Extreme-ultraviolet spectra of highly charged Pt ions with several valence-shell electrons: Observation and accurate calculations

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Previous observations of Cu- through Ge-like high-Z ions have demonstrated that accurate measurements and theory agree well for ions with a single valence electron but that additional electrons in the valence shell cause progressively worsening computational problems. We have obtained highly resolved euv spectra of Pt ($Z = 78$) ions in an electron-beam ion trap. The measured wavelengths are compared to the results of a number of recent large-scale calculations, including our own multireference Møller-Plesset computations. The latter calculations match the best for Cu- and Zn-like ions and represent an order-of-magnitude improvement in predictive accuracy for Ga- and Ge-like ions.

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I. INTRODUCTION

For the resonance lines in one-valence-electron ions of the Cu isoelectronic sequence, there is excellent agreement of electron-beam ion trap (EBIT) measurements and highly developed theory, including QED, for high-Z elements up to $Z = 92$ [1–7]. These findings for Cu-like ions are part of the success story of EBIT measurements on one-valence-electron ions, which also comprise Li-like [8–12] and Na-like [13–15] ions. The Livermore EBIT was set up for spectroscopy more than two decades ago [16] and has since produced the largest share by far of such spectroscopic data [17]. Measurements by the Heidelberg group of Li-like ions of Fe and Cu [18,19] using photoexcitation at the Free-electron LASer in Hamburg (FLASH) light source have recently ushered in a new phase of EBIT work. Measurements of high-Z Na-like ions have also been reported from the NIST EBIT [20,21]. In contrast, laser-produced plasma spectra [22–24] and the relativistic calculations adjusted to match them show a trend different from that of the aforementioned calculations, a trend that in hindsight has been associated with systematic errors in the experiment on a high-density plasma.

EBIT studies of Zn-like ions [6,7,25] have demonstrated that, for ions with more than one electron in the valence shell, theory evidently has much larger problems. This work and associated investigations of Ga- and Ge-like spectra [3,4, 25,26] have instigated improvements of several calculational approaches, such as multiconfiguration Dirac-Fock (MCDF), many-body perturbation theory (MBPT), and relativistic configuration interaction (RCI) calculations. Whereas some of the earlier calculations only addressed spectra already measured (jeopardizing the notion of the predictive power of theory), there now are several large-scale calculations that present systematic coverage of a range of elements, including those for which experiment has not yet provided spectra. We compare the results of the recent calculations with our high-resolution euv observations of the element Pt ($Z = 78$) in an EBIT.

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II. EXPERIMENT

Measurements on highly charged ions of platinum were performed at the EBIT-I [27] at Lawrence Livermore National Laboratory. The device has been optimized for spectroscopic studies of highly charged ions [28], reaching from the aforementioned investigations of simple atomic spectra of fundamental physics interest to the study of spectra needed for diagnosing high-energy-density plasmas and tokamaks [29–31]. Pt was introduced to EBIT-I as a wire probe [32,33]. The tip of a Pt wire was moved inward from the side of the EBIT vacuum vessel until it was eroded by ion sputtering. Evaporated Pt atoms drifted toward the ion trap, where they were quickly ionized by the electron beam. Every 20–60 s, the content of the trap was dumped to halt the accumulation of possible contaminants, such as barium and tungsten, and then the trap cycle was repeated. The measurements were run along with another experiment that involved many different electron-beam energy settings. Only those data sets were considered for the present analysis that had an electron-beam energy in the range 2–5 keV; in these data sets, Ni-like ions (Pt^{50+}, $V_{ion} = 4354$ eV [34]) and Cu-like ions (Pt^{49+}, $V_{ion} = 2878$ eV) represented the highest charge states while Ga- and Ge-like ions (Pt^{48+}, Pt^{47+}) were present in a higher abundance. The charge balance was monitored by an x-ray microcalorimeter [35].

The observations employed a grazing-incidence flat-field spectrograph [36] which is equipped with an $R = 44.3$ m variable spacing grating and a cryogenic CCD detector. The same instrument has been used for some of our earlier work on Cu- and Zn-like heavy ions [7]. It depends on the narrow (diameter about 50 μm [37]) cylindrical volume of ions excited by the electron beam instead of an entrance slit. Since the earlier observations, the spectrograph has undergone several mechanical modifications; the working range now extends to beyond 100 Å.

