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K-shell hole production by light ions in the region $\eta^{1/2} \lesssim 1$[†]

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An accurate numerical calculation of K-shell hole production by light ions is presented for nonrelativistic target atoms in the projectile energy region $\eta^{1/2}$ $(=\hbar v/Z_n e^2) \lesssim 1$. Both hydrogenic and Hartree-Fock target-atom potentials were investigated. After correcting a computer error in some of our initial results published recently, good agreement is found for proton impact absolute cross sections and for the projectile atomic-number dependence of the cross section. We also present simple analytical formulas which fairly well reproduce our results for target atomic numbers ranging from $Z_n = 13$ to 30. This will hopefully facilitate the use by experimentalists of this rather involved calculation, which includes up to 54 basis functions. An interesting result of the comparison between the calculated and experimental atomic-number dependence of the cross section is that it suggests charge-transfer contributions to K-shell hole production are at least as large as given by the Brinkman-Kramers (BK) approximation. This requires that a great deal of electron stripping occurs as the projectile tries to leave the atom with its captured electron, because experimentally measured cross sections for charge transfer are considerably less than the BK. A simple model is presented which shows that this is to be expected.

I. INTRODUCTION

K-shell hole production has been the subject of an intensive theoretical and experimental[1-9] investigation recently. The projectile atomic number Z_p dependence of the cross section has received particular attention. Only when this is understood is there any hope of calculating absolute cross sections for x-ray production. In this paper we present an accurate numerical calculation for nonrelativistic atoms in the region $\eta^{1/2}$ $(=\hbar v/Z_n e^2) \lesssim 1$.

In K-shell hole production two types of processes can occur: direct ionization (or excitation to a bound unoccupied state) and charge transfer to the projectile. Measured charge transfer cross sections[10] indicate that for protons, this process is a factor of 2 or 3 smaller than the Brinkman-Kramers[11] (BK) estimate.[12] One might conclude therefore, that for light ion projectiles charge transfer effects are only a few percent of the cross section when the target atom has nuclear charge greater than 10 or so. We shall thus be initially concerned with the direct process.

Until the work of Basbas, Brandt, and co-workers,[2] the most familiar methods of calculating K-shell ionization were the plane-wave Born approximation[13] (PWBA), the semiclassical approximation[14] (SCA), and the binary encounter approximation[15] (BEA). The references cited give good descriptions of these methods so here we limit ourselves to two observations pertinent to this work.

(i) If the deflection of the projectile from its constant-velocity straight-line path is negligible then the SCA and PWBA are formally equivalent and give the same answer as long as no further approximations are applied to either method, for example, treating different angular momentum states in the ejected electron configuration, or approximating integrals, and so on.

(ii) All these methods give cross sections proportional to Z_p^2. It follows, as the only deficiency in the PWBA (SCA) is the neglect of terms higher order in Z_p, that the BEA can only be an approximation to the PWBA. Thus to observe that the PWBA, or SCA, give different answers for absolute cross sections than the BEA and further to note that sometimes the BEA gives better agreement with experiment is to miss the real point, i.e., the inadequacy of the PWBA.

The PWBA can be incorrect for only two reasons: the wave functions describing the atom might not be sufficiently accurate, or second and higher Born terms might be important. In everything we discuss here we are assuming the projectile follows a straight-line path. There is thus no difference between the various Born terms obtained from the wave treatment and the semiclassical treatment. In fact, all our calculations are carried out in the framework of the latter, i.e., the projectile is considered only to provide a time-dependent perturbation of the atom.

By changing the atomic number Z_p of the projectile one can explore the behavior of the higher-order Born terms. It was established experimentally[2,3,5] in this manner that K-shell hole production $\sigma(Z_p)$ did not follow a Z_p^2 behavior. The ratio r_{12} defined by

$$
\begin{aligned}
r_{12} &= Z_2^2\sigma(Z_1)/Z_1^2\sigma(Z_2)\\
&= (1+Z_1\rho+\cdots)/(1+Z_2\rho+\cdots)\\
&\approx 1+(Z_1-Z_2)\rho+\cdots\,, \qquad (1)
\end{aligned}
$$

where

$$\sigma(Z_p)=\alpha Z_p^2(1+Z_p\rho+\cdots)$$

was found to be less than unity in the region $\eta^{1/2}\leqslant 0.3$ and to rise above unity at higher energies. An approximate but quantitative explanation was also presented for this.[2,3] It was suggested that two effects were involved: an increased binding because at low energies the circulating K electron sees an extra charge Z_p added to the nuclear charge Z_n; and a polarization effect in which the electron density near the projectile is increased by the projectile's attractive force. The former process decreases the cross section as compared to Born, the latter increases it. A review has been given by Brandt[16] on the success of this model.

It should be observed that this model makes no reference to the presence of the other electrons in the atom. It turns out that it is crucial in this regard for the independent particle or Hartree-Fock description of the atom to be accurate.[4] We shall certainly assume that to be the case. This brings us though to another deficiency present in the standard treatment of the PWBA, the approximations used to describe the target atom. Most authors have used the procedure described by Merzbacher and Lewis,[13] a screening model with hydrogenic wave functions. Hartree-Fock is a better description of an independent particle model atom but suffers from the problem that one would have to calculate scattering wave functions from a nonlocal potential, and then perform integrals numerically even to calculate the first Born term. Thus, the effects of antisymmetrization and screening have never been properly treated for this problem.

In summary then, the PWBA has been shown to be inaccurate both theoretically and experimentally. A semiquantitative understanding of the higher-order Born corrections has been given, but no exhaustive numerical study has ever been presented of the various approximations commonly used to calculate absolute K-shell hole production cross sections.

