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Energy and angular distributions of electrons ejected from CH_4 and C_3H_8 under 16.0 keV electron impact

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Abstract. Relative cross sections, differential in energy and angle, for electrons ejected from CH_4 and C_3H_8 molecules under 16.0 keV electron impact have been measured. Electrons were analyzed by a 45° parallel plate electrostatic analyzer at emission angles varying from 60° to 135° with energies from 50 eV to 1000 eV. The angular distributions of electrons exhibit structures which are found to arise from Coulomb and non-Coulomb interactions. Furthermore, the double differential cross sections of electrons ejected from C_3H_8 molecule are found to be higher in magnitude than those from CH_4. This result supports the fact that the number of ejected electrons participating in collisions with C_3H_8 molecules is more than that in CH_4. Also, the angular distributions of C–K-shell Auger electrons emitted from the target molecules have been studied and shown to be isotropic within the experimental uncertainty.

Keywords. Double differential cross sections; Auger electrons; binary peak; polarization.

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1. Introduction

When a charged particle of high velocity collides with an atom or a molecule, ejection of one or more electrons from the target takes place. This process leads to the ionization of the bombarded particle. A number of different mechanisms have been identified by which the ionization of a target element takes place. Study of these mechanisms is of practical importance in a variety of areas, including plasma physics, radiation physics, atmospheric physics, astrophysics and in the study of penetration of charged particles through matter [1]. Earlier studies were directed to both theory and experiment for obtaining the total ionization cross sections [2], however, a little and incomplete information are available on the study of ionization mechanisms in molecules. The elucidation of several of these mechanisms has been based on detailed studies of the energy and angular distributions of ejected electrons from collisions between energetic electrons and the atoms/molecules. Since 1930s, several groups started studying the energy and angular distributions of ejected electrons both theoretically and experimentally [3–6]. While some interesting features were found in these studies, most of them were subjected to several defects particularly

on the experimental conditions. So, some refined techniques were required to obtain more reliable results. Over the last few decades, a number of measurements of the energy and angular distributions of ejected electrons from interaction of energetic charged particles with atom/molecule have been performed using more refined experimental techniques [7–11]. However, these studies have not provided experimental data for collisions of high keV electrons with targets, in particular with molecules. Further, in order to understand the phenomena of many electron ejection processes and to interpret them in light of the available theoretical models, more extensive data on energetic electron–atom/molecule collisions are needed. In this effort, the present work is part of a program to produce secondary electrons by ionizing some light hydrocarbon molecules by keV electron impact and to determine energy and angular distributions of the ejected electrons from such collisions.

Secondary electrons produced in electron–atom/molecule collisions are composed of the primary electrons scattered by atoms/molecules and those of ejected electrons from the target. Electrons ejected in the interaction are generally found to originate from direct and indirect ionization processes. Those ejected in the direct ionization process (called 'δ-electrons') have a continuous energy distribution corresponding to the continuum states in the atoms or the molecules, while the electrons indirectly ejected from the highly excited states of target atoms/molecules have the sharp energy spectra. Typical examples of the indirect ionization are the 'auto-ionization' and the 'Auger process'. Sharp discrete lines are believed to arise from the auto-ionization and the Auger processes additively superimposed on the continuous spectra of δ-electrons.

The study of energy and angular distributions of ejected electrons from atoms and molecules finds importance from different aspects: firstly, the scalability of molecular cross sections [12] shows that the weakly bound electrons dominantly take part in the ionization of molecules. This result is further confirmed by the comparison between the total ionization cross sections and the innershell ionization cross sections (see e.g., Lynch *et al* [13]). Secondly, the angular distributions of direct ionization cross sections give a dynamical details of electron–atom/molecule interaction [14,15]. Occurrence of a peak on the angular dependence curve of double differential cross sections corresponds to the binary encounter (BE) peak, which is discussed in the literature [16]. Thirdly, the angular dependence of innershell ionization cross sections of the Auger peak sheds light on the polarization and alignment of the initial state with which the peak is associated. Because of the poor detection and transmission efficiencies of our electron energy analyzer, the detection of low energy electrons ($E_e < 50$ eV) was relatively less reliable and no analysis of such electrons has been included in the analysis. Normally, the auto-ionization peaks are found to arise in the low-energy region of the energy distributions. These peaks are, however, not observed in the present measurements. A detailed and systematic study of the electron energy spectra arising due to indirect ionization, for example, due to the Auger process will be discussed later in §4.3. In this paper, we present and discuss the experimental results of energy and angular distributions of continuous electrons resulting due to the direct ionization process and of characteristic (Auger) electrons due to indirect ionization process from methane (CH_4) and propane (C_3H_8) molecules by the impact of 16.0 keV electrons.

