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Angular Distribution of the Outgoing Electrons in Electronic Ionization*

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The information which can be obtained from measurements on both the outgoing particles in single ionization is discussed for the case of high-energy electrons incident on neutral one- and two-electron atoms. The Born approximation is used to analyze a particular experiment suggested by studies of the nuclear $(p, 2p)$ reaction. It is concluded that the momentum distribution for the single-electron orbitals of atomic systems can be determined by experiments of this type. The effects of identity and correlations are investigated, especially for the case of target helium atoms.

I. INTRODUCTION

In the single ionization of an atomic system by an incident electron, two electrons are emitted. The differential cross section for this process, $\sigma\Delta\Omega_1\Delta\Omega_2\Delta E$, specifies the number of ionization events (per unit incident flux and target atom) in which the two outgoing electrons have directions within the solid angles $\Delta\Omega_1$ and $\Delta\Omega_2$ and a total energy within the range ΔE. No direct experimental information on this differential cross section exists, i.e., only integrated cross sections have been measured in which the kinematics of the outgoing electrons were not separately determined. A full understanding of the ionization process requires differential cross-section measurements of this more general type. In this paper, we analyze the possibilities that would be opened up by such measurements at high incident energies.

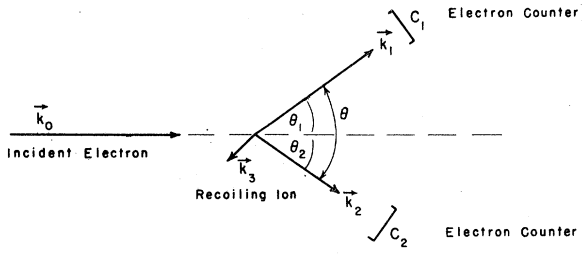

FIG. 1. Proposed experiment of the Chamberlain-Segrć type for single ionization of an atomic system by a high-energy electron. The angular variation of the coincidence counting rate is determined by the momentum distribution of the target system evaluated for $\vec{k}_1+\vec{k}_2-\vec{k}_0$.

Recently, we proposed a specific experiment of this type, using high-energy electrons, which could provide a test of the "direct interaction" mechanism in atomic reactions.[1] A schematic representation of the experiment is given in Fig. 1. An electron is incident on an atomic system with kinetic energy E_0 greater than the ionization potential B. Two electron detectors, C_1 and C_2, capable of determining direction and energy, are employed in coincidence. If the ionization occurs according to the direct interaction model, i.e., as a simple "knock-out" process, the angular distribution of the outgoing electrons will have special features. To see this most easily, consider the special case $E_0 \gg B$, so that the target electron may be considered to be at rest in first approximation. Then the two outgoing electrons are emitted at an angle of $\pi/2$ with respect to each other, since this is a collision between two particles of equal mass, one of which is initially at rest. Because the target electron does have a finite binding energy and is in motion, there will be a distribution in the separation angle θ. This distribution will be concentrated near the angle $\theta_0 = \pi/2 - B/E_0$ with an angular breadth $\Delta\theta \simeq (B/E_0)^{1/2}$.

This type of measurement was first made by Chamberlain and Segrć for the nuclear $(p, 2p)$ reaction.[2] Tyren, Maris, and Hillman made the important improvement of measuring the sum of the outgoing electrons' energies.[3] According to energy conservation

$$E_0 - B = (E_1 + E_2) + E_3 , \qquad (1.1)$$

if the recoil energy E_3 is ignored, the sum E_1+E_2 determines the binding energy of the knocked-out particle. Thus if the target system is described

by an independent-particle model – as is appropriate for atomic as well as nuclear physics – the outgoing electrons may be expected to be emitted in well-defined energy groups if the shell structure is well defined. This implies that the angular distribution of the outgoing particles should be studied as a function of the shell from which the target particle is knocked out. Experiments of this kind have already become an important tool in nuclear spectroscopy.[4]

In this paper we explore the results to be expected from measurements on both of the outgoing electrons in the atomic $(e,2e)$ reaction, i.e., single ionization by an incident electron. For purposes of simplicity in this first discussion, the analysis is restricted to high incident energies and to the experimental arrangement depicted in Fig. 1. Measurements of this type are of interest at any energy, although it is only at very high energies that a theoretical framework now exists for interpreting such experiments. Variations in the experimental arrangement should also be worthwhile. Not only might one of the counters be moved out of the plane of the other counter and the incident beam, but detection of the recoil ion, the use of projectiles other than electrons, and the use of various molecules and ions as targets, should be considered. Our primary objective at this time is to stimulate interest in the simplest experiment of this type, the $(e,2e)$ reaction at high energies with neutral atoms as the target.[5]

In Sec. II we use the crudest possible theory, the plane-wave Born approximation, to discuss the knockout of an electron which singly occupies the weakest bound orbital of an atom. As is expected from the simple discussion given above, the angular distribution of the outgoing electrons is given directly by the momentum distribution of the orbital. Section III is devoted to the application of this theory to atomic hydrogen and the alkali atoms. In Sec. IV, we extend the theory to two-electron atoms, paying attention to the requirement of overall antisymmetry of the wave function with respect to electron exchange. A detailed analysis is given for He as the target and for the effect of electron correlations in its ground state. The results of the paper are then briefly summarized in Sec. V.

II. SIMPLE THEORY FOR ONE-ELECTRON SYSTEMS

The starting point of our theory is the familiar expression for the transition probability per unit time according to first-order perturbation theory

$$w(\alpha_i,\alpha_f)=(2\pi/\hbar)|\langle\alpha_f|H'|\alpha_i\rangle|^2\,\delta[E(\alpha_f)-E(\alpha_i)]. \tag{2.1}$$

Here α_i and α_f are quantum numbers for the initial and final states of the Hamiltonian H_0 describing the incident electron and the target system

$$H_0|\alpha\rangle=E(\alpha)|\alpha\rangle, \tag{2.2}$$

and H' is the interaction between these two systems: the sum of Coulomb interactions between the incident electron and all the electrons and nuclei of the target. In order to obtain a reaction cross section from (2.1), we must suitably average over α_i, sum over α_f, and divide by the incident flux. The states $|\alpha_i\rangle$ and $|\alpha_f\rangle$ describe one and two unbound electrons, respectively, which we here approximate by plane waves. Thus our results are meant to apply only to high energies where the following conditions are satisfied: (1) large incident and exit speeds – relative to characteristic speeds for the target and residual systems – and (2) large incident and exit kinetic energies – relative to the potentials generated by the target and residual systems. The first condition permits the use of (2.1), and the second the use of plane, rather than distorted, waves.