It is our aim in this paper to rectify this situation. We present a numerical semiclassical approach in which we solve the time-dependent perturbation of the atom by expanding the wave function in a truncated set of Hilbert functions centered about the target. We keep s, p, and d angular momentum states. We have mainly concentrated on the second Born term in our analysis, but have also studied the effect of higher-order terms for a few selected energies and atoms. Both hydrogenic and Hartree-Fock models have been studied and the results compared with each other. We have calculated both r_{12} and absolute cross sections $\sigma(K)$ for aluminum, titanium, and nickel. We have also fitted our results with simple formulas to allow interpolation to other atoms. Finally, we have reconsidered the importance of charge transfer in K-shell hole production in the light of our calculation.

Our method of calculation is described in detail in Secs. II and III. Section IV gives our results and compares them to experiment. Finally, in Sec. V we discuss the results and give our conclusions.

II. METHOD

The method of calculation, which has been briefly described previously,[1] consists of treating the fast projectile as moving with constant velocity past the atom and perturbing it with a time-dependent potential $V(\vec{r},t)$. The time-dependent Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\psi(\vec{r},t)=[H_e(\vec{r})+V(\vec{r},t)]\psi(\vec{r},t) \qquad (2)$$

is then solved by expanding $\psi(\vec{r},t)$ in a truncated space of Hilbert functions $U_j(\vec{r})$. To facilitate later manipulations we first of all diagonalize $H_e(\vec{r})$, the single-particle atomic Hamiltonian, in a basis set, producing a set of $\chi_\lambda(\vec{r})$, eigenfunctions to PH_eP with eigenvalues λ. Here P is the projection operator onto our basis. We chose the $U_j(\vec{r})$ one-electron basis functions to be orbital angular momentum eigenfunctions, and as the notation implies they are centered about the target atom. They thus contain no time dependence as a two-centered basis would have, i.e., one which includes an expansion about the projectile.

This method is not new,[17] but has fallen out of fashion for the proton-hydrogen problem, which is the paradigm of such a computational procedure. The reason for this is that in the proton-hydrogen problem charge transfer plays an important role. This has the consequence that the wave function has to describe the electron being pulled away a rather long distance by the projectile. Such an elongated distortion is rather difficult to reproduce by a one-centered expansion without using a prohibitively large number of partial waves.

The trouble with the two-centered expansion is that it involves matrix elements of the potential which cannot be evaluated explicitly but need numerical integration. Further, they depend on so many of the problem's variables; the impact parameter B, the time t, and the velocity v of the projectile, that they cannot be conveniently stored in a computer. The result is that many proton-hydrogen codes[18] take 10 to 20 min to integrate a single impact parameter on an IBM 360-65, and this is in a basis often so small that not even the first four bound states of the system are accurately reproduced and the continuum is poorly represented. Thus ionization cannot be accurately treated.

For a light ion projectile (a proton, deuteron, or α particle) charge transfer, although present, should not play a dominant role as long as the target atom has a sufficiently large atomic number Z_n. For $Z_n = 13$ charge transfer, as computed in the BK approximation, is less than 3% of the K-shell hole production for protons and deuterons and less than 25% of the cross section for K-shell hole production for α particles. For large Z_n, the percentage contribution from the BK charge transfer is reduced by approximately Z_n^3. Correspondingly, we find when computing the contribution from the Born terms past the first, that inclusion of only s and p states in our one-centered expansion is quite sufficient in the present energy range ($\eta^{1/2}$ less than 1.0). For the first Born cross sections only s, p, and d states need be included.

With a one-centered expansion, the matrix elements of the potential $V(\vec{r}, t)$[19] are

$$\langle\lambda | V(\vec{r}, t) | \lambda'\rangle = \int \chi_\lambda(\vec{r}) \left(\frac{Z_p e^2}{R} - \frac{Z_p e^2}{|\vec{R} - \vec{r}|} \right) \chi_{\lambda'}(\vec{r}) d^3 r . \quad (3)$$

Here

$$\vec{R} = (\vec{B}, vt) \equiv (\vec{B}, Z) ,$$

with $\vec{B}$ the (two-dimensional) impact parameter; Z_p is the projectile atomic number. These matrix elements are functions of the internuclear distance R only, apart from quickly calculable terms such as $Z/R, B/R$, etc. They do not depend on the projectile velocity. This still does not mean they can be stored on a computer because it turns out that to integrate out Eq. (2) takes a rather large number of points, of the order of a few hundred. Counting quickly shows that for a 54-state basis the storage space (e.g., $54 \times 54 \times 400$) is excessive. The reason for this problem is that ψ is a rapidly oscillating function of t at low velocities. This is due to the fact that it contains factors like $e^{iH_e Z/\hbar v}$, as can be read off from Eq. (1). We circumvent this difficulty by diagonalizing H_e in our basis. We can thus write the second Born approximation for our initial target function as

$$\psi_2(\vec{r}, Z) = e^{-i\mu Z/\hbar v} \chi_\mu(\vec{r}) - \frac{i}{\hbar v} \sum_\lambda \chi_\lambda(\vec{r}) e^{-i\lambda Z/\hbar v} \times \int_{-\infty}^{Z} \langle\lambda | V | \mu\rangle e^{i(\lambda-\mu)Z'/\hbar v} dZ' . \quad (4)$$

Here $\chi_\mu(\vec{r})$ is the target ground-state wave function.

