Theoretical approaches to electron-impact ionization

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Outline of talk

- Review of the various theoretical approaches to electron-impact ionization (EII) – brief descriptions
  - Close-coupling methods
  - Distorted-wave (DW) approaches
  - Other approaches
  - Relativistic considerations
- EII cross sections: very brief overview of calculations performed to date
  - Light atoms (Z=1-10)
  - Moderate Z atoms (Z~11-54)
  - Heavy atoms
  - Inner-shell contributions
- Tungsten
  - What has been computed so far for this system?
  - Can we gauge the accuracy of calculations performed to date?
  - How can we benchmark such work?
- A tentative proposed benchmark:
  - Tungsten ions of importance in fusion modeling, (around 3 ions) could be selected, of various stages of ionization
  - Theory and experiment (such as crossed-beams methods) both work towards this and a joint publication prepared to analyze quality of resulting cross sections and the associated uncertainty

Theoretical approaches to EII: close-coupling

- Close-coupling approaches aim to treat the interaction between the outgoing (scattered & ejected) electrons in a non-perturbative manner in an effort to more accurately model the ionization process
- Several close-coupling approaches have been developed in recent decades:
  - Convergent close-coupling (CCC)
  - R-matrix (with pseudo states) (RMPS) and variants thereon
  - Time-dependent close-coupling (TDCC)
  - Exterior complex-scaling (ECS)
- **Each of these approaches has advantages/disadvantages, some of which are discussed in talks at this workshop**
- **Most approaches are quite computationally expensive**
- Fully relativistic versions of many of these approaches are also available
- Accepted view is that one requires such close-coupled approaches for low-energy, neutral & near-neutral systems for which the electron-electron couplings are strong and cannot be treated perturbatively

Theoretical approaches to EII: distorted-wave

- A distorted-wave (DW) approach treats the electron-electron interactions perturbatively
  - The incident, scattered, and ejected electronic wavefunctions are all ‘distorted’ by the potential of the target atom (including nuclear and electron potential terms)
  - One can choose the potential in which the wavefunctions are computed (eg ‘post’ or ‘prior’ forms of the interaction potential)
- Several sets of DW codes are available world wide, such as LANL codes (available to run online[1]), FAC, etc.
- DW approaches are **much less computationally expensive** compared to close-coupling approaches
- They are accepted to be less accurate for neutral/near-neutral targets, but accuracy appears adequate for moderately and highly-ionized systems
- Fully relativistic DW versions are also available, as well as ‘semi-relativistic’ versions (often based on the Cowan code) in which one-electron terms are added into the non-relativistic Hamiltonian

[1] *http://aphysics2.lanl.gov/tempweb/lanl/*

Theoretical approaches to EII: other approaches

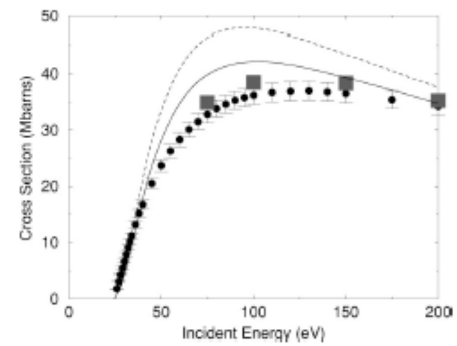
- Other approaches have been discussed in the literature that can produce EII cross sections
- *Binary-Encounter Bethe (BEB)* method pioneered by YK Kim[1]
  - Shows generally very good agreement with measurement for near-neutral atoms and requires really only knowledge of the binding energy of the target atom
- *Statistical* approach has been recently discussed by Lisitsa's group[2]
  - Based on collective excitations of atomic electrons with the local plasma frequency using Thomas-Fermi models for the electron density
- Such methods are constructed with a view to provide very (computationally) fast estimates of the cross section
  - **These methods take essentially no computer resources to run**
- Accuracy is difficult to judge; while comparisons to measurement sometimes looks good, the predictive ability of such approaches is not clear

[1]*Kim & Rudd, PRA 950, 3954 (1994)*
[2]*Demura et al, JETP Letters 101, 85 (2015)*

Light atoms: sampling of cross sections

- For H, He, & Li close agreement exists between theory & experiment
- Recent study on Be also reports good agreement between a variety of close-coupling methods
  - Recent study highlighted close agreement between RMPS, CCC, and TDCC methods
  - DW approaches ~ 20-30% higher for neutral atom but quickly approaches close-coupling results by two-times ionized
- Although studies of other light atoms (Z=1-10) are not comprehensive, where comparisons exist, agreement between different close-coupling approaches is generally satisfactory

