

Radiative lifetime of the 1S_0 metastable state in doubly charged Kr ions

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The radiative lifetime of the $4s^24p^4\ ^1S_0$ metastable state in Kr^{2+} ions has been experimentally determined using an electrostatic ion trap of the ion-beam storage type. The radiative lifetime was obtained by measuring the number of 350-nm photons emitted during the $^1S_0 \rightarrow ^3P_1$ magnetic dipole transition of Kr^{2+} as a function of the storage time. The storage lifetime of the Kr^{2+} beam in the trap, which is a correction term in the determination of the radiative lifetime, was measured by detecting Kr neutrals produced by electron capture collisions with residual gases in the trap. We have obtained a lifetime value of 17.4 ± 0.4 ms, which is larger than the previous experimental results, but consistent with theoretical predictions.

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I. INTRODUCTION

Radiative lifetimes for forbidden transitions of ions are important not only in furthering our understanding of the atomic structure, but also for applications such as the diagnostics of density and temperature in astrophysical and laboratory plasmas [1]. The radiative lifetimes of the 1S_0 metastable state in the ground-state configuration (ns^2np^4) of doubly charged rare-gas ions (Ne^{2+} , Ar^{2+} , Kr^{2+} , and Xe^{2+}) have been investigated experimentally and theoretically by several groups [2]. This state decays primarily to the 3P_1 state by the magnetic dipole ($M1$) transition or to the 1D_2 state by the electric quadrupole ($E2$) transition. $M1$ and $E2$ transition probabilities are typically several orders of magnitude lower than the allowed electric dipole transitions, and hence the 1S_0 state of these ions has a long lifetime on the order of milliseconds to hundreds of milliseconds.

In this work, we present radiative lifetime measurements of the 1S_0 metastable state of Kr^{2+} ions. Figure 1 shows the energy-level diagram of the $4s^24p^4$ ground-state configuration of Kr^{2+} . As shown in the figure, the dominant radiative decay channel of the 1S_0 metastable state of Kr^{2+} is the decay to the 3P_1 state via the $M1$ transition. Compared to this channel, the radiative decay to the 1D_2 and 3P_2 states via the $E2$ transition is minor. The radiative wavelengths for $^1S_0 \rightarrow ^3P_1$, $^1S_0 \rightarrow ^1D_2$, and $^1S_0 \rightarrow ^3P_2$ transitions are approximately 350, 542, and 302 nm, respectively.

Several theoretical calculations have derived values for the radiative lifetime of the 1S_0 metastable state of Kr^{2+} . The transition probabilities calculated by Garstang [3] using a semiempirical approach resulted in a lifetime of 17.2 ms. Subsequent calculations by Biémont and Hansen [4] using the Hartree-Fock-Roothaan method indicate a lifetime value of 17.3 ms. These values agree well with each other.

Experimentally, the radiative lifetime can be obtained by measuring the photons emitted during the transition. Because of the long lifetime of more than 10 ms of the 1S_0 metastable state of Kr^{2+} , several groups have applied this method to the ions in ion traps. Lifetimes of 13.1 ± 0.6 and 14.8 ± 0.8 ms were determined by Walch and Knight [5] using a cylindrical

radio-frequency ion trap and by Calamai and Johnson [6] using a Kingdon-type ion trap, respectively. These experimental values are mutually inconsistent even considering their errors, and are a few tens of a percent smaller than the theoretical values. The ion source used in their experiments was installed internally in the ion trap. As described in Ref. [6], the uncertainties in the measurements stemmed largely from the correction for the destruction of the trapped ions by collisions with parent gases from the ion source.

Recently, Bhushan *et al.* [7] experimentally determined the radiative lifetime of the 1S_0 metastable state of Xe^{2+} ions with a small error using an electrostatic ion trap of the ion-beam storage type. Such traps can sustain ultrahigh vacuum during measurements because the ion source is placed far from the trap, thus minimizing the influence of collisional losses of the stored ions. Furthermore, the variation in the number of stored ions over time can be monitored readily, which enables correction for the collisional losses of the stored ions. We have previously measured the radiative lifetime of the 1S_0 metastable state of Kr^{2+} ions using an electrostatic ion trap. Our preliminary result [8] had larger than the expected uncertainty due to statistical error and ion losses unassociated with residual gas collisions. This paper presents the details of the measurements and the final result, the accuracy of which was improved by increasing the number of measurements and using a more detailed correction for the observed ion losses.