The oscillatory difficulty with the integrand is now avoided by using a modified Simpson's rule. Normally a Simpson's rule replaces the integrand over three consecutive points by a parabola. Thus,

$$\int_{Z_1}^{Z_3} f(Z) dZ \approx \int_{Z_1}^{Z_3} (a + bZ + cZ^2) dZ . \quad (5)$$

The coefficients a, b, and c are found from a knowledge of $f(Z)$ at Z_1, Z_2, and Z_3. Note that it is not necessary that these points be equally spaced. Our procedure is simply to evaluate the integrals that arise from ψ_2 of Eq. (3) according to

$$\int_{Z_1}^{Z_3} e^{i\alpha Z} f(Z) dZ \approx \int_{Z_1}^{Z_3} e^{i\alpha Z} [a + bZ + cZ^2] dZ . \quad (6)$$

The convergence of the integral is now not governed by how large α is.

This simple device reduces the number of integration points needed to find ψ_2 to as little as 30, which in turn allows storage of the matrix elements. This speeds up the calculation by two orders of magnitude. As we only store the matrix elements as a function of R, the points for the Z integration depend on B and are not equally spaced. It further turns out that with light ions ($Z_p = 1$ or 2), the second Born approximation is quite accurate ($\approx 2\%$) in the energy range considered, so having worked out $\psi_2(\vec{r}, t)$ we can use it to evaluate the cross section for transition to any state σ_λ. Thus

$$\sigma_\lambda = \frac{2}{(\hbar v)^2} \int d^2 B \left| \int_{-\infty}^{+\infty} e^{i\lambda Z/\hbar v} \langle\lambda | V | \psi_2\rangle dZ \right|^2 . \quad (7)$$

Here too we can save time by using the time reversal properties[20] of the matrix elements to write the limits on Z as zero and infinity, and those of Z' in the definition of ψ_2 as zero and infinity for some terms and Z and infinity for others. The latter integrals are of the form

$$\int_0^\infty e^{i\alpha_1 Z} f(Z) dZ \int_Z^\infty e^{i\alpha_2 Z'} g(Z') dZ' .$$

In performing the Z integration, one has the problem of choosing the oscillatory phase to be used. We employed $(\alpha_1 + \alpha_2)$ on the grounds that if α_2 were large an integration by parts would give a Z-dependent oscillation of this form. It apparently works very well. No increase in the number of points to work out the second Born terms was needed over working out the first Born terms.

The important feature of our basis is that it is bounded at infinity. It leads to a discrete set of orthogonal and normalizable eigenfunctions. Thus we now have to answer the question of how from σ_λ, Eq. (7), are we to calculate K-shell hole production. There are several ways of calculating ionization to a continuum set of eigenstates of the true Hamiltonian H_e once one has the transition probabilities to the discrete pseudostates of PH_eP. One method is to write down the wave function ψ at $t=\infty$ and project onto the true states of H_e. This method has been used for example by Wilets and Gallaher.[18] It has some difficulty because the eigenstates of H_e are not eigenstates of PH_eP and the projected amplitude varies with time even as $t\rightarrow\infty$. Another approach is to recognize that one can construct the true continuum eigenstate from the pseudostates. This method has been used for example by Bassichis, Reading, and Scheerbaum.[21]

In fact, we used neither method here. Our method is to make sure that our occupied pseudostates are very accurate reproductions of the true occupied states. As all the occupied states are bound this is of course possible to achieve. We are thus guaranteed that our unoccupied pseudostates, which by construction are orthogonal to the occupied pseudostates, are very accurately orthogonal to the true occupied eigenstates. They therefore are already in the subspace of the continuum states and no further projection is needed such as with Wilets and Gallaher. To calculate a cross section for K-shell hole production then, we sum σ_λ over the unoccupied states. Naturally this includes all contributions from excitations to unoccupied bound states and is not the pure ionization cross section. It is in fact the cross section we need, i.e., that for K-shell hole production. It is interesting to calculate the pure ionization cross section. This may be done by choosing a basis which has sufficient accuracy to reproduce enough of the lowest bound states so that the cross section from higher excited bound states is very small and its contribution, though included in the "ionization" calculation, is negligible. We give an example of this in the next section when we check our pseudostate calculation for pure ionization against the Born calculation in the hydrogenic model.

Lastly it should be noted that with this method the Hartree-Fock Hamiltonian, once it is diagonalized, is just as easy to treat as the hydrogenic case. Thus it is straightforward for us to calculate K-shell hole production keeping the important effects of screening and antisymmetrization present in the Hartree-Fock potential. The calculations with the Hartree-Fock potential do however have the disadvantage that they must be repeated for each target atom, whereas the hydrogenic potential cross sections can be scaled from one target atom to another.

III. BASIS

We diagonalize the atomic Hamiltonian H_e on a finite Hilbert basis. The representation which we used was one invented by one of us (J. F. R.); it was first used by Zimmerman in his thesis[20] on proton-hydrogen scattering. A typical set of radial parts of the basis function $u_j^l(r)$ is given by

$$U_j(\vec{r}) \equiv u_j^l(r) Y_{lm}(\theta,\phi),$$
$$u_j^l(r) = r^l \exp[-c_l r/a_n(1-\epsilon e^{i\phi_j})], \quad |\epsilon|<1, \tag{8}$$

where ϕ_j are points chosen at equally spaced intervals from 0 to 2π and l is the angular momentum. Also, a_n is the Bohr radius for a hydrogenic atom of nuclear charge Z_n. That such a representation is complete can be seen from the following argument. From a knowledge of the generating function[22] for the Laguerre polynomials we know that

$$e^{-\rho/(1-s)} = \sum_{n=0}^{\infty} e^{-\rho}(s^n/n!)L_n(\rho). \tag{9}$$

Replacing s by $\epsilon e^{i\phi}$ it can be seen that

$$\frac{n!}{2\pi\epsilon^n}\int_0^{2\pi} e^{-in\phi}\exp[-\rho/(1-s)]\,d\phi = L_n(\rho)e^{-\rho}. \tag{10}$$

If we replace the integral in ϕ by a sum of terms taken around the unit circle we see that it is possible to represent any $L_n(\rho)$. As the $L_n(\rho)$ are complete, so therefore is our basis. This method incidently can be used for many orthogonal polynomials, it also gives an integral representation for them.

