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Differential cross sections for single ionization of H_2 by 75keV proton impact

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We have calculated Triply differential cross sections (TDCS) and doubly differential cross sections (DDCS) for single ionization of H_2 by 75 KeV proton impact using the molecular 3 body distorted wave eikonal initial state (M3DW-EIS) approach. Previously published measured DDCS (differential in the projectile scattering angle and integrated over the ejected electron angles) found pronounced structures at relatively large angles which were interpreted as an interference resulting from the two-centered potential of the molecule. Theory treating H_2 as atomic H multiplied by a molecular interference factor only predict the observed structure when assumptions are made about the molecular orientation. Here we apply the M3DW-EIS method, which does not rely on such an ad hoc approach, but rather treats the interference from first principles.

I Introduction

For the past several decades, the problem of ionization of atoms and molecules by charged particles has been studied extensively [1-17]. The triply differential cross section (TDCS) represents the most sensitive test of theory as it determines the energy and the location of all the final state particles. Previously it was difficult to measure projectile scattering angles for a fast heavy ion projectile as the deflection is immeasurably small. Only for light ion impact at intermediate energies have experimental TDCS been obtained by measuring the projectile scattering angle directly [18]. With the use of Cold Target Recoil Ion Momentum Spectroscopy [COLTRIMS][4-6], TDCS measurements have now become possible even for fast heavy ions by deducing the projectile scattering angle from momentum conservation [19].

Theoretically many different approaches have been used to calculate TDCS for ion-impact ionization of atoms such as the First born approximation-Hartree Fock (FBA-HF) [7], the continuum distorted wave (CDW) [20], the continuum distorted wave-Eikonal initial state (CDW-EIS) [2,11], the three coulomb wave Hartree Fock (3C-HF) [7], the 3DW-EIS (3-body distorted wave- Eikonal initial state) [1, 8], and a non-perturbative TDCC (time dependent close coupling) approach [21]. The FBA is only valid at sufficiently large projectile energies for higher-order contributions to be small. The CDW-EIS/3DW-EIS approach accounts for higher order effects in both the initial and final states, but it is effectively a 2-state approximation neglecting couplings to excited states other than the continuum states of interest. Both higher-order contributions and couplings to multiple final states are incorporated in the TDCC method, but current versions use a single center base not incorporating any bound projectile states. Therefore, capture is completely neglected.

In spite of all the work that has been done for ionization of atoms, the literature on theoretical work for ion-impact ionization of molecules is much less extensive. The simplest approach is to approximate the ionization of H_2 as ionization of atomic H multiplied by an interference factor to approximate scattering from the two nuclei [22]. Recently Pindzola *et al.* [23] reported a TDCC calculation for single and double ionization of H_2 . However, only total cross sections were reported in that work. In the present work, we present a molecular version of the CDW-EIS and use it to calculate the TDCS for proton-impact ionization of H_2 which are then integrated over the ejected electron's solid angle to obtain the double differential cross sections (DDCS). We will call the DDCS differential in projectile scattering angle and energy loss DDCS-P to distinguish it from DDCS-E which are differential in ejected electron observation angles and integrated over projectile scattering angles.

The atomic CDW-EIS model was introduced by Crothers and McCann [11] for the single ionization (SI) of hydrogenic targets. Fainstein *et al.* [24] used it for multi-electronic targets. The CDW-EIS has been used very successfully for many years in heavy ion collisions with atoms for studies of DDCS-E [25-28]. In the original CDW-EIS approach [11], the projectile is treated classically, the projectile-ion interaction has no effect on the TDCS, and the projectile trajectory is approximated as a straight line [2]. Later, this method was refined to include the projectile-ion interaction by Rodriguez *et al.*[29], Sanchez *et al.* [30], and Ciappina *et al.*[31]. Madison *et al.* [2] introduced a fully quantum mechanical version of the CDW-EIS which they called the 3DW-EIS approach for heavy-ion collisions with atoms. For the initial state, the 3DW-EIS uses a HF wave function for the bound electron and an Eikonal wave for the projectile. For the final state, a distorted wave is used for the ejected electron, a Coulomb wave is used for the projectile in the field of a point charge, and the Coulomb distortion between the projectile and the ejected electron is also included. The Eikonal approximation for the initial state contains the asymptotic Coulomb interactions between the projectile and target [11]. The 3DW-EIS model has previously produced good agreement with experimental TDCS results [1, 8] for single ionization of helium. A detailed comparison between the 3DW-EIS and CDW-EIS for ionization of helium was given by Madison *et al.* [2]. Here we extend the 3DW-EIS approach for atoms to treat ionization of molecules and we call the molecular version M3DW-EIS.

