitation synchronously to the electricity, and/or improve the magnetic shield. Cancellation of the magnetic field fluctuation has been successfully developed by combination of feedback and feed-forward control of the current of the coils. [43]

In addition, it is necessary to further improve the linewidth and stability of the clock laser. The linewidth or short-term frequency stability can be improved by increasing the servo gain used in the FM sideband technique. We evaluated the linewidth to be 29 Hz relative to the resonance of the high-finesse cavity by integrating the PSD of the frequency fluctuation in a large frequency range. However, we particularly need to evaluate and improve the close-to carrier components because the absolute linewidth has been achieved to be 100 Hz. And also, it is inevitable to introduce an anti-vibration table and improve the method of supporting the optical cavity. Long-term frequency stability or frequency drift can be improved by controlling the temperature of the high-finesse cavity to that of the zero thermal coefficient of the ULE glass, which is the material of the spacer of the high-finesse cavity.

(Highly accurate optical frequency comparison is required for clock operation. Therefore, an optical frequency comb locked to another optical clock is required.) By solving the above problems, an uncertainty of 10^{-17} can be achieved in single-ion clock using $^{138}\text{Ba}^+$ ions. Note that the uncertainty of 1.2×10^{-17} has been achieved in single $^{88}\text{Sr}^+$ ions, [44] which has the energy levels similar to $^{138}\text{Ba}^+$

8.1.2 multi-ion clock using odd isotopes

First, we can trap only single $^{137}\text{Ba}^+$ ions and have not observed more than two $^{137}\text{Ba}^+$ ions. This is because the natural width of the $^1\text{S}_0 - ^1\text{P}_1$ transition, currently used for the first excitation of the photoionization, is greater than

the isotope shift between the two odd isotopes and selective ionization of the specific odd isotopes is impossible. To selectively load the one of the two odd isotopes $^{135}\text{Ba}^+$ and $^{137}\text{Ba}^+$, a method for odd-isotope selective excitation of Ba atoms is required. We need to investigate other schemes including narrower transitions such as $^1\text{S}_0 - ^3\text{P}_1$ transition.

Second, if is necessary to develop a clock laser at the wavelength of $2.05\ \mu\text{m}$ to drive $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition. This can be achieved by improving the clock laser based on the difference frequency generation developed in this study. If we change the ECDL at 664 nm , which is one of the fundamental waves used in the difference frequency generation, to another ECLD at 701 nm , we can achieve a narrow-line radiation at $2.05\ \mu\text{m}$.

Third, it is difficult or impossible to decouple the $F=0$ hyperfine state in the $^2\text{D}_{3/2}$. The hyperfine intervals in the $^2\text{D}_{3/2}$ is smaller than those in $^{171}\text{Yb}^+$, although the $F=2$ in the $^2\text{D}_{3/2}$ state in $^{171}\text{Yb}^+$ successfully decoupled from the cooling cycle and single-ion spectroscopy of the $^2\text{S}_{1/2}(F=0) - ^2\text{D}_{3/2}(F=2)$ transition has been performed. [45,46] One possible solution is state detection by depleting the ions remaining in the $^2\text{S}_{1/2}$ after the excitation of the $^2\text{S}_{1/2} - ^2\text{D}_{3/2}$ transition. This scheme has been realized using $^{138}\text{Ba}^+$, [21] and driving the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition can be used for this purpose [12] using the clock laser developed in this study.

By solving the problems above, I believe that a multi-ion clock using the odd isotope Ba^+ will realize. Then, it is essential to increase the number of ions to achieve similar frequency stability to that of the optical lattice clock using atoms. This can be achieved, for example, by increasing the number of trap electrodes in the axial direction.

Bibliography

- [1] J.A. Sherman, W. Trimble, S. Metz, W. Nagourney, and N. Fortson. Progress on indium and barium single ion optical frequency standards. In *Digest of the LEOS Summer Topical Meetings, 2005.*, pages 99–100. IEEE, New York, 2005.
- [2] Norval Fortson. Possibility of measuring parity nonconservation with a single trapped atomic ion. *Phys. Rev. Lett.*, 70:2383–2386, Apr 1993.
- [3] I. V. Inlek, C. Crocker, M. Lichtman, K. Sosnova, and C. Monroe. Multispecies trapped-ion node for quantum networking. *Phys. Rev. Lett.*, 118:250502, Jun 2017.
- [4] David Hucul, Justin E. Christensen, Eric R. Hudson, and Wesley C. Campbell. Spectroscopy of a synthetic trapped ion qubit. *Phys. Rev. Lett.*, 119:100501, Sep 2017.
- [5] W. Neuhauser, M. Hohenstatt, P. Toschek, and H. Dehmelt. Optical-sideband cooling of visible atom cloud confined in parabolic well. *Phys. Rev. Lett.*, 41:233–236, Jul 1978.
- [6] W. Neuhauser, M. Hohenstatt, P. E. Toschek, and H. Dehmelt. Localized visible ba^+ mono-ion oscillator. *Phys. Rev. A*, 22:1137–1140, Sep 1980.
- [7] N. Yu, W. Nagourney, and H. Dehmelt. Radiative lifetime measurement of the ba^+ metastable $d_{3/2}$ state. *Phys. Rev. Lett.*, 78:4898–4901, Jun 1997.
- [8] Carolyn Auchter, Thomas W. Noel, Matthew R. Hoffman, Spencer R. Williams, and Boris B. Blinov. Measurement of the branching fractions and lifetime of the $5D_{5/2}$ level of ba^+ . *Phys. Rev. A*, 90:060501, Dec 2014.

