Non-perturbative investigation of the interference effects in the ionization of the H$_2$ by charged particle impact

S. Borbély, K. Póra and L. Nagy
Faculty of Physics, Babeş-Bolyai University, str. Kogálniceanu nr. 1, 400084 Cluj-Napoca, Romania
E-mail: lnagy@phys.ubbcluj.ro

Abstract. The interference effects in the double differential ionization cross section of H$_2$ are studied theoretically. Theoretical, non-perturbative models (Volkov model, numerical solution of the TDSE) developed for the ionization of atomic systems by intense ultrashort laser pulses are applied with success to study the ionization by fast charged particle impact. These results are compared with those obtained by a perturbative impact parameter method.

1. Introduction
Interference effects in the ionization of the hydrogen molecule due to the two-center character of the target have been observed experimentally and studied theoretically both for charged particle [1, 2, 3, 4] and photon [5, 6, 7, 8, 9] impact. Just like in the Young experiments in optics, the atoms in the molecule can be considered as separate coherent sources of electrons, leading to interference patterns in the ionization cross sections. The presence of such interference effects in the ionization spectra of diatomic molecules by highly charged particle impact was observed experimentally for the first time by Stolterfoht et al [1].

Experiments usually do not detect the orientation of the molecular axis. However, recently it has become possible to observe experimentally the alignment dependence of the ionization in the case of diatomic homonuclear molecules [10] and important experimental and theoretical work is done to achieve better alignments of the molecular targets [11]. These efforts suggest, that in the near future there will be available angular resolved ionization cross sections for given molecular alignments. Unlike in the case of photoionization, there are only a few theoretical studies [4, 12, 13] in the case of the ionization by charged particle impact or by ultrashort laser pulses, which produce reliable angular resolved theoretical data for fixed molecular orientation.

In the present paper we use a method developed for the ionization by short laser pulses [14] for the ionization by charged particle impact and compare with the previous results obtained with a perturbative impact parameter method [2].

2. Theory
The Coulomb interaction between the projectile and electron is expanded into multipole series. It is known that at high velocities of the projectile the ionization is dominated by distant collisions, and in the multipole series of the interaction the dipole term is the most important [15]. The
electric field is decomposed into a component parallel and a component perpendicular relative to the projectile trajectory. After basic calculations can be proved, that for high projectile velocities (more than 50 a.u. in the present calculations) the overall momentum transfer toward the electron is along the perpendicular direction, because the parallel components for the approaching and departing parts of the trajectory cancel out. The electron system does not change significantly in the short time interval of the interaction. In these conditions the electron transitions are mainly driven by the perpendicular component of the electric field, and it is safe to neglect the parallel component during our calculations. This approximation for high projectile velocities is valid also for atoms, and in case of molecules for any orientation of the molecular axis. The remaining perpendicular component can be approximated by a sine-square half-cycle electromagnetic pulse [16].

The time evolution of the active electron in the $H_2$ in the presence of an external electromagnetic field is described using the time dependent Schrödinger equation. The screening effect of the passive electron is included into the Coulomb potential $V(\vec{r})$, which acts between the active electron and the nuclei of the molecule. In the present approach the time dependent wave function is expressed as a superposition of Volkov wave functions [14]

$$\Psi(\vec{r},t) = \int d\vec{p} c(\vec{p},t)\Psi_V(\vec{p},t).$$

(1)

Using the time dependent Schrödinger equation for the expansion coefficients, the following integro-differential equation is obtained (for details see [14])

$$\frac{\partial}{\partial t} c(\vec{p},t) = -\frac{i}{(2\pi)^3} \int d\vec{s} c(\vec{s}+\vec{p},t)e^{-\frac{i}{2} \int_0^t dt' \vec{A}(t')} \int d\vec{r} V(\vec{r})e^{i\vec{s}\cdot\vec{r}},$$

