Dipole Magnus approximation for electron-atom collisions: Excitation of the resonance transitions of Li, Na, and K

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The Magnus expansion, to second order, has been used to solve the coupled-channel, symmetrized impact-parameter equations for electron-atom scattering. Collision integrals are evaluated in the dipole approximation, allowing both first- and second-order terms to be written in closed, analytic form. The numerical work is therefore essentially reduced to a matrix exponentiation for each value of the impact parameter, which can be efficiently carried out by the well-known diagonalization procedure. It thus becomes computationally feasible to handle problems involving a large number of closely coupled states. As a test case, the theory has been applied to electron-impact excitation of the resonance transitions of Li, Na, and K. The calculated cross sections were found to be in good agreement with experimental data over most of the intermediate-energy range. Thus far, the present method appears to be competitive with more sophisticated approaches and is readily applicable to complex processes, such as electron collisions with atoms in excited states.

I. INTRODUCTION

In the present paper, we consider the theoretical description of electron-atom collisions, involving many channels coupled together by strong, long-range (dipole) forces. This situation is likely to occur, for example, in the treatment of inelastic electron scattering from excited atoms, where there are typically a large number of closely spaced, strongly interacting states. Excitation of the resonance transitions of the alkali-metal atoms represents a much simpler problem, but one that is nevertheless a good prototype for the more complex processes of interest. The objectives of this work are therefore to develop a computationally practical method for many-channel, close-coupling problems and to test the theory on the lighter alkali metals, for which extensive data exits in the literature.[1–6]

Our approach is based upon the Magnus solution[7,8] to the semiclassical, impact-parameter equations for the system. The major advantage of the Magnus expansion is that it guarantees unitarity, independent of where the series is truncated, and therefore conserves probability; it also agrees with both perturbation theory and the sudden approximation as limiting cases. In order to evaluate the collisional matrix elements, we make the dipole approximation, as originally introduced by Seaton[9] and subsequently extended and applied to a variety of problems in atomic collision theory.[10–12] Use of the dipole approximation allows both first- and second-order terms to be given as closed-form, analytic expressions. The numerical work is therefore essentially reduced to a matrix exponentiation for each value of the impact parameter, which can be efficiently carried out by the well-known diagonalization procedure. Computational efficiency is important not only for handling a large number of coupled channels, but also for integrating the scattering equations out to large impact parameters, as would be necessary, for example, in applications involving electron—excited-atom collisions.

The work described in this paper is closely related to two other theories appearing in the literature. Mandelberg[13] has used the Callaway-Bauer method,[14] together with the dipole approximation, to investigate the excitation of atomic hydrogen by electrons. The Callaway-Bauer solution turns out to be equivalent to the first-order term of the Magnus series (see below). Since we include second-order, time correlation effects, our theory extends Mandelberg's work, while also differing from it in several specific ways (e.g., the treatment of the small-impact-parameter limit, the implementation of detailed balance). A much more elaborate theory, referred to as the second-order diagonalization method, has been developed by Baye and Heenen[15] and applied by them to the excitation of hydrogen and helium by both electrons and protons. This method is also based upon a unitary, exponential expansion, includes second-order effects, and more significantly retains the full electron-electron Coulomb potential. It is therefore not limited to dipole interactions as is the present theory, and can be applied to cases for which optically forbidden processes are of particular importance. The drawback to using the full Coulomb interaction is an enormous increase in numerical complexity, especially in the evaluation of the second-order matrix elements. To summarize, the present theory appears to lie between these other two methods in both complexity and potential accuracy.

II. THEORY

We treat the electron-atom collision semiclassically and take as our starting point the symmetrized impact-parameter equations,[16,17] which can be written in matrix notation as

$$i\hbar\frac{\partial}{\partial Z}\underline{c}(\rho,Z)=\underline{H}(\rho,Z)\underline{c}(\rho,Z) \tag{1}$$

with the interaction matrix defined by

$$H_{\nu\mu}(\rho,Z)=\frac{1}{(v_\nu v_\mu)^{1/2}}\langle\nu\,|\,V(\rho,Z;\mathbf{r})\,|\,\mu\rangle e^{i(k_\mu-k_\nu)Z}\,. \tag{2}$$

