Measurement of the $3s_{1/2}$-$3p_{3/2}$ resonance line in Na-like U^{81+}

P. Beiersdorfer,* E. Träbert, H. Chen, M.-H. Chen, M. J. May, and A. L. Osterheld
Department of Physics and Advanced Technologies, Lawrence Livermore National Laboratory, Livermore, California 94550-9234
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The wavelength of the $3s_{1/2}$-$3p_{3/2}$ transition of Na-like U^{81+} was determined to be $9.499\,85 \pm 0.000\,15$ Å (1305.12 ± 0.02 eV), using the EBIT-I electron beam ion trap. The measurement is many times more sensitive to the radiative contributions from quantum electrodynamics than earlier measurements for Pt^{67+} and Pb^{71+}. Our result strongly deviates from various predictions employing scaled hydrogenic quantum electrodynamic corrections and establishes a benchmark for multielectron QED calculations that agrees well with the trend established by *ab initio* calculations.

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I. INTRODUCTION

Tests of quantum electrodynamics (QED) increasingly have shifted from one-electron to multielectron systems, as the theoretical tools to perform many-body QED calculations are advancing [1,2]. Of special interest have been those multielectron ions with a closed shell and a single valence electron: Li-like, Na-like, and Cu-like ions. These systems at first glance mimic the one-electron ion. As a result, many of the results developed for hydrogenlike ions can be carried over to these systems by simple scaling techniques to provide approximate transition energies, while more sophisticated approaches or truly *ab initio* many-body QED calculations are being developed.

Experimental results have provided highly accurate data for comparison with calculations. For example, the $2s_{1/2}$-$2p_{3/2}$ transition of Li-like ions has been measured as high as U^{89+} [3–6]. The accuracy achieved in these measurements combined with recent theoretical developments was sufficient to isolate the contributions from second-order Feynman diagrams, notably the two-loop self-energy term [7].

Recently, we have reported data for the $4s_{1/2}$-$4p_{3/2}$ transitions of Cu-like ions up to uranium that greatly improved upon earlier measurements [8]. These measurements revealed notable discrepancies with theory that relied on scaled hydrogenic QED corrections for the two highest-Z ions studied, Th^{61+} and U^{63+}.

The most accurate datum for a $3s_{1/2}$-$3p_{3/2}$ transition in a high-Z ion was reported for Pt^{67+} [9]. A value of 5.32 ± 0.05 eV was inferred for the radiative contributions from this measurement. Two subsequent measurements focused on the $3s_{1/2}$-$3p_{3/2}$ transition in Na-like Pb^{71+} [10,11]. The best of these measurements, performed at the Unilac heavy-ion accelerator at GSI Darmstadt, achieved a four times worse accuracy than the platinum measurement.

In this study, we extend the measurements for Na-like ions to the heaviest naturally occurring element and report data for the $3s_{1/2}$-$3p_{3/2}$ transition in U^{81+}. Our measurement improves the experimental sensitivity to radiative effects by a factor of 5 over that of the earlier platinum measurement. This improvement is sufficient to establish discrepancies with theory that could not be resolved with the platinum measurement. As is the case for the Cu-like U^{63+} $4s_{1/2}$-$4p_{3/2}$ transition, the measured Na-like U^{81+} $3s_{1/2}$-$3p_{3/2}$ transition energy deviates markedly from the energy predicted by theory that relies on scaled hydrogenic QED corrections. Our result thus establishes a benchmark for the calculation of many-body QED in high-Z multielectron ions.

II. EXPERIMENT

The present measurements were carried out using the EBIT-I electron beam ion trap [12]. This device was put into service in 1986 at the University of California Lawrence Livermore National Laboratory. It had been converted to the SuperEBIT electron beam ion trap, but it was reconfigured to the original design after an internal move in 2001. Its utility for making accurate QED measurements was first demonstrated in 1990 [13].

We employed two vacuum crystal spectrometers with differing spectral resolving powers to perform the measurement of the $3s_{1/2}$-$3p_{3/2}$ transition in U^{81+}. A survey spectrum of the $3s_{1/2}$-$3p_{3/2}$ emission in various ions of uranium is shown in Fig. 1. The spectrum was obtained with the very same flat-crystal spectrometer that was used earlier to obtain a spectrum of Na-like Pb^{71+} [11] and has been described there. For the present measurements, the spectrometer used a cesium acid phtalate (CsAP) crystal with a $2d$ spacing of 25.68 Å. The measurements were carried out in second-order Bragg reflection at a Bragg angle of about 48°.

