PHYSICAL REVIEW A VOLUME 36, NUMBER 3 AUGUST 1, 1987

Correlation in atomic scattering

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(Received 19 February 1987)

Correlation due to the Coulomb interactions between electrons in many-electron targets colliding with charged particles is formulated, and various approximate probability amplitudes are evaluated. In the limit that the electron-electron, $1/r_{ij}$, correlation interactions are ignored or approximated by central potentials, the independent-electron approximation is obtained. Two types of correlations, or corrections to the independent-electron approximation due to $1/r_{ij}$ terms, are identified: namely, static and scattering correlation. Static correlation is that contained in the asymptotic, e.g., bound-state, wave functions. Scattering correlation, arising from correlation in the scattering operator, is new and is considered in some detail. Expressions for a scattering correlation amplitude, static correlation or rearrangement amplitude, and independent-electron or direct amplitude are derived at high collision velocity and compared. At high velocities the direct and rearrangement amplitudes dominate. At very high velocities, v, the rearrangement amplitude falls off less rapidly with v than the direct amplitude which, however, is dominant as electron-electron correlation tends to zero. Comparisons with experimental observations are discussed.

I. INTRODUCTION

The many-body and many-electron problem is central to basic understanding in various areas of physics, chemistry, and biology. In atomic physics there is an advantage in studying the many-body problem since the two-body and parts of the three-body problem are well understood. A generally useful concept in the study of the many-body problem is correlation, the problem of how particles depend on one another. This interdependence may be understood as deviation from an independent-particle picture. In atomic physics the physical cause of correlation in the many-electron problem is usually the Coulomb interaction between electrons. It is this problem of correlation applied to atomic collisions that is considered in this paper.

The many-electron problem may be studied directly in atomic physics by considering multiple-electron transitions. Over the past several years there have been a number of experimental studies[1-17] of two-electron transitions in simple targets. These include studies of double ionization of helium[1-8] by various projectiles, including most recently an experiment[1] with antiprotons at CERN (European Organization for Nuclear Research) by Andersen and co-workers. Multiple ionization in molecular hydrogen has been observed[10-12] by Edwards and co-workers for both electron and proton impact. Simultaneous ionization and electron capture, first investigated[13] by Horsdal-Pedersen and Larsen, has now been studied[14-17] for various projectiles. The independent-electron approximation has generally provided a useful conceptual approach for understanding[17-21] these observations, although the need for including correlations is becoming evident especially at high collision velocities. Analysis[8,14-17] of the experimental results above has been done using direct (uncorrelated) and rearrangement (simply correlated) mechanisms. This analysis follows studies,[18-21] begun earlier, of multiple inner-shell ionization at moderately high velocities where it was shown that the uncorrelated independent-electron picture could be used. Other studies[22-26] of outer-shell multiple ionization have shown that correlation can play a dominant role as well.

In this paper we address the problem of correlation in atomic collisions, giving a common foundation for the examples listed above. We start with the uncorrelated independent-particle picture,[27-35] or direct mechanism,[15] and add correlation. It is useful, we believe, to recognize two kinds of correlation: static and scattering correlation. Static correlation is correlation carried in the asymptotic, e.g., bound-state, wave functions. It is this static correlation that gives rise to the rearrangement mechanisms[15] as well as most multiple ionization in outer shells at high collision velocities. Scattering correlation is correlation intrinsic to the scattering process itself, i.e., correlation occurring during the collision. Since our development of scattering correlation is new, we devote extra attention to it in this paper.

In Sec. II we begin with a formulation of the many-electron scattering problem in terms of the evolution operator, $U(t,t_0)$, in the impact-parameter picture. Static and scattering correlation terms are generally defined after Eq. (11) as specific corrections to the independent-electron approximation. The independent-electron approximation then is rederived detailing approximations for further consideration. A simple expression for the direct mechanism is given. Then static and scattering correlation terms are considered. In the subsection on static correlation (Sec. IV A) a simple expression for the rearrangement mechanism is presented. A general expression for scattering correlation for two electrons is reduced to an approximate form at high velocities. A simple closed-form expression valid for large impact parameters is derived. In Sec. IV B a two-electron example is considered and the correlated and uncorrelated contributions to the total probability amplitude are compared and discussed. Finally, some of the physical and mathematical features and some of the limitations of this approach are considered.

II. FORMULATION

The Hamiltonian for scattering of neutral atom of nuclear charge Z_T by a particle of charge Z_P and mass M is (using atomic units)

$$H=-\frac{\nabla^2}{2M}+\frac{Z_PZ_T}{R}-\sum_{j=1}^{Z_T}\frac{Z_P}{|\mathbf{R}-\mathbf{r}_j|}+\sum_{j=1}^{Z_T}\left[\frac{-\nabla_j^2}{2}-\frac{Z_T}{r_j}+\sum_{\substack{k,j\\(k>j)}}\frac{1}{|\mathbf{r}_k-\mathbf{r}_j|}\right] \tag{1}$$

$$=K+V+H_0=K+H_{\text{el}}\ , \tag{2}$$

where

$$K=-\frac{\nabla^2}{2M} \tag{3}$$

and

$$V=\frac{Z_PZ_T}{R}-\sum_{j=1}^{Z_T}\frac{Z_P}{|\mathbf{R}-\mathbf{r}_j|}=Z_P\sum_{j-1}^{Z_T}\left[\frac{1}{R}-\frac{1}{|\mathbf{R}-\mathbf{r}_j|}\right]\equiv\sum_{j=1}^{Z_T}V_j \tag{4}$$

and

$$H_0=\sum_{j=1}^{Z_T}\left[-\frac{\nabla_j^2}{2}-\frac{Z_T}{r_j}+\sum_{\substack{k,j\\(k>j)}}|\mathbf{r}_k-\mathbf{r}_j|^{-1}\right]\ . \tag{5}$$

Here $\mathbf{R}$ is the internuclear coordinate and $\mathbf{r}_j$ the coordinate to the jth electron. The $|\mathbf{r}_k-\mathbf{r}_j|^{-1}$ electron-electron interaction gives rise to spatial electron-electron correlations present in the unperturbed (static) atomic Hamiltonian H_0.

