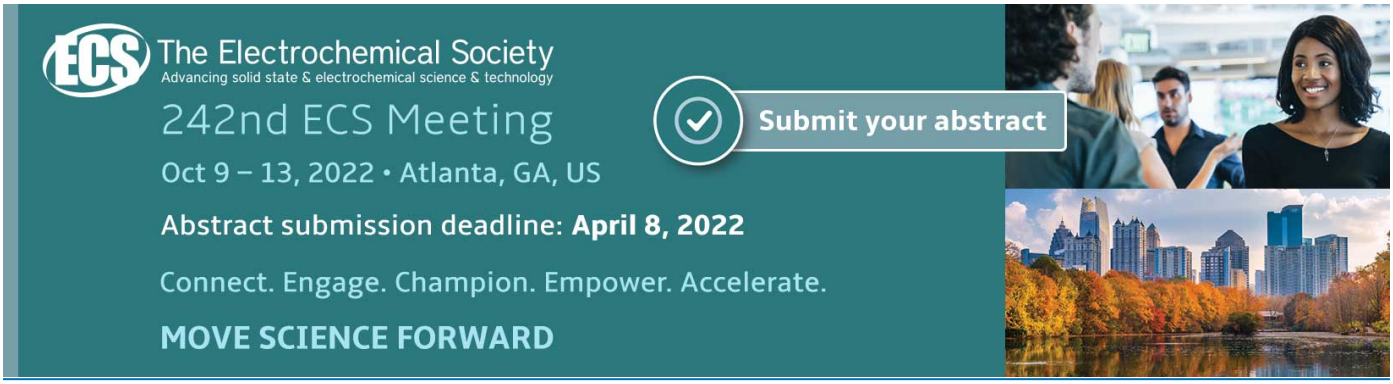
Electron-impact ionization of atomic ions: Theoretical results



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Electron-Impact Ionization of Atomic Ions: Theoretical Results

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Abstract. A brief overview is given of theoretical results for electron-impact ionization of atoms and ions. A description is given of the main theoretical methods, along with the databases where the data are archived. It is shown that for light species, ground and metastable ionization cross sections are in reasonable agreement with experiment when non-perturbative data are used for the near neutrals and distorted wave data are used for ions greater than a few times ionized. Some discrepancies between theory and experiment still remain for systems with open d and open p subshells. The sensitivity of ionization rate coefficients to the near threshold part of the ionization cross section is shown. The role of excited states in effective ionization rate coefficients is demonstrated and recent excited state ionization cross section results for H, He, He^{+}, B^{2+} and Ne are presented.

1. Introduction

In recent years significant advances have been made in the calculation of electron-impact ionization cross sections using non-perturbative methods. This has supplemented long-standing efforts using distorted-wave methods and has allowed comprehensive sets of ionization rate coefficients to be generated for a range of systems. The data used by the modeling community are often still taken from the older archives of Arnaud and Rothenflug [1], Arnaud and Raymond [2] and Mazotta et al. [3]. For electron-impact ionization there have been two recent reviews by Dere [4] and Mattioli et al. [5]. The impact made by the new data presented in these recent reviews on ionization balance curves was studied by Bryans et al. [6]. There are also online archives of electron-impact ionization rate coefficients, we list here just a few of the most commonly used ones. The ADAS database, *http://www.adas.ac.uk*, contains rate coefficients for electron-impact ionization and derived coefficients referred to as Generalized Collisional Radiative (GCR) coefficients which include the density dependent effects of ionization from excited states, see Summers et al. [7]. The ADAS consortium recently made a large portion of their atomic data publicly available online through the OPEN-ADAS web site, *http://open.adas.ac.uk*. The NIFS web site, *https://dbshino.nifs.ac.jp*, provides a search engine for various atomic and molecular databases. The IAEA web page, *http://www-amdis.iaea.org*, provides a means of simultaneously searching a wide range of currently used databases. The ORNL CFADC database contains electron-impact ionization rate coefficients and an archive of experimental

measurements, see *http://www-cfadc.phy.ornl.gov*. Also of interest are the Los Alamos distorted-wave codes that can be run through a web interface at *http://aphysics2.lanl.gov/tempweb/lanl*. The CHIANTI database is commonly used in astrophysical studies and can be accessed via *http://www.damtp.cam.ac.uk/user/astro/chianti*. Of common use in astrophysics is the ATOMDB database *http://cxc.harvard.edu/atomdb*. The Convergent Close-Coupling (CCC) cross section results can be accessed at *http://atom.curtin.edu.au/CCC-WW/index.html*. Results from the Binary Encounter Bethe (BEB) method from the NIST group are archived at *http://physics.nist.gov/PhysRefData/Ionization/atom_index.html*.

