Extreme ultraviolet spectra of highly charged Xe ions

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Using the SuperEBIT electron-beam ion trap and a flat-field spectrometer, we observed extreme ultraviolet spectra of highly charged ions of Xe and measured the wavelengths of prominent lines from Li-, Be-, B-, Na-, and Mg-like ions. Our results for Li- and Na-like ions are as precise as the best available elsewhere. The results for Be-, B-, and Mg-like ions are much more precise than available data or extend those available from lower-Z ions and reveal significant shortcomings of the various theoretical predictions.

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I. INTRODUCTION

Basic science interest has long focused on the spectra of ions with a single electron (H-like ions), because they can be calculated most precisely. There also is interest in ions with a single electron outside a core of closed electronic shells, such as in the ions of the Li, Na, and Cu isoelectronic sequences [1–4], because these can not only be calculated, but also measured rather precisely [5,6] (for a compilation of earlier data, see Ref. [3]). One of the major driving factors has been the quest for a determination of the quantum electrodynamical (QED) contributions to atomic structure. However, the same magnitude of QED effects is present in ions with more electrons in the same open valence shell. The structure of these ions can be measured with an accuracy that is comparable to that of measurements of single valence shell electron ions.

Assuming that highly charged ions can be calculated best (because the central potential is best defined, and fully relativistic calculations are not only necessary [7], but converge well for highly charged ions), one might consider studying the role of the electron-electron interaction that is brought about by several electrons in the same valence shell. However, highly precise calculations of Be- and Mg-like ions (see below) have not yet reached the accuracy of their counterparts for Li- and Na-like ions [1–4]. Nevertheless, recent calculations have taken into account the QED effects of several electrons. Assuming that these QED calculations are sufficiently accurate, precise experimental data can then be used to test the calculational quality of the often less accurately determined non-QED part of the treatment.

We aimed at obtaining observations on suitable ions (of Xe) that would provide benchmarks for such calculations. Our light source is an electron-beam ion trap. This is a low-density device (in contrast to laser-produced plasmas) with ions practically at rest (in contrast to the accelerator-based beam-foil spectroscopy technique). In contrast to tokamak work, higher charge states can be obtained.

II. 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 higher-energy device, SuperEBIT [8], was employed. Xe was injected into SuperEBIT by means of a ballistic gas injector with a reservoir pressure of 10^{-8} Torr or less. Inside the SuperEBIT vessel, the gas expands to a density that is lower by about two orders of magnitude at the position of the trap. 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 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 Li-like ions of Xe is about 9.6 keV, while the production of Na-like ions requires 3.25 keV [9,10].

In our measurements at 20 keV electron-beam energy, the highest charge state identified from spectral lines was Na-like Xe^{43+}. Lines of ions up to Xe^{51+} (Li-like) were identified in spectra recorded at electron-beam energies of 106 and 133 keV. Because Xe gas was being injected continually, ions in lower charge states were always present.

The present measurements employed a flat-field spectrometer (FFS) [11]. The spectrometer was equipped with a 1200 lines/mm variable line spaced concave grating [12] and a cryogenically cooled back-thinned charge-coupled device (CCD) camera. The camera chip had 1024×1024 pixels on a square area of about 25 mm each side. The FFS imaged the light from the ion trap, using the 70 μm diameter electron beam [13] as the source, onto the CCD chip where it resulted in the geometrically expected width of about 2 pixels. The total area of the CCD chip was binned by a factor of four in the nondispersive direction to create an effective CCD array of 256×1024 pixels. Due to spectral aberrations, the image of each line was slightly curved at the CCD surface. Simple summing across the dispersion direction would, therefore, result in broadened spectral features. Instead, the files obtained in the typically 20 minutes of exposure time were

