Atoms **2015**, *3*, 76-85; doi:10.3390/atoms3020076

OPEN ACCESS
atoms
ISSN 2218-2004
www.mdpi.com/journal/atoms

Article

Collisional-Radiative Modeling of Tungsten at Temperatures of 1200–2400 eV

James Colgan [1,*], Christopher J. Fontes [2], Honglin Zhang [2] and Joseph Abdallah, Jr. [1]

[1] Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA; E-Mail: abd@lanl.gov

[2] Computational Physics Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA; E-Mails: cjf@lanl.gov (C.J.F.); zhang@lanl.gov (H.L.Z)

* Author to whom correspondence should be addressed; E-Mail: jcolgan@lanl.gov; Tel. +1-505-665-0291.

Academic Editor: Bastiaan J. Braams and Alfred Müller

Received: 4 March 2015 / Accepted: 17 April 2015 / Published: 30 April 2015

Abstract: We discuss new collisional-radiative modeling calculations of tungsten at moderate temperatures of 1200 to 2400 eV. Such plasma conditions are relevant to ongoing experimental work at ASDEX Upgrade and are expected to be relevant for ITER. Our calculations are made using the Los Alamos National Laboratory (LANL) collisional-radiative modeling ATOMIC code. These calculations formed part of a submission to the recent NLTE-8 workshop that was held in November 2013. This series of workshops provides a forum for detailed comparison of plasma and spectral quantities from NLTE collisional-radiative modeling codes. We focus on the LANL ATOMIC calculations for tungsten that were submitted to the NLTE-8 workshop and discuss different models that were constructed to predict the tungsten emission. In particular, we discuss comparisons between semi-relativistic configuration-average and fully relativistic configuration-average calculations. We also present semi-relativistic calculations that include fine-structure detail, and discuss the difficult problem of ensuring completeness with respect to the number of configurations included in a CR calculation.

Keywords: tungsten, fusion plasma, collisional-radiative modeling

---

1. Introduction

The properties of tungsten are of prime importance for the future of magnetic fusion modeling. Tungsten has been proposed as a plasma-facing device for the ITER machine [1] due to its high melting point, and its low affinity for tritium. However, the high radiative losses of tungsten mean that its presence in the fusion plasma must be limited to as small as possible. It is also therefore critical to understand the atomic properties of tungsten and the plasma kinetics that give rise to the radiative losses over the large temperature ranges that will be found in ITER.

In this submission, we report on collisional-radiative modeling calculations for tungsten made using the Los Alamos ATOMIC plasma kinetics code. We focus on a relatively small temperature range of 1200 to 2400 eV as tungsten plasma at similar temperatures was investigated experimentally using ASDEX Upgrade in recently reported work [2]. In this temperature range, the most abundant tungsten ions contain many electrons in open N-shell configurations, resulting in potentially vast numbers of atomic levels that require consideration. This makes the calculation of atomic data and the resulting plasma kinetics modeling extremely complex and computationally intensive. However, the investigations conducted on ASDEX Upgrade revealed potentially important emission in the $\sim$6 nm wavelength region that was not predicted in the accompanying modeling, motivating further calculations for tungsten plasma at conditions of relevance. The modeling of tungsten in this temperature range was the focus of a recent non-LTE workshop (NLTE-8) held in November 2013 [3], which formed part of a series of workshops devoted to comparison of codes dedicated to non-LTE kinetics modeling [4]. Tungsten has been a test case for many of the non-LTE kinetics workshops [5,6], and also the subject of various investigations using EBIT measurements (for example, [7]).

2. Calculation of Atomic Data for Tungsten Using the Los Alamos Suite of Codes

We performed several sets of atomic data calculations for tungsten in an effort to gauge sensitivity to various approximations inherent to collisional-radiative modeling. Several sets of calculations were made using the Los Alamos suite of semi-relativistic codes, which has been described in [8,9]. Calculations were made using differing lists of configurations to test the convergence of the tungsten plasma properties to the completeness of the number of configurations included. We also performed semi-relativistic calculations that include fine-structure detail in the predicted emission using our mixed-unresolved-transition-array (MUTA) approach [10]. This approach allows significantly more detail to be captured in the emission with a modest increase in computational requirements.

