

Chapter 1

Introduction of electron collision processes

This chapter describes the fundamentals and significance of the study of electron collision mechanism with the target systems. Various collision processes occurring during the electron scattering with the target has been also introduced in this chapter. Further, the importance of the study of electron collision with the condensed or solid phase target compounds has also been discussed here.

1.1 Introduction

The understanding of the fundamentals of science and how they are used in everyday life is becoming increasingly important as a result of the high pace of technological advancement that has occurred over the past few decades. Most of the scientific and engineering fields depend heavily on physics, which is the foundation of the sciences. It is not an exaggeration to say that knowledge of the physics is essential for any scientist/researcher who wants to make headway in their chosen subject/field. Among all the fields of physics and in the scientific curriculum, atomic and molecular physics takes centre stage. This field of physics was at the forefront of scientific research at the turn of the twentieth century.

The study of atomic and molecular collisions has seen a significant increase in recent years, both in terms of experimental effort and theoretical speculation. The exploration of collision phenomena is one of the most important aspects of research into the microscopic structure of matter. Experiments that involve scattering event, provide the foundation for the vast majority of our understanding of the interactions and forces that are generated when incident charged particles collides with the target molecular/atomic structures.

The necessity of having in-depth knowledge of scattering from target species infiltrates the space sciences, condensed physics, plasma sciences, astrophysics, laser sciences, accelerator

technology, and fusion research etc. The expansion of our understanding of star evolution relies heavily on the knowledge gleaned from the various collision mechanisms. The composition of the atmosphere, starting from lower layer to upper layer, is governed by the processes as a result of scattering phenomena between the charged particle and molecular systems. Such investigations aid in comprehending the realities underlying the thinning of ozone layer caused by a chain of collisions of atomic and molecular systems. It is crucial to examine the collision of electron and molecules in order to ascertain the transport and charge-balance characteristics of electrons in the low-temperature plasmas. When estimating the rate coefficients of the many distinct transitions that are engaged in the various plasmas, it is of the utmost significance to have accurate cross-section data of the electron/positron collisions with target systems.

Deep research into the dynamics of scattering event is thus necessary given the potential applications of atomic or molecular scattering data in numerous scientific and technological sectors.

1.2 Electron collisions

Current study focuses on the electron collisions with various applied molecules, which find the significance in variety of applied and research fields.

1.2.1 Electron: A superior probe for collision mechanisms

Since an electron is a fundamental particle that permeates the entire cosmos, it would make an excellent probe for studying the matter in its various forms. Collisions between the electrons and the target species are the main cause of many natural occurrences. Enthralling phenomena like these are not exclusive to Earth; they occur on all the planets and even in the remote reaches of the galaxies.

The advent of quantum mechanics in the early 20th century marked the beginning of the study of electron collision with atomic and molecular systems. The Frank-Hertz experiment, which was conducted in 1914 by James Franck and Gustav Hertz, was the first electron scattering experiment. Discrete excited states in Hg-atom (mercury) were first detected by this Frank-Hertz experiment. Another exceptional experiment was that of conducted by Ramsauer [1].

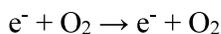
At the course of this experiment, a noteworthy feature known as the "Ramsauer-Townsend minimum" was discovered, which is the characteristic phenomenon of low energy electrons scattering. Resonances are another intriguing aspect of low energy collisions that has been noticed [2]. These experimental ventures then successfully fostered an interest in scattering physics. Following that, several experiments were carried out for investigating the scattering of electrons by a variety of target species [3–6]. The ability of the electron to be diffracted by crystals was found in 1927 by George Paget Thomson; and Davisson and Germer, providing experimental support for the wave-particle duality of matter. Over the time, with the advent of new technologies like electron microscopy, and electron diffraction, the scattering experiments advanced significantly. These have enabled the researchers for more in-depth research on molecular structure and characteristics.

1.2.2 Electron induced molecular processes

When a projectile collides with an object, the collision between them can be understood as a simple "binary encounter". There are multiple possible pathways, mainly categorised in elastic and inelastic events, which occur when a collimated beam of any ionized or charged particle interacts with a target system.

