

State-selective double-electron capture in $\text{He}^{2+} + \text{He}$ collisions at intermediate impact energies

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We report calculations of cross sections for electron capture to individual doubly excited states for 150–500-keV $^3\text{He}^{2+}$ ions colliding with helium atoms. By adopting an independent-electron approximation, the capture probability to each doubly excited state is calculated in terms of single-capture amplitudes to each substate which are properly weighted with configuration-interaction coefficients. The validity of such an approximation is checked by comparing the results with calculations in which the electron correlation was considered explicitly. The calculated final results for double capture to the $(2s^2)^1S$, $(2s2p)^1P$, and $(2p^2)^1D$ states are in fair agreement with the recent experiment of Zouros *et al.* [Phys. Rev. A **35**, 1963 (1987)].

I. INTRODUCTION

Recently, Zouros *et al.*¹ have reported absolute cross sections for the production of doubly excited $(2s^2)^1S$, $(2p^2)^1D$, and $(2s2p)^1P$ states of helium by the double-electron-capture process in the 150–500-keV $^3\text{He}^{2+} + \text{He}(1s^2)$ collisions. These state-selective double-electron-capture (SSDEC) cross sections are measured using the technique of high-resolution zero-degree Auger electron spectroscopy.² Earlier studies on the state-selective double-electron capture for other colliding systems are limited to very low energies.^{3–8} For high-energy collisions often only the n distributions of the final states are measured.⁹

A straightforward yet much complicated theoretical method for treating two-electron processes is to solve the time-dependent wave functions for the two-electron or quasi-two-electron problems using the close-coupling method using either the atomic-orbital¹⁰ (AO) or molecular-orbital expansions.¹¹ For example, in the AO-expansion method, one would have to diagonalize the two-electron atom in truncated products of single-particle states and expand the time-dependent wave functions in terms of these states to solve the transition amplitudes. Such calculations have been carried out for a number of collision systems at the relatively lower energies^{10,12} where the number of final states populated in the collision is small. It has been applied to another collision system¹³ for transfer excitation processes where the role of electron correlation has been observed explicitly in the form of resonant transfer excitation (RTE) peaks. At the higher collision energies where inelastic processes become less selective, the question then arises how many channels have to be included in the calculation for reasonable accuracy of the results. This question becomes particularly severe if interest is in the “small” channels like capture into doubly excited states which are populated with relatively small cross sections. In the present work, we take the measurements by Zouros *et al.* as motivation for a study on whether double-electron capture to individual

projectile states at higher energies can be treated in some form of an independent-electron model.¹⁴ The latter model is clearly much preferable computationally and has been widely used in multiple processes for ionization,¹⁵ transfer ionization,¹⁶ and double capture to ground states.¹⁷ To obtain capture amplitudes to individual doubly excited states, the present model of the independent-electron approximation accounts for the electron correlations in the final states, i.e., the independent-electron model is applied only during the time evolution of the collision.

In the present calculations we evaluate the single-electron-capture amplitude using the modified two-center atomic orbital expansion model (AO+).¹⁸ This model, when augmented with united-atom (UA) orbitals or pseudostates, has been shown to provide accurate single-electron-capture cross sections for a wide range of (quasi-) one-electron collision systems in the intermediate energy region.^{18–23} Together with a suitable combination of configuration-interaction coefficients, from these amplitudes we can derive double-electron-capture amplitudes to individual states (Sec. II). Calculations based on such a model are then compared with experiments of Zouros *et al.*

At two selected energies, we also performed calculations of double-electron capture within the two-electron version of the close-coupling model with AO+ basis sets.¹⁸ In this calculation, the full Hamiltonian of the two-electron system is included, and the basis is chosen in close analogy to the basis choice in the independent-electron model. Therefore comparison of results from both model calculations allows for an estimate of the validity of the independent-electron model while limitations of the basis-set choice should affect both calculations in similar ways. Comparison to experiment should then indicate whether the size of the chosen basis set is sufficient.

