PHYSICAL REVIEW A, **66**, 032709 (2002)

Single ionization of helium by antiprotons: A case study by self-interaction-free time-dependent density-functional theory

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(Received 26 June 2001; revised manuscript received 17 May 2002; published 19 September 2002)

Single ionization of helium by antiprotons in a wide energy range (5–1000 keV) has been studied by a self-interaction-free time-dependent density-functional theory. Our calculated single-ionization cross sections are in agreement with the experiments in the high-energy regime. The calculated single-ionization cross sections around the peak position are 20% lower than the recent close-coupling results as well as the experiment. In the low-energy regime, our results are lower than the close-coupling results, but still larger than the experimental one. A detailed comparison between our method and the close-coupling method is discussed. Our estimated ratios of double-to-single ionization are in agreement with the experimental measurements within a factor of 2 over the whole energy range. The limitation of the present method is discussed by comparing with the more complicated theoretical methods.

DOI: 10.1103/PhysRevA.66.032709 PACS number(s): 34.50.Fa, 34.10.+x, 36.10.−k

I. INTRODUCTION

The collision between antiprotons and atoms has been a subject of interest in atom-ion collision physics. Although the collision between antiprotons and H atoms is the simplest system in the theoretical study, the experimental measurement in the lower-energy regime is not available yet due to the low H atom density in the experiment. Since He atoms are the next simple atomic system that can provide high gas density, there are some single- and double-ionization measurements in the medium to low-energy regimes [1–3]. From a theoretical point of view, the collision between He atoms and antiprotons is a fundamental two-electron system including both static and dynamic correlations. There are many theoretical studies [4–6] on the single ionization by various approximations. Although for the single ionization, the experimental data [3] were in good agreement with the continuum-distorted-wave–eikonal-initial-state (CDW-EIS) calculations [4], that should be valid in the high-energy regime, recent studies [7,8] by the close-coupling (CC) method showed that the existed experimental data could be too small in the low-energy regime. Apart from the single ionization, the ratio of double-to-single ionization provides more physical insight about the electron-electron dynamic correlation, which is not well studied yet. There are only a few studies [9–13] of the double ionization. Meanwhile, a new experimental study on the collision between atoms and antiprotons in the low-energy regime will be available in the near future by using a slow antiproton beam at Antiproton Decelerator of CERN [14]. All these stimulate us to study the ionization of He atoms by antiprotons in a wide energy range (from 5–1000 keV) by the time-dependent density-functional theory (TDDFT) [15,16]. The TDDFT with a conventional exchange-correlation functional form has already been applied to atom-ion collisions [17] and the results were not in agreement with the experiment. Like the static DFT with a conventional exchange-correlation functional form, the TDDFT with a local exchange-correlation functional form contains a spurious self-interaction, which should be removed. The TDDFT with an optimized effective potential and a self-interaction correction (TDDFT/OEP-SIC) [18,19] circumvents the self-interaction by use of a local exchange-correlation functional form with a self-interaction correction [20]. Such a method has been applied to the high-order harmonic generation of complex atoms in the intense laser fields [21]. Now, we will apply the TDDFT/OEP-SIC method to atom-ion collision processes to explore the limitation of the TDDFT in such a two-electron system. In the TDDFT/OEP-SIC equations, it is very important to have a good time propagator to solve the single-electron-like time-dependent Schrödinger equation. Here, we will use the time-dependent generalized pseudospectral method with the second-order split operator in the energy representation [22]. Such a method has been successfully applied to the high-order harmonic generation in the intense laser field [23], high-resolution spectroscopy of H atoms in the crossed electric and magnetic fields [24], and collision of H-like ions by protons [25,26] and antiprotons [27].

Based on the TDDFT/OEP-SIC method, we have calculated the single-ionization cross sections of He atoms by antiprotons in a wide energy range (from 5 to 1000 keV). We found that for the single-ionization cross sections, (1) our calculated results are in good agreement with the experiment in the high-energy regime ($E>300$ keV), (2) our results confirm that the experimental results are too small in the low-energy regime, which was reported in the recent theoretical studies [7,8], but our results are also smaller than the recent theoretical results in the low-energy regime, and (3) in the peak region, our results are smaller than the recent CC results [7,8], which are in agreement with the experiment. While our calculated ratios of double-to-single ionization

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cross sections in this regime are greater than the experimental one by a factor of 2. Our estimated ratios of double-to-single ionization are in agreement with the experimental measurements in other energy regimes. The possible reasons of the discrepancies between our results and the recent close-coupling results are the limitation of the present theoretical model and/or the way how to describe in the single and double ionizations in the TDDFT. In principle, we only know the time-dependent density in the TDDFT. A comparison between the present theoretical model and the close-coupling method is discussed. A further experiment as well as a theoretical study is needed to understand the discrepancies among the various theoretical calculations, and the discrepancies between the theoretical calculations and the experimental measurements.