Elastic collision processes

It is the process by which a beam of projectile deflected from an object in such a way that no energy is transferred from the projectile to the object. The internal structure of the target system remains unchanged in elastic processes. The momentum and kinetic energy are conserved. Following is the example of elastic electron scattering of oxygen molecule.



Inelastic collision processes

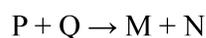
Inelastic scattering occurs when a portion of the incoming particle energy is transmitted to the target system's internal energy leading to the ionizations or excitations of the target atom or molecule. Following are few illustrations of various inelastic processes induced by electrons,



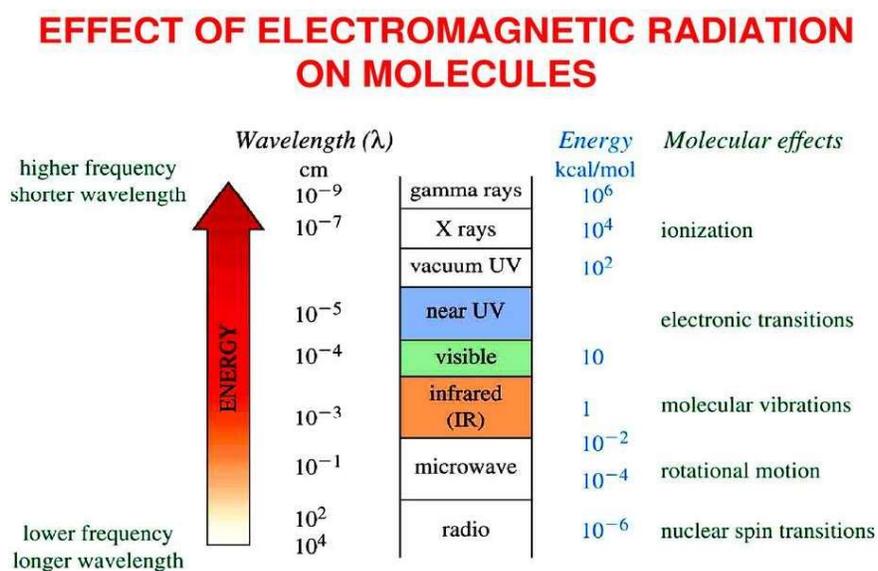


Reactions

The process by which the composite system (P + Q) separates into two particles M and N, which are distinct from the target particles (P and Q), or many particles, is referred to as a reaction.



Or



Graphics source: Wade, Jr., L.G. *Organic Chemistry*, 5th ed. Pearson Education Inc., 2003

Figure 1.1 Effect of Electromagnetic waves on molecular systems

Credit: Wade, Jr., L.G. *Organic Chemistry*, 5th edition Pearson Education Inc., 2003

Figure 1.1 provides a graphical illustration of the aforementioned processes. Any incident energy will result in the possibility of elastic scattering. Energy of the order of meV is needed for excitation of rotational energy levels, while vibrational levels excite at energies, around 0-1 eV. For scattering dynamics below the threshold for ionisations to occur, the excitation of electronic energy levels is the predominant inelastic channel, while above it, ionisation takes the control. A complete picture of the scattering mechanism can be obtained by including all of these inelastic as well as elastic events.

1.3 Previous studies on electron collisions

For the electron collision study, specifically for the determination of the various cross sections, a variety of experimental and theoretical approaches are utilised. The observational research of certain target systems has been hampered by barriers including the expense of the necessary instrumentation, the dependability of the findings, and the rate at which they are produced. Data that cannot be determined by experiment (such radicals and chemicals that are difficult to prepare into gaseous phase, as well as biomolecules) necessitate the use of theoretical approaches. The inherent physics of electron scattering could not be explained by classical conceptions, thus during 1960s scientists began building the theoretical methodologies for the scattering of electrons from target atomic/molecular species based on quantum mechanical concepts. Let's have a look at a few different experimental and theoretical scattering strategies.

