

Electron Impact Ionization Cross Section- Semi-empirical approach

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Abstract

This review pertains to the various semi-empirical methods used to calculate collision cross-section of atomic particles (from mono to polyatomic particles having mass from 1 amu to about 120 amu) by electron impact ionization process have been discussed. The semi-empirical methods mostly used for finding the precise cross-section data theoretically, where experimental data is not available and/or have limited data. Moreover, the paper describes advancement of these methods for calculating cross-section for complex molecules also. Some remarkable applications in applied sciences are also discussed. The paper discusses the various theoretical formalisms used by different authors with different parameters such as Born Approximation, Binary Encounter Bethe, R-matrix method, Spherical complex optical potential and among others. Those were improved time to time for the better result to determine total cross-section data.

Keywords: Scattering processes, electron impact ionization cross-section and semi-empirical approach.

1. INTRODUCTION

Atoms and molecules are of fundamental importance in physics and chemistry. They have different properties which can be obtained by collision studies. The collision occurs between the projectile like an electron, ion or photon, and target (atom or molecule). During the collision, many processes take place and may be elastic scattering (where the kinetic energy of the projectile does not change), inelastic scattering (where the projectile loses energy and the target recoils or changes mass) or absorption (where the projectile disappears in the target). During inelastic scattering, many channels can occur like dissociation, ionization, excitation

etc. The probability occurring of a particular process can be determined in term of the quantity known as a cross-section. In other words, the cross-section is defined as "the ratio of the number of events per unit time and per unit scatterer, to the flux of the incident particles with respect to the targets". In a simple way, we can say, the cross-section is the effective area of the target that interacts with the incident beam and scatters it (C J Joachain, 1983).

$$\sigma_{tot} = P \times N_A / \Phi_A \times n_B$$

Where

σ_{tot} = Scattering cross section

P = Probability that an incident particle interacts with the target and thereby gets removed from the incident flux by scattering

N_A = Number of particles A reaching the target per unit time

Φ_A = Flux of A with respect to target B

n_B = number of target particles B that intercept the incident beam

There are various methods of inelastic scattering such as Electron impact ionization, Electrospray ionization, Fast-atom bombardment, Field ionization, Laser ionization, Thermal ionization etc. But electron impact ionization is the most used method for inelastic scattering processes. The collision depends on the different parameters of projectile and targets like incident energy, solid angle, impact parameter etc. So, the scattering phenomenon gives ample information about the atomic and molecular system. For example, the strength of electron-electron and electron-nucleus interaction inbound state system that can be obtained by collision experiments. The study of electron-atom or molecular collision has also been strongly motivated by the need of data for testing and developing suitable theories of the scattering and collision processes (Taj, Manaut, and Oufni, 2011), which provides a tool for obtaining details information as the structure of target atoms or molecules (Weigold, Hood, & Teubner, 1973; Furness & McCarthy, 1973).

Different theoretical formalisms are used for calculating total ionization cross-section depending on the quantum collision theory used. Oftenly, the semi-empirical approaches are used for calculating the Electron Impact Ionization Cross Section(EIICS) theoretically inconveniently applied to limited types of targets and/or limited energy range (from threshold to few KeV).

Therefore, here we will discuss the development of some semi-empirical approach for evaluating the electronic structure information and all other atomic properties which are used to determine the absolute ionization cross-section for different molecules.

2. SEMI-EMPIRICAL APPROACH

There are many theoretical models from which the EIICS can be determined more accurately or correlate to the experimental results. These methods are helpful in calculating the cross-section of atoms, molecules, clusters, ions and radicals. The theoretical or empirical formulas were given by Thomson (1912), Bethe (1930), Elwert (1952), Knorr (1958), Gryzinski (1959), and Drawin (1961) but among these, the best approximation has been given by Drawin (1963). Later on, this approximation has been modified by different authors from time to time. Rudge and Schwartz (1965) used these theoretical approaches for analysis for He⁺. Green and Barth (1965) proposed the following expression for calculating the cross-section:

$$\sigma(E) = \frac{q_0 A}{W^2} \left(1 - \frac{W}{E}\right)^\nu \left(\frac{W}{E}\right)^{1-\omega} \quad (1)$$

Where

E = incident energy of the electron,

W = excitation energy,

$q_0 = \pi a_0^2 (2 R_e)^2 = 6.513 \times 10^{-14} \text{ cm}^2 \text{ eV}^2$, and

A, ν , and ω are constants adjusted to reproduce the experimental data.

