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Wavelengths of the $4s_{1/2}$ - $4p_{3/2}$ resonance lines in Cu- and Zn-like heavy ions

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Using an electron beam ion trap and a high-resolution flat-field spectrometer, the EUV resonance lines $4s_{1/2}$ - $4p_{3/2}$ of the Cu- and Zn-like ions of Os, Bi, Th, and U (Z = 76 - 92) have been observed and their wavelengths measured. Our experiments remove systematic errors from line blends encountered in earlier work. Our results on Cu-like ions are in good agreement with recent *ab initio* calculations for all ions that include QED. Our results for Zn-like ions corroborate and extend our earlier findings, but consistently good theoretical values for comparison are lacking.

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I. INTRODUCTION

The study of ions along the Cu isoelectronic sequence provides important benchmarks through both precision experiments and modern atomic structure theory. Production and charge-state isolation of Cu-like ions are more easily achieved than for ions of many other charge states. Precision wavelength determinations by both experiment and calculation can yield information on atomic structure under high-field conditions, when fully relativistic calculations take into account radiative corrections and nuclear structure effects.

Plasmas produced by the then most powerful laser systems, such as the NOVA and OMEGA lasers, have reached Cu-like ions of the heaviest naturally occurring elements years ago. However, the laser-produced plasma data in the high-nuclear charge (high-Z) regime [1] show a systematic deviation from calculation (for example, by Kim *et al* [2]) by several hundred ppm for the heaviest elements. Lower-Z ions measured in lower-density tokamaks do not exhibit any noticeable difference with theory. In a recent paper [3], we have presented data from observations of the same $4s_{1/2}$ - $4p_{3/2}$ lines in the low-density environment of the EBIT-II electron beam ion trap. Our measurements continued the good agreement with theory set by the tokamak measurements and disagreed with the high-Z trend set by the laser-produced plasma measurements. Deviations between our data and theory were only found at the two highest-Z ions. Here, our measured wavelengths were slightly longer than predicted by the calculations by Kim *et al* [2]. The data point for Th (Z=90) agreed with the trend of the *ab initio* calculation by Blundell [4], but the data point for U (Z=92) did not. Since then, theory has been checked by new high-precision calculations of relativistic correlation energies and QED corrections [5]. The latest calculations confirm the trend of Blundell's calculations, and they agree with our data except for Z=92. Using a new spectrometer that affords a sixfold increase in spectral resolution over our previous instrument, we have revisited Th (Z=90) and U (Z=92), and we have added observations on Os (Z=76) and Bi (Z=83). The new observations corroborate the latest theoretical results all the way.

The $4s_{1/2}$ - $4p_{3/2}$ resonance line ($4s^2\ {}^1S_0$ - $4s4p\ {}^1P^o_1$ in LS coupling notation) of the Zn-like ions appears in the same spectra. Experiment should be able to provide equally precise wavelength data for Zn-like ions as for the Cu isoelectronic sequence. Theory, however, faces the problem of treating an additional electron in the valence shell. As demonstrated elsewhere [6], various calculations have been tried to this effect, but none has reached satisfactory results. In our previous study, the lines of interest for Zn-like ions were partly blended with lines from other charge states. With the higher resolution of our new spectrometer, this problem has been overcome, and the associated error bars have been reduced markedly. We provide new data on the aforementioned four high-Z elements in order to provide benchmarks that theory will need to match in the quest for a satisfactory description of electron-electron interaction in few-electron systems.

