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Observation of interference effects in ejected electrons from 16.0-keV e^--SF_6 collisions

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The relative energy- and angle- dependent cross sections for emission of electrons from SF_6 molecule by impact of 16 keV electrons have been measured. The angular distributions of ejected electrons are shown to exhibit an oscillatory structure which is suggested to arise due to an interference effect. The condition for interference effect for the present collision system has been examined and it is shown that the appearance of interference pattern takes place above a threshold energy of 65 eV for the ejected electrons. The ejected electrons producing an interference structure are suggested to originate from two atomic centers of a transiently formed doubly ionized parent molecule, namely, SF_6^{2+}. This extremely unstable ion suffers a Coulomb explosion and gives rise to many singly charged stable radical and atomic ions. The time of flight mass spectrometric results of our earlier work [Phys. Rev. A **67**, 022704 (2003)] on partial ionization of SF_6 molecule by impact of 16 keV electrons are found to support the existence of these stable ions.

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Ionization processes in molecular collisions with energetic charged particles provide an ideal test ground to describe the fundamental interactions occurring in a multiparticle system. These interactions are found to play an important role in astrophysics, plasma physics, thermonuclear fusion, and in studies of surface and materials [1]. In the present work an effort is made to understand the ionization processes occurring in molecules by impact of energetic electrons. Study of energy- and angular distributions of ejected electrons from molecules by impact of energetic charged particles is one of the commonly used techniques to shed light on ionization processes in such collisions [2–5]. Depending on the projectile's 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 [6,7]. The soft collision at high projectile energies can be attributed to three body collisions [8]. These fast projectiles may be regarded as a source of virtual photons which gives rise to the dipole transitions involving the transfer of a unit angular momentum. The violent collisions at low projectile velocities give rise to binary encounter collision due to a large momentum transfer of the projectile to the target electrons. A general understanding of the electron ejection processes in collisions with low velocity charged particles with simple diatomic molecules is available [9–14], wherein a "binary collision process" plays a dominant role. When momentum transfer by the incident particle to the target electrons assumes a minimum value, the peaking approximation in the dipole interaction is generally applied. Under these conditions, the ejected electrons carry the momentum transfer in the backward direction in order to conserve the total momentum of the collision system. On one hand, 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. [6], 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. On other hand, as the binary encounter process involves a large momentum transfer to a "single" individual electron of the target and produces a peak structure in the forward direction $(\theta<\pi/2)$ in the angular distributions, obviously, it is not expected to produce two coherent electrons to cause an interference effect.

60 MeV/u Kr^{34+}-ion-induced interference effect has been observed experimentally by Stolterfoht *et al.* [6] in the double differential ionization cross sections of ejected electrons from H_2 molecule. In their work, they have shown that an oscillatory pattern on the energy distributions of the double differential cross sections of the ejected electrons arises due to the interference effect and that it is independent of the ejection angle of the electrons. They have been able to see this oscillatory pattern when they divided their double differential cross sections by the corresponding theoretical results of Fainstein *et al.* [15]. In contrary to their own observation, recently Stolterfoht *et al.* [16] have shown that the double differential ionization cross section of H_2 molecule by impact of 68-MeV/u Kr^{33+} ions follows an oscillatory pattern due to the interference effect among the ejected electrons from the target. This pattern was found to vary with the ejection angle of the electrons in agreement with their own theoretical predictions [16].

