

Direct two-photon double ionization of H₂A. S. Simonsen,^{*} S. A. Sørngård, R. Nepstad, and M. Førre[†]*Department of Physics and Technology, University of Bergen, N-5007 Bergen, Norway*

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We have studied the process of direct (nonsequential) two-photon double ionization of molecular hydrogen (H₂). Solving the time-dependent Schrödinger equation by an *ab initio* method, total (generalized) and single-differential cross sections are obtained at photon energies from 26 to 33 eV. Both parallel and perpendicular orientation of the molecule with respect to the laser polarization direction are considered, and the results are compared with previously calculated cross sections at 30 eV, as well as the predictions of a simple model.

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I. INTRODUCTION

The problem of direct, as opposed to sequential, two-photon double ionization of xenon [1] and helium [2,3] was introduced some time ago. Since then, the direct (nonsequential) process in helium has been the focus of great interest, both theoretically [4–21] and experimentally [22–27]. These investigations were partly triggered by the development of high-order harmonic [28,29] and free-electron laser (FEL) [30,31] light sources, as well as the development of sophisticated numerical methods, capable of tackling correlated motions in few [4,32] and multiphoton [33] ionization processes. The study of fundamental breakup processes in nature is important and paves the way for further investigations of the role of correlations in few and multiphoton multiple ionization processes in atoms and molecules. More recently, the four-body breakup of H₂ by two-photon impact has received considerable attention, both in the direct [34–37] and sequential regime [38,39].

Employing the time-dependent close coupling method, Colgan *et al.* [34] studied the direct two-photon double ionization of H₂ by 30 eV photons. Total and triple-differential cross sections for the process were obtained for both parallel and perpendicular orientation of the molecule with respect to the linear laser polarization direction. In 2009, Morales *et al.* [35] revisited the problem employing a time-independent approach and the method of exterior complex scaling. Clear discrepancies in the respective triple-differential cross sections were found. Even more recently, Guan *et al.* [36,37] calculated the corresponding cross sections using a fully *ab initio*, nonperturbative approach, solving the time-dependent Schrödinger equation in prolate spheroidal coordinates for a 10 cycle laser pulse of peak intensity 10¹⁴ W/cm². The cross sections were obtained projecting the final continuum wave function onto a set of uncorrelated two-center Coulomb waves, and the results were found to differ from those reported by both Colgan *et al.* [34] and Morales *et al.* [35].

In the present work we again consider the problem of direct two-photon double ionization (TPDI) of H₂. For this purpose, we solve the time-dependent Schrödinger equation in spherical coordinates employing a recently developed *ab initio* numerical framework [17,40]. The framework has here been further developed in order to take into account the two-center nature of the problem. Following [34–37] we

assume the fixed-nuclei approximation in the calculations, that is, the nuclei are considered being fixed at their equilibrium internuclear distance at $R = 1.4$ a.u. throughout the interaction with the laser pulse. Since the electrons are ejected almost instantaneously in the direct TPDI process and move apart much faster than the Coulomb exploding nuclei, the approximation is expected to be very accurate, concordant with earlier findings in the corresponding one-photon double ionization process in H₂ [41,42].

Using a 15 cycle laser pulse of sine-squared shape, total (generalized) and single-differential cross sections are obtained for photon energies in the interval 26 to 33 eV. The TPDI cross sections are calculated by subtracting the bound and single continuum states from the total wave packet. In order to obtain sufficiently converged results and to minimize the effect of the Coulombic repulsion between the electrons, the wave function is propagated some additional optical cycles after the pulse before the projections are performed. The results of the full calculations are compared with the previous ones [34–37], and differences and similarities are noted. Furthermore, they are compared with the predictions of a simple approximate model, recently proposed for the corresponding TPDI process in helium [12,18].

Atomic units, where m_e , \hbar , and e are scaled to unity, are used throughout unless stated otherwise.

