

Double K -shell electron capture for ion-atom collisions at intermediate energies

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A three-state, two-center atomic expansion method is used to investigate double-electron capture from the K shell of multielectron atoms to the K shell of projectiles in the energy region where the projectile velocity is nearly equal to the orbital velocity of the target K -shell electron. By adopting an independent-electron approximation, the Coulomb interaction between the two electrons is replaced by monopole terms with a screening charge. The validity of this approximation is checked by comparing the cross sections for double capture in p -He and He^{2+} -He collisions obtained from this method with other theoretical calculations and experiments. Double-electron-capture cross sections from the K shell of Ne atoms by F^{9+} , O^{8+} , and N^{7+} ions are then calculated; the results are shown to compare well with the neon Auger hypersatellite intensities measured by Woods *et al.*

I. INTRODUCTION

Single- and multiple-electron transfer cross sections in ion-atom collisions have been studied experimentally over a wide energy range throughout the years. A great deal of experimental data has been accumulated in the higher-energy region in conjunction with the study of the penetration of α particles and fission fragments through matter. Recent attention has been stimulated in connection with the design of powerful heavy-ion accelerators in which charge transfer plays a prominent role. The results of these studies have been reviewed recently by Betz¹ and by Tawara and Russek.² Charge transfer is also an important mechanism for the production of inner-shell vacancies in violent ion-atom collisions. It is well recognized now that target K -shell vacancy production cross sections increase drastically when the projectiles carry K -shell vacancies into the collision, particularly if the projectile's nuclear charge is comparable to the nuclear charge of the target.³⁻¹¹ This increase in vacancy production cross sections is understood as due to the opening of the K - K charge transfer reaction channel.

Theoretical studies for multiple charge transfer, whether from outer or inner shells, are very scarce. One obvious reason for the lack of theoretical investigations is the difficulty encountered in the single-charge-transfer process. The simple first-order perturbation theory is found to be inadequate for charge transfer¹²; other methods¹³ which are more accurate turn out to be too complicated to be generalized to multielectron atoms. Adding to the complexity, from the theoretical viewpoint, is that most experiments often give total capture cross sections from the target to the projectile without differentiating the contribution from one subshell to another subshell.

In a recent article,¹¹ it was shown that a simple two-state two-center atomic expansion (TSAE) method is capable of predicting accurate single-electron-transfer cross sections from the K shell of multielectron atoms to the K shell of projectiles when the projectile's velocity is comparable to the K -shell orbital velocity of the target. In this article we generalize the TSAE method to multiple-electron capture. Specific attention is directed at the double-electron capture from the K shell of the target to the K shell of the projectile. For the method to be simple and applicable to double capture in multielectron ion-atom collisions, a simplified atomic model is proposed. By replacing the interelectronic Coulomb interaction $1/r_{12}$ by monopole terms with a screening charge, the complicated integrals involving $1/r_{12}$ are eliminated. To account for the two-step mechanism in which double capture proceeds through two successive single-electron transfers, an atomic basis function describing one K -shell electron in the target and the other in the K shell of the projectile is also included in the atomic expansion; this basis function, together with wave functions of the initial and final states, makes the present model a three-state two-center atomic expansion calculation. This simplified model is applied to obtain double-electron-capture cross sections in p -He and He^{2+} -He collisions to assess the importance of the approximations introduced in our model. Applications of this model to the double K -shell electron capture from neon atoms by F^{9+} , O^{8+} , and N^{7+} ions are then made; the results of this calculation are compared with the intensities of hypersatellite lines in the Auger spectra of neon measured by Woods *et al.*¹⁴ These hypersatellites, lying on the high-energy side of the diagram lines, are due to the filling of double K -shell vacancies by outer electrons.