The origin of interference effect of electrons ejected from a diatomic molecule can be explained by considering the fact that two atomic centers in a diatomic molecule represent a double slit assembly. When electrons of same energy are ejected simultaneously from two atomic centers separated by a finite distance $\mathbf{R}_0$, they may interfere with each other, giving rise to an oscillatory structure in their angular distributions. Such an electron interference pattern may be compared with Young's double slit experiment. The visibility of such an interference effect has been discussed in detail by Walter

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and Briggs [17]. When two electrons are ejected with a momentum $\mathbf{k}$ from each of two atomic centers of a diatomic molecule having a nuclear separation $\mathbf{R}_0$, they will see the nuclear splitting only when $\mathbf{k} > \mathbf{R}_0^{-1}$, i.e., when the de Broglie wavelength associated with the two electrons is smaller than $\mathbf{R}_0$; otherwise, they will see essentially the two atomic centers as one point. This consideration suggests that the ejected electrons of high energy associated with smaller values of de Broglie wavelengths are expected to show an interference structure more favorably in their angular distributions than those of a low energy. In order to explain the interference effect observed for the ejected electrons from two centers of a polyatomic molecule SF_6 in the present experiment, we determine the double differential ionization cross sections (DDCS) similar to the double ionization of a H_2 molecule by one photon which are given by an expression following the model of Walter and Briggs [17] as

$$\frac{d\sigma}{d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{K}} = \frac{4\pi^2 \alpha h}{E} |T_{\mathrm{fi}}|^2, \tag{1}$$

where α is the fine structure constant, E is the incident energy of the projectile, $\mathbf{k}_1$ and $\mathbf{k}_2$ are the momenta of two ejected electrons relative to the molecular center of mass and $\mathbf{K}$ is the relative nuclear momentum. $|T_{\mathrm{fi}}|$ is the Born-Oppenheimer transition dipole matrix element including both nuclear and electronic parts. The angular distribution of ejected electrons is governed by the electronic part of T_{fi} and it is proportional to $\hat{\boldsymbol{\varepsilon}}(\mathbf{k}_1+\mathbf{k}_2)2\cos[\frac{1}{2}(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}]$ (see Ref. [17]). Hence, in the angular distribution, the term $\cos^2[\frac{1}{2}(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}]$ gives an oscillatory structure.

In the case of a multiple ionization of the symmetric polyatomic molecules, the momenta of two electrons ejected from two dissimilar constituent atoms of the molecule may add coherently to give rise to an interference effect. A large number of experimental works have been devoted to the ionization of diatomic molecules [9–12] and of polyatomic molecules [13,14] by charged particle impact, however, no obvious signature of interference effect in ejected electrons from polyatomic molecules has ever been reported in the literature. In this communication, we report for the first time an oscillatory structure in the angular distribution of electrons ejected from a polyatomic SF_6 molecule under the impact of 16 keV electrons. This structure is attributed to an interference effect of the electrons emitted from such collisions.

The present experiment was carried out in our laboratory using a 50 keV electron-atom and electron-molecule collision experimental facility [18]. A detailed description of the scattering chamber and that of the electron spectrometer has been given elsewhere [19]. The collisionally induced continuum and characteristic electrons emitted from a target gas of SF_6 in the energy range of 40–1000 eV by impact of 16 keV electrons were measured. The ejected electrons were energy analyzed by a 45° parallel plate electrostatic analyzer (FWHM ~ 12%) equipped with a channel electron multiplier which could be positioned around the collision center at different angles of emission (60°–135°). The energy spectra of the ejected electrons as a function of their energy for different emission angles with respect to incident beam direction were recorded in a multichannel scaling (MCS) mode of a Pentium based 4K multi-channel analyzer (MCA). Relative double differential cross sections for electron emission were determined by using a method similar to that described in our previous work [19]. The experimental uncertainty in the measurements of relative DDCS is estimated to be about 20%.

