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Evidence for interference effects in both slow and fast electron emission from D_2 by energetic electron impact

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Interference patterns due to the coherent electron emission from the atomic centers of a D_2 molecule by fast electron impact are experimentally observed in doubly differential cross sections. This behavior is supported by theoretical predictions. Measurements are given for the impact of 2.4 keV electrons. The emitted electrons are detected in the energy range from 2 to 2000 eV, at observation angles between 30° and 130°. In addition to the interference phenomenon in electron emission following soft collisions, it is shown that interference effects appear for ejected electron energies corresponding to the binary-encounter mechanism.

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Interference phenomena have been of crucial importance in the foundation of quantum mechanics. Analogies with the Young two-slit experiment [1] played a fundamental role in the description and comprehension of the dual nature of quantum objects such as electrons (see for instance Ref. [2]). In 1961 the experiment of electrons passing through a two-slit arrangement was accomplished, validating the previous assumptions of such thought experiments [3]. Since then particle interference has been investigated with neutron, atom, and molecule beams [4–6].

A fascinating alternative way of observing interference patterns is provided by the electron spectra resulting from the ionization of molecular diatomic targets. In a Young-type experiment the two molecular centers of the target can play the role of slits so that the coherent scattering of the projectile should be observed. It is clear that in order to obtain interference patterns in such experiments, the projectile wavelength must be of the order of the length of well-separated scatterers. The difference with the case of interest in the present work resides in the fact that the projectile beam acts only causing the electron ionization of the target and the possible existence of interference patterns is analyzed in the spectra of ionized electrons. The effect is due to the two-center character of the initial bound state in which the target electrons are distributed preferably around both nuclei of the molecule. Thus, a coherent emission from these two indistinguishable centers is obtained giving rise to oscillations in the differential cross sections of emitted electrons [7]. It should be expected that if the existence of interference patterns is governed by the initial bound state, the effect should appear for any enough energetic projectile colliding with the same target.

Previous works were devoted to the description of the interference phenomenon for several kinds of reactions. For instance, predictions were done in the case of photoionization of H_2^+ and H_2 molecules [7,8]. Similarly, recent measurements of the electron emission spectra in collisions of fast krypton ions [9,10], carbon ions [11], and protons [12] impacting on H_2 provide evidence for the interference effects, renewing the interest in these subjects [13–16]. Very recently, the possible existence of these interference effects have been also predicted theoretically for $(e,2e)$ reactions involving molecular hydrogen as targets [17].

In all these studies, molecular transition amplitudes were shown to be described as one-center contributions modulated by an interference factor, in analogy with experiments involving light waves. However, this analogy should be taken with caution because the coherent electron emission from the two molecular centers is considered and not the interferences caused by the scattering of the projectile beam from the two diffraction centers. As will be seen below, the interference factor depends, for massive particles, on the ejected electron momentum, the momentum transferred by the projectile to the electron, and the separation of the scattering centers (i.e., the internuclear distance). This is in contrast with photoionization processes, where only the ejection momentum and the internuclear distance are relevant as the transferred momentum is zero.

The interference patterns may also depend on the nature of the massive projectiles involved. For instance, differences in the emission spectra corresponding to ion or electron beams appear, according to the kinematical conditions imposed by the large difference of the projectile masses. For example, at identical projectile velocities the binary encounter peak where single collisions between the projectile and target electrons are produced, are present at much larger final electron momenta in the ion case. As a consequence, interference patterns are expected to be different. Moreover, for multicharged ions, the presence of a high projectile charge can also strongly modify the profiles of low-energy emitted electrons [18,19]. It must be also noted that in the case of ion impact, the projectile can be considered as moving classically from low to high collision energies, in the sense that the corresponding de Broglie wavelength is small compared to the characteristic target dimensions. The situation is completely different for electron projectiles, because they can be

considered as moving classically only for energies larger than a few tens of keV. Thus, at lower energies their quantum-mechanical behavior has to be taken into account.

In this work, interference effects in the single ionization of D_2 following 2.4-keV e^-+D_2 are measured. Besides its intrinsic interest, the present results are also useful to gain information about particle interference patterns in which finer details are not yet understood [10]. Moreover, previous experiments for ion impact were done only for ejected electron energies much smaller than the corresponding ones to the binary encounter peak, where essentially electron ionization is produced by a dipolar interaction with the projectile. The use of electrons as projectiles allows to observe that the interference phenomenon must appear also for larger ejected electron velocities, where the mechanisms that induce the electron emission are completely different. Full quantum theoretical results are provided supporting the measurements.