It is now assumed that the internuclear motion may be separated[36] from the electronic motion and that the internuclear motion may be treated classically, so that the internuclear trajectory $\mathbf{R}(t)$ is well defined. The resulting equation[37] for the electron motion is now time dependent, namely,

$$i\frac{d\psi_{\text{el}}}{dt}=H_{\text{el}}\psi_{\text{el}}\ . \tag{6}$$

Because

$$H_{\text{el}}=H_0+V \tag{7}$$

is now explicitly time dependent since V depends on $\mathbf{R}$ which is explicitly time dependent, the evolution operator U is not simply given by $\exp[-iH_{\text{el}}(t-t_0)]$, but rather by a more complicated expression containing time ordering, namely, Eq. (10) below.

In order to recover standard results, including the semiclassical Coulomb approximation (SCA) result,[38,39] it is useful to work in the intermediate presentation, where one takes full advantage of the fact that the eigenfunctions of H_0 are known (or nearly known). In the intermediate representation, the evolution operator $U(t,t_0)$ is governed[40] by

$$i\frac{d}{dt}U_I(t,t_0)=V_I(t)U_I(t,t_0)\ , \tag{8}$$

where

$$V_I(t)\equiv e^{-iH_0t}V(t)e^{iH_0t}\ . \tag{9}$$

Equation (8) may be formally solved[37] to yield (with T as the time-ordering operator)

$$\begin{aligned}U(t,t_0)&=T\exp\left[-i\int_{t_0}^{t}V_I(t_1)dt_1\right]\\&=1+(-1)\int_{t_0}^{t}dt_1V_I(t_1)+(-i)^2\int_{t_0}^{t}dt_2V_I(t_2)\int_{t_0}^{t_2}dt_1V_I(t_1)\\&\quad+(-i)^3\int_{t_0}^{t}dt_3V_I(t_3)\int_{t_0}^{t}dt_2V_I(t_2)\int_{t}^{t_2}dt_1V_2(t_1)+\cdots\ ,\end{aligned} \tag{10}$$

namely, Dyson's equation.

The probability amplitude for scattering from the asymptotic initial state ϕ_i to the asymptotic final state ϕ_f is given by

$$a^{if}=\langle\phi_f|\psi_i\rangle=\langle\phi_f|U(+\infty,-\infty)|\phi_i\rangle\ . \tag{11}$$

From this basic expression we identify two kinds of correlation: static correlation coming from correlation in the asymptotic states ϕ_f and ϕ_i, and scattering correlation arising from correlation in the evolution operator U. In this paper a quantity is generally defined to be uncorrelated if that quantity is determined from a single sum of uncorrelated single-electron Hamiltonians even though the quantity obeys the Pauli exclusion principle and appropriate symmetry requirements such as rotational symmetry (conserving total angular momentum) and parity. Correlation is the difference between the exact and uncorrelated quantities. Usually static correlation corresponds to deviations from a product wave function for ϕ and scattering correlation corresponds to deviation of the evolution operator U from a product of single-particle operators. As we explain in Sec. IV, both types of correlation arise physically from the electron-electron Coulomb interaction, often called the correlation interaction in atomic physics. An explicit expression for scattering correlation valid to first order in the correlation interaction is given by Eq. (29) in Sec. IV. Simple examples of static correlation and scattering correlation are discussed in Sec. V.

III. INDEPENDENT-ELECTRON APPROXIMATION

Since we define correlation as the deviation from the independent-electron approximation, we now review briefly the independent-electron approximation for atomic scattering. In the independent-electron approximation,[27–35] the probability amplitude for a multielectron transition in an ion-atom collision is simply a product of independent single-electron transition probabilities amplitudes. The independent-electron approximation is exact under the following conditions.[29]

(i) The projectile is a point charge.

(ii) The internuclear motion is separated from the electronic motion and treated as elastic classical scattering.

(iii) The $|\mathbf{r}_k-\mathbf{r}_j|^{-1}$ electron-electron interactions are approximated by single-electron potentials.

Condition (i) is fully satisfied in collisions of atoms with electrons or bare ions. Condition (ii) is well satisfied[36] for incident heavy ions and may be satisfied for high-velocity incident electrons[41] whose de Broglie wavelength is small compared to atomic distances. Constraints due to condition (iii), which corresponds to ignoring the effects of electron-electron correlations during the collision, are considered in further detail in Sec. IV. The Pauli exclusion principle is usually included in the definition of an independent-particle model in the static many-body problem. However, in the scattering problem, electron identity can be difficult to include and is often ignored in practice. This important question of Pauli exclusion effects in summing over final states in many-electron ($n\geq 3$) targets has been well addressed by Reading and Ford,[30] and is therefore bypassed here, except to note that there are some systems in which these exclusion effects play no role. Attention here is therefore focused on condition (iii), namely, the question of scattering correlations.

Electron-electron correlation potentials appear in Eq. (5) where H_0 may be written as a sum of operators H_0^j which reduce to single-particle operators when correlations are ignored. Specifically, from Eq. (5),

$$H_0=\sum_{j=1}^{Z_2}\left[-\frac{\nabla_j^2}{2}-\frac{Z_2}{r_j}+\sum_{\substack{k,j\\(k>j)}}|\mathbf{r}_k-\mathbf{r}_j|^{-1}\right]\equiv\sum_{j=1}^{Z_2}\tilde{H}_0^j\ . \tag{12}$$

Because

$$\sum_{\substack{k,j\\(k>j)}}|\mathbf{r}_k-\mathbf{r}_j|^{-1}$$

is not a single-electron operator, the operators $\tilde{H}_0^j$ are not single-electron operators. However, if the

$$\sum_{\substack{k,j\\(k>j)}}|\mathbf{r}_k-\mathbf{r}_j|^{-1}$$

electron-electron interactions which give rise to correlations are approximated by an average potential, i.e.,

$$\sum_{\substack{k,j\\(k>j)}}|\mathbf{r}_k-\mathbf{r}_j|^{-1}\simeq v_j(r_j)\ ,$$

then the H_0^j terms are indeed single-electron operators H_0^j and correlation disappears.