The incident electron is considered to move along a straight-line trajectory, its position being described by the cylindrical polar coordinates (ρ,Z,Φ) centered on the target atom. The Z axis lies parallel to the trajectory, and ρ represents the impact parameter [the Φ dependence has been factored out of Eqs. (1) and (2) as in Ref. 17]. $\underline{c}(\rho,Z)$ is a column vector whose elements represent the amplitudes of the internal states $(\nu,\mu,\ldots)$ of the target atom, v_ν and k_ν are the asymptotic velocity and wave number in the νth channel, respectively, and $V(\rho,Z;\mathbf{r})$ is the interaction between the incident electron and the target, with internal coordinates denoted collectively by $\mathbf{r}$. The symmetry in the channel velocities guarantees that detailed balance is satisfied, a feature missing from the ordinary impact-parameter method. The corresponding cross section for the transition $0\rightarrow\nu$, with the boundary condition $c_\nu(Z=-\infty,\rho)=\delta_{0,\nu}$ is

$$Q_{\nu 0}=2\pi\frac{v_\nu}{v_0}\int_0^\infty |\,c_\nu(Z=\infty,\rho)\,|^2\rho\,d\rho\,. \tag{3}$$

The Magnus solution to Eq. (1) is given by[7,8] (with the variable ρ suppressed)

$$\underline{c}(Z)=e^{\underline{A}(Z,-\infty)}\underline{c}(-\infty) \tag{4}$$

with

$$\underline{A}(Z,-\infty)=\sum_{n=1}^{\infty}\underline{A}^{(n)}(Z,-\infty) \tag{5}$$

and

$$\underline{A}^{(1)}(Z,-\infty)=\frac{1}{i\hbar}\int_{-\infty}^{Z}dZ_1\underline{H}(Z_1)\,, \tag{6}$$

$$\underline{A}^{(2)}(Z,-\infty)=\frac{1}{2}\left[\frac{1}{i\hbar}\right]^2\times\int_{-\infty}^{Z}dZ_1\int_{-\infty}^{Z_1}dZ_2[\underline{H}(Z_1),\underline{H}(Z_2)]\,, \tag{7}$$

etc. The nth term is a sum of integrals of n-fold multiple commutators of $\underline{H}$. The $\underline{A}^{(n)}$ are each anti-Hermitian, so that Eq. (4) gives a unitary approximation no matter where the infinite series (5) is truncated. The first-order term represents instantaneous effects, while the second- (and higher-) order terms introduce time correlation (recalling that time $\sim Z/v$ along the trajectory). Retaining only the $n=1$ term is exactly equivalent to the Callaway-Bauer method,[13,14] as mentioned in the Introduction.

In order to proceed with the evaluation of the A matrix, we introduce the dipole approximation[9]

$$V(\mathbf{R};\mathbf{r})\approx\frac{4\pi}{3}e^2\sum_{i=1}^{N}\sum_{m=-1}^{1}\frac{Y_{1m}^*(\hat{\mathbf{R}})}{R^2}r_iY_{1m}(\hat{\mathbf{r}}_i) \tag{8}$$

with $\mathbf{R}$ the position vector of the incident electron (for a straight-line path $R^2=\rho^2+Z^2$) and the target taken to have N electrons. It should be noted that there are actually two separate approximations involved in the use of Eq. (8). The first is to assume that the dipole term gives the dominant contribution in the multipole expansion of the Coulomb potential. Second, it is assumed that only distant collisions, for which $R>r_i$ (all i), are important. This latter condition is also consistent with the use of straight-line trajectories intrinsic to the impact-parameter approach. Substituting Eqs. (8) and (2) into the first- and second-order matrices, Eqs. (6) and (7), and collecting terms, we find

$$A_{\nu\mu}^{(1)}(\infty,-\infty)=\frac{1}{i\hbar}\frac{4\pi}{3}e^2\sum_i\sum_m\frac{\langle\nu\,|\,r_iY_{1m}(\hat{\mathbf{r}}_i)\,|\,\mu\rangle}{(v_\nu v_\mu)^{1/2}}\times T_m^{(1)}(\beta_{\mu\nu})\,, \tag{9}$$