II. THEORETICAL MODEL

The aim of this article is to investigate double-electron capture from the K shell of multielectron atoms to the K shell of bare projectiles. We will adopt a simple atomic model by disregarding the electrons which do not involve directly in the transitions. By neglecting the interaction of the two K -shell electrons with other outer electrons, the motion of these two electrons, described by the time-dependent wave function $\Psi(\vec{r}_1, \vec{r}_2, t)$, is governed by

$$\left(H - i \frac{\partial}{\partial t}\right) \Psi(\vec{r}_1, \vec{r}_2, t) = 0, \quad (1)$$

$$\Psi(\vec{r}_1, \vec{r}_2, t) = a(t)\psi_1(\vec{r}_{1A})\psi_1(\vec{r}_{2A})\exp\{-i[\frac{1}{2}\vec{v} \cdot (\vec{r}_1 + \vec{r}_2) + \frac{1}{4}v^2t + E_A t]\} + b(t)\psi_2(\vec{r}_{1B})\psi_1(\vec{r}_{2A}) \\ \times \exp\{-i[\frac{1}{2}\vec{v} \cdot (-\vec{r}_1 + \vec{r}_2) + \frac{1}{4}v^2t + E_B t]\} + c(t)\psi_2(\vec{r}_{1B})\psi_2(\vec{r}_{2B})\exp\{-i[\frac{1}{2}\vec{v} \cdot (-\vec{r}_1 - \vec{r}_2) + \frac{1}{4}v^2t + E_C t]\}. \quad (3)$$

In Eq. (3) $a(t)$, $b(t)$, and $c(t)$, respectively, are the probability amplitudes at time t for elastic scattering, single-electron transfer, and double-electron transfer. The velocity-dependent phase factors are introduced to ensure that the electrons are moving either with the target or with the projectile.¹⁵ Approximate independent-electron wave functions are used in expansion (3); they are written as product of hydrogenic wave functions with certain effective charges. The total two-electron energies of the three atomic states are given by E_A , E_B , and E_C , respectively.

The simple approximate wave functions used in Eq. (3) are introduced to simplify the calculations for evaluating double-electron-capture cross sections. The expansion in Eq. (3) itself introduces three major approximations. First, it is a truncated expansion in an otherwise complete eigenfunction expansion (actually the two-center expansion is an overcomplete basis expansion) using atomic functions. Second, the three atomic basis functions used in the expansion are approximate wave functions of the Hamiltonian (2) in the separated-atom limit; more accurate eigenfunctions for the two electrons are available but are too complicated to use for the general study here. Thirdly, the intermediate-state wave function used in Eq. (3) is not properly symmetrized with respect to the two electrons.

By substituting Eq. (3) into Eq. (1) and (2), a set of coupled first-order differential equations for the transition amplitudes, written in matrix form

$$i\dot{S}y = Hy \quad (4)$$

where the two-electron nonrelativistic Hamiltonian is

$$H = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z_A}{r_{1A}} - \frac{Z_A}{r_{2A}} - \frac{Z_B}{r_{1B}} - \frac{1}{r_{12}}. \quad (2)$$

Atomic units are used in Eqs. (1) and (2) and throughout this paper. The index $i=1, 2$ refers to the two electrons and $i=A, B$ refers to the target and projectile, respectively. In the above expression, Z_A (Z_B) is the nuclear charge of the target (projectile); $r_{ij} = |\vec{r}_i - \vec{r}_j|$ is the distance between the two particles i and j , with the radius vectors defined with respect to the midpoint of the internuclear axis. In the three-state two-center atomic expansion of $\Psi(\vec{r}_1, \vec{r}_2, t)$, we write

is obtained. The column vector y consists of the three amplitudes $a(t)$, $b(t)$, and $c(t)$. The matrix S consists of overlap matrix elements between nonorthogonal orbitals and H consists of Coulomb interactions between these orbitals. Because of the assumption of product functions in the expansion in Eq. (3), the matrix elements of S and H are all expressible in terms of simple one-electron integrals except integrals involving $1/r_{12}$. These latter integrals can be evaluated by some elaborate methods but we choose to ignore these integrals altogether by replacing the Hamiltonian (2) by

$$H^* = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z_A^*}{r_{1A}} - \frac{Z_A^*}{r_{2A}} - \frac{Z_B^*}{r_{1B}} - \frac{Z_B^*}{r_{2B}}, \quad (5)$$

through the introduction of effective charges $Z_i^* = Z_i - \frac{5}{16}$ ($i=A, B$). This choice of Hamiltonian gives the best variational ground-state wave functions for the two-electron atoms when the wave functions are assumed to have the simple product form. It approximates the true two-electron Hamiltonian H of Eq. (2) well in the asymptotic region when both electrons are centered at the projectile or at the target, but not so well when the two electrons are associated with different centers. In this latter case the bare nuclear charge Z_A and Z_B should be used in the asymptotic region. Since we are interested in double-electron capture, we prefer to choose the approximate Hamiltonian (5) which provides a better description of initial and final states. By choosing the Hamiltonian (5), the basis functions used in Eq. (3) are the exact solutions of the two-electron atoms in the separated-

atom limit.