- [9] K. J. Arnold, R. Kaewuam, S. R. Chanu, T. R. Tan, Zhiqiang Zhang, and M. D. Barrett. Precision measurements of the $^{138}\text{Ba}^+ 6s^2S_{1/2} - 5d^2D_{5/2}$ clock transition. *Phys. Rev. Lett.*, 124:193001, May 2020.
- [10] Ravid Shaniv, Nitzan Akerman, Tom Manovitz, Yotam Shapira, and Roee Ozeri. Quadrupole shift cancellation using dynamic decoupling. *Phys. Rev. Lett.*, 122:223204, Jun 2019.
- [11] Nati Aharon, Nicolas Spethmann, Ian D Leroux, Piet O Schmidt, and Alex Retzker. Robust optical clock transitions in trapped ions using dynamical decoupling. *New Journal of Physics*, 21(8):083040, aug 2019.
- [12] M. R. Dietrich, N. Kurz, T. Noel, G. Shu, and B. B. Blinov. Hyperfine and optical barium ion qubits. *Phys. Rev. A*, 81:052328, May 2010.
- [13] Dahyun Yum, Debashis De Munshi, Tarun Dutta, and Manas Mukherjee. Optical barium ion qubit. *J. Opt. Soc. Am. B*, 34(8):1632–1636, Aug 2017.
- [14] L. Slodička, G. Hétet, N. Röck, S. Gerber, P. Schindler, M. Kumph, M. Hennrich, and R. Blatt. Interferometric thermometry of a single sub-doppler-cooled atom. *Phys. Rev. A*, 85:043401, Apr 2012.
- [15] Warren Nagourney, Nan Yu, and Hans Dehmelt. High resolution Ba^+ monoion spectroscopy with frequency stabilized color-center laser. *Optics Communications*, 79(3):176–180, 1990.
- [16] X. Zhao, N. Yu, H. Dehmelt, and W. Nagourney. Isotope-shift measurement of the $6^2s_{1/2} - 5^2d_{5/2}$ transition in Ba^+ . *Phys. Rev. A*, 51:4483–4486, Jun 1995.
- [17] K. B. MacAdam, A. Steinbach, and C. Wieman. A narrow band tunable diode laser system with grating feedback, and a saturated absorption spectrometer for Cs and Rb. *American Journal of Physics*, 60(12):1098–1111, 12 1992.
- [18] V. V. Flambaum and V. A. Dzuba. Search for variation of the fundamental constants in atomic, molecular, and nuclear spectra. *Canadian Journal of Physics*, 87(1):25 – 33, 2009.
- [19] R. M. Godun, P. B. R. Nisbet-Jones, J. M. Jones, S. A. King, L. A. M. Johnson, H. S. Margolis, K. Szymaniec, S. N. Lea, K. Bongs, and P. Gill.

