Indian J. Phys., **Vol. 85,** No. 12, pp. 1705-1715, December 2011

Probing the dynamics of collision induced processes in atoms, molecules and solid surfaces with impact of keV electrons

R Shanker
Department of Physics, Atomic Physics Laboratory, Banaras Hindu University, Varanasi-221 005, Uttar Pradesh, India

E-mail : shankerorama@gmail.com

Abstract : Collision induced excitation and ionization of atoms and molecules by energetic electrons play an important role in thermonuclear fusion plasmas, and in atmospheric science. In this paper, a brief review of the recent experimental works that have been carried out in the Atomic Physics Laboratory at Banaras Hindu University, Varanasi is presented. Some selected examples of the collision induced processes involved in impact of keV electrons with atoms, molecules and solid surfaces is highlighted. In particular, the results obtained on the multiple- and dissociative ionization of Ar atom and SF_6 molecule, the electron backscattering from a pure thick *W* target and on the electron bremsstrahlung emission from gaseous Ar and thick solid Ti targets are presented and discussed.

Keywords : Excitation; multiple dissociative ionization; electron backscattering; electron-bremsstrahlung; Auger electrons; Doppler effect.

PACS Nos. : 33.20.-t, 34.80.Dp, 79.20.Rf

1. Introduction

In this paper, we have discussed the experimental results that have been obtained in recent years on the collision induced processes in atoms and molecules under impact of keV electrons in Atomic Physics Laboratory at Banaras Hindu University (BHU), Varanasi, India; no efforts is made to review the works of other investigators in this area except for direct comparison. Primarily we have concentrated our goal on three different types of problems, namely, (i) the multiple and dissociative ionization of atoms and molecules, for instance, Ar and SF_6 in impact of energetic electrons (a few tens of keV) in single collision conditions using a slow/fast coincidence technique, (ii) the study of electron backscattering from metallic surfaces, for example, *W* and (iii) measurement of electron bremsstrahlung spectra emitted from atoms and thick solid targets, for example, Ar and Ti. It may be mentioned here that the experimental setup for these investigation initially was developed in a collaborative research program between BHU and University of Greifswald, Germany under Indo-German joint cooperation around 1996 and later these studies have been continuously supported by DST, New Delhi. As a matter of fact this Atomic Physics Laboratory is one of the best existing functional laboratories in the university system of the country.

2. Setup for study of multiple and dissociative ionization of atoms and molecules

Figure 1 shows a schematic diagram of the experimental arrangement for measuring the doubly-differential cross sections (DDCS) of multiply ionized atoms and molecules using an electron-ion coincidence technique [1]. In this crossed beam type experiment, a beam of mono-energetic electrons interacts with a dilute beam of chosen gaseous target of atoms and molecules. For this, a well defined beam of atomic/molecular gas is introduced in the interaction zone by an effusing gas through a hypodermic needle or a multi capillary tube. Currently we are replacing it by a supersonic gas jet system for obtaining the target gas atoms at a sub-kelvin temperature with high number densities (~10^{11} atoms/cm^3).

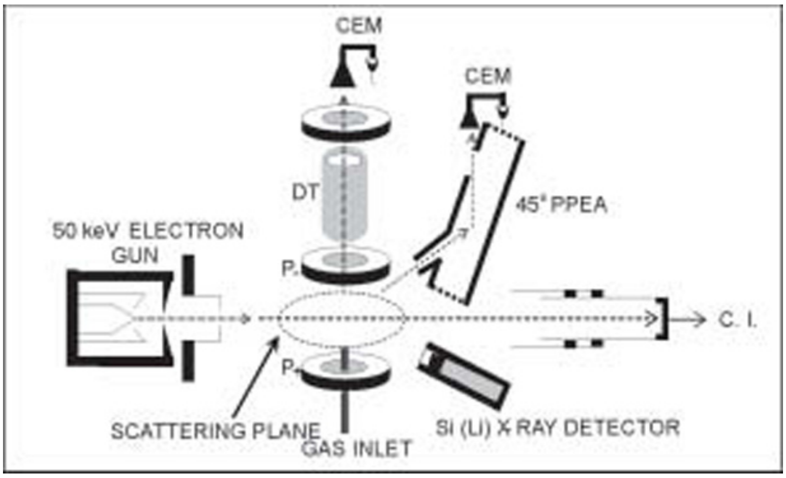

Figure 1. Schematic diagram of the experimental setup. DT : Drift tube; P+, P- : Extraction plates; CEM : channel electron multiplier; PPEA : Parallel plate electrostatic analyser; C.I. : Current integrator.

