Theoretical and experimental investigations of electron emission in $He^{2+}+H_2O$ collisions

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Very recently, Ohsawa *et al.* have presented a new experimental apparatus for measuring doubly differential cross sections of electron emission from water vapor with fast heavy-ion impact (6–25 MeV). Successful measured energy and angular distributions of secondary electrons were produced for a collision of 6.0 MeV/u He^{2+} ions with vapor water [Nucl. Instrum. Methods Phys. Res. B **227**, 431 (2005)] and for 10.0 MeV/u He^{2+} ions [Phys. Rev. A **72**, 062710 (2005)]. The present theoretical work aims to reproduce these very recent measurements as well as more former existing data dedicated to low-energy alpha particles. Doubly, singly differential, and total ionization cross sections have then been studied.

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I. INTRODUCTION

With the more and more regular use of ionizing radiations in medicine, it seems today necessary to better appraise the biological consequences of radiological examinations, particularly to know, with the highest degree of accuracy, the energy deposits induced by all the radiations commonly used in radiotherapy and even in medical imaging (light and heavy ions, electrons and positrons, x rays and γ rays). This implies to describe, at the nanometric scale, each collision induced by the projectile in order to quantify the full spectra of molecular damage radio induced and then to access to a cellular dosimetry. To do that, the preferential method is the Monte Carlo simulation, which consists in simulating, step-by-step, interaction after interaction, the history of each ionizing particle created during the irradiation of the biological matter. Each projectile-target interaction is then described by means of multidifferential and total cross sections in order to access to the complete kinematics of the collision. In these conditions, we clearly understand the necessity for radiobiologists, radiotherapists, and nuclear doctors to access to accurate differential and total cross sections to know the fine structure of the ionizing particle in the living matter, this latter being commonly described by water.

To date, there are a number of Monte Carlo track structure codes devoted to light ions (protons and alpha particles), which have been developed independently (PTRAN [1], PETRA [2], STBRGEN [3], DELTA [4], TRION [5], LEPHIST [6], and LEAHIST [7]). However, most of them are based on a semiempirical description of the ionization process by means of least-squares fittings of experimental measurements of differential as well as total cross sections.

Thus, for alpha particles (LEAHIST), Uehara and Nikjoo have used a polynomial fit of the experimental data of Rudd *et al.* measured between 10 and 300 keV/u [8] and of Toburen *et al.* measured between 75 and 500 keV/u [9] to express the total ionization cross sections. At energies below 10 keV/u, smooth extrapolations were also made whereas for energies greater than 500 keV/u extrapolations were performed by taking into account both the reproducibility of stopping powers and the scaling of proton ionization cross sections. To treat the energy spectra of the secondary electrons, the authors used the Rudd's model, initially developed for protons, which provides a parametrization of the electron spectrum after ion impact. This approach is based on a binary encounter model modified to agree with the Bethe theory at high energies and with the molecular promotion model at low energies [10]. Relatively good agreements were then found between the experimental singly differential cross sections (SDCS) and the Rudd's results by means of a large number of fitting parameters. Thus, Uehara and Nikjoo reported in Ref. [7] that this model satisfactorily reproduced the average energies of the secondary electrons ejected by He^{2+} ions for ion energies lower than 300 keV/u, but that it underestimated them for greater energies (up to about 30% at 2 MeV), which could be corrected by using suitable scaling factors (see Ref. [7] for more details). Furthermore, concerning the angular distributions for secondary electrons (namely, the doubly differential cross sections, DDCS), Uehara and Nikjoo mentioned that ICRU [11] has recommended the use of a semiempirical formula given by Hansen, Kocbach, and Stolterfoht (called the HKS model [12]) for determining the DDCS for electron emission by heavy ions. However, as underlined by the authors, these semiempirical DDCS give only limited agreements with the experimental data. Under these conditions, Uehara and Nikjoo preferred to use a random sampling of ejected directions among the experimental data. However, these latter are limited and there has been a very small amount of cross section data on electron emission from water vapor produced by charged particle impact.

On the experimental side, the literature remains very poor. We can, for example, cite the extensive graphical representation given by Toburen *et al.* [9], and very recently the doubly differential cross sections for the energy and angular distributions of secondary electrons produced in collisions of 6.0 and 10.0 MeV/u He^{2+} ions in water vapor [13,14] and even more recently for 15.0 MeV/u alpha particles. In this latter, DDCS were presented and compared in the low-energy region to the Toburen's data by means of Fano plots and very good consistency was found. SDCS were finally

deduced and compared to the semiempirical model introduced by Rudd *et al.* [10].

Describing the ionization process at the multidifferential level needs sophisticated theoretical calculations that do exist for atomic targets but that remain scarce for molecules, and more particularly for water molecule. In fact, one of the basic difficulties in describing the ionization process in ion-molecule collisions at intermediate to high energies (i.e., the energy regime considered in this work) arises from the long range of the Coulomb interaction between all the charged particles. This problem can be overcome by distortion effects introduced into the initial and final channels as performed in the theoretical model reported by Olivera *et al.* for calculating the contribution from the inner shell of water vapor dose profile [15,16]. In this work, the authors have calculated doubly differential cross sections for single ionization of vapor water by ion impact as a function of electron energy and angle by using the *continuum-distorted-wave-eikonal-initial-state* (CDW-EIS) approximation, initially developed by Fainstein *et al.* for protons traversing H, He, and simple molecular targets [17,18]. This model was a first-order model of a distorted-wave series, which considered distortions (due to the long-range Coulomb potential in both the initial and final channels) introduced as multiplicative factors in the initial bound and final continuum states of the molecular target [19]. In this way, it was then possible to account for two-center effects, which were not included in the first Born or plane-wave approximations. Furthermore, there is an additional difficulty in modeling the ionization process of the molecule target, namely, the description of the molecular states of the target. We therefore distinguish different convenient ways to treat this problem: a first one, called Bragg's additivity rule, which consists in expressing each molecular cross section (differential as well as total) as a linear combination of atomic cross sections weighted by the number of atoms in the molecule [16,19] and a second one, called *complete neglect of differential overlap* (CNDO), where the molecular orbitals are written in terms of atomic orbitals of the atomic constituents [20,21]. However, as underlined by Galassi *et al.* [19], in both these models the calculated doubly differential cross sections exhibit unsatisfactory agreement with the experimental data at small angle regions. The authors linked these discrepancies to the fact that the electronic populations were not correctly reproduced in these two descriptions, especially for the calculations within the Bragg's rule framework. Finally, a third method to calculate the populations of the target is that of molecular orbitals constructed from a linear combination of atomic orbitals in a self-consistent field (MO-LCAO-SCF) [22], whose quality has already been highlighted by Galassi *et al.* [19] for low-Z molecule ionization by proton impact at intermediate and high energies. In previous works, we have used this kind of description, and more precisely that given by Moccia [23] who expressed molecular wave functions of small molecules like H_2O, NH_3, and CH_4 by linear combinations of Slater-type functions. Numerous studies have then been produced about ionization by electron impact in terms of multidifferential as well as total cross sections [24–28]. Very recently, we have extended our full-differential model to the water ionization by proton impact, in a large impact energy range (0.1–100 MeV) and provided differential and total cross sections, which compared very satisfactorily with a large set of experimental data (see Ref. [29] for more details).

