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Influence of Young-type interference on the forward-backward asymmetry in electron emission from H_2 in collisions with 80-MeV bare C ions

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We use the forward-backward angular asymmetry in the electron emission cross sections in fast ion impact ionization of H_2 as a probe of the inversion symmetric coherence in homonuclear diatomic molecules. The electron energy dependence of the asymmetry parameter for H_2 exhibits oscillatory structure due to Young-type interference in contrast to atomic targets such as He. The asymmetry parameter technique provides a self-normalized method to reveal the interference oscillation independent of theoretical models and complementary measurements on atomic H target.

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Angular distribution of various types of radiations (particles and photons) is known to be quite sensitive to various effects associated with different physical processes in atomic, nuclear, plasma physics and other branches of physics. In fast ion-atom ionization, the long range Coulomb interaction of the final state electrons with the target and the projectile ions influences the evolution of the electron wave function and thereby the angular distribution of electron emission. Such two-center effect is known to cause a large forward-backward asymmetry [1–4] in the electron emission spectrum. The electron emission spectrum from the simplest diatomic molecule H_2 manifests yet another important aspect of interference [5] in ion-atom ionization besides the well-known mechanisms such as soft collision, two-center effect and binary encounter [1–4,6–8]. Since the two indistinguishable H atoms in the H_2 molecule may be considered as the coherent emission sources of phase coupled electrons in a large impact parameter collision, their contributions add coherently and an interference effect should be observed. Therefore, the electron emission from H_2 may be viewed as a natural coherent system which is similar to Young's double slit interference phenomenon [5]. We demonstrate here that the additional mechanism of Young-type interference plays a major role in the angular asymmetry of electron double differential cross section (DDCS) and asymmetry parameter itself would be a sensitive test to study the interference for a diatomic molecular target.

Following the initial theoretical studies on the interference effect in electron scattering [9] and photoionization [5], very recently the evidence of Young-type interference was found in the fast-ion collisions with H_2 [10–12]. Ideally one would have expected an oscillation in the DDCS spectrum due to interference. But a steep fall of the DDCS by about four or five orders of magnitude (see below) does not allow one to observe the oscillation directly. The oscillations, thereby, were observed in the DDCS ratios (H_2-to-2H) which was explained due to the interference. However, the experiments using H are rare due to the experimental constraint and oscillations in the DDCS ratios were observed [4,12] in such experiment with H. Theoretical DDCS for atomic, or effective atomic H have also been employed [10,11] in the absence of an atomic H target. In such cases, the shapes of the oscillations are sensitive to the atomic parameters such as the effective atomic number (Z_{eff}) which is model dependent. So far oscillations have not been observed based on the H_2 DDCS data only. In this work we present a method based on the analysis of the asymmetry parameter which is independent of normalization procedures and permits therefore to observe the interference effect directly in the H_2 data only. By eliminating the need for the data on atomic H is an important step forward in the study of this effect.

Various aspects of the interference effect in fast and slow ion collisions have been investigated both theoretically and experimentally [13–18]. The importance of this effect in other related fields have been already noted earlier, such as, in photoionization [5,19–21] and electron capture [22] and energy loss of H_2 in solid targets [23]. Recently Becker and co-workers [24] have investigated the effect of isotope substitution on interference in case of N_2 to explore the effect of two dissimilar "slits." Therefore, a better understanding of this process is important in different branches of physics.

We measure the energy and angular distribution of the electron emission from H_2 and He in collisions with fast bare C ions, obtained from the BARC-TIFR 14 MV tandem Pelletron accelerator in Mumbai. The beam energy was chosen to be 80 MeV so that the projectile velocity was sufficiently large (v_p=16.4 a.u.) such that the binary encounter peak is well separated from the soft collision one for the extreme forward angles studied. An electron spectrometer equipped with a hemispherical electrostatic analyzer and a PC based data acquisition system was used. The energy and angular distributions of the electron-DDCS were studied for several forward (20°, 30°) and backward angles (150°, 160°) for electron energies between 1 and 500 eV for H_2. We show, in Figs. 1(a) and 1(b), the measured electron DDCS for the emission angles 20° and 160° for H_2 target. The DDCS be-