From the interaction region electrons ejected at 90° to the electron beam direction in anarrow solid angle are accepted by an indigenously built 45° parallel plate electrostatic analyser (PPEA). The energy analysed electrons are detected by a channel electron multiplier (CEM). Ions produced by collision process are extracted from the collision zone in a 4π solid angle by a small electric field (50–150 V per centimetre) depending on the type of gas used and are finally detected into a time-of-flight (TOF) mass spectrometer (home-built). Inside the ion analyser, ions are further accelerated by a potential difference up to 100 V depending upon the gas under consideration and allow them to drift in a field free region of about 20 mm length. At the end of drift tube, the ions are accelerated by a negative potential of about 3.0 kV at the mouth of CEM to insure efficient detection of all ions species [2].

From the time spectrum after subtraction of random coincidences, the number of true coincidence N_c^n is related to the n+ partial DDCS by,

$$\frac{d^2\sigma^{(n)}}{dEd\Omega} = \frac{N_c^n \sigma_i}{N_i \Delta E \Delta\Omega \in_\delta} \tag{1}$$

where, σ_i is the total cross section for ion production, N_i the number of detected ions, ε_δ the efficiency of the electron detection system and ΔE and $\Delta\Omega$ are respectively the energy band width and solid angle associated with the electron-analyser.

The DDCS ($d^2\sigma/dEd\Omega$) for ionisation can be found from the data of the partial doubly differential cross sections ($d^2\sigma^n/dEd\Omega$) using the relation,

$$\frac{d^2\sigma}{dEd\Omega} = \sum_n \left(\frac{d^2\sigma^n}{dEd\Omega} \right) \tag{2}$$

The partial double differential cross sections (PDDCS) of atoms, for example, of Ar ions are measured by detecting coincidences between produced Ar-recoil ions and electrons of different energies ejected at angle θ_e = 90° to the incident beam direction. This angle of ejection was chosen due to formation of a well-defined interaction zone which is viewed by the analyser compared to the case at forward or backward ejection angles. A typical TOF spectrum of multiply charged Ar ions detected in coincidence with the ejected electrons of energy E_δ = 190 eV (Ar L-MM Auger electrons) from 12 keV e^- + Ar collisions is shown in Fig. 2 (also see, Ref. [3]). This study has shown that the

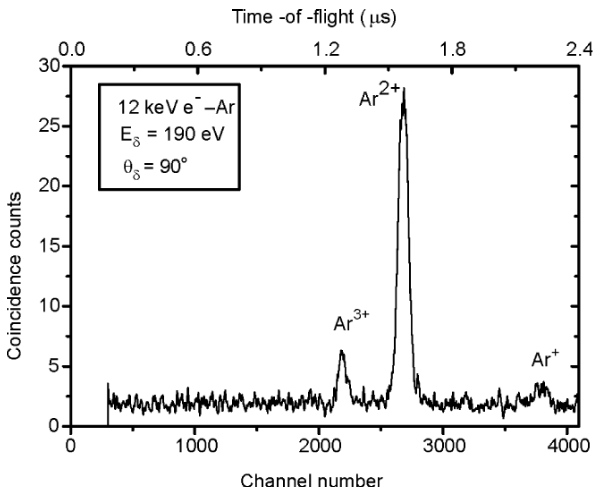

Figure 2. Time-of-flight spectrum of argon ions produced from coincidences between the electrons with energy 190 eV ejected at 90° with respect to incident beam direction and the recoil ions in collisions of 12 keV electrons with argon atoms.

Ar^+ ions are produced due to directly ejected electrons of energy 190 eV with a very small probability; while the multiply charged argon ions are produced due to cascade Auger- and through the shake-off processes.

