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March 5, 2008

Physical Review A

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# Electric-dipole 5s - 5p transitions in promethiumlike ions

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(Dated: March 3, 2008)

## Abstract

The 5s - 5p electric-dipole resonance transitions in highly ionized promethiumlike ions have been studied applying relativistic multi-reference Møller-Plesset second-order perturbation theory. The transition wavelengths are determined to within 0.2 Å in the more highly charged ions, where the level degeneracies are small. For somewhat lighter ions a very large reference space was used in order to account for the many degeneracies. In order to calculate transition probabilities and lifetimes, correlation corrections have been added to the transition operator in the next order. The contributions from the higher orders of the theory, that is, frequency-dependent Breit correction, Lamb shift, and mass shifts, have been estimated. The results are used to re-assess spectroscopic data from beam-foil, electron beam ion trap, and tokamak observations.

PACS numbers: 32.70.Jc, 31.15.Md, 34.50.Fa

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## I. INTRODUCTION

Historically, the prominent  $ns\ ^2S - np\ ^2P^o$  resonance lines of alkali elements and their isoelectronic spectra have been investigated very intensively by both theoretical and experimental means. Spectral analysis and systematization have led to the development of Grotrian diagrams which - owing to a single valence electron outside a closed shell - signaled the (relatively) simple structure of the alkalis. The prominence of the resonance lines opened up the field of spectral analysis and spectrochemistry some 150 years ago, and it intrigued theory to turn to these transitions as a test bench on which to try modeling gross and fine structure effects as well as a number of important as well as subtle atomic structure effects. Among these are relativistic effects in highly charged ions, transcending by far the fine structure seen and named as such in atoms and low-charge state ions, and quantum electrodynamics (QED) corrections. With a single valence electron, it was also assumed that it was easy to calculate the transition probabilities reliably. However, it has taken more than two decades after the first precise transition rate measurements on Li and Na atoms (in the late 1970s) to have theory and experiment converge on accurate values of these entities. Although we will refer to transition probabilities and level lifetimes in specific alkali sequence ions in the course of the discussion, our atomic structure calculations of specific high nuclear charge  $Z$ , many-electron alkali sequence ions show that those transitions at best play a secondary role in the present context, contrary to earlier assumptions.

The adequate fully relativistic treatment of alkali sequence ions has been pursued for decades at Notre Dame, by W. R. Johnson and colleagues. For example, in some earlier papers Johnson, Blundell and Sapirstein used relativistic many-body perturbation theory (MBPT) to calculate energy levels in Li-like, Na-like and Cu-like ions [1–3]. In these papers they accounted for Coulomb and Breit corrections up to third-order. The discrepancy between theoretical predictions and experimental measurements was attributed to QED effects. In 1991, Kim *et al.* [4] obtained correlation energies from the differences of MBPT and multi-configuration Dirac Fock (MCDF) calculations, and combined these with MCDF results for the energy levels as well as a semiempirically corrected estimate of QED effects. In 1993, Blundell [5] extended the Notre Dame study of alkalilike ions to include an *ab initio* calculation of the self-energy and vacuum polarization. The agreement with experiment was within two experimental standard deviations. The quality of both of these calculations has

been corroborated by recent experiments reported from the Livermore electron beam ion traps EBIT-I and SuperEBIT [6–9] which reach up to U ( $Z=92$ ) for all three isoelectronic sequences and find good agreement with these calculations. For Li-like ions, the alternative experimental approach of using a foil-excited fast ion beam as the light source has reached up to Xe ( $Z=54$ ) [10] with data that since have been corroborated by an experiment again at SuperEBIT [11]. These reports on the experiments also discuss earlier experimental and theoretical efforts, which we prefer not to repeat here.

A common theme in the pursuit of accurate calculations is the finding of many-body effects in what originally was perceived as a single electron system and what is still approximated well - but not well enough for present day requirements - by a single valence electron model. The Kaiserslautern group of H. Schmoranzer [12] has found in their study of alkali atom resonance transition probabilities that theory and experiment agree well only for Li and Na (to much better than 1%), but that there are increasing discrepancies (10 to 20%) in the heavier alkali atoms of K, Rb, and Cs. They employed the very same experimental techniques to the various atomic species; hence it is likely that the causes of discrepancy lie in the theoretical description of such many electron systems. Similar problems have been encountered in rare gas atoms from Ne through Rn.

The ordering of inner electronic shells in heavy elements can be subject to the ionic charge (or rather the core charge, that is, the nuclear charge minus the number of electrons in the innermost electronic shells). For example, the ground state of neutral Pm ( $Z=61$ ) has the (abbreviated) electron configuration  $4f^5 5s^2 5p^6 6s^2$ , which does not match that of any alkaline atom. In 1980, however, Curtis and Ellis performed Hartree-Fock calculations [13] and then suggested that Pm-like ions with a nuclear charge  $Z$  larger than 73 have alkali structure with a ground configuration  $4f^{14} 5s$ . Since important relativistic corrections were omitted by Curtis and Ellis, Theodosiou and Raftopoulos performed additional Dirac-Fock calculations [14] and predicted the turning point where  $4f^{14} 5s \ ^2S_{1/2}$  rather than  $4f^{13} 5s^2 \ ^2F_{7/2}^o$  becomes the ground state to happen at  $Z=77$  instead of  $Z=74$ . Since these theoretical studies in the 1980's, there appear to have been done no further high-accuracy calculations in highly ionized Pm-like ions.

Accurate theoretical predictions are required to make positive single-line assignments of the  $4f^{14} 5p$  level decays that in an alkali spectrum would be expected as resonance lines of some prominence. This task is made difficult by the fact that in the light of the advanced

calculations the lines of interest are expected to appear among a fair number of others and do not offer individual signatures (fine structure, decay mode, etc.) that might help with the identification. This situation increases the need for accurate, reliable predictions. We recently developed a version of a relativistic state-specific multi-reference Møller-Plesset perturbation theory [15–17] using the common set of radial spinors obtained by a state-averaged multiconfigurational self-consistent-field (MCSCF) procedure [18]. This approach has been very successful in identifying lines in the spectra of ions of a variety of isoelectronic sequences.

