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Measurement of an unusual $M1$ transition in the ground state of Ti-like W^{52+}

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Using a high-efficiency, transmission grating spectrometer the $3s^23p^63d^4$ $J=2$–3 ground-state transition was measured in Ti-like W^{52+}. The measurement has achieved 20 times better precision than the only previously reported high-Z measurement. As such, it tests recent structure calculations for complex highly charged ions. Discrepancies of around 100 Å are found. This is true even for semi-empirical calculations. The measurement, however, validates a very recent calculation with which agreement is within 2 Å.

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I. INTRODUCTION

Accurate measurements of transition energies within the ground state of highly charged ions serve as benchmarks to gauge the accuracy of atomic structure theory. Additionally, the identification of intense lines in the near UV and visible region of the spectrum is important from the perspective of plasma diagnostics to be used in the measurement of such parameters as the plasma temperature and magnetic-field strengths. As plasma devices achieve hotter temperatures, of particular interest are transitions from the ground state of highly charged, high-Z ions with ionization potentials between 5 and 10 keV. As such, Feldman, Indelicato, and Sugar carried out a broad range theoretical survey of ions with a $3s^23p^63d^l$ ground configuration in hopes of predicting such a useful transition [1]. Through this survey was discovered a transition that not only satisfied this need, but showed an unusual wavelength stability as a function of Z in the range $Z=60$ to $Z=92$. Using refined calculations that included retardation to all orders, screened radiative corrections, and nuclear size corrections, they predicted that the fine-structure transition $3s^23p^63d^4$ $J=2$–3 within the Ti-like ground state would have a wavelength of $\lambda_{\mathrm{Nd}}=3557$ Å at $Z=60$, change very little to $\lambda_{\mathrm{W}}=3546$ Å at $Z=74$, and slowly decline in wavelength to $\lambda_{\mathrm{U}}=3200$ Å at $Z=92$.

Soon after publication by Feldman *et al.*, the first measurements of this transition in Xe^{32+} and Ba^{34+} were reported [2], with results that differed from *ab initio* calculations by about 200 Å, a discrepancy that can be attributed to the complexity of modeling a 22-electron system. Over the course of the next few years, measurements were made of this transition in the ions Nd^{38+} and Gd^{42+}, as well as I^{31+} and Cs^{33+} [3]. The measured wavelengths were again found to be about 5% longer than predicted. Only a single measurement has yet been reported of ions beyond Gd^{42+}, the element at which a competing transition, the $J=4$–3, begins to become the more probable decay path [1]. This measurement of Au^{57+} was performed with modest resolution and measured the transition wavelength to be about 50 Å, or 1.5%, longer than predicted [4].

New calculations were performed by Indelicato that attempted to improve upon the significant disparity between theory and experiment by doing multiconfiguration Dirac-Fock (MCDF) calculations with a larger basis set [5]. These calculations failed to improve the agreement since it was found that the extra configurations contributed equally to the $J=2$ and $J=3$ levels. A separate effort of Beck in 1997 produced *ab initio* calculations for four of the previously measured mid-Z elements with somewhat improved results. However, these extensive calculations made no predictions for any unmeasured transitions [6]. Subsequently, Serpa *et al.* used the measured values from Xe, Ba, Nd, and Gd as a guide to predict adjusted values for higher-Z elements up to $Z=76$ (Os^{54+}) [7]. An even more recent set of calculations by Kato *et al.* [8] made an attempt to explain the cause of the anomalous stability of the wavelength in terms of a concept of a critical atomic number beyond which relativistic effects dominate electron correlation in atomic structures, but was unable to improve upon the predictions of wavelengths from even the original set by Feldman *et al.* Ensuing calculations by Kato *et al.*, have recently been reported that significantly reduce the disagreement between theory and the available measured values to a level better than 1% [9]. These calculations were made with the extended-optimal-level mode of the MCDF method and included lowest-order QED corrections as well as a nuclear charge distribution represented by a spherically symmetric Fermi model. Most importantly, the calculations predicted new values for the high-Z elements.

