

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

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(Received 11 September 2002; published 10 March 2003)

Using the EBIT-II electron-beam-ion trap and a flat-field spectrometer, the extreme UV resonance lines  $4s_{1/2}$ - $4p_{3/2}$  of the Cu-like ions of Yb, W, Pb, Th, and U have been observed and their wavelengths measured. Our results differ substantially from earlier measurements performed with plasmas produced by the OMEGA and NOVA lasers and have higher accuracy. Our results are in good agreement with theory in all ions but those of the highest charge states.

DOI: 10.1103/PhysRevA.67.032502

PACS number(s): 32.30.Jc, 34.50.Fa, 34.80.Dp, 31.30.Jv

### I. INTRODUCTION

The study of ions along the Cu isoelectronic sequence provides important benchmarks through both precision experiments and modern atomic structure theory. From the experimental standpoint, the production and charge-state isolation of Cu-like ions is easier than for ions of many other charge states. Plasmas produced by the most powerful laser systems, such as the NOVA and OMEGA lasers, can reach Cu-like ions of the heaviest naturally occurring elements. Precision wavelength determinations by both experiment and calculation can then yield information on atomic structure under high-field conditions, where fully relativistic calculations have to take into account radiative corrections and nuclear structure effects. 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.

We undertook to observe the same  $4s_{1/2}$ - $4p_{3/2}$  lines in the low-density environment of the electron-beam-ion trap EBIT-II. With an accuracy increased from threefold to eightfold, our measurements suggest that the high- $Z$  trend set by the laser-produced plasma measurements was spurious. Our measurements continue the good agreement with theory. Deviations are only found at the highest- $Z$  ions. Here, our measured wavelengths are longer than predicted, in contrast to the trend suggested by the laser-produced plasma data.

### II. OVERVIEW OF THEORY VS EXPERIMENT

Cu-like spectra feature prominent lines associated with a single, easily excited valence electron outside a closed  $3d^{10}$  core. Calculationally, the situation is much more complex than this: for the  $4s$ - $4p$  resonance transition, the angular momentum  $l$  of the valence electron is smaller than that of all electrons in the  $3d$  shell, and hence the electron—in a clas-

sical picture—penetrates the electron core. Starting out from the closed-shell model, experimental systematization efforts and theory have tried to approximate the physical situation by introducing the concept of core polarization. However, in the high- $Z$  regime, relativistic effects play a dominant role [3] over nonrelativistic contributions to atomic structure, and the semiclassical descriptions that work well for lighter ions are no longer appropriate. Theory faces the task of describing a system that consists of a nucleus of finite size and of 29 electrons. Such a many-body system cannot be calculated exactly. Nevertheless, quite a number of calculations have been presented in the last 20 years to predict reliably the lowest-energy levels and thus the transition energies of Cu-like ions.

The first measurements of the wavelengths of Cu-like transitions in heavy elements (Au, Pb, Bi, Th, and U) have been reported by Seely *et al.* using the OMEGA laser [4]. The authors compared the measured wavelengths to theoretical values calculated with a multiconfiguration Dirac-Fock (MCDF) package (the GRANT code [5,6]), using the extended average level method, with transverse Breit and QED corrections. The significant differences between the measured and calculated values were attributed to “a departure from the scaled hydrogenic QED contribution to the  $4s$  energy,” that is, to a deficiency in the theoretical treatment of the QED. Cheng and Wagner [7] tried to understand the causes of the difference between theory and experiment by using another variant of the MCDF package, the optimal level (OL) approximation, thus accounting for core relaxation at each energy level, and also including finite-nuclear-size effects. With results that agreed better with experiment than the previous calculations, they concluded that the residual discrepancy noted by Seely *et al.* [4] was probably due to the omission of the finite-nuclear-size effects which are important for low- $n$  and  $-l$  states, and that their improved treatment of QED corrections was good enough to give good estimates of its contributions.

