Chapter 1

FUNDAMENTALS AND SCOPE OF ELECTRON SCATTERING

We have addressed the fundamental phenomenology of scattering theory, a variety of cross sections expressing the likelihood of occurrence of various phenomena, and their relevance to many domains of current science and industry. We also evaluated prior research on electron-induced processes on atomic/molecular species conducted by various experimental and theoretical groups.

1.1 Introduction

The study of electron scattering (or collisions) with an atom/molecule target dates back more than a century. The scattering process is an important tool for determining the ultimate elements of matter and understanding the behaviour of various types of matter. The examination of negative slope sections that emerged in the voltage-current properties of a mercury vapour carrying cell in the experiment of Franck and Hertz (1914) provided strong direct proof for the quantization of atomic energy levels. In addition, this was the very first investigation into inelastic thresholds in electron-atom scattering phenomena. Several important features of plasma, environmental, biological, pharmacological, and industrial processes result from this kind of scattering of electrons by atoms, molecules, and radicals.

The phenomena of electron interaction have relevance in the field of plasma chemistry and other applications. "Low-temperature plasmas (LTPs) are particularly important in a variety of areas such as semiconductor fabrication, the destruction of volatile organic molecules, modifying the mechanical characteristics of surfaces, and nuclear fusion reactor wall chemistry" [1]. In flame sampling and Knudsen-cell thermos-chemistry, absolute ionization cross sections are also necessary to calculate quantitative gas densities from mass spectrometric observations [1]. In sectors like plasma-assisted microstructure etching and high-quality thin-film deposition, diverse electron-molecule scattering cross sections like excitation, ionization, dissociation, and so on have become crucially important [2].

1.2 Electron: An Excellent Probe For Scattering Processes

With a negative electric charge and no interior structure, electrons are subatomic fundamental particles. In states of matter, free electrons with a variety of energies are readily produced. The most frequent and well-known occurrence is the collision of an electron with other particles and molecules. As a result, the best and most capable probe for examining the structure and physical characteristics of matter is an electron. Due to their low mass, electrons require a smaller potential difference than other charged particles in order to accelerate to the necessary energy level. Electrons can simply be propelled to the energy value range under investigation. Consequently, electrons are the easiest to manage and control, making them ideal as a projectile source.

1.3 Electron Collision Processes

A multitude of kinetic events occur when a free projectile particle, such as the electron, interacts with a target (atom/molecule) during scattering phenomena. These events could then be separated into two mechanisms:(1) Elastic Scattering; (2) Inelastic Scattering. The total kinetic energy of the electron – molecule system is conserved in the elastic scattering. In inelastic scattering, a significant portion of the kinetic energy of the incoming electron is lost in various electron driven molecular processes.

1.3.1 Fundamental scattering processes

Figure 1.1 depicts a scattering process conceptually. A narrow stream of particles 'A' collides with a target 'B' (which might be an atom or a molecule) in a standard scattering experiment. .

