Coherence and partial localization in electron emission from asymmetric diatomic molecules

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Abstract. The presence of interference patterns in the spectra of emitted electrons from \(\text{HeH}^+\) molecular ions due to the impact of bare ions is analysed. It is shown that these oscillations, which are explained in terms of the coherent emission of electrons from the vicinities of the target nuclei, are preserved even after averaging over all molecular orientations. The influence of partial localization of electrons around the alpha particle center is studied. The relationship between electron ionization by ion impact and photoionization is investigated.

1. Introduction

Interference effects due to coherent emission of electrons from the neighbourhood of the nuclei of diatomic targets have been studied during the last 50 years. The first theoretical predictions concerning this phenomenon were made by Cohen and Fano in the decade of 1960 for photoionization of \(\text{H}_2\) molecular ions [1]. After this pioneering work, many theoretical studies were carried out to investigate the presence of interference in ionization of single diatomic molecules by impact of photons and electrons [2–5]. Nevertheless, the existence of this effect was confirmed forty years later, when cross sections for single electron ionization from \(\text{H}_2\) targets by \(60\ \text{MeV}/\text{u} \ \text{Kr}^{34+}\) ion impact were measured [6]. Since then, numerous theoretical [7–20] and experimental [21–35] investigations were carried out for impact of photons, electrons and ions on diatomic molecules. These investigations were mainly oriented to the study of coherent electron emission from homonuclear targets such as \(\text{H}_2\), \(\text{N}_2\) and \(\text{O}_2\), and to a lesser extent, for heteronuclear molecules.

The aim of the present work is to present the results obtained for single ionization of the heteronuclear molecular ion \(\text{HeH}^+\) by impact of bare ions. It has been shown that, in spite of the highly asymmetry of the electron distribution of this target, fingerprints of coherent emission of electrons from the vicinities of the molecular centres were clearly seen in the angular distributions [36] and that interferences are preserved even after averaging the corresponding cross sections over all possible molecular orientations [37]. Details concerning the theoretical model have already been given previously; thus, only a general description of the method will be presented in the following sections. The straight line version of the impact parameter method will be employed.

Atomic units are used throughout unless otherwise stated.
2. Theory

Single ionization of HeH\(^+\) molecular ions by impact of bare ions at intermediate and high collision energies is investigated. The incident particle moves with a velocity \(v\) with respect to a reference frame whose origin is fixed at the centre of mass of the target. At the high impact energies here considered, the vibrational and rotational times of the molecule are much larger than the collision one, so it can be assumed that the nuclei of the target remain frozen in their initial positions during the reaction. Also, we consider that the residual electron remains in its initial state, in such a form that the two-electron problem can be reduced to the analysis of a one-electron system. It is possible to show that this hypothesis is equivalent to the one of an independent electron model where the passive electron may occupy any state of HeH\(^2+\).

With these assumptions in mind, the five-fold differential cross section (FDCS) as a function of the electron energy \(\varepsilon_k\) and the orientation of the ionized electron \(\Omega_k\) and of the molecule \(\Omega_b\) can be obtained as:

\[
\sigma^{(5)}(\varepsilon_k, \Omega_k, \Omega_b) = 2k \int d\tilde{\rho}|a_{i,f}^+(\tilde{\rho})|^2
\]

with \(a_{i,f}^+\) the post-form of the transition amplitude for the active electron. In equation (1) \(k = \sqrt{2\varepsilon_k}\) is the modulus of the electron momentum \(\tilde{k}\), with \(\varepsilon_k\) the electron emission energy, and \(\tilde{\rho}\) is the impact parameter vector. The double differential cross sections (DDCS) as a function of the energy and solid angle of the emitted electron can be then obtained by averaging equation (1) over all possible molecular orientations:

\[
\sigma^{(2)}(\varepsilon_k, \Omega_k) = \frac{2k}{4\pi} \int d\tilde{\rho}\Omega_b |a_{i,f}^+(\tilde{\rho})|^2
\]

