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Authors
Mittleman, Marvin H.
Watson, Kenneth M.

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UNIVERSITY OF CALIFORNIA

Ernest O. Lawrence

Radiation Laboratory

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BY ATOMS AT HIGH ENERGIES[*]

Marvin H. Mittleman

Lawrence Radiation Laboratory
University of California
Livermore, California

and

Kenneth M. Watson

Lawrence Radiation Laboratory and Department of Physics
University of California
Berkeley, California

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ABSTRACT

Some consequences of the Pauli principle for the elastic scattering of electrons by atoms are studied. The contributions both from the exchange integrals and from the Hartree-Fock condition that the scattered wave be orthogonal to the bound-state wave functions are expressed in a simple approximate form. For high-energy electrons these corrections are very small.

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I. INTRODUCTION

In two previous publications[1a,1b] we have introduced a method for treating the scattering of charged particles by atoms with the use of an equivalent scattering potential. In the present discussion we apply this method to a study of the role of the Pauli principle in high-energy electron scattering by neutral atoms. In a later paper the corresponding problems will be discussed for low-energy electrons.

It is recognized that the Pauli principle plays a secondary part in scattering at high energies. On the other hand, the possibility of an analytic discussion in the limit of high energy permits one to obtain some insight into the effects of electron exchange and into its relative importance. Much of our discussion is also relevant for scattering at low energies.

II. FORMULATION

The scattering of an electron having initially a momentum $\underset{\sim}{p}_0$ and an energy ϵ_0 is considered. The scatterer is a neutral atom having Z electrons. The kinetic-energy operator for the scattered electron is

$$K = -\frac{\hbar^2}{2m}\nabla^2 \quad , \tag{1}$$

where m is the electronic mass. The scattering atom is described in the Hartree-Fock approximation, the orbital states for the bound electrons being written as $g_\nu (\nu = 1, 2, \cdots Z)$.

The wave function $\emptyset(x)$ of the scattered electron satisfies the Schrodinger equation[1]

$$[K + v - \epsilon_0]\emptyset = \sum_{\nu=1}^{Z} \lambda_\nu g_\nu \quad . \tag{2}$$

Here v is the effective scattering potential and the λ_ν are Lagrangian multipliers chosen to establish the condition that $\emptyset$ be orthogonal to the g_ν's. That is, the conditions

$$(g_\nu, \emptyset) = 0 , \qquad \nu = 1, 2, \cdots Z \tag{3}$$

determine the λ_ν's just as for the Hartree-Fock equations.

To second order (see 1a, 1b) the potential v is

$$v = v_1 + v_{1\,ex} + v_2 \ , \tag{4}$$

where

$$v_1(x) = \int d^3y \ [\sum_\nu g^*_\nu(y) \ g_\nu(y)] \frac{e^2}{|\underset{\sim}{x} - \underset{\sim}{y}|} - \frac{Z \ e^2}{x} \ , \tag{5}$$

$$v_{1\,ex}(x) = - \int d^3y \ \sum_\nu g^*_\nu(y) \ [\frac{e^2}{|\underset{\sim}{x} - \underset{\sim}{y}|} \ g_\nu(x) \ ]P_{xy} \ , \tag{6}$$

and

$$v_2 = - \frac{e^2 \ \alpha}{(x^2 + d^2)^2} \quad . \tag{7}$$

Here P_{xy} is an operator which interchanges $\underset{\sim}{x}$ and $\underset{\sim}{y}$, to give $P_{xy}\phi(\underset{\sim}{x}) = \phi(\underset{\sim}{y})$, etc. In Eq. (7) α is the atomic polarizability and may be taken approximately as[2]

$$\alpha = \frac{4}{9} \frac{1}{a} \sum_{i=1}^{Z} (\bar{x}_i^{\,2})^2 , \tag{8}$$

where $a = \dfrac{\hbar^2}{me^2}$ is the Bohr radius and $\bar{x}_i^{\,2}$ is the mean-square distance from the nucleus of the $\underline{i}$th orbital electron. Rather than to use the variational expression[1] to obtain v_2, we have introduced the more customary cutoff parameter d. An approximate value of d is given by[3]

$$d^4 \simeq a \, \alpha \, Z^{-1/3} . \tag{9}$$

Hammerling, Shine and Kivel[4] have used an expression similar to Eq. (4) for v. They have replaced $v_{1\ ex}$ by an approximation due to Slater, however.[5]

