# Experimental study of the x-ray transitions in the heliumlike isoelectronic sequence: Updated results

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# Experimental study of the x-ray transitions in the heliumlike isoelectronic sequence: New results

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## Abstract

We revisit the discrepancy between experiments and theory for the x-ray transitions of heliumlike ions and report on a measurement of the $n = 2 \rightarrow n = 1$ x-ray transitions of heliumlike $Cu^{27+}$. These measurements were carried at the Livermore electron beam ion trap facility and achieved an accuracy of 18 ppm. The measured values show reasonable agreement with theory, but they do not follow the trend established by other measurements with similar uncertainties.

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## I. INTRODUCTION

Two-electron, or so-called heliumlike, ions are the simplest multi-electron systems, and they represent a testbed for the development of different aproaches to treating relativistic and QED effects in the presence of electron "screening," including all-order calculations in the parameter $\alpha Z$, as discussed by Artemyev et al. [1]. Although the parameter $\alpha Z$ does not approach unity until high values of the atomic number $Z$ are reached, experiments studying x-ray transitions connecting to the heliumlike $1s^2$ ground state may already be sensitive to different approaches or uncalculated terms in low- and mid-$Z$ ions. In other words, because of the complexity introduced by the additional electron, experiments involving heliumlike ions in this range of $Z$ are more likely to test and distinguish among theoretical predictions than experiments studying one-electron ions, which are unencumbered by electron correlations and thus are 'easiest' to describe theoreticallly. Indeed, published experimental data for one-electron ions have shown agreement with theory throughout the range of atomic numbers. The level of agreement for hydronlike ions, though, is both unexpected and disconcerting [2]: The uncertainty limits of essentially *all* x-ray measurements overlap with the theoretical values, despite the expectation from the meaning of the experimental uncertainties that about a third of all measurements should produce values that disagree with predictions – if the uncertainties truly represented the often quoted 1-$\sigma$ error bars. This has led to speculation that the theory for hydrogenlike ions [3] is so well established that authors may not publish results that would be seen as being in disagreement [2]. An exception has been the measurement of hydrogenlike $\mathrm{Ge}^{31+}$ by Chantler et al. [4], which was published subsequent to the analysis in [2]. The potentially self-selecting experimental results for hydrogenlike ions give added importance to testing the predictions of the heliumlike x-ray transition whose theoretical predictions are inherently less well trusted than those for hydrogenlike ions.

Twenty-five years ago Beiersdorfer et al. published experimental data from crystal spectrometer measurements of the x-ray lines of heliumlike ions of several elements between potassium and iron [5]. The measurements were performed on the Princeton Large Torus (PLT) tokamak with an accuracy of about 40 ppm. The PLT results for the $1s2p\ ^1P_1 \rightarrow 1s^2\ ^1S_0$ heliumlike resonance line were combined with then-existing wavelengths obtained from experiments studying ions between sulfur and krypton from heavy-ion accelerators [6–10] to show that the measured values for ions with atomic number $Z \geq 19$ were somewhat shorter than theories predicted at the time. Although the uncertainties of all PLT measurements overlapped with the best available theory at the time, the consistent trend that they noted led the authors to suggest a need to include additional terms in the calculations [5].

For reference, we reproduce the figure presented by Beiersdorfer et al. [5] in Fig. 1. The comparison with calculations encompassed theoretical values from three approaches available at the time: the non-relativistic variational approach augmented with an elaborate treatment of QED effects used by Drake [11], the multi-configuration Dirac-Fock method used by Indelicato [12], and the Z-expansion method of Vainshtein and Safronova [13]. The comparison shows a systematic difference between the measurements and the three theories, whereby the largest differences were seen with the theory of Vainshtein and Safronova in Fig. 1(c). The difference with the theory of Vainshtein and Safronova suggested an almost linear increase of the fractional wavelength, or energy, differences (and thus a cubic increase of the absolute energy differences with $Z$). Note, though, that the differences appear to vanish, if the trend is extrapolated to about $Z = 10$, and to reverse sign for values of $Z$ below. The comparisons in Fig. 1(a) and Fig. 1(b) suggest a crossover between $Z \approx 16 - 18$, i.e., they suggest that the differences change sign for atomic numbers below these values.