Since a Hydrogen molecule has two nuclear centers, the projectile can scatter from either center which leads to the possibility of observing interference effects. This kind of interference, which is closely related to the Young type double slit experiment of light, was first discussed by Tuan and Gerjuoy [32] a long time ago for ionization of molecular targets. However, only later was it observed experimentally [3,33,34]. Earlier reports found evidence for interference in the ejected electron distribution [35]. There have been several DDCS-E measurements reported which found indication for double-slit interference, but the effects were so weak that they could only be seen indirectly in ratios between molecular and atomic cross sections

In contrast, significant structures were seen in measured DDCS-P by Alexander *et al.* [3]. These structures are due to the type of interference predicted by Tuan and Gerjuoy [32]. Alexander *et al.* [3] found that this interference in the diffracted projectile wave is a much more sensitive probe as it was observed directly in the absolute DDCS-P cross sections without having to take ratios to atomic H cross sections. Another interesting feature in the data of Alexander *et al.* [3] was that the interference structure disappeared for energy losses corresponding to an ejected electron speed equal to the projectile speed. The velocity of the projectile in that work is 1.73 a.u which matches with the velocity of the ejected electron with an energy of 41.6eV. It is

well known that for matching velocities, the post-collision interaction (PCI) between the ejected electron and the outgoing projectile, which was investigated by Crooks and Rudd [36] several decades ago, is maximized. Alexander *et al.* [3] therefore discussed the possibility that there may be a correlation between the phase factor giving rise to the interference and PCI. In this article we employ the M3DW-EIS method to study whether the structures in the experiment can be reproduced by a calculation that treats the target as a molecule ab initio and to obtain a better understanding about the origin of these structures. This method accounts for interference from first principles since any structure must result from the molecular wavefunctions used in the calculation (rather than being imposed on an atomic calculation through an interference factor). We will also investigate the TDCS to see what insight can be gained from the more differential level. We will use atomic units for all calculations.

III Theory

Rayleigh Interference Factor

A commonly used approximation for diatomic molecules is to multiply the atomic cross section by a two-center interference factor. Rayleigh [37] used the interference factor more than 100 yrs ago in the study of acoustic power originating from a pair of point sources. The same factor arises in Moore's 1955 calculation of elastic coherent scattering from diatomic molecules [38], Fano [39] over 40 years ago used it for treating photoionization of molecules, and Stia *et al.* [22] applied it to particle ionization. In this approximation the cross section for non-oriented molecules is

$$\sigma_{H_2} = 2\sigma_H \left(1 + \frac{\sin(\omega)}{\omega}\right), \quad (1)$$

where σ_H is the TDCS for atomic Hydrogen, and σ_{H_2} is the TDCS for molecular Hydrogen. The interference factor $\left(1 + \frac{\sin(\omega)}{\omega}\right)$ depends on the inter-nuclear separation D , and the recoil momentum of the ion p_{recoil} ,

$$\omega = p_{recoil} D. \quad (2)$$

Assuming that electronic transition only occurs in the Franck-Condon region, for the vibrational and electronic ground state of the neutral Hydrogen molecule, D can be anywhere from approximately $1.15 a_0$ to $1.7 a_0$. At very small projectile energies, where the collision time t_c is much larger than the vibration time t_v , a Franck-Condon transition is not necessarily a good assumption. However, at the projectile energy studied here $t_c \ll t_v$ is realized and the equilibrium distance of $1.4 a_0$ is normally used for D .

This approximation has been moderately successful for electron-impact ionization of molecules averaged over all orientations [40-42] as well as for ion impact [34-35].

M3DW-EIS

The 3DW-EIS model has previously been applied to heavy particle ionization of atoms [1, 8] and here we generalize the method to ionization of molecules. The triply differential cross section [1, 8] is given by

$$\frac{d^3\sigma}{d\Omega_p d\Omega_e dE_e} = N(2\pi)^4 \mu_{ie} \mu_{PM}^2 \frac{k_a k_b}{k_i} |T_{fi}|^2, \quad (3)$$

where μ_{ie} is the reduce mass of the target ion-electron system, μ_{PM} is the reduce mass of the projectile-target system, N is the number of indistinguishable electrons in the target, k_a is the final momentum of the projectile after the collision, k_b is the momentum of the outgoing electron, and k_i is the initial momentum of the projectile before the collision. The exact transition matrix was given by Jones and Madison *et al.* [2] using the two potential formulation

$$T_{fi} = \langle \chi_f^- | V_i | \beta_i \rangle + \langle \chi_f^- | W_f^+ | (\psi_i^+ - \beta_i) \rangle. \quad (4)$$

Here β_i is the asymptotic initial state of the system, V_i is the initial state interaction between the projectile and the target, χ_f^- is an approximate final state wave function, W_f^+ is the final state perturbation, and ψ_i^+ is the exact initial-state wave function which must be approximated. In terms of perturbation series, the first term of Eq. (4) represents first order and the second term represents all higher orders.