(2)

where $\vec{A}(t) = -\int_0^t \vec{E}(t') dt'$ is the vector potential of the external electromagnetic field. The initial condition for the expansion coefficients is obtained from the initial state wave function $\psi_i(\vec{r})$. The molecular orbital describing the the initial state of the active electron is written as a linear combination of two atomic orbitals with effective charge $\alpha$. Denoting by $S$ the overlap integral between the two atomic orbitals, the initial condition is

$$c^0(\vec{p}) = \sqrt{\frac{\alpha^5}{2\pi^5(1+S)} \frac{2\cos(\bar{p}\bar{R}_0)}{(\alpha^2+p^2)^2}},$$

(3)

where $\bar{R}_0$ is the half of the nuclear axis. The simplest approximate solution of Eq. 2 is obtained by neglecting the Coulomb potential ($V(\vec{r}) = 0$). In this case for the expansion coefficient we have $c_V(\vec{p},t) = c^0(\vec{p})$. This approximation, also called Volkov model, provides good results only for high field intensities. In improving this approximation a possibility is the first order iterative solution of Eq. 2, which in the case of atomic systems was used successfully [14], however the most accurate results can be obtained by the full numerical solution. In the present work Eq. 2 is solved on a FEDVR (finite element discrete variable representation) numerical grid using fourth order Runge-Kutta method. The obtained numerical solution further on will be called TDSE. From the time dependent wave functions calculated in the framework of Volkov or TDSE model the transition probability amplitude to a plane wave final state can be calculated as $a_{i f} = (2\pi)^{3/2}c(\vec{k} - \vec{A}(\tau),\tau)$. We compare these results with those obtained by a perturbative impact parameter method, developed previously [2]. In this model we consider the interaction of the projectile with the active electron as a perturbation. In the calculation of the the transition amplitude the peaking approximation was applied. For the transition amplitude we get [2]

$$a(b,\bar{R}_0) = \frac{2\sqrt{3}Z_pN\alpha}{\sqrt{\pi}e^{\frac{1}{2}}(k^2\gamma^2)}\sqrt{\pi}e^{-i\bar{b}\cdot\bar{k}} \left[ b_+K_1(b_+\gamma)e^{-i(q-k)R_0} + b_-K_1(b_-\gamma)e^{i(q-k)R_0} \right],$$

(4)
Figure 1. Ionization probability densities in the collisional plane as a function of electron momentum ($k$) and electron ejection angle ($\theta_k$ is measured relative to the direction of the perpendicular momentum transfer) for different molecular axis orientations ($\theta_R_0$) represented on a logarithmic scale.

\[ \gamma = \sqrt{\alpha^2 + (q - k_\parallel)^2}, \]  

Here $b_+$ and $b_-$ are the impact parameters relative to the two nuclei, while $R_{0\parallel} = R_0 \cos \beta_{R_0}$, $\beta_{R_0}$ being the angle between the molecular axis and the projectile beam direction. The parallel component of the ejected electron momentum $k$ is $k_\parallel = k \cos \beta_k$, with $\beta_k$ the electron ejection angle. $K_1(x)$ denotes the modified Bessel function, $N$ a normalization factor. $q = \Delta E/v$ is the minimum value of the momentum transfer, where $\Delta E = E_i + k^2/2$ is the energy transfer to the electron.
3. Results and discussions

Calculations were performed for 68 MeV/u ($v = 52.18$ a.u.) K$^+{}^{33+}$ projectile at fixed impact parameter ($b = 4a_0$) value. The electric field corresponding to these parameters has a strength of $E_0 = -Zp/b^2 = 2.0625$ a.u. and duration of $\tau = 4b/v = 0.306$ a.u. The probability densities were obtained for different molecular axis orientations ($\theta_R$ is measured relative to the direction of the perpendicular momentum transfer) in the framework of the Volkov, TDSE and perturbative models. Their values in the collisional plane are presented on Fig. 1. A good agreement between Volkov and TDSE results is observed. For both models well defined deep minima in the probability densities may be observed, which appear due to the interference. The minimum is defined by the zero values of expression $\cos[(\mathbf{k} - \Delta\mathbf{p})\mathbf{R}_0]$, where $\Delta\mathbf{p} = \mathbf{A}(\tau)$ is the momentum transferred from the projectile to the electron. The same formula was also obtained empirically by Stolterfoht et. al. [1]. The good agreement between the TDSE and Volkov models is due to the short duration and high intensity of the electric pulse, i.e. the sudden approximation applied in the Volkov model is valid in accordance with the findings of Duchateau et al [17].

