Chapter 8
Summary

In conclusion, we have presented an intensive study for ionization of atoms, small molecules and large biomolecules in collisions with fast electrons, protons and highly charged ions. A detailed experimental study has been performed to understand the collision dynamics of the different projectiles interacting with the target species. The well known electron spectroscopy technique has been employed to understand the intricate details of the different aspects of the collision mechanisms taking place when a projectile collides with the neutral target species. The double differential cross section (DDCS) of the electrons emitted from the target atom/molecule were measured as a function of their emission energy and emission angles. An electron gun capable of generating keV energy electrons was used to perform the experiments using fast electrons. The 14.5 GHz ECR ion accelerator at TIFR was used to produce the keV energy protons and the 14 MV Pelletron accelerator was used to carry out the experiments involving MeV energy C^{6+} ions.

For performing experiments with the electron gun, the existing beamline was revamped in order to improve the beam quality and beam transmission. Several apertures were added in the beamline to generate a collimated parallel electron beam. The filament of electron gun was aligned with respect to the anode plate aperture and along with all the apertures in the beamline. Further, a pair of magnetic coils were introduced which increased the beam transmission through the electron spectrometer drastically to $\sim 90\%$. The electron gun was used to generate electrons having energies between 3 and 8 keV which collided with the diatomic molecules N_2 and O_2. The 50^0 north beamline of the ECR ion accelerator was aligned with respect to the ion source, initially with water level and then with laser beam. Two effusive jet source were used to perform the experiments for gaseous targets and biomolecules in collisions with 200 keV protons. The nozzle of the effusive jet source used for gas target experiments was initially aligned with respect to the beamline by performing mechanical alignment and then further alignment was carried out by keeping the beam "ON" and checking the electron count rate by moving the nozzle tip in a direction perpendicular to the beamline. An effusive jet oven assembly was designed and fabricated as a part of this thesis work for producing vapour of biomolecules which are commercially available in powder form. A water cooled jacket was prepared and the metallic oven was housed inside the jacket so as to avoid the production of dark counts due to heat transfer from the oven to the detector. The biomolecules studied in the present thesis are uracil, a nucleobase of RNA and bromouracil, one of the members of the class of halouracil molecules. Here too the effusive jet source was aligned with the beam using the same method as described above. The oven temperature was increased gradually to

140 - 200 ^{0}C over a time period of 24 hrs. This exercise was performed to ensure a stable rate of flow of the vapour effusing out from the oven assembly. The e-DDCS measurements were performed for 200 keV proton impact on uracil and bromouracil using the ECR ion accelerator. The Pelletron accelerator was used to carry out the experiments for uracil and bromouracil in collisions with 66 MeV bare C ions and further for 42 MeV C^{6+} ions impacting on bromouracil. In addition to the experiments with biomolecules, the ECR ion accelerator was also used to study the collision dynamics for 200 keV H^+ ions colliding with the atomic target, He and small molecules CH_4 and O_2. Measurements were also carried out for 66 MeV C^{6+} ions in collisions with O_2 molecules. All the measurements were compared with state-of-thye-art theoretical models. The ion impact ionization studies of He, CH_4 and O_2 were compared with the CDW-EIS calculations. Similarly the ionization of uracil and bromouracil by 200 keV protons and 42 and 66 MeV bare C ions were also compared with the CDW-EIS model. The electron impact ionization studies were compared with the CB1 and the CTMC model. The CTMC model showed an excellenet agreement with the measured DDCS for N_2. Further, the CSP-ic model was used to compute the total ionization cross section of electron impact on N_2.

A major part of the present work dealt will investigating the evidence of interference oscillations from the ionization channel for the multi-electronic targets N_2 and O_2 when collided with 7 keV electrons. For this purpose, the DDCS measurements were performed for electron emission from the two targets. The cross sections were measured for several forward and backward emission angles. Theoretical DDCS for atomic nitrogen and oxygen were calculated based on the B1 model. For nitrogen, the calculations were performed considering two different effective target charges i.e., Z_T = 1 and Z_T = Z_{bk}, whereas for oxygen an effective charge of unity was considered. The experimental-to-theoretical DDCS ratios (i.e., N_2/2N) were obtained using both the values of target effective charge. The ratios revealed clear oscillatory structures due to the Young type interference effect for all the emission angles, although the shape of oscillation was found to be dependent on the choice of Z_T. For Z_T=1, the DDCS ratios revealed clear oscillations, however, in case of Z_T=Z_{bk}, the ratios had to be normalized by a linear function to deduce the oscillations clearly. The ratios were further fitted with the Cohen-Fano model for interference in molecular double-slit. The forward-backward asymmetry parameter which was obtained only by using the measured DDCS for the molecular target (N_2), displayed clear evidence of interference oscillations and the fitting function based on the Cohen-Fano model matched well with the experimentally obtained $\alpha(k)$. However, periodic deviations were seen in the asymmetry parameter which indicated the presence of higher order scattering mechanism. In the similar manner, experimental-to-theoretical DDCS ratios were also obtained for the O_2 target and here too oscillations were seen for the different emission angles. To provide a quantitative estimate of the frequency of oscillations, the same were fitted with the Cohen-Fano model and the fitting matched well with the data points. The angular distribution of the DDCS spectra showed forward-backward asymmetry. This phenomena was exploited to generate the asymmetry parameter. The asymmetry parameter further showed signature of interference os-

cillations when derived for a pair of complementary angles as well as non-complementary angles. $\alpha(k)$ was fitted by a function derived on the basis of the Cohen-Fano model. The fitting matched quite well with the experimentally measured data, thus providing an unambiguous evidence of the interference effect. To make a comparative study of the variation of $\alpha(k)$ for molecular and atomic target, asymmetry parameter was also calculated for Helium, which showed a monotonically increasing behaviour as compared to the oscillations observed for N_2 and O_2 molecules. The remarkable result obtained in our work showing a clear signature of interference patterns in the electron emission from the diatomic molecules N_2 and O_2 induced by fast electrons is possibly due to the fact that simultaneous multiple ionization of different orbitals is much less in case of fast electron impact ionization, compared to that for heavy-ion collisions.

