



# Study on laser diode excitation of Sr<sup>+</sup> Ions and Ca atoms for frequency standards

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# 神戸大学博士論文

Study on Laser Diode Excitation of  $\text{Sr}^+$  Ions and Ca Atoms for Frequency Standards  
原子周波数標準に用いる Sr イオンと Ca 原子の半導体レーザー励起光源の研究

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## Abstract

In this thesis, a series of studies on a laser diode light sources for exciting  $\text{Sr}^+$  ions and Ca atoms are summarized.  $\text{Sr}^+$  ions and Ca atoms are considered candidates for the next-generation atomic frequency standards of microwave frequency and optical frequency respectively. The accuracy of the Cs atomic clock, which is the current atomic frequency standard, has been improved at the rate of one order of magnitude every 10 years. However, the accuracy improvement is approaching the limit due to the processing accuracy of the system and the properties of the material used. In the field of atomic frequency standards, such technologies as ion trap and laser cooling have been utilized to improve the accuracy of standards. In the development of  $\text{Sr}^+$  ions and Ca atoms frequency standards, a laser diode system is introduced for these purposes. This leads to the possibility of a future easy-to-handle, compact, transportable apparatus. In the system of frequency standards in the microwave range, in addition to  $\text{Sr}^+$  ions,  $\text{Hg}^+$  ions are being studied at the National Institute of Standards and Technology (NIST) in the United States and  $\text{Yb}^+$  ions are being studied at the National Measurement Laboratory (CSIRO) in Australia. However, since the wavelengths of optical pumping are 194 nm for  $\text{Hg}^+$  ion and 369 nm for  $\text{Yb}^+$  ion, there a lot of problems still remain in the reliability of the light source. As for Ca atoms, the natural linewidth of the transition at 657 nm is as narrow as 400 Hz, and the hyperfine structural transition is affected only slightly by external fields. So, the research groups such as Physikalisch-Technische Bundesanstalt (PTB) in Germany and NIST have used the transition of Ca atoms at 657 nm as an optical frequency standard. However, in the field of frequency standards, it is also important to develop standards with different designs and compare mutually the output frequency of each standard to improve their reliability. In addition, establishing national standards for optical frequency is significant in related precision fundamental sciences.

This thesis consists of five chapters.

In chapter 1, the background, objectives and technological significance of this study along with the required performance of the excitation light source for use in frequency standards are described.

In chapter 2, the development of a laser diode light source for exciting  $\text{Sr}^+$  ions is described. To ensure long-term frequency stability, absorption line of the  $\text{Sr}^+$  ion at 421.671 nm in a hollow cathode lamp was used as the reference. In this study, the excitation light source was produced by second-harmonic generation (SHG) of the 844nm diode laser. The development of this system is the first attempt to lock the frequency of the diode laser at 844 nm to the frequency of the  $5s^2S_{1/2}-5p^2P_{1/2}$   $\text{Sr}^+$  ion transition.

In chapter 3, the development of an ion trap system for confining ions in a vacuum is described. The ion trap was designed a large size so that many ions are confined within the Lamb-Dicke region.  $\text{Sr}^+$  ions fluorescence from  $5s^2S_{1/2}$  to  $5p^2P_{1/2}$  is detected, and the temperature of the ions was estimated from the full width at half maximum of the fluorescence. As a result, the number of confined ions was in the range of  $2.5 \times 10^6 \sim 2 \times 10^7$ ,

the fluorescence intensity was 2000 c/s or higher, and the sufficient S/N for detecting double resonance fluorescence signals was realized. The elimination of the first-order Doppler shift was also realized in the 98% of the trapped ions. Since  $\text{Sr}^+$  ion has metastable state, when the intensity of the pumping laser becomes higher than a certain level, excited electrons are accumulated in the metastable state and fluorescence is not detected. Therefore, we observed the change in the fluorescence intensity using a pump-back laser and estimated the collisional quenching rate from the metastable state of the  $\text{Sr}^+$  ion. In addition, various buffer gases, such as He,  $\text{N}_2$  and  $\text{CH}_4$ , were introduced into the ion trap system and their effects on the fluorescence intensity were observed. As a result, it was clarified that He is highly effective in reducing the kinetic energy of trapped ions, and that it is the best buffer gas for improving frequency stability.

In chapter 4, the development of a light source for cooling Ca atoms at 423 nm is described, with the aim of realizing an optical frequency standard using the Ca transition at 657 nm. One of the major technical challenges in building a frequency standard is the generation of tunable single-frequency radiation near the cooling transition. We have developed a blue master/slave/slave laser for cooling Ca atoms. It consists of an infrared external cavity diode laser (master laser), a high-power solitary diode laser (slave laser), the frequency of which is locked to that of the master laser by an injection seeding technique, and  $\text{KNbO}_3$  crystals for SHG, which are mounted in a power build-up (enhancement) cavity. The light from the slave laser is injected into the  $\text{KNbO}_3$  crystals. By injecting the output light from SHG into a blue diode laser, a master/slave/slave laser light source in the blue-wavelength region was developed. The spectral linewidth of the injection-locked blue laser was approximately 0.4 MHz. It is concluded that since the natural linewidth of the cooling transition of Ca atoms at 423 nm is 34 MHz, the obtained output power and linewidth are sufficient to cool Ca atoms.

Chapter 5 is the conclusions, in which the data obtained from this study and the issues that remain to be solved are summarized.

# Study on Laser Diode Excitation of $\text{Sr}^+$ Ions and Ca Atoms for Frequency Standards

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# Chapter 1

## Introduction

### 1.1 Frequency Standards

In principle, there exists two different sets of systems for realizing a frequency standard: one is a quantum mechanical system utilizing atoms, molecules and ions, and the other is classical system like the astronomical clock, i.e. the earth orbit around the sun, or recent systems like pulsars and binary star systems. The observation of such astronomical clocks is a very interesting topic for astrophysical and fundamental physical reasons but probably not for future applications of defining the unit of time. Because of the quantum structure of matter, the indistinguishable reproduction of atoms, molecules and ions is the most important advantage of these systems and it is already applied for the present definition of second by Cs clock.

At present, the period of time known as the second is defined with reference to a microwave transition in atoms of cesium. The duration of 9 192 631 770 periods of the radiation corresponding to the transition between two hyperfine levels of the ground state of the cesium-133 atom. This definition is realized by a cesium atom beam clock or cesium atomic fountain scheme.

On the other hand, various atoms and ions are presented as future primary frequency standards. Frequency standards using different atomic species and different techniques will be needed in wide range of spectrum. The rf ion trap technique has been used in various field of study, such as ultra-high resolution spectroscopy and frequency standards. The trapped-ion candidates can achieve extremely narrow linewidths with high stability, and they have excellent prospects for extremely high accuracy. Neutral atom/molecule candidates have the potential for extremely high stability due to the large number of participating atoms/molecules, and they should achieve good accuracy as well. Due to their narrow linewidths and insensitivity to external perturbations, the intercombination lines of the alkaline earth atoms are among the most promising and practical neutral candidates.

Among various species, we chose  $\text{Sr}^+$  ion and Ca atom. Since their cooling and clock wavelengths are accessible with diode laser systems. This leads to the possibility of a future easy-to-handle, compact, transportable apparatus. In the field of frequency standards, diode lasers are used extensively. The advantage of using diode lasers in these applications are low cost, reliability, tenability, low amplitude noise, and ability to modulate at high frequencies. The choice of  $\text{Sr}^+$  ion was also due to the easy realization of the Lamb-Dicke region. Ca atom was selected since the intercombination transition  $^3\text{P}_1\text{-}^1\text{S}_0$  of atomic  $^{40}\text{Ca}$  at 657 nm is an excellent reference for the development of an optical frequency standard. The advantages result from its low natural linewidth of 400Hz, the

absence of hyperfine structure, the small influence of external fields on the transition frequency.

The international standard for time and frequency metrology is the Coordinated Universal Time (UTC) time scale maintained by the Bureau International des Poids et Mesures (BIPM) in Paris, France. The mission of the BIPM is to ensure international uniformity of measurements and traceability to the International System of Units (SI). The BIPM maintains a time scale known as International Atomic Time (TAI) based upon data from more than 200 atomic oscillators of 50 National Metrology Institute's (NMI) of the member states. Most of the oscillators are cesium based, but some hydrogen masers also contribute to TAI. Data from each contributing oscillator is submitted to the BIPM through the common view observations of GPS satellites. The scale unit of TAI is kept as close as possible to the SI second. As a service to end users, the NMIs distribute signals referenced to their UTC time scale thus complete the traceability chain.

## 1.2 Laser Cooling

Laser cooling techniques lead to considerable improvement in the frequency standards. A number of laboratories have developed novel frequency standards based on cold atoms<sup>1)</sup>. It is now evident that the use of laser-cooled atoms and ions will lead to orders of magnitude improvement in stability and accuracy for the next generation of frequency standards<sup>2,3)</sup>. Enhanced stability results primarily from the increased atom light interaction time provided by slow atoms, which leads to narrower transition linewidths. Increased accuracy results from narrower lines and the reduction of critical velocity-dependent systematic frequency offsets, such as the second-order Doppler shift. In the microwave region, cold atoms are the great success in Cs atomic fountain<sup>3)</sup>. In the optical domain, improvements due to laser cooling may prove even more significant. Indeed, over the past few years there has been excellent progress in the development of the next generation of optical frequency / wavelength standards, due in large part to the use of laser-trapped atoms and ions.

Laser cooling is a technique that uses light to cool atoms or ions to a very low temperature. The simplest form of laser cooling is the so-called Optical Molasses. This technique works by tuning the frequency of light slightly below an electronic transition in the atoms or ions (Fig.1.1). Because the light is detuned to the red of the transition, the atoms or ions will absorb more photons if they move towards the light source, due to the Doppler effect. By using counter propagating sets of laser beams in all three Cartesian coordinates, we get a force which drives the velocity of all the atoms or ions to zero. In this way, the atoms or ions are cooled. The lowest temperature one can reach with this technique is the so called Doppler temperature. This temperature limit is due to the fact that the light not only cools the atoms or ions, but also heats them. The light that is absorbed is emitted by spontaneous emission into a random direction. This means that the atoms or ions get a lot of random momentum kicks from the light, which causes heating.

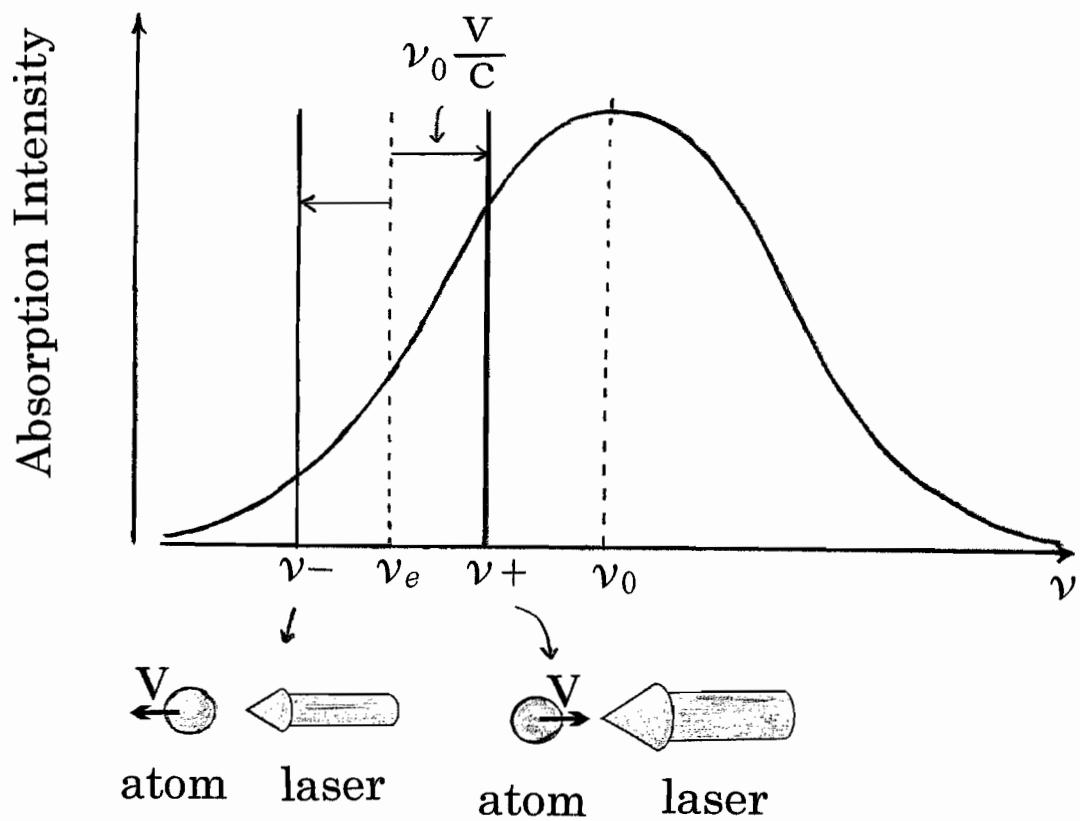


Fig. 1.1. Atomic absorption spectrum in the weak excitation limit. When an atom is moving with velocity  $v$  in the two counter propagating lasers with a frequency  $\nu_e$ , the frequencies felt by atom are  $\nu_e \pm \nu_0(v/c)$ . The photons of the laser which propagates in the opposite direction will be absorbed more frequently, which leads to net decelerating force.

The lowest temperature where these two opposing mechanisms balance each other is called Doppler temperature. The cooling process carries on until the Doppler-broadened width of absorption line is reduced to the natural linewidth  $\Gamma$ <sup>4)</sup>. A detailed calculation shows that the minimum kinetic energy achievable is  $E_{\text{min}} = \hbar \Gamma/4$ <sup>5)</sup>. This is known as the Doppler cooling limit.

### 1.3 Lamb-Dicke Regime

If a single ion is tightly confined such that the amplitude of its motion is less than the wavelength of the probing radiation, the first-order Doppler width of the probing transition is effectively eliminated<sup>6)</sup>. With the Doppler broadening removed, the very narrow natural linewidth of the transition can be observed. This can be simply seen for a one dimensional model. Fig. 1.2 shows the one dimensional model. The ion is interrogated by microwave radiation with an electric field

$$E(t) = E_0 \sin(\omega_0 t - kx) \quad 1.1)$$

where  $\omega_0$  is the angular frequency,  $t$  is the time,  $k$  is the radiation wave vector,  $x$  is the coordinate.

The motion of the ion can be given by

$$x = (a/2) \cos \Omega t \quad 1.2)$$

where  $x$  is the ion's coordinate,  $a/2$  is the half amplitude of the ion's motion,  $\Omega$  is the angular frequency of the ion.

Therefore, the ion experiences an electric field

$$E(t) = E_0 \sin(\omega_0 t - k(a/2) \cos \Omega t) \quad 1.3)$$

which can be rewritten in terms of Bessel functions. Substituting  $m = ka/2$  into 1.3) we obtain

$$\begin{aligned} E(t) = E_0 [ & J_0(m) \sin \omega_0 t \\ & - J_1(m) \{ \cos(\omega_0 + \Omega)t + \cos(\omega_0 - \Omega)t \} \\ & - J_2(m) \{ \cos(\omega_0 + 2\Omega)t + \cos(\omega_0 - 2\Omega)t \} \\ & \dots\dots\dots ] \quad 1.4) \end{aligned}$$

The spectrum obtained from such a confined ion consists of a main resonance at  $\omega = \omega_0$  with equally spaced sidebands at  $\pm n\Omega$ . The relative strengths of these components are given by the  $[E_0 J_n(m)]^2$ . Hence the modulation index  $m$  characterizes the confinement

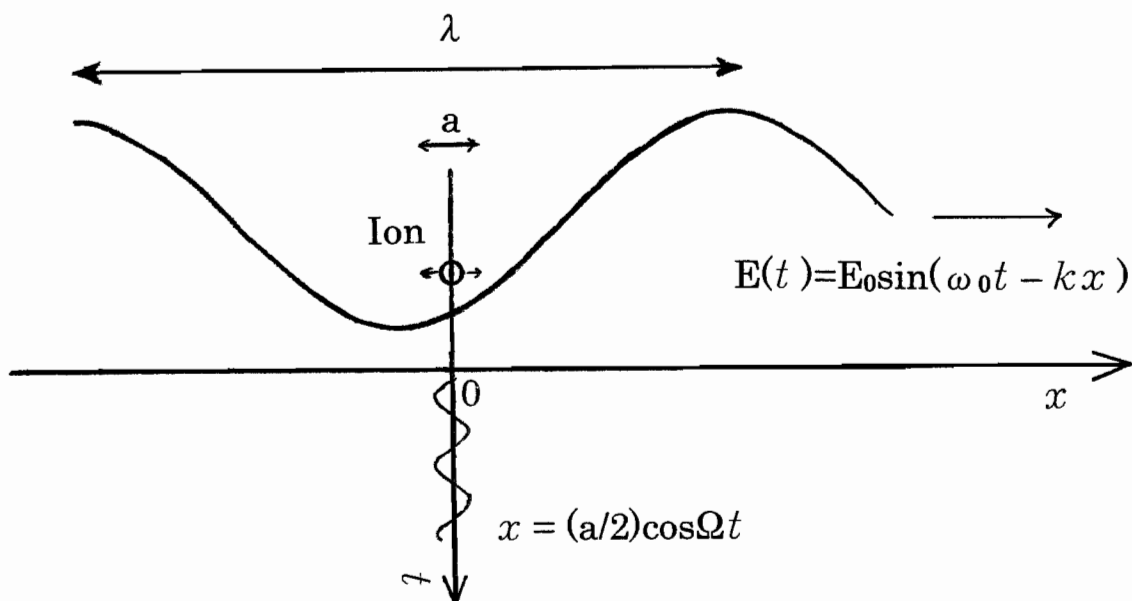


Fig. 1.2. One dimensional model of the Lamb-Dicke region. The motion of the ion can be given by  $x = (a/2) \cos \Omega t$ . If a single ion is tightly confined such that the amplitude of its motion is less than the wavelength of the probing radiation, the first-order Doppler width of the probing transition is effectively eliminated.

of the ion. For  $m \gg 1$  there are many sidebands. However for  $m \leq 1$  the only significant term is that for  $m = 0$ ; that is, a single resonance. This is known as the Lamb-Dicke regime. The first order Doppler width of the resonance is effectively removed leaving a profile dominated by a main carrier at  $\omega = \omega_0$  and a few weak sidebands at multiples of  $\Omega$  from the carrier. The long wavelength of the radiation means that the Lamb-Dicke criterion is easily satisfied. In the case of radiation at 5 GHz and radius of confinement is 5 mm, the value  $m$  becomes 0.26. The intensity ratio of main carrier becomes 0.98.

## 1.4 Laser Microwave Double Resonance Technique

Resonance absorption of a microwave photon is difficult to detect directly because the energy is too small and the transition rate is too low. The combination of laser spectroscopic technique with rf spectroscopy method, however, the microwave photon is, so to speak, converted to a visible photon which has sufficient energy to be detected; the mechanism can be understood as a quantum amplification of  $10^6$  in energy. The detection of the microwave transition does not rely on the small absorption of the microwave but can use the higher sensitivity of optical detectors. Therefore the optical-rf double-resonance method has now become a very powerful technique for high-precision measurements of fine or hyperfine splitting in atoms and ions.

This can be seen for a simple model. In Fig. 1.3 we consider two different states  $^2S_{1/2}$  and  $^2P_{1/2}$ .  $^2S_{1/2}$  may be split into closely spaced sublevels  $F=4$  and  $F=5$ . A narrow band laser which is tuned to the transition  $F=4$  and  $^2P_{1/2}$  selectively depletes the level  $F=4$  and increases the population of the level  $F=5$ . If the optically pumped sample is placed inside an rf field with the frequency  $\omega_{rf}$  tuned into resonance with the transition  $F=5 \rightarrow F=4$  the level population  $F=4$  which had been depleted by optical pumping, will increase again. This leads to an increased absorption of the optical pump beam, which may be monitored by corresponding increase of the laser-induced fluorescence intensity.

## 1.5 Organization of Dissertation

This thesis consists of five chapters.

In chapter 1, the background, objectives and technological significance of this study along with the required performance of the excitation light source for use in frequency standards are described.

In chapter 2, the development of a laser diode light source for exciting  $\text{Sr}^+$  ions, which are used as a microwave frequency standard, is described. To ensure long-term frequency stability, the transition frequency of the  $\text{Sr}^+$  ion in a hollow cathode lamp is used as the reference.

In chapter 3, the development of an ion trap system for confining ions in a vacuum for a long time is described. In order to confine a large number of ions within in the

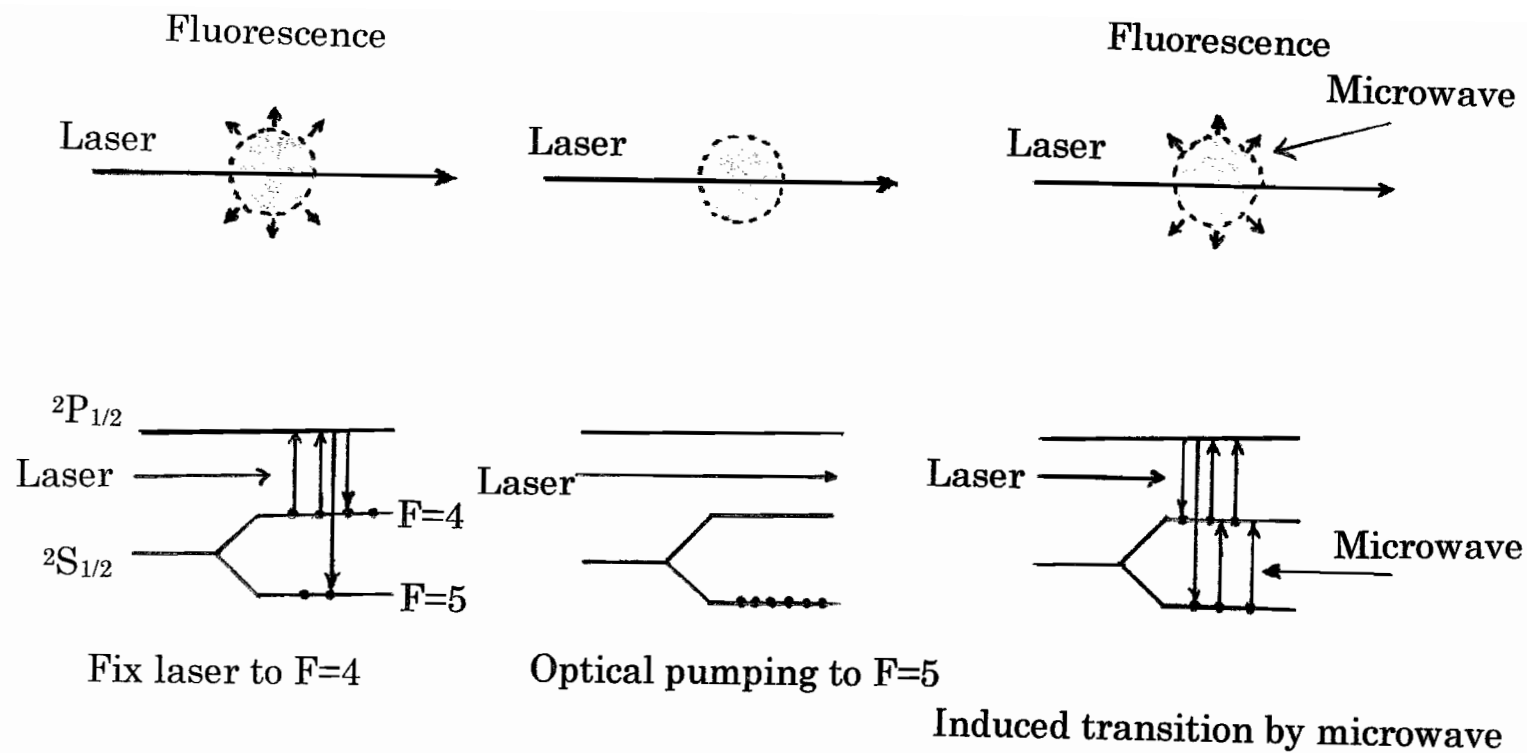


Fig. 1.3. Optical radio-frequency double resonance scheme.

Lamb-Dicke region and to obtain a good signal-to-noise ratio (S/N), the ion trap is fabricated by using a relatively large electrode. Since  $\text{Sr}^+$  ions can be metastable, when the intensity of the pumping laser becomes higher than a certain level, excited electrons are accumulated in the metastable state and fluorescence is not detected. Therefore, we observe the change in the fluorescence intensity using a pump-back laser and estimate the collisional quenching rate from the metastable state of the  $\text{Sr}^+$  ion. In addition, various buffer gases, such as He,  $\text{N}_2$  and  $\text{CH}_4$ , are introduced into the ion trap system and their effects on the fluorescence intensity are observed. In Fig. 1.4 the position of Chapter 2 and Chapter 3 in the microwave frequency standard is illustrated, where the required performance for each system are also summarized.

In chapter 4, the development of a light source for cooling Ca atoms at 423 nm is described, with the aim of realizing an optical frequency standard at 657 nm. A blue master/slave/slave laser is developed for cooling Ca atoms. The developed experimental set-up consists of an infrared external cavity master laser diode, a high-power solitary slave laser diode,  $\text{KNbO}_3$  crystals for SHG, and a blue diode laser diode. In Fig. 1.5 the position of Chapter 4 in the optical frequency standard is illustrated, where the required performance for each system are also summarized.

Chapter 5 is the conclusions, in which the results obtained from this study and the subjects to be solved are summarized.

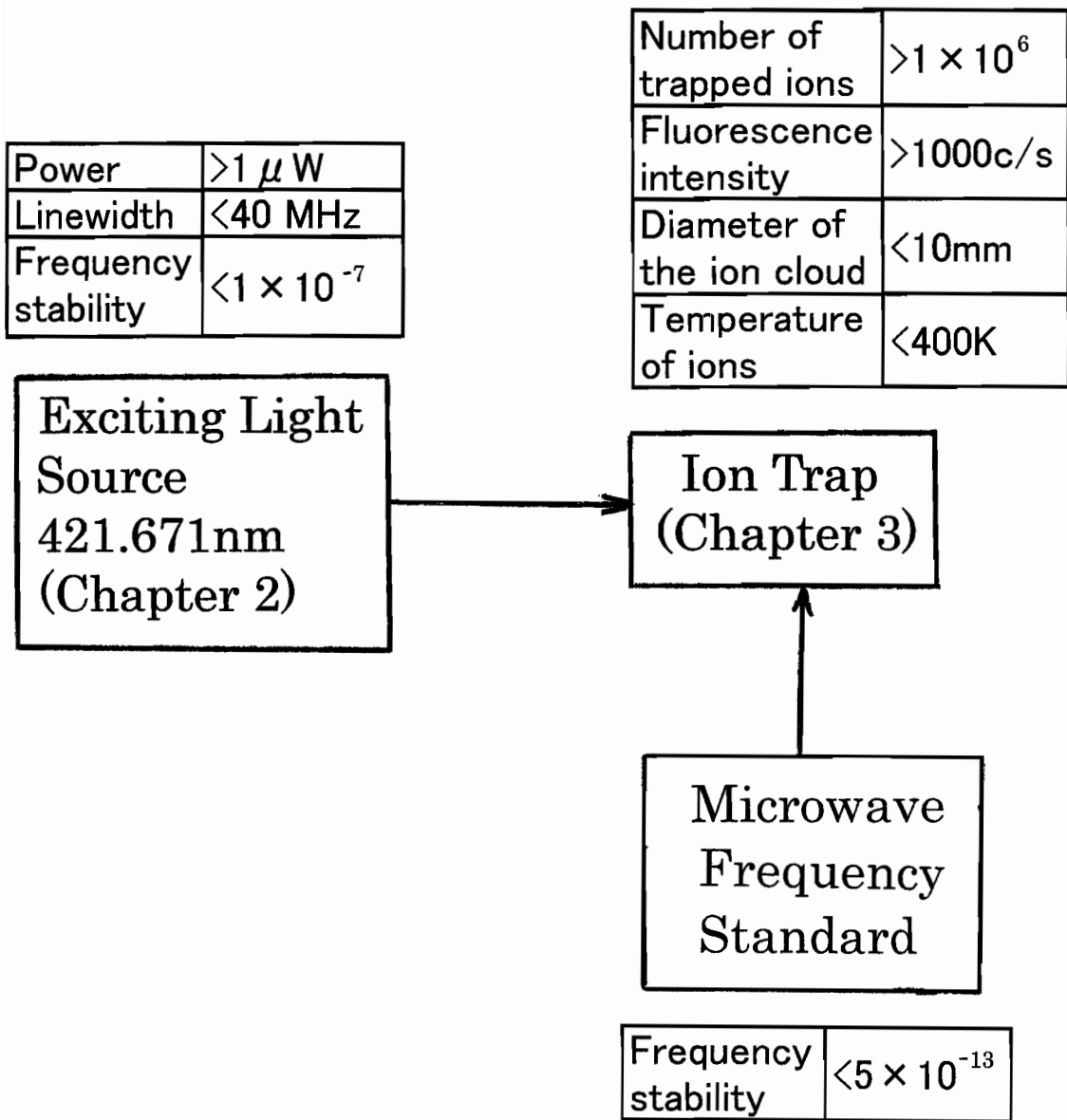


Fig. 1.4.  $\text{Sr}^+$  ions microwave frequency standard system and its required performances.

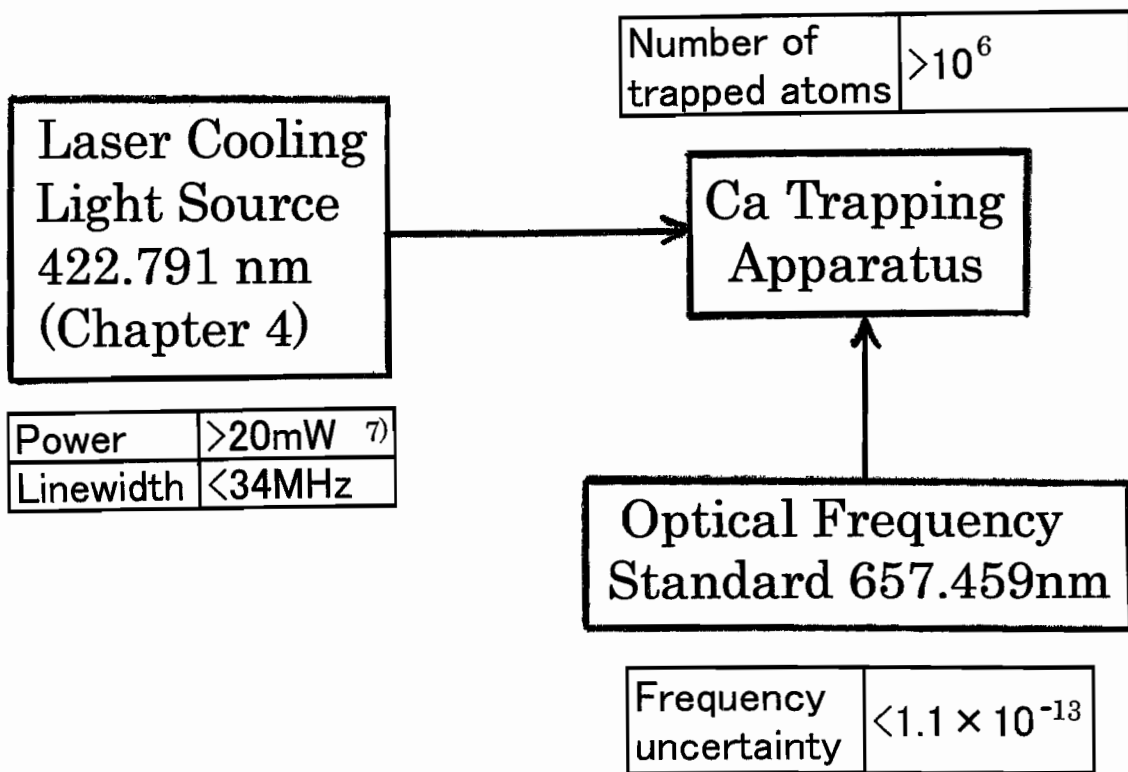


Fig. 1.5. Ca atoms optical frequency standard system and its required performances.

