Double excitation in Li and Be by electron impact: A distorted-wave approach

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The distorted-wave approximation is used to calculate the double-electron-excitation cross section of $\text{Li}(^2S^{-4}P)$ and $\text{Be}(^1S^{-3}P)$ by electron impact. The distortion in the incident and exit channels incorporates the distortion effects of the static, polarization, and exchange potentials appropriately. The results are compared with the Born-Oppenheimer approximation.

I. INTRODUCTION

Single-electron excitation in various atoms (ions) is well studied.¹⁻⁴ Atoms (ions) with two active electrons may have two electrons excited. Such doubly excited states lie in the continuum and have been known spectroscopically for some time.^{5,6} These states are short lived and mostly decay to the continuum. With the availability of sophisticated experimental techniques it is now becoming possible to study such states in more detail and to explore their utility in a broad sense. A few theoretical approximations have been used to find the production cross section of some doubly excited states in helium atoms.^{7–12} These included the simple Born-Oppenheimer,^{7–9} close-coupling,¹⁰ Glauber,¹¹ and distorted-wave (DW) approximation¹² methods. For helium, there has been even an effort¹³ to measure the cross section for the $2p^{33}P$ excitation state from its ground $1s^{21}S$ state. It has been shown¹² that the DW approach is quite useful when applied to double excitation in helium. Similar conclusions are also drawn in similar DW studies of single innershell-electron excitations of autoionizing levels in lithium.⁴ In lithium, a few doubly excited autoionized states have been observed in addition to the prevalent single inner-shell excited autoionizing states. These doubly excited states in lithium are of fundamental importance because their excitation cross-section knowledge could be used to understand electron capture by lithium (viz., Listudies). There is only one calculation for the ${}^{2}S{}^{4}P$ double excitation in Li using the simple Born-Oppenheimer (BO) theory. This was done by Kulander and Dahler.⁹ However, better approximations are now available to give reliable estimates of the double-excitation cross sections. Motivated by this fact, as well as by our experience with the useful application of the DW theory,9,14-16 encouraged us to reexamine this problem using the DW theory.

In this paper we calculate the $\text{Li}({}^{2}S^{-4}P)$ excitation cross section and compare the results to the BO theory.⁹ In addition to calculating this cross section we extend our DW method to the calculation of ${}^{1}S^{-3}P$ double excitation in Be. These cross sections are again compared with those of the BO theory. Our choice of these doubly excited states in Li and Be is based on two facts: (1) being parityunfavored transitions⁹ these are relatively longer-lived states and are suitable to be treated as bound states, and (2) the BO calculations⁷⁻⁹ are available for comparative purposes.

II. THEORY

The T matrix in the distorted-wave (DW) approximation (with full allowance of exchange symmetries) for any inelastic electron and an N-electron target-atom scattering process is given by¹⁵⁻¹⁷

$$T_{if} = \langle F^{-}(\mathbf{k}_{f}, 0)\psi_{f}(1, \dots, N) | V(0, \dots, N) \\ - U_{i(f)}(0) | \psi_{i}(1, \dots, N)F^{+}(\mathbf{k}_{i}, 0) \rangle , \qquad (1)$$

where

$$V = -\frac{Z}{r_0} + \sum_{i=1}^{N} \frac{1}{r_{i0}} .$$
 (2)

V is the interaction potential between the target and the projectile electron. The properly antisymmetrized initial (final) atomic wave function is $\psi_i(\psi_f)$, the distorted-wave function of electron in the incident (exit) channel is $F^+(\mathbf{k}_i,\mathbf{r}_0)$ or $F^-(\mathbf{k}_f,\mathbf{r}_0)$, and \mathbf{r}_{i0} is the distance between the electron whose coordinate is \mathbf{r}_i and the free electron having position vector \mathbf{r}_0 . The superscript + (-) refers to the usual outgoing (ingoing) wave-boundary conditions. Z is the target nuclear charge. \mathbf{k}_i and \mathbf{k}_f are chosen respectively to be initial and final wave vectors of the projectile electron; $U_{i(f)}$ is defined to be initial (final) spherically averaged distorting potential for the projectile electron in the incident (exit) channel. (Atomic units will be used throughout.) For $U_{i(f)}$ the following choice was made:

$$U_{i(f)} = V_{\text{stat}}^{i(f)} + V_{\text{exch}}^{i(f)} + V_{\text{pol}}^{i(f)} .$$
(3)