The ionization process is treated theoretically as a pure electronic transition by applying closure relations over all possible final rotational and vibrational states, and considering that the electronic transition matrix element depends weakly on the internuclear distance. Therefore, only vertical transitions at fixed equilibrium internuclear distance ρ_0 from the electronic ground state of the D_2 target to the electronic ground state of the residual D_2^+ molecular ion are considered in the calculations. This theoretical treatment is justified by recent theoretical and experimental results where it has been shown that, for the same system than the one considered here, autoionization of doubly excited states lying above the first and second ionization thresholds and interferences between direct ionization and autoionization are produced only at ejected electron energies smaller than 20 eV [20]. In particular, in this previous calculation the final state results from a close-coupling expansion that includes the four lowest ionization thresholds of the residual target, double excited states, and the corresponding nuclear states that describe vibration and dissociation. Thus, the model accounts for interferences between electronic and nuclear channels. For larger ejected electron energies only direct ionization is observed. The interference patterns of interest in our study appear in a much larger ejected electron energy range than the low one discussed above, and the use of the Born-Oppenheimer approximation is justified.

In this work, observables of the collision system are the differential cross sections for electron emission. As experiments do not distinguish the initial orientation of the molecules, averages over all internuclear axis angles are done. Thus, doubly differential cross sections (DDCSs) are given by

$$\frac{d^2\sigma_{\mathrm{mol}}}{d\Omega_e dE_e} \cong 2\frac{1}{4\pi}(2\pi)^4\frac{k_e k_s}{k_i}\int d\Omega_s \int d\Omega_\rho |t_{fi}^e(\rho_0)|^2, \quad (1)$$

where the emitted electron is ejected with momentum $\boldsymbol{k}_e$ (or equivalently, with energy $E_e=k_e^2/2$) into the differential solid angle Ω_e with respect to the incidence direction, and the projectile is scattered with momentum $\boldsymbol{k}_s$ into the solid angle Ω_s. The wave vector $\boldsymbol{k}_i$ of the incoming electron defines the incident direction, $\boldsymbol{\rho}_0$ denotes the equilibrium internuclear vector of the molecular target, Ω_ρ is the solid angle subtended by the internuclear vector, and t_{fi}^e represents the electronic transition matrix element.

The initial wave function is chosen as a product of a plane wave describing the motion of the incident particle and the initial two-electron bound state that in this work is described by a Heitler-London-type wave function [21]. Cohen and Fano [7] have also used a simple linear combination of effective atomic orbitals to represent the initial bound state and a single-center continuum wave function to describe the exit channel in the case of photoionization of H_2^+ molecular ions, obtaining an analytical form to describe the interference patterns. In a recent work [22] it has been shown that for photoionization of H_2 molecules and for the case that the residual H_2^+ molecules remain in the ground state, the use of this formula gives an adequate description of interference patterns as compared with calculations obtained with exact electronic bound and continuum wave functions, except at low ejected electron energies. It supports the use of a simple Heitler-London wave function for electron impact, as it has been previously done with success for ion impact [13].

In order to describe the final electronic wave function, the two-effective center (TEC) model [23] is employed. This first-order approximation is based on the localized nature of the initial electronic density around the molecular nuclei. Therefore, 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 corresponding to the region from which ionization is not produced. Thus, the final continuum wave function of the ejected electron is chosen as an effective one-center Coulomb wave function taking into account the interaction of the emitted electron with one or the other molecular nucleus [23].

With all these assumptions and following Ref. [17], the molecular DDCSs given by Eq. (1) may be rewritten as

$$\frac{d^2\sigma_{\mathrm{mol}}}{d\Omega_e dE_e} \cong \int d\Omega_s \left[1+\frac{\sin(\chi\rho_0)}{\chi\rho_0}\right]\frac{d^3\sigma_{2\mathrm{H}}}{d\Omega_e d\Omega_s dE_e}, \quad (2)$$

where $\boldsymbol{\chi}=\boldsymbol{k}_e-\boldsymbol{K}$, and $\boldsymbol{K}=\boldsymbol{k}_i-\boldsymbol{k}_s$ is the momentum transferred to the ionized electron. The triply differential cross section $d^3\sigma_{2\mathrm{H}}/d\Omega_e d\Omega_s dE_e$ refers to two *effective* H atoms located at the position of each molecular nuclei. The expression in parentheses appearing in Eq. (2) is identified as the interference factor due to coherent emission from the different scattering centers in the molecule. It is worth mentioning that oscillations persist even after performing the integration over all molecular orientations.

The experiments were carried out at CIRIL in Caen, France, using an electron gun of simple design. The electron beam was directed into an effusive D_2 gas jet and collected in a Faraday cup (FC), after passing through an exit slit (ES). Typical currents of $\sim 30\ \mu$A were measured. The average target pressure was estimated to be $\sim 10^{-4}$ mbar (corresponding to a density of $\sim 10^{13}$ cm^{-3}), whereas the background pressure was $\sim 10^{-7}$ mbar. In order to reduce the background due to electron reflection and secondary electrons emitted from surfaces, the current on the exit slit was minimized.