In the present study we report on our investigation of the term energies in Pm-like ions using second-order single-reference Møller-Plesset (MP2) and multi-reference Møller-Plesset (MR-MP) theory. We have undertaken such calculations both as a demonstration of the capabilities of our calculational approach and in order to compare with experimental observations that have been made over the years. Moreover, a reassessment of the experimental situation seems necessary, because most searches for the lines of interest have been futile. Evaluating the conditions under which to expect a reliable signal might help to judge whether the experiments done so far could have produced substantial evidence, and to guide any future experimental search for the  $4f^{14}5s - 4f^{14}5p$  transition lines in heavy Pm-like ions.

## II. COMPUTATIONAL RESULTS

Our computational procedures have been amply described elsewhere [15–17]. In addition to the excitation of valence electrons, our computations comprise up to double core excitation. Indeed, in the beginning of the Pm sequence, it is not clear what the actual ground configuration and thus the valence electron would be, and we have included all candidate configurations along with their respective core excitations. One of our results is that the  $4f^{14}5s \ ^2S_{1/2}$  level is the ground state from  $Z=78$  onwards (see figure 1). The general trend of the low-lying configurations as it evolves along the isoelectronic sequence towards uranium is shown in figure 2.

From the quality of the results obtained for other atomic systems and from the energy value convergence observed with our large basis sets, we estimate that these multi-reference Møller-Plesset (MR-MP) calculations can produce transition wavelengths of Pm-like ions for  $77 \leq Z \leq 92$  with an accuracy of  $0.2 \text{ \AA}$ . In these more highly charged ions, the amount of

level degeneracies is small. For somewhat lighter ions a very large reference space was used in order to account for the many degeneracies, and we assume a slightly larger wavelength uncertainty there. In order to calculate transition probabilities and lifetimes, correlation corrections have been added to the transition operator in the next order. The contributions from the higher orders of the theory, that is, frequency-dependent Breit correction, Lamb shift, and mass shifts, have been estimated.

For the present topic of interest, we have extracted the wavelengths of the  $4f^{14} 5s - 5p$  transitions in Pm-like ions with  $74 \leq Z \leq 92$  and present them together with the results of earlier calculations and with various experimental data in table I.

Table II lists the first (lowest) 80 levels of the Pm-like ion  $W^{13+}$ . Among these, the  $4f^{14} 5s$  level, the idealized “ground state” of an alkalilike system, is number 24, while the  $5p$  resonance levels of the alkalilike system would be at positions 112 and 243 (not listed here). Evidently, for Pm-like  $W^{13+}$ , the alkalilike model is not appropriate.

Nevertheless, we have calculated (tables III and IV) the strongest E1 transitions (estimated wavelength uncertainty  $0.25 \text{ \AA}$ ) to be expected in the two wavelength ranges that were covered in observations of W spectra at the Berlin electron beam ion trap [19] (which will be discussed below). The tabulations do not represent any collisional-radiative modeling; considering that the excitation cross sections tend to decrease with higher principal quantum number and excitation energy, lines from high-lying levels are bound to appear more weakly than suggested by transition probabilities alone.

As expected, our calculations show the general atomic structure and its changes along the isoelectronic sequence as they have been obtained and discussed before. However, based on our experience with somewhat simpler ions, we expect our calculational results to be more accurate than any of the calculations of Pm-like spectra that have been published elsewhere. Before we discuss details, however, we would like to stress a key observation: Our calculations do not predict that there should be one or two dominant resonance lines in the spectra of any of the ions investigated experimentally so far. In fact, the richness of the calculated spectra should not allow to make immediate, unambiguous and valid identifications in most of the Pm-like spectra. Apart from the heaviest ions (for which there are very few observations), the spectra are no longer expected to be dominated by just a few lines that arise from  $ns - np$  resonance transitions as is the case in the lighter alkali-like ion spectra. The question then arises to what has been seen and assigned in the few experiments, and what may be

expected from the various light sources in a future, more educated search.

### III. PAST EXPERIMENTS

The experimental search for 5s-5p transitions in Pm-like ions has been based on the expectation that these transitions would produce prominent resonance lines, as is the case in lighter alkali-like systems in which the resonance levels are the lowest excitation levels. In those atomic systems, cascades are funneled to these very same levels, thus boosting the level population and consequently the line intensities. However, due to the fact that the  $4f^{14}5p$  states are actually not the lowest odd-parity levels in the ions up to  $Pb^{21+}$ , those levels are affected by numerous crossings with  $4f^{12}5s^25p$  levels. The admixture of those states is so significant that it requires an extensive multi-reference treatment of the correlation effects in the theoretical approximations. Ions as heavy as  $U^{38+}$  have first excited levels belonging to the  $4f^{14}5p$  and  $4f^{14}5d$  configurations, and those levels are well separated from the  $4f^{13}5s^2$ ,  $4f^{13}5s5p$ , and  $4f^{12}5s^25p$  levels. In those cases, single-reference correlation calculations are sufficient; in the lighter ions of the Pm sequence, they are not.

Another difficulty in the identification of the  $4f^{14}5s - 4f^{14}5p$  transitions in ions with nuclear charge  $74 < Z < 92$  is the fact that large arrays of transitions with one or two vacancies in the core fall into the same wavelength range (this was recognized by Kaufman *et al.* [20]). The transitions  $4f^{13}5s^2 - 4f^{13}5s5p$  and  $4f^{12}5s^25p - 4f^{12}5s5p^2$  produce numerous strong lines that are expected as strong as the  $4f^{14}5s - 4f^{14}5p$  lines or even stronger. As we discuss below with the example of the Bochum beam-foil spectra, such line clusters may actually hide the lines of interest in high-density light sources, whereas spectra obtained at electron beam ion traps (EBIT) are expected to suffer less from multiple excitations. We note that using an EBIT (a very low particle density device) - at that time a truly novel device [21] - has already been suggested in the 1990 report on the Bochum beam-foil study [20].

