Young-type interference effect on angular distribution of secondary electrons emitted from H_2 in collisions with fast electrons

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The Young-type interference arising due to the spatial coherence has been investigated in the electron emission spectrum from fast electron impact ionization of the inversion symmetric homonuclear diatomic molecule H_2. The evidence of the interference effect in the angular distribution of the double differential spectrum of the secondary electron is found. The signature of constructive interferences has been identified in the soft-collision regions as well as in binary encounters. The observed oscillation in the forward-backward asymmetry parameter is explained in terms of the Cohen-Fano-type interference coupled with the angular dependence of oscillation frequency. A comparative study indicates a marked difference between the angular asymmetry in the case of fast heavy ion (F^{9+}) and electron collisions with H_2 at a similar velocity.

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Young-type interference phenomena observed with light as well as with a single electron in a double-slit experiment [1,2] have been of crucial importance in the foundation of quantum mechanics. An atomic scale alternative of such a double-slit experiment could be feasible by looking into electron emission from a two-center homonuclear molecule, excited by a photon or fast charged particle. It was predicted about 40 years ago by Cohen and Fano in photoionization [3] and was first observed only recently in the electron spectra resulting from the ionization of molecular hydrogen by GeV energy heavy ions in a pioneering investigation by Stolterfoht *et al.* [4] and by MeV energy heavy ions [5–7]. The oscillations observed in the ratio of double differential cross section (DDCS) of H_2 and 2H, as a function of electron energy, i.e., de Broglie wavelength of electrons was termed as the signature of Young-type interference. Such ratios were fully measured by Misra *et al.* [5] and Tribedi *et al.* [8,9] by measuring the DDCS of H_2 as well as atomic H. Since then, there has been a steady flow of new investigations of this process using different projectiles as probes. In photoionization processes, interference patterns were theoretically investigated for H_2^+ and H_2 targets [10–13]. Also, the effect was studied for the first time on core electrons of N_2 by Rolles *et al.* [14], later on by Liu *et al.* [15], and recently by Kreidi *et al.* [16]. In the two former experiments, emphasis was given to study the symmetry of sites from where coherent electron emission leads to the interference effect. The importance of the interference studies on the fundamental quantum mechanics was revealed through the very recent study of double photoionization of H_2 [17] which focused on the quantum to classical transition in an entangled system by observing the loss of coherence. The second-order process in such electron interference has also been predicted and observed [18,19] in fast-ion collisions.

Electrons as projectiles are fundamentally different from heavy ions because they are dimensionless, negatively charged, light mass particles with larger de Broglie wavelengths at identical velocities. Therefore, electron impact dynamics is principally different from highly charged heavy ions and, as shown below, can provide a better benchmark system to study interference oscillation in the asymmetry parameter. The existence of interference effects has been predicted theoretically for $(e,2e)$ reactions involving H_2 as the target [20]. Evidence of interference patterns in the cross-section differential in the final energy and polar angle of the emitted electron was shown in an e^-+D_2 experiment [21]. The effect became also evident from the structure of triple differential cross section (TDCS) measured recently [22,23] (see also Ref. [24]).

So far the interference effect, in ionic collisions, has been investigated in the DDCS-ratio spectrum (H_2-to-2H) as a function of electron emission velocity which showed oscillatory structure. It may be noted that in Young's two-slit experiment the variation of intensity of light was observed at different positions on the screen (i.e., at different observation angles) which was due to constructive and destructive interferences. The corresponding situation in ion-molecule collision will require the measurement of electron intensity oscillation as a function of electron emission angle, which has not been reported so far (except TDCS in *e*-2*e*-type experiments [22,23]). In this paper, we show evidence of the interference effect in the electron DDCS spectrum as a function of emission angle and demonstrate that the constructive interference prevails in the soft-collision regions as well as the electrons emitted in binary encounters. This investigation, thereby, can be viewed as a more direct comparison with Young's double-slit experiment.

Apart from the methods of deriving interference pattern from the ratio of molecular DDCS to that of the atomic one, another way to look into it comes from the analysis of the angular asymmetry of molecular DDCS, which is derived

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"only" from molecular DDCS and is free from any normalization. This procedure, which was introduced by Misra *et al.* [25], can be used self-consistently to study the interference [25–28]. In heavy-ion collisions, it is known that the long-range Coulomb interaction of the final state electrons with the target and high projectile charge influences the evolution of the electron wave function and thereby the angular distribution [29–35]. Such a two-center effect is known to cause a forward focusing of electron emission resulting in a large forward-backward asymmetry in the emission spectrum [28–35]. However, for electrons as projectiles, the behavior of the asymmetry parameter, which describes the angular asymmetry, is not known. Here, we have performed a comparative study of interference patterns derived from the asymmetry parameter using bare fluorine ions (F^{9+}) and electrons as projectiles colliding with H_2 at comparable velocities.