In the following, we provide a test of the new calculation by Kato *et al.* [9] by presenting a high-accuracy measurement of the $3s^23p^63d^4$ $J=2$–3 transition with Ti-like W^{52+}. The measurement was performed using an alternative transmission grating spectrometer that provides the very high efficiency necessary to obtain high spectral accuracy. As a result, our measurement achieved a precision that is 20 times that of the Au^{57+} measurement, i.e., of the only other measurement in the very-high-Z region. We find that the recent calculation has indeed improved the predictive quality of theory for high-Z ions by more than a factor of 50 over existing calculations.

II. EXPERIMENTAL SETUP AND MEASUREMENT

The present measurement was performed at the EBIT-II facility at the Lawrence Livermore National Laboratory. This is one of the two original electron beam ion traps, a description of which is given, for example, in Ref. [10]. It

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has been predicted that for Z larger than 64, the $3s^23p^63d^4$ $J=4-3$ transition is more favorable than the anomalous $J=2-3$ transition. For W^{52+} only 16% of the $J=3$ population is expected to go to the $J=2$, and for U^{70+} this drops to only 5% [1]. In order to make precision measurements from high-Z elements it is necessary to have both a high-resolution and high-efficiency optical spectrometer system. We used a newly developed transmission grating spectrometer consisting of a matched pair of $f/4.6$ achromatic lenses, a very high-efficiency, 6-in.-diam planar quartz transmission grating, and a back-thinned, cryogenically-cooled charge-coupled-device (CCD) detector. The use of two identical lenses and a lack of an external slit results in a one-to-one image of the photon emitting ions within EBIT at the two-dimensional detector plane. Slitless operation is possible because in EBIT the ions emit only in a narrow region defined by the 70-μm-diam electron beam.

Feldman *et al.*, predicted the wavelength of the Ti-like transition from W to be $\lambda_W = 3546.1$ Å; however, each of the previous measurements of other elements determined wavelengths on the order of 5% longer than predicted. Therefore, observations were made in the range from 3420 to 3900 Å in search of the line. Calibration of the spectrometer is performed *in situ*. Calibration using external sources, such as standard spectral lamps shining though a port opposite the spectrometer, as has been reported by the groups at NIST [2] and Oxford [11], requires significant effort towards alignment and still retains the possibility of adding additional systematic errors. In fact, external spectral lamps do not fill the optics the same way the EBIT ions do. This has led to systematic uncertainties that cannot be accounted for in a way that is satisfactory to us, which is why we do not use this method. Well-known lines in the UV/visible emitted from highly charged ions within EBIT are uncommon, and reported wavelengths do not have the precision required for calibration. There is, however, a third option. Atoms injected by the gas injector and streaming through the chamber pass through the electron beam with a good chance of colliding with beam electrons and not becoming trapped. The collisions may strip none to a few electrons from the atoms while leaving the remaining atomic electrons possibly in an excited configuration. These electrons can then make fast transitions to lower states, many of which are in the wavelength range of our setup. These lines provide excellent, well-known calibration references. In fact, precision measurements of lines emitted from low-charge ions and atoms have been performed for many years on sources other than EBIT and values for the wavelengths of these lines are well established [12].

The spectral lines from low-charge-state ions and atoms appear in the CCD image regardless of the trap depth, i.e., whether or not an axial trapping potential is applied to the top and bottom drift tubes. By raising the potential of the middle drift tube to a higher value than the top drift tube, spectral features from low-charge ions can be distinguished from those of highly charged ions. This is called ''inverting the trap'' and has proven to be very helpful in spectral charge-state identification. Since no trapping and ensuing ionization occur, the spectrum of untrapped atoms and ions is fairly independent of the electron-beam energy.

