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Calculation of electron-impact ionization cross sections: Bottom-up inductive vs. top-down deductive approaches

H. Deutsch[a], F.X. Bronold[a], K. Becker[b,c,*]

[a] Institut für Physik, Universität Greifswald, Germany
[b] Department of Applied Physics, NYU Polytechnic School of Engineering, Brooklyn, NY, USA
[c] Department of Mechanical and Aerospace Engineering, NYU Polytechnic School of Engineering, Brooklyn, NY, USA

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ABSTRACT

This paper attempts to elucidate the relationship between bottom-up inductive and top-down deductive approaches to the calculation of electron-impact ionization cross sections for atoms. Specifically, the ionization cross sections for atomic hydrogen and helium derived from the various approaches are compared in detail.

1. Introduction

The production of charged particles by electron impact is among the most fundamental processes in collision physics. Cross sections for electron-impact ionization of atoms (as well as molecules) have been measured and calculated since the early days of collision physics (see e.g., [1–5]) because of their fundamental importance to our understanding of collision physics and their relevance in many applications. Electron-impact ionization cross sections are important quantities in a variety of applications and technologies as diverse as low-temperature processing plasmas, fusion edge plasmas, gas discharges, planetary, stellar, and cometary atmospheres, radiation chemistry, mass spectrometry, and chemical analysis [2].

Much progress has been made in the experimental determination of cross sections for atomic and molecular targets [1–6] in the past three decades. Rigorous quantum mechanical calculations of ionization cross sections have only appeared in the literature recently and only for some simple atoms in their electronic ground state and in very limited energy ranges (see e.g. Ref. [7]). The rigorous theoretical treatment of ionization cross sections for a wide range of impact energies from threshold to thousands of electron volts (hereafter referred to as "deductive approach", since they start from general principles and deduce expressions for special cases under well-defined conditions) of an atom is beyond the capability of current quantum-mechanical electron collision theory [8–10]. The need to incorporate ionization cross sections for these targets in modeling codes for various applications (see Refs. [11–16]) has stimulated interest in the use of less rigorous approaches such as semi-rigorous methods that incorporate aspects of established collision theories and some quantum mechanically calculated target properties to the calculation of the ionization cross section (hereafter referred to as "inductive approaches", since they start from special cases and attempt to derive expressions of general applicability) as well as various fitting formulas (see e.g. Ref. [17]).

The most frequently used deductive approaches to the calculation of single ionization cross sections of atoms (i.e. the removal of a single target electron as a result of the impact of an incident electron) are based on the Born approximation [18] or the Bethe approximation [19], whose origins date back to the early part of the 20th century. These approaches are well suited to represent the ionization cross sections in the regime of high impact energies (typically for energies above about 50 times the ionization threshold, which is similar to what was observed for excitation cross sections (see e.g. [20]). The inductive approaches gained prominence starting in the late 1980s. The first semi-rigorous ionization cross section calculation approaches appeared in the literature in 1987 by Khare and co-workers [21] and by Deutsch and Märk (DM formula) [22,23] and, a few years later, the binary–encounter dipole (BED) and binary-encounter Bethe (BEB) approaches of Kim, Rudd, and co-workers [24] emerged. The inductive DM formula is based on a fitting procedure using a few reliable experimentally determined

* Corresponding author at: Department of Applied Physics, NYU Polytechnic School of Engineering, Brooklyn, NY, USA. Tel.: +1 718 260 3608.
E-mail address: kurt.becker@nyu.edu (K. Becker).

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ionization cross sections in conjunction with quantum mechanically calculated target properties, general assumptions, scaling laws, and similarity arguments to arrive at an easy-to-use cross section formula that can be applied to a variety of targets, not just to atoms. Lastly, we mention the empirical Lotz formula [25,26], which is a comparatively simple, easy-to-use 3-parameter fit formula for the calculation of single ionization cross section for the electron-impact ionization of ground-state atoms. The agreement between the prediction of the Lotz formula and measured cross sections was found to be remarkably good for light atoms (low atomic number Z), however, the Lotz cross section overestimates the cross section for heavy atoms [10].

Until the late 1960s, Born-type models were widely used with various attempts to increase their range of validity and accuracy to lower energies by introducing electron-electron exchange between the projectile and target electron, but with varying success as discussed by Rudge [8] and addressed in more detail in Section 3.4 of this paper. At the end of the 1970s, significant progress had been made in the application of optical potential [27], coupled-channels and R-Matrix methods [28–30] to describe electron-atom discrete inelastic scattering. Furthermore, powerful computers that allowed solutions of the time-dependent Schödinger equation provided, in principle, a convergent expansion of the scattering wave function in terms of a complete set of target states. Bray and Stelbovics [31] were the first to show that their formalism – a convergent method to treat the continuum states – can be used to extract total ionization cross sections with electron exchange fully included. These authors also applied this method to study the fundamental three-body problem of electron hydrogen scattering.