The asymptotic initial state wavefunction β_i is given by

$$\beta_i = \phi_{PW}(\vec{k}_i) \psi_{\text{target}}, \quad (5)$$

where $\phi_{PW}(k_i)$ is an initial state plane wave for the incident projectile, and ψ_{target} is the initial state wave function of the molecular target which can be of arbitrary size (the present formalism is not limited to H₂). In the usual manner we assume that the passive electrons do not participate in the collision so that their wavefunctions do not change during the collision. If we let (ξ) represent the coordinates of all the passive electrons, the final state ion wave function for the

molecule $\psi_{ion}(\xi, \mathbf{R})$ will depend on (ξ) and the orientation of the molecule (\mathbf{R}) , while the initial target wave function $\psi_{target}(\xi, \mathbf{R}, \mathbf{r}_b)$ will depend both on (ξ, \mathbf{R}) and the active electron coordinate (\mathbf{r}_b) (we assume that the collision time is sufficiently short that the final-state orientation is the same as the initial-state orientation). The effective one-electron wavefunction for the active electron is given by

$$\langle \psi_{ion}(\xi, \mathbf{R}) | \psi_{target}(\xi, \mathbf{R}, \mathbf{r}_b) \rangle = \psi_{Dyson}(\mathbf{r}_b, \mathbf{R}), \quad (6)$$

where $\psi_{Dyson}(\mathbf{r}_b, \mathbf{R})$ is the so-called Dyson orbital. Most of the experimental data reported so far for heavy particle ionization represent an average over all molecular orientations and the proper way to calculate an average over orientations would be to evaluate the T-matrix for a sufficiently large number of orientations that a numerically accurate average could be calculated. Due to the excessive computer time required for this process, Gao *et al.* [43] proposed the OAMO (orientation averaged molecular orbital) approximation. The essence of the OAMO approximation is to average the molecular orbitals instead of averaging the cross sections. Gao showed both analytically and numerically that the OAMO approximation was valid for ionization of ground σ_g states of molecules as long as the momentum transferred to the nucleus is less than 1 a.u (which is the case for many existing experiments) and it has been very successful for electron-impact ionization of H₂ [44]. In this approximation, the calculation of molecular cross sections reduces to the same level of difficulty as calculating atomic cross sections. Consequently, the asymptotic initial state can be written as

$$\begin{aligned} \beta_i &= \phi_{PW}(\vec{k}_i) \psi_{Dyson}^{OAMO}(r_b) \\ &= \frac{e^{i\vec{k}_i \cdot \vec{r}_a}}{(2\pi)^{3/2}} \psi_{Dyson}^{OAMO}(r_b), \end{aligned} \quad (7)$$

where $\psi_{Dyson}^{OAMO}(r_a)$ is the Dyson orbital averaged over all orientations.

To perform a theoretical calculation using Eq. (4) requires making a choice for the final state wavefunction χ_f^- (which determines the perturbation W_f) and making an approximation for the exact initial state wavefunction ψ_i^+ . The simplest approximation for the initial state wavefunction is to approximate it as the asymptotic initial state $\psi_i^+ \approx \beta_i$ and this approximation is normally called the Plane wave Born Approximation or the first Born approximation (FBA). In the FBA, the higher order terms in Eq. (4) cancel and the FBA T matrix [12] becomes

$$T_{fi}^{FBA} = \langle \chi_f^- | V_i | \beta_i \rangle. \quad (8)$$

The final state wavefunction χ_f^- must also be approximated and, in the FBA, the scattered heavy projectile is normally also represented as a plane wave. However, a plane wave would not be a good choice for the ejected electron since the energies are typically fairly small. Consequently, a better choice would be a distorted wave in the field of the molecule

$$\chi_f^- = \phi_{PW}(\vec{k}_a) \phi_{DW}(\vec{k}_b), \quad (9)$$

where $\phi_{DW}(\vec{k}_b)$ is a distorted wave for the ejected electron in the field of the molecular target. The distorted waves are solutions of the Schrödinger equation

$$\left(-\frac{1}{2}\nabla^2 - U_{ion} + \frac{k_b^2}{2}\right) \phi_{DW}(\vec{k}_b) = 0, \quad (10)$$

where U_{ion} is a spherically symmetric potential for the molecular ion. The spherically symmetric distorting potentials for molecules are calculated similar to the atomic case. The starting point is the molecular charge density for the molecular ion which is obtained from the Dyson orbitals

$$\rho(\mathbf{r}, \mathbf{R}) = \sum_{k=1}^m n_k \left| \phi_{Dyson}^k(\mathbf{r}, \mathbf{R}) \right|^2, \quad (11)$$