2. Experimental procedures

Measurement of the cross sections differential in energy and angle for emission of electrons from methane (CH_4) and propane (C_3H_8) by the impact of 16.0 keV electrons has

been carried out on a newly developed experimental set-up in our laboratory. The set-up essentially consists of a vacuum system for evacuating a compact scattering chamber as well as an electron gun housing in a differential mode, a 45°-parallel plate electrostatic energy analyzer (PPEA), a Faraday cup for beam monitoring and a data acquisition system. Since, the details of design, fabrication, assembly, optimization and test results of different components of the experimental set-up are described elsewhere [17], we shall give here only a brief description of the components of direct relevance in the present measurements. A schematic diagram of the present experimental set-up is shown in figure 1.

The scattering chamber used for the study of cross sections differential in energy and angle of ejected electrons from interaction of 16.0 keV electrons with molecules was evacuated by a TM pump (240 l/s) and a fore pump, while the electron gun enclosure was evacuated by a combination of a small TM pump (60 l/s) and a fore pump. Two compact full range dual gauge heads: PKR 251 (Pirani and cold cathode gauges) coupled with their respective control units monitored the pressures in scattering chamber and in electron gun housing simultaneously. The base pressure of the scattering chamber and that of the electron gun enclosure without a target gas load was found to be better than 1.6×10^{-6} Torr. During measurements, the target gas pressure was maintained at 4.4×10^{-4} Torr to ensure a 'single collision' condition.

Electron gun used in the present experiment is a custom built unit and it provides a focused electron beam with energy accuracy of much less than 1% at the considered energy. The electron beam spot size has been found to lie between 50 μm and 3 mm depending upon the working distance (10–500 cm) from the mounting flange of the electron gun. The beam was monitored on a biased (-60 V) Faraday cup after it was transmitted through the target. A well-defined beam of the target was obtained by effusing the molecular gas at a thermal velocity through a multicapillary tube. Each capillary has a length of 5 mm and internal diameter of 0.05 mm. Energy distributions of electrons ejected with energies from

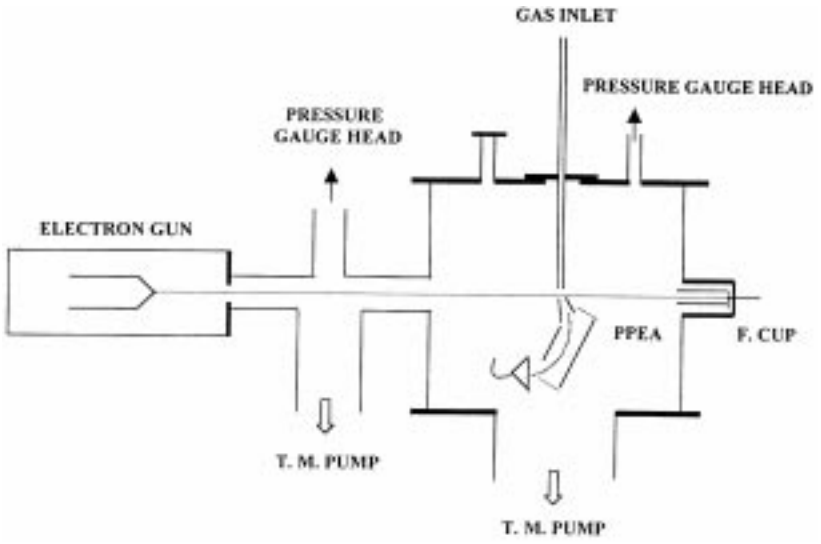

Figure 1. Schematic diagram of the experimental set-up.

50 eV to 1000 eV in impact of 16 keV electrons with target molecules were obtained by recording the spectra at a chosen angle of their emission with the help of a 45°-parallel plate electrostatic analyzer (FWHM: $\Delta E/E = 12\%$) equipped with a channel electron multiplier (CEM). Detection of ejected electrons at different angles was facilitated by keeping the electrostatic analyzer at a chosen angle with respect to the incident beam direction. A remote control device for the rotation of PPEA around the collision centre made it possible to put the PPEA at a desired angle with an angular accuracy of $\pm 1°$.

