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Electron-impact-ionization cross sections of H_2 for low outgoing electron energies from 1 to 10 eV

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Theoretical and experimental fully differential cross sections are presented for electron-impact ionization of molecular hydrogen in a plane perpendicular to the incident beam direction. The experimental data exhibit a maximum for 1-eV electrons detected 180° apart and a minimum for 10-eV electrons. We investigate the different physical effects which cause back-to-back scattering and demonstrate that, over the energy range from 10 to 1 eV, a direct transition is observed from a region where Wannier threshold physics is essentially unimportant to where it completely dominates.

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Low electron energy (near-threshold) ionization has been studied for atoms over the years and it is now well understood [1–3]. There have been several experiments and theories reported for near-threshold ionization for hydrogen [4], helium [5], and heavier inert gases [6], and the agreement between experiment and theory is generally very good. By contrast, $(e,2e)$ studies for ionization of molecules at low energies has received relatively little attention until recently. Current models are now in reasonable agreement with experimental data for H_2, providing an understanding of the collision dynamics under the conditions used in the experiments [7–9]. These collisions provide direct information about the importance of three-body effects, including electron-electron correlation, polarization, and multiple collisions in the ionization process.

Al-Hagan *et al.* [7] compared experimental and theoretical $(e,2e)$ results for ionization of H_2 and He (having the same number of electrons and protons) in a plane perpendicular to the incident beam direction (the *perpendicular plane*). The experimental measurements were performed with both final-state electrons having 10-eV energy and the fully differential cross section (FDCS) was measured as a function of the relative angle between outgoing electrons. The experiments revealed that both H_2 and He had peaks in the cross sections at relative angles around 90° and 270°. In contrast, for back-to-back scattering at 180°, helium showed a very strong peak (the largest cross section) while H_2 had a very small minimum. It was demonstrated that the 90° and 270° peaks for both H_2 and He resulted from elastic scattering of the projectile from the target into the perpendicular plane, followed by a binary collision between projectile and target electrons. Since the binary collision occurs between particles with equal mass and energy, the mutual angle between the electrons is then 90° (or 270°). This process occurs for both atomic and molecular targets. For helium, it was shown that the large maximum resulted from one of the scattered electrons being very close to the nucleus, so that it elastically backscattered at 180° from the point nuclei. For the case of H_2, the highest probability of electron-electron collision occurs *between* the two hydrogen nuclei where on average the net attractive force cancels, resulting in almost no backscattering and hence a minimum at 180°.

We have now extended the (10 eV, 10 eV) measurements for H_2 in the perpendicular plane to lower equal-energy pairs down to (1 eV, 1 eV). We discovered that the deep 180° *minimum* for (10 eV, 10 eV) became decreasingly shallow as the energy lowered and eventually developed into a *peak* at 180° for (1 eV, 1 eV). The purpose of this Rapid Communication is to identify the physical effects responsible for the minimum changing into a maximum. We show that at 1 eV the maximum is *not* related to nuclear scattering as was the case for 10 eV He but rather is due to final-state electron-electron repulsion [normally called postcollision interaction (PCI)].

This finding is reminiscent of the Wannier law, which predicts that, at threshold, the electrons will emerge at 180° due to PCI. An interesting and unresolved question concerns the range of validity for the Wannier threshold law, and we show here that this starts to break down for electrons with energy (0.5 eV, 0.5 eV). However, we are close enough to this region at (1 eV, 1 eV) so that PCI is still dominant for the FDCS. Martinez *et al.* [10] very recently showed that PCI was not dominant for these same energies for atomic targets, so this finding appears to be a phenomena associated with molecules. To our knowledge, this is the first direct observation of the transition from Wannier physics to nonthreshold physics for fully differential cross sections of H_2. Surprisingly, the dominance of PCI becomes unimportant very quickly after 1 eV and is found to be of no consequence for back-to-back scattering by 10 eV.

The apparatus used for the experimental studies in Manchester has been described in detail elsewhere [8,9,11]. Briefly, the spectrometer is fully computer controlled and computer optimized, and it can access geometries from coplanar to the perpendicular plane. All results presented here were carried out in the perpendicular plane using an unselected energy electron gun and hemispherical energy analyzers to detect scattered and ejected electrons. The energy resolution was ∼1 eV, and the angular resolution was around ±3°. Different electron beam currents were used at each energy so as to optimize the coincidence signal-to-noise ratio, so all results are renormalized to unity at the peak of the data for comparison to theory.