In the same state of mind, we propose by the present work to test our model to the alpha particle case and to compare the theoretically calculated DDCS and SDCS to the experimental measurements recently obtained by Ohsawa *et al.* [14] for 6.0 and 10.0 MeV/u He^{2+} ions as well as the last measurements performed for 15.0 MeV/u He^{2+} ions. To be more complete, we have also reported a comparison with more former measurements, taken from Toburen *et al.* [9], Rudd *et al.* [8], and Rudolph and Melton [30] concerning low-energy alpha particles.

In the present paper, Sec. II deals with the experimental setup and the measurement conditions. In Sec. III we briefly present the theoretical model and in Sec. IV, the calculated doubly and singly differential cross sections (DDCS and SDCS, respectively) as well as the total cross section (TCS) are successively presented and compared to a large part of the existing experimental measurements. Finally, in Sec. V, a conclusion about the modeling of the ionization of a water molecule is given. Atomic units are used throughout unless otherwise indicated.

II. EXPERIMENT

An outline of our apparatus [13] is given below. The used method is one of the well-known cross-beam techniques, in which incident ions interact with a vertically emitted water-vapor molecular beam. Ejected secondary electrons are detected by a Chevron-type microchannel-plate (MCP) assembly after being analyzed by a 45° inclined parallel-plate electrostatic spectrometer, which is rotatable from 20° to 160° with respect to the incident-beam direction. Water vapor is vertically emitted into the interaction region from a nozzle of 1×15 mm^2 aperture, and is instantly frozen and trapped as ice on a stainless-steel panel, which is cooled by liquid nitrogen. With this water-vapor generation and collection system, a stable water-vapor jet (10^{-2}–10^{-3} Torr) was obtained without deteriorating the pressure in the scattering chamber: $\sim4\times10^{-7}$ Torr with a vapor flow of 40.0 cm^3/min. Some new techniques allowed us to improve the estimated uncertainty (systematic error) to be on the order of ±13%, except for high-energy electrons.

In a recent measurement with 10.0 MeV/u He^{2+} ions, we tried to evaluate the DDCS at electron energy down to 1 eV. However, the measured values were considerably fluctuated due to the magnetic field produced by the ON/OFF control in the dc current for a heater surrounding the nozzle for the emission of water vapor, at which the temperature was stabilized to 100 °C.

In addition, we recently found a significant problem. During 2–3 years of measurements, our chamber (made of iron) was magnetized due to the effects by both the heater and the opening/closing process of the upper lid of the chamber for maintenance. In the latter process, slight but frequent collisions between the lid and chamber would produce some magnetization, resulting in a considerable increase in the residual magnetic field (by a factor of ~5) at around the col-

lision center. The lower limit in measurable energies with a small uncertainty has thus been unfortunately changed to ~20 eV in recent measurements from ~7 eV in the early measurements. Meanwhile, the higher limit was slightly improved (increased) from 10 to 12–14 keV by careful operation of the deflector electrode against sparks, though it is still lower than the maximum energy (22.0 keV) in collisions of 10 MeV/u projectiles.

The following modifications were made for the 15 MeV/u He^{2+} experiments: the chamber was demagnetized and the power supply for heaters changed to an ac type; the metal panel (made of Cu at present) surrounding the interaction region is also changed to that made of μ-metal, in order to suppress the effects of stray magnetic fields, the details of which will soon be published elsewhere.

III. THEORY

The DDCS presented in this work have been calculated in the Born approximation framework by testing several theoretical models already used to treat the water ionization by protons. Therefore, the theory will be briefly reported in the present section and for more details we refer the reader to our previous work [29].

A. Cross section calculation

Contrary to the *plane wave Born approximation* (PWBA) in which all particles are described by plane wave, the ejected electron is, in the *first Born approximation* (FBA), described by a Coulomb wave (we speak of the FBA-CW model), the incident ion and the scattered electron remaining described by plane waves. In these conditions, the nonrelativistic triple differential cross section (TDCS), without exchange, is simply given by

$$\frac{d^3\sigma}{d\Omega_e d\Omega_s dE_e} = 16M_p^2 Z_{\text{ion}}^2 \frac{k_e k_s}{k_i}|T|^2, \tag{1}$$

where Ω_s and Ω_e represent the solid angles of detection for the scattered ion and for the ejected electron, respectively, M_p the proton mass, and Z_{ion} the ion charge ($Z_{\text{ion}}=2$ in the present work). The momenta $\mathbf{k}_i$, $\mathbf{k}_s$, and $\mathbf{k}_e$ are related to the incident projectile, to the scattered ion, and to the ejected electron, respectively. They depend on the corresponding energy through the relations $k_i^2=8M_pE_i$, $k_s^2=8M_pE_s$, and $k_e^2=2E_e$. The matrix element T describes the transition of the system from the initial state to the final state.

However, as detailed in Refs. [28,29], several improvements can be brought into the FBA-CW model to describe the ionization process in the Born approximation.

(1) The ejected electron can be described by means of a distorted wave function calculated by the numerical resolution of the Schrödinger equation where distortion effects between the ejected specie and the ionized target are introduced (one speaks of *distorted wave Born approximation*, DWBA).

