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Fully Correlated Electronic Dynamics for Antiproton Impact Ionization of Helium

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We present total cross sections for single and double ionization of helium by antiproton impact over a wide range of impact energies from 10 keV/amu to 1 MeV/amu. A nonperturbative time-dependent close-coupling method is applied to fully treat the correlated dynamics of the ionized electrons. Excellent agreement is obtained between our calculations and experimental measurements of total single and double ionization cross sections at high impact energies, whereas for lower impact energies, some discrepancies with experiment are found. At an impact energy of 1 MeV we also find that the double-to-single ionization ratio is twice as large for antiproton impact as for proton impact, confirming a long-standing unexpected experimental measurement.

The double ionization of helium by ion impact has been a long and fruitful field of study in atomic collision physics. As a fundamental four-body system, it provides stringent tests of any theoretical description of charged particles moving in a Coulomb field. Moreover, the sign and magnitude of the ion charge can be experimentally varied relatively simply to explore interesting physical effects. New antimatter collision experiments are planned at the Facility for Antiproton and Ion Research (FAIR) [1]. FAIR is an international collaboration on atomic and molecular physics that intends to investigate antiproton driven ionization processes and even kinematically complete antiproton collision experiments. These experimental efforts complement the recent intense activity in antihydrogen studies of recent years [2]. In the past 20 years, experimental measurements showed, unexpectedly, that the ratio of double-to-single ionization of helium from proton impact was around a factor of 2 lower than that from antiproton impact [3], a feature which has not yet been observed in a fully converged theoretical calculation. Subsequent experimental measurements [4] of the total double ionization cross section by antiproton impact not only revealed a larger cross section than for proton impact, but also that at low impact energies, the double ionization cross section for antiproton impact does *not* monotonically decrease, but shows an increasing cross section as the impact energy is lowered.

In response to this renewed experimental activity, in this Letter we present fully converged calculations of single and double ionization of helium from antiproton impact and find similar unexpected ratios of the double-to-single ionization of helium compared with proton impact. We also provide converged single and double ionization cross sections for antiproton impact over a wide energy range from 10 keV to 1 MeV. Theoretical calculations for single ionization of helium by antiproton impact cross sections have suggested that the experimental measurements [4]

below 30 keV may be inaccurate [5,6]. Experimental efforts are under way to remeasure the single and double ionization cross sections [7], and preliminary results [8] suggest that the single ionization cross section does not decrease as rapidly as reported by Hvelplund *et al.* [4]. These preliminary results may bring closer agreement between theory and experiment for single ionization.

For double ionization of helium by antiproton impact, no theoretical description has been used to calculate the cross section over the full energy range of the measurements. Barna *et al.* [9] calculated the double ionization cross section at a single energy of 3.6 MeV which agreed well with experiment. A close-coupling method was used by Díaz *et al.* [10] to calculate double ionization cross sections for impact energies above 200 keV, with limited success. The calculations reported were around a factor of 2 lower than the measurements of [4]. Keim *et al.* calculated the single and double ionization of helium by antiproton impact using the framework of time-dependent density-functional theory [11]. These results produced reasonable agreement for the single ionization cross sections. However, the double ionization cross sections were considerably higher than the experimental results for all energies. A similar approach was used by Tong *et al.* [12], who calculated single ionization cross sections that were lower than the experimental measurements, and double ionization probabilities for 15 and 100 keV that yield much larger cross sections compared with experiment. The multicut forced impulse method has been the only calculation for double ionization of helium by antiproton impact below 200 keV [13] which has compared well with experiment. However, at these low impact energies the multicut forced impulse method predicts single ionization cross sections that are in less than satisfactory agreement with previous calculations [5,6].

In this work, we apply our time-dependent close-coupling (TDCC) method to treat the double ionization

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of helium by antiproton impact [14]. The six-dimensional time-dependent Schrödinger equation for the outgoing electrons is reduced to a coupled set of two-dimensional partial differential equations through a partial-wave expansion of the two-electron wave function in spherical coordinates centered on the target atom. The two-electron wave function is subjected to a time-dependent projectile interaction which is included through a multipole expansion. We assume a straight-line motion for the impacting ion.

Our previous study using this approach explored alpha-particle collisions with helium. For high impact energies, excellent agreement between theory and experiment was found for single and double ionization of helium. At low energies, charge transfer to the impacting ion may become significant, so that a description using a two-electron wave function centered on the target is unsuitable. However, in the case of antiproton impact, charge transfer cannot occur, so our method is appropriate for treatment of the antiproton-atom collision at any impact energy.