Equation (2.1) is essentially the same formula used to describe the nuclear $(p,2p)$ reaction.[4] In that case, however, one must invoke a further implication of the "impulse approximation,"[6] and replace the matrix elements of H' by those of a suitable scattering operator. This is an essential change because the potential between two nucleons differs greatly from the operator describing their scattering. In atomic physics, on the other hand, the amplitude for Coulomb scattering is correctly given, except for phase, by the Born approximation, i.e., by the matrix elements of the potential. Thus the distinction between potential and scattering operator is unimportant in using the impulse approximation for atomic scattering if the phase of the relevant two-body scattering amplitude does not enter. Moreover we will show below that the kinematics for the experiment depicted in Fig. 1 are such that the matrix element of the two-body scattering operator is a constant. Therefore in both the nuclear $(p,2p)$ and the $(e,2e)$ reactions at high energies, any difference between potential and scattering operator affects only the absolute normalization of the calculated cross section. The theory we have been describing originated, of course, with Bethe.[7]

In this section we consider the case where the knocked-out electron is initially in a single-particle orbital nl, and the reaction does not alter the structure of the rest of the atomic system. Of course, hydrogenic systems are the only ones which exactly satisfy this description, but this model should provide a good first approximation for other targets such as the alkali atoms. In the beginning we will ignore the spins and the identity of all the electrons. The unperturbed Hamiltonian is

$$H_0=T_1+(T_2+T_3+V_{23}), \tag{2.3}$$

and the interaction Hamiltonian is

$$H'=V_{12}+V_{13}, \tag{2.4}$$

where 1 stands for the incident electron, 2 for the target electron, and 3 for the rest of the target; T is the kinetic energy and V is the potential energy of interaction. The constant Hamiltonian for the unaffected part of the target system has been dropped from (2.3). We shall also assume that only the interaction between the incident and target electron is important, i.e., V_{13} will be ignored in H'.

In configuration space, the initial state is

$$\langle\vec{r}_1\vec{r}_2\vec{r}_3|\alpha_i\rangle=\frac{e^{i\vec{k}_0\cdot\vec{r}_1}}{\Omega^{1/2}}\left[\frac{e^{i\vec{K}\cdot\vec{R}}}{\Omega^{1/2}}\,\phi_{nlm}(\vec{r})\right]_{\vec{K}=0}, \tag{2.5}$$

where the first factor refers to the incident electron and the second to the target. Here $\vec{r}_1, \vec{r}_2$, and $\vec{r}_3$ are the respective positions of the incident electron, the target electron, and the center of mass of the rest of the target – which is also the origin of the central force for the orbital nlm – i.e.,

$$\vec{r} = \vec{r}_2 - \vec{r}_3 \ . \tag{2.6}$$

Also $\vec{k}_0$ is the incident electron's wave number, and $\vec{k}$ is the same for the target atom of mass M located at

$$\vec{R} = (m\vec{r}_2 + M\vec{r}_3)/(m+M) \ , \tag{2.7}$$

where m is the electron mass. We use normalization in a box of volume Ω for the plane-wave states. The energy of the wave function (2.5) is

$$E(\alpha_i) = \epsilon(k_0) - B_{nl} \ , \tag{2.8}$$

where $$\epsilon(k) = \hbar^2 k^2/2m \tag{2.9}$$

and B_{nl} is the binding energy of the orbital mln. The final wave functions will be of the type

$$\langle r_1 r_2 r_3 | \alpha_f \rangle = \Omega^{-3/2} e^{i(\vec{k}_1\cdot\vec{r}_1 + \vec{k}_2\cdot\vec{r}_2 + \vec{k}_3\cdot\vec{r}_3)} \ , \tag{2.10}$$

where now $\vec{k}_1$ and $\vec{k}_2$ are the wave numbers for the two outgoing electrons and $\vec{k}_3$ is that for the residual atomic system (usually an ion). The corresponding energy is

$$E(\alpha_f) = \epsilon(k_1) + \epsilon(k_2) + E(k_3) \ , \tag{2.11}$$

where $$E(K) = \hbar^2 K^2/2M \tag{2.12}$$

The matrix element of H' in (2.4) between the initial and final states (2.5) and (2.10) may now be easily obtained by integrations over $\vec{r}_1$, $\vec{r}_2$, and $\vec{r}_3$. Each of these leads to a Kronecker δ function if the potential $V(\vec{r}_1 - \vec{r}_2)$ and the spatial wave function $\phi_{nlm}(\vec{r})$ are first replaced by their respective Fourier transforms

$$V(\vec{r}) = [(2\pi)^3/\Omega] \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}} v(\vec{q})$$

$$\xrightarrow[\Omega\to\infty]{} \int d\vec{q}\, e^{i\vec{q}\cdot\vec{r}} v(\vec{q}) \ , \tag{2.13}$$

$$\phi_{nlm}(\vec{r}) = [(2\pi)^{3/2}/\Omega] \sum_{\vec{\kappa}} e^{i\vec{\kappa}\cdot\vec{r}} \psi_{nlm}(\vec{\kappa})$$

$$\xrightarrow[\Omega\to\infty]{} (2\pi)^{-3/2} \int d\vec{\kappa}\, e^{i\vec{\kappa}\cdot\vec{r}} \psi_{nlm}(\vec{\kappa}) \ . \tag{2.14}$$

The result is

$$\langle \alpha_f | H' | \alpha_i \rangle = [(2\pi)^3/\Omega]^{3/2} \delta_{\vec{k}_0 - \vec{k}_3, \vec{k}_1 + \vec{k}_2} \times v(\vec{k}_1 - \vec{k}_0) \psi_{nlm}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0) \ . \tag{2.15}$$

The δ function expresses momentum conservation; in particular, the recoil momentum must be supplied by the target electron. We also note that the amplitude for the process, $\alpha_i = \vec{k}_0, nlm \to \alpha_f = \vec{k}_1\vec{k}_2\vec{k}_3$, is proportional to the electron-electron scattering amplitude $v(\vec{k}_1 - \vec{k}_0) = v(\vec{k}_2 - (-\vec{k}_3))$ and the probability amplitude $\psi_{nlm}(-\vec{k}_3)$ for the target electron to have the required momentum $-\vec{k}_3 = \vec{k}_1 + \vec{k}_2 - \vec{k}_0$. Of course the residual ion retains the same momentum $\vec{k}_3$ because we have assumed that it does not interact during the collision.