1.3.1 Theoretical methods

Mott and Massey [7] are often regarded as the forefathers of the theoretical scattering physics. In addition, numerous theories, the most employed ab-initio techniques are the Schwinger multichannel (SMC), R-matrix, and Kohn variational approach, have been devised. Since an endless number of pathways are potentially available for electron-target collisions with impact energies over the ionisation threshold of the target, they cannot all be explicitly encompassed in the expansion of total wave function. In order for tackling the collisions at this energy, a number of methods, including, pseudostate methods, optical potentials, and Born approximation extensions to lower energies by incorporating higher-order terms in the born series, have also been developed.

To determine total cross sections in the energy regime beyond the target's ionisation threshold, the spherical complex optical potential (SCOP) method have been used by many groups including Jiang and team [8], Jain and Baluja [10,11], and Lee and his team [9]. In addition, most of the present work being done here is focused on computations using SCOP with certain modifications. The ab-initio R-matrix approach has also been used effectively by various groups, such as Tennyson and co-workers [12], and Baluja and co-workers [13]. We have also utilised the Quantemol-N program [14], which is based on the UK R-matrix code, to evaluate total cross sections for some of targets with kinetic energies below 15 eV.

In addition to it, specialised methods for the investigation of ionisations were established. Methods such as Deutsch-Mark (DM) formalism, Complex spherical potential ionisation contribution (CSP-ic) method, Binary-encounter-Bethe (BEB) method are some of the most common and widely used approaches for ionisation computations. A thorough explanation is available in Carsky and Curik [15]. With the development of these various quantum theoretical models, scientists were able to precisely compute scattering cross-sections and other molecular properties, offering a robust foundation for interpreting experimental results.

1.3.2 Experimental techniques

Measurements of scattering can be carried out using a variety of different approaches. The most popular method for measuring energy loss involves holding the scattering angle and the impact energy constant. In this mode, the scattering signal is detected as a function of the energy that was lost by the electron through the utilisation of techniques such as pulse counting and multi-channel scaling. An energy-loss spectrum is the end product of such experiment. The only feature that manifests at zero energy loss is the elastic scattering feature; all other characteristics relate to ionisation and different excitation processes.

The following pieces of equipment were utilised in the investigation of electron collisions with target system:

- (1) A source of single-energy (monoenergetic) electrons, such as an electron gun
- (2) A system for producing the gaseous target and determining the number density of it
- (3) A system that allows the target gas to be introduced into the collision chamber
- (4) An arrangement of detectors for tracking the electrons that are emitted upon collisions

Numerous texts, such as "Atomic and Molecular Radiation Physics" by Christophorou [16], Bransden and Joachain [18], and Massey and Burhop [17] provide literature on experimental procedures and results.

There are three main types of electron collision experiments:

- I. Electron beam experiment
- II. Electron swarm experiment
- III. Appropriate combination of Electron beam and swarm experiments

The swarm experiments [19] allowed for the measurement of three different types of cross sections: electron attachment, inelastic, and momentum transfer. Impact energies of 1 eV or less are well-suited to the electron swarm experiments. The energy range where they have made their most significant contributions is less than a few tens of an eV. In such experiment, a number of different factors coming together to make it simpler to carry out swarm measurements, while the opposite is true for beam experiments. From 1 eV to high energies, beam measurements look good.

Numerous research organizations from all around the world have reported their measurements on the total cross sections (Q_T) produced from the impact of projectile electrons. Experiments for determining the Q_T have been carried out in Japan by Kimura, Sueoka, and their colleagues [20,21] at low and intermediate energy levels. To determine the cross sections, they employed time-of-flight device. The Ramsauer type spectrometer has been utilised by Zecca and colleagues in Italy [22]. They measured the Q_T of a number of different compounds at energies ranging from 100 to 3000 eV. Experiments to determine Q_T in the region of intermediate to higher energy are carried out by the Mason and co-workers [24] along with Garcia group [23] independently at different places. Experiments conducted by Szmytkowski and his colleagues [25] at Poland, have reported Q_T at energies, ranging from 1 eV to 250 eV.