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# Chapter 2

## 2. Laser Diode Source for Exciting $\text{Sr}^+$ Ions

### 2.1 Introduction

Frequency standards in microwave region based on high precision measurement of hyperfine transition of trapped ions are realizable. An ion-trap technique combined with a laser-microwave double resonance spectroscopy has a great advantage in precision and sensitivity to measure the hyperfine structure of ground state ions. Among various species, we chose  $\text{Sr}^+$  ion for microwave frequency standard, since its cooling wavelength is accessible with diode laser system. This leads to the possibility of a future easy-to-handle, compact, transportable apparatus. In the system of frequency standards in the microwave range, in addition to  $\text{Sr}^+$  ions,  $\text{Hg}^+$  ions are being studied at the National Institute of Standards and Technology (NIST) in the United States and  $\text{Yb}^+$  ions are being studied at the National Measurement Laboratory (CSIRO) in Australia. However, since the wavelengths of optical pumping are 194 nm for  $\text{Hg}^+$  ion and 369 nm for  $\text{Yb}^+$  ion, there are a lot of problems still remain in the reliability of the light source.

The choice of  $\text{Sr}^+$  ion was also due to the easy realization of the Lamb-Dicke region in the microwave region. If ions are confined within the Lamb-Dicke region, the first order Doppler width of the probing transition is effectively eliminated.

In this chapter, the development of a laser diode light source for exciting  $\text{Sr}^+$  ions, which are used as a microwave frequency standard, is described. To ensure long-term frequency stability, absorption line of the  $\text{Sr}^+$  ion at 421.671 nm in a hollow cathode lamp was used as the reference. Studies have been published on using the Fabry-Perot interferometer as an external frequency reference; however, this method is vulnerable to long-term drift due to fluctuations in the length of the Fabry-Perot cavity. In this study, the excitation light source was produced by injecting light from a high-power laser diode into  $\text{KNbO}_3$  crystals and locking the output light obtained by second-harmonic generation (SHG) to the absorption line of the  $\text{Sr}^+$  ion in a hollow cathode lamp. The development of this system is the first attempt to lock the frequency of the diode laser at 844 nm to the frequency of the  $5s^2S_{1/2}-5p^2P_{1/2}$   $\text{Sr}^+$  ion transition.

### 2.2 Second Harmonic Generation

In order to develop the light source for exciting  $\text{Sr}^+$  ions, it is necessary to obtain the specific desired wavelength. The second-harmonic generation (SHG) of high-power output from the semiconductor laser was utilized. We used  $\text{KNbO}_3$ , since it possesses the largest

nonlinear coefficients for wavelength conversion of all commercially available inorganic materials. Strong, natural birefringence ( $n_e \cdot n_o = 0.22$ ), combined with high nonlinearity make  $\text{KNbO}_3$  one of the most versatile materials for Type 1 Second Harmonic Generation (SHG). The nonlinear coefficients of  $\text{KNbO}_3$  crystals are 3 times as large as those of KTP crystals.  $\text{KNbO}_3$  is widely used for the generation of red-green-blue laser light via SHG, tunable near infrared (NIR) light via OPO/OPA, and also for electro-optic and photo-refractive studies. Efficient angle and temperature-tuned wavelength conversion is possible throughout the optical bandwidth from  $0.4 \mu\text{m}$  to  $4.7 \mu\text{m}$ .

The theoretical SHG output power is given by<sup>1,2)</sup>

$$P_{2\omega} = (2\omega^3 d_{32}^2 / \pi \epsilon_0 n_\omega^2 c^4) P_\omega^2 l \quad (2.1)$$

Here,  $P_{2\omega}$  is the SHG output power,  $P_\omega$  is the output power at the laser fundamental mode,  $d_{32} = 21 \times 10^{-12} \text{m/V}$  is the crystal's nonlinear optical constant,  $\epsilon_0$  is the permittivity of free space,  $n_\omega$  is the index of refraction of  $\text{KNbO}_3$ ,  $c$  is the velocity of light,  $l$  is the length of the crystal, and  $\omega = 2\pi c / \lambda$ , where  $\lambda = 843 \text{ nm}$  is wavelength of the fundamental mode.

The SHG experimental setup is shown in Fig. 2.1. 461.671 nm light is generated by focusing a high power semiconductor laser (SDL5410-G1) onto a  $\text{KNbO}_3$  crystal. A thin glass plate is placed 2 mm in front of the laser's front facet, so that light weakly reflected off the plate is fed back to the semiconductor laser. The glass plate is attached to a piezoelectric ring. Changes in a bias voltage applied to this ring generate shifts in the position of the plate. This arrangement makes it possible to select longitudinal modes of the semiconductor laser<sup>3)</sup>. The laser beam was collimated by an aspheric lens ( $f = 6.24 \text{ mm}$ , 0.4 NA). The full width and half maximum of the laser output emission line were 5 MHz and 46 mW, respectively.

The standard method of wavelength selection is to place a diffraction grating external to the semiconductor laser and return the diffracted light to the laser. In this work, however, we chose the method described above. One advantage of our method is that power losses in the semiconductor laser are low. If we compare the performance of a setup based on our monolithic chip with regard to optical feedback and power extraction with that of setups based on an external diffraction grating, we find that the performance of a Littrow cavity is 67% and the performance of a Littman cavity<sup>4)</sup> ranges from 50% to 100%. After passing through an optical isolator, the laser beam is focused onto a  $\text{KNbO}_3$  crystal by a lens with a 125 mm focal length. The  $\alpha$ -cut  $3 \times 3 \times 10 \text{ mm}$   $\text{KNbO}_3$  crystal is placed at the center of a vacuum chamber in order to prevent condensation of moisture. In order to obtain phase matching in the crystal, we mounted it on top of a Peltier element, which was placed in a slot cut into a copper heat sink. It is well known that good results are obtained when the plane of polarization of the laser is oriented so that the fundamental wave follows the  $\alpha$ -axis<sup>1)</sup>.

In order to obtain the optimum temperature value for the doubling crystal, it is necessary to measure the temperature tuning curve. Since the sensitivity of the second harmonic power to the temperature variations of the crystal is very sharp. The SHG power

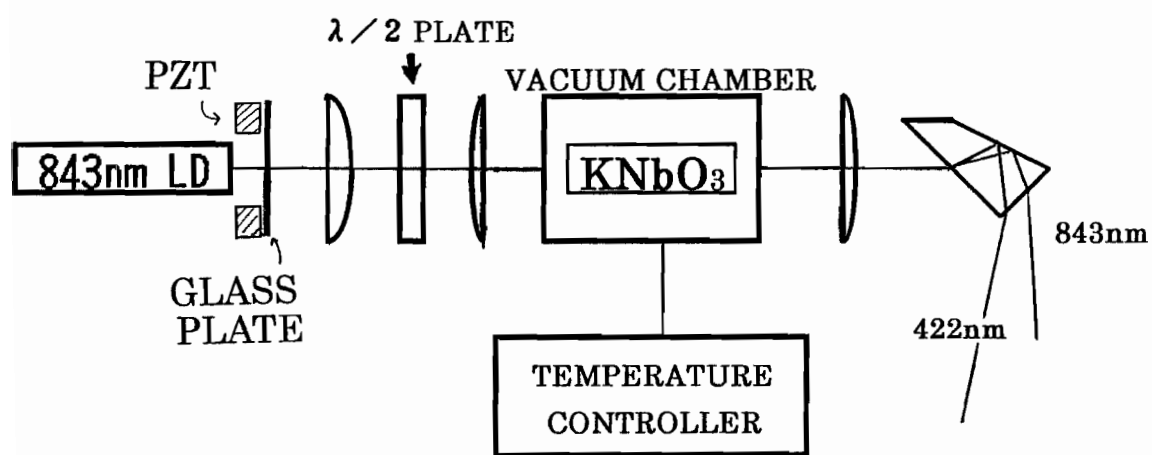


Fig. 2.1. SHG experimental set-up.

drops sharply when the temperature of the  $\text{KNbO}_3$  increases or decreases from the optimum value. SHG is obtained when the temperature of the cooled crystal reaches  $-22.93^\circ \text{C}$ . The rate of change of the SHG output frequency with the laser injection current is  $7.5 \text{ GHz/mA}$ . The temperature dependence of the SHG power is shown in Fig. 2.2. When the oscillation wavelength of the semiconductor laser is  $844.704 \text{ nm}$ , at which the output power is  $86.1 \text{ mW}$ , the power incident on the crystal is  $81 \text{ mW}$  and a SHG output power of  $61 \mu\text{W}$  is obtained. (In the fluorescence measurements, because the absorption line wavelength corresponding to the  $\text{Sr}^+$  ion is  $421.671 \text{ nm}$ , it is necessary to tune the fundamental mode wavelength of the laser to  $843.342 \text{ nm}$ . When this is done by controlling the laser injection current and temperature, the resulting output power of the fundamental mode is  $46 \text{ mW}$ ). For the length of crystal we used, i.e.,  $10 \text{ mm}$ , Eq. (1) gives a computed value of  $591 \mu\text{W}$  for  $P_{2\omega}$ . On the other hand the actual output power was  $62 \mu\text{W}$ . The measured dependence of the second-harmonic power on  $843 \text{ nm}$  incident power is shown in Fig. 2.3. It agrees with the general  $P_{\omega}^2$  dependence. In our view, the reason why the experimental value is lower than this value is incomplete alignment of the crystal axis and imperfect material quality, which causes the incident beam shape to approach that of an ellipse.

## 2.3 Frequency Stabilization

In order to stabilize the frequency of a CW tunable LD to the center of the required transition frequency of ions, it is valuable to use the absorption line of a hollow-cathode lamp. Y. C. Chung and R. W. Tkach reported the frequency stabilization of a  $1.3 \mu\text{m}$  DFB laser to an argon line using the optogalvanic effect<sup>5)</sup>. M. Musha et al. detected an optogalvanic signal of the  $5s^2S_{1/2}-5p^2P_{1/2}$  transition of  $^{88}\text{Sr}^+$  ion<sup>6)</sup>. We used the absorption signal of a hollow-cathode lamp. It is easier to use differential detection than the optogalvanic effect, since the former method directly detects the absorption signal before the electrical signal goes to the complicated apparatus. This is important to initially determine the optimum parameter of the laser diode. Hollow-cathode discharge tubes are commercially available for nearly all the stable elements. These devices normally consist of a gas discharge such as argon or neon and a cathode made of the element of interest. The element is introduced into the discharge by sputtering from the cathode walls.

Preparatory to frequency stabilization it was necessary to observe the absorption signal of  $\text{Sr}^+$  ions in a hollow-cathode lamp. By observing the absorption signal, we can estimate the line width of the Doppler profile of  $\text{Sr}^+$  ions in a hollow-cathode lamp. From the line width (FWHM) of the Doppler profile, the modulation depth of the semiconductor laser can be estimated. The increase of the modulation depth of the semiconductor laser results in the increase of the intensity of the first-derivative signal. The intensity of the first-derivative signal becomes maximized when the modulation depth of the semiconductor laser is in accordance with the line width of the Doppler profile. After the modulation depth exceeds this value the intensity of the first-derivative signal decreases. From this we can adjust the modulation current of the semiconductor laser to obtain the

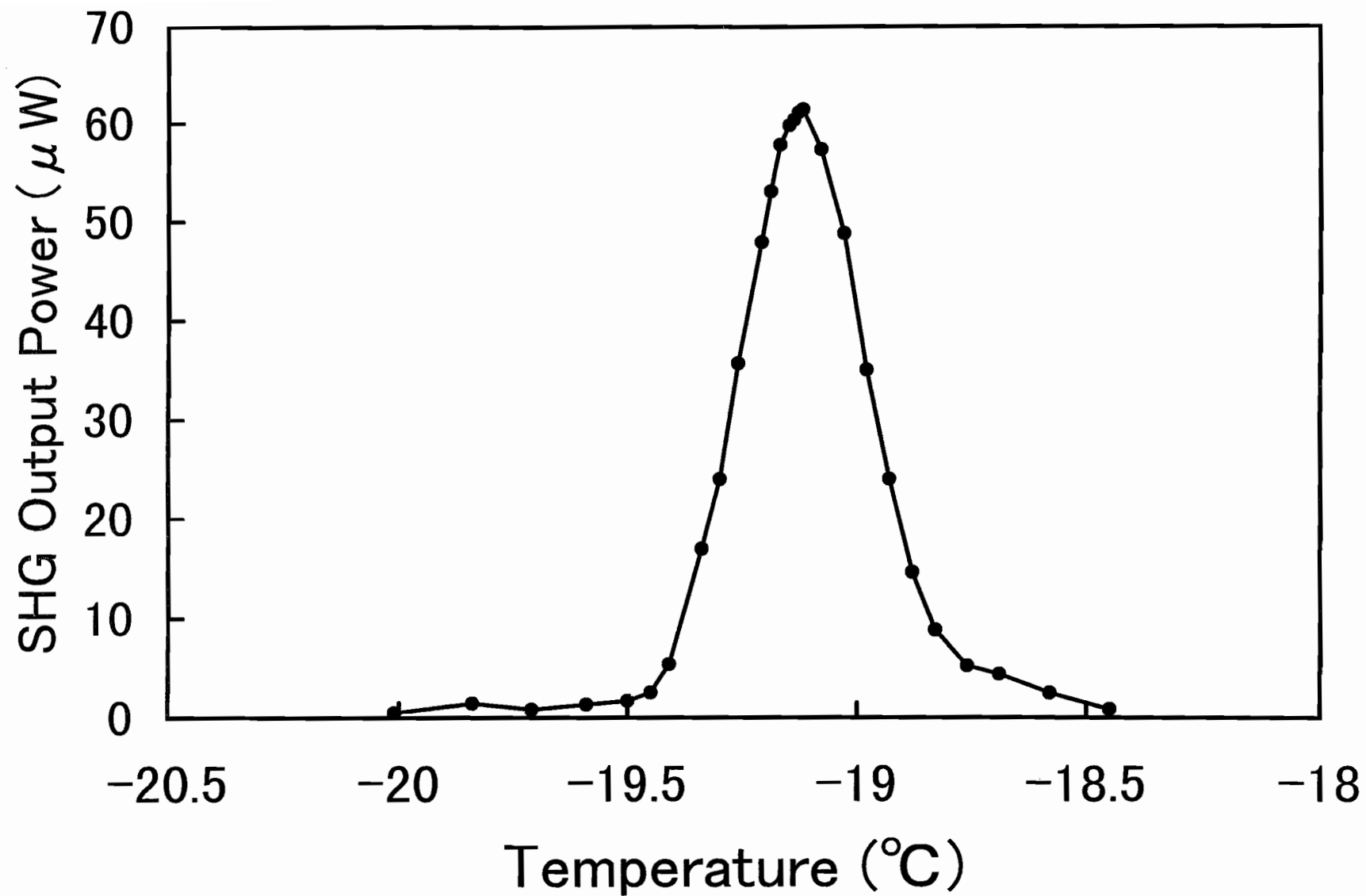


Fig. 2.2. Temperature tuning characteristics of second-harmonic output power.

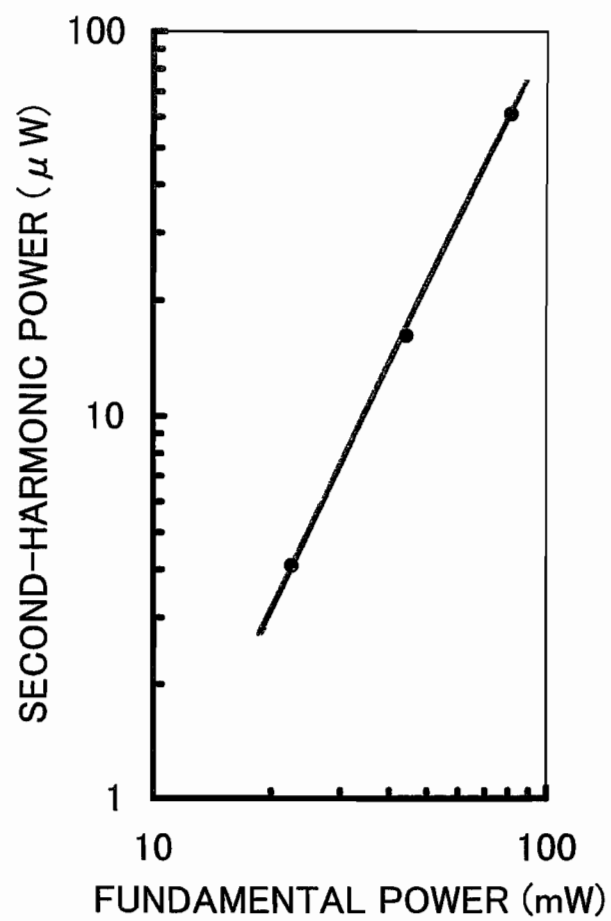


Fig. 2.3. Measured dependence of the second-harmonic power on 843 nm incident power.

good control signal for frequency stabilization. The experimental set-up is shown schematically in Fig. 2.4. In order to match the semiconductor laser wavelength to the absorption line of the  $\text{Sr}^+$  ion, it is necessary to adjust the temperature and injection current. For an injection current of 84 mA, an output power of 46 mW is obtained at an oscillation wavelength of 843.342 nm, which leads to an SHG output power of 5  $\mu\text{W}$ . In order to match the SHG wavelength of 421.671 to the absorption line corresponding to the  $5s^2S_{1/2}-5p^2P_{1/2}$  transition of the  $\text{Sr}^+$  ion, we first use a wavelength meter to monitor the wavelength of the fundamental mode. Although optical source technology for second harmonic generation generally calls for placing the nonlinear crystal in a resonator in order to resonantly amplify the fundamental mode<sup>7)</sup>, our experiments involve only fluorescence detection, so that we use the simple single-pass method for reasons of simplicity. We used a differential amplifier to detect the SHG, in order to increase sensitivity. It is necessary to place an optical filter at the photodetector front end in order to eliminate large amounts of light from sources such as luminescence from the hollow-cathode lamp (Hamamatsu photonics L2783), which span a wide frequency range compared to the SHG. In order to equalize light intensities at the front ends of the photodetectors in the absorption line detection and reference light detection arms respectively, we adjusted the intensity by rotating the Glan-Thompson polarizer in the hollow-cathode lamp path. In order to set the discharge current of the hollow-cathode lamp to 20 mA, we used a 30 k $\Omega$  ballast resistor. Before the frequency stabilization is carried out, it is necessary to detect the  $\text{Sr}^+$  ion absorption line using the hollow cathode lamp. Figure 2.5 (a) shows the observed absorption line, in which we observe a dip due to absorption by the ions. The hollow cathode lamp gives rise to an absorptivity of 0.022 for the laser beam. The full width at half power of the absorption line is 1.4 GHz. In trace (b) we have introduced a chopper in order to eliminate optical shot noise. The signal was detected at the chopping frequency using a lock-in amplifier. The chopping frequency was 175 Hz. The time constant of the lock in amplifier was 10 s.

The advantage of the derivative spectroscopy with a frequency modulation of the laser is the possibility for phase sensitive detection, which restricts the frequency response of the detection system to a narrow frequency interval centered at the modulation frequency. Frequency independent background absorption from cell windows and background noise due to the levels of fluctuation of the laser intensity or of the density of absorbing ions are essentially reduced. Regarding the signal to noise ratio and achievable sensitivity, the frequency modulation technique is superior to an intensity modulation of the incident radiation. The frequency of a single mode laser can readily be modulated when an AC voltage is applied to the DC voltage of the laser diode. A diagram of frequency stabilization experimental set-up is shown in Fig. 2.6<sup>8)</sup>. The frequency stability of the SHG was estimated from the error signal of the lock-in amplifier. In order to obtain a zero-crossing error signal, which provided a frequency discriminator slope, the injection current of the diode laser was modulated and thus the frequency of the SHG was modulated. The change in the absorptivity produces a change in the transmitted light intensity, which we detected by scanning the frequency of the laser light over the range of

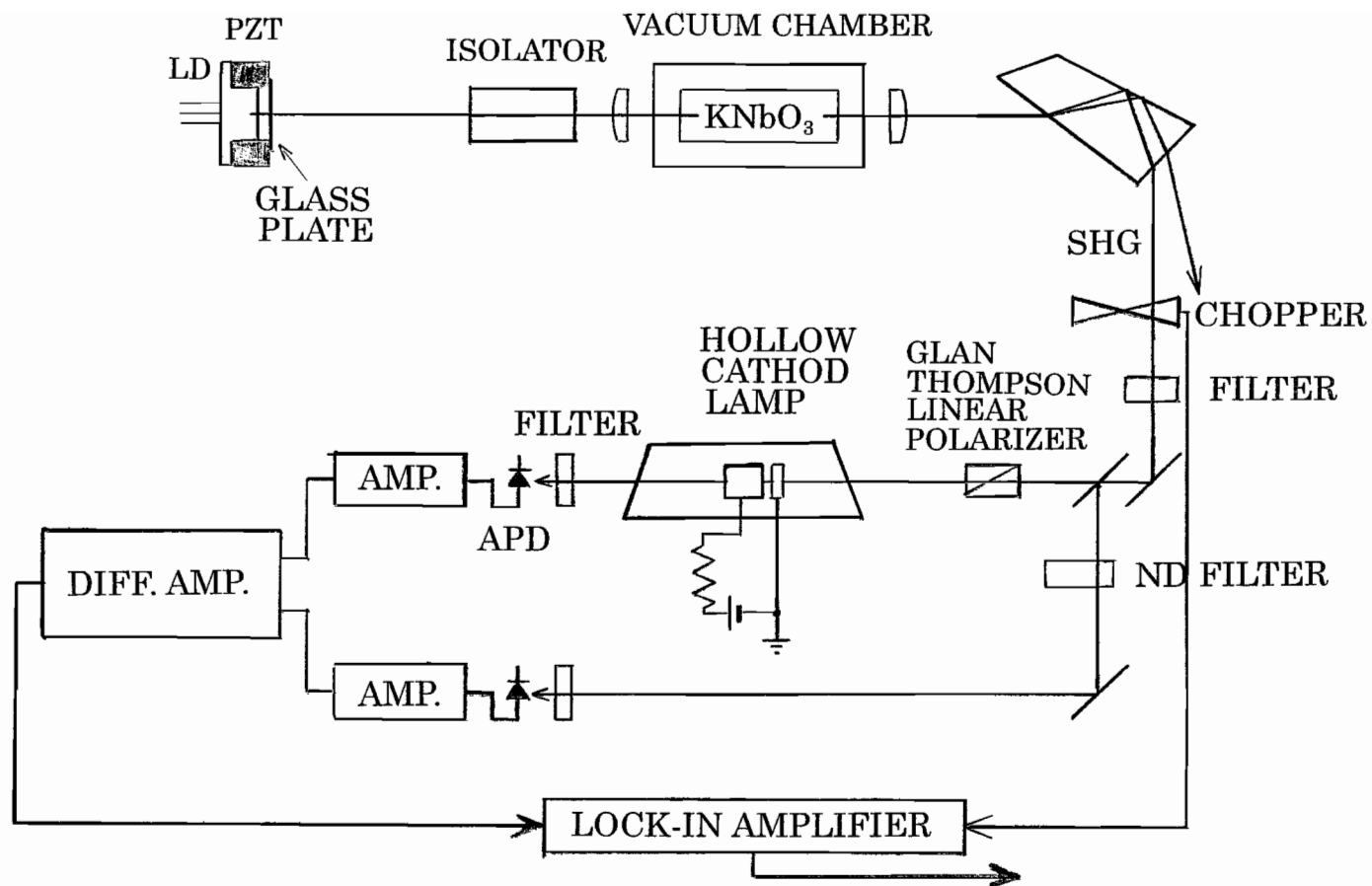


Fig. 2.4. Experimental set-up to observe the absorption signal of  $\text{Sr}^+$  ions.

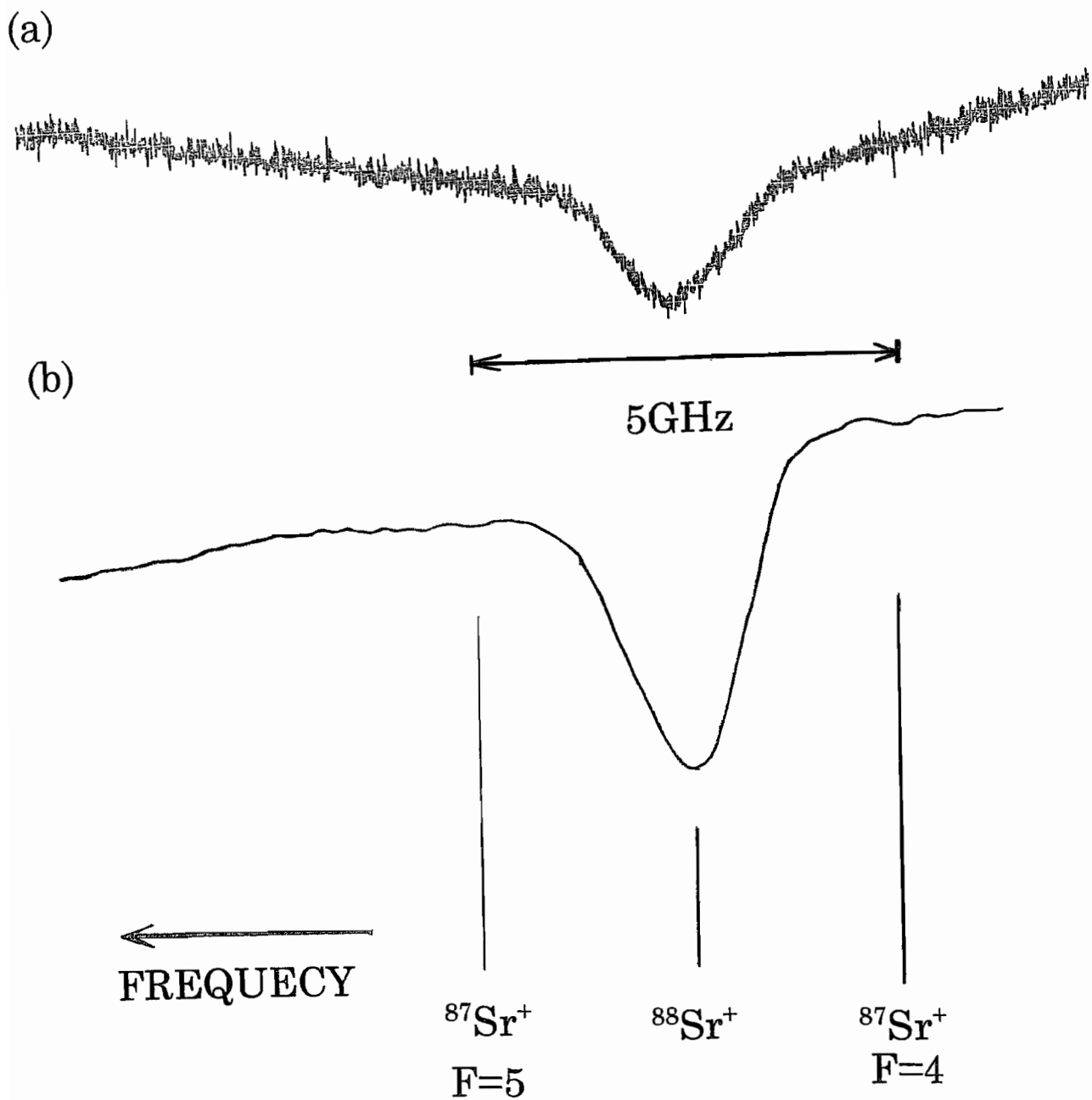


Fig. 2.5. (a) Absorption line of  $^{88}\text{Sr}^+$  ions in hollow-cathode lamp. (b) shows reduction of the influence of shot noise by introduction of a chopper. The chopping frequency was 175 Hz, and the time constant of the lock-in amplifier was 10 s.

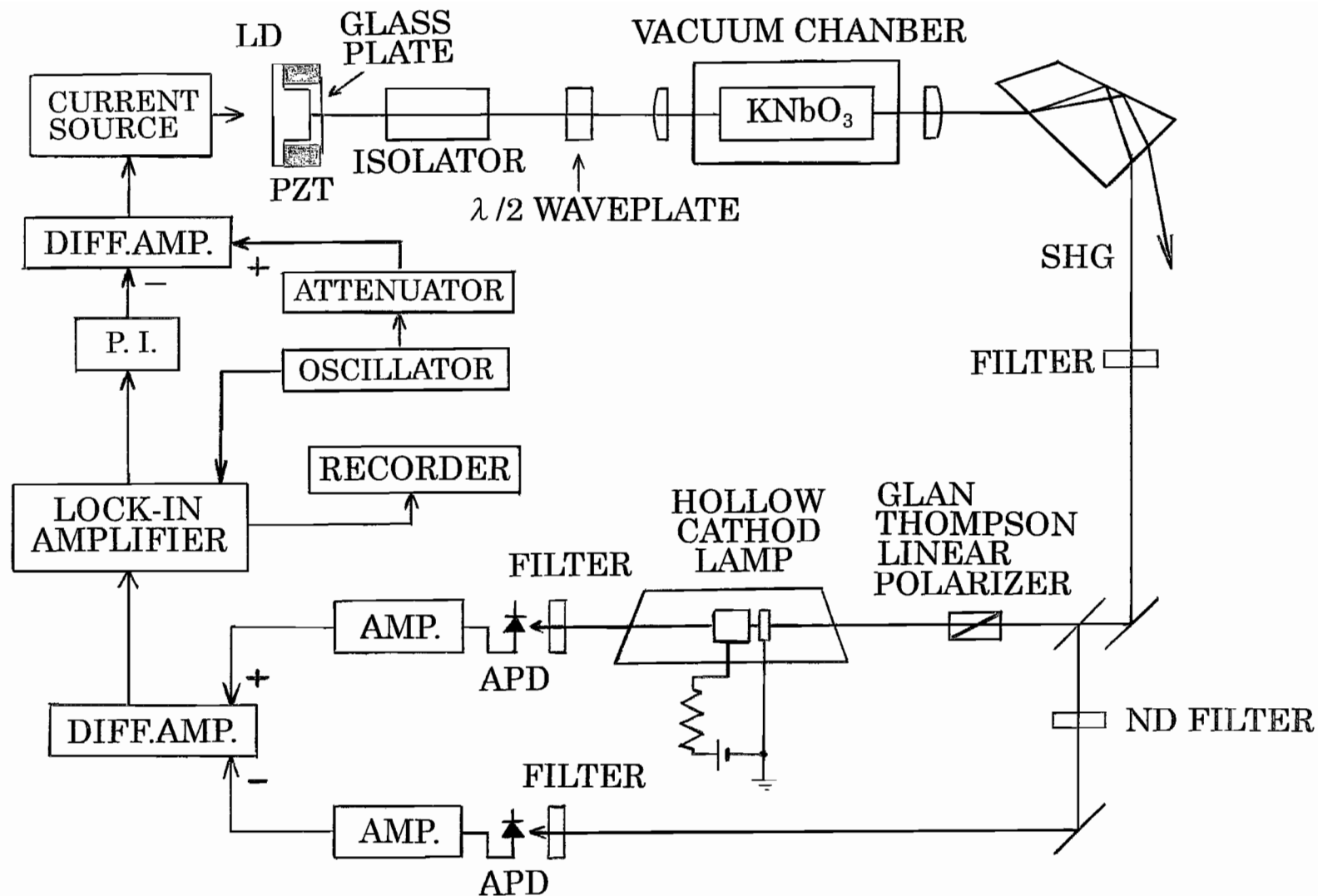


Fig. 2.6. Experimental setup for frequency stabilization. The letters P and I denote proportional amplifier and integrator, respectively.

the absorption spectrum. By performing synchronous detection with a lock-in amplifier and symmetrizing about the center frequency of the absorption spectrum, we obtain an inverted electronic signal. By adding the 41 Hz signal from an oscillator to the SHG, which was frequency-modulated at the semiconductor laser with a modulation depth of 480 MHz, a first-derivative signal was obtained by detection in a differential lock-in amplifier. In Fig. 2.7 we show the trace of the first derivative of the absorption line. Because the power output at the second harmonic that was usable for photodetection was below 1  $\mu$ W, an avalanche photodiode was used as the photodetector in order to increase the amplification factor. In addition, the op-amp we used was chosen for low noise and high temperature stability. A sweep was imposed on the current to the semiconductor laser in order to use this signal for frequency stabilization by the method of zero-crossing frequency discrimination.