II. EXPERIMENT

A. Electrostatic ion trap

The experiments were performed using an electrostatic ion-beam trap at the Kyoto Prefectural University that has been described in detail elsewhere [9]. Figure 2(a) shows a schematic view of the ion trap. The electrostatic ion trap consists of two sets of cylindrical electrodes that work as reflectors (E1–E3) and einzel lenses (E4–E6), respectively. The overall trap length is 600 mm. The space between the two lenses is a field-free region of approximately 250 mm length. An ion-beam pulse is injected into the trap along its axis from the outside. The potentials are continuously applied to the electrodes on the exit side of the trap. All potentials of the electrodes on the entrance side are set to zero initially and then raised within 200 ns after injection of the pulse beam using fast, high-voltage switches. This rise time is much shorter than

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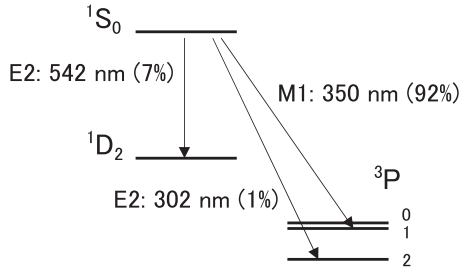


FIG. 1. Energy-level diagram of the $4s^2 4p^4$ ground-state configuration of Kr^{2+} ions. The radiation wavelengths and the branching ratios in the figure refer to the energy levels in [12] and the transition probabilities in [4].

the circulation time of the ion (typically over $10 \mu\text{s}$) in the trap. Through appropriate combinations of the reflector and lens potentials, the beam is multiply reflected between the two reflectors and is confined in a transverse direction to the axis by the lenses, resulting in the storage of the ion beam in the trap. The potentials are determined to satisfy the condition of $ls/(l + 2s) \leq f \leq s$ according to the stability criteria for a lens waveguide in ray transfer matrix analysis [9], where f represents the focal length of the lens, l is the distance between the two lenses, and s is the distance between the lens

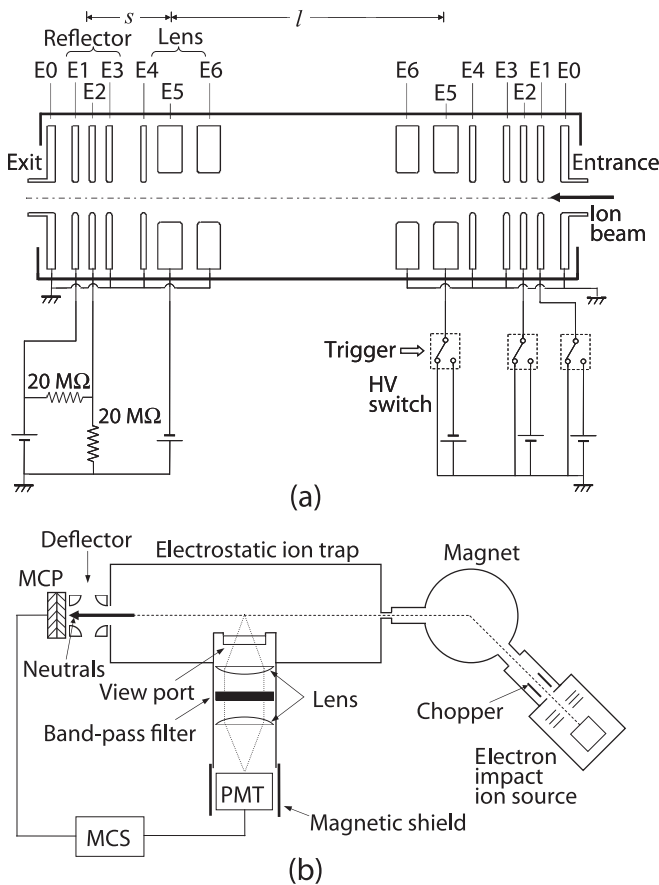


FIG. 2. (a) Schematic view of the ion trap and its power supply. (b) Apparatus configuration of this experiment with the optical system for the photon measurement.

and the reflection point of the ion. For the actual parameters of this trap ($l = 310 \text{ mm}$ and $s = 45 \text{ mm}$), the condition results in $35 \text{ mm} \leq f \leq 45 \text{ mm}$. Accordingly, for example, potentials of 1870, 935, and -4200 V are applied to the electrodes of E1, E2, and E5, respectively, in the case of a 2.4 keV-Kr^{2+} ion beam.

