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# Distorted wave calculations for electron loss process induced by bare ion impact on biological targets

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## HIGHLIGHTS

- Distorted wave models are used to investigate ion-molecule collisions.
- Differential and total cross-sections for capture and ionization are evaluated.
- The influence of dynamic screening is determined.
- Capture reaction dominates the mean energy deposited by the projectile on the target.

## ARTICLE INFO

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## ABSTRACT

Distorted wave models are employed to investigate the electron loss process induced by bare ions on biological targets. The two main reactions which contribute to this process, namely, the single electron ionization as well as the single electron capture are here studied. In order to further assess the validity of the theoretical descriptions used, the influence of particular mechanisms are studied, like dynamic screening for the case of electron ionization and energy deposition on the target by the impacting projectile for the electron capture one. Results are compared with existing experimental data.

## 1. Introduction

Research on electronic reactions involved in collisions between ions and molecules are of prime interest in different areas, like radiobiology and medical physics for possible applications in radiotherapy with protons (protontherapy) and heavy ions (hadrontherapy).

Two main processes contribute to the single electron loss from atomic/molecular targets, namely, the electron capture and the electron ionization which dominates at low and high impact velocities, respectively. In recent works, both reactions were theoretically studied for biological targets by using different perturbative approximations (Champion et al., 2010, 2012; Galassi et al., 2012). In particular, water molecules as well as adenine, cytosine, guanine, thymine, uracil and sugar–phosphate backbone molecules were considered. The continuum distorted wave–eikonal initial state (CDW-EIS) approach (Crothers and McCann, 1983; Fainstein et al., 1988) was used for describing the single electron ionization at intermediate and high impact energies. Previous calculations using this model were employed with success to characterize this reaction for collisions involving numerous atoms and molecules as collision aggregates (Fainstein et al., 1991; Stolterfoht et al., 1997; Galassi et al., 2000). More recently, electron loss from molecular targets due to electron capture by the projectile was also investigated using the CDW-EIS and the continuum distorted wave (CDW) approximations (Galassi et al., 2010; Champion et al., 2012 and references therein). In general, an adequate description of the existing experimental data was obtained, even considering that they are very scarce for biological targets, in particular for DNA nucleobases and uracil. In this context, new theoretical predictions appear of prime importance in many fields like (micro)dosimetry.

In order to further investigate the adequacy of the mentioned distorted wave models, the present work is focused on the analysis of different physical effects which will help at the same time for a better understanding of the reactions analyzed. In particular, isolated water and adenine molecules are here considered, the last one being seen as a test case to predict some general behaviors common to all DNA molecular compounds.

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Atomic units will be used in the following except where otherwise stated.

## 2. Theory

Let us consider the single electron capture and the single electron ionization processes induced by bare ion impact on an atomic or molecular target. For these processes, it has been shown that multi-electron collision systems may be reduced to the study of a three body reaction composed by the projectile, the active electron and the residual target (Rivarola and Salin, 1984; Fainstein et al., 1988; Corchs et al., 1993; Galassi et al., 2004). In such a representation it is assumed that the passive electrons (the not promoted ones) remain in their orbitals during the collision. This is valid at high enough impact velocities for which the collision time is smaller than the one corresponding to the relaxation of the passive electrons. For molecular targets it will correspond additionally to collision times smaller than the vibrational and rotational ones.

The reaction is described from a reference frame fixed on the target nucleus and the straight line version of the impact parameter approximation is employed. We focus here our analysis on the use of two distorted wave models, the continuum distorted wave (CDW) and the continuum distorted wave-eikonal initial state (CDW-EIS). The internuclear interaction is not included in the following description because integration over all projectile angular distributions is considered.

In the $\alpha$-entry channel, the initial one-active electron distorted wavefunction is chosen as

$$\chi_\alpha^+ = \varphi_\alpha(\vec{x})\exp(-i\varepsilon_\alpha t)\mathcal{L}_\alpha(\vec{s}) \tag{1}$$

where the super-index $(+)$ indicates that it preserves correct outgoing boundary conditions, being the distortion factor

$$\mathcal{L}_\alpha(\vec{s}) = \exp[-i\nu\ln(vs+\vec{v}\cdot\vec{s})] \tag{2}$$

in the CDW-EIS model, whereas

$$\mathcal{L}_\alpha(\vec{s}) = N(\nu)\,{}_1F_1(i\nu;1;ivs+i\vec{v}\cdot\vec{s}) \tag{3}$$

in the CDW one. In Eq. (1), $\varphi_\alpha(\vec{x})$ represents the non-perturbed initial electron bound orbital and $\varepsilon_\alpha$ the corresponding orbital energy, $\vec{x}$ and $\vec{s}$ are the electron position vectors as measured from the target and projectile nuclei, respectively, $Z_P$ is the projectile nuclear charge, $\vec{v}$ is the impact velocity, and $\nu = Z_P/v$. The distorted functions included in the entrance channel depending of the $\vec{s}$ coordinate, take into account the fact that the electron bound to the target is simultaneously travelling in a continuum state of the projectile field, being this one considered in an eikonal approximation in CDW-EIS. This approximation in CDW-EIS is proposed in order to avoid the non-normalization of the initial distorted wavefunction, which provokes in the CDW model the overestimation of the total cross-sections at intermediate collision velocities. The function $N(a) = \exp(\pi a/2)\,\Gamma(1-i\,a)$ gives the normalization of the corresponding continuum factor.