In this paper we review some of the main theoretical methods used to calculate electron-impact ionization cross sections, showing their areas of validity. We start by looking at ionization from ground and metastable states, then progress to ionization from excited states and their role in effective ionization rate coefficients, commonly used in plasma impurity transport codes.

2. Theoretical methods

The theoretical methods that are used to calculate electron-impact ionization cross sections can be split into the following broad categories.

- Semi-empirical methods (e.g. Lotz [8], Exchange Classical Impact Parameter (ECIP) [9], Binary Encounter Bethe [10, 11])
- Perturbative methods, such as the distorted-wave (DW) approach
- Non-perturbative methods such as
  - R-matrix with pseudostates (RMPS) [12]
  - Time-Dependent Close-Coupling (TDCC) [13]
  - Convergent Close-Coupling (CCC) [14]
  - Exterior Complex Scaling (ECS) [15]

In general, the non-perturbative methods are in good agreement with each other and are normally required for the near neutral systems where correlation effects are strongest. Distorted-wave methods perform well for ground and metastable ionization of systems more than a few times ionized.

For electron-impact single ionization the dominant mechanisms are direct ionization and excitation-autoionization. For low charge states, Auger rates are much larger than radiative rates, and excitation to an autoionizing level will overwhelmingly lead to an ionization. For more highly charged systems, the radiative rates for the autoionizing levels become competitive with the Auger rates and radiative branching must be taken into account. For example, Loch et al. [16] found that radiative branching became significant for W ions greater than 28 times ionized. Note that processes such as resonant excitation double autoionization (REDA) are also possible, but will not be considered in this article as they usually contribute only a small amount to the total ionization cross section.

3. Ground and metastable ionization

For ground and metastable ionization of near neutral systems, distorted-wave methods tend to overestimate the ionization cross section. This is shown in Fig. 3 for neutral neon. The distorted-wave results are higher than the experiment, while the recent R-matrix with pseudostates results of Ballance et al. [17] are in good agreement with experiment. The distorted-wave method improves as one progresses up the iso-nuclear sequence (i.e. with increasing ion charge). For example, the calculations of Loch et al. [18] showed that distorted-wave calculations agreed with experiment for Kr^{2+} and higher charge states. Although the agreement with increasing ion charge is often not as quick as in the Kr case, distorted-wave results are usually in good agreement with experiment by 4 or 5 times ionized.

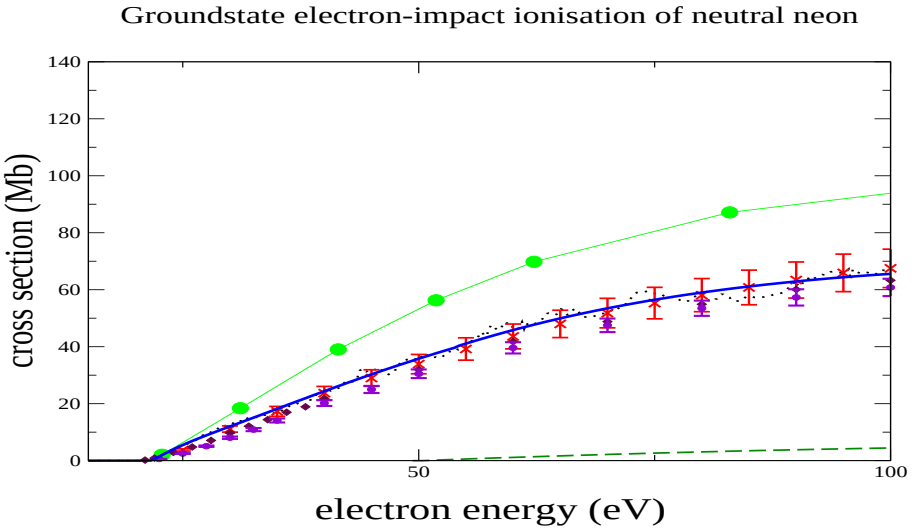

Figure 1. Electron-impact ionization cross section for neutral neon. The dotted black curve represents the raw RMPS results for the 2p ionization, the solid blue curve is the fit to the raw RMPS results. The connected green circles show configuration-average distorted-wave results for the 2p direct ionization and the dashed line shows configuration-average distorted-wave results for the 2s direct ionization. The red crosses show the measurements of Krishnakumar and Srivastava [19]. The diamonds show the measurements of Wetzel et al. [20] and the maroon circles show the measurements of Rejoub et al. [21]. Figure taken from Ballance et al. [17].