For the parallel orientation of the molecular axis relative to the projectile trajectory (i.e. $\theta_R = 90^\circ$) the same minima are obtained also with the perturbative model. For this orientation $b_+ = b_-$ and the amplitude given by Eq. 4 is proportional to $\cos[(k_j - q)\mathbf{R}_0]$, leading to a similar dependence as for the other two methods (3). For the other orientations of the molecular axis the impact parameters relative to the two nuclei ($b_+$ and $b_-$) are different, causing one term of the amplitude (4) being much larger than the other. Consequently, in this model, the interference is unobservable for molecular orientations $\theta_R = 0^\circ$ and $\theta_R = 45^\circ$.

Another difference between the models is, that in the case of the non-perturbative models the electrons are predominantly ejected in the direction of the momentum transfer, while in the perturbative model also the ejection in the opposite direction has the same probability, leading to a dipole character.

We assume, that in the calculated cross sections these important differences between the models will diminish, because of the integration over all possible impact parameters. The calculation of the differential cross section with the Volkov and TDSE models will be the topic of our future work.

Acknowledgments

This work was supported by the Romanian Academy of Sciences (grant No. 31/2008) and by the Romanian National Plan for Research (PNII) under contract No. ID 539.

References

[1] Stolterfoht N et al 2001 Phys. Rev. Lett. 87 023201.
[2] Sarkadi L 2003 J. Phys. B 36 2153.
[3] Laurent G et al 2002 J. Phys. B: At. Mol. Opt. Phys. 35 L453.
[4] Cohen H D and Fano U 1966 Phys. Rev. 150 30.
[5] Nagy L, Kocbach L, Póra K and Hansen J P 2002 J. Phys. B: At. Mol. Opt. Phys. 35 L495.
[6] Borbely S and Nagy L 2007 Rad. Phys. Chem. 76 516.
[7] Baier S, Becker A and Plaja L 2008 Phys. Rev. A 78 013409.
[8] Nicolas Sisourat, Jérémie Caillat, Alain Dubois and Pablo D. Fainstein 2007 Phys. Rev. A 76 012718.
[9] Póra K and Nagy L 2008 Nucl. Instr. Meth. B, doi:10.1016/j.nimb.2008.10.059, in press
[10] Borbely S, Tókési K and Nagy L 2008 Phys. Rev. A 77 033412.
[11] Sakai H, Minemoto S, Nanjo H, Tanji H and Suzuji T 2003 Phys. Rev. Lett. 85 083001.
[12] Nicolas Sisourat, Jérémie Caillat, Alain Dubois and Pablo D. Fainstein 2007 Phys. Rev. A 76 012718.
[13] Póra K and Nagy L 2008 Nucl. Instr. Meth. B, doi:10.1016/j.nimb.2008.10.059, in press
[14] Nicolas Sisourat, Jérémie Caillat, Alain Dubois and Pablo D. Fainstein 2007 Phys. Rev. A 76 012718.
[15] Borbely S and Nagy L 2007 Rad. Phys. Chem. 76 516.
[16] Nicolas Sisourat, Jérémie Caillat, Alain Dubois and Pablo D. Fainstein 2007 Phys. Rev. A 76 012718.
[17] Duchateau G, Illescas C, Pons Z, Cormier E and Gayet R 2000, J. Phys. B: At. Mol. Opt. Phys. 33 L571.