$$A_{\nu\mu}^{(2)}(\infty,-\infty)=\frac{1}{2}\left[\frac{1}{i\hbar}\frac{4\pi}{3}e^2\right]^2\times\sum_{i,j}\sum_{\alpha}\sum_{mm'}\frac{\langle\nu\,|\,r_iY_{1m}(\hat{\mathbf{r}}_i)\,|\,\alpha\rangle}{(v_\nu v_\alpha)^{1/2}}\times\frac{\langle\alpha\,|\,r_jY_{1m'}(\hat{\mathbf{r}}_j)\,|\,\mu\rangle}{(v_\alpha v_\mu)^{1/2}}\times T_{m,m'}^{(2)}(\beta_{\alpha\nu},\beta_{\alpha\mu}) \tag{10}$$

with

$$T_m^{(1)}(\beta_{\mu\nu})=\int_{-\infty}^{\infty}dZ_1\frac{Y_{1m}^*(\hat{\mathbf{R}}_1)}{R_1^2}e^{i(k_\mu-k_\nu)Z_1}\,, \tag{11}$$

$$T_{m,m'}^{(2)}(\beta_{\alpha\nu},\beta_{\alpha\mu})=\int_{-\infty}^{\infty}dZ_1\frac{Y_{1m}^*(\hat{\mathbf{R}}_1)}{R_1^2}e^{i(k_\alpha-k_\nu)Z_1}\times\int_{-\infty}^{Z_1}dZ_2\frac{Y_{1m'}^*(\hat{\mathbf{R}}_2)}{R_2^2}e^{i(k_\mu-k_\alpha)Z_2}-\int_{-\infty}^{\infty}dZ_1\frac{Y_{1m'}^*(\hat{\mathbf{R}}_1)}{R_1^2}e^{i(k_\mu-k_\alpha)Z_1}\times\int_{-\infty}^{Z_1}dZ_2\frac{Y_{1m}^*(\hat{\mathbf{R}}_2)}{R_2^2}e^{i(k_\alpha-k_\nu)Z_2}\,, \tag{12}$$

where $\beta_{\mu\nu}=(k_\mu-k_\nu)\rho$ and α represents the complete set of intermediate states. An important feature of the A-matrix elements is that each term factors into two distinct parts, one depending on the properties of the target atom and the other describing the dynamics of the collision. This results directly from the use of the asymptotic form of the dipole potential (see also Ref. 12). Thus, while the factor involving the target will depend upon the particular atomic system of interest, the dynamical part of the problem can be solved in general by evaluating the collision integrals $T^{(1)}$ and $T^{(2)}$.

Recalling that $R^2=\rho^2+Z^2$, $Y_{10}^*(\hat{\mathbf{R}})=(3/4\pi)^{1/2}(Z/R)$, and $Y_{1\pm1}^*(\hat{\mathbf{R}})=\mp(3/8\pi)^{1/2}(\rho/R)$, where we have neglect-

ed the Φ dependence consistent with Eqs. (1) and (2), we can integrate Eq. (11) to obtain

$$T_m^{(1)}(\beta_{\mu\nu})=\left[\frac{3}{\pi(1+|m|)}\right]^{1/2} e^{i\pi(m+1)/2}\frac{1}{\rho}\beta_{\mu\nu}K_m(\beta_{\mu\nu}) , \tag{13}$$

where K_m is the modified Bessel function of order m. Using a contour integration technique, Klarsfeld[18] has been able to calculate the integral

$$J(\gamma_1,\gamma_2)=\tfrac{1}{2}\int_{-\infty}^{\infty} dX_1 \frac{e^{i\gamma_1 X_1}}{(1+X_1^2)^{3/2}} \int_{-\infty}^{X_1} dX_2 \frac{e^{-i\gamma_2 X_2}}{(1+X_2^2)^{3/2}} \tag{14}$$

in closed form, and with some manipulation the $T^{(2)}_{m,m'}$ can be expressed in terms of this J integral. To simplify the notation, we first set $\beta_1=\beta_{\alpha\nu}$ and $\beta_2=\beta_{\alpha\mu}$. We also note that the various m,m' combinations reduce to four independent expressions, since $T^{(2)}_{-1,-1}=T^{(2)}_{1,1}$, $T^{(2)}_{1,-1}=T^{(2)}_{-1,1}=-T^{(2)}_{1,1}$, $T^{(2)}_{-1,0}=-T^{(2)}_{1,0}$, and $T^{(2)}_{0,-1}=-T^{(2)}_{0,1}$. Due to certain symmetry relations (see below), only two cases need be considered: (1) $\beta_1>0$, $\beta_2<0$ and (2) $\beta_1\geq\beta_2>0$. Using Klarsfeld's results,[18] we find for case (1)

