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H_2O double ionization induced by electron impact

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Double ionization of water molecules remains, still today, rarely investigated on both the experimental and the theoretical side. In this context, the present work reports on a quantum mechanical approach providing a quantitative description of the electron-induced double ionization process on isolated water molecules for impact energies ranging from the target ionization threshold up to about 10 keV. The cross section calculations are here performed within the first Born approximation framework in which the initial state of the system includes a molecular ground-state wave function expressed as a single-center linear combination of atomic orbitals while the final state of the system is characterized by two independent Coulomb wave functions used for describing the two ejected electrons coupled by a Gamov factor used for modeling the electron-electron repulsion. Besides, in order to go beyond the first Born approximation, the scattered electron is considered as a particle being in the Coulomb field of the nucleus—whose charge is screened by the ejected electrons—and then treated by an approximate Coulomb wave function. In this *perturbative*-type description, let us add that the passive (not ionized) electrons are considered as frozen in their molecular orbitals during the collision which permits to reduce the electron-target interaction potential to a *two-active-electron* problem. Comparisons with rare available experimental data are reported as well an energetic analysis in terms of mean secondary energy transfer during the double ionization process in order to demonstrate the relevance of the electron-induced double ionization process.

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Electron-induced interactions in water are of great importance in many fields of research ranging from astrophysics to cellular biology with significant impacts in radiobiology, medical imaging, and radiotherapy, essentially due to the fact that water is commonly used as a surrogate of the living matter. In this context, it is nowadays well recognized that the "physics" stage that takes place at the first postirradiation femtoseconds plays a key role in the avalanche of events occurring throughout the water radiolysis and then appears as a decisive step in the induction of the radiation cellular damages [1]. Under these conditions, accurate cross sections—related to the different electron-induced interactions on water molecules—appear as crucial input data for the numerical codes devoted to the electron track-structure description in biological matter [2]. In this context, the single collisional processes induced by electron impact in water (including the elastic and inelastic channels) have been intensively investigated on both the theoretical [3,4] and the experimental [5,6] sides. Comparatively, the electron-impact double ionization (DI) of water molecules is less documented and therefore generally neglected in the majority of existing numerical track-structure codes arguing a presupposed minor contribution in the total energy deposit pattern. However, whether it is in the water radiolysis process for predicting the free radical production consecutive to the fragmentation of the ionized water molecules [7] or in the radiobiology framework for pointing out the key processes at the origin of the cellular death [8], a deeper understanding of the DI process induced by electron impact remains still required.

Contrary to the photo-double-ionization experiments, which have been extensively reported (see Refs. [9–11]), electron-double-ionization measurements remain up to now scarce and limited to the work of Jones *et al.* [12] where $(e,3-1e)$ momentum spectroscopy experiments have been reported for 2055-eV incident electrons (see Ref. [13] for more details). From a "macroscopic" point of view, namely, at the total (integrated) scale, the investigation of the double ionization of water molecules is also very poor since to the best of our knowledge we only find studies on electron-induced fragmentation of water molecules resulting from the formation of doubly charged ions. In this context, let us cite the work of Frémont *et al.* [14] where double vs single ionization ratios have been reported for impact energies ranging from 20 to 200 eV. More recently, Montenegro and co-workers published a series of works [15–17] where fragmentation branching ratios of multiply charged water molecules have been analyzed in order to propose a general scheme for the fission and autodissociation processes of doubly charged water molecules. On the theoretical front, the situation is more critical since with the works of Dal Capello and co-workers, who successively treated the DI process within the first and second Born approximation [18–20], our recent publications [21,22] appear as the only one existing theoretical support for the electron-induced DI process of water molecules with, in particular, a detailed analysis in terms of angular and energy distributions of the ejected electrons as well as total cross sections for single *oriented* water molecules [23].