Other calculations on the state-selective double-electron-capture cross sections for this system and energy range are the continuum-distorted-wave (CDW) (in the

independent-particle approximation) model by Salin *et al.*²⁴ In their calculations, Salin *et al.* employed a form of the independent-electron model similar to the present one, i.e., the double-capture probability amplitude to each doubly excited state is expressed as suitable linear combinations of products of two single-electron-capture amplitudes which are evaluated using the CDW model. These calculations are in large discrepancy (within a factor between 6–75) with the measurements of Zouros *et al.* above 200 keV. Below 200 keV the discrepancy is even larger. Since the CDW theory is basically a high-energy approximation which is known to break down in the intermediate-energy region where the experiment was carried out, it is not clear to what extent the discrepancy between the experiment and their calculations is due to the limitation of the independent-electron model or to the CDW theory for the single-electron-capture process.

There are a number of possible *variants* of the independent-particle model for applications to multiple processes. Assuming there is no correlation and that there is no change of screening after the first electron is captured, we can obtain the double-capture cross section from the single-electron-capture probability $P(b)$,

$$\sigma_{20} = 2\pi \int P^2(b) b db . \quad (1)$$

If we assume that the second electron has relaxed and thus there is a change in screening before it is captured, then there is a change of binding energy for the second electron from -0.904 to -2.0 a.u. and hence a different capture probability $P'(b)$, from the remaining target He^+ ion. In this model, the double-capture probability is calculated by employing the relation

$$\sigma_{20} = 2\pi \int P(b)P'(b) b db . \quad (2)$$

In Sec. II, we provide a brief description of the present theory and the results are discussed in Sec. III. Concluding remarks are made in Sec. IV.

II. THEORY

A. Independent-electron model

In the independent-electron approximation, if $a_i(b)$ is the scattering amplitude corresponding to the i th state, then the total cross section for double-electron capture is determined from $P^2(b)$, where $P(b) = |a_i(b)|^2$. This simple expression is valid for double capture to the ground state ($1s^2\ ^1S$) or double capture to singly excited states.

The situation for doubly excited states is somewhat different. It is well known that doubly-excited-state wave functions are not adequately represented by the product of two simple one-electron orbitals. Consider the $(2l, 2l')$ doubly excited states formed in a collision. In the independent-electron model the electronic wave function for the first electron in the $n=2$ subspace after the electron capture is given by

$$\psi_1(\mathbf{r}_1; b) = \sum_{l,m} a_{2lm}(b) \phi_{2lm}(\mathbf{r}_1) , \quad (3)$$

where $\phi_{nlm}(\mathbf{r})$ are hydrogenic orbitals and the sum is over

all allowed l and m values for $n=2$. A similar expression can be written for the second electron after the capture. The two-electron wave functions are the symmetrized products (for singlet states) of single-electron wave functions $\psi_1(\mathbf{r}_1; b)$ and $\psi_2(\mathbf{r}_2; b)$,

$$\psi(1, 2) = \frac{1}{\sqrt{2}} [\psi_1(\mathbf{r}_1; b)\psi_2(\mathbf{r}_2; b) + \psi_1(\mathbf{r}_2; b)\psi_2(\mathbf{r}_1; b)] . \quad (4)$$

To obtain the scattering amplitude $A(b)$ for each doubly excited state within the $2l2l'$ configuration, we reexpand (4),

$$\psi(1, 2) = \sum_{L, M} A_{LMS}(b) \phi_{LMS}(\mathbf{r}_1, \mathbf{r}_2) , \quad (5)$$

in terms of the two-electron eigenfunctions $\phi_{LMS}(\mathbf{r}_1, \mathbf{r}_2)$. We assume that configuration interaction (CI) within the intrashell ($n=2$) is included, i.e.,

$$\phi_{LMS}^{n=2}(\mathbf{r}_1, \mathbf{r}_2) = \sum_i c_i \Phi^{n=2}(1, 2) , \quad (6)$$

where

$$\Phi^{n=2}(1, 2) = |nl_1 nl_2 LMS\rangle$$

is the *symmetrized* two-electron product wave function.