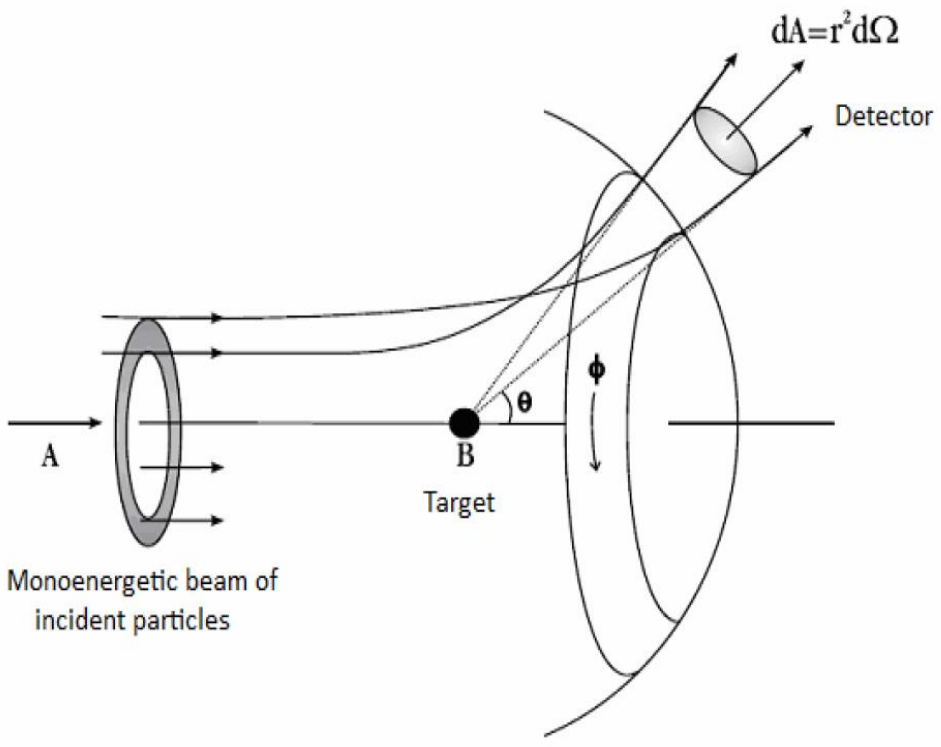

Figure 1.1 Schematic Illustration of Scattering mechanism [3,4]

The distance between the source of incident particles and the target are greater than de Broglie wavelength associated with incident particles. The experimental settings are so chosen that

each target scatterer acts independently [5]. A number of scattering processes that might occur are listed below.

Elastic Scattering

Total kinetic energy and total momentum are conserved in elastic scattering.

$A + B \rightarrow A + B$

Illustration:

$e^- + XY_3 \rightarrow e^- + XY_3$

Here 'XY_3' denotes any Molecule and 'e^-' denotes electron.

Inelastic Scattering

When two particles A and (or) B collide, inelastic scattering occurs, and the internal quantum state of one or both particles changes. Although total kinetic energy is not conserved in this operation, momentum is.

$A + B \rightarrow A^* + B$

$A + B \rightarrow A + B^*$

$A + B \rightarrow A^* + B^*$

Examples:

$e^- + XY_3 \rightarrow e^- + XY_3^*$ [Electronic excitation]

$e^- + XY_3(j) \rightarrow e^- + XY_3(j')$ [Rotational excitation]

$e^- + XY_3(v) \rightarrow e^- + XY_3(v')$ [Vibrational excitation]

$e^- + XY \rightarrow 2e^- + XY_3^+$ [Parent ionization]

$e^- + XY_3 \rightarrow 2e^- + XY_2^+ + Y$ [Dissociative ionization]

$e^- + XY_3 \rightarrow 2e^- + Y^+ + XY_2$ [Dissociative ionization]

$e^- + XY_3 \rightarrow 2e^- + X^+ + Y_3$ [Dissociative ionization]

$e^- + XY_3 \rightarrow 2e^- + Y_3^+ + X$ [Dissociative ionization]

$e^- + XY_3 \rightarrow 3e^- + X^{++} + Y_3$ [Dissociative ionization]

$e^- + XY_3 \rightarrow XY_3^-$ [Electron attachment]

$e^- + XY_3 \rightarrow XY^- + Y_2$ [Dissociative electron attachment]

$e^- + XY_3 \rightarrow e^- + XY + Y_2$ [Neutral dissociation]

1.3.2 Cross sections for scattering mechanism

The study of atomic and molecular physics pertains to numbers much smaller than humans typically observe. For instance, the length's order of significance is about equivalent to the Bohr radius ' a_o ' (radius of the H atom, $a_0 = 52.9 \times 10^{-10}$ m). The atomic unit (a.u.) system is used in the present work. In the a.u. system,

Velocity of light, $c \cong 137$;

Charge of electron, $e = 1$

Mass of electron, $m_e = 1$;

$\hbar = 1$ ($\hbar = \frac{h}{2\pi}$, where $h =$ Plank's constant); and $4\pi\varepsilon_0 = 1$ (where $\varepsilon_0 =$ free space's permittivity).