This article has a three-fold purpose. First, we show that the inductive DM formalism can be related to the deductive Born/Bethe approximations. We show in particular how the ad hoc weighting factors g_{nl} in the DM formalism relate to the ionization factors b_{nl} in the Bethe approximation. Second, we demonstrate that calculations of electron-impact ionization cross sections using the Bethe, DM, BED, and Lotz approaches have a basic common feature: a calculation of the high-energy behavior of the cross section is "corrected" in the regime of low impact energies by introducing interference or exchange terms. Third, we show that the "low-energy correction" in all four cases results in very similar cross section shapes.

2. Theoretical background

Electron impact ionization is a reactive break-up collision with two indistinguishable electrons in the exit channel that interact with each other and the residual ion core. The fundamental concepts controlling this process as well as the general expressions for the collision cross sections have been known for many years [8,32–34], yet reliable quantitative calculations of ionization cross sections beyond perturbation theory (see e.g. Ref. [8] for references to work performed up to the 1960s and Refs. [35–41] for more recent work) have been performed only recently using large-scale computational methods [42–53]. The collision-theoretical background of electron impact ionization is well established in the literature [8,32–34,54–59]. We refer the reader in particular to the excellent paper of Rudge [8] and the work of Friedrich and co-workers [60,61]. A review of the literature shows that top-down calculations of ionization cross sections starting from the N-electron Schrödinger equation are extremely demanding (deductive approach). The challenges of the top-down approach is most clearly seen when one expresses the ionization cross section in terms of the ionization amplitude. A rigorous treatment requires knowledge of the exact break-up wave function, which even in the simplest case of the ionization of atomic hydrogen is unknown

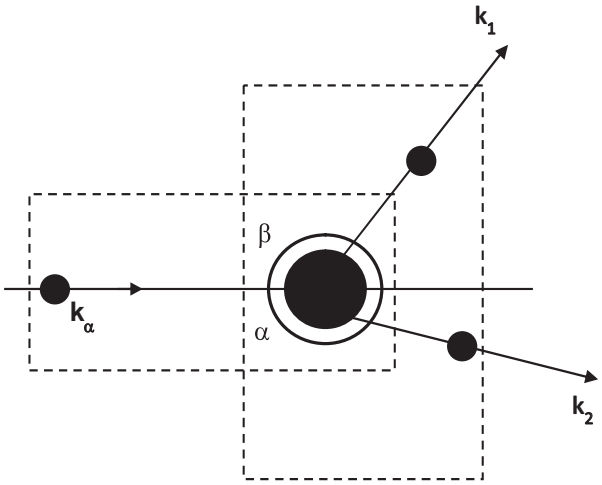

Fig. 1. Schematic diagram of the collision break-up geometry. The incident electron, with momentum represented by the vector $\boldsymbol{k}_\alpha$ hits the target (initially in state "α") leaving it behind in state "β". The two electrons in the exit channel are represented by their momentum vectors, $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$, respectively.

because of the long-range Coulomb interaction between the scattered and ejected electrons and the residual ion core [58,59]. Despite occasionally successful theoretical work, it is safe to say that the assertion by Gerjoy expressed in 1965 [62] that brute force computations of ionization cross sections are most likely to fail still holds largely true today. This, in turn, makes phenomenological bottom-up methodologies using cross section formulae which have been fitted to a varying extent to a selected set of experimental data to as yet unexplored situations [10] (inductive approach) very appealing and an invaluable tool.

2.1. The Born approximation

The Born approximation has been discussed extensively in the literature and we will limit the following discussion to the bare minimum that is required as a basis for the subsequent discussions. Fig. 1 illustrates the general break-up collision with two particles in the entrance channel and three particles in the exit channel. The incident electron hits the target, which is initially in a state "α", with momentum k_α leaving behind the target in state "β" and two electrons leaving the interaction zone with momenta k_1 and k_2, respectively. Accounting for exchange, the spin averaged total ionization cross section as a function of energy E, $\sigma_{\alpha\to\beta}$ (E), has the form (see Ref. [60]):

$$\sigma_{\alpha\to\beta}(E) = \int_0^{E_{\max}} dT_2 \int d\omega_1 d\omega_2 \cdot \frac{k_1 \cdot k_2}{4\pi \cdot k_\alpha} \cdot |f_\beta^{av}(\omega_1, \omega_2, T_2)|^2 \qquad (1)$$

where $E_{\max} = (E - E_\beta)/2$. Here ω_1 and ω_2 are unit vectors in the direction of respectively k_1 and k_2, T_2 is the energy of the ejected electron and E_β is the energy of the target after the collision when it misses one electron. $f^{av}{}_\beta(\omega_1, \omega_2, T_2)$ is the spin-averaged ionization amplitude implicitly defined by

$$|f_\beta^{av}(\omega_1, \omega_2, T_2)|^2 = |f_\beta^0(\omega_1, \omega_2, T_2)|^2 + 3|f_\beta^1(\omega_1, \omega_2, T_2)|^2 \qquad (2)$$

where S = 0,1 is the total spin of the two electrons.

The cross section in equation (1) is given in units of "$\pi a_o{}^{2}$", where a_o is the Bohr radius and all energies are measured in Hartrees (atomic units). We note that throughout the remainder of this paper, we will use SI units whenever possible.

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