where m is the number of orbitals in the molecule, n_k is the occupation number of each orbital, and the density depends on the orientation of the molecule. To obtain the spherically symmetric distorting potential, we average Eq. (11) over all orientations to form the average radial charge density

$$\rho^{ave}(r) = \langle \rho(\mathbf{r}, \mathbf{R}) \rangle, \quad (12)$$

where the brackets denote taking an average over all orientations. The spherically symmetric static distorting potential representing the interaction between the projectile and the target molecular electrons is then found in the standard way using the average radial density

$$U_{ele}(r_a) = \left\langle \int \frac{\rho^{ave}(r) d\mathbf{r}}{|\mathbf{r}_a - \mathbf{r}|} \right\rangle, \quad (13)$$

where now the brackets denote taking an average over all angular locations for \mathbf{r}_a . The final state static distorting potential is the sum of the electronic contribution plus the nuclear contribution

$$U_{ion} = U_{ele} + U_{nuc}. \quad (14)$$

Here U_{nuc} is the contribution from the molecular nuclei. Just as we need to average over all orientations to obtain the potential for the molecular electrons, we also need to average over all orientations for the nuclei. Averaging a nucleus over all orientations is equivalent to placing the nuclear charge on a thin spherical shell which has a radius equal to the distance from the nucleus to the center-of-mass. Consequently, U_{nuc} is a sum of potentials for concentric spheres for each nucleus centered at the center-of-mass. For H_2 , we have a charge of +2 on a shell with a radius of $0.7 a_0$.

With these choices, the T matrix for the FBA becomes

$$T_{fi}^{FBA} = \left\langle \phi_{PW}(\vec{k}_a) \phi_{DW}(\vec{k}_b) | V_i | \phi_{PW}(\vec{k}_i) \psi_{Dyson}^{OAMO}(r_a) \right\rangle. \quad (15)$$

If the exact initial state wavefunction is approximated as something better than a plane wave, the second term of Eq. (4) does not vanish and we get contributions from all higher order terms in perturbation series. One approximation that has been very successful for heavy particle ionization is the Eikonal initial state wave function which was introduced by Crothers and McCann [11]. In this approximation, the asymptotic form of the Coulomb interaction between the projectile-active electron and projectile-ion are taken into consideration. The Eikonal initial state is given by

$$\begin{aligned} \psi_i^+ &\approx \phi_{EIS}(\vec{k}_i) \psi_{Dyson}^{OAMO}(r_b) \\ &= \frac{e^{i\vec{k}_i \cdot \vec{r}_a}}{(2\pi)^{3/2}} \left\{ \exp\left[i \frac{Z_p}{v_i} \ln\left(\frac{v_i r_a - \vec{v}_i \cdot \vec{r}_a}{v_i r_{ab} - \vec{v}_i \cdot \vec{r}_{ab}} \right) \right] \psi_{Dyson}^{OAMO}(r_b) \right\}, \end{aligned} \quad (16)$$

where Z_p is the charge of the projectile (in this case a proton), $r_{ab} = |\vec{r}_a - \vec{r}_b|$, and \vec{v}_i is the velocity of the projectile with respect to the target molecule

$$\vec{v}_i = \frac{\vec{k}_i}{\mu_{p1}}. \quad (17)$$

In the standard CDW-EIS (coulomb distorted wave-Eikonal initial state) approximation that has been used extensively for heavy particle scattering from atoms, the projectile is treated semi-classically and the terms involving β_i in Eq. (4) are neglected. In our approach which we have labeled M3DW-EIS, the projectile is treated fully quantum mechanically and we evaluate all terms in the T-matrix. For the final 3-particle state, we include the Coulomb interactions for all 2-particle subsystems as shown in Fig. 1

The final state is approximated [1, 8] as

$$\chi_f^- = \phi_{CW_p}(\vec{k}_a) \phi_{DW}(\vec{k}_b) C_{e-p}, \quad (18)$$

where $\phi_{CW_p}(\vec{k}_a)$ is a Coulomb wave for an effective charge of +1 and C_{e-p} is the Coulomb interaction between the electron and heavy projectile. The final state interaction between the projectile and ejected electron is normally called the post collision interaction (PCI), and including C_{e-p} in the final state wavefunction means that PCI is contained to all orders of perturbation theory. Explicitly the ϕ_{CW_p} is given by

$$\phi_{CW_p}(\vec{k}_a) = (2\pi)^{-3} \exp(i \vec{k}_a \cdot \vec{r}_a) C^-(\gamma_a, \vec{k}_a, \vec{r}_a), \quad (19)$$

where the Sommerfeld parameter $\gamma_a = \frac{Z_p}{v_a}$ with v_a final relative velocity of the projectile and target ion and

$$C^-(\gamma, \vec{k}, \vec{r}) = N(\gamma) {}_1F_1(i\gamma, 1, -i(kr + \vec{k} \cdot \vec{r})). \quad (20)$$

Here ${}_1F_1$ is the confluent hypergeometric function, and the Gamow factor $N(\gamma) = e^{-\pi\gamma/2} \Gamma(1 - i\gamma)$ where Γ is the gamma function. Likewise the C_{e-p} interaction is given by

$$C_{e-p} = C^-(\gamma_{ab}, \vec{k}_{ab}, \vec{r}_{ab}), \quad (21)$$

where $\gamma_{ab} = -\frac{Z_p}{v_{ab}}$ with v_b and v_{ab} is the final relative velocity of the ejected electron and target ion subsystem, and the projectile and ejected electron subsystem respectively.

