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STUDY OF MOLECULAR PROCESSES THROUGH CHARGED PARTICLE SCATTERING

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Introduction

The present work examines and explain the scattering processes of intermediate to high energy electron collision with variety of molecular targets. It will cover the impact energy range from ionization energy of most molecules (about 10-15 eV) to energy at or above 5000 eV. Since the response of an atomic and molecular target to the incident electron depends on its impact energy and the structural properties of the target itself [1]. Since the beginning of the 20th century, both theoretical and experimental molecular physics communities have studied electron molecule collisions.

Our interest in the present collision calculations arises in view of the applications of relevant cross sections data in both pure and applied sciences. The selected targets find applications in atmospheric science, plasma physics / chemistry and radiation physics. In addition to photo-induced processes in the ionosphere, atmospheric ionization is also produced by high energy particles emitted from the Sun and the cosmic rays. These particles may also play a role in cloud formation and climate change. Therefore, it is important to be able to determine the electron impact total and ionization cross sections of Aeronomy species. Different techniques to characterise, describe, and model various processes in low-temperature plasmas, which are increasingly useful in a variety of quickly evolving high-tech applications (such as the manufacture of microelectronic chips) and semiconductor physics, heavily rely on electron-molecule collisions [2,3]. The schematic representation of scattering phenomena is shown in figure 1.

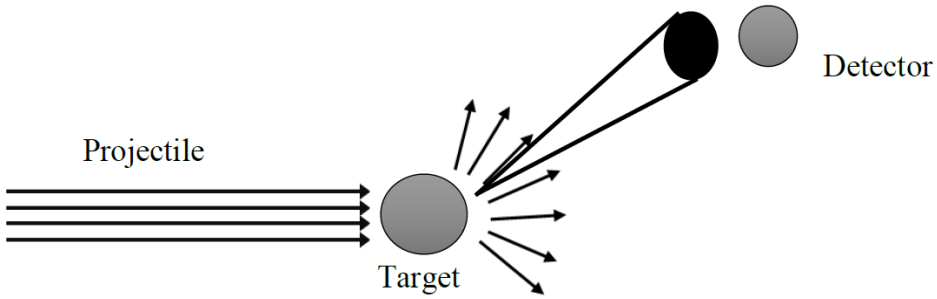

Figure:1 Schematic of scattering phenomena

Recently it has been recognized that electron driven processes also play an important role in life sciences, for example by initiating single and double strand breaking in DNA. DNA double-strand breaks (DSB) and single strand breaks are not formed as a consequence of the direct absorption of UV radiation by DNA. Rather, they occurred as the consequence of the

attempted repair of UV radiation-induced base damage in DNA. Radiation therapy is one of the major cancer treatment techniques other than chemical and surgical therapy. During radiation therapy electrons are produced with a wide range of energies from the irradiated areas. The secondary electrons produced can collide with DNA molecules in human cells, causing damage that destroys the cancer cells [4,5].

Electron scattering experiments with complex molecules e.g. bio-molecules (thymine, adenine, guanine, 3-hydroxytetrahydroFuran, α-Tetrahydrofurfuryl alcohol etc.), industrial relevance molecules (Furfural, para-Benzo quinone, Fluoronitrile etc.) and reactive radicals in the gas phase are challenging because of the practical difficulties involved in the preparation of well-characterized pure gas targets of these molecules and in the subsequent quantitative determination of the target densities. Hence, there is a need for more comprehensive theoretical investigations of electron scattering studies evaluating various quantitative and qualitative cross sections for these molecules/radicals. Therefore, there has been an ever - increasing emphasis on the development of theoretical methods to provide data using simpler approximate theories, capable of delivering cross sections accurately and quickly over a wide range of energy so as to encompass many important phenomena, e.g. ionization of the target.

Brief research methodology

In modeling such processes there is a need for accurate knowledge of electron collision cross sections for relevant biological molecules. All these fundamental as well as technological reasons motivated us to take up this study. In this work we investigate the molecular processes induced by the electrons and quantify them through various cross sections e.g. ionization, excitation, inelastic, elastic, total cross sections etc. For the intermediate and high energy calculations we have employed the Spherical Complex Optical Potential (SCOP) approach and Complex Scattering Potential-ionization contribution (CSP-ic) method.

The study of electron-molecule interactions conducted for the thesis is presented in the following chapters:

Chapter I: Introduction

The scattering of particles provides a wealth of information about the structure of matter. Collision techniques have been effectively used to analyze the internal structure of the atoms and even of their constituent particles and also their nature of interactions. The basic scattering

phenomena are explained in this chapter, along with the different electron interaction processes that can be observed in daily life. It also seeks to provide a summary of the history of electron-driven processes from different atoms and molecules. The study of electron interactions with biomolecules, astromolecules, plasma molecules or technological applied molecules is essential in the various applied fields, such as plasma sciences, radiation dosimetry, astrophysics and semiconductor industries etc. [6]. This chapter provides a brief summary of the significance of the target molecules chosen for the present investigation.