He



Be

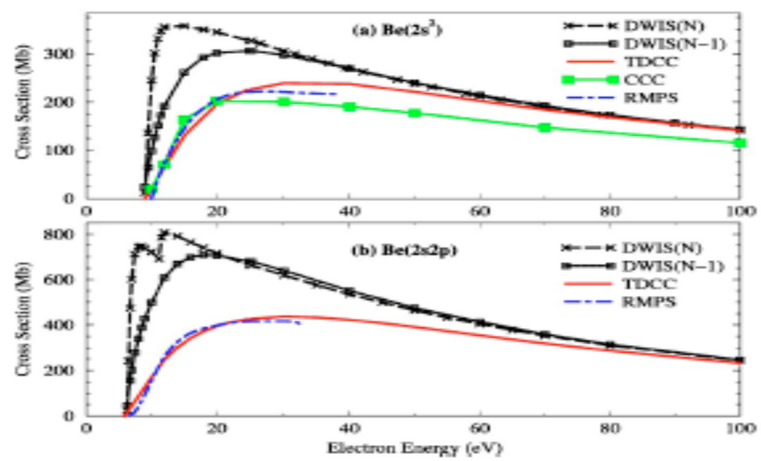


FIG. 1. Electron-impact ionization cross sections for neutral Be, from (a) the ground $1s^2 2s^2$ configuration and (b) the first excited $1s^2 2s 2p$ configuration. The solid lines are the time-dependent close-coupling calculations. The dot-dashed lines are the RMPS calculations and the short-dashed (with crosses) and dotted lines (with squares) are the DWIS(N) and DWIS($N-1$) calculations, respectively. The solid line with squares are CCC calculations from Ref. [7]. In (b) all calculations include ionization from both the $2s$ and $2p$ subshells. Also, the RMPS calculations are for ionization from the $1s^2 2s 2p$ ^3P term only (1.0 Mb$=1.0\times10^{-18}$ cm^2).

Pindzola & Robicheaux, PRA 61 052707 (2000)
Zatsarinny et al, PSST submitted (2016)

Moderately heavy atoms: Z~11-36

- Far fewer close-coupling studies exist – some studies have been published for several noble gas atoms and quasi one-electron (Na) and two-electron (Mg) targets
- DW approach over-estimates the cross section (compared to measurement) for neutral Si (not unexpected)
  - Although we note that at higher energies, the DW approach appears reasonable

Si

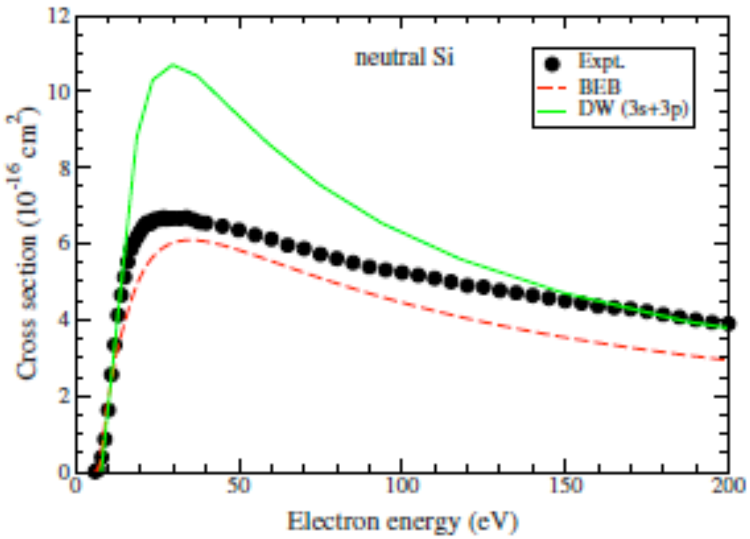


FIG. 2. (Color online) Electron-impact ionization cross sections for neutral Si. The partial cross sections from the $3s$ and $3p$ subshells of the ground $3s^23p^2$ configuration have been summed to compare with experiment. We compare our present distorted-wave calculations (DW) with the experimental measurements of Freund *et al.* [11] and binary-encounter-Bethe (BEB) calculations of Stone and Kim [32].

Colgan et al, PRA 77, 062704 (2008)

Moderately heavy atoms: Z~11-36

- DW calculations for Si^{2+} appear of acceptable accuracy

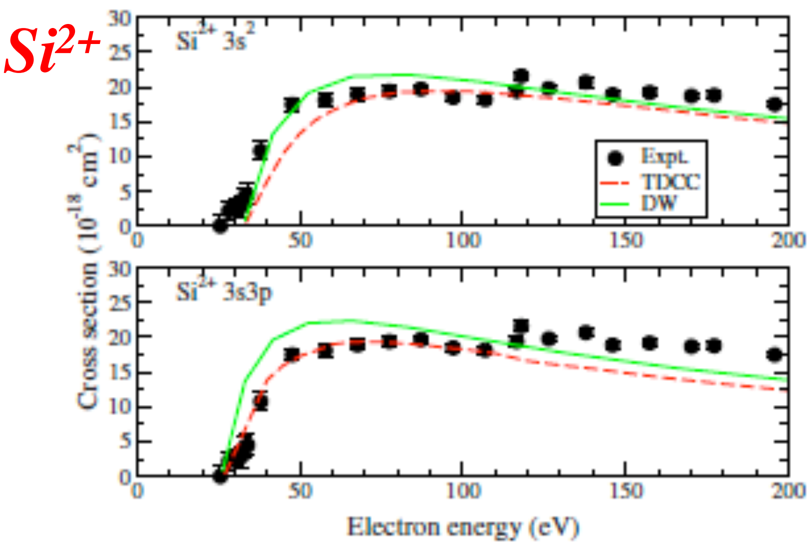


FIG. 3. (Color online) Electron-impact ionization cross sections for Si^{2+} from the ground $3s^2$ and excited $3s3p$ configurations. In both cases, we compare our present distorted-wave calculations (DW) and time-dependent close-coupling (TDCC) calculations with the experimental measurements of Djurić *et al.* [12].