We will present our theoretical method in Sec. II, in which we will emphasize on the working equations instead of a general introduction about the TDDFT, which can be found elsewhere [15,16,18,19]. The calculated results and discussion will be presented in Sec. III, followed by a summary.

II. SELF-INTERACTION-FREE TIME-DEPENDENT DENSITY-FUNCTIONAL THEORY

In the framework of the TDDFT/OEP-SIC, He atoms in an external field can be studied by solving the following time-dependent Schrödinger equation (atomic units $\hbar = m = e = 1$ are used throughout the paper unless indicates otherwise):

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = H(t)\psi(\mathbf{r},t), \tag{1}$$

with

$$H(t) = -\frac{\nabla^2}{2} + V(\mathbf{r},t) + V^{ext}(\mathbf{r},t), \tag{2}$$

$$\rho(\mathbf{r},t) = \psi^*(\mathbf{r},t)\psi(\mathbf{r},t), \tag{3}$$

$$V_c(\mathbf{r},t) = \int \frac{\rho(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}', \tag{4}$$

$$V_n(r) = -\frac{2}{r}, \tag{5}$$

and

$$V(\mathbf{r},t) = V_n + V_c + V_{xc}[\rho,\rho] - V_{xc}[\rho,0]. \tag{6}$$

Here, V_{xc} is the local-spin exchange-correlation potential [28] and $\psi(\mathbf{r},t)$ is the single-electron wave function. We assume that the total two-electron wave function is a product of the two single-electron wave functions (single determinate approximation). Note that the time-dependent effective potential in Eq. (6) is a special case of the optimized effective potential with self-interaction correction for a two-electron system. Equation (1) can be solved by the second-order split-operator method with a generalized pseudospectral grid in the energy representation [22,25,26] as

$$\psi(t+\Delta t) = e^{-iH_0\Delta t/2}e^{-i\tilde{V}(t)\Delta t}e^{-iH_0\Delta t/2}\psi(t), \tag{7}$$

where

$$H_0 = -\frac{\nabla^2}{2} + V_o(\mathbf{r}), \tag{8}$$

$$\tilde{V}(t) = V(\mathbf{r},t) - V_o(\mathbf{r}) + V^{ext}(\mathbf{r},t). \tag{9}$$

Here, $V_o(\mathbf{r})$ is an effective potential without the time-dependent external field, which is of a spherical symmetry. To propagate the wave function in Eq. (7), we use spherical coordinates and the radial part is discretized by the generalized pseudospectral grid method [29]. The first step is to map the semi-infinite domain $[0,\infty]$ or $[0,r_{max}]$ into the finite domain $[-1,1]$ using a nonlinear mapping $r = r(x)$, followed by the Legendre pseudospectral discretization. A suitable algebraic mapping for atomic structure calculations is provided by the following form:

$$r = r(x) = L\frac{1+x}{1-x+\alpha}, \tag{10}$$

where L and $\alpha = 2L/r_{max}$ are mapping parameters. The introduction of nonlinear mapping usually leads to either an asymmetric or a generalized eigenvalue problem. Such undesirable features can be removed by the use of a symmetrization procedure [29]. Thus by defining

$$\phi_l(r) = \sqrt{r'(x)}\chi_l[r(x)], \tag{11}$$

one finds the transformed Hamiltonian possesses the following symmetrized form:

$$\hat{H}_l^o(x) = -\frac{1}{2}\frac{1}{r'(x)}\frac{d^2}{dx^2}\frac{1}{r'(x)} + V_l[r(x)], \tag{12}$$