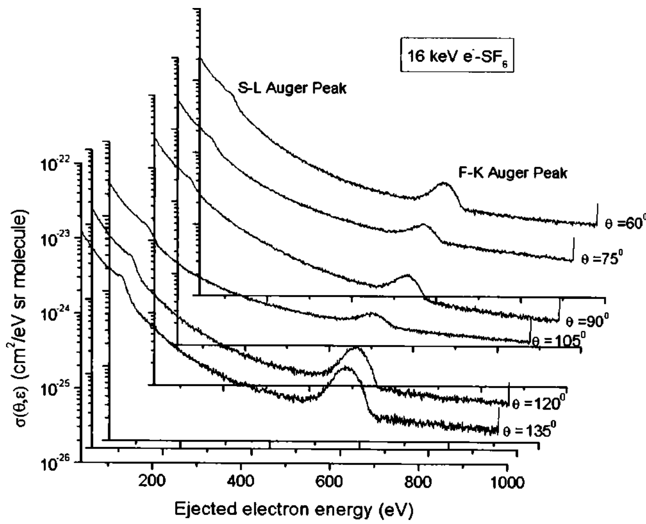

FIG. 1. Relative double differential cross sections of ejected electrons from SF_6 by impact of 16 keV electron as a function of ejection energy.

The relative double differential cross sections of electrons ejected from 16 keV e^--SF_6 collisions have been measured as a function of energy and their emission angles. Figure 1 shows that magnitudes of these relative electron emission cross sections are different for different angles of emission. The energy of ejected electrons varied from 40 to 1000 eV, while their emission angles varied from 60° to 135°. Two distinct peaks are found to arise in each spectrum of energy distributions: one at about 120 eV and the other at 630 eV. These peaks are attributed respectively to S-L and F-K shell Auger transitions.

Angular distributions of electrons ejected with different energies are shown in Fig. 2, which exhibit an oscillatory structure. This structure becomes more pronounced as the energy of the ejected electrons is increased. It is noted that this oscillatory structure is well outside the experimental uncertainties of the relative cross sections which are typically about 20%. Such a structure shows two peaks: one in the forward direction ($\theta < \pi/2$) and the other in the backward direction ($\pi/2 < \theta < \pi$), where θ is the angle of ejection of electrons with respect to the direction of the incident beam. The peak observed, in the forward direction, viz., at $\theta \sim 75°$, is believed to arise from the "binary encounter" events between the projectile electrons and the orbital electrons of the target molecule. The position of the binary encounter peak appearing at angle θ_{BP}, can be determined by using a relation [20]

$$\theta_{\mathrm{BP}} = \cos^{-1}\left(\frac{v_e}{v_p}\right), \tag{2}$$

where v_e and v_p are the velocities of the ejected and the projectile electrons respectively. Using the values of the

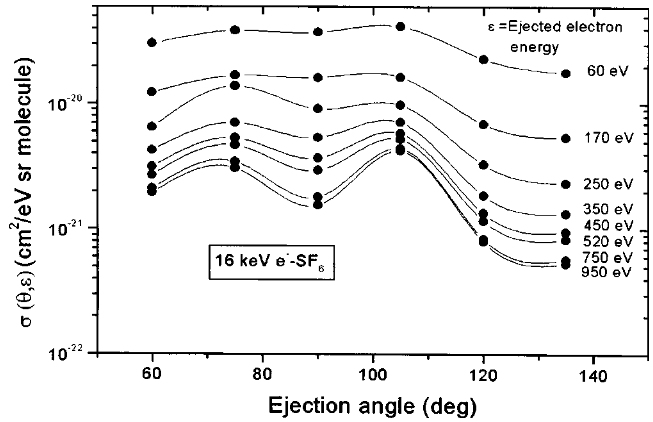

FIG. 2. Angular distributions of electrons of selected energies ejected from collisions of 16 keV electrons with SF_6 molecule.

ejected electron's velocity corresponding to energy of 950 eV and that of the incident electron for our collision system, the value of θ_{BP} is estimated to be 76°. The present experimental binary encounter peak is found to lie at 75°. These two values are found to be in a good agreement within the experimental uncertainty of angular measurements (±1°).