B. Experimental method

The experimental setup is schematically shown in Fig. 2(b). The Kr^{2+} ions were produced using a Nier-type electron impact ion source. Electron energy of 400 eV was used in the ion source, resulting in the production of the Kr^{2+} ions in the metastable states as well as in the ground state. Although the fractional population of the produced 1S_0 state was not measured in this work, it was previously reported to be 0.063 at the electron energy of 400 eV using the same type of ion source as in our work [10]. The ion beam was accelerated to 2.4 keV and was pulsed to a width of $12 \mu\text{s}$ using an electrostatic chopper. The pulse width corresponds to the circulation time of the 2.4-keV Kr^{2+} ion in the trap. The pulse beam was momentum analyzed in a 45° deflecting magnet, and was injected into the trap. The flight time of the ion between the ion source and the trap was $21 \mu\text{s}$, which is much shorter than the expected lifetime of the 1S_0 state of Kr^{2+} , so that most of the ions in the 1S_0 state entered the trap before their decay. After the ion confinement in the trap was established, the 350-nm photons emitted during the $^1S_0 \rightarrow ^3P_1$ transition were measured with an optical system installed in the field free region. The count rate of the photons as a function of time was recorded using a multichannel scalar (MCS) with a dwell time of 0.5 ms . After the predetermined storage time (0.85 s), which was long enough to measure the lifetime of the 1S_0 state of Kr^{2+} , the ions were dumped from the trap by switching off the voltages on the entrance side. The sweep duration of the MCS was set to 1 s according to the storage time. The ion injection and trapping cycle was repeated and the data accumulated over 10^5 such injections. The total measurement time was restricted by the life of the filament in the ion source ($\sim 70 \text{ h}$).

The composition of the optical system is illustrated in Fig. 1(b). The photons were focused through a Kovar-glass viewport on the detection plane of a photomultiplier tube (PMT) (R585; Hamamatsu Photonics) by two optical planoconvex lenses of fused silica (focal length 35 mm , effective diameter 22 mm). Photons of a desired wavelength were chosen by a bandpass filter [center wavelength 349 nm , full width at half maximum (FWHM) 10 nm , effective diameter 22 mm , HQBP350-UV; Asahi Spectra) placed between the two lenses. The PMT was located in a magnetically shielded case to eliminate the influence of an external magnetic field. The dark noise of the PMT was about 4 counts per second. The transmittance values of the optical components from the manufacturers' data are 0.60 for the view port, 0.93 for the lens, and 0.87 for the bandpass filter, and similarly the quantum efficiency is 0.15 for the PMT for a wavelength of 350 nm . From these efficiencies and the solid angle of the lens (0.43 sr), the detection efficiency of the optical system for a wavelength of 350 nm can be roughly estimated as 2.3×10^{-3} .

The number of stored Kr^{2+} ions decreases with the trapping time because of electron capture or elastic collision with residual gases. When the ion changes its charge to $+1$ or

0 by single- or double-electron capture, it is not reflected by the electric field of the reflector electrodes, and hence it escapes from the trap after the single- or double-capture collision. For elastic scattering, most scattered ions collide with the electrode or the trap wall immediately, and cease to exist in the trap. The influence of this collision-induced decay of the trapped ions must be corrected for in the measured photon data [Eq. (3) in Sec. III]. The number of stored ions as a function of the trapped time was monitored according to the following procedure. The number of neutral atoms (or singly charged ions) produced at a storage time between t and $t + \Delta t$ (Δt corresponds to the dwell time of the MCS) is expressed as $N(t) = \rho \int_t^{t+\Delta t} \sigma_{\text{cap}}(v)vI(t)dt$, where ρ is the density of residual gas atoms, v is the ion velocity, $I(t)$ is the number of stored ions at time t , and $\sigma_{\text{cap}}(v)$ is the cross section of double-electron capture (or single-electron capture) at v . When Δt is much longer than the ion's circulation time in the trap, the $\sigma_{\text{cap}}(v)v$ can be replaced by the averaged value over the circulation time $\bar{\sigma}_{\text{cap}}\bar{v}$. The count rate of the neutrals, defined as the number of neutral atoms per constant Δt , is then proportional to the average number of ions present in the trap between t and $t + \Delta t$:

$$R(t) \approx \rho \bar{\sigma}_{\text{cap}} \bar{v} \int_t^{t+\Delta t} I(t)dt \propto \int_t^{t+\Delta t} I(t)dt / \Delta t. \quad (1)$$

