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# Europium (Z=63) n=3-3 lines in the extreme ultraviolet: Na- through Si-like ions

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## Abstract

Using the SuperEBIT electron beam ion trap and two high-resolution flat-field spectrographs, we have observed EUV spectra of highly charged ions of Eu and measured the wavelengths of prominent lines from Na-, Mg-, Al-, and Si-like ions with better than 100 ppm precision, the highest of any such ions with atomic number between $Z$=38 and $Z$=92. Our result for Na-like ions compares well with the trend set by two other measurements with comparable accuracy performed at much higher nuclear charge ($Z$=78, 92), affirming the accuracy of *ab initio* calculations. It, however, disagrees with the trend set by a measurement of the neighboring element Gd ($Z$=64) obtained from laser-produced plasmas that had favored different theoretical approaches. There appear to be no similarly accurate multi-element calculations for the Mg-, Al-, and Si-like ions of Eu.

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## I. INTRODUCTION

Basic atomic physics interest has long focused on the spectra of ions with a single electron (H-like ions), because they can be calculated most accurately. 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, because these cannot only be calculated [1–3], but also measured rather accurately (see [4–11] and references therein). In many observations of hot plasmas and other light sources the resonance lines of alkali-like (and alkali earth -like) ions are seen brightly and hence should be measurable with high precision. One of the major driving factors of precision measurements of the wavelengths of such spectral lines 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, which can be measured with comparable accuracy. Here, however, computation is challenged with additional electrons, be they in the valence (outer) shells (Mg-, Al-, Si-like ions etc.) or, to a lesser extent, by electrons in additional closed inner shells (Li-, Na-, Cu-like ions).

Highly charged ions can be calculated best, because the central potential is best defined. Fully relativistic calculations are necessary [12], and they converge well for highly charged ions. However, the experimental data base is best for low to moderate charge states, as a result of the historical technical development of light sources and spectroscopic equipment. When high-$Z$ ions came into reach (by laser-produced plasmas, heavy-ion accelerators, and electron beam ion traps (EBIT)), the competition aimed for the highest values of the nuclear charge $Z$. This goal, with the largest QED contributions, has been reached for various isoelectronic sequences. Because of the effort required, there are not many such very-high $Z$ measurements, and there are wide gaps in the middle of the periodic table of elements, where the QED effects are smaller, but so are the uncertainties in the calculations that are due to nuclear parameters. Data are rich up to nickel ($Z$=28) and then taper off rapidly towards Xe ($Z$=54). There are only few data available between Mo ($Z$=42, formerly of fusion reactor vessel interest) and W ($Z$=74, of present fusion reactor vessel interest), and many of them are not very accurate.

The theoretical predictions of the resonance transition energy for Na-like U ($Z$=92) span a band of 6000 $cm^{-1}$, and differ by about 1000 $cm^{-1}$ for elements with $Z\approx60$. The best

calculations of transition energies in heavy, one valence electron ions (i. e., one electron outside of a closed shell) ions have been found to differ from good experiments by no more than some 100 to 200 ppm, and the first calculations have reached a comparable accuracy also for two-electron systems [13–16]. Electron beam ion traps are particularly well suited for such measurements, because the ion cloud in the trap is stationary and thus does not cause Doppler shifts as do expanding laser-produced plasmas or fast ion beams. The low density (ultrahigh vacuum) in electron beam ion traps leads to little collisional excitation, and thus the level populations are almost exclusively in the ground configurations, and consequently ground state transitions are by far the brightest lines. The NIST EBIT group has published transition energy data on Na-, Mg-, Al- and Si-like ions of several elements in the range $Z$=72-79, but with uncertainties on the order of 1000 ppm the measurements [9] are of limited use. Moreover, the NIST EBIT group results scatter by about 10000 cm$^{-1}$ relative to the theory trend and thus add no new information, as these data cannot distinguish among any theory. Seely *et al.* [17] have extended laser measurements up to Gd ($Z$=64), achieving a precision of 1300 cm$^{-1}$. Träbert *et al.* [7] and Clementson and Beiersdorfer [10] have measured transitions of xenon and tungsten ions at the LLNL electron beam ion trap, and their data are more precise by a factor of about three than the NIST EBIT data.

