Measurement of the Two-Loop Lamb Shift in Lithiumlike U^{89+}

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(Received 8 April 2005; published 1 December 2005)

Using the SuperEBIT electron beam ion trap, we have measured the $2s_{1/2}$-$2p_{1/2}$ transitions in U^{88+} and U^{89+}. The measured value of 280.645 ± 0.015 eV for Li-like U^{89+} improves the available precision by nearly an order of magnitude and establishes a new benchmark for testing the total QED contribution to the transition energy within a fractional accuracy of 3.6×10^{-4}. We infer a value for the $2s$ two-loop Lamb shift in U^{89+} of -0.23 eV, from which we estimate a value of -1.27 eV for the $1s$ two-loop Lamb shift in U^{91+}.

DOI: 10.1103/PhysRevLett.95.233003 PACS numbers: 31.30.Jv, 12.20.Fv, 32.30.Rj

The Coulomb field of heavy nuclei provides a strong-field environment for testing bound-state quantum electrodynamics (QED) not afforded by low-Z atoms or ions. Tests of one-loop QED (self-energy and vacuum polarization) in high-Z ions have confirmed theory, and theoretical interest has shifted to the evaluation of higher-order QED. Particular focus has been directed to the two-loop self-energy correction, which has only recently been successfully evaluated for highly charged ions [1]. For the $1s$ ground level in U^{91+}, the calculated value is -1.56 eV. Combined with the remaining two-loop terms, this yielded a contribution of -1.26 eV to the $1s$ Lamb shift [2,3]. Measurements sensitive to the $1s$ QED term of hydrogen-like uranium U^{91+}, the heaviest naturally occurring element, have achieved an accuracy of 4.6 eV [4]. This corresponds to a fractional accuracy of 1.7% when compared to the total $1s$ QED contribution of about 267 eV, but it is insensitive to two-loop corrections. By contrast, a measurement of the $2s$ QED energy in lithiumlike Bi^{80+} achieved an accuracy of 0.039 eV [5], providing a fractional accuracy of 1.5×10^{-3}. Measurements of lithium-like systems thus promise to be more sensitive to higher-order QED terms than those of hydrogenic systems. However, the calculation of QED terms for lithiumlike ions is more complex than for hydrogenlike ions due to the presence of two additional electrons. This complexity has been overcome by recent successful calculations of the two-photon exchange correction to the $2s_{1/2}$-$2p_{1/2}$ transition in lithiumlike ions [6], and only the two-loop Lamb shift contributions remain uncalculated in second-order QED. This fact and the assumption that three-photon physics can be neglected have been used by Sapirstein and Cheng to estimate the two-loop Lamb shift correction in lithiumlike Bi^{80+} from the experimental data, resulting in a value of 0.175 eV, which is 4 times larger than the experimental error limits [7].

Lithiumlike uranium represents the ion of choice for testing bound-state strong-field QED, but the available accuracy has remained the same for over a decade. A measurement of the 280-eV $2s_{1/2}$-$2p_{1/2}$ transition by Schweppe *et al.* using Doppler-tuned spectroscopy on the Bevalac heavy-ion accelerator achieved an accuracy of 0.10 eV [8]. Relying on the fact that calculations of dielectronic recombination resonance energies equal those of the $2s_{1/2}$-$2p_{1/2}$ transitions energy, provided a small experimentally measured correction is added, Brandau *et al.* recently reported a value with an accuracy of 0.099 eV based on a measurement carried out using the heavy-ion accelerator and storage ring facility at Darmstadt, Germany [9]. In this Letter, we report a direct measurement of the $2s_{1/2}$-$2p_{1/2}$ transition energy in lithiumlike U^{89+} based on extreme ultraviolet (EUV) emission spectroscopy. Our accuracy is 0.015 eV, which improves the available accuracy for this ion by nearly an order of magnitude and provides a 360 ppm test of the approximately 42 eV total QED contribution to this transition. It is also more than an order of magnitude more precise than a Bragg-crystal spectrometer measurement of the 4.1-keV U^{89+} $2s_{1/2}$-$2p_{3/2}$ transition, which achieved an accuracy of 0.26 eV [10]. The new benchmark allows us to infer the value of the two-loop Lamb shift. We use this value in turn to estimate the value of the $1s$ two-loop Lamb shift in U^{91+}, providing the first test of recent two-loop Lamb shift calculations in high-Z hydrogenlike ions, and remarkably good agreement is obtained. Our measurement approach has the advantage that the close-by $2s^2\,{}^1S_0$-$2s_{1/2}2p_{1/2}\,{}^3P_1$ transition in berylliumlike U^{88+} is observed concurrently. It is similarly sensitive to QED corrections as the lithium-like $2s_{1/2}$-$2p_{1/2}$ transition and, thus, provides another benchmark for testing multielectron QED calculations.