where $V_l = [l(l+1)]/2r^2 + V_o(\mathbf{r})$, leading Eq. (12) to a symmetric eigenvalue problem. In the Legendre pseudospectral method, the collocation points $\{x_i\}$ are the roots of the polynomials $P'_{N+1}(x)$, where N is the total number of grid points used in the discretization. In such a discretized scheme, the Hamiltonian $\hat{H}_l^o(x)$, Eq. (12), can be represented by the following matrix form:

$$[H_l^o]_{ij} = (D_2)_{ij} + V_l(x_i)\delta_{ij}, \tag{13}$$

with

$$(D_2)_{ij} = \frac{1}{r'(x)}\frac{(N+1)(N+2)}{6(1-x_i^2)}\frac{1}{r'(x)}, \quad i=j, \tag{14}$$

$$(D_2)_{ij} = \frac{1}{r'(x)}\frac{1}{(x_i-x_j)^2}\frac{1}{r'(x)}, \quad i \neq j. \tag{15}$$

The eigenvalues and eigenfunctions of $\hat{H}_l^o$ will be denoted by $\{\varepsilon_k(l)\}$ and $\{\chi_{ki}(l)\}$, respectively. The propagation of the wave function can be performed in Eq. (7).

The advantages of the numerical method are that (1) we use a nonequal space grid with a denser grid in the physically important region (interaction region) and wider grid in the outer region to save the computer time; (2) we propagate the time-dependent wave function in the energy representation that is more effective and accurate than that in the kinetic representation [30,31]. The detailed numerical procedure can be found in Refs. [22,25,26]. With the impact-parameter approximation, the interaction between the antiproton and electrons is written as

$$V^{ext}(\mathbf{r},t)=\frac{1}{|\mathbf{R}(t)-\mathbf{r}|}. \tag{16}$$

Here, we assume that the trajectory of the antiproton is a straight line. We propagate the wave function from $t=0 \rightarrow T$, with an impact parameter b along the x direction and a projectile velocity v along the z direction starting from z_0. The initial wave function is located in the target ground state $1s^2$. When the projectile passes through the target or is far from the target, we can obtain the single-electron survival probability as

$$P(T,b)=\sum_i |\langle \psi(T)|\varphi_i\rangle|^2, \tag{17}$$

with φ_i, the time-independent bound-state wave function centered at the target He atom. The corresponding single and double cross sections can be estimated as

$$\sigma^{+}=2\pi\int P^{+}(b)b\ db, \tag{18}$$

$$\sigma^{2+}=2\pi\int P^{2+}(b)b\ db, \tag{19}$$

with

$$P^{+}(b)=2[1-P(T,b)]P(T,b), \tag{20}$$

$$P^{2+}(b)=[1-P(T,b)]^2. \tag{21}$$

Note that the limitation of Eqs. (20) and (21) will be discussed in Sec. III for a comparison of our present method with the close-coupling method. If we choose

$$\tilde{V}(t)=V^{ext}(\mathbf{r},t) \tag{22}$$

in Eq. (9), we will return to the single active electron (SAE) model [8,32,33]. In the SAE model, the electron-electron dynamic correlation, $V(\mathbf{r},t)-V_o(\mathbf{r})$ in Eq. (9) is ignored. To study the dynamic correlation effect, we will compare the single- and double-ionization cross sections calculated with TDDFT/OEP-SIC and SAE/OEP-SIC methods.

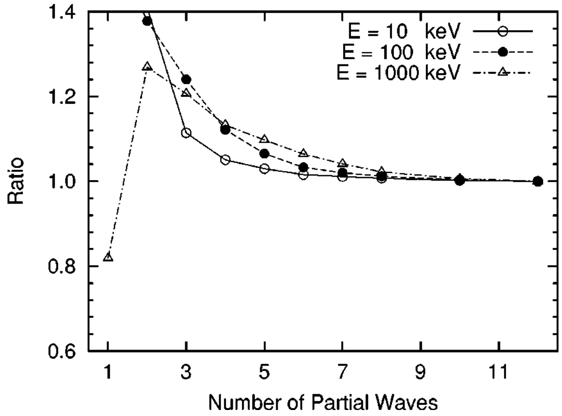

FIG. 1. Single-ionization cross sections of He atoms by antiprotons as a function of the number of partial waves included in the calculation by the TDDFT/OEP-SIC method.

