PHYSICAL REVIEW A **78**, 060701(R) (2008)

# Interference effects due to projectile target nucleus scattering in single ionization of $H_2$ by 75-keV proton impact

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(Received 6 July 2008; published 3 December 2008)

Doubly differential cross sections (DDCSs) for single ionization of molecular hydrogen by 75-keV proton impact have been measured and calculated as a function of the projectile scattering angle and energy loss. Interference structures are observed in the scattering angular dependence of the DDCSs, which disappear, however, at electron speeds near the projectile speed. The comparison to our calculations shows that the projectile-target nucleus interaction plays a central role. Furthermore, our data suggest that for a given scattering angle, ionization favors well-defined molecular orientations.

DOI: 10.1103/PhysRevA.78.060701 PACS number(s): 34.50.Gb, 34.50.Bw, 34.50.Fa

Fragmentation processes in atomic and molecular collisions, such as ionization of the target atom, are particularly well-suited to the study of the fundamentally important few-body problem [1–3]. Consequently, multiple differential single ionization cross sections have been measured for a broad range of collision systems [3]. For small perturbations $\eta$ ($\eta$=projectile charge/velocity ratio) and simple atomic targets a qualitative understanding of the reaction dynamics has begun to emerge [4]. However, for increasing perturbation serious discrepancies between theory and experiment remain [5].

Theoretical difficulties not only increase with increasing perturbation, but also with increasing target complexity. For molecular targets, the complexity of the wave function makes theoretical analysis more cumbersome. Conversely, the two (or multiple) center potential of the molecule also makes the physics more interesting. Since one cannot distinguish from which center the scattered wave is diffracted, both contributions need to be treated coherently which may lead to observable interference patterns. The fundamental importance of interference effects in the general context of quantum mechanics has been pointed out by several authors, e.g., [6,7]. Interference structures have been reported in double differential electron energy spectra for the simplest neutral molecular system $H_2$ [6]. Since then coherence effects in ionization of molecular targets have been studied extensively, e.g., [7–12]. More recently, such effects were also investigated for molecular ionic projectiles colliding with a neutral target atom [13]. The structures in the electron energy spectra of Ref. [6] were difficult to discern in the absolute cross sections; only in ratios to theoretical or experimental [6,8,10,12] atomic hydrogen cross sections could they be clearly identified.

The interference term contains the phase angle which depends on three quantities: the molecular orientation, the electron momentum $\mathbf{p_e}$, and the momentum transfer $\mathbf{q}$ from the projectile to the target. It was shown that averaging over the molecular orientation does not completely destroy the interference pattern [6,14]. However, integration over $\mathbf{q}$, which is inherent to the double differential electron spectra, tends to "wash out" the phase factor such that an interference pattern is not easily detectable in the cross sections. On the other hand, double differential cross sections as a function of $\mathbf{q}$, or equivalently the projectile scattering angle, and ejected electron energy are not integrated over $\mathbf{q}$, but instead over the electron solid angle. If the phase angle is more sensitive to $\mathbf{q}$ than it is to $\mathbf{p_e}$, an oscillating interference pattern may be more pronounced in the projectile scattering angle dependence of the cross sections than in the ejected-electron angle dependence.

In this Rapid Communication we report on measured and calculated doubly differential cross sections for single ionization of $H_2$ by 75-keV proton impact as a function of the scattered projectile angle $\theta$ and ejected electron energy $E_e$. Indeed, pronounced structures are observed in the projectile scattering angle dependence.

The experiment was performed at the Missouri University of Science and Technology. A 5-keV proton beam was generated from a hot cathode ion source, accelerated to 75 keV, and collimated by a set of slits 0.1 mm $\times$ 0.1 mm in size. The protons crossed a cold ($T \approx 2$ K) molecular hydrogen target beam from a supersonic jet. The recoil ions were extracted from the interaction region by a weak electric field and arrived on a channel plate detector, but were not momentum analyzed in this experiment.