The present measurements were carried out using the SuperEBIT high-energy electron beam ion trap [11] at the University of California Lawrence Livermore National Laboratory. Lithiumlike uranium ions were produced and excited by successive collisions with a 150-keV, 200 mA electron beam and confined in a 200-V potential applied to the upper and lower trap electrodes and the approximately 10-V radial space charge of the electron beam. The emission from the uranium ions was monitored in the x-ray regime to assess the ionization balance with a high-purity

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germanium detector focusing on the radiative recombination signal and a high-resolution microcalorimeter focusing on the $2s_{1/2}$-$2p_{3/2}$ x-ray transitions [10,12]. The ionization balance typically peaked around boronlike U^{87+} and carbonlike U^{86+}, depending on the specific run conditions.

Observations in the EUV were made with a grazing-incidence spectrometer specifically developed for this purpose. The instrument employed a 44.3 m radius of curvature, 2400 ℓ/mm grating, and a 1340×1300 pixel LN_2-cooled charge-coupled device (CCD) detector, as described in Ref. [13]. The resolving power of the instrument covering the wavelength range 35–47 Å was about $\lambda/\Delta\lambda \approx 1600$. A single spectrum was acquired by integrating for 30 min. The flux from the uranium lines was low, resulting in typically about two to ten counts in either the lithiumlike or berylliumlike lines in a given spectrum. Typically, ten 30-min spectra were added to produce a statistically meaningful spectrum, as illustrated in Fig. 1(a).

The spectrum in Fig. 1 shows the lithiumlike $2s_{1/2}$-$2p_{1/2}$ and the berylliumlike $2s^2\,{}^1S_0$-$2s_{1/2}2p_{1/2}\,{}^3P_1$ transition. It also shows the $1s^2\,{}^1S_0$-$1s_{1/2}2p_{3/2}\,{}^1P_1$ resonance transition in heliumlike C^{4+}, labeled w in common notation. The wavelength of this transition is known to better than 1 mÅ [14,15] and serves as a wavelength standard for our measurement. We made sure that the carbon line was visible in each uranium spectrum by continuously injecting a small amount of CO_2 into SuperEBIT. CO_2 also served as a coolant for trapping the uranium ions as described in Refs. [10,11]. Possible emission from heliumlike oxygen, which may blend in second order with the lithiumlike uranium line, has been suppressed by the energy discrimination afforded by the CCD detector. The presence of the carbon line anchored the wavelength scale and allowed us to account for any drift in the position of the spectral lines in time. A total of 17 spectra similar to that in Fig. 1 showing the U^{89+} line and 20 spectra showing the U^{88+} line were separately analyzed.