It is now straightforward to derive amplitudes $A_{LMS}(b)$ as

$$A_{LMS}(b) = \sqrt{2} \sum_i c_i \sum_{\substack{l_1, m_1 \\ l_2, m_2}} C_{m_1 m_2 M}^{l_1 l_2 L} a_{2l_1 m_1}(b) a_{2l_2 m_2}(b) , \quad (7)$$

where the C 's are the Clebsch-Gordan coefficients. The double-electron-capture cross section for each doubly excited state with usual quantum numbers L and M and spin $S=0$ is then obtained from the amplitude A_{LMS} .

For the $n=2$ shell, each intrashell excited state is represented by a single configuration except for $2s^2$ and $2p^2\ ^1S$ states. Introducing CI, we can write the correlated states (denoted by a bar over the symbol) in terms of uncorrelated ones as

$$^1S; |\bar{2s^2}\rangle = \alpha |2s^2\rangle + \beta |2p^2\rangle , \quad (8)$$

and

$$^1S; |\bar{2p^2}\rangle = -\beta |2s^2\rangle + \alpha |2p^2\rangle , \quad (9)$$

where the coefficients α and β are calculated to be 0.8796 and 0.4756, respectively, for helium.

B. Coupled-channel atomic orbital expansion method

The coupled-channel atomic-orbital expansion method was used to calculate the transition amplitudes. In the independent-electron approximation, we treat the two electrons independently. Each electron evolves independently in the field of the target and the projectile poten-

tials. The electron-target interaction is described by a model potential which gives the correct ionization energy of the ground state of helium. This AO+ model for one-electron systems has been discussed previously.¹⁸ The active electron's time-dependent wave function is expanded in terms of traveling atomic and pseudostate orbitals on both centers. This gives rise to a set of first-order coupled differential equations for the expansion coefficients which are solved to obtain the scattering amplitudes for each single particle state $i = nlm$ at each impact parameter and energy.

The present AO+ basis set for the $\text{He}^{2+} + \text{He}(1s^2)$ system consists of a total number of 19 states (12 states on the projectile, 10 atomic orbitals for $n = 1, 2$, and 3, and 2 $1s$ UA's; 7 states on the target, i.e., 2 $1s$ orbitals describing He ground state, 3 $n = 2$ AO's representing excited channels of the target and two UA orbitals, same as on the projectile). The UA orbitals give the continuum pseudostates.

Two versions of the independent-electron approximation were investigated. In the first model, we assume that the second electron is completely equivalent to the first one. Thus the wave function ψ_1 in Eq. (3) applies to both electrons. The state-selective double-electron capture (SSDEC) cross sections thus calculated are denoted as SSDEC (1). In another model, we assume that the second electron is fully relaxed after the first electron has been captured. In other words, the second electron sees a He^{2+} potential. With the target wave functions properly modified, a similar AO+ calculation was carried out to obtain the transitions amplitudes. The resulting double-capture cross sections are denoted as SSDEC (2).

To assess the validity of the independent-electron approximation in the formulation described above, we have also performed calculations within the two-electron version^{10,12} of the AO-expansion method with full account of the electron-electron interaction. In these calculations, the He target ground state and the 1S and 1P first excited states are represented by four symmetrized products of hydrogenic wave functions $(n_1 l_1 m_1 Z_1)(n_2 l_2 m_2 Z_2)$, where Z_1 and Z_2 are fixed by minimizing the respective electronic energy of each state. One-electron capture is represented by a set of ten similar product states, with the target electron residing in the $1s$ He^+ orbital and the projectile electron in the $n = 1-3$ He^+ orbitals at the projectile center, or the captured electron in $1s$ He^+ and the target electron excited to the $n = 2$ He^+ orbitals. Finally, double-electron capture to the ground state and to the singly excited states is represented by four projectile-centered product states similar to those given above for target excitation, while capture to doubly excited states is represented by another nine states which are of type $(2s2s')$, $(2s2p)$ and $(2s2p')$. For the $(2p2p')$ states, configurations with total angular momentum 0 and 2 have been constructed. With this basis set of two-electron configurations, it is believed that the two types of calculations for two-electron processes, within the one-electron and within the two-electron model, start from similar assumptions about the importance of channels for the considered processes. The comparison of results between the two model calculations may then allow