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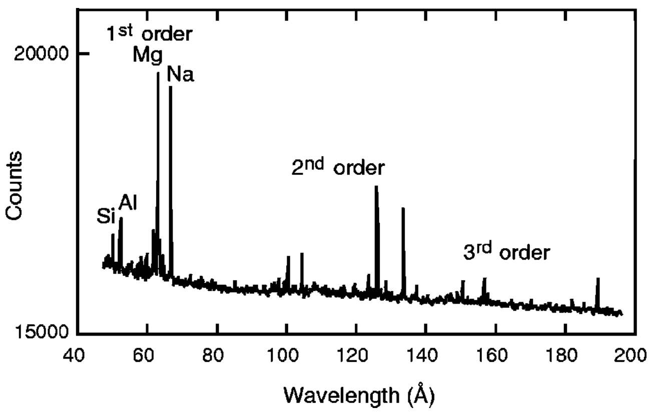

FIG. 1. EUV Spectrum of Xe recorded with a flat-field spectrometer. The data were recorded with 20 keV electron-beam energy. The spectrum shows $3s$-$3p$ lines from Na- and Mg-like ions as the strongest ones, and appearing in several diffraction orders. Al- and Si-like ions are expected to show more lines than the two labeled ones; some are part of the line cluster near 60 Å. Most others are below the C absorption edge and thus do not show in higher diffraction orders either.

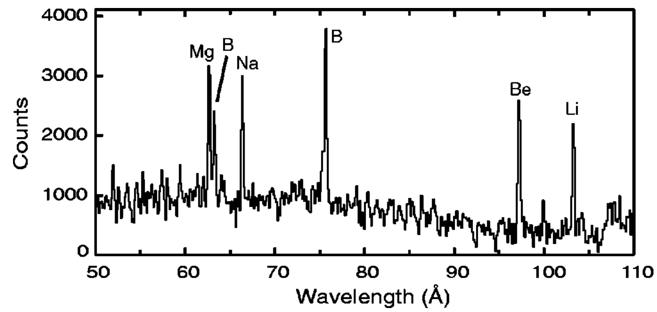

FIG. 2. Spectrum of Xe, restricted to the range of first diffraction order observations of the lines of interest. The spectrum shown was accumulated from more than 40 individual spectra that had been obtained at electron-beam energies between 106 to 133 keV. An experimentally determined background has been subtracted from the data. In addition to the resonance lines of Na- and Mg-like Xe ions, lines from the Li-, Be-, and B-like ions are shown. Any prominent line from ions of charge states in between (C- to Ne-like) would be expected below the short-wavelength end of the recording.

individually filtered for cosmic rays, and then only a central section evaluated.

Calibration was performed by recording spectra when injecting CO_2 or N_2 instead of Xe, determining the central position (in channels) of a number of well-known transitions in H- and He-like ions of C, N, and O, and then fitting a fifth-order polynomial to the calibration data. For reference line wavelengths, we use the calculations of one-electron ions by Garcia and Mack [14], as well as the accurate calculations of He-like ions by Drake [15]. Observation of several orders of diffraction was possible for some of the lines (Fig. 1); this yielded additional line positions for calibrations as shown in Ref. [16]. The calibration spectra alternated every few hours with groups of Xe spectra. Thus any possible shifts would be recognized and corrected for.

The background light emission recorded by the CCD was determined by running EBIT with an ''inverted trap'' [17], that is, with the middle drift tube at a higher potential than the top drift tube, so that no trapping occurred and any ion 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. Examples of the spectra are shown in Figs. 1 and 2.

The peaks were fit with Gaussian functions. The quoted wavelength errors result from a statistical analysis of the scatter of the individual measurements, which is much more significant than the very small statistical uncertainty based on signal statistics. Our line position errors combine (in quadrature) the errors of the calibration and of the reproducibility of the actual measurements. Almost fifty spectra were evaluated to obtain the wavelength results given in Table I.