Now if electron-electron correlations are ignored, as in Eq. (12), then $[H_0^i,H_0^j]=0$ since the H_0^j are single-particle operators. Recalling from Eq. (4) that V is a sum of single-particle potentials V_j, and noting that $[H_0^k,V_j]\sim\delta_{kj}$, then

$$\begin{aligned}V_I(t)&=e^{-iH_0t}Ve^{iH_0t}\\&=\sum_j\exp\left[-i\sum_k H_0^k t\right]V_j\exp\left[i\sum_l H_0^l t\right]\\&=\sum_j e^{-iH_0^jt}V_je^{iH_0^jt}=\sum_j V_I^j(t)\ .\end{aligned} \tag{13}$$

That is, without electron-electron correlations, $V_I(t)$ is a sum of single-particle operators $V_I^j(t)$, and as a consequence the evolution operator reduces[29,35] to a product of evolution operators, i.e.,

$$\begin{aligned}U(t,t_0)&=T\exp\left[-i\sum_j\int_{t_0}^{t_1}V_I(t_1)dt_1\right]\\&=\prod_j T\exp\left[-i\int_{t_0}^{t_1}V_I(t_1)dt_1\right]\\&=U_0(t,t_0)=\prod_j U_j(t,t_0)\ .\end{aligned} \tag{14}$$

The electrons evolved independently during the collision.

Finally, if Pauli exclusion[27] is ignored, then the probability amplitude is a product of independent-electron amplitudes, namely,

$$a^{if}=\langle\phi^f|\Psi^i\rangle=\langle\phi^f|U|\phi^i\rangle=\prod_j\langle\phi_j^f|U_j|\phi_j^i\rangle=\prod_j a_j^{if}\ , \tag{15}$$

where Ψ represents the full electron wave function and ϕ the asymptotic wave function. Consequently, the transition probability $|a^{if}|^2$ is a product of independent-electron probabilities in the independent-electron approximation, thus simplifying considerably the many-body problem.

The direct mechanism[15] corresponds to the use of Eq. (15) which leads to a binomial distribution[29] of single-electron probabilities, $P_j(B)=|a_j^{if}|^2$, where B is the impact parameter of the projectile. It is not necessary to use perturbation theory to evaluate the single-particle probabilities, $P_j(B)$, since the independent-electron approximation does not depend on the strength of the interaction potential V. However, it is often convenient and at high velocities appropriate to use perturbation theory for the $P_j(B)$. For a target with two electrons both undergoing transitions we have from Eqs. (11) and (14) using first-order perturbation theory:

$$\begin{aligned}a_D^{if}&=\langle\phi_2^f\phi_2^f|U_1(-\infty,+\infty)U_2(-\infty,+\infty)|\phi_1^i\phi_2^i\rangle\\&=\langle\phi_1^f|U_1|\phi_1^i\rangle\langle\phi_2^f|U_2|\phi_2^i\rangle=a_1a_2\\&=\left[-i\int\langle\phi_1^f|V_{I1}|\phi_1^i\rangle dt\right]\\&\quad\times\left[-i\int\langle\phi_2^f|V_{I2}|\phi_2^i\rangle dt\right]\ .\end{aligned} \tag{16}$$

Since this amplitude is second order in the interaction potential, it corresponds to a second Born amplitude expressed as a product of two first Born amplitudes. Further reduction of this simple example is given in Sec. V.

IV. CORRELATION

Now let us consider correlation, i.e., corrections to the independent-electron approximation due to the electron-electron interactions. As we have noted after Eq. (11), there are two types of correlation, namely, static and scattering correlation.

A. Static correlation

Static correlation is that included in the asymptotic wave functions, ϕ_i and ϕ_f. Usually these asymptotic wave functions are expressed as bound-state wave functions of the target in the initial and final states, respectively. Sometimes distortions due to the projectile are included. In any case, the electron-electron correlation is that of the target wave functions. This problem of correlation in static atomic wave functions has received more attention than the more difficult problem of correlation in atomic scattering. As a consequence, wave functions including static correlation are available. It is these wave functions, such as correlated Hartree-Fock wave functions, which may be used for ϕ_i and ϕ_f in the transition amplitudes of Eq. (11).

Sometimes the asymptotic states ϕ_i and ϕ_f are eigenfunctions of different asymptotic Hamiltonians because the correlation in the initial and final asymptotic states are different. For example, if an electron is removed from helium, then the initial wave function of helium contains electron-electron correlation, but the final wave function may be uncorrelated asymptotically. Then ϕ_i and ϕ_f are nonorthogonal. This nonorthogonality can result in nonvanishing matrix elements giving finite results for certain, e.g., multiple, transitions. Such calculations have been in use for some time. As Bransden and Dalgarno[42] have pointed out, the physical cause for such transitions is the electron-electron correlation interaction v. Since this correlation effect is expressed in the asymptotic wave functions, we regard it as a static correlation.

An example of such a static correlation effect is the rearrangement[15] or shakeoff mechanism. Shakeoff is caused by a change in correlation following removal of an electron by the projectile leading to rearrangement in the final state which results in the ionization of a subsequent electron. A simple example can be easily obtained by including nonorthogonal asymptotic wave functions in Eq. (15) and assuming that the projectile interacts with only one target electron. Then we have

$$a_R = \left\langle \phi^f \middle| T \exp\left[-i \int_{-\infty}^{\infty} V_I^1(t) dt \right] \middle| \phi_i \right\rangle . \tag{17}$$

If we further approximate ϕ_i and ϕ_f as product wave functions, then the simple expression that results is

$$a_R = \langle \phi_1^f \,|\, U_1 \,|\, \phi_1^i \rangle \prod_{j=2}^{n} \langle \phi_j^f \,|\, \phi_j^i \rangle . \tag{18}$$

If first-order perturbation theory is used for a nth-order effect, then a_R is first order in the interaction potential and $(n-1)$th order in the correlation interaction. This amplitude is further discussed in Sec. IV.

B. Scattering correlation

In this section we consider the influence of the electron-electron Coulomb interactions on the scattering process itself. We start with a set of basic states, $\{\phi_j\}$, which are eigenfunctions of the uncorrelated H_0^j and express the interaction operator of the projectile and the jth electron as

$$V_I^j(t) = V_I^{j0}(t) + V_I^{jC}(t) , \tag{19}$$

where V_I^{j0} is the limit of V_I^j as the correlation potentials, $|\mathbf{r}_i - \mathbf{r}_j|^{-1}$ go to zero, corresponding to the independent-electron approximation.