We have now measured corresponding data of yet higher spectral resolution for Eu ($Z$=63), achieving an uncertainty of 200 cm$^{-1}$, which is more than a factor of six lower than that of the Seely datum for the neighboring element Gd. This new measurement joins the best of the earlier data (for U [6], Pt ($Z$=78) [4], and W ($Z$=74) [10]) and forms a data set that is internally consistent and compatible with the trend of the best calculations. Our measurements extend the same data quality to key lines in the spectra of Mg-, Al-, and Si-like ions of Eu, for which theory is less well developed so far.

## II. EXPERIMENT

The experiment was done at the Lawrence Livermore National Laboratory EBIT facility [18]. Of the laboratory's two electron beam ion traps, the higher-energy device, SuperEBIT [19, 20], was employed. Eu was injected into SuperEBIT as atomic vapor, making use of its high vapor pressure when heated to several hundred degrees Celsius via a tungsten filament [21]. 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, negative 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 Na-like ions of Eu is about 4.7 keV, while the production of Mg-like ions requires 4.5 keV [22].

In our experiment the electron beam energies were varied from about 2 keV to 40 keV in order to explore a wide range of charge states and to identify the prominent lines of interest. These energies are appropriate for measurements of Cu- through Ne-like ions, but not all of these have prominent lines in the wavelength range our spectrographs were set to. For the sake of signal statistics, we co-added groups of spectra that were recorded at low electron beam energies (below 2.5 keV), at 2.6 to 3 keV, at 3.1 to 3.8 keV, and at 4 keV or higher. (The spectra at the lower electron beam energies will be presented elsewhere [23].) The relative intensities of lines in such spectra relate to the ionization potentials of the ions in the trap, which helps with the identification of likely charge states. The highest charge state identified from spectral lines was Na-like $Eu^{52+}$. Because Eu vapor was being injected continually, ions in lower charge states were always present.

The present measurements employed two flat-field spectrographs [24, 25], each equipped with a 2400 $\ell$/mm variable line spaced concave grating and a cryogenically cooled back-thinned CCD camera. The camera chip had 1340 x 1300 pixels of 20 $\mu$m $\times$ 20 $\mu$m each. The concave gratings imaged the light from the ion trap, using the $\leq$60 $\mu$m diameter electron beam [26] as the source, onto the CCD chip where it resulted in a width of about 3 pixels expected from the source size and the prevalent temperature in SuperEBIT [27, 28]. The files obtained in the typically 60 minutes of exposure time were individually filtered for cosmic rays. Calibration was performed by recording spectra when injecting $CO_2$, the resonance line series of C V and the C VI $Ly_\alpha$ line spanning almost the full width of the camera, which covered the wavelength range 32 to 43 Å (see the full spectrum in figure 1 top). The carbon reference lines comprised C VI $Ly_\alpha$ at 33.734 Å and the C V lines $1s^2$ - 1snp (n=3 to 5) at 32 to 35 Å in the lower wavelength half and the C V $1s^2$ - 1s2p 'w' and 'y' lines at 40.267 and 40.730 Å, respectively, in the upper half. The wavelengths of these lines are known to better than 1 mÅ from calculations [29, 30] and were readily employed to determine a slightly quadratic calibration curve.

Because the count rate was sufficiently low, we could detect individual photons in the CCD image. As a result, we applied a filter routine that suitably selected individual counts while discriminating against background and cosmic rays. The peaks in the resulting spectra were fit with Gaussian functions. The quoted wavelength errors result from a statistical analysis of the data, of the calibration, and of the scatter of the individual measurements. The line width (FWHM) of one instrument was about 24 mÅ, of the other about 32 mÅ, the difference arising from the quality of focusing the instrument. We note that several of the carbon lines persisted even after the $CO_2$ injection ended.

## III. RESULTS AND DISCUSSION

The lines shown in Fig. 1 are readily identified as 3s - $3p_{3/2}$ transitions in Na- and Mg-like ions as well as n=3 to n=3 transitions in Al- and Si-like ions of Eu. In addition we see several carbon reference lines. All of the strong Eu lines visible in Fig. 1 are absent at electron beam energies below 4 keV and are replaced by emission lines from lower charge state ions. There, a multitude of charge states differ by only small amounts in ionization potential. At the electron beam energy settings as low as 2.2 keV, lower charge states down to Cu-like ions are the highest that can be produced, and the spectrum accordingly changes drastically. Systematic, small-step variations of the electron beam energy have been used with other elements such as Fe [31] to identify the likely charge state of origin for each spectral line. This was not feasible here, and we leave the lower charge state ion spectra until suitable calculations become available.