We also may determine the units in $a.u.$ such as:

Linear momentum, $p = \hbar k = k = \frac{2\pi}{\lambda}(a_0^{-1})$($k =$ wave vector and $\lambda =$ wave length);

Energy, $E = \frac{\hbar^2 k^2}{2m} = \frac{k^2}{2}(a_0^{-2})$

$1\ \text{a.u. of energy (Hartree)} = 27.2114\text{eV}$; and

$1\ \text{a.u. of energy (Rydberg)} = \frac{1}{2}\ \text{Hartree} = 13.6057\text{eV}$;

Moreover, CSs are described in a.u. as:

Differential cross section, DCS in $\frac{a_0^2}{Sr}$ (Sr = Steradian);

TCS and MTCS in a_0^2 or $Å^2$.

1.4 Quantification Of Processeses

Theoretical calculations and scattering experiment findings are typically expressed in terms of distinctive values referred to as "cross sections." To determine the likelihood that a given event will occur, cross-sections are used. Several cross sectional types are covered in this section.

1.4.1 Differential cross sections

We now turn our attention to the elastic process, in which a dN(number of particles) are elastically dispersed in a solid angle $d\Omega$ per unit of time. For appropriately thinner targets, the number of particles distributed per unit time per unit solid angle correlates to the incoming flux (Φ_A), and the number of target scatterers (n_B) may be expressed as,

$$dN \propto \Phi_A n_B d\Omega \tag{1.1}$$

$$dN = \frac{d\sigma}{d\Omega}(\theta, \phi)\Phi_A n_B d\Omega \tag{1.2}$$

DCS, or differential cross section, for elastic scattering is defined as the proportionality factor $\frac{d\sigma}{d\Omega}(\Omega)(\Omega = \theta, \phi)$. The amount of particles released into a solid angle defined by the polar angles in figure 1.1 is quantified by DCS. Hence, from equation 1.1, we obtain

$$\frac{d\sigma}{d\Omega}(\Omega) = \frac{dN}{\Phi_A n_B d\Omega} \tag{1.3}$$

We can write from Joachain [3],

$$\frac{d\sigma}{d\Omega} = |f(\Omega)|^2 \tag{1.4}$$

Where, $f(\Omega)$ is the scattering amplitude.

1.4.2 Momentum transfer cross section

The momentum transfer cross section (MTCS) is defined by,

$$Q^M = \int f(1 - cos\theta)\frac{d\sigma}{d\Omega}d\Omega \tag{1.5}$$

The momentum transfer cross section is obtained using the differential cross sections.

1.4.3 Total elastic and inelastic cross sections

By integrating the differential cross section across each solid angle$(d\Omega)$, one may derive the total or integral elastic cross section.

$$\sigma_{el} = \int \frac{d\sigma}{d\Omega}(\Omega)d\Omega \tag{1.6}$$

This can also be written as,

$$\sigma_{el} = \int_0^{\pi}\int_0^{2\pi}\frac{d\sigma}{d\Omega}(\theta,\phi)\sin\theta \cdot d\theta \cdot d\phi \tag{1.7}$$

In the absence of any inelastic process the σ_{el} is purely elastic. However, a scattering event may involve a number of different inelastic processes in addition to the elastic processes.

1.4.4 Total ionization cross sections

The losses of incident particles in the outgoing channel is caused by the overall inelastic cross section. The total of all discrete electronic excitations and ionization are described by the total inelastic cross sections denoted by Q_{inel}. Each of these mechanisms can be given as follows:

$$Q_{inel} = \Sigma Q_{exc} + Q_{ion} \tag{1.8}$$

The second term in equation (1.8) represents the total cross sections (TCS) for all acceptable ionization processes.