While in general one finds reasonable agreement between experiment and distorted-wave theoretical predictions for systems more than a few times ionized, there are some notable exceptions, mostly for open p- and d-shell systems. For example, Ge^{7+} through to Ge^{9+} show discrepancies between DW results and experiment [22].

Ionization cross section measurements can have significant metastable fraction in their ion beams. This complicates the comparison with theory, but also provides a useful means of testing theoretical models of metastable ionization cross sections, which are more sensitive to correlation effects in the collision process. For systems where the ionization potential for the metastable is significantly below the ionization potential for the ground state, one can measure the pure metastable ionization cross section by looking at energies below the ground state threshold. This was recently done by Borovik et al. [23] for Li^+. They found that their measurements of the metastable cross section were in good agreement with convergent close-coupling results, and with recent R-matrix with pseudostates and time-dependent close-coupling calculations [24]. Another approach is to independently measure the metastable fraction. Fogle et al. [25] recently showed that one can measure the metastable fraction in Be-like systems using a gas attenuation method. This method uses the fact that the cross section for charge exchange from a gas such as He to the metastable state in Be-like ions is greater than the cross section for charge exchange into the ground state of the same ions. Thus, by passing the Be-like beam through a He gas cell, and measuring the attenuation of the Be-like ions, they were able to calculate the metastable fraction. Comparison with theoretical models then found that distorted-wave results overestimated the total cross section, while R-matrix with pseudostates results were in good agreement with the measured cross section, when the measured metastable fraction was assumed, see Fig. 2. Knowledge of the metastable fraction was invaluable in this case, as one could easily obtain good agreement with the experimental measurement using the distorted-wave

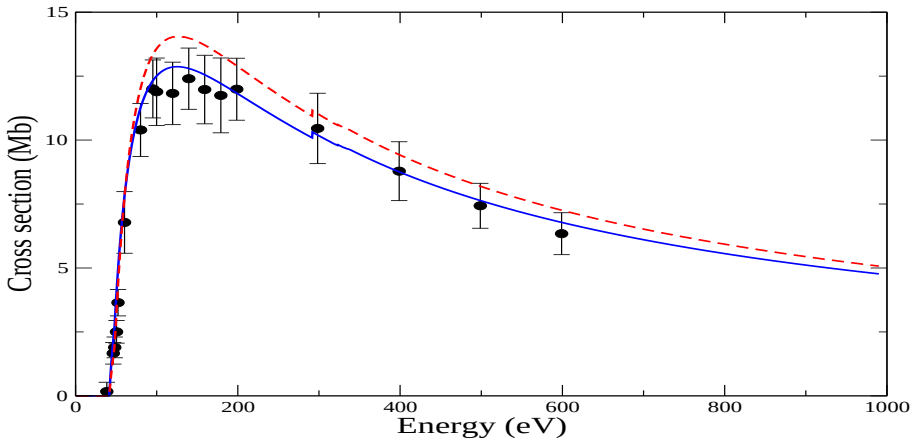

Figure 2. Electron-impact ionization cross section for C^{2+}. Experimental results are shown by the solid circles, the distorted-wave results are shown by the dashed line and the solid line show the R-matrix with pseudostates results. Both the distorted-wave and the R-matrix with pseudostates results use the experimentally measured metastable fraction. Figure adapted from Fogle et al. [25].

results by assuming a smaller metastable fraction, leading to cross sections that were 10-15% too high.