Prominent $4s$-$4p_{3/2}$ transitions in all Pt ions mentioned are expected at wavelengths near 50 Å. In this wavelength range, calibration lines of B, N, and Ar are available, although not conveniently close. Therefore, an attempt was made to see and calibrate the Pt lines in second diffraction order, using

first-order lines of oxygen (O VI) and neon (Ne VI–VIII) [38–42] for calibration. The low-Z gases CO_2 and Ne were used on their own to establish the wavelength scale in the wavelength range covered. However, Ne was also bled into the electron-beam ion trap during the wire probe insertion of Pt; such a light element contaminant is beneficial for the evaporative cooling of the cloud of heavy ions. In this experiment, the low-density admixture of Ne provided wavelength references (mostly Ne VII and Ne VIII) in the actual Pt spectra. Even when oxygen was not injected (as CO_2), the spectra also showed some oxygen lines.

In the Kelly wavelength tables [38], many of the wavelength entries on Ne are taken from a paper by Hermansdorfer [43]. The number of decimals given there suggests overall uncertainties of a few milliangstroms, but the paper states an overall uncertainty as large as 50 mÅ. Tondello and Paget [39] give Ne wavelength data with 5–20-mÅ uncertainties; Kramida and Buchet-Poulizac [40,41] report and reevaluate data on Ne VIII, some of which are good to a few milliangstroms; Kramida *et al.* [42] reevaluate earlier data on Ne VI. In the wavelength range of present interest, there are no significant changes from the Hermansdorfer numbers. From the consistency of the database, it seems that the errors stated by Hermansdorfer may actually be much smaller. For oxygen, the wavelength uncertainties are generally smaller, probably because the correspondingly lower charge states have been reached in many light sources that were feeding precision spectrographs.

III. LINE IDENTIFICATION

Figure 1 shows a section of the observed euv spectrum; the data shown have been combined from several 60-min exposures. The linewidth (full width at half maximum) is 50 mÅ, corresponding to a resolving power of $\lambda/\Delta\lambda \approx 2000$. The

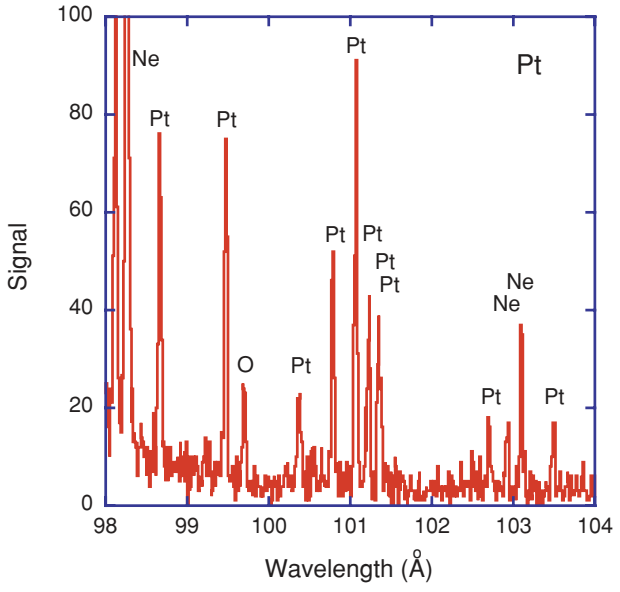

FIG. 1. (Color online) Section of an euv spectrum of Pt recorded at the Livermore electron-beam ion trap. The data for this spectrum have been combined from several spectra recorded at different electron-beam energy settings. Several lines are identified with Ne used for calibration and evaporative cooling of the trapped ion cloud.

TABLE I. Apparent wavelength values (Å) and uncertainties for those lines in Fig. 1 that have not been identified with the calibration spectra elements.

Wavelength (assume first order)	Uncertainty	Wavelength[a] (assume second order)	Uncertainty	Ident.[b] (order)
97.386	0.008	48.693	0.004	
98.658	0.006	49.329	0.003	
99.468	0.006	49.734	0.003	Ga (I)
100.360	0.008	50.180	0.004	Ga (II)
				Ge (II)
100.781	0.006	50.390	0.003	Zn (II)
101.061	0.006	50.530	0.003	Ge (I)
101.222	0.006	50.611	0.003	Ge (II)
101.345	0.006	50.672	0.003	Ga (II)
102.701	0.006	51.350	0.003	Cu (II)
103.485	0.008	51.742	0.004	

[a]Column states the wavelength for the case that the recording in the first column did correspond to an observation in second order of diffraction.

