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# Measurement of the two-loop Lamb shift in lithiumlike $U^{89+}$

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## Abstract

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 value of $280.645 \pm 0.015$ eV for Li-like $U^{89+}$ improves the available precision by nearly an order of magnitude and establishes a new benchmark for testing QED, including higher-order contributions, within a fractional accuracy of better than $3 \times 10^{-4}$. From our measurement, we infer a value for both the $2s$ and $1s$ two-loop Lamb shift, yielding excellent agreement with recent calculations of the 1.26 eV $1s$ two-loop Lamb shift in $U^{91+}$.

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The Coulomb field of heavy nuclei provides a test of bound-state quantum electrodynamics (QED) in a strong-field environment 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 1.55 eV. Measurements sensitive to the $1s$ QED term of hydrogenlike uranium $U^{91+}$, the heaviest naturally occurring element, have achieved an accuracy of 13 eV [2]. This corresponds to a fractional accuracy of 5% when compared to the total $1s$ QED contribution of about 270 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 [3], providing a fractional accuracy of $1.5 \times 10^{-3}$. Measurements of lithiumlike systems thus promise to be more sensitive to higher order QED terms than 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 [4], 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 four times larger than the experimental error limits [5].

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 [6]. 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 GSI heavy-ion accelerator and storage ring facility [7]. 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. Moreover,

it is more than two orders 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 [8]. The new benchmark allows us to infer the value for 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 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 lithiumlike $2s_{1/2}$ - $2p_{1/2}$ transition, and thus provides another benchmark for testing multi-electron QED calculations.

The present measurements were carried out using the SuperEBIT high-energy electron beam ion trap [9] 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 germanium detector focussing on the radiative recombination signal and a high-resolution microcalorimeter focussing on the $2s_{1/2}$-$2p_{3/2}$ x-ray transitions [8, 10]. 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 $\ell$/mm grating and a 1340×1300 pixel $LN_2$-cooled charge-coupled device (CCD) detector, as described in [11]. The resolving power of the instrument covering the wavelength range 35 to 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 $2s_{1/2}$ - $2p_{3/2}$ 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 [12, 13] and serves as a wavelength standard for our measurement.

We made sure that the carbon line was visible in each uranium spectrum by injecting a small amount of $CO_2$ into SuperEBIT. The $CO_2$ also served as a coolant for trapping the uranium ions as described in [8, 9]. 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 seventeen spectra similar to that in Fig. 1 showing the $U^{89+}$ line and twenty spectra showing the $U^{88+}$ line were separately analyzed.

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 [12–14] and readily establish the wavelength scale and dispersion based on a quadratic fit.

The variation of the wavelengths inferred from each of the seventeen measurements of the $U^{89+}$ $2s_{1/2}$-$2p_{1/2}$ transition is shown in Fig. 2. The uncertainty limits 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 $O^{6+}$ 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 seventeen spectra (or twenty in the case of berylliumlike 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 seventeen $U^{89+}$ 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 $44.1783 \pm 0.0024$ Å. This corresponds to $280.645 \pm 0.015$ eV, using the conversion factor $hc = 12398.42$ eVÅ[15]. Our value is in good agreement with the value of $280.59 \pm 0.10$ eV obtained with Doppler-tuned spectroscopy [6]. It is somewhat larger than the value of $280.516 \pm 0.099$ 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 [7].

Similarly, we determine a wavelength value for the $U^{88+}$ $2s^2$ $^1S_0$-$2s_{1/2}2p_{1/2}$ $^3P_1$ transition of $41.6335 \pm 0.0017$ Å, or $297.799 \pm 0.012$ 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. It is, however, slightly affected by blending with the forbidden transition of heliumlike $C^{4+}$. Because both lines are measured in the same order, energy discrimination cannot be used to diminish the effect of the blend.

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 $\alpha^2$ have recently been completed, including the two-photon exchange term as well as estimates of higher-order photon exchange contributions [4, 5, 16]. Adding these to the one-photon exchange, first order QED, nuclear recoil, nuclear polarization, and one-electron finite size contributions yields 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. [4], Sapirstein and Cheng [5], and Andreev et al. [16] is listed in Table II. The differences among the three values in Table II arise from differences in the calculated values of the two-electron self energy and the estimated size of the three-photon exchange term, as discussed by Sapirstein and Cheng [5]. Error limits are purely theoretical estimates and are dominated by the uncertainty in the estimate of the three-photon exhange contribution. No uncertainty estimate was given by Sapirstein and Cheng. Subtracting these values from our measured transition energy yields the two-loop Lamb shift. It ranges from 0.175 eV to 0.305 eV, depending on the theoretical value used, as given in Table II. The results show that, at a minimum, the two-loop Lamb shift is more than an order of magnitude larger than the uncertainty of our measurement.