The scattering matrix for a final momentum $\underset{\sim}{p}$ for the electron is

$$T = (X_p, v \phi) , \tag{10}$$

where

$$X_p = (2\pi)^{-3/2} e^{i \underset{\sim}{p} \cdot \underset{\sim}{x}} . \tag{11}$$

Finally, the differential scattering cross section is

$$\sigma = (2\pi)^4 m^2 |T|^2 . \tag{12}$$

III. SIMPLIFICATION OF $v_{1\ ex}$

For high-energy electron scattering, the principal contributions to exchange corrections come from the interior of the atom. For this reason,

we shall use the Fermi-Thomas approximation in obtaining the exchange term.

Consider now that contribution to T in Eq. (10) which is

$$T_{1\ ex} = (\chi_p, \ v_{1\ ex}\ \emptyset)$$

$$= -\sum_{\nu=1}^{Z} \int d^3x\ d^3y\ \chi_p^*(x)\ [\ \tfrac{1}{2}\ g_\nu(x)\ g_\nu^*(y)\ ]\ \frac{e^2}{|\underset{\sim}{x} - \underset{\sim}{y}|}\ \emptyset(y)\ .$$

$$(13)$$

Rather than to introduce spin wave functions in this equation, we have simply inserted a factor of $\tfrac{1}{2}$ before $g_\nu g_\nu^*$ to take account of the fact that we should have kept only those orbitals ν for which the spin is parallel to that of the incident electron.

Now, in the Fermi-Thomas approximation,

$$\tfrac{1}{2} \sum_{\nu=1}^{Z} g_\nu(x)\ g_\nu^*(y) \approx \int\limits_{\ell< P_F(y)} \frac{d^3\ell}{(2\pi)^3}\ e^{i\ \underset{\sim}{\ell}\cdot(\underset{\sim}{x} - \underset{\sim}{y})}\ . \qquad (14)$$

Here $P_F(y)$ is the Fermi momentum at $\underset{\sim}{y}$. If we replace the variable $\underset{\sim}{x}$ by $\underset{\sim}{\rho} \equiv \underset{\sim}{x} - \underset{\sim}{y}$ and observe

$$\chi_p^*(x) = \chi_p^*(y)\ e^{-i\ \underset{\sim}{p}\cdot\underset{\sim}{\rho}}\ ,$$

there results

$$T_{1\ ex} = -\int d^3y\ \chi_p^*(y) \left\{ \int d^3\rho \int\limits_{\ell< P_F(y)} \frac{d^3\ell}{(2\pi)^3}\ e^{i(\underset{\sim}{\ell} - \underset{\sim}{p})\cdot\underset{\sim}{\rho}}\ \frac{e^2}{\rho} \right\} \emptyset(y)\ .$$

We now define

$$I(y) \equiv -\int\limits_{\ell< P_F(y)} \frac{d^3\ell}{(2\pi)^3} \int d^3\rho\ e^{i(\underset{\sim}{\ell} - \underset{\sim}{p})\cdot\underset{\sim}{\rho}}\ \frac{e^2}{\rho}$$

$$(15)$$

$$= -\frac{2e^2}{(2\pi)^2} \int\limits_{\ell< P_F(y)} \frac{d^3\ell}{(\underset{\sim}{\ell} - \underset{\sim}{p})^2}$$

Thus,

$$T_{1\ ex} = \int d^3y\ \chi^*_p(y)\ I(y)\ \emptyset(y)$$

(16)

$$= (\chi_p,\ I\ \emptyset)\ .$$

Since we anticipate that I will be small at high energies, we shall keep only first-order terms in I in Eq. (16), and thus may neglect $v_{1\ ex}$ in calculating $\emptyset$.

A better approximation may be obtained as follows, however. A typical matrix element of $v_{1\ ex}$ in the perturbation expansion of $\emptyset$ is

$$(\underset{\sim}{k}\ |\ v_{1\ ex}\ |\ \underset{\sim}{k}'\ ) = (\chi_k,\ v_{1\ ex}\ \chi_{k'})\ .$$

At high energies $k \simeq p$, however. Thus, following the steps leading from Eq. (13) to Eq. (16), we obtain

$$(\underset{\sim}{k}\ |\ v_{1\ ex}\ |\ k'\ ) \approx (\chi_k,\ I\ \chi_{k'})\ .$$

This means that under the stated conditions we may replace the interaction $v_{1\ ex}$ in Eq. (4) by I. Then Eq. (4) is replaced by

$$v \approx v_1 + I + v_2\ .$$

(17)

The nonlocal interaction $v_{1\ ex}$ is now replaced by a simple potential $I(y)$.