- And by Si^{7+} DW appears very accurate

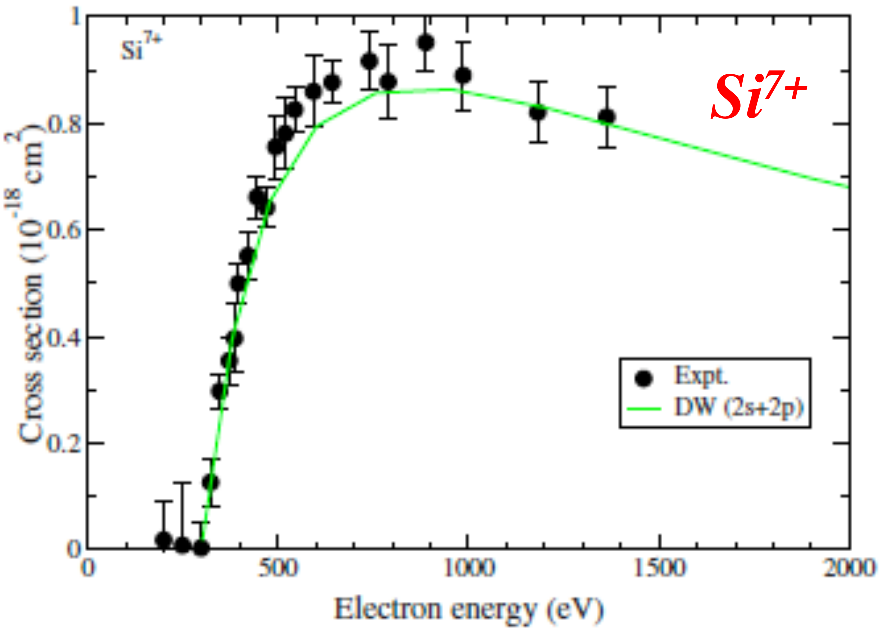


FIG. 5. (Color online) Electron-impact ionization cross sections for Si^{7+}. The partial cross sections from the 2*s* and 2*p* subshells of the ground $2s^22p^3$ configuration have been summed to compare with experiment. We compare our present distorted-wave calculations (DW) with the measurements of Zeijlmans van Emmichoven *et al.* [15].

Moderately heavy atoms: Z~11-54

- Transition metals?
  - Not aware of (m)any close-coupling calculations
- Close-coupling approaches generally have difficulty with targets in which the atomic structure is complex
  - TDCC, ECS, and CCC are restricted to (at best) quasi one-electron and two-electron targets
    - And even these calculations may involve approximations with respect to structure
  - R-matrix approach **can** be applied to complex targets, but can calculations be converged?
  - Relatively little ionization calculations have been performed with R-matrix calculations for heavy atoms – excitation is in some sense easier
  - Preliminary calculations were performed on Mo^+ in 2005
    - Correlation and term-dependence in the initial state was explored

Mo^+

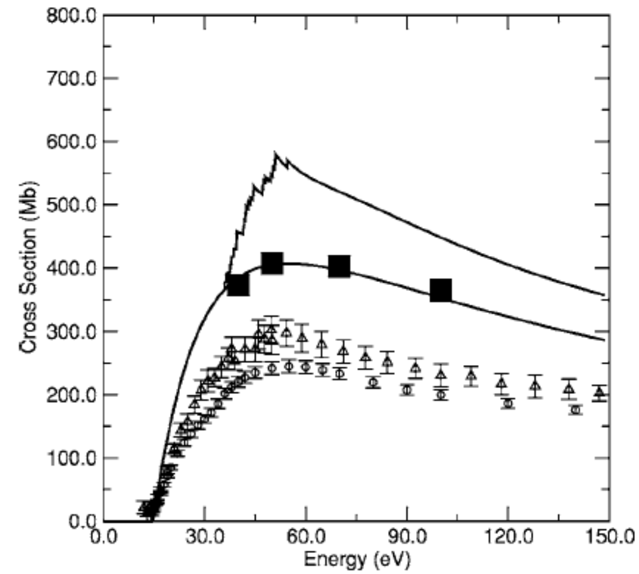


FIG. 4. Electron-impact single ionization cross section for Mo^+ in the $4d^5$ ground configuration. Closed squares connected by solid curve: TDCC calculation for direct ionization, solid curve: TDCC calculation for direct ionization plus TIDW calculation for $4p \rightarrow nl$ excitation-autoionization, open triangles: experimental measurements [6], and open circles: experimental measurements [20].