We diagonalized two Hamiltonians in our basis, a hydrogenic one

$$H_e = -(\hbar^2/2m_e)\nabla_r^2 - Z_n e^2/r, \tag{11}$$

and a Hartree-Fock one,

$$H_e = -(\hbar^2/2m_e)\nabla_r^2 + V_{\mathrm{HF}}. \tag{12}$$

The Hartree-Fock potential was taken from the compilation of Clementi and Roetti.[23] The Hartree-Fock H_e includes the effect of screening, which means the "natural" radius of a $4s$ state of nickel for example is a factor of 20 times larger than the K-shell radius. To include this in our basis when the Hartree-Fock H_e was used we deformed the "circle" in the complex ξ plane, $\xi = 1-\epsilon e^{i\phi}$, into an ellipse, which was fixed by requiring that $\xi=1$ be a focus, that $\xi = 1-\epsilon$ be its nearest extremity, and that the furthest extremity of ξ be set at approximately the radius of the highest-lying bound orbital. We tried various values of ϵ ranging from 0.5 to 0.95 to establish covergence. We also included the hydrogenic orbital which most accurately represented the $1s$ or $2p$ state as a special basis function. Our basis sets were chosen to always give very accurate representations of the lower Hartree-Fock bound states.

Having diagonalized our basis, it was possible to see that various states, such as the point at $\xi = 1 - \epsilon$, had the effect of producing a χ_λ with very high λ, which contributed little to the end result. It was found that this point could be left out with no loss of accuracy. This also points out the danger of false convergence criteria. A blind increase in the number of states used, though producing a stable answer, may not have coverged if the increase in the number of basis states only serves to add eigenfunctions of high λ.

The computationally attractive feature of this basis is that the matrix elements, which rapidly increase in complexity as n increases in an expansion in $L_n(\rho)$, are the same in form for different ξ. The advantage in program speed and simplicity cannot be overemphasized.

In our calculations we felt that the important feature of the basis and potential was that the lower bound states and the continuum be well represented. As will be seen in Table I, the higher bound states have very small first Born matrix elements. Hence the open-shell Hartree-Fock potential, as defined by Clementi and Roetti, was not used. Instead the high open shells were treated as closed, but with the proper occupation number. This results in great simplification in the Hartree-Fock Hamiltonian diagonalizations, and had very little effect on the filled orbital eigenvalues or wave functions.

An important test of the completeness of our basis sets, in particular their ability to represent the continuum, was comparison with exact numerical calculations of the K-shell ionization cross section, with a hydrogenic target potential.[13] Some representative results are given in Table I. The continuum is well represented in the first basis set. The comparison with the exact ionization calculation requires that the contribution from the bound states be subtracted. This bound-state contribution was estimated from the results of the second basis, which gives a very good representation of the first seven hydrogenic bound states. Summing over the $2s$ through $8s$ states gives a cross section for $1s \rightarrow$ bound-state transitions of 4674 b. Subtracting this from the total K-shell excitation cross section of 6575 b from the first basis gives an ionization cross section of 1901 b. The exact result is 1852 b. The small discrepancy could well be due to additional contribution from higher bound states that needs to be subtracted out. The K-shell hole production cross section from the first basis is given by subtracting the $2s$ and $3s$ contributions from the total, as the $2s$ and $3s$ orbitals are filled in aluminum. The result, 2275 b, is a factor of 2 smaller than the corresponding result for a Hartree-Fock target potential. That the hydrogenic potential gave K-shell hole production cross sections approximately a factor of two smaller than the Hartree-Fock potential was a general feature present in all our calculations. It is caused by the fact that the ionization energy is too large in the hydrogenic model. There is no way as in Merzbacher and Lewis[13] to alter the Hamiltonian with a *constant* screening

TABLE I. Aluminum first born calculation for K-shell hole production $E = 1.0$ MeV/amu. S final states. Hydrogenic target atom.

Pseudostate quantum number	Basis which gives even distribution of eigenvalues[a] λ/Z_n^2 (Hartrees)[c]	σ_λ(b)	Basis which gives good bound-state representations[b] λ/Z_n^2 (Hartrees)[c]	σ_λ(b)
$2s$	0.3750	3569	0.3750	3565
$3s$	0.4457	732	0.4444	632
$4s$	0.4933	727	0.4688	225
$5s$	0.5751	770	0.4800	106
$6s$	0.7157	519	0.4861	59
$7s$	0.9632	233	0.4898	39
$8s$	1.4367	25	0.4932	48
$9s$	2.4931	1	0.4988	91

[a] The first eigenstate $1s$ is omitted, as are the last two. This basis has 11 elements and parameters $\epsilon = 0.5$ and $c_0 = 1.0$.

[b] The first and last six states are omitted. This basis has $\epsilon = 0.75$ and $c_0 = 0.25$.