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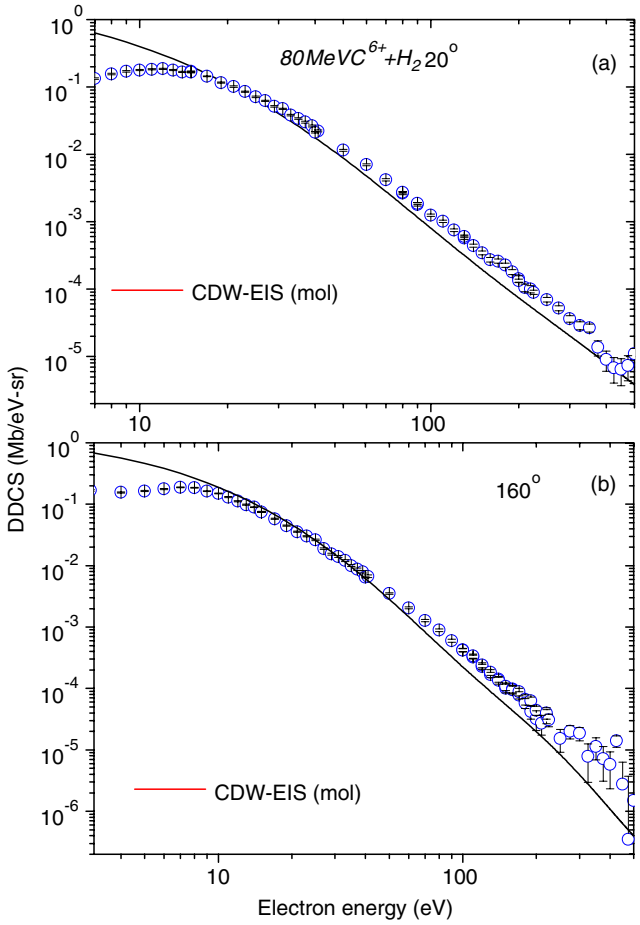

FIG. 1. (Color online) The DDCS spectra observed for the emission angles 20° and 160° in the case of H_2 target.

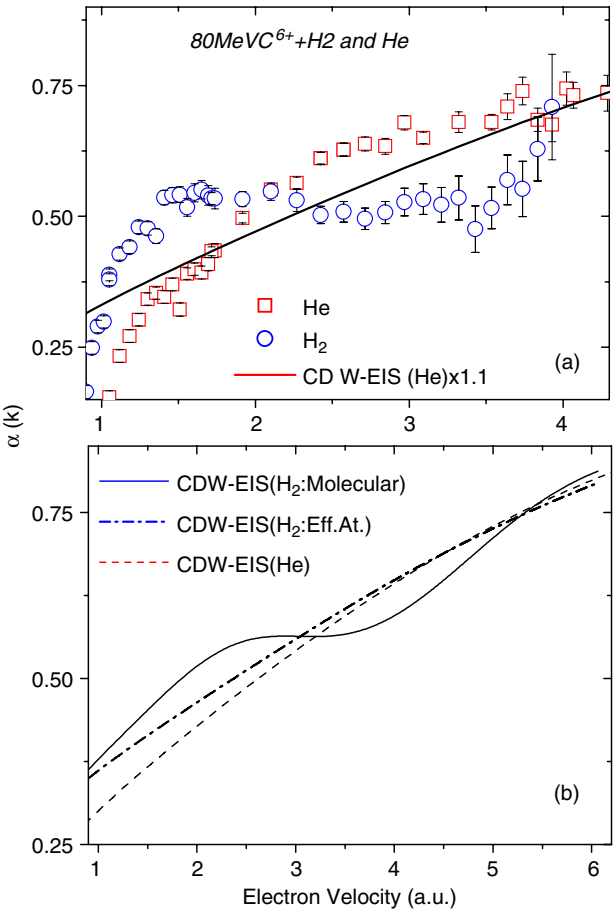

FIG. 2. (Color online) (a) The derived values of $\alpha(k)$: (a) for H_2 and He. The line corresponds to the CDW-EIS (multiplied by 1.1) for the He. (b) The solid and the dashed-dotted lines correspond to the CDW-EIS (molecular), the CDW-EIS (effective atomic type) calculations for H_2, and the dashed line is the model calculation for the He.

low 5–10 eV are not used in some cases since they are very sensitive to instrumental errors. Qualitatively an overall good agreement with the molecular CDW-EIS model can be seen with small deviations at higher energies.