The fully correlated wave function Ψ^{LM} for the single and double ionization of a two-electron target atom by antiproton collision is obtained by the evolution of the time-dependent Schrödinger equation in real time:

$$i\frac{\partial \Psi^{LM}(\vec{r}_1, \vec{r}_2, t)}{\partial t} = H_{\text{system}}\Psi^{LM}(\vec{r}_1, \vec{r}_2, t), \quad (1)$$

where the nonrelativistic Hamiltonian is given by

$$H_{\text{system}} = \sum_i^2\left(-\frac{1}{2}\nabla_i^2 - \frac{Z_t}{r_i}\right) + \frac{1}{|\vec{r}_1 - \vec{r}_2|} - \frac{Z_p}{|\vec{r}_1 - \vec{R}(t)|}$$
$$- \frac{Z_p}{|\vec{r}_2 - \vec{R}(t)|} \quad (2)$$

and Z_p is the projectile atomic number and Z_t is the target atomic number. For straight-line motion, the magnitude of the time-dependent projectile position is given by

$$R(t) = \sqrt{b^2 + (d_0 + vt)^2}, \quad (3)$$

where b is an impact parameter, d_0 is a starting distance ($d_0 < 0$), and v is the projectile speed. If we expand Ψ^{LM} in coupled spherical harmonics and substitute into Eq. (1), the resulting close-coupled equations for the radial expansion functions, $P_{l_1l_2}^{LM}(r_1, r_2, t)$, are then given by

$$i\frac{\partial P_{l_1l_2}^{LM}(r_1, r_2, t)}{\partial t} = T_{l_1l_2}(r_1, r_2)P_{l_1l_2}^{LM}(r_1, r_2, t) + \sum_{l_1',l_2'}V_{l_1l_2,l_1'l_2'}^{L}(r_1, r_2)P_{l_1'l_2'}^{LM}(r_1, r_2, t) + \sum_{L'M'}\sum_{l_1',l_2'}W_{l_1l_2,l_1'l_2'}^{LM,L'M'}(r_1, R(t))P_{l_1'l_2'}^{L'M'}(r_1, r_2, t)$$
$$+ \sum_{L'M'}\sum_{l_1',l_2'}W_{l_1l_2,l_1'l_2'}^{LM,L'M'}(r_2, R(t))P_{l_1'l_2'}^{L'M'}(r_1, r_2, t), \quad (4)$$

where $V_{l_1l_2,l_1'l_2'}^{L}(r_1, r_2)$ is the electron-electron potential and $W_{l_1l_2,l_1'l_2'}^{LM,L'M'}(r_i, R(t))$ represents the interaction between the projectile and the electrons. The initial value boundary condition for Eq. (4) is given by

$$P_{l_1l_2}^{LM}(r_1, r_2, t=0) = \delta_{L,L_0}\delta_{M,M_0}\bar{P}_{l_1l_2}^{L_0M_0}(r_1, r_2, \tau \rightarrow \infty), \quad (5)$$

where $\bar{P}_{l_1l_2}^{L_0M_0}$ is the radial portion of the ground state wave function. The two-electron ground state wave function of the helium atom is obtained by relaxation of the time-dependent Schrödinger equation in imaginary time ($\tau = it$). The fully correlated ground state is expanded in coupled spherical harmonics resulting in a set of close-coupled equations for the $\bar{P}_{l_1l_2}^{L_0M_0}(r_1, r_2, \tau)$ radial expansion functions with the initial boundary conditions given by a product of single particle bound radial orbitals for the one-electron target ion with $L = M = 0$ [14].

We solve the TDCC equations, Eq. (4), using a lattice technique to obtain a discrete representation of the radial expansion function and all operators on a two-dimensional grid. The single and double ionization probabilities, $\mathcal{P}(E, b)$, are calculated using the time-dependent radial wave function propagated for a suitable time. The radial wave function is then projected onto both bound and continuum single particle orbitals, as discussed in detail in [14].

The TDCC calculations employed a 384×384 point radial lattice with a uniform mesh spacing of $\Delta r = 0.20$,

giving a converged ground state of helium on the lattice. In order to achieve accurate double ionization cross sections it was found that up to 101 coupled channels had to be included in the expansion of the final state two-electron wave function. The time-dependent projectile potential was expanded into monopole, dipole, quadrupole, and octopole terms, all of which were required to converge the total cross sections. The time-dependent wave function was propagated from a time when the projectile was a distance $d_0 = -50a_0$, through closest approach, to a time when the projectile was at a distance $d_0 = +40a_0$. The total cross sections calculated at the lowest impact energy of 10 keV had to be propagated for 142 a.u. of time to reach this distance using a number of time steps greater than 28 000.