For the experimental arrangement of Fig. 1, we have to sum the transition probability over finite angular and energy ranges for the two outgoing electrons and over all recoil momenta $\vec{k}_3$, since the residual system is not observed. In addition, we must average over the initial magnetic quantum number m corresponding to an unpolarized target. Using (2.15), the total transition probability per unit time divided by the incident flux, $I_0 = \hbar k_0/m\Omega$, is

$$I = \frac{m\Omega}{\hbar k_0} \frac{1}{2l+1} \sum_{m=-l}^{l} \sum_{\vec{k}_1}{}' \sum_{\vec{k}_2}{}' \sum_{\vec{k}_3} \frac{2\pi}{\hbar} \left(\frac{(2\pi)^3}{\Omega}\right)^3 \times \delta_{\vec{k}_0 - \vec{k}_3, \vec{k}_1 + \vec{k}_2} \left| v(\vec{k}_1 - \vec{k}_0) \right|^2 \left| \psi_{nlm}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0) \right|^2 \times \delta(\epsilon(k_0) - B_{nl} - \epsilon(k_1) - \epsilon(k_2) - E(k_3)) \ , \tag{2.16}$$

where the sums over $\vec{k}_1$ and $\vec{k}_2$ are restricted so that $\vec{k}_1$ and $\vec{k}_2$ are within the intervals $\Delta\vec{k}_1$ and $\Delta\vec{k}_2$, respectively. The symbol $\Delta\vec{k}$ represents the three-dimensional element in the $\vec{k}$ space of an electron that is detected by one of the counters C:

$$\Delta\vec{k} = k^2 \Delta k \Delta\hat{k} = \tfrac{1}{2}(2m/\hbar^2)^{3/2} \epsilon^{1/2} \Delta\Omega \Delta\epsilon \ ,$$

where $\Delta\Omega$ is the solid angle subtended and $\Delta\epsilon$ is the energy resolution. This expression for I is somewhat idealized because we have assumed that the counters have 100% efficiency for detecting electrons in the interval $\Delta\vec{k}$. The evaluation of the sums in (2.16) is complicated by the dependence of the recoil energy $E(k_3)$ (in the energy-conserving δ function) on the sum variables $\vec{k}_1$ and $\vec{k}_2$. The result of the calculation is that I has the form

$$I = \sigma(k_0 nl, \vec{k}_1\vec{k}_2) \Delta\Omega_1 \Delta\Omega_2 \Delta\epsilon \ , \tag{2.17a}$$

where σ is a quantity with dimensions of area/energy and is defined as the cross section for the process. It is given by the expression

$$\sigma(\vec{k}_0 nl, \vec{k}_1\vec{k}_2) = C 8\pi^5 (\epsilon_1\epsilon_2/\epsilon_0)^{1/2} f_B(\vec{k}_1 - \vec{k}_0)|^2 \times |F_{nl}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0)|^2 \tag{2.17b}$$

A number of new quantities appear on (2.17a) and (2.17b). First of all, special units have been adopted in (2.17b) in which lengths are measured in terms of the Bohr radius $a_0 = \hbar^2/me^2$ and energies in terms of the Rydberg $R = e^2/2a_0$. Next the Fourier transform of the Coulomb potential v has been replaced by the Rutherford scattering amplitude whose absolute value is

$$|f_B(\vec{q})| = 2/q^2 \,. \tag{2.18}$$

Also the radial part of $\psi_{nlm}(\vec{\kappa})$ has been replaced by

$$\psi_{nlm}(\vec{\kappa}) = F_{nl}(\kappa) Y_{lm}(\hat{\kappa}) \,, \tag{2.19}$$

and the following identity has been used

$$\frac{1}{2l+1} \sum_{m=-l}^{l} |\psi_{nlm}(\kappa)|^2 = \frac{1}{4\pi} |F_{nl}(\vec{\kappa})|^2 \,. \tag{2.20}$$

The factor C in (2.17b) contains the restrictions imposed by energy conservation. If the two electron counters detect electrons within the intervals $(\epsilon_1 - \frac{1}{2}\Delta\epsilon_1,\ \epsilon_1 + \frac{1}{2}\Delta\epsilon_1)$ and $(\epsilon_2 - \frac{1}{2}\Delta\epsilon_2,\ \epsilon_2 + \frac{1}{2}\Delta\epsilon_2)$, then we may define the quantity

$$\Delta\epsilon = \epsilon(k_0) - B_{nl} - [\epsilon(k_1) + \epsilon(k_2) + E(|\hat{k}_1 k_1 + \hat{k}_2 k_2 - \vec{k}_0|)] \tag{2.21}$$

which is the difference between the given total energy and that measured by the counters. Then C is expressed as

$$C = \begin{cases} 1 & |\Delta\epsilon| < \frac{1}{2}(\Delta\epsilon_1 + \Delta\epsilon_2) \\ 0 & |\Delta\epsilon| > \frac{1}{2}(\Delta\epsilon_1 + \Delta\epsilon_2) \end{cases} \times \begin{cases} 1 & |\Delta\epsilon| < \frac{1}{2}(\Delta\epsilon_2 - \Delta\epsilon_1) \\ 1 - \dfrac{2|\Delta\epsilon|}{\Delta\epsilon_1 + \Delta\epsilon_2}, & \frac{1}{2}(\Delta\epsilon_2 - \Delta\epsilon_1) < |\Delta\epsilon| < \frac{1}{2}(\Delta\epsilon_2 + \Delta\epsilon_1) \end{cases} \tag{2.22}$$

where we have taken $\Delta\epsilon$ in (2.17a) to be $\Delta\epsilon = \Delta\epsilon_1 < \Delta\epsilon_2$. To derive this result we have had to make use of the fact that $m \ll M$ and to assume that all other factors in (2.17) do not vary much within $\Delta\epsilon$.

