

# The 1s Lamb Shift in Hydrogenlike Rhodium Measured with an Electron Beam Ion Trap

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(Received March 3, 2003)

The 1s Lamb shift in hydrogenlike rhodium has been measured by observing radiative recombination into the 1s vacancy of bare rhodium produced and trapped in the Tokyo electron beam ion trap. The 1s binding energy was determined by measuring the radiative recombination X-ray energy and subtracting the electron beam energy from it. The electron beam energy was measured by simultaneously observing radiative recombination into the 1s vacancy of bare krypton, for which the level energy is precisely known from an existing experiment. The 1s Lamb shift in hydrogenlike rhodium has been found to be  $28.7 \pm 3.4$  eV. At the same time, the two electron contribution to the ground state energy of heliumlike rhodium and krypton have been determined. Experimental results are compared with existing theories.

KEYWORDS: highly charged ion, electron beam ion trap, Lamb shift

DOI: 10.1143/JPSJ.72.1650

## 1. Introduction

The Lamb shift of a single state is defined as the energy difference between the true binding energy and the point-nucleus Dirac eigenvalue with the nonrelativistic reduced mass correction. For hydrogenlike ions, the Lamb shift can be written as

$$E_{\text{LS}} = \frac{\alpha (\alpha Z)^4}{\pi n^3} F(\alpha Z) m_e c_0^2, \quad (1.1)$$

where  $\alpha$  is the fine structure constant,  $Z$  the atomic number,  $n$  the principal quantum number,  $m_e$  the electron rest mass,  $c_0$  the velocity of light in vacuum, and  $F(\alpha Z)$  a dimensionless slowly varying function. As understood from the eq. (1.1), the Lamb shift is largest for the ground state and rapidly increases with  $Z$ . It is thus useful to measure the ground state energy of hydrogenlike heavy ions to test the quantum electrodynamics (QED) theory. In calculation of the Lamb shift for low- $Z$  ions, perturbative methods based on the  $\alpha Z$ -expansion of the Coulomb interaction between the electron and the nucleus are very useful. However, since  $\alpha Z \sim 1$  for high- $Z$  ions, the perturbative methods based on the  $\alpha Z$ -expansion break down. It is thus necessary to develop nonperturbative methods for calculation of the Lamb shift in high- $Z$  hydrogenlike ions. To date, several theoretical attempts have been done. Refer to ref. 1 and references therein for the recent developments of theoretical methods.

To examine these theoretical methods, systematic comparison with experiments is needed. The 1s Lamb shift in atomic hydrogen has been precisely measured with an accuracy of several ppm.<sup>2-4</sup> This accuracy has been achieved by the two photon laser spectroscopy of the 1S–2S energy separation with a frequency doubled dye laser. However, this method can not be applied for hydrogenlike ions with  $Z \geq 2$  because the 1S–2S energy separation becomes so large that the transition can not be excited with an existing laser. The usual way to measure the 1s Lamb shift in hydrogenlike ions is to measure the wavelength of the Lyman- $\alpha$  ( $2p \rightarrow 1s$ ) transitions which fall in the X-ray region for  $Z \gtrsim 10$ . In these measurements, the 1s Lamb shift can be deduced by taking account of theoretical energy of the  $2p$  level, which can be calculated very accurately. For

intermediate to heavy elements, measurements have been done for fast-moving ions with accelerators. In those experiments, accuracy was limited by the Doppler correction, i.e. the uncertainties in the observation angle and the ion velocity. Attempts to decrease the Doppler uncertainties were done for hydrogenlike gold and uranium in experiments with an electron cooler at GSI (Gesellschaft für Schwerionenforschung).<sup>5</sup> In these experiments, radiative recombination (RR) between the bare target and the cooler electron whose velocity is the same as that of the target ion were observed at an observation angle of almost  $0^\circ$ . With this arrangement, the error arising from the observation angle uncertainty became negligible. Although they achieved very high accuracy ( $\Delta E/E_{\text{LS}} \sim 4 \times 10^{-2}$ ), the error arising from the velocity uncertainty still made large contribution to the total experimental uncertainty.