Into the straight line version of the impact parameter approximation, the expression for \(a_{i,f}^+\) is:

\[
a_{i,f}^+ (\tilde{\rho}) = -i \int_{-\infty}^{+\infty} dt \left\langle \chi_f^+ | W_f^+ \chi_i^+ \right\rangle
\]

where \(W_f^+\) is the perturbation operator corresponding to the final channel, and \(\chi_i^+ (\chi_f^-)\) are the initial (final) wavefunction with correct asymptotic outgoing (incoming) boundary conditions. For the continuum distorted wave-eikonal initial state (CDW-EIS) approximation, these wavefunctions are chosen as:

\[
\begin{align*}
\chi_i^{+,EIS} (\tilde{x}, t) &= \varphi_i(\tilde{x}) \exp(-i \varepsilon_i t) L_i^{+,EIS} (\tilde{s}) \\
\chi_f^{-,CDW} (\tilde{x}, t) &= \varphi_f(\tilde{x}) \exp(-i \varepsilon_k t) L_f^{-,CDW} (\tilde{s})
\end{align*}
\]

In the preceding expressions, \(\varphi_i\) and \(\varphi_f\) are bound and continuum states of the active electron in the entrance and exit channels, respectively, \(\varepsilon_i\) is the initial orbital energy, and \(\tilde{x}\) \((\tilde{s})\) is the electron coordinate with respect to the centre of mass of the target (projectile nucleus). Also in equations (4) and (5), \(L_i^{+,EIS}\) and \(L_f^{-,CDW}\) represents projectile eikonal and continuum distortion functions, respectively.

In order to describe the initial molecular orbital of the HeH\(^+\) ion, a basis set of Slater-type orbitals (STO) \(\phi_h^{STO}\) centred on each molecular nuclei is employed:

\[
\varphi_i(\tilde{x}) = \omega_1 \phi_1^{STO} (\tilde{x}_1) + \omega_2 \phi_2^{STO} (\tilde{x}_2)
\]

where the subscripts 1 and 2 denote the He\(^{2+}\) and H\(^+\) nuclei, respectively, and \(\tilde{x}_h\) represents the electron coordinate with respect to the nucleus \(h\) \((h = 1, 2)\). The values of both the STO
exponents and the coefficients $\omega_h$ of equation (6) were optimized by employing the quantum chemistry program Gaussian 98 [38]. These quantities were calculated by using a STO-6G basis set within the Hartree-Fock (HF) approximation, where each Slater-type orbital is described as a linear combination of 6 Gaussian-type orbitals (GTO) (see references [39,40] for more details). The equilibrium internuclear distance $b$ and the orbital energy $\varepsilon_i$ were calculated by using a $6 - 311G^*$ basis set, also within the HF formalism.

The final continuum wavefunction in the exit channel is described by employing the CDW-EIS model and the two-effective centre (TEC) approximation [7, 10], so that the expression for

\begin{equation}
\varphi_f(x) = (2\pi)^{-3/2} \exp[iK \cdot (\vec{x} + \vec{b}_h)] N^*(\xi) F_1(-i\xi; 1; -ikx_h - ik\cdot\vec{x}_h)
\end{equation}

when the component $\phi^{STO}_h$ of the initial orbital wavefunction (see equation (6)) is considered. In equation (7), $\vec{b}_h$ represents the position of the $h$-nucleus with respect to the centre of mass of the molecular nuclei, and $\xi = k^{-1}\sqrt{2\varepsilon_i}$.

Considering the scattering matrix element $\mathcal{R}^+_i(\vec{\eta})$, which is the Fourier transform of the transition amplitude $a^+_i(\vec{\rho})$, and the well-known Parseval-Plancherel identity, the expressions for the differential cross sections of equations (1) and (2) can be rewritten as it follows:

\begin{equation}
\sigma^{(5)}(\varepsilon_k, \Omega_k, \Omega_b) = \frac{2k}{4\pi} \int d\vec{\eta} d\Omega_b |\mathcal{R}^+_i(\vec{\eta})|^2
\end{equation}

\begin{equation}
\sigma^{(2)}(\varepsilon_k, \Omega_k) = \frac{2k}{4\pi} \int d\vec{\eta} d\Omega_b |\mathcal{R}^+_i(\vec{\eta})|^2
\end{equation}

respectively, where $\vec{\eta}$ is the component of the total momentum transfer $\vec{K}$ perpendicular to the direction of the incident particle.