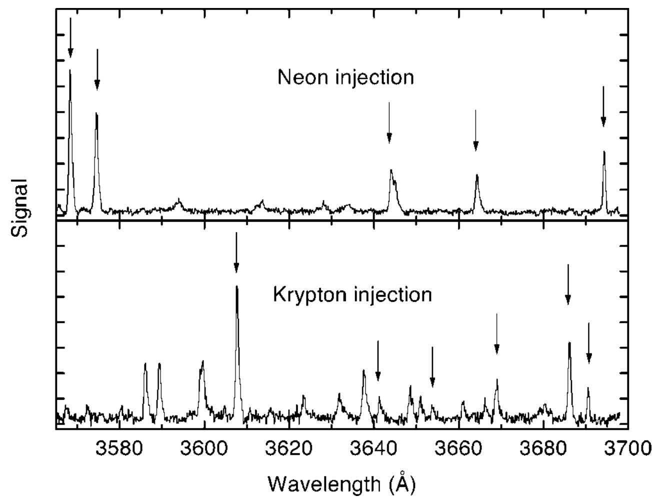

FIG. 1. *In situ* calibration spectra using low charge states of neon and krypton. For both cases shown EBIT was operated with an inverted trap so that no highly charged ions were formed. ↓ indicate lines that were used in the calibration. Unmarked lines are unidentified in the standard reference tables.

Neon and krypton gases were introduced into EBIT for the *in situ* calibration of the spectrometer. Figure 1 shows the spectra of untrapped Ne and Kr resulting from many hours of summed data for each gas at a single position of the CCD. The strongest lines are mostly from singly and doubly charged ions. The arrows (↓) in the figure point out the 11 lines that were used to calibrate this section; unmarked, isolated, intense lines are not identified in the reference tables. Many transitions from neutral atoms are found, but, though these lines are the most precisely measured reference lines, they are insufficiently intense to be used for calibration.

The electron-beam energy necessary to create the Ti-like

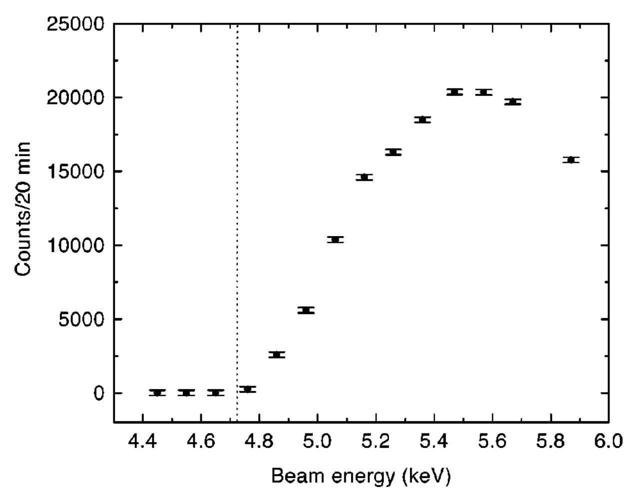

FIG. 2. Total number of counts above background collected by the spectrometer per 20 minutes as a function of electron-beam energy. The energy scale incorporates the potentials applied to the drift tube assembly and the middle drift tube, as well as an estimation for the space charge. The energy necessary to create the Ti-like charge state is 4.72 keV (dotted line).

charge state — 22 remaining bound electrons — is 4.72 keV [13]. To search for the line, EBIT-II was set to an electron-beam energy $E_{\mathrm{beam}}=4.86$ keV and current $I_{\mathrm{beam}}=130$ mA. The detector accumulated signal for 20-min integrations for each spectrum, was then moved towards shorter wavelengths, and the procedure repeated until the region 3420–3900 Å was covered: only a single line was detected. To ascertain the emitting charge state, a scan over the electron-beam energy was performed (see Fig. 2). The disappearance of the line after lowering the beam energy below that necessary to create the Ti-like charge state and the relatively close agreement of its wavelength with the theoretical treatment by Feldman *et al.* [1] verified its identification as the $J=2-3$ transition. The electron-beam energy was then adjusted until an optimal count rate was achieved in the line. Data were accumulated for many hours, alternating the W injection with either the Ne or Kr gas injection. Figure 3 shows the signal plotted along the calibrated wavelength axis. The wavelength was measured to be 3627.13 Å (in air) with a full width at half maximum of 1.32 Å.