The molecular distorted wave Born approximation (MDW) has been presented previously [12] so only a short summary is presented here. The FDCS is given by

$$\frac{d^5\sigma}{d\Omega_a \Omega_b dE_b} = \frac{2}{(2\pi)^5}\frac{k_a k_b}{k_i}|T|^2, \tag{1}$$

where $\vec{k}_i$, $\vec{k}_a$, and $\vec{k}_b$ are wave vectors for the initial, scattered, and ejected electrons, respectively. The T matrix for the MDW is given by

$$T^{\mathrm{MDW}} = \langle \chi_a^-(\vec{k}_a,\mathbf{r}_1)\chi_b^-(\vec{k}_b,\mathbf{r}_2)|V - U_i|\phi_j^{\mathrm{OA}}(\mathbf{r}_2)\chi_i^+(\vec{k}_i,\mathbf{r}_1)\rangle, \tag{2}$$

where $\mathbf{r}_1$, $\mathbf{r}_2$ are coordinates of the incident and bound electrons, χ_i, χ_a, and χ_b are distorted waves for the incident, scattered, and ejected electrons, respectively, and $\phi_j^{\mathrm{OA}}(\mathbf{r}_2)$ is the initial bound-state wave function approximated by the orientation-averaged molecular wave function. The initial state interaction V is the potential between the incident electron and neutral molecule, and U_i is a spherically symmetric molecular potential used to calculate the initial-state distorted wave for the incident electron, $\chi_i^+(\vec{k}_i,\mathbf{r}_1)$.

The final state is approximated as a product of distorted waves for the two continuum electrons, which are calculated as with the initial state, except the spherically symmetric static distorting potential of the molecular ion is used instead of U_i.

The molecular three-body distorted wave approximation (M3DW) [13,14] is similar to the MDW except an electron-electron Coulomb repulsion factor is included in the final-state wave function. Here we adopt the Ward-Macek average Coulomb-distortion factor between the two final-state electrons [15]. When the Coulomb interaction is included in the final-state wave function, PCI is included to all orders of perturbation theory and when only included in the perturbation, PCI is included only to first order.

The time-dependent close-coupling (TDCC) approach to electron-impact ionization of H_2 has also been described in detail previously [8,9]. For small impact electron energies, fewer partial waves are usually required than at higher impact energies, but the spatial grids required to fully converge the calculation may become very large. Since the TDCC calculations must also be run for each impact energy separately, the computational cost associated with deriving the required amplitudes for each energy is considerable. We therefore only present TDCC calculations for three outgoing energies, as detailed in the following.

Figure 1 compares experimental and theoretical FDCS for electron-impact ionization of H_2 for the case of equal final-state electron energies in the perpendicular plane. The data are compared with MDW and TDCC theories, both theory and experiment being normalized to unity as previously noted. The TDCC results are presented only where the two electrons have energies of 2, 5, and 10 eV, respectively. The shape of the experimental data changes dramatically as the energy decreases. The binary scattering peaks are at ~90° and ~270° and minimum at 180° for 10-eV electrons becomes a single 180° maximum at 1 eV. Agreement between experiment and both calculations is good at 10 eV for the angular range of the experimental measurement. We note, however, that the MDW predicts unphysically large cross sections for small and large angular separations. Since 0° (or 360°) scattering corresponds to two equal-energy electrons traveling in the same direction, it is clear that these cross sections should be very small as the TDCC predicts. For the MDW, as the energy decreases, there is a minimum at 180° for all energies. Although the MDW 180° minimum becomes less shallow with decreasing energy,