In this paper, we present 1s Lamb shift measurement for hydrogenlike rhodium ( $Z = 45$ ) performed with the Tokyo electron beam ion trap (Tokyo-EBIT).<sup>6-8</sup> In an EBIT, trapped ions are at rest, so that no Doppler correction is needed. Higher accuracy is thus expected compared to experiments with fast-moving ions. However, spectra from an EBIT usually contain contribution from several charge states, so that Lyman- $\alpha$  can not be clearly resolved from the K-lines of other charge states when an usual Ge detector is used. To overcome this problem, Beiersdorfer *et al.*<sup>9</sup> observed the spectrum from highly charged xenon ions in the magnetic trapping mode of an EBIT. Since there is no electron-impact excitation in the magnetic trapping mode, only the hydrogenlike and bare ions can contribute to the spectrum. Thus the obtained spectrum was so simple that they could determine the energy of the Lyman- $\alpha$  transition. Meanwhile in the present measurement, RR X-rays were observed in the normal operation mode. RR spectra for capture into the  $n = 1$  vacancy are simple since only the hydrogenlike and bare ions can contribute to them. In addition, since there is no bremsstrahlung in the RR region, clear lines without a background can be obtained. Since the energy of RR X-rays is the sum of the electron beam energy and the binding energy of the captured level, binding energy can be experimentally obtained by measuring the energy of the RR X-ray if the electron energy is known. However, it is

generally difficult to know the electron beam energy in an EBIT. In the present measurement, RR X-rays for highly charged krypton ions, for which the level energy is accurately known, were observed simultaneously to determine the electron beam energy. In §2, the experimental principle and the procedure will be described in detail. The experimental results and discussion will be given in §3.

## 2. Experiments

### 2.1 Principle

In an EBIT, a nearly monoenergetic electron beam with a width of  $\sim 50$  eV interacts with trapped ions. When the target ion is bare, the beam electron can be captured into the  $1s$  vacancy with emitting an RR X-ray whose energy ( $E_{RR}$ ) is the sum of the electron energy ( $E_e$ ) and the  $1s$  binding energy ( $E_{1s}$ ) of the recombined hydrogenlike ion:

$$E_{RR} = E_e + E_{1s}. \quad (2.1)$$

Thus, if  $E_{1s}$  is known,  $E_e$  can be determined by measuring  $E_{RR}$ . For example, the  $1s$  energy of hydrogenlike krypton is known with an accuracy of 0.5 eV.<sup>10)</sup> Thus, by measuring the RR X-ray energy for bare krypton, the electron beam energy can be determined with an accuracy of up to 0.5 eV. The other way around, once the electron energy is determined, the  $1s$  binding energy of hydrogenlike heavy ions can be obtained by measuring the RR X-ray energy for the bare ion.

In the present measurement, the RR transitions into the  $1s$  vacancy of bare krypton and rhodium were observed simultaneously:

$$E_{RR}^{Kr} = E_e + E_{1s}^{Kr} \quad (2.2)$$

$$E_{RR}^{Rh} = E_e + E_{1s}^{Rh}. \quad (2.3)$$

The difference between the above equations gives

$$E_{RR}^{Rh} - E_{RR}^{Kr} = E_{1s}^{Rh} - E_{1s}^{Kr}. \quad (2.4)$$

Thus, the  $1s$  binding energy of hydrogenlike rhodium ( $E_{1s}^{Rh}$ ) can be determined by measuring the difference in RR X-ray energy between rhodium and krypton because  $E_{RR}^{Kr}$  is accurately known.