3. Data acquisition and reduction

Electrons of different energies were detected by a CEM operated in a pulse counting mode. The energy spectra of the ejected electrons as a function of their energy for different emission angles (60°–135°) with respect to the incident electron beam direction were recorded in a multichannel-scaling (MCS) mode of a Pentium-based 4K channel multichannel analyzer (MCA). The physical limitation of the scattering chamber prevented from recording of the electron energy spectra at extreme lower and higher angles. After subtracting the background counts arising from the residual gas, the numbers of actual ejected electrons from the target gas were ascertained. The number (N_e) of electrons ejected from the gas target at a particular electron energy (ε) and at a given emission angles (θ) was converted to the corresponding relative cross sections, differential in emission angle and ejection energy by using the equation

$$\frac{d^2\sigma}{d\varepsilon\, d\Omega} = \sigma(\theta,\varepsilon) = \frac{N_e e^{\alpha P x}}{N_p P\, ds \Delta E\, 3.23 \times 10^{16}} \text{cm}^2/\text{eV}\cdot\text{sr}\cdot\text{molecules} \qquad (1)$$

where N_p is the number of projectile electrons registered by the Faraday cup, P the target gas pressure in Torr, ΔE the energy spread of the electrons transmitted through the analyzer, ds the product of the solid angle subtended by the analyzer and the electron path length observed within the solid angle, α the absorption coefficient for electrons of energy ε in the target gas and x the distance traveled by electrons of a given energy from their birth place to the detector. Taking the typical values of α for electrons in CH_4 and C_3H_8 from the work of Brode [18] and the experimental values of pressure P and x, the term $e^{\alpha P x}$ was calculated. As the transmission efficiency of our PPEA is poorly ($< 10\%$) known for low energy electrons, no effort was made to analyze the energy spectra of ejected electrons below 50 eV. Further, since in our case, the 'scattering volume' is formed by an overlap of the electron beam ($\Phi = 3$ mm) and that of the gas beam effusing from a multicapillary tube ($\Phi = 5$ mm), the scattering zone is not strictly a 'point scattering centre'; it was, therefore, necessary to apply a '$\sin\theta$-correction factor' to the data of angular distributions. All energy spectra of ejected electrons were finally treated with the transmission and dispersion corrections of the analyzer.

4. Results and discussion

4.1 *Energy distributions*

The relative double differential cross sections (DDCS) of ejected electrons from CH_4 and C_3H_8 molecules as a function of ejection energy by the impact of 16.0 keV electrons were

measured and the results are displayed in figures 2 and 3 respectively. The energy distribution of ejected electrons covers the energy range from 50 eV to 1000 eV for different angles of emission. These distributions of double differential cross sections are found to vary by about three decades of magnitude over a wide range of angles from 60° to 135° for both CH_4 and C_3H_8 molecules. The experimental uncertainty in the measurements of relative DDCS is estimated to be about 23% as the square root of the sum of squares of the individual uncertainties. This uncertainty basically stems from the uncertainties in ds, E, N_e and N_p (see eq. (1)) whose individual contributions are estimated to be 19, 12, 2 and 0.2% respectively.

The general features of both sets of the curves shown in figures 2 and 3 are found to be similar. The qualitative shape of the curves resembles the energy distributions of ejected electrons from CH_4, NH_3, SF_6 and TeF_6 by 0.25–2.0 MeV proton impact (see for example, Toburen *et al* [12] and Lynch *et al* [13]). The velocity of protons at highest energy used by Toburen *et al* is however much lower compared to our electron velocity. So there is no possibility to compare their cross sections with ours at least in the case of CH_4. The prominent structures of these spectra exhibit a sharp peak at a relatively low electron energy for all measured angles and a broad maximum at a large energy and for a small ejection angle. The broad maximum in the high-energy region is believed to arise from the binary encounter events between the projectile electrons and the orbital electrons of the target molecules. The broad energy distribution arises due to the binding of the orbital electrons of target molecules and has been discussed in detail in §4.2. The sharp peak appearing in