Ludlow et al, PRA 72 032729 (2005)

Heavy atoms

- Beyond transition metals the available data thins out considerably
- Some work on tungsten, that we will discuss soon
- Also extensive work on inner-shell ionization of heavy systems
  - K-shell and L-shell ionization of neutral atoms often resembles ionization of highly-charged ions
  - Fully relativistic approach is often required due to high energies of incident electrons
- Distorted-wave approach seems completely adequate for such purposes

K-shell ionization of Fe

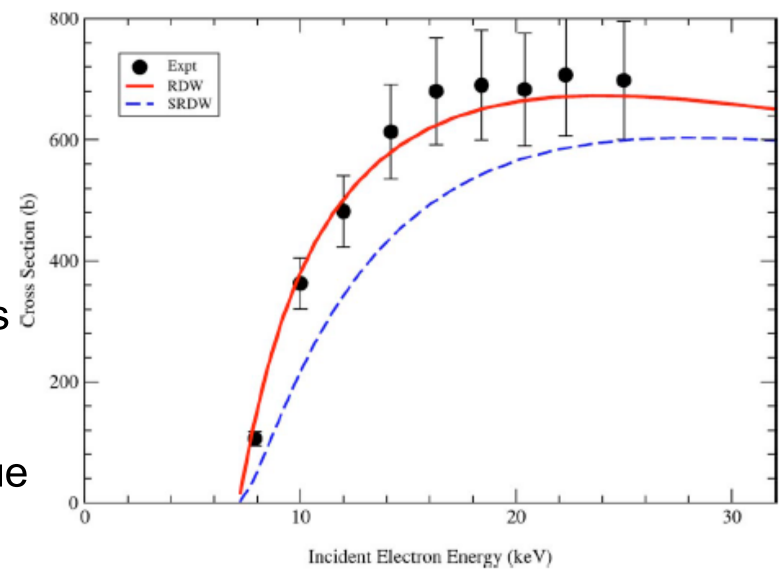


FIG. 2. (Color online) *K*-shell electron-impact ionization of Fe. We compare the RDW calculations with our experimental measurements. The dashed curve is a SRDW calculation.

Colgan et al, PRA 73, 062711 (2006)

Electron-impact ionization of W and its ions

- Complete EII calculations for all of the isonuclear sequence of W performed by Loch et al in 2005
  - Configuration-average distorted-wave approach used
  - Probably inaccurate for neutral W, but thought to be of acceptable accuracy around W^{4+}
  - Investigated role of radiation damping in reducing indirect excitation-autoionization contributions
  - Explored when fully relativistic calculations were required (around Ne-like)
- These data **all** incorporated into ADAS modeling of fusion plasmas and used in modeling by Putterich et al

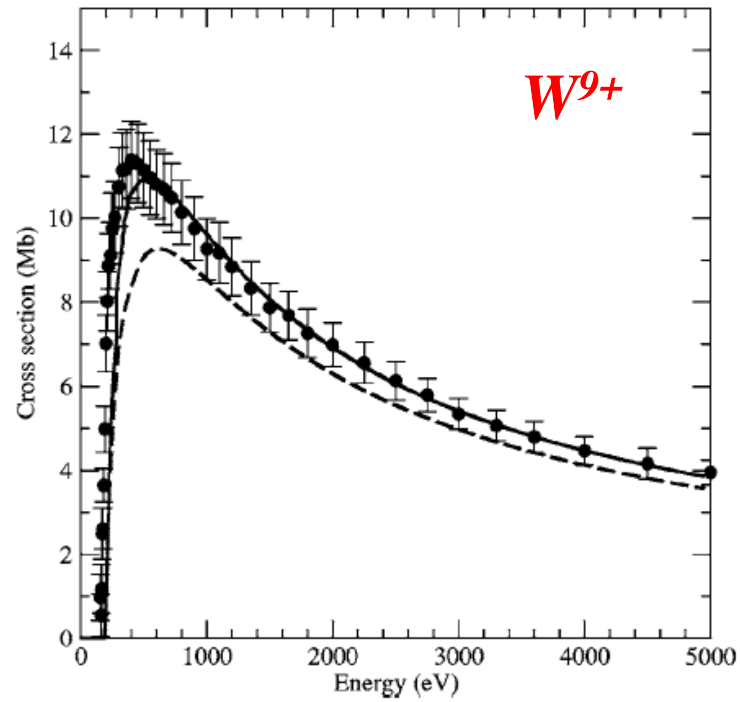
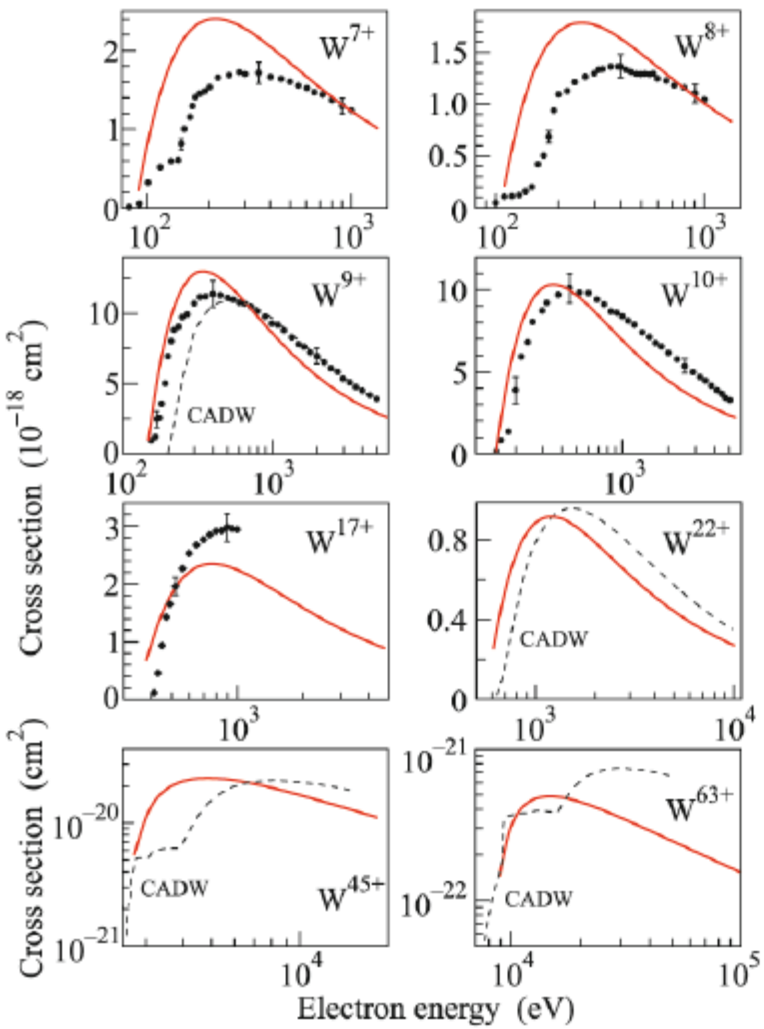


