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Mechanisms of double excitation in ion-atom collisions at high velocities

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Double-excitation cross sections of helium atoms by bare heavy-ion projectiles at high energies are calculated within the one-center atomic-orbital expansion method including the electron-electron interaction between correlated helium wave functions. The resulting excitation cross sections to $2s^2\,{}^1S^e$, $2p^2\,{}^1D^e$, $2s2p\,{}^1P^o$, and $2p^2\,{}^1S^e$ states are analyzed. We conclude that double excitations at high energies by low-Z_p projectiles are dominated by a first-order process in which the two-electron transition proceeds through one electron-projectile interaction in conjunction with shakeup or rearrangement due to electron-electron interactions. For high-Z_p projectiles at a given velocity, on the other hand, the excitation is dominated by a second-order process in which the transition proceeds through two successive electron-projectile interactions.

I. INTRODUCTION

Mechanisms of two-electron transitions are currently of great interest in atomic collisions. These effects have been studied, e.g., in double ionization,[1-7] double capture,[8-10] and transfer excitation[11] in the keV/u to MeV/u region. Of particular interest is the collision at high energies where the projectile speed is much faster than the speed v_0 of the orbiting electrons in the target. For such collisions, it is well known that the first-order Born approximation or its equivalents are adequate in describing single-excitation and single-ionization processes, particularly for optically allowed transitions. In the first-order Born approximation the cross sections scale quadratically with respect to the charge of the projectile and thus there is no difference in the total cross sections between particle and antiparticle projectiles. This has been illustrated recently for the single-ionization cross sections of He by protons and antiprotons.[1]

The situation is less clear for two-electron transitions. Measurements of He excitation by protons and antiprotons in a broad energy range around 1–2 MeV show that double-ionization cross sections by antiprotons are about twice as large as those seen with equivelocity protons.[1] To understand these differences, various qualitative interpretations[3,4] have been made and some quantitative calculations have been carried out by Reading and Ford.[2] Of particular interest is the assessment of the role of electron correlation and the importance of double-collision mechanisms for the two-electron transitions. In this respect, total double-ionization cross sections are difficult to analyze since many integrations over the angles and energies of the ejected electrons and the scattering angles of the projectile have to be carried out before theory and experiment can be compared.

In order to unravel the different mechanisms for two-electron transitions, it was argued[3] that it is more convenient to study double-excitation processes to specific doubly excited states at high velocities. This has motivated experimentalists to look at double-excitation processes recently.[12,13] However, as we will see, the study of double excitation is complicated by its interference with the direct single-ionization process, and the analysis and interpretation of double-excitation cross sections are possible only if the interference can be neglected.

In the limit that there is no interference between double-excitation and single-ionization processes, the dependence of double-excitation cross sections can be analyzed from the perturbation-series viewpoint. If the two-electron transition is determined by the first-order electron-projectile interaction, the resulting cross section scales as Z_p^2. Such a transition will be called a first-order process that involves the electron-projectile interaction only once. If the transition proceeds through a double collision mechanism, where two-electron transitions result from two successive electron-projectile interactions, then such a process is called a second-order process and the cross section scales as Z_p^4. If the two mechanisms are of comparable importance, then the cross section will have a Z_p^3 dependence term resulting from the interference of the two mechanisms.[3] Experimental studies have been carried out recently by Pedersen and Hvelplund[12] using fast equivelocity electrons, protons, and carbon ions (charge state 4–6) at 1.84 MeV/u and by Giese *et al.*[13] using similar ions plus F^{q+} ($q=7-9$) at 1.5 MeV/u, with the goal of understanding mechanisms of double excitations. It is the purpose of this work to analyze the calculated double excitations of He by various projectiles obtained from theoretical calculations and to assess the relative importance of the first-order and the second-order mechanisms.

II. METHODS OF CALCULATIONS

Although at high energies $E > 1$ MeV/u, and at least for low-Z_p projectiles, a perturbation expansion approach (at least up to the second Born term) for the calculation of double-excitation cross sections is fully justified, it was found more convenient to calculate the same using an existing coupled-channel code developed previously[14] for low-energy two-electron collision systems. Since we are concerned with collisions at high velocities where charge-transfer channels are small, only two-electron target atomic states $1s^2\,{}^1S^e$, $1s2s\,{}^1S^e$, $1s2p\,{}^1P^o$, $2s^2\,{}^1S^e$, $2p^2\,{}^1D^e$, $2s2p\,{}^1P^o$, and $2p^2\,{}^1S^e$ are included in the close-coupling expansion. Although independent particle notations are used in the designation of states, actually each wave function is obtained from a diagonalization of the helium Hamiltonian within the given set of two-electron states.[11] These two-electron states are products of hydrogenic functions with effective charges chosen variationally to minimize the energy of the states included. The effective charges and the resulting energies thus obtained are shown in Table I of Ref. 11. Better but more complicated wave functions can be obtained from the structure calculation but were not pursued because of the large increase of computing time needed. The two-electron time-dependent Schrödinger equation is solved numerically within this basis set and excitation amplitudes to each doubly excited state are extracted. The total cross section for each state is obtained by integrating the excitation probabilities over impact parameters.