The main conclusion to be drawn from (2.17) is that the intensity I is proportional to the probability distribution in momentum space $|F_{nl}(\kappa)|^2$, where

$$\vec{\kappa} \equiv \vec{k}_1 + \vec{k}_2 - \vec{k}_0 = -\vec{k}_3 \,. \tag{2.23}$$

To understand how the functional dependence of $|F_{nl}|^2$ can be measured, we consider the energies of the outgoing electrons, $\epsilon(k_1)$ and $\epsilon(k_2)$, to be within the energy ranges $\Delta\epsilon_1$ and $\Delta\epsilon_2$. Such particles will be registered by counters gated at the same energy ranges so that $C(\Delta\epsilon)$ will be nonzero. That is, ionization from the particular shell with binding energy B_{nl} may be studied by proper choice of the outgoing energies. Then varying the directions $\hat{k}_1$ and $\hat{k}_2$ will map out a range in the argument $\kappa = |k_1\hat{k}_1 + k_2\hat{k}_2 - \vec{k}_0|$. As a special but informative case, we consider $\hat{k}_1$, $\hat{k}_2$, and $\hat{k}_0$ coplanar, $\hat{k}_1$ and $\hat{k}_2$ at equal angles with respect to the incident direction $(\theta_1 = \theta_2 \equiv \theta/2)$, and $k_1 = k_2 \equiv k$. In this "symmetrical situation," the recoil momentum is always parallel to the incident momentum, i.e.,

$$\vec{\kappa} = (2k\cos\tfrac{1}{2}\theta - k_0)\hat{k}_0 \,. \tag{2.24}$$

If we assume that k has been chosen so that $\Delta\epsilon = 0$ [or equivalently that $\Delta\epsilon$ is negligible compared with the various terms in (2.21)], then energy conservation implies that

$$2k^2 = k_0^2 - \gamma^2 - (m/M)\kappa^2 \,, \tag{2.25}$$

where $\gamma^2 = (2m/\hbar^2)B_{nl}$. Now if we consider the particular angle θ_0 corresponding to no recoil, i.e., to the ionization of an electron with zero momentum $\kappa(\theta_0) = 0$, we find this angle to be given by

$$\cos\tfrac{1}{2}\theta_0 = k_0/2k \,. \tag{2.26a}$$

In this same case we also have

$$k = (k_0/\sqrt{2})(1 - \gamma^2/k_0^2)^{1/2} \tag{2.26b}$$

and, therefore,

$$\cos\theta_0 = \gamma^2/(k_0^2 - \gamma^2) \,. \tag{2.27}$$

At very high energies $\gamma^2/k_0^2 = B_{nl}/E_0 \ll 1$, and so (2.27) is, up to first order in γ^2/k_0^2,

$$\theta_0 \cong \pi/2 - B_{nl}/E_0 \,. \tag{2.28}$$

Thus, when the experiment is arranged to observe the ionization of target electrons which are at rest, they will be observed at a slightly smaller angle than $\pi/2$ because of the finite binding.

We may use (2.26a) to write $\vec{\kappa}$ in (2.24) for the symmetrical situation as

$$\vec{\kappa} = [(\cos\tfrac{1}{2}\theta/\cos\tfrac{1}{2}\theta_0) - 1]\vec{k}_0 \tag{2.29}$$

without any approximation. Also we note that the (2.26b) is always an excellent approximation because κ^2 will never be much bigger than γ^2 – otherwise $|F_{nl}(\kappa)|^2$ becomes too small – and the ratio m/M makes the recoil term in (2.25) negligible.[8]

The remaining factors in (2.17b) may be ignored in the symmetrical situation, at least as far as the variation with κ or θ is concerned. The smallness of the recoil energy implies that $\Delta\epsilon$, and therefore the factor C, is constant. Of course $(\epsilon_1\epsilon_2)^{1/2} = \epsilon$ is also fixed. The magnitude of the momentum transfer $\vec{q} = \vec{k}_1 - \vec{k}_0$ is

$$q = \frac{k_0}{\sqrt{2}} \left(1 \pm 2\frac{\kappa}{k_0} - \frac{\gamma^2 + (m/M)\kappa^2}{k_0}\right)^{1/2} , \tag{2.30}$$

where the upper sign refers to $\hat{\kappa}$ parallel to $\hat{k}_0$, and the lower sign to $\hat{\kappa}$ antiparallel to $\hat{k}_0$. Thus the scale of variation for f_B is k_0 whereas that for F_{nl} is typically γ; hence $f_B(q)$ may be replaced by $f_B(k_0/\sqrt{2})$ to a very good approximation. Therefore the important variation in (2.17) for the symmetrical situation arises from the factor $|F_{nl}(\kappa)|^2$ for $\kappa \ll k_0$. We express this fact by writing the relative cross section (the ratio of the differential cross section to its maximum value) for the symmetrical situation σ_S as

$$\sigma_S = |F_{nl}(\kappa)/F_{nl}(\kappa_m)|^2 \tag{2.31a}$$

or, using (2.29),

$$\sigma_s(\theta)=\frac{1}{|F_{nl}(\kappa_m)|^2}\left|F_{nl}\left(\left|\frac{\cos(\theta/2)}{\cos(\theta_0/2)}-1\right|k_0\right)\right|^2 . \tag{2.31b}$$

The subscript s identifies the symmetrical situation under discussion, and θ_m is the first maximum in $|F_{nl}|$; θ_m differs from θ_0 for $l\neq 0$, in which cases $F_{nl}(0)=0$.

Up to now we have ignored the identity of the initial and target electrons. Because we are dealing with an essentially free collision and a single target electron, the effects of the identity are easy to treat. For unpolarized beams, (2.17) has to be replaced by

$$\sigma(\vec{k}_0 nl,\vec{k}_1\vec{k}_2)=C8\pi^5(\epsilon_1\epsilon_2/\epsilon_0)^{1/2}|F_{nl}(\vec{k}_1+\vec{k}_2-\vec{k}_0)|^2$$
$$\times[\,|f_B(\vec{k}_1-\vec{k}_0)|^2+|f_B(\vec{k}_2-\vec{k}_0)|^2$$
$$-\mathrm{Re}f_B^*(\vec{k}_1-\vec{k}_0)f_B(\vec{k}_2-\vec{k}_0)]\ . \tag{2.32}$$

This expression is invariant under interchange of the two electron counters, i.e., $\vec{k}_1\rightleftarrows\vec{k}_2$. Equation (2.32) can be understood in terms of the familiar formula for unpolarized scattering of two free electrons,

$$\tfrac{3}{4}|f_1|^2+\tfrac{1}{4}|f_0|^2=|f|^2+|g|^2-\mathrm{Re}f^*g\ , \tag{2.33}$$

in terms of the singlet and triplet amplitudes f_S, $S=0$, and 1, and the direct and exchange amplitudes

$$f_1=f-g\ , \tag{2.34a}$$

$$f_0=f+g\ . \tag{2.34b}$$

In our high-energy Born approximation, $f\cong f_B(\vec{k}_1-\vec{k}_0)$ and $g\simeq f_B(\vec{k}_2-\vec{k}_0)$ so that (2.32) agrees with (2.33). For the symmetrical situation the identity of the electrons leads only to a change in the absolute cross section because both momentum transfers, $\vec{k}_1-\vec{k}_0$ and $\vec{k}_2-\vec{k}_0$, are given by (2.30); i.e., they are essentially constant. Therefore (2.31), which gives the angular variation of the cross section, remains valid when the identity of the incident and target electron are included.