1.4.5 Total excitation cross sections

The first component ΣQ_{exc} in equation (1.8) denotes the sum of all excitation cross sections, which includes all attainable electronic excitation channels. These channels show the discontinuous occurrence of all electron transitions that are electrically allowed.

1.4.6 Total (complete) cross sections

The total Q_T obtained by combining total Q_{el} and Q_{inel}. This relates to the influence of all possible scattering channels in every scattering process. The Q_T represents the likelihood of contact among an incident electron and a target particle. Thus,

$$Q_T = Q_{el} + Q_{inel}$$

1.4.7 Rotational cross sections

The Q_T are approximated using spherical interactions. The spherical potential was utilized to employ the partial wave approach to figure out the Schrödinger problem. Because atoms do not have a permanent dipole moment, this technique works well for them; but, when it comes to molecules, Non-spherical potential and concomitant cross-sections must also be considered, especially for those having a permanent dipole moment and/or quadrupole moment. Rotational cross sections become relevant mostly at low energies, as represented by Q_{rot} in our study. We have not examined Q_{rot}.

1.4.8 Grand total cross sections

The sum of total cross sections arising from spherical and non-spherical collisions, as given by ΣQ_{TOT}, is the grand total cross section. The non-spherical component of the grand TCS [6] is represented by Q_{rot}, and the spherical part is represented by Q_T.

$$\Sigma Q_{TOT} = Q_T + Q_{rot}$$

1.5 Electron Collision Studies

Different cross sections are extracted using various techniques both experimental and theoretical. Several constraints have hampered the investigative examination of particular objectives, including the cost of apparatus, the dependability of the outcomes, and the time necessary to produce the output results. In experimental investigation, the reactivity as well as the possible toxicity and the smaller life span of the targets can pose various challenges. In such situations, theoretical approaches are necessary to investigate electron – molecule scattering.

1.5.1 Experimental investigations

The following are often included in the basic experimental setup for determining cross sections:

- An electron gun equipped with the appropriate technology to deliver a narrow beam of the requisite energy range as a projectile source.
- An automated arrangement to generate the target species in the gaseous form and measuring its number density.
- Transfer of the gaseous molecules into the collision chamber and energetic beam of electrons to execute the scattering phenomenon.
- A detection system is constructed to monitor the electrons that are scattered following scattering events and to investigate the products in terms of angular distribution and loss of energy.

According to a review of the literature [5,7,8], there are several types of electron collision investigations as follows:

(i) Electron beam experiments
(ii) Electron swarm experiments and

(iii) An appropriate combination of both

The following cross sections are typically determined via swarm investigations:

(i) Inelastic cross sections,
(ii) MTCS (Momentum transfer cross sections) and
(iii) EACS (Electron attachment cross sections)

Electron swarm experiments are suitable for dispersion at low energies of 1eV or fewer, although electron beam studies are advantageous from 1eV to exceptionally large energy values. Several groups all throughout the world have conducted out scattering tests.

1.5.2 Theoretical studies

Apart from so many experimental groups, numerous theoretical teams are focusing on electron interactions with atoms/molecules.

Because of the difficulties found in trials, cross section computations employed a complicated optical potential known as SCOP (Spherical Complex Optical Potential) [9]. The SCOP has been utilized effectively by several groups [9–12]. For computing single electron collision ionization cross sections for compounds, there are two helpful quasi methods: (1) Kim and Rudd's Binary-encounter-Bethe (BEB) technique [13,14]. (2) Formalism of Deutsch-Märk (DM) [15,16]. Both approaches were established for determining atom ionization cross sections until being employed to examine neutral molecule compounds. After that, both groups modify their ideas for investigating molecular ions.

The Bethe cross section and the Mott cross section are combined in the BEB (Binary-Encounter-Bethe) theory, that exhibits high incident energy characteristics. Here, the BEB concept is a condensed form of BED (Binary-Encounter-Dipole). Mott cross section formalism [17] describes the collision between two "free" electrons.