By using the linear portion of the first-derivative signal, we locked the laser frequency to this transition by means of negative feedback. The proportional and integral control was used. The proportional gain was 5.6 and the integral time was 0.69 s. In Fig. 2.8 we show the error difference signal of the lock-in amplifier when the frequency was stabilized by the  $\text{Sr}^+$  ion absorption line. Before the control was imposed, the frequency variation was 100 MHz, but once the control was turned on the frequency variation did not exceed 10 MHz<sup>9)</sup>. Although other researchers<sup>5,6)</sup> have used methods such as the optogalvanic effect to lock the oscillation wavelength of the laser to the hollow-cathode lamp absorption line, in our experiments we were able to detect the locking signal with good sensitivity by using the difference between the absorption line and the laser line.

Considering the Doppler line width of the  $\text{Sr}^+$  ions ( more than 1 GHz ) which were confined in an rf ion trap, this frequency fluctuation affected less than  $7 \times 10^{-4}$  fluorescence intensity of  $\text{Sr}^+$  ions. In order to study the effect of laser linewidth on the Doppler width in the optical region of the fluorescence signal, theoretical calculation of induced transition rate was performed in the Appendix. In our case the line-width of SHG was 10 MHz, we could admit that the linewidth of the laser had negligible effect on the Doppler width in the optical region of the  $\text{Sr}^+$  ions.

## 2.4 Conclusion

In this chapter, the development of a laser diode light source for exciting  $\text{Sr}^+$  ions is described. The frequency of a SHG of a high power 844nm laser diode has been stabilized to the absorption line of  $^{88}\text{Sr}^+$  ion at 421.671 nm in a hollow cathode lamp. The frequency fluctuation never exceeded 10MHz, which is much narrower than the Doppler line width of the  $\text{Sr}^+$  ions (more than 1 GHz) confined in an rf ion trap. To our knowledge, the development of this system is the first demonstration of the frequency stabilization of the 844 nm laser light to the  $5s^2S_{1/2}-5p^2P_{1/2}$  transition line of  $^{88}\text{Sr}^+$  ions at 421.671 nm. As a result, more than 1 $\mu$ W of SHG can be used to fluorescence of  $\text{Sr}^+$  ions, which is enough to deplete one of the hyperfine levels of the ground state of  $^{87}\text{Sr}^+$  ions which for laser microwave double resonance.

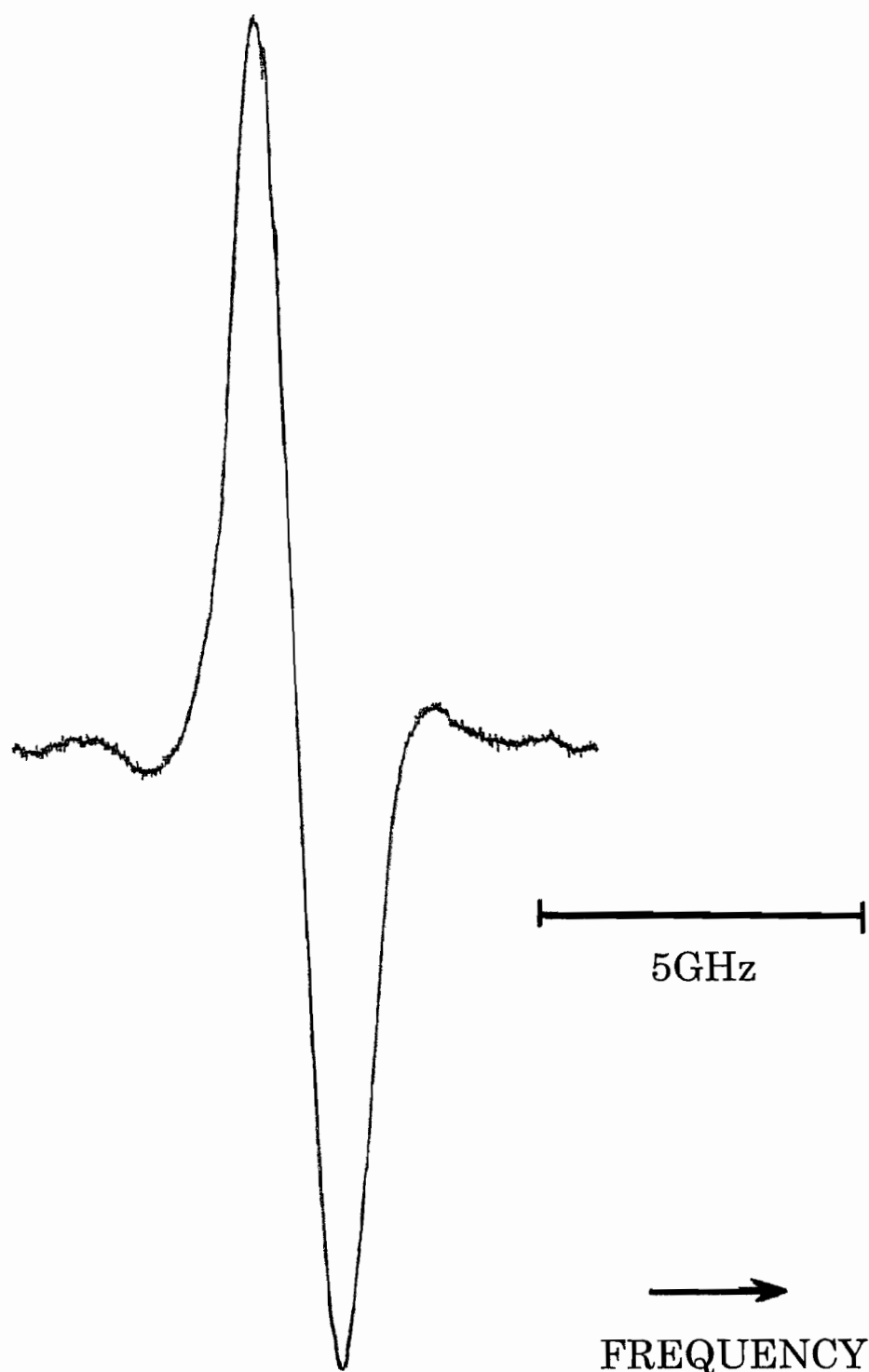


Fig 2.7. Signal corresponding to first derivative of the absorption line of  $^{88}\text{Sr}^+$  ions in a hollow-cathode lamp. The second-harmonic output power was modulated by modulating the injection current to the semiconductor laser. The second-harmonic modulation frequency and modulation depth were 41 Hz and  $\pm 480$  MHz, respectively. The time constant of the lock-in amplifier was 100 ms.

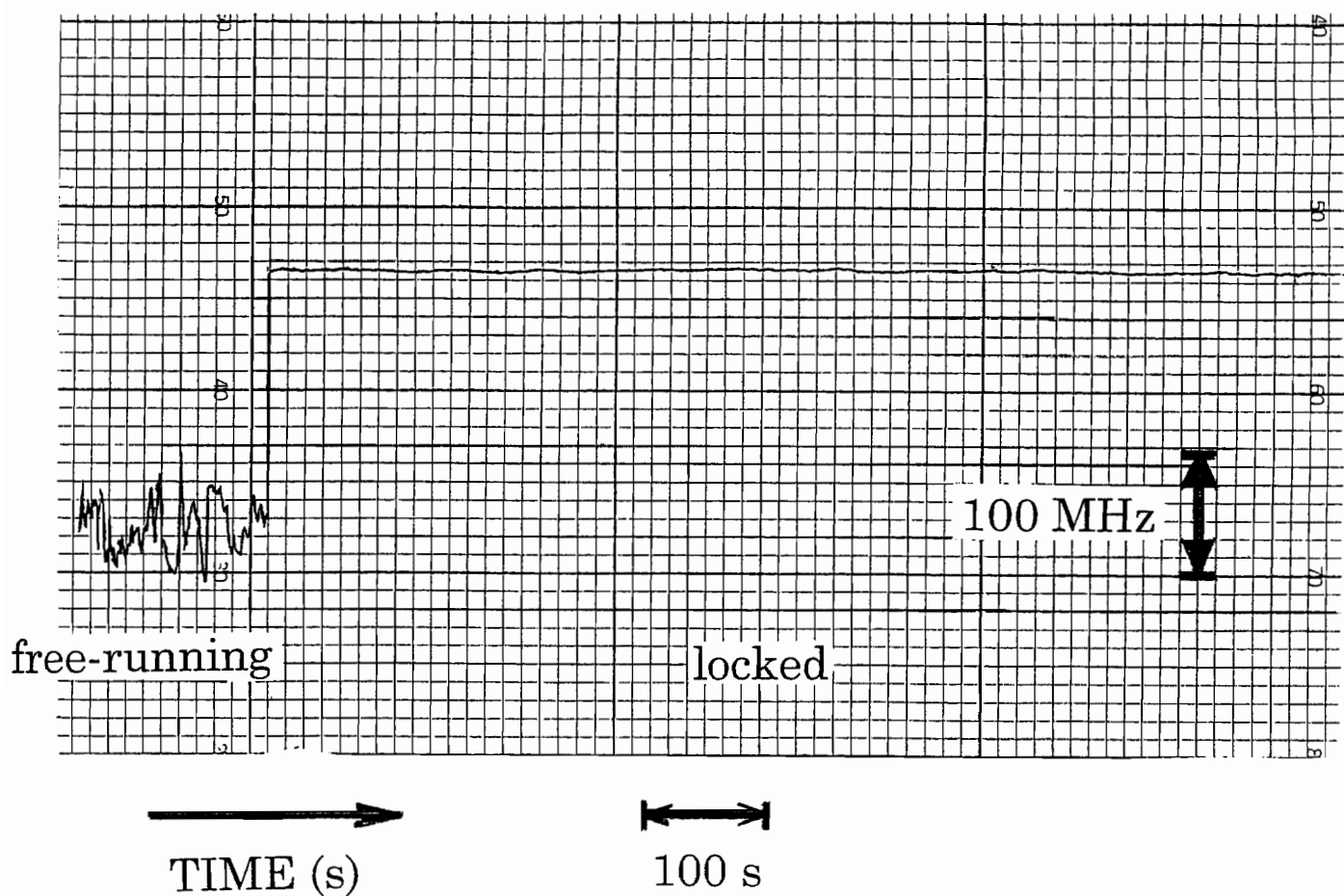


Fig. 2.8. Error signal of lock-in amplifier. The second-harmonic signal was locked to the absorption line of the  $^{88}\text{Sr}^+$  ions. The second-harmonic modulation frequency and modulation depth were 90 Hz and  $\pm 480$  MHz, respectively.

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## Appendix

### A2.1 Induced transition rate

The theory of induced transition rate in spectroscopy is a well-developed subject, treated in too many texts, monographs and reviews. Induced transition rate is of considerable interest in solving rate equations. The common method to calculate the value is executed under the assumption that the incident beam is perfectly monochromatic. However if the light source is a semiconductor laser, the spectral line-width vary from kHz to several tens of MHz. In this section, I analyze the effect of the line-width of a weak incident light. The absorption saturation phenomenon is not treated here.

The rate of an electric dipole transition is given by

$$W = I\sigma/h\nu \quad (2A.1)$$

where  $\sigma$  denotes an absorption cross section, and  $I$  and  $\nu$  are the intensity and the frequency of an interacting light, respectively, and  $h$  is the Plank's constant. The intensity of the light is given by

$$I = h\nu\rho_s c \quad (2A.2)$$

where  $\rho_s$  is the photon density [ $\text{m}^{-3}$ ] and  $c$  is the speed of light. Substitution of (2A.2) into (2A.1) gives

$$W = \sigma\rho_s c \quad (2A.3)$$

The absorption cross-section is given by

$$\sigma(\omega) = (\pi^2 / \omega^2) [(\Delta\omega_L)^2 / \{(\omega - \omega_0)^2 + (\Delta\omega_L)^2\}] \quad (2A.4)$$

where  $\omega_0$  is the stationary transition frequency of atom and  $\Delta\omega_L$  is the half width at half maximum(abbreviated to HWHM) of the Lorentzian profile due to spontaneous emission. The transition frequency  $\omega_0$  of atom is the Doppler-shifted due to the velocity  $v$  of the atom. The Doppler broadening is represented by

$$(\ln 2 / \pi)^{0.5} (1 / \Delta\omega_G) \exp[-\ln 2 \{(\omega - \omega_0) / \Delta\omega_G\}^2] \quad (2A.5)$$

where the  $\Delta \omega_G$  is the HWHM of the Gaussian profile given by

$$\Delta \omega_G = (\omega_0 / c) \sqrt{8kT \ln 2 / m} \quad (2A.6)$$

where  $k$  is the Boltzman's constant,  $T$  is the temperature and  $m$  is the mass of atom. When the light frequency is equal to  $\omega_0$  the induced transition rate at  $\omega_0$  is given as

$$W(\omega_0) = \int_{\omega_0 - n\Delta \omega_G}^{\omega_0 + n\Delta \omega_G} (\pi c^2 / \omega^2) [(\Delta \omega_L)^2 / \{(\omega - \omega_0)^2 + (\Delta \omega_L)^2\}] \rho_S c \\ (\ln 2 / \pi)^{0.5} (1 / \Delta \omega_G) \exp[-\ln 2 \{(\omega - \omega_0) / \Delta \omega_G\}^2] d\omega \quad (2A.7)$$

The integration limits are chosen considering the velocity of atoms.

The transition rate at another frequency  $\omega_1$  is given by

$$W(\omega_1) = \int_{\omega_0 - n\Delta \omega_G}^{\omega_0 + n\Delta \omega_G} (\pi c^2 / \omega^2) [(\Delta \omega_L)^2 / \{(\omega - \omega_1)^2 + (\Delta \omega_L)^2\}] \rho_S c \\ (\ln 2 / \pi)^{0.5} (1 / \Delta \omega_G) \exp[-\ln 2 \{(\omega - \omega_0) / \Delta \omega_G\}^2] d\omega. \quad (2A.8)$$

When, we consider the line-width of the incident light, the profile of the effective absorption cross-section at frequency  $\omega_1$  is given by:

$$\sigma_e(\omega_1) = \int_{\omega_0 - n(\Delta \omega_L + \Delta \omega_S)}^{\omega_0 + n(\Delta \omega_L + \Delta \omega_S)} (\pi c^2 / \omega^2) [(\Delta \omega_L) / \{(\omega_1 - \omega)^2 + (\Delta \omega_L)^2\}] \\ [(\Delta \omega_S / \pi) / \{(\omega - \omega_0)^2 + (\Delta \omega_S)^2\}] d\omega \quad (2A.9)$$

where  $\Delta \omega_s$  is the HWHM of the Lorentzian profile of the incident light spectrum. Fig.2.A1 shows the calculated effective absorption cross-section  $\sigma_e$  of  $\text{Sr}^+$  ions for various  $\Delta \omega_s$  and  $\Delta \omega_L = 2\pi \times 20.2$  [Mrad/s]. Here the curve for 5MHz is the reference used for  $5s^2S_{1/2} - 5p^2P_{1/2}$  transition of  $\text{Sr}^+$  ions. It is found from Fig.2A.1 that the effective absorption cross-section become wider as the line-width of incident light source becomes wider. Simiarly, the induced transition rate at  $\omega_1$  is represented by

$$W(\omega_1) = \int_{\omega_0 - n(\Delta \omega_G + \Delta \omega_L + \Delta \omega_S)}^{\omega_0 + n(\Delta \omega_G + \Delta \omega_L + \Delta \omega_S)} (\pi c^2 / \omega^2) [(\Delta \omega_L) / \{(\omega_1 - \omega)^2 + (\Delta \omega_L)^2\}] \\ [(\Delta \omega_S / \pi) / \{(\omega - \omega_0)^2 + (\Delta \omega_S)^2\}] (\ln 2 / \pi)^{0.5} \\ (1 / \Delta \omega_G) \exp[-(\ln 2) \{(\omega_1 - \omega_0) / \Delta \omega_G\}^2] d\omega_1 \quad (2A.10)$$

The calculated transition rate is shown in Fig 2A.2 for various  $\Delta \omega_S$ ,  $\Delta \omega_G = 0.5$  GHz, and  $\Delta \omega_L = 40.4$  MHz.  $\Delta \omega_G = 0.5$  GHz is the reference of the Doppler width of the  $\text{Sr}^+$  ions in

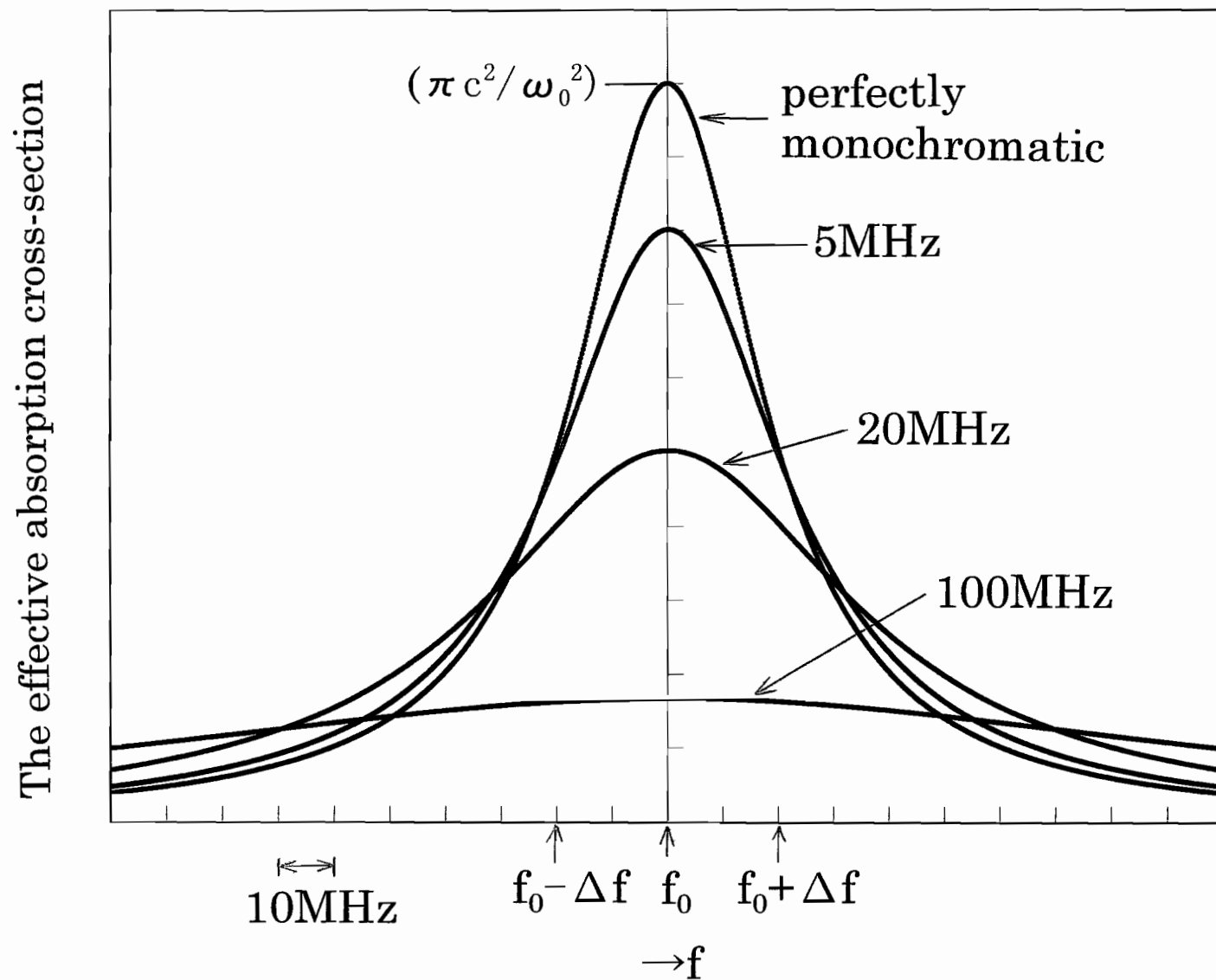


Fig. 2.A1. Profiles of absorption cross-section for various line-width of incident light

our rf trap. In the figure, the curve for the linewidth of 10MHz corresponds to our experiment, which is much smaller than the Doppler width of the  $\text{Sr}^+$  ions.

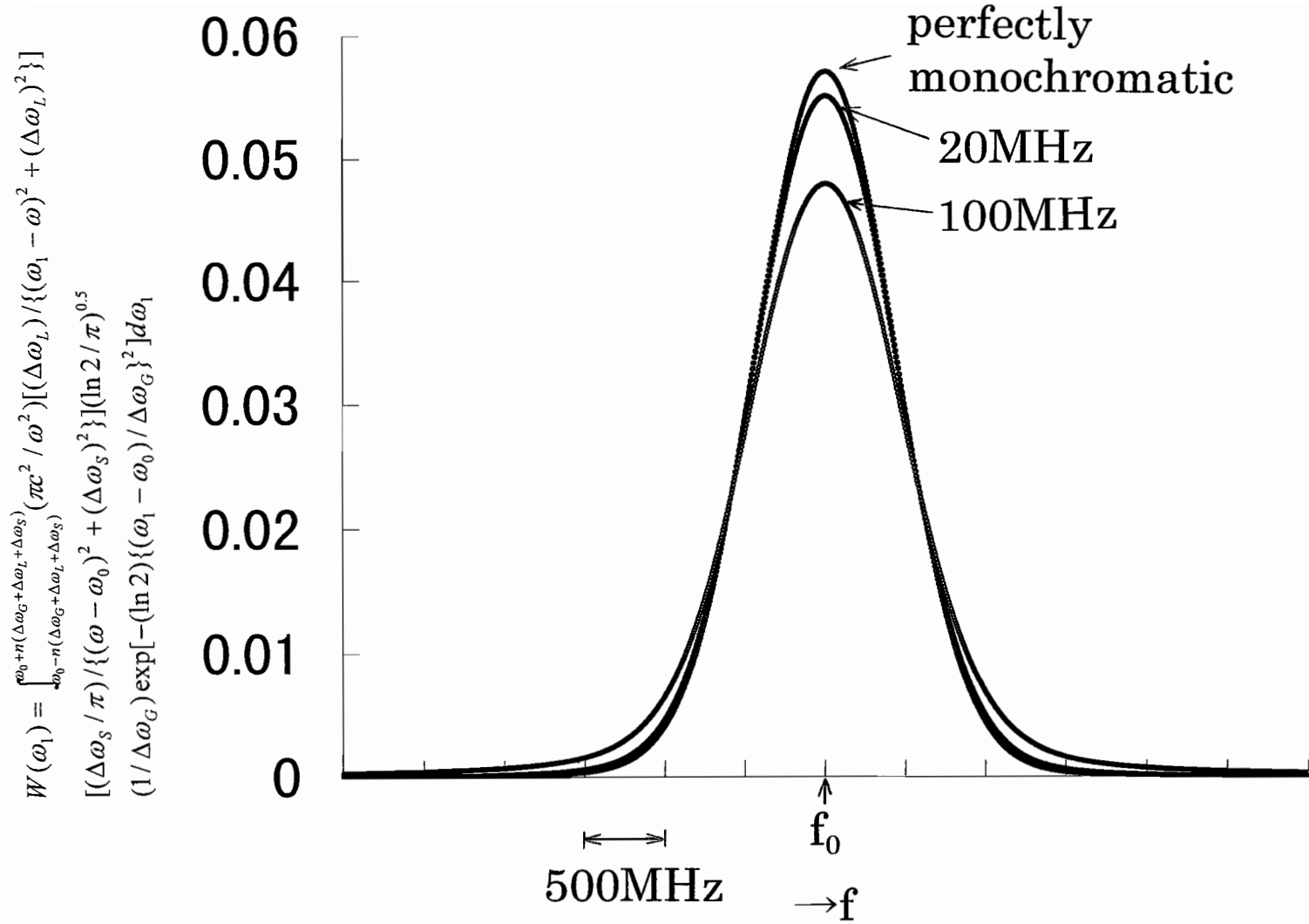


Fig. 2.A2. Induced transition rate for various line-width of incident light.  $f_0$  is the resonance frequency.

## Chapter 3

### Fluorescence of $\text{Sr}^+$ Ions

#### 3.1 Introduction

The use of ion storage technique for frequency standard is motivated by the fact that ion can be confined by electric and magnetic field for long periods of time without suffering the large perturbations such as the collisions with the wall. The linewidth of a spectrum  $\Delta \omega$  is limited by uncertainty law  $\Delta \omega \Delta t \simeq 1$  where  $\Delta t$  is the interrogation time. Therefore, a long interrogation time results in a high resolution or high Q of the spectra, where Q is defined by  $Q = \Delta \omega / \omega_0$  and  $\omega_0$  is the transition angular frequency. When ions are confined within the region of  $\lambda / 2\pi$  (where  $\lambda$  is the wavelength of the interrogated electromagnetic field), the observed spectra are free from the first-order Doppler shift. The region  $\lambda / 2\pi$  is called the Lamb-Dicke region. Confinement within the Lamb-Dicke region is easily realized in the microwave region. For example,  $\lambda / 2\pi$  is 10mm at 5GHz. Among various species, we chose  $\text{Sr}^+$  ions for microwave frequency standard, since its cooling wavelength is accessible with diode laser system, leading to the possibility of simple, compact and ultimately transportable system.

In this chapter, the development of an ion trap system for confining ions in a vacuum for a long time is described. In order to obtain a good signal-to-noise ratio (S/N), the ion trap was fabricated by using a relatively large electrode for confining a large number of ions.  $\text{Sr}^+$  ion fluorescence from  $5p^2P_{1/2}$  to  $5s^2S_{1/2}$  was detected, and the temperature of the ions was calculated from the full width at half maximum of the fluorescence. In the case where  $\text{Sr}^+$  ions are irradiated with a laser, whose wavelength is equal to the allowed transition  $5s^2S_{1/2} \rightarrow 5p^2P_{1/2}$ , the ions in the laser-pumped  $5p^2P_{1/2}$  state also decay to the  $4d^2D_{3/2}$  metastable state with a branching ratio of 13:1<sup>1)</sup>. This effect reduces the fluorescence intensity. Therefore, we observed the change in the fluorescence intensity using a pump-back laser and estimated the collisional quenching rate from the metastable state of the  $\text{Sr}^+$  ion. In addition, various buffer gases, such as He,  $\text{N}_2$  and  $\text{CH}_4$ , were introduced into the ion trap system and their effects on the fluorescence intensity were observed.

#### 3.2 Ion Trap

Electric and magnetic field are used to exert relatively large forces, giving traps with large depth (much greater than  $kT$  at room temperature), easy loading and long storage times. The simplest arrangement which could be envisaged is one which gives rise

to a three-dimensional potential well for the electrical potential  $\phi$ . However, application of Gauss's law shows that this is not possible (the Earnshaw's theorem). The result is that a three-dimensional trap requires a more complicated arrangement. The system generally used is one with cylindrical symmetry. The three-dimensional radio-frequency quadrupole ion trap is one of a family of devices which utilize path stability as means of separating ions according to their mass-to-charge number ( $m/z$ ) ratio. The geometry of the quadrupole mass spectrometer is shown in Fig. 3.1. The analyzer consists of a parallel array of four rod electrodes mounted in a square configuration. The ideal geometry dictates that each electrode should be hyperbolic in cross section, but in practice, for ease of manufacture, round cylindrical rods often are employed, with the spacing optimized to approximate the ideal electric field. The field within the analyzer is created by coupling opposite pairs of rods together and applying radio-frequency and direct-current potentials between the pairs. Ions created within the source are injected through the parallel array, and under the influence of the fields they describe complex trajectories. Some of these trajectories are unstable in that they tend toward infinite displacement from the center so that the ions are lost, for example, through collisions with an electrode. Ions which are successfully transmitted through the analyzer are said to possess stable trajectories, and these are recorded on the detection system.

The radio-frequency quadrupole ion trap is directly related to the quadrupole mass filter so that it can be visualized as being a solid of revolution generated by rotating the hyperbolic rod electrodes about an axis perpendicular to the  $z$  axis and passing through the centers of two opposing rods. This results in one pair of electrodes joining up to form a doughnut-shaped ring electrode and other two forming end-cap electrodes which are moved closer together as shown in Figure. 3.2. The field is generated by applying the RF and DC voltages between the ring electrode and the pair of end-cap electrodes, it is generally more convenient to maintain the end-cap electrodes at ground potential and simply supply power to ring electrode. The motion of a positive ion is described in Appendix 1. In the case of the rf trap, a heating mechanism exists. The energy of the micro motion, caused by the rf driving field, of a trapped ions is converted to a secular motion via ion-background gas or ion-ion collisions. This is called rf heating. As the number of trapped ions increases, a cloud of trapped ions spreads outwards from the trap center due to the Coulomb repulsion force caused by their charges. The rf driving field becomes stronger as the distance from the trap center increases. Therefore, an increase of the number of trapped ions results in increase in the energy of micro motion, and consequently, of secular motion due to the rf heating. In order to reduce the kinetic energy of the trapped ions, their number must be decreased. If we would like to cool the ion as low as possible, it is necessary to trap only a single ion and locate it at the trap center where no trapping rf field exist. On the other hand, the restoring force acts in all directions.

### 3.3 Strontium Ion

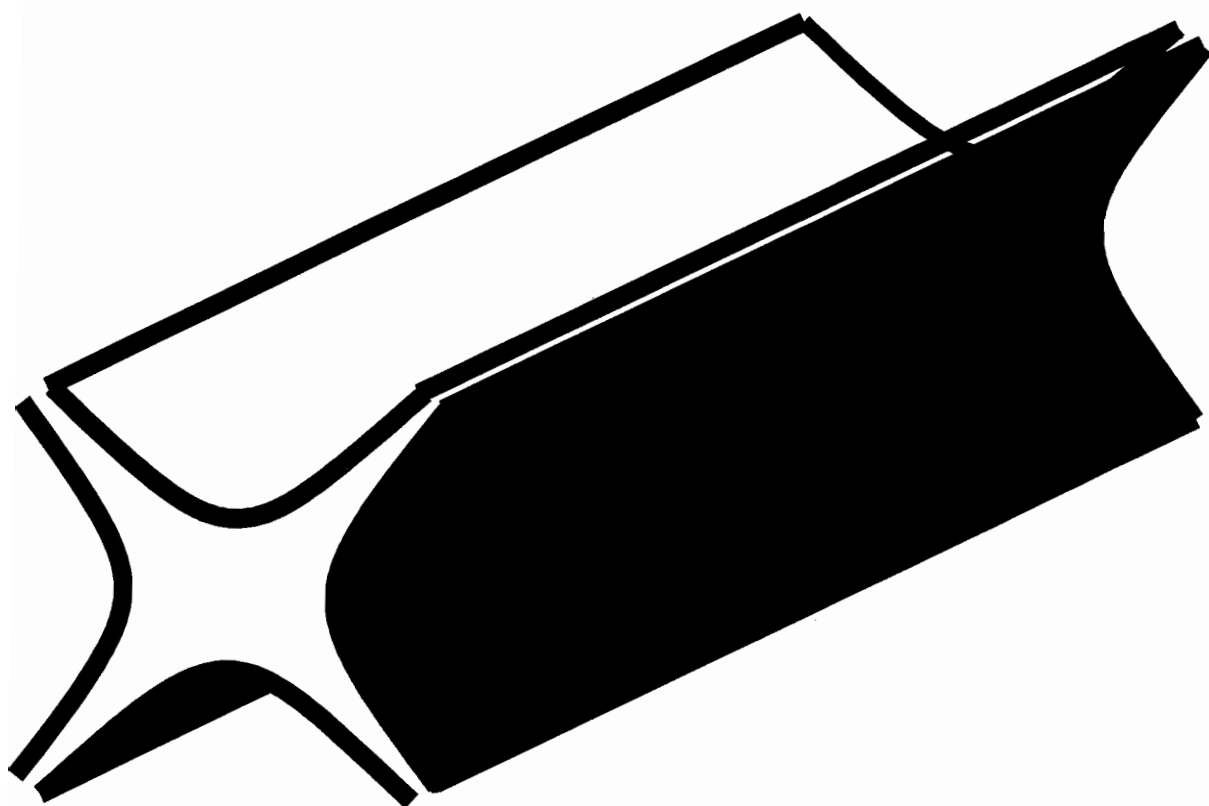


Fig. 3.1. Geometry of the quadrupole mass spectrometer.