Multiple measurements of the transition with various beam currents and energies were made with each of the lines accumulating a statistically significant number of counts. Using a purely statistical approach, the error in each measurement is found to be about 0.051 channels, or equivalently, 0.0071 Å. However, the scatter of the various measurements was much larger, the standard deviation of the fits being 0.72 channels (18 μm) — about 0.10 Å of the calibrated wavelength scale. Factors that can contribute to such a scatter are the shifting of the electron beam within EBIT or movement of the entire optical system during the measurement. The spectrometer optical system produces a one-to-one image of the electron beam at the detector; therefore, a measured shift of, for instance, a single channel at the detector could be caused by the translation of the electron beam by 25 μm. However, systematic studies of the stability of the electron beam have shown that during steady-state operation there is no measurable shift in the electron beam's position [14]. The beam's position has been shown to change slightly as the beam energy is changed (about 6 μm maximum), but only with changes in energy that required the retuning of EBIT. Throughout these measurements, no retuning was required.

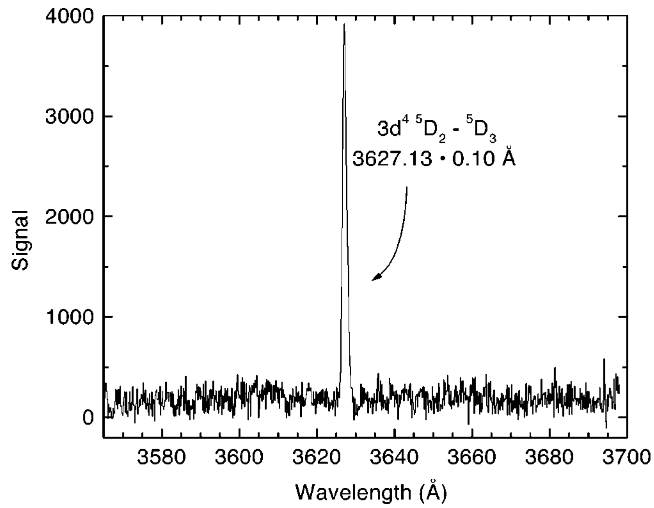

FIG. 3. Spectrum from one setting of the transmission grating spectrometer. The single line, the only one detected in the spectral band from 3430 to 3880 Å, is identified as the Ti-like W ground level $J=2-3$ transition.

TABLE I. Predicted and measured values for the wavelengths of the anomalous $3s^2 3p^6 3d^4$ $J=2-3$ transition along the Ti isoelectronic sequence.

Element	Z	Predictions (Å) Feldman[a]	Serpa[b]	Kato[c]	Measured wavelengths (Å)
I	53	4109.5		4307.1	4303.3(0.8)[d]
Xe	54	3952.5	4130	4156.4	4138.8(0.7)[d], 4139.4(2.0)[e]
Cs	55			4028.5	4021.4(1.2)[d]
Ba	56		3932	3936.5	3930.8(1.8)[d], 3932.4(2.0)[e]
Nd	60	3556.8	3749	3753.8	3753(2)[b]
Gd	64		3686	3712.0	3713(2)[b]
Tb	65	3548.3			
Er	68		3636		
Yb	70	3564.5		3676.7	
Hf	72		3562		
W	74	3546.1	3524	3625.7	3627.13(0.10)[f]
Os	76		3507		
Au	79			3531.0	3532(2)[g]
Pb	82	3427.3		3462.7	
U	92	3199.7		3209.0	

[a]Feldman *et al.* [1].
[b]Serpa *et al.* [7].
[c]Kato *et al.* [9].
[d]Yamada *et al.* [3].
[e]Morgan *et al.* [2].
[f]Present work, in air wavelength.
[g]Träbert *et al.* [4].