- Frequency Ratio of Two Optical Clock Transitions in Yb + 171 and Constraints on the Time Variation of Fundamental Constants. *Physical Review Letters*, 113(21):210801, November 2014.
- [20] N. Huntemann, B. Lipphardt, Chr. Tamm, V. Gerginov, S. Weyers, and E. Peik. Improved Limit on a Temporal Variation of m_p / m_e from Comparisons of Yb + and Cs Atomic Clocks. *Physical Review Letters*, 113(21):210802, November 2014.
- [21] Adam Kleczewski, Matthew R. Hoffman, J. A. Sherman, Eric Magnuson, B. B. Blinov, and E. N. Fortson. Coherent excitation of the $6S_{1/2}$ to $5D_{3/2}$ electric-quadrupole transition in $^{138}\text{Ba}^+$. *Phys. Rev. A*, 85:043418, Apr 2012.
- [22] A. Bartels, C. W. Oates, L. Hollberg, and S. A. Diddams. Stabilization of femtosecond laser frequency combs with subhertz residual linewidths. *Opt. Lett.*, 29(10):1081–1083, May 2004.
- [23] Masashi Abe, Keisuke Takahata, and Hiroyuki Sasada. Sub-doppler resolution 3.4 μm spectrometer with an efficient difference-frequency-generation source. *Opt. Lett.*, 34(11):1744–1746, Jun 2009.
- [24] Hideyuki Sera, Masashi Abe, Kana Iwakuni, Sho Okubo, Hajime Inaba, Feng-Lei Hong, and Hiroyuki Sasada. Sub-doppler resolution mid-infrared spectroscopy using a difference-frequency-generation source spectrally narrowed by laser linewidth transfer. *Opt. Lett.*, 40(23):5467–5470, Dec 2015.
- [25] Kazuhiko Sugiyama and Jun Yoda. Stabilization of sum or difference frequency by controlling one of the fundamental lasers of frequency mixing. *Optics Communications*, 95(1-3):77–80, January 1993.
- [26] Kazumoto Hosaka, Hajime Inaba, Yoshiaki Nakajima, Masami Yasuda, Takuya Kohno, Atsushi Onae, and Feng-Lei Hong. Evaluation of the clock laser for an yb lattice clock using an optic fiber comb. *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, 57(3):606–612, 2010.
- [27] R. W. P. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward. Laser phase and frequency stabilization using an optical resonator. *Applied Physics B Photophysics and Laser Chemistry*, 31(2):97–105, June 1983.

- [28] Mark Notcutt, Long-Sheng Ma, Jun Ye, and John L. Hall. Simple and compact 1-hz laser system via an improved mounting configuration of a reference cavity. *Opt. Lett.*, 30(14):1815–1817, Jul 2005.
- [29] D. S. Elliott, Rajarshi Roy, and S. J. Smith. Extracavity laser band-shape and bandwidth modification. *Phys. Rev. A*, 26:12–18, Jul 1982.
- [30] Shuhei Hatanaka, Kazuhiko Sugiyama, Masatoshi Mitaki, Masatoshi Misono, Sergey N. Slyusarev, and Masao Kitano. Phase locking of a mode-locked titanium–sapphire laser-based optical frequency comb to a reference laser using a fast piezoelectric actuator. *Applied Optics*, 56(12):3615, April 2017.
- [31] G. Leschhorn, T. Hasegawa, and T. Schaetz. Efficient photo-ionization for barium ion trapping using a dipole-allowed resonant two-photon transition. *Applied Physics B*, 108:159–165, June 2012.
- [32] J. A. Sherman, A. Andalkar, W. Nagourney, and E. N. Fortson. Measurement of light shifts at two off-resonant wavelengths in a single trapped Ba^+ ion and the determination of atomic dipole matrix elements. *Phys. Rev. A*, 78:052514, Nov 2008.
- [33] T.W. Haensch and B. Couillaud. Laser frequency stabilization by polarization spectroscopy of a reflecting reference cavity. *Optics Communications*, 35(3):441–444, December 1980.
- [34] U. Schünemann, H. Engler, R. Grimm, M. Weidemüller, and M. Zielonkowski. Simple scheme for tunable frequency offset locking of two lasers. *Review of Scientific Instruments*, 70(1):242–243, 01 1999.
- [35] Kaduki Komori, Yosuke Takasu, Mitsutaka Kumakura, Yoshiro Takahashi, and Tsutomu Yabuzaki. Injection-locking of blue laser diodes and its application to the laser cooling of neutral ytterbium atoms. *Japanese Journal of Applied Physics*, 42(8R):5059, aug 2003.
- [36] K. J. Arnold, S. R. Chanu, R. Kaewuam, T. R. Tan, L. Yeo, Zhiqiang Zhang, M. S. Safronova, and M. D. Barrett. Measurements of the branching ratios for $6P_{1/2}$ decays in $^{138}\text{Ba}^+$. *Phys. Rev. A*, 100:032503, Sep 2019.
- [37] R. K. Raj, D. Bloch, J. J. Snyder, G. Camy, and M. Ducloy. High-frequency optically heterodyned saturation spectroscopy via resonant degenerate four-wave mixing. *Phys. Rev. Lett.*, 44:1251–1254, May 1980.