[b]Column indicates isoelectronic sequence and diffraction order of our line identifications detailed in Table II.

observed wavelengths of lines that are not from the calibration spectra of oxygen and neon are given in Table I. The individual spectra were obtained at electron-beam energy settings that favored charge states $q = 46+$ (Ge-like ions) and lower. As a result, the resonance line of the Cu-like ion ($q = 49+$) appears rather weak, but the counting statistics is good enough to determine the (first-order) wavelength to 51.350 ± 0.003 Å. This wavelength is in excellent agreement with the isoelectronic trend of the previous Livermore EBIT data [7] relative to the predictions by Kim *et al.* [1]. The calculations by Blundell [2] deviate from those by Kim *et al.* at very high Z and were found to be even more accurate in that range [7], but Blundell has not provided results for $Z = 78$. In a way, this finding of agreement with already corroborated highly accurate calculations for Cu-like ions also corroborates the above assumption that the true uncertainty of the Ne wavelength data of Hermansdorfer [43] might be significantly smaller than stated by their originator.

The line at 100.781 Å is identified with the second diffraction order of the 50.390 ± 0.003 Å resonance line in the Zn-like ion of Pt. In principle, this identification is feasible on the basis of earlier identifications along the isoelectronic sequence (see [7,26]) in combination with the results of calculations, even if those are not particularly accurate, as long as they can be adjusted to the experimental data; then the predicted isoelectronic trend can be exploited. However, the many resolved lines in the small wavelength interval 100–102 Å pose the serious problem of finding out which line is which, within the scatter of various predictions. Fortunately, there are two highly accurate calculations: the RCI calculations by Chen and Cheng [44] and our own multireference Møller-Plesset MBPT calculations [45] that point to our candidate line almost within the experimental error bar. We discuss such calculations in the next section. This line and its place in the Zn isoelectronic sequence are presented in more detail elsewhere [45].

Another eight lines in the wavelength range 98–112 Å show moderate line intensities but do not coincide with lines seen in the calibration spectra. We therefore assume that these lines arise from Pt ions of the next-lower charge states, which would primarily be Ga-, Ge-, and As-like ions. The two aforementioned identified lines are from $4s_{1/2}$-$4p_{3/2}$ transitions observed in second order of diffraction. Similar transitions are expected in the Ga- through As-like ions (and beyond), but the wavelength interval studied is also close to that of the majority of $4s_{1/2}$-$4p_{1/2}$ transitions appearing in first order of diffraction. In such heavy ions, the fine structure intervals of the ground and first excited configurations are so large that the range of levels can overlap. What at low Z may be clearly discernible line multiplets therefore can spread by a factor of 2 or more in wavelength at high Z. A line group in high-Z element spectra may resemble a line multiplet from a transition array but instead arise from similar (or even dissimilar) transitions in a variety of ion charge states.

Such extremely large fine-structure spreads were also observed in the NIST EBIT studies of W ($Z = 74$) in the x-ray spectral range [46] and of Hf, Ta, W, and Au in the euv spectral range [20]. In these works, collisional-radiative modeling was used to predict not only the wavelengths but also the relative intensities of the lines observed, and this proved important for interpreting some of the line blends. Here we employ a spectrometer which has higher resolution than the NIST instrument [47] in order to avoid line blends in most—but not all—cases of interest.

IV. ATOMIC STRUCTURE CALCULATIONS

When previously presenting euv spectra of Cu-, Zn-, Ga-, and Ge-like heavy ions observed in an EBIT, the agreement of measured wavelengths with some calculations was within 50 ppm for Cu-like ions [7], but some 5000 ppm for *ab initio* calculations of Zn-like ions (300–1000 ppm for semiempirically adjusted calculations) [6,26], and yet the agreement was progressively poorer for ions with more than two electrons in the valence shell [3,4,26]. We are happy to see that meanwhile the call for better calculations has been heeded.

Such high-Z ions as discussed here feature massive relativistic effects. The first fully relativistic treatment has been accomplished by employing MCDF codes. In the range of atomic systems of present interest, such calculations have been highly successful for Cu-like ions [1,2]. New MCDF calculations [48] have been undertaken for Cu-like ions, which in the present context are not so much needed on their own but serve as a quality reference for similar calculations of Zn- through Ge-like ions by the same Mons-Liège collaboration [49–51]. MCDF calculations by Cheng and Wagner [52] have also been shown to be among the best of Zn-like ions at the time. However, they cover only a narrow selection of elements along the isoelectronic sequence.