Two different calculations have been carried out: One includes all the basis functions described above and the other is truncated calculation where $1s2s\,{}^1S^e$ and $1s2p\,{}^1P^o$ singly excited states are *not* included. In a second-order process, doubly excited states are populated through two successive one-electron transitions if electron correlation is neglected; the first step is to populate one of the singly excited states, $1s2s\,{}^1S^e$ or $1s2p\,{}^1P^o$, and in the second step the remaining $1s$ electron is further excited to $2s$ or $2p$. By removing these singly excited states from the basis set, two such successive one-electron transitions can no longer proceed. By comparing the results from these two calculations, the importance of the double collision process for populating each doubly excited state is assessed.

III. RESULTS AND DISCUSSION

A. Total cross sections

Calculations of double excitations of He have been carried out for various bare projectiles at two fixed velocities corresponding to energies of 1.5 and 6 MeV/u. Before presenting the results, it is appropriate to point out how the calculated results can be compared with experimental data. Since each doubly excited state studied here decays almost exclusively via autoionization, experimental ejected electron spectra at each ejected angle are expected to display Fano resonance behavior,[15]

$$\frac{d^2\sigma}{d\theta\,dE} = C(\theta,E) + \frac{A_j(\theta)\varepsilon_j + B_j(\theta)}{1+\varepsilon_j^2}\ , \tag{1}$$

near each resonance showing effects of interference with direct single-ionization continuum. In (1), Shore's parametrization was used.[16] For each resonance j, $\varepsilon_j = 2(E-E_j)/\Gamma_j$, where E_j is the resonance energy and Γ_j its width. If the electron spectra are integrated over the resonance, the result is

$$\frac{d\sigma}{d\theta} = \int C(\theta,E)\,dE + \tfrac{1}{2}\pi\Gamma_j B_j(\theta)\ , \tag{2}$$

where the term containing $A_j(\theta)$ is integrated out. Equation (1) "appears" to consist of a first term coming from the direct single ionization, a third term containing $B_j(\theta)$ from the double excitation, and a second term containing $A_j(\theta)$ from the interference of the two. Thus one associates the last term in (2) with the cross section arriving from the doubly excited state.[17,18] However, as have been shown in the literature,[15,19] such an identification is not correct when the *ejected electron* spectra are observed since the coefficient $B_j(\theta)$ contains terms that are products of single-ionization and double-excitation amplitudes. [If the energy loss of the *scattered* particles are measured, then the term containing $B_j(\theta)$ in (2) gives the contribution of the double-excitation process alone; see Refs. 15 and 19.] Despite of this limitation, the last term in (1) or (2) has often been treated in the literature as the cross sections from the double-excitation process. If such a procedure is adopted, then integration of the last term in (2) over angles gives an "experimental" total double-excitation cross section which can be compared with the present calculation. In a rigorous test of the theory, comparison between calculation and experiment should be carried out by comparing double differential cross sections directly, i.e., at the level given by Eq. (1). Theoretically this cannot be done at present since one does not know how to incorporate continuum states into a coupled-channel calculation. There is another further complication in comparing the theory with experimental results. The ejected electron spectra are modified by the post-collision interaction[20,21] effect which makes the extraction of experimental total double-excitation cross sections for each state even more dubious. However, the goal here is to identify the different mechanisms for double excitations over a large energy region and for many different projectiles. A rough comparison with experimental cross sections derived using the procedure described above will be made despite the inherent problem with such a comparison.

With the above comments in mind, we now compare the calculated double-excitation cross sections with experimental data in Table I while the full set of calculated results are given in Table II. In the measurement of Pedersen and Hvelplund, unresolved structures were observed for the $2p^2\,{}^1D^e$ and $2s2p\,{}^1P^o$ states in the ejected electron spectra at different angles using 1.84-MeV/u ions. In the similar measurement of Giese *et al.* at 1.5 MeV/u, these two states were partially resolved in the electron spectra, and the individual cross section for each state was extracted. In Table I, the calculation for negatively charged particles was carried out for antiprotons, but the measurements were for electrons at the same velocity. We note that for e^- and p projectiles the calcula-

TABLE I. Experimental and theoretical double-excitation cross sections of He by various bare projectiles. The entries are the sum of double excitation to $2p^2\,{}^1D^e$ and $2s2p\,{}^1P^o$ states in units of 10^{-19} cm^2.