Were we to average Eq. (15) over all momenta p within the Fermi sea, the resulting $I(y)$ would reduce to the Slater potential.[5] This procedure seems to have little justification for the scattering problem, since p is the momentum of the incident particle and is quite unrelated to the momenta within the Fermi sea.

Evaluation of the integral (15) leads to

$$I(y) = -\frac{e^2}{\pi p} \left\{ p\, P_F(y) - \frac{1}{2} [\, p^2 - P_F^{\,2}(y)\,]\, \ln \left| \frac{p + P_F(y)}{p - P_F(y)} \right| \right\} .$$

$$(18)$$

In the limit with $p \gg P_F$, this reduces to

$$I(y) \approx -\frac{e^2}{(2\pi)^2\, p^2}\, n_e(y) ,$$

$$(19)$$

where n_e is the electron density at y .

IV. THE CONDITION OF ORTHOGONALITY FOR $\emptyset$

We now turn our attention to the solution of Eq. (2). This has the form

$$\emptyset = \emptyset_0 + \sum_{\nu=1}^{Z} \lambda_\nu\, \emptyset_\nu .$$

$$(20)$$

Here $\emptyset_0$ is the solution of the homogeneous equation

$$[K + v - \epsilon_0]\emptyset_0 = 0 .$$

$$(21)$$

That is, $\emptyset_0$ is the solution of the scattering problem if we ignore the orthogonality condition (3).

The $\emptyset_\nu$'s are solutions of the inhomogeneous Schrodinger equations

$$[K + v - \epsilon_0]\emptyset_\nu = g_\nu , \qquad \nu = 1, 2, \cdots Z .$$

$$(22)$$

It has been argued above that the Fermi-Thomas approximation may be used, since the inner-shell electrons contribute the most to our exchange

corrections. Since by Eq. (17) v is now a local potential, we may solve Eq. (22):

$$\emptyset_{\nu}(x) = \frac{1}{[K(\ell) + v(x) - \epsilon_0]} \, g_{\nu}(x) \, . \qquad (23)$$

Here $\underset{\sim}{\ell}$ is the momentum of the νth orbital electron at the point $\underset{\sim}{x}$, therefore $K(\ell) = \ell^2/2m$. We recall that it is in the spirit of the Fermi-Thomas and WKB approximations to assign a local momentum in the vicinity of a point in space. Indeed, we may suppose that the volume of the atom is divided into small elements $\delta\tau_j$ $(j = 1, 2, \cdots)$. In each of these volume elements we introduce a set of plane-wave states

$$g_{\ell,j} = \frac{1}{\sqrt{\delta\tau_j}} \, e^{i \, \underset{\sim}{\ell} \cdot \underset{\sim}{x}} \, .$$

The index ν in Eq. (23) then specifies both $\underset{\sim}{\ell}$ and j .

From the above discussion, we see that in our approximation the $\emptyset_{\nu}$'s have the form

$$\emptyset_{\nu} = \frac{1}{[K(\ell) + v_j - \epsilon_0]} \, g_{\ell,j} \, .$$

Thus $\emptyset_{\nu}$ is __proportional__ to g_{ν} and we have the condition

$$\Lambda^0 \, \emptyset_{\nu} \simeq \emptyset_{\nu} \quad , \qquad (24)$$

where

$$(x \mid \Lambda^0 \mid x') \equiv \sum_{\nu} g_{\nu}(x) \, g_{\nu}^*(x') \qquad (25)$$

is the projection operator on the ground-state orbitals. Equation (23) is not valid outside the atom, of course.

From Eqs. (3), (20), and (25) we now obtain

$$\Lambda^0 \phi = 0 = \Lambda^0 \phi_0 + \sum_{\nu} \lambda_{\nu} \phi_{\nu} \quad ,$$

or   (26)

$$\sum_{\nu} \lambda_{\nu} \phi_{\nu} = - \Lambda^0 \phi_0 \quad .$$

With this result we can write Eq. (20) as

$$\phi(x) = (1 - \Lambda^0)\phi_0 \quad .$$ (27)

This is not valid for x outside the atom, since Eq. (23) omits the asymptotic wave in ϕ_{ν} . Thus we cannot use Eq. (27) directly to find the scattered electron wave, but must instead insert Eq. (27) into Eq. (10).