II. THEORY AND NUMERICAL APPROACH

A. Numerical model

The Hamiltonian for fixed-in-space H₂ interacting with a laser field consists of two parts,

$$\hat{H} = \hat{H}_{\text{mol}} + \hat{H}_{\text{field}}, \quad (1)$$

where the first term is the field-free Hamiltonian for the hydrogen molecule,

$$\hat{H}_{\text{mol}} = \sum_{i=1}^2 \left(\frac{\mathbf{p}_i^2}{2} - \frac{1}{|\mathbf{r}_i + \mathbf{R}/2|} - \frac{1}{|\mathbf{r}_i - \mathbf{R}/2|} \right) + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}, \quad (2)$$

\mathbf{R} being the internuclear vector. The second term is the interaction with the laser field, which in a semiclassical approximation takes the form

$$\hat{H}_{\text{field}} = \mathbf{A}(t) \cdot (\mathbf{p}_1 + \mathbf{p}_2), \quad (3)$$

when the velocity gauge and the dipole approximation have been assumed. The laser field is modeled by the classical

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electric field $\mathbf{E} = -\partial_t \mathbf{A}$, with the time-dependent vector potential given as

$$\mathbf{A}(t) = A_0 \sin^2\left(\frac{\pi t}{T}\right) \cos(\omega t) \hat{\mathbf{u}}. \quad (4)$$

Here the unit vector $\hat{\mathbf{u}}$ defines the polarization of the linearly polarized field, $A_0 = \frac{E_0}{\omega}$, E_0 is the peak electric field amplitude, ω is the central frequency, and T is the pulse duration.

The standard procedure for solving a partial differential equation numerically is to expand the solution in a convenient basis, and subsequently solve the resulting system of ordinary differential equations. In our approach, the angular rank of the two-electron wave function is expanded in coupled spherical harmonics, and the radial ranks in B splines [43,44],

$$\Psi(r_1, r_2, \Omega_1, \Omega_2, t) = \sum_{i,j,k} c_{i,j,k}(t) \frac{B_i(r_1)}{r_1} \frac{B_j(r_2)}{r_2} \mathcal{Y}_{i_1, i_2}^{L, M}(\Omega_1, \Omega_2). \quad (5)$$

Here $k = \{l_1, l_2, L, M\}$ is a combined index for the angular indices. In this basis the radial and angular kinetic energy operators have a very sparse structure with few nonzero elements.

The H_2 molecule is a two-center system, and is in many respects best represented in prolate spheroidal coordinates, in which the H_2^+ molecule is separable. Nevertheless, it is still possible to utilize the single-center approach at the cost of being able to fully exploit the symmetry properties of the two-center system. In our case, the electron-nucleus interaction operator is expanded in spherical harmonics using the multipole expansion,

$$\begin{aligned} & \frac{1}{|\mathbf{r}_i + \mathbf{R}/2|} - \frac{1}{|\mathbf{r}_i - \mathbf{R}/2|} \\ &= - \sum_{j \in 2\mathbb{Z}^*} \sum_{m=-j}^j \frac{4\pi}{2j+1} \frac{r_{<}^j}{r_{>}^{j+1}} Y_{j,m}^*(\theta, \phi) Y_{j,m}(0, 0), \end{aligned} \quad (6)$$

where $r_{>}$ and $r_{<}$ are the largest and smallest of r_i and $R/2$, respectively, the internuclear vector is assumed to lie along the z axis, and the index j runs over even integers. The single-center approach in H_2 deviates in general very little from the formalism commonly used for two-electron atomic systems like helium. The only difference, as evident from Eq. (6), is the need for terms beyond the monopole term to support the two-center nature of the system, making the calculations more demanding regarding memory requirements and run time.

As opposed to helium, the total angular momentum L is not a conserved quantity in the hydrogen molecule. While the helium ground state comprises only the $L = 0$ symmetry, the nonspherical symmetric electron-nucleus interaction entails the H_2 ground state to be composed of several even L components. In addition to $L = 0$ the most prominent components are those of $L = 2, 4$, and 6 . When exposing the molecule to a linearly polarized laser field, it should be noted that the projection of the total angular momentum $M = m_1 + m_2$ onto the z axis is conserved and equal to zero as long as the axis of polarization is parallel to the internuclear vector (z axis). Whenever the polarization axis points elsewhere, the cylindrical symmetry of the system is broken and hence the M quantum number is not conserved.

Our numerical scheme for solving the two-electron time-dependent Schrödinger equation (TDSE) in the basis (5) was presented earlier [17,40]. The framework has already been used for single and double ionization studies in helium [17,18,45,46] and the negative hydrogen ion [47].