3. Ionic fragmentation of SF_6 molecule

The relative partial differential ionization cross sections for the production of ionic fragmentation of SF_6 molecule under impact of 10–20 keV electrons have been measured using an ejected electron-ion coincidence technique in a crossed beam apparatus as mentioned above with a time-of-flight analysis of the ions. The detection angle of the ejected electrons of un-discriminated energies was kept at 90° with respect to the incident electron beam direction. The 14-ionic fragments, namely SF_5^+, SF_4^+, SF_3^+, SF_2^+, SF^+, S^+, F^+, SF_4^{++}, SF_3^{++}, SF_2^{++}, S^{++}, F^{++}, SF_3^{+++} and S^{+++} resulting from the dissociative ionization of the SF_6 molecule were identified and their relative production cross sections were measured (Fig. 3). The branching ratios of 8-ionic fragments SF_m^{n+} (m = 1–5; n = 1–3) as a function of impact energy were determined. These ratios are found to have an almost a constant value over the considered impact energies, no previous data or theoretical calculations exist for a direct comparison with the present results. For a more detailed description of this work, see, Ref. [4,5].

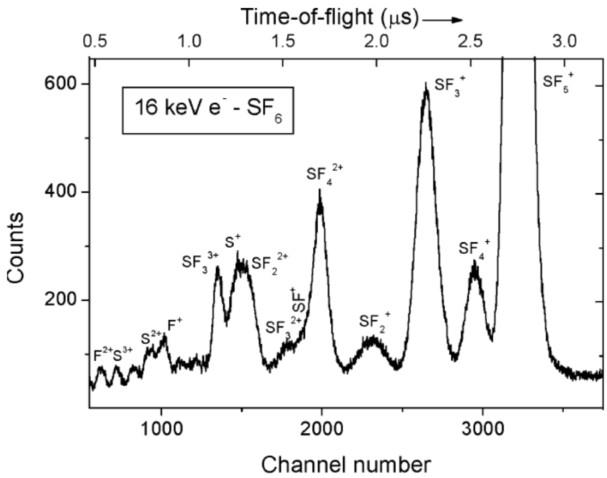

Figure 3. Time-of-flight spectrum of the SF_6 ionic fragments produced in 16 keV e^- + SF_6 collisions.

4. Nuclear dynamics probed by ejected electrons from SF_6 molecule under keV electron impact

Ionic fragmentation of molecules induced by energetic charged particles has been the subject of current interest in basic research of collision physics. During the molecular dissociative ionization, if a core electron of an atom in the molecule is promoted to an unbound molecular state, then the produced core hole may decay *via* auto ionization [6]. In contrary if the core electron is directly ionized, then the unbalance created in the molecular charges induces the Coulomb explosion which in turn breaks the molecular bonds and results into ionic fragmentation in a time-scale of femto seconds. The fragmented ions may acquire the relative kinetic energies of large values [7] and finally decay through the Auger emission in the time-scale less than 10^{-14} s. As a consequence, the atom-like auger decay feature can coexist in molecular Coulomb explosion with its core ionization. Hence, the nuclear motion of the core ionized atom may influence the characteristics of the emitted Auger electrons. The obvious influence is due to the Doppler effect on Auger electrons emitted from the core excited or the core-ionized moving particles. The study of such Doppler effect in the electron spectra of a core-ionized molecule may prove to be a unique tool in studying the details of dynamics of nuclear motion as well as Auger emission in a given collision reaction.

We have produced an experimental evidence which shows that the Fluorine *K*-Auger line produced from a core-ionized polyatomic SF_6 molecule suffers the Doppler effect under impact of keV electrons. This effect comes into play due to the motion of F^+ ions which gain a large kinetic energy as a result of Coulomb explosion of a transiently formed SF_6^{++} ion. A schematic diagram of this reaction is shown in Fig. 4. Results obtained on intensity variation and energy shift of the *F-K* Auger line have confirmed the above mentioned conclusions. For a detailed discussion and description of the experimental procedures, [8].