The scattered proton beam was decelerated by 70 keV and energy analyzed by an electrostatic parallel plate analyzer [15] yielding the projectile energy loss $\Delta E$. The projectiles were then detected by a position sensitive detector where the projectile scattering angle was determined from the position on the detector. The projectile and recoil-ion detectors were set in coincidence. The coincidence setup was important as it cleaned the recorded data essentially completely from background signals resulting, e.g., from the residual gas in the vacuum. The absolute energy resolution was approximately $\pm 1.5$ eV and the scattering angle resolution was better than $\pm 50$ $\mu$rad. Since $\Delta E$ is equal to the ejected electron energy $E_e$ plus the ionization potential of the $H_2$

 

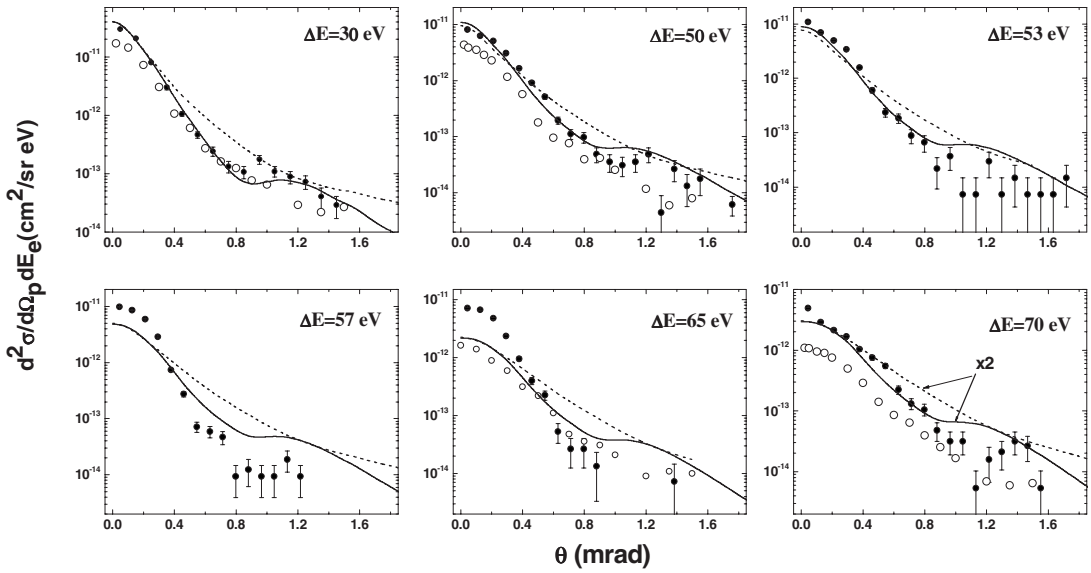


FIG. 1. DDCSs plotted as a function of the projectile scattering angle $\theta$ for fixed energy losses $\Delta E$ of 30, 50, 53, 57, 65, and 70 eV, respectively. Closed symbols: experimental data for $H_2$; open symbols: experimental data for helium; dashed curves: CDW-EIS calculation; and solid curves: CDW-EIS calculation with fixed molecular orientation (for details see text).

molecule $I_P$=15.4 eV, the coincident projectile position spectrum is directly proportional to the doubly differential cross sections $d^2\sigma/d\Omega_p dE_e$ (DDCSs), where $\Omega_p$ is the projectile solid angle.

In Fig. 1 the DDCSs are plotted as a function of the projectile scattering angle for fixed energy losses of 30, 50, 53, 57, 65, and 70 eV, respectively. The DDCSs were normalized to recommended total cross sections [16]. The data (solid circles) at each fixed energy loss generally show the typical behavior of such cross sections: they steeply decrease with increasing scattering angle. However, apart from this expected trend at larger scattering angles structures appear for energy losses of 30 and 50 eV and again at an energy loss of 70 eV. At energy losses near 57 eV, however, the large scattering angle structure disappears. For a helium target, in contrast, no structures were found in the DDCSs at any energy loss (open circles in Fig. 1).

In order to analyze the appearance and disappearance of the large-angle structure further we present in Fig. 2 the ratios $R$ between the measured DDCSs for $H_2$ and twice the theoretical DDCSs for atomic hydrogen for energy losses of 30, 50, 57, and 70 eV. The latter cross sections were calculated using the continuum distorted wave eikonal initial state (CDW-EIS) approach including the projectile-residual target ion (PI) interaction [17]. The structures already observed in the cross sections of Fig. 1 become even more prominent in $R$ and, in fact, a second structure at smaller angles (around 0.3 mrad) becomes visible. Likewise, the (nearly) complete absence of the structure at large scattering angles around an energy loss of 57 eV is more evident in $R$ as well.