The advent of electron beam ion traps for studying the x-ray emission of highly charged ions [14, 15] has enabled measurements that, in principle, might have fewer systematic uncertainties than tokamak and heavy-ion accelerator experiments. Unlike in plasma sources, dielectronic satellite transisitions that tend to blend with the heliumlike resonance line, which is commonly referred to as line $w$, are absent. Such satellites are present in tokamak measurements, but at a reduced level compared to high-density plasma sources such as vacuum sparks or laser-produced plasmas, which have also been used to measure the x-ray lines of heliumlike ions [16–18]. Opacity effects, which affect high-density plasma sources, do not exist in electron beam ion trap plasmas, nor are these measurements affected by relativistic Doppler shifts associated with many heavy-ion accelerator measurements.

Maclaren et al. and Widmann et al. have used the Livermore electron beam ion traps to measure the energies of the x-ray transitions of heliumlike $Ge^{30+}$ and of heliumlike $Kr^{34+}$ [19, 20]. These new values are plotted together with the older values in Fig. 2. Again, they found values that were slightly larger than those predicted by Drake [11]. For example, the measurement of $Kr^{34+}$ by Widmann et al. was $13114.68 \pm 0.36$ eV [20], while Drake predicted 13114.33 eV, i.e., the difference was within error limits and similar to what was found in the PLT measurements. However, Widmann et al. could not confirm the value obtained earlier on a heavy-ion accelerator. The heavy-ion accelerator value was $13115.45 \pm 0.30$ eV [10], i.e., 0.77 eV larger than the value of Widmann et al., suggesting that the heavy-ion accelerator measurement could be an outlier.

Chantler et al. used the Gaithersburg electron beam ion trap to measure the x-ray lines of heliumlike $V^{21+}$ [21]. For line $w$ they reported a value of $5205.10 \pm 0.14$ eV. This compares to a value of $5205.27 \pm 21$ eV obtained by Beiersdorfer et al. [5]. The new measurement, thus, had a reduced uncertainty of about two third of that of the older PLT value, and the two values overlapped within their respective uncertainties. The Gaithersburg vanadium value, however, agreed better than the PLT value with theory, as shown in Fig. 2, leading Chantler et al. to conclude that "we therefore find no evidence of the earlier reported trend that experimental values are greater than theory."

Although Chantler et al. [21] did not find evidence for the observation by Beiersdorfer et al. [5] that experimental values were larger than theory, the theorists did. Theoretical approaches [1, 22–24] developed subsequent to those referenced by Beiersdorfer et al. all have produced energies larger than the values calculated by Drake [11]. For example, in Fig. 2 we include the values reported by Cheng et al. [24], which have come close to wiping out the discrepancy with experimental values, especially when the $Kr^{34+}$ accelerator measurement is no longer allowed to dominate the the high-$Z$ trend. The most recent theoretical values, which are from Artemyev et al. [1], fall between the theoretical values of Drake [11] and Cheng et al. [24].

Additional measurements have extended the original plot reported by Beiersdorfer et al. [5] both to higher and lower values of $Z$. For example, Thorn et al. [25] have used the Livermore SuperEBIT electron beam ion trap to measure the x-ray transitions of heliumlike $Xe^{52+}$. They find a value of $30631.2 \pm 1.2$ eV for the $w$ line. While this value differs outside the error bar from Drake's value of 30629.28 eV, it is in marginal agreement with the 30630.05 eV from Artemyev et al. and in good agreement with the prediction of 30630.64 eV by Cheng et al. Applying x-ray excitation by a synchrotron beam to $Fe^{24+}$ produced and confined in an electron beam ion trap has also produced data with very low uncertainties. Rudolf et al. [26] measured a value of $6700.55 \pm 0.07$ eV, which agrees exactly with the predictions of

Cheng et al., but has a slightly higher energy than the prediction by Artemyev et al., and, as expected from Fig. 1(b) is significantly higher than Drake's value.