### 2.2 Experimental procedure

The present experimental setup and procedure are similar to those used by Marrs *et al.*<sup>10)</sup> in the measurements of two-electron contributions to the ground state energy of helium-like ions. The present experimental arrangement is shown in Fig. 1. Highly charged rhodium and krypton were produced and trapped in the Tokyo-EBIT.<sup>6-8)</sup> In an EBIT, an electron beam emitted from a cathode is accelerated towards an ion trap with magnetic compression by a superconducting magnet. The ion trap consists of three successive drift tubes (DTs), in which positive ions can be trapped axially by applying a positive bias to the two outer DTs. The electron beam successively ionize the trapped ions, and finally highly charged ions are produced and trapped in the middle DT. Rhodium was injected from a metal vapor vacuum arc (MEVVA) source<sup>11)</sup> installed at the top of the EBIT, while krypton was continuously injected from a gas injector installed at the same level as the middle DT. The injection methods for gas and metal ions are described in detail

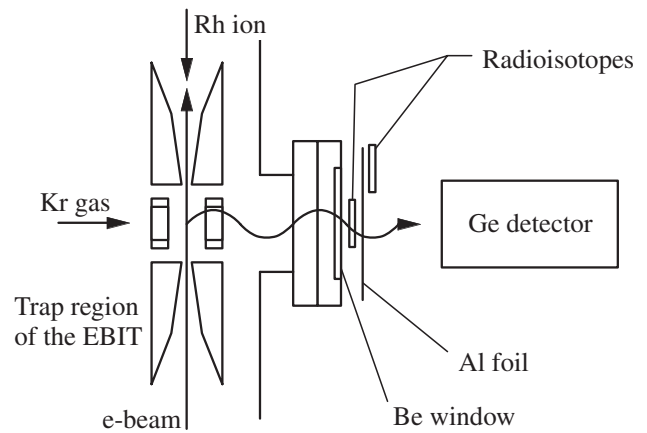


Fig. 1. Schematic layout of the present experimental setup. Rhodium ions were injected in a pulsed mode from a MEVVA source, while krypton was continuously injected from a gas injector. The distance from the center of the trap to the beryllium window was about 300 mm, and that between the window and the germanium detector was about 100 mm.

Table I. Energy of the reference lines used in the present measurements.<sup>12)</sup>

| Radioisotope      | Energy (eV) | $\Delta E$ (eV) |
|-------------------|-------------|-----------------|
| <sup>109</sup> Cd | 88 033.60   | 1.03            |
| <sup>57</sup> Co  | 122 060.65  | 0.12            |
| <sup>57</sup> Co  | 136 473.56  | 0.29            |

elsewhere.<sup>11)</sup>

RR X-rays were observed with a planar germanium detector (EURISYS MESURES, EGP500-15) through a beryllium window with a thickness of 1 mm and a radius of 25 mm. The two radioisotopes <sup>109</sup>Cd and <sup>57</sup>Co were used to calibrate X-ray energy. Table I lists the  $\gamma$ -ray energy of the references.<sup>12)</sup> Radiations from these isotopes were measured simultaneously with the RR X-rays to examine the pulse height drift arising from the instability of the detector and the electronics. An aluminum foil 0.3 mm thick was placed between the window and the detector to attenuate intense characteristic X-rays from krypton and rhodium ions and prevent the piling up of the signal pulses. The intensity of radiations from the isotopes was adjusted by covering them with an aluminum sheet and by adjusting the distance from the detector.

RR spectra were acquired as follows. After injecting rhodium ions from the MEVVA source, trapping potential was applied to the trap. Data acquisition was then started after waiting for 1.5–3.5 s for ion production. Observation was continued for 8–12 s, and then the trapped ions were dumped by removing the trapping potential and rhodium ions were injected again. The dumping was needed to avoid the accumulation of contaminants such as Ba and W evaporated from the cathode. One run was terminated after repeating this cycle for 1–1.5 h. More than 10 runs were integrated to obtain the final spectrum. Several runs which were found to show large drift were excluded from the integration. As a result, the drift of each peak was restrained within  $\pm 35$  eV, which is much small compared to  $\pm 200$  eV in ref. 10.

In the present measurements, the electron–ion interaction

energy for rhodium and krypton ions must be the same. It was thus essentially important to trap and observe rhodium and krypton simultaneously. If rhodium and krypton were observed alternately, the degree of the neutralization of the electron beam can change, and thus the electron-ion interaction energy can change. The simultaneous observation is also important to prevent the experimental uncertainty arising from the instability of the electronics and so on. It was thus confirmed that krypton and rhodium ions existed in the trap at the same time by observing the time evolution of the RR X-rays.