Under the assumption given above, all the matrix elements of S and H are expressible in terms of one-electron integrals. The explicit forms of these matrix elements are given in the Appendix.

III. RESULTS AND DISCUSSIONS

A. Double capture from two-electron atoms

For two-electron atoms, the Hamiltonian (2) is exact in the nonrelativistic theory. The simplified Hamiltonian H^* of Eq. (5) is expected to be valid for $Z_A, Z_B \gg 1$. To investigate the effect of replacing $1/r_{12}$ by effective monopole terms as given in H^* , we compute double-electron-capture cross sections in p -He and $\text{He}^{2+} + \text{He}$ collisions; the results are compared with calculations which do not impose such an approximation.

1. $p + \text{He} \rightarrow \text{H}^- + \text{He}^{2+}$

This is obviously not a system where the screening approximation of our model is expected to work very well. Two drawbacks of this approximation are obvious: the electron-electron interaction in H^- is the same order as the electron-proton interaction; and the product function $\psi_2(\vec{r}_{1B})\psi_2(\vec{r}_{2B})$, where ψ_2 is the hydrogenic $1s$ wave function with effective charge $Z_2^* = \frac{11}{16}$, is known to be not representing the ground state wave function of H^- very well. Nevertheless, calculations based upon our present model are performed to check the energy dependence of the deviation from other calculations and experiments.

Experimental data for reaction (6) have been obtained by Fogel *et al.*,¹⁶ by Williams,¹⁷ and by Schryber¹⁸ at low energies and by Toburen and Nakai¹⁹ at higher energies. Their results are shown in Fig. 1 together with theoretical calculations. Among the theoretical predictions, the results of Gerasimenko²⁰ are obtained using first Born approximation which disagree with experiments by more than two orders of magnitude. The deficiency of the first Born theory for charge transfer has been examined before¹¹ and will not be discussed further. The other three theoretical calculations, including the present one, all employ the three-state two-center atomic expansion method similar to the one outlined in Sec. II, the difference being in the important fine details involved in carrying out the calculation. Among the three, the calculation of Roy *et al.*,²¹ shown in dash-dotted lines in Fig. 1, probably involves the least approximations beyond the truncated atomic expansion mentioned above. They solve the time-dependent Schrödinger Eq. (1) with the full Hamiltonian (2). The basis functions used in the atomic expansion are the accurate correlated wave func-

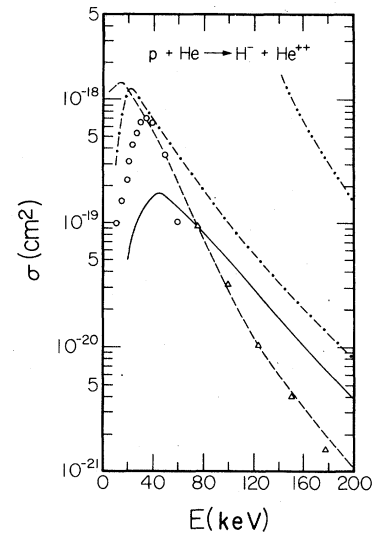


FIG. 1. Total double-electron-capture cross sections in proton-helium collisions. Experimental: \circ , Williams; Δ , Toburen and Nakai. Theoretical: solid line, present; dashed line, Biswas *et al.*; dash-dotted line, Roy *et al.*; dash-double-dotted, Gerasimenko.