In the above expression [Eq. (3)] on the right-hand side the first term is the static potential of the target, and can be written as

$$V_{\text{stat}}^{i(f)} = \langle \psi_{i(f)} | V | \psi_{i(f)} \rangle .$$
(4)

If $\psi_{i(f)}$ is written as a single determinant of spin orbitals

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 $(\phi_{i(f)}^{j})$, then

$$\psi_{i(f)} = \frac{1}{\sqrt{N!}} \det \left| \phi_{i(f)}^{1} \phi_{i(f)}^{2} \cdots \phi_{i(f)}^{N} \right|$$
(5)

and hence static potential [Eq. (4)] can be rewritten as

$$V_{\text{stat}}^{i(f)} = \frac{Z}{r_0} - \sum_{j=1}^{N} \int \frac{|\phi_{i(f)}^j|^2}{|r_j - r_0|} d\mathbf{r}_j d\sigma_j , \qquad (6)$$

 σ_j is the spin coordinate of the *j*th electron. The second term [in Eq. (3)] is the nonlocal exchange potential which takes into account the exchange of the projectile electron with the bound electrons in the target. The third term [in Eq. (3)] is the polarization potential which includes the polarization effect of the target by the projectile electron. The form and choice of these potentials for different atoms will be mentioned later in the text when we consider each transition separately. Expanding F(k,r) in partial waves, we have (assuming $k = k_i$ or k_f and $U = U_i$ or U_f)

$$F^{\pm}(k,r) = \frac{1}{\sqrt{k}} \sum_{l=0}^{\infty} (2l+1)i^{l} e^{\pm i\delta_{l}(k^{2})} \frac{u_{l}(k,r)}{r} P_{l}(\hat{\mathbf{k}}\cdot\hat{\mathbf{r}}) ,$$
(7)

where δ_l is the phase shift of the *l*th partial wave and $u_l(k,r)$ is the solution of the equation

$$\left[\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} - 2U(r) + k^2\right] u_l(k,r) = 0.$$
 (8)

The boundary conditions are

$$u_l(k,r)_{r\to 0} = 0 \tag{9}$$

and

$$u_{l}(k,r)_{r\to\infty} = k^{-1/2} \sin \left[kr - \frac{l\pi}{2} + \delta_{l}(k^{2}) \right].$$
 (10)

The equations are solved numerically using the procedure adopted by McDowell et al.,¹⁸ and Srivastava et al.¹⁵ The procedure consists of using the noniterative method of Marriot¹⁹ to obtain the solution of Eq. (8). The normalization of the radial wave equation and evaluation of the phase shift is done by matching the JWKB solution as suggested by Burgess.²⁰ Total cross sections are calculated from the T matrix [Eq. (1)] in the conventional manner after selecting a specific transition for a particular target atom. Appropriate atomic wave functions are used, and the distorted waves are obtained from Eq. (8). In Sec. III we describe our procedure for the evaluation of the T matrix for specific cases. In each case we use atomic wave functions which are chosen to be suitable combinations of single-particle orthonormal wave functions. The L-S coupling scheme is assumed.

III. APPLICATION TO SPECIFIC ATOMS

A. Lithium $(1s^2 2s S \rightarrow 1s 2p^2 P)$

The ground-state $1s^22s^2S$ function is written in the following form:

$$\psi_i \equiv \psi_{00}(1,2,3) = \frac{1}{\sqrt{3}} \left[R_{1s}(1) R_{1s}(2) R_{2s}(3) Y_{00}(\hat{1}) Y_{00}(\hat{2}) Y_{00}(\hat{3}) \eta_i(1,2,3) + \cdots \right],$$
(11)

where the ellipse represents two cyclic permutations of electron labels [in Eq. (12) also]. We used $\phi_{nlm}(\mathbf{\hat{r}}) = R_{nl}(r)Y_{lm}(\hat{\mathbf{r}})$ where R_{nl} and $Y_{lm}(\hat{\mathbf{r}})$ refer to the radial and spherical harmonic part of an orbital wave function. Here the spin function is