For simplicity we consider here a two-electron system. It is straightforward to generalize the result to a system with many electrons. For a two-electron system the unperturbed Hamiltonian from Eq. (5) is simply

$$\begin{aligned} H_0 &= \left[\frac{-\nabla_1^2}{2} - \frac{Z_2}{r_1} \right] + \left[\frac{-\nabla_2^2}{2} - \frac{Z_2}{r_2} \right] + r_{12}^{-1} \\ &= \left[\frac{-\nabla_1^2}{2} - \frac{Z_2}{r_1} + v_1(r_1) \right] + \left[\frac{-\nabla_2^2}{2} - \frac{Z_2}{r_2} + v_2(r_2) \right] \\ &\quad + [r_{12}^{-1} - v_1(r_1) - v_2(r_2)] \\ &\equiv H_0^1 + H_0^2 + v , \end{aligned} \tag{20}$$

where H_0^1 and H_0^2 are true one-electron operators and v is a correlation potential, e.g., $|\mathbf{r}_i - \mathbf{r}_j|^{-1}$.

The interaction potential, corresponding to Eq. (9), is

$$V_I(t) = e^{-iH_0 t} V(t) e^{iH_0 t} = e^{-i(H_0^1 t + H_0^2 t + vt)} [V_1(t) + V_2(t)] e^{i(H_0^1 t + H_0^2 t + vt)} = V_I^1 + V_I^2 = \sum_{j=1}^{2} V_I^j , \tag{21}$$

where V_I^j corresponds to the interaction between the projectile and the jth electron. Since $[H_0^j, v] \neq 0$, now the propagators $e^{\pm iH_0 t}$ in V_I^j do not factor into a product of independent-electron propagators, and V_I^j contains operators acting on electrons other than the jth electron. That is, because $[H_0^j, v] \neq 0$, it follows that V_I^j is a many-body operator.

The many-body operator for scattering correlation V_I^{jC} is the difference of the full operator of Eq. (20) and the independent-electron operator with $v=0$ in Eq. (20).

Next we proceed to expand Eq. (20) to first order in the correlation potential v to obtain a first-order expression for the scattering correlation operator V_I^{jC}.

1. First-order operator

In principle the full many-electron operator V_I^j of Eq. (21) may now be used in Eqs. (10) and (11) to evaluate fully correlated scattering amplitudes. In practice this is not easily done. However, if we consider cases where the correlation potential v is weak, we may expand terms in Eq. (10) in a perturbation series in v, and identify and evaluate the resulting first-order scattering correlation. We now proceed to expand the many-electron operator V_I^j into an uncorrelated part V_I^{j0} and a correlated part V_I^{jC} to lowest order in v in the form of Eq. (19), i.e., $V_I^j = V_I^{j0} + V_I^{jC}$.

We begin our expansion by defining commutator operators, linear in v, that generate scattering correlation, namely,

$$\beta_1 \equiv [H_0^1, v] \tag{22a}$$

and

$$\beta_2 \equiv [H_0^2, v] \; . \tag{22b}$$

These β commutators, discussed in Sec. V, may now be used to factor the $e^{iH_0 t}$ propagators using

$$e^{A+B} = e^{A} e^{B} e^{-1/2[A,B]} \ldots \tag{23}$$

corresponding[43] to the Baker-Hausdorff theorem.

At this point only terms to first order in βt^2 are retained. Since an nth-order commutator carries a factor of t^{n+1}, this expansion is convergent, for short collision times, as discussed further in Sec. V. Now, noting that $[H_0^1, H_0^2] = 0$, we have

$$\begin{aligned} V_I^1(t) &= e^{-i(H_0^1+H_0^2+v)t} V_1 e^{i(H_0^1+H_0^2+v)t} \\ &= e^{-i^2/2[H_0^1,v]t^2} e^{-iH_0^1 t} e^{-i(H_0^2+v)t} \\ &\quad \times V_1 e^{i(H_0^2+v)t} e^{iH_0^1 t} e^{i^2/2[H_0^1,v]t^2} + O([\beta_1, H_0]t^3) \; . \end{aligned} \tag{24}$$

Since V_1 is simple function of $\mathbf{R}$ and $\mathbf{r}_1$, e.g., $V_1 = -Z_P / |\mathbf{R} - \mathbf{r}_1|$, therefore $[H_0^2, V_1] = [v, V_1] = 0$ and

$$V_I^1(t) = e^{(1/2)\beta_1 t^2} V_I^{10}(t) e^{-(1/2)\beta_1 t^2} \; , \tag{25}$$

where

$$V_I^{10}(t) \equiv e^{-iH_0^1 t} V_1 e^{iH_0^1 t} \tag{26}$$

is the single-electron operator to which V_I^j reduces as v (and therefore β_j) goes to zero. Expanding to first order in β, t^2,

$$V_I^1(t) = V_I^{10} + \tfrac{1}{2}[\beta_1, V_I^{10}]t^2 \; . \tag{27a}$$

Similarly,

$$V_I^2 = V_I^{20} + \tfrac{1}{2}[\beta_2, V_I^{20}]t^2 \tag{27b}$$

so that

$$V_I^j = V_I^{j0} + \tfrac{1}{2}[\beta_j, V_I^{j0}]t^2 \; . \tag{27c}$$

This is of the form of Eq. (19) with $V_I^{jC} \simeq \frac{1}{2}[\beta_j, V_I^{j0}]t^2$. Hence, the many-electron operator V_I^j is written as a sum of a one-electron operator V_I^{j0} (which produces no correlations) and a many-electron scattering operator given to first order by V_I^{jC}, which goes to zero as the correlation v disappear.