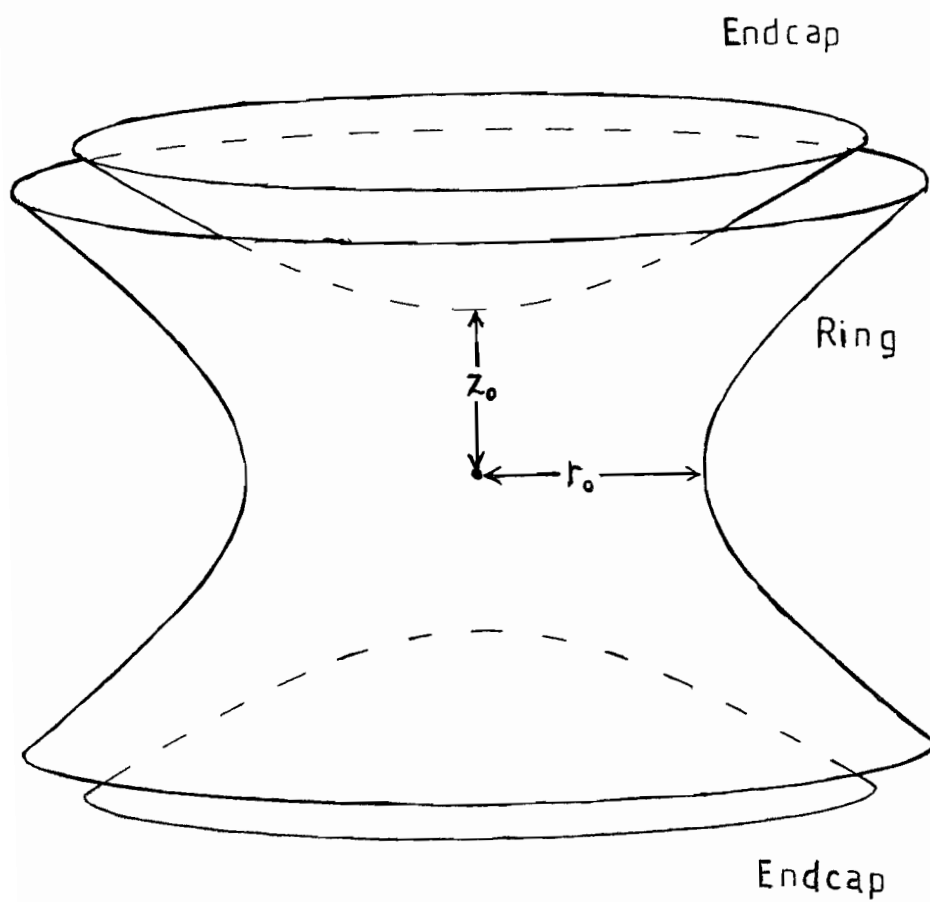


Fig. 3.2. Electrode structure of an ion trap.

Sr atom is involved a rare earth and it's singly ionized  $\text{Sr}^+$  ion has a weakly bound outer electron, the so called valence electron, all other 36 electrons are in closed shells. Some of the basic characteristics of Sr atom are listed in Table 3.1.  $\text{Sr}^+$  ion has a term scheme similar to that of alkali atoms. The energy level diagram of  $^{88}\text{Sr}^+$  ion is shown in Figure 3.3. The natural linewidth of the  $5s^2S_{1/2}$ - $5p^2P_{1/2}$  transition is 40.4 MHz.<sup>1)</sup> Ions are excited by pumping with light from a laser source with a wavelength of 421.671 nm, which matches the excitation of the ion from the  $5s^2S_{1/2}$  ground state to the  $5p^2P_{1/2}$  excited state. Ions in the  $5p^2P_{1/2}$  state relax to the  $5s^2S_{1/2}$  ground state by emitting fluorescence in a time of order  $10^{-8}$  s. It is this fluorescence that we observe. In addition, ions in the  $5p^2P_{1/2}$  state can relax to the metastable  $4d^2D_{3/2}$  level by emitting fluorescence, with a branching ratio of 1 in  $14^1$ . Ions in this state remain there for about 0.3 s, after which they return to the ground state, again by emitting fluorescence. If the laser intensity at 421.671 nm that excites the ions from the  $5s^2S_{1/2}$  ground state to the  $5p^2P_{1/2}$  excited state is high, ions are likely to accumulate in the metastable  $4d^2D_{3/2}$  level, which acts to weaken the intensity of the 421.671 nm fluorescence. In order to prevent this, it is necessary to pump the ions in the metastable  $4d^2D_{3/2}$  level back up to the  $5p^2P_{1/2}$  level. We accomplish this by using a 1091.788 nm fiber laser (IPG Laser Ytterbium Laser Model YLD-100B-1091) to re-excite the ions in the metastable  $4d^2D_{3/2}$  level back up to the original higher level. The buffer gas is also used to quench the metastable state. The electric quadrupole allowed  $4d^2D_{5/2}$ - $5p^2P_{1/2}$  transition at 674nm has a narrow line width of 0.4Hz.<sup>2)</sup> The transition has been recommended to one of the frequency standard by CIPM. Some of the basic characteristics of  $\text{Sr}^+$  ions are listed in Table 3.2.  $\text{Sr}^+$  ion has odd isotopes, i.e.,  $^{87}\text{Sr}^+$  ion with a nuclear spin  $i = 9/2$  which has a hyperfine structure.

The energy diagram of  $^{87}\text{Sr}^+$  ion is shown in Figure 3.4. The transition between the hyperfine splitting in the  $^2S_{1/2}$  ground state provides the reference in a microwave frequency standard. The frequency of the transition is 5.002 GHz. A particularly narrow transition between  $|F = 4, m_f = 0\rangle$  and  $|F = 5, m_f = 0\rangle$ , whose frequency is independent of magnetic field to first order, can be utilized to determine the hyperfine splitting or frequency standard.

### 3.4 RF Resonance Method

In order to obtain a good signal-to-noise ratio (S/N), the ion trap was fabricated by using a relatively large electrode for confining a large number of ions. In order to determine the number of trapped ions we used rf resonance absorption method. In Fig. 3.5 we show the trap structure schematically. The trap is made up of a diameter 40 mm single-ring electrode and two end-cap electrodes. Sr atoms ejected from an oven located close to the electrodes were injected toward the trap center from a hole bored in the ring

# Atomic number 38

## Electron configuration

| Shells |  |     |   |       |   |    |     |   |   |       |
|--------|--|-----|---|-------|---|----|-----|---|---|-------|
| K      |  | L   |   | M     |   |    | N   |   |   | O     |
| n=1    |  | n=2 |   | n = 3 |   |    | n=4 |   |   | n = 5 |
| s      |  | s   | p | s     | p | d  | s   | p | d | s     |
| 2      |  | 2   | 6 | 2     | 6 | 10 | 2   | 6 | 0 | 2     |

First ionization potential            5.69 eV

## Natural abundance

| Isotope | mass       | abundance % |
|---------|------------|-------------|
| 84      | 83.913426  | 0.51        |
| 86      | 85.9092647 | 9.86        |
| 87      | 86.9088816 | 7.00        |
| 88      | 87.9056167 | 82.58       |

Table 3.1. Basic characteristics of Sr atom .

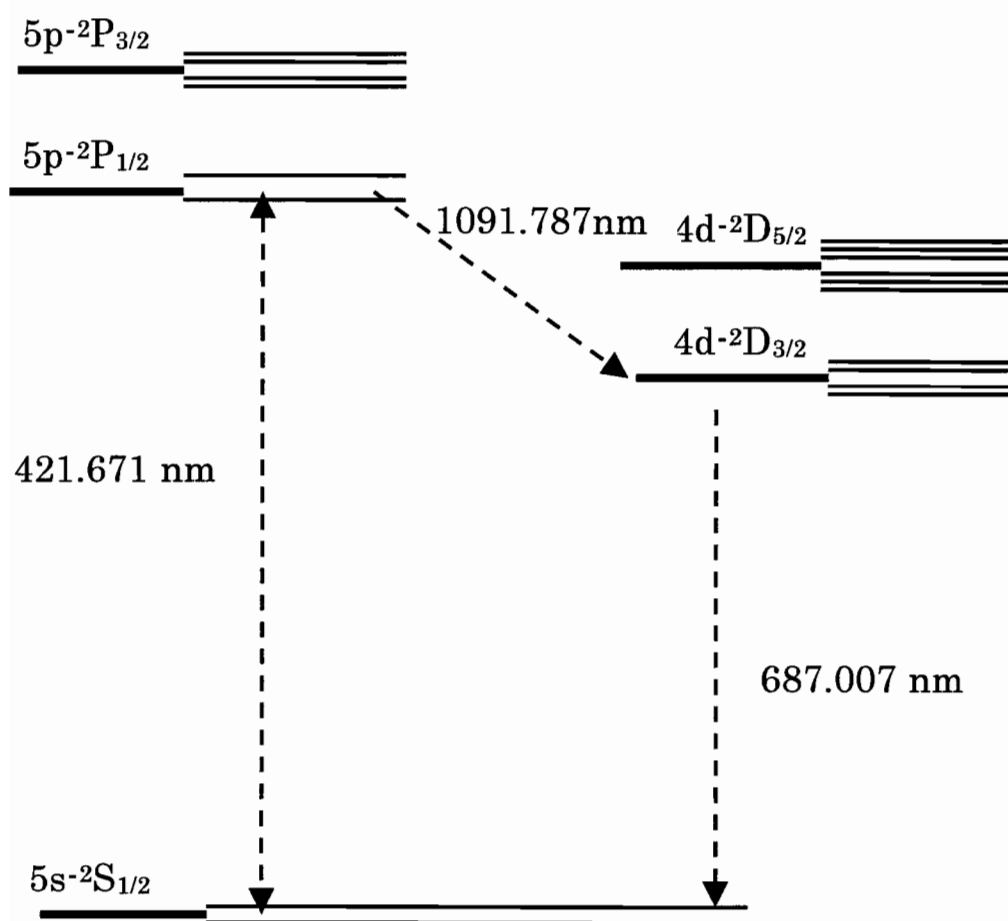


Fig. 3.3. Energy level diagram of  $^{88}\text{Sr}^+$ .

| Transition                  | Probabilities<br>A (10 <sup>6</sup> /sec) | Wave-length<br>( nm ) | Frequency<br>( THz ) |
|-----------------------------|-------------------------------------------|-----------------------|----------------------|
| $5p^2P_{3/2} - 5s^2S_{1/2}$ | $143 \pm 6$                               | 407.886               | 734.991              |
| $5p^2P_{1/2} - 5s^2S_{1/2}$ | $127 \pm 5$                               | 421.671               | 710.964              |
| $5p^2P_{3/2} - 4d^2D_{5/2}$ | $8.7 \pm 1.5$                             | 1033.014              | 290.211              |
| $5p^2P_{3/2} - 4d^2D_{3/2}$ | $1.0 \pm 0.2$                             | 1003.940              | 298.616              |
| $5p^2P_{1/2} - 4d^2D_{3/2}$ | $9.5 \pm 2$                               | 1091.788              | 274.589              |
| $4d^2D_{3/2} - 5s^2S_{1/2}$ |                                           | 687.007               | 436.375              |
| $4d^2D_{5/2} - 5s^2S_{1/2}$ | 1.3                                       | 674.025               | 444.779              |

Table 3.2. Basic characteristics of Sr<sup>+</sup> ion.

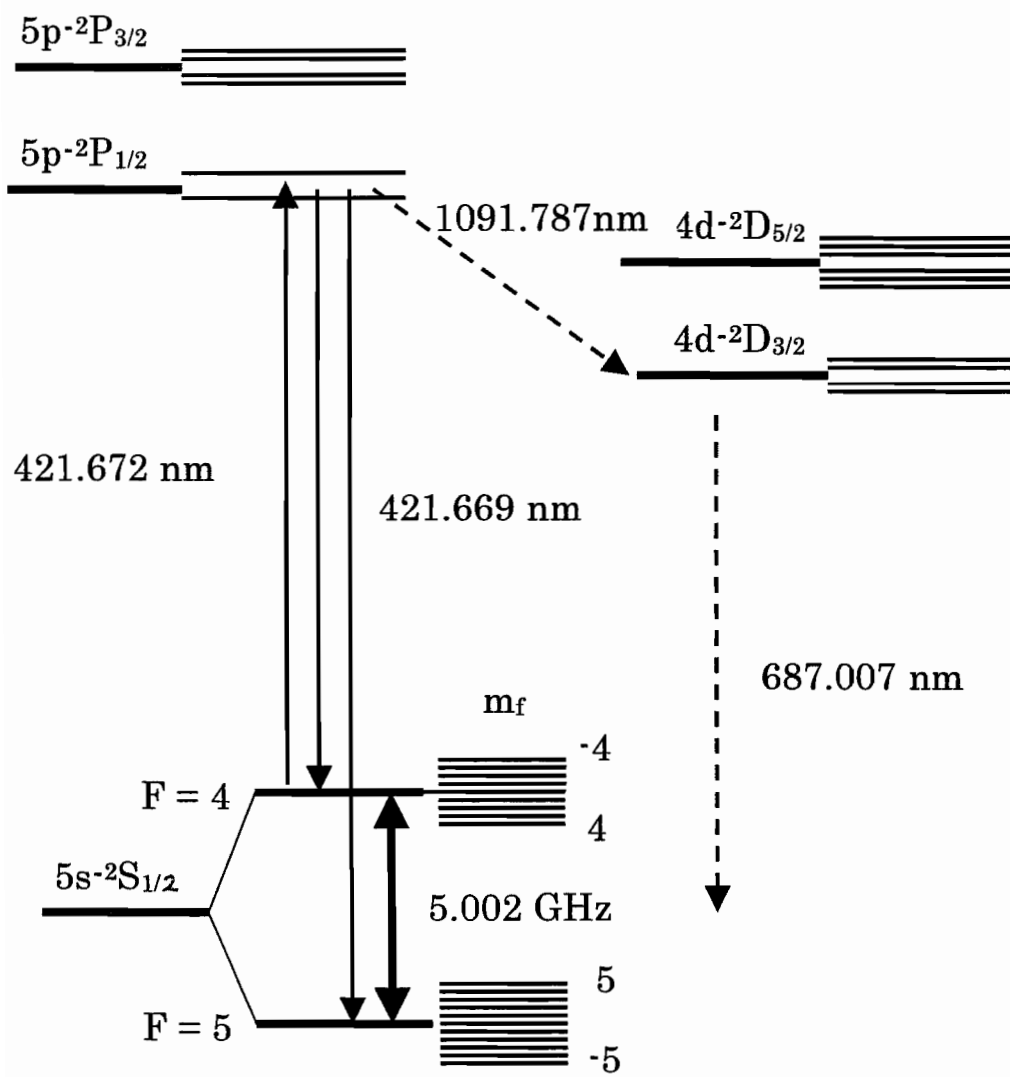


Fig. 3.4. Energy level diagram of  $^{87}\text{Sr}^+$ .

electrode. The trap was set up at the center of the vacuum chamber, where the vacuum could reach values of  $10^{-7}$  Pa when an ion pump was used. Neutral gas was transported to the center of the vacuum chamber by a leak valve. The peak-to-peak amplitude of the applied ac voltage was 512 V at a frequency of 400 kHz, and  $q_z = 0.45$ ,  $\alpha_z = 0.02$ .

Confining the ions at the center of the trap does not bring them to rest: although they are confined, they still execute three-dimensional harmonic oscillations. Although the frequency of harmonic oscillations of a single ion obtained by solving the Mathieu equation is 69 kHz, in practice the number of confined ions is in the range  $10^6 \sim 10^7$ , and these ions interact by way of their mutual Coulomb forces. These interactions affect the trapping potential so as to lower the harmonic oscillation frequency from the value obtained by solving the Mathieu equation<sup>3)</sup>. Current experiments give a value of 55 kHz for this frequency.

When an external probe oscillator is used to excite a resonant (tank) circuit connected in parallel across the two end-cap electrodes, those ions that oscillate along the  $z$ -axis direction in-phase with the oscillator signal absorb energy from it. By sweeping the dc voltage  $V_{dc}$  shown in the Fig. 3.5, we can vary the depth of the potential of the ion trap, and hence the frequency of the ion's harmonic oscillations. The LC tank circuit was designed to have a resonance frequency of 55 kHz. The rf probe oscillator, which adds a resistance  $R_0$  to the LC tank circuit, was designed to have a frequency of 55 kHz as well. When the swept dc voltage  $V_{dc}$  passes through the value corresponding to ion's harmonic oscillation frequency of 55 kHz, the ion absorbs energy and a dip in the detector voltage is observed. Detecting this absorption signal allows us to verify the presence of the ions. In addition, in order to find the total number of confined ions we swept the frequency of the rf probe oscillator from 54 kHz to 56 kHz, and used the measured LC tank circuit voltage for detection. The sweep time was taken to be 50 seconds. We sketch this voltage line shape in Fig. 3.6. When it contains no ions, the trap can be treated as a simple capacitor. Hence, if we sweep the frequency of the rf probe oscillator, only the resonance curve attributable to the LC tank circuit connected between the end caps is obtained (Fig. 3.6a). In contrast, when ions are confined in the trap, they act like a second resonant circuit in parallel with the real LC tank circuit<sup>4)</sup>. This second "effective" resonant circuit creates a dip in the line shape of the LC tank circuit (see Fig. 3.6b). However, it is only when the two resonance frequencies are equal that a dip is observed. When this is the case, the confined ions resonantly absorb power from the rf probe oscillator. We may regard the excitation of these ion oscillations as giving rise to the observed decrease in the voltage across the LC tank circuit. From the depth ( $v_{10} \sim v_1$ ) of the dip in Fig. 3.6 and the width  $\Delta\omega$  we can calculate the total number of confined ions<sup>5)</sup>. Appendix 2 describes how to calculate the number of trapped ions. Figure 3.7 shows how the total number of ions varies as we vary the pressure of the buffer gas  $N_2$ . We explain this result as follows: when the buffer gas pressure is high, the number of collisions between the ions and buffer gas molecules becomes high, causing the ions to cool by losing energy, which in turn causes a large number of ions to accumulate in the trap. From the calculated total number of ions we can determine the size of the ion cloud. A method for finding the spatial distribution of the ions from the ion temperature is

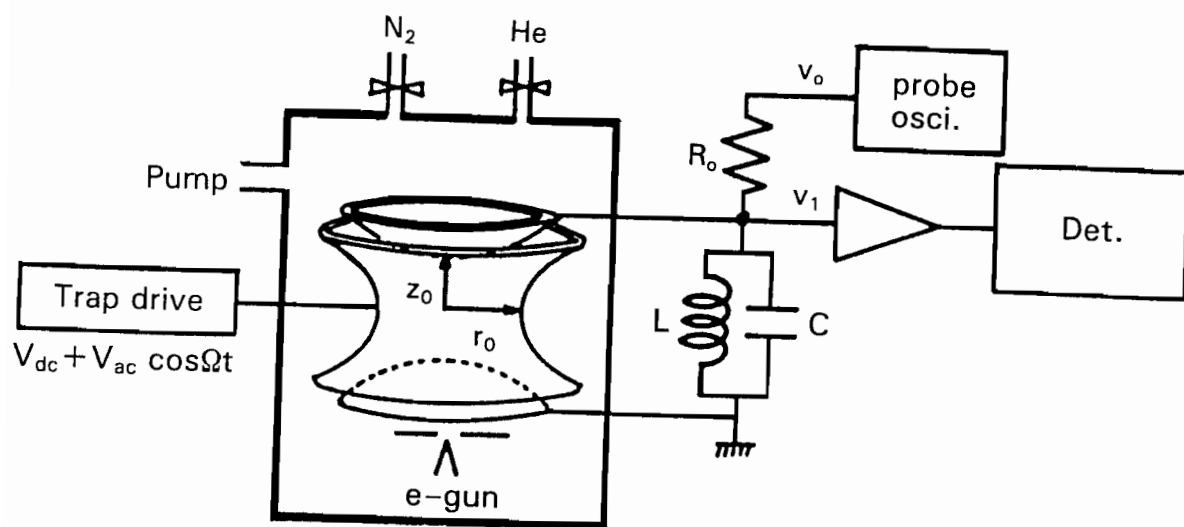


Fig. 3.5. Experimental setup for ion trap.

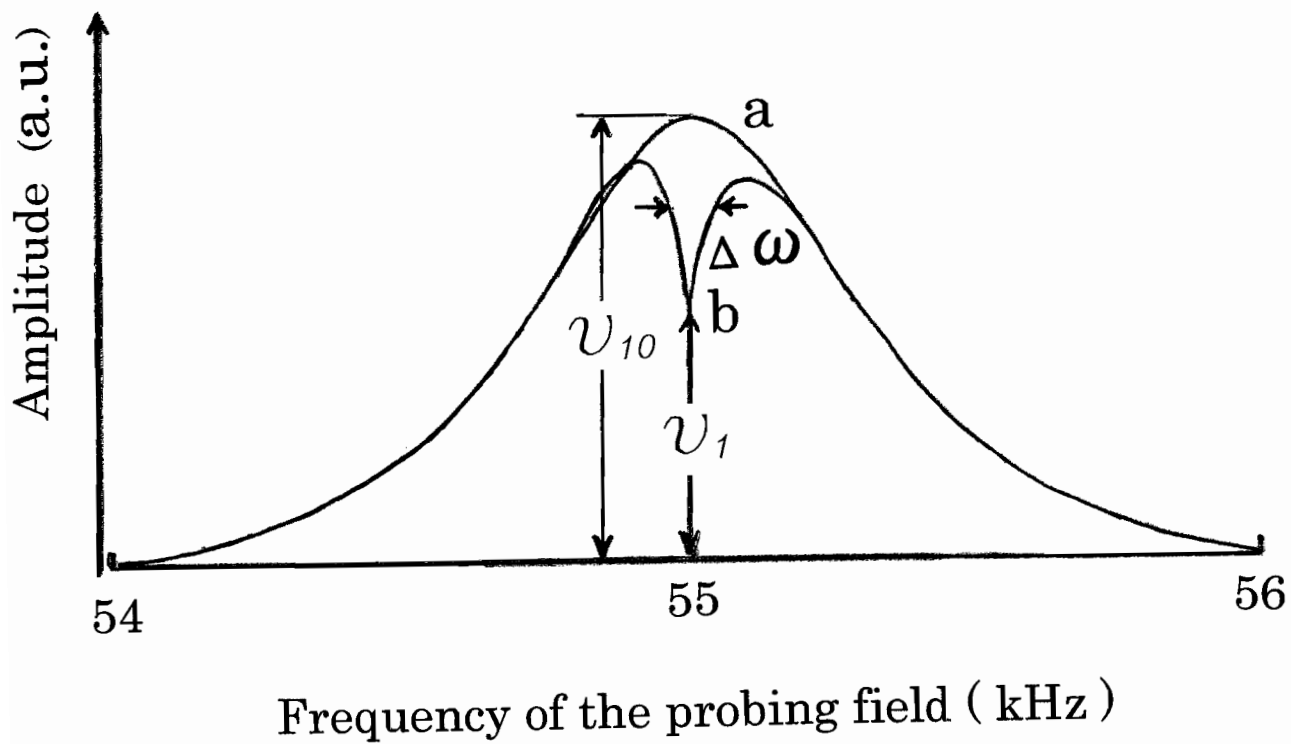


Fig. 3.6. Absorption signal obtained by sweeping the dc voltage  $V_{dc}$  applied to the ring electrodes.

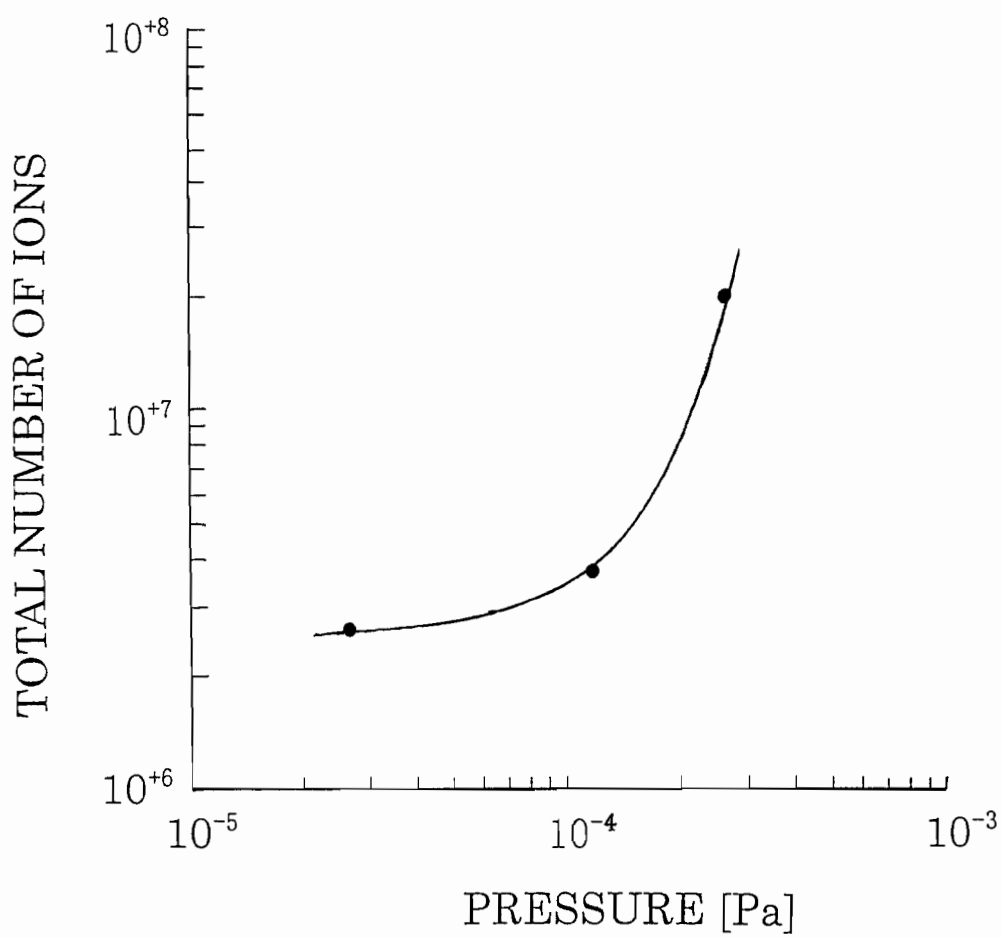


Fig 3.7. Change in the number of trapped ions as the buffer gas pressure is varied.

discussed in the article by Cutler et al<sup>3)</sup>. In the situations we encountered most often, the inferred size indicates an approximate diameter of around 7 mm.

### 3.5 Detection of Fluorescence

In order to estimate the second order Doppler shift of  $\text{Sr}^+$  ions in the microwave region, we must know the speed of ions. The speed of ions is derived from the temperature of ions. The temperature of the ions is calculated from the width of the fluorescence signal. The above-mentioned reason we observed the fluorescence of trapped  $\text{Sr}^+$  ions. Figure 3.8 shows the experimental apparatus for observing the fluorescence. The 1091.788nm light was obtained by the Ytterbium fiber laser (Model YLD-100B-1091). The fiber laser has gain at many longitudinal modes, which lie under an envelope with a full width at half maximum of 68 GHz. Out of a total output power of 90 mW, we used 2.6 mW for fluorescence detection. As a result of this re-pumping light, an increase in the fluorescence intensity was observed. For a 1.86  $\mu\text{W}$  intensity of 421.671 nm laser light at the position of the ion cloud over a cross sectional area of about 2.5 mm<sup>2</sup> ( $\phi = 1.8$  mm), we set the intensity of the 1091.788 nm fiber laser light at 2.6 mW over a cross sectional area of about 20 mm<sup>2</sup> ( $\phi = 5$  mm). From the fact that the aperture had a solid angle of  $0.06 \times 4\pi$ , the lens and filter transmission was 0.69, and the photomultiplier tube electron efficiency was 0.22, a product of 0.009 is derived for the fluorescence detector efficiency. At very low incident radiation power it is advantageous to use the photo-multiplier for counting photoelectrons emitted at a rate  $n$  per second rather than to measure the photocurrent.

One of the major contributions to the spectral line-width is the Doppler width, which is due to the thermal motion of the ions. We observed two or three Gaussian profiles with the irradiation of two laser beams, in which the wavelength of the 843nm laser light was swept, though that of the 1092nm laser light was fixed, and then we observed two or three profiles with cutoff of the 1092nm laser light beam. This procedure was repeated twice. In all cases where we observed the fluorescence from the trapped ions, the gate time of the photon counter was fixed to be 1s. The fluorescence signals which were obtained by the photon counting method are shown in Fig. 3.9. From the half-width at half maximum (HWHM) of the Gaussian profiles, that is, the Doppler width, the temperatures of trapped ions were obtained.

In order to determine the proper buffer gas for cooling the  $\text{Sr}^+$  ions, we examined several gases. In addition we changed the pressure of each buffer gas. Fig. 3.10 shows the temperature of  $\text{Sr}^+$  ions with buffer gases ( $\text{He}$ ,  $\text{N}_2$  and  $\text{CH}_4$ ) at several different pressures. He gas was effective in reducing the kinetic energy of the trapped ions. The ions were cooled to 400 K at a He pressure  $2.7 \times 10^{-4}$  K which reduced the second order of the Doppler shift below  $5 \times 10^{-13}$  in the microwave region.