	1.84 MeV/u		1.5 MeV/u	
	Reference 12	Present	Reference 13	Present
$e^-(\bar{p})$	0.191±0.04	0.32	0.305±0.011	0.35
p	0.231±0.058	0.32	0.245±0.088	0.35
C^{6+}	8.305±1.744	25.6	20.1±7.20	31.8
F^{9+}			48.99±17.66	111.6

tions agree with experiments to better than a factor of 2. For C^{6+}, the two sets of experimental data have large discrepancies. Our calculation agrees better with the data of Giese *et al.*, but the difference becomes larger for F^{9+}. This discrepancy could be due to either the difficulty of extracting experimental double-excitation cross sections from the ejected electron spectra or to the inaccuracy in the present limited coupled-state calculation where single-ionization channels were not included in the basis set and the convergence of the calculation for each system was not fully explored.

We next analyze the calculated cross sections to establish the mechanisms of double excitations at high velocities. The entries in Table II also contain calculations for antiprotons and two fictitious antiparticles with charge $Z=-2$ and -6. The cross sections in parentheses are from the truncated calculations where $1s2s\,{}^1S^e$ and $1s2p\,{}^1P^o$ singly excited states are not included in the basis set. The remaining goal of this paper is to analyze these calculated results in terms of the first-order and second-order mechanisms, bearing in mind that the first-order process is characterized by the Z_p^2 dependence in the integrated cross section. We emphasize that in our analysis first-order or second-order processes are defined with respect to the electron-projectile interactions. The so-called TS-1 where doubly excited states are populated once through electron-projectile interaction with a subsequent electron-electron interaction, has been called a two-step process in the literature.[1] In our formulation, TS-1 is considered to be included in the first-order process which cannot be separated from the so-called shakeup (or shakeoff for ionization) process since there is no well-defined mathematical procedure of separating the two amplitudes. We stress that both shakeup and TS-1 processes give Z_p^2 dependence in the integrated cross sections. Similarly, in a second-order process the electron interacts with the projectile twice, thus resulting in cross sections which vary like Z_p^4. If the amplitudes from the first-order and second-order processes are comparable, then the interference will contribute a Z_p^3 term.

The identification of the relative importance of the second-order mechanism versus the first-order mechanism can be carried out in different ways. First, if the second-order mechanism is important, the cross sections

TABLE II. Double-excitation cross sections (in units of 10^{-19} cm^2) of He at 1.5 and 6 MeV/u obtained from the coupled-channel calculations. Quantities in parentheses are from the truncated set where singly excited states were not included in the basis set.

Z	$2s^2\,{}^1S^e$	$2p^2\,{}^1D^e$	$2s2p\,{}^1P^o$	$2p^2\,{}^1S^e$
At 1.5 MeV/u				
1	0.074(0.071)	0.048(0.034)	0.30(0.30)	0.045(0.039)
2	0.32(0.27)	0.36(0.17)	1.26(1.14)	0.22(0.15)
4	1.56(0.99)	3.80(1.09)	6.00(3.87)	1.36(0.55)
6	5.16(2.06)	15.6(3.50)	16.2(6.86)	4.86(1.08)
9	19.8(4.4)	60.3(10.9)	51.3(9.47)	18.9(2.1)
−1	0.073(0.074)	0.027(0.022)	0.33(0.32)	0.041(0.040)
−2	0.294(0.30)	0.186(0.083)	1.47(1.29)	0.19(0.16)
−6	2.52(2.37)	9.55(1.43)	24.0(11.9)	3.77(1.41)
At 6.0 MeV/u				
1	0.020(0.018)	0.0098(0.0089)	0.144(0.144)	0.0125(0.012)
2	0.084(0.081)	0.052(0.040)	0.59(0.58)	0.053(0.049)
4	0.35(0.33)	0.38(0.20)	2.48(2.30)	0.24(0.19)
6	0.89(0.75)	1.5(0.59)	6.1(5.1)	0.72(0.42)
9	2.45(1.75)	6.20(1.93)	15.5(10.8)	2.30(0.88)
−1	0.020(0.020)	0.078(0.0076)	0.14(0.14)	0.012(0.012)
−2	0.079(0.080)	0.035(0.029)	0.588(0.572)	0.050(0.048)
−6	0.729(0.710)	0.823(0.277)	6.23(5.08)	0.65(0.43)