$$\eta_i(1,2,3) = (1/\sqrt{2})(\alpha_1\beta_2 - \alpha_2\beta_1)\alpha_3$$

The excited state $(1s 2p^{24}P)$ can be written as

$$\psi_f \equiv \psi_{1,M_1}(1,2,3)$$

$$=\frac{1}{\sqrt{3}}\sum_{m_1,m_2}C(1,1,1,m_1,m_2,M_L)[R_{2p}(1)R_{2p}(2)R_{1s}(3)Y_{1m_1}(\hat{1})Y_{1m_2}(\hat{2})Y_{00}(\hat{3})+\cdots]n_f(1,2,3), \qquad (12)$$

where

$$\eta_f(1,2,3) = \frac{1}{\sqrt{2}} (\alpha_1 \beta_2 + \alpha_2 \beta_1) \alpha_3$$
.

Here L=1, $M_L=(0,\pm 1)$, and the radial wave functions R_{1s} , R_{2s} , and R_{2p} for different orbitals as well as excitation energy, ($\Delta E=4.54$ Ry) are taken to be the same as used by Kulander and Dahler.⁹ $C(l_1, l_2, l_3, m_1, m_2, m_3)$ is the Clebsch-Gordan coefficient.²¹

With these atomic wave functions used in the T matrix [Eq. (1)] and performing integration on the spin coordinates we can write simply (the direct matrix as well as most of the other terms in the exchange matrix become zero due to orthogonality of the atomic orbital wave functions)

$$T_{if}^{M_{L}} = \frac{1}{\sqrt{6}} \sum_{m_{1},m_{2}} C(1,1,1,m_{1},m_{2},M_{L}) \left[\left\langle F^{-}(\mathbf{k}_{f},0)R_{2p}(1)R_{2p}(2)R_{1s}(3)Y_{1m_{1}}(\widehat{1})Y_{1m_{2}}(\widehat{2})Y_{00}(\widehat{3}) \times \left| \frac{1}{r_{02}} \right| F^{+}(\mathbf{k}_{i},1)R_{1s}(3)R_{1s}(0)R_{2s}(2)Y_{00}(\widehat{0})Y_{00}(\widehat{2})Y_{00}(\widehat{3}) \right\rangle - \left\langle F^{-}(\mathbf{k}_{f},0)R_{2p}(1)R_{2p}(3)R_{1s}(2)Y_{1m_{1}}(\widehat{3})Y_{1m_{2}}(\widehat{1})Y_{00}(\widehat{2}) \times \left| \frac{1}{r_{03}} \right| F^{+}(\mathbf{k}_{i},0)R_{1s}(2)R_{1s}(0)R_{1s}(3)Y_{00}(\widehat{0})Y_{00}(\widehat{2})Y_{00}(\widehat{3}) \right\rangle \right].$$
(13)

Further,

$$T_{if}^{M_L} = -\frac{1}{\sqrt{12}} I_1 I_2 (\delta_{M_L,1} + \delta_{M_L,-1})$$
(14)

with

$$I_{1} = \left\langle F^{-}(\mathbf{k}_{f}, 0) R_{2p}(2) Y_{1, M_{L}}(\widehat{2}) \right. \\ \left. \times \left| \frac{1}{r_{02}} \right| R_{1s}(0) R_{2s}(0) Y_{00}(\widehat{0}) Y_{00}(\widehat{0}) \right\rangle$$
(15)

and

$$I_2 = \langle R_{2p}(\hat{1}) Y_{10}(\hat{1}) | F^+(\mathbf{k}_i, 1) \rangle , \qquad (16)$$

and with the use of spherical harmonic expansions for $F^{\pm}(k,r)$, as given by equation,⁷ and for $1/r_{02}$, given by the following:

$$\frac{1}{r_{02}} = \sum_{l} \frac{4\pi}{2l+1} \frac{r_{<}^{l}}{r_{>}^{l+1}} Y_{lm}(\hat{\mathbf{0}}) Y_{lm}^{*}(\hat{\mathbf{2}}) .$$
(17)

We can reduce both I_1 and I_2 to the form of onedimensional comfortable integrals which are evaluated using Simpson's method. The total cross section σ is then obtained using conventional integration over all scattering angles in space to give

$$\sigma = 2 \frac{k_f}{k_l} \int \left| \frac{1}{2\pi} T_{if}(M_L = 1) \right|^2 d\Omega . \qquad (18)$$