III. RESULTS AND DISCUSSION

The following discussion is ordered by the increasing number of electrons. Li-like ions have two prominent resonance transitions, $2s$-$2p_{1/2}$ and $2s$-$2p_{3/2}$, while Na-like ions feature $3s$-$3p_{1/2}$ and $3s$-$3p_{3/2}$ transitions. Correspondingly, in Be- (Mg-) like ions there are the $ns^2\,{}^1S_0$-$nsnp\;{}^{1,3}P^o_1$ resonance and intercombination transitions. For elements as heavy as Xe, the fine structure splitting in the one-electron spectra and the electrostatic energy splitting in the two-electron spectra are so large that the wavelengths of these transition pairs differ by roughly a factor of two. For this reason, only one component of each pair shows in our spectra. For Li-like ions, this is the long wavelength component $2s\;{}^2S_{1/2}$-$2p\;{}^2P^o_{1/2}$ (near 103.4 Å), and for Be-like ions the intercombination transition $2s^2\,{}^1S_0$-$2s2p\;{}^3P^o_1$ near 97.4 Å. For Na- and Mg-like ions the selection works out the other way round: we see the $3s\;{}^2S_{1/2}$-$3p\;{}^2P^o_{3/2}$ transition near 66.6 Å and the $3s^2\,{}^1S_0$-$3s3p\;{}^1P^o_1$ resonance line near 62.9 Å. The other lines of each pair are expected between 120 and 130 Å, where they would lie amidst a number of second diffraction order lines. In our spectra they are too weak to be identified.

A. Li-like ions

The wavelength measurements on the resonance lines of many Li-like ions have recently been summarized by Bosselmann *et al.* [18]; the latest measurements, on Xe^{51+}, have been reported by Feili *et al.* [19] who employed one of the largest heavy-ion accelerators (the UNILAC accelerator at GSI Darmstadt) and an $R=5$ m grazing incidence spectrometer with a position-sensitive microchannel plate detector. The spectrum observed from the fast-ion beam was calibrated with a stationary light source, requiring elaborate procedures to determine the Doppler corrections with sufficient reliability. Our present measurement yields a wavelength value of 103.483 ± 0.010 Å. This is slightly less precise than the result (103.475 ± 0.007 Å) given by Feili *et al.*, but it is in excellent agreement with that value. Our result thus similarly tests the QED contribution at a level of better than 0.2%. Since our result has been obtained with a stationary

TABLE I. Predicted and measured wavelengths of various transitions in the few-electron spectra of Xe. All values are given in units of angstroms.

Ion charge	Isoelectronic sequence	Transition	Theory	Wavelength Semiempirical	Experiment
51+	Li	$2s\ ^2S_{1/2}$-$2p\ ^2S_{1/2}$	103.806 [a]		103.35±0.085 [b]
			103.2 [c]		103.475±0.007 [d]
			103.476 [e]		103.483±0.010 [f]
50+	Be	$2s^2\ ^1S_0$-$2s2p\ ^3P_1^o$	97.74 [c]		97.9±0.4 [g]
			97.4205 [h]		97.426±0.020 [i]
			97.3945 [j]		97.430±0.009 [f]
49+	B	$2s^2\,2p\ ^2P^o_{1/2}$-$2s2p^2\ ^4P_{1/2}$	77.29 [c]		75.868±0.009 [f]
49+	B	$2s2p^2\ ^4P_{1/2}$-$2s^22p\ ^2P^o_{3/2}$	63.34 [c]		63.474±0.020 [f]
43+	Na	$3s\ ^2S_{1/2}$-$3p\ ^2P_{3/2}$	66.146 [k]	66.623 [l]	66.58±0.03 [m]
			66.632 [e]		66.574±0.020 [f]
42+	Mg	$3s^2\ ^1S_0$-$3s3p\ ^1P_1^o$	62.19 [n]	62.8947 [o]	62.875±0.012 [f]
			62.66 [p]		
			62.447 [q]		