The measurements were carried out for (i) an 8 keV ($v_p \sim 24$ a.u.) electron beam obtained from a commercially obtained e-gun and (ii) a 95 MeV ($v_p \sim 14$ a.u.) F^{9+} beam (obtained from the BARC-TIFR Pelletron accelerator facility) colliding on H_2 and He gaseous targets. In both types of experiments the electron beam as well as ion beams were highly collimated by using several apertures. The emitted electrons were detected using an electron spectrometer equipped with a hemispherical electrostatic energy analyzer and a channel electron multiplier. The energy resolution of the spectrometer was about 6% of the electron energy, limited by the entrance and exit apertures. Experiments were done by flooding the chamber with target gas keeping pressure of 0.15 mTorr for ejected electron energies 1–40 eV. For higher energy electrons the gas pressure was 0.3 mTorr. The front and exit apertures of the spectrometer were biased to small voltages of +6 V in order to help the lowest energy electrons be detected. Background pressure was kept at 1×10^{-7} Torr. The energy and angular distributions of the electron DDCS were derived from the electron counts suitably normalized using the known experimental parameters and geometry. The DDCSs were studied for different angles ranging from 30° to 150° and for electron energies between 1 and 500 eV. Further details of the experimental setup are described in [28,37].

Figures 1(a) and 1(b) show the measured absolute DDCS of electrons emitted in 8 keV $e^- + H_2$ collision at angles 45° and 135°, respectively. Full curves are representing theoretical molecular DDCSs calculated with two-effective center (TEC) approximation, where the ionization of one of the target electrons may be considered as produced preferably from the vicinity of either molecular center, whereas the other electron screens completely the nucleus from which ionization is not produced. A detailed discussion on this theory is given in Refs. [20,21]. A good agreement between TEC calculations and measurements is obtained. Also in the same figures, the dashed lines represent calculations corresponding to two effective H atomic centers, where an effective hydrogen with an effective nuclear charge is equal to the one used in the Heitler-London wave function and an effective energy equal to the molecular bound energy has been used [20,21]. Experimental DDCSs divided by 2 times the theoretical cross sections for two effective H atoms, as a function of the ejection velocity, are shown in Figs. 1(c) and 1(d), respectively. These ratios yield first-order oscillations due to interference. The dashed lines denote complete theoretical ratios. However, in the ratio spectra there are some discrepancies between the experiment and theory with respect to the amplitude and phase. This may indicate that a better approximation is required for the molecular wave function. A fitting is done to the ratio plots using a function $a+bk+f\sin(cdk)/cdk$ denoted by solid lines in Figs. 1(c) and 1(d), where a, b, f, and c are the adjustable parameters. The quantity k is the ejected electron momentum and d is the internuclear separation (1.41 a.u. for H_2). The linear function $a+bk$ takes care of the increasing discrepancy (with energy) between the theory and experiment in the DDCS level. The parameter c represents the frequency of oscillation. In accordance with previous studies, we observe angular dependence of the frequency of oscillation. As it is apparent in Fig. 1, the frequency of the backward angle (135°) is higher than the complementary forward angle (45°). The experimental-to-theoretical ratio drops below 1 a.u. The origin of such fall may be due to electron correlation and/or the screening effect. Indeed, at low ejection momentum, the ejected electron is more sensitive to details of the potential near the nuclei [12]. However, this effect is not included in the current theory. Relative statistical uncertainties of the ratio spectra are ranging from 5%–10% below an ejection velocity of 4 a.u. and higher above that velocity.