Apart from Q_T , Nishimura in Japan and Kim in the United States are doing experiments on electron impact ionisation cross sections (Q_{ion}) for energy ranging from target ionisation energy to 1 keV [26]. The Harland Group [27,28] work on a modified version of an external fourier transform mass spectrometer. Experiments on scattering of electrons and ionisation have been performed by Tarnovsky and co-workers in the United States as well as Deutsch in Germany [29] at energies ranging from the ionisation threshold to around 200 eV. Total and

partial Q_{ion} was measured by Stebbings and colleagues [30] in a study conducted in the United States. Mason and coworkers in the UK [31] have also determined Q_{ion} at moderate and high energies for molecules like O_3 . Indian researchers Subramanian and coworkers [32] have measured the ion impact Q_{ion} . The cross section of electron impacts on targets of interest have been measured by Tribedi and coworkers [33] and Jagatap and coworkers [34].

1.4 Applications of electron scattering study

The universe has always held a certain allure for humankind, making the investigation of its many unsolved puzzles a highly treasured endeavour. Electron collision mechanisms drive the entire Universe's evolution and are crucial to understand it, which is now entrenched fact.

1.4.1 Planetary sciences

The majority of the physicochemical phenomena within the cosmic plasmas are governed by charge transfer and collisional ionisation processes because of the low density of interstellar space. In addition, the ionospheres of all the planets, including Earth, serve as a fantastic natural laboratory for collisional processes between the electrons and various celestial species. Iconic missions like Rosetta and Cassini-Huygens have shed light on the significance of electron collisions in various astronomical objects. The Martian airglow [35], and aurorae, identical to those seen in Earth's atmosphere, also found on Saturn, Jupiter, Neptune, and Uranus [36,37] are caused by electron induced ionizations.



Figure 1.2 Earth aurora
Credit: NASA [38]

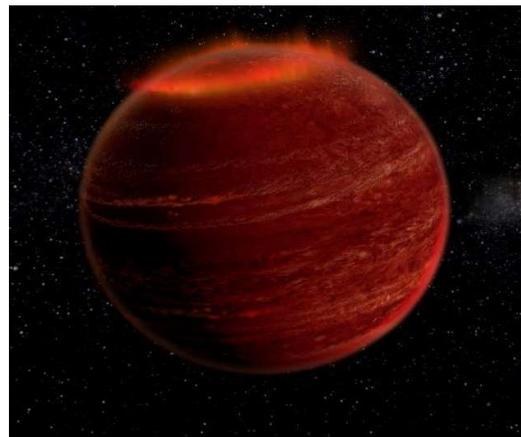


Figure 1.3 Aurora on brown dwarf
Credit: Chuck Carter and Gregg Hallinan/Caltech [39]

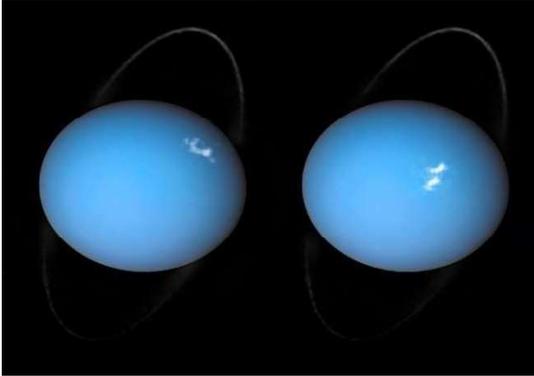


Figure 1.4 Aurora on Uranus
 Credit: ESA/Hubble & NASA, L. Lamy/Observatoire de Paris [40]



Figure 1.5 Aurora on Rogue planet
 Credit: Chuck Carter, Caltech, NRAO/AUI/NSF [41]