The survey spectrum is dominated by $3s_{1/2}$-$3p_{3/2}$ transitions in Mg-like U^{80+} and Si-like U^{78+}. Transitions from Al-like U^{79+} and P-like U^{77+} as well as a few lower-charge states are also seen. The emissions from different charge states were identified by varying the ionization balance and observing which features increase or decrease and in what proportion. Moreover, we were guided by calculations of the relative intensities for electron beam excited 3-3 emission lines performed with the Hebrew University Lawrence Livermore atomic code (HULLAC) set of computer codes [14]. These calculations were originally performed to identify the Na-like transition of Pt^{67+} among the 3-3 transitions mea-

*Electronic address: beiersdorfer@llnl.gov

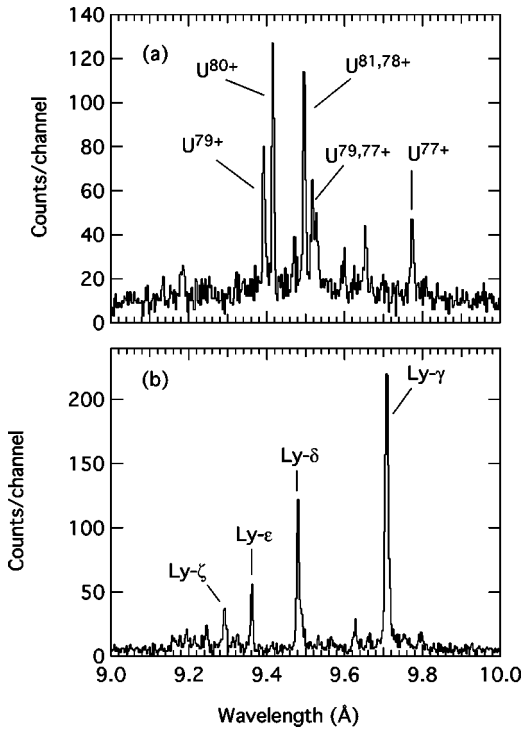

FIG. 1. Spectra obtained with a CsAP crystal in second-order Bragg reflection. (a) Spectrum of the $3p_{3/2} \rightarrow 3s_{1/2}$ transitions in highly charged uranium ions. The transitions are labeled by the emitting charge state. The spectrum was obtained at an electron beam energy of about 20 keV and a current of about 200 mA. (b) Spectrum of the $1s$-np Lyman series of Ne^{9+} used for calibration.

sured [9] on the EBIT-II electron beam ion trap at the University of California Lawrence Livermore National Laboratory. We also made use of the energy values of the various 3-3 transitions given by the multiconfiguration Dirac-Fock (MCDF) code of Grant *et al.* [15,16]. The MCDF energies differed by several eV from the measured values, but together with the intensity predictions from the HULLAC code they provided reasonable guidance for identification.

Identification of the $3s_{1/2}$-$3p_{3/2}$ transition of Na-like U^{81+} is not readily achieved. We found that the line is nearly coincident with the $3s^2_{1/2}3p^2_{1/2}\ ^1S_0 - 3s_{1/2}3p^2_{1/2}3p_{3/2}\ ^1P_1$ transition in Si-like U^{78+}. As a result, it cannot be resolved without the aid of line-fitting procedures. Fortunately, the Si-like uranium line emanates from an ion three charge states lower than the Na-like ion. Experimental variations of the ionization balance, therefore, can unequivocally determine which line in the blend results from which charge state.

Calibration of the uranium spectrum was accomplished by recording the $1s$-np $(n \geqslant 4)$ transitions in hydrogenlike Ne^{9+}. A spectrum of the Ne^{9+} Lyman series is shown in Fig. 1(b). The wavelengths of these lines are well known [17,18], and they may readily serve as calibration references. Calibration spectra were recorded interspersed in time with measurements of the uranium lines.

Two detector and crystal settings were employed to check for systematic deviations. Spectra from each setting were added and analyzed separately. The results are given in Table I. The $3s_{1/2}$-$3p_{3/2}$ transition of Na-like U^{81+} could be measured within an accuracy of 0.2 eV, where much of the uncertainty is due to the difficulty of resolving the Na-like line from the Si-like line. This, unfortunately, is not much better than the accuracy of our earlier proof-of-principle measurement of the $3s_{1/2}$-$3p_{3/2}$ transition of Na-like Pb^{71+} [11], which was measured with an uncertainty of 0.26 eV.

TABLE I. Measured $3s$-$3p$ transitions energies (in eV) from different experiments. The quadrature sum average is given in the last row. The crystals used in the measurements are cesium hydrogen phthalate (CsAP) and ammonium dihydrogen phosphate (ADP).