#### A. Beam-foil spectroscopy

The experimental side of the Pm sequence case began in the late 1970es, with wide humps being observed in EUV spectra of fusion plasma devices like ORMAK at Oak Ridge. The hope was for the identification of individual spectral lines in such spectra, so that



the ionization potential of the associated ion species would yield a measure of the plasma temperature. (A sample of such spectra (in this case, of W) has been shown by Sugar and Kaufman [22] who found a lower degree of ionization from identifying spectral structures than had been assumed before.) Experiments with fast Au ( $Z=79$ ) ions were therefore performed at Brookhaven, sending ion beams through thin carbon foils and measuring the spectra of EUV light emitted by the ions upon leaving the exciter foil [23]. By varying the ion beam energy, in this case from 31 to 238 MeV, the charge state distribution in the ion beam after ion-foil interaction was systematically varied. In this way, the approximate charge state of the ion species that emits a given spectral line might be followed through several energy steps and thus be determined. As it turned out, the beam-foil spectra were also dominated by wide humps. Although no clean lines appeared, the  $5s - 5p$  transitions in the Pm-like spectrum AuXIX were deemed to be possibly present. Other (fruitless) experimental searches for these lines (by laser-produced plasma or in vacuum sparks) have been mentioned by Kaufman *et al.* [20].

After theory had been improved by Theodosiou and Raftopoulos (see above), a new beam-foil measurement was tried at Bochum [24]. In order to handle the very heavy Au ions, a second gas stripper had to be introduced to the Bochum accelerator and its beam transportation system which had been designed to serve only low energy, light ion nuclear physics. Owing to its powerful ion source, the machine nevertheless produced an Au ion beam that was probably more intense than had been available at Brookhaven (where the accelerator otherwise offered the more suitable run parameters), but the ion beam energy could not be varied notably. The Bochum spectroscopic equipment was based on a conventional scanning grazing-incidence monochromator. Spectra were recorded at different foil positions (for different time delays after excitation) and showed two large humps - no surprise there (see figure 3). However, with the instrumental line width well established, several individual lines showed clearly in positions away from the humps, and the humps could be recognized as dense clusters of unresolved lines, now at least known to belong to Au and to a certain range of charge states - whereas in the aforementioned fusion plasma device neither the element nor the charge state range had been determined with any reliability. Taking guidance from the available calculations, two rather weak lines in the Bochum beam-foil spectra (at  $205.8 \text{ \AA}$  and at  $313.7 \text{ \AA}$ ) were tentatively associated with the AuXIX  $5s - 5p_{1/2,3/2}$  transitions. These lines persisted in the delayed spectra that had been recorded

at times of up to 400 ps after excitation. Actually, the Pm-like resonance levels were not expected to be so long lived, but all lighter alkali-like ions show massive cascade tails after foil-excitation of fast ion beams, and if these were the right lines, such cascade repopulation might explain the temporal persistence.

Another four years later, V. Kaufman from NIST Gaithersburg visited the Bochum beam-foil group for an attempt at finding and measuring the  $5s^2 - 5s5p$  intercombination transitions in Sm-, Eu- and Gd-like ions of Os, Ir, Pt, and Au ( $Z=76-79$ ) [20]. The Bochum group had by that time gained experience in identifying the decays of long-lived levels by studying delayed spectra. Taking guidance from new (somewhat adjusted) Cowan code (Hartree-Fock with statistical exchange) calculations by Kaufman, and intercomparing lines in the spectra of four elements, several intercombination transitions were, indeed, assigned. The intercombination transition in Sm-like Au XVIII, however, was found at 313.6 Å, and thus took the place of what earlier on had been assumed to be one of the  $5s - 5p$  transitions in the Pm-like Au XIX spectrum. The general systematics of such  $\Delta n=0$  transitions implicitly displaced the likely position of the Pm-like ion line ( $5s - 5p_{1/2}$ ) towards shorter wavelengths, into the range of one of the humps (and the other Pm-like ion resonance line ( $5s - 5p_{3/2}$ ) to the other hump), but no new wavelengths were given for lack of an identifying signature.

However, the identification of the line in the Sm-like ion, by wavelength (in comparison to improved Cowan code calculations) and by its decay curve characteristics (level lifetime and typical cascade tail) corroborated the general assumptions on the charge state distribution in the Bochum beam-foil experiment. If the Sm-like spectrum was excited (somewhat) prominently, then the Pm-like spectrum (a neighbouring charge state) should show up as well. Judging from the same beam-foil experiment (figure 3), the only prominent line emission in the prompt spectrum occurs in line clusters on the short-wavelength side close to the predicted positions of the two  $5s - 5p$  resonance transitions. Beam-foil excitation takes place in a high-density environment, and multiple excitation, core excitation, and displaced terms figure prominently in beam-foil spectra. One has to expect a multitude of  $5s - 5p$  lines from a variety of core configurations, and from several charge states, forming two line clusters, one for  $5s - 5p_{1/2}$  and the other for  $5s - 5p_{3/2}$  transitions and their respective companion lines. The calculated level lifetimes are well below 100 ps [13], while the observation was for about the first 150 ps after excitation. The relative line intensities in such practically time-integrated observations are then expected to reflect the level populations rather than

individual transition probabilities. If the two Au XIX 5s - 5p lines of interest should appear prominently at all (moderately so at best), they can only have wavelengths that are slightly shorter than predicted by our calculations - there are no nearby unassigned lines at longer wavelengths.

The report by Hutton *et al.* [19] includes one line (5s - 5p<sub>1/2</sub>) from a beam-foil experiment on Pb (Z=82) done at RIKEN (Japan), with a wavelength a little shorter than predicted, just as observed with the candidate lines in the Bochum beam-foil spectrum. The recent paper by Wu and Hutton [26] displays a spectrum recorded in that old and otherwise unpublished experiment. The experiment apparently was performed at the low limit of the accelerator performance envelope, and the spectrum evidently is poor. Unfortunately, the experiment did not yield information on the other line of the pair. Hutton *et al.* also mention an attempt (which had failed) at measuring the lines of interest in an ion-gas collision experiment.