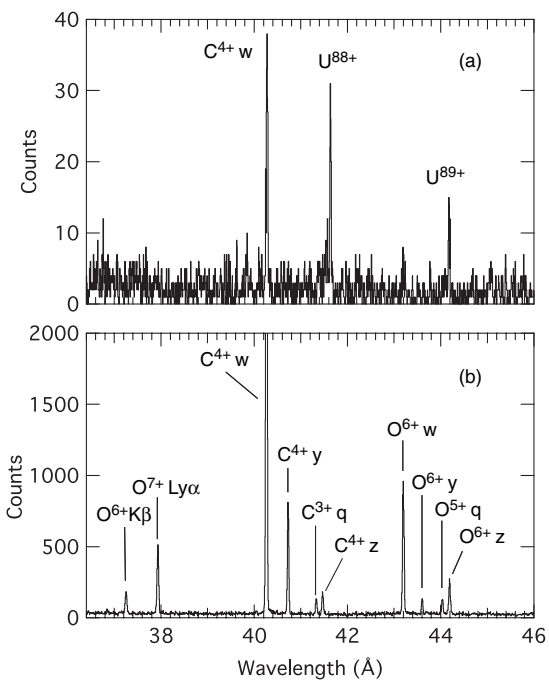

FIG. 1. Spectra obtained with the high-resolution SuperEBIT grating spectrometer. (a) Spectrum of the $2s_{1/2}$-$2p_{1/2}$ transitions in U^{88+} and U^{89+} representing the accumulation of ten 30-min exposures. (b) Calibration spectrum showing the emission of heliumlike and hydrogenlike carbon (first order) and oxygen (second order). The spectrum results from the addition of four 30-min exposures.

The wavelength dispersion of the spectrometer was determined by dedicated measurements of the K-shell emission of carbon in first order and oxygen in second order. A typical spectrum is shown in Fig. 1(b). The spectrum was produced by puffing large amounts of CO_2 into the trap; the beam current and beam energy, however, were kept the same as for the uranium measurements. The spectrum in Fig. 1(b) shows a variety of heliumlike and hydrogenlike lines of carbon and oxygen. These lines are all well known [14–16] and readily establish the wavelength scale and dispersion based on a quadratic fit.

The variation of the wavelengths inferred from each of the 17 measurements of the U^{89+} $2s_{1/2}$-$2p_{1/2}$ transition is shown in Fig. 2. The uncertainty limit of each measurement point is given by the quadrature sum of the statistical uncertainty associated with the determination of the centroids of the U^{89+} and C^{4+} reference lines, as well as an estimate of the possible error due to line blending given by the fact that the U^{89+} line is nearly coincident with the $1s^2$-$1s2s\,{}^3S_1$ forbidden line (labeled z) in O^{6+}. The amount of blending was determined by the (near) absence of the strong O^{6+} resonance line in the spectra after energy discrimination against second-order lines. A summary of the contributions to the overall uncertainty of the energy of the U^{89+} line is given in Table I.

A total of seven CO_2-injection calibration runs were recorded during the two-month period of this experiment. Each of the 17 spectra (or 20 in the case of berylliumlike

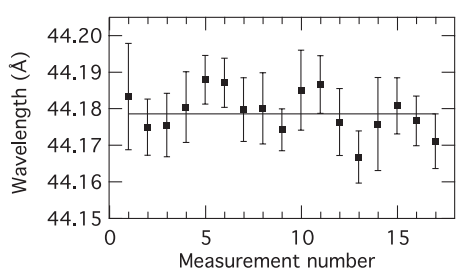

FIG. 2. Determinations of the $2s_{1/2}$-$2p_{1/2}$ wavelength in U^{89+} from 17 separate spectra for one of several wavelength calibrations. The error bars represent statistical uncertainties of each individual measurement. The weighted average is indicated by the solid line.

TABLE I. Contributions to the uncertainty of the U^{89+} $2s_{1/2}$-$2p_{1/2}$ transition energy.

Type	Magnitude (Å)
Position of U^{89+} line	0.0017
Position of C^{4+} line	0.0009
Blending with O^{6+} line	0.0004
Wavelength standards	0.0008
Wavelength dispersion	0.0011
Quadrature sum	0.0024

uranium) was calibrated against each of the seven calibration runs. The wavelength dispersion determined from each calibration was remarkably reproducible during this period, indicating few, if any, unaccounted-for systematic effects. As a result, the variation of the average wavelengths determined from the 17 U^{89+} spectra for the seven different calibrations is small, as shown in Fig. 3. The uncertainty in the wavelength dispersion is included in the overall uncertainty of the measurement, as given in Table I.