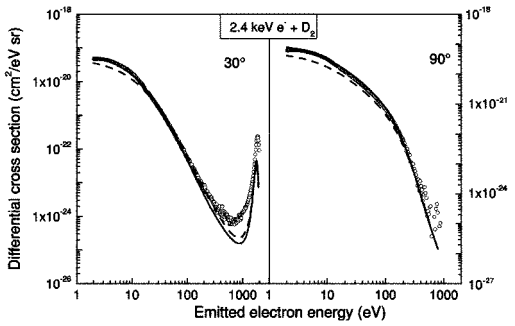

FIG. 1. Cross sections for electron emission in 2.4 keV e^-+D_2 collisions as a function of the emitted electron energy, for detection angles of 30° (left-hand side) and 90° (right-hand side). Experimental data, open circles; twice theoretical cross sections for two effective H atoms, dashed curves; theoretical molecular cross sections, full curves.

Typical ratios of ~1000 between both FC and ES currents were obtained, and were found to be sufficiently high to measure spectra from 2 eV up to 500 eV for backward angles or 2000 eV for forward angles.

To perform the experiment, the electron-spectroscopy apparatus from the Hahn-Meitner Institut in Berlin [24] was used. The emitted target electrons were energy analyzed using a single-stage spectrometer, which consists of a 45° parallel-plate analyzer. The electron energy was determined with a resolution of 5% full width at half maximum (FWHM). The angular resolution is ~2° FWHM. Since the main experimental *relative* uncertainties are due to statistical errors, care was taken to obtain a sufficient counting rate so that the relative uncertainties are smaller than ~30%. To evaluate cross sections, we have used a method previously described in detail [25]. Cross sections for electron emission were obtained by subtracting spectra with the target-gas jet and in the uniform-target-gas mode. The latter spectra were achieved by moving the gas jet upwards far away from the electron beam and flooding the scattering chamber uniformly with the D_2 gas target (below 10^{-5} mbar).

In Fig. 1, measured spectra for electron emission from D_2 molecules are shown (open circles) as a function of the ejection energy. The impact energy is $E_i=2.4$ keV and the emission angle θ_e is fixed at 30° and 90°. All experimental results were normalized to the theoretical ones by taking the corresponding values for the detection angle of 90° as reference. The experimental data present typically a monotonic decrease of several orders of magnitude as the emission energies increase, showing that the bound electron is predominantly ejected with low energies. For the $\theta_e=30°$ case, a peak is observed at around 1850 eV. This peak may be related to binary collisions in which all the momentum is transferred to the ejected electron.

Theoretical molecular DDCSs obtained with the TEC approximation are also shown in Fig. 1 (full lines). A very good agreement between experimental and calculated cross sections is found in almost the whole range of ejection energies presented here. However, some discrepancies in absolute values appear at large ejected electron energies. It could be due to exchange effects, not included in the present formulation, which might be important at increasing values of the ejection energies.

DDCSs corresponding to two *effective* H atomic centers are also displayed in Fig. 1 (dashed lines). These atomic cross sections were obtained by neglecting the damping oscillatory term in Eq. (2), i.e., by considering an *effective* hydrogen with an effective nuclear charge equal to the one used in the Heitler-London wave function and an effective energy equal to the molecular bound energy. This procedure is useful to avoid the presence of additional structures related to the different Compton profiles of H and H_2 targets [13]. In particular, for low (high enough) values of the ejection energy, atomic results are smaller (larger) than the molecular TEC ones, which give a correct description of the measurements at low and intermediate energies. According to Eq. (2), differences between molecular and effective atomic results should be more evident in both the soft collision (small K and k_e values) and binary-encounter ($\boldsymbol{K}\cong\boldsymbol{k}_e$) regions, where interferences are constructive. However, it should be noted that in the region of small ejected electron velocities, autoionization also contributes to the ionization spectra [20], thus making more difficult the identification of interference effects in the soft collision region.

To make more noticeable the presence of interference terms coming from the two-center geometry of the molecule, the experimental cross sections were divided by twice the DDCSs corresponding to the *effective* H atom calculations. The resulting ratios (referred to as experimental ratios in the following) were fitted using a function of the type $a_0+a_1k_e+a_2\sin(ck_e)/(ck_e)$, where a_0, a_1, a_2, and c are adjustable parameters. The parameter c plays the role of an effective frequency which may depend on the emission angle [10]. The spherical zero-order Bessel function describes the oscillating part of the ratios, while the linear component of the fitting function takes into account the increasing discrepancies with increasing emitted electron energy between experiment and theory (Fig. 1). The experimental ratios and the corresponding fitted curves are presented in Fig. 2 as a function of the electron velocity for different detection angles, after subtraction of the linear function $a_0+a_1k_e$. To avoid the presence of contributions due to multielectron processes, the figures are presented for ejected electron velocities larger than 1 a.u. As the ejection velocity increases, fluctuations associated with experimental relative uncertainties (essentially due to statistical errors) appear in the experimental ratios. Nevertheless, the oscillations around unity are clearly visible except for the $\theta_e=70°$ case, which will be discussed below. The ratios take a maximum value for the smaller ejection velocities considered, accordingly to the constructive interferences predicted by the theory, and then decrease until they reach a minimum value.