### B. Electron beam ion trap work

Twelve years after the second set of Bochum beam-foil studies of Au, electron beam ion traps had been set up in several laboratories, and Hutton *et al.* [19] reported on identifications of two 5s - 5p lines in the Pm-like spectrum of W (Z=74) as observed in the Berlin EBIT. (The aforementioned recent paper by Wu and Hutton [26] tells more about this and other unpublished old experiments than the original paper, but otherwise adds no new physics information.) Pm-like W has an ionization potential of 310 eV, according to calculations by Scofield [25], whereas Kramida and Reader [27] give 290.7 eV and our own calculations suggest 298.75 eV. None of these values has an experimental corroboration. With an ionization potential of this magnitude in mind, the electron beam energy in the Berlin EBIT was varied in 30 eV steps between 190 and 360 eV in order to find the prominent 5s - 5p lines by a variation of the charge state balance. However, the multi-electron ion spectra are not otherwise known (yet), and predictions of the charge state distribution in the electron beam ion trap are vague. There is no benchmark for telling which, say, five charge states are being reached, produced amply, or already ionized away and burned out. The calculated ionization potential of the Sm-like ion (62 electron) is 287 eV (or about 258 eV, or 251 eV), and that of the Nd-like ion (60 electrons) is 313 eV (or 325 eV, or 335 eV). The electron beam energy width of 30 to 50 eV easily spans the ionization energies of more than

one charge state; optimum excitation occurs not at the ionization limit of the lower charge state, but at some energy above threshold. As soon as Pm-like ions are produced with some efficiency at an electron beam energy above the threshold, it is likely that they are beginning to be also ionized, and so may be the next higher charge state, too. In short, there is no better technique than the recording of spectra with step-wise increases of the electron beam energy yet (as has been practiced at the Berlin EBIT [28]), but in the low electron beam energy range and with such narrowly spaced ionization potentials and such many-electron spectra, this technique cannot produce conclusive evidence that the right charge state has been reached (and not already overstepped), unless spectral lines that belong to a given charge state can be identified. There is no valid corroboration yet of the tungsten line identities assumed as guidance for this process at the Berlin EBIT.

Hutton *et al.*, however, have interpreted the spectroscopic evidence as showing both W XIV lines of primary interest. The  $5s - 5p_{1/2}$  transition is claimed to lie at a wavelength slightly shorter than predicted (0.3% away), whereas the  $5s - 5p_{3/2}$  transition is reported at a wavelength about 2% shorter than calculated (see table I). This seems inconsistent, considering our experience with our calculations when applied to the spectra of simpler ions. Beyond this inconsistency, the question arises whether maybe only one of the two lines is from Pm-like W, or possibly none at all.

As discussed above, the  $4f^{14}5s$  level is by far not the ground state in Pm-like W ions. EBIT as a low-density light source is known to populate almost exclusively levels than can be excited directly from the ground state, because there is plenty of time for relaxation towards the ground state ( $4f^{13}5s^2 \ ^2F_{7/2}$  and  $\ ^2F_{5/2}$  in this case, see figures 1 and 2). We have calculated the line pattern (line intensity pattern) for W XIV lines that we would expect in the vicinity of the  $5s - 5p_{1/2}$  transition - the spectral section shown by Hutton *et al.* [19] - and we see no obvious similarity. The same holds for the vicinity of the  $5s - 5p_{3/2}$  transition that is shown in the paper by Wu and Hutton [26], but on a poor scale. (We had larger scale plots of the Berlin EBIT data at our disposal [28].)

Consequently, we are not convinced that those EBIT observations show the lines of interest at all. We have therefore undertaken calculations of other charge state spectra as well, a systematic presentation of those calculational results is in progress), and we find that some of the lines reported by Hutton *et al.* [19] match the wavelengths of transitions predicted for the Gd-like (W XI) through Nd-like ions (W XV), but not the “alkalilike”  $5s - 5p$  lines in the

Pm-like spectrum (W XIV) - which are not ground state transitions and therefore should not be expected to show strongly in EBIT spectra. Nevertheless, our calculations indicate that Pm-like spectra of W have, indeed, been produced at the Berlin EBIT [28], and that several of the prominent lines in those spectra can be identified (see table V). These are just not the elusive signature lines of ions in the alkali isoelectronic sequence. Further line identifications are expected from ongoing calculations of the spectra of neighboring charge states of W.

Considering the importance of tungsten as a wall material in the ITER fusion energy project under construction (see [29, 30]) a better experimental coverage of the EUV spectra in an EBIT which is run at the appropriately low electron beam energies is highly desirable (and is underway at Berlin).

### C. Tokamak observation

The heaviest element of which a line has been assigned to the  $5s - 5p_{3/2}$  transition in a Pm-like ion so far is uranium. Contrary to the claim by Hutton *et al.* [19] that this observation was from a (high density) laser produced plasma, the cited work by Fournier *et al.* [31] actually concerned a (low density) tokamak plasma. Based on the atomic structure, the resonance line character of the  $5s - 5p$  lines should be most pronounced in the heaviest ions. Only one candidate line is considered, and the associated wavelength is slightly longer than predicted. However, in a tokamak there is no easy way of actually determining the charge state of a highly charged ion, and thus the proximity of the single line wavelength to a calculated one of a specific ion may be fortuitous. Fournier *et al.* clearly state the practical problems, and they note that according to their collisional-radiative model calculations, even in uranium the Pm-like ion resonance lines are expected to be accompanied by stronger lines from the  $4d^{13} 5s^2 - 5s5p$  array and other, similar satellites. Their spectra contain many unassigned lines, including such in the vicinity of our calculated line positions, but these lines are not clear enough to venture positive identifications.

## IV. DISCUSSION

So, what do we learn from all this about the Pm isoelectronic sequence?

To our present judgment, the only three promising line identifications of  $5s - 5p$  “alkaline ion resonance lines” are those in beam-foil spectra of Au [24] (two lines, based on the revised identifications [20]) and Pb [19] (one line). The calculations presented by Hutton *et al.* [19], some of which were using an early version of the presently employed algorithm, were clearly closer to the newly defined small data set than the original calculational results given by Curtis and Ellis [13], more than 20 years earlier. (Those predictions are now superseded by our present calculations.) The data for the two W lines put forward by Hutton *et al.*, however, differ from prediction by very different amounts, and the data point for U differs from prediction with a different sign (longer wavelength than predicted). Hutton *et al.* also listed the 1982 Bochum results, noting the discrepancy from the isoelectronic trend in comparison to theory, but overlooking the fact that the lines had been reassigned to another ion twelve years before their analysis. Only this last error of judgment has been corrected by Wu and Hutton [26]. Evidently, the mixed batch of suggested identifications presented by Hutton *et al.* is not intrinsically consistent and therefore hardly convincing; of the six lines used by Hutton *et al.* for a comparison to theory, five are known to be wrongly associated with  $4f^{14} 5s - 5p$  transitions or at least strongly suspected to be.