The lowest-order effect of the scattering correlation operator on the evolution operator is easily seen by expanding U in Eq. (10) to first order in the interaction potential $V_I^j(t)$ using Eq. (27c), namely,

$$\begin{aligned} U(t,t_0) &= T \exp\left[-i \int_{t_0}^{t} V_I(t_1) dt_1 \right] \\ &\simeq 1 - i \int_{t_0}^{t} V_I(t_1) dt_1 \\ &= 1 - i \int_{t_0}^{t} V_I^0(t_1) dt_1 - \frac{i}{2} \int_{t_0}^{t} \sum_{j=1} [\beta_j, V_I^{j0}(t_1)] t_1^2 dt_1 \\ &= 1 - i \int_{t_0}^{t_1} V_I^0(t_1) dt_1 - i \int_{t_0}^{t} V_I^C(t_1) dt_1 \; , \end{aligned} \tag{28}$$

where $V_I^C(t) = \sum_j [\beta_j, V_I^{j0}(t)]t^2$. Since only the last term contributes to scattering correlations, one has an explicit expression valid to first order in the correlation potential v for scattering correlation, namely,

$$\begin{aligned} U - U_0 &= -\frac{i}{2} \int_{t_0}^{t} \sum_j [\beta_j, V_I^{j0}(t_1)] t_1^2 dt_1 \\ &= -i \int_{t_0}^{t} V_I^C(t_1) dt_1 \; , \end{aligned} \tag{29}$$

where β_j, V_I^{j0}, and v are defined by Eqs. (22), (26), and (20), respectively. The right side of Eq. (29) expresses the lowest-order correction to the independent-electron approximation due to scattering correlations, as defined after Eq. (11).

2. First-order matrix element

The first-order scattering correlation amplitude is found by taking the matrix element of Eq. (29) above as $t_0 \to -\infty$ and $t \to +\infty$, namely,

$$\begin{aligned} a_{\mathrm{SC}} &= -i \sum_{j=1}^{2} \int_{-\infty}^{\infty} \langle \phi_f | \tfrac{1}{2}[\beta_j, V_I^{j0}(t_1)] | \phi_i \rangle t_1^2 dt_1 \\ &= -i \sum_{j=1}^{2} \int_{-\infty}^{\infty} \langle \phi_f | V_I^{jC} | \phi_i \rangle dt \; . \end{aligned} \tag{30}$$

Using $V_j = -Z_P / |\mathbf{R} - \mathbf{r}_j|$ and $v = |\mathbf{r}_1 - \mathbf{r}_2|^{-1}$, then it is straightforward to show that

$$\begin{aligned} V_I^{jC} &= \tfrac{1}{2}[\beta_j, V_I^{j0}]t^2 \\ &= \frac{1}{2} \left[\left[-\tfrac{1}{2}\nabla_j^2, \frac{1}{r_{12}} \right], e^{-iH_0^j t} \frac{Z_P}{|\mathbf{R} - \mathbf{r}_j|} e^{iH_0^j t} \right] t^2 \\ &= -e^{-iH_0^j t} \frac{Z_P}{2} \left(\boldsymbol{\nabla}_j \frac{1}{r_{12}} \right) \cdot \left(\boldsymbol{\nabla}_j \frac{1}{|\mathbf{R} - \mathbf{r}_j|} \right) e^{+iH_0^j t} t^2 \\ &\quad + O([\beta_j, H_0]t^3) \; . \end{aligned} \tag{31}$$

As in Eq. (24) we again ignore the $O([\beta_j, H_0]t^3)$ contribution. Then, with ϕ_i and ϕ_f as eigenfunctions of the uncorrelated H_j^0, we have

$$a_{\mathrm{SC}}=-i\frac{Z_P}{2}\sum_{j=1}^{2}(-1)^j\int_{-\infty}^{\infty}dt\,t^2e^{-i\omega t}\left\langle\phi_f\left|\frac{(\mathbf{r}_1-\mathbf{r}_2)}{|\mathbf{r}_1-\mathbf{r}_2|^3}\cdot\frac{(\mathbf{R}-\mathbf{r}_j)}{|\mathbf{R}-\mathbf{r}_j|^3}\right|\phi_i\right\rangle\,, \tag{32}$$

where $\omega=(E_i-E_f)$. If ϕ_i and ϕ_f are expanded in spherical harmonics of $\hat{\mathbf{r}}_1$ and $\hat{\mathbf{r}}_2$, then the angular integrations are easily done applying the integral of Deb *et al.*[44] twice. Further reduction depends on the choice of radial wave functions chosen for ϕ_i and ϕ_f.

A simple expression for the scattering correlation amplitude of Eq. (32) may be obtained by considering large impact parameters. We then approximate $|\mathbf{R}-\mathbf{r}_j|^{-3}\simeq R^{-3}(1+3\mathbf{r}_j\cdot\mathbf{R}/R^2)$. Using this in Eq. (32) after some algebra, we obtain

$$a_{\mathrm{SC}}=iZ_P\int_{-\infty}^{\infty}dt\,t^2e^{-i\omega t}\left\langle\phi_f\left|\frac{P_2(\hat{\mathbf{r}}_{12}\cdot\hat{\mathbf{R}})}{R^3r_{12}}\right|\phi_i\right\rangle\,, \tag{33}$$

where P_2 is the second-order Legendre polynomial. Using $R^2=B^2+Z^2$, $Z=vt$, and $q_0=\omega/B$, we obtain

$$\begin{aligned}a_{\mathrm{SC}}&=-i\frac{Z_P}{v^3}\int_{-\infty}^{\infty}e^{iq_0Z}\left\langle\phi_f\left|\frac{P_2(\mathbf{r}_{12}\cdot\hat{\mathbf{R}})}{r_{12}}\right|\phi^i\right\rangle\\&\quad\times\frac{Z^2}{(B^2+Z^2)^{3/2}}dZ\\&=-2i\frac{Z_P}{v^3}Q^{if}[K_0(q_0B)-(q_0B)K_1(q_0B)]\,,\end{aligned} \tag{34}$$

where $Q^{if}=\langle\phi_f\,|\,P_2(\hat{\mathbf{r}}_{12}\cdot\hat{\mathbf{R}})/r_{12}\,|\,\phi_i\rangle$ is a quadrupole matrix element, K is a modified Bessel function, and now v is the collision velocity. This amplitude, further discussed in Sec. V, is first order in the interaction potential and first order in the correlation potential.