In Fig. 2, theoretical ratios obtained from Eq. (2) are also presented for comparison. The overall agreement between experiments and theory confirms the presence of interference patterns due to the coherent emission from both scattering centers. Moreover, the frequencies predicted by the theory are close to those of the fitted ratios. In accordance with Refs. [10,13,17], it appears that the frequency of the oscillations varies with the ejection angle. Moreover, it is noted that the frequency increases at backward emission as θ_e increases.

Extra features appear at $\theta_e=70°$. For instance, the theoret-

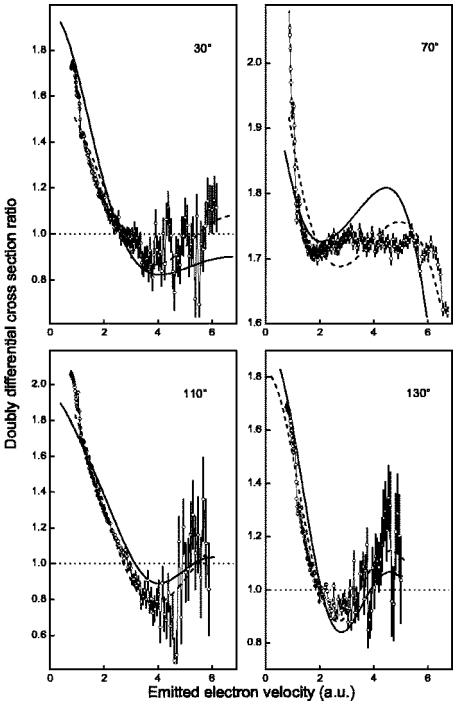

FIG. 2. Experimental (open circles) and theoretical (full curves) doubly differential cross-section ratios (see text) for electron emission in 2.4 keV e^-+D_2 collisions as a function of the emitted electron velocity, for detection angles of 30°, 70°, 110°, and 130°. Due to the strong decrease of electron emission cross sections with increasing emitted electron velocity (see Fig. 1), the statistical uncertainties for the experimental cross-section ratios range from a few percent at electron velocities lower than 2 a.u. to (typically) 20 %–30 % at velocities larger than 5 a.u. Experimental ratios are fitted by using a zero-order Bessel function (dashed lines).

ical ratio is found to oscillate, presenting a minimum and a maximum at velocities of $\sim$2 a.u. and $\sim$4.5 a.u., respectively. This maximum comes from the oscillatory term in the integrand of Eq. (2), which gives a maximum contribution for $\boldsymbol{K}\cong\boldsymbol{k}_e$ (i.e., under binary-encounter conditions). This maximum in the theoretical ratios should not be confused with the binary-encounter peak itself. Instead, it is the signature of constructive interference in the binary-encounter region. Experimentally, the maximum in the ratio is not observed. However, as in the theoretical ratio, at a velocity of $\sim$2 a.u. a strong change in the ratio slope is found so that the ratio becomes rather constant at larger electron velocities. In the velocity range considered in Fig. 2, the theory presents a cross-section ratio oscillating around a value of $\sim$1.7 (and not around the unity as in all the other angles considered here), in good agreement with the constant value of experimental ratio for $k_e \gtrsim 2$ a.u. A detailed analysis of the theoretical predictions clearly shows that it is a consequence of the overlapping of the structures related to constructive interferences at small ejection velocities and in the binary-encounter region. Thus, experiment supports the existence of constructive interferences in the binary-encounter region. For larger ejection velocities (not shown in Fig. 2) the theoretical ratio presents damping oscillations around the unity as it occurs for the other ejection angles considered.

In conclusion, experimental evidence of the existence of interference effects due to the coherent electron emission from the two indistinguishable molecular centers of the target was provided for the case of fast electrons impacting on D_2. The electron spectra were recorded at emission energies up to 2000 eV. These measurements allow a more detailed discussion about the interference phenomenon, which is shown to appear at ejected energy regions corresponding to different ionization mechanisms. While for ion impact the measurement of constructive interference in the binary-encounter region is an extremely difficult task because high ejected electron energies are necessary, in the present electron impact study constructive interferences were experimentally accessible to observation in the binary-encounter region, as well as for small ejected electron velocities. The present measurements for electrons are moreover supported by their fairly good agreement with theoretical predictions.

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