[c] With our choice of the $1s$ state as the zero of energy the ionization threshold starts at $\lambda/Z_n^2 = 0.5$.

potential, or what is equivalent to the introduction of the parameter θ. An addition of a constant (independent of r) potential has no effect on our calculation. The correct way to proceed is to add a screening potential that does depend on r as we have done in the Hartree-Fock model. Nevertheless, there is a great deal to be learned from the hydrogenic calculation. It certainly allows checks of the code as we have described. It also served to show that r_{12} as a function of velocity v was to a large extent independent of whether the Hartree-Fock Hamiltonian for a particular atom or a scaled hydrogenic model was used. This indicated that r_{12} for the Hartree-Fock model might scale as a function of η, a point in fact verified as we shall show.

Note also that in the first basis the last few states have very small σ_λ, indicating a sufficiently high energy range for our eigenvalues and their ability to represent the emitted electron states important at this energy.

IV. RESULTS

We applied the methods described above to two problems. Firstly we calculated the absolute cross section including the second Born term for proton-impact K-shell hole production. As has been noted by others,[17] to be consistent in keeping all terms thru Z_p^3 in the Born series, we included the second Born amplitude only in its interference term with the first Born amplitude

$$\sigma = |f_{B1}|^2 + f_{B1} f_{B2}^* + f_{B1}^* f_{B2} . \tag{13}$$

Here f_{B1} and f_{B2} are the first and second Born amplitudes, respectively. The basis used had up to 54 states, with as many as 10 s states, 20 p states, and 24 d states. Reasonable convergence could be obtained with as few as 6 s states, 12 p states, and 15 d states. We found the inclusion of d states essential for accuracy in the first Born cross section, but they had negligible effect on the second Born amplitude. In the d state part of the second Born amplitude only s-d and p-d virtual transitions involving d states were included; the d-d transitions were left out as being probably of less importance. A more thorough study of the role of d states in the second Born, to include the d-d interaction at higher energies, is in progress.

We also wrote an independent code using a time development U-matrix approach to give us the effect of higher Born terms. This method consisted of writing the U matrix which connects the wave function at time t_1 to that at time t_3 as

$$\underline{\psi}_I(t_3) = \underline{U}(t_3, t_1)\underline{\psi}_I(t_1)$$

$$\approx \exp\left(-\frac{i}{\hbar v}\int_{t_1}^{t_3} \underline{V}_I(t')dt'\right)\underline{\psi}_I(t_1) .$$

Here $\underline{\psi}_I$ is the vector representing the wave function in the interaction picture, and $\underline{V}_I$ is the matrix of the interaction potential. We now solve the scattering problem, which is to calculate $\underline{\psi}_I$ at $t = \infty$ by taking products of the matrices.[24] A U-matrix method was first used in Zimmerman's thesis[20] for the proton-hydrogen problem and has also been applied by Shakeshaft.[25] In our application it was at least an order of magnitude slower than the second Born calculation.

The U-matrix code also served as a test of the code which explicitly calculated the first and second Born interference term. This code was further tested by repeating the calculations of Kingston *et al.* and Holt and Moiseiwitsch[17] of $1s$-$2s$ and $1s$-$2p$ excitation of atomic hydrogen by proton impact, on a basis including only the $1s, 2s, 2p_0, 2p_\pm$ hydrogen atom target states. Complete agreement was obtained.

Some typical results are shown in Tables II–IV for aluminum, titanium, and nickel. Firstly we list σ_M from a calculation performed in the fashion

TABLE II. Absolute cross sections for Al (in units of 10^{-20} cm^2).

E(MeV/amu)	σ_M	σ_{HF}	$\sigma_{\mathrm{HF}}^{(2)}$	THIM[a]	% diff. from $\sigma_{\mathrm{HF}}^{(2)}$
0.75	1.260	1.019	0.878	0.829 ± 12%	−6%
1.0	1.704	1.406	1.298	1.34	+3
1.25	2.052	1.726	1.643	1.72	+5
1.5	2.320	1.979	1.912	1.96	+2
2.0	2.669	2.326	2.317	2.27	−2
2.5	2.856	2.518	2.518	2.65	+5
3.0	2.940	2.608	2.608	2.78	+7
3.5	2.962	2.635	2.635	2.80	+6
4.0	2.942	2.623	2.623	2.73	+4

[a] Reference 8. A fluorescence yield of 0.0357 from Ref. 26 was used. The quoted uncertainty in the experimental values is that present in the x-ray production cross sections.

TABLE III. Absolute cross sections for Ti (in b).

E(MeV/amu)	σ_M	$\sigma_{\rm HF}$	$\sigma_{\rm HF}^{(2)}$	AJ [a,c]	% diff. from $\sigma_{\rm HF}^{(2)}$	BG [b,c]	% diff. from $\sigma_{\rm HF}^{(2)}$
0.5	59	55	30				
1.0	308	276	206				
1.5	653	580	495	597 ± 11%	+21%		
2.0	1011	893	806	983	+22	1010 ± 13%	+25%
2.5	1344	1186	1111				
3.0	1639	1451	1389	1750	+26	1680	+21
4.0	2111	1891	1842				
5.0	2445	2209	2194	2660	+21	2520	+15
6.0	2675	2426	2426	2970	+22	2860	+18
7.0	2827	2570	2580	3230	+25	3030	+17
9.0	2975	2710	2729	3410	+25	3180	+17
11.0	3007	2729	2751	3480	+27	3160	+15

[a] Reference 9.

[b] Reference 6.

[c] A fluorescence yield of 0.219 was used (Ref. 26)

of Merzbacher and Lewis. Secondly, the Hartree-Fock first Born $\sigma_{\rm HF}$, and thirdly, a full calculation with the Hartree-Fock potential and including second Born effects, $\sigma_{\rm HF}^{(2)}$ was performed. In the last columns $\sigma_{\rm HF}^{(2)}$ is compared to some recent measurements of absolute cross sections. Table V shows the separate contributions to $\sigma_{\rm HF}$ for aluminum from s, p, and d final states.