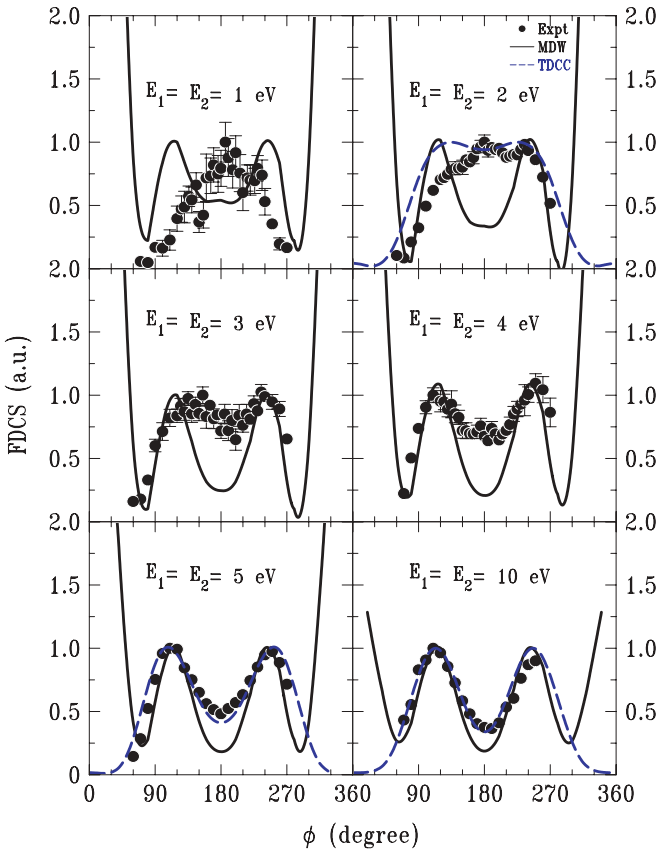

FIG. 1. (Color online) FDCS for ionization of H_2 using perpendicular plane kinematics. The FDCS are plotted as a function of φ (the angle between the two final-state electrons in the detection plane). The energies of the outgoing electrons are shown on the respective plots. The experimental measurements are compared with MDW calculations (the solid curve) and the TDCC calculations (the dashed curve). For each energy, the experimental and theoretical data are normalized to unity at the experimental maximum.

disagreement between experiment and theory increases with decreasing energy, the MDW predicting a minimum at 1 eV in contrast to the data. It is important to note that Martinez *et al.* [10] found very good agreement with the comparable 1-eV data for He using the atomic equivalent of the MDW, so the MDW is good for atoms at this energy but not molecules! The lowest energy calculated using the TDCC theory was at 2 eV, and the TDCC still exhibits a shallow minimum at this energy while the data indicate a maximum at 180°.

To investigate the physical effects causing the change in shape of the FDCS as the energy decreases, we tested the importance of both nuclear scattering and electron-electron interactions. In [7] we investigated the effect of nuclear scattering for electron impact ionization of H_2 in the perpendicular plane where the two outgoing electrons each had 10 eV. For this case, we demonstrated that the 180° minimum in H_2 became a maximum when the two nuclei were brought together to form a point charge while the electronic distribution was left unchanged. Consequently, we decided to see whether nuclear scattering could be causing the peak in the data at 1 eV. In Fig. 2, MDW results are presented where the size of the nuclear separation is reduced from $1.4a_0$ to a point charge, keeping

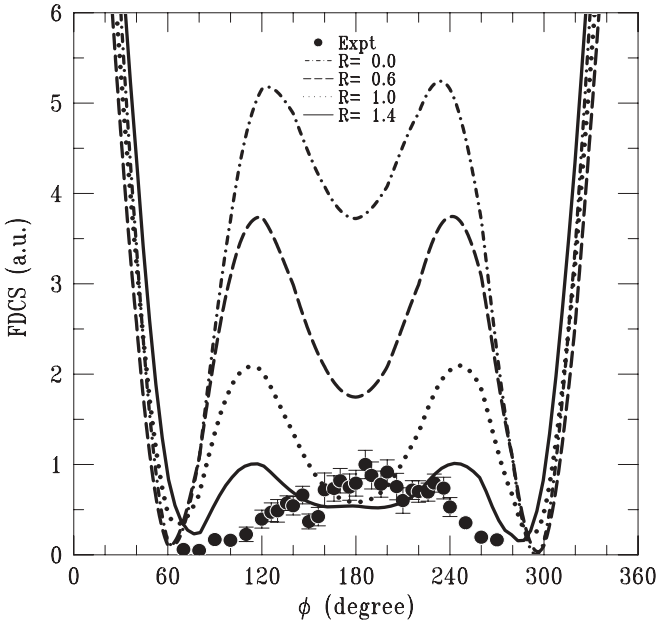

FIG. 2. The dependence of the FDCS as a function of φ (the angle between the two final-state electrons in the detection plane) for various nuclear separations. Both ejected electrons have energy of 1 eV. The MDW calculations are for different nuclear separations $R = 0.0a_0$, $0.6a_0$, $1.0a_0$, and $1.4a_0$ as shown.

