Cross section data for electron collisions in plasma physics

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Abstract. We present a survey of cross section data for electron collisions used in plasma physics. Needs for cross section data have been identified in different fields of plasma physics and a brief review of existing data on electron/radical data has been presented. Experimental capabilities and recent results obtained in the Belgrade Laboratory for Atomic Collision Processes have been discussed.

1. Introduction
In scattering processes the strength of interactions among atomic particles could be viewed through observables called cross sections. The differential cross section, \(d(E, \theta, \varphi)\), is usually derived from the division of the flux of scattered particles through a large sphere of radius \(r\) within the solid angle \(d\Omega = \sin \theta \ d\varphi \ d\theta\) and the flux of incident particles. It has dimension of area and is energy dependent observable. It could be related with the scattering amplitude \(A_{\beta}\) by the following expression:

\[
d\sigma(E, \theta, \varphi) = \frac{j_f r^2 d\Omega}{|j_i|} = \frac{k_f}{k_i} |A_{\beta}|^2 d\Omega
\]

where \(j_f\) and \(j_i\) are current densities of scattered and incident particles, and \(k_f\) and \(k_i\) are corresponding wave vectors.

Essential input for modelling the processes in plasma physics consists of complete cross section data sets for all interactions. Subset of these data are collisional electron cross section data that include elastic and all inelastic processes (electronic and vibrational excitation, partial and total ionization, dissociative electron attachment, etc.) Data are needed for ground state species as well as metastables and other excited atomic particles and/or radicals.

In The Laboratory for Atomic Collision Processes at The Institute of Physics (LACPIP), Belgrade, the concentrated effort has been put into cross section measurements funded by two subsequent
national research projects (2002-2005 and 2006-2010) [1] in order to establish data sets needed for collisional community and modelling purposes. These measurements cover rare gas atoms (He [2], Ar [3], Kr [4], Xe [5]) metal atoms (Cd [6], Zn [7], Ca [8], Yb [9], Mg [10], Pb [11], Bi [12]), small molecules and molecules of biological interest (O$_2$ [13], H$_2$O [14], THF [15], THFA [16], glycine and alanine [17]). Differential (DCS) and integrated (integral and momentum transfer) cross sections for elastic scattering and excitation processes have been obtained for numerous species. Energy loss spectra have been recorded together with threshold electron spectra. Several different techniques have been exploited in the experimental set-ups for production of monochromatic beams and detecting the scattered electrons.

2. Electron collision cross section data in plasma physics

The needs for electron cross section data depend on the subfield of plasma physics and parameters and characteristics of plasma. Generally, one can observe diverse conditions of plasmas i.e. densities, temperatures, energy distributions of atomic particles, particle compositions, etc. Depending of what is the major subject of the study, whether it is astrophysical or fusion plasmas, plasmas in gas discharge lasers or plasma etching processes in semiconductor manufacturing, or just optical plasma diagnostics, one should consider appropriate set of elementary atomic particle processes and accordingly adequate cross sections.

Numerous atomic and molecular species are present in plasmas and are used in applications such as etching and deposition. Very often rare gas atoms are present and considered as plasma constituents together with di-, tri-, and polyatomic molecules as well as radicals and other fragments. These species could be in their ground state or excited (and metastable) states. In the study of electron interaction with these atomic particles, the variety of processes is possible and each of them is characterized by cross section that is energy and angular dependent value.

2.1. Needs for cross sections in different types of plasmas

Regarding the needs for atomic and molecular data in astrophysical plasmas, Jorissen [18] identified two major purposes where data are used: (i) in computing opacity of the stellar matter and (ii) in determining abundances for specific chemical elements. He had recognized several current problems where atomic data are important for appropriate solutions: iron problem and the question of estimating stellar metallicity, oxygen problem, the importance of accurate knowledge of energies, transitions, and oscillator strengths for heavy elements, problem of dating the oldest galactic stars, etc.

In fusion plasmas accurate atomic and molecular data sets are used to model conditions in tokamak edge radiation and particle behavior in cold divertor regions (Kubo [19]). For controlling the impurity particle transport and edge plasma radiation losses, data for effective ionization and recombination rate coefficients are required. Currently Ar atoms are injected for radiation loss power enhancement but the role of Kr atoms is envisaged in this process. Also study of W and other heavy atoms is a priority due to their use as divertor plates. Collisions of hydrogen molecule, hydrocarbon molecules and He atoms have been extensively studied in cold divertor plasmas. Regarding the electron collision processes major investigations are directed toward the understanding of the role of vibrationally excited H$_2$ molecules [20].

