LETTER TO THE EDITOR

Interference effects in the ionization of H_2 by fast charged projectiles

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Abstract
A theoretical investigation of the experimentally observed (Stolterfoht N *et al*
2001 *Phys. Rev. Lett.* **87** 023201) interference effects in the double differential
cross sections for ionization of the hydrogen molecule by fast ion impact is
reported. The $H_2/2H$ cross section ratios as a function of the ejected electron
velocity show an oscillating pattern, for which Stolterfoht *et al* propose a
formula $C + G \sin(k\ D)/(k\ D)$, where k is the electron momentum and D the
internuclear separation in H_2. Our analysis in its simplest form leads instead to
a formula $C + G \sin(k_\parallel\ D)/(k_\parallel\ D)$ where $k_\parallel$ is the component of k parallel to
the projectile velocity. The presented theoretical model thus explains why
at 90° the interference pattern will be strongly suppressed. In addition to
the simplified analysis a numerical evaluation of a more accurate model is
presented, confirming the latter qualitative prediction.

The interference effects expected due to the two-centre character of a molecular target (H_2
or H_2^+) have been analysed both for collisions with electron transfer [1–3] and for the
photoionization processes [4, 5]. These analyses have shown that for both processes the
transition amplitudes can be expressed as coherent superpositions of one-centre amplitudes,
displaying interference patterns in the cross sections. These remain present even for cross
sections averaged over the orientation of the axis of the target molecule [2, 5].

Recently Stolterfoht *et al* [6] reported experimental results on electron emission induced by
fast charged ions impinging on H_2 molecular targets, demonstrating such a type of interference
effect also in the double differential cross sections for an ionization process. The interference
effect has been displayed by comparing the experimental differential ionization cross sections
for molecular targets with theoretical cross sections for the hydrogen atom (multiplied by two).
For the measured electron ejection angles, the ratio of differential cross sections measured for
H_2 and calculated for two hydrogen atoms oscillates as a function of increasing velocity of the
ejected electron.

In this letter we report a theoretical analysis of such a type of interference based on a simplified evaluation of ionization amplitudes. A qualitative theoretical model for the observed interference has already been given in the original paper of Stolterfoht *et al* [6], explaining the oscillations without any dependence on the electron ejection angle. Here we attempt a more detailed analysis of the interference patterns, leading to a prediction of electron ejection angle dependence of the oscillation shapes of these ratios. The present analysis is based on the impact-parameter formulation. For 60 MeV u^{-1} Kr^{34+} projectiles, used in the experiments by Stolterfoht *et al* [6], the ratio between the charge and the velocity of the projectile $Z_p/v \approx 0.7 < 1$, justifying the use of a first-order approximation at least in model calculations and a qualitative analysis.

The theoretical formulation follows closely the atomic target model of Hansen and Kocbach [7], here modified for molecular targets. The first-order transition amplitude for a projectile with impact parameter b and velocity v and a certain orientation of the molecular axis D can be written as

$$a(b, \hat{D}) = \frac{iZ_p}{v} \int_{-\infty}^{+\infty} dZ \, e^{iqZ} \left\langle \psi_k(r) \left| \frac{1}{|r - R|} \right| \psi_i(r, D) \right\rangle, \tag{1}$$

where r is the position vector of the active electron and $\psi_i(r, D)$ and $\psi_k(r)$ its initial and final states where k is the ejected electron momentum. The position of the projectile along the trajectory is $R = be_x + Ze_z$ with $Z = vt$. The origin is in the centre of the molecule. The well known minimum momentum transfer is $q = \Delta E/v$ where $\Delta E = E_i + k^2/2$ is the energy transfer to the electron.