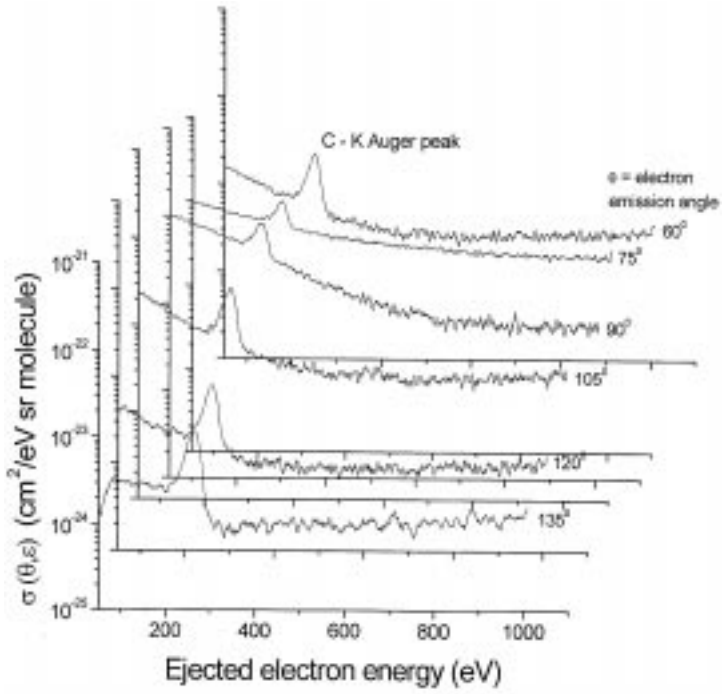

Figure 2. Double differential cross sections (DDCS) of ejected electrons from CH_4 as a function of ejection energy by the impact of 16.0 keV electrons.

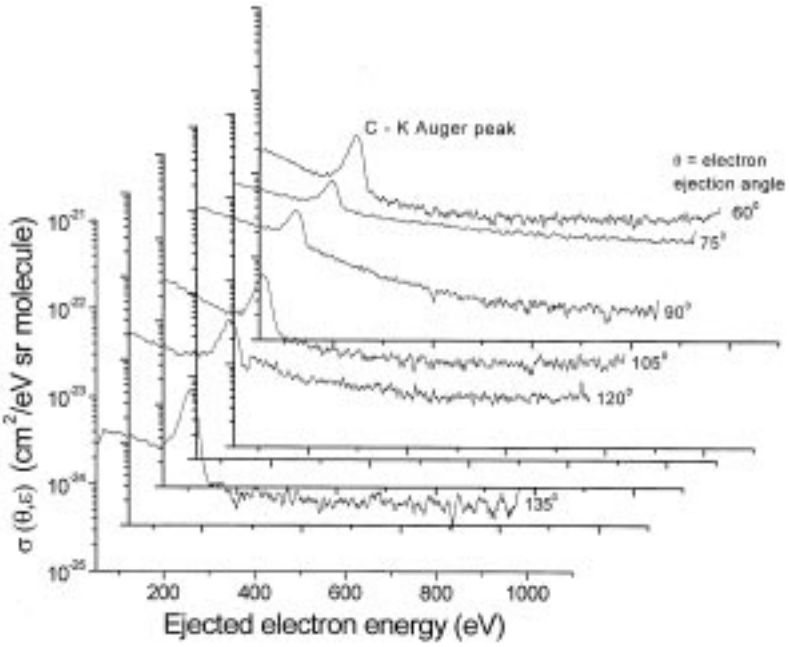

Figure 3. Same as given in figure 2 except for C_3H_8 molecule.

the low energy region arises due to the carbon K-shell Auger transitions at about 250 eV in the target molecules.

4.2 *Angular distributions*

The variation of double differential cross sections of ejected electrons per molecule from CH_4 and C_3H_8 by 16.0 keV electron impact as a function of ejection angle θ, is shown in figures 4 and 5 respectively for different electron energies. A qualitative variation of the intensity of electrons ejected from CH_4 and C_3H_8 at a particular electron energy is similar. Emission cross sections of low energy electrons (less than hundred eV) are much more nearly isotropic for both the considered hydrocarbon molecules. Quantitative comparisons of angular distributions have been made for ejected electrons from CH_4 and C_3H_8 and are shown in figure 6. Comparisons for three selected energies of ejected electrons (75 eV, 350 eV and 850 eV) in the entire angular range show that cross sections of C_3H_8 are larger than the corresponding cross sections of CH_4 molecule. This result suggests that larger numbers of electrons are taking part in ionization events in C_3H_8 than in CH_4 molecule. Further, figures 4 and 5 exhibit two broad peak structures in the DDCS function, one in the forward direction ($\theta < 90^\circ$) and other in the backward direction ($\theta > 90^\circ$). The occurrence of these peaks can be understood by knowing the nature of the initial and final states of the interacting particles as it is discussed in the last paragraph of this section. The variation of DDCS peak with the energy of ejected electrons in the forward direction shows that the cross sections for ejected electrons of low energy appear with a broad maximum.