(2) The scattered ion and the ejected electron can both be described by a Coulomb wave (2CW model) (see Refs. [29,31] for more details).

In these two models, the effective charges seen by the escaping electron and ion are defined in this work as the effective ionic charges [29] and have been taken to be equal to 1, whereas some authors like Galassi *et al.* [19] defined them as $Z_T=\sqrt{-2n\epsilon_i}$, where n is the principal quantum number corresponding to each atomic subshell that constitutes the initial bound state of the active electron in the molecule: however, let us note that this discrepancy essentially affects the inner subshells of the target for which the cross sections have minor contribution into the ionization process.

(3) Finally, one of the most sophisticated models used to treat the ionizing process is that introduced by Brauner, Briggs, and Klar (called the BBK *model*) [32], which consists in exhibiting a correct asymptotical Coulomb three-body wave function by describing the final state by the product of three Coulomb waves taking into account the interaction between the scattered ion and the ionized target, the interaction between the ejected electron and the ionized target, and the scattered ion-ejected electron interaction.

However, as explained in Refs. [28,29], all these sophisticated descriptions are essentially needed for experimental configurations in which the ejected velocity V_e matches the scattered velocity V_s and consequently do not affect the present investigations. Indeed, all the experimental results provided by Ohsawa *et al.* and reported in the present work consider particles with an ejected velocity $V_e<7$ a.u. ($E_e<673$ eV) and a scattered velocity $V_s>15.5$ a.u. ($E_s>6$ MeV/u). Thus, the differential cross sections have always been calculated in the FBA-CW model except for the experimental conditions reported by Toburen *et al.* [9] (corresponding to an ejected velocity close to the velocity of the alpha particle) for which the 2CW model and the most sophisticated BBK model were used.

B. Target description

To describe the water molecule in vapor phase, we have used the molecular description proposed by Moccia [23], who developed each of the five molecular wave functions in terms of Slater-type functions, centered at a common origin, namely, upon the heaviest nucleus, i.e., the oxygen atom. Let us note that these functions refer, for a particular molecular orientation given by the Euler angles (α,β,γ), to the calculated equilibrium configurations, which agree well with the experimental data (see Ref. [24] for a summary). Under these conditions, the ten bound electrons of the water molecule are distributed among five one-center molecular wave functions $\upsilon_j(\vec{r})$ (with j ranging from 1 to 5) corresponding to the orbitals $^1B_1$, $^3A_1$, $^1B_2$, $^2A_1$, and $^1A_1$ whose binding energies are 0.4954 a.u., 0.5561 a.u., 0.6814 a.u.,1.3261 a.u., and 20.5249 a.u., respectively. Each of them is expressed by linear combinations of Slater-type functions and is written as

$$\upsilon_j(\vec{r}) = \sum_{k=1}^{N_j} a_{jk}\Phi_{n_{jk}l_{jk}m_{jk}}^{\xi_{jk}}(\vec{r}), \tag{2}$$

where N_j is the number of Slater functions used in the development of the jth molecular orbital and a_{jk} the weight of each real atomic component $\Phi_{n_{jk}l_{jk}m_{jk}}^{\xi_{jk}}(\vec{r})$ written as

$$\Phi_{n_{jk}l_{jk}m_{jk}}^{\xi_{jk}}(\vec{r}) = R_{n_{jk}}^{\xi_{jk}}(r) S_{l_{jk}m_{jk}}(\hat{r}), \tag{3}$$

where the radial part $R_{n_{jk}}^{\xi_{jk}}(r)$ is given by

$$R_{n_{jk}}^{\xi_{jk}}(r) = \frac{(2\xi_{jk})^{n_{jk}+1/2}}{\sqrt{(2n_{jk})!}} r^{n_{jk}-1} e^{-\xi_{jk}r}, \tag{4}$$

and where $S_{l_{jk}m_{jk}}(\hat{r})$ is the so-called real solid harmonic [33] expressed by

$$\text{if } m_{jk} \neq 0 \quad S_{l_{jk}m_{jk}}(\hat{r}) = \left(\frac{m_{jk}}{2|m_{jk}|}\right)^{1/2} \left\{ Y_{l_{jk}-|m_{jk}|}(\hat{r}) + (-1)^{m_{jk}} \left(\frac{m_{jk}}{|m_{jk}|}\right) Y_{l_{jk}|m_{jk}|}(\hat{r}) \right\}$$

$$\text{if } m_{jk} = 0 \quad S_{l_{jk}0}(\hat{r}) = Y_{l_{jk}0}(\hat{r}). \tag{5}$$

IV. RESULTS AND DISCUSSION

As mentioned above, the water bound state provided by Moccia [23] corresponds to a particular orientation of the molecular target given by the Euler angles (α, β, γ). Therefore, the differential cross sections accessible in first by our code correspond to *eightfold differential cross sections* (8DCS) [27], which describe the ionizing process in terms of scattered and ejected angular distributions, ejected energy distribution, and for a given oriented water molecule. The TDCS are then analytically obtained by integration of the 8DCS over the Euler solid angle Ω_{Euler} (with $d\Omega_{\text{Euler}} = \sin\beta\, d\alpha\, d\beta\, d\gamma$) whereas the DDCS are obtained by numerical integration of the TDCS over the scattered detection angle Ω_s. Finally, the SDCS are obtained after integration of the DDCS over the ejected detection angle Ω_e. This procedure is obviously performed for each of the five orbitals of the water molecule and the differential cross sections presented here correspond to "global" differential cross sections obtained by summing up all the subshell contributions weighted by the number N_{elec} of electrons per orbital, i.e., $N_{\text{elec}}=2$.