Equation (10) now reads

$$T = (X_p, v(1 - \Lambda^0)\phi_0) \quad .$$ (28)

The exchange part is

$$T_{ex} = -(X_p, v \, \Lambda^0 \, \phi_0)$$ (29)

$$= - \int d^3x \; d^3y \; X_p^*(x) \, v(x) \, (x \mid \Lambda^0 \mid y) \, \phi_0(y) \quad .$$

From Eq. (14) we have

$$(x \mid \Lambda^0 \mid y) = \int_{\ell < P_F(y)} \frac{d^3\ell}{(2\pi)^3} e^{i \, \underset{\sim}{\ell} \cdot \underset{\sim}{\rho}}$$

and

$$X_p^* = X_p^*(y) \, e^{-i \underset{\sim}{p} \cdot \underset{\sim}{\rho}} \quad , \qquad \text{where} \quad \underset{\sim}{\rho} \equiv \underset{\sim}{x} - \underset{\sim}{y} \quad .$$

Thus

$$T_{ex} = - \int d^3y \ \chi^*_p(y) \left\{ \int_{\ell < P_F(x)} \frac{d^3\ell}{(2\pi)^3} \int d^3\rho \ e^{i(\underset{\sim}{\ell} - \underset{\sim}{p})\cdot\underset{\sim}{\rho}} \ v(\underset{\sim}{y} + \underset{\sim}{\rho}) \ \emptyset_0(y) \right\} . \tag{30}$$

Let us define

$$J(y) \equiv + \int_{\ell < P_F(y)} \frac{d^3\ell}{(2\pi)^3} \int d^3\rho \ e^{i(\underset{\sim}{\ell} - \underset{\sim}{p})\cdot\underset{\sim}{\rho}} \ v(\underset{\sim}{y} + \underset{\sim}{\rho}) \ , \tag{31}$$

so

$$T_{ex} = - \int d^3y \ \chi^*_p(y) \ J(y) \ \emptyset_0(y) \tag{32}$$

$$= - (\chi_p, \ J \ \emptyset_0) \ .$$

Thus

$$T = (\chi_p, \ [v - J]\emptyset_0) \ . \tag{33}$$

We may re-express this more concisely as follows. Let us introduce the Fourier transform $\bar{v}(k)$ of $v(x)$:

$$v(x) = (2\pi)^{-3/2} \int d^3k \ e^{i \underset{\sim}{k}\cdot\underset{\sim}{x}} \ \bar{v}(k) \ . \tag{34}$$

Then J may be written as

$$J(y) = (2\pi)^{-3/2} \int_{|q-p| < P_F(y)} d^3q \ e^{i \underset{\sim}{q}\cdot\underset{\sim}{y}} \ \bar{v}(q) \ . \tag{35}$$

Finally,

$$\Delta(y) \equiv v(y) - J(y)$$

$$= (2\pi)^{-3/2} \int_{|\underset{\sim}{q}-\underset{\sim}{p}|>P_F(y)} d^3q \ e^{i \ \underset{\sim}{q}\cdot\underset{\sim}{y}} \ \bar{v}(\underset{\sim}{q}) \tag{36}$$

defines an "equivalent potential" for Eq. (33).

For sufficiently high energies, J may be evaluated in a very simple way. We have

$$v(\underset{\sim}{y} + \underset{\sim}{\rho}) = v(\underset{\sim}{y}) + \underset{\sim}{\rho}\cdot\nabla \ v(\underset{\sim}{y}) + \cdots \ . \tag{37}$$

Now, $\rho \approx O(\frac{\hbar}{p})$, therefore for

$$\left| \frac{\hbar}{p} \frac{dv}{dy} \right| \ << \ v(y) \tag{38}$$

we may set

$$J = v(y) \int_{\ell < P_F(y)} d^3\ell \ \delta(\underset{\sim}{\ell} - \underset{\sim}{p})$$

$$= v(y) \quad \text{for} \quad p < P_F(y) \tag{39}$$

$$= 0 \quad \text{for} \quad p > P_F(y) \ .$$

The expression (33) is finally

$$T = \int_{p > P_F} d^3y \ \chi^*_p(y) \ v(y) \ \emptyset_0(y) \ . \tag{40}$$

The integral over y extends here only over those portions of the atom for which $p > P_F(y)$.