B. Extracting physical information

Extracting physical quantities for a two-photon double ionization process is a nontrivial task, because of the complications that arise in separating the single and double continuum. In the present work, an approximation to the double continuum component is obtained by applying complementary projection operators to the final wave function. The complementary projection operators are constructed to remove the population from bound and single continuum channels. First the bound component is removed. Then the single continuum is removed by subtracting all components from the wave function corresponding to one electron remaining in a bound H_2^+ state, as follows:

$$\begin{aligned} |\Psi_{\text{DC}}\rangle &= |\Psi_f(\mathbf{r}_1, \mathbf{r}_2, t)\rangle - \sum_m \langle \phi_m^{\text{H}_2^+}(\mathbf{r}_1) | \Psi_f(\mathbf{r}_1, \mathbf{r}_2, t) \rangle | \phi_m^{\text{H}_2^+}(\mathbf{r}_1) \rangle \\ &\quad - \sum_{m'} \langle \phi_{m'}^{\text{H}_2^+}(\mathbf{r}_2) | \Psi_f(\mathbf{r}_1, \mathbf{r}_2, t) \rangle | \phi_{m'}^{\text{H}_2^+}(\mathbf{r}_2) \rangle \\ &\quad + \sum_{m, m'} \langle \phi_m^{\text{H}_2^+}(\mathbf{r}_1) | \langle \phi_{m'}^{\text{H}_2^+}(\mathbf{r}_2) | \Psi_f(\mathbf{r}_1, \mathbf{r}_2, t) \rangle \\ &\quad \times | \phi_m^{\text{H}_2^+}(\mathbf{r}_1) \rangle | \phi_{m'}^{\text{H}_2^+}(\mathbf{r}_2) \rangle. \end{aligned} \quad (7)$$

Here m and m' sum over all H_2^+ bound states, which are obtained by numerical diagonalization of the H_2^+ single-particle Hamiltonian. Furthermore, Ψ_{DC} represents the double continuum wave packet, and $\Psi_f(\mathbf{r}_1, \mathbf{r}_2, t)$ is the total (single + double) continuum wave function at some time t after the pulse. It should be noted that in this approximation to the double continuum, the electron-electron interaction is completely disregarded. Thus, for the method to be valid, it is important that the electrons are as far apart as possible. This is achieved by letting the wave packet propagate for some additional time after the end of the laser pulse, typically 5–10 optical cycles, in order for the ionized wave packet to reach near-asymptotic distances before the projections are performed. The disadvantage of this approach is of course the necessity of using correspondingly larger radial boxes.

After the double continuum wave packet Ψ_{DC} is found using Eq. (7), the generalized cross section is easily obtained,

$$\sigma = \left(\frac{\omega}{I_0}\right)^2 \frac{P_{\text{ion}}}{T_{\text{eff}}}, \quad (8)$$

where

$$P_{\text{ion}} = \langle \Psi_{\text{DC}} | \Psi_{\text{DC}} \rangle \quad (9)$$

is the double ionization probability. Here ω is the photon energy, I_0 is the laser intensity, and T_{eff} is the effective pulse duration. The latter quantity depends on the pulse shape, and for a sine squared envelope it is given by $T_{\text{eff}} = \frac{35}{128} T$ [14].

The electron energy distributions are derived from the double continuum wave packet by projecting it onto pairs of H_2^+ field-free continuum energy eigenstates—one for each