In order to examine the relationship between the total number of trapped ions and the He buffer gas pressure, we changed the He buffer gas pressure and measured the fluorescence intensity. Since the increase of the number of trapped ions results in the

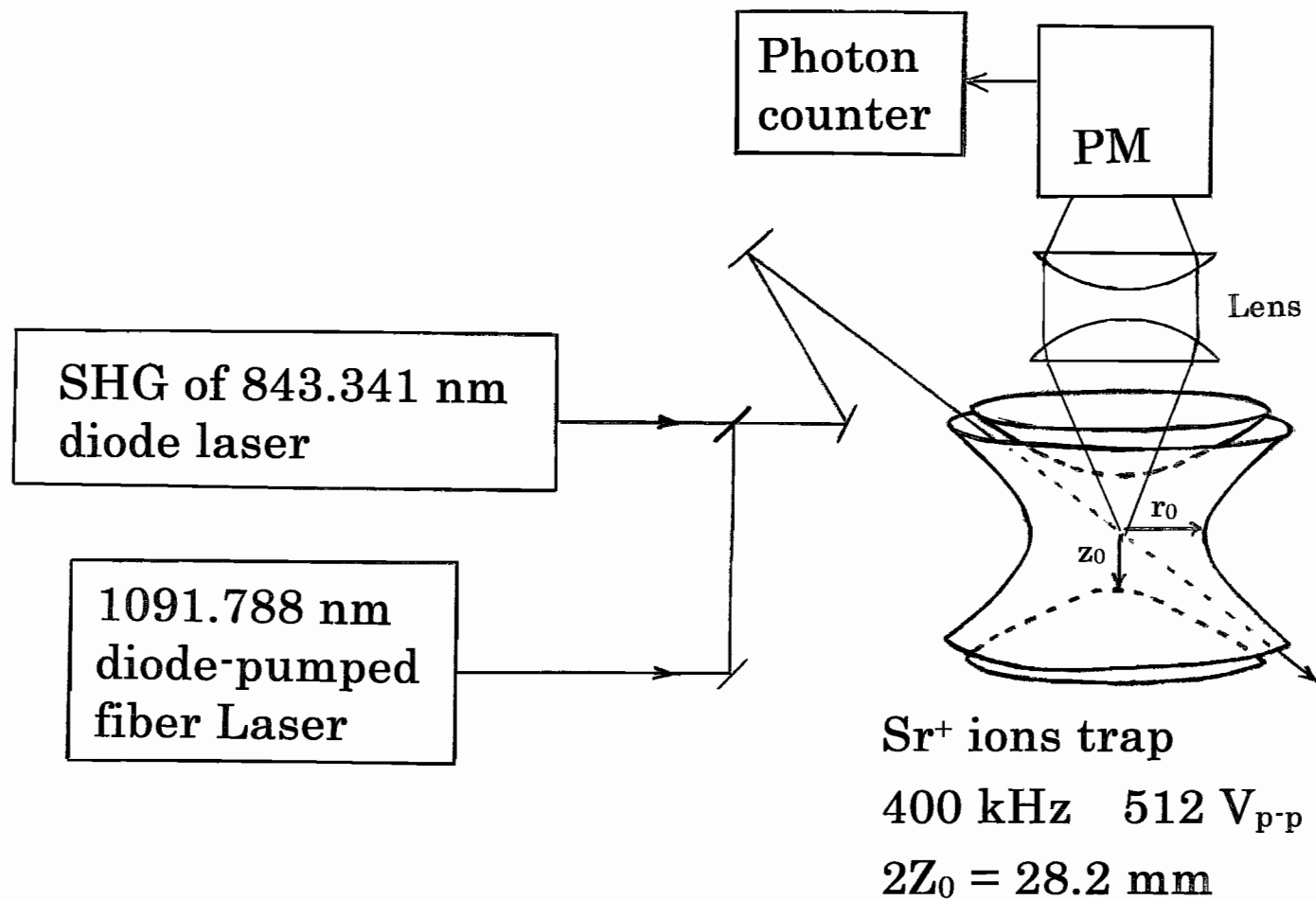


Fig. 3.8. Experimental setup for fluorescence detection.

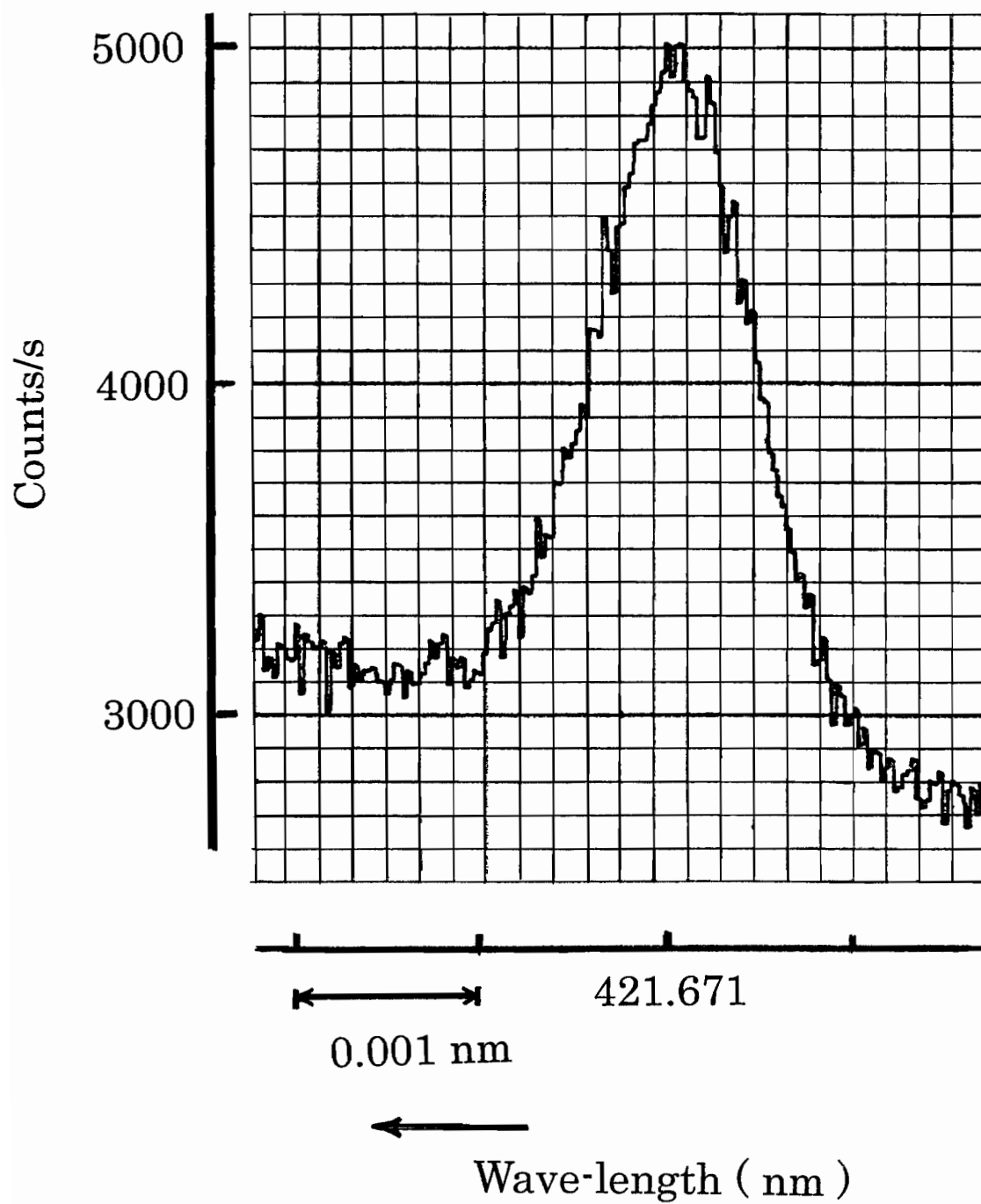


Fig. 3.9. Measured fluorescence signal.

increase of the S/N ratio. The increase of the trapped ions can be deduced by the increase of the fluorescence intensity. Figure 3.11 shows the He buffer gas pressure versus the fluorescence intensity. As described in section 3.4, when the buffer gas pressure is high, the number of collisions between the ions and buffer gas molecules becomes high, causing the ions to cool by losing energy, which in turn causes a large number of ions to accumulate in the trap.

In order to study the relationship between the total number of trapped ions and the fluorescence intensity, we drew a graph. Figure 3.12 shows the total number of trapped ions versus the fluorescence intensity. The increase of the number of trapped ions results in the increase of fluorescence. The increase ratio of fluorescence intensity versus the number of trapped ions decreased. As the number of trapped ions increased, a cloud of trapped ions spread outwards from the trap center due to the Coulomb repulsion force caused by their charges. As the size of the ion cloud became large, the ratio of the fraction of the ion cloud, which was irradiated with the laser beam decreased.

### 3.6 Fluorescence Intensity with Pump-back Light

It is important to know the quenching rate from the  $4d^2D_{3/2}$  metastable state of  $Sr^+$  ions. Since, in the case where  $Sr^+$  ions are irradiated with a laser, whose wavelength is equal to the strongly allowed transition  $5s^2S_{1/2}-5p^2P_{1/2}$ , the ions in the laser-pumped  $5p^2P_{1/2}$  state also decay to the  $4d^2D_{3/2}$  metastable state with a branching ratio of 13:1.<sup>1)</sup> This effect reduces the fluorescence intensity. From the ratio of the peak intensity with irradiation of laser light at 1092nm and that without 1092nm, we can obtain the quenching rate for the  $4d^2D_{3/2}$  state of  $Sr^+$  ion with He,  $N_2$  and  $CH_4$  gases<sup>6,7)</sup>. Fig. 3.13 shows the typically observed time-dependent peak intensity of the Gaussian profiles at different pressures of  $N_2$  gas. The decay of the intensities of the fluorescence was caused by the escape of the trapped ions from the ion trap. A UV filter with transmission ratio of 0.67 for 422nm was introduced after recording several signals to obtain signals under a condition of the lower light intensity. From these data, it was confirmed that the saturation effect was not induced with the  $1.86 \mu W$  laser power. In order to obtain the ratio of the fluorescence with the pump-back light to that without the pump-back light, let  $N_s$ ,  $N_p$ ,  $N_d$ , be the numbers of ions in the S, P, and D states, and  $r_1$  and  $r_2$  the percentages relative to the ion cloud as a whole of ions illuminated by light at the 421.671 nm and 1091.788 nm laser wavelengths. Then the following rate equations can be written:

$$dN_s/dt = -W_1(f)r_1N_s + \{A_1+W_1(f)r_3\}N_p + (A_3+q)N_d \quad (3.1)$$

$$dN_p/dt = +W_1(f)r_1N_s - \{A_1+W_1(f)r_3+A_2+W_2(f)\}N_p + W_2(f)r_2N_d \quad (3.2)$$

$$dN_d/dt = +\{A_2+W_2(f)\}N_p - \{W_2(f)r_2+A_3+q\}N_d, \quad (3.3)$$

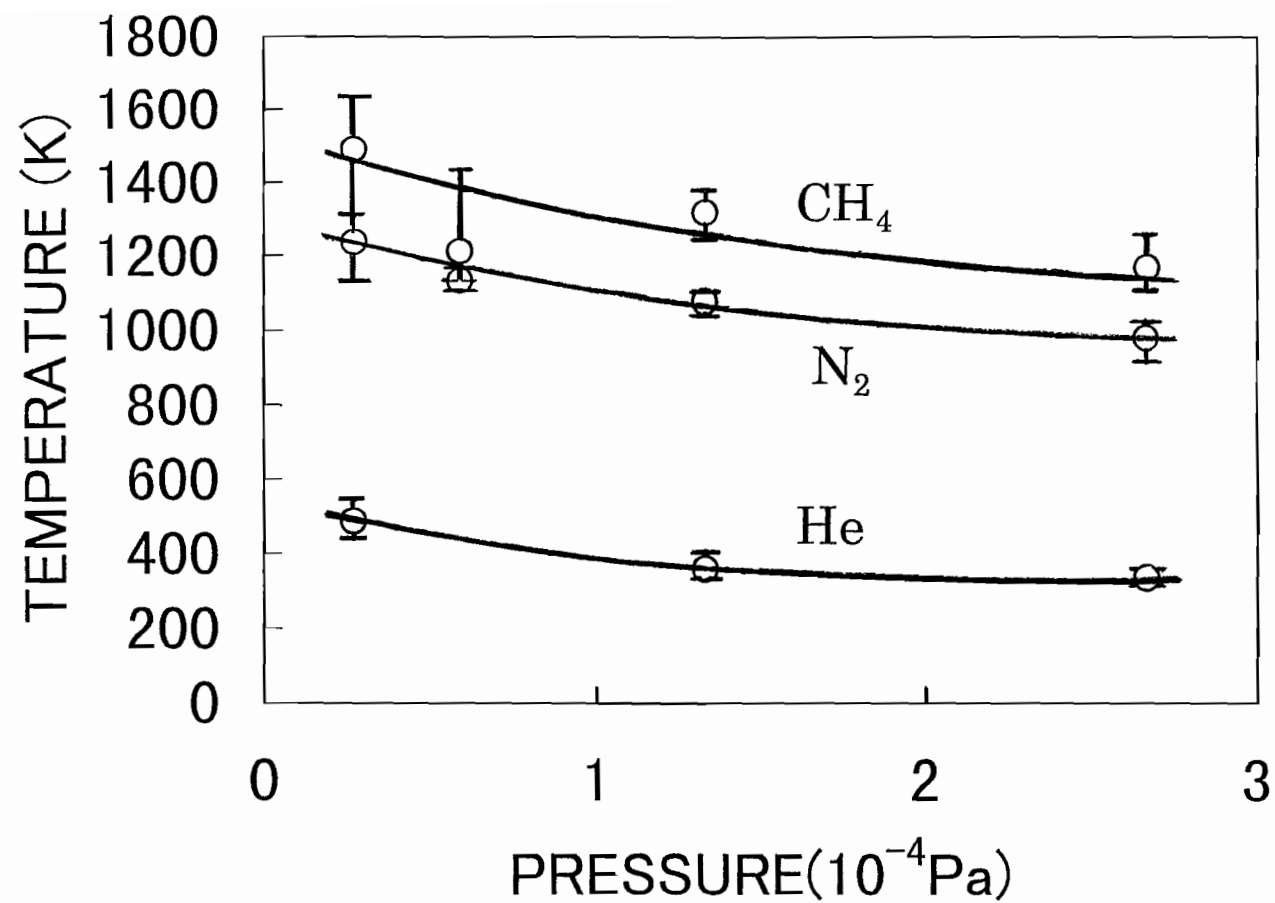


Fig. 3.10 Temperature of trapped  $\text{Sr}^+$  ions.

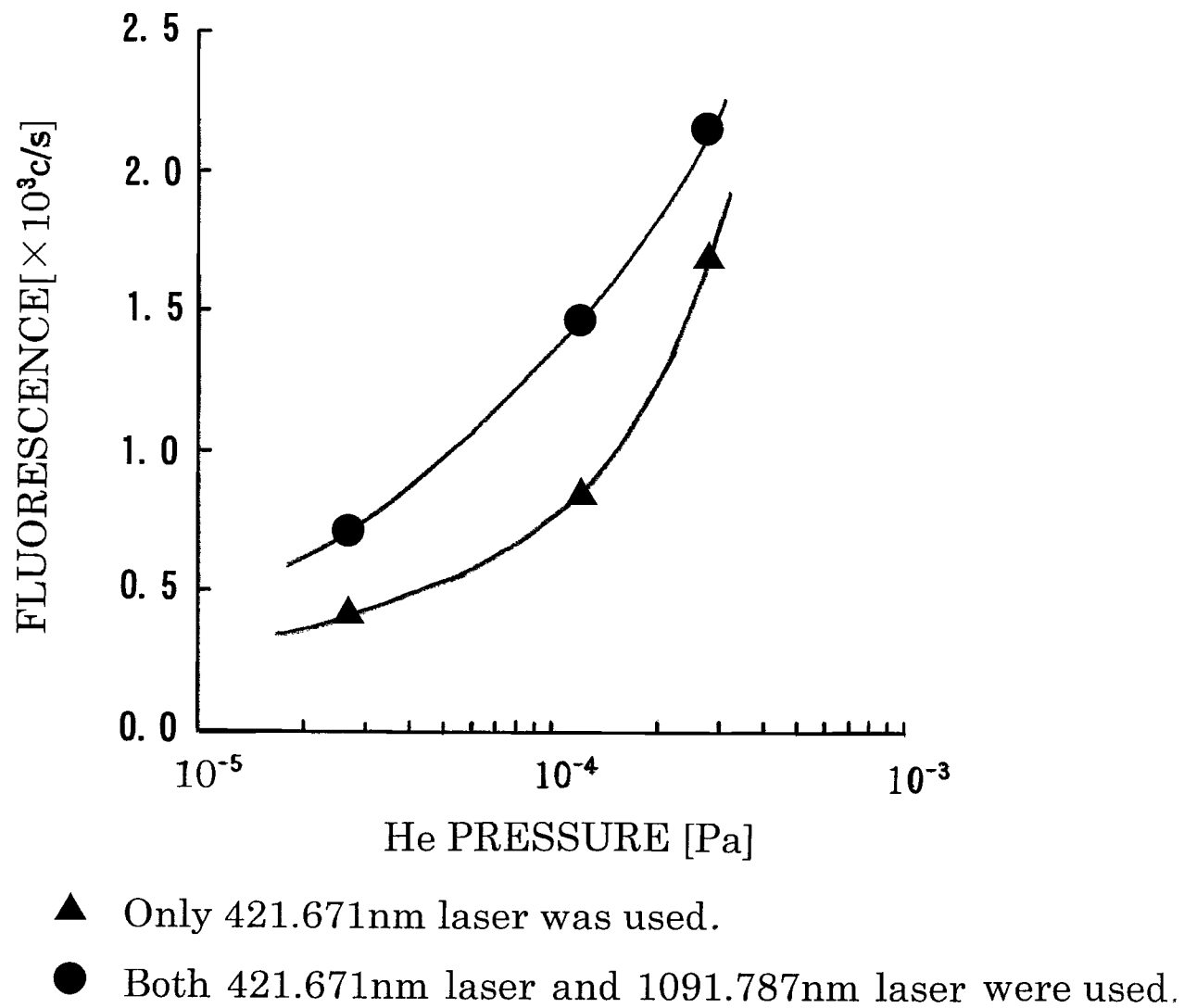


Fig. 3.11 He buffer gas pressure versus fluorescence intensity.

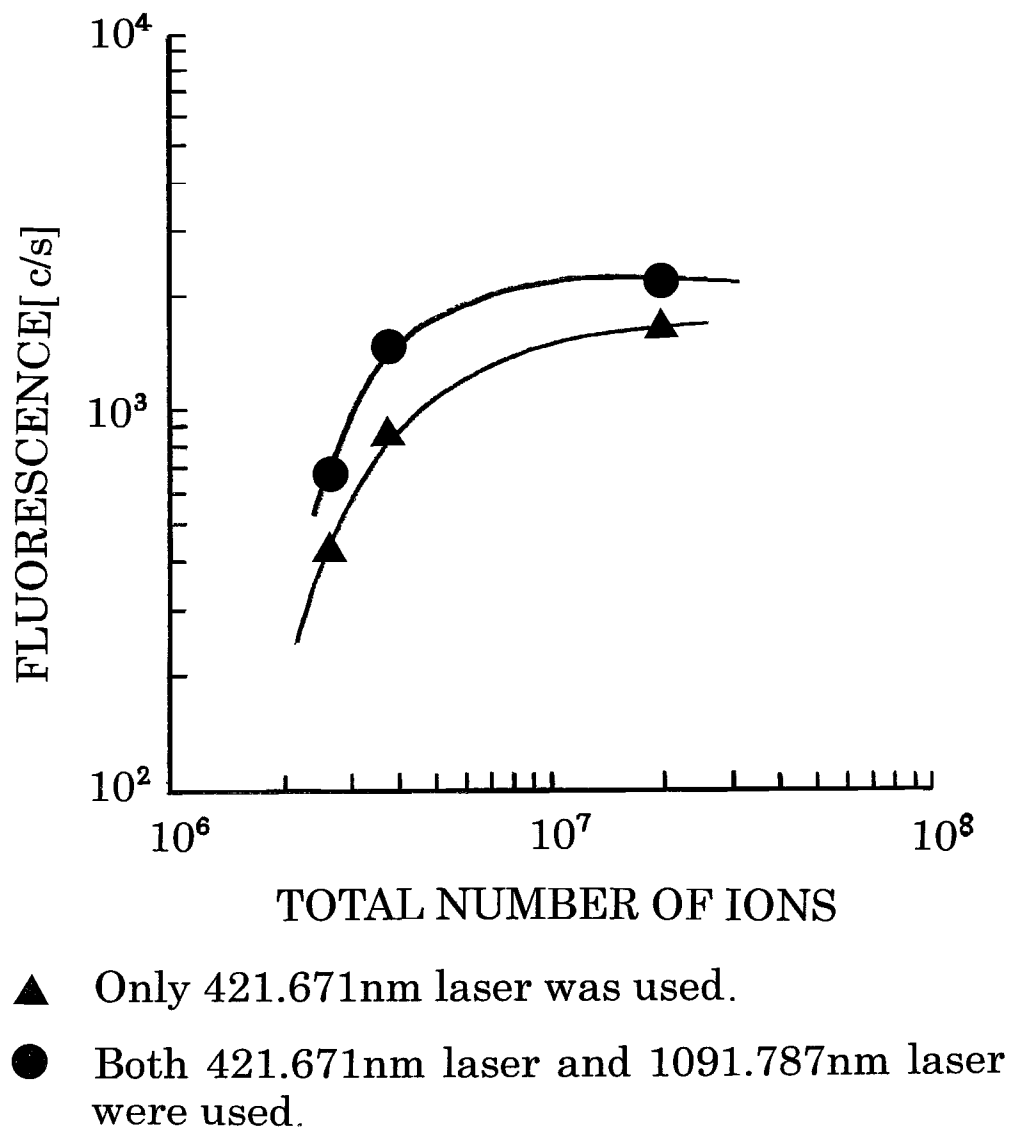


Fig. 3.12 Total number of trapped ions versus the fluorescence intensity.

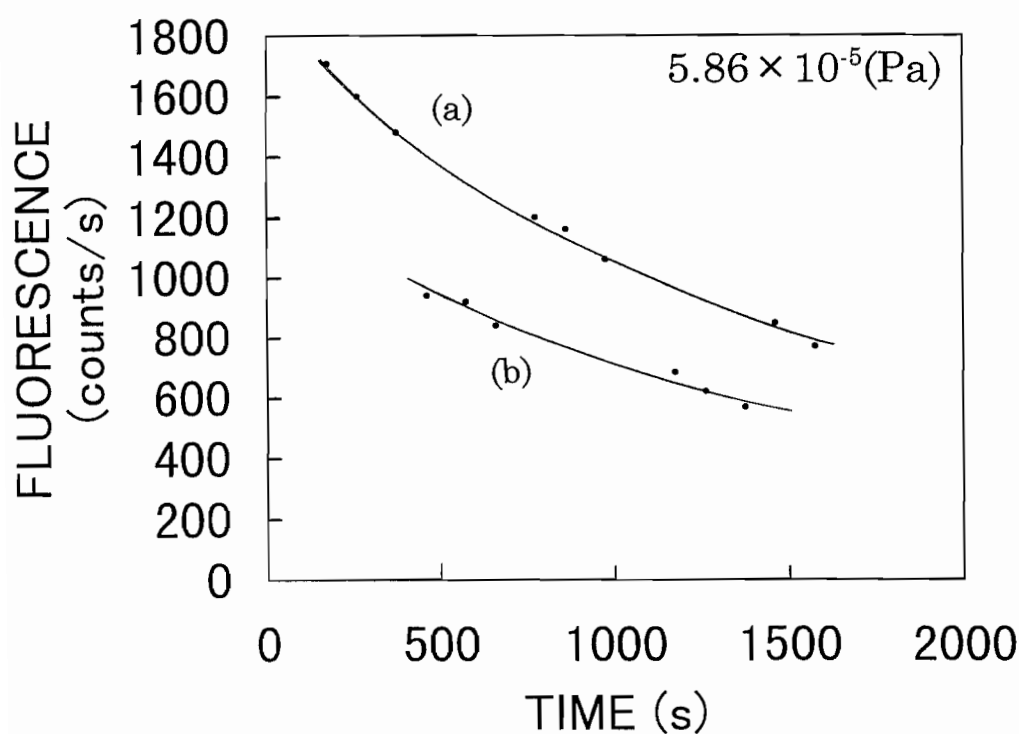
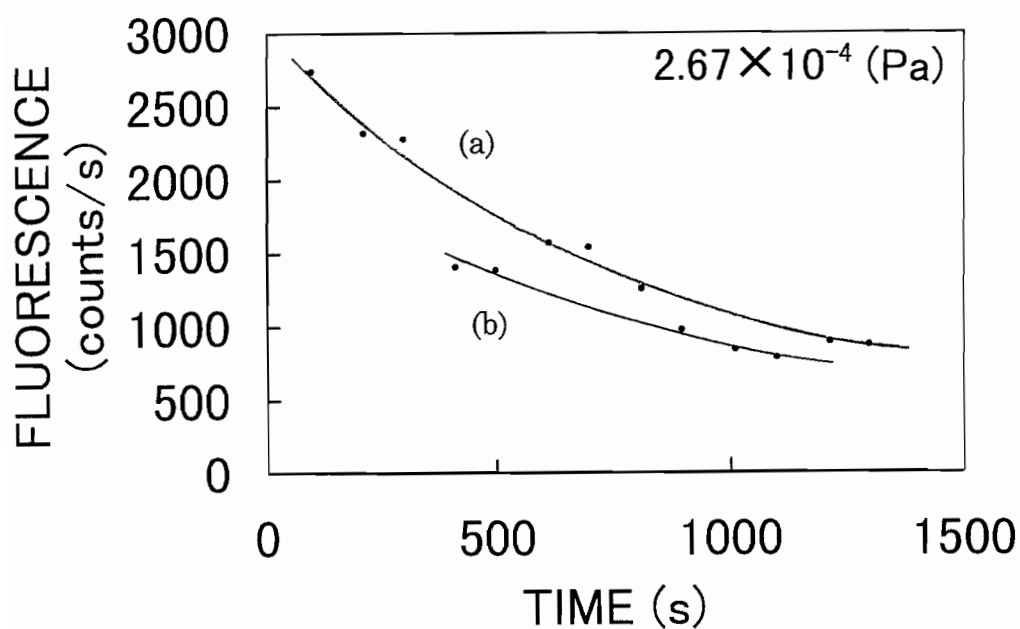


Fig. 3.13. Fluorescence decay curve when nitrogen is used as a buffer gas. (a) Illumination by both the 421.671 nm laser and the 1091.788 nm laser, (b) Illumination by the 421.671 nm laser alone.

Here  $W_1(f)$  and  $W_2(f)$  are the probabilities for transitions stimulated by the 421.671 nm and 1091.788 nm lasers, and  $A_1$ ,  $A_2$ , and  $A_3$  are the transition probabilities for spontaneous emission involving P-S, P-D, and D-S transitions, respectively. In addition, we introduce the quenching rate  $q$  for transitions mediated by collisions between the ions and the buffer gas that empty the  $4d^2D_{3/2}$  ion level and return the ion to the ground state. We do not consider the structure mixing due to the collisions with buffer gas since Gerz et. al. confirmed experimentally that there is no fine structure mixing between  $4d^2D_{3/2}$  and  $4d^2D_{5/2}$  of  $Sr^+$  due to a collision of He buffer gas<sup>8)</sup>. In the present work, the obtained value of  $r_2$  was larger than that of  $r_1$ , since the 1092nm laser light was more loose-focused at the trap center than the 422nm laser light as shown in Fig. 3.14. Because the 0.3 s lifetime of the  $4d^2D_{3/2}$  level is so long, the  $N_d$  and  $N_s$  populations are assumed to be distributed everywhere within the ion cloud. The fact that the  $N_d$  population is distributed everywhere within the ion cloud is corroborated by our measurements of the fluorescence Doppler width (about 2 GHz), which imply an ion temperature somewhat in excess of 1000 K, from which we conclude that the ion velocities are about 700 m/s. From this fact, and the fact that the lifetime of the  $4d^2D_{3/2}$  level is about 0.3 s, we can conclude that the population  $N_d$  of ions is distributed everywhere within the cloud. Because the excited-state lifetime is very short when the excited state is  $5p^2P_{1/2}$ , the spatial distribution of these ions in the region within the ion cloud that is illuminated by the repumping laser alone differs from their spatial distribution in the region illuminated by both the repumping laser and the pumping laser. This is because in the region illuminated by the repumping laser alone the only transfers between populations are between  $N_p$  and  $N_d$ . In contrast, we assert that transitions between both  $N_p$  and  $N_d$  and  $N_s$  and  $N_p$  take place in the region illuminated by both types of laser. This means that in order to compute the number of interactions with the pumping laser within  $N_p$ , it is necessary to introduce a coefficient  $r_3$ . This coefficient  $r_3$  is given by

$$r_3 = [r_1 W_1(f) N_s + \{r_1/r_2\} r_2 W_2(f) N_d] / \{r_1 W_1(f) N_s + r_2 W_2(f) N_d\}. \quad (3.4)$$

Moreover, we have not introduced the percentage  $r_2$  in the term  $W_2(f) N_p$  that appears in Eqs. (3.2) and (3.3), since the excited-state lifetime is very short when the excited state is  $5p^2P_{1/2}$ . Hence, we can assume that the population  $N_p$  is to a good approximation zero in regions where there is no illumination by the repumping laser. In these experiments, it was usually permissible to solve these equations under the following conditions:

A gate time of the photon counter of 1s is longer than  $(1/A_1)$  and  $(1/A_2)$

1.  $q, A_3 \ll W_1(f) \ll W_2(f) \ll A_2, A_1$  Here,  $A_3 = 2.69 s^{-1}$ <sup>8)</sup>,  $q$  is of the same order as  $A_3$ ,  $W_1(f) \doteq 1 \times 10^3 s^{-1}$ ,  $W_2(f) \doteq 2 \times 10^4 s^{-1}$ ,  $A_2 = 9.5 \times 10^6 s^{-1}$ <sup>1)</sup>, and  $A_1 = 1.27 \times 10^8 s^{-1}$ <sup>1)</sup>.
2.  $r_1$  and  $r_2$  are constant.  
We used the following equation to find  $W_1$ :

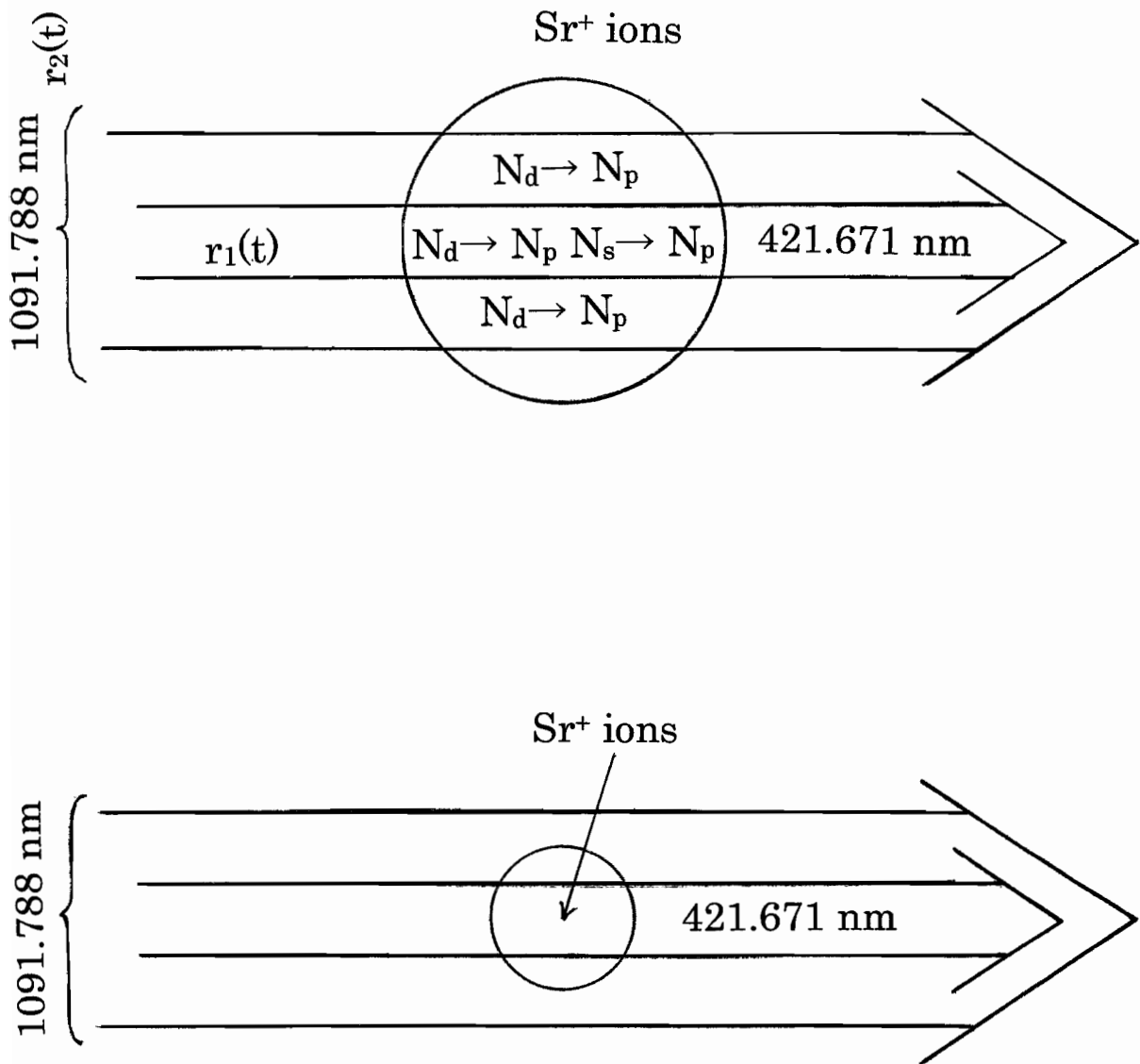


Fig. 3.14. Ions irradiated by two laser beams.