Consequently, we can obtain information about the time variation of the number of stored ions through a measurement of the count rate $R(t)$ of the neutrals or singly charged ions escaping from the trap. Neutrals and singly charged ions were measured using a microchannel plate detector (MCP) installed downstream of the trap. The count rate of these particles was recorded using a MCS with a dwell time of 0.5 ms. However, it was observed that the background secondary electrons repelled by the negative high voltage of the lens electrodes (-4200 V) escaped from the trap's exit and were collected by the MCP. These secondary electrons were rejected using an electrostatic deflector in front of the MCP. Similarly, the singly charged ions were also rejected by the deflector and only the neutrals were detected by the MCP.

The trap is evacuated using a turbo molecular pump (900 l/s) and a nonevaporable getter pump (100 l/s for H_2). To limit the conductance of the gas flow from the beam-transport line into the trap, a 40-mm-long pipe with an internal diameter of 4 mm is installed at the trap chamber's entrance. The beam-transport line from an ion source is evacuated using two turbomolecular pumps (360 l/s) and the upstream beginning of the trap is evacuated using a cryopump (1500 l/s) for differential pumping. The trap is maintained under ultrahigh vacuum using these pumps, yielding a background pressure of 5×10^{-8} Pa during measurements.

III. ANALYSIS AND RESULTS

Figure 3 shows the counts of photons as a function of the storage time corresponding to 240 000 injections of the pulse beam. The plotted data in the figure are the counts combined into time bins of 1 ms to reduce fluctuations. It can be seen that the photon counts decrease exponentially, and are thus interpreted as the desired 1S_0 metastable decay, and then reach a flat background. The count rate of this background

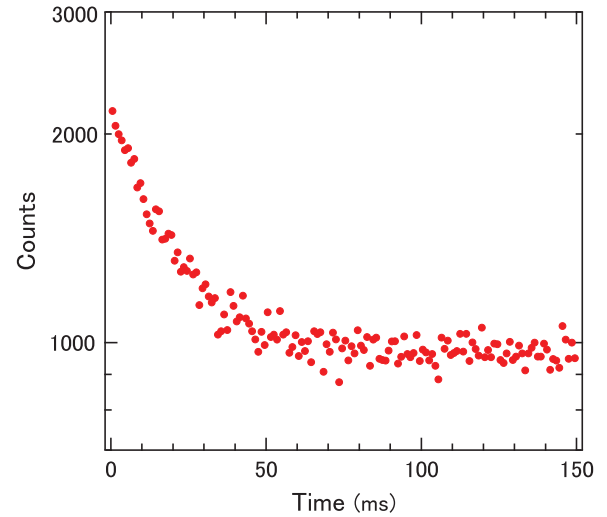


FIG. 3. (Color online) Counts of photons for the $4s^24p^4\ ^1S_0 \rightarrow 4s^24p^4\ ^3P_1$ transition of Kr^{2+} ions (350 nm) as a function of storage time. The plotted data are the counts combined into time bins of 1 ms.

(~ 4 counts per second) is very close to the dark count rate of the PMT, indicating that the background is mostly due to the dark noise of the PMT. The number of trapped 1S_0 metastable ions can be roughly calculated using the total photon counts in Fig. 3 (2×10^5 counts after background subtraction). The ratio of the number of ions within the region viewed by the optical system to the total number of trapped ions is 0.02. Considering these values and the detection efficiency of the optical system, it can be estimated that about 2000 1S_0 ions were stored in the trap per beam pulse injection. It should be mentioned that the yield of photons above the background level was not observed using other bandpass filters with neighboring shorter or longer central wavelength (312 ± 4 and 379.5 ± 4.5), thus verifying that only the photons emitted by the transition of interest ($^1S_0 \rightarrow ^3P_1$, 350 nm) were detected.