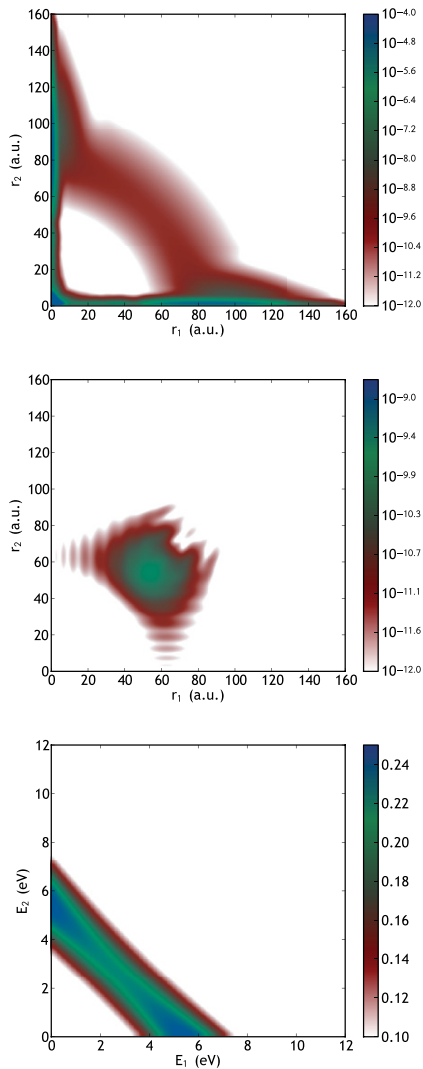


FIG. 1. (Color online) Upper panel: Radial wave function density (in a.u.) obtained at seven optical cycles after the end of the pulse, for a 15-cycle laser pulse of central frequency $\omega = 1.05$ a.u. (corresponding to the photon energy 28.6 eV), and intensity 10^{13} W/cm². The molecule is oriented parallel to the polarization axis of the laser field. The scale is logarithmic. Intermediate panel: Radial wave function density (in a.u.) of the double continuum wave packet, after the bound and single ionized populations have been removed. Lower panel: Corresponding energy distribution of the outgoing electrons in units of 10^{-5} a.u.

electron, as obtained by numerical diagonalization of the full one-electron H₂⁺ Hamiltonian imposing zero boundary condition at the edge of the radial box. Although these energy eigenstates do not fulfill the incoming-wave boundary condition [48], which would be a prerequisite for calculating angular-differential cross sections, they do produce accurate energy-differential quantities, provided the electron-electron interaction can be neglected.

III. RESULTS AND DISCUSSION

In our calculations, and in the case of parallel alignment of the molecule, we have used a radial box extending to $r_{\max} = 160$ a.u., covered by 147 B splines. The B splines are distributed on a grid such that the density of splines is exponentially decreasing away from the center of mass. After

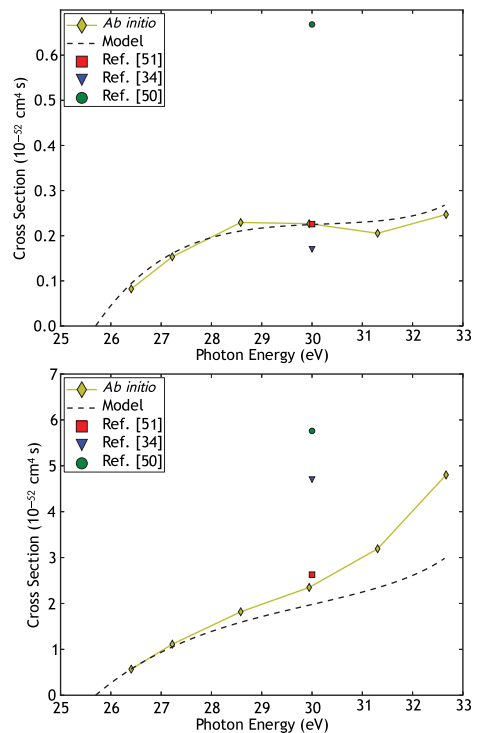


FIG. 2. (Color online) Total (generalized) cross section for the process of direct (nonsequential) two-photon double ionization of H₂. Upper panel: molecule oriented parallel with the laser polarization axis. Lower panel: molecule oriented perpendicular to the laser polarization axis. Golden line with diamonds: present *ab initio* result. Dashed line: model result Eq. (10). Blue triangle: theoretical result by Colgan *et al.* [34]. Green circle: theoretical result by Morales *et al.* [35,50]. Red square: theoretical result by Guan *et al.* [37,51].

