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Shell corrections to electronic stopping powers from orbital mean excitation energies

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Shell corrections to electronic stopping powers are obtained from a theory which utilizes different orbital mean excitation energies for the atomic shell and which determines the shell correction from knowledge of the atomic velocity distribution. When summed over all shells, and taking into account the "effective" occupation of the shells, the orbital mean excitation energies yield the total mean excitation energy I. Orbital mean excitation energies are much larger than I for K shells and less than I for the outer valence shells. The atomic velocity distributions are obtained from numerical Hartree-Fock calculations. We report shell corrections for Al and Ar. We find very good agreement between other theoretical and experimental shell corrections and the present calculation. We find that the L shell gives the dominant contribution to the shell corrections for low projectile velocities while K-shell corrections dominate for larger velocities. The total shell correction is relatively insensitive to the choice of the total mean excitation energy. We have also investigated the validity of expanding the shell correction in powers of v^{-2}.

I. INTRODUCTION

The stopping of swift charged particles impinging on foils is, for large projectile velocities, determined by the mean excitation energy I of the target material through the logarithmic term in the Bethe formula.[1] However, for the lower velocities at which most experiments are carried out, the electronic shell corrections to the Bethe formula are important. Thus, in order to obtain the quantity of main interest in stopping theory, the mean excitation energy from measurements, one must have estimates of the velocity-dependent electronic shell corrections which are generally taken either from Walske[2,3] or from Bonderup.[4] Walske applies the formalism of Livingston and Bethe[5] to the calculation of shell corrections from K and L electrons, using screened hydrogenic orbitals.

Bonderup's[4] calculation is based on the statistical treatment of the target electrons proposed by Lindhard and Scharff[6] and by Lindhard and Winther.[7] The logarithm of the total mean excitation energy is, in this formalism, determined by

$$\int \ln[\rho(r)^{1/2}]\rho(r)r^2dr ,$$

where $\rho(r)$ is the total electronic density obtained from Thomas-Fermi-like models. The expressions given by Bonderup[4] were also used by Rousseau *et al.*[8] to compute shell corrections for a beam of 0.8- and 2.0-MeV α particles using Herman-Skillman[9] atomic electronic densities for the target material. Rather similar results are obtained in the three calculations by Walske,[2,3] Bonderup,[5] and Rousseau *et al.*[8] However, the Walske shell corrections are smaller than those obtained with the Bonderup formalism. This difference is mainly due to the fact that the screened hydrogenic approximation only works well for K shells, thus yielding L-shell[3] and M-shell[10] corrections that are too small.

Sigmund[11] has recently derived a relation between the stopping cross section $S(v)$ of moving target electrons and the cross section $S^{(0)}(v)$ for scatterers at rest. If the simple Bethe formula is inserted for $S^{(0)}(v)$, it means that one common mean excitation energy is used for all shells. We will show that this proposal leads to unrealistically large shell corrections for small projectile velocities, much larger than those obtained both by Walske[2,3] and by Bonderup.[5] However, we will also demonstrate that we obtain shell corrections which agree in magnitudes with both earlier theoretical and experimental shell corrections using the Sigmund formalism provided we apply this formalism *shellwise* and use different *orbital* mean excitation energies in $S^{(0)}(v)$ for the various shells. The orbital mean excitation energies are obtained from the Hartree-Slater calculations of oscillator strength moments by Dehmer *et al.*[12] and by Inokuti *et al.*[13,14]

In the next section we discuss how to obtain orbital mean excitation energies from the oscillator strength moments. We also briefly review the formalism which we use to calculate the electronic shell corrections. Calculations are performed for Al and Ar and these calculations are discussed in Sec. III, while Sec. IV contains some concluding remarks.

II. THEORY

The electronic[15] stopping of swift charged particles traversing matter is proportional to the stopping cross section $S(v)$. Disregarding relativistic and "density-effect" corrections, $S(v)$ can be obtained from the approximate[1,16,17] expression for velocities $\gtrsim 2v_0$, v_0 being the atomic unit of velocity $e^2/\hbar$. One generally expresses $S(v)$ as

$$S(v)=\frac{4\pi e^4 Z_1^2 Z_2}{mv^2}L(v)\ , \tag{1}$$

where v is the beam velocity and $L(v)$ is the stopping number defined as

$$L(v)=\ln(2mv^2/I)\theta(2mv^2-I)-\frac{C(v)}{Z_2} +Z_1L_1(v)+Z_1^2L_2(v)\ , \tag{2}$$

where $\theta(t)$ is the Heaviside step function.