[a]Johnson *et al.* [1], with Lamb shift from Ref. [54].
[b]Martin *et al.* [20].
[c]Cheng *et al.* [26].
[d]Feili *et al.* [19].
[e]Kim *et al.* [3].
[f]This work.
[g]Möller *et al.* [31].
[h]Safronova *et al.* [27].
[i]Feili *et al.* [32].
[j]Chen and Cheng [28].
[k]Johnson *et al.* [4].
[l]Reader *et al.* [44].
[m]Seely *et al.* [48].
[n]Cheng and Johnson [52].
[o]Ekberg *et al.* [46].
[p]Ivanova *et al.* [53].
[q]Zou and Froese Fischer [60].

light source, the complexity of our spectroscopic effort can be much lower, while the sources of systematic error are notably different. The precision was reached even with our much smaller experimental setup: using diffraction gratings of similar radius of curvature as ours, the larger GSI instrument working in second and third order of diffraction had geometrical advantages. Our wavelength result is, however, clearly more precise than the fast-ion beam value reported by Martin *et al.* [20] and is, corroborated by the closeness of our result to the Feili value, also more accurate. Among the calculational values listed in Table I, the predictions by Kim *et al.* [3] match our result best. The latest QED evaluations have been listed in the report by Feili *et al.* [19] and need not be repeated here.

B. Be-like ions

The low-lying energy levels of Be-like ions have been measured and systematized repeatedly, as data for progressively heavier ions (up to Mo, $Z=42$) have become available from the observation on magnetic fusion devices, such as tokamaks [21–25], in addition to calculations, many of which, unfortunately, covered only the low-Z range of the isoelectronic sequence (among others, see Refs. [26–29]). In the Be-like ion Xe^{50+}, the $2s^2\ ^1S_0$-$2s2p\ ^3P_1^o$ intercombination transition gives rise to a line near the $2s$-$2p_{1/2}$ resonance line of the Li-like ion. The same intercombination line appeared in the beam-foil spectrum of Xe obtained by Martin *et al.* [20], but no precise value of the wavelength was given. The beam-foil study by Möller *et al.* [30,31] comprised no wavelength calibration effort and instead relied on predicted wavelength values to find the line, and then measured the decay rate of the upper level. Feili *et al.* [19] also observed the line, but opted not to present it in that publication, because in the stationary calibration light source they had no good reference lines that would frame this particular line. Subsequently a wavelength value of 97.426±0.020 Å was reported informally [32]. The present work provides a measured wavelength value for this transition, of 97.430 ±0.009 Å, that is twice as precise. Among the calculations, the latest ones, a many-body perturbation theory treatment by Safronova *et al.* [27] and a relativistic configuration interaction approach by Chen and Cheng [28] come closest to our experimental result (Fig. 3). However, we note that the calculation by Safronova *et al.* does not agree well with the experimental findings for low-Z ions, and that experimental data and calculation again disagree around $Z=30$. The calculations by Chen and Cheng agree reasonably well with the experimental data up to $Z=32$, but then different trends evolve. Clearly, both more experimental data in the mid-Z range and calculations that do not report on just a few elements, but give continuous coverage of many elements, would be valuable to help clarify the situation.

C. B-like ions

B-like ions contribute two lines to the present spectra. These lines might appear as a curiosity (earlier discussed by

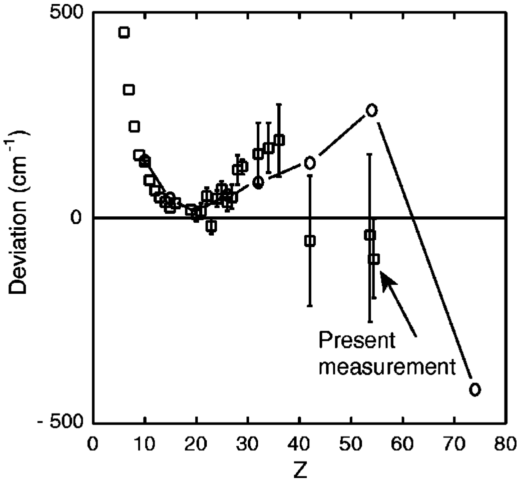

FIG. 3. Comparison of experimental data and calculations for mid-*Z* ions of the Be isoelectronic sequence. The level position of the $2s2p\ ^3P_1^o$ level, relative to the calculation by Safronova *et al.* [27]. ○, calculational results by Chen and Cheng [28] (connected with a line to guide the eye); □, experimental results (for references, see text). For low nuclear charges *Z*, the experimental error bars are smaller than the symbol size.