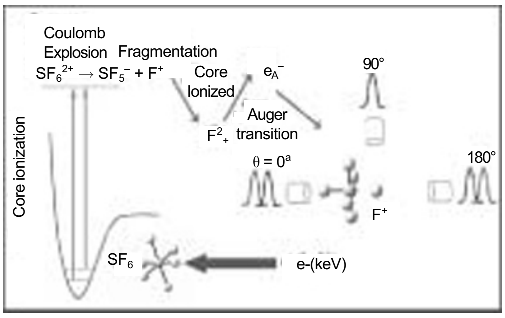

Figure 4. Schematic diagram showing F-K Auger line produced from a core ionized fluorine atom of a SF_6 molecule and suffering with the Doppler effect under impact of keV electrons.

5. Observation of interference effect in ejected electrons

Further the study of energy and angular distributions of ejected electrons from molecules under impact of energetic charged particles is one of the commonly used techniques to shed light on ionization processes [9]. Depending on the projectile velocity, structures in energy and angular distributions of ejected electrons are found to change according to different roles played by various collision processes in electron ejection from the target molecules. The simultaneous ejection of electrons from constituent atoms of a molecule *via* 'soft' collisions may add coherently and produce an interference pattern at the backward angles $\theta(\pi/2 < \theta < \pi)$. Hence, for high velocity of incident particles $v_p \sim 50$ a.u., the momentum transfer becomes minimum to the target electrons and the soft collision gives rise to an oscillatory structure due to interference effect among the ejected electrons. In this context, we carried out an experiment to measure the relative energy and angle dependent cross sections for emission of electrons from SF_6 molecule by impact of 16 keV electrons. The angular distribution of ejected electrons shows an oscillatory structure which is suggested to arise due to an interference effect (Fig. 5). The condition for occurring an interference effect in the considered collision system is found to set in above a threshold energy of 60 eV for the ejected electrons. The ejected electrons producing an interference structure are suggested to originate from two atomic centres (S and F) of a transiently formed doubly ionized parent molecule, namely, SF_6^{++}. This extremely unstable ion suffers a Coulomb explosion and gives rise to many singly and multiply charged stable radical and atomic ions. The time-of-flight mass spectrometric results of our earlier work [4] on partial ionization of SF_6 molecule by impact of 16 keV electrons are found to support the existence of these stable ions [10].

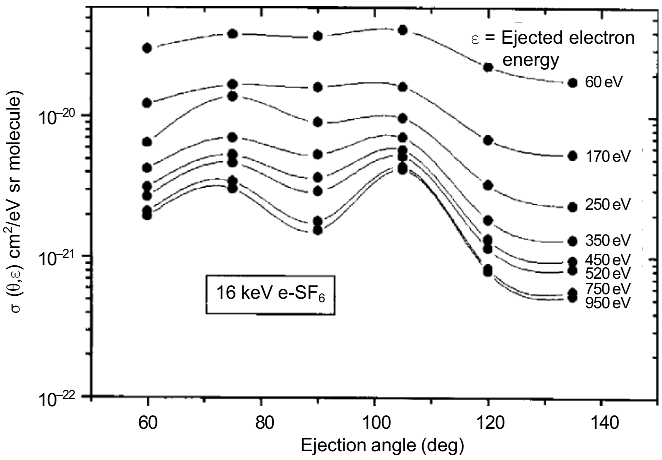

Figure 5. Angular distributions of ejected electrons from collisions of 16 keV electrons with SF_6 molecule. An oscillatory structure due to interference effect is seen for electrons having energies greater than 60 eV.