Relativistic many-body perturbation theory (RMBPT) has evolved in the quest for higher accuracy. The technique has, for example, been employed by the Notre Dame group on Zn-like ions [53–55]. While Blundell *et al.* [53,54] addressed only the ion species that had already been subject to measurement at the time, Safronova and Safronova have provided a more extensive coverage of the high-Z element range [55]. However, there also are significant calculational differences. The paper by Safronova and Safronova basically reproduces the results of the earlier paper by Blundell, Johnson, Safronova, and Safronova [53] (and adds more, including data on Pt) but misses the major improvements in accuracy made in the meantime by Blundell [54] (who does not cover Pt for the Zn isoelectronic sequence).

Configuration interaction (CI) calculations have long been known for their accuracy, but they are also notorious for their high demand on computing power. Since computing has become much cheaper, CI calculations have been extended to cover relativity in the form of RCI computations. Very recently, such calculations have addressed Zn-like ions [44] with high accuracy.

A variant of MBPT is the relativistic multireference Møller-Plesset (MR-MP) code to treat the structure of many-electron ions with high accuracy [56–59]. For ions of present interest, the code was first applied several years ago to Zn-like ions by Vilkas and Ishikawa [60], and their results at the time came much closer to the experimental data than any other *ab initio* computation. However, at that time, an "optimized" set of Gaussian basis functions was used in order to accommodate the limited computing resources. Such a choice of basis functions requires a tedious process of fine-tuning the basis exponents, which on its own is a possible source of uncertainty. The procedure can now be simplified by using a much bigger basis set, the so-called universal Gaussian basis set [61]. With this approach, the Ishikawa group has calculated the levels of Zn-like Pt^{48+} ions and recalculated the resonance transition energies of Zn-like ions from $Z = 70$ to $Z = 92$. For Pt, the calculated wavelength of 51.351 Å matches the measured value of 51.350 ± 0.003 Å extremely well. The results of these calculations beyond the resonance transition of Pt^{48+} ions are presented elsewhere [45]. Because of the success with Zn-like ions, the same type of calculation has also been applied to Ga- and Ge-like ions of Pt, as well as to Pt ions with more $n = 4$ electrons up to the Kr isoelectronic sequence. However, beyond the Ge isoelectronic sequence, no candidate lines were found to fall into the presently studied wavelength interval.

V. COMPARISON OF MEASUREMENTS WITH CALCULATIONS

Table II lists our line identifications ordered by isoelectronic sequence. The measured wavelengths are juxtaposed with the results of recent calculations. This process has unequivocal results only for Cu- and Zn-like ions of Pt. The problems with the other ions are discussed below. .

Recently Palmeri *et al.* [48] have compared the results of various calculations to experimental data in the high-Z range, and there is no need to repeat this exercise here. The calculations by Palmeri *et al.* deviate by about 75 mÅ or 1500 ppm from our experimental findings on Pt^{49+}. However, the deviation is about the same as for other ions in this range of nuclear charge Z, so there is no doubt about the line identification and the observational results. In Fig. 2 we compare the experimental data from $Z = 36$ to $Z = 92$ to the smooth trend of the calculations by Kim *et al* [1]. The experimental data are from high-resolution observations at the Livermore electron-beam ion trap ([7] and this work) and show

TABLE II. Wavelength values (Å) for the $Pt^{q+}4s^k4p^l - 4s^{k-1}4p^{l+1}$ transitions studied in this work.[a]

Experiment (this work)	Theory	Ref.
Pt^{49+} $3d^{10}4s\ ^2S_{1/2}$–$3d^{10}4p\ ^2P^o_{3/2}$		
51.350 ± 0.003	51.322	[22]
	51.343	[1]
	51.426	[48]
Pt^{48+} $4s^2\ ^1S_0 - 4s4p\ ^1P^o_1$		
50.390 ± 0.003	50.103	[24]
	50.196	[49]
	50.360	[55]
	50.3857	[44]
	50.3869	This work
Pt^{47+} $4s^24p\ J = \frac{1}{2} - 4s4p^2\ J = \frac{1}{2}$		
50.180 ± 0.004 bl	49.832	[50]
	50.1802	This work
Pt^{47+} $4s^24p\ J = \frac{1}{2} - 4s4p^2\ J = \frac{3}{2}$		
50.673 ± 0.003 bl	50.428	[50]
	50.6617	This work
Pt^{47+} $4s^24p - 4s^24d$		
50.673 ± 0.003 bl	50.6765	This work
Pt^{47+} $4s^24p\ J = \frac{1}{2} - 4s4p^2\ J = \frac{5}{2}$		
99.468 ± 0.006	99.416	This work
Pt^{46+} $4s^24p^2\ J = 2 - 4s4p^3\ J = 2$		
50.180 ± 0.004 bl	50.1840	This work
Pt^{46+} $4s^24p^2\ J = 0 - 4s4p^3\ J = 1$		
50.611 ± 0.003	50.555	[51]
	50.6017	This work
Pt^{46+} $4s^24p^2\ J = 0 - 4s^24p^2\ J = 2$		
101.061 ± 0.006	101.101	This work