We assume the initial state of the electron in the hydrogen molecule to be a linear combination of two 1s type atomic orbitals centred at each of the nuclei

$$\psi_i(r, D) = N(e^{-\alpha|r - D/2|} + e^{-\alpha|r + D/2|}) \tag{2}$$

where N is a normalization factor and α the effective charge. The final state wavefunctions for electrons of momentum k are approximated by plane waves

$$\psi_k(r) = \frac{1}{(2\pi)^{\frac{3}{2}}} e^{ikr}. \tag{3}$$

The evaluation of the amplitude equation (1) is based on the use of the Bethe integral

$$\frac{1}{|r - R|} = \frac{1}{2\pi^2} \int d^3s \, \frac{e^{-isR} e^{isr}}{s^2}. \tag{4}$$

Inserting the potential (4) and the final state (3) in the amplitude (1), and changing the momentum variable in the Bethe integral according to $s = p + k$, we get

$$a(b, \hat{D}) = \frac{iZ_p}{v} \frac{1}{2\pi^2} \frac{1}{(2\pi)^{3/2}} \int_{-\infty}^{+\infty} dZ \, e^{iqZ} \int d^3r \, e^{-ikr} \int d^3p \, \frac{e^{-i(p+k)R} e^{i(p+k)r}}{|p + k|^2} \psi_i(r, D). \tag{5}$$

Because of the simplified final state (3) the r-integral is simply the momentum space initial-state wavefunction $\phi_i(p, D)$

$$\frac{1}{(2\pi)^{3/2}} \int d^3r \, \psi_i(r, D) e^{ipr} = \phi_i(p, D), \tag{6}$$

and the transition amplitude becomes

$$a(b, \hat{D}) = \frac{iZ_p}{v} \frac{1}{2\pi^2} \int_{-\infty}^{+\infty} dZ \, e^{iqZ} e^{-ikR} \int d^3p \, \frac{\phi_i(p, D) e^{-ipR}}{|p + k|^2}. \tag{7}$$

Further analytical evaluation is made possible by the next approximation. If one considers only fast ejected electrons, their momentum $k \gg p$ in the region of p contributing significantly to the last integral in equation (7). Thus a peaking approximation,
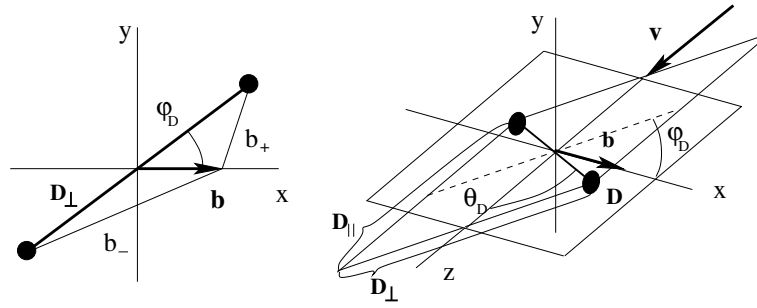
$$|p + k| \approx k \tag{8}$$

Figure 1. Definitions of the quantities, distances and angles.

is used, moving the denominator out. The remaining integral over p can be used as an inverse of the Fourier transform equation (6)

$$\frac{1}{(2\pi)^{3/2}} \int d^3p \, \phi_i(p, D) e^{-ipR} = \psi_i(R, D). \tag{9}$$

We decompose the electron momentum according to

$$kR = k_\perp b + k_\parallel Z, \tag{10}$$

where $k_\perp$ and $k_\parallel$ are the perpendicular and parallel components of the final momentum k, relative to the z axis, respectively. With this notation we obtain

$$a(b, \hat{D}) = \frac{iZ_p}{vk^2} \sqrt{\frac{2}{\pi}} e^{-ibk_\perp} \int_{-\infty}^{+\infty} dZ \, e^{i(q-k_\parallel)Z} \psi_i(R, D). \tag{11}$$

With the definition $R = be_x + Ze_z$, the initial state equation (2) becomes

$$\psi_i(R, D) = N(e^{-\alpha[(Z+D_\parallel/2)^2+b_+^2]^{1/2}} + e^{-\alpha[(Z-D_\parallel/2)^2+b_-^2]^{1/2}}), \tag{12}$$

where b_+ and b_- are the lengths of impact parameters relative to the two nuclei

$$b_\pm = (b^2 + D_\perp^2/4 \pm bD_\perp \cos\varphi_D)^{1/2}, \tag{13}$$

the components $D_\parallel$ and $D_\perp$ of the internuclear separation D and its azimuthal angle φ_D as indicated in figure 1.