Twenty five years ago, one of the measurements of $Ar^{16+}$ has had the lowest uncertainty of any of the measurements (12 ppm) and was in excellent agreement with the prediction of Drake [9]. This is true also today, as several very high precision measurements of $Ar^{16+}$ have been recently reported with uncertainties as low as 2 ppm [27–29], although very recently some of these uncertainties have been revised upward by a factor of two [30]. These measurements again were performed using an electron beam ion trap. They confirm what was seen 25 years ago, namely that the argon values appear to be in excellent agreement with the theory of Drake as well as with Artemyev et al. They are not, however, in agreement with the predictions of Cheng et al. Finally, we note that there are also new values for heliumlike ions with atomic number lower than argon. Engström and Litzén reported values for the $w$ line of $C^{4+}$, $N^{5+}$, and $O^{6+}$ [31]. As intimated by the original plots by Beiersdorfer et al. [5] (cf. Fig. 1), the differences between theory and experiment from such low-$Z$ ions should change sign, and indeed they do. Whether this change in sign is real or just an artifact of unknown systematic errors in the measurements remains to be seen.

A new value for the $w$ line of $Ti^{20+}$ was measured recently by Chantler et al. [32]. Like the measurement of $V^{21+}$ by Chantler et al., the $Ti^{20+}$ measurement was performed at the Gaithersburg electron beam ion trap and the resultant value overlaps with the old value from the PLT tokamak. But unlike their previous measurement of $V^{21+}$, which had an energy *less* than that predicted by Drake, their new measurement of $Ti^{20+}$ has *more* energy than Drake's prediction. Consequently, the authors have now "discovered" a $Z$-dependent discrepancy between experiment and theory for which they did not find evidence earlier. They fitted the discrepancy with a $Z^3$ dependence. Such a power law fit, unfortunately, does not reproduce the excellent agreement between experiment and theory for $Ar^{16+}$, as noted in a pointed criticism of this fit in a Comment by Epp [33]; nor does it predict the change in sign afforded by the data from Engström and Litzén for the $w$ lines of $C^{4+}$, $N^{5+}$, and $O^{6+}$.

The controversy ignited by the analysis of the heliumlike x-ray data of Chantler et al. [32–34] has prompted us to analyze our measurements of heliumlike $Cu^{27+}$ recorded on the Livermore SuperEBIT and EBIT-II electron beam ion traps. The uncertainties of our measurements are not as good as those of the best argon or iron measurements, but because they are of a higher-Z ion they add a point in a region of atomic number where no data are so far available. We find $Cu^{27+}$ energies that are somewhat *smaller* than predicted.

## II. EXPERIMENT

The $Cu^{27+}$ K-shell emission had been recently employed as reference lines to calibrate some of our measurements of the L-shell transitions of highly charged tungsten [35]. The $Cu^{27+}$ lines in turn can be calibrated by a set of hydrogenlike lines. We note that the $Cu^{27+}$ lines have twice the energy of the K-shell Rydberg transitions of hydrogenlike $Ar^{17+}$. Consequently, $Ar^{17+}$ lines can serve as calibration lines, as we demonstrated when we measured the structure and QED contributions to the $2s_{1/2}$-$2p_{3/2}$ transitions in neonlike $Th^{80+}$ through lithiumlike $Th^{87+}$ [36] as well as to the electric dipole forbidden $2p_{1/2}$-$2p_{3/2}$ transitions in neonlike $U^{82+}$ through berylliumlike $U^{88+}$ [37]. For the present measurements, we recorded the $Ar^{17+}$ lines in first order to calibrate the $Cu^{27+}$ lines measured in second order.

Similar to our measurements of the L-shell lines of tungsten [35], the spectra were recorded with a von Hámos-type crystal spectrometer [38]. This spectrometer is reasonably matched to the narrow line source formed by the electron beam and provides high photon throughput with moderately high spectral resolution, as demonstrated in many earlier measurements [39–42]. In order to have the spectral coverage to detect multiple reference lines, the instrument has a resolving power less than that afforded by the temperature of the ions, which in our electron beam ion traps has been measured to be a few hundred eV for typical operation conditions that maximize photon yield, i.e., trap depths of 100 to 300 V and beam currents above 150 mA [43–46].