III. RESULTS AND DISCUSSION

Based on the TDDFT/OEP-SIC method, we have calculated the single-ionization cross sections of He atoms by antiprotons over a wide energy range (from 5 to 1000 keV). All the calculations were performed in a 4 PC Linux cluster with a Pentium III 400-MHz CPU. To check the numerical stability, we first turn off the external potential V_{ext} and propagate the wave function. The normalization of the time-dependent wave function changes less than 10^{-9} after 5000 time steps. The numerical convergence has also been checked by changing the number of the partial waves and the number of grid points. Figure 1 shows the convergence of the single-ionization cross sections against the partial waves at 10-, 100-, and 1000-keV impact energies, respectively. For the lower impact energy (10 keV), we can get the converged results with a few partial waves ($l<5$). For the higher impact energy (1000 keV), we need more partial waves to get the converged results. All these are consistent with our common sense that for a higher impact energy, we need more partial waves in the calculation. The final results presented here are calculated with 12 partial waves and 128 radial grid points with 1000–3000 time steps. For the lower impact energy, we use more time steps.

Figure 2 shows the single-ionization cross sections calculated by the TDDFT/OEP-SIC (thick solid line) and SAE/OEP-SIC (thick dashed line) methods. It is very interesting to see that the two results are in agreement with each other in the high-energy regime ($E>200$ keV). The SAE/OEP-SIC results are larger than that of the TDDFT/OEP-SIC as the impact energy decreases. This can be understood as for the high impact energy, the interaction time is short and the electron-electron dynamic correlation is smaller than that for the low impact energy case. This observation is in agreement with the comparison of the independent particle model and one active electron model reported by Igarashi *et al.* [8] as also shown in Fig. 2. But our results are significantly lower than their results in the medium-to-low energy regimes. Physically speaking, due to the partial ionization of electrons, the remaining electrons are bounded tightly by the nucleus. The SAE/OEP-SIC calculation does not take into account such dynamic correlation effects. Therefore, the

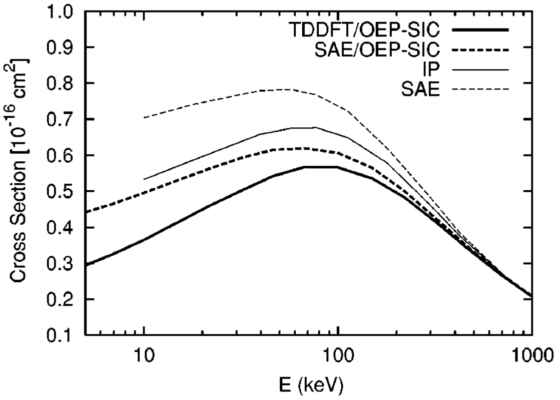

FIG. 2. Single-ionization cross sections in the collision between He atoms and antiprotons calculated by TDDFT/OEP-SIC and SAE/OEP-SIC. The calculations of the independent particle model and the single active electron model from Ref. [8] are also presented for comparison.

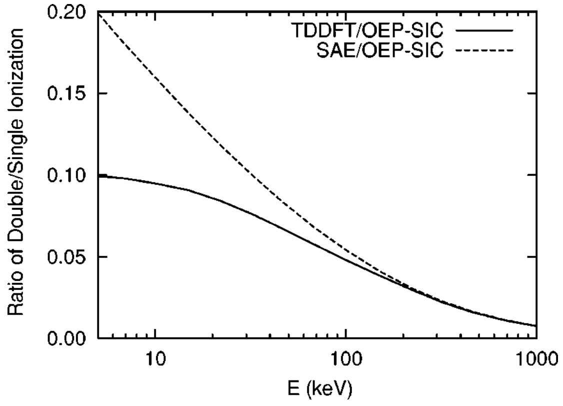

FIG. 4. Ratio of double to single ionization of He atoms by antiprotons calculated by TDDFT/OEP-SIC and SAE/OEP-SIC methods.

SAE/OEP-SIC results are larger than the TDDFT/OEP-SIC results as shown in Fig. 2. A similar behavior has also been observed in the high-order harmonic generation (HHG) of He atoms in intense laser fields [18]. The HHG yield of TDDFT/OEP-SIC and SAE/OEP-SIC shows a large discrepancy in an intense laser field than in a relative weak field.