As highlighted above, the ionizing process for charged particles has already been investigated in radiobiological studies to appreciate the real biological risks induced during (un)controlled irradiations. However, these works are, in a major part, essentially limited to singly differential and total cross section calculations, these latter being performed by means of semiempirical analytic expressions deduced from experimental data (see, for example, Ref. [34]). Among them, we can cite the work of Uehara and Nikjoo [7] who have recently presented the development of the new generation of the Monte Carlo track-structure code called LEAHIST in which the ionization process for bare, dressed, and neutral helium ions were essentially obtained from experimental data when they exist and from fitting expressions and extrapolations otherwise. Thus, for He^{2+} ions, the authors made several approximations whose main characteristics can be summarized as follows: (i) the total cross sections were either deduced from polynomial functions fitting the experimental data of Rudd *et al.* [8] (measured between 10 and 300 keV/u) and the data of Toburen *et al.* [9] (measured between 75 and 500 keV/u) or extrapolated (for greater energies) by taking into account both the reproducibility of stopping powers and the scaling property between the proton and alpha particle ionization cross sections; (ii) to treat the energy distribution of the secondary ejected electrons the authors used, as a first approximation, the Rudd's model initially developed for protons and then applied for studying the case of He^{2+} ions. Thus, by using a large number of fitting parameters (namely, ten parameters), the model of Rudd reproduced the SDCS of He^{2+} ions of energy lower than 300 keV/u. For greater energies, scaling factors were used to correct the underestimated cross sections and then to reproduce the good average energies, which were underestimated up to about 30% at 2 MeV/u; (iii) considering now the angular distributions of the secondary electrons, the authors followed the recommendation of the ICRU [11] for using the semiempirical formula of Hansen-Kocbach-Stolterfoht (or the HKS model), which provides readily calculable DDCS with reasonably good agreement with the experimental angular distributions for the ejected electrons with various energies for high-energy projectiles. In fact, experimental DDCS measurements are very rare and essentially limited to low-energy He^{2+} ions (see the experiments of Toburen *et al.* [9]). Therefore, to cover the broad ion energy range, the DDCS were also deduced from the more abundant experimental data for protons by assuming that the differences between the angular distributions of these two ions were minor.

Contrary to this work, the models we propose in the present paper need *neither experimental adjustments nor fitting parameters*. They are only based on quantum-mechanical developments performed in the *Born approximation*. In the major part of the experimental conditions reported in this work, we used the FBA-CW model where the incident and scattered particles are described by a plane wave and the ejected electron is described by a Coulomb wave. However, when the velocities of the incident alpha particle and of the ejected electron are very close, we used the BBK model. Moreover, it is also important to remember that the water target is described by means of molecular wave functions expressed as a linear combination of atomic orbitals in a self-consistent field (MO-LCAO-SCF), whereas it was treated in the works previously cited [16,19–21] either by using the Bragg's additivity rule to express the total molecular cross sections or in the CNDO approximation where the molecular orbitals were written in terms of atomic orbitals of the atomic constituents.

Thus, in view of the foregoing, the present study appears as the first theoretical work dedicated to fully differential as well as total cross section calculations for the water ionization by alpha particles.

A. Doubly differential cross sections

In Fig. 1, we compared the calculated DDCS to the experimental results provided by Ohsawa *et al.* [14] for 6 MeV/u He^{2+} ions ejecting electrons of 19.2, 38.5, 96.2, 192, 385, and 673 eV, respectively. Very good agreements

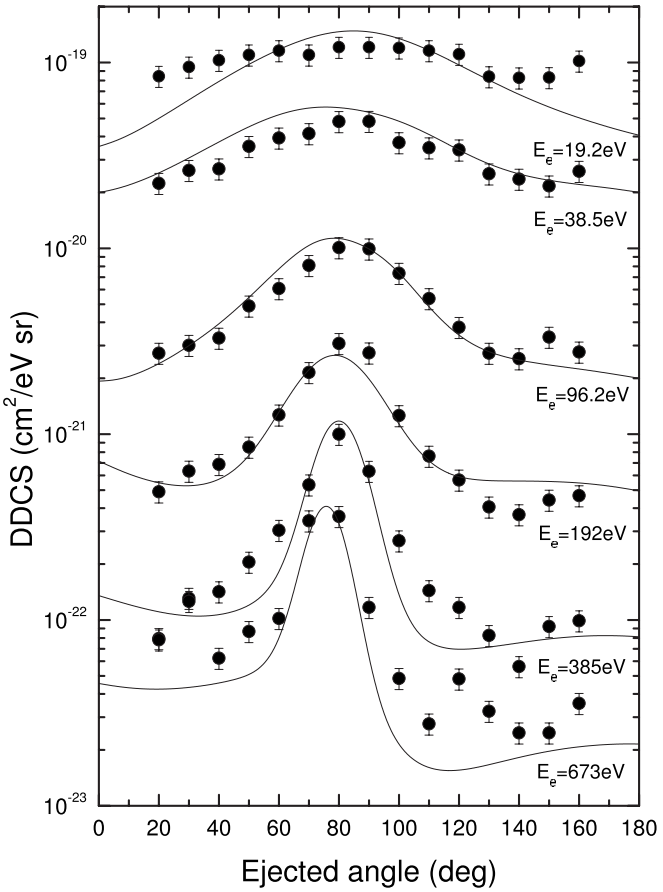

FIG. 1. DDCS for water ionization by alpha particles of 6.0 MeV/u. The experimental data (solid circles) of Ohsawa *et al.* [14] are compared to the theoretical results (FBA-CW model) for six different ejected electron energies: E_e=19.2, 38.5, 96.2, 192, 385, and 673 eV.

can be observed between the experimental and theoretical results in all cases, except for the lower ejected energy reported, i.e., for E_e=19.2 eV for which our DDCS overestimate the experimental data essentially for $\theta_e<60°$ and $\theta_e>120°$. This discrepancy may be due to the distortion induced by the incident ion, which is not taken into account in the present work. For more details, we refer the reader to our previous work [29] where several sophisticated models are tested for proton-water collision, and particularly, the Brauner, Briggs, and Klar (BBK) model, which represents a real improvement, essentially for electron velocities matching the scattered ones. For $\theta_e>60°$, the agreement becomes very good for each ejected energy considered and we theoretically observe the appearance of the binary peak located at a critical value of θ_e, denoted $(\theta_e)_{\text{crit}}$ of about 85.3°, 76.3°, 79.6°, 79°, 80.1°, and 75.8°, respectively, for the different ejected energies investigated, which agrees well with the experimental observations (see Table I). Theoretically, these values correspond to binary collisions in which the energy lost by the incident particle is completely transferred to the target molecular electron with the residual ion acting as a spectator [35,36]. This region, called Bethe ridge, can be simply defined by considering the collision of an incident projectile with a target electron at rest. After the collision, the electron recoils at an angle θ_e with kinetic energy E_e and corresponding momentum $\mathbf{k}_e$, while the incident ion is scattered at an angle θ_s with kinetic energy $E_s=E_i-E_e-I(j)$ and corresponding momentum $\mathbf{k}_s$, where $I(j)$ represents the binding energy of the molecular subshell ionized. Thus, from momentum conservation, we obtain