calculated with or without the inclusion of singly excited states in the coupled-channel calculation should be quite different. This is understood as follows. In the limit that the two electrons can be approximated as independent, a second-order mechanism for double excitation to $2s^2$, for example, can be perceived as two successive one-electron transitions, $1s^2 \rightarrow 1s2s \rightarrow 2s^2$, via electron-projectile interactions at each collision. (Note that other intermediate states would need two-electron transitions in the second collision, possible only via electron-electron interactions.) In other words, the intermediate $1s2s$ state is essential in the second-order mechanism for the population of $2s^2$ if the electron correlation effect is neglected. If the inclusion or the removal of singly excited states in the coupled-channel calculation shows no effects on the calculated double-excitation cross sections, the second-order mechanism can be considered not important. Second, if the second-order mechanism is not important, then the cross sections for particles and antiparticle projectiles should be the same. With these two criteria in mind, we note from Table II that the first-order mechanism appears to dominate for $|Z_p|=1-2$ at 1.5 MeV/u and for $|Z_p|=1-4$ at 6 MeV/u to better than a few percent. In all cases, the $2p^2\,{}^1D^e$ state shows the largest deviation from the prediction of the first-order mechanism.

We can also determine whether the mechanism is primarily first order or not by examining the Z_p dependence of the excitation cross sections. To this end, we define $\bar{\sigma}(Z_p)=\sigma(Z_p)/Z_p^2$, and the ratio $\gamma=\bar{\sigma}(Z_p)/\sigma(Z_p=1)$. If Z_p^2 scaling is exact, then γ is unity. In Table III we show the γ for each doubly excited state calculated for $Z_p=1-4$. We note that at each energy the ratios γ for $2s^2\,{}^1S^e$, $2s2p\,{}^1P^o$, and $2p^2\,{}^1S^e$ states stay reasonably close to unity, but for $2p^2\,{}^1D^e$ the deviation is much larger, indicating that the second-order or other high-order effects are relatively more important for this state. Among the four states, the ratio γ for the optically allowed $2s2p\,{}^1P^o$ state is closest to unity. At 6 MeV/u the γ for each state is in general closer to unity than at 1.5 MeV/u, indicating that the first-order mechanism becomes even more dominant at higher energies.

As the projectile charge increases, the second-order mechanism and other higher-order effects become more important if the velocity stays fixed. This can be easily seen by noting that in Table II the cross sections calculated with or without the inclusion of singly excited states at 1.5 MeV/u are drastically different for higher Z_p projectiles, while such differences are smaller at 6 MeV/u. Since the cross sections for particle and antiparticle impacts are different, one cannot conclude that the second-order mechanism alone is responsible for the double excitations. We conclude that at the energies studied both the first-order and second-order, and possibly higher-order, mechanisms are important for excitations to doubly excited states. Only at much higher energies will the first-order mechanism become dominant for these higher-Z_p ions.

We also note that from Table II, double-excitation cross sections by negatively charged particles are larger than by positively charged particles, except for the $2s2p\,{}^1P^o$ state where the opposite is true. This is to be compared with double ionization where the negative charged particles have been shown to have larger cross sections.[1,2]

B. Impact-parameter dependence

We next analyze the impact-parameter dependence of the calculated results. In Fig. 1 we show $bP_i(b)$ versus b, where b is the impact parameter and $P_i(b)$ is the excitation probability at 1.5 MeV/u. The projectiles are F^{9+} in Fig. 1(a). In Fig. 1(b) the projectiles are protons, but antiproton results for $2s2p\,{}^1P^o$ are shown as well. For comparison, single-excitation probabilities by protons are given in Fig. 1(c). Two features are obvious from these graphs: (i) In Fig. 1(b) the range of b for $2s2p\,{}^1P^o$ is much larger than for $2s^2\,{}^1S^e$, and (ii) comparing Fig. 1(a) with Fig. 1(b), the range of b for $2s^2\,{}^1S^e$ stays about the same, while for $2s2p\,{}^1P^o$ and $2p^2\,{}^1D^e$ both appear to shift to smaller b for higher Z_p. This second feature is particularly surprising since one would intuitively expect that the range of b for excitations to extend further out for higher-Z_p projectiles. We will note, however, that this is consistent with the observation made earlier that double excitation is dominated by the first-order mechanism for small Z_p and by the second-order mechanism for higher Z_p, as explained further below.