The peak appearing at a backward angle, that is, at $\theta \sim 105°$ in the oscillatory structure, may be explained by considering the cause of interference effect produced by two electrons ejected simultaneously during the collision from two atomic centers of a doubly charged molecular ion of SF_6 by impact of 16 keV electrons with SF_6 molecules as discussed in the following paragraphs. Similar angular distribution of ejected electrons from SF_6 molecule was measured by Toburen *et al.* [14] for proton impact with energy of 0.3–1.8 MeV. However, their spectra did not show up any peak at a backward angle. Recalling the condition for appearance of interference effect, a peak appears in the backward angle on the angular distribution due to interference effect caused by zero momentum transfer of the fast charge particle (say $v_p \sim 50$ a.u.) [6] to the target electrons. It may be noted that in the present experiment, the 16 keV incident electrons correspond to a velocity of 38 a.u., whereas the proton projectile's velocity used by Toburen *et al.* [14] corresponds to only 8.4 a.u. At such a low impact velocity, the occurrence of interference structure in the angular distribution of ejected electrons seems to be improbable.

From Fig. 2, it is also seen that the oscillatory structure does not occur for electrons of very low energies, say, 60 eV. The electrons ejected with energies of 170 eV or greater show an oscillatory peak which becomes more pronounced as the energy of ejected electrons is increased to higher values. Also, another condition for appearance of an interference effect for the ejected electrons is that the wavelength associated with them should be of the order of or less than the internuclear distance of the considered molecule from which they are ejected. In the present case, the electrons are considered to be ejected from two atomic centers, namely, of S, and that of F of a SF_6 molecule (S–F bond length =1.54 Å), the interference effect in the spectrum of angular distribution of ejected electrons, is expected to occur for those electrons which have energies larger than 65 eV.

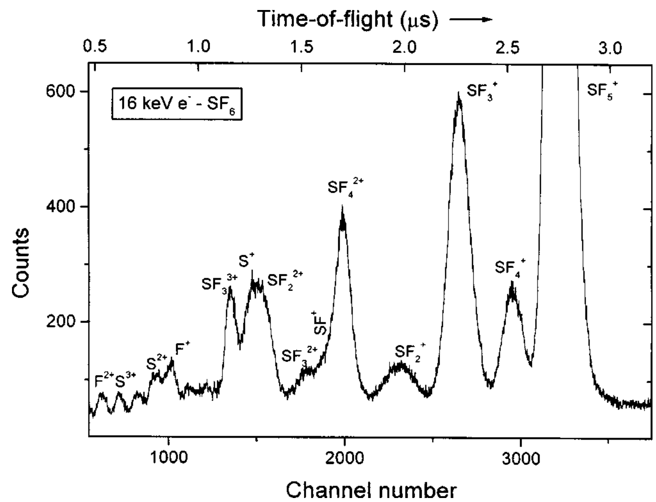

FIG. 3. Time of flight spectrum of the SF_6 ionic fragments produced in 16 keV electron–SF_6 collisions (Ref. [21]).

Hence, the peak observed in the backward angle in the angular distributions for electrons having energies 170 eV or above is suggested to arise from interference effect.

The interference effect observed in the angular distributions of ejected electrons from SF_6 molecules is qualitatively explained as follows: The two interfering electrons are considered to originate from two atomic centers of a SF_6 molecule during its double dissociative ionization (DDI) events. The DDI processes responsible to yield different stable radical ions of SF_6 molecule are supported by the findings of the work of Singh *et al.* [21]. In their work, they have obtained a time of flight (TOF) spectrum of various radical ions produced in 16 keV e^--SF_6 collisions (as shown in Fig. 3). The TOF spectrum was recorded by observing the coincidences between the produced fragment ions and the simultaneously ejected electrons of indiscriminated energies at an angle of 90° with respect to the incident electron beam direction in a single collision condition. During the collision, it is suggested that an extremely unstable SF_6^{2+} radical ion is formed for a very short time, much less than picosecond, by impact of 16 keV electrons with the SF_6 molecule. It may be pointed out here that during the ionic fragmentation of SF_6, ionization of the parent molecule takes place first before it gets dissociated [22]. The weak S–F bond is known to have a binding energy of about 4.38 eV. The time required to break this bond is of the order of picoseconds [23,24]. When the SF_6^{2+} ion is formed, it gets dissociated immediately via a Coulomb explosion (CE) yielding different stable fragment ions, namely, SF_5^+, SF_4^+, SF_3^+, SF_2^+, SF^+, S^+, and F^+ following the first reaction channel:

$$SF_6 + e_i^- \rightarrow SF_6^{2+} + 2e_{ej}^- + e_s^- \xrightarrow{\text{CE}} SF_m^+ + F^+ + (n-1)F + 2e_{ej}^- \tag{3a}$$

and in the second reaction channel, SF_4^{2+}, SF_3^{2+}, SF_2^{2+} and SF^{2+} ions are formed:

$$SF_6 + e_i^- \rightarrow SF_6^{2+} + 2e_{ej}^- + e_s^- \xrightarrow{CE} SF_m^{2+} + nF + 2e_{ej}^-, \quad (3b)$$

where, e_i^-, e_{ej}^- and e_s^- are incident, ejected and primary scattered electrons, respectively, $m+n=6$. It is to be noted that the double ionization of constituent S and F atoms of SF_6 molecule requires a larger amount of energy (the ionization potentials of S^{2+} and F^{2+} are 33.70 and 52.39 eV, respectively) than for their simultaneous single ionizations (the ionization potentials of S^+ and F^+ are 10.36 and 17.42 eV, respectively). Hence, before the S–F bond of the SF_6 molecule is broken, the transient formation of SF_6^{2+} ions takes place in a most favored channel by simultaneous ejection of two electrons: one from the S atom and the other from the F atom attached to S–F bond of a SF_6 molecule representing a double- slit system. This conjecture is also supported by Eq. (3a). These two electrons interfere with each other after they are emitted from a double-slit-like system. The pairs of electrons ejected from S and F atoms in creation of the SF_6^{2+} ion give interference pattern when they fulfill the condition for a constructive interference for a particular orientation between $\mathbf{R}$ and $\mathbf{k}_1$ and $\mathbf{k}_2$. It is also observed from Eq. (3b) that in a less favored channel the unstable SF_6^{2+} ion after Coulomb explosion yields different doubly charged ions ejecting two electrons from the S atom of SF_6 alone (see explanation in Ref. [21]). These ejected electrons do not take part in the formation of an interference pattern because they originate from a single center of the S atom during formation of various ions. The existence of stable fragment ions mentioned above, viz., SF_m^+ ($m=0-5$) and F^+; SF_m^{2+} ($m=0-5$) ions, can be verified and identified from the TOF spectrum (see Fig. 3). In the second channel, the $SF^{2+}$and SF_5^{2+} ions are however not observed in the TOF spectrum; this may be due to either their low probability of occurrence or due to their very short life times. It may be mentioned here that, though the formation of stable ions, viz. SF_5^+, SF_3^+, SF_4^{2+}, SF_2^{2+}, and S^{2+}, is well explained by a hybrid model (see Ref. [21]), it can not explain the formation of SF_4^+, SF_2^+, SF^+, S^+, and SF_3^{2+} ions, however. In contrast, the double dissociative ionization and multi-electron dissociative ionization mechanism explain the formation of all these ions.

In conclusion, we have observed for the first time an oscillatory structure in the angular distributions of electrons ejected from SF_6 under the collisions of 16 keV electrons. This oscillatory structure provides an evidence for an interference effect occurring between two electrons simultaneously ejected from two centers of the constituent atoms of SF_6 molecule. For the present collision system, no theoretical calculations are available to predict the interference structure on the energy or angular distribution of the ejected electrons. However, a theoretical model for double ionization of a diatomic molecule by photon impact qualitatively supports the occurrence of oscillatory structure for the observed angular distributions of ejected electrons.

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