As in the quoted paper [7], the integrations over Z can be performed analytically

$$\int_{-\infty}^{+\infty} dZ \, e^{iQZ} e^{-\alpha\sqrt{(Z\pm L)^2+b_\pm^2}} = e^{\mp iQL} \frac{2\alpha b_\pm}{\sqrt{\alpha^2+Q^2}} K_1(b_\pm\sqrt{\alpha^2+Q^2}) \tag{14}$$

where $K_1(x)$ denotes the modified Bessel function, $(q-k_\parallel)$ is represented by Q and the length $\frac{1}{2}D_\parallel$ by L. With this result and with the notation

$$\gamma = \sqrt{\alpha^2 + (q-k_\parallel)^2}, \tag{15}$$

the transition probability equation (11) for a given impact parameter and orientation of the molecular axis becomes

$$w(b, \hat{D}) = |a(b, \hat{D})|^2 = \frac{Z_p^2}{v^2k^4} \frac{2}{\pi} N^2 \frac{4\alpha^2}{\gamma^2} [(b_+K_1(b_+\gamma))^2 + (b_-K_1(b_-\gamma))^2$$

$$+ b_+K_1(b_+\gamma)b_-K_1(b_-\gamma)(e^{-i(k_\parallel-q)D_\parallel} + e^{+i(k_\parallel-q)D_\parallel})]. \tag{16}$$

For comparison with experiment the probabilities must be averaged over molecular orientation:

$$w(b) = \frac{1}{4\pi} \int d\varphi_D \int \sin\theta_D \, d\theta_D \, w(b, \hat{D}). \tag{17}$$

The averaged probabilities can be integrated over the impact parameter to obtain the differential cross sections

$$\frac{d\sigma}{d\Omega_k \, dk} = 2\pi \int b \, db \, w(b). \tag{18}$$

The integrations in equations (17) and (18) must be performed numerically and the results are discussed below.

However, the interference pattern can be analysed approximately, by recognizing that the most significant angular variation in equation (16) is contained in the fast oscillating factor of the interference term

$$\tfrac{1}{2}(e^{-i(k_\parallel - q)D_\parallel} + e^{+i(k_\parallel - q)D_\parallel}) = \cos[(k_\parallel - q)D\cos\theta_D], \tag{19}$$

where we used $D_\parallel = D\cos\theta_D$ (cf figure 1). We average thus only this factor over the orientation of the molecular axis, assuming the other variations to be unimportant. This gives

$$\tfrac{1}{2} \int_0^\pi \cos[(q - k_\parallel)D\cos\theta_D] \sin\theta_D \, d\theta_D = \frac{\sin[(k_\parallel - q)D]}{(k_\parallel - q)D}. \tag{20}$$

Hence, the thus-approximated transition probability may be written in the form

$$w(b) = c(b) + g(b)\frac{\sin[(k_\parallel - q)D]}{(k_\parallel - q)D}, \tag{21}$$

and the corresponding cross section equation (18) is then

$$\frac{d\sigma}{d\Omega_k \, dk} = C + G\frac{\sin[(k_\parallel - q)D]}{(k_\parallel - q)D}. \tag{22}$$

This result can be contrasted to that obtained by Stolterfoht *et al* [6],

$$\frac{d\sigma}{d\Omega_k \, dk} = C + G\frac{\sin kD}{kD}. \tag{23}$$

Since generally (for moderate values of ejected electron velocities) $q \ll k$, our approximate formula can be further approximated by

$$\frac{d\sigma}{d\Omega_k \, dk} = C + G\frac{\sin k_\parallel D}{k_\parallel D}. \tag{24}$$

This qualitative formula applicable for less energetic electron ejection can be instructively compared with that of Stolterfoht *et al* (equation (23)). For the forward direction these two simple forms agree, while for electrons ejected at 90° the interference pattern disappears. The suppression of the interference pattern at 90° seems to be confirmed by preliminary experimental results [9]. The more precise approximate formula equation (22) still predicts some interference pattern at 90°, but with a very slow initial variation. Note that at angles close to the right angle the oscillations are governed only by the increase of the momentum transfer q and are not present in equation (24). The difference between the present theory and the model calculations of Stolterfoht *et al* [6] can be traced to the fact that their arguments are valid only for forward directions.