The condition (38) for the validity of the approximate Eq. (40) is

$$\frac{p^2}{\hbar^2} >> \left[\left(\frac{d \ln v(y)}{dy} \right)_{y_c} \right]^2 \quad , \tag{41}$$

where y_c is defined by

$$p = P_F(y_c) \quad . \tag{42}$$

For the Fermi-Thomas potential,

$$v_1(y) = - \frac{Z e^2}{y} X(s) \quad , \tag{43}$$

where

$$s = \frac{Z^{1/3}}{(0.88)a} y \tag{44}$$

and $a = \hbar/me^2$, as before. Here $X(s)$ is the dimensionless Fermi-Thomas potential function. We have then, from Eq. (41),

$$\frac{p^2/2m}{Ry} \equiv \frac{\epsilon_0}{Ry} >> \frac{Z^{2/3}}{[0.77]} \left[\left(\frac{d \ln \left(\frac{X(s)}{s} \right)}{ds} \right)_{s=s_c} \right]^2 \equiv r_0 \quad , \tag{45}$$

where

$$s_c = \frac{Z^{1/3}}{(0.88)a} y_c \quad , \tag{46}$$

and $Ry = e^2/2a$ is the Rydberg constant.

When the condition (45) is satisfied, we expect the approximate Eq. (40) to be valid.

V. APPLICATION TO THE BORN APPROXIMATION

In this section we illustrate the above with a calculation using the Born approximation and taking

$$X(s) = e^{-0.8 s} \ .$$
(47)

The condition (45) becomes now

$$\frac{\epsilon_0}{Ry} >> r_0 \equiv [\frac{Z^{2/3}}{0.77}](\frac{1}{s_c} + 0.8)^2 \ .$$
(48)

Values of s_c and r_0 are given in Tables I and II, respectively.

We must also, of course, satisfy the usual condition for the validity of the Born approximation, which is

$$\frac{Z e^2}{\hbar} (\frac{m}{p_0}) << 1 \ .$$

This may be rewritten as

$$\frac{\epsilon_0}{Ry} >> r_1 = Z^2 \ .$$
(49)

Values for r_1 are also shown in Table II. It is evident that, except for the lightest atoms, the condition (49) is more restrictive than is (48).

To simplify the integration, we replace Eq. (40) by introducing into the integrand the factor $(1 - e^{-y/y_c})$ and then integrating over all y:

$$T = \int d^3y \ \frac{e^{-i \underset{\sim}{\kappa} \cdot \underset{\sim}{y}}}{(2\pi)^3} (1 - e^{-y/y_c}) \ v(y) \ ,$$
(50)

where $\underset{\sim}{\kappa} \equiv \underset{\sim}{p} - \underset{\sim}{p_0}$. For v we use Eq. (17), with v_1 given by Eqs. (43) and (47), I given by Eq. (19), and v_2 given by Eq. (7).

TABLE I

Values for the Cutoff Radius

$\epsilon_0(\text{Ry})$	s_c			
	$Z = 10$	$Z = 18$	$Z = 36$	$Z = 54$
3	1.34	1.62	1.95	---
6	1.16	1.37	1.70	1.90
9	1.02	1.25	1.56	1.74
12	0.94	1.15	1.45	1.64
15	0.89	1.08	1.38	1.57
20	0.80	1.01	1.29	1.47
30	0.71	0.89	1.16	1.33
50	0.60	0.77	1.01	1.16
90	0.48	0.64	0.86	0.99

TABLE II

Values for the Limiting Energy Ratios v_0 and v_1

Z	v_0	v_1
10	25	100
18	31	324
36	39	1300
54	48	2900

The electron density in Eq. (19) is taken as

$$n_e \approx (0.1) \frac{Z^2}{a^3} \frac{e^{-1.2\,s}}{s^{3/2}} \quad . \tag{51}$$

