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N. Bachi

G. S. Otero

S. Otranto

Ronald E. Olson
Missouri University of Science and Technology, olson@mst.edu

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The Role of Multiple Electron Processes for Fast Ion H_2O Collisions

N. Bachi[1,a)], G. S. Otero[1], S. Otranto[1] and R. E. Olson[2]

[1]*Instituto de Física del Sur (IFISUR), Departamento de Física, Universidad Nacional del Sur (UNS), CONICET, Av. L. N. Alem 1253, B8000CPB-Bahía Blanca, Argentina.*
[2]*Physics Department, Missouri University of Science and Technology, Rolla MO 65409, USA.*

[a)]Corresponding author: nicolas.bachi@uns.edu.ar

Abstract. In this work, collision processes between C^{6+}, O^{8+} and Si^{13+} ions and H_2O molecules that lead to target ionization are studied by means of the classical trajectory Monte Carlo method. We employ an 8-electron model in which the H_2O electron densities and energies dynamically adapt during ionization processes to those corresponding to the H_2O^{q+} ions by assuming vertical transitions between the different molecular ionic states. Net total ionization cross sections and single differential cross sections in energy and angle are shown for 4 MeV/u projectiles and compared to available experimental data and theoretical predictions from distorted wave models.

INTRODUCTION

Ionization studies involving charged particles and H_2O molecules have been subject of many experimental and theoretical studies in the last few decades. Nowadays, complementary sets of data from different groups give insight on the ionization mechanisms acting on light particle [1-5] as well as on heavy particle collisions [6-15]. Besides the conceptual richness in terms of basic underlying physics, these collision systems provide benchmarks to improve our present theoretical and experimental capabilities towards an accurate description of collision processes involving molecular systems of increasing complexity.

In addition, by taking the H_2O molecule as the archetype compound of biological media, the obtained cross sections are of potential relevance for radiation therapies at their planning stages, in which Monte Carlo based codes are used to simulate the spatial extension for the irradiation procedure. These codes keep track of the history followed by the charged particles in the beam using collisional cross sections to statistically determine each individual step as they transit the media, and the possible generation of secondary electrons which eventually lead to subsequent ionization events.

In this work, we theoretically study ionization processes resulting in the emission of one or more electrons in 4 MeV/u C^{6+}, O^{8+} and Si^{13+} collisions on H_2O molecules. In Section II, we detail the main features of the semiclassical model hereby used, that is based on the classical trajectory Monte Carlo (CTMC) method. In Section III we show the obtained total cross sections and single differential cross sections in energy and angle and compare them to the recently reported experimental data of Bhattacharjee [13]. Total net ionization cross sections are calculated and compared to the predictions of the continuum distorted wave-eikonal initial state (CDW-EIS) calculations reported in its prior and post formalism for a one-active electron target [13]. Finally, conclusions are drawn in Section IV.

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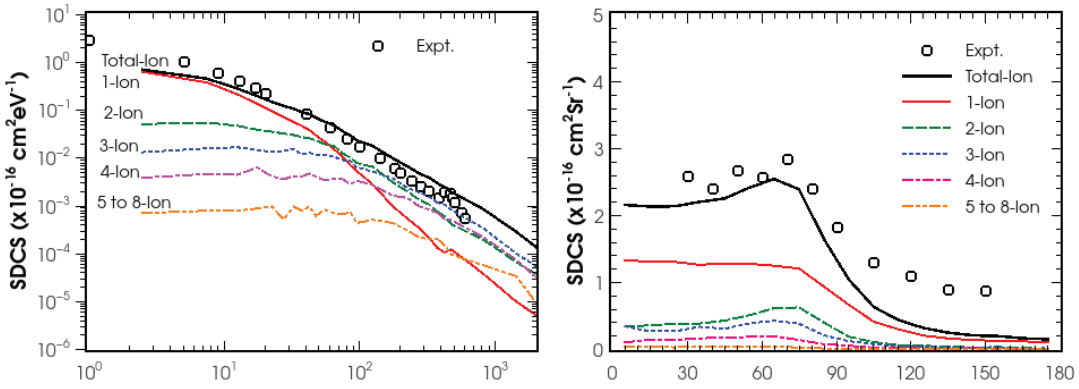

FIGURE 1. Pure ionization SDCS in energy and angle for 4 MeV/u Si^{13+} collisions on H_2O.