In 1989, Seely *et al.* [1,8,9] reported the observation of  $4s_{1/2}$ - $4p_{3/2}$  transitions in Cu-like ions from mid- $Z$  elements (including Sn, Xe, La, Nd, Eu, Gd, Dy, and Yb) made on the Princeton Large Torus (PLT) tokamak. They attributed the remaining discrepancies to electron correlation corrections

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that were not accounted for in the calculations by Cheng and Wagner. So in the course of two years time, there flared some debate over whether there was good agreement between theory and experiment, and about the role of QED, finite nuclear size, and correlation corrections in these calculations.

Between 1988 and 1990, Johnson *et al.* published a series of three papers on the topic of relativistic many-body perturbation theory (MBPT) studies of energy levels of ions with a single valence electron outside of a closed shell [10–12]. The third of these reported calculations of energies of  $4s_{1/2}$  and  $4p_{3/2}$  states of Cu-like ions with nuclear charges in the range  $Z=29-92$  [12]. These calculations were based on Dirac-Fock wave functions and included Coulomb correlation corrections (second and third order), retarded Breit interaction, correlation corrections to the Breit interaction (second and third order), finite-nuclear-size corrections, and reduced mass and mass polarization corrections. They did not include QED corrections such as the electron self-energy and vacuum polarization, but have otherwise been considered to be the most complete theoretical treatment to date.

Two calculations have sought to provide the QED corrections that are necessary for a comparison between experiment and these theoretical results. In the first, Kim *et al.* [2] calculated the self-energy corrections via three independent approximations—the  $\langle r \rangle$  method, the  $\rho$  method, and the Welton method (see Ref. [2] for a description of these methods). Whereas the results from the  $\rho$  and the Welton methods were in close agreement with each other, the  $\langle r \rangle$  method (the default choice in the GRANT code used by Seely *et al.* [4]) was found to lead to transition energies with poor agreement to experiment, especially at high  $Z$ . Moreover, the authors subtracted their calculated MCDF level energies from the MBPT results of Johnson *et al.* in order to obtain relativistic correlation energies.

In order to compare the theoretical results to the measured values, a total energy was determined by summing the MCDF energy, the relativistic correlation energy, and the QED correction from the Welton method. Kim *et al.* [2] then computed *predicted values* based on the total theory minus the difference between the theory and experiment. Comparison of their predicted values to the measured values led to the interesting conclusion that high- $Z$  experimental values obtained from spectra of laser-produced plasmas (at the OMEGA and NOVA laser facilities) had a tendency to be too high in energy as compared to the calculations, while most of the data (from less high  $Z$  ions) obtained from tokamak plasmas [PLT tokamak and the Texas Experimental Tokamak] were in excellent agreement with their predicted values (Fig. 1).

The second calculation of the QED correction terms, by Blundell [13], involved *ab initio* calculations of the screened self-energy and the vacuum polarization. A comparison of the energies of the  $4s_{1/2}-4p_{3/2}$  transitions as predicted by Blundell and by Kim *et al.* shows that other than for low  $Z$ , the two nearly independent predictions have very similar results, with the Kim *et al.* predictions having slightly higher energies.

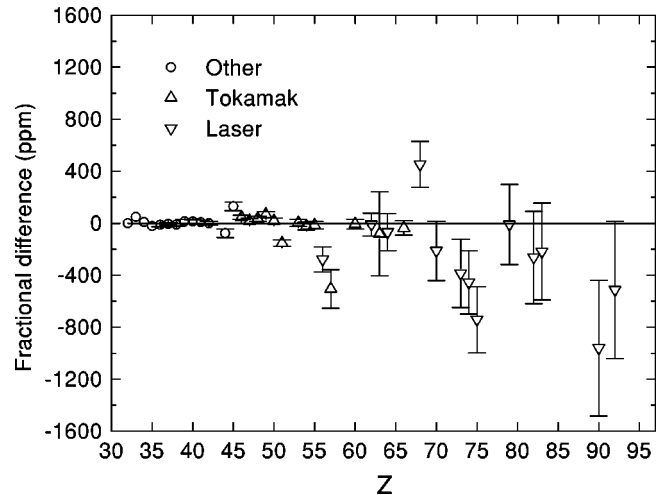


FIG. 1. Fractional difference of the  $4s_{1/2}-4p_{3/2}$  transition wavelengths predicted by Kim *et al.* [2] and the measured values from high-temperature plasma sources.