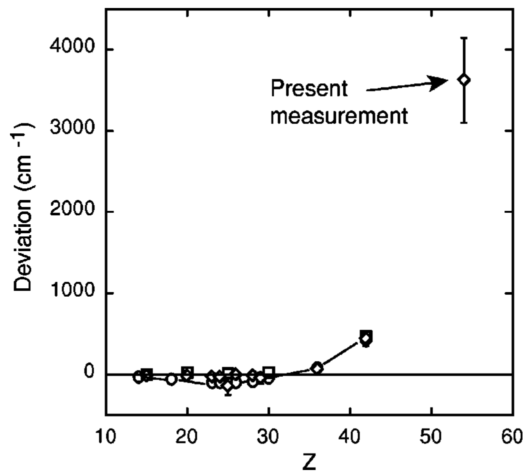

FIG. 4. Comparison of experimental data and calculations for mid-*Z* ions of the B isoelectronic sequence. The level position of the $2s2p^2\ ^2P_{3/2}^o$ level, relative to the calculation by Safronova *et al.* [38]. ○, calculational results by Huang *et al.* [36] (connected by an eye-guiding line); □, calculations by Vilkas *et al.* [39]; ◇, experimental results (for references, see text). For low nuclear charges *Z*, the experimental error bars are smaller than the symbol size.

Möller *et al.* [31]) when considering the B-like spectra of low-Z ions. There, the $M1$ transition (with a weak $E2$ contribution) within the ground term is the prominent transition that appears in many low-density plasma light sources (including tokamaks and the solar corona [21]) and that has been systematized over a wide range of elements by Edlén [24,33,34] and by Myrnäs *et al.* [35]. Recent calculations by various techniques have covered different parts of the isoelectronic sequence [26,36–41]. A transition between the levels of the ground term of B-like Xe ions corresponds to a wavelength of about 35 Å, which is below the range of our present spectra. In a heavy ion such as Xe, however, the "fine-structure" intervals of the $2s^2\,2p\ ^2P^o$ ground term and the $2s2p^2\ ^4P$ excited term (scaling as Z^4) are of a similar size as the electrostatic $2s$-$2p$ interval (which scales as Z^1), and the lowest of the $2s2p^2\ ^4P_J$ levels ($J=1/2$) then happens to lie lower than the upper one ($J=3/2$) of the ground term levels. Thus a considerable part of the $2s^22p\ ^2P_{3/2}^o$ level population (theory predicts about 30% [26]) decays toward the $2s2p^2\ ^4P_{1/2}$ level [an *E1* spin-changing (intercombination) transition] and from there (by another intercombination transition) to the true $2s^22p\ ^2P_{1/2}^o$ ground level. Both lines appear in our spectra, and the fact that the one line (at 63.474 Å) replenishes the upper level of the other (at 75.868 Å) immediately renders the second line the stronger one and thus facilitates identification. The combination of both transition energies yields the position of the upper doublet level ($2\,893\,527\pm53$ cm^{-1}, corresponding to a ground-state decay wavelength of 34.56 Å) and thus provides for a long-range extension to Edlén's energy systematization of the ground term intervals in the B-like ions [24,33–35].