The different ranges of impact parameters in Fig. 1(a) for the different states can be explained by a second-order mechanism. Based on the calculated $P_{1s-2s}(b)$ and $P_{1s-2p}(b)$ for single excitations to $1s2s\,{}^1S^e$ and $1s2p\,{}^1P^o$ states, we can estimate that the $P(b)$ for $2s^2\,{}^1S^e$ is ap-

TABLE III. Check of Z_p^2 dependence of double-excitation cross sections at equivelocity collisions for low-Z_p projectiles. The entries give the parameter $\gamma=\bar{\sigma}(Z_p)/\bar{\sigma}(Z_p=1)$ where $\bar{\sigma}(Z_p)=\sigma(Z_p)/Z_p^2$. If each doubly excited state is populated by the first-order electron-projectile interaction, γ will be unity for each Z_p.

State	$2s^2\,{}^1S^e$		$2p^2\,{}^1D^e$		$2s2p\,{}^1P^o$		$2p^2\,{}^1S^e$	
E (MeV/u)	1.5	6.0	1.5	6.0	1.5	6.0	1.5	6.0
Z_p/γ								
1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
2	1.08	1.05	1.88	1.32	1.05	1.02	1.22	1.06
3	1.26		3.20		1.11		1.46	
4	1.32	1.09	4.95	2.42	1.25	1.08	1.89	1.20

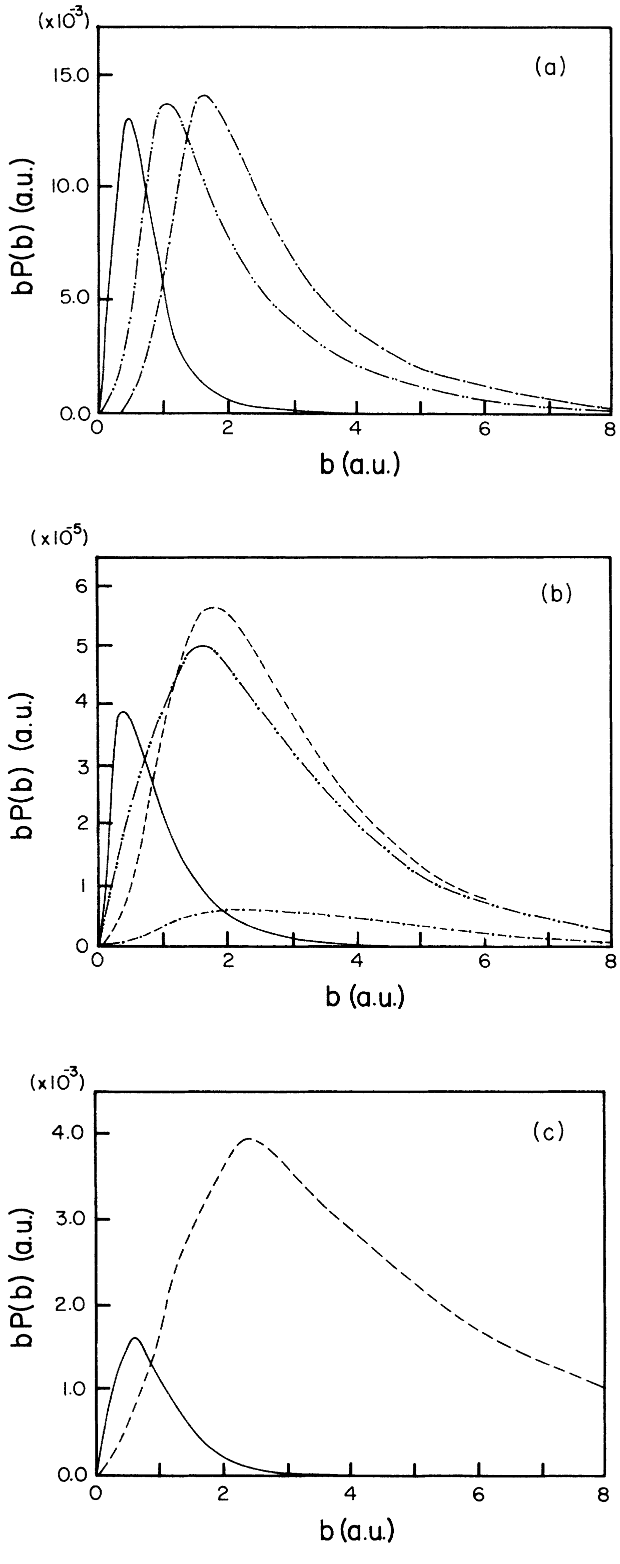

FIG. 1. Impact-parameter weighted excitation probabilities of helium atoms by bare projectiles at 1.5 MeV/u vs impact parameters: (a) double excitations by F^{9+} impact: $2s^2\,{}^1S^e$ (solid lines), $2s2p\,{}^1P^o$ (dash-dot-dot lines), and $2p^2\,{}^1D^e$ (dash-dot lines). (b) Similar to (a) but by protons. Double excitations by antiprotons to $2s2p\,{}^1P^o$ are shown in dashed lines. (c) Single excitations to $1s2s\,{}^1S^e$ (solid lines) and to $1s2p\,{}^1P^o$ (dashed lines) by protons.