THEORETICAL MODEL

In the present CTMC code, Hamilton equations for the projectile, the three molecular centers and eight non-interacting electrons are numerically solved using a Runge-Kutta-Gill integrator with adaptive-step size. Each of the electrons evolves under a 3-center potential which can be represented as the sum of terms corresponding to the O and the H centers:

$$V_{eO}(r) = -\frac{8-(N_O-1)(1-e^{-\alpha_O r})}{r}, \qquad V_{eH}(r) = -\frac{8-(N_H-1)(1-e^{-\alpha_H r})}{r} \tag{1}$$

Here, r represents the electron distance to the respective molecular center. Within this parameterization, the electron asymptotically sees the molecule as a core charge of +1 and a Coulomb functional form applies.At closer distances, on the other hand, the potential value depends on how close the electron is situated with respect to each molecular center. How this transition takes place as the electron position is varied depends on the parameters N_O, N_H, α_O and N_H. In our case, the parameters $N_0 = 8.18$, $N_H = 1.91$, $\alpha_O = 3.06$ and $\alpha_H = 5.07$ are determined by a least squares fitting procedure over the molecular anisotropic potential derived from the $\varphi_j(\mathbf{r})$ molecular orbitals provided by Moccia [16] and that is given by,

$$V^{j}_{H_2O}(r) = -\sum_{l=1}^{3}\frac{Z_l}{|r-R_l|} + \sum_{j=1}^{N_{MO}} N_{ij}\int dr' \frac{|\varphi_j(r')|^2}{|r-r'|} \tag{2}$$

In this expression, Z_l are the nuclear charges of the atoms making up the molecule. The sum index j runs over the valence molecular orbitals $1b1$, $3a1$, $1b2$ and $2a1$ and $N_{ij} = 2$ for $i \neq j$ and $N_{ij} = 1$ for $i = j$. The molecular centers are kept fixed at their equilibrium relative distances $r_{OH} = 1.814$ a.u. and $r_{HH} = 2.908$ a.u. The present least squares procedure provides the best overall fit by minimizing $\chi^2 = \chi_{1b1}^2 + \chi_{3a1}^2 + \chi_{1b2}^2 + \chi_{2a1}^2$.

The eight non-interacting electrons are sorted with the ionization potentials of the H_2O ground state molecular orbitals. As one of the electrons is ionized and acquires a positive relative energy with respect to the target center of mass, the remaining electrons are spatially re-sorted considering the ionization potentials of the H_2O^+ orbitals assuming a vertical transition for the molecule. The re-sorted electrons keep their original momenta and see the asymptotic charge of the O-atom increased by one unit in this step. This procedure is repeated as new electrons are emitted from the target. By doing so, a dynamical adaptation of the target is achieved which leads to a proper description of the energetic electron density in the H_2O^{q+} molecular ion at all times, together with correct ionization potentials which are particularly relevant for multiple electron removal. An asymptotic energy and momentum analysis for all the emitted electrons following each collision event allows determining the total net cross section and the associated Single Differential Cross Sections (SDCS) in energy and angle.

RESULTS

First, we show the present SDCS in energy and angle for 4 MeV/u Si^{13+} collisions on H_2O and compare to the experimental data recently reported by Bhattacharjee *et al* [15]. By inspecting the collected events, we conclude that single ionization dominates the emission of low energy electrons, while multiple ionization dominates the energetic emission. This clearly reflects the fact that multiple ionization is associated to low-impact parameter collisions, for which head-on-like collisions are much more probable. Overall, the present theoretical results seem to underestimate the soft electrons region, a fact that could be partially due to two well known limitations of this classical procedure: i) the classical cutoff in the radial electron distributions which limits the contribution arising from large impact parameters, and ii) the lack of quantal tunneling that leads to the incorrect asymptotic dependence $1/E$ for the single ionization cross section instead of the correct $log(E)/E$ [17].

Regarding the SDCS in angle, the present model leads to good agreement with the experimental data for ($\theta_e < 90°$), exhibits a maximum at about $\theta_e = 65°$ in good agreement with the experimental data and underestimates the large angle emission ($\theta_e > 90°$). By analyzing the separate contributions from the single and multiple ionization channels, we note that the single ionization mechanism exhibits a decreasing behavior which accentuates for ($\theta_e > 80°$). In contrast, the multiple ionization channels are the ones that provide the peak-structure that is also suggested by the data.