The wavelength value for the U^{89+} $2s_{1/2}$-$2p_{1/2}$ transition determined by our measurements is 44.1783 ± 0.0024 Å. This corresponds to 280.645 ± 0.015 eV, using the conversion factor $hc = 12\,398.42$ eVÅ [17]. Our value is in good agreement with the value of 280.59 ± 0.10 eV obtained with Doppler-tuned spectroscopy [8]. It is somewhat larger than the value of 280.516 ± 0.099 eV inferred from measurements of $1s^2 2p_{1/2} n\ell$ dielectronic resonance peaks and calculated values of the binding energy of the $n\ell$ Rydberg electron [9].

Similarly, we determine a wavelength value for the U^{88+} $2s^2\,{}^1S_0$-$2s_{1/2}2p_{1/2}\,{}^3P_1$ transition of 41.6335 ± 0.0017 Å, or 297.799 ± 0.012 eV. The uncertainty of this measurement is smaller because the signal rate of the U^{88+} transition is more than twice that of the U^{89+} transition. Moreover, it is closer to the C^{4+} reference line, making it less sensitive to errors in the wavelength dispersion.

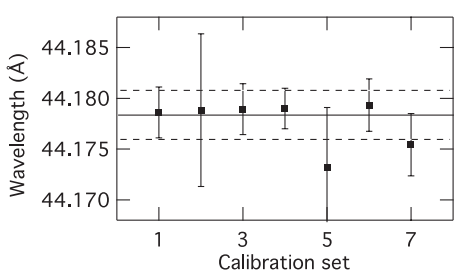

FIG. 3. Variation of the U^{89+} $2s_{1/2}$-$2p_{1/2}$ wavelength determinations for seven spectral calibrations taken at different times interspersed among the uranium measurements. Error bars represent the statistical uncertainty of each individual calibration. The weighted average is indicated by the solid line. The dashed lines mark the uncertainty limits of the final wavelength result.

Our measurement of the U^{89+} $2s_{1/2}$-$2p_{1/2}$ transition energy can be used to determine the two-loop Lamb shift. Rigorous calculations of all two-electron contributions of order α^2 have recently been completed, including the two-photon exchange term as well as estimates of higher-order photon exchange contributions [6,18,19]. Adding these to the one-photon exchange, first-order QED, nuclear recoil, nuclear polarization, and one-electron finite size contributions yield a value for the $2s_{1/2}$-$2p_{1/2}$ transition energy that misses only the two-loop Lamb shift contribution. The sum of these contributions, as given by Yerokhin *et al.* [6], Andreev *et al.* [18], and Sapirstein and Cheng [19], is listed in Table II. The differences among the three values in Table II are small and arise from differences in the estimated size of the three-photon exchange term. Error limits are purely theoretical estimates and are dominated by the uncertainty in the nuclear finite size correction to the binding energies and, in the case of Ref. [6], by the estimate of the three-photon exchange contribution. Subtracting these values from our measured transition energy yields the two-loop Lamb shift. It ranges from 0.175 to 0.215 eV, depending on the theoretical value used, as given in Table II. The results show that the two-loop Lamb shift is more than an order of magnitude larger than the uncertainty of our measurement. Once calculations of all QED terms of order α^2 are complete, the accuracy of the theoretical predictions will be limited by the uncertainty of the nuclear radius of uranium, as pointed out by Yerokhin *et al.* [20]. Calculations presented in Table II use the value of $\langle r^2 \rangle^{1/2} = 5.860 \pm 0.002$ fm [21]. The uncertainty of this value limits the accuracy of the QED calculations, in principle, to 0.02 eV. This is comparable to the uncertainty in our measurement and means that tests of QED to our level of accuracy should by possible in the future.