proximately given by $P^2_{1s-2s}(b)$, and for $2s2p\,{}^1P^o$ and $2p^2\,{}^1D^e$ are approximately $P_{1s-2s}(b)P_{1s-2p}(b)$ and $P^2_{1s-2p}(b)$, respectively. Since the range of $P_{1s-2s}(b)$ is much smaller than the range of $P_{1s-2p}(b)$, the range for the products is smallest for $2s^2\,{}^1S^e$ and largest for $2p^2\,{}^1D^e$, with $2s2p\,{}^1P^o$ in between. Using the actual $P_{1s-2s}(b)$ and $P_{1s-2p}(b)$ calculated, we have checked that the shape and the range of Fig. 1(a) are indeed reproduced by this model, while the actual magnitudes are a factor of about 3 too high. The overestimate in the magnitude is easily understood. In the second-order model, the first collision from $1s^2\,{}^1S^e$ to $1s2s\,{}^1S^e$ has an excitation energy of 20.6 eV, while the second collision from $1s2s\,{}^1S^e$ to $2s^2\,{}^1S^e$ involves an excitation energy of 37.3 eV. The estimate based on the simple product of single-excitation probabilities does not account for this increasing excitation energy in the second step and thus tends to overestimate the cross sections. Such an estimate also does not account for electron correlation in the wave functions.

We next interpret Fig. 1(b). From the Z_p^2 scaling of the total cross sections, we have concluded that doubly excited states in this case are populated primarily by the first-order process, except for $2p^2\,{}^1D^e$. If this is the case, then the $bP(b)$ dependence for $2s^2\,{}^1S^e$ should be similar to that for $1s2s\,{}^1S^e$ since each involves a single electron-projectile interaction for exciting from $1s$ to $2s$. Similarly, this should hold true for $2s2p\,{}^1P^o$ and $1s2p\,{}^1P^o$ since each involves a single electron-projectile interaction for exciting from $1s$ to $2p$. This is precisely the case. In Fig. 1(c) we show that $bP(b)$ for single excitations by protons to $1s2s\,{}^1S^e$ and $1s2p\,{}^1P^o$ states do have the same shape as the $bP(b)$ for $2s^2\,{}^1S^e$ and $2s2p\,{}^1P^o$, respectively. We also show the $2s2p\,{}^1P^o$ state populated by antiprotons in Fig. 1(b). The peak of $bP(b)$ in this case shifts slightly further out, similar to the curve (not shown) for the $1s2p\,{}^1P^o$ single excitation by antiprotons.

In the first-order mechanism, the projectile interacts once with one of the electrons; the other electron can be excited through the shakeup process or through the so-called TS-1 process where the other electron is excited through the electron-electron interaction. As stated earlier, in our formulation we cannot separate uniquely these two processes since shakeup is a change of screening, also due to the electron-electron interaction. For our purpose here, we call the excitation of the second electron due to the electron-electron interaction a shapeup process. To estimate the shakeup probability, we calculate the ratio $S_1=P(2s2s)/P(1s2s)$ and $S_2=P(2s2p)/P(1s2p)$, where $P(i)$ is the probability for excitation to state i at each impact parameter b. In the present calculations, the shakeup probabilities S_1 and S_2 are of the order of a few percent (1–5 %) and larger than the single-excitation probabilities by protons at these energies. It is clear why the double-excitation cross section for $2p^2\,{}^1D^e$ state is small since a strict shakeup is not allowed from $1s$ to $2p$. The somewhat larger double-excitation cross section for $2p^2\,{}^1S^e$ is due to the configuration mixing of this state with the $2s^2\,{}^1S^e$ state.

