Resonance-to-intercombination-line ratios of neonlike ions in the relativistic regime

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We report measurements of the intensity ratio of the $1s^22s^22p^5_{1/2}3d_{3/2} \rightarrow 1s^22s^22p^6$ resonance line to the $1s^22s^22p^5_{3/2}3d_{5/2} \rightarrow 1s^22s^22p^6$ intercombination line in neonlike Kr^{26+} and Mo^{32+}. The measurements were performed at the EBIT-I electron beam ion trap facility at the Lawrence Livermore National Laboratory and utilized an x-ray microcalorimeter. The measured ratio for Mo^{32+} is in four times closer agreement with theoretical predictions than earlier measurements of ions with lower atomic number. Our measurement thus suggests a narrowing of the disagreement with atomic number, which had not been observed in the previously existing data. This implies that the disagreement with theory may be localized to ions within a range of atomic numbers in which intermediate coupling dominates.

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I. INTRODUCTION

The $1s^22s^22p^5_{1/2}3d_{3/2}\ ^1P_1 \rightarrow 1s^22s^22p^6\ ^1S_0$ resonance (commonly denoted $3C$) and the $1s^22s^22p^5_{3/2}3d_{5/2}\ ^3D_1 \rightarrow 1s^22s^22p^6\ ^1S_0$ intercombination (commonly denoted $3D$) electric-dipole transitions are of great scientific interest because they dominate the x-ray emission of neonlike ions and, especially in the case of neonlike iron, Fe^{16+}, are important for the interpretation of astrophysical observations [1,2]. The observed ratio of the two lines does not readily agree with theoretical predictions. Over the past 20 years, it has been shown that this disagreement extends over a wide range of neonlike ions between Ar^{8+} and Kr^{26+} [3–9]. In the case of Fe^{16+}, it was shown that both the absolute and relative electron-impact excitation cross sections [10] as well as the relative oscillator strengths [11] differ from theory. The latter measurement was used to speculate about potential limitations in present-day quantum-mechanical calculations that appear to limit theory from reaching agreement with experimental observations [11].

Correlation effects are largest for the ions with the lowest atomic number Z. Therefore, discrepancies between experiment and theory, if caused by uncalculated correlation effects, should be largest for Ar^{8+} and diminish for Kr^{26+}. Indeed, this is seen in a recent study presented by Santana *et al.* [9] in which they employed configuration-interaction (CI) calculations with a limited set of configurations. When using second-order many-body perturbation theory (MBPT), which accounts for a very large fraction of the correlation effects, the discrepancy between theory and experiment disappeared as expected for the lowest-Z neonlike ions. Moreover, all calculations, including those with a small basis set and those using MBPT, converged to the same value for the highest-Z ions, suggesting that correlation effects are no longer important for those ions. Surprisingly, however, even the MBPT calculations no longer matched the experimental values. Instead, the difference with experiment grew as a function of Z to about 20% at $Z = 30$ and appeared constant up to the last measured point of $Z = 36$.

In this paper we present a measurement of the intensity ratio of the resonance to intercombination line of neonlike Mo^{32+}, which is six atomic numbers higher than the highest ion reported so far. Understandably, the intercombination line is weak for the low-Z neonlike ions. However, it increases in strength due to the increasing effects of relativity until its intensity surpasses that of the resonance line above about $Z = 36$. This means that the intensity ratio of neonlike Mo^{32+} is dominated by relativistic effects, putting our new measurement firmly in the relativistic regime.

Our present measurements employ an x-ray calorimeter. The calorimeter has a much higher counting efficiency than the crystal spectrometers used to make most of the earlier measurements [3,4] of the x-ray spectra of neonlike ions. As a result, our measurements have smaller statistical uncertainties than the earlier measurements. Taking advantage of this property, we also present a calorimeter measurement of the intensity ratio of the Kr^{26+} lines, which allows us to compare our result with that obtained earlier using a crystal spectrometer [4,12–14].