The following potentials have been chosen to solve Eq. (8) for the initial (final) distorted waves $u_l(k_{i(f)}, r)$

$$V_{\text{stat}}^{i}(r) = -\frac{Z}{r_{0}} + A_{1} \int \frac{|\phi_{1s}(\mathbf{r})|^{2}}{|\mathbf{r} - \mathbf{r}_{0}|} d\mathbf{r} + A_{2} \int \frac{|\phi_{2s}(\mathbf{r})|^{2}}{|\mathbf{r} - \mathbf{r}_{0}|} d\mathbf{r} , \qquad (19)$$

$$V_{\text{stat}}^{f}(\mathbf{r}) = -\frac{Z}{r_{0}} + A_{1}^{1} \int \frac{|\phi_{1s}(\mathbf{r})|^{2}}{|\mathbf{r} - \mathbf{r}_{0}|} d\mathbf{r} + A_{2}^{1} \int \frac{|\phi_{2p}(\mathbf{r})|^{2}}{|\mathbf{r} - \mathbf{r}_{0}|} d\mathbf{r} , \qquad (20)$$

with $A_1 = 2$, $A_2 = 1$ and $A_1^1 = 1$, $A_2^1 = 2$.

The second term in Eq. (19) is not spherically symmetric in nature so we have used the conventionally adopted $^{16-22}$ spherically symmetric part of it.

The following form of the polarization potential is utilized:

$$V_{\rm pol}^{i} \equiv V_{\rm pol}^{f} = -\frac{\alpha}{2r^{4}} \left[1 - \sum_{i=1}^{8} b_{i}r^{i}e^{-ar} \right].$$
(21)

 α is the dipole polarizability of lithium in the ground state, and b_i 's and α are known constants and are taken as described in the literature.^{23,24}

The exchange potential is chosen to be that suggested by Furness and McCarthy 25 and used by several others, 17,26

$$V_{\text{exch}}(r) = \left(\left[\frac{1}{2} k_i^2 - V_{\text{stat}}^i(r) \right] - \left\{ \left[\frac{1}{2} k_i^2 - V_{\text{stat}}^i(r) \right] + 4A_1 |\phi_{1s}|^2 + 4A_2 |\phi_{2s}|^2 \right\}^{1/2} \right) / 2.$$
 (22)

Our choice in selecting the same exchange and polarization potentials in the incident and exit channel is not unique. This has been done with success in previous works^{15,17} also.

B. Beryllium atom $(1s^22s^2 S - 1s^22p^2 P)$

In the Be atom there are two valence electrons $(2s^2)$ which lie outside a spherically symmetric core. The $1s^2$ electrons are weakly coupled to the $2s^2$ electrons. We therefore consider the $2s^2$ electrons to be the active electrons which are excited by the projectile electron into the $2p^2$ orbital. We also find in ${}^{1}S^{-3}P$ excitation that these two 2s electrons get excited to 2p orbital. It would therefore be adequate in the evaluation of the T matrix [Eq. (1)] to consider only these two electrons to take part in the collision dynamics. Thus, we treat the Be atom to behave as a simple two-electron system represented by its valence electrons in the evaluation of the t matrix. Define ϕ_{1s} , ϕ_{2s} , and ϕ_{2p} to be the orthonormal wave functions of an electron in the Be atom in 1s, 2s, and 2p orbitals. We choose these to be the same as described by Becker and Dahler.⁷ The wave function of the Be atom in the 1s ground state and excited 3p state can be expressed as

$$\psi_i({}^{1}S) \equiv \psi_{00}(1,2)$$

= $R_{2s}(1)R_{2s}(2)Y_{00}(\hat{1})Y_{00}(\hat{2})\eta_i(1,2)$, (23)

with the spin function $\eta_i(1,2) = (1/\sqrt{2})(\alpha_1\beta_2 - \alpha_2\beta_1)$. Similarly,

with the spin function $n_f(1,2) = (1/\sqrt{2})(\alpha_1\beta_2 + \alpha_2\beta_1)$. Substituting these wave functions into Eq. (1) for the transition matrix and proceeding in a similar way as for Li we find after the spin coordinate integration:

$$T_{if}^{M_{L}} = -\sqrt{3}C(1,1,1,m_{1},m_{2},M_{L}) \left\langle F^{-}(\mathbf{k}_{f},0)R_{2p}(1)R_{2p}(2)Y_{1m_{1}}(\widehat{1}) \times Y_{1m_{2}}(\widehat{2}) \left| \frac{1}{r_{02}} \right| R_{2s}(0)R_{2s}(2)F(\mathbf{k}_{i},1)Y_{00}(\widehat{0})Y_{00}(\widehat{2}) \right\rangle.$$
(25)