We now move to the total net cross section for pure ionization $\sigma_{net} = \sigma_{1\text{-}ion} + 2\sigma_{2\text{-}ion} + 3\sigma_{3\text{-}ion} + ..+8\sigma_{8\text{-}ion}$. In Figure 2 we show σ_{net} as a function of the projectile charge Z_P. The experimental data from Bhattacharjee *et al* and the CDW-EIS predictions in the post and prior formalisms are included for comparison. While data for C^{6+} and Si^{13+} have been reported at 4 MeV/u, for O^{8+} measurements were made at 3.75 MeV/u and 4.5 MeV/u. Hence, the 4 MeV/u value is estimated from an interpolation between these two impact energies. Explicit values for σ_{net} are given in Table I. Present theoretical results are in reasonable agreement with the experimental data for C^{6+} and Si^{13+} projectiles. The general trends of the theoretical results lead to a smooth increasing behavior with projectile charge state Z_P, in contrast to the experimental data which display a maximum for O^{8+}. On the other hand, and in concordance with the present CTMC results, both formulations of CDW-EIS also suggest a smooth increasing behavior with Z_P but exhibit a consistent overestimation of the present results by a factor 2 to 3. As a result, the CDW-EIS theories provide results in good agreement with the experimental data for O^{8+} but clearly overestimate those corresponding to C^{6+} and Si^{13+} [14].

TABLE 1. Total net cross sections σ_{net} (in 10^{-16} cm^2) for 4 MeV/u C^{6+}, O^{8+} and Si^{13+} collisions on H_2O. (*) Interpolated data from reported cross sections at impact energies 3.75 MeV/u and 4.5 MeV/u.

	Present Theory	**CDW-EIS prior**	**CDW-EIS post**	**Expt.**
C^{6+}	4.98	12.5	10.6	7.2
O^{8+}	7.89	23.0*	20.0*	18.4*
Si^{13+}	17.5	50.9	43.3	23.1

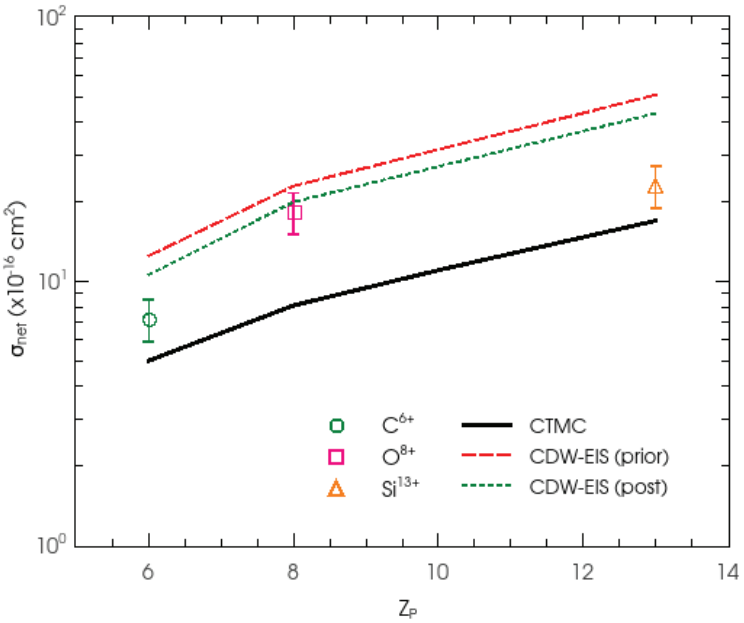

FIGURE 2. Total net cross section σ_{net} for 4 MeV/u C^{6+}, O^{8+} and Si^{13+} collisions on H_2O. The experimental data and CDW-EIS theoretical results in prior and post forms of ref. [14] are included for comparison.

CONCLUSIONS

In this work a semiclassical analysis of ionization of the H_2O molecule by ion impact has been performed identifying the comparative role of the single ionization and multiple ionization channels. In contrast to formulations based on one-active electron descriptions, the present model allows for the simultaneous treatment of the eight electrons in the valence molecular orbitals. Despite the fact that electron-electron correlation terms cannot be explicitly included since these would turn the target unstable, a dynamic adaptation of the target has been conceived to include the vertical transitions and the change of ionization potentials of the remaining electrons as the molecule is ionized.

According to the present description, energetic electron emission is associated to multiple ionization in closer encounters between the projectile and the H_2O molecule, while low energy electrons are mostly generated in single ionization processes which correspond to larger impact parameters.

Further work is currently under way involving larger projectile charges to gain insight on the role of multiple ionization processes in a collision regime for which the Bohr parameter $Z_P/v_P > 1$, that is beyond the accepted range of validity for perturbative methods.

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