Green (1966) has shown that this semi-empirical method is used to predict the cross-section approximately within 5% experimental error from threshold up to about 100eV. Mort and Massey (1965) applied Born Approximation for the determination of inelastic collisions cross-section. They use the Bohr radius, $a_0 = \hbar^2/m_e^2 = 0.5292 \times 10^{-8} \text{ cm}$, as a unit of length and the Rydberg Energy, $R_e = \hbar^2/2ma_0^2 = m_e^4/2 \hbar^2 = 13.60 \text{ eV}$, as a unit of energy. A dimensionless momentum transfer parameter was introduced, which is defined by

$$X = a_0^2 K^2 \quad (2)$$

Simple mathematical expression results the following equation:

$$X = \frac{2E}{R_e} \left[1 - \cos \theta \left(1 - \frac{W}{E} \right) \frac{1}{2} - \frac{W}{2E} \right] \quad (3)$$

Here

$K = k - k' \rightarrow$ difference between incident and outgoing propagation vectors

$W = E_n - E_0 \rightarrow$ excitation energy for the n^{th} state from the ground state.

This gives the symmetric cross-section with respect to the excitation energy of the incident particles.

Wolfgang Lotz (1967) proposed the new formula to determine single ionization cross section of atoms or ions. He states that there will be higher kinetic energy (E) of the scattered particles than the binding energy (P_i) of the target atoms/ions. He used three constants (a, b, c) in cross-section measurement. The formula for cross section is as (Green, 1967)-

$$\sigma = a \left[1 - b \exp \{ -c(U-1) \} \right] \sum_{i=1}^N q_i \frac{\ln \left(\frac{E}{P_i} \right)}{EP_i} \quad (4)$$

Where $E \geq P_i$

This gives the single ionization cross-section data of different atoms within 5-10% experimental error from the ground state to 10 KeV.

Deutsch et al. (2000) give the related details to this method and proposed the simple expression for total cross-section :

$$\sigma = \sum_{i=1}^N \sigma_i; \quad \sigma_i = q_i \sigma^{ei} \quad \text{where } N = \text{number of subshells.}$$

3. BINARY ENCOUNTER BETHE (BEB)

The collision may be elastic or inelastic. The inelastic collision may be a hard collision or soft collision depending on the impact parameters. Mott and co-worker (1930) gave the 'Mott cross section formalism', using Rutherford cross-section (Landau Lifshitz, 2000) where electron exchange effect takes place, which is considered as hard collisions. On the other side, Bethe gives 'Bethe cross-section formalism' developed through first Born approximation (Inokuti, 1971), for dipole interaction which involves fast incident electrons accounting soft collisions. Among these, the most widely used method is the Binary Encounter Bethe theory of Kim, Rudd, and co-workers (1997). Binary Encounter Bethe (BEB) model combines the Mott cross-section and the Bethe cross-section. It gives the particular molecular orbital total cross-section by electron impact ionization (Huber, Sukuba, Urban, Limtrakul, & Probst, 2016) as

$$\sigma = \frac{s}{t+u+1} \left[\frac{\ln(t)}{2} \left(1 - \frac{1}{t^2} \right) + 1 - \frac{1}{t} - \frac{\ln(t)}{t+1} \right] \quad (5)$$

After that, they refined Binary Encounter Bethe theory and suggested the new expression. As

$$\sigma_{BEB} = \frac{s}{t+(u+1)/n} \left[\frac{Q \ln(t)}{2} \left(1 - \frac{1}{t^2} \right) + (2 - Q) \left(1 - \frac{1}{t} - \frac{\ln(t)}{t+1} \right) \right] \quad (6)$$

Here

$t = T/B$, $u = U/B$, $a_0 = \text{Bohr radius } (0.52918 \text{ Å})$, $S = 4\pi a_0^2 N R^2 / B^2$ and R , T , N , B , U , and Q represent the Rydberg energy (13.6057eV), incident electron energy, occupied electron number, binding energy, average kinetic energy and dipole constant respectively.

Kim and Rudd (1994) further improved the previous formalism in different types. They combined the modified form of the Mott cross-section and the Bethe cross-section and developed some other approximations including single differential ionization cross-section for a particular subshell based on Binary Encounter Dipole (BED) model.

This theory discussed the knowledge of molecular wave functions in details. Deutsch and co-worker (1999) compared the different 31 molecules and free radicals (Deutsch, Becker, Matt, & Märk, 2000) including both BED and BEB model calculation of experimental data with Khare and co-workers. The result showed the best agreement between theoretical and experimental data within 5-10% experimental error from threshold to few KeV.