We interpret the line pattern in the spectra observed at higher electron beam energies with the help of insight from our earlier EBIT studies of xenon (Xe) and tungsten (W) that used one of the present spectrographs [7, 10]. Actually, the appropriate section of the Xe spectrum has played only a minor role in our 2003 Xe publication [7], but it was shown as an example by Vilkas *et al.* in 2006 [32] who demonstrated the high accuracy of their Multi-Reference Møller-Plesset calculations. The n=3 to n'=3 transition energies are expected to scale linearly with the nuclear charge $Z$ and the ion core charge $\zeta = Z - m + 1$ ($m$ being the number of electrons) seen by the valence electron; hence our analysis represents an interpolation process between our earlier observations of Xe and W. A summary of our line identifications and of the measured wavelengths is given in Table I.

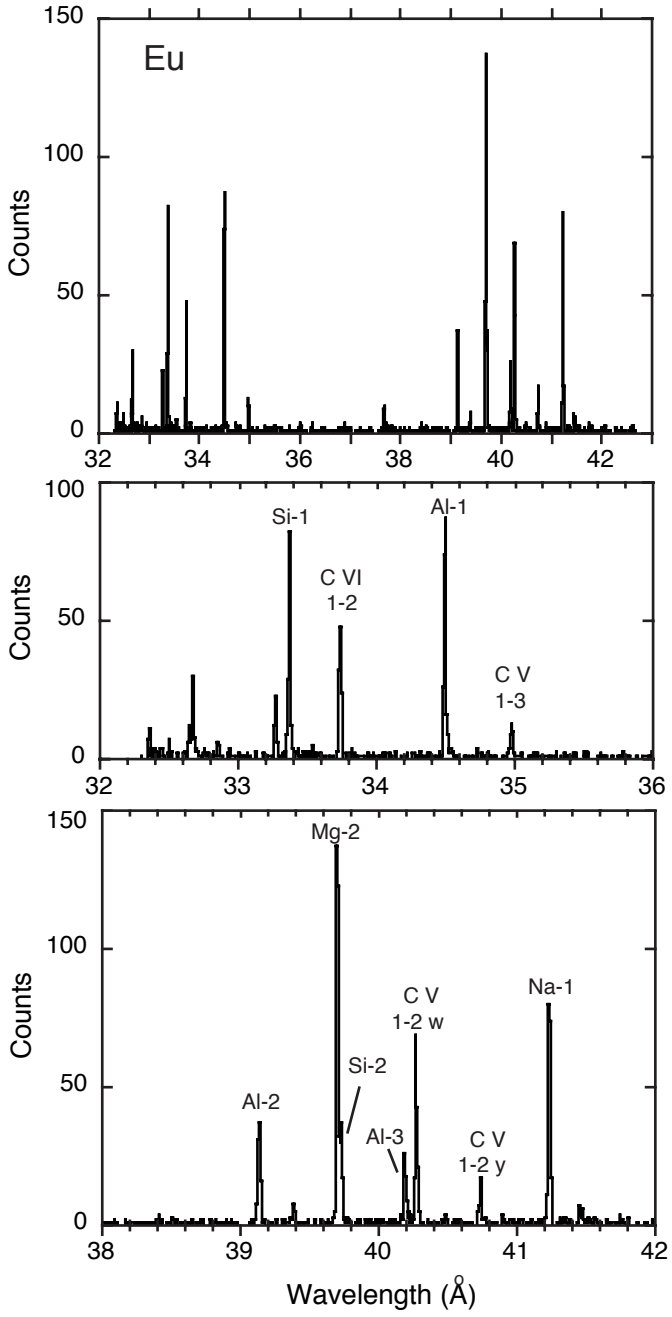


FIG. 1: EUV spectrum of Eu recorded with a flat-field spectrometer at an electron beam energy of about 8 keV. The spectrum shows n=3 to n=3 lines from Na-, Mg-, Al-, and Si-like ions as the strongest ones of europium, as well as calibration lines of H- and He-like ions of carbon. Top: full range of a typical exposure, middle and bottom: parts of the same spectrum with labels that are explained in Table I.

### A. Na-like

The 3s $^2S_{1/2}$ - 3p $^2P^o_{3/2}$ transition in $Eu^{52+}$ (Na-like ion) appears at 41.232 Å close to the position of 41.228 Å predicted decades ago by Kim *et al.* [2] (see Table I and Fig. 2). The difference is about 100 ppm and exceeds the experimental error bar only slightly. The calulations by Seely *et al.* [17] are clearly poorer, and the semiempirical correction proposed by Seely *et al.* on the basis of their laser-produced plasma experiments makes agreement with experiment worse. The isoelectronic trend derived from those laser experiments has been discredited by previous observations, and our measurement also disagrees with that trend. In fact, our measurement of Eu is only one atomic number away from Gd measured on the NOVA laser [17]; yet our data corroborate a very different trend. Figure 2 shows the isoelectronic trends of measurements and calculations.