Calculations were also performed using the Los Alamos suite of fully-relativistic atomic structure codes. The Los Alamos fully relativistic approach to collisional-radiative modeling has been recently described in detail [9]. In this work, we used a relativistic configuration-average approach and tested whether the addition of higher-order multipole transitions (beyond the inclusion of dipole E1 radiative transitions) were significant for tungsten at the temperatures and densities considered here. Our previous study of tungsten [9] showed that the inclusion of higher-order multipole transitions makes a significant difference to the plasma kinetics and subsequent emission spectrum at an electron temperature of 5 keV.

2.1. Semi-Relativistic Calculations

The semi-relativistic Los Alamos calculations begin from the CATS atomic structure code [8], that are based on Cowan's codes [11]. CATS computes common atomic structure quantities such as configuration (or level) energies, transition probabilities, and statistical weights, and also computes plane-wave-Born collision strengths for electron-impact excitation. Ionization cross sections (*i.e.*, including photoionization, electron-impact ionization, and autoionization) are computed using the GIPPER code [12] that takes as input the semi-relativistic wavefunctions generated from CATS. The ion stages of relevance to the tungsten collisional-radiative modeling discussed here contain between 26 and 46 electrons. Many of these ion stages have open p or d sub-shells in their ground configuration and also these ion stages may have many configurations that are relatively closely spaced in energy. The first of these considerations can imply that the configurations likely spawn very large numbers of fine-structure levels, and the second consideration implies that one must include a large number of configurations to ensure that all populations are properly accounted for. Thus, the collisional-radiative modeling of such ion stages quickly becomes a formidable computational challenge.

We first performed calculations in configuration-average mode in an effort to test whether convergence has been attained with respect to the number of configurations included in our calculations. The number of configurations included in our calculations was considerable, and the list of configurations included is best summarized symbolically via super-configuration notation. For example, for Zr-like W (40 electrons), we retain configurations of the type $3^{18}4^{12}$, $3^{18}4^{11}(5-10)^1$, $3^{18}4^{10}5^2$, $3^{18}4^{10}5^1(6-10)^1$, $3^{18}4^95^3$, $3^{18}4^95^2(6-10)^1$, $3^{17}4^{13}$, and $3^{17}4^{12}(5-10)^1$. Here, 4^{10} represents all possible permutations of 10 electrons in the $n=4$ shell [*i.e.*, $4s^24p^64d^2$, $4s^24p^64d^14f^1$, $4s^24p^64f^2$, $4s^24p^54d^24f^1$, $\cdots$, $4f^{10}$]. The only restriction imposed on this list is that when more than one electron is excited out of the $n=4$ shell, no more than 6 electrons are allowed to occupy the $4f$ subshell. This choice of parameters resulted in more than 110,000 configurations for this single ion stage. Similar lists of configurations were created for the other ion stages of interest here (and we label this set of calculations as Model A). Although this choice of configurations resulted in extremely large atomic datasets that are time-consuming to create (as well as occupying significant disk space), there is no guarantee that this list is converged, *i.e.*, that the addition of more configurations would not change the ion populations significantly. To test for convergence, we performed several additional calculations. We first removed some configurations from the original model by limiting the range of single-electron excitations to $n=8$, not $n=10$, as in the original model (we label this set of calculations Model B). We also constructed a new model calculation that included two-electron promotions from the $n=3$ shell, where both electrons may be placed into the $n=4$ shell or one in the $n=4$ shell and one in the $n=5-10$ shells (Model C). We finally constructed another model calculation which was based on Model C, but also allowed two-electron promotions from the $n=4$ shell into the $n=6$ shell (as well as one electron into $n=6$ and one into $n=7-10$), labelled as Model D. We investigated the change in the average ionization and the radiative power loss of these various calculations, again all in the configuration-average approximation. We found that the average ionization from the Model B calculations increased by around 1% and that the radiative power loss decreased by around 4% compared to the Model A calculations. The Model C calculations resulted in average ionization and radiative power loss values that are almost identical to those from Model A,

indicating that the two-electron excitations from the M-shell had little or no effect at the temperatures considered here. The Model D calculations produced average ionization values that were well within 1% of the average ionization values from Model A, and produced radiative power loss values that were just 1%–2% higher than those from Model A.

