EXECUTIVE SUMMARY

Name of the Principal Investigator: **Dr. Chetan G Limbachiya**

Title:
Calculations on collisions of electrons with Atoms and Molecules of Applied Interest and ionization of bio-molecular targets

Abstract: The project aimed at theoretical study of interaction of electrons with different important atomic and molecular targets including simple biomolecules. The calculations of various total cross-sections (TCSs), e.g. total (complete) cross sections (Q_T), total inelastic cross sections (Q_{inel}), total elastic cross sections (Q_{el}), total ionization cross sections (Q_{ion}) and summed total excitation cross sections ($\sum Q_{exc}$) find applications in several fields of science and technology. The results are compared with available theoretical and experimental data and published in reputed journals.

Introduction:

Atomic - molecular collision study has a history of more than 100 years now and has found important place in the exploration of basic processes like excitation, ionization and dissociation or fragmentation at the subatomic level. While the problem of electron scattering with atoms and molecules has been addressed by many theoretical and experimental scientists in recent past, here the main intention is to find theoretically, various cross-sections which are the measures of the probabilities of different processes to occur in the regime of electron - atom / molecule interactions.

These processes have gained much importance in environmental sciences, study of aurora, ionosphere of the earth, chemistry of radiation and other astrophysical and biological environments. The basic knowledge gained in the electron scattering studies has a lot of potential for applications in the

pure and applied fields of Lasers, atmospheric sciences, aeronomy, astrophysics etc. Growing interests in the study of interaction of electrons with organic compounds is inspired by their wide application in microelectronics, biology and medicine.

Electron-molecule collisions play key role in different methods to characterize, describe and model the complex processes in low-temperature plasmas, which are of ever-increasing utility in many rapidly developing high-tech applications (e.g. fabrication of microelectronic chips) and in semiconductor physics.

The development of the next generation of micro and nano systems technologies (MST) remains essential to the commercial success of the global information industry. MST devices are silicon based and control of the processing methodology to produce such MST can only occur through the understanding of the properties of plasma, which etch the Silicon wafers.

In the life sciences the role of electron-driven processes is being recognized as crucial to our understanding of radiation damage of cellular materials. Secondary electrons with energies up to 20 eV are thought to induce DNA lesions mainly through resonant scattering. Recently the mechanisms by which such degradation occurs have been the subject of considerable research effort with genotoxic effects of ionising radiation in living cells being commonly attributed to direct impact of high energy quanta or by complex radical chemistry triggered by the production of OH species by the primary ionising radiation. In our investigation we study water and the OH radical on electron impact along with other radicals. Collision data is needed to develop models of the track structures in irradiated media; these models, in turn, can help in understanding the effects on humans of low radiation doses, both from natural causes and those employed in medicine for diagnostics and treatment.

In this project the aim was to address in detail, the problem of electron scattering from atoms, molecules and radicals and present calculations to obtain various total cross sections for these targets of applied interest. In our theoretical approach, which is quantum mechanical, we use the optical potential in the Schroedinger equation and solve it numerically. The targets are approximated to spherical symmetry so that partial wave analysis can be employed to calculate the phase shifts, which are used to calculate the total cross sections. For the large targets such as biomolecules, group additivity approximations are employed. We calculated total (complete) cross sections (Q_T), total inelastic cross sections (Q_{inel}), total elastic cross sections (Q_{el}), total ionization cross sections (Q_{ion}) and summed total excitation cross sections ($\sum Q_{exc}$) for targets of applied interest. We are able to investigate the relative proportions of these cross sections and hence the corresponding physical phenomenon dynamically.