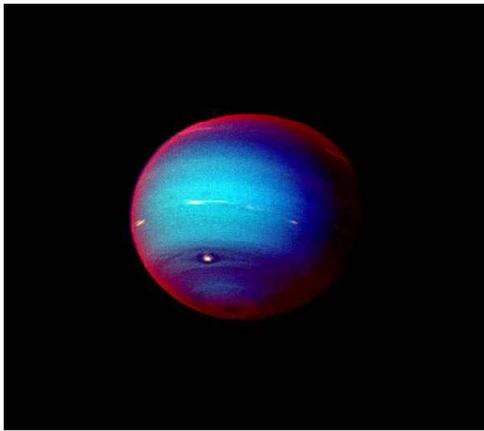


Figure 1.6 Aurora on Neptune
 Credit: NASA/JPL [42]



Figure 1.7 Aurora on Jupiter
 Credit: NASA, ESA, and J. Nichols (University of Leicester) [43]

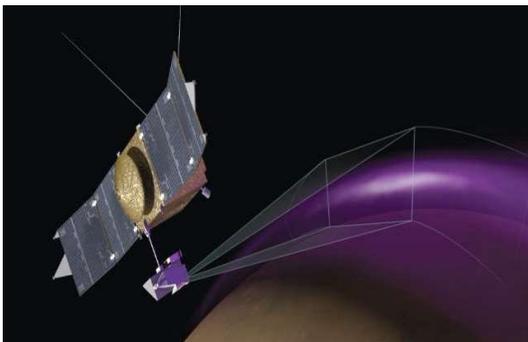


Figure 1.8 Aurora on Mars
 Credit: University of Colorado [44]

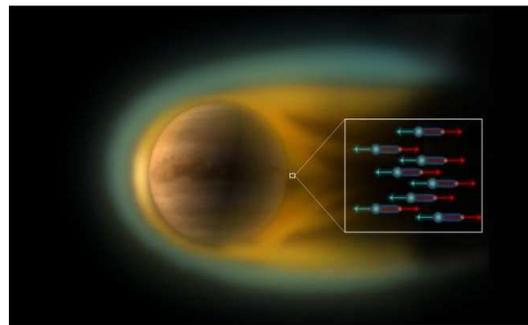


Figure 1.9 Aurora on Venus
 Credit: ESA/C. Carreau [41]

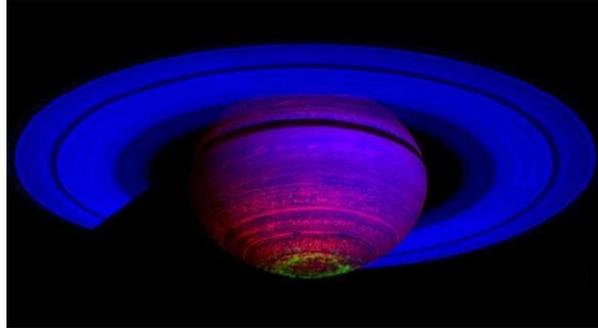


Figure 1.10 Aurora on Saturn

Credit: VIMS Team, U. Arizona, U. Leicester, JPL, ASI, NASA [45]

In order to comprehend the behaviour of upper atmospheres of stellar bodies, auroras and plasmas, it is necessary to have the data pertaining to the cross-section of electron scattering.

1.4.2 Biomedical research field

Investigations by Sanche and his group [46,47] demonstrated that DNA damage resulting in strand breaks are induced by low to intermediate energetic electrons. Theoretically, this type of damage can be examined through Monte Carlo simulations to pinpoint the radiation damage [48]. At this point in time, it is not possible to conduct experiments that would describe the early physical activity caused by ionising radiation at the cellular level. Collision cross-sections are a vital piece of information that need to be provided as an input to the simulation codes PARTRAC [49], NOREC [50], and CPA100 [51]. It is also necessary to have them in order to calculate the average distance between two successive collision sites. In light of these facts, the study of electron-biomolecule interactions has recently garnered a much of attention from the communities of atomic and molecular physics. This paved the path for the advancement of new therapeutics in the medical field.