This sensitivity study led us to tentatively conclude that the list of configurations included in our Model A calculations for tungsten is reasonably well converged, at least with respect to the ionization balance and integrated radiative power loss quantities. Based on this, we then performed MUTA structure calculations using the Model A list of configurations. These calculations compute fine-structure resolved transition probabilities for all dipole-allowed transitions between any two configurations in the list. The fine-structure calculations are based on an intermediate-coupling approach, where one includes interactions between levels arising from the same configuration, but does not include interactions between levels arising from different configurations (*i.e.*, full configuration-interaction). This set of calculations was quite time-consuming (although not prohibitively so) and also produced large sets of data, for which care must be taken in terms of storage and data access. This set of MUTA calculations resulted in several billions of level-resolved transitions that were then included in the emission calculations presented in Section 3. In constructing these models, we note that we still perform our collisional-radiative modeling calculations based on the configuration-average approach, with the level populations used in the fine-structure calculations being obtained via a statistical splitting approach. This approach may not be accurate if, for example, metastable populations are important in the kinetics of the plasma under consideration.

2.2. Fully-Relativistic Calculations

The fully-relativistic calculations start from the RATS Dirac-Fock-Slater atomic structure code that is based on the Penn State University codes of Sampson and co-workers [13]. The GIPPER ionization code [9,12,14] computes the required ionization cross sections using the fully-relativistic wavefunctions generated by RATS. Calculations were performed in the relativistic configuration-average mode. Although the input to the fully-relativistic path in the Los Alamos codes begins with a list of non-relativistic configurations, such a list spawns a much larger number of relativistic configurations, making this path quite computationally intensive. We thus used a model that retains a smaller number of non-relativistic configurations than used in the semi-relativistic calculations. This model includes configurations with sub-shells up to and including $n = 8$ and contains a limited number of doubly excited configurations. The fully relativistic configuration-average calculations were performed in two modes; the first retained only dipole-allowed (E1) radiative transitions in the collisional radiative modeling, and the second set of calculations included all possible radiative transitions from electric dipole through magnetic octupole, *i.e.*, E1 through M3. Such calculations were made due to the unexpectedly prominent M3 transition that was observed in fusion measurements at ASDEX Upgrade at higher temperatures [2].

3. Collisional-Radiative Modeling Using ATOMIC

The Los Alamos ATOMIC code [15,16] can accept data from either the semi-relativistic or fully-relativistic calculations that have just been described. The generated atomic data are stored in a

platform-independent binary format that allows large datasets to be handled in an efficient manner by ATOMIC, allowing us to create quite large atomic models. ATOMIC calculations were performed at an electron density of 10^{14} cm^{-3}, the usual density found in magnetic fusion plasmas. Calculations were made at electron temperatures ranging from 1200 to 2400 eV and no radiation temperature. These conditions require the inclusion of ion stages of tungsten from Fe-like (26 electrons) to Pd-like (46 electrons).

We first present in Figure 1 the ionization distribution of tungsten at the two temperatures of 1200 eV and 2400 eV. We compare the ionization balance computed from the ATOMIC semi-relativistic calculations (labelled ATOMIC SCA) with the ATOMIC fully-relativistic calculations (labelled (ATOMIC RCA). At the higher temperature of 2400 eV, we find that the two ATOMIC calculations are in quite good agreement for the ionization distribution, with an average ionization ($\overline{Z}$) that agrees to around 1 %.