### III. EXPERIMENT

The experiment was done at the University of California Lawrence Livermore National Laboratory EBIT facility. Of the laboratory's two electron-beam-ion traps, the lower-energy device, EBIT-II, was employed.

The measurements covered the six elements Yb ( $Z=70$ ), W ( $Z=74$ ), Au ( $Z=79$ ), Pb ( $Z=82$ ), Th ( $Z=90$ ), and U ( $Z=92$ ). Ions of the respective species were introduced into EBIT-II by means of a metal vapor vacuum arc ion source. 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 of 2.0–4.5 keV for the elements considered here [14], which is but a small fraction of the working range of EBIT-II. 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 very “clean” extreme-ultraviolet (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 the Cu-like ion were employed to learn about possible contamination of the spectra by lines from ions in lower charge states.

Cu-like ions have two prominent resonance transitions,  $4s-4p_{1/2}$  and  $4s-4p_{3/2}$ . In the very heavy ions of present interest, both lines are in the EUV range of the spectrum, but the wavelengths differ by more than a factor of 2. Only the  $4s^2S_{1/2}-4p^2P_{3/2}^o$  transition falls into the detection range of our spectroscopic setup. However, the upper level of the  $4p$  configuration,  $4p^2P_{3/2}^o$ , is arguably the more interesting one, as here the wave function is more strongly affected by relativistic effects than it is for the  $4p^2P_{1/2}^o$  level (the  $4p_{1/2}-4p_{3/2}$

“fine structure” interval, a relativistic effect, being as large as the  $4s-4p_{1/2}$  electrostatic energy difference). We note that a measurement of the  $4s_{1/2}-4p_{1/2}$  energy splitting with improved accuracy has recently been reported using electron-ion interactions at the CRYRING heavy-ion storage ring [15].

The present measurements employed a flat-field spectrometer system (FFS) [16]. The spectrometer was equipped with a 2400 lines/mm variable line spaced concave grating [17] and a cryogenically cooled back-thinned charge-coupled device (CCD) camera. The camera chip had  $1024 \times 1024$  pixels on an area of about  $25 \text{ mm}^2$ . The FFS imaged the light from the ion trap, using the  $50\text{-}\mu\text{m}$ -diameter electron beam [18] as the source, onto the CCD chip, where it resulted in the geometrically expected width of about two pixels. The total area of the CCD chip was binned by a factor of 4 in the nondispersive direction to create an effective CCD array of  $256 \times 1024$  pixels. Due to spectral aberrations, the image of each line is slightly curved at the CCD surface. Simply summing across the dispersion direction would, therefore, result in broadened spectral features. Instead, the files were individually filtered for cosmic rays and collapsed into one, three, or eight columns depending on the line intensity available for the various elements in the typically 20 min of exposure time for individual spectra. Each of the sections was independently calibrated, effectively increasing the number of only weekly dependent results by a factor equal to the number of columns. Calibration was performed *in situ* by determining the central position (in channels) of a number of well-known transitions in H- and He-like ions of C, N, and O, of Li-like Ne, and of Ne-like Ar, then fitting a third-order polynomial to the calibration data. For reference line wavelengths, see the on-line databases [19] and the references therein, as well as the accurate calculations of He-like ions by Drake [20].

The background light emission recorded by the CCD was determined by running EBIT with an “inverted trap” [21], that is, with the middle drift tube at a higher potential than the top drift tube, so that no trapping occurred and any ions would be expelled. Since there is some random noise associated with the readout of each CCD image, several background spectra were averaged and smoothed before they were subtracted from the data files. The peaks were fit with Gaussian functions. An example of the spectra and calibration is shown in Fig. 2 for the case of uranium. A summary of the results from each of the measurements of Cu-like ions is found in Table I. For more details, see Ref. [22].

Yb ( $Z=70$ ), as the lowest- $Z$  element covered here, serves as a useful connection between the high- $Z$  EBIT-II measurements and those made of middle- $Z$  elements at tokamaks. With a wavelength of near  $75 \text{ \AA}$ , this transition fits well into the optimal working range of the 2400 lines/mm grating. The Yb spectra were recorded at an electron-beam energy of  $E_{beam}=2.51 \text{ keV}$  and a beam current of  $I_{beam}=57 \text{ mA}$ . Nine hours of observation resulted in 27 two-dimensional spectra of this transition, each of which was evaluated as three individual spectral strips. Neon and carbon calibration spectra were taken in between each 2 h of Yb data, whereas nitrogen was injected before and after the 9 h of Yb injection.