tions of Schull and Löwdin²² for He and of Löwdin²³ for H^- ; and the antisymmetrization of the two electrons in the intermediate state is properly accounted for in the expansion. Their only further approximation is that the two-center exchange integrals are evaluated approximately, though the influence of this approximation has been claimed to be unimportant. The results of their calculation are in reasonable agreement with experimental data near where the cross section peaks, but much too high at higher energies. The calculations of Biswas *et al.*,²⁴ shown in dashed lines, agree with experimental results very well at higher energies; however, the reason for this agreement is not clear. In essence, the physical model of Biswas *et al.* is not different from that of Roy *et al.* except that the former uses plane waves to describe the motion of the heavy projectiles while the latter uses an impact parameter approximation. The difference in using the classical and quantum-mechanical approach in describing the motion of heavy projectiles in ion-atom collisions has been shown by Bates¹⁵ to have no effect on the total-capture cross sections; thus the difference in capture cross sections in the two calculations must be attributed to the further approximations introduced in the model of Biswas *et al.* In their model, the close-coupling equations are converted to integral equations. In solving these integral equations, they adopt the method of Bhadra *et al.*²⁵ by neglecting the principal-value parts of the inte-

grals. These further approximations result in significant improvement for the calculated single- and double-capture cross sections, particularly at higher energies. On the other hand, the physical basis for neglecting the principal-value parts of the integrals and the improvement of the model are not established.

The results of the present simple model are shown in a solid line in Fig. 1. Obviously the agreement with experiment is poor, particularly at low energies. This is not very surprising since the work of Bransden and Sin Fai Lam²⁶ and others²⁷ have shown that proper antisymmetrization of the total two-electron wave function is important at low collision energies (< 50 keV). Our results at higher energies are closer to experiments than those of Roy *et al.*; however, the better agreement is probably fortuitous unless the numerical approximation used by Roy *et al.* introduces substantial error. On the other hand, the small deviation from Roy *et al.* in this energy region indicates that the approximations adopted in our model do not produce significant errors. The parameters used in our calculations are $Z_A^* = 1.6875$; $Z_B^* = 0.6875$; $E_A = Z_A^{*2}$; $E_C = Z_B^{*2}$; E_B

$= \frac{1}{2}(E_A + E_C)$. Experimental two-electron total energies for E_A and E_C have also been used in the calculation, but no substantial differences in the cross sections are noticed.

2. $\text{He}^{2+} + \text{He}(1s^2) \rightarrow \text{He}(1s^2) + \text{He}^{2+}$

This is the simplest symmetric resonant-double-electron-transfer process, the cross sections are expected to fall off smoothly with increasing collision energies. Experimental and theoretical results for this reaction are shown in Fig. 2. Data points at low energies, shown by triangles, are due to Bayfield and Khayrallah²⁸; their results are very close to those of Shah and Gilbody²⁹ and of Berkner *et al.*³⁰ (within 35%). At higher energies, the data shown in circles are due to Allison,³¹ and the data shown in a solid line, are due to Pivovar *et al.*³² The three theoretical calculations are all performed using the three-state two-center atomic expansion method. The calculation of Fulton and Mittleman³³—their results shown in dash-double-dotted lines, solves the full two-electron Hamiltonian (2) but uses the simple product of hydrogenic wave functions as basis functions. Our calculation also uses the same simple product function but solves the approximate Hamiltonian (5). The work of Fulton and Mittleman also includes proper symmetrization of the intermediate state which we do not include. Both calculations agree with experimental data reasonably well, indicating our approximations do not produce significant errors for this reaction. The dashed lines are due to Mukherjee *et al.*³⁴ where an approximation similar to that used by Biswas *et al.*²⁵ for *p*-He collisions has been employed; their results agree with experiments very well.

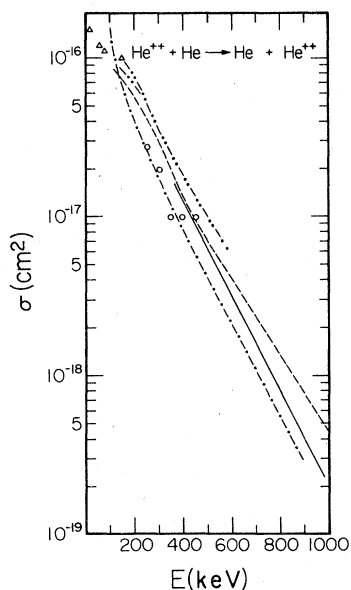


FIG. 2. Total double-electron-capture cross sections in $\text{He}^{2+} + \text{He}(1s^2)$ collisions. Experimental: Δ , Bayfield and Khayrallah; \circ , Allison; solid line, Pivovar *et al.* Theoretical: dash-dotted line, present; dash-double-dotted line, Fulton and Mittleman; dashed line, Mukherjee *et al.*