III. APPLICATION TO HYDROGEN AND THE ALKALI ATOMS

For hydrogenic orbitals, the momentum distribution is given in closed form in terms of Gegenbauer polynomials.[9] For example, the relative cross section (2.31) for the $n=1$, $l=0$ state becomes

$$\sigma_s(1,0)=[1+(\kappa/Z)^2]^{-4} \tag{3.1a}$$

or

$$\sigma_s(1,0)=\left\{1+\left[\left(\frac{\cos(\theta/2)}{\cos(\theta_0/2)}-1\right)k_0Z^{-1}\right]^2\right\}^{-4} . \tag{3.1b}$$

As expected, (3.1b) is strongly peaked about θ_0 with a width of order $(B/E_0)^{1/2}=Z/k_0$; the actual half-width in this case is $\Delta\theta_{1/2}\simeq\frac{1}{3}Z/k_0$. A plot of $\sigma_S(1,0)$ versus κ for $Z=1$ is given in Fig. 2. It is interesting to compare (3.1) with the corresponding cross section for the $n=2$, $l=0$ state,

$$\sigma_s(2,0)=\{[2\kappa/Z)^2-1]/[(2\kappa/Z)^2+1]^3\}^2 \tag{3.2}$$

which is also plotted in Fig. 2 for $Z=1$. In addition to the central maximum at $\kappa=0$, there is a zero at $\kappa=Z/2$ and an extremely small subsidiary maximum at $\kappa=Z/\sqrt{2}$. The intensity from this maximum is down by $3^{-6}\cong 10^{-3}$ from the central peak. On the other hand, the absolute cross sections stand in the ratio $|F_{20}(0)|^2:|F_{10}(0)|^2=2^5:1$. The essential difference between (3.1) and (3.2) is the compression of the $n=2$ cross-section curve towards smaller momenta, corresponding to the smaller binding energy of this orbital.

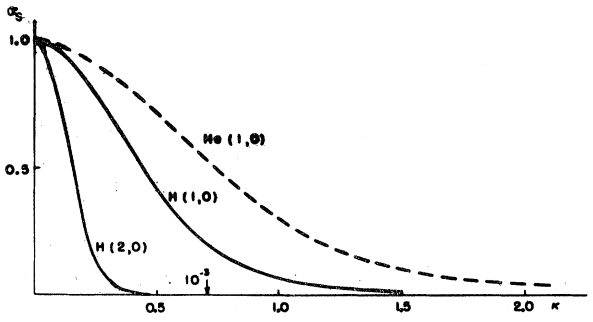

FIG. 2. The relative cross section for the symmetrical situation ($\theta_1=\theta_2=\theta/2$, $k_1=k_2=k\approx k_0/\sqrt{2}$) plotted versus κ [Eq. (2.29)] for $1s$ and $2s$ states of H (solid curves) and the $(1s)^2$ state of He (dashed curves). The arrow indicates the position of an extremely small subsidiary maximum for the $2s$ state of H.

In order to discuss the situation for the alkalis, we first recall some general properties of central-field wave functions in momentum space. According to (2.14) and (2.19), the radial parts of the momentum- and configuration-space wave functions are related by the spherical Bessel transform

$$F_{nl}(\kappa)=C_l\int_0^\infty dr\,r^2 j_l(\kappa r)R_{nl}(r)\ , \tag{3.3}$$

with $C_l=(-)^l4\pi(2\pi)^{-3/2}$. In analyzing this expression, we recall the characteristic lengths of R_{nl}: $n-l-1$ zeros $r_{nl}^{(\nu)}$ and the exponential decay lengths $a_{nl}=(2mB_{nl}/\hbar^2)^{1/2}$. Then from (3.3) we can easily determine that

(*i*) $F_{nl}(\kappa)=0(\kappa^l)$, for $\kappa\ll a_{nl}^{-1}$,

(*ii*) $F_{nl}(\kappa)$ has $n-l-1$ zeros,

(*iii*) $F_{nl}(\kappa)=0(\kappa^{-(l+4)})$, for $\kappa\gg(r_{nl}^{(1)})^{-1}$.

We can also study the changes in F_{nl} due to changes in R_{nl} by rewriting (3.3) as[4]

$$\delta F_{nl}(\kappa)/\delta R_{nl}(r)=C_l r^2 j_l(\kappa r)\ . \tag{3.4}$$

For s states, $j_l(kr) = (kr)^{-1}\sin kr$, and (3.4) implies that a change in $R_{no}(r)$ at r affects $F_{nl}(\kappa)$ only for $\kappa \lesssim r^{-1}$. Crudely speaking, changes at large r affect small κ and changes at small r affect all κ.

For the ns ground states of the alkalis, we may therefore expect a central peak (in $|F_{nl}(\kappa)|^2$ or in σ_S) plus $n-1$ subsidiary maxima. The properties of the central peak will be mainly determined by the asymptotic behavior of the radial function $R_{nl}(r)$, which is largely hydrogenic in character. In fact, we expect the width of this central peak to change fairly slowly from one alkali to the next, because of the relatively slow variation in their ionization potentials. The most interesting features of the cross section will be connected with the subsidiary maxima and minima, i.e., with the zeros of F_{n0}. These characteristics are determined by the deviations of the actual (Hartree-Fock) central field from its asymptotic Coulomb form. Unlike the situation discussed above for the pure Coulomb potential, the subsidiary maxima for the alkalis are expected to yield cross sections which are quite comparable with that from the central peak. Indeed this is one of the most important reasons for carrying out ionization experiments of this type, that is to measure directly and from point to point the wave functions F_{nl} of the atomic self-consistent field.

IV. TWO-ELECTRON ATOMS

We consider here the case of targets with two electrons in a $^1S_0$ state, the prototype being the normal helium atom in its ground state. However, in line with the point of view adopted with regard to the alkali atoms, we expect the analysis also to apply to knockouts from the valence shell of the alkaline earths. The theoretical basis will be essentially the same as in Sec. II, except that the antisymmetric nature of the three-electron wave functions will be incorporated from the outset of the calculation.