It has been shown that, to a good approximation, the triple differential cross sections $d^3\sigma/d\Omega_p d\Omega_e dE_e$ (TDCSs) for $H_2$ averaged over all molecular orientations can be expressed in terms of the TDCSs for atomic hydrogen H by [6,14,17]

$$(\mathrm{TDCS}_{\mathrm{H}_2}) = 2\mathrm{TDCS}_{\mathrm{H}}\left(1+\frac{\sin(\chi)}{\chi}\right). \quad (1)$$

Here, the phase factor in the interference term $(1+\frac{\sin(\chi)}{\chi})$ is $\chi=p_{\mathrm{rec}}D$, where $p_{\mathrm{rec}}$ is the magnitude of the recoil-ion momentum and $D$ is the internuclear distance in the molecule. Therefore the double differential ratios of Fig. 2 can be viewed as the interference term averaged over all recoil-ion momenta.

The structures in $R$ can be interpreted as an interference due to coherent scattering from the two centers of the molecule. One important question to answer is which quantities mainly determine the phase factor. The recoil-ion momentum, the magnitude of which enters in $\chi$, is given by $\mathbf{p}_{\mathrm{rec}}=\mathbf{q}-\mathbf{p}_{\mathbf{e}}$. It has been argued that the double differential electron energy spectra are dominated by small $q$ collisions, so that there the interference pattern is basically determined by the ejected electron [6]. Here, we observe interference maxima at scattering angles larger than 1 mrad corresponding to momentum transfers of larger than 3–3.5 a.u. (depending on $\Delta E$), while the electron momentum ranges from 1 to 2 a.u. The occurrence of interference maxima at large scattering angles thus suggests that the PI interaction plays an important role in the interference pattern.

The primary role of the PI interaction in the interference is supported by our theoretical calculations. The dotted curves in Fig. 2 were computed using Eq. (1) with the atomic hydrogen cross section calculated in the plane-wave-Born (PWB) approximation. The postcollision interaction (PCI) between the scattered projectile and the ejected electron is accounted for following the procedure of Salin [18], so we label this curve as PWB-PCI. However, the PI interaction is not included at all in the PWB-PCI since the projectile is treated as a plane wave. In contrast, the CDW-EIS calculation (dashed curves) [17] includes both PCI and the PI inter-

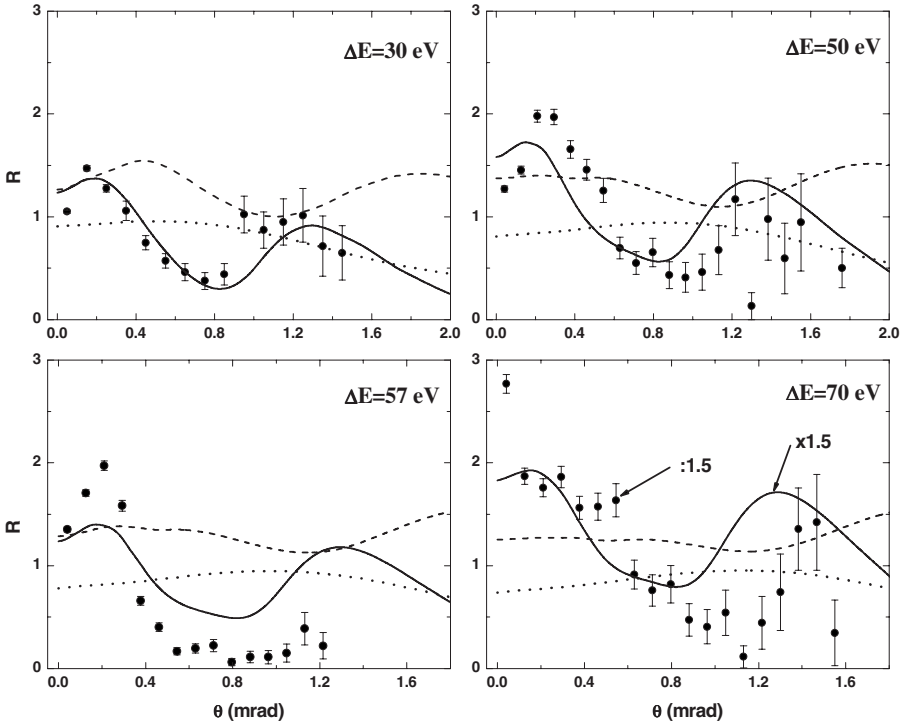


FIG. 2. Ratios between measured DDCSs for $H_2$ and twice the theoretical DDCSs for atomic hydrogen for energy losses of 30, 50, 57, and 70 eV. Dotted curves: PWB-PCI calculation; dashed curves: CDW-EIS calculation; and solid curves: CDW-EIS calculation with fixed molecular orientation (for details see text).