The spectrometer uses a 12 cm $\times$ 5 cm $\times$ 0.02 cm LiF(200) crystal, which has been bent cylindrically to a radius of curvature of 30 cm in the case of the SuperEBIT measurements and to a radius of curvature of 75 cm in the case of the EBIT-II measurements. The $2d_\infty$ spacing of this crystal plane is equal to 4.027 Å [47]. The spectrometer is equipped with a multiwire proportional counter with a sensitive area of $10 \times 3$ cm$^2$ [48].

The second-order region between 1.4 and 1.6 Å, which contains the K-shell lines of heliumlike $Cu^{27+}$ and, in first order, the Ly-$\gamma$, Ly-$\delta$, and Ly-$\epsilon$ lines of $Ar^{17+}$ was observed in two different spectrometer settings using the high-energy SuperEBIT electron beam ion trap and in one spectrometer setting using the EBIT-II electron beam ion trap. Copper was injected into the trap with a metal vapor vacuum arc (MeVVA) injector [49], while argon was introduced via ballistic gas injection.

A typical spectrum of heliumlike $Cu^{27+}$ is shown in Fig. 3. We use the labels $w$, $x$, $y$, and $z$ introduced by Gabriel [50] to denote the heliumlike transitions from levels $1s2p\ ^1P_1$, $1s2p\ ^3P_2$, $1s2p\ ^3P_1$, and $1s2s\ ^3S_1$ to the $1s^2\ ^1S_0$ ground state, respectively.

We note that because of the hyperfine interaction the $1s2p\ ^3P_0$ level is also allowed to decay to the ground state. The resulting x-ray transition blends with line $y$, as illustrated before [51]. Thus, our transition energy measurement of line "$y$" is really that of a blend of two lines. The emission from innershell satellite lines, i.e., the K-shell lines from $Cu^{25+,26+}$, is rather weak, but some of these weak satellite lines may blend with line $z$. There is no line blending with dielectronic satellite lines [52, 53] in our measurements, as such lines are not excited at the energies of the electron beam in our measurements.

The wavelengths of hydrogenlike reference lines are known from measurement ($\leq 5$ ppm [27]) and theory ($\ll 1$ ppm) to a high degree of accuracy. In the present case we have used the wavelengths calculated by Johnson and Soff [3] and by Garcia and Mark [54] for the hydrogenlike argon lines as reference standards. The fact that our experiment employs an electron beam means that the $np_{3/2} \rightarrow 1s_{1/2}$ transitions are polarized, while the $np_{1/2} \rightarrow 1s_{1/2}$ transitions remain unpolarized [55]. Because we cannot resolve the two components in each reference line, we need to model the relative contributions from each component based on the calculated angular emission, x-ray polarization, and crystal reflectivity [56, 57]. This introduces an uncertainty of the wavelength of the reference lines that translates to an uncertainty of about 5 ppm when measuring the copper lines.

Because the copper energies were determined by comparing first and second order spectra, we need to account for the fact that crystals have a different index of refraction depending on the order of reflection $n$. In general, the wavelength $n\lambda$ of a given line is given by Bragg's law [58]

$$n\lambda = 2d_\infty(1 - (2d_\infty)^2\delta/(n^2\lambda^2))\sin\theta \tag{1}$$

where $d_\infty$ is the afore-mentioned lattice spacing of the crystal, $\theta$ is the Bragg angle, and $\delta$ is the deviation of the index of refraction from unity. The value of $\delta/\lambda^2$ is taken to be independent of wavelength and equal to $3.14 \times 10^{-6}$ Å$^{-2}$ for LiF [58]. Hence, we can use the more familiar form of Bragg's law

$$n\lambda = 2d_n \sin\theta \tag{2}$$

by realizing that in first order reflection $2d_1 = 4.0267949$ Å and in second order reflection $2d_2 = 4.02694873$ Å.

## III. RESULTS AND CONCLUSIONS

The energies of the heliumlike transitions observed in the three different experiments are listed in Table I. The experiment labeled "Run 1" was conducted on the EBIT-II device, while the experiments labeled "Run 2" and "Run 3" were performed on SuperEBIT.