B. Double-electron capture from the *K* shell of multielectron atoms by bare projectiles

For ion-multielectron-atom collisions, the two-electron Hamiltonian (2) is no longer rigorous because the interaction of *K*-shell electrons with outer electrons is not included. The approximate Hamiltonian (5) includes the mutual screening of *K*-shell electrons well but not the outer screening due to outer electrons. This later effect will be partially included in our model by using experimental binding energy for E_A in Eq. (3) in the actual calculations instead of the hydrogenic values obtained from using the effective charge Z_A^* .

A great deal of multiple-charge-transfer data has been accumulated, but measurements of double capture from one specific subshell to another are

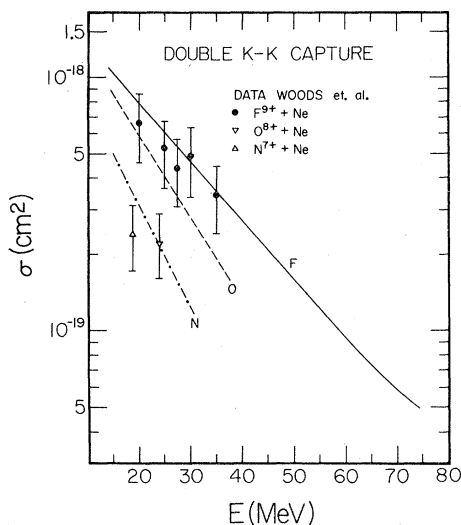


FIG. 3. Total double-electron-capture cross sections from neon atoms by F^{9+} , O^{8+} , and N^{7+} projectiles. The experimental data are taken from Woods *et al.* The theoretical results are from the present calculation; solid line, F^{9+} ; dashed line O^{8+} ; dash-dotted line, N^{7+} .

scarce. Recently Woods *et al.* measured the intensities of Ne Auger hypersatellite lines by bombarding neon gas targets by 1–2-MeV/amu projectiles of F^{9+} , O^{8+} , and N^{7+} ions. These hypersatellites are due to the filling of double K -shell vacancies by outer electrons and are observed *only* when the projectiles are *bare* nuclei. This latter fact implies that these double vacancies are produced by the double K - K charge transfer process; contributions from direct Coulomb ionization are negligible.

Experimental data of Woods *et al.* also established that double-capture cross sections in F^{9+} -Ne collisions drop by a factor of 2 within the narrow energy region of 1–2 MeV/amu, while in the same energy region, the single-capture cross sections remain almost constant. To investigate the velocity and projectile-charge dependence of the double capture-cross section, we employ the simplified model of Sec. II to compute the double K - K capture cross sections in F^{9+} -, O^{8+} -, and N^{7+} -Ne collisions. Our model is expected to work reasonably well for $Z_A, Z_B \gg 1$, as in the collision systems studied here. The approximation of replacing $1/r_{12}$ by monopole terms, the representation of two-electron wave functions by product hydrogenic functions are becoming more valid with increasing Z_A and Z_B .

In Fig. 3, we show the results of our simple calculations and the comparison with the data of Woods *et al.*¹⁴ The energy dependence and actual values of double K - K capture cross sections for F^{9+} -Ne collisions calculated in our model are in

good agreement with data. For N^{7+} and O^{8+} projectiles, there is only one experimental datum point for each case; but the agreement is not as good. There is no other theoretical data for comparison except the simple estimate from the approximation of Oppenheimer,³⁵ Brinkman and Kramers³⁶ (OBK) given in Ref. 14. The OBK values are two orders of magnitude higher than experimental values.