II. EXPERIMENT

The measurements were done using the EBIT-I electron beam ion trap at the Lawrence Livermore National Laboratory [15]. Operating at electron beam energies below 30 keV, EBIT-I has been used for experiments in the realms of optical UV, EUV, and x-ray spectroscopy. Overviews of the history and experimentation carried out at EBIT are provided by Beiersdorfer [16] and Marrs [17].

Mo or Kr was injected into the trapping region, where it was ionized to the desired state through collisions with the monoenergetic electron beam. The resultant ions were electrostatically confined by the charge of the electron beam and the voltage applied to the top and bottom drift tubes,

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respectively acting in the radial and axial directions. We used different methods of injection for Kr and Mo. Kr, being a noble gas, was injected into the trap through a gas injector. Mo, by contrast, was injected as a compound material, i.e., molybdenumhexacarbonyl, which was injected using a sublimator at a pressure of $\leqslant 1.6 \times 10^{-6}$ torr [18]. No cooling gas was used with the sublimator.

In order to maximize the presence of neonlike ions, we adjusted the electron-beam energy to be well above the threshold for obtaining neonlike ions yet sufficiently below the energy required to make fluorinelike ions. In the Kr experiment this meant using a beam energy of 2.7 keV, which is about 200 eV below the 2925 eV ionization potential of Kr^{26+}. In the Mo experiment the beam energy was set to 4.0 keV, which is about 200 eV below the 4235 eV ionization potential of Mo^{32+} [19]. Transition energies for lines $3C$ and $3D$ discussed here are known from both theory and previous experiments [4,20], and there is no ambiguity in line identifications.

The line emission spectra of interest were measured using the EBIT calorimeter spectrometer (ECS) [21–24]. In short, the ECS consists of a variant of the Suzaku/XRS spaceflight detector system. It is connected to a low-maintenance cryogenic system for cooling to 51 mK. The ECS employs a 32-pixel x-ray calorimeter array, where each pixel is a thermal x-ray detector. The absorber material that constitutes part of a given pixel absorbs an incident x ray, whence the integrated thermometer is able to sense the heat released by the x-ray absorption process.

The ECS uses multiple thermal shields, which also function as light blocks. The filters are made out of aluminium and polyimide with a total thickness of 1460 Å for aluminium and 2380 Å for polyimide. At the x-ray energies of interest for the krypton measurement, i.e., between 1800 and 1850 eV, the combined polyimide filter material transmits 98%, while the aluminum transmits $\geqslant$89% [25]. For the range of energies of interest for the Mo measurement, which is 2580–2680 eV, the transmission coefficients are respectively 99% and 95% [25]. In all these cases there is a less than 1% variation within the respective energy bands.

The ECS detector array is divided into a midenergy and a high-energy band subarray. The pixels used in our experiment all belong to the midband subarray, covering energies in the range from 0.1 to 10.0 keV. Specifically, the midband pixels in our experiment utilize HgTe absorbers, measuring 625 μm $\times$ 625 μm in area, and 8 μm in thickness. These pixels have previously been shown to have a quantum efficiency of 95% at 6 keV [22]. The quantum efficiency in the two energy regions of interest here is 100%. The ECS provides the benefit of an autonomous, real-time monitoring and analysis system for data acquisition. The ECS has been used at our facility since its original implementation in 2007; earlier models had been in use since 2000 [26].

For the Mo experiment an additional light block was added to the setup to reduce the flux of low-energy x rays. This block consisted of two Be windows with a combined thickness of 139.7 μm. This thickness is large enough that we need to account for the change in the transmissivity across the energy range of interest for the two neonlike Mo^{32+} lines. According to the transmission data from the Center for X-Ray Optics, the transmission coefficients for our Be windows are 45.86% at 2580.58 eV (line $3D$) and 41.72% at 2677.61 eV (line $3C$) [25]. We accounted for this effective 4% difference in the transmissivity by applying a transmissivity correction to the obtained ratios.