In a previous work [36], it was shown that, in the TEC approximation, the scattering matrix element $\mathcal{R}^+_i(\vec{\eta})$ can be expressed as a linear combination of effective one-centre scattering matrix elements $\mathcal{R}^{+\text{eff}}_k$ associated with the complete basis of STO centred on each molecular nuclei. According to that, and within the CDW-EIS model, the following expression for the square modulus of $\mathcal{R}^+_i(\vec{\eta})$ is obtained:

\begin{equation}
|\mathcal{R}^{+\text{CDW-EIS}}_{i,f}(\vec{\eta})|^2 = \omega_1^2 |\mathcal{R}^{+\text{eff}}_{1}(\vec{\eta})|^2 + \omega_2^2 |\mathcal{R}^{+\text{eff}}_{2}(\vec{\eta})|^2 + 2\omega_1\omega_2 \cos[(k + \vec{K}) \cdot \vec{b}] \left[ \text{Re} \mathcal{R}^{+\text{eff}}_{1}(\vec{\eta}) \text{Re} \mathcal{R}^{+\text{eff}}_{2}(\vec{\eta}) + \text{Im} \mathcal{R}^{+\text{eff}}_{1}(\vec{\eta}) \text{Im} \mathcal{R}^{+\text{eff}}_{2}(\vec{\eta}) \right] \\
+ 2\omega_1\omega_2 \sin[(k + \vec{K}) \cdot \vec{b}] \left[ \text{Re} \mathcal{R}^{+\text{eff}}_{1}(\vec{\eta}) \text{Im} \mathcal{R}^{+\text{eff}}_{2}(\vec{\eta}) - \text{Im} \mathcal{R}^{+\text{eff}}_{1}(\vec{\eta}) \text{Re} \mathcal{R}^{+\text{eff}}_{2}(\vec{\eta}) \right]
\end{equation}

It can be seen that if in equation (10) we set $\omega_1 = \omega_2$ and $\mathcal{R}^{+\text{eff}}_{1}(\vec{\eta}) = \mathcal{R}^{+\text{eff}}_{2}(\vec{\eta})$, the expression corresponding to the homonuclear case is recovered [7].

3. Results and discussions

Our main interest is focused on interference effects in the single ionization reaction of the HeH$^+$ molecular target by impact of protons and bare carbon ions. We will analyse firstly the angular distribution of target electrons, considering the case of a coplanar geometry in which the molecule, the emitted electron and the projectile are all in the same plane. In order to reveal the oscillatory structures associated with the coherent emission of electrons from both molecular centres, we calculate the ratios:
\[ R_{HeH^+}(\varepsilon_k, \Omega_k) = \frac{\sigma^{(5)}}{2k\omega_1^2 \int d\Omega R^{-s}_{1,1}(\vec{\Omega})^2} \] (11)

In the preceding expression, the five-fold differential cross sections \( \sigma^{(5)} \) of equation (8) is divided by a FDCS obtained by integrating the first addend of equation (10). This choice is supported by the fact that the electron density around the helium nucleus largely dominates the molecular electron density and it is not very different from that of the atomic helium orbital [36].

In figures 1 and 2 we present the ratios obtained for impact of C\(^6^+\) at collision energies of 1, 13.7 and 100 MeV/u, when the molecule is aligned parallel and perpendicular to the direction of the projectile, respectively. These results are compared with the angular distributions produced by two emitters of spherical waves of amplitudes \( \omega_1 \) and \( \omega_2 \) separated a distance \( b \):

\[ I(\theta) \sim \omega_1^2 + \omega_2^2 + 2\omega_1\omega_2 \cos(\vec{k} \cdot \vec{b}) \] (12)

where the coefficients \( \omega_h \) (\( h = 1, 2 \)) are defined in equation (6). This last formula describes correctly the ionization of HeH\(^+\) molecular ions by circularly polarized light at high photon energies, with the incidence direction of the light perpendicular to the plane formed by the molecular axis and the electron ejection direction. A similar expression was employed by Akoury et al. [34] to describe the photoionization of H\(_2\) molecules.