IV. SUMMARY

We have applied the two-center atomic expansion method to the double-electron-capture processes in multielectron ion-atom collisions at intermediate energies by adopting a simple independent-electron approximation in which the $1/r_{12}$ interelectronic Coulomb interaction is replaced by monopole terms with a screening charge. Applications of the present model to the simple p -He and He^{2+} -He double-capture processes indicate that the present model is capable of providing a reasonable estimate of the capture cross sections. Extension of the present simple model to more complicated multielectron ion-atom collisions for the double K -shell vacancy production in neon atoms by F^{9+} , O^{8+} , and N^{7+} projectiles gives cross sections in agreement with the intensities of hypersatellite lines of neon measured by Woods *et al.*

It is appropriate to comment on the region of validity of the present model. As discussed in Ref. 12, two-center atomic expansion method is expected to be a valid approximation for electron-transfer processes when the projectile's velocity is nearly identical to the orbital velocity of the particular subshell from which electron capture occurs. Only a very limited number of basis functions will be needed in the atomic expansion if the energy levels of initial and final states are very close such that there are not additional states with energy levels lying in between.

It is generally known that vacancy production in ion-atom collisions is dominated by direct Coulomb ionization at high collision energies while electron-capture processes is more important when the projectile and orbital-electron velocities are nearly equal; this latter process is particularly important when the charge of the projectile is not small. For ion-atom collisions with a sufficiently high projectile velocity, the target atoms will lose their outer electrons primarily by direct-Coulomb-ionization mechanism while the inner-shell electrons are lost by electron-capture process. It is interesting to see if the two different electron-loss processes can be treated independently. By combining the model of McGuire and Richard³⁷ for multiple Coulomb ionization with the multiple-

capture probabilities obtained using the present model, a simple *ab initio* theory for multiple-electron loss can be derived. Such a model for multiple-electron loss is probably desirable in view of the difficulty in formulating a multichannel scattering theory which incorporates both multiple-direct-Coulomb ionization and electron-capture processes simultaneously.

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APPENDIX: REDUCTION OF TWO-ELECTRON INTEGRALS IN DOUBLE K-K CAPTURE CALCULATIONS

By using products of hydrogenic functions as basis in the atomic expansion (3) and employing the simplified Hamiltonian (5), the S and H matrix elements of Eq. (4) are expressible in terms of single-electron integrals. For matrix elements of S ,

$$S_{ii} = 1, \quad i = 1, 2, 3$$

$$S_{12} = S_{ab}, \quad S_{13} = S_{ab}^2, \quad S_{23} = S_{ab}, \quad (\text{A1})$$

and

$$S_{ji} = S_{ij}^*, \quad i, j = 1, 2, 3;$$

and for matrix elements of H ,

$$H_{12} = h_{ab} + s_{ab}h_{aa}, \quad H_{21} = h_{ba} + s_{ba}h_{aa}, \quad H_{13} = 2s_{ab}h_{ab},$$

$$H_{31} = 2s_{ba}h_{ba}, \quad H_{23} = h_{ab} + s_{ab}h_{bb}, \quad H_{32} = h_{ba} + s_{ba}h_{bb},$$

$$H_{11} = 2h_{aa}, \quad H_{22} = h_{aa} + h_{bb}, \quad H_{33} = 2h_{bb}, \quad (\text{A2})$$

where the one-electron matrix elements are defined as

$$s_{ab} = \int d\tau \phi_A^* \phi_B e^{i\vec{v}\cdot\vec{r} + i\omega t},$$

$$h_{ab} = \int d\tau \phi_A^* \left(-\frac{Z_A^*}{r_A} \right) \phi_B e^{i\vec{v}\cdot\vec{r} + i\omega t},$$

$$h_{ba} = \int d\tau \phi_B^* \left(-\frac{Z_B^*}{r_B} \right) \phi_A e^{-i\vec{v}\cdot\vec{r} - i\omega t}, \quad (\text{A3})$$

$$h_{aa} = \int d\tau \phi_A^* \left(-\frac{Z_B^*}{r_B} \right) \phi_A,$$

$$h_{bb} = \int d\tau \phi_B^* \left(-\frac{Z_A^*}{r_A} \right) \phi_B.$$

The one-electron excitation energy ω is defined as

$$\omega = \epsilon_A - \epsilon_B,$$

with ϵ_A (ϵ_B) the hydrogenic 1s binding energy with effective nuclear charge Z_A^* (Z_B^*). The total two-electron energies E_A , E_B , and E_C in Eq. (3) are related to ϵ_A and ϵ_B by $E_A = 2\epsilon_A$, $E_B = \epsilon_A + \epsilon_B$, and $E_C = 2\epsilon_B$.

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