Figure 4 shows the time variation of the number of stored ions using the counts of neutrals that were measured using the MCP. The plotted data are the values after the subtraction of a constant background value. The background value was determined from the MCP counts after dumping the ions from the trap. It can be clearly seen that the stored ions decayed exponentially with a lifetime of the order of seconds. The ion loss rate in the trap due to collisions with the residual gases is given by $dI(t) = -\rho\sigma(v)vI(t)dt$, where $\sigma(v)$ is the total cross section of the ion loss by elastic scattering and electron capture at ion velocity v . Assuming that the $\sigma(v)v$ can be replaced by the averaged value over the circulation time $\bar{\sigma}\bar{v}$ as in Sec. II B, the number of stored ions decreases exponentially with storage time as

$$I(t) = I(0) \exp(-\rho\bar{\sigma}\bar{v}t) = I(0) \exp(-t/\tau_c), \quad (2)$$

where $\tau_c = 1/\rho\bar{\sigma}\bar{v}$ denotes the storage lifetime of the ion beam due to collision-induced decay. Adopting $\bar{\sigma}\bar{v} \approx 10^{-8} \text{ cm}^3/\text{s}$ [11] and with a background pressure of 5×10^{-8} Pa, Eq. (2) gives exponential decay with τ_c of the order of seconds, thus explaining the observed time variation of the counts of the neutrals in Fig. 4 well. However, as shown in the

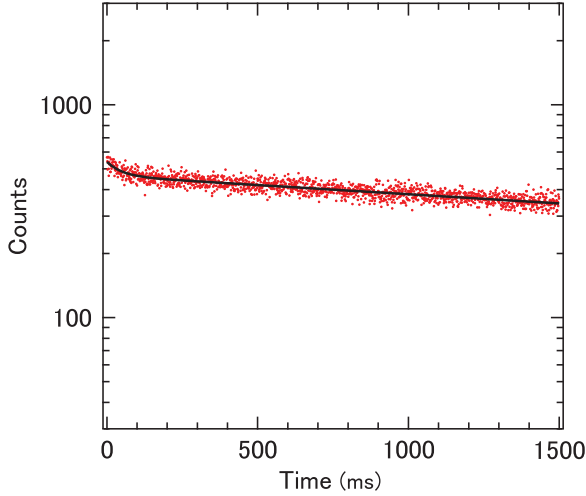


FIG. 4. (Color online) Neutral Kr counts as a function of the storage time. The background counts were subtracted. The solid curve depicts a fit to the data using Eq. (4).

figure, a smaller initial rapid decay was observed at time t less than about 100 ms. We interpret this rapid decay as attributable to ions with an injection angle that is larger than the acceptance angle of the trap because its decay time depends strongly on defocusing of the incident ion beam by the ion-source lens or on the incident-beam alignment to the trap. The ion loss by ion-ion collisions may be considered as another possible cause of the rapid decay. This process can be most prominent near the turning points in the ion trajectories where the ion density is highest. However, we did not experimentally observe any change in the rapid decay rate dependent on the number of Kr^{2+} ions injected into the trap, indicating that the contribution of ion-ion collisions to the observed rapid decay is negligibly small.

Considering the ion loss in the trap as mentioned above, the measured photon decay spectrum after the background subtraction is described by a function of the form,

$$\alpha \left[N_c(0) \exp \left\{ - \left(\frac{1}{\tau} + \frac{1}{\tau_c} \right) t \right\} + N_r(0) \right. \\ \left. \times \exp \left\{ - \left(\frac{1}{\tau} + \frac{1}{\tau_r} \right) t \right\} \right], \quad (3)$$

where τ is the desired radiative lifetime of the metastable state and α denotes a normalization factor. τ_c , $N_c(0)$ and τ_r , $N_r(0)$ represent the storage lifetime and the neutral intensity at time zero for the slow (collision-induced) decay component and those for the rapid decay component of the neutral species' spectrum in Fig. 4, respectively. These four values were determined by a fit to the neutral spectrum with two exponentials as

$$N_c(0) \exp(-t/\tau_c) + N_r(0) \exp(-t/\tau_r) \quad (4)$$

(the solid curve in Fig. 4). τ_c , $N_c(0)$, τ_r , and $N_r(0)$ of 5028 \pm 114 ms, 464 \pm 2, 47.8 \pm 7.1 ms, and 76.8 \pm 8.3, respectively, have been obtained from the fit.