The aim of the present work is to go beyond this series of *orientation-dependent* studies and then to report a theoretical description of the double ionization process for isolated water molecules in considering all the final water target configurations, namely, the two-electron removal from

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similar orbitals or not. Total cross sections as well as mean energy transfers are here calculated within the first Born approximation by considering a single-center description of the molecular ground-state wave function and by assuming that the passive (not ionized) electrons remain as frozen in their molecular orbitals during the collision and then reducing the electron-target interaction potential to a two-active electron problem. In addition, we assume that the remaining electrons in the doubly charged ion core are unaffected by the ionization process. Under these conditions, the initial state of the system is characterized by the product of an incident plane wave with a molecular target wave function while the final state is described by the product of a scattered plane wave with two independent Coulomb wave functions as well as an approximate expression of the Gamov factor for modeling the electron-electron repulsion [24,25]. Finally, in order to go beyond the domain of validity of the first Born approximation and then to extend the current water molecule double ionization modeling down to the intermediate energy regime, we followed the recommendations of Behati *et al.* who suggested to use an *approximate* Coulomb wave function of effective charge Z^* to represent the scattered electron in the field created by the water nucleus together with the two ejected electrons [26–28]. In their approach, the authors simply replaced the scattered electron Coulomb wave function by the first term of the whole hypergeometric series that may then be seen as a multiplicative factor, which characterizes the influence of the effective Coulomb field on the scattered electron being not far from the target nucleus. The effective charge Z^* is finally expressed thanks to the analytical approach suggested by Berakdar *et al.* [27,28]. However, it is worth noting that the current *perturbative* treatment of the projectile-target interaction brings significant simplifications and may be questionable even in the high-energy domain. Thus, and in view of future comparisons with experiments, higher-order theoretical treatments going beyond the simple perturbative regime should be preferred in particular for predicting the main mechanisms involved in the double ionization process (see Refs. [29,30] for more details). However, up to now, only very few second-order Born models have been developed for describing the electron-induced double ionization process for molecular targets, the only one case available in the literature corresponding to the H_2 molecule [31,32]. In this context, we have recently reported a detailed comparison between first- and second-order treatments of the double ionization of isolated water molecules and clearly pointed out particular kinematics where a second-order theory was needed [20]. However, there is no denying that such calculations are very computer-time consuming and they remain still nowadays limited to the calculations of multiple differential cross sections, in particular for molecular targets. In this sense, the current study must be seen as an attempt of theoretical predictions for the total double ionization cross sections of water molecules and will have to be confirmed by higher-order calculations as well as future experiments.

Under these conditions, the first Born *orientation-dependent* fivefold differential cross sections, namely, differential in the direction of the scattered electron Ω_s, differential in the directions of the two ejected electrons Ω_1 and Ω_2, and differential in the ejected energy transfers E_1 and E_2, may be simply expressed as

$$\begin{aligned}\frac{d^5\sigma}{d\Omega_1 d\Omega_2 d\Omega_s dE_1 dE_2}&\\ \equiv \sigma^{(5)}&(\Omega_1,\Omega_2,\Omega_s,E_1,E_2)\\ = \sum_{j_1=1}^{N_{orb}}&\sum_{j_2\geqslant j_1}^{N_{orb}} \sigma^{(5)}_{j_1 j_2}(\Omega_1,\Omega_2,\Omega_s,E_1,E_2)\\ = (2\pi)^4&\frac{k_1k_2k_s}{k_i} g_G(k_1,k_2)\sum_{j_1=1}^{N_{orb}}\sum_{j_2\geqslant j_1}^{N_{orb}} |T_{j_1 j_2}|^2, \end{aligned}\qquad (1)$$

where $g_G(k_1,k_2)$ refers to the approximate expression for the repulsive Gamov factor given by Defrance and co-workers [24,25]:

$$g_G(k_1,k_2) = \begin{cases} \frac{2\pi}{k_1}\frac{e^{-2\pi/k_1}}{1-e^{-2\pi/k_1}} & \text{when} \quad k_1 > k_2 \\ 0 & \text{when} \quad k_1 = k_2\ , \\ \frac{2\pi}{k_2}\frac{e^{-2\pi/k_2}}{1-e^{-2\pi/k_2}} & \text{when} \quad k_2 > k_1 \end{cases} \qquad (2)$$