6. Electron backscattering from thick metallic targets

When energetic electrons collide with a solid metallic target, they lose their energy by a variety of mechanisms; some electrons are backscattered without any energy loss. This elastic electron backscattering process plays an important role in many experimental techniques, like, low energy electron diffraction (LEED), scanning electron microscopy (SEM), electron probe microanalysis (EPM), Auger electron spectroscopy (AES), electron lithography physics, radiation damage and elastic peak technique. Observable properties of backscattered electrons (BEs) consist of their absolute yield per incident electron (the BE coefficient η), their angular and energy distributions. It is clear that backscattering process must be sensitively determined by the details of the elastic scattering interaction but it is not clear as to what extent inelastic process determines the properties of the returning projectile. Even less is known about the interaction volume for the process; how deep does the typical electron penetrate before returning back to surface? More exact experimental information about the macroscopic effect, especially the energy and angular distribution of backscattered electrons from a thick solid target would be required to examine the validity of existing theories. It was considered, therefore, worthwhile to measure the backscattering coefficient and the energy distribution of electrons backscattered from a thick target at high impact energy of electrons. Such an experiment has been carried out in our laboratory for a thick target *W* under impact of 8 keV electrons. The energy range of backscattered electrons is considered between 70 eV to 1700 eV. The angle of incidence α and the take-off angle θ were chosen to have values α = 0°, 10° and 20° and θ = 110°, 120° and 130° respectively (Fig. 6). The energy distribution function exhibits two sharp peaks, which are found to appear at 216 eV and 548 eV. These are identified as Auger peaks of tungsten arising due to electron transitions 4*d*–6*s*6*p* and 4*s*–6*s*6*p*, respectively. The measured energy spectra are compared with two different theoretical models [11,12].

The energy distributions of backscattered electrons as a function of reduced impact energy ($\varepsilon = E/E_0$, where, E and E_0 are the energies of backscattered and that of incident electrons respectively) is found to increase with incidence angle α for a given value of ε. The predictions of two theoretical models are found to be in satisfactory agreement with our experiment [13].

7. Electron bremsstrahlung from thin- and thick targets

7.1. Bremsstrahlung spectra from thin (gas) targets :

When energetic electrons impact with target atoms, both characteristic- and non-characteristic X-rays are produced. The continuous part (bremsstrahlung) of the X-ray is found to extend upto the high photon energy limit (which is equal to the incident electron energy) with distinct characteristic lines of the target atom superimposed on the continuum, The production of characteristic X-rays from collisions between electrons and 'free' atoms is used as an important experimental tool to measure the ionization cross sections for inner shells of the target atoms and to test collision theories. The origin of the continuous part of X-ray spectrum is understood to lie in the process of acceleration or deceleration of the incident electrons in the Coulomb field of the target atom (molecule). Electron bremsstrahlung (EB) is the radiation of a photon emitted in the scattering of an electron from an atom and it is only one particular consequence of the general coupling of the electromagnetic field and matter fields. The process is closely related to both elastic electron scattering and to direct radiative recombination.

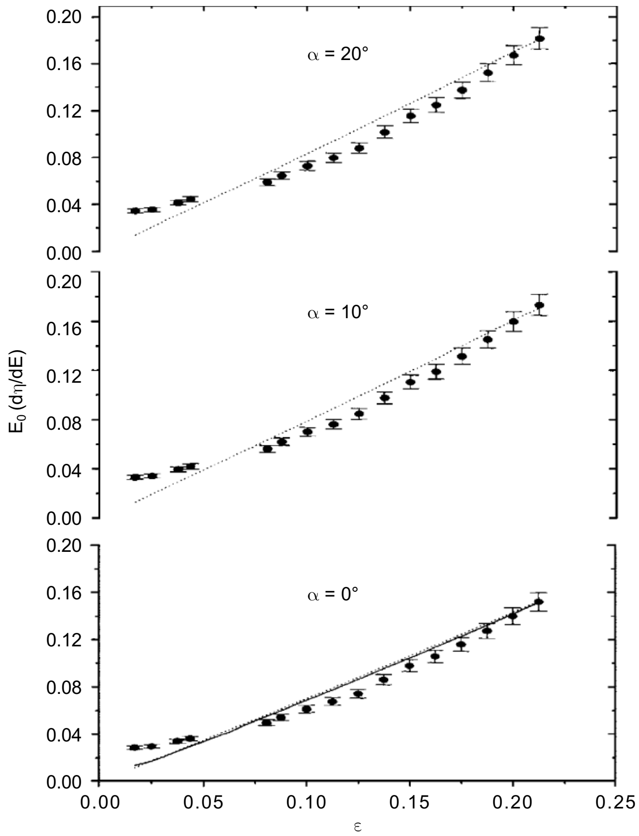

Figure 6. Energy distributions of backscattered electrons from a thick tungsten target under impact of 8-keV electrons as a function of $\varepsilon = E/E_0$ and angle of incidence α for integrated take-off angle θ. (•) : present experiment, the solid and the dotted line curves are the theoretical results [11,12].