We determined the 1S_0 metastable lifetime using Eq. (3). Figure 5 shows the photon decay spectrum after subtracting a constant background value from the data shown in Fig. 3. The background value was determined by averaging the counts

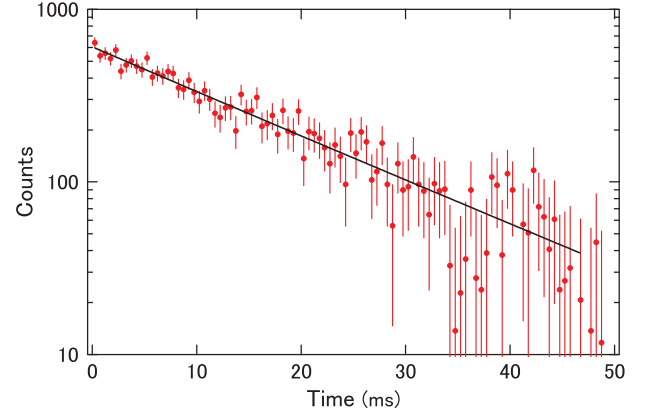


FIG. 5. (Color online) The same as in Fig. 3, but with the background counts subtracted and the error bars plotted at each data point. A fit to the data using Eq. (3) is shown by the solid curve.

for $t > 200$ ms. The error bar at each data point reflects both the statistical counting error and the error in the background subtraction. The radiative lifetime τ of the metastable state was obtained by fitting Eq. (3) to these data using τ and α as the fitting parameters. The fit was done to the data points between $t = 0$ and a zero-crossing point where a negative data value first emerged ($t = 47$ ms in this case). The result is shown by the solid curve in the figure, yielding a radiative lifetime of $\tau = 17.7 \pm 0.7$ ms. It was confirmed that the calculated value of τ had a tolerance within ± 0.1 ms with a shift in the starting or ending bin towards lower or higher time by a few bins. Incidentally, the fit ignoring the rapid decay of the stored ions [$1/\tau_r = 0$ in Eq. (3)] and the fit ignoring both the decays [$1/\tau_c, 1/\tau_r = 0$ in Eq. (3)] yielded $\tau = 17.0 \pm 0.6$ ms and $\tau = 16.9 \pm 0.6$ ms, respectively, which shows that the influence of the rapid decay cannot be ignored in the determination of τ .

Since a major fraction of the ions injected into the trap are expected to be in the 1D_2 and $^3P_{1,2}$ states [10], the observed storage lifetime due to collisional loss ($\tau_c = 5028$ ms) predominantly reflects the loss of ions not in the 1S_0 state. It can be approximated as $\tau_c \sim 1/\sigma^{D,P} v \rho$, where $\sigma^{D,P}$ represents the collisional loss cross sections for the 1D_2 and $^3P_{1,2}$ states. This value was applied to Eq. (3) in the determination of the radiative lifetime, where the storage lifetime due to collisional loss of the 1S_0 state must be used. This may cause an additional systematic error. We roughly estimated the difference in the collisional loss cross sections for the 1S_0 state and other states in the following manner. Considering the 1S_0 state, the quotient between the photon spectra measured at different residual gas densities (vacuum pressures) of ρ_1 and ρ_2 is given by Eqs. (2) and (3) as

$$\frac{\alpha^{(1)} N_c^{(1)}(0) \exp\{-\sigma^S v \rho_1 t\} + \alpha^{(1)} N_r^{(1)}(0) \exp\{-\left(\frac{1}{\tau_r^{(1)}}\right)t\}}{\alpha^{(2)} N_c^{(2)}(0) \exp\{-\sigma^S v \rho_2 t\} + \alpha^{(2)} N_r^{(2)}(0) \exp\{-\left(\frac{1}{\tau_r^{(2)}}\right)t\}}, \quad (5)$$

where σ^S is the cross section of the collisional loss of the ions in the 1S_0 state and the upper index (i) indicates the measurement at the vacuum pressure of ρ_i . Regarding $\tau_r^{(i)}$, it would not be necessary to take the ion states of 1S_0 , 1D_2 , and $^3P_{1,2}$ into account, because the loss affected by the injection process (rapid decay in Fig. 4) is independent of the difference