$$\begin{aligned}
T^{(2)}_{1,1}(\beta_1,\beta_2)&=\frac{3i}{8}\frac{1}{\rho^2}(\beta_1+\beta_2)e^{-(\beta_1-\beta_2)} ,\\
T^{(2)}_{1,0}(\beta_1,\beta_2)&=\frac{3}{4\sqrt{2}}\frac{1}{\rho^2}[1+(\beta_1+\beta_2)]e^{-(\beta_1-\beta_2)} ,\\
T^{(2)}_{0,1}(\beta_1,\beta_2)&=\frac{-3}{4\sqrt{2}}\frac{1}{\rho^2}[1-(\beta_1+\beta_2)]e^{-(\beta_1-\beta_2)} ,\\
T^{(2)}_{0,0}(\beta_1,\beta_2)&=\frac{-3i}{4}\frac{1}{\rho^2}(\beta_1+\beta_2)e^{-(\beta_1-\beta_2)}
\end{aligned} \tag{15}$$

and for case (2)

$$\begin{aligned}
T^{(2)}_{1,1}(\beta_1,\beta_2)&=\frac{3i}{2}\frac{1}{\rho^2}[\tfrac{1}{4}(\beta_1+\beta_2)e^{-(\beta_1-\beta_2)}\\
&\qquad -\beta_1\beta_2K_1(\beta_1)I_1(\beta_2)] ,\\
T^{(2)}_{1,0}(\beta_1,\beta_2)&=\frac{3}{\sqrt{2}}\frac{1}{\rho^2}\{\tfrac{1}{4}[1+(\beta_1+\beta_2)]e^{-(\beta_1-\beta_2)}\\
&\qquad -\beta_1\beta_2K_1(\beta_1)I_0(\beta_2)\} ,\\
T^{(2)}_{0,1}(\beta_1,\beta_2)&=\frac{-3}{\sqrt{2}}\frac{1}{\rho^2}\{\tfrac{1}{4}[1-(\beta_1+\beta_2)]e^{-(\beta_1-\beta_2)}\\
&\qquad +\beta_1\beta_2K_0(\beta_1)I_1(\beta_2)\} ,\\
T^{(2)}_{0,0}(\beta_1,\beta_2)&=-3i\frac{1}{\rho^2}[\tfrac{1}{4}(\beta_1+\beta_2)e^{-(\beta_1-\beta_2)}\\
&\qquad -\beta_1\beta_2K_0(\beta_1)I_0(\beta_2)] ,
\end{aligned} \tag{16}$$

where again K_0, K_1, I_0, and I_1 are modified Bessell functions. For all other cases we apply the symmetry relations

$$\begin{aligned}
T^{(2)}_{m,m'}(-\beta_1,-\beta_2)&=[T^{(2)}_{m,m'}(\beta_1,\beta_2)]^*\\
T^{(2)}_{m,m'}(\beta_2,\beta_1)&=-[T^{(2)}_{m',m}(\beta_1,\beta_2)]^*
\end{aligned} \tag{17}$$

which can be most easily derived by making the change of variables $y_{2,1}=\frac{1}{2}(X_1\pm X_2)$, with $X=Z/\rho$, in the original $T^{(2)}$ integral. Equations (15)—(17) have checked for a variety of cases by comparison with direct numerical integration of Eq. (12).

Having specified the A matrix, we must now exponentiate it according to Eq. (4). The standard diagonalization procedure is used for this purpose (see, for example, Ref. 13). Briefly, we first form the Hermitian matrix $\underline{A}'=i\underline{A}$, which is diagonalized by the unitary transformation $\underline{U}^\dagger\underline{A}'\underline{U}=\underline{\Lambda}$. The elements of $\underline{\Lambda}$ are the eigenvalues (λ_p) of $\underline{A}'$, while $\underline{U}$ is constructed from the properly ordered, normalized eigenvectors of $\underline{A}'$. We can then write $e^{\underline{A}}=e^{-i\underline{A}'}=\underline{U}e^{-i\underline{\Lambda}}\underline{U}^\dagger$, where the elements of $e^{-i\underline{\Lambda}}$ are given by $e^{-i\lambda_p}\delta_{p,q}$. The major numerical effort is therefore reduced to the diagonalization of a Hermitian matrix $\underline{A}'$ for each value of the impact parameter ρ.