Measurements were done at two different electron energies 74 keV and 106 keV to examine systematic uncertainties. The electron beam currents were 155 mA and 135 mA respectively.

### 3. Results and Discussion

Figure 2(a) shows a spectrum obtained at an electron energy of 106 keV. The peak width was dominated by the detector resolution, which is  $\sim 550$  eV full width at half maximum (FWHM). The energy spread of the electron beam ( $\sim 50$  eV) was too small to contribute to the peak width. Although the observation was performed for  $\sim 17$  h, data

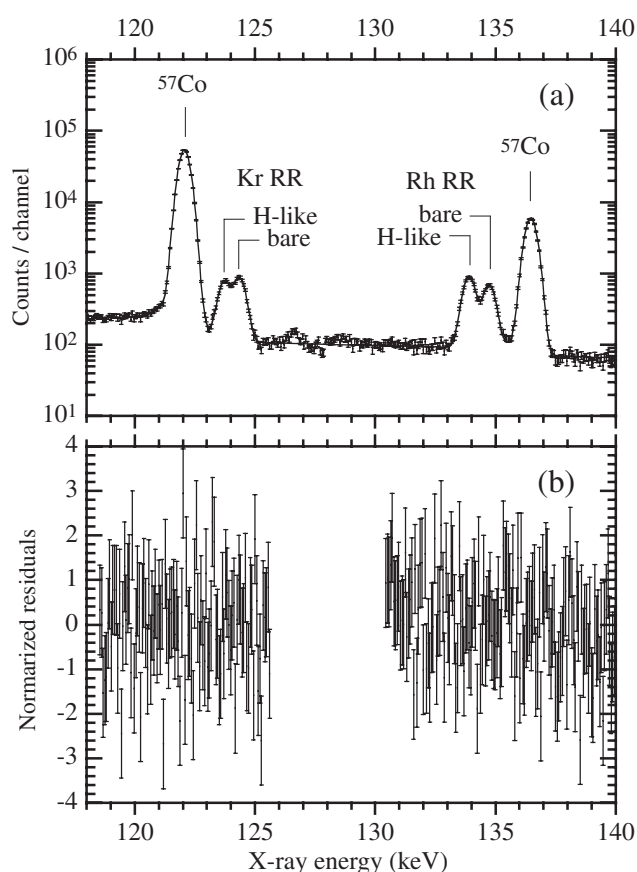


Fig. 2. (a) RR spectra obtained at an electron energy of 106 keV. Peaks around 124 keV and 134 keV corresponds to RR for krypton and rhodium ions, respectively. There are two peaks for each element; one of them, which appear in the higher energy side, corresponds to RR into the  $1s$  vacancy of the bare ion, and another corresponds to RR into the  $1s$  vacancy of the hydrogenlike ion. Peaks at 122 keV and 136 keV are the reference lines from the radioisotope  $^{57}\text{Co}$ . The solid line is the peak function fitted to the data. (b) Normalized residuals of the fit. The region between 125.7 and 130.4 keV were excluded from the fit procedure.

showing large drift were excluded from integration to obtain the final spectrum shown in Fig. 2(a). As a result, the practical live time was  $\sim 11$  h. Two reference lines from  $^{57}\text{Co}$  were recorded simultaneously. The value of interest in this experiment is the difference between krypton and rhodium in the X-ray energy for RR into the  $1s$  vacancy of the bare ion, i.e. the difference in the  $1s$  binding energy of the hydrogenlike ion. To determine the center of each peak, the following peak function  $F(x)$  presented by Longoria *et al.*<sup>13)</sup> was fitted to the experimental spectrum,

$$F(x) = F_1 + F_2 + F_3 + F_4 \quad (3.1)$$

$$F_1 = P_1 \exp\left[-\frac{1}{2}z^2\right] \quad : \text{Gaussian function}$$

$$F_2 = \frac{P_6}{[1 + \exp(z)]^2} \quad : \text{Step function}$$

$$F_3 = \frac{P_4[\exp(P_5z)]}{[1 + \exp(z)]^4} \quad : \text{Lower exponential}$$