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**Figure 1.** Five-fold differential cross section ratios and photoionization model calculations (equation (12)) for the HeH\(^+\) aligned parallel to the incident beam, for different values of the electron energy: (a) 100 eV, (b) 400 eV, (c) 700 eV and (d) 1000 eV. The alpha particle is considered to be placed on the semi-axis coinciding with the emission angle \( \theta = 0^\circ \).
Figure 1 shows clearly the presence of interference patterns. For each impact velocity the effect is more noticeable as the energy of the ionized electron increases. Moreover, for parallel orientation, the differential cross section ratios tend to the angular distributions predicted by the photoionization model as the impact energy increases. This behaviour can be easily explained from equation (10) considering that the component of the momentum transfer \( \vec{K} \) perpendicular to the impact velocity does not contribute to the oscillatory factors and the parallel one depends as \( v^{-1} \). In consequence, for high energy impact, the arguments of the trigonometrical functions presents in equation (10) can be reduced to \( k \cdot b \), and the resulting expression for \( |R_{i,f}^{CDW-\psi_{IS}}(\vec{n})|^2 \) is analogue to the one corresponding to the photoionization model of equation (12).

On the other hand, when the molecular ion is perpendicular to the direction of incidence, interference patterns are observed in the FDCS ratios. However, they do not show a clear tendency to the photoionization model results as the impact energies increases, as for parallel orientation. This behaviour can be explained again considering that for this particular molecular orientation, the transverse momentum transfer does not become negligible with respect to the electron momentum \( k \) at high energy impact.

![Five-fold differential cross section ratios and photoionization model calculations](image)

**Figure 2.** Five-fold differential cross section ratios and photoionization model calculations (equation (12)) for the HeH+ aligned perpendicular to the incident beam, for different values of the electron energy: (a) 100 eV, (b) 400 eV, (c) 700 eV and (d) 1000 eV. The alpha particle is considered to be placed on the semi-axis coinciding with the emission angle \( \theta = 90^\circ \).

In order to test the validity of the explanation given in the preceding paragraph, we have plot in figure 3 the five-fold differential cross section ratios for perpendicular orientation but taking the total momentum trasfer \( K = 0 \) in the trigonometric functions of the third and fourth addends of equation (10). As it can be seen, there is a very good qualitative agreement between
the new angular spectra and the photoionization results. The number of lobes is the same, and their positions are almost coincident. The small differences between ion and photon impact can mainly be attributed to the different mechanisms leading to electron ionization of the target, being for ion impact the momentum transfer from the projectile the most important one.

|                  | \(v\) |
|------------------|--------|
|                  | (a)    | (b)    | (c)    | (d)    |
| \(C^+(1\text{ MeV/u})\) | ![Image](image1.png) | ![Image](image2.png) | ![Image](image3.png) | ![Image](image4.png) |
| \(C^+(13.7\text{ MeV/u})\) | ![Image](image5.png) | ![Image](image6.png) | ![Image](image7.png) | ![Image](image8.png) |
| \(C^+(100\text{ MeV/u})\) | ![Image](image9.png) | ![Image](image10.png) | ![Image](image11.png) | ![Image](image12.png) |
| Photoionization Model | ![Image](image13.png) | ![Image](image14.png) | ![Image](image15.png) | ![Image](image16.png) |

**Figure 3.** Same as figure 2, but considering \(K = 0\) in the calculation of the trigonometric factors of equation (10).

In order to determine if interference patterns are preserved after averaging on all possible molecular orientations, we study the behaviour of double differential cross sections \(\sigma^{(2)}(\varepsilon_k, \Omega_k)\). Employing equations (5) and (10), they can be expressed as:

\[
\sigma^{(2)}_{HeH^+}(\varepsilon_k, \Omega_k) = 2k \omega_1^2 \int d\tilde{\eta} \left| R^{+,eff}_1(\tilde{\eta}) \right|^2 + 2k \omega_2^2 \int d\tilde{\eta} \left| R^{+,eff}_2(\tilde{\eta}) \right|^2 + 2k \omega_1 \omega_2 \int d\tilde{\eta} \left[ \text{Re} R^{+,eff}_1(\tilde{\eta}) \text{Re} R^{+,eff}_2(\tilde{\eta}) \right] \sin \left( \frac{[k + \hat{K}] b}{|k + \hat{K}|} \right)
\]