In Fig. 1, we present our TDCC calculations for single and double ionization of helium by antiproton collisions. The top panel of Fig. 1 shows the single ionization cross section measurements [4,15], our TDCC results (red squares), and the calculations of Schultz and Krstić [5]. The two sets of calculations are in good agreement over a wide energy range down to around 30 keV. Below 30 keV, the TDCC calculations are higher than the experimental measurements, although new preliminary measurements of this single ionization cross section [8] suggest that the previous measurements [4] may be too low.

The lower panel of Fig. 1 shows the double ionization of helium by antiproton impact for ion energies from 10 keV
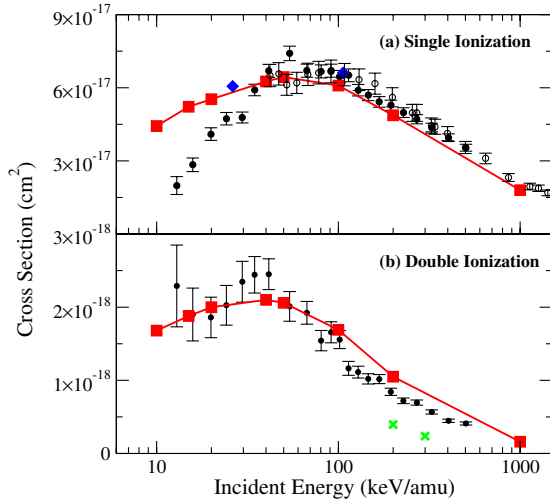
FIG. 1 (color online). (a) Cross sections for single ionization of helium by antiproton impact. Filled squares: TDCC calculations; filled diamonds: calculations of Schultz and Krstić [5]; filled and open circles: experimental measurements of [4,15]. (b) Cross sections for double ionization of helium by antiproton impact. Filled squares: TDCC calculations; crosses: calculations of Diaz *et al.* [10]; filled circles: experimental measurements of [4].
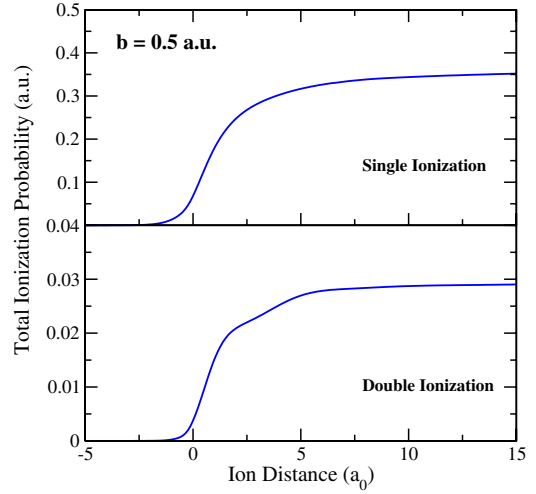

FIG. 2 (color online). Evolution of the ionization probability, $\mathcal{P}(E, b = 0.5a_0)$, for a 50 keV antiproton collision with a helium atom as a function of the impacting ion distance. Upper panel: single ionization probability summed over all partial waves. Lower panel: double ionization probability. The helium atom is located at the origin of the collision system ($d_0 = 0$).

to 1 MeV. The TDCC (red squares) calculations are in excellent agreement with the experimental measurements above 20 keV. Between 10 and 20 keV, the TDCC calculations decrease monotonically. However, the experimental measurements show a sharp increase as the impact energy decreases. The TDCC results are in excellent agreement with the experimental data points at 15 and 20 keV, suggesting that perhaps only the lowest energy measurements are in error.

A time-dependent propagation of the two-electron wave function can be a powerful tool in understanding the evolution of the collision system. Figure 2 shows the ionization probability $\mathcal{P}(E, b)$ for both single and double ionization for a 50 keV antiproton collision with a helium atom. The initial ion starts at $d_0 = -50a_0$, then propagates through distance of closest approach ($b = 0.5a_0$) to a final distance of $d_0 = +40a_0$. The total propagation time for the 50 keV collision system is 63 a.u. of time. The helium atom is placed at $d_0 = 0a_0$. Figure 2 shows the evolution of the ionization probability for one impact parameter ($b = 0.5a_0$) and for an ion distance from $d_0 = -5a_0 \rightarrow 15a_0$. The choice of the impact parameter equal to $b = 0.5a_0$ represents the impact distance of maximum double ionization probability. The total single and double ionization cross sections for a given energy are obtained by the relation

$$\sigma(E) = 2\pi \int_0^\infty \mathcal{P}(E, b) b\, db. \qquad (6)$$

The ionization probabilities for both single and double ionization of helium by antiproton impact increase rapidly just before the antiproton reaches the helium atom (posi-

tioned at $d_0 = 0a_0$), and then tend to a constant value beyond $d_0 \sim 10a_0$. The results in Fig. 2 show that the antiproton collision with the helium atom occurs in approximately $t_{\mathrm{collision}} = 4.24$ a.u. of time or 0.1 fs. Such fast collision times are currently just beyond the pulse lengths of experimental ultrafast laser techniques which are being considered for use in probing electronic behavior of atomic systems.