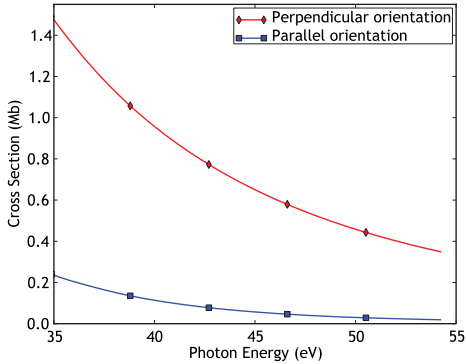


FIG. 3. (Color online) Photoionization cross section obtained for fixed-in-space H_2^+ molecules at $R = 1.4$ a.u. Blue line with squares: molecule oriented parallel to the laser polarization axis. Red line with diamonds: molecule oriented perpendicular to the laser polarization axis.

some distance (8 a.u. in this work), the density of splines is held constant. The higher density close to the origin enables resolving the ground state to a satisfactory level. Furthermore, the angular basis is truncated at $l_{\max} = 6$, $L_{\max} = 12$, and $M = 0$. With this basis we obtain the ground state energy -1.8852 a.u., which is in satisfactory agreement with the calculated benchmark value -1.8888 a.u. [49]. Due to memory limitations, a somewhat smaller basis has been applied for the perpendicular geometry. In that case, $l_{\max} = 4$, $L_{\max} = 8$, $M = -2, -1, 0, 1, 2$, and 101 B splines are distributed in a radial box extending to $r_{\max} = 150$ a.u. Varying the size of the basis sets, it is found that the cross sections are fairly well converged, both for the parallel and perpendicular geometries.

The upper panel in Fig. 1 displays the radial wave function density seven optical cycles after the interaction with a 15-cycle laser pulse. The photon energy is 28.6 eV. The corresponding double continuum wave packet, as obtained by the subtraction procedure described in Sec. II B, is shown in the intermediate panel. From this wave packet, the electrons' energy distribution is derived and depicted in the lower panel.

Figure 2 depicts our results for the total cross section, both for parallel (upper panel) and perpendicular (lower panel) orientation of the molecule with respect to the laser polarization axis. The results are obtained for a 15-cycle laser pulse of intensity 10^{13} W/cm². The wave packet is propagated for about seven optical cycles after the action of the pulse before the projections are performed. It should be noted that the calculated total cross sections vary by less than 3% from the time just after the pulse up to this point.

Also shown in Fig. 2 is the result of Colgan *et al.* [34] (blue triangle), Morales *et al.* [35,50] (green circle), and Guan *et al.* [37,51] (red square) at the photon energy 30 eV. The agreement between our calculated cross section at 30 eV and the value in [37,51] turns out to be excellent, whereas clear discrepancies with the results of the two other studies are found. It is not clear to us what is the origin of these

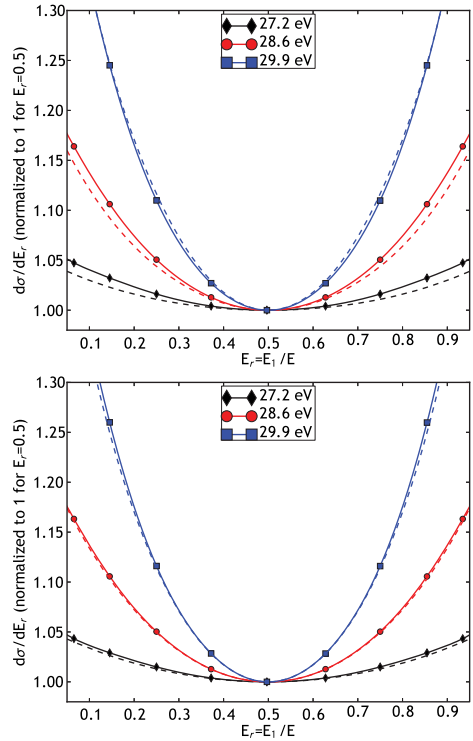


FIG. 4. (Color online) Single differential cross section (SDCS) for direct two-photon double ionization of H_2 at the photon energies 27.2, 28.6, and 29.9 eV. Full lines are the *ab initio* results and dashed lines indicate the prediction of the model Eq. (10). Upper panel: molecule oriented parallel with the laser polarization axis. These SDCSs are scaled, in order to align them for equal energy sharing, with the factors, 6.88, 4.82, and 5.31 for the photon energies 27.2, 28.6, and 29.9 eV, respectively. Lower panel: molecule oriented perpendicular to the laser polarization axis. Similarly, the SDCSs are scaled with the factors 0.95, 0.61, and 0.52 for the three photon energies 27.2, 28.6, and 29.9 eV.

discrepancies, but we nevertheless point out two possible explanations. First, Colgan *et al.* [34] used a 12-cycle (flat-top) laser pulse of peak intensity 10^{15} W/cm², which supports three and higher-order photon processes to a greater extent compared to our pulse, and that could possibly lead to a nonnegligible depletion of the population in the initial state. Second, as already pointed out by Colgan and co-authors [34], the doubly excited states of H_2 , the first one lying only about 30 eV above the H_2 ground state at $R = 1.4$ a.u. [48], may have some influence on the results. This could possibly explain the difference between the results of the time-dependent and time-independent approaches, respectively.