We can also use the first-order perturbation theory to complement the qualitative interpretation in the preceding paragraph. The transition amplitude from the ground state $|\Psi_i\rangle$ to the doubly excited state $|\Psi_f\rangle$ is

$$\langle\Psi_f|-Z/r_{1p}-Z/r_{2p}|\Psi_i\rangle \ . \tag{3}$$

To define each electronic orbital uniquely for both the initial and final states, we construct, in this consideration, the helium ground and doubly excited states using products of hydrogenic wave functions with $Z=2$. The screening is accounted for by introducing configuration-interaction wave functions. For the ground state we obtained

$$|\Psi_i\rangle=0.95|1s^2\rangle-0.28|1s2s\rangle-0.10|1s3s\rangle -0.03|2p2p\rangle \ . \tag{4}$$

For $2s2p\ ^1P^o$,

$$|\Psi_f\rangle=0.865|2s2p\rangle-0.19|2s3p\rangle-0.31|2p3s\rangle -0.34|2p3d\rangle \ , \tag{5}$$

and for $2p^2\ ^1D^e$,

$$|\Psi_f\rangle=0.865|2p2p\rangle+0.28|2s3d\rangle+0.07|2s4d\rangle -0.39|2p3p\rangle \ . \tag{6}$$

If we use these wave functions into (3), the nonzero contribution to the first-order term comes from configurations of initial and final states that differ by one orbital only. The resulting dominant first-order matrix element to the $2s2p\ ^1P^o$ state is

$$M=-0.21\langle 1s|T|2p\rangle+0.05\langle 1s|T|3p\rangle \tag{7}$$

and to the $2p^2\ ^1D^e$ is

$$M=-0.08\langle 1s|T|3d\rangle-0.02\langle 1s|T|4d\rangle \ , \tag{8}$$

where $T=-Z_p/r_p$. The results for the two $^1S^e$ states can be similarly obtained.

The expression above explains the results shown in Fig. 1(b). At high energy, the dominant single excitation is the optically allowed 1*s*-2*p* transition. This is a dipole transition and extends over a large range of *b*, as shown in the single excitation $bP(b)$ in Fig. 1(c). For $2p^2\ ^1D^e$, the first-order probability is small, partly due to the small quadrupole transition amplitude from 1*s* to 3*d* in Eq. (8) and partly because of the small coefficient. For $2s^2\ ^1S^e$, we would have a term from the smaller 1*s*-2*s* monopole transition which also extends to small *b* only.

C. Comparison with double ionizations

It is interesting to compare the ratio $R_{\rm exc}$, of double-excitation to single-excitation cross sections, with respect to the ratio $R_{\rm ion}$, of double ionization with respect to single ionization, particularly between proton and antiproton projectiles. At 1.5 MeV/u, $R_{\rm ion}=0.6\%$ for antiprotons and 0.25% for protons where the difference is more than a factor of 2. For $R_{\rm exc}$ between double excitation to $2s2p\ ^1P^o$ with respect to $1s2p\ ^1P^o$, the ratio is 0.85% for antiprotons and 0.77% for protons. The same ratio for double excitation to $2s^2\ ^1S^e$ with respect to $1s2s\ ^1S^e$ is 2.6% for antiprotons and 2.2% for protons. In both double-excitation cases, the difference between protons and antiprotons is of the order of 15% or less.

Large interference in two-electron transition probabilities occurs when the first-order and second-order mechanisms are of comparable importance, as shown by the excitation to $2p^2\ ^1D^e$ by protons and by antiprotons (see Table II), where the difference between particle and antiparticle impact at 1.5 MeV/u is about a factor of 2, except that in this case protons give a larger cross section. The large difference in $R_{\rm ion}$ between protons and antiprotons can be similarly interpreted. It is expected that the first-order mechanism eventually will dominate at very high energies. The fact that double excitation to $2p^2\ ^1D^e$ and double ionization by protons and antiprotons are so different is attributable to the relatively small contribution from the first-order mechanism. From Eq. (8) we note that the first-order matrix element for excitation to $2p^2\ ^1D^e$ contains no contribution from the 1*s*-2*p* transition even when "correlated" wave functions are used. From the conventional shakeup mechanism, the second electron cannot be excited from 1*s* to 2*p*. This explains why it has a much smaller cross section as compared to $2s2p\ ^1P^o$. Similarly, for double ionizations we note that according to Eq. (3), the first-order contributions have to come from the terms in the ground-state configuration-interaction (CI) coefficients for configurations that contain at least one continuum orbital. Such CI coefficients are expected to be small but they cannot be easily evaluated. To estimate the relative magnitude of this term, we evaluate instead the conventional shakeoff probability. If we assume that the initial He ground state is given by the product of two hydrogenic orbitals with an effective charge of 1.675, the shakeoff probability was evaluated to be about 0.9%. If we use Eq. (7) to estimate the shakeup probability for $2s2p\ ^1P^o$, it is about 4%. Thus the first-order mechanism is relatively smaller for double ionization and may be comparable to the second-order mechanism at 1.5 MeV/u, thus giving strong interference effect and a large difference in the cross sections between particle and antiparticle impacts.