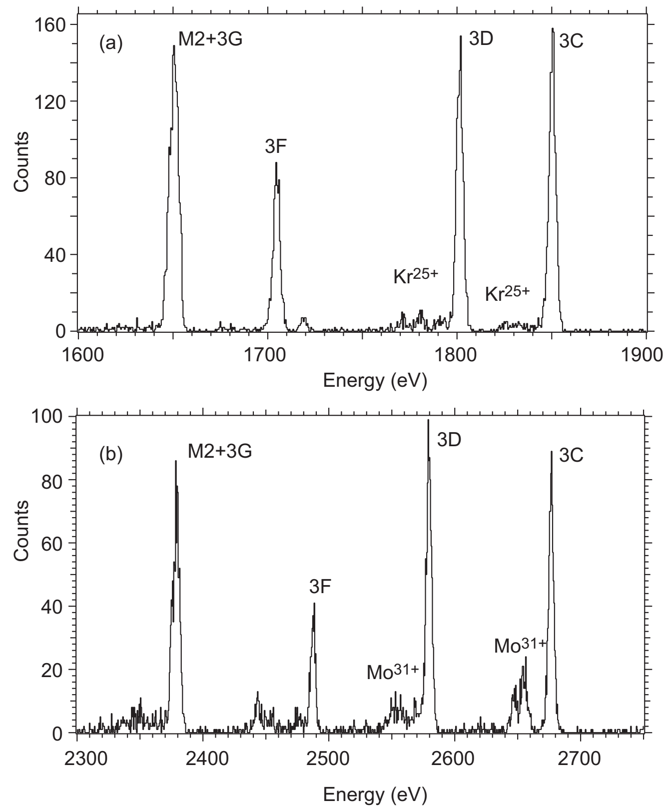

FIG. 1. Spectra of (a) neonlike Kr^{26+} and (b) neonlike Mo^{32+} obtained with a single pixel of the ECS calorimeter on the EBIT-I electron beam ion trap at Livermore. In standard notation, $M2$, $3G$, $3F$, $3D$, and $3C$ denote the transitions from the $(1s^22s^22p^5_{3/2}3s_{1/2})_{J=2}$, $(1s^22s^22p^5_{3/2}3s_{1/2})_{J=1}$, $(1s^22s^22p^5_{1/2}3s_{1/2})_{J=1}$, $(1s^22s^22p^5_{3/2}3d_{5/2})_{J=1}$, and $(1s^22s^22p^5_{1/2}3d_{3/2})_{J=1}$ upper levels to the $(1s^22s^22p^6)_{J=0}$ ground level, respectively. Weaker lines are from lower charge states, i.e., predominantly from sodiumlike ions.

Typical x-ray spectra obtained by the ECS of Kr^{26+} and Mo^{32+} obtained with the ECS on EBIT-I are shown in Fig. 1. Specifically, the figure displays for each ion the spectrum obtained with a single pixel. As mentioned above, there are 14 pixels in the midband subarray, all of which collect spectra of similar quality.

III. ANALYSIS AND RESULTS

Intensities were obtained by performing a multipeak fit to the spectra using Gaussian trial functions. The spectra exhibit several weak features (cf. Fig. 1), which are emission from charge states lower than neonlike, in particular from sodiumlike ions. The existence of sodiumlike peaks in the spectra made it necessary for us to include fits of these peaks in the overall line fitting procedure. The sodiumlike lines are relatively weak. This constrains the contributions from unfitted lines that blend with the neonlike lines of interest.

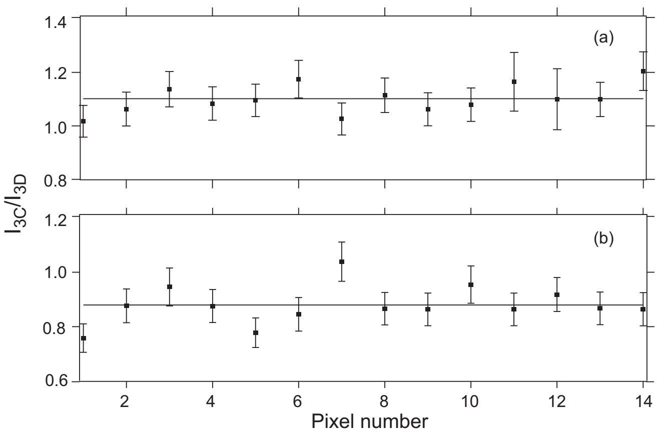

FIG. 2. Ratio of the intensity of line $3D$ relative to line $3C$ inferred from the spectra recorded by each calorimeter pixel: (a) neonlike Kr^{26+}; (b) neonlike Mo^{32+}. The values have not been corrected for the filter transmission.