Finally, we consider the calculation of the cross section from Eq. (3). Although the dipole potential of Eq. (8) is singular as $R\to 0$, the use of the unitary, exponential solution prevents the cross section from diverging, and, in principle, no lower cutoff on the impact parameter need be invoked.[13] However, even though the cross section does indeed remain finite for an arbitrarily small lower limit on the ρ integration, it will not in fact converge to the Born approximation at high energies. We have therefore chosen to replace the lower limit of integration in Eq. (3) by a value of ρ_0, determined such that the cross section calculated by the present method agrees with the Born approximation at high energies.[9,12] Although the use of a cutoff is perhaps not entirely satisfactory, our prescription for specifying its value is at least well defined and relatively easy to implement. Furthermore, for many strong-coupling cases (see Sec. III) the cross section is only weakly dependent on ρ_0. The unitary property of the Magnus approximation, of course, obviates the need for any additional strong-coupling cutoff, as is required in perturbation theory.[9]

III. RESULTS AND DISCUSSION

In this section results are presented for electron-impact excitation of the transitions Li($2s\to 2p$), Na($3s\to 3p$), and K($4s\to 4p$), which should provide a good testing ground for the theory. Since we are using a semiclassical approach, we have concentrated primarily on the intermediate-energy regime, defined loosely as being above the ionization threshold but below the region where the Born approximation becomes valid. For the lighter alkali metals, this would imply incident energies $\gtrsim 5$ eV, but somewhat more conservatively the lower limit of the intermediate-energy range for these atoms is usually taken to be about 10 eV according to the literature.[1]

Before describing the calculations, it is useful to consider the main qualitative features of the problem. Excita-

tion of the resonance transitions of the alkali metals is dominated by the very strong s-p dipole interaction. Back coupling (i.e., $s \rightarrow p \rightarrow s$) must therefore be taken into account in order to obtain reasonable results, which in turn requires a theory that ensures conservation of probability. The failure of perturbation theory in this regard is demonstrated by the fact that the Born approximation gives peak cross sections which are a factor of about 3 too high for the lighter alkali metals.[19] Indirect coupling to intermediate states also proves to be important. Extensive quantum-mechanical close-coupling calculations have been carried out[3,20] for Na and K to investigate the effects of including various intermediate levels. It was found that by far the most important indirect process is $3s \rightarrow 3p \rightarrow 3d$ for Na, and similarly $4s \rightarrow 4p \rightarrow 3d$ for K, which transfers population from the p to the d state, thereby leading to a sizable reduction in the $s \rightarrow p$ cross section. Only minor changes resulted from the addition of more states to the close-coupling expansion. In addition, exchange effects are small at intermediate energies. The problem is therefore basically controlled by the intense s-p and p-d dipole interactions and should thus be amenable to the present theoretical treatment.

From the close-coupling results of Refs. 3 and 20, we know that a three-state expansion is adequate. Our calculations therefore included the basis states: $2s,2p,3d$ for Li; $3s,3p,3d$ for Na; and $4s\,4p,3d$ for K. Two additional approximations were made. The first was to consider the alkali metals to be one-electron systems (i.e., the so-called frozen core approximation), which eliminates the sums over i and j in Eqs. (9) and (10). Second, the sum over intermediate levels, α, in Eq. (10) was restricted to the same basis states as were used in the coupled-state expansion. This latter approximation is justified *a posteriori* by the fact that second-order effects turn out to be small in the energy range of interest. The numerical input required to perform the calculations consists of the energy levels, the dipole matrix elements, and the impact-parameter cutoff, ρ_0. Energy levels and oscillator strengths were taken from the Natl. Bur. Stand. (U.S.) tables.[21–23] The sign of the matrix element cannot be obtained from the value of the oscillator strength; to determine the sign we therefore used the Bates-Damgaard method[24] as corrected by Bebb.[25] The ρ_0 parameter was found by matching the calculated cross section to the Born approximation at energies of about 1 keV, the Born values being taken from the papers of Gallagher and co-workers.[2,26,27] These various quantities are given in Table I. It is interesting to compare ρ_0 with the physical size of the atom, which can be estimated from the radial expectation value $\langle r \rangle$ of the valence, s electron. From relativistic Hartree-Fock-Slater calculations,[28] $\langle r \rangle = 3.75$, 4.02, and 4.94 (in units of the Bohr radius) for Li, Na, and K, respectively, which are within a few percent of the ρ_0 values listed in the table. Integration over the impact parameter [see Eq. (3)] was carried out with gradually increasing step size from ρ_0 to a value ρ_{max}, typically $\sim 40a_0$, beyond which we used a perturbation-theory solution. The range $\rho_{max} \leq \rho \leq \infty$ contributed only a few percent to the total integral.