The study of Fogle et al. [25] also demonstrated the sensitivity of ionization rate coefficients to the near threshold part of the ionization cross section. Atoms/ions typically start to ionize at temperatures below their ionization potential. Thus, the most important rate coefficients for ionization balance calculations are the low temperature values which sample the near threshold part of the ionization cross section. As was shown by Juric et al. [26], using just the Younger fitting expression [27] can lead to large errors in the near threshold fit to the cross section. Fogle et al. [25] showed that errors of up to 50% could be introduced into the low temperature rate coefficients for C^{2+} if the Younger expression was used to fit the cross section. They recommended instead that a Chebychev polynomial fit or the Rost and Pattard [28] expression be used. The method shown in Dere [4] of scaling the rate coefficients and temperature grid also provides a useful means of accurately accounting for the near threshold part of the cross section. Thus, while some differences between existing ionization databases may be due to the underlying atomic cross section data, it is likely that many differences are due to the choice of fitting function used to fit the cross sections.

4. Excited state ionization

Recent work has shown that while DW methods become more accurate with increasing ion charge, they become less accurate with increasing n-shell. At the same time, classical or semi-classical results should perform better for excited state ionization but become less accurate with increasing ion charge. Thus, recent investigations have focused on determining ion charge and n-shell ranges where DW and semi-classical methods can be used for excited state (i.e. non-ground or metastable state) ionization cross sections. Excited state ionization processes often have very small ionization potentials, involving relatively slow moving electrons with a large amount of correlation in the collision process. As such they represent challenging theoretical calculations. Other than for excited state ionization cross section measurements of rubidium [29] and cesium [30] using an atom trapping technique, there are no measurements for excited state ionization cross sections. Thus, non-perturbative methods provide the only approach for

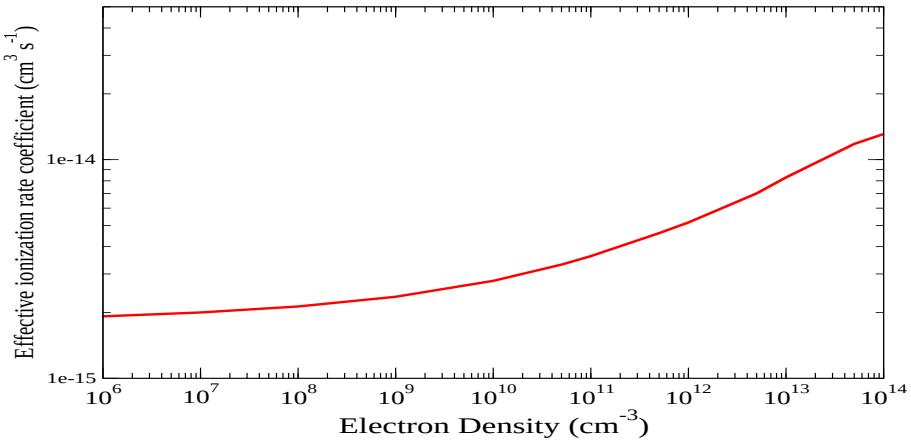

Figure 3. Effective ionization rate coefficient for neutral He as a function of electron density at Te = 1.8×10^4K.

calculating accurate excited state ionization cross sections, and for determining the ion stage at which perturbative methods become valid and at what n-shell classical/semi-classical methods are appropriate. Excited states play an important role in ionization for moderate to high density plasmas, where processes such as collisional excitation from the ground to an excited state can be followed by collisional ionization of the excited state. Thus, plasma modelers often use derived Generalized Collisional Radiative (GCR) coefficients when modeling emission and ionization balance. The GCR coefficients are described in detail by Summers et al. [7]. These rate coefficients are both temperature and density sensitive and include both effective ionization and recombination rate coefficients. We note that for ion stages that contain metastables there exist metastable coupling coefficients as well as effective ionization and recombination rate coefficients, see [7].

Considering ionization, the effective ionization rate coefficient that connects the ground (σ) of ion stage X^N with the ground (ν) of ion stage X^{N+1}, with connections via the set of excited states (i and j) in X^N , one obtains the effective ionization rate coefficient:

$$S_{CD,\sigma\rightarrow\nu} = \mathscr{S}_{\nu\sigma} - \sum_{j=1} \mathscr{S}_{\nu j} \sum_{i=1} \mathcal{C}^{-1}_{ji} \mathcal{C}_{i\sigma} \tag{1}$$

where $\mathscr{S}$ is the ionization rate coefficient, $\mathcal{C}$ is the collisional-radiative matrix and the summations are over the set of excited states in the ion stage. Fig. 3 shows the effective ionization rate coefficient for neutral He, from a recent compilation by Loch et al. [31]. The data presented here were generated using the ADAS suite of codes [32]. Other collisional-radiative codes, such as the Los Alamos suite, also produce GCR coefficients, though care must be taken when comparing different GCR datasets as the definition of the various GCR coefficients can vary.