All three “surviving” experimental wavelengths are about 1% shorter than our calculations predict. That is less of an agreement than we have found in our work on fewer-electron ions, but we also note that the three experimental wavelengths are not particularly accurate (the  $1 \text{ \AA}$  measurement uncertainty corresponds to about 0.3 to 0.5%). For most of the identifiable lines in the Berlin EBIT spectra of W [28], the difference between our calculated results and their observed line positions is much less than that.

There certainly is room for improvement on the experimental side. However, some of the earlier options have gone away, since beam-foil spectroscopy, with its advantages of an adjustable charge state distribution and mono-isotopic spectra, has gone out of fashion. The only practical option nowadays seems to be the use of an electron beam ion trap. This device also offers an adjustable charge balance similar to the beam-foil interaction process, but EBITs work better for more highly charged ions. Owing to the atomic structure changes of Pm-like ions along the isoelectronic sequence (see figure 2), such studies in search for the alkalilike ion  $5s - 5p$  transitions would best start with U and then work their way towards lighter elements. Pm-like  $U^{31+}$  has an ionization potential of about 1.1 keV [25] and is accessible at all present-day EBITs.

## Acknowledgments

We thank C. Biedermann (Berlin) and R. Hutton (now at Shanghai) for access to unpublished material, and P. Beiersdorfer (Livermore) for helpful advice on the manuscript. This research was supported in part by the U.S-Israel Binational Science Foundation. ET acknowledges travel support from the German Research Association (DFG). Some of this work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory in part under Contract W-7405-Eng-48 and in part under Contract DE-AC52-07NA27344.

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TABLE I: Theoretical and experimental wavelengths (in Å) of the  $5p - 5s$  electric dipole transitions. Lamb shifts and frequency-dependent first-order Breit corrections are included in MP2 and MR-MP. Scaled hydrogenic Lamb shifts are included in the Dirac-Fock (DF) calculations.

Z	DF <sup>a</sup>	HFR <sup>b</sup>	Curtis <sup>c</sup>	RCI	MP2	MR-MP	MR-MP <sup>f</sup>	Expt
<hr/>								
$4f^{14}5s^2S_{1/2}$			$-4f^{14}5p_{-2}P_{1/2}$					
74	370.79	366.2	379	372.25	369.45	369.14	367.41	365.3 <sup>d</sup>
75				354.16	350.22			
76		334.9 <sup>c</sup>		336.65	332.26			
77	319.21	318.9 <sup>c</sup>	325	319.69	320.78	319.81		
78		304.4 <sup>c</sup>		305.05	305.16	305.13		
79	290.33	291.0 <sup>c</sup>	297	291.60	291.53	291.49		289*
80				279.19	279.12	279.08		
81				266.47	267.68	267.66		
82	255.70	251.5	263	257.05	257.07	257.05		
92	179.63	175.1	186	180.96	181.36	181.37		175.4 <sup>d</sup>
<hr/>								
$4f^{14}5s^2S_{1/2}$			$-4f^{14}5p_{-2}P_{3/2}$					
74	262.42	263.7	277	262.30	261.06	259.62	258.77	258.2 <sup>d</sup>
75				240.49	237.82			
76		227.8 <sup>c</sup>		224.24	221.42			
77	212.38	212.5 <sup>c</sup>	226	212.49	212.94	212.30		
78		198.6 <sup>c</sup>		198.78	199.26	198.77		
79	186.43	185.9 <sup>c</sup>	199	186.24	186.95	186.14		184*
80				174.72	174.69	174.67		
81				164.30	164.12	164.11		
82	154.98	154.5	166	154.30	154.36	154.35		153.0 <sup>d</sup>
92	87.25	86.2	96	87.16	87.44	87.44		

<sup>a</sup> Dirac-Fock calculations with 1s, 2s and 2p analytical Lamb shifts [14]

<sup>b</sup> Hartree-Fock Relativistic calculation using semiempirical methods [19, 20]

<sup>c</sup> Hartree-Fock calculation with semiempirical relativistic corrections [13]

<sup>d</sup> Hutton unpublished identifications of the experimental wavelengths [19]

<sup>e</sup> Semiempirical calculations from [20]

<sup>f</sup> Calculations based on a much larger Relativistic Configuration Interaction model

\* Observation in [24], identification suggested on the basis of this work

TABLE II: Multi-reference Møller-Plesset (MR-MP) calculated energies and lifetimes  $\tau$  of some of the  $4f^{14}5s$ ,  $4f^{14}5p$ ,  $4f^{13}5s^2$ ,  $4f^{13}5s5p$ ,  $4f^{13}5p^2$ ,  $4f^{12}5s^25p$ ,  $4f^{12}5s5p^2$ , and  $4f^{13}5p^3$  levels in Pm-like tungsten. The term values are listed relative to the  $4f^{13}5s^2\ ^2F_{7/2}^o$  ground state. Five numbers in the first and fifth columns denote occupation numbers (Occ) of the relativistic one-electronic shells  $4f_{5/2}$ ,  $4f_{7/2}$ ,  $5s_{1/2}$ ,  $5p_{1/2}$  and  $5p_{3/2}$ . The alkalilike  $4f^{14}5s$ ,  $5p_{1/2}$  and  $5p_{3/2}$  levels have occupation numbers 68100, 68010 and 68001, respectively, and are at the positions 24, 112 and 243 of a table of which the first 80 entries are listed below. Column "Key" denotes quantum number  $J$  and the sorting number of the RCI energy level followed by a parity symbol (star indicates odd parity).