Finally we need to find the perturbation W_f which depends on the choice for χ_f^- and which can be calculated from

$$W_f = \frac{1}{\chi_f^-} (H - E) \chi_f^-, \quad (22)$$

where H is the total Hamiltonian for the system and E is the total energy of the whole system.

For χ_f^- of Eq. (18) the perturbation is given by [16]

$$W_f = \bar{K}(-\frac{Z_p}{v_{ab}}, \vec{k}_{ab}, \vec{r}_{ab}) \cdot [\bar{K}(-\frac{1}{v_b}, \vec{k}_b, \vec{r}_b) - \bar{K}(\frac{Z_p}{v_a}, \vec{k}_a, \vec{r}_a) / \mu_{PI}], \quad (23)$$

where the perturbation factor is given as [1, 8]

$$\bar{K}(\gamma, \vec{k}, \vec{r}) = \gamma k \left[\frac{{}_1F_1(1 + i\gamma, 2, -ikr - i\vec{k} \cdot \vec{r})}{{}_1F_1(i\gamma, 1, -ikr - i\vec{k} \cdot \vec{r})} \right] (\hat{k} + \hat{r}). \quad (24)$$

To get DDCS-P, we have to numerically integrate the TDCS over the ejected electron scattering angles.

IV. Results

In Figure 2 we present three dimensional TDCS for an ejected electron energy of 14.6 eV and scattered projectile angles increasing from 0.24 mrad to 0.84 mrad. The scattering plane, is determined by the incident and scattered momentum vectors for the projectile (\vec{k}_i, \vec{k}_a) . The momentum transfer vector $\vec{q} = \vec{k}_i - \vec{k}_a$ lies in the same plane. For these four cases, the cross sections exhibit the characteristic double lobe structure well known from (e,2e) studies [45]. The large lobe on the right is known as the binary peak, since it has a maximum either at or near the angle corresponding to a classical collision between the projectile and an atomic electron at rest with the residual ion playing a passive role (i.e. near the direction of \vec{q}). The small lobe on the left near the direction of $-\vec{q}$ is known as the recoil peak and it is formed due to backscattering from the nucleus [2, 17]. For this peak, the electron is initially moving in the $+\vec{q}$ direction and then is backscattered by 180° from the nucleus. However, the projectile – residual target ion interaction can also contribute to the recoil peak [17]. For atomic targets, the binary lobe is usually completely dominant when the projectile scattering angle is large. Surprisingly, for the molecular hydrogen target studied here, we observe the exact opposite behavior in the calculated TDCS: For case (a), there is very small recoil peak when the projectile scattering angle is small, but its intensity relative to the binary peak intensity increases with increasing scattering angle

In Fig. 3, TDCS results are presented for an ejected electron which has the same velocity as the final state projectile and a small scattering angle for the proton. For this case, all one sees is a huge cross section in the forward direction which looks more like a “shooting star”. During the 1970’s Crooks and Rudd [36] experimentally found an enhancement in the DDCS-E cross sections when the ejected electron’s velocity matched the projectile’s velocity. They attributed this observation to the strong attraction an electron would have when moving close to the proton and with the same speed. They called this enhancement electron capture to the continuum (ECC) since the electron appears to be ‘captured’ but it is not bound. Furthermore, Vajnai *et al.* [46] and Schulz *et al.*[47] showed that, in this process, both the projectile and the ejected electron are strongly focused along the initial projectile beam axis leading to a strong narrowing of the angular distribution. From Fig. 3, it is seen that ECC is a completely dominant effect for the velocity matching case since all the electrons are drawn into a very narrow cone in the forward direction where the proton is located. The cross section does not have the traditional binary and

recoil lobe shape. The binary peak would be expected to be at about 15° and the observed peak is at 0° so this peak may be a shifted binary peak or perhaps a new peak that completely swamps the binary peak (see below).