Experiment	Result
CsAP	1305.18 ± 0.21
CsAP	1304.95 ± 0.34
ADP	1305.11 ± 0.04
ADP	1305.12 ± 0.03
Average	1305.12 ± 0.02

To determine the $3s_{1/2}$-$3p_{3/2}$ transition of Na-like U^{81+} with higher accuracy, we employed the higher-resolution vacuum crystal spectrometer described in [19]. Unlike the survey spectrometer, this spectrometer is able to record spectra at Bragg angles exceeding 50°. Moreover, the path between EBIT-I, crystal, and detector is nearly twice as long.

For the present measurements, the high-resolution spectrometer utilized an ammonium dihydrogen phosphate (ADP) crystal with lattice spacing $2d = 10.64$ Å. It measured the lines of interest in first-order Bragg reflection at a Bragg angle of about 63°. The observed resolving power $\lambda/\Delta\lambda$ was about 2600. The linewidth $\Delta\lambda$ of about 3.6 mÅ represents a convolution of the resolving power of the crystal, ion temperature, and detector resolution. It is about four times better than the resolution achieved with the survey instrument.

A spectrum of the $3s_{1/2}$-$3p_{3/2}$ transition of Na-like U^{81+} observed with the high-resolution vacuum spectrometer is presented in Fig. 2(a). The line is now resolved from the neighboring Si-like U^{78+} transition. In the (near) absence of blending, the line's centroid could now be determined within one part in 12–17 of the linewidth, where the low counting statistics was the main limitation.

As in the lower-resolution measurement, the Lyman lines of Ne^{9+} were used for calibration, as shown in Fig. 2(b). Because of the trade-off between higher resolution and broader spectral coverage, only the $5p \rightarrow 1s$ Lyman-δ and the $6p \rightarrow 1s$ Lyman-ϵ transitions were available for calibration. As illustrated in Fig. 2, the Na-like uranium line is close to the Ne^{9+} Lyman-δ line; the difference is about 19 mÅ (2.6 eV). Therefore, the accuracy of the present measurement depends strongly on the quality of the reference wavelength assigned to the Lyman-δ line. The two components $1s_{1/2}$-$5p_{1/2,3/2}$ of the Lyman-δ line could not be resolved. Their energies differ by only 0.03 eV. Using the 2:1 statistical weight for the Lyman-δ_1 and Lyman-δ_2 component, the average energy is 1307.748 eV [17]. Weights of 3:1 or 1:1 change this value by no more than 0.005 eV. The systematic error associated with the choice of the reference line is thus negligible.

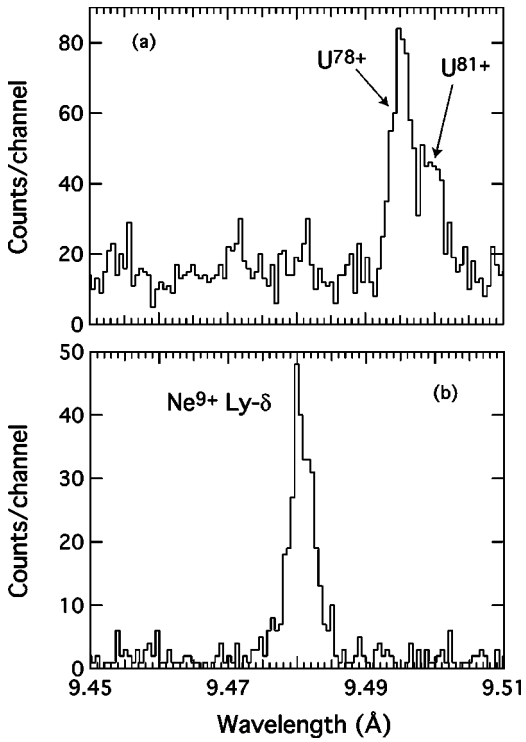

FIG. 2. Spectra obtained with an ADP crystal in first-order Bragg reflection. (a) Close-up of the $3p_{3/2} \rightarrow 3s_{1/2}$ emission from Na-like U^{81+} and Si-like U^{78+}. The spectrum was obtained at an electron beam energy of about 24 keV and current of about 160 mA. (b) Close-up of the Lyman-δ line of H-like Ne^{9+}. The spectrum was recorded at a beam energy of 2.5 keV and a current of 120 mA.

As for the survey measurements, two detector and crystal settings were employed to check for systematic deviations, and the spectra from each setting were added and analyzed separately. The results are given in Table I. With the high-resolution spectrometer, the $3s_{1/2}$-$3p_{3/2}$ transition of Na-like U^{81+} could be measured within a 1σ accuracy of 0.03 eV.