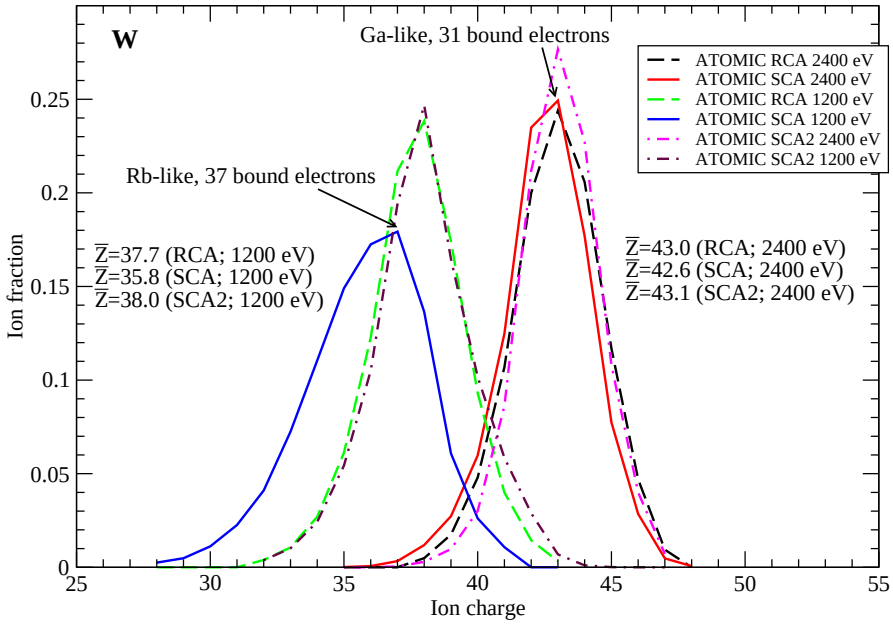

Figure 1. Ionization distribution of tungsten at two temperatures and an electron density of 10^{14} cm^{-3}: The left-most curves represent the ionization balance at a temperature of 1200 eV and the right-most curves represent the ionization balance at 2400 eV. We compare the ATOMIC semi-relativistic calculations (ATOMIC SCA) (solid lines) with the ATOMIC fully-relativistic calculations (ATOMIC RCA) (dashed lines). We also present an ATOMIC calculation (labelled SCA2, dot-dashed lines) that used the same configuration list as used in the ATOMIC RCA calculations.

However, at the lowest temperature considered of 1200 eV, we find that the ATOMIC RCA calculations predict a somewhat higher average ionization ($\overline{Z}$) and a narrower ionization distribution compared to the ATOMIC SCA calculations. The difference in average ionization is around 5% and implies that the ATOMIC SCA calculations will have a larger contribution from less-ionized tungsten ions compared to the ATOMIC RCA calculations. The differences between the two sets of calculations is due to the differing numbers of configurations included in each calculation, rather than due to relativistic structure effects. This is demonstrated by an additional semi-relativistic ATOMIC calculation that used

the same configuration list as was used in the ATOMIC RCA calculations. This calculation (labelled ATOMIC SCA2 in Figure 1) produces an ionization balance that is quite close to the ATOMIC RCA results. This shows that relativistic effects on the ionization balance do not appear to be significant.

We now examine the emission spectrum of tungsten in the few-nm wavelength range that was investigated in the ASDEX Upgrade measurements of Pütterich *et al.* [2]. We first present the ATOMIC RCA calculations at a temperature of 2400 eV in Figure 2, and compare the ATOMIC RCA calculations that retain only E1 transitions (ATOMIC RCA E1) with ATOMIC RCA calculations that retain all multipole transitions up to M3 (ATOMIC RCA MULT). We find that, although the spectra are quite similar and produce radiative power loss values that agree to within 2%, some of the strong E1 lines found in the ATOMIC RCA E1 calculation are significantly modified by the addition of the multipole transitions. As illustrated in [9] for a tungsten plasma at the higher temperature of 5000 eV, the inclusion of extra radiative transitions in the ATOMIC RCA MULT calculations can lead to a redistribution of population, underscoring that multipole transitions should be included when emission spectra are important for plasma diagnostics. This conclusion can be somewhat unexpected since many previous studies have found that multipole transitions are only important for much lower density plasmas (as found in astrophysical plasmas). The importance of multipole transitions here in medium-density fusion plasmas reflects the complexity of the configurations of relevance for these ions of tungsten as well as the high Z value of tungsten, since the multipole transition probabilities scale strongly with Z. Open p and d-shell configurations may undergo multipole radiative transitions that redistribute population that would otherwise be retained in excited states.