TABLE I. Dipole matrix elements and impact-parameter cutoff values ρ_0 for Li, Na, and K.

	Radial dipole matrix elements (in atomic units) $\langle s \mid r \mid p \rangle$	$\langle p \mid r \mid d \rangle$	Impact-parameter cutoff ρ_0 (in units of the Bohr radius)
Li	−4.08	4.48	3.65
Na	−4.36	−5.79	3.87
K	−5.02	7.22	4.84

The cross sections are shown in Figs. 1–3, where QB represent the Born approximation,[19] $Q2$ stands for the present dipole Magnus approximation (DMA) with a two-state (s,p) expansion, and $Q3$ represents the DMA with the three-state (s,p,d) expansion. Results are given for both first-order (solid $Q2$ and $Q3$ curves) and second-order (dashed $Q2$ and $Q3$ curves) calculations. The other symbols represent the experimental data, which will be discussed below. It should be noted that the standard (nonunitary) impact-parameter solution to Eq. (1) was found to reproduce the Born approximation (QB) curves in Figs. 1–3 to $\sim 10\%$, and we have therefore not explicitly included these values.

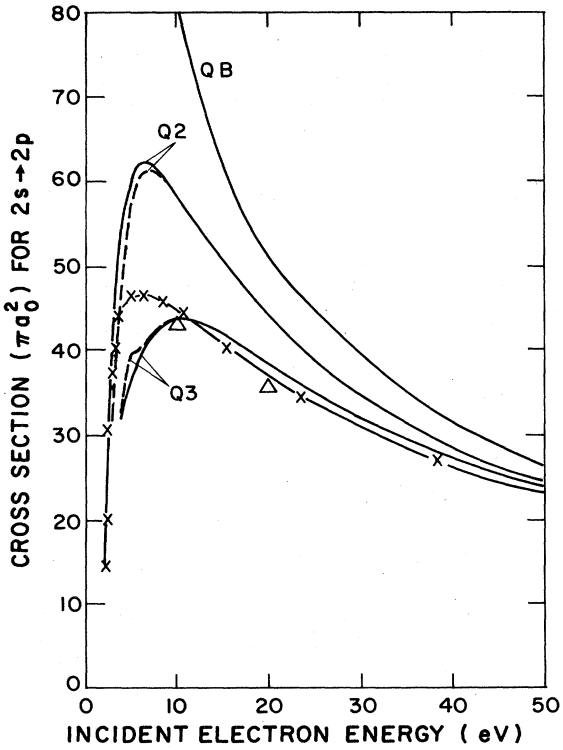

FIG. 1. Electron-impact excitation cross section for the resonance transition of lithium. (——, QB) Born approximation (Ref. 19); (——, $Q2$) present theory, two-state, first-order; (— — —, $Q2$) present theory, two-state, second-order; (——, $Q3$) present theory, three-state, first-order; (— — —, $Q3$) present theory, three-state, second-order; (—— × ——) data of Leep and Gallagher (Ref. 27); (△) data of Vuskovic, Trajmar, and Register (Ref. 6).