We define a quantity $\alpha(k)$ as

$$\alpha(k,\theta_1) = \frac{\sigma(k,\theta_1) - \sigma(k,\pi-\theta_1)}{\sigma(k,\theta_1) + \sigma(k,\pi-\theta_1)}, \tag{1}$$

where, electron energy $\varepsilon_k = k^2/2$ (in a.u.) and θ_1 is chosen to be low forward angle, 20°. However, by expanding the $\sigma(k,\theta)$ in terms of the Legendre's polynomials, it was shown by Fainstein *et al.* [8] that, the $\alpha(k)$ would represent the angular asymmetry parameter if $\theta_1 = 0$. Since angular distribution vary slowly near 0 and π [4] the measured $\alpha(k,20°)$ approximately represent the angular asymmetry parameter (however, this approximation is not necessary for the rest of the analysis below). The derived values of $\alpha(k)$ [i.e., $\alpha(k,20°)$] show a smooth variation, i.e., increasing with the electron velocity for an atomic target such as He [see Fig. 2(a)]. This behavior is expected based on the two center electron emission which is qualitatively well represented by the CDW-EIS model. On the contrary, for C^{6+} colliding with H_2 [Fig. 2(a)] the asymmetry parameter shows an oscillatory structure superimposed on a smoothly varying function. This difference in the behavior between an atomic and molecular target at such high energy collision was unexpected in the framework of independent electron approximation and two-center effect alone. We show below that the interference effect and the difference in the oscillation frequency for forward and backward angles are the sources of such structures.

To understand this effect qualitatively, we performed a molecular CDW-EIS calculation [15]. The main feature of this model is the representation of the initial bound state by a two-center molecular wave function. Within the impact parameter approximation the transition amplitude reduces to a coherent sum of atomic transition amplitudes for each molecular center. In fast ion collisions the ionization is much faster than the typical vibrational or rotational periods which could be important only near ionization threshold [25] and therefore the nuclear degrees of freedoms are generally neglected, as shown by Fojón *et al.* [21]. The CDW-EIS model, however, reproduces the Young-type interference effect on the electron emission from H_2 and predicts higher number of oscillations for backward angles than for the forward ones. The difference in the frequency for forward and backward angles causes the oscillatory structure in the $\alpha(k)$. It can be

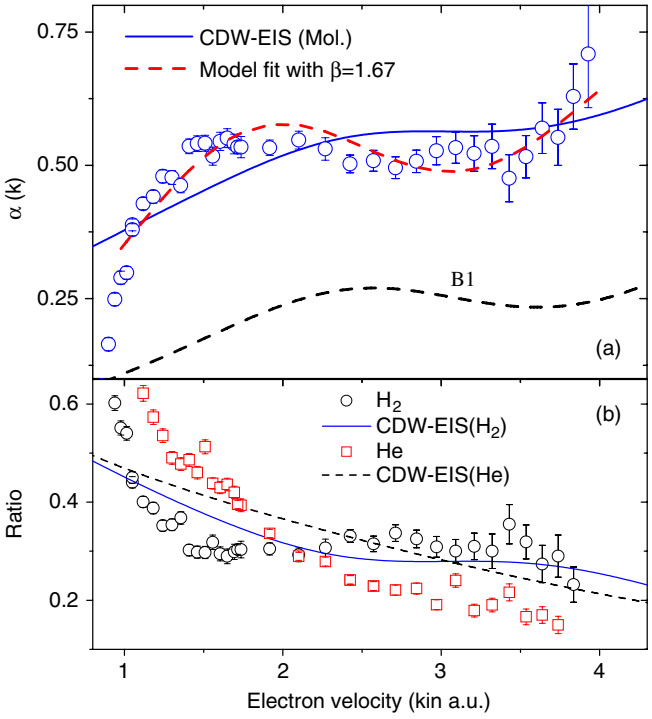

FIG. 3. (Color online) (a) The $\alpha(k)$ values for the H_2 along with the CDW-EIS calculations (solid line) and the model fit (dashed line, see text). (b) The ratios DDCS(160)/DDCS(20) for the H_2 and He along with the CDW-EIS calculation.