The uncertainties are dominated by the uncertainties that arise from the drift of the copper and argon line positions throughout the course of a run day, i.e., the uncertainties reflect the reproducibility, or lack thereof, for the different spectra in which a given line was observed. Variations can also be seen between the three runs listed in Table I. Statistical uncertainties and uncertainties in the energies of the calibration lines have been considered but contribute a rather negligible amount, even to the weakest line, i.e., line $x$.

The average experimental value is given in Table I for each observed line. It represents the weighted average of the three measurements. The combined uncertainty of the average value is 0.15 eV or 18 ppm. A statistical averaging of the uncertainties appears justified, as the drifts in the line positions are random and can be assumed to average out as more measurements are added. The uncertainties, thus, can be thought of as 1-$\sigma$ limits.

For comparison, Table I also lists the transition energies calculated by Drake [11] and Artemyev et al. [1]. The measured values for $w$, $x$, and $y$ agree with Drake's values within their uncertainty limits, although they are on average somewhat smaller. The predictions made by Artemyev et al. are about 50 meV larger than those made by Drake, and the experimental values therefore differ from these more than from Drake's. The disagreement is, however, just barely outside the uncertainty limits. By contrast, the experimental value for $z$ differs by several $\sigma$ from the values of Drake and of Artemyev et al. This may mean that the line is blended with one or more innershell satellite lines.

In Fig. 4 we plot the $Z$-dependent difference between the values for the $w$ transition calculated by Drake and those measured in various experiments. The figure shows the experimental results from tokamaks, electron beam ion traps, and heavy-ion accelerators. However, for clarity, only three $Ar^{16+}$ data are shown; the value obtained by Briand et al. [8] was omitted because its uncertainty is about two orders of magnitude larger than that of the most recent measurement.

Our $Cu^{27+}$ datum falls into a region between iron ($Z = 26$) and germanium ($Z = 32$) where so far no mesurements have been made. Although it is in reasonable agreement with the predictions of Drake and of Artemyev et al., it does not agree with the bulk of the experimental data above $Z = 18$, albeit it adds to the trend set by the low-$Z$ data from Engström and Litzén [31]. Given the general scatter of the experimental data, it is not unexpected, however, that some experimental results will be found that fall outside the general trend. It is worthwhile noting that the result from Run 1, which utilizes the higher-resolution spectrometer, agrees well with the predictions of Cheng et al. [24] and thus 'confirms' the trend established by the measurements of the neighboring ions. However, there is no *a priori* reason to discard the results from Runs 2 and 3, and, thus, they need to be included in the averaged value we have plotted in Fig. 4.

Our datum also falls well outside the $Z^3$ scaling proposed by Chantler et al. [32, 34], which is also shown in Fig. 4. The scaling by Chantler et al. treats the two highly precise measurements of $Ar^{16+}$ [27, 28] as outliers [34]. Our datum will undoubtedly be treated similarly, but no more so than they treat their own datum for $V^{21+}$ [21], the $Ar^{16+}$ datum from Deslattes et al. [9], and the three points from Engström and Litzén [31] as outliers. However, our measurement, together with another new result presented recently for $Fe^{24+}$ [26], will draw the statistical fit of Chantler et al. closer toward zero, making it essentially coincident with the calculations of Cheng et al. [24], which are also shown in Fig. 4. Thus, the 'novelty' of the $Z$-scaling by Chantler et al. [32] is reduced to the question which of the available theoretical approaches best describes the experimental data. This is not a novel question, but it has been the subject of investigation since our orignal paper [5] more than 25 years ago.

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TABLE I: Comparison of experimental and theoretical data for the four observed x-ray transitions in heliumlike $Cu^{27+}$. All values are in eV.

| Line | Experiment | | | | Theory | |
|---|---|---|---|---|---|---|
| Label | Run 1 | Run 2 | Run 3 | Weighted average | Drake [11] | Artemyey et al. [1] |
| w | $8391.12 \pm 0.20$ | $8390.31 \pm 0.30$ | $8390.61 \pm 0.30$ | $8390.82 \pm 0.15$ | 8390.98 | 8391.03 |
| x | $8371.23 \pm 0.20$ | $8371.13 \pm 0.30$ | $8371.09 \pm 0.30$ | $8371.17 \pm 0.15$ | 8371.26 | 8371.32 |
| y[a] | $8347.22 \pm 0.20$ | $8346.45 \pm 0.30$ | $8346.82 \pm 0.30$ | $8346.99 \pm 0.15$ | 8346.95 | 8346.99 |
| z | $8310.87 \pm 0.20$ | $8310.76 \pm 0.30$ | $8310.81 \pm 0.30$ | $8310.83 \pm 0.15$ | 8311.29 | 8311.35 |

[a]blended with $1s2p\ ^3P_0 \to 1s^2\ ^1S_0$ decay.