These GCR coefficients can make a large difference to modeling results. Allain et al. [33] found that the effective ionization rate coefficient for neutral Li was more than an order of magnitude above the ionization rate coefficient for the ground state only, in good agreement with the GCR ionization data of Loch et al. [34]. These GCR data were then used in Li transport studies on the DIII-D tokamak. Doyle et al. [35] found that GCR data for Li-like systems was able to resolve a long standing discrepancy in Li-like emission lines in the solar atmosphere. These GCR coefficients represent a concise set of atomic coefficients that can be used in plasma transport codes without explicitly resolving of the collisional-radiative equations.

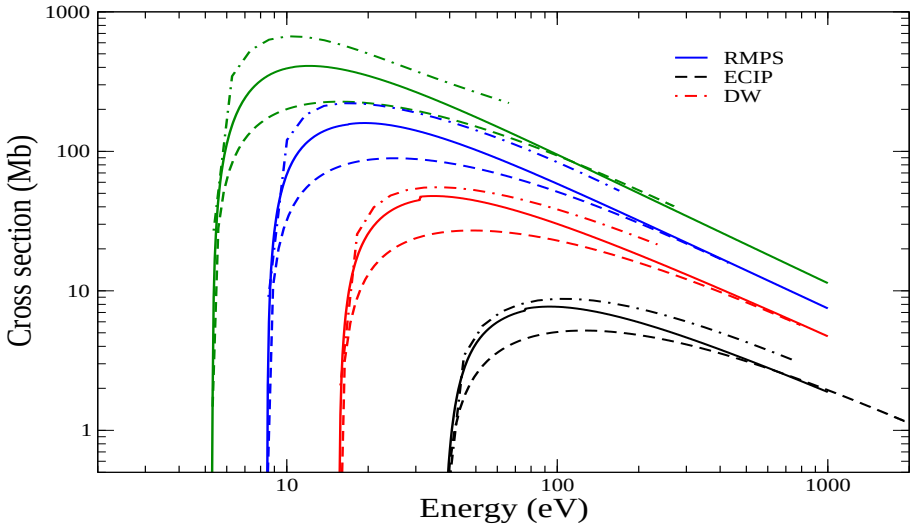

Figure 4. Electron-impact ionization cross section of $1s^2ns$ ionization cross sections for B^{2+}. The solid lines show the RMPS results, the dashed lines shows the ECIP results and the dot-dashed lines show the distorted-wave results. The lowest set of lines correspond to $1s^22s$, with the next higher ones being the $1s^23s$, $1s^24s$, and $1s^25s$ cross sections.

Thus, the remainder of this article will focus on recent efforts at calculating accurate excited state ionization cross sections using non-perturbative methods and comparing the results with distorted-wave and semi-classical calculations. For neutral H, Griffin et al. [36] used RMPS and TDCC calculations to show that distorted-wave results became worse with increasing n-shell, while classical trajectory Monte-Carlo results become accurate by n=4. Loch et al. [31] showed that for neutral H, the semi-classical ECIP method provides results in reasonable agreement with non-perturbative calculations.

For He, there is a discrepancy between measurements of the 1s2s ^3S metastable ionization cross section and all of the non-perturbative calculations. For the excited states, Loch et al. [31] used CCC [37] and RMPS results to show that ECIP calculations for excited state ionization were sufficient for modeling purposes for neutral He, while distorted-wave methods overestimated the cross section. For He^+ on the other hand, distorted-wave results produced accurate cross sections for the excited state ionization while ECIP results underestimated the cross section.

For systems beyond H and He, it is likely that there exists a range of near neutral ion stages where neither distorted-wave or semi-classical results are valid for ionization from excited states. Recent RMPS calculations of B^{2+} showed that neither ECIP or DW cross sections produced accurate excited state ionization cross sections, see Fig. 4. For neutral Ne, Ballance et al. [17] showed that neither DW or ECIP cross sections produce accurate cross sections for the excited states of neutral neon. The DW results continue to diverge from non-perturbative results with increasing n-shell, being up to a factor of three higher than the RMPS results at the peak of the n=5 cross section. The ECIP results stay consistently lower than the RMPS cross section results for all of the calculated excited states. This is shown in Fig. 5.