The ionization cross sections of H_2 by 60 MeV u^{-1} Kr^{34+} impact for electron ejection angles 30° and 150° calculated numerically from formulae (16)–(18) are plotted along with the experimental data [6] in figure 2. In the calculations the initial state of the hydrogen molecule has been described by a Heitler–London-type wavefunction [8] with the effective charge $\alpha = 1.165$ and the internuclear distance $D = 1.42$. Because of the simplified final-state described by equation (3) and the peaking approximation (8) valid only for high values of momentum k, for low electron energies these calculated cross sections are not expected
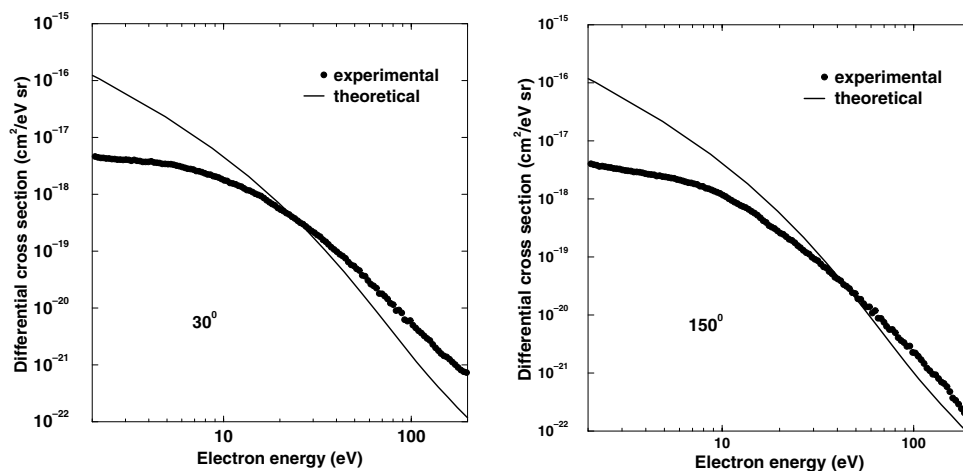
Figure 2. Double differential cross sections for the ionization of H_2 by 60 MeV u^{-1} Kr^{34+} ion impact for 30° (left) and 150° (right) ejection angles as a function of the electron energy. Present calculated values are compared with the experimental data of Stolterfoht *et al* [6].
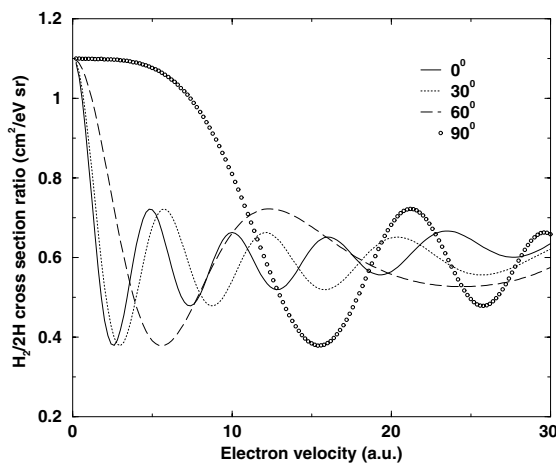

Figure 3. Calculated double differential cross section ratios for the ionization of H_2 and 2H by 60 MeV u^{-1} Kr^{34+} ion impact as a function of ejection angle and electron velocity for selected angles.

to agree with the experiment. Some disagreement is found even at high electron energies. However, the model is still useful for the model calculations allowing understanding of the interference patterns.