FIG. 1. Electron-impact ionization cross section for W^{9+}. Dashed curve: CADW calculations for direct ionization; solid curve: CADW calculations for direct and indirect ionization; solid circles: experimental measurements [3] ($1.0\ \text{Mb} = 1.0 \times 10^{-18}\ \text{cm}^2$).

Loch et al, PRA 72, 052716 (2005)

Electron-impact ionization of W and its ions

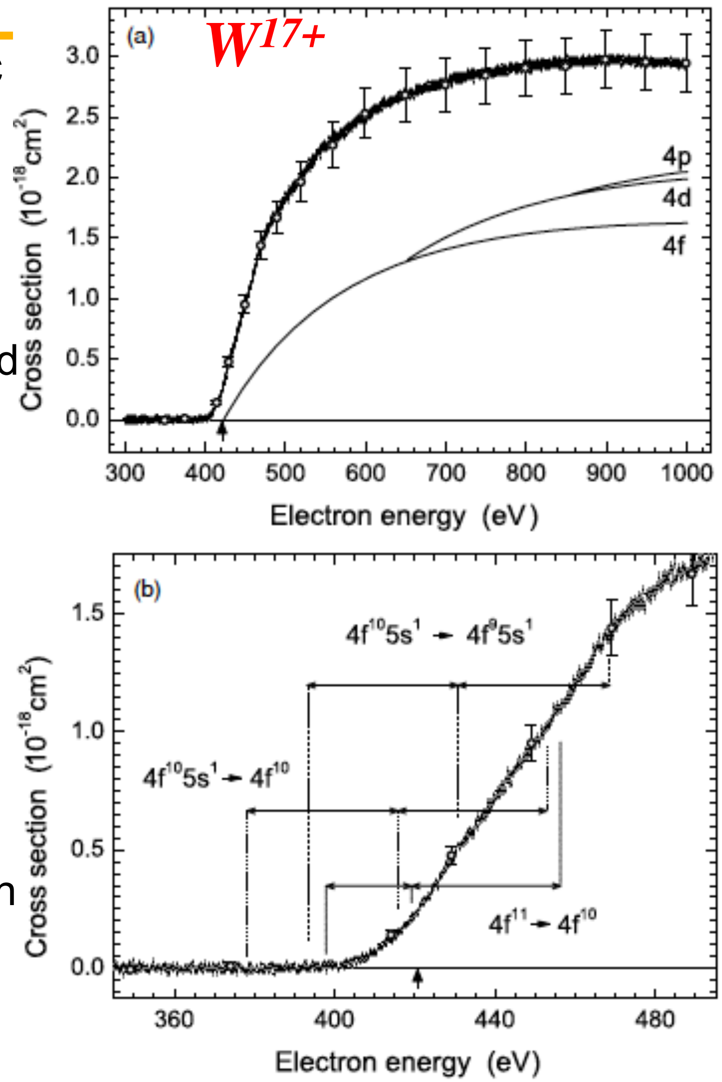
- Complete EII calculations for all of the isonuclear sequence of W also recently published by Demura et al
  - Using a statistical (Thomas-Fermi) approach
  - Agreement with available experiment is mixed
  - Comparison with CADW shows some significant differences for some ions
  - Also published ionization rates which can be convenient for collisional-radiative modeling



Demura et al, JETP 101, 85 (2015)