No calculations exist to compare the inferred $2s_{1/2}$-$2p_{1/2}$ two-loop Lamb shift in lithium-like $U^{89+}$ with theory. However, the two-loop Lamb shift of the $1s$ level in hydrogenlike

$U^{91+}$ has recently been calculated to be $1.26 \pm 0.33$ eV [1, 17]. 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 comparing $U^{89+}$ with $U^{91+}$. First, we note that the one-loop Lamb shift of the $2s$ level is about 13% 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 determine the $U^{91+}$ $1s$ two-loop Lamb shift by multiplying by 6.33 the inferred two-loop Lamb shift from our measurement. The results are given in Table II. The error limits are the scaled uncertainties of the theroretical 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, 17] and those inferred from our measurement yields remarkably good agreement for two of the three values. The value inferred from the $2s_{1/2}$-$2p_{1/2}$ transition energy calculated by Yerokhin et al. [4] is in near-perfect agreement, while that inferred from the energy calculated by Andreev et al. is well within the uncertainty of the calculated $1s$ two-loop Lamb shift. The value inferred from the $2s_{1/2}$-$2p_{1/2}$ transition energy calculated by Sapirstein and Cheng is in less good agreement, which may reflect problems with their estimated three-photon exchange contribution. Ignoring the latter value, we obtain an average $U^{91+}$ $1s$ two-loop Lamb shift of 1.2 eV.

Calculations of the berylliumlike transition energies are by far less advanced than those for 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 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 [18] and that of 298.177 eV by Safronova et al. [19] reveals agreement within $-0.055$ eV 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 size of the $2s_{1/2}$-$2p_{1/2}$ two-loop Lamb shift in lithiumlike $U^{89+}$ to be about 0.20 eV. 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 1.2 eV, which is in excellent agreement with the calculated value of $1.26 \pm 0.33$ eV.

## Acknowlegments

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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 |

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 $280.645 \pm 0.015$ eV. All values are in eV.

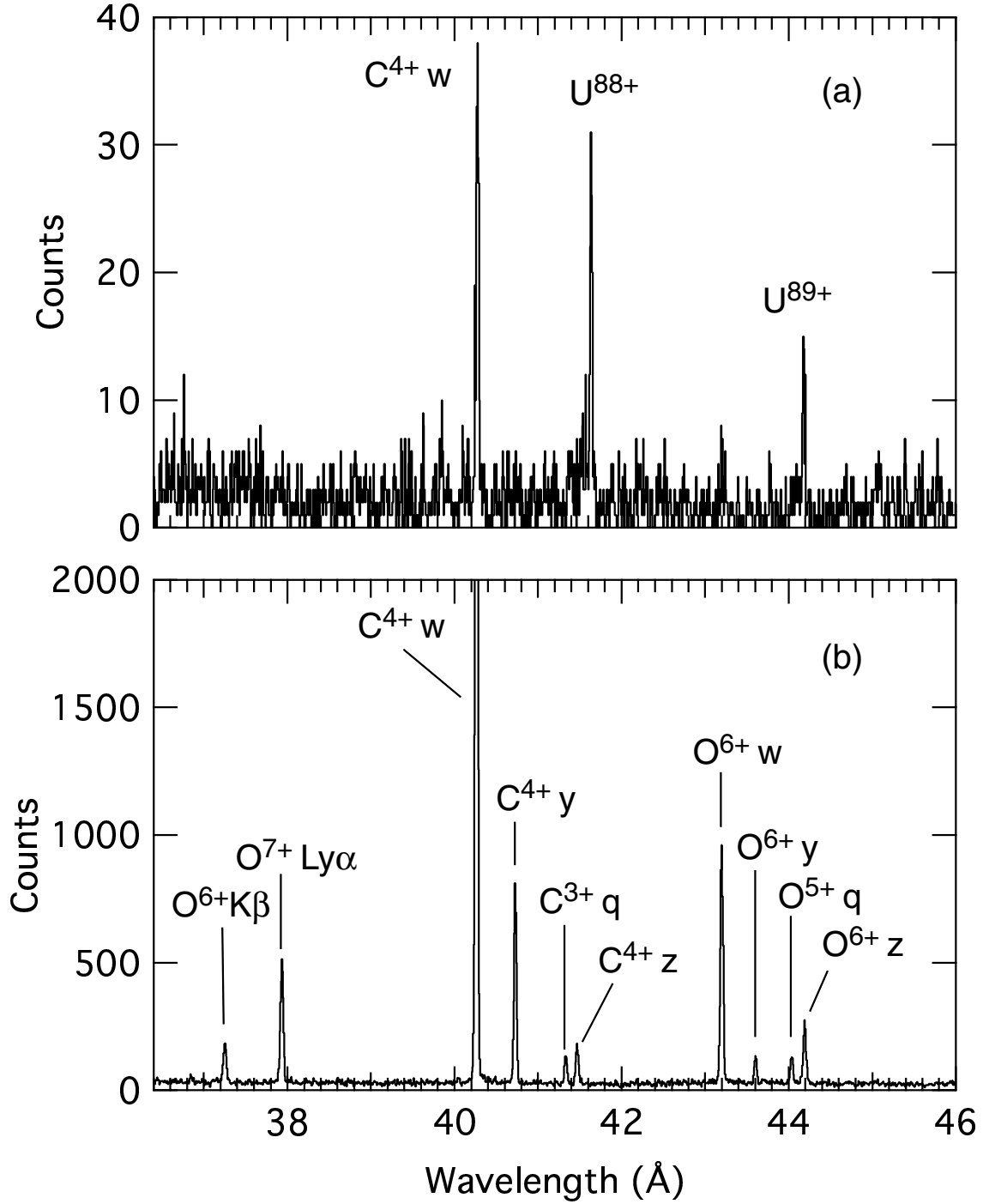| Transition energy | Two-loop Lamb shift | |
|---|---|---|
| | $2s_{1/2}$-$2p_{1/2}$ | $1s$ |
| | $(U^{89+})$ | $(U^{91+})$ |
| 280.44(10) [4] | 0.205 | 1.29(63) |
| 280.47(7) [16] | 0.175 | 1.12(44) |
| 280.34 [5] | 0.305 | 1.90 |