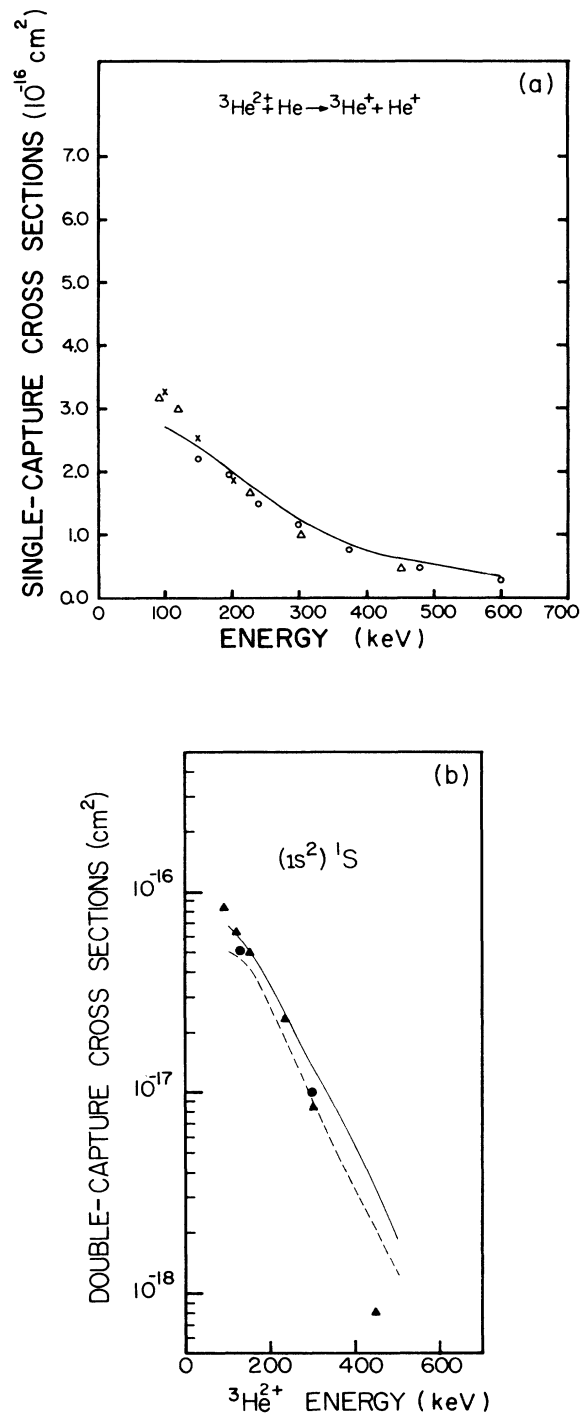


FIG. 1. (a) Total cross sections for the $\text{He}^{2+} + \text{He}(1s^2) \rightarrow \text{He}^+ + \text{He}^+$ collisions at 100–600-keV impact energies. Present one-electron calculations are shown by the solid curve. Experimental points: crosses, DuBois (Ref. 32); open circles, Shah and Gilbody (Ref. 35); open triangles, Rudd *et al.* (Ref. 34). (b) Total cross sections for the double-electron capture into the projectile ground state [$\text{He}^{2+} + \text{He}(1s^2) \rightarrow \text{He}(1s^2 \ ^1S) + \text{He}^{2+}$]. Present one-electron model calculations: solid curve, using Eq. (2); dashed curve, using Eq. (2); Results from the two-electron model are shown by closed circles. Experiment: triangles, Ref. 34.

some conclusion about the validity of the one-electron model, while comparison to experiment may show whether the basis sets chosen are adequate.