The Bethe cross section formula [18] accounts for the dipole interactions that includes rapid incident electrons in soft impact processes. In the BED concept, a straightforward equation for the OOS (Optical Oscillator Strength) is utilized in accordance with the findings of the hydrogen atom, helium atom, and hydrogen molecule. The BEB and BED theories both depend on variables that were discovered either through experimentation or by studying ion wave functions and objective.

Calculations based on either of the two models and the experimental findings are in excellent accordance for incident energies values few keV over the threshold energy value. Calculations based on the BED model, which outperform the BEB concept most of the time, indicate steady variation of 5% to 15% at the maximum from experiment. Bethe cross section was employed in the BED system, which was created by Khare et al. to represent long-range dipole interactions [19,20].

Deutsch and Märk [21] established the DM approach for computing atomic ionization cross sections, which was subsequently refined and enlarged several occasions. The atomic Q_{ion} is defined by the DM approach as the total of all partial Q_{ion} connected to the discharge of just one electron from a certain atomic subshell. [15]. While doing the Mulliken or perhaps another molecular orbital population research, that depicts molecular orbitals compared to component atoms' orbitals, the DM approach can preferable be adapted to molecular ionization cross section estimates. BEB and DM formalisations each come with a set of disadvantages.

The CSP − ic (Complex Scattering Potential − ionization contribution) technique for estimating TICS (Total Ionization Cross Section) using total inelastic cross sections was developed by Joshipura and colleagues [22]. Vinodkumar et al [23] created the Enhanced CSP-ic approach.

1.6 Areas Of Electron Scattering Applications

The investigation of electron collisions with objects has a broad spectrum of uses in Astrophysics, Plasma, Industries, and Atmospheric and Biological sciences, as mentioned below:

1.6.1 Astrophysics

On titan, the largest moon of the Saturn, UV radiation, cosmic rays and electron bombardment of the Saturn's magnetosphere cause dissociation of N_2 and CH_4, leading to complex organic chemistry at higher altitudes, which results into solid organic aerosols responsible for Titan's brownish colour [24]. Cosmic rays, UV light, and electrons hitting Saturn's aurora all help this process along. These are the most advanced alien organic substances found in the Planetary System. Their chemical yield is generally described as beginning with the photolytic induction of the precursor chemicals for nitrogen and methane, followed by the formation of hydrocarbon and nitrile monomers in the gaseous phase, followed by polymerization procedures which

result in solid materials large enough to condense, aggregate, and fall to the ground. Many studies have been carried out to replicate and investigate such a complex environment in a lab environment [25].

While examining the polymeric chemical composition of the aerosols, the plasma device PAMPRE offered crucial cues [25]. By creating lab equivalents of these aerosols, the PAMPRE research aims to mimic the atmospheric composition of Titan, especially the chemical interaction that produces aerosols. A spectrometer on board the Cassini probe has shown that tholins are generated in Titan's atmosphere at elevations more than 1000 km. Many heavy compounds, ions, as well as other unique scenarios of Titan's chemistry have been discovered by the sensors on board the current Cassini/Huygens mission (NASA/ESA) [26].

1.6.2 Atmospheric science

According to the discipline of atmospheric physics, high-energy particles released by the Sun and cosmic radiation are what generate atmospheric ionization in contrast to photo-driven phenomena in the ionosphere. These particles may lead to cloud formation and climate change, according to research [27,28], and a connection between ozone depletion and cosmic rays is also anticipated [29]. So, it is important to study the electron interaction, ionization and total cross sections (TCS) with atmospheric molecules. Calculations related to general atmospheric constituents like O_2, N_2 and H_2O are usually well developed, but results of ionization cross sections of trace species (OClO, Cl_2O, N_2O_5) are generally unavailable [29]. The presence of excitation and fluorescence caused by electron collisions in the solar system's atmosphere species is demonstrated by the presence of auroras in the Arctic and Antarctic, electro glow in the ionospheres of the large planets, UV radiation released by the plasma Torus, and UV emissions from Titan and Triton (a satellite of Neptune).