V. DISCUSSION

Although the independent-electron approximation has provided a useful starting point for understanding the nature of multiple-electron transitions in atomic collisions,[17–20,28–35] it is becoming increasingly clear that correlation is required for more complete understanding. A good example is provided by studies[1–8] of single and double ionization of helium by high velocity electrons, protons, ions, and antiprotons where double-ionization cross sections predicted[15,34] by the independent-electron approximation can be an order of magnitude or more too small at velocities above several MeV/amu. Predictions using static correlation using our methods[8,15] are within a factor of 2 of these observations and the energy dependence of the calculations is also significantly improved when static correlation is included. A major difficulty here is our lack of knowledge of the continuum wave function for two correlated electrons. The most complete calculation at this time is the recent result of Reading and Ford[45] who use pseudostates for the continuum and include correlation in a coupled channel calculation. Their results are within 50% of observation. Another example illustrating the applicability of our methods is[14] capture plus ionization in helium by protons, α particles, and fully stripped lithium ions. Again the independent-electron approximation fails by more than an order of magnitude, e.g., above 1 MeV/amu for proton impact, while including correlation[13,14] gives agreement within a factor of 2 and improves the energy dependence of the cross sections significantly. At lower collision velocities correlation is also important[46] and scattering correlation, as discussed below, may be significant.

To illustrate the relative importance and nature of uncorrelated independent-electron approximation or direct contributions, static correlation or rearrangement, and scattering correlation, let us consider a two-electron target undergoing a transition from initial state ϕ_i to final state ϕ_f via an interaction with a particle of charge Z_P and velocity v. The exact probability amplitude given by Eq. (11) may be separated into two terms,

$$\begin{aligned}a&=\langle\phi_f\,|\,U\,|\,\phi_i\rangle=\langle\phi^f\,|\,U_0\,|\,\phi^i\rangle+\langle\phi^f\,|\,U-U_0\,|\,\phi^i\rangle\\&=a_{\mathrm{static}}+a_{\mathrm{SC}}\,.\end{aligned} \tag{35}$$

The a_{static} amplitude contains only static correlation. Both static correlation, from ϕ_i and ϕ_f, and scattering correlation, from $U-U_0$, are included in a_{SC} which contains all the scattering correlation. Within the impact-parameter picture this expression is exact.

The direct and rearrangement amplitudes may both be obtained in the simplest case by expanding $a_{\mathrm{static}}=\langle\phi_f\,|\,U_1U_2\,|\,\phi_i\rangle$ in both v and V. If v is small $\phi\simeq\phi_1\phi_2$ and if V is small $U\simeq 1-i\int V_I dt$. Then

$$\begin{aligned}a_{\mathrm{static}}&=\langle\phi_{f1}\,|\,\phi_{i1}\rangle\langle\phi_{f2}\,|\,\phi_{i2}\rangle-\left[i\left\langle\phi_{f1}\left|\int V_{11}dt\right|\phi_{i1}\right\rangle\langle\phi_{f2}\,|\,\phi_{i2}\rangle+i\left\langle\phi_{f2}\left|\int V_{I2}dt\right|\phi_{i2}\right\rangle\langle\phi_{f1}\,|\,\phi_{i1}\rangle\right]\\&\quad-\left\langle\phi_{f2}\left|\int V_{11}dt\right|\phi_{i2}\right\rangle\left\langle\phi_{f2}\left|\int V_{12}dt\right|\phi_{i2}\right\rangle\\&=\langle\phi_f\,|\,\phi_i\rangle+a_R+a_D\,.\end{aligned}$$

We ignore the first term since correlation alone cannot cause a transition. It is sensible to retain the shakeoff amplitudes, $\langle\phi_{fj}\,|\,\phi_{ij}\rangle$ in a_R, nevertheless, since the potential V does change the system, e.g., by removing an electron, so that correlation may effect subsequent change. Hence if both v and V are small, we have

$$\begin{aligned}a_{\mathrm{static}}&\simeq a_1a_2+a_1\langle\phi_{f2}\,|\,\phi_{i2}\rangle+a_2\langle\phi_{f1}\,|\,\phi_{i1}\rangle\\&=a_D+a_R\,,\end{aligned} \tag{36}$$

where $a_j=-i\langle\phi_{fj}\,|\,\int V_{Ij}dt\,|\,\phi_{ij}\rangle$. Equation (35) is exact. Equation (36) is valid when a_D and a_R are both much smaller than 1 in magnitude.

Some comparison of the amplitudes for the independent-electron approximation, static correlation, and scattering correlation is possible. For simple two-electron targets using first-order perturbation theory for both the projectile and correlation interactions, we have from Eqs. (35) and (36):

$$a = a_D + a_R + a_{\mathrm{SC}} \ , \tag{37}$$

where from Eqs. (18), (20), and (34) with $t = Z/v$,

$$a_D = a_1 a_2 = -\frac{Z_P^2}{v^2} \prod_{j=1}^{2} \int_{-\infty}^{\infty} dZ\, e^{iq_0 Z} \left\langle \phi_j \left| \frac{1}{|\mathbf{R}-\mathbf{r}_j|} \right| \phi_j^f \right\rangle \ , \tag{38a}$$

$$a_R = a_1 \langle \phi_{f2} | \phi_{i2} \rangle = -\frac{iZ_P}{v} \int_{-\infty}^{\infty} dZ\, e^{iq_0 Z} \left\langle \phi_1^i \left| \frac{1}{|\mathbf{R}-\mathbf{r}_j|} \right| \phi_1^f \right\rangle \langle \phi_2^f | \phi_2^i \rangle \ , \tag{38b}$$

and

$$a_{\mathrm{SC}} = -i\frac{Z_P}{2v^3} \sum_{j=1}^{2} (-1)^j \int_{-\infty}^{\infty} dZ\, Z^2 e^{-iq_0 Z} \left\langle \phi_f \left| \frac{(\mathbf{r}_1-\mathbf{r}_2)\cdot(\mathbf{R}-\mathbf{r}_j)}{|\mathbf{r}_1-\mathbf{r}_2|^3 |\mathbf{R}-\mathbf{r}_j|^3} \right| \phi_i \right\rangle \ . \tag{38c}$$

All of the above expressions based on perturbation theory are valid at high velocities, v. At the highest velocities the rearrangement amplitude, corresponding to static correlation, is dominant. As the velocity decreases (or as the projectile charge Z_P increases), the direct amplitude becomes larger. Scattering correlation is not important until the velocity, v, is relatively low.