$$F_4 = \frac{P_9[\exp(P_{10}z)]}{[1 + \exp(-z)]^4} \quad : \text{Higher exponential}$$

$$z = (x - P_2)/P_3,$$

where  $P_1$  to  $P_{10}$  are the fitting parameters. However, only  $F_1$  was considered for the RR peaks because the counting statistics of these peaks were so small that the contribution of  $F_2$ ,  $F_3$ , and  $F_4$  were negligibly small. The higher exponential  $F_4$  was also ignored for the reference line at 136 keV because the contribution of it was considered to be small compared with the background. In the fitting procedure, the ratios  $P_4/P_1$  and  $P_6/P_1$  (i.e., the contribution of  $F_3$  and  $F_2$  to  $F_1$ ) for the two reference lines were constrained to be the same. The peak width of the RR lines for the hydrogenlike and bare ions is also constrained to be the same. The background was assumed to be a linear function. The solid line in Fig. 2(a) represents the results of the fit, and Fig. 2(b) shows the residuals of the fit. No significant residual is found, suggesting that the peak functions used in the fitting procedure was appropriate. The values of interest obtained are listed in the first column of Table II. The experimental errors represent the statistical ones. The second and third rows are the two electron contribution to the ground state energy of the heliumlike ions, which will be discussed later.

Similar observation was performed also at an electron energy of 74 keV to examine systematic errors. The spectrum is shown in Fig. 3(a). It took 13.5 h to obtain this spectrum. The peak function (3.1) was fitted to the spectrum in a way similar to the spectrum obtained at  $E_e = 106$  keV, but the higher exponential  $F_4$  was ignored both for the  $^{109}\text{Cd}$  line at 88 keV and for the  $^{57}\text{Co}$  line at 122 keV. At first, an ordinary least square fit was tried, i.e. the data was weighted by the statistical error in the fitting procedure. From the result of the ordinary fit, small contribution of RR for the  $n = 2$  orbitals of tungsten ions were found to exist in the region of RR for krypton. The contribution of tungsten was estimated from the ordinary fit to be 70 counts per channel at the maximum. Thus the final fit was performed with assuming the counting error to be the root sum square of 70 and the statistical error for the region between 84.5 and

Table II. Experimental results. 1st row: difference in X-ray energy for RR into the 1s vacancy of the bare ion between rhodium and krypton; 2nd row: two electron contribution to the ground state energy of heliumlike krypton, which corresponds to the difference in X-ray energy between RR into the 1s vacancy of the bare ion and that of the hydrogenlike ion; 3rd row: similar to the 2nd row, but for heliumlike rhodium. Results obtained at  $E_e = 106$  keV are listed in the column “present 1”, and that at  $E_e = 74$  keV in “present 2”. All values are given in eV.

|                             | Present 1           | Present 2           | Weighted mean       | Theory            | Difference     |
|-----------------------------|---------------------|---------------------|---------------------|-------------------|----------------|
| $E_{RR}^{Rh} - E_{RR}^{Kr}$ | $10\,380.0 \pm 7.0$ | $10\,369.7 \pm 3.9$ | $10\,372.1 \pm 3.4$ | $10\,375.76^{a)}$ | $-3.6 \pm 3.4$ |
| Kr 2-e contribution         | $636.9 \pm 7.9$     | $642.2 \pm 4.9$     | $640.7 \pm 4.1$     | $639.94^{a,b)}$   | $0.8 \pm 4.1$  |
| Rh 2-e contribution         | $833.2 \pm 6.3$     | $823.4 \pm 2.7$     | $824.9 \pm 2.5$     | $825.35^{a,b)}$   | $-0.4 \pm 2.5$ |