II. EXPERIMENT

The experiment was done at the University of California Lawrence Livermore National Laboratory electron beam ion trap facility. Of the laboratory's two electron beam ion traps, the higher-energy device, SuperEBIT [7], as well as its low-energy configuration, EBIT-I, were employed. Much of the experimental procedure has been explained before [3, 6] and does not need to be repeated here. The new measurements covered the elements Os (Z=76), Bi (Z=83), Th (Z=90), and U (Z=92). Ions of Bi, Th, and U, respectively, were introduced into the electron beam ion trap by means of a Metal Vapor Vacuum Arc ion source (MeVVA); for Os, a volatile compound, osmium tetroxide, was used by ways of the low-pressure, ballistic gas injector, instead. Upon breaking the molecule by collisions with the fast electrons of the beam, the light ion radicals evaporate from the trap volume and provide evaporative cooling to the Os ion cloud. For easier trapping of the other elements, nitrogen (in

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some cases carbon dioxide) was bled into the vacuum vessel via the ballistic gas injector. Ions were trapped by the combination of a strong (3 T) magnetic field for radial confinement, electric fields in a drift tube arrangement for axial confinement, and the attractive potential offered by the intense electron beam.

Bombarded by the electron beam, the ions are being ionized in a stepwise fashion. Ionization ends when the charge state reached has a higher ionization energy than is available as kinetic energy in the electron beam. The electron beam energy necessary to create Cu-like ions is of the order 2.0 − 4.5 keV for the elements considered here [8], which is but a small fraction of the working range of SuperEBIT. The ionization energy of the next higher charge state, Ni-like ions, is relatively high (and the prominent lines lie in other ranges of the spectrum). Consequently, a wide range of electron beam energies is available to create a charge state balance that is dominated by Cu- and Ni-like ions, by burning out the lower charge states, and thus creating 'clean' EUV spectra with mostly lines from Cu-like and Zn-like ions remaining. On the other hand, electron beam energies below the production threshold of these two ion species were employed to learn about possible contamination of the spectra by lines from ions in lower charge states.

The present measurements employed a new flat-field spectrometer system (FFS) that represents an improvement in spectral resolution by a factor of six over our previous instrument [9]. The spectrometer is equipped with a 2400 ℓ/mm variable line spaced concave grating [10] of R=44.3 m radius of curvature and with a cryogenically cooled back-thinned charge-coupled device (CCD) camera. The older CCD camera contains a chip that has 1024 × 1024 pixels on a square area of about 25 mm edge length. The newer camera used has about 1340 × 1300 pixels on the same area. The grating imaged the light from the ion trap, using the 60 μm diameter electron beam [11] as the source, onto the CCD chip where it resulted in the geometrically expected width of about 3 to 4 pixels (older CCD) to 4 to 5 pixels (newer CCD), respectively.

With the older CCD camera, the total area of the CCD chip was binned by a factor of four in the non-dispersive direction to create an effective CCD array of 256 × 1024 pixels. Due to spectral aberrations, the image of each line is slightly curved at the CCD surface. Simple summing across the dispersion direction would, therefore, result in spectral features broadened to about five channels. As the signal level was quite high, we elected to evaluate only the center part of the spectra where the line curvature is less of a problem. Spectra were recorded of Os, Th, and U, with typical exposure times of 20 minutes per spectrum. Calibrations were done before and after some 4 to 6 hours of data taking.

With the second (newer) CCD, the spectral image area was subdivided into three parallel strips along the direction of dispersion. The spectra of each strip were separately calibrated and evaluated, resulting in 48 individual measurements for Bi, 24 for Th, and 30 for U. The 30-minute exposures were interspersed with calibration spectra every hour or two. Moreover, in several spectra of Bi and Th, the calibration was effected in the very spectra, by increasing the amount of cooling gas until the lines of the light elements appeared along the ones of the heavy element.

Calibration was performed by reference to a number of well known transitions in H- and He-like ions of C, N, and O (mostly from separate exposures, using CO_2 or N_2 gas injection and short timing cycles), then fitting a 2^{nd}-order polynomial to the calibration data. For reference line wavelengths, we used the calculations by Garcia and Mack for H-like ions [12] and those by Drake for n=2 levels of He-like ions [13]. For both sets of calculations, the accuracy is assumed to be better than 1 mÅ. For some singlet lines in He-like ions of N and O, Engström and Litzén [14] provide even more accurate reference wavelengths, and they correct significant errors in the Kelly tables [15].