while the transition matrix element $T_{j_1 j_2}$ refers to the simultaneous ejection of two electrons from two molecular orbitals labeled j_1 and j_2, respectively. Let us add that $k_i = \sqrt{2E_i}$, $k_1 = \sqrt{2E_1}, k_2 = \sqrt{2E_2}$, and $k_s = \sqrt{2(E_i - E_1 - E_2 - I^{2+})}$, where E_i denotes the incident electron energy while I^{2+} refers to the double ionization threshold (see Table 1 in Ref. [23]). Let us remark here that similarly to our previous works, we use here the single-center *orientation-dependent* description of the water molecule provided by Moccia [33] where the ten bound electrons of the water target molecule are distributed among $N_{orb}(=5)$ molecular wave functions corresponding to the five molecular orbitals denoted $1b_1$, $3a_1$, $1b_2$, $2a_1$, and $1a_1$.

Then, benefiting from the selectivity rules of the complex harmonics used in the description of the molecular wave function, we easily access to triply differential cross sections, which are then numerically integrated over the solid angle Ω_s as well as the ejected energies E_1 and E_2 to provide *orientation-dependent* total cross sections defined as [26]

$$\sigma = \int_0^{E_{1max}}\int_0^{E_{2max}}\int |f(k_s)|^2 \frac{d^3\sigma}{d\Omega_s\, dE_1\, dE_2} d\Omega_s\, dE_1\, dE_2, \qquad (3)$$

where

$$|f(k_s)|^2 = \frac{2\pi Z^*}{k_s}\frac{1}{(1-e^{-2\pi Z^*/k_s})}$$

with

$$Z^* = \begin{cases} 0 & \text{when} \quad k_s \geqslant k_1,k_2 \\ 1 - k_s/k_1 & \text{when} \quad k_2 < k_s < k_1 \\ 1 - k_s/k_2 & \text{when} \quad k_1 < k_s < k_2 \\ 2 - k_s/k_1 - k_s/k_2 & \text{when} \quad k_s < k_1,k_2 \end{cases}.$$

In Eq. (3), let us note that the upper limits of the integration $E_{1\,\max}$ and $E_{2\,\max}$ are defined by $E_{1\,\max} = (E_i - I^{2+})$ and $E_{2\,\max} = E_{1\,\max} - E_1$ with $E_i = I^{2+} + E_s + E_1 + E_2$ [26]. The integration over the target orientations is finally carried out analytically thanks to the rotation matrix properties.

Double ionized water molecules exhibit a high instability leading to a dissociation pattern, which results from several possible combinations of two-electron removal. Thus, we report in Fig. 1, the *absolute* contribution to the total DI cross

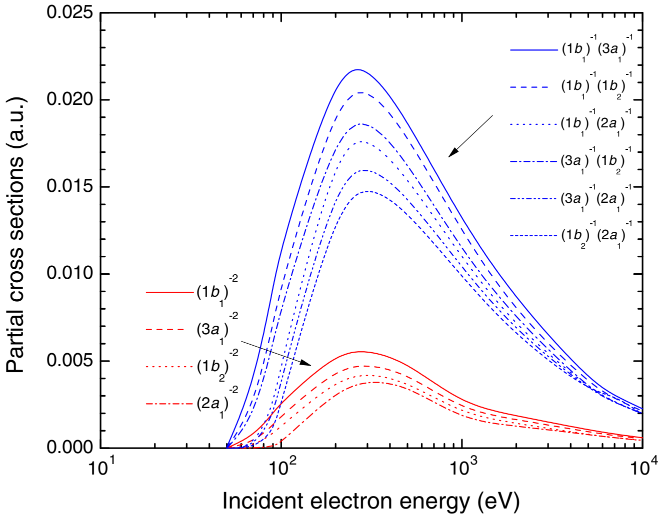

FIG. 1. (Color online) Partial cross sections of the double ionization of water molecules.