The total ionization cross sections are calculated by integrating the ionization probabilities as defined in Eq. (6). Figure 3 shows the weighted probabilities [the integrand of Eq. (6)] for both proton (black dashed line) and antiproton (red solid line) ionization as a function of the impact distance from the helium atom at an incident energy of 1 MeV. For the antiproton impact, the peak of the single ionization weighted probability is at an impact parameter of $b \approx 1.0a_0$, while the double ionization weighted probability peaks at a smaller impact parameter ($b \approx 0.8a_0$). The proton's maximum ionization probability is similar to the antiproton for single ionization but shifted to smaller impact parameters for double ionization. However, of much greater significance is the difference between the proton and antiproton double ionization probabilities. As also observed in the experimentally measured double-to-single ratios [3,4], our calculations find that for antiprotons the double ionization cross section is larger than the proton double ionization by approximately a factor of 2, but that the single ionization processes have approximately the same total cross section. At this impact energy, the total single and double ionization cross sections are in good agreement with experiment for antiproton impact (as shown in Fig. 1) and for proton impact (compared with the measurements of [16]). The reasons for this difference between the double ionization cross sections are
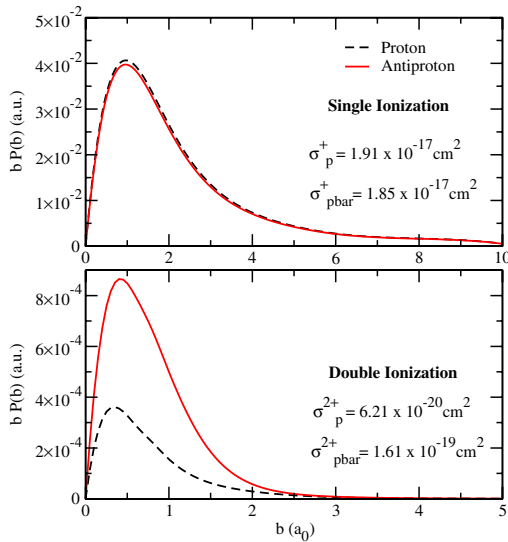
FIG. 3 (color online).   Weighted probabilities for single (upper) and double (lower) ionization of helium by antiproton (red solid line) and proton (black dashed line) impact at 1 MeV impact energy.

still not well understood. Clearly, the change of sign of the projectile affects the double ionization mechanism in a much greater manner than the single ionization mechanism. This could be partially explained by arguments arising from a Born-approximation description [17], where interference between various mechanisms for double ionization depended on the sign of the ion charge, resulting in different double-to-single ratios for protons and antiprotons. We note that following a rapid ejection of a single electron, which based on the upper panel of Fig. 3 is equally probable for the proton impact and antiproton impact, the remaining transient collision complexes are quite different. For proton impact, the remaining electron sees a positive charge of $+3$, while for the antiproton impact the remaining electron sees a positive charge of $+1$. Thus, the second electron may find it easier to escape in the antiproton impact case. Similar effects have been observed for double photoionization of helium by large photon energies, where the shakeoff mechanism becomes dominant at high photon energy [18,19]. Although complicated by the multipole potential in this case, a similar mechanism may be responsible for the large double ionization probability from the antiproton impact. Also, the impact distance b can provide useful insight into understanding the dynamics involved in the collision. Clearly, double ionization is most probable when the antiproton significantly penetrates the electron cloud of the helium atom. This suggests the possibility that the antiproton undergoes multiple collisions with the electrons to doubly ionize. Single ionization is most probable at a somewhat larger impact parameter, and still is probable even when the ion is 3 or 4 a.u. from the atom, at which distance there is almost no probability of double ionization. Thus the

single ionization mechanism is likely dominated by a more standard "binary" collision process.

In conclusion, the TDCC method has been shown to accurately compute single and double ionization total cross sections for antiproton impact ionization of helium over a wide energy range. The single ionization total cross section calculations are in excellent agreement with experiment and with previous calculations. The double ionization total cross section calculations also agree well with experiment above 20 keV. The antiproton double ionization cross sections are 2 times larger than the proton double ionization cross sections at the same impact energy, whereas the single ionization cross sections are similar. We aim to further exploit our method to extract the differential cross sections for the two outgoing electrons after ion impact. Such calculations may allow comparison with the large set of experimental measurements of differential cross sections for ion impact which currently exist (e.g. [20]) and should yield more insight into the nature of the double ionization process.

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