Figure 1.2 electron collision-induced excitation and fluorescence of auroras

(Image Courtesy: Images.google.com)

1.6.3 Plasma Physics

Atomic and molecular ionization caused by electron impacts is a key process in a plasma system. The following are some areas of applications based on electron assisted ionization cross sections of compounds.

- Plasmas for low-temperature processes
- Plasma edge fusion
- Gas discharges
- Radiation chemistry
- Mass spectroscopy
- Chemical analysis

Low-temperature plasmas (LTP) signify a distinctive state of matter which is composed of neutral molecules, atoms, radicals, excited states, ions and electrons [30]. The characteristic electron energies of low temperature plasmas (LTP) range from intermediate to higher energies that can produce radicals, charged molecules and excited states [30].

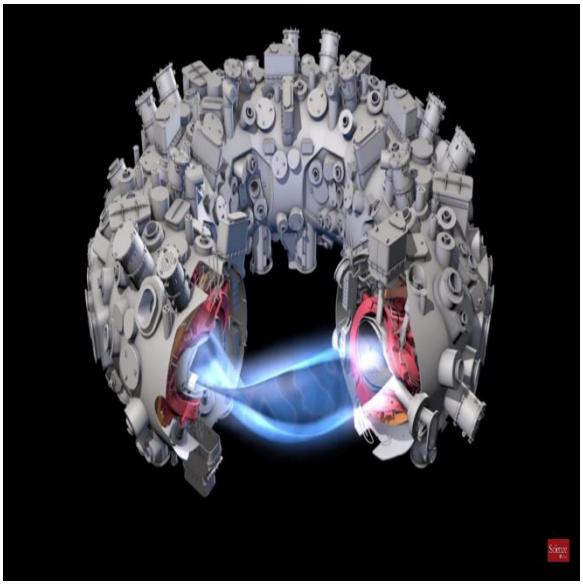

Figure 1.3 Plasma fusion reactor

(Image Courtesy: Images.google.com)

The plasma interface's space charge sheaths accelerates and produce ions fluxes with varying energies between a few eV and hundreds of eV when the pressure is low [30]. Through sputtering, etching, activating, and depositing materials for microelectronic devices and surgical devices, ion fluxes can modify substrates. The microwave, arc, and inductively linked plasma discharges, which operate almost at thermodynamic equilibrium, provide the basis for the industrial applications of plasma [30]. The temperature of electrons (Te) is higher than the temperatures of heavier particles and gases [31,32]. Even the most thermal materials can be effectively and non-destructively exposed to highly reactive plasma species using LTP emitters. The whole microelectronics industry, which is the technological foundation of contemporary civilization, is made possible, by the advantageous plasma surface interactions that collect and erase nanoscale precision components in the manufacturing of microprocessors [33]. Plasma treatment was created as a result of this beneficial interaction with surfaces that also included liquids, organic tissues, and wounds [34]. LTPs can also interact positively and productively with interior plasma interfaces, such as in a dusty plasma that is packed with particles or aerosols [35].

1.6.4 Biological viewpoint

Secondary electrons with energies that vary from 0 to 20 eV produce single-strand and double-strand DNA breaks, giving researchers a clearer understanding of how ionizing radiation damages live tissues and cells. [36]. Because of this, there is a great deal of curiosity in the mechanisms involved in scattering electrons off biological molecules. A number of theoretical and experimental initiatives aimed at enhancing our knowledge of the processes behind radiation-induced DNA damage, including direct phenomena like electronic, ionization, etc. as well as complex mechanisms like resonances (dissociation and dissociative electron attachment) [37].