While calculations do not yet exist to compare the inferred $2s_{1/2}$-$2p_{1/2}$ two-loop Lamb shift in lithiumlike U^{89+} with theory, the two-loop Lamb shift of the $1s$ level in hydrogenlike U^{91+} has recently been calculated to be -1.26 ± 0.33 eV [1–3]. We can infer a value for the two-loop Lamb shift of the $1s$ level in hydrogenlike U^{91+} from our measurement by assuming the two-loop Lamb shift scales in similar fashion as the one-loop Lamb shift when

TABLE II. Calculated U^{89+} transition energies and two-loop Lamb shift for the U^{89+} $2s_{1/2}$-$2p_{1/2}$ transition and for the U^{91+} $1s$ level inferred from the measured energy of 280.645 ± 0.015 eV. All values are in eV.

Calculated transition energy	Reference	Inferred two-loop Lamb shift $2s_{1/2}$-$2p_{1/2}$ (U^{89+})	$1s$ (U^{91+})
280.44(10)	[6]	0.205	−1.31(64)
280.47(7)	[18]	0.175	−1.12(45)
280.43(7)	[19]	0.215	−1.37(45)

comparing U^{89+} with U^{91+}. First, we note that the one-loop Lamb shift of the $2s$ level is about 15% larger than that of the $2s_{1/2}$-$2p_{1/2}$ transition, because the $2p_{1/2}$ is also affected by QED effects. Second, we note that the U^{91+} $1s$ first-order Lamb shift is about 5.6 times larger than that of the U^{89+} $2s$ level. As a result, we estimate the U^{91+} $1s$ two-loop Lamb shift by multiplying the inferred two-loop Lamb shift from our measurement by -6.33. The results are given in Table II. The error limits are the scaled uncertainties of the theoretical estimates for the theoretical $2s_{1/2}$-$2p_{1/2}$ transition energies.

A comparison between the $1s$ two-loop Lamb shift calculated by Yerokhin *et al.* [1–3] and those estimated from our measurement yields very good agreement. The values inferred from the $2s_{1/2}$-$2p_{1/2}$ transition energy calculated by Yerokhin *et al.* [6] and Sapirstein and Cheng [19] are in near-perfect agreement, while that inferred from the energy calculated by Andreev *et al.* [18] is well within the uncertainty of the calculated $1s$ two-loop Lamb shift. From these, we obtain a weighted-average U^{91+} $1s$ two-loop Lamb shift of -1.27 ± 0.45 eV, where error limits reflect the scaled uncertainty of the calculated $2s_{1/2}$-$2p_{1/2}$ transition energies.

Calculations of the berylliumlike transition energies are by far less advanced than those of lithiumlike ions. In part, this is due to the increased complexity of these ions. Moreover, no experimental values for the $2s^2\,{}^1S_0$-$2s_{1/2}2p_{1/2}\,{}^3P_1$ transition energy in high-Z berylliumlike ions above xenon have been available to guide theory. Nevertheless, a comparison of our measured value with the calculated value of 297.744 eV by Chen and Cheng [22] and that of 298.177 eV by Safronova *et al.* [23] reveals agreement within -0.055 and 0.378 eV, respectively.

In summary, we have presented a benchmark for testing high-field QED in uranium based on passive emission spectroscopy that improves recent results based on an indirect measurement of dielectronic resonances by almost an order of magnitude. The accuracy of our measurement is more than an order of magnitude better than the estimated size of the two-loop self-energy correction, which has not yet been calculated. We infer the weighted average of the $2s_{1/2}$-$2p_{1/2}$ two-loop Lamb shift in lithiumlike U^{89+} to be 0.20 eV (or -0.23 eV for the $2s$ two-loop term). Our measurement also provides the first test of the recent two-loop Lamb shift calculation for the $1s$ level in hydrogenlike U^{91+}. We obtain a value of -1.27 eV, which is in excellent agreement with the calculated value of -1.26 ± 0.33 eV.

We are grateful to Jonathan Sapirstein, Kwok-Tsang Cheng, and Mau Chen for helpful discussions and communicating their calculated results before publication. We thank Ed Magee and Merv Lawrence for their technical support. This work was performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

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