Additional algebraic reduction is possible for large impact parameters. Using the amplitude a_1 in first-order perturbation theory, Eqs. (38a) and (38b) may be written as

$$a_1 = -\frac{iZ_P}{v} \int_{-\infty}^{\infty} \frac{e^{iq_0 Z}}{R^2} \langle \phi_j^f | rP_1(\hat{\mathbf{r}}\cdot\hat{\mathbf{R}}) | \phi_j^i \rangle dZ$$

$$= -i\pi \frac{Z_P}{v} D_1^{if} e^{-q_0 B}/B \ , \tag{39}$$

where $D^{if} = \langle \phi_{f1} | rP_1(\hat{\mathbf{r}}\cdot\hat{\mathbf{R}}) | \phi_{i1} \rangle$ corresponding to a dipole matrix element. Using Eqs. (38a)–(38c) and Eq. (34), we have

$$a_D = -\pi^2 (Z_P^2/v^2) D_1^{if} D_2^{if} e^{-(q_0^1+q_0^2)B}/B^2 \ , \tag{40a}$$

$$a_R = -i\pi (Z_P/v) D_1^{if} e^{-q_0^1 B}/B \langle \phi_2^f | \phi_2^t \rangle \ , \tag{40b}$$

$$a_{\mathrm{SC}} = -2i\frac{Z_P}{v^3} Q^{if} [K_0(q_0 B) - (q_0 B) K_1(q_0 B)] \ . \tag{40c}$$

These expressions are valid only at large impact parameter B. This completes our simple discussion of correlation in a two-electron target.

Conceptually, as well as computationally, it is possible to distinguish between scattering correlation and static correlation depending on whether or not correlation occurs during the collision. The concepts are illustrated in the collision diagrams of Figs. 1–3. In Fig. 1 the independent-electron approximation is illustrated. Here the electrons do not interact with each other. Static correlation is included in Fig. 2. Here the electrons may interact on the way in and on the way out, but they do not interact during the collision. Because of the change in correlation during the collision, the electrons may readjust somewhat on the way out in the final state, corresponding to the rearrangement mechanism. Scattering correlation is included in Fig. 3 where the electrons interact during the collision. As we discuss further below, whether a correlation term may be classified as static or scattering correlation depends on how the calculation is done. If the correlation is included on the asymptotic wave function, it is regarded as static correlation. Otherwise, it must occur during the collision and is a scattering correlation.

Mathematically, correlation is expressed in this paper by noncommuting terms in the time evolution of the system. The commutator $\beta_j = [H_0^j, v]$ generates correlation in both the asymptotic ϕ_i and ϕ_f and in the scattering operator U. When ϕ_i and ϕ_f are fully correlated, the β_j terms are fully included in ϕ_i and ϕ_f, giving complete static correlation. Scattering correlation in U is generated

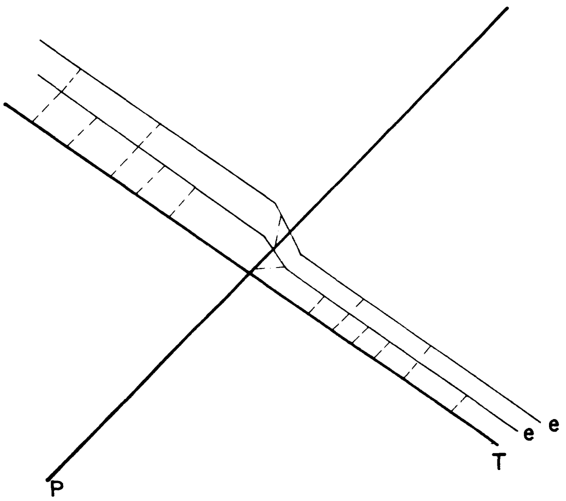

FIG. 1. Scattering event for a two-electron target (T) interacting with a projectile (P) in the independent-electron approximation. Time increases in the upward direction. Electron-electron correlation is not included.

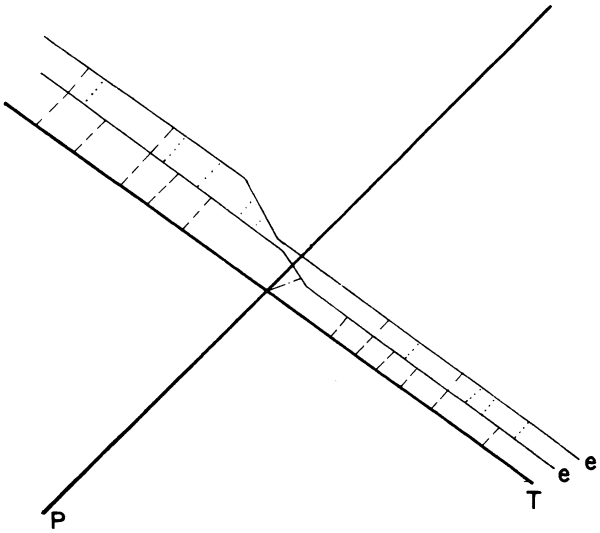

FIG. 2. Scattering event with static correlation included. Correlation is included in the asymptotic states, but no correlation is included during the collision.

by a coupling of the correlation commutator β_j with the interaction potential V_I corresponding to the double commutator $[\beta_j, V_I^{j0}]$ in Eq. (28). As seen from Eq. (31), coupling between v and V_I is done via the ∇_j operator in the electronic kinetic energy. Coupling between electrons occurs via the correlation potential v contained in β_j.