An unscreened hydrogenic calculation gives a cross section roughly a factor of 2 smaller than σ_M or $\sigma_{\rm HF}$. This indicates the importance of including the correct binding in the calculation. We further note significant deviations between σ_M and $\sigma_{\rm HF}$. In this work we are trying to calculate ionization to within 2%. We believe we have achieved this within the context of the independent particle model. We therefore conclude from σ_M and $\sigma_{\rm HF}$ that the extra trouble of doing a Hartree-Fock calculation is well worth it. The effect of screening and exchange on the outgoing continuum states as represented by the Hartree-Fock model is clearly important.

The experimental data σ_E shows good agreement with $\sigma_{\rm HF}^{(2)}$. It is interesting that $\sigma_E/\sigma_{\rm HF}^{(2)}$ for the Akselsson and Johansson data on nickel and titanium is fairly constant over a wide range of energies. This suggests possible errors in the experimental normalization or inaccurate fluorescent yields. It is possible that the situation could be improved on the theoretical side by using configuration interaction wave functions. We are investigating this.

The U-matrix code was fairly expensive to run and we only did a few spot checks to ensure that

TABLE IV. Absolute cross sections for Ni (in b).

E(MeV/amu)	σ_M	$\sigma_{\rm HF}$	$\sigma_{\rm HF}^{(2)}$	AJ [a,c]	% diff. from $\sigma_{\rm HF}^{(2)}$	BG [b,c]	% diff. from $\sigma_{\rm HF}^{(2)}$
1.0	36.7	30.7	21.0				
1.5	94.7	80.3	62.3	67 ± 11%	+8%		
2.0	169	144	121	134	+10		
2.5	250	214					
3.0	331	284	258	284	+10		
4.0	486	412					
5.0	621	531	513	562	+9	710 ± 12%	+38%
6.0	733	631					
7.0	826	716	707	778	+10		
9.0	958	839	836	926	+10		
11.0	1041	914	916	1010	+10	1150	+26%
14.0	1103	980	981			1270	+29%
17.0	1120	993	993			1250	+26%

[a] Reference 9.

[b] Reference 6.

[c] A fluorescence yield of 0.414 was used (Ref. 26).

TABLE V. Separate contributions to σ_{HF} from s, p, and d states for Al.

E(MeV/amu)	σ_{HF} (in units of 10^{-20} cm^2) s	 p	 d	 Total
0.25	0.090	0.059	0.007	0.157
0.50	0.264	0.290	0.023	0.577
1.0	0.468	0.851	0.087	1.406
1.5	0.533	1.279	0.167	1.979
2.5	0.517	1.700	0.301	2.518
4.0	0.428	1.818	0.377	2.623
6.0	0.332	1.708	0.367	2.407
8.0	0.267	1.556	0.328	2.151

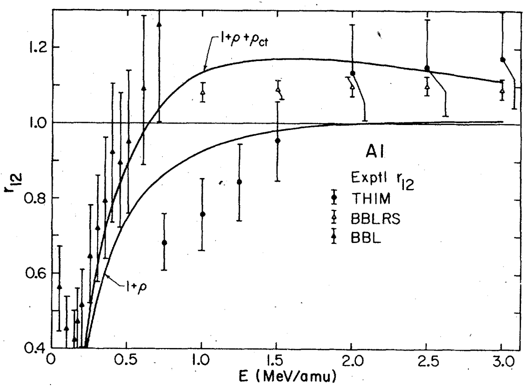

FIG. 1. Projectile atomic number dependence of r_{12} for Al. Experimental points are taken from Ref. 8 (THIM), Ref. 2 (BBLRS), Ref. 3 (BBL). Solid curves are the present theoretical calculations with, $1+\rho+\rho_{CT}$, and without, $1+\rho$, the approximate charge transfer contribution described in the text.

the accuracy of our calculations was not being drastically affected by higher order Born terms. For 0.5-MeV/amu deuterons on aluminum (a low energy, with the largest Z_p/Z_n ratio), we found an absolute cross section of 0.428×10^4 b from the second Born code and 0.420×10^4 b from the U-matrix code. Similar results were obtained at higher energies and with different atoms.

The deuteron projectile calculations give us values for the parameter ρ, defined by

$$\sigma_{\mathrm{HF}}^{(2)}=Z_p^2\alpha(1+Z_p\rho). \tag{14}$$

The parameter α is the first Born cross section divided by Z_p^2. We found that our result for $1+\rho$ could be fit to approximately 2% accuracy, over the range of Z_n and energies $(0.05\leq\eta^{1/2}\leq1)$ we considered, by the simple formula

$$\rho=-(Z_p/Z_I)c/[1+(\eta_I/\eta_0)^2],\qquad c=11.54,\quad \eta_0=0.093. \tag{15}$$

Here $\eta_I^{1/2}=\hbar v/Z_Ie^2$ and the parameter Z_I is found by requiring $Z_I{}^2e^2/2a_0=I_K$, where I_K is the K-shell ionization energy. Also, our first Born absolute cross sections for proton impact were found to accurately scale (2%) between Ti and Ni and between Ti and Al in the energy range given by $0.08\leq\eta^{1/2}\leq1$ according to the simple formula

$$\sigma_{\mathrm{HF}}(\eta_I)=(Z_I'/Z_I)^{3.92}\sigma_{\mathrm{HF}}(\eta_I'). \tag{16}$$

Equations (15) and (16), combined with our tabulated σ_{HF} values for Al, Ti, and Ni, should allow accurate calculation of $\sigma_{\mathrm{HF}}^{(2)}$ for any atomic target with Z_n between 13 and 30.