An evaluation of Eq. (50) leads to

$$T = T_1 + T_2 + T_3 \quad . \tag{52}$$

Here T_1 arises from v_1. It is

$$T_1 = -\frac{Z\,e^2}{2\pi^2}\left(\frac{1}{\kappa_0^2 + \kappa^2}\right)\left\{1 - \frac{\kappa_0^2 + \kappa^2}{(\kappa_0 + \frac{1}{y_c})^2 + \kappa^2}\right\} \quad , \tag{53a}$$

where

$$\kappa_0 = \frac{Z^{1/3}\,\lambdabar}{(0.88)\,a} \quad . \tag{54}$$

The contribution T_2 comes from the potential term v_2. The exact integral may be expressed in terms of known functions, but the term associated with the "cut-off" ($-e^{-y/y_c}$) is cumbersome and not more than qualitative, anyway, since we have used a crude expression for v_2 at small distances. We thus give this part of T_2 only for the limit that $d \gg y_c$:

$$T_2 = -\frac{e^2\alpha}{8\pi d}\left\{e^{-\kappa d} - \frac{8}{\pi}\frac{(y_c\,d^3)^{-1}}{[(\frac{1}{y_c})^2 + \kappa^2]^2}\right\} \quad . \tag{53b}$$

Finally, T_3 arises from the expression I in v. It is

$$T_3 = -\frac{Z^{3/2} e^2}{5(2\pi)^4} \frac{(0.88 \, a)^{3/2}}{p^2 \, \kappa} \sqrt{\pi} \left\{ \text{Im} \left[\frac{1}{\sqrt{\frac{3}{2} \kappa_0 - i \kappa}} - \frac{1}{\sqrt{\frac{3}{2} \kappa_0 + \frac{1}{y_c} - i \kappa}} \right] \right\} .$$

(53c)

Here "Im[...]" means "imaginary part of."

The contributions to T which result from the Pauli principle "cut off" for $y < y_c$ are easily recognizable in Eqs. (53), since they involve the parameter y_c. The entire quantity T_3 represents an exchange correction, of course.

When the condition (49) is satisfied, the exchange corrections are seen to be small, except for large scattering angles (for which the scattering cross section is itself very small).

We are now in a position to compare the relative sizes of the various correction terms. First, the ratio of the second to the first Born approximation is

$$R \simeq Z \sqrt{\frac{Ry}{\epsilon_0}} .$$

(55)

The ratio of $\frac{T_3}{T_1} \equiv R \left(\frac{v_{ex}}{v_1} \right)$ is from Eqs. (53)

$$R \left(\frac{v_{ex}}{v_1} \right) \equiv \frac{T_3}{T_1} \sim Z^{2/3} \left(\frac{Ry}{\epsilon_0} \right)$$

(56)

$$= Z^{-1/3} \left(\frac{Ry}{\epsilon_0} \right)^{1/2} R .$$

Finally the ratio of the correction arising from J to that from v_1 is

$$R(\frac{J}{v_1}) \approx \kappa_0^2 y_c^2 \approx z^{4/3} (\frac{Ry}{\epsilon_0})$$

$$= z^{-1} R .$$

(57)

Thus, the exchange corrections seem never to exceed the corrections to the first Born approximation.

In the next Section we shall re-examine $R(J/v_1)$ in the classical, phase integral approximation.

VI. THE PHASE INTEGRAL APPROXIMATION

We consider again

$$T = \int d^3y \; \chi^*_p(y) \; [v(y) - J(y)]\emptyset_0(y) ,$$

(58)

with

$$\emptyset_0 = (2\pi)^{-3/2} \; e^{i \, P(y)} ,$$

(59)

where $P(y) = \int [p^2 - 2M \, v]^{1/2} \, ds$ is the phase integral along the classical orbit of the scattered electron. A simple consideration shows that only for those orbits for which the classical orbit is parallel to p and for which $v \approx 0$ does the product $[\chi^*_p \, \emptyset_0]$ not oscillate rapidly. This can happen only near the boundary of the atom, where, by Eq. (39), $J = 0$. Thus we obtain no correction from J in the phase integral approximation.

VII. CONCLUSIONS

We have obtained approximate expressions for two leading exchange corrections, expressed in terms of the "quasi-potentials" I and J. These two approximations were evaluated on the basis of the Fermi-Thomas model. At high energies these have a very small effect on the scattering. At lower energies, these expressions are still applicable if we are content with the Fermi-Thomas model. In this case a numerical evaluation of $\emptyset_0$ seems to be necessary, however.

FOOTNOTES AND REFERENCES

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