\[
= S_{d_{HeH^+}}(\varepsilon_k, \Omega_k) + S_{d_{H^+}}(\varepsilon_k, \Omega_k) + S_{i_{HeH^+}}(\varepsilon_k, \Omega_k)
\]

where \(S_{d_{HeH^+}}(\varepsilon_k, \Omega_k)\) and \(S_{d_{H^+}}(\varepsilon_k, \Omega_k)\) represents the direct terms related to the ionization process of each one of the molecular nuclei, whereas \(S_{i_{HeH^+}}(\varepsilon_k, \Omega_k)\) represents the interference term and contains the information arising from coherent electron emission [37]. For the particular case of the homonuclear H\(_2\) molecule, the DDCS results in:
\[
\sigma^{(2)}_{\text{H}_2}(\varepsilon_k, \Omega_k) = 4k \omega^2 \int d\eta |R^{+,\alpha\beta}(\eta)|^2 + 4k \omega^2 \int d\eta |R^{+,\alpha\beta}(\eta)|^2 \frac{2\sin|\vec{k} + \vec{K}|b}{|\vec{k} + \vec{K}|b} 
\]
(14)

where, analogously to the HeH\(^+\) case, \(S_{dH^+}\) represents the direct term corresponding to the electron emission from the proximities of both \(H^+\) nuclei and \(S_{iH^2}\) in the interference term.

To put in evidence the characteristic undulations associated to interference effects, we calculate the ratios:

\[
\tilde{R}_{\text{H}_2}(\varepsilon_k, \Omega_k) = \frac{\sigma^{(2)}_{\text{H}_2}}{2S_{dH^+}} = 1 + \frac{S_{dH^+}}{S_{iH^2}} + \frac{S_{iH^+}}{S_{iH^2}} 
\]
(15)

for HeH\(^+\) molecular ions, and:

\[
\tilde{R}_{\text{H}_2}(\varepsilon_k, \Omega_k) = \frac{\sigma^{(2)}_{\text{H}_2}}{2S_{dH^+}} = 1 + \frac{S_{iH^+}}{S_{dH^+}} 
\]
(16)

for hydrogen molecules.

It is evident from equation (15) that only the term \(S_{iH^+}/S_{dH^+}\), which will be henceforth denoted as \(R_{iH^+}\), can give contributions to coherent emission oscillations in the ratios \(\tilde{R}_{\text{H}_2}\). Analogously, in equation (16), the important term is the quotient \(S_{iH^+}/2S_{dH^+}\) which will be henceforth denoted as \(R_{iH^2}\). In figure 4 we plot the ratios \(R_{iH^+}\) and \(R_{iH^2}\) for proton impact at 13.7 and 100 MeV/u, at electron emission angles \(\theta_k = 30^\circ\) and \(\theta_k = 150^\circ\). It can be seen that smooth undulations appear in the \(R_{iH^+}\) spectra. If we consider a certain value of the electron emission angle \(\theta_k\) and a fixed projectile energy, we can observe that the frequency of the oscillations increases as \(\varepsilon_k\) increases. Moreover, for each impact velocity, the ratio frequency is larger for the backward emission angles than for the forward ones as it has previously been observed for \(H_2\) targets.

The corresponding ratios for the \(H_2\) target have an oscillatory behaviour analogous to that of the HeH\(^+\) case. It can be observed in figure 4 that the amplitudes of \(R_{iH^+}\) are smaller than those of \(R_{iH^2}\), especially at low electron emission energies. This difference can be attributed to the partial localization of the emitted electron around the alpha particle. The frequency of the oscillations in these ratios are similar for the homonuclear and for the heteronuclear molecules; in particular, the positions of the respective maxima and minima are almost coincident.

Thus, in spite of the highly asymmetry of the electronic distribution of the HeH\(^+\) molecular ion, interference patterns due to coherent emission of electrons from the proximities of the target nuclei can be observed in the doubly differential cross sections, even after averaging over all molecular orientations. These oscillations are less pronounced than the ones corresponding to the homonuclear case.