The Hamiltonian for the problem is

$$H = T_1 + (T_2 + T_3 + T_4 + V_{23} + V_{24} + V_{34}) + (V_{12} + V_{13} + V_{14}) , \tag{4.1}$$

where 4 refers to the nucleus (plus inert inner shells), 2 and 3 refer to the target (valence) electrons, and 1 refers to the incident electron. The perturbing Hamiltonian to be used in the transition probability (2.1) is $V_{12} + V_{13}$, i.e., the interaction between the incident electron and the target nucleus will be again ignored. The initial and final wave functions, prior to antisymmetrization with respect to the incident electron, are as follows:

$$\langle \vec{r}_1\vec{r}_2\vec{r}_3\vec{r}_4 | \alpha_i \rangle = \Omega^{-1/2} e^{i\vec{k}_0\cdot\vec{r}_1} \phi_i(\vec{r}_2 - \vec{r}_4, \vec{r}_3 - \vec{r}_4) \times \Omega^{-1/2} |m_0\rangle |S=0, M=0\rangle , \tag{4.2}$$

$$\langle \vec{r}_1\vec{r}_2\vec{r}_3\vec{r}_4 | \alpha_f \rangle = 2^{-1/2}(1 - P_{23})\Omega^{-3/2} e^{i(\vec{k}_1\cdot\vec{r}_1 + \vec{k}_2\cdot\vec{r}_2 + \vec{k}_3\cdot\vec{r}_{34})} \times \phi_{nlm}(\vec{r}_{34}) |m_1\rangle |m_2\rangle |m_3\rangle . \tag{4.3}$$

We introduced some new quantum numbers beyond those used in Sec. II: m_0, the spin projection of the incident electron; S, and M, the total spin quantum numbers of the target electrons; m_1 and m_2, the spin projections of the outgoing electrons; m_3, the spin projection of the recoil ion; and l and m, its orbital angular momentum quantum numbers. In addition, $\vec{r}_{34} = (m\vec{r} + M\vec{r}_4)(m+M)^{-1}$, ϕ_i is the (space- and exchange-symmetric) wave function for the two target electrons, ϕ_{nlm} is the final electron's orbital, and P_{ij} is an exchange operator (for both space and spin variables). The presence of the operator $2^{-1/2}(1 - P_{23})$ guarantees that both of the target electrons can be knocked out.

Were we to immediately substitute the above wave functions into (2.1), we would not obtain any exchange scattering amplitudes associated with the identity of the incident and target electrons. This would be a serious error in our analysis, because we are particularly interested in the "symmetrical situation," where the two outgoing electrons have the same dynamical properties. Therefore it is essential to multiply (4.3) by the operator $(1 - P_{12} - P_{13})$, which thereby generates a completely antisymmetric final state. The amplitudes included in this way are represented schematically in Figs. 3a–d: (a) and (c) represent the direct knockout of particles 2 and 3, respectively, and (b) and (d) are the corresponding exchange amplitudes. Figure 3e depicts a process which may be ignored at high energies ($k_0, k_1, k_2 \gg 1$), i.e., it is very improbable that a knockout collision with particle 3 will leave it bound and eject particle 2 with a large momentum. Finally, after the entire matrix element between initial and final states is evaluated, we will have to average over m_0 and sum over m_1, m_2, m_3, and m, because we consider only the situation where the initial state is unpolarized and no polarization analysis is made of the final state.

The calculation we have just outlined is quite standard, so we omit the details and give only the final result. The appropriate cross section [of the type defined by (2.17)] is

$$\sigma(\vec{k}_0 i, \vec{k}_1\vec{k}_2 nl) = C 8\pi^5 (\epsilon_1\epsilon_2/\epsilon_0)^{1/2} 2(2l+1) \times |\gamma_{nl}^{(i)}(\vec{k}_1 + \vec{k}_2 - \vec{k}_0)|^2 \times [\,|f_B(\vec{k}_1 - \vec{k}_0)|^2 + |f_B(\vec{k}_2 - \vec{k}_0)|^2 - \mathrm{Re} f_B^*(\vec{k}_1 - \vec{k}_0) f_B(\vec{k}_2 - \vec{k}_0)\,] . \tag{4.4}$$

The only essential change from (2.32) is the appearance of the factor $2(2l+1)|\gamma_{nl}^{(i)}|^2$ rather than $|F_{nl}|^2$, where $\gamma_{nl}^{(i)}$ is defined as follows:

$$\Gamma_{nlm}^{(i)}(\vec{k}) = \gamma_{nl}^{(i)}(\kappa) Y_{lm}(\hat{\kappa}) , \tag{4.5}$$

$$\Gamma_{nlm}^{(i)}(\vec{k}) = (2\pi)^{-3/2} \int d\vec{r}\, e^{+i\vec{k}\cdot\vec{r}} \times \int d\vec{r}\,' \varphi_{nlm}(\vec{r}\,') \; \phi_i^*(\vec{r}, \vec{r}\,') \tag{4.6a}$$

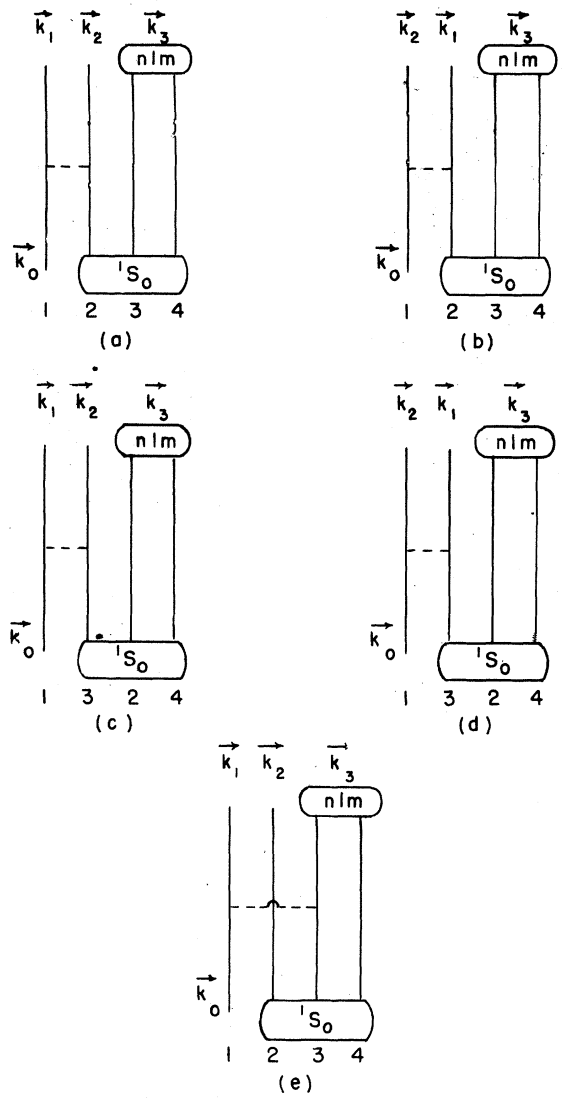

FIG. 3. Processes relevant to the calculation of the amplitude for the ionization of an electron in a $^1S_0$ two-electron state leaving one electron in the state nlm. Initial states are at the bottom, and final states at the top of each diagram. The Coulomb interaction is represented by a dashed line and particle states by solid vertical lines. The upper four diagrams are included in this calculation; the last is typical of those which are omitted.