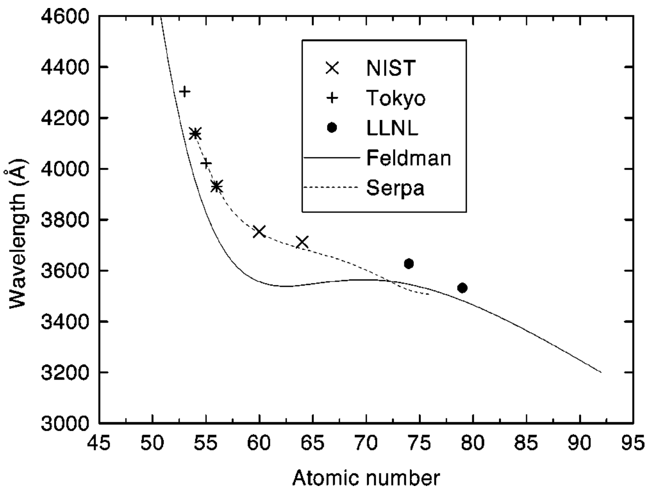

FIG. 4. Comparison of the wavelengths of the Ti-like $3s^2 3p^6 3d^4$ $J=2-3$ ground-state transition predicted by Feldman *et al.* [1] (solid line) and Serpa *et al.* [7] (dashed line) with those measured at various EBITs. ● mark the measurements reported by the NIST EBIT group [2,7]; $+$ depict the points measured at the Tokyo EBIT [3]; $\times$ show the previous and present measurements from the LLNL EBIT at highest Z [4]. Not shown is a measurement from the Oxford EBIT of the transition in Ba ($Z=56$) [11], which agrees with the other two reported measurements. The error for each of these measurements is much smaller than the relative size of the markers.

Movement of the spectrometer system would have to be traced to some sort of external interaction. The optical table is rigidly mounted to its stand, which in turn is bolted onto the concrete floor, reducing the possibility of motion due to vibrations or accidental bumping. The internal components are also rigidly attached to the table making even slight movements unlikely. However, thermal expansion of the optical table, resulting from temperature fluctuations in the room may have a significant effect. A temperature change in the EBIT-II laboratory room of 1 °C to 3 °C throughout a 24 hour period has been found to be common. This is particularly significant since measurements at EBIT are performed day and night. An analysis of the spectrometer's geometry has estimated the thermal shift to be approximately 4 μm/°C at the detector. Temperature fluctuations, therefore, account for most of the measured 18-μm scatter of the line positions noted in the analysis and presently limit the precision of our measurements. The final result of our measurement is 3627.13 ± 0.10 Å. The majority of the calibration lines are known to better than 0.01 Å [12] and, therefore, add no significant adjustments to the error budget.

III. CONCLUSIONS

In Table I we list several predictions and all available data of this unique transition. As shown in the table the only two earlier predictions for the wavelength of the Ti-like transition at high Z differ considerably from our measurement. The predictions made by Feldman *et al.*, and Serpa *et al.*, are shown in Fig 4. It is interesting to note that the recent semiempirical prediction by Serpa *et al.*, disagrees even more with our measurement than that by Feldman *et al.* Our wavelength measurement of 3627.13 Å (3628.16 Å vacuum wavelength) is 82 and 104 Å, respectively, longer than these two predictions. The value predicted by the new calculation by Kato *et al.* [8] is 3625.7 Å. This value is remarkably close to our measured value, improving the accuracy of the predictions by nearly two orders of magnitude. The close agreement to our measurement suggests that the new calculations may well fill the void of unmeasured elements between Gd and W. It would be interesting and useful to continue such measurements at even higher Z. Since these transitions should be weak due to the competition of the $J=4-3$ transition, the success of these measurements will depend even more on the use of high-efficiency optics. The present measurement shows that this is possible even if the emitted intensity decreases by an order of magnitude.

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