The lines of interest in Bi and Th lie very close to the strong lines that result from $1s^2$ - 1s2p transitions in He-like carbon and nitrogen, respectively, which renders the best situation for the calibration effort. Examples of the heavy-ion spectra and light-ion calibration spectra are shown in Fig. 1. The lines of H-like and He-like N were used in [3] for the calibration of the U lines. However, these are relatively far apart and the U lines are situated well in between. Therefore, these lines provide a reference wavelength scale that is less accurate than in the case of the Th lines, which are almost coincident with the K-shell lines of He-like N (see Fig. 1). Because of this situation, we employed an additional set of reference lines given by the 1s - np Lyman series of H-like C. The series limit of 1s - np transitions in H-like ions of carbon is near the U lines of present interest. These calibration lines are well known, but weak. By their multitude they nevertheless provide a reliable reference grid. For Os, second diffraction order lines of nitrogen were used that were not so close in position and not so tightly spaced as in the other cases, thus affording a somewhat lower calibration precision.

Both CCD chips have different background distributions, but none had any prominent features that would perturb the determination of line positions unless corrected for. Therefore a flat background (representing most of the read-out noise) was subtracted from the raw data before evaluation, and the rest was approximated by a low-order polynomial function. The peaks were fit with Gaussian functions.

The line width of such a grating spectrometer as that employed here is largely source limited and thus practically constant over the working range. The resolving power $\lambda/\Delta\lambda$ consequently is lowest at the shortest wavelengths covered. We therefore demonstrate our resolution with data for the most affected case, uranium, in Fig. 2. Clearly the profile of the line labeled "Zn" has some overlap with a weaker line of other origin, but the wavelength difference is large enough to permit a reliable multi-peak fit analysis. The line structure is basically similar in all ions studied here. In [3], the line group of Fig. 2 was much less well resolved, which limited the attainable accuracy in spite of excellent data statistics.

While the blending problem has been largely overcome with the new instrument, the remaining principal sources of error are signal statistics and measurement reproducibility. By straightforward statistical analysis, the positions of the lines of interest can be determined to a very small small fraction of the line width (one percent and less). However, this precision is not decisive, since the spectrometer is not stable enough (vibrations, thermal fluctuations, etc.) to reproduce line positions to anywhere near this precision. The scatter of the line positions from repeat measurements was such that it permitted to determine a mean wavelength to better than $\pm 0.5\,$mÅ. This number, however, is inappropriate as a meaningful error estimate, since the data distribution was dominated by systematic variations (drifts), not by pure (random) statistics. In order to represent the systematic error evident in the reproducibility of the individual line positions, we instead quote an error margin that encompasses two thirds of all data points, and this amounts to 1 to 2 mÅ.

III. RESULTS AND DISCUSSION

A summary of the results from each of the measurements of Cu-like ions is found in Table I. Results on Zn-like ions are listed in Table II. In both cases we also list some selected theory values, leaving out most of those shown previously to be less satisfactory. We also list our earlier results, as well as representative data from other experiments [1, 16–19]. Compared to our previous work, the much better spectral resolution practically removes line blends as a significant source of systematic error. This improvement results in some slight shifts and some notably reduced error estimates. Almost all of our previous results are corroborated within their error estimates.

The exception is the Cu-like U^{63+} result. We measure 26.4233±0.0015 Å. The earlier result obtained on the EBIT-II electron beam ion trap was 26.4325±0.0019 Å [3]. The difference may be due to a calibration difference. The present measurement uses the Lyman series of H-like C as reference lines in addition to the H-like and He-like N lines used as references in [3]. The present result is now also closer to the result of 26.400±0.015 Å given by Seely et al. [1] and almost overlaps with their uncertainty limits.