The values of ρ may also be used to predict the projectile atomic number dependence ratios, as defined in Eq. (1). In that equation, $\sigma(Z_p)$ is the K-shell ionization (and excitation to unoccupied states) cross section for a projectile of atomic number Z_p. Taking the ratio for deuterons (or protons) and alpha particles r_{12} to be given by

$$r_{12}=\sigma(2)/4\sigma(1)=(1+2\rho)/(1+\rho)\approx1+\rho, \tag{17}$$

i.e., neglecting all terms involving Z_p^4 in the ratio, gives the results shown in Figs. 1–3. The most important thing to note is ρ is negative at our lower energies and goes only slightly above one as $\eta^{1/2}$ is increased. This implies almost exact cancellation of the "binding" and "polarization" terms. This differs from the results presented in Ref. 1, which were in error.

An interesting feature of the calculation of r_{12} is that it is practically independent of whether we use a hydrogenic (not shown) or Hartree-Fock model. This is in spite of the fact that the first Born cross section can differ by a factor of 2 in the two cases.

We once again checked with our U-matrix code to see if the effect of higher Born terms invalidates our results for r_{12}. The U-matrix code cal-

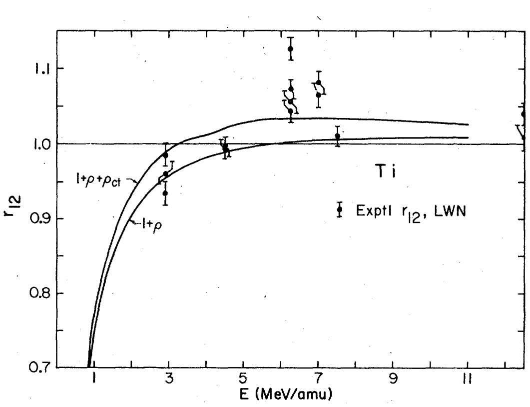

FIG. 2. Projectile atomic number dependence of r_{12} for Ti. Experimental points are taken from Ref. 5 (LWN). Solid curves are as in Fig. 1.

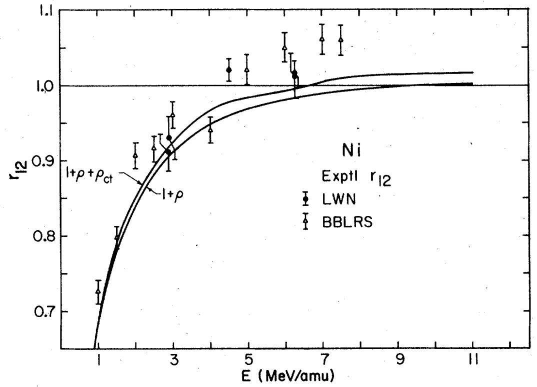

FIG. 3. Projectile atomic number dependence of r_{12} for Ni. Experimental points are taken from Ref. 5 (LWN), Ref. 2 (BBLRS). Solid curves are as in Fig. 1.

culates all terms in Z_p both for the α particle and deuteron. Thus we could not expand r_{12} in ρ but instead calculated r_{12} directly as $\sigma(2)/4\sigma(1)$. For aluminum at 0.5 MeV/amu we obtained 0.741 for $(1+\rho)$ from the second Born code and 0.730 for r_{12} from the U-matrix code. Similar results were obtained for nickel at higher energies. The differences were never more than 2%.

To explain why the experimental r_{12} rises appreciably above unity we have to now include charge transfer in our calculation. In the Brinkman-Kramers (BK) approximation the charge transfer is less than 3% of the total cross section for proton impact, but as it increases as Z_p^5, it is not negligible for alpha particles. We have calculated $\sigma_{\rm BK}$, the Brinkman-Kramers cross section for charge transfer into the projectile ground state, using the Hartree-Fock representation of the target atom. For protons on argon we get a $\sigma_{\rm BK}$ identical to that of Halpern and Law.[12]

We are at present calculating the second Born term for charge transfer, but for now we may only estimate the effect this might have.

We are at rather low energies here. The idea of Basbas[2,3] *et al.* is that there are two effects competing here of "increased binding" and "polarization." These effects should play roughly the same role for charge transfer as they do in ionization. Thus one might estimate the cross section for this process as

$$\sigma_{cT}=\sigma_{\rm BK}(1+\rho)\,. \tag{18}$$

As ρ differs appreciably from zero only at the lower values of $\eta^{1/2}$ considered here, not much difference is observed in using $\sigma_{\rm BK}$ or σ_{cT} in the total cross section for α-particle charge transfer. We assume that charge transfer into excited projectile states follows an n^{-3} law and that this effect in r_{12} for α particles approximately cancels that given from $\sigma_{\rm BK}$ for deuterons or protons. We thus get an additional contribution to r_{12} of $\rho_{\rm BK}$, i.e., $r_{12}=1+\rho+\rho_{\rm BK}$, where $\rho_{\rm BK}=\sigma_{\rm BK}/4\alpha$. We found that $1+\rho_{\rm BK}$ was fitted to 2% accuracy by the simple formula

$$\rho_{\rm BK}=\frac{c\eta_I/Z_I^3\eta_0}{1+(\eta_I/\eta_0)^2}\,, \tag{19}$$

with $c=572$, $\eta_0=0.022$. If σ_{cT} from Eq. (18) is used instead of $\sigma_{\rm BK}$, the contribution to r_{12} is changed to $\rho_{cT}=(1+\rho)\rho_{\rm BK}$. The effect of including ρ_{CT} is shown in Figs. 1–3. It is seen that it certainly gives an effect of the right order of magnitude and sign to remove the remaining small discrepancy between theory and experiment. Only a detailed calculation of the second Born term though can definitely settle the matter. However, the point is that certainly an effect of this order of magnitude is needed. This is in complete disagreement with *experimental measurements* of charge transfer, which typically give charge transfer cross sections as two or three times smaller than obtained with $\sigma_{\rm BK}$.[10] If one uses these experimental values to estimate the effect on r_{12}, as we did before starting this calculation, they show charge transfer has little or no effect on r_{12}.