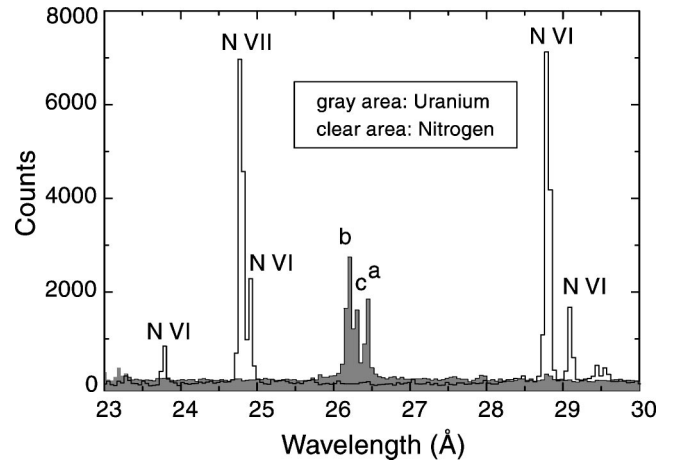


FIG. 2. EUV spectra obtained with uranium injection (shaded area) and nitrogen injection (unshaded area). The marked N lines are some of those used in the calibration. The uranium lines are *a*, Cu-like  $U^{63+} 4s^2 S_{1/2}-4p^2 P_{3/2}^0$ ; *b*, Zn-like  $U^{62+} 4s^2 {}^1S_0-4s4p^1 P_1^0$ ; *c*, blend of the Ga-like  $U^{61+} 4s^2 4p^2 P_{1/2}^0-4s4p^2 P_{3/2}^0$  and Ge-like  $U^{60+} 4s^2 4p^2 {}^3P_0-4s4p^3 D_1^0$  transitions. U was ionized and excited with  $E_{beam}=6.8 \text{ keV}$  and  $I_{beam}=113 \text{ mA}$ .

(Similar procedures were applied for the other elements.) As a final check of systematic errors, a transition to the ground state of the Be-like ion of Ne was measured, the  $1s^2 2s^2 {}^1S_0-1s^2 2s4p^1 P_1^0$  line with a reported wavelength value of  $75.765(5) \text{ \AA}$  [23]. The wavelength measured using the above method of calibration and six independent measurements was  $75.7665(36) \text{ \AA}$ , where the error in the last two digits listed in parentheses is the statistical deviation of the six measurements. A similar statistical spread was found in the measurement of the Cu-like lines. The accuracy of this measurement was limited by the lack of precise calibration lines near the long-wavelength end of the spectrum.

The W measurement was part of a study of the EUV spectra of a number of charge states that will be discussed in detail elsewhere [25]. An independent study of the EUV spectrum of tungsten in an electron-beam-ion trap has been presented by Radtke *et al.* [26]. In order to investigate the production of the various ions and to identify their individual spectra, the electron beam was varied in a series of energies from 1.7 keV to 3.0 keV, that is, both above and below threshold for the production of the Cu-like ion.

The production of intense lines from Au was readily achieved. The Au data were recorded at the optimal energy for the production of the Cu-like charge state. The wavelength of the transition of interest in the Cu-like ion of Au, near  $48.9 \text{ \AA}$ , warranted the use of two transitions to the ground state of Ne-like Ar, having wavelengths of  $48.730$  and  $49.180 \text{ \AA}$ , respectively, for calibration. However, the reference line wavelengths are known up to only  $\pm 0.002 \text{ \AA}$  [24]. This increased uncertainty was somewhat compensated for by the use of the C, N, and O spectral lines in the shorter-wavelength region. Figure 3 shows a sample spectrum of Au from this investigation. Lower-precision data on Cu- and Zn-like Au ions have been reported from an independent survey of Au spectra from many charge states using the same electron-beam-ion trap and detection system, but less elabo-



TABLE I. Predicted and measured wavelengths of the  $4s^2S_{1/2}-4p^2P_{3/2}^o$  transition in Cu-like ions of the six ions covered in these experiments. Of the earlier experimental results, only a representative one of the laser-produced plasma studies is quoted. All values are given in units of angstroms. The numbers in brackets are the errors of the previous measurements as stated in the literature. For the present measurements, the numbers in parentheses are the statistical and systematic errors, respectively. The brackets denote the combined uncertainty limits.