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.
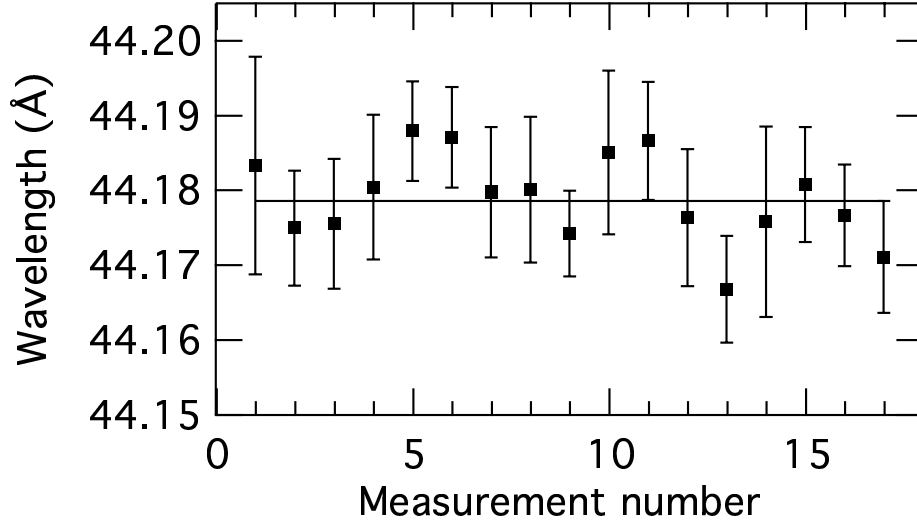
10

FIG. 2: Determinations of the $2s_{1/2}$-$2p_{1/2}$ wavelength in $U^{89+}$ from seventeen 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.
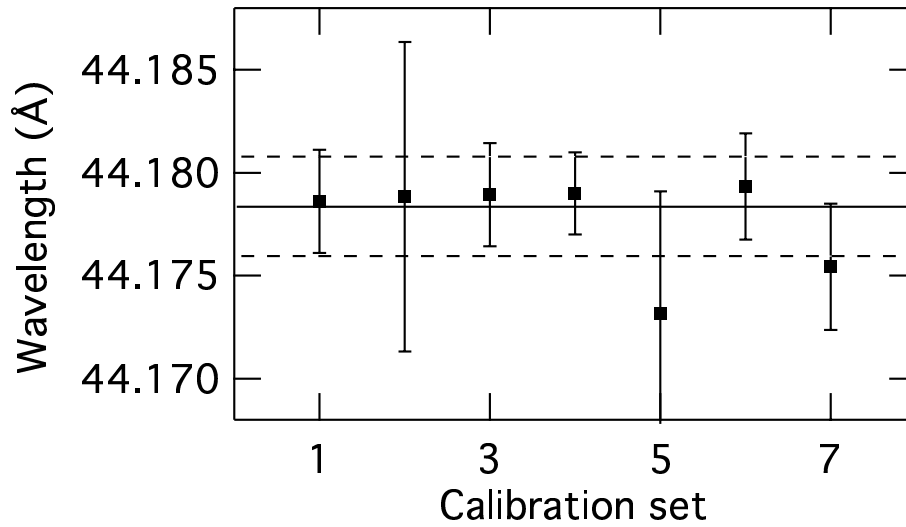


FIG. 3: Variation of the $U^{89+}$ $2s_{1/2}$-$2p_{1/2}$ wavelength determinations for seven spectral calibrations taken at different times interspersed among the uranium measurements. Error bars represent the statistical uncertainty of each individual calibration. The weighted average is indicated by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