The *ab initio* calculations by Blundell [3] are in the same accuracy class as those by Kim *et al.* and diverge only for elements well above europium. Unfortunately, Blundell's calculations do not explicitly cover europium. The older calculations by Ivanov and Ivanova [33] are close to experiment at low $Z$, but the results diverge from experiment at higher atomic numbers and are so far away from our experiment that they cannot be included in the plot shown in Fig. 2. Overall, the experimental data with the smaller error bars (with the exception of the range $Z = 53 - 55$ in which data from various light sources and measurements cluster in a separate location that suggests a common systematic error, perhaps an unrecognized spectral blend) agree best with the calculations by Blundell who describes the high-$Z$ trend better than Kim *et al.* Incidentally, the recent NIST EBIT data [9] are claimed to favor the Blundell calculations over the Kim *et al.* ones. Considering the scatter and the error bars of the NIST EBIT data in Figure 2, however, their 3s-$3p_{3/2}$ transition data encompass all of these predictions without any clear discrimination. In contrast, the LLNL EBIT observations for Eu (this work), W [10], Pt [4], and U [6] all have much smaller uncertainties and clearly exclude the predictions by Seely *et al.* The uranium datum from Livermore is the only one of the present sample of Na isoelectronic sequence data that indeed clearly points to the calculations by Blundell as the best of the pack, because only at the highest $Z$ values the difference between Blundell and Kim *et al.* is notable.

TABLE I: Measured (this work) and calculated wavelengths (in Å) of $n$=3 to $n$=3 transitions in the EUV spectra of Na- through Si-like ions of Eu. The line numbering follows the example of Clementson and Beiersdorfer for W [10].

| Ion charge | Isoelectronic sequence | Line label | Transition | Wavelength Experiment | Wavelength Theory |
|---|---|---|---|---|---|
| 52+ | Na | Na-1 | $3s_{1/2}$ - $3p_{3/2}$ | 41.232 ± 0.003 | 41.228 [a] |
| | | | | | 41.182 [b] |
| | | | | | 41.211 [c] |
| | | | | | 40.900 [d] |
| 51+ | Mg | Mg-2 | $(3s^2)_0$ - $(3s_{1/2}3p_{3/2})_1$ | 39.698 ±0.003 | 39.622 [e] |
| | | | | | 39.372 [f] |
| | | | | | 39.694 [g] |
| 50+ | Al | Al-1 | $(3s^23p_{1/2})_{1/2}$ - $(3s^23d_{3/2})_{3/2}$ | 34.493 ± 0.003 | 34.411 [h] |
| 50+ | Al | Al-2 | $(3s^23p_{1/2})_{1/2}$ - $(3s_{1/2}3p_{1/2}3p_{3/2})_{1/2}$ | 39.133 ± 0.003 | 38.860 [h] |
| 50+ | Al | Al-3 | $(3s^23p_{1/2})_{1/2}$ - $(3s_{1/2}3p_{1/2}3p_{3/2})_{3/2}$ | 40.186 ± 0.003 | 39.991 [h] |
| 49+ | Si | Si-1 | $(3s^23p^2_{1/2})_0$ - $(3s^23p_{1/2}3d_{3/2})_1$ | 33.368 ± 0.003 | 33.280 [i] |
| 49+ | Si | Si-2 | $(3s^23p^2_{1/2})_0$ - $(3s_{1/2}3p^2_{1/2}3p_{3/2})_1$ | 39.726 ± 0.003 | 39.524 [i] |

[a] Kim *et al.* [2]
[b] Seely *et al.* [17]
[c] Seely *et al.* prediction with semiempirical correction [17]
[d] Ivanov and Ivanova *et al.* [33]
[e] Marques *et al.* [34]
[f] Ivanova *et al.* [35]
[g] Santana [36]
[h] Huang [37] with different identifications
[i] Huang [38] with different identifications

### B. Mg-like

Ekberg *et al.* [39] have reviewed the experimental work on Mg-like ions up to the middle of the periodic table. There are various calculations of Mg-like ions [34, 35, 40–49], but most do not reach as high as $Z$=63 or they do not include europium. For the $ns^2\ ^1S_0$ - $nsnp\ ^1P^o_1$ resonance transition in the Mg-like ion $Eu^{51+}$ there is no such reliable literature prediction