between these states. The $\alpha^{(i)}N_c^{(i)}(0)$ and $\alpha^{(i)}N_r^{(i)}(0)$ terms depend only on the number of injected ions, their injection angle, the ratio of the 1S_0 states in the injected ions, and the detection efficiency of the optical system. Hence, the values of $\alpha^{(i)}$, $N_c^{(i)}(0)$, $N_r^{(i)}(0)$, and $\tau_r^{(i)}$ obtained using the neutral spectrum at ρ_i can be assigned without modification. The ρ_2 term can be replaced by $(\tau_c^{(1)}/\tau_c^{(2)})\rho_1$, where $\tau_c^{(i)}$ ($\sim 1/\sigma^{D,P}v\rho_i$) is also obtained from the neutral spectrum at ρ_i . Then, the value of $\overline{\sigma^S v\rho_1}$ is given by a fit of Eq. (5) to the quotient between the photon spectra measured at two different vacuum pressures. Finally, the ratio of the collisional loss cross sections for the 1S_0 state and the 1D_2 and $^3P_{1,2}$ states, $\sigma^S/\sigma^{D,P}$, is given by

$$\frac{\sigma^S}{\sigma^{D,P}} \approx \frac{\overline{\sigma^S v\rho_1}}{\overline{\sigma^{D,P} v\rho_1}} = \overline{\sigma^S v\rho_1} \times \tau_c^{(1)}. \quad (6)$$

We measured the photon and neutral spectra under the reduced vacuum condition ($\sim 2 \times 10^{-6}$ Pa) by slowing the speed of the turbo pump in the trap [in this case, the photon spectrum yielded $\tau = 14.6 \pm 0.6$ ms, ignoring $1/\tau_c$ and $1/\tau_r$ in Eq. (3)]. From these spectra and the present data at the higher vacuum condition ($\sim 5 \times 10^{-8}$ Pa), we obtained the ratio $\sigma^S/\sigma^{D,P} = 1.4 \pm 0.6$. This reduces the storage lifetime due to collisional loss used in the determination of the radiative lifetime by a factor of 2 at a maximum, but leads to an increase in the error of the radiative lifetime of only less than 1%.

Repetition of the experiment to improve precision yielded radiative lifetime values of $\tau = 17.2 \pm 0.7$ and 17.3 ± 0.7 ms in two more runs. Thus, the lifetime of the 1S_0 metastable state of Kr^{2+} was determined to be 17.4 ± 0.4 ms from a weighted average of these values.

The radiative lifetime of the metastable 1S_0 state obtained in the present study is compared with previously reported theoretical and experimental lifetimes in Table I. Our result is compatible with the theoretical predictions [3,4] within the experimental error. Compared with the two earlier measurements [5,6], our result yields a larger lifetime even with the experimental error taken into account. The reason for this discrepancy remains unclear, but there are differences in the experimental methods between this and the earlier works. The ion source in our experiment is placed at a distant position outside the trap while the ion sources in the earlier experiments are located inside the trap. The external ion source enables us to maintain the ultrahigh vacuum ($\sim 5 \times 10^{-8}$ Pa) of the

TABLE I. Comparison of theoretical and experimental values of the lifetime of the 1S_0 metastable state of Kr^{2+} .

Lifetime (ms)	Reference	Theory or Experiment
17.2	Garstang [3]	Theory
17.3	Biemont and Hansen [4]	Theory
13.1 ± 0.6	Walch and Knight [5]	Experiment
14.8 ± 0.8	Calamai and Johnson [6]	Experiment
17.4 ± 0.4	This work	Experiment

trap during measurements, which is two to three orders of magnitude higher than the vacuum levels in the earlier experiments. In addition, the external ion source also enables us to select and inject only the Kr^{2+} ions into the trap. Another difference is in the method for the correction of the loss of stored ions. In the present experiment, the ion loss as a function of the storage time was monitored directly through the measurement of the neutrals escaping the trap. On the other hand, the earlier experiments measured the photon spectra at various pressures of the Kr^{2+} -ion parent gas introduced in the trap, and the corrections were applied by extrapolating the results to the zero pressure of the parent gas.

In summary, we have determined the radiative lifetime of the $4s^24p^4\ ^1S_0$ metastable state of Kr^{2+} using an electrostatic ion trap. The lifetime was measured by detecting the photons emitted by the $M1$ transition of the 1S_0 state to the 3P_1 state (350 nm). The loss of the stored ions in the trap as a function of the storage time was monitored by measuring the Kr neutrals, and two decay components of the number of stored ions were observed: one due to the collision-induced decay with residual gases in the trap and another associated with the stabilization of the ions in the trap. The measured lifetime was corrected for these decays. Our experimental lifetime is in fair agreement with the previously reported theoretical values. On the other hand, our value is about 20%–30% larger than the earlier measurements using other types of traps with internal ion sources and a different correction method for the loss of stored ions.

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