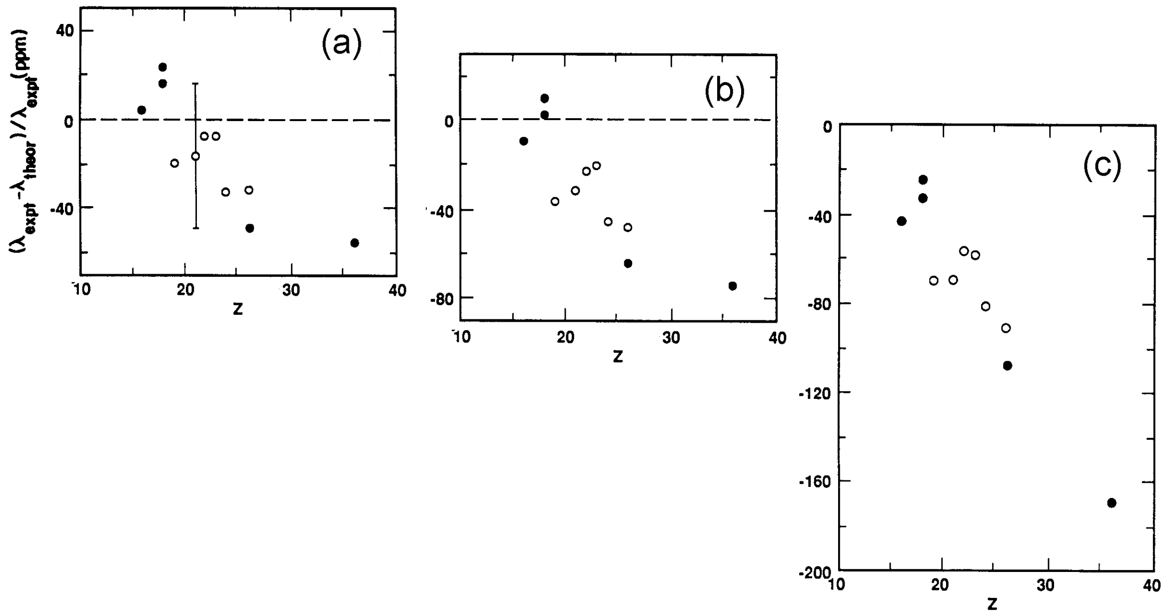


FIG. 1: Comparison of the experimental and calculated values of the $1s2p\ ^1P_1 \to 1s^2\ ^1S_0$ wavelength in different heliumlike ions. The theoretical wavelengths $\lambda_{theor}$ are given by (a) Indelicato [12], (b) Drake [11], and (c) Vainshtein and Safronova [13]. The solid points are from measurements on heavy-ion accelerators [6–10]; the open points are from measurements on a tokamak [5]. A value of 12398.54 eVÅ was used to convert between energy and wavelength at the time the experimental and theoretical data were produced. [Figure adapted from [5].]