The constants have their usual meaning and Z_1 and Z_2 refer to the beam and target particles, respectively. L_1 and L_2 are correction terms often called the Barkas and the Bloch corrections, respectively. I is the mean excitation energy which quantum mechanically is given as

$$\ln I=\frac{\displaystyle \sum\!\!\!\!\!\!\int \frac{df}{dE}\ln E\,dE}{\displaystyle \sum\!\!\!\!\!\!\int \frac{df}{dE}dE}\ , \tag{3}$$

where df/dE is the density of optical dipole oscillator strength per unit energy of excitation E above the ground state. $\sum\!\!\!\!\!\!\int$ indicates summation over discrete and integration over continuum states. The quantity $C(v)/Z_2$ in Eq. (2) is the so-called shell correction, the calculation of which we shall be concerned with in the present work. In an independent-particle picture, for instance Hartree-Fock, we may split $\ln I$ up into shellwise contributions. We will define the orbital mean excitation energies through the relation

$$\ln I=\frac{1}{Z_2}\sum_k w_k \ln I_k\ , \tag{4}$$

where the summation extends over all occupied shells and where w_k are "orbital weight factors" which for a neutral atom fulfill

$$\sum_k w_k=Z_2\ . \tag{5}$$

We will utilize two different expressions for w_k which both fulfill Eq. (5) and we thus obtain from Eq. (4) two sets of I_k's for a given value of I. The actual definition of w_k is given in Eqs. (27)–(34). Introducing Eq. (4) in Eq. (2) and disregarding the Barkas and the Bloch corrections, we have the following relation between the orbital shell correction $C_k(v)$ and the orbital stopping numbers $L_k(v)$:

$$C_k(v)=w_k\ln(2mv^2/I_k)\theta(2mv^2-I_k) -Z_2L_k(v)\ , \tag{6}$$

provided we define $L_k(v)$ such that

$$L(v)=\sum_k L_k(v)\ . \tag{7}$$

If one combines Eqs. (2) and (4)–(7), it becomes apparent that the decomposition of the shell correction into orbital components must be

$$C(v)=\sum_k\left[C_k(v)+\frac{w_k}{Z_2}\ln\left[\frac{2mv^2}{I_k}\right]\times(\theta(2mv^2-I)-\theta(2mv^2-I_k))\right]\ . \tag{8}$$

All calculations reported here are performed at velocities for which $2mv^2>I_k$ for all k. Thus, the second term in Eq. (8) vanishes and is omitted in the following discussion.

Sigmund[11] has recently derived an expression which determines $S(v)$ in terms of the electronic velocity distribution $\rho(v_2)$ of the scatterer (i.e., the target electrons) and the stopping cross section $S^{(0)}(v)$ for a system in which the scatterers are at rest. The main assumptions which enter into Sigmund's derivation are the following: (1) that the stopping of the charged particles is accomplished by collisions with nonrelativistic electrons, (2) that these collisions are elastic and single binary collisions, and (3) that the collision cross section has azimuthal symmetry. By simply using energy and momentum conservation in various coordinate systems, it is thus possible to relate $S(v)$ to $S^{(0)}(v)$. Expressed in terms of $L(v)$ and $L^{(0)}(v)$ Sigmund[11] finds

$$L(v)=\pi\int_0^\infty f(v_2)v_2dv_2 \times\int_{|v-v_2|}^{v+v_2}L^{(0)}(v_1)\left[1+\frac{v^2}{v_1^2}-\frac{v_2^2}{v_1^2}\right]dv_1\ , \tag{9}$$

where $L^{(0)}(v)$ is obtained from $S^{(0)}(v)$ as $L(v)$ from $S(v)$ in Eq. (1) and where $f(v_2)$ is the isotropic velocity distribution of the target electrons normalized as

$$4\pi\int_0^\infty f(v_2)v_2^2dv_2=1 \ . \tag{10}$$

Equation (9) gives the stopping cross section from the whole electronic distribution of the atom. If we use the same arguments which led to Eq. (9) for each atomic shell, we obtain