Following Stolterfoht *et al* [6], we have calculated the cross section ratios for the hydrogen molecule to two hydrogen atoms. The cross sections for the atomic targets are evaluated with the same ionization potential and effective charge as for molecular targets. In figure 3 we plot the differential cross section ratios for selected ejection angles up to large values of electron velocity. For these large velocities the ratio oscillates with a variable period, since the momentum transfer $q = (E_I + k^2/2)/v$ becomes comparable to k as the latter increases. It would be interesting to confirm these predictions for varying ejection angles experimentally. We can also note that below $k = 5$ au, i.e. in the region studied up to
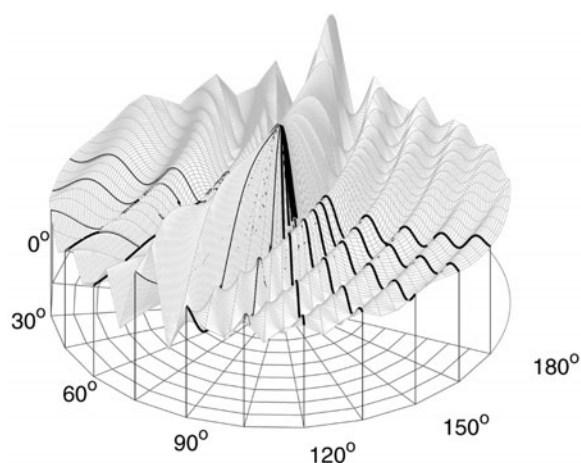
Figure 4. Calculated double differential cross section ratios for the ionization of H_2 and 2H by 60 MeV u^{-1} Kr^{34+} ion impact as a function of ejection angle and electron velocity, where the velocity is used as radial coordinate. The maximum value of the electron velocity is 20 au.
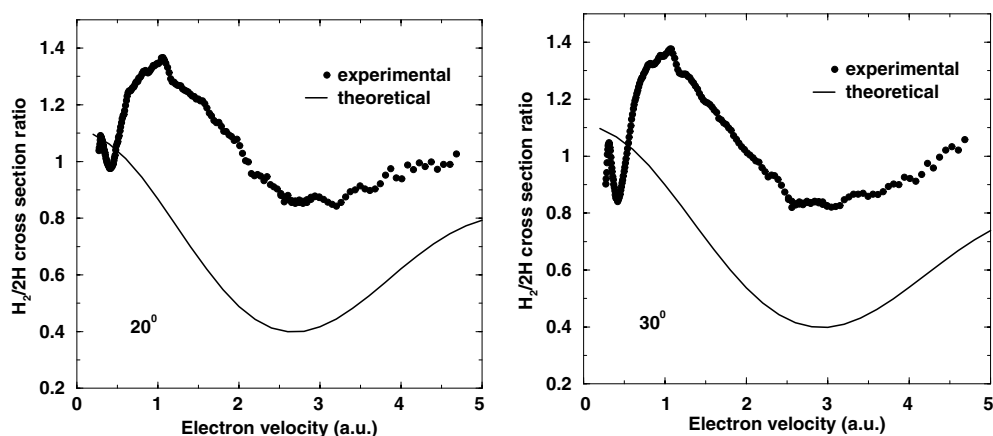

Figure 5. Double differential cross section ratios for 20° (left) and 30° (right) ejection angles compared with the normalized experimental to theoretical ratios of Stolterfoht *et al* [6].

now in the experiments, the differences between the curves for 0° and 30° are negligible. The differential cross section ratios as a function of ejection angle and momenta are plotted in figure 4. As the simplified formula (22) predicts, the oscillations for smaller values of k at 90° ejection angle are suppressed, and the period of oscillations varies with the angle. The 'waves' on this three-dimensional plot have an approximately constant 'wavelength' only in the z direction.

Figure 5 compares the cross section ratios obtained from the full integration of equations (16)–(18) with the ratios of experimental (H_2) to the theoretical (2H) cross sections reported by Stolterfoht *et al* [6] at 20° and 30°. The fact that the magnitudes of these ratios are not fitted perfectly is not surprising in view of the overall quality of the cross sections discussed above, and also because the experimental ratio was obtained through several steps of correcting the theoretical cross sections for atoms [6]. More important is that the oscillations

in cross section as a function of the ejected electron velocity have period and phase very close to the experimental values. This good agreement, as expected, is obtained for higher values of the ejected electron velocity ($v > 1.5$ au).

In conclusion, we have shown that the oscillatory pattern observed in the electron ejection cross section of the hydrogen molecule as a function of the electron velocity has a period changing with the electron ejection angle, which can be summarized by a very simple formula (24), as well as a more precise one (22). At 90° these oscillations are strongly suppressed, as preliminary experimental results [9] seem to confirm.

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