a) Ref. 15 b) Ref. 17

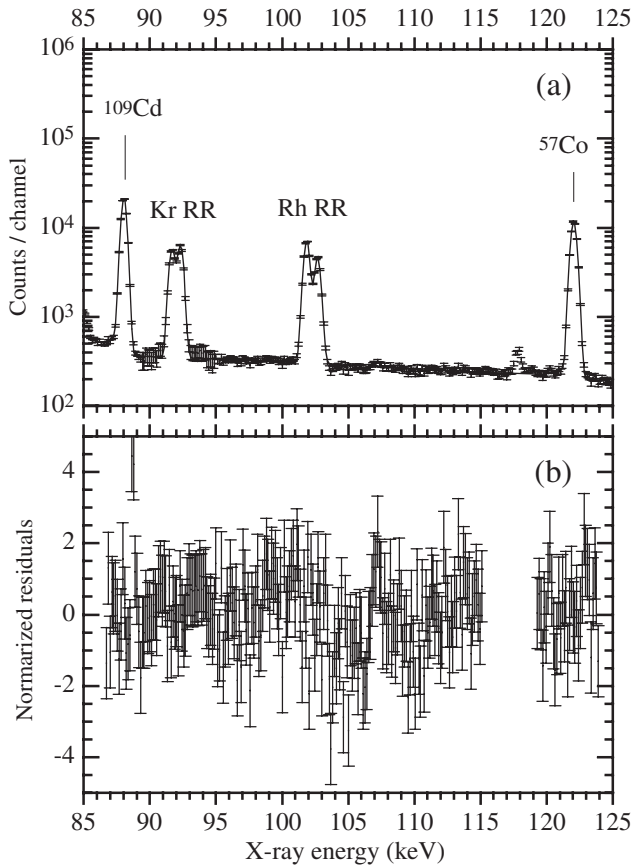


Fig. 3. (a) RR spectra obtained at an electron energy of 74 keV. Peaks around 92 keV and 102 keV corresponds to RR for krypton and rhodium ions, respectively. Peaks at 88 keV and 12 keV are the reference lines from the radioisotopes  $^{109}\text{Cd}$  and  $^{57}\text{Co}$ . (b) Normalized residuals of the fit. The region between 115.2 and 119.1 keV were excluded from the fit procedure.

90.0 keV. The solid line in Fig. 3(a) represents the result of the fit, and in Fig. 3(b) the residuals of the fit are shown. Slightly significant residuals are found around 88.7 keV. It may be the contribution of the higher exponential  $F_4$  for the  $^{109}\text{Cd}$  reference line. A fit with including  $F_4$  was thus tried, but the parameters did not converge to a reasonable value. Thus  $F_4$  was excluded in the present analysis. The values of interest obtained are listed in the second column of Table II.

Since there is no inconsistency between the two results, the final values were obtained as a weighted mean of the two results (the reciprocal of the square of the each experimental error was used as the weight) as shown in the third column of Table II.

The energy of the  $2p_{3/2}-1s$  transition (Lyman- $\alpha_1$ ) in hydrogenlike krypton was precisely measured by Tavernier

Table III. Present result for the 1s binding energy and the 1s Lamb shift in hydrogenlike rhodium.

|                                     | Present           | Theory <sup>a)</sup> | Difference     |
|-------------------------------------|-------------------|----------------------|----------------|
| Rh <sup>44+</sup> 1s binding energy | $28308.4 \pm 3.4$ | $28311.96 \pm 0.03$  | $-3.6 \pm 3.4$ |
| Rh <sup>44+</sup> 1s Lamb shift     | $28.7 \pm 3.4$    | $25.13 \pm 0.03$     | $3.6 \pm 3.4$  |

a) Ref. 15

*et al.*<sup>14)</sup> to be  $13508.95 \pm 0.5$  eV. Taking account of the theoretical  $2p_{3/2}$  energy, which was accurately calculated by Johnson and Soff<sup>15)</sup> to be  $-4427.28$  eV including the Lamb shift, the 1s binding energy of hydrogenlike krypton is estimated to be  $-17936.23 \pm 0.5$  eV. Using this value, we determined the 1s binding energy of hydrogenlike rhodium from eq. (2.4) to be  $-28308.4 \pm 3.4$  eV. By subtracting the point-nucleus Dirac eigenvalue, which is 28337.09 eV including the nonrelativistic reduced mass correction, we have obtained the 1s Lamb shift of hydrogenlike rhodium to be  $28.7 \pm 3.4$  eV. The present results are summarized in Table III.