Figures 3 and 4 put the results into perspective by displaying the isoelectronic trends. The new data on Os and Bi fall into the same smooth isoelectronic trend as our previous data on Yb, W and Th, and the new data on Th and U. The new result, for Cu-like U, that differs from earlier results by more than the estimated error, is no longer at variance with the trend of the most advanced calculations [4, 5]. Overall, our previous and present data from the electron beam ion trap corroborate fully the quality of calculations of Cu-like ions. The *ab initio* calculations by Blundell [4], that suggested a deviation with opposite sign than the MCDF calculations that were semi-emprically corrected by reference to laser-produced plasma measurements, are fully corroborated by the present data. The experimental accuracy of the new data is of the order of 40 ppm.

Concerning Zn-like ions, our data provide an equally accurate data set as for the Cu-like ions. So far, theory has not been able to match this. The disagreement between theoretical predictions and experimental findings is considerable, as is illustrated in Fig. 4. The figure includes only the most accurate theoretical results (for a full set, see [6]); we note that the offset from *ab initio* calculational results is about 0.5%, significantly more than is seen in the Cu isoelectronic sequence. Clearly, electron-electron interactions in ions with more than one electron in the valence shell deserve more theoretical attention, now that experimental benchmark data are available. Until such calculations are made, we note that the semi-empirical predictions [19, 21] also are in need of revision. The better of the two will need to be shifted by about 300 ppm in the range $70 \leq Z \leq 85$ and by more than 1000 ppm for uranium.

On a final note, we point out that the present results bode well for future measurements of the $3s_{1/2}$ - $3p_{1/2}$ and $2s_{1/2}$ - $2p_{1/2}$ valence transitions in very high-Z Na-like and Li-like ions, respectively, which also fall into the wavelength region spanned by the present measurements. Recent measurements of xenon with a lower-resolution instrument have shown the feasibility of such experiments on SuperEBIT [20].

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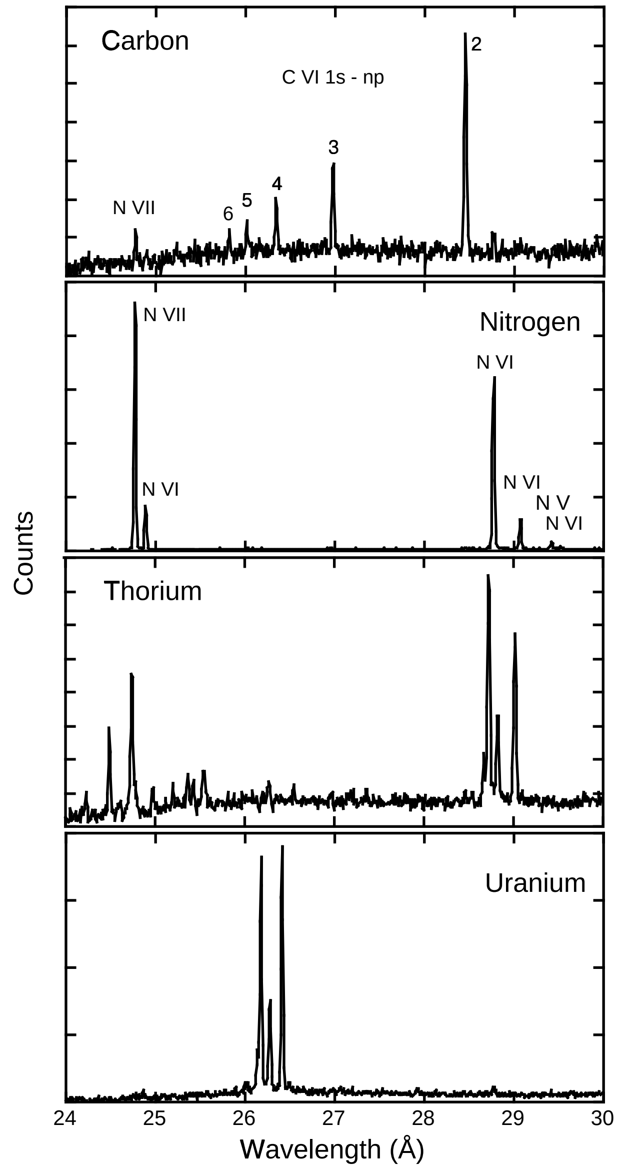