In Fig 4 the TDCS is shown as a function of the electron ejection angle in the scattering plane on a semi-log scale. For an ejected electron energy of 14.6eV and projectile scattering angle of 0.34 mrad, the classical angle for a binary collision (direction of \vec{q}) is 60° and there is a very broad maximum in the TDCS near this angle. However, for the case of the equal velocities (i.e. 41.6eV) and a scattering angle of 1.03 mrad, the classical angle for a binary collision is 70° . Although the TDCS exhibits a broad shoulder for larger angles, there is no structure at 70° and the cross section for 0° is two orders of magnitude larger. This shoulder in the vicinity of the classical peak is not even observable on a linear plot! This again dramatically illustrates the importance of ECC. It also shows that ECC does not just lead to a narrowing and shift of the binary peak, but it actually results in a separate peak structure apart from the binary and recoil lobes that, to the best of our knowledge, has not been predicted yet by any other theory. Experimental triply differential data for ion impact close to the matching velocity are not yet available.

In Fig 5, the DDCS-P results are compared with absolute experimental data for fixed ejected electron energies of 14.6eV, 34.6eV, 37.6eV, 41.6eV, 49.6eV and 54.6eV respectively. The experimental data shows some structure for ejected electron energies of 14.6eV, 34.6eV and 54.6eV. The other energies do not have any particular structure and the cross sections fall off rapidly. These structures were attributed by Alexander *et al.* [3] to the presence of interference effects resulting from the two-nuclear scattering centers - similar to the double slit experiment for light. Each figure contains FBA results calculated using Eq. (15), CDW-EIS-Rayleigh, and M3DW-EIS. Recall that CDW-EIS-Rayleigh are results obtained for ionization of atomic hydrogen multiplied by the interference factor in Eq (1). It is seen that the FBA results do not show any structure and are in very poor agreement with the experiment even though two-center molecular bound states and molecular distorted waves were used for the ejected electron. Since the FBA contains no interaction of the projectile with the nuclei, and no multiple interactions of the projectile with the active electron, this strongly indicates that the observed structure must come from higher order interactions between the projectile and the target.

The CDW-EIS-Rayleigh results are in much better agreement with experiment, at least for energies of 14.6, 34.6, 37.6, and 54.6 eV. However, the CDW-EIS-Rayleigh calculation does not predict any of the structure seen in the experimental data.

The present M3DW-EIS results predict a structure almost identical to the experiment for 14.5 eV ejected electrons and a small structure for 34.6 eV ejected electrons. However, the magnitude of the M3DW-EIS cross sections were consistently lower than the data. We noticed that if we multiply the M3DW-EIS results by a factor of 4.5, we get very good agreement with the experimental data for energies 14.6eV, 34.6eV, 37.6eV and 54.6eV. For the two other energies, multiplying the DDCS by the 4.5 factor produces good agreement for the smaller scattering angles while overestimating experiment for the larger angles. However the important point is that the experimental data contains statistically significant structure only for electron ejection angles of 14.6, 34.6 eV, and possibly 54.6 eV and the M3DW-EIS predicts the same structure in exactly the same angular range for the two smallest energy losses. The lack of structure for 37.6, 41.6 and 49.6 eV is attributed to the structure being eliminated due to the large effect of ECC for the velocity matching and nearly matching conditions. It is interesting to note that the worst agreement between the M3DW-EIS and experiment occurs for the velocity match of 41.6 eV where our TDCS show a huge ECC effect to the extent that we overestimate experiment for the larger scattering angles (after normalization). Although experiment does show some structure again for 54.6 eV which was not found in the M3DW-EIS calculation, the structure occurs where the cross sections are very small and the error bars are significantly larger than for 14.6 and 34.6 eV. It is thus not clear how significant these discrepancies between experiment and theory are.

Concerning the 4.5 difference in magnitude, the experimental data were normalized by integrating the doubly differential cross sections over projectile scattering angles and the singly differential cross sections were normalized to the absolute values recommended by Rudd *et al.* [48]. The theoretical results integrated over projectile scattering angles differ from the Rudd *et al.* [48]. recommended values by a factor of about 3.7. Consequently, it appears that the M3DW-EIS yields the proper shape but not magnitude. On the other hand, one would have to get the important physics of the collision correct to get the proper shape whereas the magnitude is just an overall constant factor. As a result, we would conclude that the M3DW-EIS contains the important physics for the molecular collision.

Alexander *et al.* [3] found that they could get a much better agreement between the CDW-EIS-Raleigh calculation and experiment if they assumed that the molecules were aligned at the time of ionization. Consequently they discussed the possibility that there were strongly preferred alignments for different projectile scattering angles. However, within the model presented here,

such a preference in orientation is not needed to explain the experimental data since we find the observed structure and our OAMO averaging procedure assumes equal weight for all orientations.