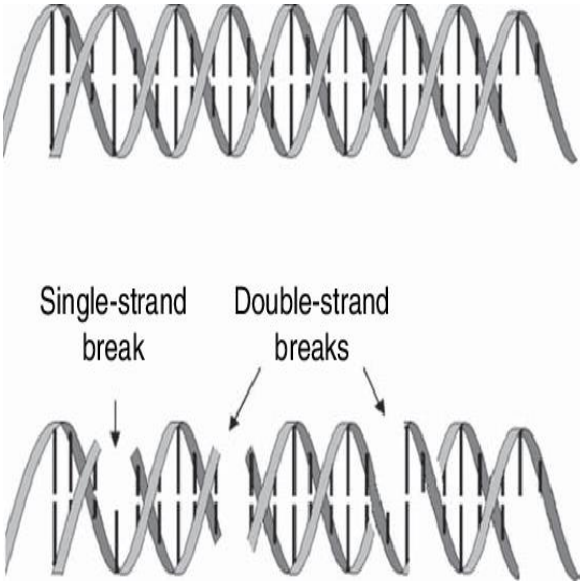

Figure 1.4 Single strand and double strand break of DNA

(Image Courtesy: Images.google.com)

Tetrahydrofuran (THF) is the most basic furanose ring type, connecting the DNA foundation's phosphate groups. It is the most basic molecular counterpart to the deoxyribose unit. A succession of THF compounds coupled to phosphate units and DNA strands can be used to represent such an essential structure [38].

Numerous bio-compounds have components that are heterocyclic atoms or their equivalents. One oxygen atom and four carbon atoms make up the five-membered ring of the heterocyclic organic molecule furan (C_4H_4O). Furan can act as a simple replica of a more complex molecule that is physiologically important [39].

1.6.5 Industrial relevance

The following are just a few of the various uses of electrical discharges:

- Air purification cation devices
- Ion implantation devices
- Chemical vapour deposition on thin films
- Semiconductor etching
- Microcircuit fabrication devices

Total electron impact cross sections play a crucial role in comprehension and modelling electrical discharge processes as they influence electron-ion recombine and reaction velocity. In the semiconductor production industry nowadays, utilizing plasma cutting to create nanometric designs is a challenging operation. Radicals are prominent constituents in plasma media having many applications in semiconductor applications. For successful modelling of these technological plasmas, a large record of ionization, excitation, dissociation, and elastic cross sections for each species in the plasma is required.

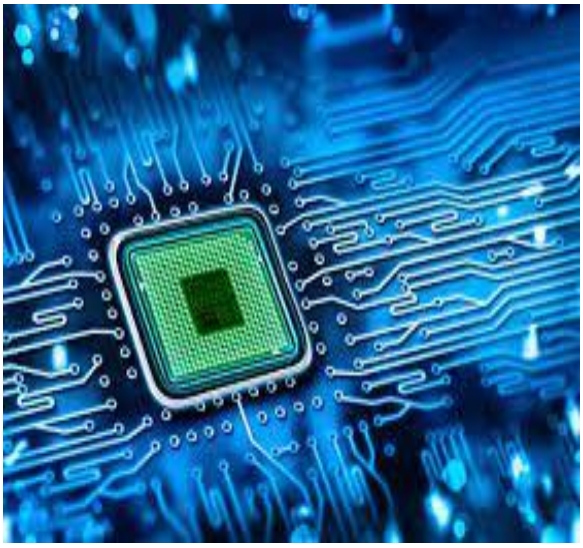

Figure 1.5 Semiconductor etching process (Image Courtesy: Images.google.com)

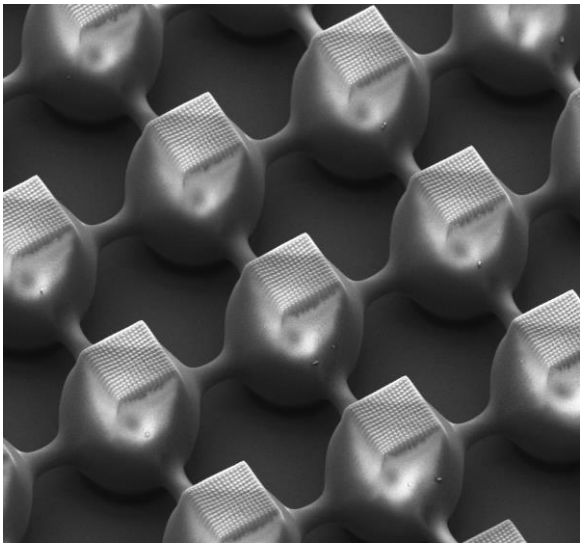

Figure 1.6 Nanofabrication (Image Courtesy: Images.google.com)