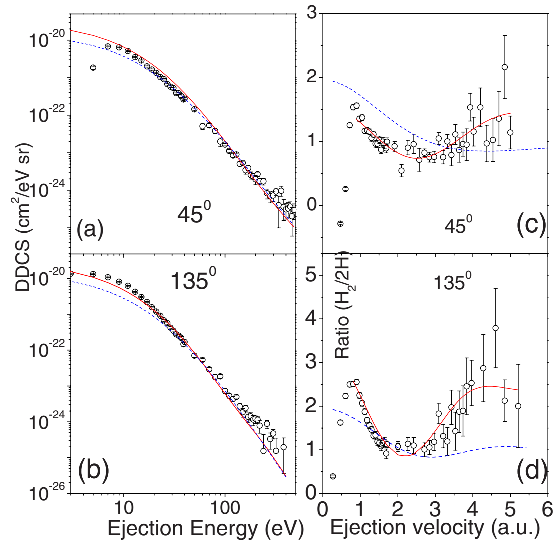

FIG. 1. (Color online) (a), (b) DDCSs of electrons emitted in 8 keV $e^- + H_2$ collision for 45° and 135°, respectively. Solid lines, theoretical molecular cross sections; and dashed lines, 2 times the theoretical cross sections for two effective H atoms. In (c) and (d) open circles represent experimental-to-theoretical DDCS ratios (H_2-to-2H), dashed lines represent theoretical DDCS ratios; solid lines, the first-order Cohen-Fano model fitting.

In Figs. 2(a)–2(c) the experimental DDCS angular distributions (symbols) are plotted along with the theoretical predictions for molecular H_2 (solid lines) and 2 times the two effective H atomic cross sections (dashed lines) for fixed emission energies of 9, 70, and 280 eV, respectively. An overall good agreement is found between theoretical molecu-

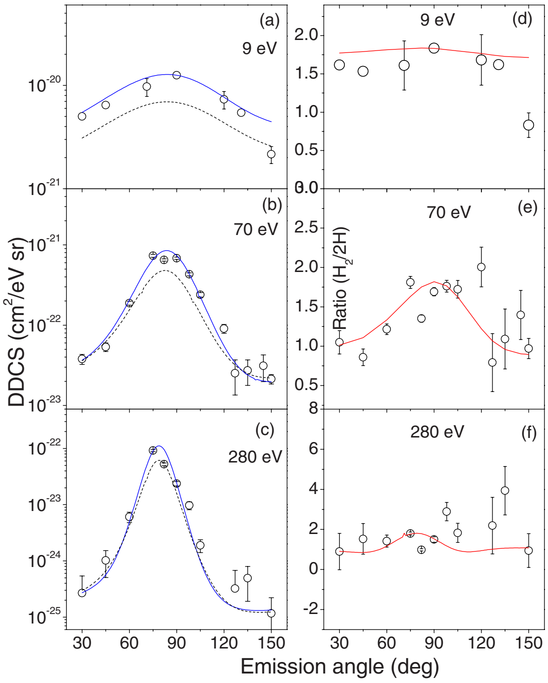

FIG. 2. (Color online) (a), (b), and (c) show angular distributions of secondary electrons emitted in 8 keV e^-+H_2 collision with energies 9, 70, and 280 eV, respectively. Solid lines, theoretical molecular cross sections; dashed lines, 2 times the theoretical cross sections for two effective H atoms. (d), (e), and (f) show experimental-to-theoretical DDCS ratios for respective emission energies. Solid lines are theoretical ratios. In (a) and (d) the typical absolute errors are shown for a few points. For the rest of the figures, only statistical errors are displayed.

lar cross sections and the experimental ones for a wide range of energies and within the angular range of 30°–150°. The apparent differences and crossover of atomic and molecular cross sections at various angles are due to constructive or destructive interferences. For 9 eV emission energy the atomic cross sections underestimate experimental molecular cross sections by about 30%–50%, whereas the absolute uncertainty of experimental data is within 20%. For 70 eV emission energy in binary-encounter regions the atomic cross sections underestimate the experimental molecular one by almost 40%–45%, whereas the absolute uncertainty of experimental data is again within 20%, statistical uncertainty being small, i.e., about 4%–6%. Similarly, for 280 eV emission energy and for emission angle around 90° (i.e., for the binary encounter region) the atomic cross sections underestimate experimental data points by 30%–40% which is again larger than the errors in the data. For example, in the case of 9 eV and emission angle 90° the difference is about 5.7×10^{-21} cm^2/eV sr. whereas the uncertainty is 2.5×10^{-21} cm^2/eV sr which is generally valid for other energies also. However, it may be seen from Figs. 2(e) and 2(f) that the statistical errors are quite large in the case of backward angles and for large emission energies.