A simple approximate model for the single-differential cross section for direct two-photon double ionization of helium was recently proposed by Horner *et al.* [12] and Førre *et al.* [18]. As a natural extension of these works, we propose the following approximate formula for the single-differential cross section in the process of two-photon double ionization of H₂,

$$\frac{d\sigma}{dE_1} \simeq \frac{1}{4} [\sqrt{f(E_1)} + \sqrt{f(2\hbar\omega - E_b - E_1)}]^2, \quad (10)$$

$$f(E_1) \equiv \frac{\hbar^3 \omega^2}{\pi} \frac{\sigma_{\text{H}_2}(E_1 + E_{\text{H}_2}) \sigma_{\text{H}_2}^+(2\hbar\omega - E_1 - E_{\text{H}_2})}{(E_1 + E_{\text{H}_2})(2\hbar\omega - E_1 - E_{\text{H}_2})(E_1 + E_{\text{H}_2} - \hbar\omega)^2}.$$

Here $E_b = 51.4$ eV is the total binding energy of H₂, the energy $E_{\text{H}_2} = 16.5$ eV corresponds to the first (single) ionization threshold of H₂, and σ_{H_2} and $\sigma_{\text{H}_2}^+$ indicate the photoionization cross section for one-photon single ionization of H₂ and H₂⁺, respectively. For further details about the models, see Refs. [18,52]. The relevant photoionization cross sections for the single ionization of H₂ was calculated by Sánchez and Martín [53]. Furthermore, the photoionization cross section of H₂⁺ at $R = 1.4$ a.u. is given in Fig. 3 for the parallel and perpendicular orientation of the molecule, respectively.

The total cross section, as obtained by applying the approximate formula Eq. (10), is depicted in Fig. 2 by dashed lines. The model seems to yield results that are in reasonable agreement with our *ab initio* findings, which is somewhat surprising given the high complexity of the problem. Quite interestingly, and in agreement with our *ab initio* calculations, the model predicts a cross section about an order of magnitude larger for the perpendicular orientation as compared to the parallel one. In the model, this difference is attributed to a corresponding difference in the photoionization cross section of H₂⁺ (cf. Fig. 3).

We now turn to the energy distributions of the ejected electrons. Figure 4 shows the energy resolved single-differential cross sections (SDCSs) at three selected photon energies, 27.2, 28.6, and 29.9 eV. The SDCSs are obtained from the respective energy distributions of the two electrons (cf. lower panel in

Fig. 1). Upper and lower panels in Fig. 4 depict the results for the parallel and perpendicular geometry, respectively. Again, the agreement between the fully *ab initio* result and the model prediction is at a quantitative level, demonstrating the strength of the simple formula. At this point it should, however, be noted that the model does not apply to angular-resolved differential cross sections. As a matter of fact, the Coulombic repulsion between the electrons turns out to play a decisive role for the movement of the electrons in the continuum, even a long time after they have been ejected. Such long-range effects are not included in the model, and it is therefore not expected to be valid for the calculation of angular distributions.

IV. CONCLUSIONS

In conclusion, we have studied the two-photon double ionization of fixed-in-space hydrogen molecules (H₂), applying a *B*-spline based numerical method. Total (generalized) and single-differential cross sections are calculated at various photon energies and compared, when possible, with previously published results [34,35,37,50,51]. Our results are in agreement with the results of Guan *et al.* [37,51], as far as the total cross section is concerned, but further theoretical and experimental investigations are required in order to settle the problem definitely. We furthermore find that our results are in quantitative agreement with the predictions of a simple model, both for the total and single-differential cross sections. These observations are concordant with previous findings in the corresponding process in helium [18,20].

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