Analogously to the FDCS case, we will compare the DDCS ratios calculated for the HeH\(^+\) ion with photoionization results obtained following the work of Walter and Briggs [3]. In this model for linearly polarized photons, whose details have been already given in [37], the expression for the doubly differential cross sections is:

\[
\sigma^{(2)}_{\nu\nu} = \frac{4\pi^2 \alpha}{\omega_{\nu\nu}} |\mathcal{T}_{\nu\nu}|^2 
\]
(17)

where \(\alpha\) is the fine-structure constant, \(\omega_{\nu\nu}\) is the angular frequency of the incident photon and \(\mathcal{T}_{\nu\nu}\) is the transition matrix element given by:
\[ T_{lf} = \langle \varphi_f | \hat{\varepsilon} \cdot \nabla | \varphi_i \rangle \]  

with \( \hat{\varepsilon} \) the polarization vector perpendicular to the \( z \) axis, \( \varphi_i \) the initial bound state (equation (6)), and \( \varphi_f \) the final continuum wavefunction (equation (7)) described within the TEC approximation and the CDW-EIS model. The ratios obtained for this photoionization model are proportional to the factor \( \sin(\sqrt{\mathbf{K}^2 + \mathbf{k}^2}b)/(|\mathbf{K} + \mathbf{k}|b) \), which is also present in the integrand of the expression to calculate \( a^{(2)} \) and thus influencing the cross section ratios of equation (15) for the ion impact case.

A comparison of the ratios obtained for 100 MeV/u energy protons impacting on HeH\(^+\) with those calculated with the photoionization model is given in figure 5. It can be seen that there is a qualitative agreement between both set of data. Still some differences are visible, in particular a shift in the oscillations with electron energy. In order to understand the origin of this discrepancy, we plot in figure 6 the cross section ratios for proton impact under the same conditions considered in figure 5, but taking the total momentum transfer \( \mathbf{K} = 0 \). The agreement with photoionization results is now very good, being the frequency of the oscillations and the positions of the maxima and minima almost the same. Thus, the differences between ion and photon impact can be attributed to the nature of the ionizing mechanism. Again, it can be concluded that the influence of the momentum transferred by the projectile to the target is very important in the ionization reaction by ion impact.

**Figure 4.** Comparison between the cross section ratios \( R_{i_{\text{HeH}^+}} \) (solid line) and \( R_{i_{\text{H}^+}} \) (dash dot dot line) for different impact energies and different values of \( \theta_k \). ECC and BE denote the electron capture to the continuum and binary encounter regions, respectively.
4. Conclusions

Ionization of the heteronuclear HeH$^+$ molecular ion by bare ion impact was theoretically analysed. Five-fold differential cross sections were calculated for the target oriented parallel and perpendicular to the incident beam of particles in a complete coplanar geometry. We have shown that, in spite of the highly asymmetry of the target, there are fingerprints of coherent electron emission in the angular spectra of emitted electrons. Also, a comparison with photoionization with circularly polarized light was made, showing that the results corresponding to ion impact tend to the photoionization ones at high energy impact, when the molecule is aligned parallel to the incident beam. Nevertheless, this tendency is not observed in the angular spectra obtained for perpendicular orientation. This effect can be attributed to the momentum transferred by the ion beam to the target.

Also, doubly differential cross sections for impact of protons on HeH$^+$ were obtained. We shown that interference patterns are preserved even after averaging the angular spectra over all possible molecular orientations. The observed interference patterns for this heteronuclear target are less pronounced than for the case of the homonuclear H$_2$ target. This effect can be attributed to the partial localization of electrons in the initial state around the alpha particle of the HeH$^+$ molecular ion and can be associated with the Young interference experiment with circular holes of different diameters. We should remark that the final state of the residual target is not discriminated in our model. Comparison of these results with those corresponding to photoionization with linearly polarized light shows a qualitative agreement at very high proton energy. The small differences in DDCSs for the proton and photon cases have their origin in the transfer of momentum induced by the charged projectile, which is absent in the photoionization case.

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