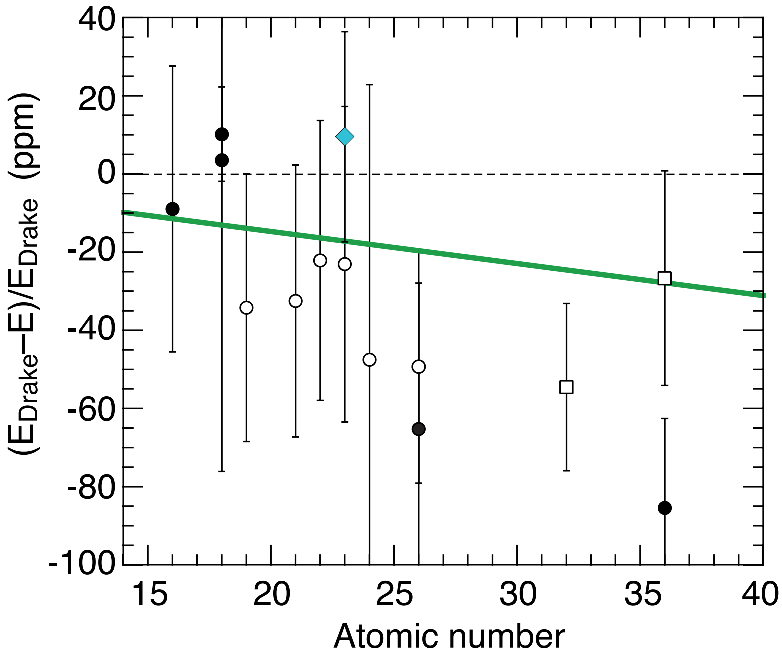


FIG. 2: Comparison of selected experimental and calculated values of the $1s2p\ {}^1P_1 \rightarrow 1s^2\ {}^1S_0$ transition energy in different heliumlike ions. All values are normalized to those calculated by Drake [11]. The calculated values from Cheng et al. [24] are given as a solid green line. Experimental values shown as black solid circles are from heavy-ion accelerators [6–10]; open circles denote tokamak results [5], open squares are results from the Livermore electron beam ion traps ([19, 20], and the blue diamond is from the NIST electron beam ion trap [21]. A value of 12398.42 eVÅ was used to convert between energy and wavelength.

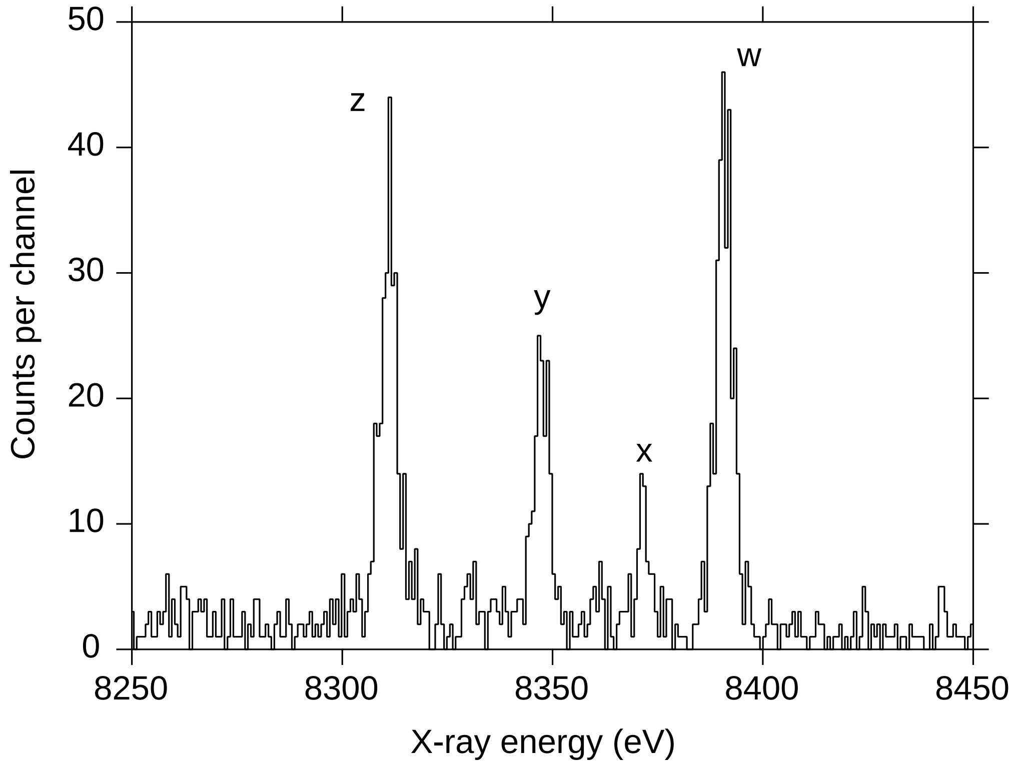


FIG. 3: Typical spectrum of heliumlike $Cu^{27+}$ showing the transitions from levels $1s2p\ ^1P_1$, $1s2p\ ^3P_2$, $1s2p\ ^3P_1$, and $1s2s\ ^3S_1$ to the $1s^2\ ^1S_0$ ground state, labeled $w$, $x$, $y$, and $z$, respectively, in the notation of Gabriel [50].