III. RESULTS

A. Total Single-Electron-Capture and Ground-State Double-Electron-Capture Cross Sections

To check the accuracy of our single-capture amplitudes, we first compare our results on the total single-electron-capture cross sections with the measured values. Then we shall compare the ground-state double-capture cross sections in the independent- and two-electron models to the experimental data. There are several measurements²⁵⁻³⁵ and calculations³⁶⁻³⁹ on the total single-electron capture and ground-state double-electron capture in the keV energy region. In Fig. 1(a) the calculated single-electron-capture cross sections (sum over all states with $n=1, 2,$ and 3) from the one-electron model are

shown to compare well with the recent experimental results of Dubois,³² Rudd *et al.*,³⁴ and Shah and Gilbody.³⁵ We remark that the model potential was chosen to represent the one-electron binding energy and the agreement between the calculated and the experimental cross sections for single-capture process is an indication of the adequacy of the independent-electron model and the coupled-channel calculations.

In Fig. 1(b) we show the integrated cross section for double capture to the ground state. The solid curve is obtained by assuming that the two electrons are completely equivalent [Eq. (1)], while the dashed line is the result of assuming that the second electron is captured from a more tightly bound He^+ core [Eq. (2)]. At two energy points, we have also depicted (closed circles) the corresponding results of the two-electron calculations. The experimental data of Rudd *et al.*³⁴ is also illustrated in Fig. 1(b). The results shown in Fig. 1(b) seem to indicate that the assumption of capturing the second electron from a more tightly bound He^+ core (where the second electron has relaxed) is the better model within the one-electron description in the lower-energy region while capturing from two equivalent electrons appear to work better at higher energies. The agreement between the theory and experiment can be considered satisfactory in view of the simplicity of the model.

Figure 1(b) also shows that the two-electron results do not differ significantly from the results of the simpler independent-electron-model calculations. This is further supported by examining the impact-parameter dependence of the capture probability. In Fig. 2(a) we show that the impact-parameter-weighted probabilities at 300 keV calculated using the independent-electron model are in good agreement with those calculated with the two-electron model where electron correlation was included. [The one-electron version was calculated with a tighter second electron, i.e., $P'(b)$.] Similar agreement was observed also at 150 keV.

B. Double-Electron Capture into the $2l2l'$ States of He

We now discuss the cross sections for the formation of $2l2l'$ doubly excited states of the projectile at impact energies of 100–500 keV. In this manifold, a total of six different singlet two-electron states, namely,

$$2s^2\ ^1S; \ 2p^2\ ^1S; \ 2s2p\ ^1P_0; \ 2s2p\ ^1P_{\pm 1}; \\ 2p^2\ ^1D_0; \ 2p^2\ ^1D_{\pm 1}; \ 2p^2\ ^1D_{\pm 2}$$

(where the subscript denotes the total magnetic quantum number) can be populated.

We have calculated the cross sections to each of these doubly excited states. In order to compare with experiment, we note that the zero-degree Auger electron spectroscopy can measure only the cross sections of the $M=0$ components (quantization axis is in the incident beam direction) and thus only the $M=0$ component from the calculations are compared with experiments.

Figure 3 provides a comparison of our calculated state-selective double-electron-capture cross sections