Plasma processes in gas discharge lasers are well understood and described in detail. Electron collisions play an important role in creating population inversion. They can easily populate metastable states as the scattering process could be viewed as an interaction of a multipole with the atomic field and electron cloud. Determining the electron excitation cross sections of optically forbidden states could serve as a test for lasing properties of the particular element. By the development of ultrafast laser technology, when the sub-femtosecond pulses are achieved, the study of the re-scattering processes of electrons emitted under strong laser field with their parent ions becomes possible with high precision [21].

Challenge in today’s semiconductor manufacturing industry is using the plasma etching processes for producing nanometric patterns. The new gas chemistry for high-performance SiO$_2$ patterning in
sub-0.1 µm has been rapidly developing. Samukawa and collaborators have demonstrated that the charging damage of devices in SiO₂ etching would be more suppressed when using C₂F₂/CF₃I chemistry than C₄F₈/Ar chemistry [22]. This is primarily due to different electron energy distribution functions associated with the certain plasma composition.

For optical plasma diagnostics the knowledge of electron cross sections for rare gas atoms is crucial. Boffard and collaborators have demonstrated that by knowing these cross sections and combining them with plasma emission measurements it is possible to extract many plasma parameters [23]. Cross section data are needed for both ground state and metastable states atoms. The role of cross sections for metastable atoms had been emphasized in the recent review on plasma electronics by Makabe and Petrović [24]

2.2. Cross sections for electron interactions with radical species

Radicals, as atomic and molecular species with unpaired electrons, are common constituents in plasma media and are seen as a key chemical component in many plasma applications in semiconductor manufacturing. Although of great practical importance, data sets of electron collisional cross sections with radical species is rather scarce in scientific literature. It is partly due to the difficulties in experimental handling of radicals as unstable and highly reactive species. That is why the available literature is mainly consisting of theoretical data.

2.2.1. CFₓ (x=1,2,3) radicals. The main feed gases used in the plasma etching processes are perfluorocarbons but these are also strong greenhouse gases [25]. CF₂ radicals are used as a gas precursor for polymer deposition, while CF₃⁺ ions are used as a dominant etchant for silicon dioxide films due to their high etching yield [26].

CF radicals have been studied by several authors. Lee et al [27] used a complex optical potential method to calculate elastic differential, integral and momentum transfer cross sections as well as total and absorption cross sections in the energy range from 1 to 500 eV. They made a comparison with the cross sections for isoelectronic NO molecule. They found that DCS data are larger for CF than NO molecule especially in the domain of smaller scattering angles (below 60°). Rozum et al [28] exploited R-matrix theory to obtain elastic and excitation cross sections at the low energy region (below 10 eV). They also found three shape resonances of different symmetries. Most recently Trevisan et al [29] investigated resonant electron – CF collision processes which could lead to production of negative ions. They studied the vibrational excitation and electron attachment processes and found several low-lying negative ion states which are expected to dominate the scattering process.

Electron - CF₂ radical cross sections have been investigated by Rozum et al [30] and Lee et al [31]. Rozum et al [30] have used R-matrix method to treat low-energy electron collisions (less than 10 eV) and calculated elastic and excitation cross sections of the six lowest-lying electronically excited states. Lee et al [31] have used an iterative Schwinger variational method combined with the distorted-wave approximation to solve the scattering equations. Cross sections are deduced in the energy range from 1 to 500 eV.