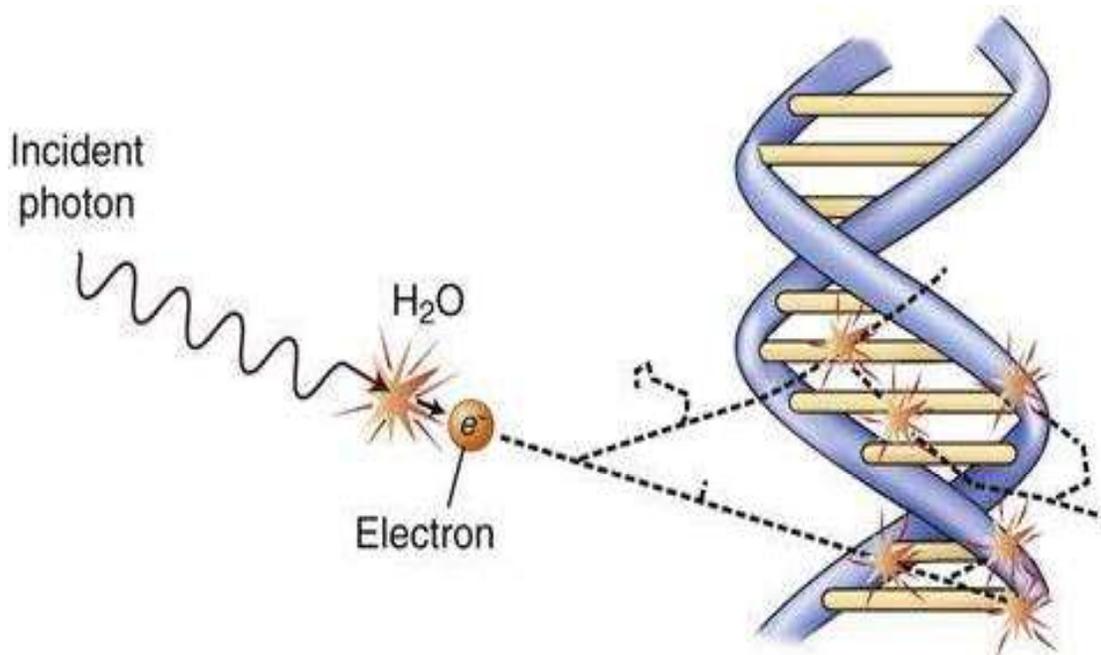


Figure 1.11 DNA strand damage caused by electrons

1.4.3 Industrial fields

Electron scattering also has had a profound impact on several fields of nanoscience and nanotechnology. The most prominent illustration of nanotechnology is the manufacturing of electrical devices, which is dependent on plasma processing technologies in which the electron collisions are the controlling processes [52]. In the semiconductor industry, fluorocarbon plasmas are by far the most common. However, despite their widespread use, fluorocarbons like SF₆ and CF₄ have extremely high GWP [53,54], with SF₆ having the highest GWP of any chemical known. There has been an ever-increasing quest for environmentally appropriate substitute for these gases with high GWP; several of these potential replacements are investigated in this thesis. In addition, electron collisions play an important part in the research on confinement fusion [55], particularly for hydrogen and its isotopes. Furthermore, hydrocarbons are significant at plasma edge [56]. Experimentally, the production of requisite number of radicals is notoriously difficult, therefore there are not many experimental data available for these species. Cross-sections, in general, are readily obtainable for large, complex molecules through the numerous theoretical calculations.

1.5 Outline of the problem

Previous research conducted by our team concentrated mostly on investigating electron scattering processes from gaseous species using the spherical complex optical potential (SCOP) formalism in conjunction with the R matrix approach. A recent modification to our SCOP computations allowed us to investigate the electron interaction with aqueous or condensed phase target systems.