Electron-impact ionization of W: comparison with measurement

- Giessen group has performed systematic measurements of EII of W ions using a crossed-beam set up
  - Compared with LANL CADW cross sections obtained from LANL website
  - Direct ionization does not account for measured cross section
  - Large indirect (excitation-autoionization) cross section is inferred
  - Ground configuration of $4d^{10}4f^{11}$ spawns 41 FS levels that span a range of > 20 eV
  - Ground configuration of W^{18+} ($4f^{10}$) spawns more than 100 FS levels
  - Contribution also expected from levels in excited $4f^{10}\ 5s^1$ configuration
  - Measurement also reported of double ionization cross sections, which are ~ two orders of magnitude lower than single ionization

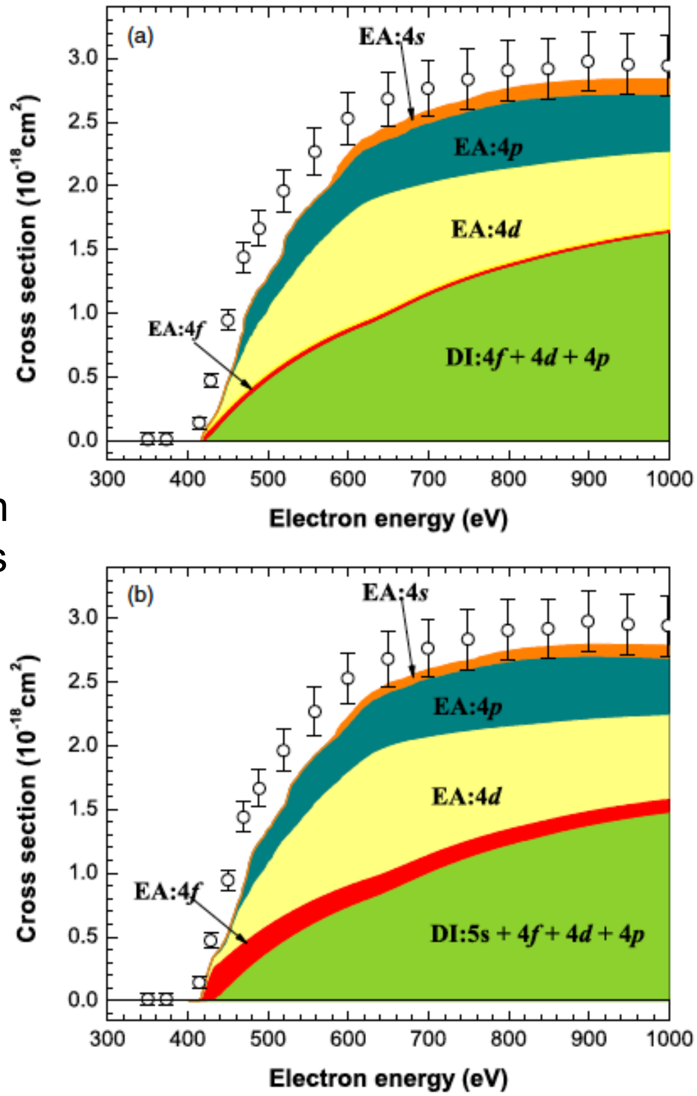


Rausch et al, JPB 44, 165202 (2011)

Electron-impact ionization of W: comparison with measurement

W^{17+}

- More recent study using distorted-wave approach (FAC code) by Zhang & Kwon added the excitation-autoionization contribution
  - Agreement with experiment is now quite good
  - Zhang & Kwon also point out that cross section from metastable $4f^{10}\,5s^1$ levels are of similar magnitude to those from ground configuration
  - Experiment likely produces an unknown fraction of ions in metastable states; so measured cross section results from some combination of contributions from levels associated with both ground and metastable states.



Zhang & Kwon, JPB 47, 075202 (2014)

Electron-impact ionization of W: comparison with measurement

- FAC code also used to investigate single ionization of W^+ ions
  - Sensitivity noted of the cross section to the choice of local central potential within the FAC calculation
  - Authors used potentials arising from either
    - W^+ [$5d^4 6s + 5d^5 + 5d^3 6s^2$] or
    - W^{2+} [$5d^3 6s + 5d^4 + 5d^2 6s^2$]
  - Different cross sections obtained depending on this choice
  - Underlines the difficulty even of the structure calculations that are required for such complex ions
  - Excitation-autoionization is also expected to contribute to the cross section in this case
  - A further complication is that the distorted-wave approximation may also be inaccurate for such low-charged ions