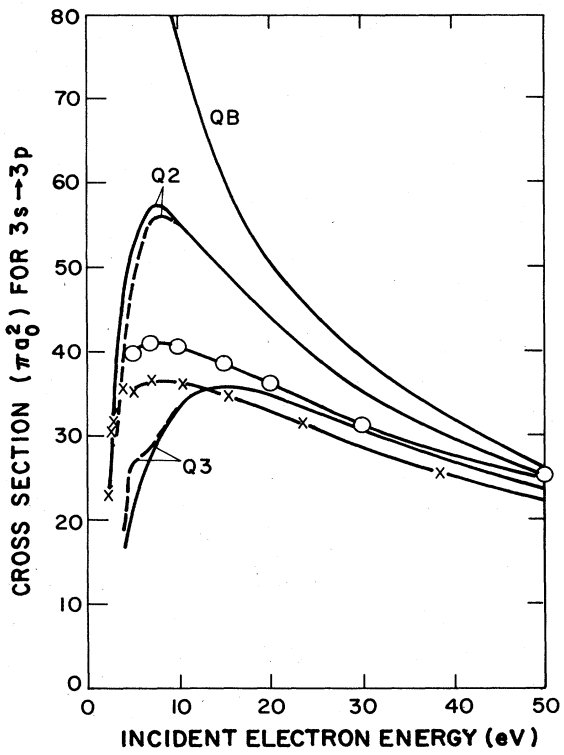

FIG. 2. Electron-impact excitation cross section for the resonance transition of sodium. (——×——) data of Enemark and Gallagher (Ref. 26); (——○——) data of Phelps and Lin (Ref. 5); all other curves have the same meaning as in Fig. 1.

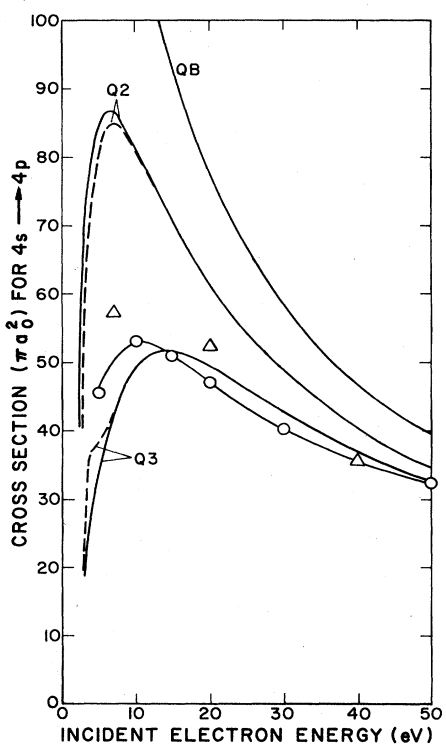

FIG. 3. Electron-impact excitation cross section for the resonance transition of potassium. (——○——) data of Phelps *et al.* (Ref. 3); (△) data of Vuskovic and Srivastava (Ref. 4); all other curves have the same meaning as in Fig. 1.

The results display a number of general features, common to all three cases. In agreement with our earlier qualitative discussion, the DMA results lie consistently below the Born approximation, which makes no allowance for conservation of probability. Coupling to the intermediate $3d$ state leads to a substantial reduction in the cross section (comparing $Q3$ with $Q2$). This effect has also been studied using the quantum-mechanical close-coupling method,[3,20] where it was found that including the $3d$ state led to a decrease in the cross section of 22% for Na and 26% for K, both at 10.52 eV. The corresponding values for the DMA are 38% for both Na and K. The present theory therefore tends to overestimate the importance of the $s \rightarrow p \rightarrow d$ coupling process. Second-order effects lead to slightly improved results at low energies but are essentially negligible beyond 10 eV. The three-state, second-order cross sections are observed to develop a shoulderlike structure in the low-energy region, which presumably would be smoothed out by the addition of higher-order terms.

We now wish to compare the present cross sections with the experimental data and with other theoretical results in the literature. Both theory and experiments for electron-impact excitation of the lighter alkali-metal atoms at intermediate energies have been reviewed through 1977.[1] We have chosen to use the data of Gallagher and co-workers[2,26,27] as a standard, since they appear to have withstood the test of time; other pre-1977 measurements are summarized in Ref. 1. More recent data[3–6] are shown in Figs. 1–3. For K, Chen and Gallagher[2] only gave the optical-excitation cross section, which included cascade contributions. Phelps *et al.*[3] also measured the optical-excitation function, which was found to be in good agreement with that of Chen and Gallagher, and were able to subtract out cascade effects to obtain the direct cross section, which is the appropriate quantity for comparison with the theory. The Chen and Gallagher optical-excitation function is therefore not shown in Fig. 3. The present theory (as represented by the $Q3$ curves) is in good agreement with the data for energies > 15 eV, while the accuracy deteriorates as one goes to lower energies. With respect to other theoretical treatments, the most extensive calculations at intermediate energies are those of Korff *et al.*[20] for Na and of Phelps *et al.*[3] for K. The multistate, close-coupling method was used in both cases, and good agreement with experiment was obtained. Several other sets of theoretical cross sections[29–31] are tabulated and reviewed in Ref. 1 and need not be reproduced here. In general, our cross sections are comparable to those calculated by more sophisticated methods throughout the intermediate-energy range. In particular, our results are quite close to those obtained in the Glauber approximation,[29] which bears some similarity to the semiclassical Magnus approach.