Figure 3 shows our calculated results compared with the experiment [3] as well as other theoretical calculations. Our results are in good agreement with the experiment for the energy above 200 keV. Large discrepancies between our calculation and the experiment appear in the peak position (around 100 keV). Comparing with other theoretical works shows that all the theoretical works agree with each other in the high-energy regime ($E > 300$ keV). Agreement between our results and the forced impulse method (FIM) [10] extends to a further lower energy ($E > 150$ keV). The CC results [7,8] are in agreement with CDW-EIS results [4] from high-energy regime to the peak position. Our results are lower than the CC results. In the low-energy regime ($E < 20$ keV), our results decrease much slower than the experiment as the impact energy decreases, and our results get closer to the CC results.

Now let us estimate the ratio of double to single ionization of He atoms by antiprotons from Eqs. (18) and (19). The double ionization can be produced through (a) the shake-off mechanisms, which are related to the static correlation and (b) dynamic correlation, which are strongly dependent on the projectile, its impact velocity, and so on. In principle, the double ionization should be studied with a highly correlated wave function. The double ionization cross section, Eqs. (19) and (21), in the TDDFT/OEP-SIC method is an approximation. We have no idea how good or bad it is so far. Figure 4 shows the ratio of double to single ionization calculated by the TDDFT/OEP-SIC and SAE/OEP-SIC methods. In the high-energy regime ($E > 200$ keV), the two results are in good agreement with each other. As the energy decreases, the SAE/OEP-SIC results increase much more rapidly than the TDDFT/OEP-SIC results. Our calculated ratios are in reasonable agreement with the experiment in the high-energy regime as well as in the lower-energy regime as shown in Fig. 5. In all other parts, our results are larger than the experimental one by almost a factor of 2, but the general trend is right. In the low-energy regime, the results of the close-

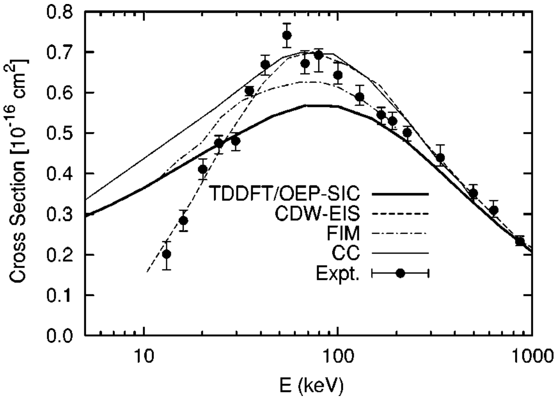

FIG. 3. The single-ionization cross sections of He atoms by antiprotons. Our calculated results are presented as TDDFT/OEP-SIC. The experimental results are from Ref. [3]. Other theoretical works: CDW-EIS calculation from Ref. [4], FIM calculation from Ref. [34], and CC calculation from Ref. [7].

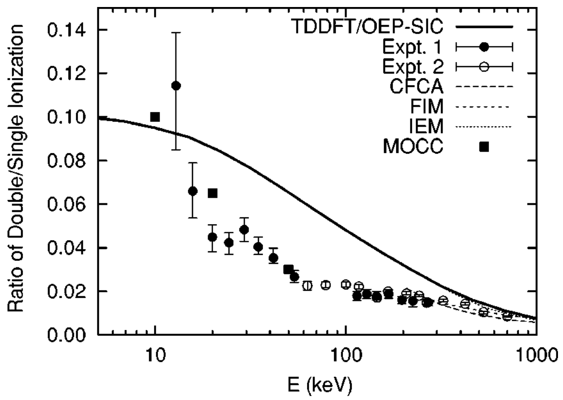

FIG. 5. Comparison of our calculated ratio of double to single ionizations with other theoretical works and the experiments. Experiment 1 from Ref. [3]; experiment 2 from Ref. [1]; CFCA and IEM from Ref. [12]; FIM from Ref. [10]; and MOCC from Ref. [9].