For both CDW-EIS and CDW models, the same final distorted wavefunctions are chosen for electron capture and electron ionization in the $\beta$-exit channel. Thus, for electron capture this wavefunction is chosen as

$$\begin{aligned}\chi_\beta^- = {} & \varphi_\beta(\vec{s})\exp(-i\varepsilon_\beta t+i\vec{v}\cdot\vec{x}-iv^2t/2)\\ & \times N^*(\varsigma)\,{}_1F_1(-i\varsigma;1;-ivx-i\vec{v}\cdot\vec{x})\end{aligned} \tag{4}$$

where $\varphi_\beta(\vec{s})$ represents the final non-perturbed bound projectile state and $\varepsilon_\beta$ its corresponding orbital energy. Also, in Eq. (4), ${}_1F_1(-i\varsigma;1;-ivx-i\vec{v}\cdot\vec{x})$ is a Coulomb continuum factor associated with the electron–residual target interaction where $\varsigma = Z_T^*/v$, with $Z_T^*$ an effective target nuclear charge defined by $Z_T^* = (-2\,n^2\,\varepsilon_\alpha)^{1/2}$ (Belkić, 1978), where the value of the principal quantum number $n$ is set to be equal to the principal quantum number of each atomic orbital used to describe the molecular orbitals (see hereafter). In such a way the interaction between the active electron and the residual target is described by an effective Coulomb potential $V_T \cong -Z_T^*/x$. The final distorted wavefunction presents also a two-center character associated with the fact that the electron evolves in the combined field of the projectile and residual target. Now, the super-index $(-)$ indicates that correct ingoing conditions are satisfied.

To describe the electron ionization process, the final distorted wavefunction is taken as

$$\begin{aligned}\chi_\beta^- = {} & (2\pi)^{-3/2}\exp(-i\varepsilon_\beta t+i\vec{k}\cdot\vec{x}-ik^2t/2)\\ & \times N^*(\xi)\,{}_1F_1(-i\xi;1;-ikx-i\vec{k}\cdot\vec{x})\\ & \times N^*(\zeta)\,{}_1F_1(-i\zeta;1;-ips-i\vec{p}\cdot\vec{s})\end{aligned} \tag{5}$$

where $\vec{k}$ and $\vec{p} = \vec{k}-\vec{v}$ are the linear momenta of the electron with respect to the target and projectile nucleus respectively, $\xi = Z_T^*/k$ and $\zeta = Z_P/p$. The first part of Eq. (5), depending on the coordinate $\vec{x}$, is a wavefunction describing the electron in a continuum state of the residual target whereas the continuum factor depending on the coordinate $\vec{s}$ corresponds to the electron–projectile interaction. Thus, the final distorted wavefunction describes the electron travelling in a continuum state of both the projectile and residual target fields, and their actions on the emitted electron are considered on equal footing. Once more, but now for ionization, the super-index $(-)$ indicates that correct ingoing conditions are satisfied.

The post- and prior-versions of the transition amplitude for both reactions above mentioned can be written as

$$A_{\alpha,\beta}^+ = -i\int_{-\infty}^{+\infty} dt\left\langle \chi_\beta^- \middle| \left(H_{el}-i\frac{\partial}{\partial t}\right)^\dagger \middle| \chi_\alpha^+ \right\rangle \tag{6}$$

and

$$A_{\alpha,\beta}^- = -i\int_{-\infty}^{+\infty} dt\left\langle \chi_\beta^- \middle| \left(H_{el}-i\frac{\partial}{\partial t}\right) \middle| \chi_\alpha^+ \right\rangle \tag{7}$$

respectively, with $H_{el}$ the electronic Hamiltonian.

Cross-sections for single ionization of water and single electron capture from adenine by impact of bare ions, are reported in the following section. In order to represent the corresponding initial orbitals a complete neglect of differential overlap (CNDO) approximation is employed. The molecular orbitals are assumed to be described by linear combinations of their atomic compound orbitals (LCAOs). The corresponding binding energies are obtained within a Hartree–Fock formalism. For more details, we refer the reader to our previous works (Olivera et al., 1996; Galassi et al., 2012).

Employing the CNDO approximation, doubly differential cross-sections (DDCSs) for ionization as a function of the energy $E_k$ and solid angle $\Omega_k$ subtended by the ejected electron and total cross-sections (TCSs) for electron capture are expressed as

$$\begin{aligned}\frac{d^2\sigma}{dE_k\,d\Omega_k} &= \sum_{j=1}^{N}\frac{d^2\sigma_j}{dE_k\,d\Omega_k}\\ &= \sum_{j=1}^{N}\sum_{i=1}^{N_j}\xi_{i,j}\frac{d^2\sigma_{at,i}}{dE_k\,d\Omega_k}\end{aligned} \tag{8}$$

and

$$\sigma = \sum_{j=1}^{N}\sigma_j = \sum_{j=1}^{N}\sum_{i=1}^{N_j}\xi_{i,j}\sigma_{at,i} \tag{9}$$

respectively. In Eqs. (8) and (9), $N$ is the number of molecular orbitals, $N_j$ is the total number of atomic components of the $j$-