seen [Fig. 2(b)] that this predicts the oscillation in the α values between 1 and 5 a.u. On the contrary, the atomic-type CDW-EIS calculation [26] based on the independent electron approximation, i.e., using an effective atomic number (Z_{eff} =1.19) for atomic H does not reproduce [dashed-dotted line in Figs. 2(b) and 3(a)] any oscillation and behaves like a single center target such as the He atom [Fig. 2(b)]. This implies that the interference process built in the CDW-EIS model using molecular wave function gives rise to the oscillations in the asymmetry parameter for H_2. The general agreement of this model is quite reasonable [see Fig. 3(a)]. The first Born (B1), like CDW-EIS, employ the two-center wave function for the initial state and thus can predict interference as can be seen from Fig. 3(a) (dashed line). However, B1 completely underestimates the $\alpha(k)$ values for HCIs, because it does not take into account the two-center effect [8]. It should be mentioned here that the observed difference with CDW-EIS is amplified here since we are comparing a very sensitive quantity, namely, the ratio of difference and sum on a linear scale. So a small factor of 1.1 was required in order to reproduce the experimental data which we consider to be reasonable. However, it may indicate that the LCAO initial state for the molecule or the "effective-center" approximation (in absence of exact molecular continuum wave functions) for the final state may not be accurate enough. This provides a future scope for improvement. Therefore we have shown that (i) the asymmetry parameter for H_2 shows an oscillatory structure due to the interference effect, (ii) the derivation of oscillations in the asymmetry parameter is independent of any model assumption, (iii) it does not even need any complementary experiment using a H target, and (iv) can be derived even from the relative DDCSs. The experiment was repeated to receive better statistics, so that, the observed structure is beyond statistical errors [due to taking differences, to derive the $\alpha(k)$ values]. However, to observe these structures with much less errors one can plot the DDCS ratios DDCS(160)/DDCS(20) [see Fig. 3(b)] Obviously the structure is present in the ratios for H_2 but missing for He.

For completeness and better representation of the data we use another model. In dipole approximation [27], following Refs. [5,10], the low-energy electron DDCS for H_2 can be written as

$$\sigma_{H_2}(k,\theta) = A(k)\left(1 + \frac{\sin[kc(\theta)d]}{kc(\theta)d}\right), \tag{2}$$

where d is the internuclear separation (1.4 a.u.) and $c(\theta)$ is the frequency parameter. The ratio of frequency at backward and forward angles is equivalent to $c(\pi-\theta)/c(\theta)$ which we define as β. Using these considerations [in Eq. (1)], i.e., replacing $c(\pi-\theta)=\beta c(\theta)$ one can fit the experimental $\alpha(k)$ data. A very good agreement is obtained between the fitted line and the data. The only main fitting parameter was β since the incoherent (nonoscillatory) part of the DDCS (smoothly decreasing function of k) could be generated fairly well from the experimental DDCS for an atomic target (which is almost exponentially decreasing function of k). The fitted value of β was found to be 1.67±.04. We repeated the experiment for $\theta_1=30°$ and $\theta_2=150°$ for which the β was found to be1.85±.05. The fitting is found to be excellent even for 95 MeV F^{9+} on H_2 (not shown here). A good fitting indicates that the simple expression of interference-influenced DDCS using peaking approximation can generally explain the phenomenon.

This may imply that the interference plays a major role in the asymmetry parameter such that this parameter itself will be a sensitive measurement of the interference. In addition, a large deviation of β from 1.0 (i.e., about 1.7 to 1.8) implies that for backward angles the frequency of oscillation cannot be governed only by the longitudinal component of the momentum i.e., the simple cos θ dependence [14] which seems to explain the forward angle data only. In addition, this provides an important tool to study the interference phenomena for molecular targets without comparing them to similar data of corresponding atomic targets. This simplifies the experimental procedure significantly. Since for other multielectronic atoms, e.g., O, N, etc., the experimental and even theoretical investigations are challenging tasks, the present technique may be unique for interference studies of such molecules (N_2, O_2, etc.). In addition it is demonstrated that the asymmetry in electron emission is influenced by this mechanism, i.e., the interference effect besides the known mechanisms mentioned above.

In conclusion, we have studied the forward-backward asymmetry parameter in electron emission spectra from two electron systems such as He and H_2 in collisions with fast bare ions of C and F. The asymmetry parameter increases

smoothly with the electron velocity for He whereas it goes through a full oscillation for H_2. The molecular CDW-EIS calculation explains the observed oscillation qualitatively quite well in terms of Young-type interference and the difference in its frequency between backward and forward angles. The exploration of oscillatory structure does not need complementary theoretical or experimental study for atomic-H target and hence is self-normalized which can be applied for other multielectronic molecular targets too. The forward-backward asymmetry in electron emission can be a sensitive tool to probe the Young-type interference also in many other diatomic molecules.

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