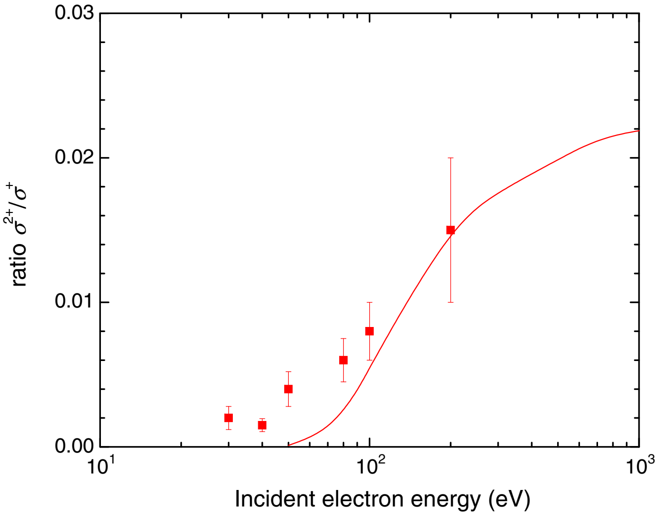

FIG. 2. (Color online) Cross section ratio relative to H_2O^+. The current theoretical predictions (red line) are compared with the experimental data taken from Frémont *et al.* [14].

section for all final channels, namely, those corresponding to the ejection of two electrons originating from the same orbital, i.e., $(1b_1)^{-2}$, $(3a_1)^{-2}$, $(1b_2)^{-2}$, and $(2a_1)^{-2}$ as well as those involving two electrons ejected from two different orbitals, i.e., $(1b_1)^{-1}(3a_1)^{-1}$, $(1b_1)^{-1}(1b_2)^{-1}$, $(1b_1)^{-1}(2a_1)^{-1}$, $(3a_1)^{-1}(1b_2)^{-1}$, $(3a_1)^{-1}(2a_1)^{-1}$, and $(1b_2)^{-1}(2a_1)^{-1}$. Let us note that the double ionization involving the inner-shell $1a_1$ is not here considered since it is negligible.

One important point to emerge from Fig. 1 is the behavior of the DI cross sections with respect to the incident energy. They all exhibit very similar energy dependence with, in particular, a peak located around 300 eV whatever the final channel considered. Besides, it is shown that the simultaneous electron ejection from two different orbitals (blue lines in Fig. 1) appears as the dominant process with an amplitude, which is 3 times higher at least than that corresponding to the two-electron removal from the same molecular orbital (red lines in Fig. 1).

As stated above, no direct comparison with experiment is nowadays possible. In fact, the only available experimental measurements of double ionization cross sections are those reported by Frémont *et al.* [14] who investigated the $e^- + H_2O$ collision at projectile energies ranging from 20 to 200 eV. Thus, via the analysis of the energy of the fragments emitted after the ionization of H_2O at a detection angle of 90° with respect to the incident beam, the authors determined ratios between multiple ($q = 2$ and $q = 3$) and single ($q = 1$) ionization. That said, we show in Fig. 2 a comparison between the experimental double vs single ionization cross-section ratio reported by the authors (denoted σ^{2+}/σ^+) and the theoretical value, the latter being deduced from the current double ionization cross section and the single ionization cross section taken from our previous study [4] where the single process of the water molecule was treated within the same *perturbative* approach. A good agreement may be observed with the experiment provided that the incident energy is greater than about 100 eV which corresponds to the energy range of validity of the current perturbative description of the double collisional process. Besides, we note that for increasing impact energies, the ratio tends to a constant value of about 2.2% as already observed by many authors for electron-induced single and double ionization on neon targets [34,35]. Let us note that similar observations were reported by Cavalcanti *et al.* for protons in neon [36] as well as by Scully *et al.* [15] in water who compared electron and equivelocity protons and concluded that the σ^{2+}/σ^+ ratio was approximately independent of the impinging energy provided that $E_i > 200$ eV.