Theoretical Methodology

We briefly describe the theoretical formalism employed to determine the Q_T, Q_{el} and Q_{ion} for the impact of electrons on the atoms, molecules and radicals studied. Our aim is to calculate various TCSs and to investigate their contributions relative to the total (complete) cross sections for the chosen targets. We employ the well-known spherical complex optical potential formalism (SCOP). The complex potential calculation for electron scattering can provide total elastic cross sections Q_{el} and its inelastic counterpart, Q_{inel} such that

$$Q_T(E_i) = Q_{el}(E_i) + Q_{inel}(E_i) \tag{1}$$

Here E_i is incident energy in eV. For a polar molecule like H_2O, one can determine the dipole rotational excitation cross section $Q_{01}(D, E_i)$. In such cases we can define the grand total cross section by,

$$Q_{TOT}(E_i) = Q_T(E_i) + Q_{01}(D, E_i) \qquad (2)$$

The dipole rotational excitation $Q_{01}(D, E_i)$ is calculated using the first Born approximation for a molecular dipole of strength D.

Our calculation for the TCSs (eqn.1) is based on a complex scattering potential, generated from spherically averaged charge densities of the target. We use single center approach for molecules like CH_4 and H_2O molecule. The charge density constituent atoms are expanded at the center of mass of the molecule. In contrast formaldehyde, HCHO and formic acid (HCOOH) are multi-centre systems. We have use the multi-center group additivity rule. Here, the charge density of lighter hydrogen atom is expanded at the center of heavier atom (carbon or oxygen). The single-center molecular charge density is then obtained by a linear combination of constituent atomic charge densities, renormalized to account for covalent molecular bonding. The justification for group additivity comes from relatively larger C–O bond length compared to C-H or O-H bond lengths in these molecules. The molecular charge density thus obtained is employed to construct a complex optical potential V_{opt}, given by

$$V_{opt}(E_i, r) = V_R(E_i, r) + iV_I(E_i, r) \qquad (3)$$

The real part V_R comprises of static potential (V_{st}), exchange potential (V_{ex}), and polarization potential (V_p) terms, as follows;

$$V_R(E_i, r) = V_{st}(r) + V_{ex}(E_i, r) + V_P(E_i, r) \qquad (4)$$

We use the analytical form of the static potential derived using the Hartree-Fock wave functions. For the exchange potential, we employ Hara's 'free electron gas exchange model'. For the polarization potential V_p, we use parameter free model of correlation polarization potential which contains some multipole non-adiabatic corrections in the intermediate region and it smoothly approaches the correct asymptotic form for large r given by Zhang et al. The imaginary part V_I in eqn. 4 also called the absorption potential V_{abs}, accounts for the total loss of scattered flux into all the allowed channels of electronic excitation and ionization. For V_{abs}, the model potential given by Staszewska is used, which is a quasifree, Pauli-blocking, dynamic absorption potential. After generating the full complex potential given in eqn. (3) for a given electron-target system, we solve the Schrödinger equation to get complex phase shifts which are used to find the cross sections given in eqn. 1. The total inelastic cross section, Q_{inel} cannot be measured directly.

Q_{inel} can be partitioned into two main contributions viz.

$$Q_{inel}\left(E_i\right)=\sum Q_{exc}\left(E_i\right)+Q_{ion}\left(E_i\right) \qquad (5)$$

where the first term is the sum over total excitation cross sections for all accessible electronic transitions. The second term is the total cross section of all allowed ionization processes induced by the incident electrons. The first term arises mainly from the low-lying dipole allowed transitions for which the cross section decreases rapidly at higher energies. The first term in eqn. 5 therefore becomes progressively smaller than the second at energies well above the ionization threshold. By definition

$$Q_{inel}\left(E_i\right)\geq Q_{ion}\left(E_i\right) \qquad (6)$$

Q_{ion} cannot be directly derived from Q_{inel} but may be estimated by the energy dependent ratio,

$$R(E_i) = \frac{Q_{ion}(E_i)}{Q_{inel}(E_i)} \qquad (7)$$

such that,

$$0 < R \leq 1$$

We require that $R=0$ when $E_i \leq I$. (I is ionization potential for the target). For a number of stable molecules like O_2, H_2O, CH_4, SiH_4 etc., for which the experimental cross sections Q_{ion} are known accurately, the ratio R rises steadily as the energy increases above the threshold, and approaches unity at high energies. Thus,

$$\begin{aligned} R(E_i) &= 0 & for \quad E_i \leq I \\ &= R_P & at \quad E_i = E_P \\ &= 1 & for \quad E_i > E_P \end{aligned} \qquad (8)$$