The investigation of electron-molecule interactions has created the groundwork for new and exciting technologies such as nanofabrication. Low energy electron driven processes have the potential to generate chemical nanoparticles on surfaces relevant to biochips based on protein immobilization or nano particle shaped materials [40]. The dissociative electron attachment of

a selected bond produces efficient chemical reaction kinetics and serves as the foundation for chemical lithography [41].

1.7 Contemporary relevance of present study

Using the spherical complex optical potential (SCOP) formalism [9] and the Complex Scattering Potential-ionization contribution (CSP-ic) technique [11], our team has previously studied electron scattering processes from gaseous species. Our ability to study electron interaction with aqueous or condensed phase target systems is thanks to a recent update to our SCOP computations.

Many of the phenomena that are associated with electron scattering from solid materials are of considerable relevance and can be applied in a range of different subfields that belong under the umbrella of the scientific discipline known as physics. In addition to the diffraction of neutrons and of x-rays, the investigation of the construction of solid substances can benefit from the addition of electron diffraction as a complementary technique. The electron microscope is proving to be a useful device in many different fields of study. It employs the electron interacting with the molecular constituents of the material. In recent years, technological advancements have made it possible to use photographic plates as a medium for researching the properties of high-energy particles such as electrons. In order for these investigations to be successful, it is essential for the researchers to possess precise data regarding the frequency of multiple scattering and the rate at which electrons lose their energy in the solid material of the plate. Determining the range of electrons present in a particular solid material has traditionally been necessary in order to accomplish the task of measuring the onset energy of electrons. In every device in which electrons are made to strike a solid, it is vitally crucial to take into account the emission of secondary electrons. The electrical resistance of semi-conductors and conductors, which is defined by the possibilities of conduction electron scattering across the substance, is another property that has important practical relevance. This property is governed by the possibility of conduction electron scattering

The current theoretical investigation uses a calculation method which is fast and yields relatively accurate cross sections of various atoms, molecules, and radicals on a wide range of energy. Because variety of information needed in the semiconductor industry as well as other sectors is tremendous, our contributions have substantial relevance in today's research and development. More precise theories like R – matrix formalism are constrain by the target size

and energy range parameters. For small compounds, the exact R- matrix approach might be beneficial for calculating cross section and estimating resonances. Nevertheless, the energy range wherein they work is quite restricted (10 eV). This approach is similarly ineffective for calculating the cross section of big compounds. For calculating molecules of any size, the R-matrix approach requires a rather lengthy time.

The entire elastic and inelastic cross sections are computed using the well-known SCOP methodology. The total Q_{ion} is then calculated using a semi-empirical method. Our formalism for determining the Q_{ion} could have certain flaws. Nevertheless, because to its great dependability and rapidity, CSP-ic concept takes the lead among the various approaches currently used to determine the total Q_{ion} [42].

The major goal of this work is to undertake theoretical study on the interaction of electrons with various molecules and radicals, which are important species in many applied domains, such as medical sciences, industrial technologies, plasma sciences, and so on.

❖ **Main Objectives of the thesis**

- To investigate the collision of electrons over a wide range of energies, from IP up to 5 keV, using a combination of SCOP and CSP-ic approaches.
- To expand the scope of our calculations so that they also take into account molecules found in the aqueous phase.
- To provide a new technique for determining the Q_T of larger, more complicated molecules for energy spectra ranging from 50 eV to 5 keV.
- To analyze the numerous connections between the cross-sections and the target properties, such as the molecular polarisability and the dielectric constant, which will lead to the prediction of those quantities for which the data is not currently accessible.

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