Occ	Key	E (cm <sup>-1</sup> )	$\tau$ (s)	Occ	Key	E (cm <sup>-1</sup> )	$\tau$ (s)
67200	7/2(1)*	0	0.000D+00	57201	9/2(8)*	217807	6.395D-05
58200	5/2(1)*	25726	3.820D-03	66201	5/2(9)*	218850	6.047D-05
66210	11/2(1)*	70613	4.281D+00	57201	1/2(4)*	222432	3.748D-05
66210	7/2(2)*	79737	9.430D-01	57201	9/2(9)*	222815	6.411D-05
66210	9/2(1)*	82904	4.488D-01	57201	3/2(6)*	223465	5.726D-05
57210	11/2(2)*	90878	1.372D-01	66201	7/2(10)*	224353	6.444D-05
57210	9/2(2)*	91228	4.733D-03	57201	5/2(10)*	226617	6.679D-05
57210	7/2(3)*	96057	2.025D-02	48201	7/2(11)*	229062	6.461D-05
66210	3/2(1)*	97235	3.092D-01	48201	11/2(8)*	229783	6.601D-05
57210	5/2(2)*	97429	2.945D-02	48201	9/2(10)*	231697	6.908D-05
57210	9/2(3)*	99745	5.324D-03	57201	5/2(11)*	235207	6.357D-05
57210	5/2(3)*	100591	8.027D-03	67110	7/2(1)	240734	1.054D-08
57210	7/2(4)*	104739	4.306D-03	48201	3/2(7)*	241271	2.025D-05
48210	9/2(4)*	111552	5.246D-03	66201	1/2(5)*	242117	5.333D-05
57210	3/2(2)*	118350	6.419D-03	66201	7/2(12)*	242647	6.328D-05
48210	7/2(5)*	119729	2.004D-03	57201	9/2(11)*	243110	5.666D-05
48210	5/2(4)*	120253	4.920D-03	66201	3/2(8)*	248422	6.131D-05
57210	1/2(1)*	128422	3.919D-02	48201	5/2(12)*	250975	5.885D-05
66210	1/2(2)*	128741	1.789D-02	57201	1/2(6)*	256389	1.132D-05
57210	11/2(3)*	131976	2.698D-03	67110	9/2(1)	256850	4.421D-10
57210	3/2(3)*	135855	3.064D-03	57201	11/2(9)*	257187	8.132D-05
48210	5/2(5)*	138285	2.622D-03	57201	5/2(13)*	258294	6.253D-05
48210	3/2(4)*	141773	1.755D-03	66201	3/2(9)*	258720	4.732D-05
68100	1/2(1)	177727	1.165D-05	67110	7/2(2)	260091	6.077D-10
66201	11/2(4)*	191771	8.330D-05	48201	7/2(13)*	260953	6.273D-05
66201	9/2(5)*	192132	6.553D-05	58110	5/2(1)	262398	2.035D-09
48210	1/2(3)*	193272	3.905D-04	67110	5/2(2)	264102	4.384D-10
66201	9/2(6)*	198170	6.983D-05	57201	3/2(10)*	264434	3.882D-05
57201	7/2(6)*	198896	7.022D-05	48201	1/2(7)*	270816	3.512D-06
66201	5/2(6)*	201930	6.564D-05	58110	5/2(3)	283410	5.582D-10
66201	7/2(7)*	203617	6.799D-05	58110	7/2(3)	284946	4.251D-10
66201	11/2(5)*	203805	7.243D-05	58110	3/2(1)	287106	4.101D-10
57201	9/2(7)*	209261	6.333D-05	48201	3/2(11)*	316058	1.195D-06
66201	5/2(7)*	210275	6.897D-05	66120	11/2(1)	341992	6.290D-10
57201	5/2(8)*	213762	6.908D-05	67101	11/2(2)	350418	4.901D-08
57201	11/2(6)*	213842	6.856D-05	66120	9/2(2)	358160	2.138D-10
57201	11/2(7)*	215100	6.509D-05	57120	9/2(3)	359121	2.598D-10

TABLE III: Compilation of strongest MR-MP electric-dipole transitions of Pm-like tungsten in the wavelength range 240-300 Å. Column “Key” denotes the upper and lower levels, respectively, by quantum number  $J$  and the sorting number of the RCI energy level followed by a parity symbol (\* indicating odd parity). The term energies of the lower levels denoted by 5/2(9), 11/2(39) and 5/2(63) are 410375, 643102 and 683492  $\text{cm}^{-1}$ , correspondingly. Remaining lower levels are listed in Table II. Level lifetime  $\tau$  (column 3) and transition probability  $A$  (column 5) can be combined to yield the decay probability weighted with the branch fraction (column 6).

$\lambda$ Å	Key upper	$\tau$ $10^{-11}$ s	Key lower	$A$ $10^{10}$ $\text{s}^{-1}$	$A^2\tau$ $10^{10}$ $\text{s}^{-1}$
240.150	3/2( 26)	1.758	3/2( 2)*	4.30	3.26
240.277	1/2( 17)	1.436	1/2( 1)*	0.55	0.044
240.461	1/2( 17)	1.436	1/2( 2)*	4.18	2.50
240.814	3/2( 27)	1.356	1/2( 1)*	0.99	0.13
241.465	1/2( 26)	1.042	3/2( 7)*	4.79	2.38
241.836	5/2( 13)	1.416	5/2( 1)*	6.92	6.78
241.866	7/2( 29)*	1.669	5/2( 9)	2.83	1.33
241.895	3/2( 21)	8.661	3/2( 1)*	0.19	0.032
242.671	3/2( 23)	2.026	5/2( 2)*	3.21	2.09
243.583	1/2( 13)	1.854	3/2( 1)*	4.72	4.14
243.765	11/2( 16)	11.77	9/2( 1)*	0.12	0.016
243.880	11/2( 24)	1.616	9/2( 4)*	3.63	2.12
244.074	11/2( 14)	17.15	9/2( 1)*	0.092	0.015
244.268	1/2( 16)	1.815	1/2( 1)*	0.84	0.13
244.483	3/2( 19)	20.22	3/2( 1)*	0.084	0.014
244.795	5/2( 10)	1.491	7/2( 1)*	6.62	6.52
245.055	9/2( 8)	1.462	7/2( 1)*	6.83	6.81
246.083	1/2( 15)	2.201	3/2( 2)*	2.74	1.65
246.102	3/2( 26)	1.758	1/2( 1)*	0.26	0.012
248.913	7/2( 13)	1.493	5/2( 1)*	5.05	3.81
249.236	11/2( 13)	18.40	9/2( 1)*	0.091	0.015
249.961	9/2( 12)*	1.701	7/2( 1)	3.47	2.05
251.384	1/2( 14)	13.35	1/2( 1)*	0.24	0.075
252.337	1/2( 15)	2.201	1/2( 1)*	1.37	0.42
252.382	3/2( 8)	1.601	5/2( 1)*	6.24	6.23
253.990	7/2( 10)	1.387	5/2( 1)*	1.30	0.24
258.773	3/2( 12)*	2.639	1/2( 1)	3.73	3.68
261.031	9/2( 5)	235.2	7/2( 1)*	0.032	0.024
262.407	3/2( 23)	2.026	1/2( 1)*	0.24	0.012
266.890	3/2( 50)*	0.796	3/2( 53)	7.76	4.80
269.109	7/2( 5)	228.2	7/2( 1)*	0.028	0.018
269.293	11/2( 37)*	1.137	11/2( 39)	4.42	2.22
271.748	5/2( 4)	185.1	7/2( 1)*	0.027	0.014
272.022	5/2( 59)*	0.881	5/2( 63)	5.66	2.82
274.396	7/2( 8)	118.9	5/2( 1)*	0.051	0.031
282.585	3/2( 4)	199.27	5/2( 1)*	0.040	0.032