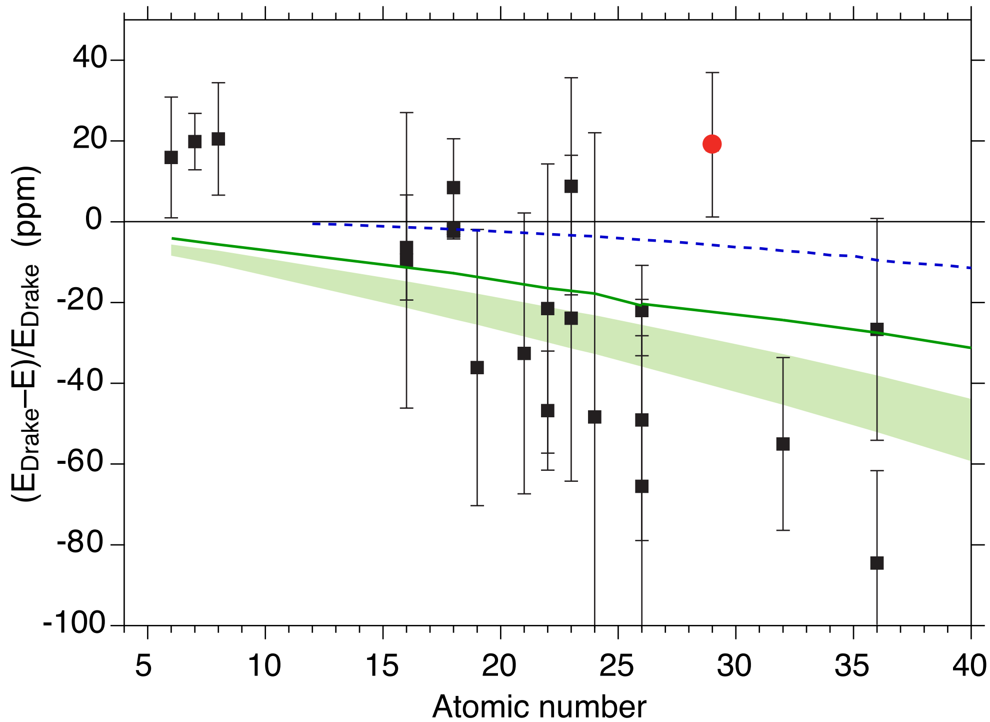


FIG. 4: Comparison of selected experimental and calculated values of the $1s2p\ ^1P_1 \rightarrow 1s^2\ ^1S_0$ transition energy in different heliumlike ions. All values are normalized to those calculated by Drake [11]. The calculated values from Cheng et al. [24] are given as a solid green line, those from Artemyev et al. [1] are given as a dashed blue line. Experimental values are shown as black solid squares and are from the following sources: $C^{4+}$, $N^{5+}$, and $O^{6+}$ - [31]; $S^{14+}$ - [6, 59]; $Ar^{16+}$ - [9, 27, 30]; $K^{17+}$, $Sc^{19+}$ - [5]; $Ti^{20+}$ - [5, 32]; $V^{21+}$ - [5, 21]; $Cr^{22+}$ - [5]; $Fe^{24+}$ - [5, 7, 26]; $Ge^{30+}$ - [19]; $Kr^{34+}$ - [10, 20]. The present value for $Cu^{27+}$ is shown as a solid red circle. A value of 12398.42 eVÅ was used to convert between energy and wavelength. The polynomial fit of the experimental data put forth by Chantler et al. [32] is shown as a green area. This fit does not include the experimental values for $Z = 6$, 7, and 8, as well as the newest values for $Z$=18, 26, and 29.