Scattering correlation has been defined as the difference between U and the uncorrelated U_0 defined in terms of single independent-electron operators

$$e^{-H_0^j t} V_j e^{iH_0^j t}$$

containing uncorrelated H_0^j operators. If fully correlated ϕ_i and ϕ_f were used, then our method should be modified because then ϕ_i and ϕ_f would not be eigenfunctions of the H_0^j which contain no correlation, but rather ϕ_i and ϕ_f would be eigenfunctions of the full H_0 which includes correlation. This would lead to difficulty in evaluation of matrix elements of $e^{-iH_0^j t} V_j e^{iH_0^j t}$. For fully correlated ϕ_i and ϕ_f it would be easier to use

$$\langle \phi_f | e^{iH_0 t} V e^{-iH_0 t} | \phi_i \rangle = \langle \phi_f | e^{iE_f t} V e^{-iE_i t} | \phi_i \rangle ,$$

thereby reducing a_{SC} to zero to first order in the interaction potential V. Then scattering correlation would be carried in Eq. (10) by matrix elements of the form $\langle \phi_m | V | \phi_n \rangle$ where ϕ_m and ϕ_n are intermediate eigenstates of H_0 including full correlation. In this paper we recognize that a fully correlated set $\{\phi\}$ often is not exactly known. Correlated continuum wave functions are especially difficult. For many problems, including the examples we have considered, the set $\{\phi\}$ is approximated by a set $\{\prod_j \phi_j\}$ of known eigenfunctions of H_0^j. It is for these uncorrelated $\prod_j \phi_j$ that our specific technique for identifying generating scattering correlation applies. With our method the action of the β commutators in generating correlation with an independent-electron basis, even in higher Born terms in Eq. (10), becomes apparent. Generalization of our approach to partially correlated ϕ_i and ϕ_f may be possible.

Now let us discuss briefly some advantages and disadvantages of our approach to the many-body problem. We first remark that there are other quite useful approaches[45–50] to the many-body problem, some of which are conceptually different, i.e., do not begin with the independent-electron approximation. However, the independent-electron approximation has provided a useful starting point for some studies of the static many-body problem and is also well defined in atomic scattering. And we are encouraged by the possibility of at least some analytic reduction of many-body matrix elements as illustrated above. Mathematical simplification sometimes leads to insight into the physical nature of a problem.

Dividing the problem of correlation into static and scattering correlation seems to us to be computationally sensible and conceptually simple. We wish to emphasize, however, that the boundary between static and scattering correlation may be varied by varying the choice of asymptotic wave functions, ϕ_i and ϕ_f. Consequently, whether a particular effect is seen as a scattering correlation or not may vary with the methods chosen (e.g., distorted-wave or partially correlated methods) to perform the calculation.

The validity of our simple expressions of Eqs. (40) depends largely on our rather extensive use of perturbation theory. While we have tried to emphasize that use of perturbation theory is not always necessary, it is central to our evaluation of the first-order scattering correlation amplitude of Eq. (32). Specifically, our commutator expansion of Sec. IV B requires that the $[\beta, V_I]t^2$ terms be much smaller than 1 but larger than $[\beta, H_0]t^3$ terms that were ignored. This basically means that the collision time must not be too large and that our methods be restricted to collision velocities higher than orbital velocities of the participating atomic electrons.

The direct and rearrangement mechanisms contained in our formulation have been useful in the analysis of vari-

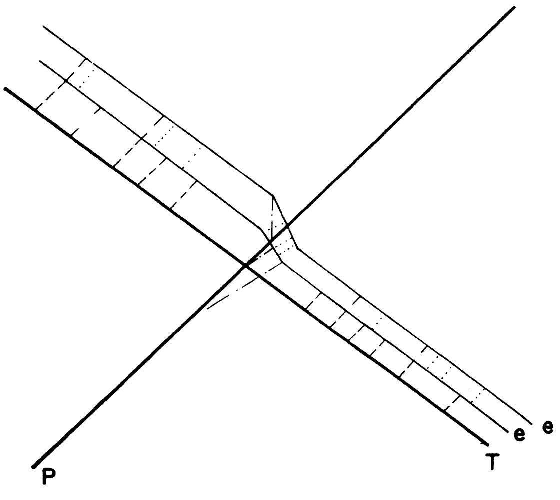

FIG. 3. Scattering event with full correlation. Scattering correlation occurs during the collision and the remaining static correlation is included in the asymptotic wave functions before and after the collision.

ous observations, two of which were discussed at the beginning of this section. Double ionization in helium,[1–8] argon, neon,[9] and molecular hydrogen[10–12] has been analyzed[8,14–17] by taking the ratio of double- to single-ionizaton cross sections. Interpretation of this ratio for protons and electrons in our formulation was reinforced by a recent measurements by Anderson and co-workers[1] at CERN using antiprotons on helium. Simultaneous capture and ionization has also been analyzed[13,14] in a similar way. Also there is evidence that this approach may give some insight into mechanisms for fragmentation of molecules[51] in collisions with charged particles at high velocities. For multiple ionization in the inner shells of atoms the well-known binomial distribution[18–21] of charge states is obtained[29] from our formulation in the limit of the independent-electron approximation. Summing over all final states but one gives[52–55] a single active electron formulation used for single-electron transitions in many-electron targets. Single and double electron capture from multielectron targets at high velocities has also been interpreted[56] in this way. Multiple ionization of outer shells[22–26] is usually due to correlation in the final state, e.g., shakeoff and Auger effects. Thus our approach gives a common formulation for various analyses previously used to interpret transitions of one or more electrons in an atomic collision. We have also introduced the concept of scattering correlation which has yet to be tested experimentally.

VI. SUMMARY

In this paper we have considered many-body correlation in atomic scattering. Here correlation, i.e., corrections to the independent-electron approximation, have been divided into two types, static correlation and scattering correlation. Static correlation is expressed in the asymptotic wave functions, while scattering correlation is expressed in the scattering operator. Scattering correlation, a new term, is pictured as correlation occurring during the scattering process itself. At high collision velocities expressions for the direct and rearrangement mechanisms have been derived from the static correlation term. In the limit of high collision velocities static correlation (or rearrangement) is dominant. At lower velocities, larger projectile charge, or weaker correlation, the uncorrelated independent-electron approximation (or direct) amplitude becomes more important. Scattering correlation is important when the collision is slow enough that electron-electron interaction contributes significantly to the collision.

ACKNOWLEDGMENTS

I gratefully acknowledge discussion with O. L. Weaver and T. M. Reeves. This work has been supported by the U.S. Department of Energy, Office of Basic Sciences, Division of Chemical Sciences.

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