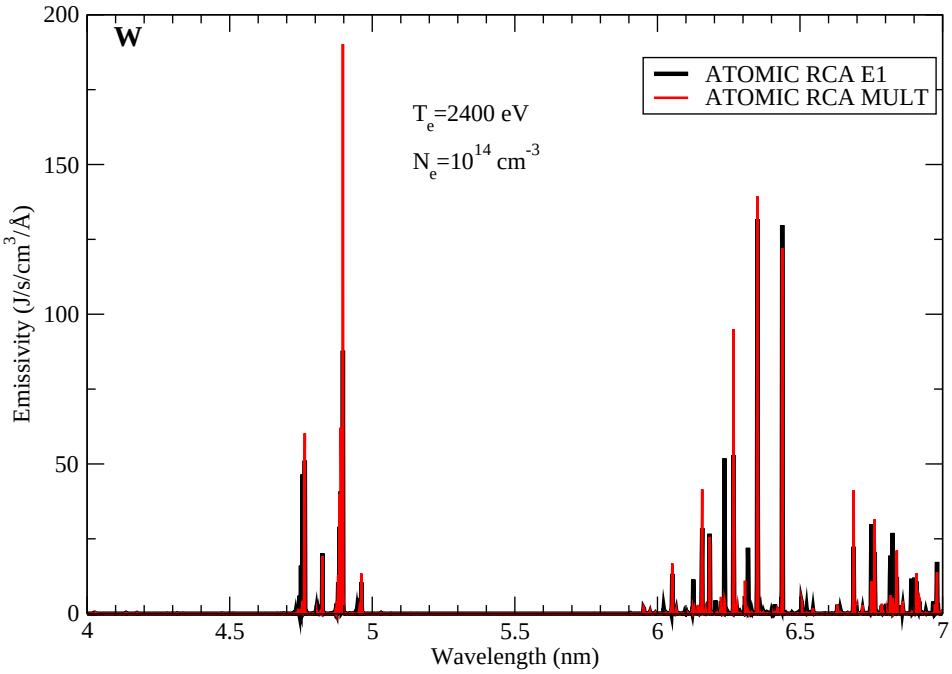

Figure 2. Emission spectrum of tungsten at a temperature of 2400 eV and an electron density of 10^{14} cm^{-3}. ATOMIC RCA E1 calculations (black curve that includes only electric dipole transitions) are compared with ATOMIC RCA MULT calculations (red curve that includes radiative transitions up to and including M3).

In Figure 3 we present the emission from tungsten at a temperature of 2400 eV from semi-relativistic calculations made using ATOMIC, as described previously. The configuration-average spectrum, which only retains natural and Doppler broadening in the calculation, is shown as the black line (labelled as ATOMIC SCA), and produces significant emission only in the 6 nm wavelength region. A calculation that employs UTA theory [17] to approximate the broadening of the bound-bound contributions to the spectrum (green curve) produces a broad featureless emission spectrum that is centered at around 5.5 nm, *i.e.*, to the left of the configuration-average results shown in Figure 3. This is to be expected, since the UTA approximation aims to mimic the effects of intermediate-coupling on the distribution of line strength within a transition array and often shifts the spectrum compared to a pure configuration-average calculation. However, the UTA broadening in this case is substantially larger than the natural and Doppler broadening used in the ATOMIC SCA calculations and also is much broader than the lines observed in the measurements of [2]. The ATOMIC semi-relativistic MUTA calculation (labelled as ATOMIC SMU), however, produces significant emission in the 4.7 nm wavelength region as well as the region around 6 nm. This is in qualitative agreement with the ATOMIC RCA calculations shown in Figure 2.