Further, to complete the database DDCS measurements were performed for keV energy electron impact ionization on N_2, for which no data exists in the literature beyond 2 keV. The measurements were performed for absolute DDCS, SDCS and TCS of the secondary electron emission in ionization of N_2 under the impact of 3, 4 and 5 keV fast electrons for emission angles between 30° and 145°. The experimental DDCSs spectra have been compared with the CB1 model calculations with two different target wave functions as well as with the CTMC model for twice of atomic nitrogen. No significant difference has been observed between the two sets of the CB1 model calculations corresponding to the two wave functions at the RHF/6-311G and CCSD/cc-pVTZ levels of theory, implying that both the description of the target wave functions work well in the present energy regime. The CTMC model provided a very good agreement with the measured data for the entire emission spectra except for certain energy-angle window region. The CB1 model predicted lower cross section values compared to the experimental data for all emission energies, with maximum discrepancy lying in the low emission energy region. The derived TCS values have been compared both with the *ab initio* CB1 and CTMC calculations as well as with the semi-empirical CSP-ic model. While qualitatively both the CB1 and the CSP-ic models show similar energy dependence, the CTMC gives the closest representation to the measured TCS values within experimental uncertainties.

In addition to carrying out the experiments for electron impact ionization and further investigating for the interference oscillations from the measured DDCS, ionization studies were also performed for ion impact ionization on atoms and molecules. We have studied the variation in the collision dynamics for atoms and molecules when ionized by keV energy protons and MeV energy bare C ions. The absolute DDCS of the electrons emitted from an atomic target He were measured for projectiles 150 keV and 200 keV protons. In addition, e-DDCS measurements were also carried out for two molecular targets CH_4 and O_2 when ionized by 200 keV/u protons along with that for O_2 in collisions with 5.5 MeV/u bare C ions. The energy of the projectiles and their charge state were chosen in such a manner that the perturbation strength were nearly the same for all of them. In case of 66 MeV bare C ions, the CDW-EIS calculations for oxygen showed an excellent agreement with the measured data for all the angles. For He, the model

provided reasonably good agreement for both 150 keV and 200 keV protons. Similarly, for 200 keV proton impact on CH_4, the calculations showed good agreement with the experimental DDCS. However, for single ionization of O_2 by keV energy protons, the model overestimates the data in case of all the emission angles. The angular distribution revealed a distinctly different character for the two different projectiles. In case of keV energy collisions, the forward backward asymmetry parameter has much higher value compared to that for MeV energy bare C ions, although the perturbation strength were nearly similar. This indicates that the perturbation strength (q_p/v_p) alone cannot characterize completely the asymmetry and two center effect. For 150 and 200 keV protons, $\alpha(k)$ showed a saturation effect for all the three targets when the electron velocity is greater than the velocity of the projectile. The single differential distributions and total cross sections are also derived. The CDW-EIS provides best agreement for the collisions with 5.5 MeV/u bare C ions whereas deviations (by a factor of 1.4 to 2.3) exist for the keV energy protons with maximum difference occurring in case of O_2, inspite of having nearly same perturbation strength for all the collisions. Further systematic investigations are required to check the efficacy of perturbation strength in characterizing the collision dynamics.

Finally ion impact ionization studies were performed for uracil and bromouracil when collided by keV energy protons and MeV energy bare C ions. DDCS measurements were performed for uracil and bromouracil in collisions with 5.5 MeV/u bare C ions and 200 keV protons. In case of bromouracil, measurements have also been extended for 3.5 MeV/u bare C ions. The DDCS measurements have been compared with the CDW-EIS calculations for both the projectiles at two different energy regimes. In almost all the cases it has been observed that theory predicts lower cross section than the measured DDCS. Further, the ratio of the DDCS for bromouracil to uracil were derived for all the emission angles for both 66 MeV C^{6+} ions and 200 keV protons. The ratios were deduced to understand quantitatively the enhancement in electron emission from bromouracil compared to uracil due to the presence of the Br atom. For 5.5 MeV/u bare C ions, an enhancement of approximately 1.25 times has been observed for majority of the emission angles. Similarly for 200 keV proton impact studies when performed on the same target species showed an enhancement of electron emission by a factor of about 1.4 to 1.5 times from bromouracil. In a similar kind of study reported recently [53], we have shown the DDCS for electron emission from iodouracil and uracil and it has been observed that the enhancement in electron emission from iodouracil is by a factor of about 2.3 times which is explained by the well known GDR feature present in iodine atom. As expected, the enhancement in electron emission from bromouracil should be less than that from iodouracil and our present experimental investigations fall in line showing an overall enhancement of about 1.4 times in case of bromouracil. This shows that addition of a single Br or I atom to a uracil molecule can increase the yield of the low energy electrons which are the main catalyst for radiation damage. The present study which provides a quantitative estimate of enhancement in low energy electron emission from halouracil is the first of it's kind and such measurements can serve as inputs for determining the doses of the incoming beam for radiation therapy.