W^+

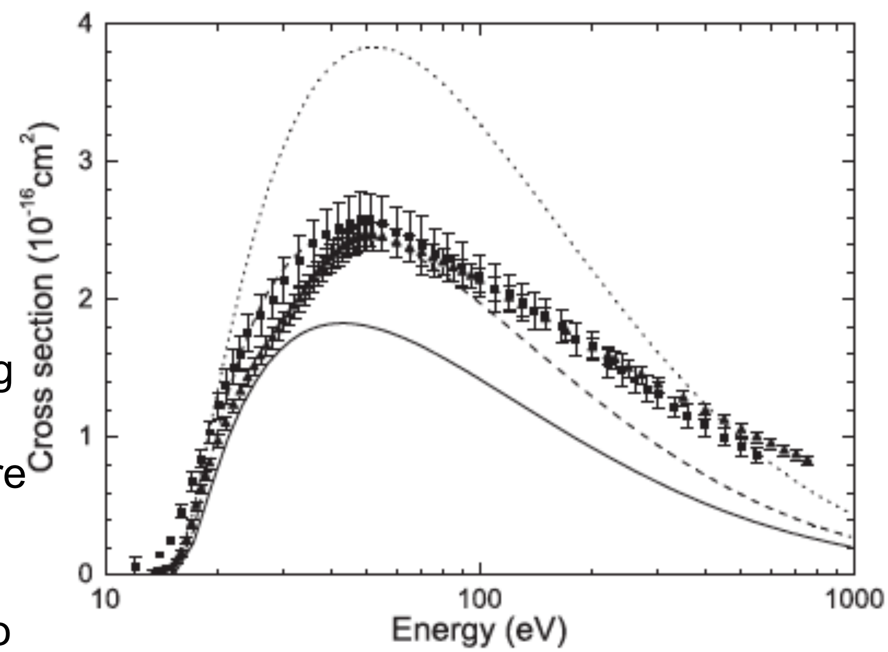


Fig. 1. DI cross section of a 5*d* and 6*s* electron for the ground level $5d^4 6s\ ^6D_{1/2}$ of W^+ forming W^{2+}. Different curves represent various configurations for the optimization of the single local central potential: dotted curve, the configurations $5d^4 6s + 5d^5 + 5d^3 6s^2$ of W^+; dashed curve, the configurations $5d^4 + 5d^3 6s + 5d^2 6s^2$ of W^{2+}; solid curve, the configurations $5d^4 + 5d^3 6s + 5d^2 6s^2 + 5d^2 5f^2 + 5d 5f^2 6s + 5f^2 6s^2 + 5d^2 5g^2 + 5d 5g^2 6s + 5g^2 6s^2$ of W^{2+}. The filled rectangles [5] and triangles [10] are the experimental results.

Zhang & Kwon, Int. J. Mass. Spect. 356, 7 (2013)

Electron-impact ionization of W: proposed benchmark studies

- We now reach the following scenario:
- Close-coupling approaches are most accurate for computing single ionization
  - But not clear if they can even be applied to heavy, complex atoms (such as low-charged W ions)
  - Or if they can, whether calculations are computationally tractable
- Distorted-wave approaches appear to work well (compared to measurement) for moderately and highly-charged ions
  - Can we quantify the uncertainty associated with this? How good is "well" !?
- Ionization of light atoms is well described, in general
- Most pressing need (in terms of fusion) is probably data for Tungsten & ions
  - But there are 74 ion stages of tungsten!
  - Which ones to prioritize?

Electron-impact ionization of W: proposed benchmark studies

- Ionization data ***does*** make a difference in fusion modeling
- When CADW data used in place of ADPAK data the ionization equilibrium changes
- Leads to different predictions of the relative abundance of various ions within the fusion plasma
  - Dielectronic recombination rates also make significant difference, but this will be discussed elsewhere…

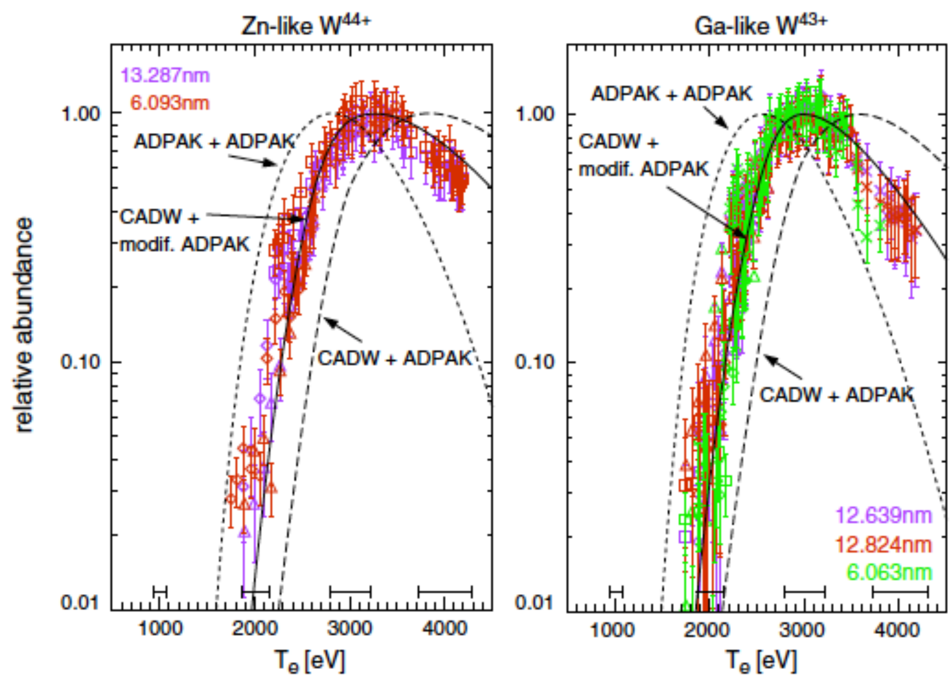


Figure 3. Relative abundances of Ni-like W^{46+} to Ga-like W^{43+} derived from theoretical data (lines) and from measurement (symbols). The measurements originate from impurity accumulation phases. Further information in the text.