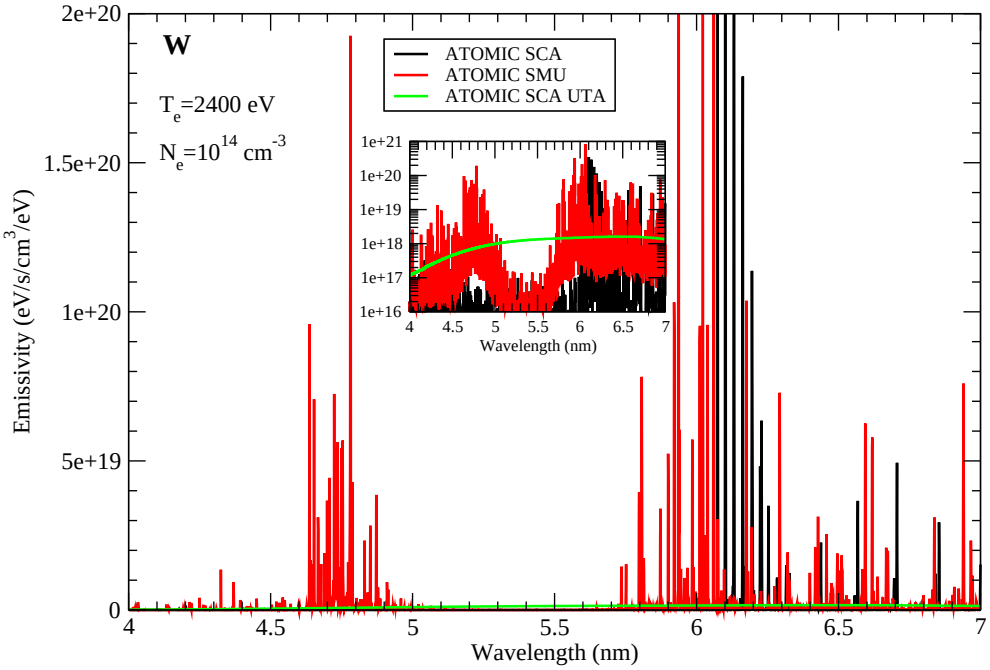

Figure 3. Emission spectrum of tungsten at a temperature of 2400 eV and an electron density of 10^{14} cm^{-3}. ATOMIC SCA calculations (*i.e.*, semi-relativistic configuration-average calculation, black curve) are compared with ATOMIC SMU calculations (*i.e.*, semi-relativistic calculations using MUTA-based fine-structure transitions, red curve). We also show an ATOMIC SCA UTA calculation (green curve) that uses UTA theory to broaden the bound-bound contribution to the spectrum. The inset figure shows the same set of spectra but on a logarithmic y-axis to more clearly show the ATOMIC SCA UTA calculation.

Figure 4 shows a similar emission spectrum, but for a temperature of 1200 eV. Similar trends to the 2400 eV case are found, with the MUTA calculations significantly redistributing the emission towards

the smaller wavelengths due to the inclusion of spin-orbit interaction. We note that the dominant emission peaks at around 4.7 nm and 6.2 nm are in qualitative agreement with the emission spectrum measured from the ASDEX Upgrade tokamak as presented in [2]. We do not attempt a more quantitative comparison because the observed emission in the tokamak measurements likely arises from plasma at a range of temperatures.

However, it is clear that a pure (semi-relativistic) configuration-average calculation is not sufficient to model the emission from tungsten plasma at such conditions. Relativistic configuration-average calculations appear qualitatively closer to the observed emission spectra. The use of the MUTA approach that uses level-resolved data for the emission (albeit with statistically obtained fine-structure populations) may be better still, since it includes significantly more detail in such calculations. It is natural to then consider whether the MUTA approach is "good enough" for the calculation of tungsten emission spectra at the temperatures considered here. Several concerns remain over such calculations. The first issue (as already mentioned earlier) is that the statistical approach used to construct the level-resolved populations from the configuration-average populations may not be sufficiently accurate. We also note that the MUTA approach does not currently include non-dipole radiative transitions, which can be important as demonstrated in Figure 2. Another issue is that the level-resolved transition energies used in the MUTA calculations may not be sufficiently accurate because configuration-interaction (CI) effects may change the transition energies and/or oscillator strengths. We have begun some exploration of this latter issue, and find that CI effects do appear to be significant for some of the transitions of interest here. Preliminary calculations indicate that the transition energies are modified by several percent by CI effects. A significant portion of the emission in the 4–7 nm wavelength range arises from $\Delta n = 0$ transitions in the $n = 4$ shell. The large number of closely-spaced configurations of this type can mix very strongly and significant CI effects on level energies and oscillator strengths are not unexpected.