$$|\mathbf{q}| = |\mathbf{k}_i - \mathbf{k}_s| = k_e \Leftrightarrow \cos(\theta_e)_{\text{crit}} = \frac{k_i^2 + k_e^2 - k_s^2}{2k_i k_e}, \qquad (6)$$

where $\mathbf{q}$ represents the momentum transfer. However, since this quantity depends on the ionized molecular orbital [via the dependence on $I(j)$], $(\theta_e)_{\text{crit}}$ will be denoted $[\theta_e(j)]_{\text{crit}}$ in the following. In Table I we have reported the analytic $[\theta_e(j)]_{\text{crit}}$ values deduced from Eq. (6) for the four outer subshells of the water molecule whereas in Table II we have compared the values theoretically and experimentally observed: good agreements are found between them. Thus, we can theoretically predict a shift of the binary peak from $(\theta_e)_{\text{crit}}=85.3°$ at E_e=19.2 eV to $(\theta_e)_{\text{crit}}=75.8°$ at E_e=673 eV, which is experimentally observed.

Similarly, in Fig. 2, we have reported the calculated DDCS for 10 MeV/u He^{2+} ions ejecting electrons of 19.2, 38.5, 96.2, 192, 385, and 673 eV, respectively. The same behavior is observed with very good agreement over the overall ejection angle range for each ejected energy considered, except one more time for E_e=19.2 eV for which our DDCS overestimate the experimental data (for $\theta_e<60°$ and $\theta_e>120°$). Thus, a binary peak can be clearly observed for $(\theta_e)_{\text{crit}}$ going from 85°–90° at E_e=19.2 eV to 79–80° at E_e=673 eV (see Tables I and II).

We present in Fig. 3 the very recent experimental DDCS measurements of Ohsawa *et al.* (private communication), which consider more energetic He^{2+} ions, namely, E_i=15 MeV/u ejecting electrons of 38.5, 67.3, 96.2, and 385 eV. In this case, the agreement is less good since, for each of the ejected energy reported, the DDCS theoretically obtained overestimate the experimental observations for $\theta_e<60°$ and $\theta_e>120°$. However, the overall behavior is well reproduced and we particularly reproduce the appearance of a binary peak, which is shifted from about 85° at E_e=38.5 eV to about 83° at E_e=385 eV (see Tables I and II).

In Fig. 4(a), we compare the results obtained in the FBA-CW model to the experiments of Toburen *et al.* [9] for low incident energy of alpha particle (E_i=2 MeV) and for ejected electron energies E_e=10, 20, 50, 100, 200, 400, and 1000 eV, respectively. We generally observe a good agreement between the FBA-CW model and the experimental data for all electron energies except at small angles, particularly, for E_e=400 and 200 eV. The increase of the DDCS at small angles is probably due to the process of charge transfer to the continuum ECC. Indeed, in this particular case, a target bound electron may be captured into a continuum state of the alpha particle and then be emitted in the moving frame of the alpha particle. In fact, this process is included in the BBK model but not in the FBA-CW model (for more details, we refer the reader to Ref. [29]). To illustrate this particularity, we have reported in Fig. 4(b) a comparison between the theoretical results obtained in the three models investigated in

TABLE I. Theoretical values of the ejection angles corresponding to the binary peak for 6.0, 10.0, and 15.0 MeV/u He^{2+} ions and for the ejected energies investigated in this work: 19.2, 38.5, 96.2, l92, 385, and 673 eV.

E_e (eV)	E_i=6 MeV/u	E_i=10 MeV/u	E_i=15 MeV/u
19.2	$[\theta_e(^1B_1)]_{crit}=86.26°$	$[\theta_e(^1B_1)]_{crit}=87.10°$	$[\theta_e(^1B_1)]_{crit}=87.64°$
	$[\theta_e(^3A_1)]_{crit}=86.07°$	$[\theta_e(^3A_1)]_{crit}=86.96°$	$[\theta_e(^3A_1)]_{crit}=87.52°$
	$[\theta_e(^1B_2)]_{crit}=85.68°$	$[\theta_e(^1B_2)]_{crit}=86.65°$	$[\theta_e(^1B_2)]_{crit}=87.27°$
	$[\theta_e(^2A_1)]_{crit}=83.66°$	$[\theta_e(^2A_1)]_{crit}=85.10°$	$[\theta_e(^2A_1)]_{crit}=86.00°$
38.5	$[\theta_e(^1B_1)]_{crit}=85.79°$	$[\theta_e(^1B_1)]_{crit}=86.75°$	$[\theta_e(^1B_1)]_{crit}=87.34°$
	$[\theta_e(^3A_1)]_{crit}=85.66°$	$[\theta_e(^3A_1)]_{crit}=86.64°$	$[\theta_e(^3A_1)]_{crit}=87.26°$
	$[\theta_e(^1B_2)]_{crit}=85.39°$	$[\theta_e(^1B_2)]_{crit}=86.43°$	$[\theta_e(^1B_2)]_{crit}=87.08°$
	$[\theta_e(^2A_1)]_{crit}=83.96°$	$[\theta_e(^2A_1)]_{crit}=85.33°$	$[\theta_e(^2A_1)]_{crit}=86.19°$
96.2	$[\theta_e(^1B_1)]_{crit}=84.39°$	$[\theta_e(^1B_1)]_{crit}=85.65°$	$[\theta_e(^1B_1)]_{crit}=86.45°$
	$[\theta_e(^3A_1)]_{crit}=84.30°$	$[\theta_e(^3A_1)]_{crit}=85.59°$	$[\theta_e(^3A_1)]_{crit}=86.40°$
	$[\theta_e(^1B_2)]_{crit}=84.13°$	$[\theta_e(^1B_2)]_{crit}=85.45°$	$[\theta_e(^1B_2)]_{crit}=86.29°$
	$[\theta_e(^2A_1)]_{crit}=83.22°$	$[\theta_e(^2A_1)]_{crit}=84.76°$	$[\theta_e(^2A_1)]_{crit}=85.72°$
192	$[\theta_e(^1B_1)]_{crit}=82.55°$	$[\theta_e(^1B_1)]_{crit}=84.23°$	$[\theta_e(^1B_1)]_{crit}=85.29°$
	$[\theta_e(^3A_1)]_{crit}=82.49°$	$[\theta_e(^3A_1)]_{crit}=84.19°$	$[\theta_e(^3A_1)]_{crit}=85.26°$
	$[\theta_e(^1B_2)]_{crit}=82.36°$	$[\theta_e(^1B_2)]_{crit}=84.10°$	$[\theta_e(^1B_2)]_{crit}=85.18°$
	$[\theta_e(^2A_1)]_{crit}=81.72°$	$[\theta_e(^2A_1)]_{crit}=83.60°$	$[\theta_e(^2A_1)]_{crit}=84.78°$
385	$[\theta_e(^1B_1)]_{crit}=79.77°$	$[\theta_e(^1B_1)]_{crit}=82.10°$	$[\theta_e(^1B_1)]_{crit}=83.55°$
	$[\theta_e(^3A_1)]_{crit}=79.72°$	$[\theta_e(^3A_1)]_{crit}=82.06°$	$[\theta_e(^3A_1)]_{crit}=83.52°$
	$[\theta_e(^1B_2)]_{crit}=79.64°$	$[\theta_e(^1B_2)]_{crit}=81.99°$	$[\theta_e(^1B_2)]_{crit}=83.47°$
	$[\theta_e(^2A_1)]_{crit}=79.18°$	$[\theta_e(^2A_1)]_{crit}=81.64°$	$[\theta_e(^2A_1)]_{crit}=83.18°$
673	$[\theta_e(^1B_1)]_{crit}=76.62°$	$[\theta_e(^1B_1)]_{crit}=79.67°$	$[\theta_e(^1B_1)]_{crit}=81.58°$
	$[\theta_e(^3A_1)]_{crit}=76.58°$	$[\theta_e(^3A_1)]_{crit}=79.65°$	$[\theta_e(^3A_1)]_{crit}=81.56°$
	$[\theta_e(^1B_2)]_{crit}=76.52°$	$[\theta_e(^1B_2)]_{crit}=79.59°$	$[\theta_e(^1B_2)]_{crit}=81.52°$
	$[\theta_e(^2A_1)]_{crit}=76.17°$	$[\theta_e(^2A_1)]_{crit}=79.33°$	$[\theta_e(^2A_1)]_{crit}=81.30°$