The ADAS database archives GCR coefficients and has data for all light species, up to Ne. These data have been undergoing review, using non-perturbative ionization data. This has led to new GCR data for Li [34], Be [38] and H/He [31]. We are currently working on B and C GCR data.

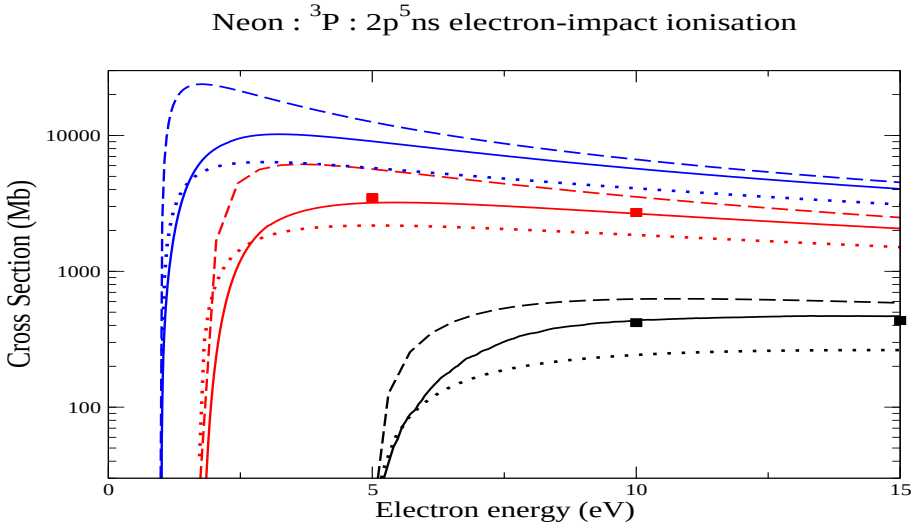

Figure 5. Electron-impact ionization cross section of the $2p^5ns\ ^P$ terms of neutral Ne. The black lines represent ionization of the $2p^53s\ ^3P$ term, the red lines represent ionization of the $2p^54s\ ^3P$ term, and the blue lines represent ionization of the $2p^55s\ ^3P$ term. The solid line shows RMPS results, the dotted line shows ECIP results and the dashed line shows distorted-wave results. The solid squares show the TDCC results for the $2p^53s\ ^3P$ and $2p^54s\ ^3P$ cross sections.

5. Conclusions

There has been much recent work on calculations of electron-impact ionization cross sections for atoms and ions. Non-perturbative methods have been shown to produce very accurate ionization cross sections and have allowed regions of validity of more approximate methods to be determined. For ground and metastable ionization cross sections non-perturbative methods have been found to produce accurate cross sections for light species, in good agreement with experiment. Distorted-wave methods usually become accurate for ground and metastable ionization for ion stages more than four or five times ionized. However, some discrepancies between experiment and distorted-wave results for ionized systems remain, mostly for open p- and d-shell systems.

There are numerous repositories of electron-impact ionization rate coefficients, many of which still show sizeable differences. Unless care is taken in the fitting of the fundamental ionization cross section, large errors can be introduced into the ionization rate coefficients. Thus, while some differences between the databases may still be due to the underlying cross sections, for ionized systems many of the differences may be due to the fitting functions used.

For plasmas where the excited states play a significant role in ionization, generalized collisional-radiative coefficients are more appropriate for modeling purposes. These data include ionization from excited states. Very little atomic data exists for these cross sections, thus modelers often have to rely on approximate methods. There is now much evidence that distorted-wave methods become increasingly inaccurate for higher n-shells. Results were presented for H, He, B and Ne where non-perturbative calculations have been used to determine excited state ionization cross sections. For H and He, it was found that semi-classical results are in good agreement with the non-perturbative calculations. For He^+, distorted-wave cross sections were in good agreement with non-perturbative calculations. However, for B^{2+} and Ne neither semi-classical or distorted-wave data produced cross sections in good agreement with non-perturbative calculations. Thus, it seems likely that for systems beyond H and He there exist a range of ion

stages where non-perturbative data have to be used. Work is continuing to further produce accurate excited state ionization cross sections for such systems.

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