A comparison of the experimental data to the three calculations [36,38,39] that extend their predictions notably beyond $Z=30$ (where Ref. [36] is seen as superceding its precursor [26]) is shown in Fig. 4. Evidently these calculations largely agree with each other and with the experimental findings up to about Kr ($Z=36$). For Mo ($Z=42$), experiment and the calculational result obtained by Huang *et al.* [36] and by Vilkas *et al.* [39] agree with each other, but deviate markedly from the prediction by Safronova *et al.* [38]. The present measurement for Xe ($Z=54$) lies much farther away from that prediction, but in the same direction as suggested by the Mo data. This seems to indicate significant shortcomings of the calculation by Safronova *et al.* [38] that become notable near $Z=40$ and rapidly aggravate for heavier ions. The oldest of the calculations mentioned, by Cheng *et al.* [26], also extend into this range of but are of little guidance value, since their predictions are far off scale (some 20 000 cm^{-1} from the experimental data point for Xe^{49+}) in Fig. 4.

The principal emission lines in the next lower charge state ions (C- to Ne-like ions) lie in the wavelength range below 50 Å and are therefore outside our present detection range. Some lines should lie in our range of view, but are expected to be weak because of unfavorable branching ratios.

D. Na and Mg-like ions

In an electron-beam ion trap, the charge state distribution can be shifted so as to comprise only a few charge states near a closed-shell system. In low-*Z* systems, for example, practically all ions but B-, Be-, and Li-like can be burned out, while not yet providing enough energy to excite He-like ions. As mentioned before, Xe was bled into SuperEBIT continually. This led to the presence of a tail of lower charge state ions that may radiate upon excitation or ionization by the electron beam. Consequently, we see in our high-electron beam energy spectra (Fig. 2) lines from Na- and Mg-like Xe ions (Fig. 1). Moreover, there also are a few lines that we can identify from predictions [42,43] with transitions in Al- and Si-like Xe ions. Excitation cross sections of these $n=3$ shell ions are much larger than those of n=2 shell ions (e.g., Li- and Be-like ions). The observed line intensities are of the same order of magnitude; this implies that there are much fewer $n=3$ shell ions than of $n=2$ shell ions in the trap.

Xe is easily added to tokamak plasmas, but tokamak data on Na-like or Mg-like ions of Xe seem not to be available. Xe as a gas is not easily used in laser-produced plasmas. Therefore Reader *et al.* [44] extrapolated their data of lower-Z elements to obtain a value for Xe^{43+}, and Ekberg *et al.* [45,46] interpolated their other measurements to derive a datum for Xe^{42+}. Only Seely *et al.* [47,48] give actual results of laser-produced plasma observations on Xe^{43+}. Their wavelength results for Xe are compatible with the extrapolated trend of lower-Z observations from tokamaks and of the best calculations (see, for example Refs. [2,3]). Several $n=3$, $\Delta n=0$ transitions in Na-, Mg-, and Al-like ions of Xe have been seen in spectra of foil-excited fast-ion beams [49,50].

Our wavelength result for the $3s$-$3p_{3/2}$ transition in the Na-like ion agrees with the value given by Seely *et al.* [48], but has a smaller uncertainty. Two weak lines near 84.85 Å and 58.24 Å coincide with the positions of the $3p_{3/2}$-$3d_{5/2}$ and $3p_{1/2}$-$3d_{3/2}$ transitions in Na-like Xe^{43+} ions [48]. In Fig. 1, only one each of the lines that arise from Al- and Si-like ions have been labeled. More (weaker) lines of the same ions cluster around the resonance line in the Mg-like ion. The observed wavelengths lie within 0.4 to 0.7 Å of the predictions by Huang [42,43]. However, given the low line intensities, we refrain from giving experimental wavelength values to those weak lines before the observations can be improved.

Our result for the Mg-like ion differs by about 500 cm^{-1} from the fitted trend presented by Ekberg *et al.* on the basis of their measurements for several ions in the neighborhood [46] (Fig. 5). The difference is just about compatible with the combined error bars of our direct measurement and of the measurements of other elements that Ekberg *et al.* base their interpolation for Xe on. The slight discrepancy between the experimental results, however, is much smaller than the deviation between the data and any of the theoretical predictions. Quite a number of predictions are available for Mg-like ions, of which we have evaluated Refs. [29,52,53,55–61]. Chen and Cheng [55] present a detailed discussion of various theoretical approaches in the low to mid-Z range. Unfortunately, most of these calculations treat only a short section of the isoelectronic sequence, or only a few ions within a wider range, resulting in little overlap with experimentally covered elements in the mid-Z range. On the basis of elemental coverage, the calculation by Ivanova *et al.* [53] seemed most suitable as a normalizing reference for a display of the competing calculations (Fig. 5).