- [38] L.-S. Ma and J.L. Hall. Optical heterodyne spectroscopy enhanced by an external optical cavity: toward improved working standards. *IEEE Journal of Quantum Electronics*, 26(11):2006–2012, 1990.
- [39] W.J. Kozlovsky, C.D. Nabors, and R.L. Byer. Efficient second harmonic generation of a diode-laser-pumped cw nd:yag laser using monolithic mgo:linbo/sub 3/ external resonant cavities. *IEEE Journal of Quantum Electronics*, 24(6):913–919, 1988.
- [40] D. J. Berkeland and M. G. Boshier. Destabilization of dark states and optical spectroscopy in Zeeman-degenerate atomic systems. *Physical Review A*, 65(3):033413, February 2002.
- [41] D. J. Berkeland, J. D. Miller, J. C. Bergquist, W. M. Itano, and D. J. Wineland. Minimization of ion micromotion in a paul trap. *Journal of Applied Physics*, 83(10):5025–5033, May 1998.
- [42] J. Keller, H. L. Partner, T. Burgermeister, and T. E. Mehlstäubler. Precise determination of micromotion for trapped-ion optical clocks. *Journal of Applied Physics*, 118(10):104501, September 2015.
- [43] B. Merkel, K. Thirumalai, J. E. Tarlton, V. M. Schafer, C. J. Ballance, T. P. Harty, and D. M. Lucas. Magnetic field stabilization system for atomic physics experiments. *Review of Scientific Instruments*, 90(4):044702, 04 2019.
- [44] Pierre Dubé, Alan A. Madej, Maria Tibbo, and John E. Bernard. High-accuracy measurement of the differential scalar polarizability of a $^{88}\text{Sr}^+$ clock using the time-dilation effect. *Phys. Rev. Lett.*, 112:173002, Apr 2014.
- [45] D Engelke and Chr Tamm. Dark times in the resonance fluorescence of trapped ^{171}Yb ions caused by spontaneous quantum jumps to the $^2D_{3/2}$ ($F = 2$) state. *Europhysics Letters (EPL)*, 33(5):347–352, February 1996.
- [46] Yasutaka Imai, Tatsuya Nishi, Masatoshi Nishizaki, Sho Kawajiri, Yuto Muroki, Rikizo Ikuta, Kai Matsumoto, Masao Kitano, and Kazuhiko Sugiyama. Single-ion spectroscopy system for the $^2S_{1/2}$ ($F = 0$) - $^2D_{3/2}$ ($F = 2$) transition in $^{171}\text{Yb}^+$. *Radio Science*, 51(8):1385–1395, August 2016.

List of publications

A. Full Length Papers with Review

1. (submitted to Journal of the Physical Society of Japan)
Hiroto Fujisaki, Takashi Moriya, Tadayuki Yoshitake, Dai Osada,
Yoshihiro Hara, Kengo Takeshita, Shinnosuke Yukawa, Yosuke Asai,
Ryohei Shibata, Yasutaka Imai, Masao Kitano, and Kazuhiko Sugiyama,
“Spectroscopy system for $^2S_{1/2}$ - $^2D_{5/2}$ clock transition at $1.76\mu\text{m}$ in
trapped barium ions using diode-laser-based light sources”

B. Conference proceedings

1. Hiroto Fujisaki, Kazuhiko Sugiyama, Yasutaka Imai,
and Masao Kitano
“Development of laser-diode based light sources for $\text{Ba}^+ ^2S_{1/2}$ - $^2D_{5/2}$
clock transition at $1.76\mu\text{m}$ and observation of quantum jumps”
*in Proceeding of the 7th International Workshop on Fundamental Physics
Using Atoms* 2014, Tokyo, Japan, Mar., pp.133-135

C. International Conferences with Review

1. Hiroto Fujisaki, Kazuhiko Sugiyama, Yasutaka Imai, and Masao Kitano
“Laser-diode-based light source for single-ion spectroscopy of the $^2S_{1/2}$
- $^2D_{5/2}$ clock transition in Ba^+ at $1.76\mu\text{m}$ ”
*The 6th International Conference on Trapped Charged Particles and
Fundamental Physics* Takamatsu, Japan
2. Hiroto Fujisaki, Yasutaka Imai, Keisuke Nishida, Masatoshi Mitaki,
Masao Kitano, and Kazuhiko Sugiyama
“Single-ion spectroscopy of the $^2S_{1/2}$ - $^2D_{5/2}$ transition in $^{138}\text{Ba}^+$ and

laser cooling of odd-isotope Ba^+ ”

The 4th European Conference on Trapped Ions 2016, Arosa, Switzerland

3. Hiroto Fujisaki, Yasutaka Imai, and Kazuhiko Sugiyama

“Spectroscopy of the clock transition and odd-isotope-selective loading of single Ba^+ ions towards realization of an optical frequency standard”

The 5th European Conference on Trapped Ions 2018, Rehovot, Israel

4. Hiroto Fujisaki, Yasutaka Imai, and Kazuhiko Sugiyama

“Progress towards realization of an optical frequency standard using trapped Ba^+ ions”

Early Career Conference in Trapped Ions 2020, Geneve, Switzerland