The *absolute* total double ionization cross section of water is displayed in Fig. 3 (left panel). In light of the absence of direct experimental validation, the current pioneer study has to be seen as prospective and one more time, we hope that its

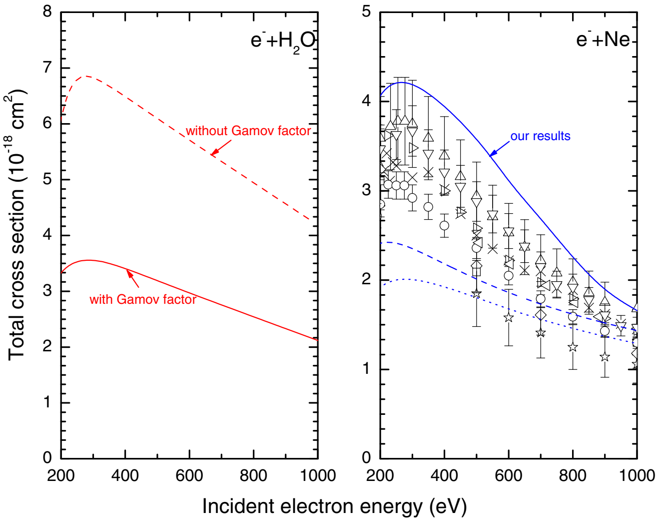

FIG. 3. (Color online) Left panel: TCS of the double ionization of the water molecule. Right panel: TCS of the double ionization of the neon target (solid line) compared with experimental data taken from diverse sources (open circles [35], open upward triangles [41], open downward triangles [42], open diamonds [43], open left triangles [34], open right triangles [44], open stars [45], open squares [46], crosses [47]). The semiempirical predictions discussed in the text are also reported for comparison (dotted line [37] and dashed line [40]).

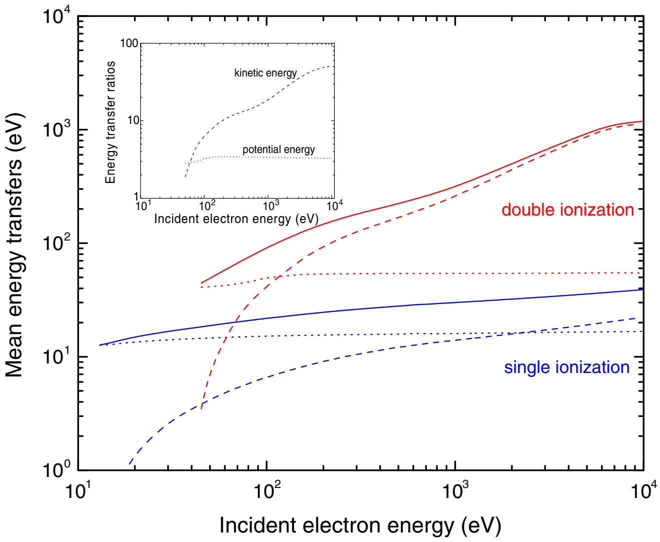

FIG. 4. (Color online) Mean energy transfers induced by electron impact during single and double ionization of water (blue and red lines, respectively). The kinetic energy (dashed lines), the potential energy (dotted lines), and the sum of both (solid lines) are reported for impact energies ranging from 10 eV to 10 keV. The double vs single ionization ratio values are reported in the inset.