$$\begin{aligned}L_k(v)=\pi\int_0^\infty &\rho_k(v_2)v_2dv_2\\ &\times\int_{|v-v_2|}^{v+v_2} L_k^{(0)}(v_1)\\ &\qquad\times\left[1+\frac{v^2}{v_1^2}-\frac{v_2^2}{v_1^2}\right]dv_1 \ ,\end{aligned} \tag{11}$$

where $\rho_k(v_2)$ now is the velocity distribution of a k-shell electron which according to Eqs. (9) and (10) must be normalized as

$$4\pi\int_0^\infty \rho_k(v_2)v_2^2dv_2=1 \ . \tag{12}$$

From Eqs. (10) and (12) we see that for a neutral atom the total and orbital velocity distributions are related as

$$f(v_2)=\frac{1}{Z_2}\sum_k n_k\rho_k(v_2) \ , \tag{13}$$

where n_k are the orbital occupation numbers obeying the sum rule

$$\sum_k n_k=Z_2 \ . \tag{14}$$

Notice that even though n_k and w_k fulfill the same sum rule [cf. Eqs. (5) and (14)] they are not the same quantity.

Both $L^{(0)}(v)$ and $L_k^{(0)}(v)$ derive from the stopping cross section of target electrons at rest. The Bethe formula without shell corrections is valid in the limit $v \gg v_e$, where v_e is the velocity of the target electrons. Thus the Bethe formula applies and

$$L_k^{(0)}(v)=\frac{w_k}{Z_2}\ln\left[\frac{2mv^2}{I_k}\right]\theta(v-\alpha_k) \ , \tag{15}$$

where

$$\alpha_k=\left[\frac{I_k}{2m}\right]^{1/2} \ . \tag{16}$$

Lindhard[18] has derived a similar expression for $L^{(0)}(v)$ for a homogeneous electron gas.

Introducing Eq. (15) in (11) we may perform the integration over v_1, analytically obtaining

$$L_k(v)=\frac{w_k}{Z_2}[A_k(v)+B_k(v)+C_k(v)] \ , \tag{17}$$

where

$$A_k(v)=4\pi\int_0^{v-\alpha_k}\rho_k(v_2)v_2^2\ln\frac{v^2-v_2^2}{\alpha_k^2}dv_2 \ , \tag{18}$$

$$B_k(v)=4\pi\int_{v-\alpha_k}^{v+\alpha_k}\rho_k(v_2)v_2^2\left[\ln\frac{v+v_2}{\alpha_k}-\frac{(\alpha_k-v)^2-v_2^2}{2\alpha_kv_2}\right]dv_2 \ , \tag{19}$$

$$C_k(v)=4\pi\int_{v+\alpha_k}^{\infty}\rho_k(v_2)v_2^2\left[\ln\frac{v_2+v}{v_2-v}-2\frac{v}{v_2}\right]dv_2 \ . \tag{20}$$

For large values of v ($v \gg v_2$) it appears that $A_k(v) \gg B_k(v), C_k(v)$. Thus

$$\begin{aligned}L_k(v)&\simeq\frac{w_k}{Z_2}A_k(v)\\ &\simeq\frac{4\pi w_k}{Z_2}\int_0^\infty\rho_k(v_2)v_2^2\left[\ln\frac{v^2}{\alpha_k^2}+\ln\left[1-\frac{v_2^2}{v^2}\right]\right]dv_2 \ ,\end{aligned} \tag{21}$$

or, expanding

$$L_k(v)=\frac{w_k}{Z_2}\left[\ln\frac{v^2}{\alpha_k^2}-\frac{\langle v_2^2\rangle_k}{v^2}-\frac{1}{2}\frac{\langle v_2^4\rangle_k}{v^4}-\frac{1}{3}\frac{\langle v_2^6\rangle_k}{v^6}-\cdots\right] \ , \tag{22}$$

where

$$\langle v_2^n \rangle = 4\pi \int_0^\infty \rho_k(v_2) v_2^{n+2} dv_2 \; . \tag{23}$$

Thus, by comparing Eqs. (6) and (22), and disregarding L_1 and L_2, we see that for large projectile velocities we may represent $C_k(v)$ as[11]

$$C_k(v) = w_k \frac{\langle v_2^2 \rangle_k}{v^2} + w_k \frac{\langle v_2^4 \rangle_k}{2v^4} + \cdots \; . \tag{24}$$

Before proceeding we would like to point out that Eqs. (9) and (11) are, in general, different. Only in the special case where I_k is the same for all shells and $w_k = n_k$, the occupation number of shell k, can Eq. (9) be derived from Eq. (17) by means of the definition in Eq. (8). We shall refer to such a situation as the "single-I" case.