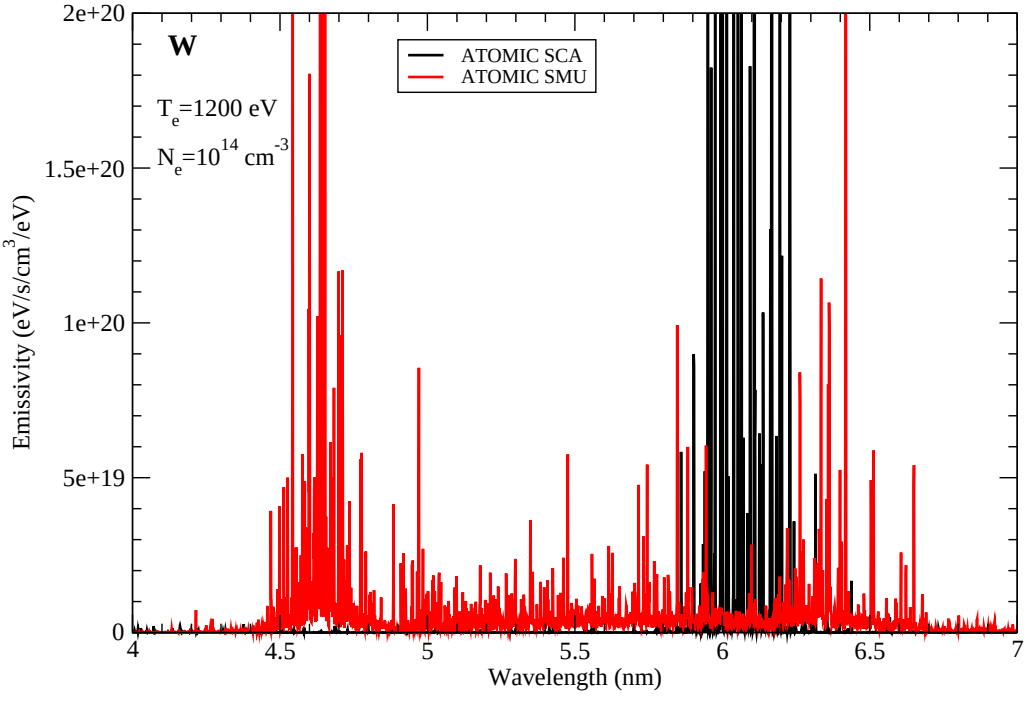

Figure 4. Same as Figure 3, except at an electron temperature of 1200 eV, and that we do not present an ATOMIC SCA UTA calculation.

4. Conclusions

In this report we have presented several collisional-radiative modeling calculations for tungsten at temperatures from 1200 to 2400 eV made using the Los Alamos ATOMIC code. We have explored the effects of semi-relativistic versus fully-relativistic atomic structure models, as well as the sensitivity of the results to number of configurations included in the calculations. We also explored how the emission from tungsten at these temperatures is modified by the use of level-resolved data.

We have found that, not surprisingly, the plasma properties of tungsten at the moderate temperatures considered here are extremely complex. If accurate emission spectra are required, it appears necessary to include significant detail in collisional-radiative modeling calculations. Any calculations should include sufficient configurations to ensure accurate ionization balances, but should also include accurate (level-resolved) atomic data (ideally including configuration-interaction effects) so that the emission is accurately computed. Non-dipole transitions may also be important for accurate emission spectra. We plan further calculations of atomic data and modeling for tungsten in this challenging regime and continue to explore ways in which such calculations can be made more tractable.

Acknowledgments

The Los Alamos National Laboratory is operated by Los Alamos National Security, LLC for the NNSA of the U.S. DOE under Contract No. DE-AC5206NA25396.

Conflicts of Interest

The authors declare no conflict of interest.