$$W_1(\omega_0) = \int \left[ \frac{(\ln 2 / \pi)^{0.5} / \Delta \omega_G \exp \{ -(\ln 2)(\omega - \omega_0)^2 / (\Delta \omega_G)^2 \}}{[(\Delta \omega_{\text{SHG}}) / \pi \{ (\omega_1 - \omega_0)^2 + (\Delta \omega_{\text{SHG}})^2 \}]} \right] \left[ \frac{(\lambda^2 / 4 \pi) c \rho \{ (\Delta \omega_L)^2 / \{ (\omega_1 - \omega)^2 + (\Delta \omega_L)^2 \} \}}{d \omega_1 d \omega} \right] d \omega_1 d \omega, \quad (3.5)$$

where  $c$  is the speed of light in vacuum,  $\rho$  [m<sup>-3</sup>] is the photon density,  $(\lambda^2 / 4 \pi)$  [m<sup>2</sup>] is the absorption cross section at the center of absorption angular frequency  $\omega_0$ ,  $\Delta \omega_L$  is the HWHM of the profiles for spectral lines of lifetime of 5p<sup>2</sup>P<sub>1/2</sub>,  $\Delta \omega_G$  is the HWHM of the Doppler width of Sr<sup>+</sup> ions, and  $\Delta \omega_{\text{SHG}}$  is the HWHM of the SHG linewidth. Since the 1092nm laser has multiple modes, eq.(3.5) is not applicable. In order to calculate  $W_2$ , we approximated the envelope of the mode spectrum by a Gaussian line shape. In the fiber laser many longitudinal modes are present, which are equally spaced on the frequency axis and fluctuate rapidly around the frequency of the 4d<sup>2</sup>D<sub>3/2</sub>-5p<sup>2</sup>P<sub>1/2</sub> ion transition in the same direction during the lifetime of the 4d<sup>2</sup>D<sub>3/2</sub> state. This assumption was supported by the fact that the fluorescence intensities did not fluctuate in the measurements. We calculated  $W_2$  by substituting the Gaussian profile for the Lorentzian laser spectrum in eq.(3.5). The solution is given in the Appendix A3.3 and  $N_p$  is given by

$$N_p = r_1 W_1 N / (13 A_2) \quad (3.6)$$

Here  $N = N_s + N_p + N_d$ . When repumping light is present, the number of photons measured with a photon counter is given by  $S_2 = A_1 N_p$ . If we take  $A_1 = 13 A_2$ ,  $S_2$  is given by

$$S_2 = r_1 W_1 N. \quad (3.7)$$

For the case of trapped ions illuminated with pump light only, we set  $W_2(f) = 0$  and use the fact that  $r_1 W_1, W_1 \ll A_2, A_1$  in the rate equation, which we then solve to obtain  $N_p$ . Doing so, we find that the photon number  $S_1$  is given by

$$S_1 = A_1 N_p = A_1 r_1 W_1 N / \{ A_1 + A_2 + r_1 W_1 A_2 / (A_3 + q) \}. \quad (3.8)$$

Therefore, the ratio  $R = S_2 / S_1$  of  $S_1$  and  $S_2$  is given by

$$R = (1 / A_1) \{ A_1 + A_2 + r_1 W_1 A_2 / (A_3 + q) \}. \quad (3.9)$$

Substituting the values of  $A_1$  and  $A_2$  into this gives

$$R = 1.077 + 0.0769 r_1 W_1 / (A_3 + q). \quad (3.10)$$

It was difficult to distinguish the values of  $r_1 W_1$  and  $A_3 + q$  when we determined the quenching rate of the 4d<sup>2</sup>D<sub>3/2</sub> metastable state for He gas by the observed  $R$  value, hence we assumed the following conditions. We first assumed that the collisional quenching rate

was proportional to the buffer gas pressure and second that  $N$  and  $r_1 W_1$  were almost the same values at the same fluorescence counting rate even if the pressure of buffer gas was different. Then we could obtain the following equations from He buffer gas at the fluorescence intensity = 1200c/s for  $2.67 \times 10^{-4}$ Pa,  $1.33 \times 10^{-4}$ Pa and  $2.67 \times 10^{-5}$ Pa, respectively,

$$1.077 + 0.0769 r_1 W_1(f)/(A_3 + 10q) = 1.58 \quad \text{at} \quad 2.67 \times 10^{-4} \text{Pa} \quad (3.11)$$

$$1.077 + 0.0769 r_1 W_1(f)/(A_3 + 5q) = 1.70 \quad \text{at} \quad 1.33 \times 10^{-4} \text{Pa} \quad (3.12)$$

$$1.077 + 0.0769 r_1 W_1(f)/(A_3 + q) = 1.85 \quad \text{at} \quad 2.67 \times 10^{-5} \text{Pa} \quad (3.13)$$

where we assume that the quenching rate was  $q$  at the pressure of  $2.67 \times 10^{-5}$  Pa. From these upper equations we can obtain 3 pairs of simultaneous equations. By solving these equations, we obtained  $q = 0.17 \pm 0.01 \text{ s}^{-1}$ . In the equation (3.13) we used  $q$  for the quenching rate and in the equation (3.11) we used  $10q$  for the quenching rate, since the quenching rate is proportional to the pressure of the buffer gas. Above mentioned reason we calculated the quenching coefficient  $R_q$  by  $q \div 2.67 \times 10^{-5} \text{ Pa}$ . This means that the quenching coefficient is  $R_q = 6400 \text{ s}^{-1} \text{Pa}^{-1}$ . (The difference in the pressure measured by the discharge current of the ion pump and that of a Bayard-Alpert ionization gauge is less than a factor 10.) The same calculations were carried out at 1000c/s, and almost identical value of  $R_q$  was obtained. For  $N_2$ , we compared the peak intensities at a counting rate of 1000 counts/s to get  $R$ . They are 1.268, 1.366 and 1.482 for the pressures of  $2.67 \times 10^{-4}$  Pa,  $1.33 \times 10^{-4}$  Pa and  $5.86 \times 10^{-5}$  Pa, respectively. From these values, we obtained a value of  $2 \times 10^4 \text{ s}^{-1} \text{Pa}^{-1}$  for  $R_q$ . From this value we obtain a value of  $0.83 \times 10^{-16} \text{ m}^3 \text{s}^{-1}$  as the quenching rate constant for the  $4d^2 D_{3/2}$  of  $\text{Sr}^+$  ions due to collisions with  $N_2$ , where the temperature of the buffer gas was assumed to equal the room temperature ( $T=300 \text{ K}$ ). Comparing the ratios for the peak intensities at 800 c/s for  $\text{CH}_4$ , we obtained  $5.6 \times 10^4 \text{ s}^{-1} \text{Pa}^{-1}$  and  $2.3 \times 10^{-16} \text{ m}^3 \text{s}^{-1}$  for quenching coefficient and the quenching rate constant for  $\text{Sr}^+ \cdot \text{CH}_4$ , respectively.

From the relation of

$$q = (n_{\text{He}}) \sigma \langle v_{\text{He}} \rangle, \quad (3.14)$$

the collisional cross section can be calculated. Where  $q$  is the quenching rate for  $\text{Sr}^+ \cdot \text{He}$ ,  $(n_{\text{He}})$  is the density of He atoms,  $\sigma$  is the collisional cross section and  $\langle v_{\text{He}} \rangle$  is the mean speed of He atoms. From this, the cross section of He quenching  $2.1 \times 10^{-20} (\text{m}^2)$  is derived. This order of the magnitude agrees with the value of the cross section calculated from the diameter of He. The quenching coefficient for the  $4d^2 D_{3/2}$  metastable state in  $\text{Sr II}$  for  $\text{H}_2$  was determined to be  $80700 \text{ s}^{-1} \text{Pa}^{-1}$  by Gerz et al.<sup>9)</sup> Our value for He is one thirteenth of that determined by them. Since the noble gases do not have the vibrational and rotational structures, this fact is not surprising. For He atom the first excited electronic state transitions are in the regions of extra ultraviolet and vacuum ultraviolet. The first excited

electronic state transitions of He is ten times higher than the  $4d^2D_{3/2}$  metastable state of  $Sr^+$  ion. When the collision occur between the  $Sr^+$  ion in the  $4d^2D_{3/2}$  metastable state and the He buffer gas, the excited energy of  $4d^2D_{3/2}$  metastable state dose not transfer to the first excited electronic state transitions of He atom<sup>9,10</sup>. The excited energy of  $4d^2D_{3/2}$  metastable state transfer only to the kinetic energy of He atom. However, our value might be somewhat higher than the quenching rate constant of the  $5d^2D_{5/2}$  metastable state in a single  $Ba^+$  ion for  $H_2$  and He obtained by Madej and Sankey<sup>10</sup>. From eqs.(3.11)and(3.12),  $r_1 W_1(f)$  was calculated to be  $27.6s^{-1}$ . On the other hand, from eq.(3.5)  $W_1(f)$  was calculated to be  $1000s^{-1}$ . Accordingly  $r_1$  was estimated to be 0.028 and from the ratio of the focus areas between two laser beams,  $r_2$  was estimated to be 0.22. Although alignment of the laser beam was difficult, if we assume that the laser light hits at the center of the ion cloud, from this value and the diameter of the laser beam light of 1.8mm, the diameter of the ion cloud was calculated to be about 10mm.

It is interesting to compare observed quenching rates to the Langevin reaction rate coefficient between  $Sr^+$  ions and atoms or molecules of buffer gases. We calculated the Langevin reaction rates using the classical ion-molecule collision theory. In this theory, when an ion approaches an atom or a molecule of the buffer gas, the ion induces a dipole moment on the atom or the molecule. Considering the mutual effect between the ion and this induced dipole moment, we can calculate the cross section of the collision. Then the Langevin rate constant  $k$  is given by<sup>10</sup>

$$k = \langle \sigma v \rangle = \{2 \pi q_{ion} / (4 \pi \epsilon_0)\} (\alpha / \mu)^{1/2}, \quad (3.15)$$

where  $\sigma$  is the cross section,  $v$  is the mean velocity of ions,  $q_{ion}$  is the charge of the ion,  $\epsilon_0$  is the permittivity of vacuum,  $\alpha$  is the polarizability and  $\mu$  is the reduced mass of the  $Sr^+$ -buffer gas molecule system. The results are shown in Table 3.3. The values in the column ratio were obtained by dividing each quenching rate by the corresponding Langevin reaction rate. A value of 0.05 for  $Sr^+ \cdot He$  indicates that one quenching event for a  $Sr^+$  ion in the  $^2D_{3/2}$  state is induced after 20 collisions with He atoms. For a He atom, the first excited electric state has extra ultraviolet and vacuum ultraviolet transition energies from the ground state. Therefore the energy of the  $Sr^+$  ions in the  $^2D_{3/2}$  state ( $14556 \text{ cm}^{-1}$ ) would not easily transfer to He atoms. However,  $N_2$  and  $CH_4$  molecules have many degrees of freedom, and thus the energy transfer from the  $Sr^+$  metastable state to the molecular energy levels is easier. Therefore these molecules have higher quenching rates. However, He gas is the most effective at reducing the temperature of the cloud of trapped ions.

### 3.7 Conclusion

In this chapter, the development of an ion trap system for confining ions in a vacuum for a long time is described. The ion trap was designed so that many ions are confined within the Lamb-Dicke region. As a result, the number of confined ions was in the

| Gas             | Polarizability<br>( $10^{-30} / (4 \pi \epsilon_0) \text{m}^3$ ) | Quenching<br>rate constant<br>( $10^{-16} \text{m}^3 \text{s}^{-1}$ ) | Langevin rate<br>constant<br>( $10^{-16} \text{m}^3 \text{s}^{-1}$ ) | Ratio | Temperature (K) |
|-----------------|------------------------------------------------------------------|-----------------------------------------------------------------------|----------------------------------------------------------------------|-------|-----------------|
| He              | 0.2                                                              | 0.27                                                                  | 5.3                                                                  | 0.05  | 400             |
| N <sub>2</sub>  | 1.7                                                              | 0.83                                                                  | 6.6                                                                  | 0.13  | 1100            |
| CH <sub>4</sub> | 2.6                                                              | 2.3                                                                   | 10                                                                   | 0.23  | 1200            |

Table 3.3. Comparison of the observed  $4\text{d-}^2\text{D}_{3/2}$  quenching rate constants with calculated Langevin reaction rates for He, N<sub>2</sub> and CH<sub>4</sub>.

range of  $2.5 \times 10^6 \sim 2 \times 10^7$ , the fluorescence intensity was 2000 c/s or higher, the sufficient S/N for detecting double resonance fluorescence signals was realized and the elimination of the first-order Doppler shift was realized in the 98% of the trapped ions. Further, we found He gas was effective in reducing the kinetic energy of the trapped ions. The ions were cooled to 400 K at a He pressure  $2.7 \times 10^{-4}$  K which reduced the second order of the Doppler shift below  $5 \times 10^{-13}$  in the microwave region.

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## Appendix

### A3.1 Motion of a Positive Ion

When a single ion experiences the quadrupole field, there is no space charge due to the presence of other charged particles. Employing cylindrical coordinate  $r$ ,  $z$ , weighting constants  $\alpha$ ,  $b$  and  $c$ , the potential  $\phi$  at any point  $(r, z)$  within the device is assumed to be given by

$$\phi(r, z) = \alpha r^2 + b z^2 + c. \quad (3A.1)$$

$\phi$  must satisfy Laplace equation given by

$$(\delta^2 \phi / \delta r^2) + (1/r)(\delta \phi / \delta r) + (\delta^2 \phi / \delta z^2) = 0. \quad (3A.2)$$

The relation given by  $4\alpha + 2b = 0$  is obtained from (3A.2), and then

$$\phi(r, z) = \alpha r^2 - 2\alpha z^2 + c. \quad (3A.3)$$

When the end caps are earthed and  $\phi_0$  is applied to the ring electrode only, the following relationships are obtained

$$\phi(r_0, 0) = \phi_0 = \alpha r_0^2 + c \quad (3A.4)$$

$$\phi(0, z_0) = 0 = -2\alpha z_0^2 + c \quad (3A.5)$$

where  $r_0$  is the minimum inner radius of the ring electrode and  $z_0$  is the minimum half-distance between the two endcap electrodes.

Using the above relations, the potential  $\phi$  is given by

$$\phi(r, z) = \alpha (r^2 - 2z^2 + 2z_0^2) \quad (3A.6)$$

$$= \phi_0 (r^2 - 2z^2 + 2z_0^2) / (r_0^2 + 2z_0^2). \quad (3A.7)$$

Introducing the relationship  $r_0^2 = 2z_0^2$  which has historically governed the physical shape of the ion trap

$$\phi(r, z) = \phi_0 (r^2 - 2z^2) / 2r_0^2 + \phi_0 / 2. \quad (3A.8)$$

The field is uncoupled in the  $r$  and  $z$  directions, and so the forces may be

determined separately. The force in the  $r$  and  $z$  directions,  $F_r$  and  $F_z$  are expressed as

$$F_r = m(d^2r/dt^2) = -e (\phi_0/r_0^2)r \quad (3A.9)$$

$$F_z = m(d^2z/dt^2) = e(2\phi_0/r_0^2)z \quad (3A.10)$$

where  $m$  is the mass of the ion.

The applied potential is a combination of a radio frequency potential  $V \cos \omega t$  and a direct current potential  $U$  of the form

$$\phi_0 = V \cos \omega t + U \quad (3A.11)$$

where the symbol  $\omega$  represents the angular frequency of an rf field. Substituting equation (3A.11) into equations (3A.9) and (3A.10) leads to the equations of motion of a singly charged positive ion in an ion trap.

$$m(d^2r/dt^2) = -e \{(V \cos \omega t + U)/r_0^2\}r \quad (3A.12)$$

$$m(d^2z/dt^2) = e \{2(V \cos \omega t + U)/r_0^2\}z. \quad (3A.13)$$

An example of the ion trajectory and ion speed, numerically calculated from eq.(3A.12) and (3A.13) is shown in Figure 3A.1 and Figure 3A.2 respectively.

There exists in the literature a second-order linear differential equation known as Mathieu equation, which was originally developed by Mathieu while investigating the motions of a vibrating membrane. Solutions of this equation and applications have been studied in detail by McLachlan<sup>11)</sup>. The final form conforming to the Mathieu equation of (3A.13) is rewritten as

$$(d^2z/d\xi^2) + (a_z - 2q_z \cos 2\xi)z = 0 \quad (3A.14)$$

where  $\xi = \omega t/2$ ,  $a_z = -8eU/(m\omega^2 r_0^2)$ ,  $2q_z = 8eV/(m\omega^2 r_0^2)$ . The solution of (3A.14) form the infinite series

$$z = A \sum C_{2n} \cos(2n + \beta_z) \xi + B \sum C_{2n} \sin(2n + \beta_z) \xi. \quad (3A.15)$$

In the solution, A and B are governed by initial conditions and  $C_{2n}$  and  $\beta_z$  are functions of  $a_z$  and  $q_z$ . If  $a_z$  and  $q_z < 1$  we can simplify  $\beta_z \doteq (a_z + q_z^2/2)^{0.5}$ . In the same way, we can solve  $r$  direction and if  $a_r$  and  $q_r < 1$ ,  $\beta_r \doteq (a_r + q_r^2/2)^{0.5}$ . To increase the signal to noise ratio in a

## Trajectories of the $^{87}\text{Sr}^+$ ion

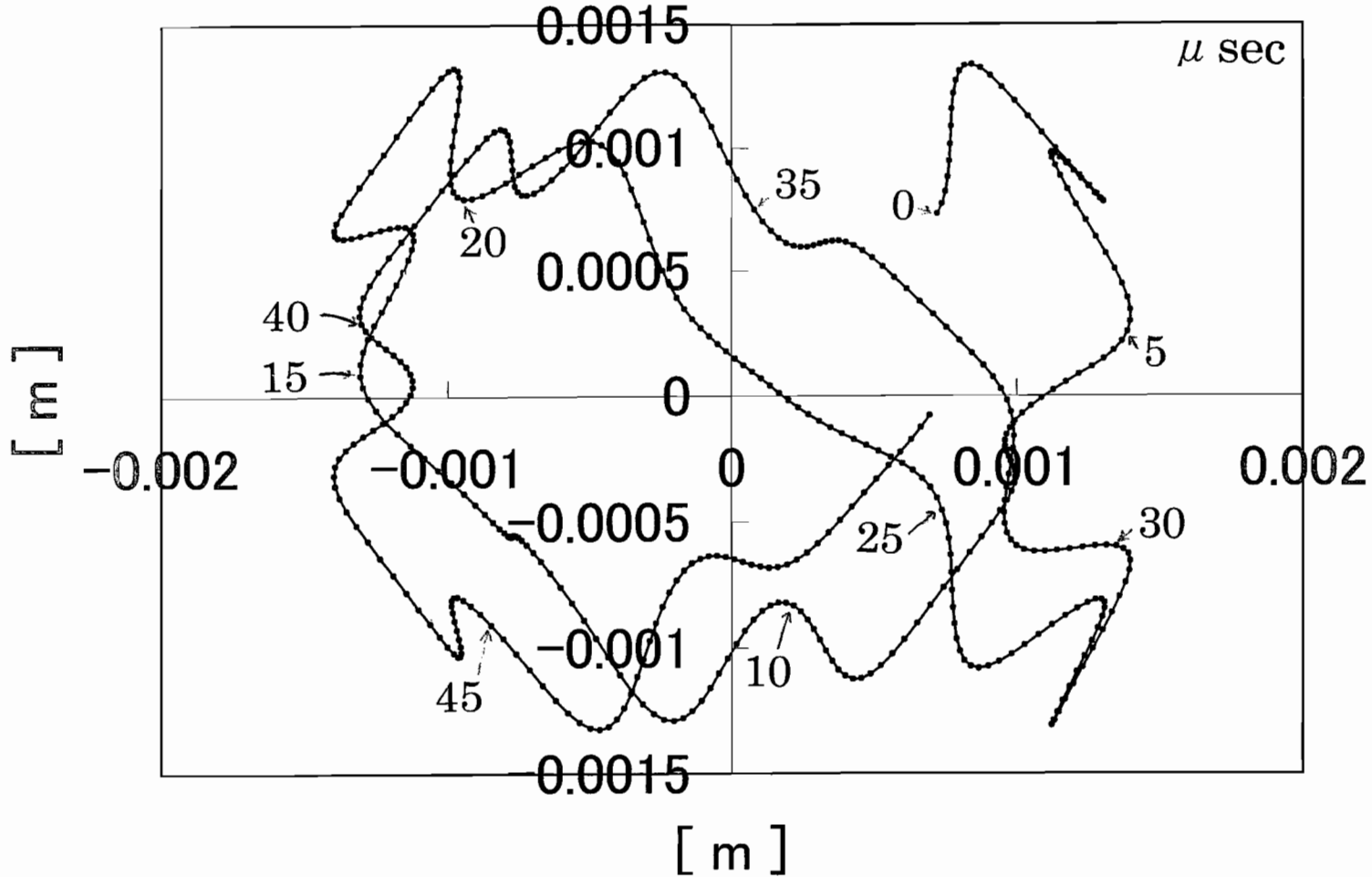


Fig. 3A.1. Trajectories of a  $^{87}\text{Sr}^+$  ion.  $V = 256 \text{ V}$ ,  $U = 7.8 \text{ V}$ ,  $f = 400 \text{ kHz}$ ,  $r_0 = 20 \text{ mm}$ ,  $z_0 = 14.1 \text{ mm}$ . Initial values are  $r = 0.7 \text{ mm}$ ,  $z = 0.7 \text{ mm}$ ,  $v_r = 245 \text{ m/s}$ ,  $v_z = 245 \text{ m/s}$ . The values 0, 5, 10 ..... are the time in  $\mu \text{ s}$ .

# Speed of $^{87}\text{Sr}^+$ ion

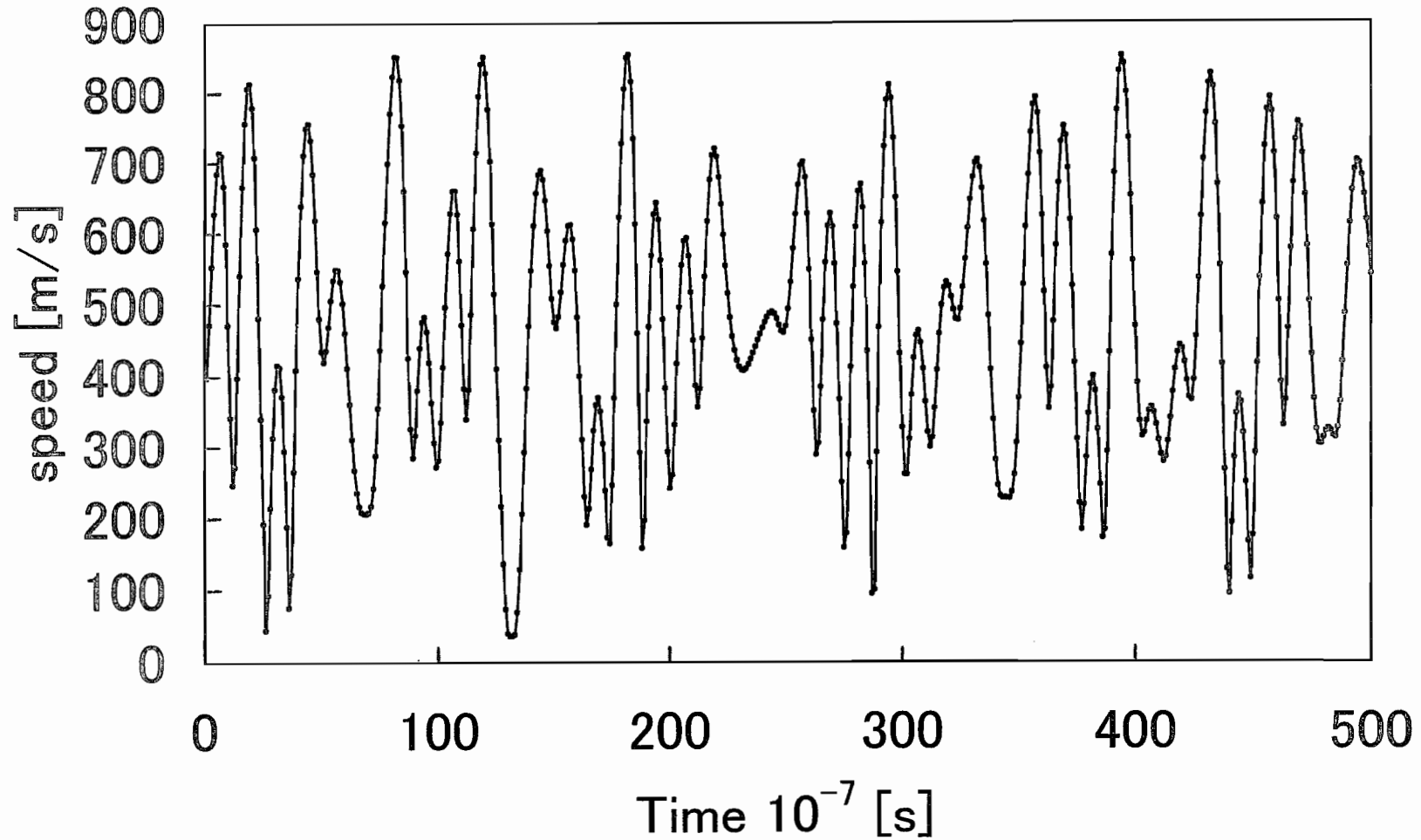


Fig. 3A.2. Speed of a  $^{87}\text{Sr}^+$  ion.  $V = 256$  V,  $U = 7.8$  V,  $f = 400$  kHz,  $r_0 = 20$  mm,  $z_0 = 14.1$  mm Initial values are  $r = 0.7$  mm,  $z = 0.7$  mm,  $v_r = 245$  m/s,  $v_z = 245$  m/s.

spectroscopic experiment it is essential to find the trap parameters to obtain maximum ion density. Figure 3A.3 represents the stable region of the Mathieu equation (3A.14). The theoretical predictions of the ion trap parameters are possible by this diagram. Ions can be stored in the ion trap only if they are stable in both the  $r$  and  $z$  directions simultaneously. Iffländer and Werth calculated and experimentally confirmed the optimum parameters in the stability diagram to increase of the number of trapped ions as many as possible, and their experimental results are  $a_z = -0.03 \pm 10\%$  and  $q_z = 0.55 \pm 15\%$ <sup>12)</sup>. We usually set our trap parameters close to these values.

### A3.2 Total Number of Trapped Ions

The equation of the macro motion of the trapped ions along the  $z$  axis is given by the forced harmonic oscillation equation

$$(d^2z/dt^2) + \gamma_z(dz/dt) + \omega_z^2 z = [\Gamma e v_1 / (2mz_0)] \quad (3B.1)$$

where  $\gamma_z$  is a reciprocal of the phase coherence time of the oscillatory response of the ions,  $\Gamma$  is a compensating factor due to the fact that the mean field is not simply given by  $v_1/2z_0$ ,  $e$  is the charge  $= +1.602 \times 10^{-19}$  C,  $v_1$  is the probing voltage appearing on the quadrupole end-electrodes,  $\omega_z$  is the characteristic ion angular frequency, and  $m$  is the mass of the ion. We can obtain (3B.1) from adding the electrical field force term to (3A.13).

$$m(d^2z/dt^2) = -e \{2(V \cos \omega t + U)/r_0\} z + [\Gamma e v_1 / (2mz_0)]. \quad (3B.2)$$

Since the mean current is given by

$$i = (Ne/2z_0)(dz/dt) \quad (3B.3)$$

where  $N$  is the total number of ions, (3B.1) is rewritten as

$$L_i(d^2q/dt^2) + R_i(dq/dt) + q/C_i = v_1(t) \quad (3B.4)$$

where  $q$  is the charge of the equivalent capacitance  $C_i$ ,  $L_i = (m/N)(2z_0/\Gamma e)^2$  and  $R_i = (m/N)\gamma_z(2z_0/\Gamma e)^2$ ,  $1/C_i = (m/N)\omega_z^2(2z_0/\Gamma e)^2$ .

Therefore, the oscillating ion system is equivalent to an LRC series resonant circuit with the above defined  $L_i$ ,  $R_i$  and  $C_i$ . The relative ion-response signal  $Y$  is defined as

$$Y = (v_{10} - v_1)/v_{10} \quad (3B.5)$$

where  $v_{10}$  is the value of  $v_1$  at the resonance frequency of the parallel circuit containing the  $L, C$  and trap capacitance when there is no ion in the trap.

We assume that the following conditions are fulfilled : (1) the parallel L C circuit

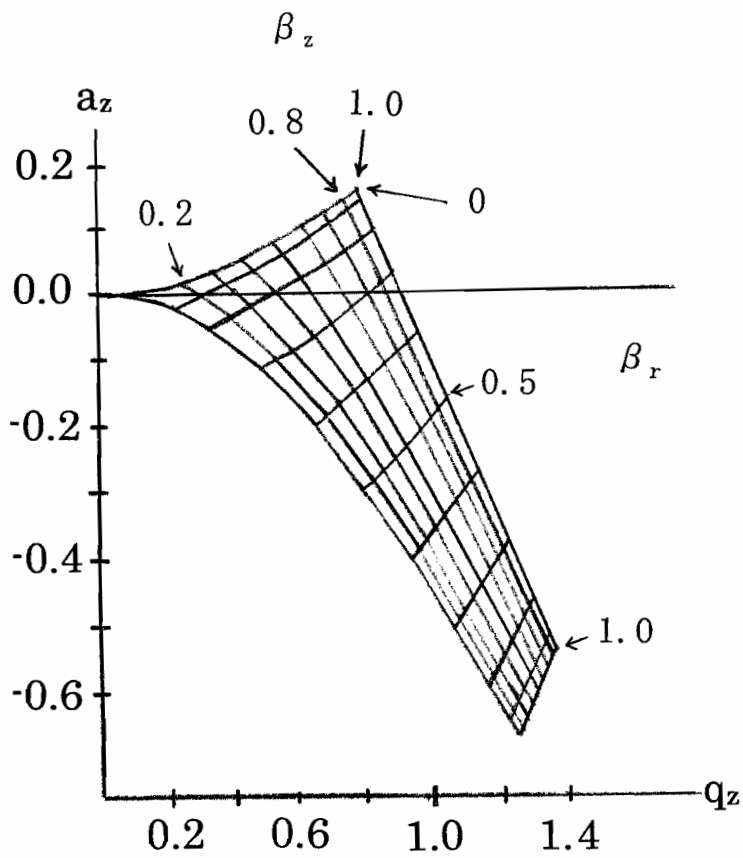


Fig. 3A.3, Stability region near the origin for the three-dimensional ion trap. The iso- $\beta$  lines are shown.  $\beta_z \equiv (a_z + q_z^2/2)^{0.5}$ ,  $\beta_r \equiv (a_r + q_r^2/2)^{0.5}$ .

which includes the capacitance of the trap, is tuned to the same frequency as the series  $L_i C_i R_i$  equivalent circuit.; and (2) the variation of the admittance of the L C circuit can be neglected in the frequency range of interest, where the imaginary part of the  $L_i C_i R_i$  circuit admittance is smaller than the real part. The admittance of the L C circuit can then be approximated by  $G_0$ , its real value at resonance. It is then easy to show that the following expression is obtained for  $v_1$ :

$$v_1 = \{v_0 - R_f(dq/dt)\} / (1 + R_f G_0) \quad (3B.6)$$

where  $v_0$  is the applied source and  $R_f$  is the high resistance which is inserted between trap and applied source.  $R_f$  corresponds to  $R_0$  in Fig. 3.5.