the present work and the experimental data of Toburen *et al.* for an incident energy E_i=2 MeV and an ejected energy E_e=200 eV. We clearly observe that the FBA-CW model (solid line) as well as the 2CW model (dashed line) are not able to reproduce the strong increasing of DDCS at small angles due to the fact that these models do not take into account the ECC process, contrary to the BBK model (dotted line), which exhibits a better agreement by reproducing the shape of the data. However, let us note that this BBK model overestimates the DDCS at smaller angles.

In Fig. 5 we compared the results of our FBA-CW model to the data of Toburen [9] for an incident energy E_i=1.2 MeV. In this case, the DDCS are reported as a function of the ejected electron energy for three fixed ejection angles. The agreement is generally good except for low energy (less than 4 eV). We also notice that a small disagree-

TABLE II. Comparison between the theoretical and the experimental angles $(\theta_e)_{crit}$ corresponding to the binary peak for the incident α-particle and the ejected electron energies reported in the present work.

	E_i=6 MeV/u		E_i=10 MeV/u		E_i=15 MeV/u	
E_e (eV)	Theoretical $(\theta_e)_{crit}$	Experimental $(\theta_e)_{crit}$	Theoretical $(\theta_e)_{crit}$	Experimental $(\theta_e)_{crit}$	Theoretical $(\theta_e)_{crit}$	Experimental $(\theta_e)_{crit}$
19.2	85.3°	85°	84.6°	90°		
38.5	76.3°	85°	81.1°	90°	84.3°	85°
96.2	79.6°	85°	80.5°	90°	80.1°	85°
192	79°	80°	86.6°	85°		
385	80.1°	80°	81.2°	80°	82.8°	85°
673	75.8°	75°	79.2°	80°		

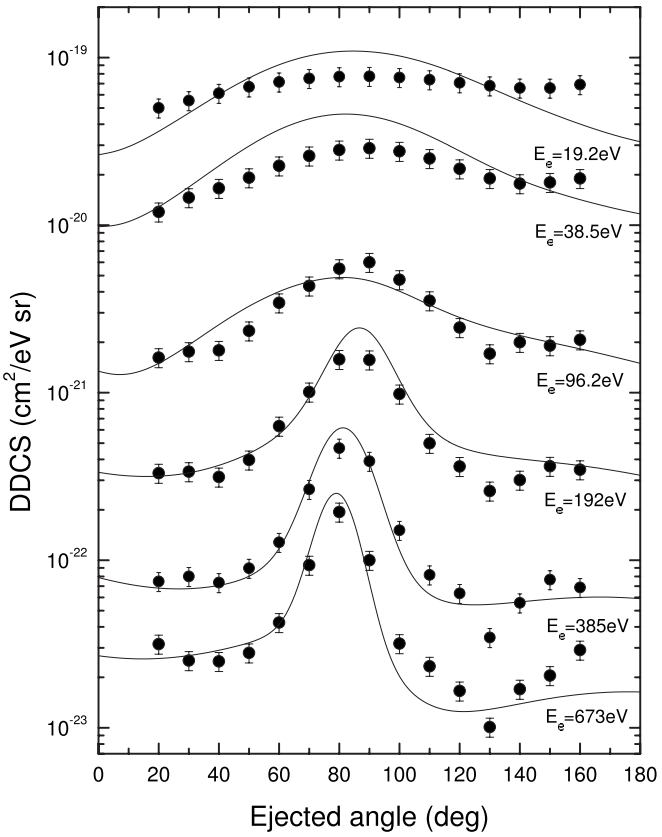

FIG. 2. Same as Fig. 1 with E_i=10 MeV/u.