The multipeak fitting process was repeated for each pixel, resulting in 14 independent ratios for each ion, as illustrated in Fig. 2. The average ratios, before adjustment for the differential absorption by the filters, are 1.102 ± 0.015 for Kr^{26+} and 0.880 ± 0.020 for Mo^{32+}. Here, the error bar represents purely the statistical uncertainty at the 68.3% confidence interval. Accounting for differential absorption produces values of 1.092 ± 0.015 and 0.793 ± 0.020, respectively. Note that the effect of the differential filter absorption is very small for Kr^{26+} as a result of the very small differential change in the transmissivity of the aluminium and polyimide filters. The much more significant effect in the Mo^{32+} case is primarily due to the relatively thick Be light block described earlier.

The fact that there are unresolved sodiumlike lines that blend with the neonlike lines introduces a systematic error. A high-resolution measurement of the x-ray emission of Kr was reported by Rice *et al.* from the Alcator tokamak [20]. It shows sodiumlike lines close to lines $3C$ and $3D$. In the krypton case, Rice *et al.* predict a Kr^{25+} transition from upper level $(1s^2 2s^2 2p^5_{1/2} 3p_{1/2} 3p_{3/2})_{J=3/2}$ to occur at 1803.5 eV, which they measure to be at 1800.5 eV. This is so close to line $3D$ that our instrument cannot resolve the two lines. Similarly, their calculations predict a sodiumlike krypton transition from upper level $(1s^2 2s^2 2p^5_{1/2} 3s_{1/2} 3d_{5/2})_{J=3/2}$, which they measured at 1846.5 eV. This line should appear as a low-energy shoulder in our measurement of line $3C$. From the size of the sodiumlike lines visibly resolved in our spectra shown in Fig. 1 we can estimate that the contribution from these unresolved lines is very small. Rice *et al.* predict two sodiumlike lines at 1794.7 eV and 1791.9 eV, which they measure to be at 1793.6 eV and 1791.3 eV, respectively. These are transitions from the $1s^2 2s^2 2p^5_{3/2} 3s_{1/2} 3d_{5/2}$ upper levels with $J = 3/2$ and $J = 1/2$, respectively, to the sodiumlike $1s^2 2s^2 2p^6 3s_{1/2}$ ground state. The two lines form a feature that is resolved in our spectra with a combined intensity of about 5% that of line $3D$. Rice *et al.* predict that these two lines are roughly 5–6 times stronger than the unresolved line that blends with $3D$ in our measurement. Thus we estimate that the unresolved line contributes about 1% to the intensity of line $3D$. Similarly, our measurements resolve the sodiumlike feature that consists of lines measured by Rice *et al.* to be at 1835.3 eV and 1831.7 eV. Their calculations predict this feature to be about twice as strong as the line that blends with line $3C$. As a result, we estimate that the unresolved line at the shoulder of line $3C$ contributes about 2% to its apparent intensity. A caveat about our estimates is that the calculated intensities shown by Rice *et al.* (Fig. 1 in [20]) match their measured sodiumlike lines only within a factor of two or larger. We thus assume a 5% uncertainty in our estimate and a differential contribution, which matters in the line ratio, half of this value. We thus linearly add a 2.5% uncertainty from line blending to the statistical uncertainties.