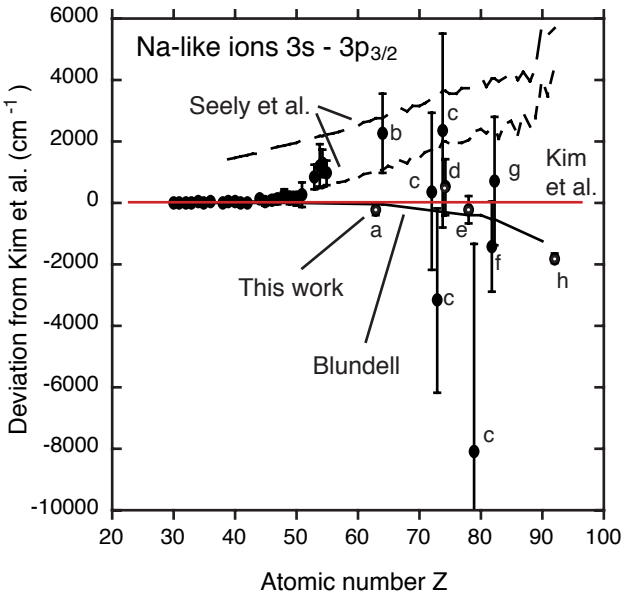


FIG. 2: (Color online only) Isoelectronic plot of the 3s-3$p_{3/2}$ transition energy in Na-like ions (from $Z = 30$ to 92). The low-$Z$ data points are from a compilation by Reader *et al.* [50]. The data are shown by their deviation from the calculations by Kim *et al.* [2] (horizontal red line). Seely *et al.* [17] give *ab initio* and semiempirically adjusted calculational results. Blundell's calculations [3] agree best with experiment, but cover only a few elements. Experiment (full circles with error bars), except for LLNL EBIT data (open circles): a) This work, b) [17], c) [9], d) [10], e) [4], f) [51], g) [52], h) [6].

of the wavelength (observed at 39.698 Å, see Table I) as for the Na-like spectra. Figure 3 shows the isoelectronic trends of measurements and of some of the calculations. The two computations that deviate drastically from the range of the experimental data are those by Cheng and Johnson [40] and by Ivanova *et al.* [35]. The primary reason for the disagreement with experiment is the neglect of radiative corrections (QED) in both computations, an effect that can be seen in the computations by Zou and Froese Fischer [47] who present results obtained with and without QED. Evidently QED is a sizable effect in high-$Z$ few-electron atomic systems, and accurate spectroscopic measurements are very sensitive to the QED contributions to atomic structure. Among the many-element calculations of ions along the isoelectronic sequence, the two best appear to be those by Zou and Froese Fischer (with QED contributions) [47] and by Marques *et al.* [34]. Neither calculation aimed primarily at the transition energies of present interest. The calculations by Zou and Froese Fischer match

within about 500 $cm^{-1}$ the trend of the low-$Z$ experimental data, but appear to deviate from the experimental trend at high $Z$ (by about 2000 $cm^{-1}$ for $Z$=63). Unfortunately, this calculation treats only a few high-$Z$ elements, and Eu is not among the selected few. The calculations by Marques *et al.* in contrast do not match the experimental trend, but over a wide range they show a constant offset on the order of 5000 $cm^{-1}$. Their predicted wavelength for Eu is 39.622 Å. Hence the difference between their computation and experiment amounts to 2000 ppm, which is more than an order of magnitude worse than the state of the art for Na-like ions. After our experiment, and without knowledge of the outcome, J. A. Santana has computed transition energies in $Eu^{51+}$ using the Multi-Reference Møller-Plesset approach [36]. The calculated resonance wavelength result differs by little more than the experimental uncertainty from our measurement. That deviation by about 120 ppm corresponds to an improvement of the computational accuracy by a factor of six over the prediction by Zou and Froese Fischer, and by a factor of 15 over the prediction by Marques *et al.* A systematic study of Mg-like ions is under way.