TABLE IV: Compilation of strongest MR-MP electric-dipole transitions of Pm-like tungsten in the wavelength range 330 - 420 Å. Column “Key” denotes the upper and lower levels, respectively, by quantum number  $J$  and the sorting number of the RCI energy level followed by a parity symbol (\* indicating odd parity). The term values of the lower levels denoted by 3/2(8) and 9/2(8) are 421951 and 408072  $\text{cm}^{-1}$ , correspondingly. Remaining lower levels are listed in Table II. Level lifetime  $\tau$  (column 3) and transition probability  $A$  (column 5) can be combined to yield the decay probability weighted with the branch fraction (column 6).

$\lambda$ Å	Key upper	$\tau$ $10^{-11}$ s	Key lower	$A$ $10^9$ $\text{s}^{-1}$	$A^2\tau$ $10^9$ $\text{s}^{-1}$
336.633	7/2( 14)*	2.77	9/2( 1)	16.55	7.63
344.378	9/2( 12)*	1.70	11/2( 2)	13.15	2.93
344.744	5/2( 14)*	2.80	7/2( 3)	14.12	5.61
347.331	5/2( 14)*	2.80	3/2( 1)	8.60	2.07
351.637	5/2( 7)	19.71	3/2( 1)*	0.93	0.17
352.846	5/2( 3)	55.82	7/2( 1)*	0.48	0.13
359.144	7/2( 4)	21.12	9/2( 1)*	3.01	1.91
359.166	9/2( 2)	21.38	7/2( 2)*	2.59	1.44
360.577	3/2( 11)	19.91	5/2( 6)*	3.97	3.15
363.298	9/2( 2)	21.38	9/2( 1)*	1.73	0.64
364.014	3/2( 14)	19.21	5/2( 7)*	2.69	1.40
364.574	3/2( 7)	20.10	5/2( 5)*	2.94	1.75
367.409	1/2( 8)*	7.49	1/2( 1)	13.34	13.33
368.244	7/2( 9)	22.20	9/2( 4)*	2.72	1.64
368.370	1/2( 4)	19.77	3/2( 3)*	2.55	1.28
372.796	9/2( 3)	25.98	11/2( 2)*	2.25	1.32
374.267	9/2( 7)	21.51	7/2( 5)*	2.36	1.20
375.157	3/2( 5)	23.85	5/2( 4)*	2.47	1.46
378.642	5/2( 2)	43.84	7/2( 1)*	1.96	1.70
378.969	3/2( 3)	25.62	3/2( 1)*	0.85	0.18
379.248	3/2( 3)	25.62	5/2( 2)*	1.63	0.69
379.606	1/2( 6)	29.23	1/2( 3)*	2.78	2.26
381.100	5/2( 1)	203.5	7/2( 1)*	0.46	0.42
382.585	3/2( 1)	41.01	5/2( 1)*	2.43	2.43
384.481	7/2( 2)	60.77	7/2( 1)*	1.57	1.49
384.497	3/2( 6)	25.11	3/2( 3)*	1.57	0.62
385.773	7/2( 3)	42.51	5/2( 1)*	2.21	2.08
387.615	1/2( 3)	28.01	1/2( 1)*	0.75	0.16
388.072	5/2( 3)	55.82	5/2( 1)*	1.31	0.96
388.095	1/2( 3)	28.01	1/2( 2)*	1.89	1.01
388.143	1/2( 9)	27.42	3/2( 6)*	2.53	1.76
389.332	9/2( 1)	44.21	7/2( 1)*	2.26	2.26
399.484	1/2( 9)*	2.43	3/2( 8)	8.16	1.62
400.094	1/2( 7)	34.53	1/2( 4)*	1.91	1.26
414.855	11/2( 10)*	2.35	9/2( 8)	7.03	1.16

TABLE V: Calculated (MR-MP) lines of Pm-like tungsten  $W^{13+}$  in the wavelength range 240 - 420 Å region and nearby lines in the Berlin EBIT spectra [28]. Column “Key” identifies upper and lower levels by quantum number  $J$  and the sorting number of the RCI energy level followed by a parity symbol (\* indicating odd parity). The branched transition probability  $A^2\tau$  is listed in the last column.