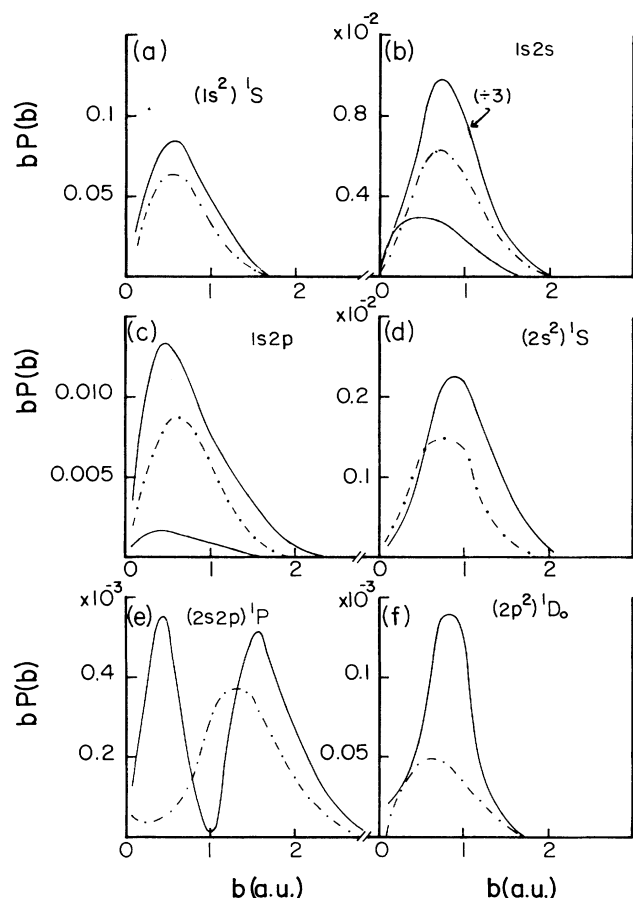


FIG. 2. Calculated weighted probabilities [$bP(b)$] for double-electron capture to the ground [(a)], singly excited [(b) and (c)] and doubly excited states of helium in ${}^3\text{He}^{2+} + \text{He}(1s^2)$ collisions at 300-keV incident energy. The dot-dashed curves are from the two-electron model. The solid curves are from various models using the independent-particle approximation (see text).

based on the independent-electron model [both SSDEC (1) and SSDEC (2)] with those of the measurements in Ref. 1. The CDW results²⁴ are not shown as they are far off the scale. From Fig. 3 we note that the results of the SSDEC (2) model are in better agreement with the trend of the data by Zouros *et al.* than the results of the SSDEC (1) model, indicating that improvement resulting from the consideration of the relaxation of the second electron. On the other hand, there is still large quantitative discrepancy with the data, with the calculated cross sections being about 2–8 times larger than the measured ones.

In Fig. 3 we also show results of calculations at 150 and 300 keV obtained using the two-electron close-coupling code, with basis set chosen as indicated earlier. The two-electron calculations give better agreement with experiments to within a factor of 2 for 1P and 1D ($M=0$) levels but it seems to miss the trend of the data for 1S . We note that the results of the one-electron and the two-electron calculations are quite close at 300 keV but rather different at 150 keV. This confirms the expectation that the one-electron model for two-electron processes should work better for higher energies, while at lower energies some explicit account of correlation is needed.

The discrepancies in Fig. 3 illustrate the difficulties of obtaining accurate SSDEC cross sections from *ab initio* calculations and the limitations of the independent electron model. (We stress that both models work quite well for the single-electron capture and double capture to the ground state, see Fig. 1.) This difficulty is mainly due to the smallness of the cross sections involved. We note that double-electron capture to doubly excited states for the present collision system is more than *two* orders of magnitude smaller than the double capture to the ground state. Such small cross sections are sensitive to the convergence of the basis set used in the coupled channel calculations and to the independent-electron model.

The discrepancy between the present two-electron and the one-electron calculations is mostly due to the limitation of the independent-electron approximation. In Figs. 2(b)–2(f) we further show the comparison of the impact-parameter dependence of the various singly and doubly excited capture probabilities (due to double capture) at 300 keV. In the one-electron version we have used the SSDEC (1) model where the relaxation of the second electron was not considered. The discrepancy between the one-electron and the two-electron model is not small, particularly for $2s2p\ ^1P$, where both the overall magnitude and the shapes are quite different. Such discrepancies are much larger than those shown in Fig. 2(a) for the probabilities for double capture to the ground state.

Accurate calculations of small cross sections for weak channels using the independent-electron model are difficult since the results are relatively sensitive to the atomic models used for each electron. At the end of the introduction we addressed the relaxation of the remaining target electron after the first electron was captured. A similar question can be raised for the screening of the projectile nucleus after the first electron has been captured. One can use different effective charge for the projectile in the calculation of $P(b)$ for the second electron.