Many of the phenomena that are allied with electron scattering from solid materials are of tremendous importance and applicability in a variety of subfields that fall under the umbrella of physics. In investigating the construction of solid substances, in addition to the diffraction of neutrons and of x-rays, electron diffraction offers a useful complement. Small angle scattering by the specimen is essential to the formation of a picture in the electron microscope, which is proving to be a useful instrument in many areas of study. Recent advances have made the photographic plate a viable medium for studying the characteristics of high-energy particles including electrons. It is crucial to these investigations that researchers have accurate data on the probability of multiple scattering and the rate at which electrons lose its energy in the plate's solid material. Measurement of the onset energy of electrons has traditionally relied on determining the range of electrons in a certain solid material. It is also important to account for emission of the secondary electrons in any device where electrons strike on a solid. Another characteristic of significant practical value is the electrical resistance of semi-conductors and conductors, which is governed by the possibilities of conduction electron scattering across the substance. - Massey 1952 [57]

In 1924 Becker first reported the loss of the electrons owing to inelastic scattering in solids. He discovered the "characteristic" energy loss spectra, which are unaffected by the energy of the projectile electron and are instead based solely on the composition of the material. In 1927 Brown and Whiddington, and in 1929 Rudberg took up this topic and continued research on it, supporting Becker's results (see to [58] and references there in). The significance of the interaction between electrons and the materials has been recognised for around 100 years. Recently, the studies on transmissions and scanning electron microscopy (SEM) investigations have benefited greatly from these kinds of research [59]. Electron beam lithography is another leading technology that places a significant emphasis on electron scattering in thin films [60]. Various surface analysis methods such as Auger electron

spectroscopy, also known as AES and X-ray photoelectron spectroscopy, generally known as XPS, are both driven by the electron-matter interactions [61]. Ionisations and excitations take place in these processes, followed by signal electron emission. Additionally, it's important to understand how electrons interact with biological species in order to learn more about the radiation damage caused by such electrons [62]. In addition to solids, liquid phase processes are of paramount significance in a variety of fields, including electrochemical reactions for energy storage, biological activity and biomineralization [63]. Image labelling structures inside of the cells [64], development of the nanoparticles in liquids [65], and electrochemical reactions [66,67] are just a few of the many uses for electron transmission methods like Transmission Electron Microscopy (TEM). In addition, the interaction of plasma-liquid is very significant in plasma discharge, making it a crucial topic to discuss. Furthermore, such studies can be applied to a wide variety of fields, including but not limited to: medicinal applications, sterilisation, chemical synthesis, material synthesis and processing, electrical switching, and environmental remediation ([68] and references therein).

As was just mentioned, electron collision in many different phases is discovered to be significant in all the multifarious applications in industrial and technological spheres. As a result, modelling such applications with the data of the gas phase is not appropriate. Independently investigated collision results from the liquids and solids are necessary to improve the fidelity of these models. When dealing with the condensed, liquid or aqueous phase, we are interested in obtaining the macroscopic characteristics such as mean free path, in addition to cross-sections. When the target travels through any substance or medium, it is subjected to a series of collisions, each of which has the potential to change either the projectile's trajectory or its amount of energy. The mean free path measures the typical distance that elapses between each of these subsequent collisions. For radiological simulations, other factors like stopping power (SP) and absorbed dose (D) are of the utmost importance [69,70]. There has been a significant amount of data on these characteristic parameters, reported for the biomolecules, including DNA constituents for electron impact in their gaseous phase up until this point. These findings are likewise now accessible for the condensed phase interaction mechanisms also. However, the aqueous phase of biological species, as opposed to the gaseous or condensed phase, offers a more realistic picture, as human body contains around 70-75% of water, the cell tissues, including DNA are always found covered with these water molecules through hydrogen bonding. The examination of

electron interaction with aqueous phase is reported in this thesis, which is an exclusive work from our group.

The primary purpose of this thesis is,

- To conduct the theoretical research on the interaction of electrons with various applied molecules and radicals, which are important species in many applied fields, such as medical sciences, industrial technologies, plasma sciences, astrophysics etc.
- To study the electron collision over a broad spread of energy, 0.1 eV to 5 keV by combining SCOP and R-matrix technique.
- To broaden the scope of our calculations to include aqueous phase molecules.
- To compute the various applied parameters, such as mean free path, stopping power and absorbed dose.
- To propose a new method for estimating the Q_T for the larger complex molecules for energy spectrum from 50 eV - 5 keV.
- To analyse the various correlations between the cross-sections and target properties, such as molecular polarisability and dielectric constant, leading to prediction of them for which the data is not available.

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