$\lambda$ (Å)	Conf	Key	$\tau$ (fs)	Conf	Key	$A^2\tau$	
EBIT	MR-MP	upper level		lower level		$10^9 \text{ s}^{-1}$	
253.84	253.99	5s5p	7/2(10)	13.9	5s <sup>2</sup>	5/2(1)*	2.4
258.38 <sup>a</sup>	258.77	5p	3/2(12)*	26.4	5s	1/2(1)	36.8
261.10	261.03	5s5p	9/2( 5)	2353	5s <sup>2</sup>	7/2(1)*	0.24
272.05	271.75	5s5p	5/2( 4)	1851	5s <sup>2</sup>	7/2(1)*	0.14
	359.14	5s5p <sup>2</sup>	7/2( 4)	211.2	5s <sup>2</sup> 5p	9/2(1)*	1.9
	360.58	5s5p <sup>2</sup>	3/2(11)	199.1	5s <sup>2</sup> 5p	5/2(6)*	3.2
	364.57	5s5p <sup>2</sup>	3/2( 7)	201.0	5s <sup>2</sup> 5p	5/2(5)*	1.8
	367.41	5p	1/2( 8)*	74.9	5s	1/2(1)	13.3
389.78	389.33	5s5p	9/2(1)	442.1	5s <sup>2</sup>	7/2(1)*	2.3

<sup>a</sup> The line at 258.38 Å corresponds to either the  $W^{12+}$  5s5p  $^1P_1^o - 5s^2$   $^1S_0$  transition predicted at 258.67 Å or the  $W^{11+}$  5s5p<sup>2</sup>  $^2S_{1/2} - 5s^2$ 5p  $^2P_{1/2}^o$  transition predicted at 258.09 Å.

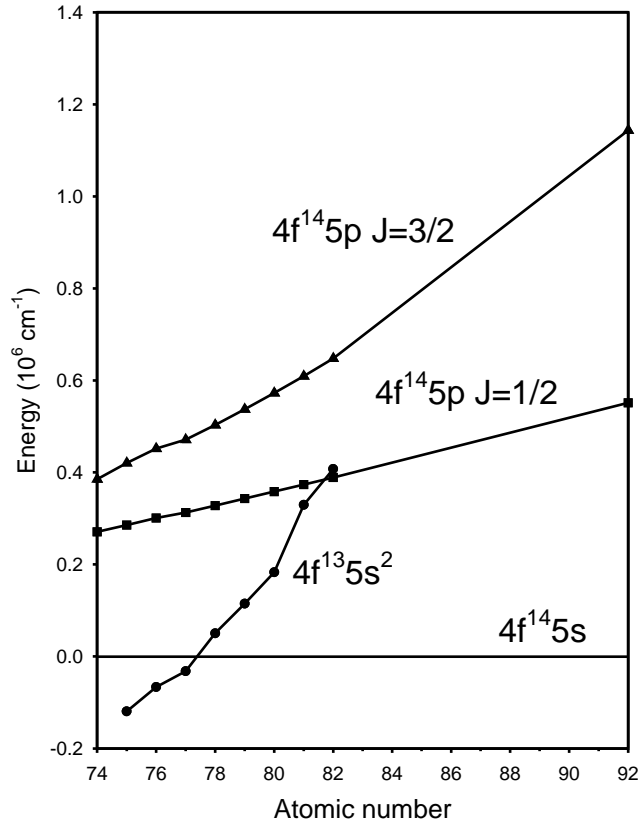


FIG. 1: MR-MP energies of the lowest levels in the Pm-like ions relative to the  $4f^{14}5s$  level.

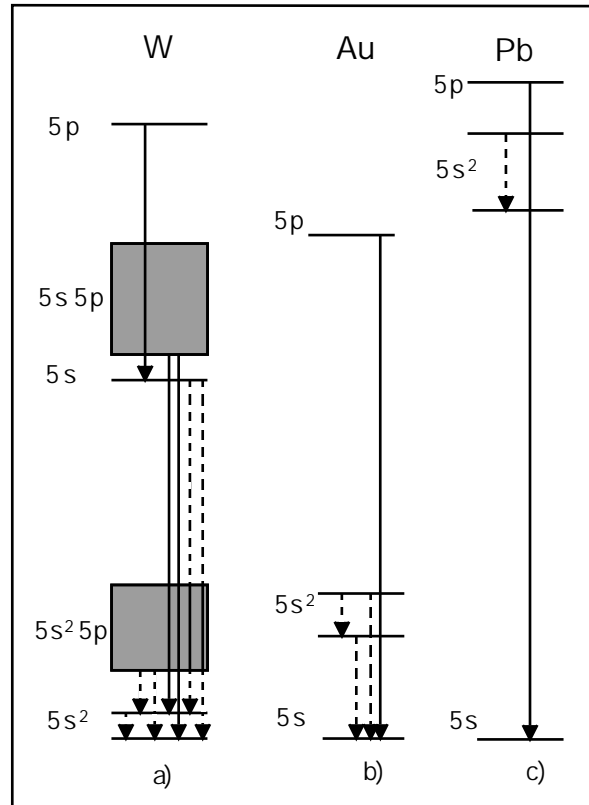


FIG. 2: Grotrian diagrams of Pm-like tungsten (plot a), gold (plot b) and lead (plot c). Solid arrows denote allowed transition and the dotted arrows denote electric-dipole forbidden transition.

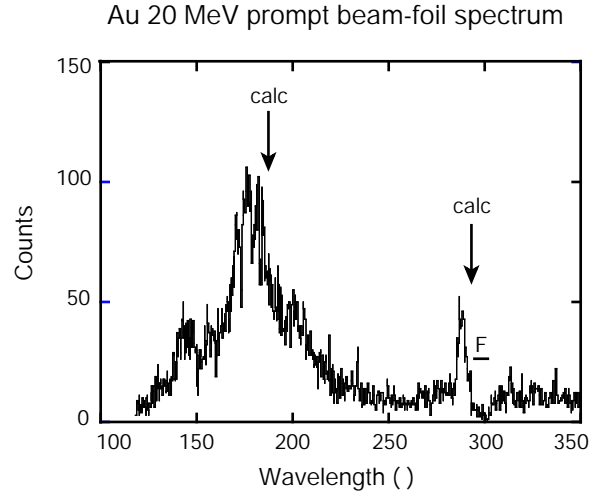


FIG. 3: Beam-foil spectrum of Au at an ion beam energy of 20 MeV (The data are the same as in [24].) The instrumental line width is less than 4 channels. Near 'F', the exciter foil suffered damage during the spectral scan and was replaced after some 30 channels. The arrows mark the calculated positions of the  $4f^{14} 5s - 5p$  transitions in the Pm-like spectrum Au XIX. The lines that we suggest to associate with these transitions are nearby at slightly shorter wavelengths.