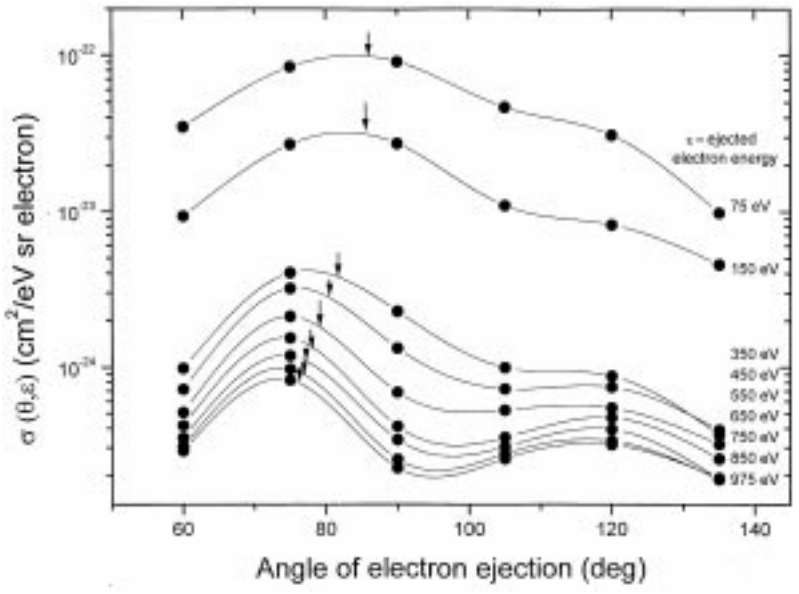

Figure 4. Angular distributions of electrons of selected energies ejected in 16.0 keV electron–CH_4 collisions. Arrows shown in the figure refer to the peak positions as calculated from the binary encounter theory [19].

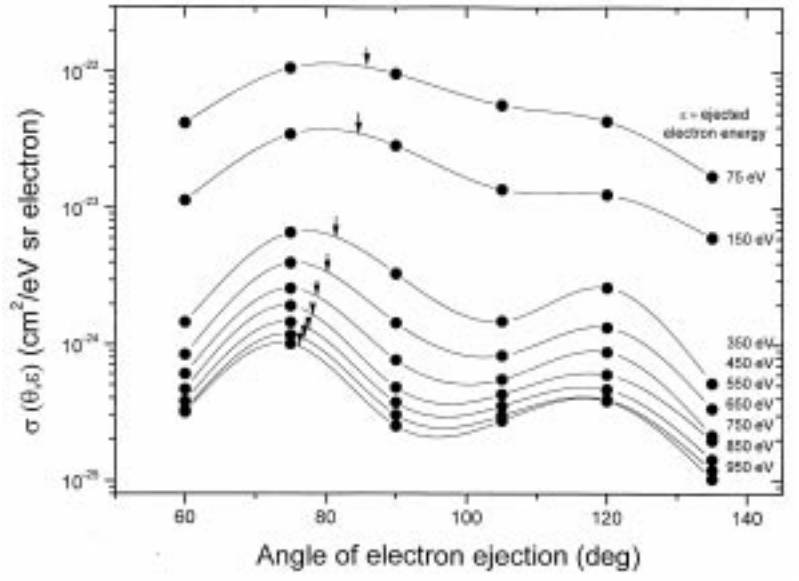

Figure 5. Same as given in figure 4 except for C_3H_8 molecule.