Chapter II: Theoretical Methodology

In this chapter the details of theoretical formalisms used for the present electron interactions study is given. We have used three different methodologies to study various molecular processes quantitively through the cross-sections.

[1] Spherical Complex Optical Potential (SCOP):

The study of scattering of electrons from the target molecules for the energies from ionization threshold (*IE*) to 5000 eV is done using the SCOP formalism. The spherically symmetric complex optical potential of the following form is employed for the calculations of inelastic and elastic cross-sections through the partial wave analysis method [7,8,9].

$$V_{opt}(r, E_i) = V_R(r, E_i) + iV_I(E_i, r) \quad (1)$$

Here, V_R includes the static potential, exchange potential and polarization potential. The imaginary part V_I of the optical potential includes the absorption potential.

[2] Complex Scattering Potential-ionization contribution (CSP-ic):

To bifurcate the continuum (Q_{ion}) and discrete (Q_{exc}) contributions of the inelastic cross-sections, this CSP-ic approach is employed. In this method we compute ionization cross section from inelastic cross sections by defining the dynamic ratio,

$$R(E_i) = \frac{Q_{ion}(E_i)}{Q_{inel}(E_i)} = 1 - C_1\left[\frac{C_2}{U + a} + \frac{\ln U}{U}\right] \quad (2)$$

The dimension less parameters C_1, C_2 and a are obtained by applying the boundary conditions of $R(E_i)$, which is given by,

$$R(E_i) = \begin{cases} 0, & for\ E_i \leq IE \\ R_P, & for\ E_i = E_P \\ 1, & for\ E_i \gg IE \end{cases} \quad (3)$$

where, E_p stands for the peak energy at which the Q_{inel} has its maximum value. From the several results of the experiments and theories the value of R_p at the E_p, is found to be around 0.7 - 0.8 [10-13].

[3] 2 parameters Semi Empirical Method (2p – SEM)

The semi-empirical method is a relatively quick and computationally cheap method for predicting the Total Cross section (TCS) of complex molecules. At the high energies, above 500 eV, which is proposed by Joshipura [14] and Garcia [15] for smaller molecules i.e., for ten electrons (Z=10) and up to Z=22 electrons systems respectively. In the present work this formula has been derived for large molecules with 55 < Z < 95. In this work we have derived two different expressions for the intermediate (50 < E_i < 500 eV) and high energy regions (E_i> 500 eV) for the complex and larger molecular systems with 55 < Z < 95. Further it might serve to audit both the theoretical approaches commonly used to estimate the TCS and also the experimental data, in the range above 50 eV.

A two-parameter expression for Q_T can be formulated for the energy range from 50-500 eV in the equation (4) and for the energy range from 500 – 5000 eV in the equation (5),

$$Q_T(E, n_e, \alpha) = \frac{n_e - 0.003 * \alpha - 22.91}{E^{0.56}} \quad (4)$$

$$Q_T(E, n_e, \alpha) = \frac{0.776 * n_e + 0.016 * \alpha - 17.88}{E^{0.77}} \quad (5)$$

Where, Q_T= Total cross section in atomic unit (a_0^2), n_e= total no. of electrons in the target molecule, α= molecular polarizability in atomic unit (a_0^3) and E is the energy of incident particles in KeV.

Chapter III: Electron collisions with analogous of DNA/RNA nucleobases

In this chapter we report the inelastic, ionization, electronic excitation, elastic and total cross-sections data of the analogous of DNA/RNA nucleobases: 3-hydroxytetrahydrofuran and α-Tetrahydrofurfuryl alcohol for the energy from ionization threshold to 5000 eV. For the calculations, SCOP and CSP-ic methods have been employed. Also, the correlation analysis between the peak of Q_{ion} and polarizability of the molecule has been done. Also, we used another formalism called complex scattering potential – ionization contribution (CSP-ic), to compute ionization cross sections (Q_{ion}) and summed excitation cross-sections ($\sum Q_{exc}$). These

cross-section data which may serve as the input parameters in predicting damage in biomolecular system induced by an electron within the energy range [16].

Chapter IV: Interaction processes for molecules of industrial relevance

In this chapter, we present the theoretical investigations of the electron scattering cross sections of the important complex molecules of industrial relevance, viz., Furfural, Para Benzoquinone and Fluoronitrile molecules for the energy range from molecular ionization energy to 5000 eV. For all of these molecules, the total (Q_T), elastic (Q_{el}), inelastic (Q_{inel}), ionization (Q_{ion}) and excitation (Q_{exc}) cross sections are reported. Also, total cross sections for Furfural, Para Benzoquinone and Fluoronitrile molecules are calculated for the energy rang 50 eV to 5000 eV using 2 parameter semi empirical model [17].