Electron - CF₃ radical cross sections have been studied by Diniz et al [32] and the calculations are performed with the Schwinger multichannel method at the static-exchange level. They have calculated elastic DCS in the incident electron energy range from 6.5 to 30 eV. Rozum et al [33] have performed R-matrix calculations to treat electron collisions with this polyatomic radical at its equilibrium geometry using a coupled states expansion. They have found no low-lying resonances and only bound state of CF₃⁻ had been detected. Antony et al [34] have calculated total ionization cross sections for a number fluorocarbon molecules and radicals in the broad energy range from threshold to 2 keV. Most recently new theoretical electron impact cross sections have been obtained for CFₓ (x=1,2,3) radicals and recommended set of data has been given for electron scattering total, excitation, momentum transfer, and elastic integral, electron impact dissociation and dissociative electron attachment processes [35].
Experimental investigation of CF$_x$ (x=1,2,3) radicals started by Becker and collaborators [36] who have measured absolute partial cross sections for the parent ionization from threshold to 200 eV as well as appearance energies. Deutsch et al [37] have measured and calculated absolute total cross sections for the single ionization of CF$_x$ radicals by electron impact.

2.2.2. NF$_x$ (x=1,2) radicals. First experimental and theoretical data of electron impact cross sections on NF$_x$ (x=1,2) radicals have been obtained by Tarnovsky et al and Deutsch et al [37]. They have determined total cross section for the single ionization process in the energy range up to 200 eV. Huo et al [38] calculated electron impact cross sections using the simplified improved binary-encounter dipole model (siBED) in the energy range up to 200 eV. They also presented the revised experimental cross sections for free CF$_x$ (x=1,2,3) radicals and free NF$_x$ (x=1,2) radicals.

2.2.3. SF$_x$ (x=1-5) radicals. Electron impact ionization of free radicals that contain sulphur atom have been studied by Tarnovsky et al [39] up to 200 eV impact energy. They have measured single ionization cross sections for the SF$_3$ and SF$_5$ free radicals and also have presented calculated cross sections for all SF$_x$ (x=1-5) radicals. A comparison of the experimentally determined cross sections, with calculated cross sections based on the modified additivity rule, showed very good agreement. Asgar Alia et al [40] have performed calculations of cross sections for SF, SF$_2$, SF$_3$, SF$_4$, and SF$_5$ by the binary-encounter-Bethe (BEB) model. The covered energy range was from threshold to 5000 eV. Joshipura et al [41] have calculated total ionization cross sections for a number of radical species: CH$_x$, CF$_x$, SiH$_x$ and SiF$_x$ (x = 1–4). Total ionization cross-sections of electron impact are calculated based on Complex Scattering Potential approach at incident energies 20 – 3 000 eV.

3. Experimental techniques and procedures in LACPIP

At the LACPIP, Belgrade interactions of electrons with molecules (atoms) have been studied in the low and medium energy ranges. In the crossed-beam arrangement where an electron beam has been perpendicularly crossed by effusive atomic/molecular beam, the scattered electrons have been analyzed in high resolution electron spectrometers: ESMA [7], UGRA [4] and SPEPRA [13].

3.1. Electron spectrometer ESMA

Electron spectrometer ESMA consists of the molybdenum hemispherical selectors in monochromator and analyser, the gold plated OFHC cylindrical lenses, the rotating plate on which the analyser is mounted and that provides assessment of scattering angles from -30$^\circ$ to +150$^\circ$ in respect to incident electron beam. Perpendicularly to the scattering plane defined by the incident and scattered view cones of the monochromator and analyser, the resistively heated oven is placed on together with the several layers of tantalum shields and copper shield in outermost radius as well as the copper cold finger cup for collecting metal atom vapours.

The ESMA spectrometer could be run in three different modes of operation: (i) energy loss mode, (ii) angular distribution mode, and (iii) incident electron energy mode. Experimental procedure follows several steps: obtaining the well collimated electron beam with the current of the order of 1 to 10 nA; tuning the monochromator and the analyser to the optimized signal to noise ratio; checking the real zero position of the scattered signal from the symmetry at positive and negative scattering angles; checking the energy scale usually from the position of the known resonance structure in the elastic cross section or the appearance of the inelastic channel (excitation threshold) in incident electron energy mode; obtaining the energy loss spectrum; determining the angular resolution through measuring the angular spread of electron beam and defining the apertures at both the monochromator and the analyser; making measurements on angular distributions of elastically and inelastically scattered electrons; measuring the intensity ratios of most pronounced inelastic feature (called resonance peak) to other peaks in the energy loss spectra at several different scattering angles. In order to obtain the relative cross sections the effective path length correction factors are applied to the angular distributions as well as the transmission factor defined from the intensity ratio measurements.
The absolute values of differential cross sections are obtained through the normalization procedure: (i) to the optical oscillator strengths or the observed transition, (ii) to the integral cross sections, or (iii) to the calculated cross section at the particular energy and scattering angle.