For our Mo measurement, we rely on the high-resolution measurement performed by Källne *et al.* [27] on the Alcator tokamak. They report a line with 1.9 eV less energy than the $3D$ line. They do not identify the configurations involved in this transition, but they have assigned it an intensity, which we can use to estimate this line's effect on our measurement. In particular, we estimate that this line has an intensity of about half the broad, resolved sodiumlike feature near 2555 eV in our spectra. Blending with this line thus enhances the apparent intensity of line $3D$ by about 4%. Källne *et al.* also measured a sodiumlike line with 2.8 eV lower energy than the $3C$ line. This line proceeds from the $(1s^2 2s^2 2p^5_{1/2} 3s_{1/2} 3d_{3/2})_{J=3/2}$ upper level to the sodiumlike ground state. Its intensity was measured by Källne *et al.* to be about one-fifth that of the sodiumlike feature that we observe at 2655 eV. From this we estimate that this line enhances the intensity of the $3C$ line by about 5%. The differential enhancement in the ratio of the molybdenum $3C$ and $3D$ lines is about 1%. As we have done for Kr, we conservatively add a 2.5% uncertainty from line blending to the statistical uncertainties.

The final values of our measurements, including the uncertainties from line blending, are 1.092 ± 0.042 and 0.793 ± 0.040 for the Kr^{26+} and Mo^{32+} line ratios, respectively.

We note that no adjustments were made for the fact that the x-ray line emission from EBIT-I is polarized [28–31]. Unlike crystal spectrometers, calorimeters are not sensitive to polarization *per se*. However, the line emission from an electron beam ion trap is not isotropic, but its angular emission pattern relative to the direction of the electron beam depends on its polarization. The relative intensity of two lines, therefore, needs to be adjusted for such polarization effects. However, calculations have shown that lines $3C$ and $3D$ have the same polarization [32], and no adjustments are needed.

The final values are listed in Table I. Moreover, they are plotted in Fig. 3 as a function of Z together with the values from the previous measurement with $Z \geqslant 28$. Here, we compare the measured values with two theoretical calculations by Santana *et al.*—one based on a CI model that considers 816 states up to triply excited $n = 3$ levels and the other based on the fully relativistic second-order MBPT method [9]. In addition, the figure includes the theoretical values from the fully relativistic distorted-wave model of Zhang and Sampson [33] and the values from a configuration-interaction calculation by Hibbert *et al.* [34].

It is interesting to note that our two values are in closer agreement with theory than any of the previous measurements shown in Fig. 3. The difference between our Mo^{32+}

TABLE I. Comparison of our measured and calculated ratios of the intensity for the 3C and 3D lines with previous measurements along with results from several theoretical models.

Z	Measured ratio (present work)	Measured ratio [4,38]	Santana (MBPT) [9]	Santana (CI) [9]	Hibbert *et al.* [34]	Zhang and Sampson [33]
28		2.30 ± 0.16	2.59	2.73	2.83	2.860
29		1.97 ± 0.14	2.27	2.39	2.47	2.473
30		1.71 ± 0.10	2.02	2.11	2.17	2.166
31					1.93	1.916
32		1.50 ± 0.14	1.63	1.69	1.74	1.701
33					1.58	1.544
34		1.12 ± 0.05	1.37	1.41	1.44	1.402
35		0.93 ± 0.07	1.27	1.30	1.34	1.285
36	1.092 ± 0.042	0.99 ± 0.07	1.18	1.21	1.25	1.189
37						1.103
38						1.034
39						0.971
40						0.918
41						0.874
42	0.793 ± 0.040					0.833

measurement and theory is only 5%, which is four times smaller than the difference found for the previously measured three highest-Z ions, as illustrated in Fig. 4. In fact, this difference is smaller than for any ion other than Ar^{8+} reported by Santana *et al.* [9]. Moreover, the uncertainty limits of our measurement for Mo^{32+} overlap with theory. The present measurement of Mo^{32+} thus indicates in a tantalizing way that the difference between theory and experiment narrows for higher-Z ions. This narrowing implies that the difference between theory and experiment is mainly confined to the range of Z in which relativistic effects start to become important but do not yet dominate so that correlation effects are still important. In other words, the disagreement might be localized to ions within a range of atomic numbers in which intermediate coupling dominates.

Our measured ratio for Kr^{26+} is closer to theory than that reported by Brown *et al.* [4]. However, our result still overlaps with the Brown *et al.* value within their respective error bars but not with theory.