[a]Only results of the most recent calculations have been listed for comparison; bl denotes blended lines.

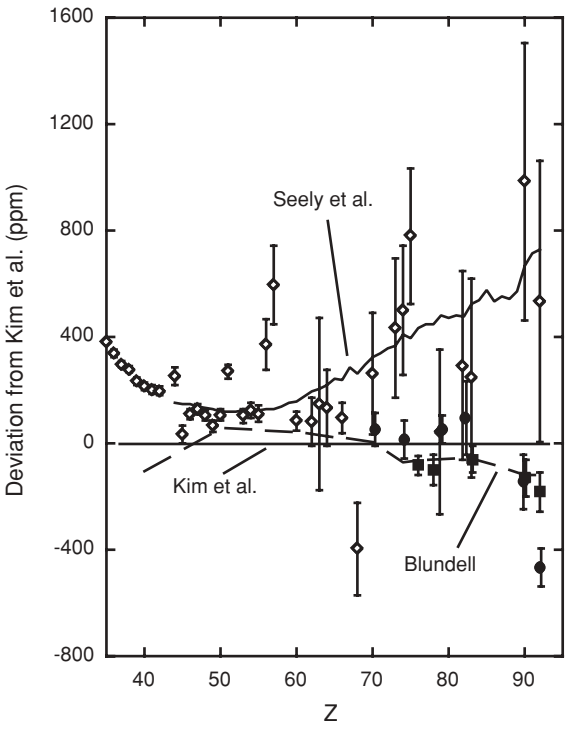

FIG. 2. Comparison of measurement and calculation for the $4s$-$4p_{3/2}$ transition energy in Cu-like ions. Horizontal line at 0, reference calculation by Kim *et al.* [1]; dashed line, Blundell [2]; solid line, Ref. [22]; open diamond, various pre-EBIT measurements including laser-produced plasma (high density) work by Seely *et al.* [22] and by Kania *et al.* [23]; solid circles, Livermore EBIT observation at moderate resolution [5]; solid squares, Livermore EBIT observations at high resolution ([7] and this work). The recent MCDF calculation by Palmeri *et al.* [48] would be almost off the scale and is not shown here.

a remarkably small scatter and excellent agreement with the calculations by Blundell [2]. We note that below $Z = 50$, both "good" calculations [1,2] deviate significantly (and differently) from experiment. The high-Z performance of both calculations is better than the low-to-medium Z quality of their results. Unfortunately, Blundell has treated only a few Cu-like ions in the high-Z range, and Pt was not among them. Our wavelength result for the Cu-like ion Pt^{49+} agrees closely (though not perfectly) with the result of the calculation by Kim *et al.*, with a deviation of merely 140 ppm, or slightly more than 2σ.

Quite a number of calculations, often semiempirically adjusted, have been published on Zn-like ions (see listing in [26]). However, having seen that most of the older calculations did not reach spectroscopic accuracy, we compare (see Table II) the new data only with the much better recent calculations. For the resonance line in Zn-like Pt ions, large-scale MCDF calculations [49] have been executed, but their results differ from experiment by about 0.2 Å (some 4000 ppm). This is better than was previously available from several *ab initio* calculations but is not always good enough for immediate line identification in the face of overlapping transition arrays from different ions. The same holds for our own (unpublished) Flexibe Atomic Code (FAC) [62] calculations, which are surprisingly accurate in view of the speed of the calculation and the relatively little effort required but do not reach sufficiently high accuracy for a meaningful comparison in the case of the transitions of present interest (and their results are not listed here). Very recent RMBPT calculations by Safronova and Safronova [55] come within 0.03 Å (600 ppm) of the experimental result, whereas the RCI calculations by Chen and Cheng [44] are closer to the experiment yet again (deviation 80 ppm), by almost an order of magnitude. Our own MR-MP calculation comes closest to our own measurement (60 ppm), practically touching the experimental error bar. This excellent agreement extends to other ions of the Zn isoelectronic sequence, as shown elsewhere [45].