V Conclusions

We have calculated M3DW-EIS (molecular 3 body distorted wave - Eikonal initial state) triply differential cross sections (TDCS) and doubly differential cross sections differential in the projectile scattering angle (DDCS-P) for single ionization of H_2 by 75 keV proton impact. In this work we demonstrated that, without assuming that any molecular orientation is favored at specific scattering angles, the interference structure found in the experimental measurements was predicted by the M3DW-EIS. For the case of equal velocity and near equal velocity, very large ECC effects were found – so large that it completely dominated normal cross section features like binary and recoil peaks. For some unknown reason, the M3DW-EIS results are a factor of 4.5 lower than experiment for all the measured ejected electron energies. Consequently, the M3DW contains the important physics determining the shape of the cross section but incorrectly predicts the magnitude which is an overall constant normalization factor.

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Figure Captions

Fig: 1. (Color online) Interactions contained in 3DW final state wavefunction

Fig: 2. (Color online) Three dimensional images of the triply differential Cross section for single ionization of the Hydrogen molecule by proton impact. The theoretical calculations are M3DW-EIS results and the ejected electron energy is 14.6eV The proton scattering angle is: (a) 0.24mrad (b) 0.34mrad (c) 0.51mrad. and (d) 0.84mrad

Fig: 3. (Color online) Three dimensional images of the triply differential Cross section for single ionization of the Hydrogen molecule by proton impact. The theoretical calculations are M3DW-EIS results. The ejected electron energy is 41.6eV and the proton scattering angle of 0.1mrad.

Fig: 4. (Color online) The Triply Differential Cross sections in the scattering plane for the ejected electron energy (a) 14.6eV ejected electron energy and 0.34mrad proton scattering angle. (b) 41.6eV ejected electron energy and 1.03mrad proton scattering angle.

Fig: 5. (Color online) DDCS-P for ionization of H_2 by 75keV proton impact as a function of projectile scattering angle. The energy of the outgoing electron are shown in each figure. Experimental results of Alexander *et al.* [3]. Theory: dash-dot (pink) CDW-EIS-Rayleigh, dash (red) M3DW-EIS times 4.5. Solid (blue) M3DW-EIS. Long dash-short dash (black) FBA calculation.