FIG. 1: Full spectra with nitrogen, carbon dioxide, thorium, or uranium injection into the electron beam ion trap. Both heavy-element spectra are calibrated by observations of lines from H- and He-like light ions.

TABLE I: Predicted and measured wavelengths (in Å) of the 4s $^2S_{1/2}$ - 4p $^2P^o_{3/2}$ transition in Cu-like ions of the 8 highest-Z ions covered in these experiments. Of the earlier experimental results, only a representative one of the laser-produced plasma studies is quoted.

Element	Z	Semiemp. [1]	Theory [2]	Theory [4]	Experiment [1]	Experiment [3]	Experiment This work
Yb	70	75.839	75.860	75.864	75.842(15)	75.8595(47)	—
W	74	62.311	62.334	62.341	62.304(15)	62.3355(45)	—
Os	76	56.534	56.558	—	—	—	56.5630(20)
Au	79	48.907	48.931	—	48.928(15)	48.9280(26)	—
Pb	82	42.358	42.377	42.381	42.349(15)	42.3740(58)	—
Bi	83	40.383	40.404	40.407	40.394(15)	—	40.4066(20)
Th	90	28.998	29.018	29.022	28.990(15)	29.0224(30)	29.0227(10)
U	92	26.401	26.420	26.423	26.400(15)	26.4325(19)	26.4233(15)

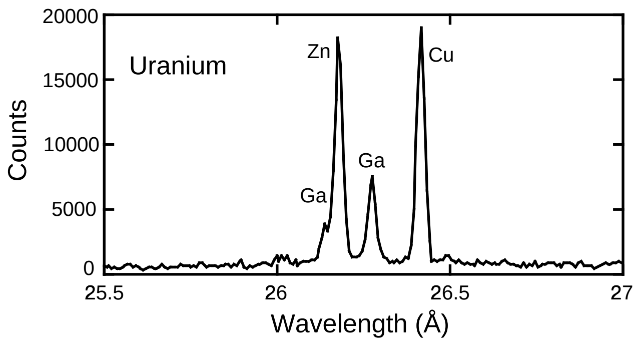

FIG. 2: Detail of an EUV spectrum obtained with uranium injection (see Fig. 1). The spectral lines originate from Cu-like, Zn-like, and Ga-like ions of uranium.

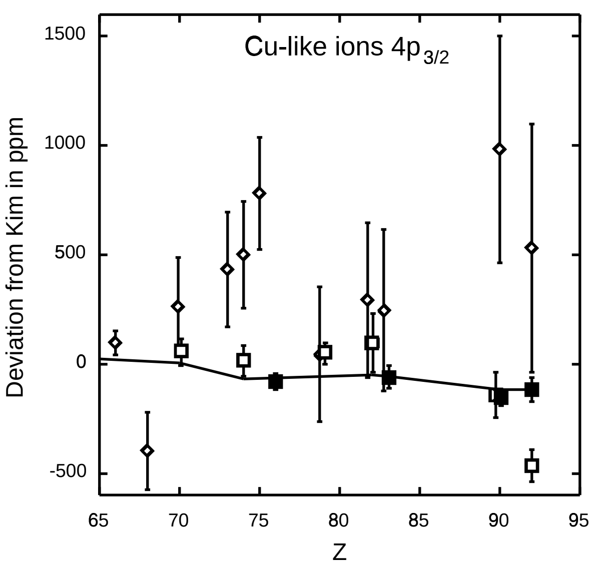

FIG. 3: Deviation of predictions and data for the 4p $^2P^o_{3/2}$ level energy in Cu-like ions from the values predicted by Kim *et al.* [2]. Full line [4], diamond data from from other sources than the electron beam ion trap, open squares [3], solid squares this work.