It is noted that the experimental uncertainties represent only the counting errors (estimated from the statistical error and the influence of the superimposed tungsten lines) because other sources of error are considered to be negligible as described in the following.

(1) The error arising from the non-linear response of the detector and the electronics was checked by examining two different methods of energy calibration for the experiment at  $E_e = 74$  keV. One of those was calibration with a linear function which was uniquely derived from the two reference lines  $^{109}\text{Cd}$  (88 keV) and  $^{57}\text{Co}$  (122 keV), and another with a linear function derived with taking account of three reference lines (the above two lines and  $^{57}\text{Co}$  at 136 keV). There was no significant difference between the two results obtained from these two calibration procedures, suggesting that the effect of the non-linearity was negligible.

(2) The amount of the trapped ions was not stable during measurement because the number of ions injected from the MEVVA ion source was not stable but different shot by shot. Thus the ratio between krypton and rhodium ions was not stable. The difference in the total charge of the trapped ions could cause the difference in the electron beam energy. However, this effect is estimated to be much less than 1 eV, and thus considered to be negligible as long as the two elements were observed simultaneously.

(3) The instability of the electronics and the detector in the observation for many hours was checked as described above, and the scattered data was excluded from the analysis. Thus the detector instability is not problem in the present

measurement.

In Table III, a theoretical result obtained with a non-perturbative numerical method by Johnson and Soff<sup>15)</sup> is also listed. The difference between the experiment and the theory is slightly larger than the experimental uncertainty. However, considering that the experimental uncertainty represents the 68% confidence limit, it can be concluded that the theoretical value agree well with the experiment. The theoretical uncertainty (0.03 eV) is the root sum square of the uncertainties in estimation of the following contribution to the Lamb shift: the self energy ( $\sim 0.01$  eV), the nuclear size effect on the self energy ( $\sim 0.01$  eV), the higher order vacuum polarization corrections ( $\sim 0.003$  eV), the higher order radiative corrections ( $\sim 0.018$  eV), the nuclear size effect on the Dirac energy ( $\sim 0.014$  eV), and the relativistic recoil correction ( $\sim 0.01$  eV). Unfortunately, the experimental uncertainties are not enough to test these theoretical uncertainties.

The difference in X-ray energy between RR into the hydrogenlike ion and that into the bare ion is also important quantity. It is equal to the difference in the ionization energy between the hydrogenlike and bare ions, which corresponds to the two electron contribution to the ground state energy of heliumlike ions.<sup>10,16)</sup> As the atomic number increases, the finite nuclear size effect on energy levels becomes very large. This effect comes to prevent ones to investigate pure QED effects. However, the finite nuclear size effect cancels out by investigating the difference in the ionization energy between the ions with the same nucleus. The second and third rows in Table II list the present results for the two electron contribution for heliumlike krypton and rhodium, respectively. The theoretical values in the table, which are the difference between the heliumlike energy obtained with the unified theory<sup>17)</sup> and the hydrogenlike energy from ref. 15, agree well with the experimental values. For the 1s energy of heliumlike krypton, there are other theoretical values obtained from the relativistic configuration-interaction calculation<sup>18)</sup> and the relativistic many-body perturbation theory.<sup>19)</sup> The former gives 639.29 eV as a two-electron contribution and the latter gives 639.74 eV; both values agree well with the present experimental value as well as that obtained from the unified theory.

In summary, we have measured the 1s Lamb shift of hydrogenlike rhodium by observing the radiative recombination X-ray emitted in electron capture into the 1s vacancy of bare rhodium produced and trapped in the Tokyo electron beam ion trap. Compared to measurements with an accelerator, the present method using trapped ions has the strong

point that the experimental uncertainty is limited only by the counting error. By applying the present method to heavier ions, it is expected that the accuracy of existing measurements will be improved. However, when the accuracy beyond the present measurement is required, it may need to develop a detector with higher energy resolution or to develop an EBIT capable of trapping more ions. The present experimental uncertainty is less than 1% of the FWHM of the peak, thus it may be difficult to increase the accuracy only by increasing observation time.

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