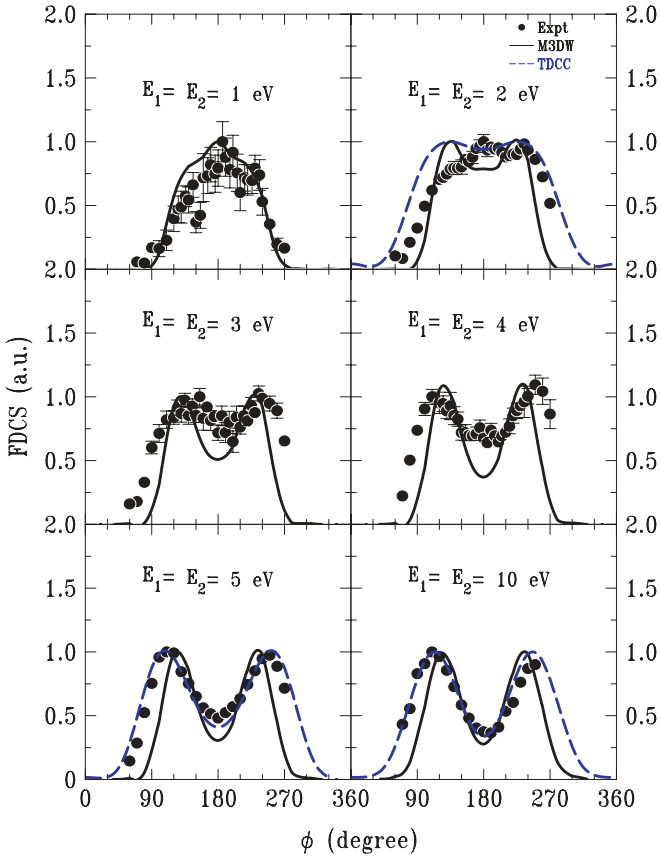

FIG. 3. (Color online) Same as Fig. 1 except now the solid curve is the M3DW.

everything else unchanged. It is clear that reducing the spacing of the nuclear separation to a point charge caused the 180° minimum to become deeper, so these results do not support the idea that the 180° peak results from nuclear scattering.

For further investigation, we added PCI to our theory to study the importance of electron-electron interactions near threshold. The Coulomb interaction in M3DW is included in the final-state wave function, so PCI is included to all orders of perturbation theory. Figure 3 shows the data compared with the M3DW approach as well as the TDCC method. As before, theory and experiment are normalized to unity at the experimental maximum. The agreement between experiment and the M3DW is now much improved. The 180° minimum for the M3DW decreases with lowering energy, in fairly good agreement with experiment, and the minimum at 2 eV is now much closer to the TDCC results. Although the experiment indicates a slight peak at 180°, a shallow minimum would nevertheless lie within the statistical uncertainty of the experiment. Also, the theoretical calculations have not been convoluted over the experimental uncertainty in energy around ±1 eV, which could also explain the small difference with theory at this energy. It is clear that inclusion of PCI is important at all energies, but it becomes much more important when both final-state electrons have 1-eV energy, since PCI turns the minimum into a maximum, and the binary peaks are much less prominent.

As noted, Al-Hagan *et al.* [7] showed that PCI did not produce a peak for back-to-back scattering at 10 eV but rather that the minimum resulted from the binary collision occurring between the two H_2 nuclei where the net attractive force producing electrons at 180° cancels on average. What is seen here is a transition from the case where PCI is *unimportant* at 10 eV to the case where PCI becomes *dominant* at 1 eV. This can be understood since the outgoing electrons have more time to interact as the energy decreases, and hence PCI forces the outgoing electrons to emerge at a mutual angle of 180°. However, it is surprising that this transition happens so quickly over a small range of energies.

The dominance of PCI at 1 eV reminds us of the Wannier threshold law. The problem of threshold ionization has been extensively studied and is now well understood [16–18]. The first theory of near threshold breakup given by Wannier [16] was extended by Peterkop and Rau, the Wannier-Peterkop-Rau (WPR) threshold law predicting that the fully differential cross section for $(e,2e)$ ionization of hydrogen should satisfy FDCS α $E_{\rm ex}^{-0.373}$ [1,2,17,18], where $E_{\rm ex}$ is the excess energy. This law has recently been confirmed by accurate numerical calculations for electron-hydrogen scattering [3,4]. If we adopt a simple double-atom model for molecular hydrogen, threshold ionization of H_2 should follow the same law. Figure 4 shows the excess-energy dependence of the FDCS for H_2 in the near-threshold energy region for backscattering at 180°. The solid line is the WPR theory normalized to the M3DW at the lowest energy and the dashed curve is the M3DW calculation. Clear differences occur only for excess energies above 1 eV (i.e., each electron has 0.5-eV energy) and significant deviations from the Wannier region are clear at higher energies.