TABLE II: Predicted and measured wavelengths of the $4s^2\ ^1S_0$ - $4s4p\ ^1P^o_1$ transition in Zn-like ions. All wavelength values are given in Å.

Element	Z	Theory	Experiment [f] i	Experiment [g]	Experiment [*]
Yb	70	73.430 [a]	73.792(20)	73.8070(66)	—
		73.8 [b]			
		73.368 [c]			
		73.784 [d]			
W	74	60.629 [a]	60.900(20)	60.9300(54)	—
		61.0 [b]			
		60.585 [c]			
		60.907 [d]			
		60.806 [e]			
Os	76	55.4 [b]	—	—	55.3840(50)
		55.084 [c]			
		55.373 [d]			
Au	79	48.0 [b]	48.063(20)	48.0583(49)	—
		47.787 [c]			
		48.038 [d]			
		47.7 [e]			
		47.991 [e]			
Pb	82	41.7 [b]	41.689(20)	41.7185(45)	—
		41.483 [c]			
		41.708 [d]			
		41.681 [e]			
Bi	83	39.8 [b]	39.792(20)	—	39.8151(20)
		39.578 [c]			
		39.796 [d]			
Th	90	28.6 [b]	28.702(20)	28.7227(67)	28.7303(11)
		28.52 [c]			
		28.704 [d]			
		28.707 [e]			
U	92	26.1 [b]	26.157(20)	26.1868(36)	26.1861(10)
		25.975 [c]			
		26.152 [d]			
		26.168 [e]			

a Multi-Configuration Dirac-Fock (MCDF) [22]
b Semiempirical analysis of experimental data [23]
c HULLAC [19]
d HULLAC plus semiempirical correction [19]
e Multi-Configuration Dirac-Fock (MCDF), with QED, including nuclear size effects [21]
f [19]
g [6]
* This work

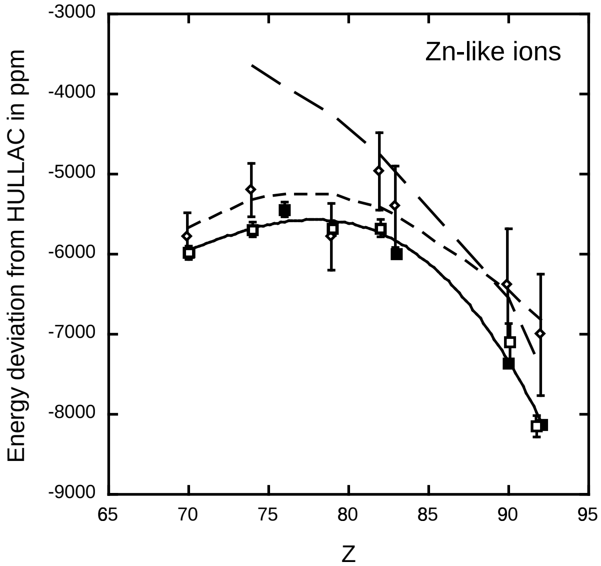

FIG. 4: Deviation of predictions and experimental data for the $4s^2$ $^1S_0$ - 4s4p $^1P_1^o$ transition wavelength in Zn-like ions from the values predicted by Brown *et al.* [19] using the HULLAC [24] code. Theory: long dashed line HULLAC plus semiempirical correction [19], short dashed line [21]; experiment: open circles [19], open squares [6], solid squares this work. On this scale, the error bars for some of the new data are smaller than the symbol size. The solid line provides a (3^{rd} order polynomial fit) guide to the eye for the electron beam ion trap data.