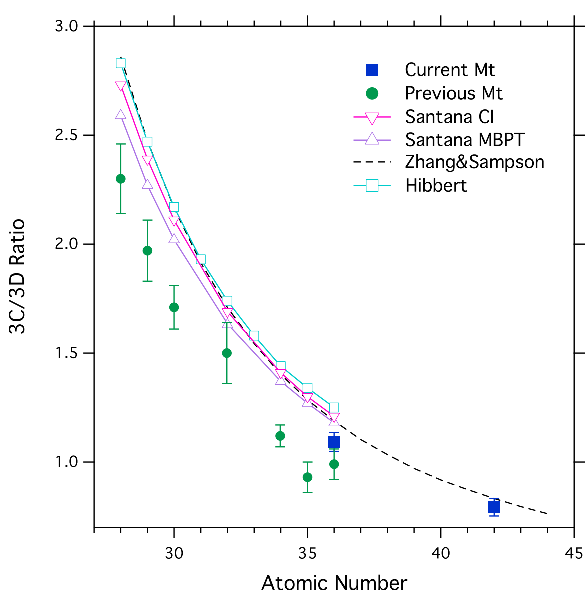

FIG. 3. Average measured 3C/3D line ratio for Kr^{26+} and Mo^{32+} (solid blue squares) compared with previous measurements by Brown *et al.* [4] and Beiersdorfer *et al.* [38] (solid green circles) and theoretical values of Santana *et al.* [9], Zhang and Sampson [33], and Hibbert *et al.* [34]. Two calculations by Santana *et al.* are shown: one is labeled CI, which is based on a 816 state configuration-interaction calculation, and another labeled MBPT, which is based on the fully relativistic second-order MBPT method.

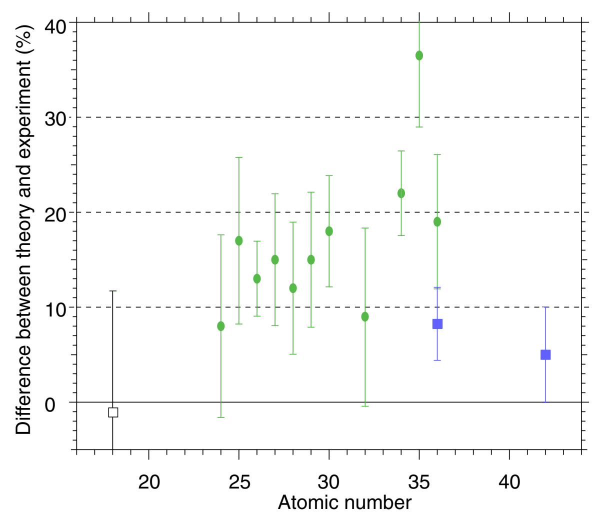

FIG. 4. Percent difference between calculated and measured values of the 3C/3D line ratio. The differences are normalized to the measured values. The theoretical value from Zhang and Sampson [33] is used for the Mo^{32+} ratio; all others are from MBPT calculations by Santana *et al.* [9]. The experimental values are from Santana *et al.* [9] (black open square), Brown *et al.* [4], and Beiersdorfer *et al.* [38] (solid green circles), and the present measurements (solid blue squares).

Clearly, measurements at even higher atomic number are needed to confirm this trend. Similarly, it may also be interesting to see how the ratio behaves at the low range of atomic numbers, where so far only one value for Ar^{8+} exists. We note that the reordering of the energy levels in neonlike ions as a function of Z [35] results in several avoided level crossings that limit the range in which the ratio varies smoothly. An avoided level crossing can have profound effects on the observed intensities and must be considered when studying ions close to such a crossing [36]. For example, Nakamura *et al.* [37] have shown that line $3D$ crosses with another electric dipole line around $Z = 54$, and the theoretical analysis by Safronova *et al.* [35] of the mixing coefficients of the upper level of line $3D$ reveals additional abrupt changes below $Z = 18$. As seen from Fig. 4, there are, however, many species between $18 < Z < 54$ that have not yet been measured and that could give information on the behavior of the relative ratio of the resonance and intercombination line in neonlike ions.

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