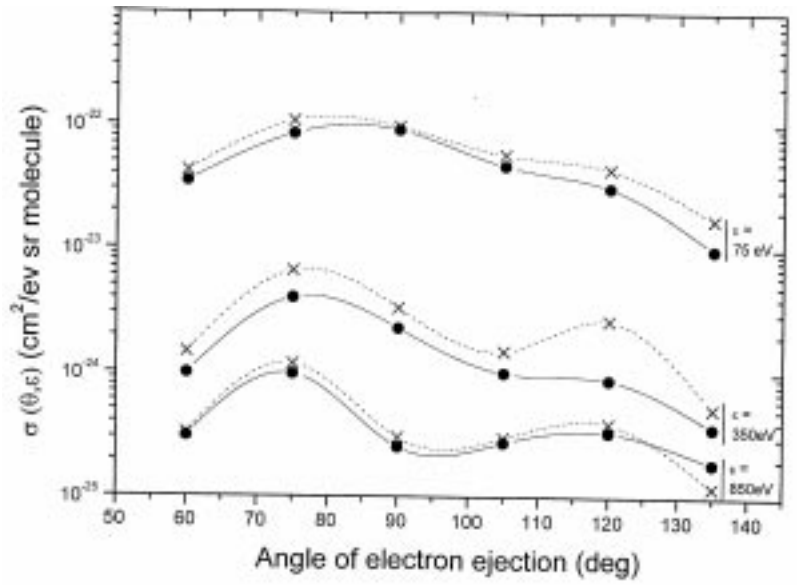

Figure 6. Comparison of DDCS of ejected electrons of selected energies from C_3H_8 ($\times$) and from CH_4 ($\bullet$) by 16.0 keV electron impact.

However, as the energy of ejected electrons increases, not only the broad maximum becomes more pronounced but also it moves in the forward direction. This maximum is often referred to as 'binary encounter peak'. The peak formation of DDCS in the forward direction can be explained by the following binary encounter mechanism. For collisions between a fast incident electron and an electron at rest, corresponding to a given amount of energy transfer, the electron goes off in a well-defined direction giving a δ-function angular distribution. If the target electron is initially bound, the binding provides its initial momentum distribution, i.e., velocity distribution in the target atom. This momentum distribution causes a change in the relative velocity of the incident and the target electrons. Consequently the relative velocities give the possibility of ejection of electrons in a broad angular range and 'smear out' the δ-function distribution of the ejected electrons. For the low momentum transfer being comparable to the binding energy of the target electrons, the low energy electrons belonging mostly to the outer shells are ejected out. For the case of momentum transfer much larger compared to the binding energy of the target electrons, the inner shell electrons are ejected out more in the forward direction compared to the low energy electrons of the outer shells along the direction of the momentum transfer. As the ejected electron energy increases, the binary peak moves in the forward direction (see figures 4 and 5). Furthermore, the high energy electrons ejected from the inner shells have initially smaller velocity-spread compared to that of the outer shell electrons. This low velocity spread reduces further the smearing out effect causing the peak to be sharper in the forward direction. This classical picture of the collision forms the basis for the binary encounter approximation (BEA) model for theoretical calculations [19]. This model is found to give a fairly reliable estimate of the angular distributions of DDCS of the ejected electrons in the forward direction.

The relation between the velocity $\vec{V}_P$ of the incident particle and the momentum transfer $\vec{P}$ by it to the target atom/molecule can be obtained by using the relation from Banks *et al* [19],

$$\hat{V}_P \cdot \hat{P} = \frac{2m_1E + P^2}{2m_1V_PP} \tag{2}$$

where E is the energy transfer by the projectile to the target atom/molecule and m_1 is the mass of the incident particle, $\hat{V}_P$ and $\hat{P}$ are the respective unit vectors of projectile's and of the momentum transfer. If θ_{BP} refers to the angle at which binary encounter (BE) peak arises, then, from eq. (2), one obtains

$$\theta_{BP} = \cos^{-1}\left(\frac{V_e}{V_P}\right) \tag{3}$$

where $\vec{V}_e$ is velocity of the ejected electron. The angle at which the BE peak occurs can be calculated using eq. (3) for our collision systems. The positions of the binary peak are found to be in close agreement with our data (see arrows in figures 4 and 5). For instance, positions of the calculated and the experimental binary peaks in 16.0 keV electron–CH_4 collisions for ejected electron energy of 975 eV are determined to be at 76° and 75° respectively. The angular uncertainty of the present measurements is estimated to be ±1°. In addition, the binary encounter peaks are found to shift towards higher angles with decreasing energy of the ejected electrons. Further, the cross sections are seen to decrease rapidly at angles greater than the binary peaks.