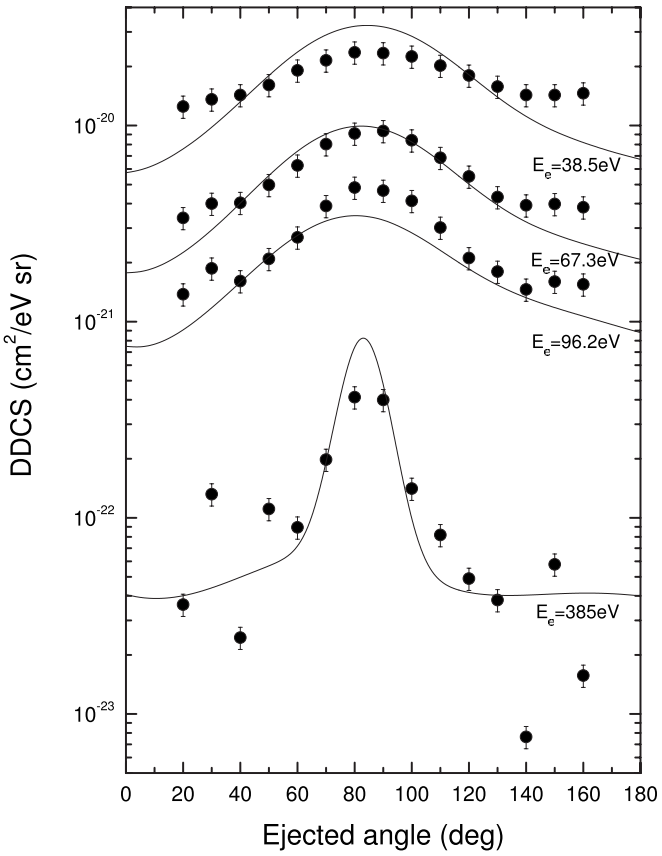

FIG. 3. Same as Fig. 1 with E_i=15.0 MeV/u and ejected electron energies E_e=38.5, 67.3, 96.2, and 385 eV.

ment is observed at θ=15° for 50 eV$<E_e<$500 eV. Indeed, the FBA-CW model underestimates the experimental DDCS since it does not include the ECC in the description of the ionization. In fact, the contribution of the ECC in the ionization process becomes important essentially at small angles (such as here 15°), which explains the best agreement observed between our FBA-CW model and the experimental measurements performed at 30° and 45°.

B. Singly differential cross sections

In Fig. 6(a), we present the SDCS calculations for the three incident energies investigated by Oshawa *et al.* The theoretical SDCS obtained in the FBA-CW model are represented by a solid line and compared to the recent experimental data obtained by Ohsawa *et al.* [14] represented by solid circles. Good agreement is observed between the experimental data and our theoretical results, which generally overestimate the experimental measurements for ejected electron energies lower than 100 eV. For higher energies, the discrepancies decrease to exhibit very good agreement with the experimental data. Finally, let us note that the results obtained by our theoretical model tend asymptotically to those obtained by the semiempirical Rudd's model [37] [dashed line in Fig. 6(a)], this latter providing a simple analytic representation of the differential cross sections over a wide range of primary and secondary energies. In Fig. 6(b), the experimental data of Toburen *et al.* [9] are compared to our FBA-CW model. A good agreement is still obtained with the two sets

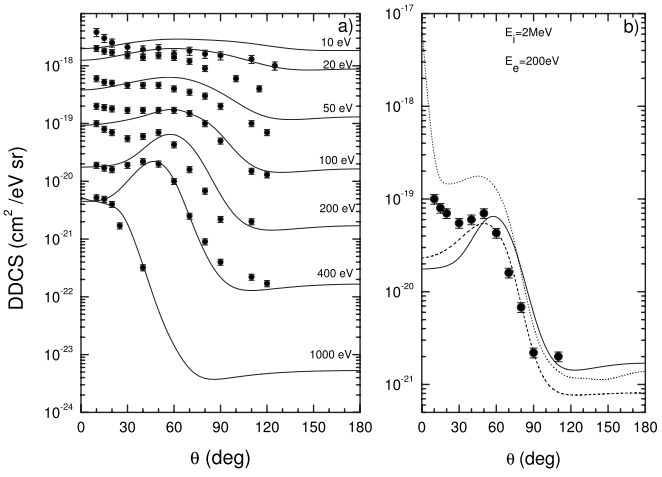

FIG. 4. (a) DDCS for water ionization by alpha particles of 2 MeV. The experimental data of Toburen *et al.* [9] are compared to the theoretical results (FBA-CW model) for 7 different ejected electron energies: E_e=10, 20, 50, 100, 200, 400, and 1000 eV. (b) DDCS for water ionization by alpha particles for E_i=2 MeV and E_e=200 eV. The experimental data are taken from Ref. [9] and compared to the theoretical results obtained in the FBA-CW model (solid line), 2CW model (dashed line), and BBK model (dotted line), respectively.

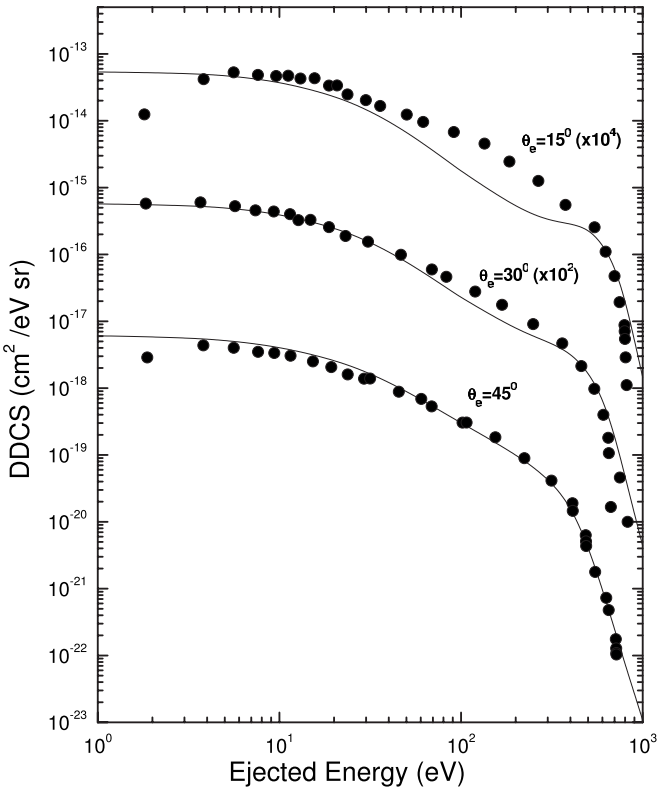

FIG. 5. DDCS for water ionization by alpha particles of 1.2 MeV reported as a function of the ejected electron energy for three fixed ejection angles (θ_e=15°, θ_e=30°, and θ_e=45°). The theoretical results (solid line) were performed in the FBA-CW model and the experimental data taken from Toburen [9]. Multiplicative factors have been used for clarity reasons.

of data (1.2 and 2 MeV) even for low ejected energies.