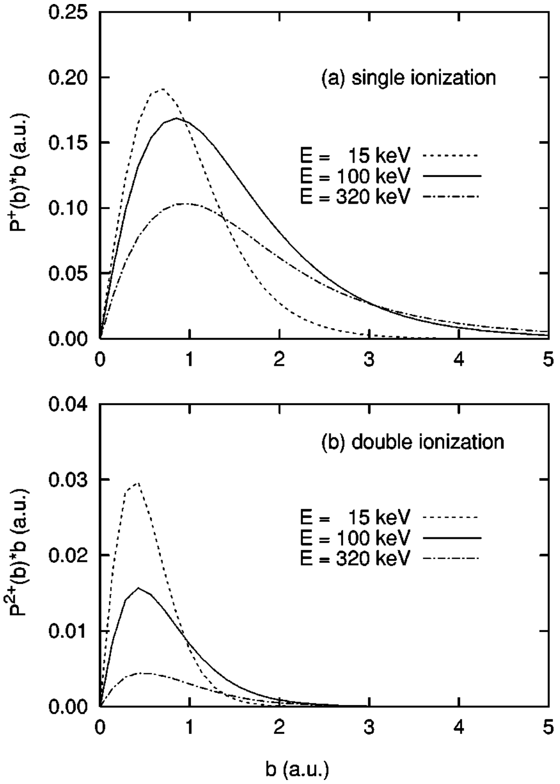

FIG. 6. (a) The single- and (b) double-ionization probabilities as a function of the impact parameter *b* with three impact energies.

coupling method with molecular-orbital one [9] are in fair agreement with the experiment, as shown in Fig. 5. In the high-energy regime, all the calculations [10,12] are in agreemental with the experiment measurement [1]. The ratio increases as the impact energy decreases, while the single-ionization cross section has a peak around 100 keV. We will study the ionization probability as a function of the impact parameter *b*, as shown in Fig. 6. We choose three impact energies, 100 keV, the peak position, 15 and 310 keV, at which the single-ionization cross section are almost the same. Figure 6 clearly shows that the single-ionization probability extends to the large impact parameter regime. The double-ionization probability is dominant in the small impact parameter regime, which explains that the ratio increases as the impact energy decreases. Such observation is consistent with intuition.

Now, let us compare our method with the close-coupling method from a more fundamental point of view. For a collision involving one electron, namely, H atoms by antiprotons or protons, our method is equivalent to the close-coupling method with one-center complete L^2 basis set. Since our one atomic orbital forms a "complete" basis set and we do not need to calculate the overlap and the interaction matrix in the time propagation, our method is more efficient than the close-coupling method. For a two-electron system, we use a single determinant wave function $\Psi(\mathbf{r}_1,\mathbf{r}_2) = \psi(\mathbf{r}_1,t)\psi(\mathbf{r}_2,t)$, with single-electron wave functions expanded over one-center complete L^2 basis set. Although all the two-electron configurations are taken into account in the single determinant, the coefficients are not independent. If we partition the single-electron wave function into

$$\psi = C_b\psi_b + C_c\psi_c, \tag{23}$$

with ψ_b and ψ_c representing the excited and continuum wave functions, the single determinant wave function will be written as

$$\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = C_b^2\psi_b(\mathbf{r}_1)\psi_b(\mathbf{r}_2) + C_c^2\psi_c(\mathbf{r}_1)\psi_c(\mathbf{r}_2) + C_bC_c[\psi_b(\mathbf{r}_1)\psi_c(\mathbf{r}_2) + \psi_c(\mathbf{r}_1)\psi_b(\mathbf{r}_2)]. \tag{24}$$

The second and third terms in Eq. (24) are the double and single ionization wave functions, that are not independent. This results that the two independent processes (single and double ionization) are described by a single parameter in Eqs. (20) and (21). Even with such a drawback, our calculated single- and double-ionization cross sections are still in reasonable agreement with the experiments. Unlike the close-coupling method, we cannot choose each individual configuration within the single-determinant approximation. To overcome the limitation of the present method, a multideterminant is needed.

To summarize, the single-impact ionization cross sections of He atoms by antiprotons has been studied by the TDDFT/OEP-SIC method in a wide energy range (from 5 to 1000 keV). Our calculated single-ionization cross sections are in good agreement with the experiment in the high-energy regime ($E > 300$ keV), but significantly lower than the recent CC calculations [7,8] in the peak region ($E \sim 100$), which were in agreement with the experiment [3]. The possible reasons of the discrepancies between our results and the recent close-coupling results are the limitation of the present theoretical model. Our estimated ratios of double to single ionization are in agreement with the experimental measurements within a factor of 2 over the whole energy range. Although the TDDFT shows an improvement over the SAE model, it still needs an improvement in considering the dynamic correlation more correctly.

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