Further simplification gives

$$T_{if}^{M_L} = -\sqrt{3/2} I_1^1 I_2^1 \delta_{M_{L,1}} , \qquad (26)$$

where

$$I_{1}^{1} = \langle R_{2p}(1)Y_{10}(\hat{1}) | F^{+}(\mathbf{k}_{i},1) \rangle$$
(27)

and

$$I_{2}^{1} = \left\langle F^{-}(\mathbf{k}_{f}, 0) R_{2p}(2) Y_{11}(\hat{2}) \times \left| \frac{1}{r_{02}} \right| R_{2s}(\hat{0}) R_{2s}(\hat{2}) Y_{00}(\hat{0}) Y_{00}(\hat{2}) \right\rangle.$$
(28)

 I_1^1 and I_2^1 are integrals which are evaluated similar to I_1 and I_2 [Eqs. (15) and (16)] and the expression given by Eq. (18) is utilized to find the total cross section for Be. The excitation threshold energy is taken to be $\Delta E = 0.543$ Ry, which is the same as used by Becker and Dahler.⁷

In the present case for Be the same form of static and exchange potentials are used. These are given by Eq. (19), (20), and (22). However, the use of the following values for the constants, as well as substituting wave function for Be must be used: $A_1=2$, $A_2=2$, $A_1^1=2$, $A_2^1=2$.

In contrast to the H, He, and Li atoms no suitable expression for the polarization potential in Be was available in the literature. However, in order to see the effect of adding a polarization potential in the distorted waves cal-



FIG. 1. Double excitation cross sections for ${}^{2}S{}^{4}P$ excitation in lithium by electron impact. \triangle , present DW results (IF); \bigcirc , present DW results (IFE); \square , present DW results (IFEP); \times , BO-theory results (taken from the figure shown in Ref. 9).

culations were performed using one of the standard forms:²⁷

$$V_{\rm pol}^{(4)} = -\frac{\alpha}{2r^4} \left\{ 1 - \exp\left[-\left[\frac{r}{r_c g} \right]^8 \right] \right\}.$$
 (29)

Here α (=37.8) is the dielectric polarizability of the beryllium atom and its experimentally measured value is taken from the literature.²⁸ r_c (=1.33) is the position of the last maximum of the absolute value of the outermost wave function, and g is an energy-dependent parameter that is taken to be unity.

IV. RESULTS AND DISCUSSION

Total cross-section results are obtained in our DW model for $1s^22s^{12}S-1s^2p^{24}P$ excitation in lithium and $1s^22s^{21}S-1s^22p^{23}P$ transition in beryllium. Incident electron energies ranged from near threshold to the asymptotic region. In order to assess the effect of the static, exchange, and polarization potentials separately we performed, for each atom, the following types of three separate calculations.

(1) The initial distorted wave is obtained by using the initial ground-state static potential while the final distorted wave in the exit channel is generated by using the final excited-state static potential. Results of this kind will be referred to as IF.

(2) The preceding first choice for the static potential, in



FIG. 2. Double excitation cross section for ${}^{1}S^{-3}P$ excitation in beryllium by electron impact. Same as in Fig. 1 (except that BO-theory results are taken from Ref. 7).