Substituting this into equation (3B.4) gives

$$L_i(d^2q/dt^2) + [R_i + R_f/(1 + R_f G_0)](dq/dt) + q/C_i = v_0/(1 + R_f G_0). \quad (3B.7)$$

Now suppose that the source voltage changes according to the equation

$$v_0 = V_0 \exp[j(\omega_z + \mu t)t] \quad (3B.8)$$

where  $\mu$  is the sweep rate of the angular frequency.

In the case where

$$\mu (2/\gamma_z) \ll \Delta \omega$$

where  $\Delta \omega$  is the line width at half-amplitude, the problem may be solved as if  $\omega$  is constant. Let  $Q$  be the complex amplitude of the charge  $q(t)$ , the equation of motion for  $Q$  becomes

$$-\omega^2 L_i Q + j\omega [R_i + R_f/(1 + R_f G_0)]Q + Q/C_i = V_0/(1 + R_f G_0). \quad (3B.9)$$

From this it may be deduced immediately that the damping term  $\gamma$  is given by

$$\gamma = \gamma_z + \gamma_1 \quad (3B.10)$$

where

$$\gamma_1 = (N/m)(\Gamma e/2z_0)^2 R_f/(1 + R_f G_0). \quad (3B.11)$$

The quantity  $\gamma_1$  describes the damping of the oscillation due to coupling to the external resistance. It depends on the ion population  $N$  as well as the coupling strength and the external resistance.

The relative ion-response signal  $Y$  is readily found to be

$$Y = 1 - [(X_i^2 + R_i^2) / \{X_i^2 + (R_i + R'_f)^2\}]^{1/2} \quad (3B.12)$$

where

$$R'_f = R_f / (1 + R_f G_0) \quad (3B.13)$$

$$X_i = \omega L_i - 1 / \omega C_i . \quad (3B.14)$$

The signal shape is symmetrical around the resonant frequency, with a peak value given by

$$Y_{max} = R'_f / (R_i + R'_f) = 1 / (1 + \gamma_z / \gamma_1) . \quad (3B.15)$$

Taking into account the proportionality of  $\gamma_1$  to  $N$ , the peak relative signal is not linearly dependent on the ion population, but rather  $Y_{max}$  saturate as  $N$  increases. This, of course, is to be expected, since  $Y = 1$  corresponds to 100% amplitude modulation of the detected RF signal. From equation (3B.14) the half-amplitude full width  $\Delta \omega$  of the relative signal is obtained as:

$$\Delta \omega = \gamma [(2 - 1.5 Y_{max}) / (2 - 0.5 Y_{max})]^{1/2} = \gamma b . \quad (3B.16)$$

The value of  $b$  is close to 1. It is minimum for  $Y_{max} = 1$  and equals 0.58.

Of particular interest is the absolute determination of the ion population number  $N$  in terms of the observables of the system. It is readily verified that while  $Y_{max}$  and  $\Delta \omega$  are both expected to be fairly complicated functions of  $N$ , their product  $Y_{max} \Delta \omega$  is simply proportional to  $N$ :

$$Y_{max} \Delta \omega = N (\Gamma^2 e^2 / 4 m z_0^2) [R_f / (1 + R_f G_0)] b . \quad (3B.17)$$

### A3.3 Steady-State Solution of the Rate Equations

$$-W_1 r_1 N_s + (A_1 + W_1 r_3) N_p + (A_3 + q) N_d = 0 \quad (3C.1)$$

$$+ W_1 r_1 N_s - (A_1 + W_1 r_3 + A_2 + W_2) N_p + W_2 r_2 N_d = 0 \quad (3C.2)$$

$$+ (A_2 + W_2) N_p - (W_2 r_2 + A_3 + q) N_d = 0 \quad (3C.3)$$

$$r_3 = \{r_1 W_1 N_s + (r_1 / r_2) r_2 W_2 N_d\} / \{r_1 W_1 N_s + r_2 W_2 N_d\} \quad (3C.4)$$

From (3C.3)

$$N_d = \{(A_2+W_2)/(W_2r_2+A_3+q)\}N_p. \quad (3C.5)$$

Substituting (3C.4) into (3C.2), we obtain

$$\begin{aligned} \{r_1W_1N_s\}^2 + 2r_1r_2W_1W_2N_dN_s - \{(A_1+A_2)r_1W_1 + r_1W_1W_2 + r_1(W_1)^2\}N_sN_p - \\ \{(A_1+A_2)r_2W_2 + r_2(W_2)^2 + r_1W_1W_2\}N_dN_p + (r_2W_2N_d)^2 = 0. \end{aligned} \quad (3C.6)$$

From the relation of  $N = N_s + N_p + N_d$  and (3C.5),

$$N_s = N - \{1 + (A_2+W_2)/(W_2r_2+A_3+q)\}N_p. \quad (3C.7)$$

In order to simplify the equations (3C.5), (3C.6) and (3C.7), the following notations are introduced.

$$\begin{aligned} \alpha &= 1 + (A_2+W_2)/(W_2r_2+A_3+q) \doteq A_2/W_2r_2, \\ \beta &= (A_1+A_2)r_1W_1 + r_1W_1W_2 + r_1(W_1)^2 \doteq (A_1+A_2)r_1W_1, \\ \gamma &= (A_1+A_2)r_2W_2 + r_2(W_2)^2 + r_1W_1W_2 \doteq (A_1+A_2)r_2W_2, \\ \delta &= (A_2+W_2)/(W_2r_2+A_3+q) \doteq A_2/W_2r_2. \end{aligned}$$

From (3C.5), (3C.6) and (3C.7), we obtain

$$\{r_1W_1N_s\}^2 + 2r_1r_2W_1W_2N_dN_s - \beta N_sN_p - \gamma N_dN_p + (r_2W_2N_d)^2 = 0, \quad (3C.8)$$

$$N_s = N - \alpha N_p, \quad (3C.9)$$

$$N_d = \delta N_p. \quad (3C.10)$$

Substituting (3C.9), (3C.10) into (3C.8) and using the condition  $r_1W_1 \ll r_2W_2$ , we obtain the equation for  $N_p$ .

$$13(A_2)^2(N_p)^2 + 12r_1W_1A_2N_p - (r_1W_1)^2\{N\}^2 = 0. \quad (3C.11)$$

Then, the solution of the equation is given by

$$N_p = r_1W_1N/(13A_2). \quad (3C.12)$$

## Chapter 4

# Laser Diode Source for Exciting Ca Atoms

### 4.1 Introduction

By virtue of their much higher oscillation frequencies, optical frequency standards have tremendous potential for improvement over their microwave counterparts, which have served as the primary standards for 50 years. In order to realize Ca stabilized laser to the one of the primary standard of time and frequency, the Ca clock, we have developed a cascade master/slave/slave injection-locking spectrometer as we prepare this system for use as a cooling laser light for Ca atoms. As for Ca atoms, the natural linewidth of the transition at 657 nm is as narrow as 400 Hz, and the hyperfine structural transition is affected only slightly by external fields. So, the research groups such as Physikalisch-Technische Bundesanstalt (PTB) in Germany and NIST have used the transition of Ca atoms at 657 nm as an optical frequency standard. However, in the field of frequency standards, it is also important to develop standards with different designs and compare mutually the output frequency of each standard to improve their reliability. In addition, establishing national standards for optical frequency is significant in related precision fundamental sciences. One of the biggest technical challenges in building a diode-based magneto-optic trap for Ca is the generation of single-frequency, tunable radiation at 423 nm with sufficient power<sup>1)</sup>.

In this Chapter, a master/slave/slave laser spectrometer comprising a master/slave laser combination coupled with a SHG external ring cavity and the SHG injected blue diode laser are described. We have developed a blue master/slave/slave laser for cooling Ca atoms. It consists of an infrared external cavity diode laser (master laser), a high-power solitary diode laser (slave laser), the frequency of which is locked to that of the master laser by an injection seeding technique, and KNbO<sub>3</sub> crystals for SHG, which are mounted in a power build-up(enhancement) cavity. The light from the slave laser is injected into the KNbO<sub>3</sub> crystals. By injecting the output light obtained by SHG into a blue diode laser, a master/slave/slave laser light source in the blue-wavelength region was developed.

### 4.2 Ca Atom

Neutral atom/molecule candidates have the potential for extremely high stability due to the large number of participating atoms/molecules, and they should achieve good accuracy as well. Due to their narrow linewidths and insensitivity to external perturbations, the intercombination lines of the alkaline earth atoms are among the most promising and practical

neutral candidates. One of the selling points of Ca-based system is that the cooling and clock wavelengths are accessible with diode laser systems, leading to the possibility of a simple, compact, and ultimately transportable apparatus. The energy level diagram of Ca atom is shown in Fig. 4.1. The 657nm  $^1S_0$  ( $m = 0$ )  $\rightarrow$   $^3P_1$  ( $m = 0$ ) transition in Ca is particularly attractive due to its narrow natural linewidth (408 Hz) and convenient laser wavelengths for spectroscopy and cooling. Since the level structures of both  $^1S_0$  and  $^3P_1$  states are simple and have a magnetically insensitive  $M = 0$  to 0 component, this transition is an excellent candidate for ultra-accurate laser stabilization and a frequency standard in the optical region<sup>2</sup>.

### 4.3 External-Cavity Laser Diode

Diffraction gratings in external cavity lasers combine the functions of the filter and external mirror. In extended cavities, the light from the grating must be retroreflected back into the gain medium. Two common retroreflecting mounting geometries for diffraction gratings in extended cavity lasers are the Littrow configuration and the grazing-incidence configuration (Littman type). In the configuration of Fig. 4.2 (Littman type), the intracavity beam makes two passes at the grating. The diffracted light from the second pass is a retroreflection of the incident from the first pass. Therefore, the angular dispersion of the retroreflected light is twice that of the light diffracted on one pass. The dispersion of the Littman type configuration is twice that of the Littrow configuration for the same angle of incidence. A properly designed ECLD will operate on a single external-cavity longitudinal mode. The use of an external grating allows tenability across the wide gain bandwidth of the semiconductor gain medium. The density of accessible mode is increased by the ratio of the external to solitary cavity length. Truly phase-continuous tuning without mode hops is also possible. The most striking feature of the stable external-cavity laser is its narrow linewidth. The linewidth of ECLD is greatly reduced in comparison to solitary diode lasers because of the longer photon lifetime of external cavity. The linewidth narrowing resulting from the increased optical length of external cavity is achieved at the cost of reduced tolerance of mechanical perturbations.

### 4.4 Injection Locking

Injection locking is a useful technique for amplifying the output power of a tunable external cavity diode laser<sup>3</sup>, is a promising method to synchronize one or more free running lasers to a master laser<sup>4,5</sup>, and is a powerful technique to transfer the frequency and the spectral purity of the master laser to a slave laser<sup>6</sup>. It can be used to ensure single mode operation, to reduce the spectral width<sup>7</sup>, to generate optical frequency and phase modulation<sup>8</sup>. Also, it can be exploited to study the static and dynamic properties of the lasers<sup>9,10</sup>. When the frequency of the master laser is close to one of the cavity modes of the slave laser, the slave laser is forced to oscillate at the same frequency as the master laser and is phase-locked in Fourier frequency range below the locking bandwidth<sup>6</sup>. There are high-power devices where

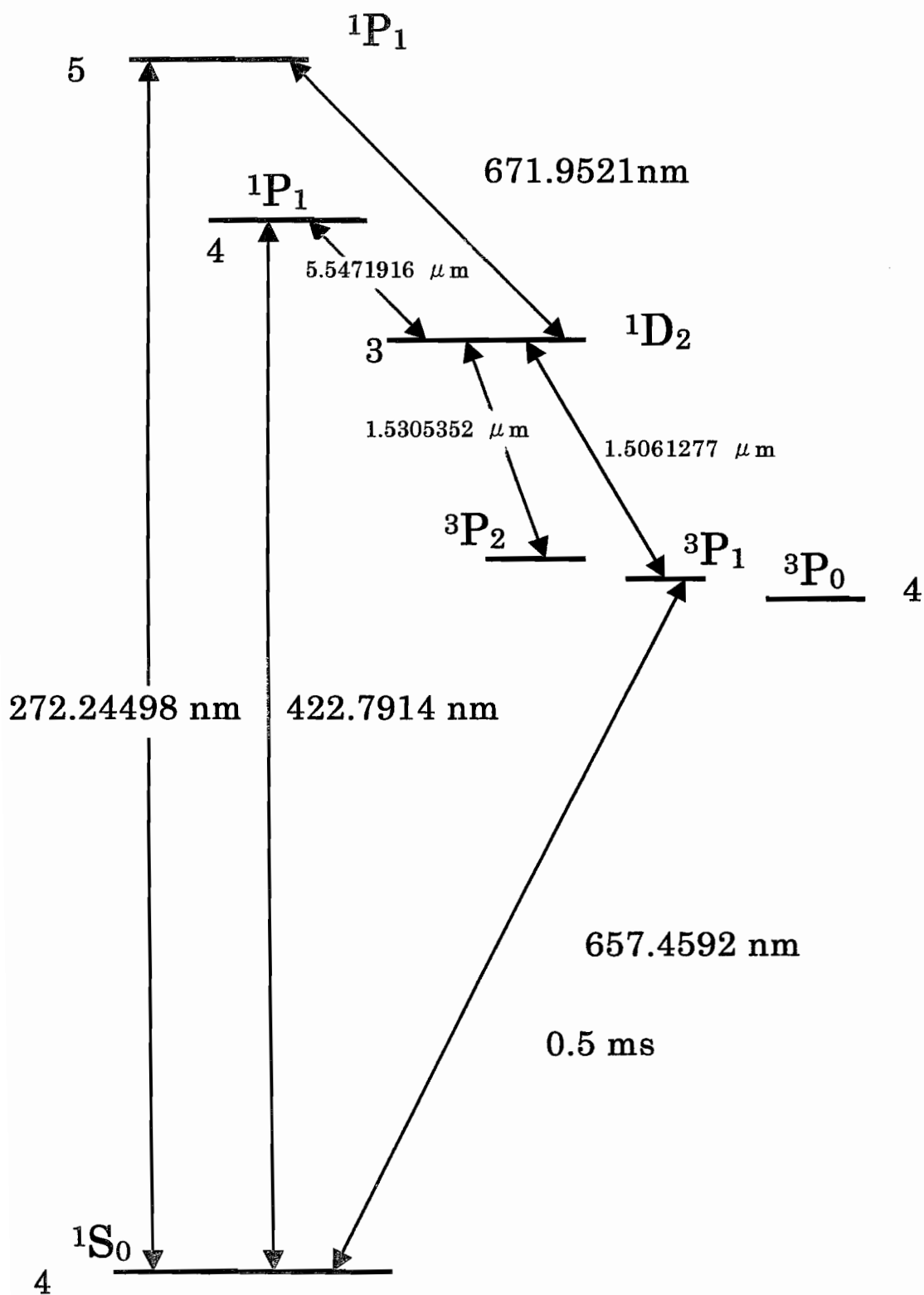


Fig. 4.1. Energy level diagram of Ca atom .

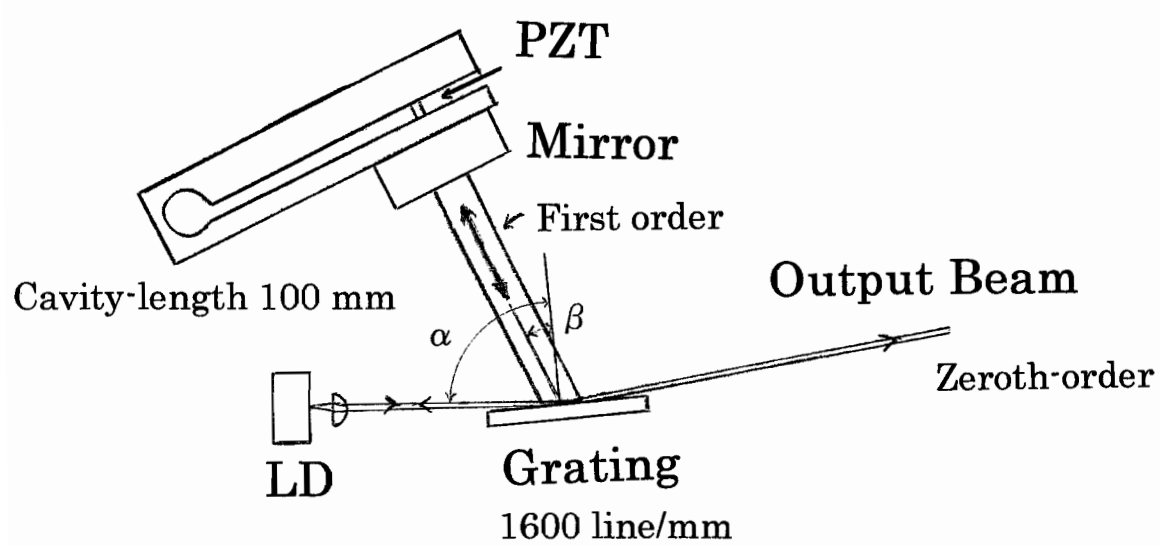


Fig. 4.2 External-Cavity Laser Diode.

the width of the active layer has been increased at the cost of a well defined transverse mode pattern. In the infrared spectral range, injection locking techniques<sup>11)</sup> have been used to combine the spectral and spatial properties of low-power single mode lasers with high power diode lasers or even diode arrays<sup>12)</sup>. In order to distinguish the input light and the output light of the slave laser, we used an isolator. Fig. 4.3 shows an isolator. The beam of the master laser is entering from the left. The light now passes through the Faraday rotator. This is made of a terbium gallium garnet (TGG) crystal which is situated in a strong homogeneous magnetic field. The crystal and the strength of the magnetic field are adjusted in such a way that the light polarization has been rotated through  $45^\circ$  on exiting the crystal (a counter clockwise rotation when viewed in the south-north direction of the magnetic field ( $+45^\circ$ )). The exit polarizer is likewise oriented at  $+45^\circ$ , so that the maximum beam intensity is transmitted. Next, consider the slave laser beam. When light of any polarization meets the polarizer it leaves it at  $+45^\circ$ . It now passes through the Faraday rotator and is again rotated through  $+45^\circ$ . The non-reciprocal nature of the Faraday effect results in the direction of rotation once again being counter clockwise as viewed from the south-north direction of the magnetic field. Thus on leaving the Faraday rotator the polarization has rotated through  $+90^\circ$  compared with the preferred polarization of the entry polarizer due to the addition of two  $+45^\circ$  rotation. In this polarization condition, it is diverted laterally by entry polarizer.

## 4.5 Stabilization Method of Ring Resonator

To lock the frequency of the master laser to the resonance of the cavity, the polarization technique by Hänsh and Couliaud<sup>13)</sup> was used. The incoming light can be decomposed into two orthogonal linearly polarized components with s polarized and p polarized. The power reflection for the S and the P components are different. Away from resonance, however, reflected wave components acquire a phase shift and the reflected beam acquires an elliptical polarization. A schematic of the setup is shown in Fig. 4.4. To detect the ellipticity, the reflected light is sent into an analyzer assembly consisting of a  $\lambda/4$  retarder and a linear polarization beam splitter. The  $\lambda/4$  retarder transforms these circular components into orthogonal linearly polarized waves which are separated by the beam splitter so that their intensities can be measured individually. Since the incoming light is linearly polarized, the two circular components have equal intensities. This fact can be very useful for balancing the sensitivity of the two photo-detectors.

## 4.6 Experiment of Master/slave/slave Injection Locking

In order to develop the laser cooling light of Ca atoms, we have developed a cascade master/slave/slave injection spectrometer. A Schematic diagram of our experimental set-up is shown in Fig. 4.5. The master laser is an external cavity diode laser (ECDL) that comprises an antireflection-coated laser diode (SDL-AR-5412-H1), collimating lens, a diffraction grating at

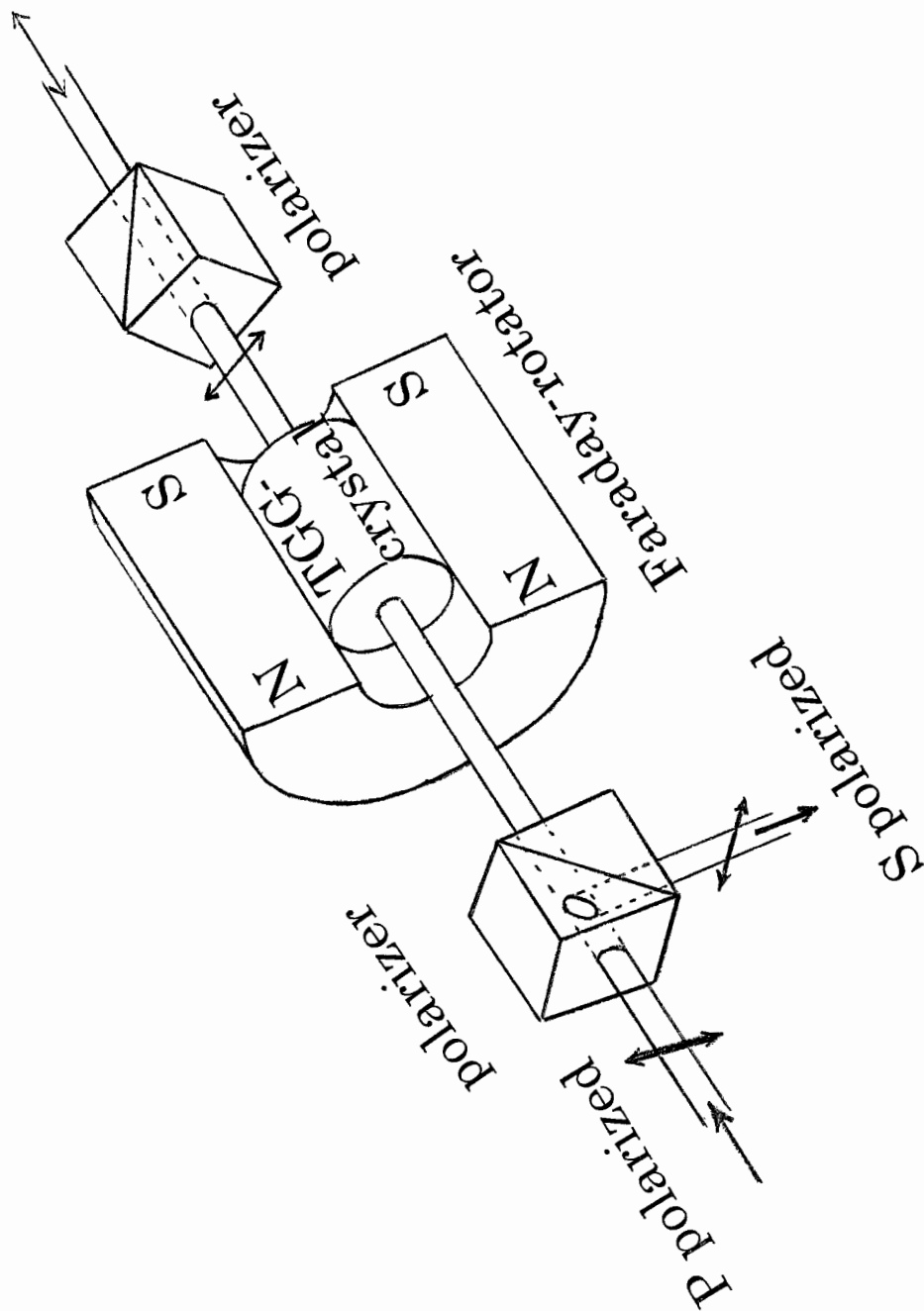


Fig. 4.3. Function of the Faraday-Isolator.

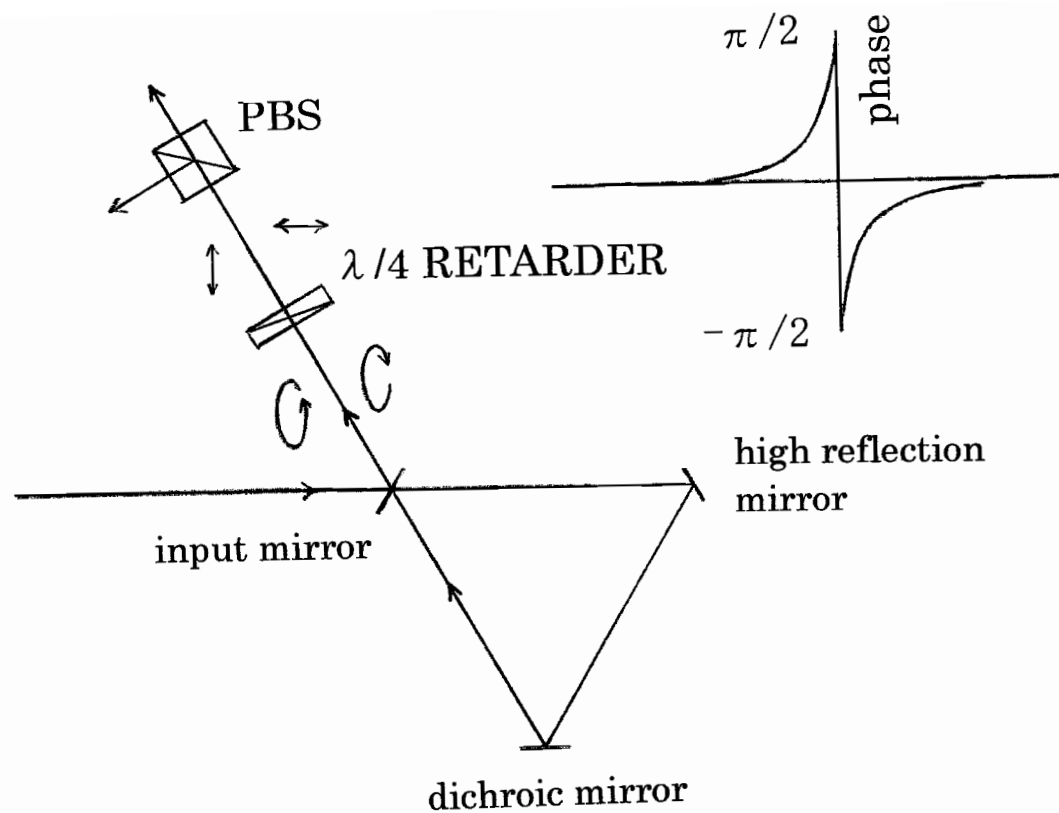


Fig. 4.4. Phase change of the light reflected by an optical cavity.

grazing incidence, and an external mirror in Littman-Metcalf configuration<sup>11</sup>. The power injected from the master to the slave laser is 5mW. The slave laser is a single-stripe high-power laser diode (SDL-5432-H1; <200 mW) without antireflection coating, followed by a collimating lens. The injection-locked slave laser provides a power of 150 mW. Cylindrical lenses with focal lengths of 80 mm and 15 mm are set before and after the isolator, respectively. They are set in order to modify the excessively oblong cross section of the collimated beam output from the slave laser as well as to reduce the beam cross section so that it will be transmitted entirely through the isolator. In front of the cavity, a mode-match lens of 300 mm focal length is used after reshaping the cross section of the slave laser beam to quasi-circular by means of an anamorphic prism pair. The beam from the slave laser is introduced to a KNbO<sub>3</sub> crystal for SHG setup within a triangular enhancement cavity. The total length of the enhancement cavity is 755 mm, and its free spectral range (FSR) is about 400MHz. A pair of lenses (f = 40 mm) provides a small beam waist. The crystal is enclosed in a small vacuum box to prevent frost formation on its surface when its temperature is lowered to -8 °C to accomplish noncritical type-I phase matching for the 4<sup>1</sup>S<sub>0</sub>-4<sup>1</sup>P<sub>1</sub> transition line. The surfaces of box windows, lenses, and crystal are all antireflection (AR) coated against both fundamental and second-harmonic radiations. If the mode matching is thorough, the power-enhancement factor is determined by  $(1-R_c)/[1-R_c^{0.5}(1-\alpha)^{0.5}]^2$ , where  $R_c$  and  $\alpha$  are the reflectivity of the input coupler mirror and an additional intracavity optical loss, respectively<sup>14</sup>. The two other mirrors have the reflectivity of 0.991 and 0.976. The transmission of the crystal is 0.991. From these values we assumed that the intrinsic loss of the cavity/crystal is 0.042. In order to increase the enhancement factor, it is necessary to consider the balance between the intrinsic loss of the cavity/crystal and the reflectivity of the input coupler. Figure 4.6 shows the enhancement factor plotted against the reflectivity of an input coupler. The dotted line shows the most efficient combination of the intrinsic loss of the cavity/crystal and the reflectivity of the input coupler. For an intrinsic loss of the cavity/crystal of 0.04, the value of the enhancement factor drops sharply when the reflectivity of the input coupler exceed approximately 0.97. On the other hand, if we choose an input coupler with reflectivity of 0.95 (a slightly smaller value than the optimum reflectivity of 0.96) the calculated enhancement is 24.7, which is a little smaller value than the maximum enhancement of 25. On the basis of these results we select the value of 0.95 for the input mirror. As  $R_c$  is about 0.95, this enhancement is expected to be 25, if we assume  $\alpha$  of 0.04. Finesse of the cavity (F; ratio of one FSR to the transmission bandwidth) is inferred to be 35 based on the observed transmission profile of the cavity. From this, the round-trip reflectivity R is estimated to be 0.91 simply based upon the relation  $F=(\pi R^{0.5})/(1-R)$ , and consequently,  $\alpha$  of approximately 0.04 is deduced<sup>13</sup>. This value is coincided with the product of the reflection ratio of the three mirrors and the transmission of the crystal. Attention was paid to the selection of an appropriate beam waist size and crystal length. In most cases the SHG efficiency is considered to be proportional to  $(l/w_0)^2$ , where  $l$  and  $w_0$  are the interaction length in the crystal and beam waist size, respectively. Up to 21 mW SHG power was obtained from the fundamental input power of 108 mW. The frequency of the master laser is stabilized to the resonance position of the enhancement cavity by means of the Hänsch-Couillaud method<sup>13</sup>. Frequency locking of the ECDL (master laser) to the cavity is performed not only through

PZT, piezoelectric transducer;  $\text{KNbO}_3$ , potassium niobate; PBS, polarizing beam splitter; PD, photodiode;  $\lambda/2$ , half-wave plate;  $\lambda/4$ , quarter-wave plate; P.P., anamorphic prism pair; H.V. amp, high-voltage amplifier.