The study of EB is of practical interest and importance in a variety of applications, for example, in interpreting astrophysical spectra, modelling hot plasmas [14], predicting plasma properties and estimating plasma parameters [15]. We have measured the emission cross sections of the characteristic and the non-characteristic X-rays produced from collisions of 10–24 keV electrons with argon atoms. The angular- and impact energy dependence of the characteristic K X-radiation of argon is found to show a net polarization of (17 ± 6)% in the investigated range of impact energy [16].

Further, the intensity of bremsstrahlung at different photon energies is measured using a photon energy window of 180 eV (k ± 0.18 keV), where, k refers to the photon energy. The data are normalized to the theoretical results of Kissel *et al* [17] at k/T = 0.5 and θ = 90°, where, T is the impact energy of electrons (Fig. 7). This figure shows a good agreement between our experimental data and the theoretical predictions from Kissel *et al* for 10 keV and 12 keV impact energies. For a detailed discussion of these results, see Ref. [18].

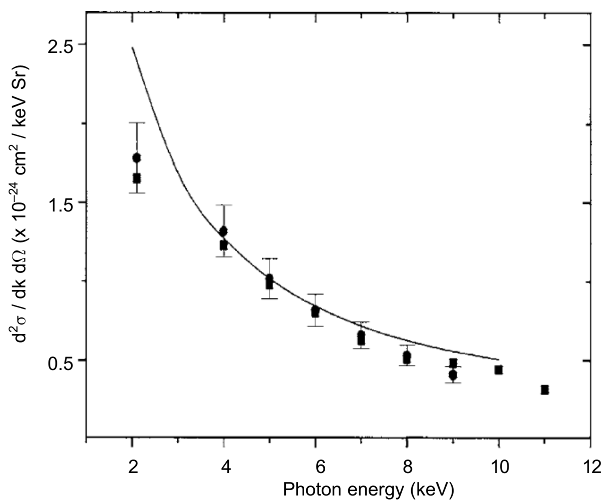

Figure 7. Double differential bremsstrahlung emission cross sections as a function of photon energy *k* produced by 10 keV and 12 keV electron impact with Ar. (•), (■), present data for 10 keV and 12 keV electron impact, respectively; ——, theoretical predictions from Kissel *et al* for 10 keV.

7.2. *Bremsstrahlung spectra from thick targets :*

The generation of X-rays from the impact of electrons with a thick target (*i.e.*, the target in which incident electrons finally come to rest) is a complex process. The production of EB is known to depend on the physical properties of the target and on the fundamental cross section for interaction of the impinging electrons with target atoms. On one hand, during interaction, the electrons lose their energy in the target mainly by the ionization process while on the other hand, the bremsstrahlung radiation is emitted from electrons having any energy less than or equal to the incident energy. The shape and intensity of the bremsstrahlung spectrum depend not only on the angle of X-ray emission but also on the scattering angle of electrons in the target. The secondary electrons that are produced in the ionization events can share the projectile electron's energy and radiate in subsequent interactions with the target. The X-rays produced in the target are self attenuated and their distance of travel depends on the location of the electron in the target while it radiates.

Measurements of X-ray emission spectra generated by 14 keV electrons impinging normally on a bulk target of Ti (Z = 22) have been carried out in our laboratory using an experimental setup described elsewhere [19]. Our data for bremsstrahlung yields as a function of impact energy have been compared with the simulation results using a general purpose Monte Carlo code PENELOPE [20]. This comparison as shown in Fig. 8 exhibits an overall agreement between experiment and theory; experiment is however, found to under estimate the simulation results below 2.5 keV; this mismatch may arise mainly due to the poorly known detection efficiency of the employed detector (Si-PIN photo diode) [21].