III. DISCUSSION

A weighted average of all four measurements results in the final value of 1305.12 ± 0.02 eV for the Na-like uranium $3s_{1/2}$-$3p_{3/2}$ transition energy. This precision corresponds to a 0.2% measurement of the 9.93 eV radiative contribution estimated by Kim *et al.* [20] using the Welton method. This represents a five times higher sensitivity to these contributions than the best previous measurement of Pt^{67+}. It is more than an order of magnitude higher than reached in the measurements of the highest-Z Na-like ion measured to date, Pb^{71+}.

In Fig. 3 we compare the best available measurements of the Na-like $3s_{1/2}$-$3p_{3/2}$ transition energies for each element with the calculations presented in [20]. Up to $Z=42$, measurements had been performed using the TEXT tokamak [21]. Measurements of the mid-Z elements up to $Z=64$ were performed using various laser facilities including the Omega and Nova lasers [21–23]. The calculations by Kim *et al.*

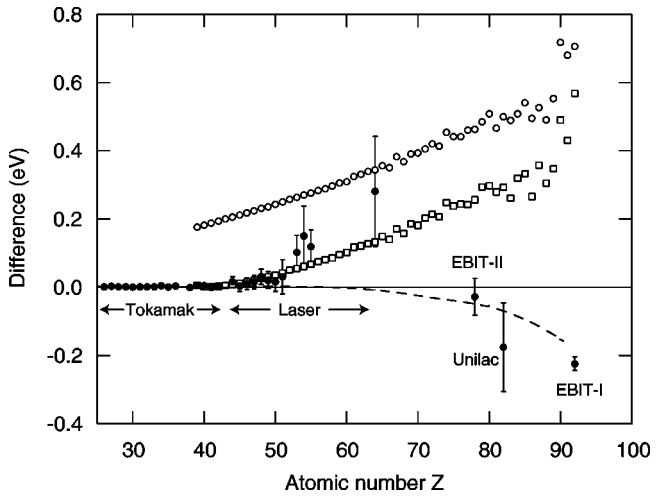

FIG. 3. Comparison of experimental and theoretical $3s_{1/2}$-$3p_{3/2}$ transition energies of sodiumlike ions. Plotted are the differences from the calculations of Kim *et al.* [20] (solid line). Open circles represent calculations of Seely *et al.* [23] using a MCDF code; open squares represent semiempirical predictions of Seely *et al.* [23]. The dashed line represents the differences from the calculations of Blundell [25]. Solid circles represent the best experimental results for each element. Tokamak data are from [21]; laser data are from [21–23]; the EBIT-II electron beam ion trap datum is from [9]; the beam-foil datum from the Unilac accelerator is from [10]. The EBIT-I electron beam ion trap measurement of U^{81+} represents the present result.

agree with the trend set by lower-Z tokamak measurements and the EBIT-II measurement of Pt^{67+}, but they differ from the laser data.

Kim *et al.* calculated QED effects by the Welton method and combined them with non-QED calculations that were partly based on MCDF and partly on many-body perturbation theory calculations. As seen in Fig. 3, these calculations, however, disagree markedly with our result for $Z=92$. The disagreement between our uranium measurement and the predictions by Kim *et al.* is similar to that found for thorium and uranium in the Cu I isoelectronic sequence [8]. Because Kim *et al.* utilized scaled, screened hydrogenic QED corrections for their predictions, the disagreement suggests a breakdown of the accuracy of such corrections at the highest Z.

Seely *et al.* [23] used the Grant code [15,16] for their MCDF calculations. In its default mode, this code employs a method for obtaining the QED corrections that Kim *et al.* identified as possibly insufficient at high Z. Indeed, Seely *et al.* added a semiempirical correction to the Grant code results that was intended to yield better predictions. This yielded better agreement with the tokamak and laser data, reproducing these data up to $Z=64$, i.e., up to the highest-Z datum available at the time. It is evident from Fig. 3 that this attempt to improve the predictions does not agree with the electron beam ion trap or Unilac data.

Blundell undertook an *ab initio* calculation of the screened self-energy and vacuum polarization effects as an extension of the relativistic many-body perturbation theory calculations by Johnson *et al.* [24], and combined the results with earlier results on correlation and nuclear effects. This

calculation matches not only the experimental (tokamak) data at lower Z but also the EBIT-II and Unilac data at high Z. Unfortunately, Blundell made no prediction for U^{81+} so that a direct comparison with our measurement cannot be made.

In summary, we have presented a measurement of the $3s_{1/2}$-$3p_{3/2}$ transition energy in sodiumlike uranium. It is the most significant one in the high-Z range so far. The observed discrepancies with theory strongly suggest a breakdown of scaled hydrogenic QED and indicates the need for *ab initio* many-body QED calculations.

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