or

$$\Gamma_{nlm}^{(i)}(\vec{\kappa}) = \int d\vec{\kappa}' \psi_{nlm}(\vec{\kappa}')\psi_i^*(\vec{\kappa},\vec{\kappa}') \quad . \qquad (4.6b)$$

In (4.6), ψ_i and ψ_{nlm} are the momentum-space wave functions for the target (two-electron) and recoil (one-electron) systems:

$$\phi_{nlm}(\vec{r}) = (2\pi)^{-3/2}\int d\vec{\kappa} e^{i\vec{\kappa}\cdot\vec{r}}\psi_{nlm}(\vec{\kappa}) \quad , \qquad (4.7a)$$

$$\phi_i(\vec{r}_1\vec{r}') = (2\pi)^{-3}\int d\vec{\kappa} \times \int d\vec{\kappa}' e^{i(\vec{\kappa}\cdot\vec{r}+\vec{\kappa}'\cdot\vec{r}')}\psi_i(\vec{\kappa},\vec{\kappa}') \qquad (4.7b)$$

According to (4.6b), $\Gamma_{nlm}^{(i)}(\vec{\kappa})$ is the joint probability amplitude for one of the electrons in state i to be in the bound orbital nlm and for the other electron (in state i) to be in the unbound plane-wave state $\vec{\kappa}$. Thus $|\gamma_{nl}(i)|^2$ is the natural generalization of the momentum distribution $|F_{nl}|^2$ for a one-electron target. Likewise the new factor $2(2l+1)$ is proper because there are now two target electrons which can be ionized and $(2l+1)$ magnetic substates available for the final ion. The symmetrized two-body cross section which appears in (4.4) has already been discussed in connection with (2.32). In contrast to elastic scattering from $^1S_0$ systems, for example, the two target electrons scatter independently into all available spin states.

The distribution in momentum space $|\gamma_{nl}^{(i)}(\kappa)|$ determines the angular variation of the coincidence counting rate in exactly the same way that $|F_{nl}(\kappa)|^2$ does for one-electron targets. Thus the relative cross section for the "symmetrical situation" for two-electron targets is given by (2.31) with F_{nl} replaced by $\gamma_{nl}^{(i)}$.

We can obtain a good understanding of the behavior of this cross section by assuming that the two electrons are in s-states

$$\phi_i(\vec{r},\vec{r}') = \chi_0(\vec{r})\chi_0(\vec{r}') \qquad (4.8)$$

Later on we shall consider more general wave functions including, in particular, correlation effects involving the target electrons. Substituting (4.8) into (4.5) and (4.6) leads to

$$\gamma_{nl}^{(i)}(\kappa) = \delta_{l,0}\beta_n G_0(\kappa) \quad , \qquad (4.9a)$$

$$\beta_n = \int d\vec{r}\,\phi_{n00}^*(\vec{r})\chi_0(\vec{r}), \qquad (4.9b)$$

where $(4\pi)^{-1/2}G_0$ is the Fourier transform of χ_0. Because β_n is the overlap of two unit-normalized orbitals, $\beta_n \leq 1$. Thus the relative cross section in this case is determined solely by the momentum distribution of χ_0, i.e.,

$$\sigma_s = |G_0(\kappa)/G_0(\kappa_m)|^2 \qquad (4.10)$$

The absolute cross section will involve, in addition, the factor $\beta_n^2 \leq 1$. It should be noted that the very reasonable assumption (4.8) does not imply that the residual ion is necessarily left in its ground state immediately after ionization. Rather one expects *every* s state ψ_{n00} to be populated with probability $|\beta_n|^2$. Thus, even if the summed energy of the outgoing electrons is not measured [recall that $E_1+E_2=E_0-(B_i-B_{nl})$], σ_s still determines the momentum distribution of the initial state if (4.8) holds. For He-like targets, this feature is quite relevant, even if the summed energy is determined, because of the accidental degeneracy of the energy levels in a pure Coulomb field.

To obtain some insight into the shape of σ_S as given by (4.10), we adopt the hydrogenic approximation

$$\chi_0(\vec{r}) = (4\pi)^{-1/2} 2 Z_i^{3/2} e^{-Z_i r}, \tag{4.11}$$

with Z_i determined variationally[10] to be $Z_i = Z_f - \frac{5}{16}$. Then we have

$$G_0(\kappa) = (32/\pi)^{1/2} Z_i^{5/2} (\kappa^2 + Z_i^2)^{-2}, \tag{4.12}$$

which leads to (3.1) when Z_i is set equal to one, and

$$\beta_0^2 = \{Z_i Z_f / [\tfrac{1}{2}(Z_i + Z_f)]^2\}^3, \tag{4.13}$$

for the probability of ionization to the ground state of the residual ion. For the case of the ground state of normal He, $Z_f = 2$, $Z_i = 1 + \frac{11}{16} = 1.688$, so that $\beta_0^2 = 0.983$. Furthermore, with (4.12), σ_S has the same shape as (3.1) and the $(n=1, l=0)$ curve in Fig. 2, but with a width increased by the factor Z_i. The peak of the cross section is also shifted in comparison with hydrogen from $\pi/2 - (R/E_0)$ to $\pi/2 - 1.695(R/E_0)$ (using the variational energy) because of the increase in ionization potential. These changes in the relative cross-section function $\sigma_S(\theta)$ are accompanied by a decrease in the absolute cross section which goes like $2\beta_0^2[G_0(0)/F_{10}(0)]^2$, all other things being equal; for He this factor is $2\beta_0^2 Z_i^{-3} \approx 2 \times \frac{1}{2} = 1$.

We will now complete our discussion of two-electron systems by evaluating the role of inter-electron correlations in the ground state of the target system. We have considerable freedom in choosing appropriate ground-state wave functions, but we are also faced with the problem of properly defining the amount of correlation. Most He wave functions are variational in character, and any measure of correlation effects will depend on the form of the wave function for a fixed value of the energy. Of course here we are only concerned with a level of accuracy far below the best available for He wave functions. The most obvious procedure would be to simply substitute wave functions of the Hylleraas type into (4.6) and attempt to carry out the integration. For example, we have done this with a very simple correlated wave function, the two-parameter Hylleraas function[10]

$$\phi(\vec{r}_1, \vec{r}_2) = \text{const} \times e^{-\frac{1}{2}[3.63(r_1 + r_2)]} \times [1 + 0.081(3.63) r_{12}] . \tag{4.14}$$

But the resulting closed form for Γ is so complex as to be essentially useless in the context of our work.