The behavior of the double differential cross sections for ejected electrons of higher energies at backward angles show another maximum at about 120°; this feature is considered to be one of the characteristics of BEA results [20,21]. In fact, our experimental results for angular distributions of DDCS of ejected electrons produced from 16.0 keV electron impact with CH_4 and C_3H_8 show such a maximum at about 120° (see figures 4 and 5). The occurrence of this peak at a large angle in our results can be explained in the following way: In a charged particle impact ionization process, the ionized electron with principal quantum number n and orbital quantum number l undergoes a transition from a bound state nl to a continuum state $\varepsilon l'$. This process, unlike the dipole transitions, has no selection rules on the final momentum l'. Thus, the ejected electron can be in εs, εp, εd, εf, εg etc. final continuum states. These continuum waves interfere with each other and give a final angular distribution. So the relative phases of each pair of continuum waves are important for DDCS. If all of the partial waves are in phase and interfere constructively, one gets the 'binary encounter' peak which essentially arises in the forward direction as of the present case discussed earlier. This is analogous to the central maximum in a diffraction pattern. At higher angles, this interference is no longer totally constructive due to the variation of phases of continuum waves with angle, and decreases rapidly. This leads to the rapid fall off of the DDCS with increasing angle beyond the BE peak. Eventually, an angle is reached where the values of phases of various partial waves interact with each other such that they interfere destructively that results to give much like the first minimum in the diffraction pattern. At still larger angles, the DDCS increases like the diffraction pattern intensity going to the second maximum, the backward angle maximum. Hence the position of the minimum and backward angle maximum of DDCS depends very sensitively on the relative phases of the continuum partial waves as a function of ejected electron energy. In the ejection of

electrons from interaction of 'low-impact energy' of electrons with atoms/molecules, the Coulomb interaction plays a dominant role. In this type of interaction, the binary encounter between the incident electrons and the orbital electrons of the target becomes semi-classical which incorporates only the Coulomb phase-shift and, thus no constructive phase relation occurs at large angles and consequently, DDCS does not show a peak at larger angles. Low energy (eV to a low keV) electron impact ionization of various atoms/molecules does not show a DDCS peak at larger angles [22]. However, the high keV electron–atom/molecule collision processes include the Coulomb interaction and also the non-Coulomb interaction giving a dissimilar nature of the variation of DDCS for ejection of electrons than that of a low energy electron–atom/molecule collisions. All these interactions change the phase of the continuum partial waves as a function of ejected electron energy. Modification of these continuum partial wave phases yields a constructive DDCS at large angles in addition to the BE maximum at low angles. The theoretical binary encounter approximation being a semi-classical model does not include the Coulomb and the non-Coulomb phase shift effects and, thus it cannot be expected to represent the DDCS accurately at large angles where they become important [20]. Also, the Born-approximation calculations include only the Coulomb phase shift and hence, they neglect the non-Coulomb phase shift. These calculations are not suitable to give the DDCS at the backward angles accurately [23,24]. The calculations, employing the Born-approximation that includes both the Coulomb and non-Coulomb phase shifts with Hartree–Slater initial discrete and final continuum wave functions are found to give a fairly well DDCS at the backward angles [16] and illustrate the importance of the non-Coulomb phase shifts. At present, no calculations are available to take account of Coulomb shifts in polyatomic molecules, such as, CH_4 and C_3H_8.

4.3 *C–K Auger peak and ionization cross sections*

Characteristic peaks observed in the energy distributions of ejected electrons from CH_4 and C_3H_8 molecules by the impact of 16.0 keV electrons correspond to the C–K Auger peaks appearing at about 250 eV (see figures 2 and 3). The innershell ionization cross sections of C–K-shell differential in energy and angle can be estimated from these spectra. Since the fluorescence yield of C–K-shell is very small (0.3%) in comparison to the Auger yield ($> 99\%$), one can equate the innershell ionization cross sections directly to the total cross sections for Auger electron production. Auger electron cross sections differential in energy and angle were obtained by the integration of C–K peak areas after the subtraction of counts contributed by continuum electrons. We have also studied the angular distributions of Auger electrons of C–K-shell in this work. Polarization P (or alignment) of the C–K-shell Auger electrons is obtained by measuring the intensity of the peak at different angles, and it is determined by using the following relation:

$$I(\theta)/I(90^\circ) = (1 - P\cos^2\theta) \tag{4}$$

where $I(\theta)$ and $I(90^\circ)$ are the intensities of Auger electrons ejected at an angle θ and at 90° respectively with respect to the incident beam direction. Ratio of the Auger electron intensities measured at different angles, $I(\theta)/I(90^\circ)$, were determined and plotted as a function of $\cos^2\theta$ (see figures 7 and 8) for CH_4 and C_3H_8 molecules respectively. The estimated experimental uncertainty for $I(\theta)/I(90^\circ)$ is about 11% as shown on each data