References

1. Pütterich, T.; Neu, R.; Dux, R.; Whiteford, A.D.; O'Mullane, M.G.; Summers, H.P.; The ASDEX Upgrade Team. Calculation and experimental test of the cooling factor of tungsten. *Nucl. Fusion* **2010**, *50*, 025012.
2. Pütterich, T.; Neu, R.; Dux, R.; Whiteford, A.D.; O'Mullane, M.G.; The ASDEX Upgrade Team. Modeling of measured tungsten spectra from ASDEX Upgrade and predictions for ITER. *Plasma Phy. Control. Fusion* **2008**, *50*, 085016.
3. Fontes, C.J.; Chung, H.-K.; Hansen, S.B.; Ralchenko, Yu. Report on the NLTE-8 Code Comparison Workshop. 2015, in preparation. Available online: http://nlte.nist.gov/NLTE8.
4. Chung, H.-K.; Bowen, C.; Fontes, C.J.; Hansen, S.B.; Ralchenko, Y. Comparison and analysis of collisional-radiative models at the NLTE-7 workshop. *High Energy Density Phys.* **2013**, *9*, 645–652.
5. Rubiano, J.G.; Florido, R.; Bowen, C.; Lee, R.W.; Ralchenko, Y. Review of the 4th NLTE code comparison workshop. *High Energy Density Phys.* **2007**, *3*, 225–232.
6. Fontes, C.J.; Abdallah, J., Jr.; Bowen, C.; Lee, R.W.; Ralchenko, Y. Review of the NLTE-5 kinetics workshop. *High Energy Density Phys.* **2009**, *5*, 15–22.

7. Ralchenko Yu.; Draganić, I.N.; Osin D.; Gillaspy, J.D.; Reader, J. Spectroscopy of diagnostically important magnetic-dipole lines in highly charged $3d^n$ ions of tungsten, *Phys. Rev. A* **2011**, *83*, 032517.
8. Abdallah, J.; Clark, R.E.H.; Peek, J.M.; Fontes, C.J. Kinetics calculations for near Ne-like ions. *J. Quant. Spectrosc. Rad. Transf.* **1994**, *51*, 1–8.
9. Fontes, C.J.; Zhang, H.L.; Abdallah, Jr., J.; Clark, R.E.H.; Kilcrease, D.P.; Colgan, J.; Cunningham, R.T.; Hakel, P.; Magee, N.H.; Sherrill, M.E., The Los Alamos suite of relativistic atomic physics codes. *J. Phys. B* **2015**, in press.
10. Mazevet, S.; Abdallah, J., Jr. Mixed UTA and detailed line treatment for mid-Z opacity and spectral calculations. *J. Phys. B* **2006**, *39*, 3419.
11. Cowan, R.D. *The Theory of Atomic Structure and Spectra*; University of California Press: Berkeley, CA, USA, 1981.
12. Clark, R.E.H.; Abdallah, J., Jr.; Mann, J.B. Integral and differential cross sections for electron impact ionization. *Astrophys. J.* **1991**, *381*, 597–600.
13. Sampson, D.H.; Zhang, H.L.; Fontes, C.J. A fully relativistic approach for calculating atomic data for highly charged ions. *Phys. Rep.* **2009**, *477*, 111–214.
14. Zhang, H.L.; Sampson, D.H. Rapid relativistic distorted-wave approach for calculating cross sections for ionization of highly charged ions. *Phys. Rev. A* **1990**, *42*, 5378–5383.
15. Magee, N.H.; Abdallah, J.; Colgan, J.; Hakel, P.; Kilcrease, D.P.; Mazevet, S.; Sherrill, M.; Fontes, C.J.; Zhang, H.L. Los Alamos Opacities: Transition from LEDCOP to ATOMIC. In Proceedings of the 14th Topical Conference on Atomic Processes in Plasmas, AIP Conference Proceedings, Santa Fe, NM, USA, 19–22 April 2004; Cohen, J.S., Mazevet, S.; Kilcrease, D.P., Eds.; pp. 168–179.
16. Hakel, P.; Sherrill, M.E.; Mazevet, S.; Abdallah, J., Jr.; Colgan, J.; Kilcrease, D.P.; Magee, N.H.; Fontes, C.J.; Zhang, H.L. The new Los Alamos Opacity Code ATOMIC. *J. Quant. Spectrosc. Rad. Transf.* **2006**, *99*, 265–271.
17. Bauche, J.; Bauche-Arnoult, C. Transition arrays in the spectra of ionized atoms. *Adv. At. Mol. Phys.* **1988**, *23* 131–195.