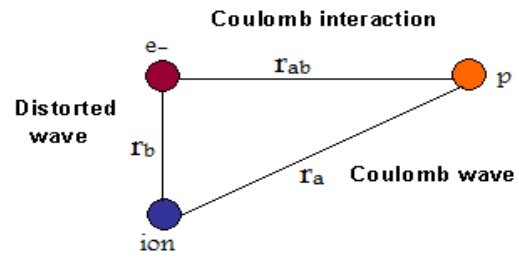


Figure 1 AZ10535 04FEB2011

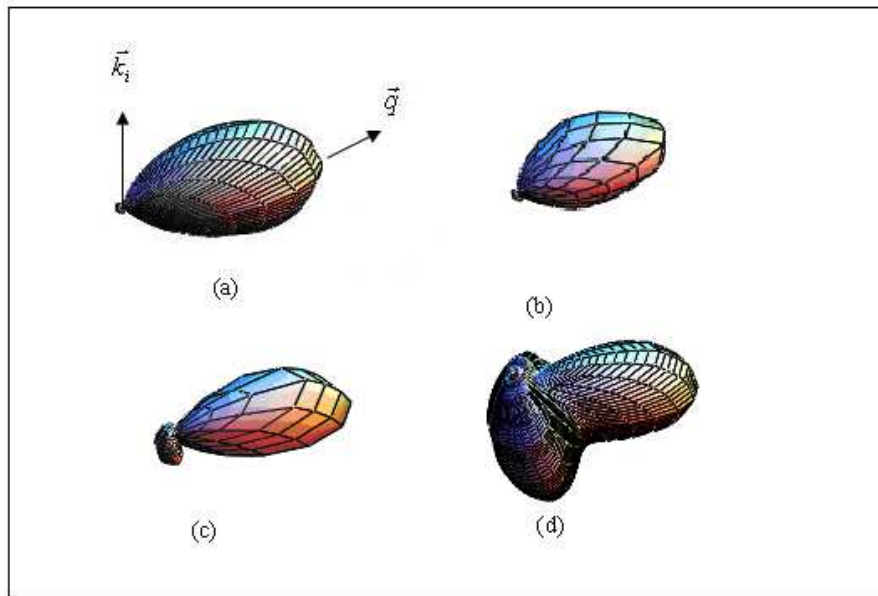


Figure 2 AZ10535 04FEB2011



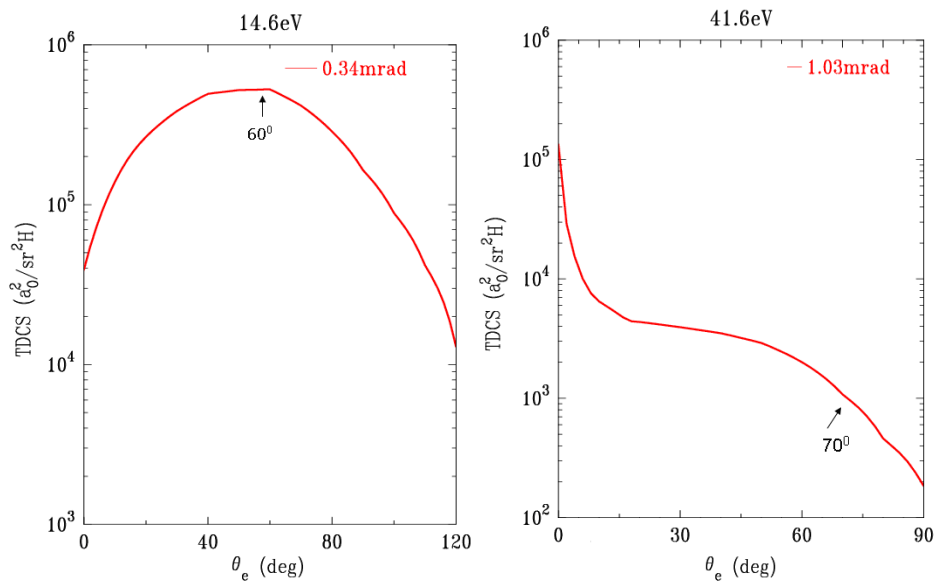


Figure 4 AZ10535 04FEB2011

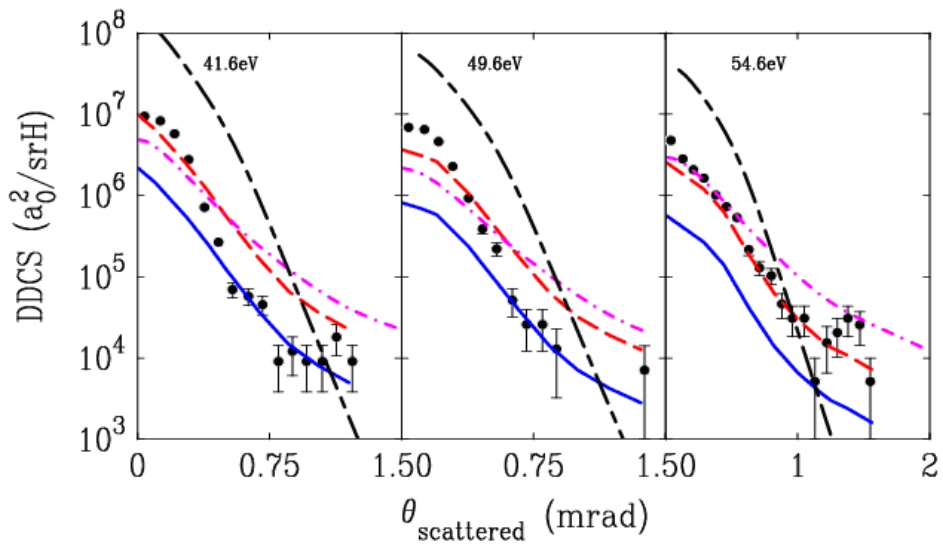
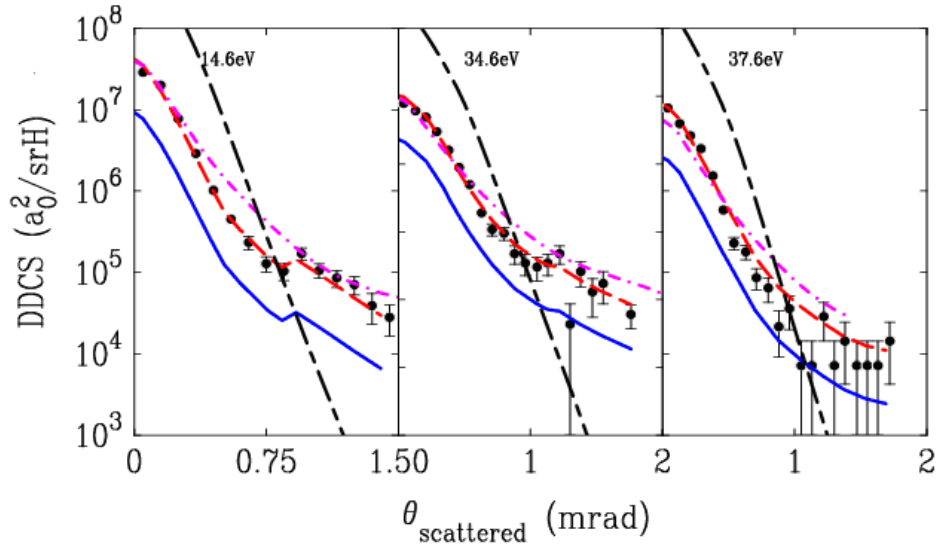


Figure 5

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