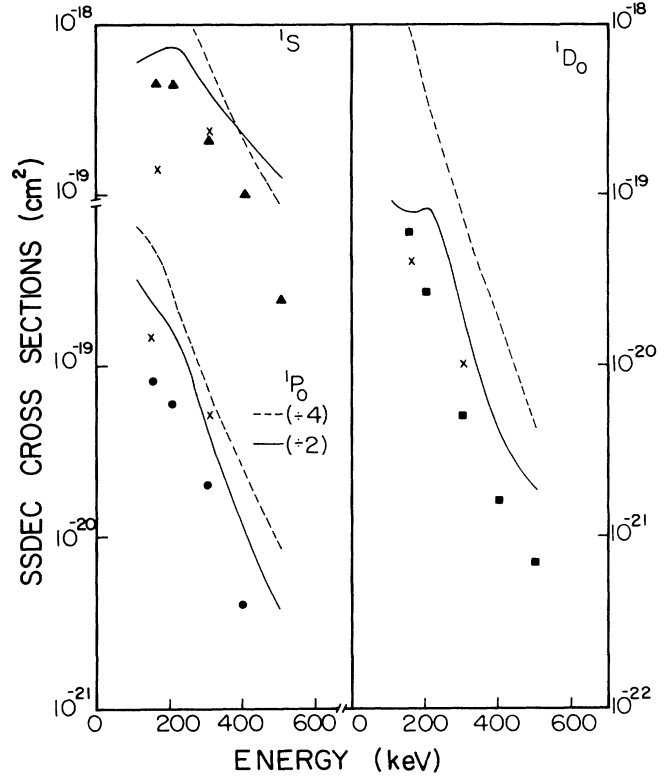


FIG. 3. State-selective double-electron-capture cross sections (for 1S , 1P_0 , and 1D_0 states) in the collision process $^3\text{He}^{2+} + \text{He}(1s^2) \rightarrow ^3\text{He}(2l2l') + \text{He}^{2+}$ at 100–500-keV energies. The theoretical curves (solid and dash) are determined respectively in the SSDEC (2) and SSDEC (1) models (for notation see the text). The results from the two-electron model are shown by crosses. The experimental data (closed symbols) are taken from Zouros *et al.* (Ref. 1).

This is illustrated in the probabilities for double capture to singly excited states, as shown in Figs. 2(b) and 2(c). In the calculation, we assumed that the second target electron was not relaxed, but two effective charges were used for the projectile in calculating $P'(b)$ for the second electron. In one model (upper solid lines) we assume that the projectile charge is 2, and that the first electron does not screen the projectile nucleus after the first electron was captured. In another model (lower solid lines), the charge for the projectile was reduced to 1, assuming screening by the first captured electron. The large discrepancy between the two calculations shows the sensitivity of the results to the models used in the calculation of probabilities in the independent particle approximation. Comparison with results from the two-electron code does not clearly favor one model or another.

IV. CONCLUSION

We reported single- and double-capture cross sections for the $^3\text{He}^{2+} + \text{He}$ system at 100–500-keV impact energy using two-center atomic-orbital expansion models and examined the validity of the independent-particle approximation. For the dominant processes such as total single-

capture cross sections to the ground state, the calculated results agreed with experimental data and the independent-particle model was shown to be valid. We also studied double capture to doubly excited states and compare the results with the recent data of Zouros *et al.*

It is shown that calculations based on the two-electron coupled-channel calculation gave reasonable agreement with experiment. For calculations carried out using the independent-particle model we found large discrepancies with experiments and with the results from the two-electron code. The results show that the great success of the independent-particle model for the cross sections of dominant channels cannot be extended to the evaluation of the cross sections of the weaker channels. The results further indicate that two-electron close-coupling calculations are needed for obtaining two-electron transitions such as the state-selective double-electron capture cross

sections studied here. (Another type of two-electron transitions, the transfer-excitation processes, were studied in Ref. 13.) In view of the large amount of computer time needed for such calculations, applications of such calculations to many systems are impractical at present. On the other hand it is not clear whether the limited validity of the independent-particle approximation is the consequence of the smallness of the charge of the projectiles used in this investigation. Similar studies with multiply charged ions are needed to answer this question.

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