action. Effective charges, both in the initial and final electronic states, were used in order to account for the presence of the "passive" remaining electron in the $H_2$ molecular target. Our CDW-EIS calculation of the DDCSs for a helium target reproduces both a calculation employing the same model [19] and measured data [20]. In accordance with the experimental data the calculation does not yield any structures for helium. The PWB-PCI calculation is in poor agreement with the data. Although the CDW-EIS is not in good quantitative agreement either, qualitatively a structure with two maxima is reproduced. The distance between the maxima is in good accord with experiment as well; however, the entire interference pattern is systematically shifted toward larger angles in the calculation and less pronounced than in the data. If the PI interaction is removed from the CDW-EIS model, poor agreement similar to the PWB-PCI calculation is obtained. Overall, it appears that the PI interaction is needed in theory in order to obtain at least qualitative agreement with the data.

In all calculations the cross sections are averaged over all molecular orientations using the approximation that each orientation contributes equally to ionization. We can get a crude estimate of the validity of this approximation from the data. If the ionization amplitude was dominated by a specific molecular orientation $\phi$, the interference term (IT) would become $\mathrm{IT}=R=[1+\cos(\chi)]$, where now $\chi=\mathbf{p}_{\mathrm{rec}}\cdot\mathbf{D}=p_{\mathrm{rec}}D\cos(\alpha)$ [6]. Here, $\alpha$ is the angle between $\mathbf{p}_{\mathrm{rec}}$ and $\mathbf{D}$. Triple differential measurements [21,22] for 75-keV $p$+He show that for a fixed scattering angle and electron energy (which is the case in the present DDCSs) the direction of the ejected electrons is well-determined within a narrow angular range. Since $\mathbf{p}_{\mathrm{rec}}=\mathbf{q}-\mathbf{p}_e$, $\mathbf{p}_{\mathrm{rec}}$ is also well-determined and a good estimate can be obtained from the triple differential data.

Using these estimated recoil-ion momenta obtained from the He data, along with the measured $R$ from Fig. 2, $\phi$ (which is composed of $\alpha$ and the recoil-ion direction) can be deduced from $R=[1+\cos(p_{\mathrm{rec}}D\cos(\alpha))]$. These estimated $\phi$ are plotted in Fig. 3 as a function of projectile scattering angle for energy losses of 30, 50, and 70 eV. These data suggest that for small scattering angles a transverse orientation (i.e., $\phi\approx 90°$) and for large scattering angles a longitudinal orientation (i.e., $\phi\approx 0°$) mostly contributes to the ionization amplitude. At the same time no significant differences between the data sets for different energy losses can be iden-

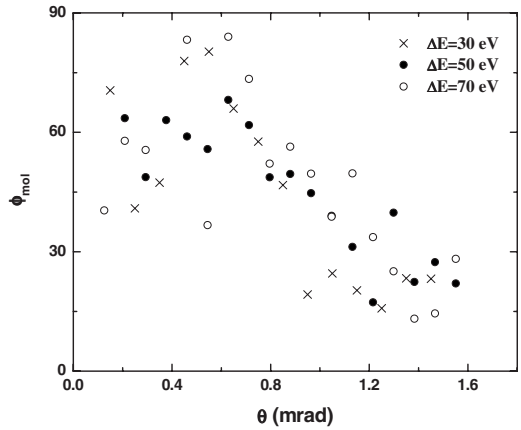


FIG. 3. Estimated molecular orientation $\phi$ plotted as a function of projectile scattering angle for energy losses of 30, 50, and 70 eV.

tified. Therefore the molecular orientation is essentially frozen for a fixed projectile scattering angle while integration over that angle, inherent to the cross-sections' differential in the energy and solid angle of the ejected electron, leaves $\phi$ largely undetermined. This could explain why the interference pattern is more pronounced in the projectile scattering angle dependence than in the electron energy dependence of the DDCSs. It could also be the reason that in the CDW-EIS calculation the structures are less pronounced than in the data. The solid curves in Figs. 1 and 2 are CDW-EIS calculations for molecules with a fixed orientation obtained from Fig. 3. It is seen that the fixed orientation calculations provide very good agreement with the data at 30 and 50 eV and significantly improved agreement with the shape of the data for the other energies (including 57 eV) compared to the calculation using averaged orientations (dashed curves in Figs. 1 and 2). The magnitudes are generally well-reproduced, as well. Only at 70 eV, both calculations underestimate the data by about a factor of 2