IV. SUMMARY AND CONCLUSIONS

In conclusion, by analyzing the results of double-excitation cross sections from coupled-channel calculations for the collision of bare ions with He, we conclude that at 1.5 MeV/u, doubly excited states are primarily populated by the first-order electron-projectile interaction with a subsequent shakeup of the second electron. This is true for the $2s^2\ ^1S^e$, $2s2p\ ^1P^o$, and $2p^2\ ^1S^e$ states analyzed and thus the excitation cross sections for these states show approximate Z_p^2 scaling. For $2p^2\ ^1D^e$ we see evidence of interference from the second-order mechanism. For higher incident charge, such as F^{9+}, we observed that all the doubly excited states studied here are populated primarily by the second-order or higher-order mechanisms where double excitation is the result of at least two successive electron-projectile interactions. We conclude that if different projectiles at a fixed velocity are used, such as the experiments of Pedersen and Hvelplund and of Giese *et al.*, the second- and higher-order mechanisms will become dominant at higher Z_p. In general, the first-order mechanism is the dominant one at high collision velocity while the second- and higher-order mechanisms become important at lower collision velocities; the energy where the transition occurs depends on the states studied.

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[1] H. K. Haugen, L. H. Andersen, P. Hvelplund, and H. Knudsen, Phys. Rev. A **26**, 1962 (1982); L. H. Andersen, H. Hvelplund, H. Knudsen, S. P. Moller, K. Elsenser, K. G. Rensfelt, and E. Uggerhoj, Phys. Rev. Lett. **57**, 2147 (1986); L. H. Andersen, H. Hvelplund, H. Knudsen, A. H. Sorensen, S. P. Moller, K. Elsener, K. G. Rensfelt, and E. Uggerhoj, Phys. Rev. A **36**, 3612 (1987).

[2] J. F. Reading and A. L. Ford, Phys. Rev. Lett. **58**, 543 (1987); J. Phys. B **20**, 3747 (1987); A. L. Ford and J. F. Reading, *ibid.* **21**, L685 (1988).

[3] J. H. McGuire, Phys. Rev. A **36**, 1519 (1987).

[4] L. Vegh, Phys. Rev. A **37**, 992 (1988).

[5] R. Olson and T. J. Gay, Phys. Rev. Lett. **61**, 302 (1988).

[6] E. Y. Kamber, C. L. Cocke, S. Cheng, and S. L. Varghese, Phys. Rev. Lett. **60**, 2026 (1988).

[7] J. P. Giese and E. Horsdal, Phys. Rev. Lett. **60**, 2018 (1988).

[8] M. Mack, J. H. Jijland, P. van der Straten, A. Niehaus, and R. Morgenstern, Phys. Rev. A **39**, 3846 (1989).

[9] A. Jain, C. D. Lin, and W. Fritsch, Phys. Rev. A **39**, 1741 (1989).

[10] T. J. M. Zouros, D. Schneider, and N. Stolterfoht, Phys. Rev. A **35**, 1963 (1987), and references therein.

[11] W. Fritsch and C. D. Lin, Phys. Rev. Lett. **61**, 690 (1988), and references therein.

[12] J. O. P. Pedersen and P. Hvelplund, Phys. Rev. Lett. **62**, 2373 (1989).

[13] J. P. Giese, M. Schulz, J. K. Swensen, H. Schöne, M. Benhenni, S. L. Varghese, C. R. Vane, P. F. Dittner, S. M. Shafroth, and S. Datz (unpublished).

[14] W. Fritsch and C. D. Lin, J. Phys. B **19**, 2683 (1986).

[15] V. V. Balashov, S. S. Lipovetskii, and V. S. Senashenko, Zh. Eksp. Teor. Fiz. **63**, 1622 (1972) [Sov. Phys.—JETP **36**, 858 (1973)].

[16] B. W. Shore, Rev. Mod. Phys. **39**, 439 (1967).

[17] D. Burch, J. Bolger, and C. F. Moore, Phys. Rev. Lett. **34**, 1067 (1975).

[18] A. Bordenave-Montesquieu, P. Beniot-Cattin, M. Rodiere, A. Gleizes, and H. Merchez, J. Phys. B **8**, 874 (1975).

[19] See also W. Mehlhorn (unpublished).

[20] P. W. Arcuni and D. Schneider, Phys. Rev. A **36**, 3059 (1987).

[21] P. van der Straten, R. Morgenstern, and A. Niehaus, J. Phys. B **21**, 1573 (1988).