The calculations by Chen and Cheng [55] and by Safronova *et al.* [56] come rather close to the experimental findings, but they do not extend beyond Mo $(Z=42)$. An apparent kink in the trends of both calculations as well as of the experimental data near $Z=31$ to 33 (Fig. 5) probably is

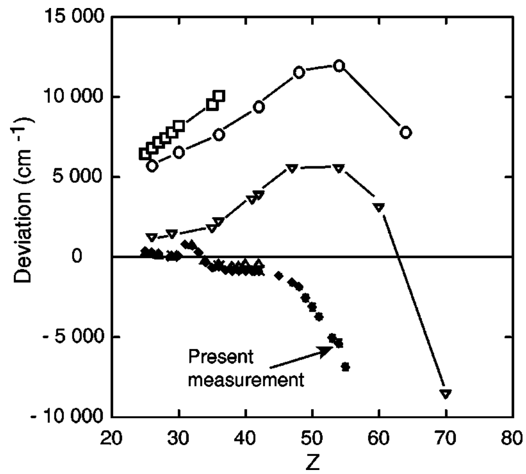

FIG. 5. Comparison of experimental data and calculations for mid-Z ions of the Mg isoelectronic sequence. The level position of the $3s3p\ ^1P_1^o$ level, relative to the calculation by Ivanova *et al.* [53]. ○, calculational results by Cheng and Johnson [52]; □, calculation by Huang *et al.* [51]; ▽, calculation by Zou and Froese Fischer [60]; X, calculations by Chen and Cheng [55]; △, show calculation by Safronova *et al.* [56]. Some of the calculational results are connected by eye-guiding lines. Filled circles are for experimental results (for references, see text). With the relatively large differences between measurement and theory, the experimental error bars are all smaller than the symbol size. The kinks (near $Z=32$) in the trends of the experimental data and of some calculational values results are probably artifacts induced by the normalization.

an artifact caused by the reference to the calculations by Ivanova *et al.* [53]. The calculations by Ivanova *et al.* [53] deviate progressively from experiment for increasing Z. For nuclear charges up to about 42 (where the difference is less than 1000 cm^{-1}), the deviation grows slowly, and then more rapidly, reaching more than 5000 cm^{-1} for Xe $(Z=54)$. Other wide-range calculations show a similar change of trend halfway through the periodic table; however, the results of the early calculations by Cheng and Johnson [52] and of the very recent calculations by Zou and Froese Fischer [60] are 17 000 cm^{-1} and 11 000 cm^{-1}, respectively, away from experiment for Xe.

Some of the calculations do not take QED corrections into account. However, considering the magnitude and the Z-dependence of the mismatch between measurement and calculation, it seems unlikely that the lack of a proper QED treatment is the only shortcoming. Rather, some imperfection in the description of the interaction among the valence-shell electrons seems to be more probable. Clearly, the calculational situation for Mg-like ions leaves much to be desired.

IV. CONCLUSION

Our wavelength measurement for the Li-like ions Xe^{51+} agrees well with the slightly more precise result from an accelerator experiment. Our results for the Be-like Xe^{50+}

ions, B-like Xe^{49+} ions, Na-like Xe^{43+} ions, and Mg-like Xe^{42+} ions are the most precise ones available. They provide the highest Z data for Be- and B-like ions and are among the highest *Z* for Mg-like ions. They reveal significant shortcomings in the calculations of all ions with more than one electron in the open valence shell. We hope that these results will spur the development of more accurate theoretical approaches for the treatment of such relatively simple multi-electron systems.

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