Element	Z	Theory [2]	Theory [13]	Experiment [1]	Experiment, this work
Yb	70	75.860	75.864	75.842[15]	75.8595(24)(40)[47]
W	74	62.334	62.341	62.304[15]	62.3355(19)(40)[45]
Au	79	48.929		48.928[15]	48.9280(21)(14)[26]
Pb	82	42.377	42.381	42.349[15]	42.3740(52)(25)[58]
Th	90	29.018	29.022	28.990[15]	29.0224(28)(10)[30]
U	92	26.420	26.423	26.400[15]	26.4325(15)(10)[19]

rate calibration procedures [27]. That work supports the spectral analysis presented here.

The wavelength of the Cu-like Pb transition of interest coincides with the carbon-edge absorption identified in the intensity calibration of the CCD at 42.4 Å, similar to other such work [28]. Consequently, the signal was relatively weak. Calibration of the Pb data files was made by fitting known C, N, and Ne lines, as was done for Yb. No separation into columns was performed for these data. Two datasets were taken with  $E_{beam}=4.53$  keV; a further seven sets had  $E_{beam}=5.08$  keV. Since the wavelength of this line is close to the well-known  $1s^2^1S_0-1s2p^1P_1^o$  transition in the He-like  $C^{4+}$  ion, the uncertainty introduced by the Ne calibration lines was not significant.

Six sets of Th data yielded intensities that were sufficient for dividing the spectra into eight columns as was done for Au, resulting in 48 measurements of the line position. The line of interest (near 29.0 Å) falls in between the He-like N resonance and intercombination lines at 28.7870 and 29.0843

Å [20], respectively, which helped to achieve high accuracy. As a test of the precision of the fit, residual oxygen lines present in the Th data were measured. The He-like resonance line of O was measured at a wavelength of  $21.601 \pm 0.006$  Å, very close to the literature value of 21.6015 Å [20] and well within the statistical error bar. Hydrogenlike  $Ly_{\alpha 1}$  was measured to be at  $18.9670 \pm 0.0061$  Å, compared to the calculated value of 18.9671 Å [29].

The wavelength of the transition of present interest in the Cu-like U ion is near 26.4 Å. Therefore the N and O lines were again used for calibration. Ne-like Ar lines were also tested in the calibration of this spectrum. Two sets of U data were obtained at  $E_{beam}=6.79$  keV, and a third at  $E_{beam}=8.10$  keV. At the higher energy, above the ionization potential of the Ni-like charge-state ion, the signal rate from the Cu-like ion was significantly lower, but nearly all of the lower charge-state lines were absent.