C. Total cross sections

Figure 7 presents a comparison between our theoretical total cross sections obtained in the FBA-CW model and experimental data available in the literature for water ionization by He^{2+} ions. Fairly good agreement is observed between our results and the experimental measurements for $_3He^{2+}$ (up triangles taken from Ref. [8]), and for $_4He^{2+}$ (circles taken from Ref. [9] and squares taken from Ref. [30]). Additionally, we have reported the analytical fit (dashed line) recently reported by Uehara and Nikjoo [7], which represents the extrapolations used in their Monte Carlo track-structure code, called LEAHIST, dedicated to low-energy alpha particle transport in water (incident energies E_i ranging from 1 keV/u to 1 MeV/u).

V. CONCLUSION

We have investigated in this work a full-differential theoretical approach to calculate doubly, singly differential, and total ionization cross sections for fast alpha particles impinging a water molecule in its vapor phase. For the first time, a detailed comparison is given between very recent experimental measurements provided by Ohsawa *et al.* and other al-

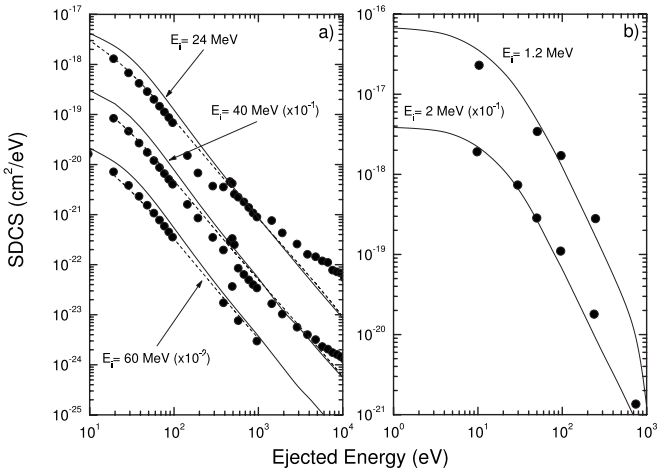

FIG. 6. SDCS for water ionization by alpha particles. Comparison between our theoretical FBA-CW model and experimental data taken from (a) Ohsawa *et al.* [14] (E_i=6, 10, and 15 MeV/u) and (b) Toburen *et al.* [9] (E_i=1.2 and 2 MeV). In both cases, multiplicative factors have been used for clarity reasons. The semiempirical results of the Rudd's model [37] have been also reported for comparison (dashed line).

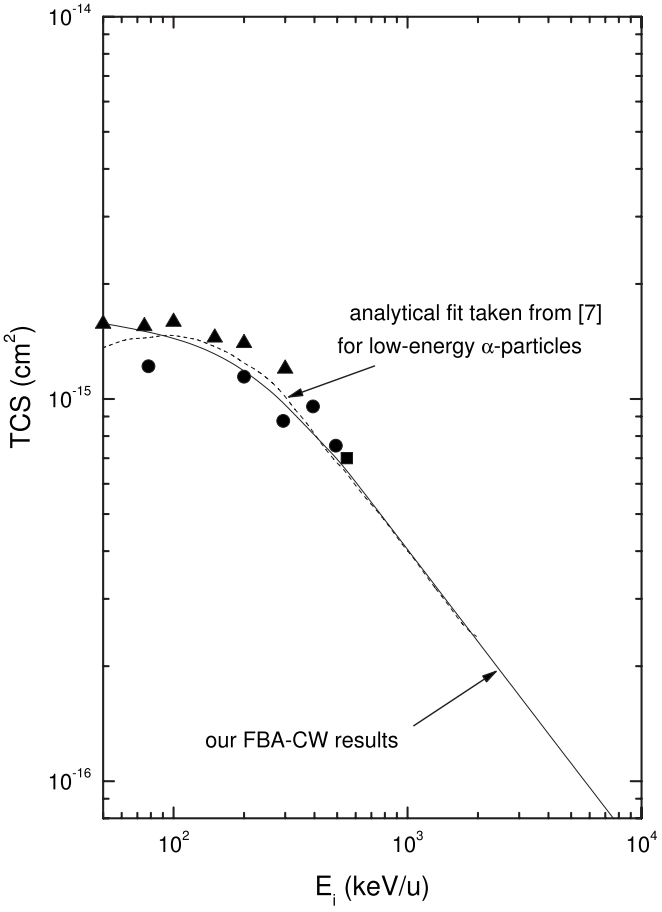

FIG. 7. TCS for water ionization by alpha particles. Comparison between the theoretical results performed in the FBA-CW model (solid line) and the experimental data taken from different sources (see text for details).

ready existing experiments (taken from Toburen *et al.*, from Rudd *et al.*, and from Rudolph and Melton) and theoretical results performed in the FBA-CW model, i.e., the FBA by describing the ejected electron by a Coulomb wave.

Contrary to the simple analytical Rudd's model, our approach requires no experimental data for adjustment and is only based on a quantum-mechanical description of the charged particle-water interaction. In this kind of study, the water target is described by means of five molecular wave functions constructed from a linear combination of atomic orbitals in a self-consistent field (MO-LCAO-SCF).

Fair agreements are observed for the differential (DDCS and SDCS) as well as the total ionization cross sections, for all the incident and ejected electron energies reported.

Finally, it is important to note that our theoretical approach may be easily introduced in numerical simulations such as Monte Carlo track-structure code for light ions in water or in matter in general. Indeed, for these codes, multiple differential calculations represent useful input data for describing in detail all the ionizing events, in terms of energy deposits and angular distributions.

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