where E_p stands for the incident energy at which the calculated Q_{inel} attains its maximum. R_p is the value of R at Ei=Ep, and we choose R_p = 0.7 to 0.8. This choice follows from the general observation that at energies close to peak of ionization, the contribution of the molecular Q_{ion} is about 70–80% in the total inelastic cross sections Q_{inel}. This behaviour is attributed to the faster fall of the first term ΣQ_{exc} in eqn.5. For calculating the Q_{ion} from Q_{inel} we need R as a continuous function of energy for Ei>I; hence we represent the ratio R in the following manner,

$$R(E_i) = 1 - f(U) \qquad (9)$$

The above ratio can be determined using the following analytical form.

$$R(E_i) = 1 - C_1 \left(\frac{C_2}{U+a} + \frac{\ln(U)}{U} \right) \tag{10}$$

where U is the dimensionless variable defined by,

$$U = \frac{E_i}{I} \tag{11}$$

The reason for adopting a particular functional form of $f(U)$ in eqn. 11 is the following. As E_i increases above I, the ratio R increases and approaches 1, since the ionization contribution rises and the discrete excitation term in Eqn. 5 decreases. The discrete excitation cross sections, dominated by dipole transitions, fall off as $\ln(U)/U$ at high energies. Accordingly the decrease of the function $f(U)$ must also be proportional to $\ln(U)/U$ in the high range of energy. However, the two-term representation of $f(U)$ given in eqn.10 is more appropriate since the first term in the square brackets ensures a better energy dependence at low and intermediate E_i. Eqn.10 involves dimensionless parameters C_1, C_2, and a, that reflect the target properties. The three conditions stated in eqn. 8 are used to determine these three parameters. To implement the third condition, we first assume $a=0$ and consider a two-parameter expression in Eqn. 10. We employ the first two conditions of Eqn. 8 to evaluate the C parameters. The two parameter expression is then used to obtain the value of R at a high energy $E_i=10E_p$, and are employed in the third condition of eqn. 8. This method discussed through eqns. (6–10) is called our Complex Scattering Potential –ionization contribution, CSP-ic.

The SCOP and CSP-ic methods discussed above enable us to determine all the cross sections to be used in various applications mentioned in earlier sections.

List of Publications

Details of the Publications resulting from the project work (please attach reprints) letter of Acceptance of paper communicated.

Sr.	Title of the paper	Journal, Vol, Year	ISSN/ISBN	Impact factor
1	Electron impact calculations of total ionization cross sections for environmentally sensitive Diatomic and Triatomic molecules from threshold to 5 keV"	J. Phys. B.,**43** (2010) 015203	1361-6455	2.031
2	Theoretical study of electron scattering processes with H_2O (ice)	Atomic Structure and Collision Processes' Pg.217(2009) Conf. Proceeding: Editor: Prof. Man Mohan	978-81 -7391- 811-3	--
3	Total and ionization cross-sections for well-known and exotic hydrocarbon molecules upon electron impact	Atomic Structure and Collision Processes' Pg.177(2009) Conf. Proceeding: Editor: Prof. Man Mohan	978-81 -7391- 811-3	--
4	"Screening-corrected electron impact total and ionization cross sections for Boron trifluoride (BF_3) and Boron trichloride (BCl_3)",	J. Phys. B **41** (2008) 245202	1361-6455	2.031
5	"Theoretical electron impact elastic, ionization and totalcross sections for silicon hydrides, SiH_x (X=1,2,3, 4) and Disilane, Si_2H_6 from threshold to 5 keV"	Euro. Phys. J. D, **48(3)** (2008) 333	1434-6079	1.513
6	"Electron impact calculationsof total and ionization cross sections for Germanium Hydrides (GeH_X; X=1-4) and DigermaneGe$_2$H$_6$"	Int. J. of Mass Spectrom., **273**(2008)145	1387-3806	2.142
7	"Computation of total electron scattering cross section for molecules of astrophysical relevance."	Journal of Phys. B-Conf. Series **115** (2008) 012013	1361-6455	2.031