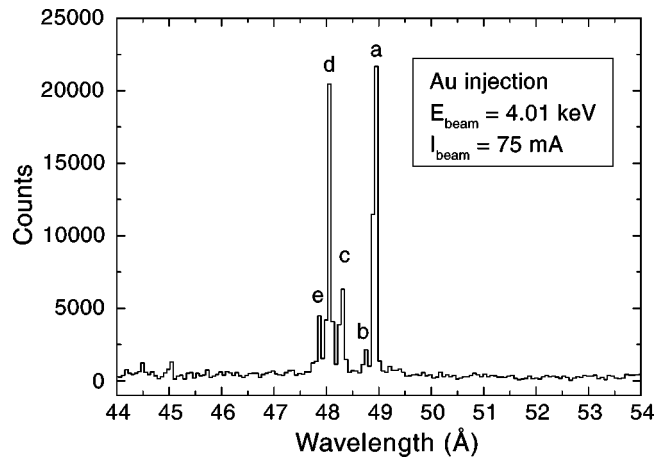


FIG. 3. Au spectrum showing a weak line at a wavelength just shorter than the line of interest from the Cu-like ion; *a*, Cu-like  $4s_{1/2}-4p_{3/2}$ ; *b*, weak transition likely from the Zn-like charge state; *c*, a blend of Ga-like  $Au^{48+} 4s^2 4p^2 P_{1/2}^o-4s 4p^2 P_{3/2}$  and Ge-like  $Au^{47+} 4s^2 4p^2 P_{1/2}^o-4s 4p^2 P_{3/2}$ ; *d*, Zn-like  $Au^{49+} 4s^2 1S_0-4s 4p^1 P_1^o$ ; and *e*, Ga-like  $Au^{48+} 4s^2 4p^2 P_{1/2}^o-4s 4p^2 P_{1/2}$ .

#### IV. ERROR ANALYSIS

A reliable error budget must include both statistical and systematic errors. The LLNL EBIT group has been using a strict definition of the statistical error, especially when measuring lines of interest for comparing with fundamental atomic structure theory, such as the determination of QED contributions in lithiumlike ions [30–32]. Whenever possible, multiple measurements are made, and the spread among individual measurements is added in quadrature to the error based on the line intensity. This procedure produces a more reliable value for the accuracy of a given measurement than can be derived from counting statistics and line-width alone, because it accounts for errors that would not be apparent otherwise, and which may only be guessed at when assessing systematic errors.

Each of the lines from the Cu-like ions had at least 3000 counts in a single spectrum, usually many more, and a full width at half maximum of  $\approx 1.7$  channels. From these, the error for each line is  $\sigma_i \approx 0.017$  channels. As detailed above, each of the Cu-like lines was measured multiple times, the least of which was during the Pb measurement, for which

nine measurements were made. From this we obtain a measure of the scatter of the multiple measurements. For the test case of Pb, a value of  $\sigma_{s.d.(\text{Pb})}=0.0864$  channels emerged, which is equivalent to  $0.0052 \text{ \AA}$ . This value is nearly an order of magnitude larger than the statistical error of  $0.0006 \text{ \AA}$  that results from the linewidth and the sum of events from all measurements of the line. The standard deviation  $\sigma_{s.d.}$  of each measurement is given in the first set of (round) parentheses for the present measurements listed in Table I.

Systematic errors are often subtle. Measurements were run at various electron-beam energies in search of lines from other charge states that might skew the peak fitting. Commonly found was a line from a lower-charge-state ion that appeared at slightly shorter wavelength, as depicted in Fig. 3 for the Au data, but this line was easily resolved from the Cu-like line, causing no notably increased uncertainty in the wavelength determination.

Data taken hours apart were compared with the intent to learn about temporal shifts of the spectra that might result from shifts in the position of the beam or shifts due to temperature fluctuations in the room—none were found.

The only significant systematic error to be noted is the error in the calibration lines. As discussed above, whenever possible, only precisely calculated or measured hydrogenlike or heliumlike transitions were used for calibration. The lines in H-like ions are known to better than  $0.001 \text{ \AA}$ , conservatively resulting in a  $1 \text{ m\AA}$  systematic error. Beyond  $45 \text{ \AA}$ , however, it became necessary to use transitions from the Li and Ne isoelectronic sequences. Specifically, the Li-like Ne transitions used in the calibration have a wavelength uncertainty of  $5 \text{ m\AA}$ , which added a systematic uncertainty of  $4 \text{ m\AA}$  to the calibration in the  $75\text{-\AA}$  region of the spectrum.

The total systematic error is listed in the second set of parentheses for the present measurements listed in Table I. It should be noted that in all cases where it was possible to measure a well-known independent transition, agreement with the known values was much better than these error ranges. For an overall error estimate, the statistical and systematic errors were combined in Gaussian quadrature; the resulting final uncertainty estimate is listed in brackets in Table I.

## V. SUMMARY OF RESULTS

A summary of the results from this series of measurements on Cu-like ions, as well as a listing of other measurements and of two theoretical predictions, is given in Table I. Instead of a complete list of all known experimental data [1,4,33–35], we only quote the results of the latest set of measurements. Our results are typically more precise than the earlier data by a factor of 3. Because of the favorable experimental conditions offered by our electron-beam-ion trap, we also assume that our data are more accurate, that is, they are suffering less from unaccounted systematic errors.