Chapter V: Electron driven processes for aqua DNA constituents

Since in the human body DNA is always covered by the water molecules, for the present investigations of electron interactions with DNA bases, we have considered the molecules in their aqueous phase. This chapter evaluates important applied quantities, such as dipole polarizability, dielectric constant and various cross sections which are critical for assessing DNA damage in biological systems. Also, we have calculated, TCS for aqua DNA constituents [18] for energy rang 50 eV to 5000 eV using 2 parameter semi empirical model.

Chapter VI: Ionization of N_2

The ionization of N_2 is presented in Chapter VI, where various cross-sections have been calculated, including ionization, excitation, elastic, and inelastic cross sections. These calculations cover a broad energy range from threshold to 5000 eV, providing comprehensive insights into the electron interactions with molecular targets. Experimentally, measurements were done in the laboratory of molecular physics at Tata institute of fundamental research (TIFR), Mumbai, as a part of our collaboration with Prof. Lokesh Tribedi, TIFR- Mumbai [19].

Chapter VII: Summary and future prospects

In this last chapter of the thesis, we summarize the present results and draw important conclusions of the present work. The future prospects of the present work in terms of latest experimental and theoretical status, will be also discussed in this chapter. We believe that the present comprehensive electron scattering studies will help in understanding of the various

electron induced molecular processes for different important applied molecules studied here.

Key findings

This thesis proposes a comprehensive approach to the calculations of electron impacts and collisions with molecular targets, focusing on applications in industrial processes, plasma research, biomedical studies, and atmospheric sciences. The theoretical study covers electron collisions with molecular targets across an energy range of 10 eV to 5000 eV, where various elastic and inelastic interaction processes are induced. Experimentally, measurements were done in the laboratory of molecular physics at Tata institute of fundamental research (TIFR), Mumbai, as a part of our collaboration with Prof. Lokesh Tribedi, TIFR- Mumbai. Due to the growing need for accurate cross sections data in both fundamental and applied research fields, interest in collision computations has recently been revitalized. This thesis introduces a method that is both efficient and simple while still delivering reliable results. The primary objective is to estimate a range of cross sections associated with electron scattering from molecules in gaseous and condensed (or aqueous) phases, including total elastic cross sections (Q_{el}), total inelastic cross sections (Q_{inel}), ionization cross sections (Q_{ion}), excitation cross sections (Q_{exc}), and total cross sections (Q_T). The calculated results are then compared with available experimental or theoretical data to validate the approach.

Conclusion

We accomplish the present research with a conclusion of the work reported here. We derive significant insights about the present theoretical frameworks employed in this investigation, concerning their significance from an application standpoint. In thesis, we have shown various electron collision processes and their relevance applications along with theoretical calculation involving various cross sections and of electron induced scattering. The theoretical investigation carried out under the electron collision range starting from threshold to 5000 eV. In order to quantify the different collision processes in terms of cross sections, we made use of different methodology: Spherical complex optical potential (SCOP), Complex scattering potential using ionization contribution (CSP-ic) and recently develop 2 parameter semi-empirical formalism (2p-SEM). To estimate elastic and inelastic cross sections, SCOP formalism is applied for intermediate to high energy regime, staring above the IP of the target

molecules. The CSP-ic is combined with a spherical complex optical potential formalism to compute ionization CSs alongside the co-produced electronic excitation CSs. Electron is the fundamental particles, interact with a molecular system which impacting numerous scientific fields. Studying electron scattering from biomolecules plays a crucial role in understanding DNA damage caused by intermediate energy electrons. This thesis focuses on analogous of DNA constituents biomolecules, 3-hydroxy-tetrahydrofuran and α-tetrahydrofurfuryl alcohol, nucleotides (Adenine, Guanine, Thymine, Uracil, and Cytosine), essential components of life. Among various electron-molecule interactions, ionization holds particular significance in plasmas and various industrial applications. Fluoronitriles (C_3F_5N and C_4F_7N) are plasma-relevant molecules have the potential to replace environmentally harmful SF_6 in plasma applications while maintaining their eco-friendly nature. Beyond plasmas and industrial applications, this study extends to furfural and para-benzoquinone - biomaterials with significant roles in green chemistry. These molecules hold promise for applications like energy storage devices and harvesting systems. This research goes beyond isolated DNA components to consider their interactions with electrons in an aqueous phase. Since living cells surround DNA with water molecules, this approach provides a more realistic picture of electron-induced damage. To achieve this, we employed a modified SCOP formalism to compute various cross section for DNA damage assessment.

Recommendation and suggestions

A significant feature of the present theory is its applicability to determine different cross-sections within the same approach. In future prospects, the suitability of the present theory in biochemistry, astrophysics, and the interstellar medium will be focused on. Also, the electron impact chemistry of the newly found astro-molecules in the astro-chemistry field will be focused. The accountability and suitability for the data obtained using present theoretical methodologies to the internationally recognized online data centers like e-Mol, the Virtual Atomic and Molecular Data Center (VAMDC) etc. will also be discussed. The extension of the present theoretical methodologies to biologically relevant molecules like proteins and also to industrial relevant molecules like carbon nanotubes and graphene, etc. will be focused.

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