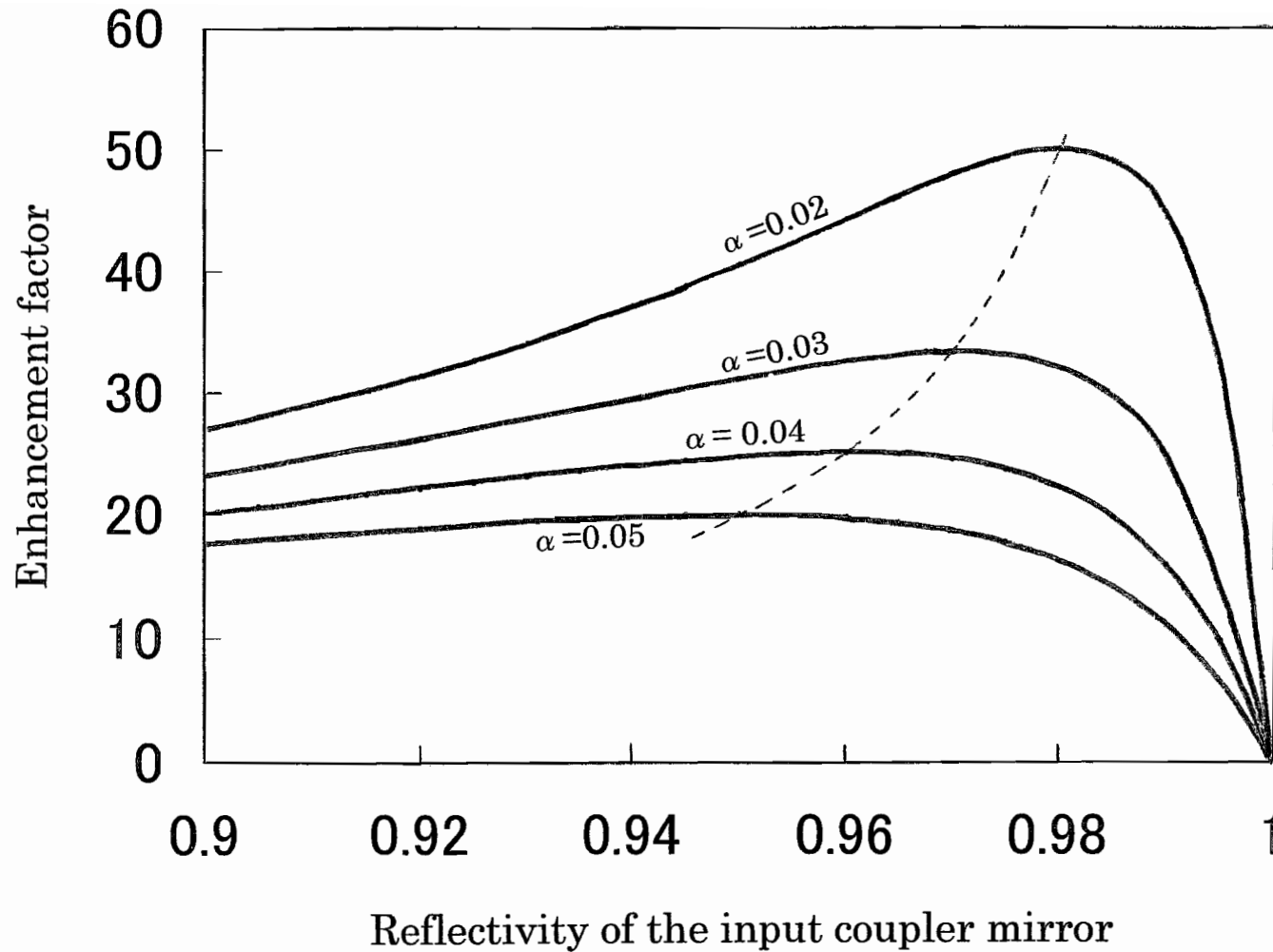


Fig. 4.6. Calculated enhancement factors plotted against the reflectivity of an input coupler for each value of internal loss ( $\alpha$ ). The dotted line shows the most efficient combination of the intrinsic loss of the cavity/crystal and the reflectivity of the input coupler.

PZT (dc component) but also through an injection current (faster component, but <16kHz bandwidth), in order to suppress the frequency fluctuation of the ECDL that is ascribed to the atmospheric and mechanical drifts induced by the surrounding acoustic noise. In order to select an efficient locking condition in the present stabilization scheme, the polarization vector of the incident light is set at a finite angle from the direction of the crystal axis, and accordingly the modes of the orthogonal polarization can be excited by the mixed polarization. The SHG can be divided by polarizing beam splitter. The power can be adjusted by the orientation of the axes of a half-wave plate. NHLV3000E (from Nichia) was used as a slave/slave laser.

In order to study the blue laser's potential to the application of the laser cooling, the part of the SHG was injected into the NHLV3000E. The output of the SHG injected blue laser was observed by using a monochromator, confocal Fabry-Perot interferometer, and wavemeter. In order to choose an efficient locking condition in the present stabilization scheme, first, we observed the very weak spectra of 422.791 nm with the original spectra of the slave/slave blue laser by using the monochromator. Next, we adjusted the incident SHG alignment by PZT so that the very weak field of the 422.791 nm spectrum became intensified. When there is an injection SHG beam, the slave/slave blue laser oscillated in a quasi-single mode as shown in Fig. 4.7<sup>15)</sup>. Since the resolution of the monochromator was 0.1 nm, fine spectra of the slave/slave blue laser were observed by using the confocal Fabry-Perot interferometer. The resolution of the Fabry-Perot interferometer was 20MHz. Fig. 4.8 corresponds to Fig. 4.7 (A) and Fig. 4.9 corresponds to Fig. 4.7 (B). In both Fig. 4.8 and Fig. 4.9, the left-hand side photo shows the spectrum for the case that the blue laser was not injection locked and the right-hand side photo shows the spectrum for the case that the blue laser was injection locked. When the blue laser was injection locked, the contrast of the transmission spectra of the confocal Fabry-Perot interferometer became clear and the value of the wave-meter changed from 421.722 nm to 422.791 nm at 72.0 mA. As shown in Fig. 4.7. (B), the output power increased approximately 70% when an injection SHG beam was used. The transmittance of the isolator was 0.62. In this system comprising of blue laser,  $\lambda/2$  wave-plate and isolator, 11.0 mW output power was obtained from the 2.2 mW input slave SHG power. Since NHLV3000E has a multi quantum-well (MQW)/GaN/AlGaIn separate-confinement-heterostructure, it oscillates in the multilongitudinal mode<sup>16)</sup>, as shown in the lower spectrum of Fig. 4.7(A). The resolution of the monochromator was 0.1 nm, which could distinguish each longitudinal mode of NHLV3000E. However, if we adequately adjust the temperature and current, the laser oscillates in a quasi-single mode, as shown in the lower spectrum of Fig. 4.7(B), in which 4 satellite longitudinal modes were observed on the short-wavelength side of the main spectrum. This phenomenon was caused by interference effects between the light reflected from the glass of the package of NHLV3000E and that from the field inside the laser diode, since the wavelength of the laser changes depending on the injection current and temperature<sup>17)</sup>. As shown in Fig. 4.7(B), when the laser was injection locked, the spectra shifted to a longer wavelength by about 1 nm. Both the main longitudinal and satellite longitudinal modes shifted to a longer wavelength by about 1 nm. The possible injection locking range was  $72.0 \pm 0.5$  mA at 23.7 °C. When the injection current exceeded this range, NHLV3000E was not injection locked to SHG. Also as shown in Fig. 4.7(A), NHLV3000E was injection locked to SHG when the injection

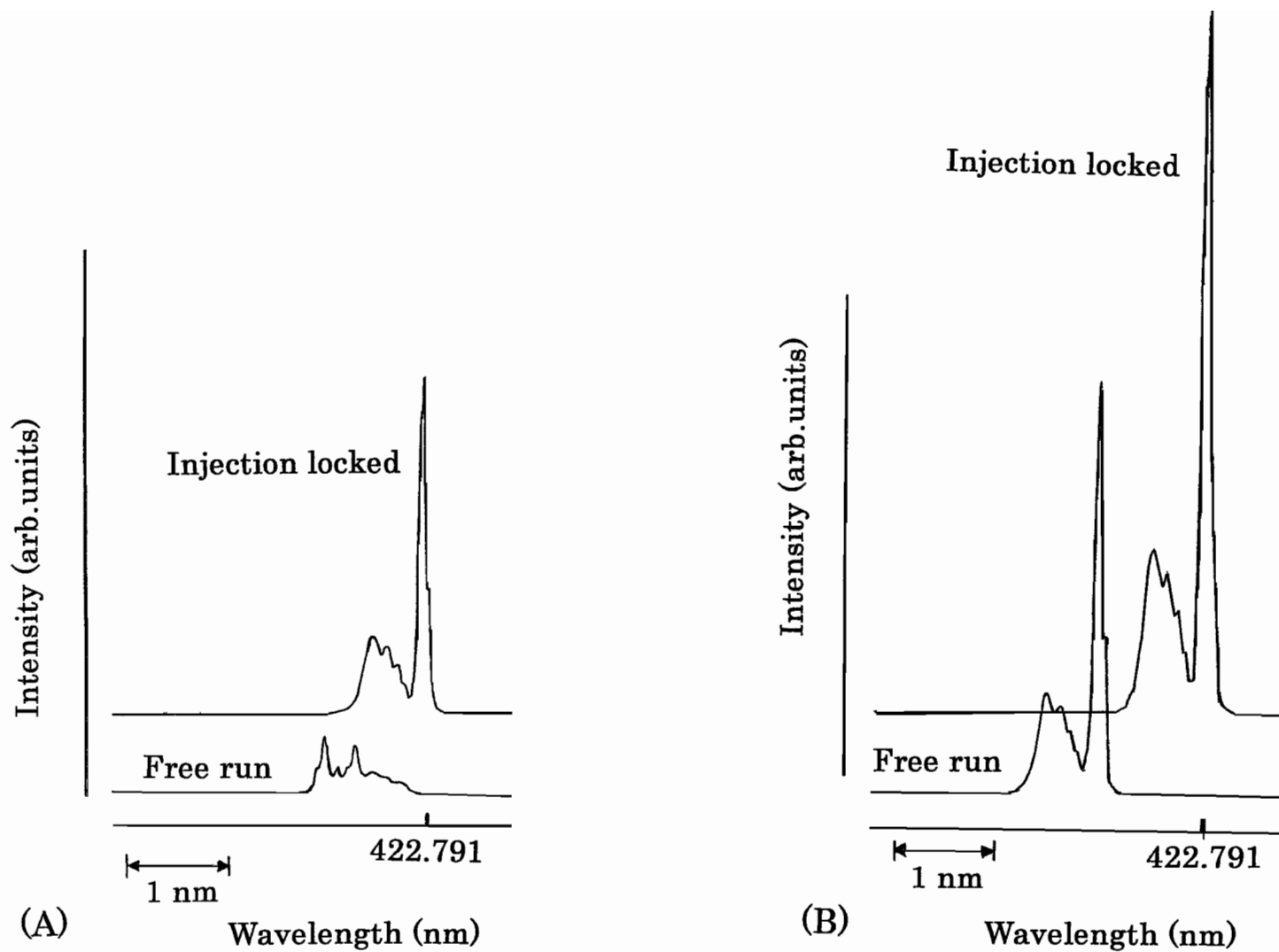


Fig. 4.7. Emission spectra of the NHLV3000E with and without the injection SHG beam.

(A)  $I = 60.9 \text{ mA}$ ,  $P_{\text{in}} = 1.2 \text{ mW}$ ,  $P_{\text{out}} = 7.7 \text{ mW}$       (B)  $I = 72.0 \text{ mA}$ ,  $P_{\text{in}} = 1.2 \text{ mW}$ ,  $P_{\text{out}} = 17.7 \text{ mW}$

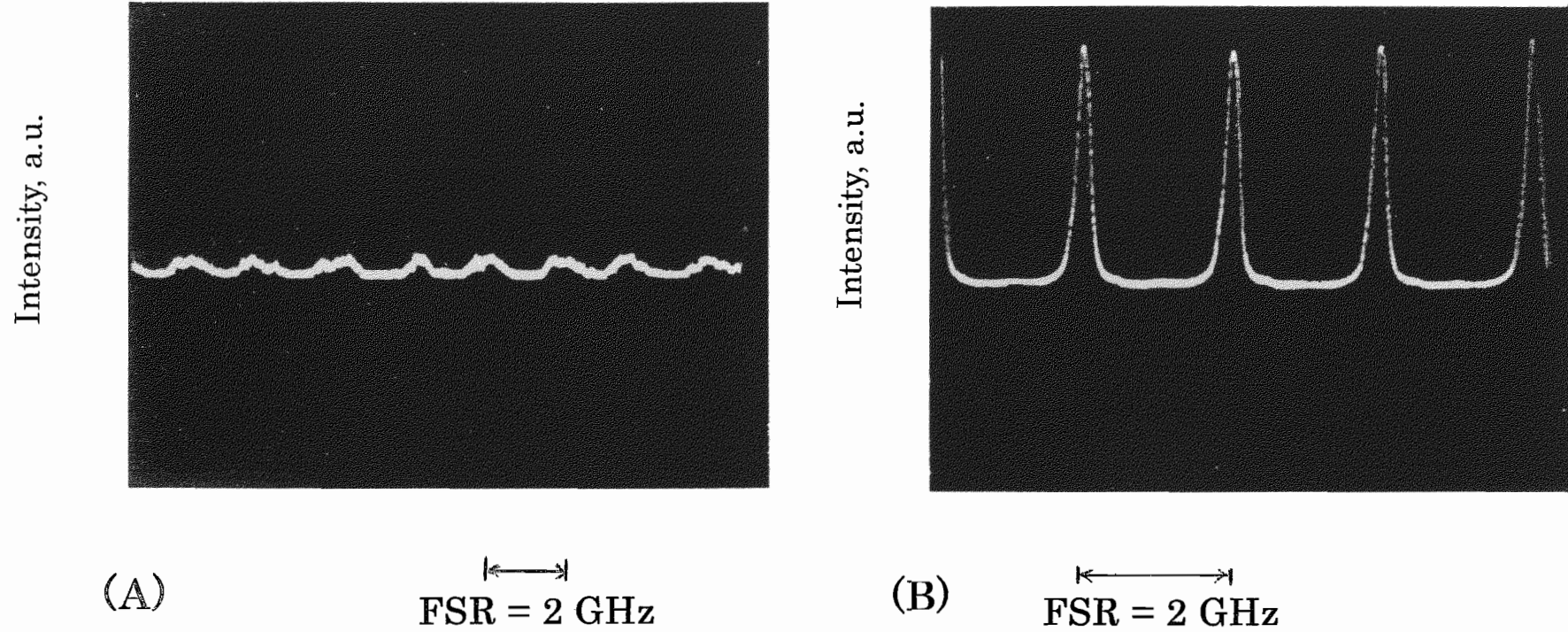


Fig. 4.8. Spectra of the slave/slave blue laser. Injection current of the blue laser was 60.9 mA, (A) free run and (B) injection locked (input power was 1.2 mW and output power was 7.7 mW).

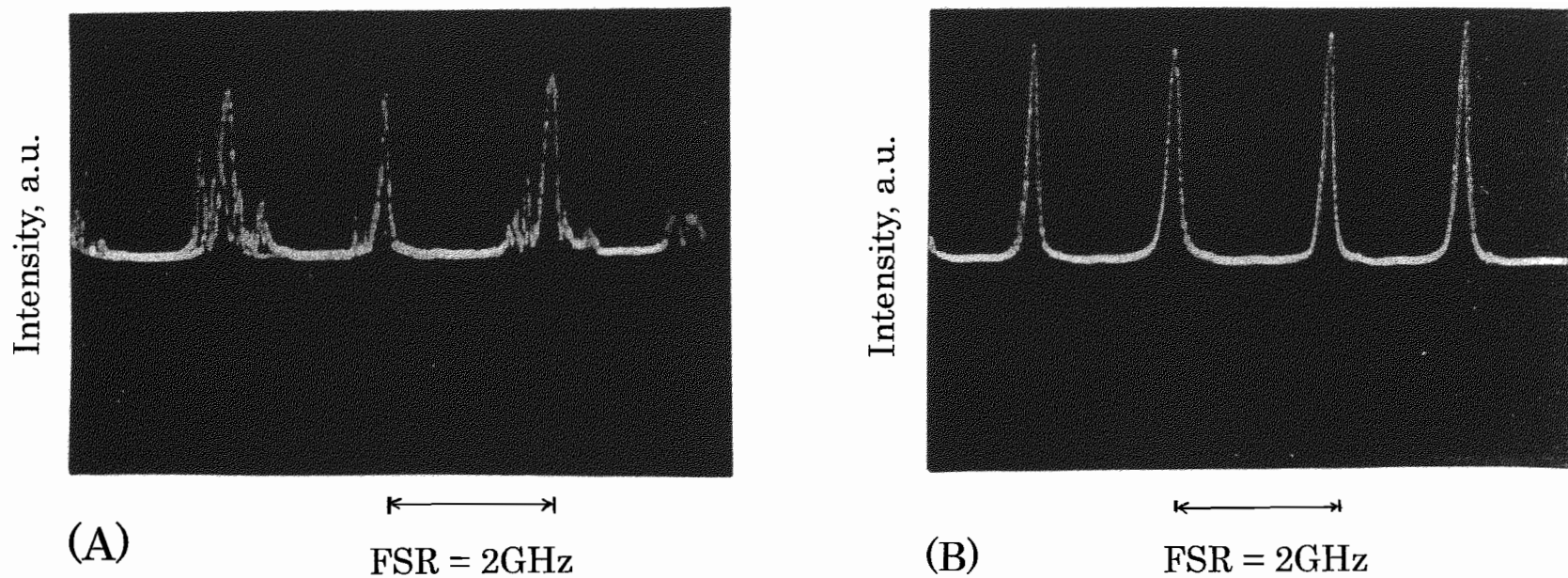


Fig. 4.9. Spectra of the slave/slave blue laser. Injection current of the laser was 72.0 mA, (A) free run and (B) injection locked (input power was 1.2 mW and output power was 17.7 mW).

current was  $60.9 \pm 0.5$  mA at  $23.7$  °C. When the injection current exceeded this range, NHLV3000E was not injection locked to SHG. In order to examine the power of the main longitudinal mode which can be utilized as the cooling light, we studied the intensity ratio of each satellite mode against the main longitudinal mode. Table 4.1 shows the intensity ratio of each satellite mode against the main longitudinal mode. The values -0.55, -0.44, -0.33, and -0.22 indicate how much shorter the wavelength was from the main longitudinal wavelength. The intensity ratio of satellite mode against the main longitudinal mode was almost constant regardless of current<sup>18)</sup>. Excluding the power of the satellite longitudinal mode of the slave/slave laser, the total power of 422.791 nm light which can be used for the magneto-optic trap (MOT) for Ca was 25.5 mW. In order to measure the precise spectral width of the injection locked blue laser and the SHG and also to confirm the injection locking to the slave/slave blue laser, we also send the injection locked blue laser through acousto-optic modulator and observed the beat signal between the SHG and the 40 MHz frequency sifted injection locked blue laser. The observed beat signal is shown in Figure 4.10. The linewidth of the beat signal was 0.75 MHz. From the linewidth of the beat signal the line width of the SHG and blue laser were both about 0.4MHz. Accordingly the linewidth of the master laser was 0.2 MHz. Since the natural linewidth of Ca atoms at the cooling transition is 34 MHz, both the power and linewidth were sufficient to cool Ca atoms.

## 4.7 Conclusion

In this chapter, the development of a light source for cooling Ca atoms at 423 nm is described, with the aim of realizing an optical frequency standard using the Ca transition at 657 nm. By injecting an SHG to a blue diode laser, a master/slave/slave laser spectrometer in the blue-wavelength region has been developed. As a result, the total power was 25.5 mW and the spectral linewidth was about 0.4 MHz. Considering the natural linewidth of Ca atoms at the cooling transition is about 34 MHz, both the power and linewidth were sufficient to cool Ca atoms by the magneto-optic trap. The effect of the second-order Doppler shift can be eliminated ( $8 \times 10^{-19}$ ) by this system. We also found first that it is possible to lock the oscillation frequency of blue laser diodes more than 1nm by injection locking technique.

| The intensity ratio of satellite mode against the main mode. |                            |               |               |               |               |
|--------------------------------------------------------------|----------------------------|---------------|---------------|---------------|---------------|
| Injection<br>current<br>(mA)                                 | SHG<br>injection<br>locked | -0.55<br>[nm] | -0.44<br>[nm] | -0.33<br>[nm] | -0.22<br>[nm] |
| 60.9                                                         | ○                          | 0.22          | 0.19          | 0.14          | 0.08          |
| 72                                                           | ×                          | 0.24          | 0.21          | 0.14          | 0.09          |
| 72                                                           | ○                          | 0.23          | 0.19          | 0.13          | 0.08          |

Table 4.1. The intensity ratio of each satellite mode against the main longitudinal mode. The values -0.55, -0.44, -0.33, and -0.22 indicate how much shorter the wavelength was from the main longitudinal wavelength.

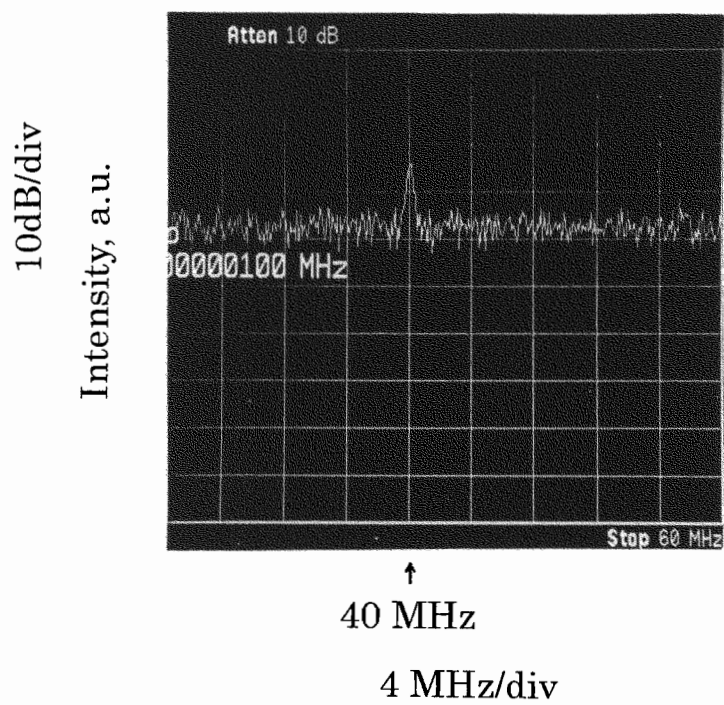


Fig. 4.10. Observed beat notes between the SHG and the 40 MHz sifted injection locked blue laser.

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## Chapter 5

### Conclusion

#### 5.1 Summary

In this thesis, a series of studies on a laser diode light sources for exciting  $\text{Sr}^+$  ions and Ca atoms are summarized.

In chapter 1, the background, objectives and technological significance of this study along with the required performance of the excitation light source for use in frequency standards are described.

In chapter 2, the development of a laser diode light source for exciting  $\text{Sr}^+$  ions is described. The frequency of a SHG of a high power 844nm laser diode has been stabilized to the absorption line of  $^{88}\text{Sr}^+$  ion at 421.671 nm in a hollow cathode lamp. The frequency fluctuation never exceeded 10MHz, which is much narrower than the Doppler line width of the  $\text{Sr}^+$  ions (more than 1 GHz) confined in an rf ion trap. To our knowledge, the development of this system is the first demonstration of the frequency stabilization of the 844 nm laser light to the  $5s^2\text{S}_{1/2}-5p^2\text{P}_{1/2}$  transition line of  $^{88}\text{Sr}^+$  ions at 421.671 nm. As a result, more than 1 $\mu$ W of SHG can be used to fluorescence of  $\text{Sr}^+$  ions, which is enough to deplete one of the hyperfine levels of the ground state of  $^{87}\text{Sr}^+$  ions which for laser microwave double resonance.

In chapter 3, the development of an ion trap system for confining ions in a vacuum for a long time is described. The ion trap was designed so that many ions are confined within the Lamb-Dicke region. As a result, the number of confined ions was in the range of  $2.5 \times 10^6 \sim 2 \times 10^7$ , the fluorescence intensity was 2000 c/s or higher, the sufficient S/N for detecting double resonance fluorescence signals was realized and the elimination of the first-order Doppler shift was realized in the 98% of the trapped ions. Further, we found He gas was effective in reducing the kinetic energy of the trapped ions. The ions were cooled to 400 K at a He pressure  $2.7 \times 10^{-4}$  K which reduced the second order of the Doppler shift below  $5 \times 10^{-13}$  in the microwave region.

In chapter 4, the development of a light source for cooling Ca atoms at 423 nm is described, with the aim of realizing an optical frequency standard using the Ca transition at 657 nm. By injecting an SHG to a blue diode laser, a master/slave/slave laser spectrometer in the blue-wavelength region has been developed. As a result, the total power was 25.5 mW and the spectral linewidth was about 0.4 MHz. Considering the natural linewidth of Ca atoms at the cooling transition is 34 MHz, both the power and linewidth were sufficient to cool Ca atoms by the magneto-optic trap. The effect of the second-order Doppler shift can be eliminated ( $8 \times 10^{-19}$ ) by this system. We also found first that it is possible to lock the oscillation frequency of blue laser diodes more than 1nm by injection locking technique.

## 5.2 Subjects to be solved

The goal of the ion storage is to produce a trapped ion frequency standard with both high accuracy and stability. Here after, by utilizing  $^{87}\text{Sr}^+$  ions in rf trap, the 5 GHz ground-state hyperfine transition will be studied as a possible microwave frequency standard. Further, introducing laser cooling to reduce the thermal or incoherent ion motion, the second-order Doppler shift will be reduced to small values. However, since trapped ions exhibit coherent motion such as the "micromotion" in rf traps, which is the largest origin of the second-order Doppler shift, it must be minimized by other means such as controlling the shape and spatial extent of the ion sample.

When construct a Ca optical frequency standard in future, we will have to develop some systems. One is a relatively compact magneto-optic trap (MOT) system. A second is a 657 nm laser system for optical frequency standard. As demonstrated in just the past year, frequency comb based on mode-locked femtosecond lasers provide a convenient, robust, and accurate means of phase-coherent linking optical frequencies to standards in microwave domain. Fig.5.1 shows a direct connection between microwave and optical frequencies. Femtosecond laser is a time domain description. Frequency comb generator is synonymous description of the same device in the frequency domain. The spectrum of the pulse train consists of many equidistant spectral lines. The spacing of the lines is equal to the repetition rate of the laser. A direct connection between the Ca optical frequency standard and Cs atomic clock will make the Ca optical frequency as a primary frequency standard.

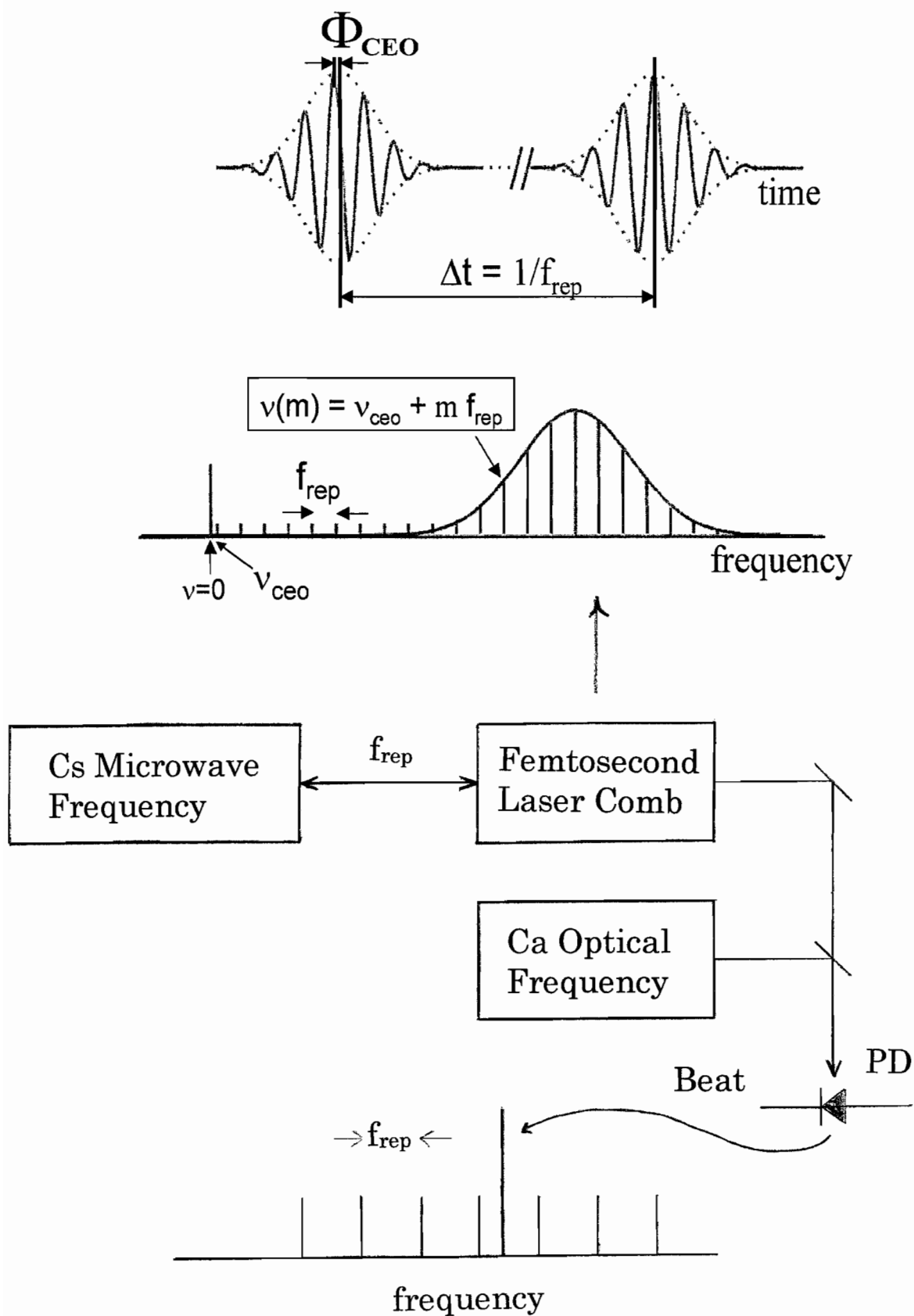


Fig. 5.1. A direct connection between microwave and optical frequencies.

## Publications

### Papers related to this work

- 1 HIRANO I, YODA J, FENG-H, OKUMURA K, ONAE A :  
Determination of Collisional Quenching Rate for the  $4D_{3/2}$  State in Sr II. Japanese Journal of Applied Physics, 37, 5767-5771, 1998
- 2 HIRANO I, YODA J, FENG-H, OKUMURA K, ONAE A : Collisional Quenching Rate by He, N<sub>2</sub> and CH<sub>4</sub> for the  $4D_{3/2}$  State in Sr II. Japanese Journal of Applied Physics, 38, 3747-3748, 1999
- 3 HIRANO I, YODA J : Frequency Stabilization of 421.671 nm Second-Harmonic Generation for Studies of  $^{88}\text{Sr}^+$  Ions Confined in an RF Trap. Optical Review, 8, 409-411, 2001
- 4 平野 育、依田 潤 : RF トラップで捕獲された  $\text{Sr}^+$  イオンの特性。電子情報通信学会論文誌、87, 303-311, 2004
- 4' HIRANO I, YODA J : Characteristics of  $\text{Sr}^+$  Ions in an RF Trap. Electronics and Communications in Japan, Part2, 87, 1-9, 2004
- 5 HIRANO I, ITO N, INABA H : Spectrometer for Ca optical frequency standard using cascade master/slave/slave injection-locking. Electronics Letters, 38, 714-716, 2002
- 6 HIRANO I, ITO N : Spectral characteristics of cascade master/slave/slave injection-locking of laser diodes. Optics & Laser Technology, 37, 81-86, 2005

### Paper not related to this work

- 1 平野育 : ビートを利用する超短光パルス列の発生。光学、21, 338-341, 1992

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