A more efficacious procedure is to expand the two-electron wave function in the manner of Green *et al.*[11]

$$\phi_i(\vec{r}, \vec{r}') = \sum_{n_1 \le n_2} \sum_{lm} (-)^m c_l(n_1 n_2) [2(1 + \delta_{n_1 n_2})]^{-1/2} \times [\phi_{n_1 l-m}^{(i)}(\vec{r}) \phi_{n_2 lm}^{(i)}(\vec{r}') + \phi_{n_1 l-m}^{(i)}(\vec{r}') \phi_{n_2 lm}^{(i)}(\vec{r})] , \tag{4.15}$$

with $\phi_{nlm}^{(i)}(\vec{r}) = R_{nl}^{(i)}(r) Y_{lm}(\hat{r})$. (4.16)

Equation (4.15) is the most general expansion in terms of a single set of central-field wave functions for a two-particle state which has zero total angular momentum and which is even under (space) exchange. Substitution of (4.15) into (4.6a) leads to

$$\gamma_{nl}^{(i)}(\kappa) = \sum_{n_1 \le n_2} [2(1 + \delta_{n_1 n_2})]^{-1/2} c_l(n_1, n_2) \times \{F_{n_1 l}^{(i)}(\kappa) \langle nl | i n_2 l \rangle + F_{n_2 l}^{(i)}(\kappa) \langle nl | i n_1 l \rangle\} , \tag{4.17}$$

where $\langle nl | i n' l \rangle = \int_0^\infty dr r^2 R_{nl}(r) R_{n'l}^{(i)}(r)$ (4.18)

is the overlap between the radial function R_{nl} for the residual ion and one of the set of radial functions used in the central-field expansion of the initial state. If this set is chosen to be the same as the complete set of final one-electron orbitals, i.e., $R_{nl}^{(i)} = R_{nl}$, (4.17) reduces to a single sum. Although such a choice also seems appropriate for the knockout process under consideration, it does not lead to a rapidly convergent expansion for the ground state of He. In order to insure satisfactory convergence with hydrogenic wave functions, it is essential to use expansions in which the leading term takes into account the screening of the nuclear charge.

By examining the three-term Hylleraas wave function for He, Green *et al.* have obtained a rapidly converging expansion using s and p orbitals. The best choices for the screened nuclear charge are close to the simple variational value used above for $l = 0$, and about 4.5 for $l = 1$. It is also found that $c_l(n_1 n_2)$ is large only for $n_1 = n_2 = l + 1$, or in the notation of Ref. 11,

$$c_l(n_1 n_2) \approx (2l+1)^{-1/2} c_l \delta_{n_1, n_2} \delta_{n_1, l+1} \tag{4.19}$$

with $c_0 \cong 1$ and $c_1 = 0.063$. The next terms, $(2s)^2$ and $(3p)^2$, involve coefficients smaller by 2 and 4%, respectively. Because these corrections must still be multiplied by the radial overlap integrals in (4.17), their effect will indeed be small. For the residual ion left in the $(n=1, l=0)$ state, this implies that (4.17) reduces to

$$\gamma_{10}^{(i)}(\kappa) = F_{10}^{(i)}(\kappa) \langle f10 | i10 \rangle , \tag{4.20}$$

which is really the same as (4.9), i.e., the same as the best uncorrelated two-electron function based on hydrogenic orbitals. In other words, the correlation effects can be ignored in this case to an accuracy of at least 1%.

Because there is only one shell in He, knockout processes in which the final ion is excited can be easily detected by energy analysis. Taking into account the degeneracy of these states, the cross section for leaving the ion in the $n=2$ states, for example, is proportional to

$$|F_{20}^{(i)}(\kappa) \langle f20 | i10 \rangle|^2 + c_1^2 |F_{21}^{(i)}(\kappa) \langle f21 | i10 \rangle|^2 .$$

The first term is the same as (4.9) for an uncorrelated initial state, but the second term is a

direct consequence of the correlations. The smallness of $c_1 = 0.063$, together with the Coulombic degeneracy in He^+, makes observation of this term very difficult for He. It might be possible, however, to do this for the alkaline earths. In this case, the observation of an np momentum distribution for a target approximately described as $(ns)^2$ – together with a summed-energy measurement indicating the residual ion to be in an np state – is clear evidence for electronic correlations. For the alkaline earths these correlations arise from interactions between the two ns electrons and, to a lesser extent, from their interactions with core electrons.

In conclusion, we find that correlations in the He ground state will be unimportant if the recoil ion is left in an s state. It also seems reasonable to expect this to be true also for alkaline earth atoms. For these systems, the effects of correlations, although small, may be directly observable by resolving processes in which the final ion is in an $l > 0$ state. This somewhat negative conclusion should not be too surprising because we are considering a *single* ionization, or knockout process. To measure correlation effects more directly, one must measure the angular distributions of the three electrons in double ionization.

V. SUMMARY

The basic conclusion of this paper is that the momentum distributions of atomic electrons can be observed directly by measuring the angular distribution of the outgoing electrons in single ionization by a high-energy electron. We have demonstrated this by analyzing in detail the simplest experiment of this type; it involves knocking out the weakest bound electron from one- and two-electron atoms and selecting processes in which the outgoing electrons have the same energy and make the same angle with the incident beam ("symmetrical situation"). Measurements of this kind can be expected to provide new information on the self-consistent field of the atomic electrons.

In this preliminary discussion, we have not covered a number of interesting variations on the basic experiment discussed here. In one category are experiments which depart from the symmetrical situation. Of particular interest would be situations involving small momentum transfers, in contrast to the large momentum transfers involved in the present discussion. In another category are experiments involving different projectiles, such as protons and other heavy ions, and other kinds of targets, for example molecules. In addition, we have not discussed the ionization from inner shells. We plan to treat these topics at the appropriate time, i.e., when experimental results become available.

Finally it must be emphasized that measurement of *both* outgoing electrons is important at any energy, and that a real understanding of the ionization process requires such experiments. We have restricted this first discussion to high incident energies because this is the only energy range where reaction theory is reliable enough to provide an interpretation of the experiments, and because this range should provide some interesting new physical information about atomic structure.

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