4. R-MATRIX FORMALISM

Wigner (1946) first time introduced the R-matrix theory. Wigner and Eisenbud (1947) work on nuclear reaction using R-matrix method theoretically. Burke *et al.* (1970) improved the method to determine the scattering cross-section of the target at each boundary. Later on, Burke *et al.* (1977) further developed these ideas for diatomic molecules and Morgan *et al.* (1997) extended these studies to polyatomic molecules. Burke *et al.* (2007) and Tennyson (2010) work on scattering theory using R-matrix method for atomic, molecular and optical processes. Recently, Kaur and Baluja (2012, 2015) studied the electron impact of NCO and SiN molecule respectively using R-matrix method and compare the elastic and inelastic total cross-section, ionization cross-section, and differential cross-section with BEB model which is in the better agreement with experimental adiabatic electronic affinity.

Hence, for low energy electron scattering calculation, Quantemol N code based on the R-matrix method is employed. The results obtained provide the better agreement for the cross-section from very low (0.01eV) to significantly high incident energies (1000 eV) for any target (including radicals and other chemically unstable molecules). Thus, using this theoretical methodology, many electron impact cross-sections can be obtained.

The R-matrix theory (Burke & Berrington, 1993) considers the behavior of an incident electron in different regions of the configuration space. This space is divided into two regions: an inner region (where electron close coupling interaction occurs) and an outer region (where the normal potential is experienced by an electron). The Schrödinger equation is used to obtain the total wave function by solving separately in each region at each boundary. In the R-matrix method an *ab initio* has been applied for more accurate results. However, for larger molecules, this method becomes increasingly complex and requires large computational efforts. Thus the R-matrix calculations are implemented only for small systems having low scattering energy (10-15 eV). So, such theories are not applicable for complex molecules. Furthermore, this involves the time-consuming computation even for small molecules.

5. SPHERICAL COMPLEX OPTICAL POTENTIAL FORMALISM

Joshiyura and co-workers (2004 and 2010) used the Spherical Complex Optical Potential (SCOP) method to compute the electron scattering cross-section with a variety of targets. These calculations are of great importance particularly for the complex molecular targets where the experimental determinations are difficult. The SCOP formalism has been successfully employed by many groups including Jain and Baluja (1992), Jiang and co-workers (2000) and Lee and co-workers (2002), to name a few to determine the electron impact total elastic and inelastic cross-section for diatomic and polyatomic molecules from intermediate to high energies.

To find out the total cross section for a large number of atomic and molecular targets, one can also use the Complex Scattering Potential-ionization contribution (CSP-ic) method (Joshiyura *et al.*, 2004). From this method, the total inelastic cross-section can be determined by combining the total ionization cross-section and total excitation cross-section (Limbachiya *et al.* 2015).

$$Q_{inel}(E_i) = \sum Q_{ion}(E_i) + Q_{exc}(E_i) \quad (7)$$

Moreover, by using the SCOP method along with CSP-ic method, one can determine the total inelastic cross-section by electron impact of various chemical molecules theoretically which agrees with the experimental data.

Vinodkumar *et al.* (2013) used the quantum mechanical theory for the calculation of total and ionization cross-section for DNA based compounds which are based on SCOP formalism and Schrödinger equation solved by the method of partial waves that gives the complex phase shifts. The phase shift is a very useful tool in the scattering theory and it often reduces the complexity of the calculation for the scattering process. These phase shifts are further used to evaluate the total elastic cross-section (Q_{el}) and inelastic cross-section (Q_{inel}) (VinodKumar *et al.* 2010)

$$Q_T(E_i) = Q_{el}(E_i) + Q_{inel}(E_i) \quad (8)$$

They calculated the ionization cross-section of Uracil ($C_4H_4N_2O_2$) which is in good agreement with available theoretical and experimental data. Therefore, by using this method, the ionization cross-section for other complex bio-molecules such as adenine ($C_5H_5N_5$), thymine ($C_5H_6N_2O_2$), guanine ($C_5H_5N_5O$), cytosine ($C_4H_5N_3O$) etc. as well as the exotic target or radicals, which are otherwise very difficult to be studied experimentally but can be calculated.

6. OTHER APPROACHES

There are many other formalisms like Complex Potential Method (CPM), Classical Trajectory Monte Carlo (CTMC) method, Classical Over Barrier (COB) method, Orientation-Averaged Molecular Orbital (OAMO) approximation method, Time-Dependent Close Coupling (TDCC) method etc. have been adopted for calculating cross-section, which are not including in this article. Further, there is lack of available precise inelastic cross-section data or limited data for the particular target as experimentally. Overall, there is 5% to 20% agreement in the calculated values from different formalisms. Khare *et al.* (1976) provided the formalism by combining the Mott and Bethe cross-section expressions, which describe ionization collisions having high and low impact parameters respectively. Later on, Khare *et al.* (1987) proposed the new expression for the cross-section as the modified Jain-Khare formalism. As