Putterich et al, Plasma Phys. Control. Fusion 50, 085016 (2008)

Electron-impact ionization of W: proposed benchmark studies

- We could focus on ions that are abundant over a wide range of temperatures of interest to fusion
- Could also select 'low-charged', 'moderately-charged' and 'highly-charged' ions
- Perform 'best possible' calculations for such ions and compare to 'best possible' measurements
  - Calculations will likely have to be distorted-wave?
  - Measurements will likely be crossed beam?

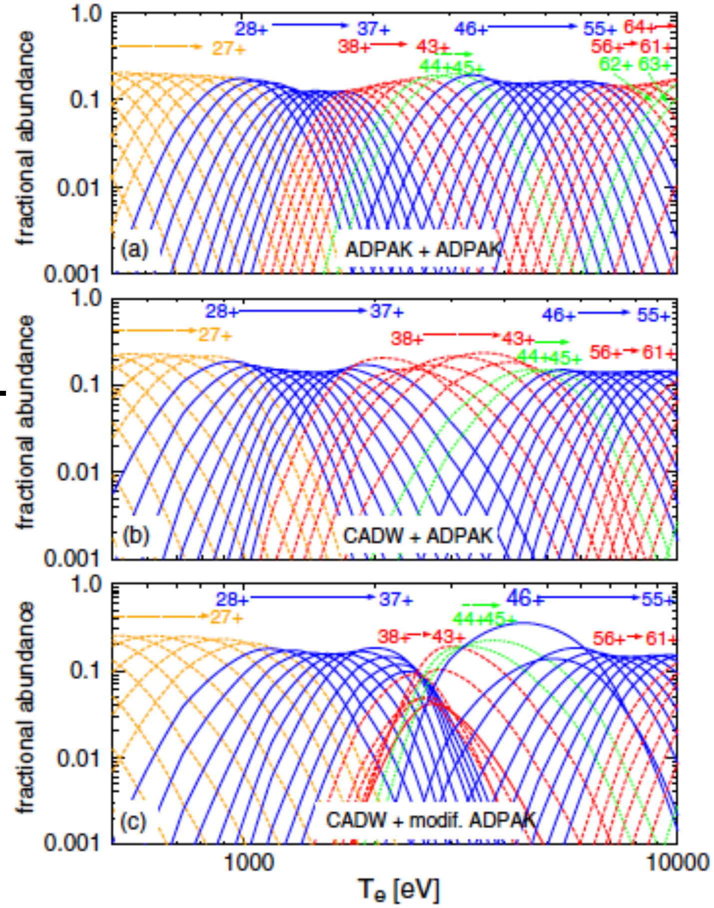
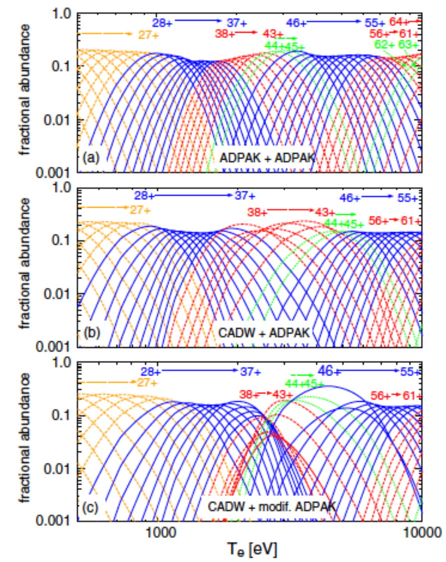


Figure 6. (*a*) Transport-free fractional abundances of tungsten ionization stages for zero electron density using ADPAK ionization and recombination data from [9], which were modified according to [4]. Numbers denote the ionization stage. Line styles and colours group ions with the same 'outermost' subshell; (*b*) same as (*a*), but ionization rates taken from [31]; (*c*) same as (*b*),while recombination rates have been modified (see table 1) according to experimental findings.

Putterich et al, Plasma Phys. Control. Fusion 50, 085016 (2008)

Electron-impact ionization of W: proposed benchmark studies

- Even distorted-wave calculations will require some planning and thought
  - Configuration-average sufficient or are level-to-level calculations required?
  - Are there sensitivities to potential used (‘post’ vs. ‘prior’ potential forms)?
  - Will fully relativistic calculations be required and for which ions?
  - How extensive is the excitation-autoionization contribution and how best to take this into account?
  - Etc
  - All such considerations should allow some measure of uncertainty of the final data



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- Measurements will also of course need planning – we will hear about this in the next session

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Putterich et al, Plasma Phys. Control. Fusion 50, 085016 (2008)

Conclusions & Future Work

- A brief overview of the tools and techniques used to compute electron-impact ionization cross sections has been given
- Close-coupling calculations are most accurate, but may be only currently applicable to light atoms
- Requirements of fusion are for complete datasets for tungsten
- Distorted-wave calculations may be only viable option for these data
- An assessment of the uncertainty of such calculations would be most welcome
- We have tentatively provided a strategy for joint calculations & measurements that could provide a measure of this uncertainty

Extra Slides