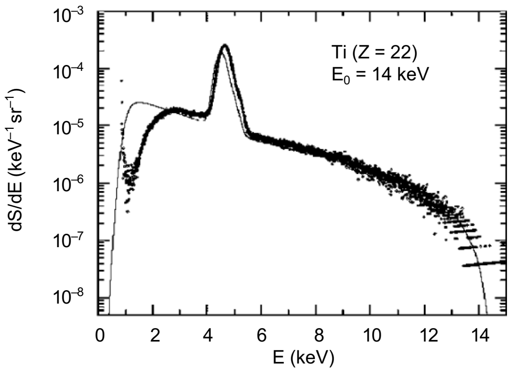

Figure 8. Comparison of X-ray spectrum produced from collisions of 14 keV electrons with Ti bulk target with calculations using the general purpose Monte Carlo simulation code PENELOPE. The solid line curve represents the results of simulation and the dots show the present experimental data. Shown on the *Y*-axis are intensities in absolute units.

8. Conclusions

It is shown that the Ar^+ ions are predominantly produced by direct ionization while Ar^{n+} are produced by Auger cascade and shake-off processes. The singly- and the multiply charged fragment species of atomic- and radical ions of SF_6 molecule are produced *via* Coulomb explosion process. Our measurements of Doppler shift in *F–K* Auger line confirm the fragmentation of SF_6 molecule *via* Coulomb explosion process.Energy- and angular distributions of S-L Auger and F-K Auger peaks are found to show their nature of being 'anisotropic' respectively. The data on energy- and angular distributions for electron backscattering from a pure thick Ti target are found to show a good agreement with the theoretical predictions. Furthermore, our study on Ti-K as well as on associated bremsstrahlung emission under keV electron impact shows a satisfactory agreement with the simulation results of PENELOPE code.

Acknowledgment

The works described in this paper have been financially supported by the Department of Science and Technology (DST), Government of India, New Delhi. The research fellowships awarded by the DST to R K Singh, S Mondal and R K Yadav during execution of the works are gratefully acknowledged. The valuable suggestions, comments and collaboration received from Profs. R Hippler and X Llovet are thankfully acknowledged.

References

[1] S Mondal and R Shanker *Phys. Rev.* **A72** 052705 (2005)

[2] J P Ravon *Nucl. Instr. Meth.* **211** 7 (1982)

[3] S Mondal and R Shanker *Phys. Rev.* **A72** 062721 (2005)

[4] R K Signh, R Hippler and R Shanker *Phys. Rev.* **A67** 022704 (2003)

[5] S Mondal and R Shanker *Phys. Rev.* **A69** 060701 (2004)

[6] E Kukk, H Aksela, S Aksela, F Gelmukhanov, H Agren and S Svensson *Phys. Rev. Lett.* **76** 3100 (1966)

[7] E M Snuder, S Wei, J Purnell, S A Buzza and A W Castleman (Jr.) *Chem. Phys. Lett.* **248** 1 (1996)

[8] S Mondal, R K Singh and R Shanker *J. Chem. Phys.* **124** 034301 (2006)

[9] W E Wilson and L H Toburen *Phys. Rev.* **A7** 1535 (1973)

[10] S Mondal and R Shanker *Phys. Rev.* **A69** 060701 (R) (2004)

[11] W S McAfee *J. Appl. Phys.* **47** 1179 (1976)

[12] P F Staub *J. Phys.* **D27** 1533 (1994)

[13] R K Yadav and R Shanker *Phys. Rev.* **A70** 052901 (2004)

[14] W H Tucker *Radiation Processes in Astrophysics* (Cambridge, MA : MIT Press) (1975)

[15] J C Weisheit *Atomic Phenomenon in Dense Plasma : Applied Atomic Collision Physics* (New York Academic) **Vol.2** p441 (1984)

[16] R K Singh and R Shanker *Phys. Rev.* **A67** 012708 (2003)

[17] L Kissel, C A Quarles and R H Pratt *At. Data Nucl. Data Tables* **28** 381 (1983)

[18] R K Singh and R Shanker *J. Phys. B : At. Mol. Opt. Phys.* **36** 3031 (2003)

[19] R Shanker *Radiation Physics and Chemistry* **75** 1176 (2006)

[20] E A Acosta, X Llovet and FSalvat *Appl. Phys. Lett.* **80** 3228 (2002)

[21] A N Agnihotri, V S Subrahmanyam, R K Yadav, X Llovet and R Shanker *J. Phys. D : Appl. Phys.* **41** 065205 (2008)