The ground terms of Ga-, Ge-, and As-like ions feature fine-structure splitting and these ions, consequently, give rise to more line-rich spectra than Cu- and Zn-like ions. Although the charge state distribution favors these former spectra, the available overall signal may be spread out over several lines in a wavelength range wider than the scope of our present observation. Hence, individual lines may appear not stronger than the resonance lines in the Cu- and Zn-like ions. When searching for candidate lines, one has to keep in mind that EBIT is a low-density light source in which the ions usually have enough time to return to the ground state (or metastable levels, if available) before any new excitation

takes place. Hence, only excitations from the ground level tend to produce lines of appreciable intensity under those low-density conditions, in marked contrast to observations after the interaction of swift ions with solid matter [63].

Our calculations of As-like and lower-charge ion spectra have not produced any lines that would be expected under EBIT density conditions in the wavelength range covered by our observations. Although about half a dozen lines calculated by our MR-MP scheme for Ga- and Ge-like Pt ions fall into our observation range, only about half of them agree closely (better than 10 mÅ or 200 ppm) with observed line positions—exactly only those that are expected under EBIT conditions. Similar to the case of Zn-like ions, the MCDF predictions for the observed lines [50,51] differ from experiment by about 0.2 Å (some 4000 ppm). Compared to these and earlier calculations, our new MR-MP calculations represent an improvement in accuracy by more than an order of magnitude. Our line identifications are listed in Table II, along with results from various calculations.

Several problems remain. According to our calculations, the 101.061 Å line that we ascribe to a first-diffraction-order line of the Ge-like ion coincides with the second-diffraction-order image of a $3d^9 4s$-$4p$ transition in the Ni-like ion Pt^{50+}, predicted at 50.534 Å. Such lines play a role in x-ray lasers based on Ni-like ions. Under the low-density conditions typical for EBIT, however, such lines are not expected to be seen with sufficient signal. Several candidates for identification with first-diffraction-order lines of Pt differ by some 50 mÅ (1000 ppm) from the results of our calculations. For these $E2$ and $M2$ quadrupole transitions, collisional-radiative modeling might be helpful to judge their line intensity as a tool for identification. Several lines of Table I that we tentatively associate with Pt remain unidentified for now.

VI. DISCUSSION

We have obtained high-accuracy wavelength data on Cu-, Zn-, Ga-, and Ge-like ions of Pt. These data tighten the isoelectronic trend of the high-quality data that are already available for Cu- and Zn-like ions. Until fairly recently, the theoretical treatment of ions with more than two electrons in the valence shell left much to be desired. MR-MP calculations have successfully treated Al- and Si-like atomic systems (three or four electrons in the $n = 3$ shell) [57,64], and the present effort on Ga- and Ge-like ions (three or four electrons in the $n = 4$ shell) demonstrates that similarly high accuracy (of a few hundred ppm) can be obtained for ground-state transitions in these ions, which marks a major step forward. At this high level of spectral resolving power and computational accuracy, the combination of electron-beam ion traps and multireference Møller-Plesset calculations appears to be well suited to provide reference markers in the *terra incognita* of multielectron spectra of highly charged ions.

The high spectral resolution of the present measurements permits us to measure line positions with high accuracy, but it also entails a number of unforeseen complications. In the quest for reliable data interpretation, higher resolution is the route of choice to be preferred over spectral modeling of unresolved line blends. Modeling based on semiempirically adjusted calculations may be very helpful for interpreting spectra, but it cannot replace detailed measurements. Alas, line blends do occur even at the high spectral resolution of our data, and then collisional-radiative modeling may give hints at the relative intensities of the blended lines. The comparison of our data with large-scale calculations demonstrates the shortcomings of most (even massive) *ab initio* calculations, which are insufficiently accurate to yield most line identifications in the present sample spectra. With less computational effort than that involved in large-scale RCI calculations, however, the MR-MP algorithm can deliver rather accurate wavelength predictions of multielectron valence shell ions that are beyond the reach of most other codes.

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