predictions will be experimentally confirmed in the near future. However, to check the ability of our model to reproduce the total double ionization cross section behavior, we have also investigated the case of the isoelectronic neon target. Thus, in comparison with the existing experimental values (see right panel of Fig. 3), we observe that our theoretical predictions show an overall good agreement both in shape and magnitude. More precisely, we note that the experimental maximal cross section is of the order of $\sigma^{2+} = (3.7 \pm 0.6) \times 10^{-18}\,\mathrm{cm}^2$ for neon, i.e., in very good agreement with our theoretical value $\sigma^{2+} = 3.55 \times 10^{-18}\,\mathrm{cm}^2$ for water, both located around $E_i^{\max} = 280\,\mathrm{eV}$. For $E_i > E_i^{\max}$, i.e., in the domain of validity of the current *perturbative* model, we observe a fairly good agreement between our theoretical predictions and the existing data. Additionally, the semiempirical predictions provided by two existing phenomenological models of electron impact direct double ionization cross sections are reported for comparison. The first one was introduced by Bélenger *et al.* [37] and is based on the semiempirical Bethe-Born type formula previously reported by Shevelko and Tawara [38] for the multiple ionization cross section for ejection of $n\,(\geqslant 3)$ electrons and a later one adapted for the double ionization process [39]. The second semiempirical formula was more recently reported by Talukder *et al.* [40]. Thus, in comparison with the experiment (see right panel of Fig. 3), we observe that contrary to our model that lies in the experimental error bars, the two semiempirical models largely underestimate the experimental observations with a disagreement reaching a factor of 2 at $E_i^{\max}$.

Finally, we show in Fig. 4 the mean energy transfers during the DI process as a function of the incident electron energy. The potential energy (dotted line)—calculated as the weighted sum of the binding energies of the different channels involved in the DI—is almost constant ($\cong$55 eV) over the whole energy range, as already reported in one of our previous works dedicated to the single ionization description [4]. In this case, an almost constant energy deposit of about 16 eV was found. Consequently, the corresponding double vs single potential energy ratio exhibits a constant value of $\cong$3.3 (see the inset in Fig. 4), which clearly demonstrates the key role played by the double ionization process in the total energy deposit pattern, even considering that the occurrence of such a process remains lower than 2% as seen above. Considering now the mean kinetic energy released—defined as the weighted sum of the mean kinetic energies of the two ejected electron energies (both deduced from the doubly differential cross sections $d^2\sigma \big/ dE_1 dE_2$)—we observe a monotonic increase with respect to the incident energy. Thus, at low impact energies, the DI process induces very small kinetic energy transfers (<10 eV) of the same order of magnitude as those observed for single ionization [4]. Then, the mean kinetic energy released during the double ionization rapidly increases to reach the keV domain for $E_i = 10\,\mathrm{keV}$. In the absence of experimental data, we have compared the obtained values with those reported by Champion in a previous theoretical study dedicated to the multiple ionization of water molecules induced by heavy charged particles impact [48]. Thus, for double ionization induced by equivelocity ions in water, we obtain a mean kinetic energy transfer of about 150 eV for 1 MeV/amu ions (vs 167 eV for 55-eV electron), of about 500 eV for 10 MeV/amu ions (vs 883 eV for 5-keV electron), and finally of about 900 eV for 20 MeV/amu ions (vs 1100 eV for 10-keV electron). These high values clearly indicate the necessity of taking into account the double ionization process in the charged particle track modeling, in particular in the high-energy regime where the kinetic energy spectra is at least 50 times higher than that observed for single ionization (see inset of Fig. 4).

We have presented here an attempt for theoretically predicting the significance of the double ionizing process of water molecules for which data are still lacking both experimentally and theoretically. Relative probabilities as well as energetic considerations have been analyzed in order to assess the role played by the double ionizing process in water radiolysis for which a real need is still needed for insight into its relevance in many fields such as radiobiology and nuclear medicine. In this context, we have demonstrated that even if the double ionization represents only a small part of the single ionization process (≤2%), it is not less important notably due to its energetic signature whose consequences have been already pointed out by many authors for predicting the radio-induced DNA damage induction. In this context, we also want to stress that our theoretical model will be extended to more complex biomolecules, for example, DNA components such as the nucleobases or the sugar phosphate backbone, which will be addressed in future studies.

We have also pointed out that no direct comparison with experiment was possible at this stage in terms of total as well as partial cross sections and we hope that the current theoretical work will be confirmed by experimental observations.

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