V. CONCLUSION AND DISCUSSION

In the preceding paragraphs we have presented the results of an attempt at a realistic calculation of absolute K-shell ionization and excitation by light ions, in an independent particle model. We have established that such a calculation can be done and convergence achieved without the large amounts of computing time associated with the similar, if more difficult, proton-hydrogen problem. We have shown by comparison with the Born-hydrogenic calculation that it is possible to treat ionization (and excitation) accurately in a truncated Hilbert space expansion. This has allowed us to solve the Hartree-Fock model too without the difficulties normally associated with the integro-differential equations of nonlocal potentials. We have established significant differences between the results of this model and the commonly parametrized hydrogenic calculation. Comparison with experiment gives good agreement with the absolute cross sections, but the theory is typically low (approximately 10–20%) when compared to experiments. The explanation for this is not clear to us. The uncertainties in the fluorescence yield certainly contribute to the situation. From the theoretical standpoint the most serious approximation we have made is in neglecting configuration interaction. However, it would be somewhat surprising if this gave 10% effects on K-shell ionization.

The projectile atomic number dependence of the cross section provides a very severe test of any theory as, at least in the Born term, the form factor containing the assumptions about the initial- and final-state wave functions divides out, and one has the advantage, both theoretically and experimentally, of a relative result. Our agreement with the experiments at low values of η is very good, but our results unequivocally do not rise appreciably above unity as we increase the energy. We have checked every possibility of error on our part in this regard and have found none. In calculations to be submitted for publication we have shown that including d states, projectile deflection, higher-order terms in Z_p and so on, does not change this result. We have even calculated proton-hydrogen ionization, compared to experiment and found good agreement at high energies. The significance of this is that as Z_n is lowered, the effective coupling constant is increased. If the calculation presented here for ionization had agreed with the measured values of r_{12}, it would through the magnification of the coupling constant have been in complete disagreement with the proton-hydrogen result. It was not.

The one effect we had in mind to neglect when we started this calculation was charge transfer, and this step has been amply justified by the speed of our calculation and its convergence with angular momentum. The d states are not at all important in this problem. They are incidently in the proton-hydrogen problem as is only to be expected. But the failure of our calculations to reproduce a series of experiments on different atoms by different experimental groups that establish that r_{12} does go above unity forces us to reexamine the role charge transfer might play. The direct measurements of charge transfer on protons indicate that BK overestimates the charge-transfer cross section by a factor 2 or 3. If this is the case for α particles, then we have overestimated the effect of ρ_{cT} in the figures shown, and charge transfer cannot be invoked to give us $r_{12} > 1$. Once again there seems to be some confirmation of this from the well studied proton-hydrogen problem. Charge transfer there also appears to be less than BK. Thus, this calculation appears to point to a difficulty in understanding the atomic number dependence of K-shell hole production. Of course this is not unwelcome. A discrepancy when resolved can lead to greater understanding.

We present a tentative explanation. We observe that the direct measurement of charge transfer in fact only measures protons which escape from the atom with an electron still attached to them. The contribution to K-shell hole production on the other hand arises from electrons which are picked up out of the K shell. If the electron were stripped off the projectile on the way out through the L and M shells then we would have an explanation of the phenomena, i.e., for K-shell hole production we count the full charge-transfer cross section, but we can explain why the measured charge-transfer cross section is less than this.

It should be noted that in invoking this process we are going beyond the independent particle model. The effect of the other electrons in a passive role is already included in the Hartree-Fock calculation. What is not included is inelastic electron-electron collisions in which the target electron is excited. Effects in which the target electron remains in its ground state while removing the electron from the projectile are already being taken into account.

A crude estimate of the importance of this stripping can be obtained by asking the question of what is the probability of an electron moving with the speed of the projectile inelastically interacting with the L and M shells. Geometrically this factor can be estimated as $\sigma/4\pi R^2$, where σ is the $L(M)$ shell ionization cross section for an electron and R is the radius of the $L(M)$ shell.

An experimental measurement[27] of L-shell ionization of argon by a 0.6-MeV proton gives a cross section of 3.8×10^{-18} cm^2. This is a little low in energy for our purposes but as the energy dependence is quite slow in this region, it is also a reasonable estimate for the cross section at say, 3 MeV. An electron moving with the speed of the proton has an energy of 1.5 keV approximately, and the cross section for high-energy electron impact ionization will roughly be of the same order of magnitude as for the proton. Assuming R is given by $2a_0/Z_n$ where a_0 is the Bohr radius gives

$$\sigma_L/4\pi R^2 \sim 1.$$

Such a crude estimate cannot be taken too seriously but it certainly indicates that multielectron effects cannot be completely ignored if one wants to attempt a serious calculation of the experimentally measured charge transfer. A similar result can be obtained for the M shell.

After submission of this manuscript for publication, a Letter by Band appeared with a similar suggestion.[28] However, his estimate of the charge-transfer cross section is an order of magnitude greater than σ_{BK}, and this would appear to be much too large for the r_{12} data.

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