### C. Al-like, Si-like

Na- and Mg-like ions each feature a single prominent line in this wavelength range; due to the large fine structure splitting of the 3p terms, the corresponding 3s-$3p_{1/2}$ transitions have a wavelength that is about three times as long as the 3s-$3p_{3/2}$ transitions. The ions with more than two electrons in the n=3 shell feature many more transitions. According to the calculations by Huang [37, 38, 53, 54], there are several dozen lines each of Al-, Si-, P-, and S-like ions of Eu in the range of our observations. This suggests that the individual lines are much weaker than the resonance lines in the Na- and Mg-like ions. Moreover, with imperfect calculations, the many predicted lines cannot easily be identified with the numerous observed lines. In addition to accurate wavelength predictions a useful calculational analysis ought to include collisional-radiative modeling to find out which transitions are likely the brightest under the conditions in a given light source. Such an effort is beyond the scope of this study. Relying on the line pattern established for Xe [7] and W [10], it is possible to identify the corresponding strongest lines in Al- and Si-like Eu (for the results see Table I). However, the comparison with calculations is problematic. We note that at face value at least some of the transitions in Al- and Si-like Eu agree closely enough between our observations (guided by

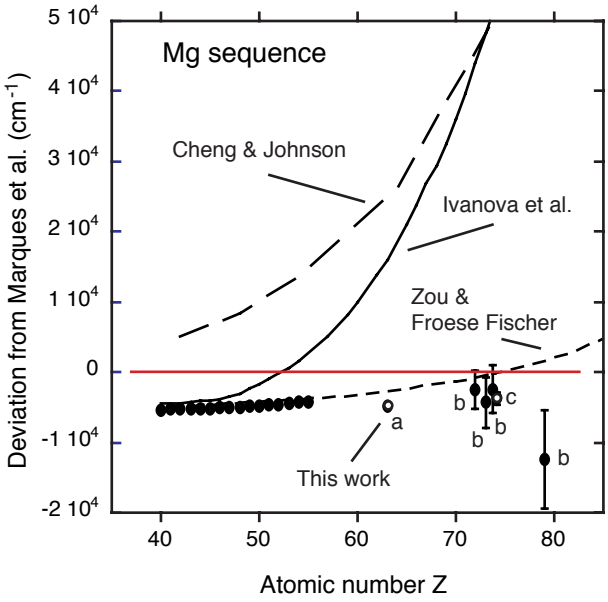


FIG. 3: (Color online only) Comparison of experimental data and calculations from $Z = 40$ upwards for ions of the Mg isoelectronic sequence. Depicted is the level position of the 3s3p $^3P^o_1$ level, relative to the calculation by Marques *et al.* [34] (horizontal red line). The other calculations shown are by Cheng and Johnson [40], by Zou and Froese Fischer [47], and by Ivanova *et al.* [35]. Experimental data (full circles with error bars) at high $Z$: a) This work, b) [9], c) [10]. For low nuclear charges Z, the experimental error bars are smaller than the symbol size.

the FAC and GRASP2 computations by Clementson and Beiersdorfer for W [10]) and the computations by Huang [37, 38] to recognize the lines in the measured spectra, while others grossly disagree. However, if one uses the transition wavelength as an identifier instead of the electron configurations, it is straightforward to find matches for all of the strongest three lines in Al-like Eu and for the strongest two lines in Si-like Eu; evidently the electron configurations are not consistently described by the various atomic structure programs. The mismatch between wavelength measurement and calculation amounts to 2000 to 5000 ppm for the individual lines. There are more calculations on Al- and Si-like ions than the three decades old ones by Huang (for example, [55, 56]), but apparently none that covers europium or many others of the rare earth elements, nor many high-$Z$ elements.

## IV. CONCLUSION

In conclusion, our results for the Na-like $Eu^{52+}$ ions and Mg-like $Eu^{51+}$ ions are the most accurate ones available in their range of the periodic table of elements, where generally atomic data on highly charged ions are scarce. The recent measurements from NIST do not shed any light on the individual theoretical trends. However, our result for Na-like Eu agrees very well with the trend set by the other two measurements of similar accuracy, on Pt and U. Together, these three results validate Blundell's calculations. The Eu measurement also rules out the validity of the Gd data point from laser-produced plasmas.

Our measurements for Mg-, Al-, and Si-like Eu ions reveal significant shortcomings in the typical calculations. We hope that these results will spur the development of more accurate theoretical approaches for the treatment of such relatively simple multi-electron systems.

The measured spectra also contain lines in the predicted range (31 to 34 Å) of P- through Ar-like ion spectra. However, these lines are relatively weak and cluster closely (similarly to the case of W [10]), so that they can be investigated sensibly (that is, with sufficient data statistics and individual charge state discrimination) only with an experimental effort beyond the window of opportunity of this work.

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