This shows that the interference effect caused by coherent emission from molecular H_2 modulates the incoherent part of the DDCS in the angular distributions. In order to quantify the effect more closely, we have taken the ratio of experimental DDCS of H_2 and 2 times the theoretical cross sections for two effective H atoms. The experimental-to-theoretical ratios are displayed by symbols in Figs. 2(d)–2(f) along with the theoretical ratios in solid lines. In the soft-collision regions, i.e., at small emission velocities, where the momentum of the ionized electron and the momentum transfer are very small, the theory predicts a purely constructive interference. For example, at 9 eV emission energy, the experimental ratio comes close to the theoretical ratio by a factor of 1.8, supporting the presence of constructive interference. The same behavior also appears in the binary collision regions around 90°, where all of the momentum is transferred to the secondary electrons. For example, in Figs. 2(b) and 2(c) the theoretical atomic cross sections (dashed lines) underestimate the molecular cross sections (both in experiment and theory) in the binary-encounter regions, implying the presence of constructive interference. Our experimental observation is reinforced by theory in the respective ratio spectra [Figs. 2(e) and 2(f)]. Interestingly, the constructive interference remains even for larger emission velocities, as it is apparent for 280 eV. In the moderate energies above 60 eV, the theoretical atomic cross sections overestimate the theoretical molecular ones by a few percent in the extreme forward and backward angles, implying a destructive interference in these regions. In Figs. 2(e) and 2(f) the crossovers between the molecular and 2 times the atomic cross sections, signifying the destructive interference, are seen at around 30° and 130° for 70 eV [Fig. 2(e)], and around 55° and 100° for 280 eV [Fig. 2(f)]. However, with the current experimental uncertainty this minor effect is difficult to resolve.

Besides first-order effects derived from the molecular to atomic cross-section ratios, the interference mechanism has been investigated in the forward-backward asymmetry parameter of the electrons emitted from He and H_2 targets. The angular distribution of the electron emission reflects the ionization mechanisms in ion-atom or electron-atom collisions. For example, in the case of heavy ion-atom collisions a large forward focusing is well known which gives rise to a large forward-backward asymmetry in electron emission. This large asymmetry is caused due to the two-center effect and post-collision interaction between the projectile and the electrons in the final state. The additional details of the physics of this process can be found in [25–28,35,36]. The asymmetry parameter can be defined as [25,35]

$$\alpha(k) = [\sigma(\theta,k) - \sigma(\pi-\theta,k)]/[\sigma(\theta,k) + \sigma(\pi-\theta,k)], \quad (1)$$

where σ refers to the DDCS, θ is small forward angle (closer to zero degrees), and k is the ejection velocity. For electron impact, the asymmetry parameter $\alpha(k)$ derived from the molecular DDCSs for emission angles 30° and 150° is plotted as a function of ejection velocity in Fig. 3(a). For molecular H_2, $\alpha(k)$ shows a clear oscillation (open circles). This behavior is in sharp contrast to the corresponding data for He (solid squares), which shows a smooth linear behavior. Such oscillation may stem from the difference of frequencies of first-order oscillations of the two complementary angles as explained earlier in Ref. [25] for heavy-ion impact. Theoretical prediction [displayed in the inset of Fig. 3(a)] shows an agreement regarding phase and frequency of the oscillation.