We can now calculate the orbital shell corrections $C_k(v)/Z_2$ from Eqs. (6) and (17)–(20) using Eq. (15), provided that we know w_k, I_k, and the orbital velocity distribution $\rho_k(v)$. The latter quantities were obtained from the nonrelativistic, numerical atomic wave functions from the Froese Fischer program system MCHF72.[19] Owing to the shellwise nature of the present approach we have used Hartree-Fock wave functions. However, we might also have used multiconfigurational wave functions where all configurations differ by at least two orbital replacements, in which case the one electron density does not have orbital cross terms. However, it has been our experience[19] that the shell corrections are nearly unaffected by inclusion of this kind of electronic correlation. The Fourier transforms of the wave functions yielding the momentum-space wave functions were performed with the Talman[20,21] method which is a combination of the fast-Fourier-transform technique and analytic evaluation of some of the momentum-space integrals. The actual details of the implementation of this method in the present context can be found elsewhere[19] and it suffices to state that it is a very accurate method for obtaining momentum-space wave functions. Sum rules for the momentum-space wave function are fulfilled to within less than 0.01%.

Using the Born approximation, Bethe *et al.*[22] derived an expression for the *velocity-dependent* part $\tilde{L}_K^{(0)}(v)$ of the stopping number $L_K^{(0)}(v)$ for the K shell. This takes the form

$$L_K^{(0)}(v) = \tilde{L}_K^{(0)}(v) - \frac{n_K}{2}(1+f_K)\ln(\hbar^2 I_K / me^4) \; , \tag{25}$$

where

$$\tilde{L}_K^{(0)}(v) = \frac{n_K}{2}(1+f_K)\ln(2\hbar^2 v^2/e^4) \tag{25a}$$

and $n_K f_K$ is the total oscillator strength of all optical transition from the K shell into unoccupied levels (continuum and discrete). Equation (25a) was derived by Bethe *et al.*[22] and, in order to preserve the standard form of the logarithmic term in the Bethe formula [cf. Eq. (6)], we have also assumed the same dependence of n_K and f_K for the last term in Eq. (25). In an independent-particle model, e.g., Hartree-Fock, where the generalized oscillator strength is a sum of orbital terms, the derivation of Bethe *et al.*[22] is also valid for all other shells which means that

$$\tilde{L}_k^{(0)}(v) = \frac{n_k}{2}(1+f_k)\ln(2\hbar^2 v^2/e^4) \; , \tag{26}$$

where the index k labels the kth shell of the atom. From Eq. (26) and the general form of $L_k^{(0)}$ in Eq. (15) it follows that the weight factor must be defined as

$$w_k = \frac{n_k}{2}(1+f_k) \; . \tag{27}$$

This derivation [Eq. (27)] does not assume any special form for the orbitals of the target atoms. It should also be recalled that neither the velocity-independent term of $L_k^{(0)}$, i.e., the mean excitation energy I_k, nor the oscillator strengths can be determined by the arguments given by Bethe *et al.*[22] Bethe *et al.*[22] and Walske[2] expressed I_K for the K shell in the form

$$I_K = \lambda Z_{\text{eff}}^2 R \; , \tag{28}$$

where Z_{eff} is the effective nuclear charge in the K shell and R is the Rydberg energy. The constant λ was computed by Bethe *et al.*[22] using screened hydrogenic orbitals for the K shell and it was found that λ was close to 1. We shall (see Sec. III A) discuss how this result compares with the I_K obtained from the present calculation.

It was suggested earlier by Livingston and Bethe[5] to use a different expression for w_k for the K shell, namely,

$$w_k' = n_k f_k \; , \tag{29}$$

and this formula was applied to higher atomic shells by Hirschfelder and Magee.[23] This result cannot rigorously be derived from the Born approximation and its validity is based mainly on less stringent arguments. However, we shall demonstrate below that the shell corrections obtained with Eqs. (27) and (29) do not deviate very much from one another and the present calculation is not able to resolve which of the two expressions give the most reliable theoretical shell corrections.