We note that for the lowest- $Z$  element, Yb, the value measured at the NOVA laser was reported as a wavelength of  $75.842(15) \text{ \AA}$  [33]. A measurement at the OMEGA laser facility yielded an even shorter wavelength of  $75.816(15) \text{ \AA}$  [36]. The measurement at the PLT tokamak at  $75.85 \text{ \AA}$  [9],

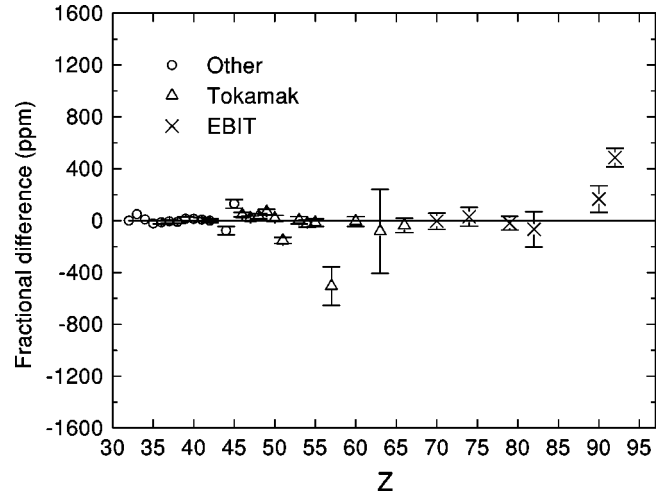


FIG. 4. Fractional difference of the wavelength predictions by Kim *et al.* [2] and experimental data from plasma sources ( $Z \leq 68$ ) and EBIT-II ( $Z \geq 70$ ).

but with a relatively large error bar of  $0.03 \text{ \AA}$ , agrees better with the present measurement.

Based on our findings, Fig. 1 has been revised into Fig. 4. Here we removed all of the laser-produced plasma results and included the six measurements of the Cu-like  $4s_{1/2}-4p_{3/2}$  transition from EBIT-II instead. The figure shows that there now is agreement between the measured and predicted values of this transition along the entire range of  $Z$ , except at very high  $Z$ . The datum for U differs from theory by about five times our uncertainty estimate. Clarification of the situation in this range may possibly arise from calculations that include two-loop QED contributions to all orders. However, such calculations have been done for Li- and Na-like ions only [37], and would need to be extended to the Cu isoelectronic sequence.

Although it is impossible to know why the transitions measured at laser-produced plasmas tended to show higher energies than either theory predicted or what was measured at the EBIT-II device, it is plausible that line blends from ions of lower charge states interfered with the proper line interpretation. From the work done on W [22,25], it is evident that intense lines from the As-, Ge-, and Ga-like charge-state ions lie between the Zn-like  $4s^2-4s4p$  and Cu-like  $4s_{1/2}-4p_{3/2}$  transitions. The Au data in Ref. [27] show a similar situation. Even with the use of an almost monoenergetic electron beam, the measurements on EBIT-II were not able to fully eliminate the Zn-like ions and still retain a significant signal from the Cu-like charge state. Laser-produced plasmas do not have the capability of an electron-beam-ion trap of isolating individual charge states by excitation energy threshold discrimination, and it is likely that lines from lower-charge-state ions were present in the data used to measure the Cu-like transition. Should these higher-energy lines not be accounted for in the fit, the measured line position of the Cu-like transition would be shifted towards the higher-energy end of the spectrum.

The disagreement between the presently reported value of

the wavelength for Cu-like U and that predicted by Kim *et al.* and by Blundell [2,13] may indicate that improvements in theory are necessary at high  $Z$ . Further measurements of this transition in U would be useful. Also, a series of measurements at high  $Z$  with five times better precision than our present measurements would enable a distinction between the calculations by Kim *et al.* [2] and by Blundell [13].

## ACKNOWLEDGMENTS

We are happy to acknowledge the dedicated technical support by Ed Magee and Phil D'Antonio. The work at the University of California Lawrence Livermore National Laboratory was performed under the auspices of the Department of Energy under Contract No. W-7405-Eng-48.

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