The accessible energy range at the spectrometer is from 5 eV to 100 eV, overall energy resolution from 40 meV to 120 meV, while the angular resolution has been estimated to be 1.5°. Differential cross sections for heavy noble gas atoms (Ar – Xe), small molecules (N₂O, H₂S) and amino acids (Gly, Ala) had been obtained at this spectrometer. But the main advantage of the current design is that both the monochromator and analyser electron optics are well shielded and differentially pumped what allows work with the metal vapours. A number of targets [6-12] have been investigated in the ESMA spectrometer, many of the transitions have been identified and numerical values of the DCSs have been determined for the first time in literature.

3.2. Electron spectrometer UGRA
Electron spectrometer UGRA consists of the high current electron gun (thoriated tungsten hairpin cathode or pure tungsten cathodes are used) and the double cylindrical mirror energy analyser (DCMA) proceeded by a four-element cylindrical electrostatic lens. After being selected by energy, the electrons are focused by a three-element cylindrical electrostatic lens into a single channel electron multiplier working in the single counting mode. The electron gun is fixed on the turntable, which can be rotated around the gas needle in the angular range of about −40° to +130° with respect to the axis of the entrance optic of the DCMA. All the elements of the system are placed inside a double metal shield, which reduces the Earth and other stray magnetic fields to less than 2×10⁻⁷ T. The whole electron spectrometer with the experimental procedure and SIMION simulations have been described in detail by Milosavljević et al (2006) [4].

The accessible energy range at the spectrometer is from 20 eV to 350 eV, overall energy resolution is 500 meV due to the initial thermal spread of the electrons, but for the DCMA alone, the obtained energy resolution is down to 30 meV. The angular resolution has been estimated to be about 2° to 3°. Critical positions of differential cross sections for noble gas atoms (Ar, Kr) has been determined in the broad scale measurements that linked both the energy dependence and angular dependence of the cross sections. This procedure turns out to be crucial for the accurate determination of the energy and angular positions where DCS values attain the minima values. A several molecular targets of biological interest, like DNA deoxyribose analogue molecules [15-16, 42] have been investigated in the UGRA spectrometer.

3.3. Electron spectrometer SPEPRA
The electron spectrometer SPEPRA is a threshold spectrometer designed for detecting electrons that have lost almost all energy in the scattering process. The interaction volume is surrounded by a transparent mesh and a small positive potential difference is supplied between the extracting electrode and the mesh. Scattered electrons with the energy distribution between zero and 10 meV are trapped by this potential and tunnelled toward the analyser. This technique is based on the improved field penetration method [43]. The obtained energy resolution is better than 30 meV.

In the threshold spectrum, the large number of features which correspond to optically forbidden states and core excited resonances have been observed and identified according to their energy positions and energy spacing between vibrational levels. Also, in the non-resonant contribution to the threshold spectrum, the mixing of Rydberg and valence states is recognized [13-14].

4. Conclusions
The need for a comprehensive data bases of electron interactions with atomic particles has been identified long ago. International Conference on Atomic and Molecular Data and Their Applications [44] (a continuing series of the conferences) has been devoted to this aspect of research activities. Recent development in the field has been summarised also by Mason [45] who had examined new
developments in electron induced processing, reviewed the current status of databases and identified the most important needs in the future electron/molecule research.

A survey of cross section data for electron collisions used in plasma physics has been presented. Needs for cross section data have been identified in the fields of plasma physics such as astrophysics and fusion plasmas, as well as in the fields of applications: gas discharge lasers, semiconductor manufacturing processes and plasma diagnostics. A brief review of existing data on electron/radical data has been presented with the accent on radicals containing fluorine atoms. The focus has been put onto electron ionization processes while dissociative electron attachment processes have not been covered since they could be found elsewhere in the literature [46].

Experimental capabilities and recent results obtained in the Belgrade Laboratory for Atomic Collision Processes have been discussed. Three different types of electron spectrometers have been presented, each having a specificity and different use in investigating electron/atomic particle interactions. Recent cross section data obtained at the LACPIP has been listed as an extensive list of references.

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