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TABLE $Li(1s)^1(2p)$	I. Electron $P^{24}P$ in units of 10 ⁻	excitation of $-4\pi a_0^2$.	$Li(1s)^2(2s)^{1/2}S$
<i>i</i> (R y)	IF	IFE	IFEP
4.6	0.0389	0.191	0.266
4.7	0.202	0.207	0.309
4.8	0.331	0.339	0.322
5.0	0.335	0.342	0.313
5.3	0.250	0.255	0.266
5.5	0.206	0.211	0.231
5.8	0.188	0.192	0.214
6.0	0.133	0.136	0.155
7.0	0.0609	0.0627	0.0690
8.0	0.0302	0.0312	0.0326

TABLE II. $Be(1s)^2(2s)^{2}S$ Electron excitation of

E_i (Ry)	IF	IFE	IFEP
4.6	0.0389	0.191	0.266
4.7	0.202	0.207	0.309
4.8	0.331	0.339	0.322
5.0	0.335	0.342	0.313
5.3	0.250	0.255	0.266
5.5	0.206	0.211	0.231
5.8	0.188	0.192	0.214
6.0	0.133	0.136	0.155
7.0	0.0609	0.0627	0.0690
8.0	0.0302	0.0312	0.0326

addition to considering the exchange potential [Eq. (22)], are used in obtaining the distorted waves for both the incident and exit channels (IFE).

(3) The second choice for the static and exchange potentials, in addition to the polarization potential [Eq. (21) for Li or Eq. (29) for Be], was included in obtaining the incident- and exit-channel distorted wave (IFEP).

All three different calculations (viz., IF, IFE, and IFEP) for each Li and Be atom are compiled in Tables I and II, respectively. These are also compared among themselves as well as with those of the BO-theory results^{7,9} in Figs. 1 and 2 separately.

In Fig. 1 and Table I our three different distorted-wave results for ${}^{2}S{}^{4}P$ excitation in Li are displayed. We find from the figure that in the near-threshold region the IF results are lowest while IFEP results are largest. The cross sections are quite sensitive to the inclusion of exchange and polarization effects in distorted-wave theory. Near the maxima IFE results are larger than the IFEP and IF result values, but all three results are within 10% variation among themselves. The position of the maximum is in each case nearly at the same energy. It is further seen that at higher energies all the results approach the same asymptotic limit as expected. Comparing our various DW results with those of BO-theory results of Kulander and Dahler⁹ is as shown in Fig. 1. It is seen that the peak value of cross section in BO theory is about the same in magnitude and location in terms of incident energy. The BO-theory results were available up to only 6.0 Ry, and it is seen that the DW calculations appear to approach asymptotic values much sooner than the BO theory.

Figure 2 and Table II show our three different DW results for $1s^22s^{21}S - 1s^22p^{23}P$ transition in beryllium. First compare our IF and IFE results which are similar in magnitude and nature to each other. We again find, similar to Li, that near the threshold region the cross sections

$-\text{Be}(1s)^2(2p)^{23}P$ in units of πa_0^2 .					
$\overline{E_i}$ (Ry)	IF	IFE	IFEP		
0.55	0.153	0.174	0.150		
0.56	0.553	0.637	0.688		
0.57	1.06	1.12	1.48		
0.58	1.62	1.81	2.03		
0.59	2.22	2.46	2.18		
0.60	2.80	3.12	1.99		
0.63	4.40	4.96	1.35		
0.65	5.19	5.90	1.03		
0.70	5.83	6.78	0.556		
0.75	4.93	5.88	0.340		
0.80	3.54	4.31	0.215		
0.85	2.33	2.91	0.141		
0.90	1.47	1.88	0.0945		
1.00	0.571	0.769	0.0448		

are sensitive to the inclusion of the effect of exchange in the distorted-wave theory. Furthermore, the IFE results are, in general, higher than the IF results in almost the entire energy region. Both cross sections approach the same asymptote. Our IFEP results obtained using the approximate form of the polarization potential [Eq. (29)] gave results somewhat indifferently for Li. It is seen that the IFEP results show a peak value much lower in magnitude as compared to the IF and IFE results. The peak is also shifted to lower incident electron energy. At high energy the IFEP results approach the IF and IFE asymptotic limit (off the graph). It appears that a further investigation of the form of the polarization potential [Eq. (29)] is desirable before any definite conclusion regarding the polarization contribution to scattering can be made.

Finally, comparing our various DW results for Be with BO-theory results⁷ (see Fig. 2) we see that BO-theory prediction is a broad maximum as compared to all DW calculations. The IF and IFE results from threshold to approximately 0.8 Ry are approximately two to three times larger than BO results. As the energy of the incident electron increases all curves approach each other. This behavior is somewhat similar to the results seen for the helium atom.⁷ Comparison of IFEP and BO results shows that the magnitude of the maximum cross sections in both is about the same, but the IFEP peak is at a lower energy.

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