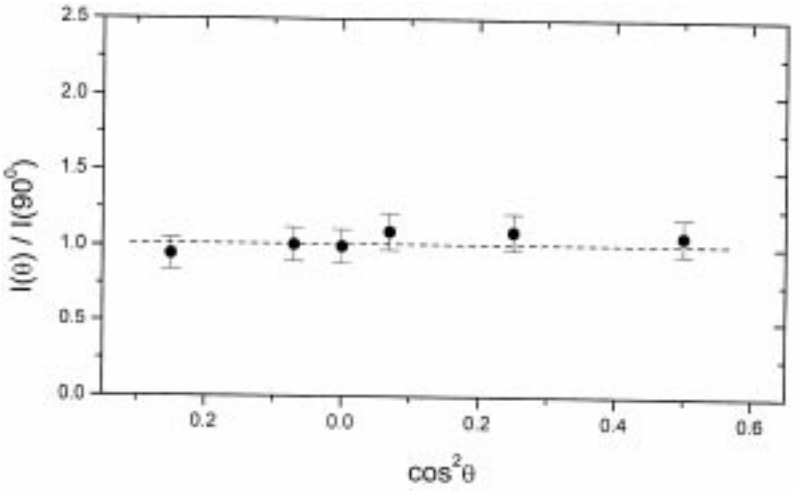

Figure 7. Angular variation of intensity ratios $I(\theta)/I(90^\circ)$ for Auger electrons emitted from C–K-shell in 16.0 keV electron–CH_4 collisions.

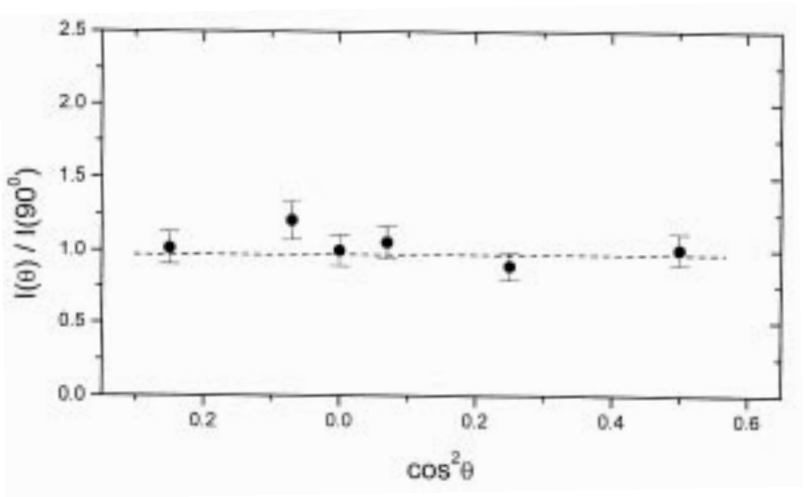

Figure 8. Same as given in figure 7 except for C_3H_8 molecule.

points with corresponding error bars. The nature of variation of the data points as a function of $\cos^2\theta$ shows that all the experimental points follow a straight line with a zero slope within the experimental uncertainty. This indicates that the carbon K-shell Auger electrons ejected from CH_4 and C_3H_8 molecules follow an isotropic distributions under the impact of 16.0 keV electrons.

5. Conclusions

Double differential cross sections of ejected electrons, differential in energy and emission angle have been measured following the interaction of 16.0 keV electrons with CH_4 and C_3H_8 molecules. These measurements were made for ejected electrons having energies between 50 eV and 1000 eV; their emission angle ranged from 60° to 135°. Due to the poor detection and transmission efficiencies of our electrostatic analyzer for low energy electrons, analysis of electrons having energies less than 50 eV was not included.

Qualitative shape of the energy distributions of ejected electrons exhibits a sharp peak of C–K Auger electrons in the low energy region (at about 250 eV) for all ejection angles and a broad maximum of the continuum high energy electrons at intermediate ejection angles. These features are found to be similar for ejected electrons from both CH_4 and C_3H_8 molecules. The binary encounter events occurring due to interaction between the projectile electrons and the orbital electrons of the target molecules result into a broad maximum for the high energy ejected electrons. Emission cross sections of low energy electrons (less than hundred eV) are much more nearly isotropic for both the considered hydrocarbon molecules. The relative magnitude of the double differential cross sections for C_3H_8 is found to be larger than the corresponding cross sections for CH_4, which suggests that relatively a larger number of electrons are available for interaction in C_3H_8 than in CH_4. Both the Coulomb and non-Coulomb interactions are found to be necessary for interpreting the broad peak seen at higher angles in the present collision systems.

Innershell ionization cross sections for carbon K-shell vacancy production in both CH_4 and C_3H_8 molecules were obtained from Auger electron yields. These cross sections are found to exhibit an isotropic distribution of Auger electrons within the experimental uncertainty.

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