$$Q_i(E, W, \theta) = \frac{a_0^2 R^2}{E} \left[\int_{k \rightarrow 0}^{E-I_i} \left\{ \frac{E-W}{E-I_i} \frac{1}{W} df_i(W, K, \theta) \times \ln[1 + C_i(E-I_i)] + \frac{E-I_i}{E(\varepsilon_0^3 + \varepsilon^3)} \times S_i \left(\varepsilon - \frac{\varepsilon^2}{E-\varepsilon} + \frac{\varepsilon^2}{(E-\varepsilon)^2} \right) \right\} 2\pi \sin \theta d\theta \right] \quad (9)$$

Here,

$W (= \varepsilon + I_i)$ is defined as energy loss suffered by the incident electron.

I_i = the ionization threshold for the production of i^{th} type of ion,

a_0 = the Bohr radius,

ε_0 = energy parameter,

C_i = collision parameter,

S_i = number of ionizable electrons,

R = Rydberg constant and

θ = the scattering angle.

From this equation, we find out the partial double-differential cross-section (PDDCS) for the production of an i^{th} type of ion by the impact of an electron of energy E with a molecule leaving secondary electron(s) of energy ε .

By summation of Partial Double Differential Cross- Section over i results in the total PDDCS:

$$Q'_i(E, W, \theta) = \sum_i Q_i(E, W, \theta) \quad (10)$$

From Lassetre's Theorem (Lassetre, Ausma, and Dillon, 1969), we can find out the linear optical oscillator strengths $df_i(W, 0)/dW$, i.e.

$$df_i(W, K, \theta) \rightarrow (1/4\pi)[1 + \beta P_2(\cos \theta)] \times df_i(W, 0)/dW, \quad (11)$$

Here

β = asymmetric parameter

$$P_2(\cos \theta) = \frac{1}{2}(3 \cos^2 \theta - 1) = 2^{\text{nd}} \text{ order Legendre polynomial}$$

Now, the oscillator strength is directly proportional to the photoionization cross section. Further integration of Eq. (9) with respect to the scattering angle θ gives the partial double-differential cross-section. Hence

$$Q_i(E, W) = \int Q_i(E, W, \theta) d\Omega, \quad (12)$$

Here $\theta = 0 \rightarrow 2\pi$ and $d\Omega = 2\pi \sin \theta d\theta$.

7. APPLICATION OF THE SCATTERING IONIZATION CROSS-SECTION

The collision cross-section is in a great interest in the investigation of various areas like plasma processing, astrophysics, astrochemistry, radiation physics, radiation chemistry and plasma chemistry (VinodKumar *et al.* 2009). The other applications of the cross-section are in the field of mass spectrometry, semiconductors, microelectronics, pollution remediation, atmospheric sciences (Antony, Joshipura, and Mason 2004). The high energy collision particles in the space play an important role in cloud formation, climate change, an important role in ozone depletion and global warming, to name a few.

A well-organized database on the electron impact collision cross-section is thus desirable due to its widespread applications which are not limited to physics but extended to life sciences also such as, initiating single and double strand breaks in DNA as radiation therapy (Glass and Varma, 1991). Radiation therapy is one of the major cancer treatment techniques in addition to chemical and surgical therapies. During this therapy, electrons are produced in a wide range of energies from the irradiated areas; secondary electrons collide with DNA molecules in human cells and eradicate the cancer cells. For such processes, we must have the accurate knowledge of electron collision cross-section for relevant biological molecules (VinodKumar *et al.* 2006). It is also applicable in the chemistry of molecules such as ionization of chemical molecules, clusters, fullerenes, polycyclic aromatic hydrocarbons, nanoparticles, etc. (Agnihotri *et al.*, 2012). Furthermore, the total cross-section plays an important role in understanding and modeling of the electrical discharge phenomenon since they determine the mobility and electron-ion recombination reactions. These characteristics are implemented in many devices as Electrical discharges devices, semiconductor etching and microcircuit fabrication devices, air purification devices etc.

8. CONCLUSIONS

The empirical formulation presented in this review gives the representation of the total ionization cross-section of atoms or molecules from the ground state by electron impact. Almost all the experimental results can be calculated by these formulas within 5-10% experimental error from threshold energy to intermediate energy. For higher energy cross-section calculations, other modified formalisms are implemented.

Although lot of papers have been published for calculating total and differential ionization cross-section for a number of atoms/molecules/ions using semi-empirical approach and other modified formalisms. However, experiments have their own difficulties and theories have their own limitations. So, a composite theoretical approach to cover large energy range is highly desired.

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