For the case of $E_1 = E_2 = 1$ eV, the M3DW is within ~20% of the WPR curve, indicating that PCI is still the dominant process producing a maximum for 180° scattering. For $E_1 = E_2 = 2$ eV (4-eV excess energy), the M3DW is

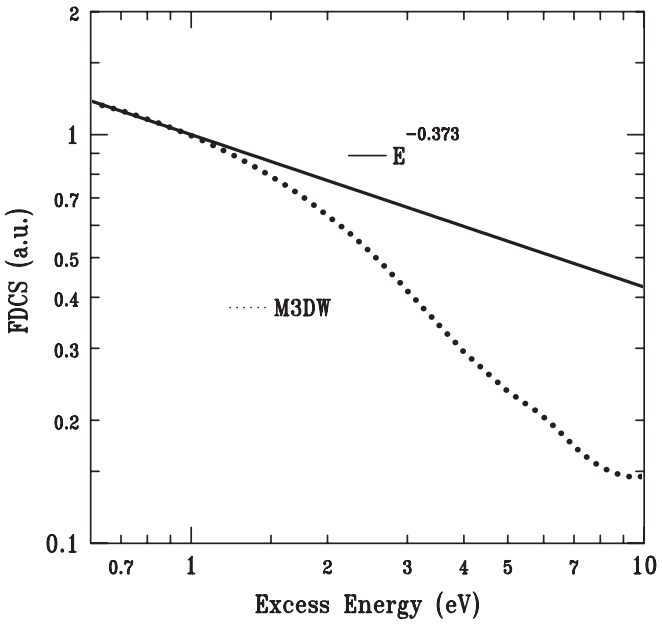

FIG. 4. Calculated H_2 FDCS for equal energy sharing and $\varphi = 180^\circ$ as a function of excess energy. The solid line gives the results of the Wannier theory normalized to the M3DW at the lowest energy and the dashed line is for the M3DW FDCS.

about 50% below the WPR curve and both M3DW and TDCC theories predict a minimum. This means that the strength of PCI is significantly reduced, and by 10-eV excess energy PCI is of little consequence for this scattering angle. As a final note, the WPR theory also predicts that the FDCS should have a Gaussian angular distribution centered around 180°. From Fig. 3 for $E_1 = E_2 = 1$ eV, we see that both experiment and theory have a Gaussian-like distribution around this angle, which is consistent with Wannier theory.

We can also compare and contrast the Wannier region for atomic hydrogen with the current molecular case. Previous studies [1] have shown that the Wannier region for atomic hydrogen extends to an excess energy of around 3.3 eV. In the molecular hydrogen case, the Wannier region has a lesser extent of about 2 eV (as indicated by Fig. 4). This can be understood by remembering that the molecular hydrogen wave function is more extended in space than the atomic hydrogen wave function, even though the two systems have similar binding energies. Wannier theory relies on classical scaling, where the distances r are replaced by r^* scaled by the excess energy E of the system: $r = r^*/E$. If a universal r^* is assumed to control the range of the Wannier region in energy, then $E = r^*/r$. Inserting $r = R_i$ with $i = \mathrm{m}$ or a for molecule and atom, respectively, yields $E_m < E_a$ since $R_m > R_a$. This explains why the threshold region for molecular hydrogen is smaller in energy than for atomic hydrogen.

In conclusion, we have compared experimental data with TDCC and M3DW calculations in the perpendicular plane for cases where the outgoing electrons have equal energies ranging from 1 to 10 eV. The data for 10 eV exhibit peaks at 90° and 270° and a minimum at 180°. We had previously shown that the ∼90° and ∼270° peaks result from elastic scattering of the projectile from the target into the perpendicular plane followed by a classical binary collision between the projectile and target electrons. For the minimum at 180°, it was shown that PCI is unimportant at this energy and that the electron-electron collision occurs between the nuclei where the net attractive force cancels on average, so that there is almost no 180° scattering.

The data presented here reveal that the shape of the FDCS completely changed from two peaks centered at 90° and 270° to a single peak at 180° as the electron energy approaches threshold. We investigated the physical effects causing this change and found that PCI changes from being unimportant at 10 eV to being the dominant physical process at 1 eV. For the lowest energy, the FDCS has a Gaussian shape centered on 180° as is predicted by the WPR threshold law. Although theory indicates that the minimum measured energies are not quite low enough for the threshold law to hold strictly, they are nonetheless close enough for the Wannier model to provide the dominant physics. It should be noted that the FDCS for helium at the same outgoing electron energies also displays a dominant single peak at 180° as seen here and that the side lobes are also eliminated. This shows that as PCI dominates the interaction, the target structure becomes decreasingly important.

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