Finally, it is of interest to investigate the effect of varying the value of ρ_0. Calculations were therefore carried

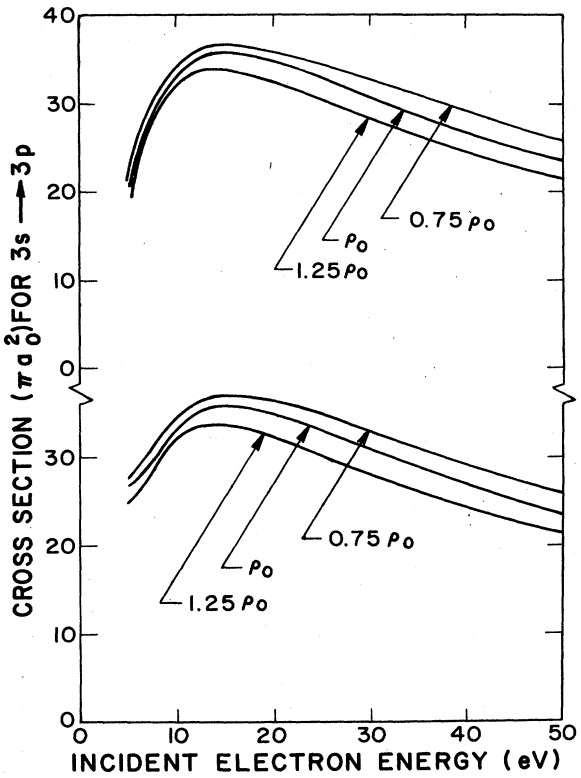

FIG. 4. Dependence of the sodium excitation cross section on variations in the impact-parameter cutoff ρ_0. The top set of curves were calculated in the three-state, first-order approximation, while the lower set are for the three-state, second-order approximation.

out for Na, using the three-state, first- and second-order approximations, with variations in ρ_0 of $\pm 25\%$ about the value specified in Table I. The resulting cross sections, shown in Fig. 4, do not change by more than $\pm 10\%$. This implies that the results do not depend sensitively on the details of the interaction at small distances ($\sim \rho_0$), which in turn suggests that the use of the asymptotic form of the dipole interaction is probably not a serious approximation, at least in the strong-coupling case.

IV. SUMMARY AND CONCLUSIONS

The Magnus approximation, to second order, has been used to solve the coupled-channel, symmetrized impact-parameter equations for electron-atom scattering. The method satisfies unitarity and accounts for a distribution of probability into the various open channels of the system. Retaining only the asymptotic dipole part of the Coulomb interaction allows the collision integrals to be written in closed, analytic form. It thus becomes feasible to handle problems involving a large number of closely coupled states.

The theory has been applied to the calculation of electron-impact excitation cross sections for the resonance transitions of Li, Na, and K. An extensive data base, both experimental and theoretical, exists for these atoms, making possible a detailed evaluation of the present approach. In addition, many of the characteristics of this fairly simple problem (e.g., long-range dipole coupling) make it representative of the more general processes of interest. The calculated cross sections were found to be in good agreement with experimental data over most of the intermediate-energy range. Second-order, time correlation effects turn out to be small. Much more important is coupling to the intermediate $3d$ state, although the theory tends to overestimate the magnitude of this effect. Nevertheless, at least for the lighter alkali metals, the present method is competitive with more complex and sophisticated approaches.

In its present form the theory can be readily applied to more complicated processes, such as inelastic electron scattering from atoms in excited states. This is an important area, where, due both to experimental and theoretical difficulties, relatively little work has been done to date. For excited atoms, contributions from states lying in the continuum may become significant. The form of the second-order A-matrix elements [see Eq. (10)] makes it straightforward to implicitly account for the continuum by using the usual closure relation together with an average excitation energy. As far as improvements and extensions to the theory are concerned, its principal drawback is the restriction to dipole interactions. A possible approach to overcome this limitation might be to use the complete Coulomb interaction in the evaluation of the first-order A-matrix elements, as has been done by Baye and Heenen,[15] but to retain the asymptotic dipole approximation for the second-order elements. As long as second-order effects do not become too large, this type of hybrid theory should be more accurate and more widely applicable than the present method without an inordinate sacrifice in computational efficiency.

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