Finally, we discuss the disappearance of the large-angle structure at energy losses around 57 eV. This energy loss corresponds to an electron speed equal to the projectile speed. It is known that effects due to PCI maximize at an electron-to-projectile speed ratio of 1 [20]. In particular, in the DDCSs as a function of scattering angle, it leads to a significant narrowing of the angular distribution. Indeed, the measured DDCSs for 57 eV are significantly narrower than at smaller and larger energy losses. It seems likely that there is a connection between the disappearance of the large-angle structure and the maximized PCI at 57 eV. One possibility is that PCI moves significant flux from large to small angles. As a result, the interference maximum at large angles could be strongly suppressed. The worse agreement between experiment and theory at energy losses around 57 eV compared to the other energy losses would then suggest that PCI is somewhat underestimated by the CDW-EIS model. This interpretation is also supported by the observation that the calculation for fixed orientation once again reproduces the shape of the experimental DDCSs at 70 eV below approximately 1 mrad, while it is much too broad at 57 eV.

In summary, doubly differential cross sections for single ionization of molecular hydrogen by 75-keV proton impact have been measured and calculated as a function of the projectile scattering angle and energy loss. Interference structures are directly observed in the scattering angular dependence of the DDCSs. The interference pattern is more sensitive to the projectile angle than to the electron energy, suggesting that the projectile-target nucleus interaction plays a central role. The large-angle structures disappear at electron speeds near the projectile speed. This may be due to a focusing effect introduced by PCI. Furthermore, our data suggest that for a given scattering angle, ionization favors relatively well-defined molecular orientations.

This work was supported by the National Science Foundation under Grants No. PHY-0652519 and No. PHY-0757749. M.F.C. acknowledges the Visitor Program of the Max-Planck-Institut für Physik Komplexer Systeme for financial support. Z.S.M. acknowledges the Fulbright Program for financial support under Grant ID No. 68431290. R.D.R. is thankful for fruitful discussions with Fernando Martin and Omar Fojon.

[1] T. N. Rescigno *et al.*, Science **286**, 2474 (1999).
[2] M. Schulz *et al.*, Nature (London) **422**, 48 (2003).
[3] M. Schulz and D. H. Madison, Int. J. Mod. Phys. A **21**, 3649 (2006).
[4] M. Schulz *et al.*, Phys. Rev. A **76**, 032712 (2007).
[5] M. Schulz *et al.*, J. Phys. B **35**, L161 (2002).
[6] N. Stolterfoht *et al.*, Phys. Rev. Lett. **87**, 023201 (2001).
[7] D. Akoury *et al.*, Science **318**, 949 (2007).
[8] N. Stolterfoht *et al.*, Phys. Rev. A **67**, 030702(R) (2003).
[9] D. S. Milne-Brownlie *et al.*, Phys. Rev. Lett. **96**, 233201 (2006).
[10] D. Misra *et al.*, Phys. Rev. Lett. **92**, 153201 (2004).
[11] D. Misra *et al.*, Phys. Rev. A **74**, 060701(R) (2006).
[12] E. M. Staicu Casagrande *et al.*, J. Phys. B **41**, 025204 (2008).
[13] L. Schmidt *et al.*, Phys. Rev. Lett. **101**, 173202 (2008).
[14] M. E. Galassi *et al.*, Phys. Rev. A **70**, 032721 (2004).
[15] A. D. Gaus *et al.*, Rev. Sci. Instrum. **65**, 3739 (1994).
[16] *Atomic Data for Fusion*, edited by C. F. Barnett (Oak Ridge National Laboratory, D16, Oak Ridge, 1990), http://www-cfadc.phy.ornl.gov/redbooks/redbooks.html
[17] M. F. Ciappina and R. D. Rivarola, J. Phys. B **41**, 015203 (2008).
[18] A. Salin, J. Phys. B **2**, 631 (1969).
[19] V. D. Rodriguez and R. O. Barrachina, Phys. Rev. A **57**, 215 (1998).
[20] M. Schulz *et al.*, Phys. Rev. A **54**, 2951 (1996).
[21] N. V. Maydanyuk *et al.*, Phys. Rev. Lett. **94**, 243201 (2005).
[22] M. Schulz *et al.*, Phys. Rev. A **73**, 062704 (2006).