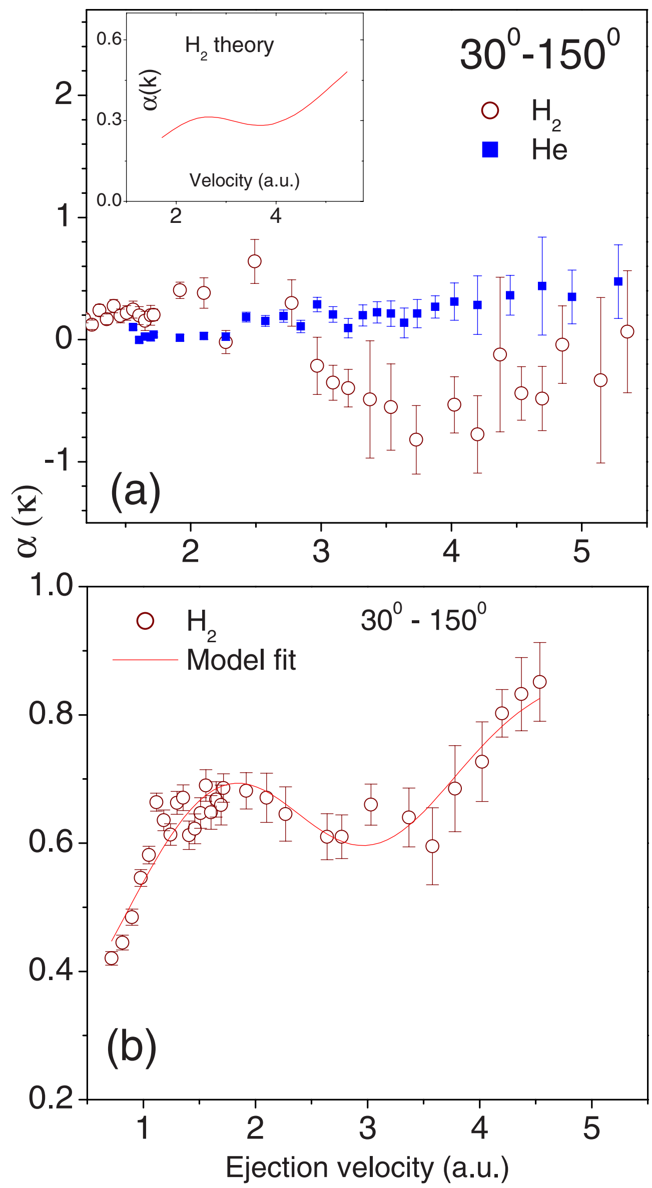

FIG. 3. (Color online) (a) The asymmetry parameter for 8 keV electron collision with H_2 (open circles) and He (squares). In the inset, the solid line is theory for H_2 showing the oscillation in $\alpha(k)$. (b) The same for 5 MeV/u F^{9+} collisions with H_2 including a model fit in the solid line (to be presented elsewhere). In both (a) and (b) we are considering two complementary forward-backward angles, namely 30° and 150°, respectively.

Hence, in this electron impact study, the oscillation of the asymmetry parameter of H_2 around merely a straight line corresponding to He reveals another facet of Young-type interference produced by two identical scattering centers.

In order to make a comparative study with heavy-ion collision, we kept identical the experimental setup and carried out another measurement with 5 MeV/u F^{9+} projectile ions colliding on H_2. For this measurement, the asymmetry parameter plotted in Fig. 3(b) shows an oscillation starting at a value α of about 0.4. However, the oscillation is seen to be around a straight line with a steep slope—in contrast to the observation for the electron projectile for which the oscillation is found around a horizontal line at zero, thereby giving positive and negative asymmetry. The steep slope for heavy-ion collision on H_2 could be attributed to the post-collisional two-center effect [25,34,35] where the influence of the field of the highly charged positive ions on the final state of the ejected electron contributes toward a continuous increment of the yield in the forward direction compared to the backward angles. The effect increases with increasing ejection velocity in the velocity range considered. Therefore, the asymmetry parameter for H_2 reveals the combined effect of two-center post-collision and interference effect. Using the Cohen-Fano-type model for the interference and considering the observed frequency difference in forward-backward angles a good fitting is obtained (see [25] for model) for heavy-ion data [Fig. 3(b)] (the details are to be presented elsewhere). For the one-center He target one observes [25] only the monotonically increasing function with a slope similar to that for H_2 (not shown here) in sharp contrast to the almost horizontal behavior in the case of electron collision [Fig. 3(a)]. The oscillations about a horizontal line in the asymmetry parameter for electron collision on H_2 is mainly caused by interference and is almost free from the two-center effect, thereby providing a benchmark system to study the interference.

In conclusion, in our very first observation, we have shown evidence of interference effect in the DDCS spectra as a function of emission angle which closely resembles the Young's double-slit experiment. Our observation supports the theoretical prediction of the presence of constructive interferences for the electrons emitted in soft and binary collisions. The signature of Young-type interference is found in the asymmetry parameter $\alpha(k)$ derived from the "only" molecular DDCS of the two complementary angles 30° and 150°, in collisions of H_2 with fast electrons. The phase and frequency of the oscillation of $\alpha(k)$ is supported by the theoretical prediction. A comparison is made with heavy-ion collision, where the post-collisional two-center effect leads to a stronger slope change of $\alpha(k)$ with increasing ejection velocity, contrary to the electron collision. Additionally, $e^- + H_2$ collision reveals a horizontal oscillation of $\alpha(k)$ about zero with positive and negative asymmetry.

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