The oscillator strengths f_k are taken from the Hartree-Slater calculation of oscillator strength moments by Inokuti and co-workers.[12–14] The authors calculated moments for all atoms with $Z \leq 38$:

$$S(0)=\sum_k n_k f_k \equiv \sum_k s_k(0) \tag{30}$$

and

$$L(0)=Z_2 \ln\left[\frac{I}{R}\right] \equiv \sum_k l_k(0) . \tag{31}$$

The authors also give the orbital terms $s_k(0)$ and $l_k(0)$.[24] By comparing Eqs. (4), (27), and (29)–(31) we arrive at the following expression for the orbital weight factors and mean excitation energies:

$$w_k = \frac{n_k + s_k(0)}{2} , \tag{32}$$

$$w'_k = s_k(0) , \tag{33}$$

and

$$I_k = R \exp\left[\frac{l_k(0)}{w_k}\right] . \tag{34}$$

In Sec. III we will consider the application of this formalism to aluminum and argon.

III. APPLICATIONS

A. Orbital parameters for aluminum

The orbital weight factors and mean excitation energies calculated from the oscillator strength moments of Inokuti and co-workers[12,24] using Eqs. (7), (32), and (34) are reported in Table I. The relative importance of the orbital contribution to ln$I$ according to Eq. (4) is indicated in the last column of Table I and we see that the $2p$ subshell gives by far the dominant contribution to I. A small value of w_k and/or a large value of I_k indicates small stopping ability of the kth shell and the entries in Table I thus imply that most stopping is accomplished by the outer electrons. For instance, for 2-MeV protons the relative contributions of $L_{1s}^{(0)}$, $L_{2s}^{(0)}$, $L_{2p}^{(0)}$, $L_{3s}^{(0)}$, and $L_{3p}^{(0)}$ to $L^{(0)}$ are 4%, 11%, 42%, 26%, and 17%, respectively. The same trend is observed when comparing the orbital weight factors with the occupation numbers: w_k is smaller than n_k for 1s and 2s, while w_{2p} (and w_{3p}) are larger than n_{2p} (and n_{3p}). However, w_k and n_k are rather similar for all k and n_k would be a good zeroth-order approximation for the weight factors. This is in fact the approach followed by Sternheimer[25,26] in his calculation of the density-effect correction δ, and our calculation thus indicates that this choice is rather good. Also, since requiring that $w_k = n_k$ is a necessary condition for obtaining the "single-I" approximation from the orbital expression, Eq. (11), we see that the most important difference between shell corrections calculated from Eqs. (9) and (11) stems from the use of orbital mean excitation energies in Eq. (11).

The value calculated by Dehmer *et al.*[12] for I (124 eV) and thus for $l_k(0)$ of aluminum is too small relative to the commonly accepted experimental value of about 163 eV.[27,28] The main reason for this discrepancy seems to be[12] that the calculations are carried out for atoms while the experiments refer to stopping by a metallic foil of aluminum. To a lesser extent, it may also be caused by the absence of electronic correlation in the Hartree-Slater calculation of Dehmer *et al.*[12] Recently, Shiles *et al.*[29] have analyzed a range of experimental optical data for metallic Al in order to determine the dielectric response function over the entire range of frequency as required by the Kramers-Kronig relations. From the dielectric response function they compute a mean excitation energy of 166 eV; very close to the best experimental estimate of I. Their data also show that the entire additional oscillator strength missing in the Hartree-Slater[12] calculation is concentrated in the valence part of the spectrum. Based on these findings we will thus increase the values of $l_k(0)$ for the valence shells (2s,3p) according to the prescription

$$l'_k(0) = l_k(0) + Z_2 \frac{w_k}{\sum_{\text{valence}} w_k} \ln \frac{I_{\text{expt}}}{I_{\text{Inokuti}}} . \tag{35}$$

TABLE I. Orbital weight factors w_k [Eq. (27)] and mean excitation energies I_k for Al calculated from $l_k(0)$ of Inokuti (Ref. 24), yielding $I = 123.6$ eV.

Shell (k)	w_k	$l_k(0)$	I_k (eV)	% of ln$I$
K	1.773	8.181	1373.0	20.5
L_1	1.677	4.397	187.2	14.0
L_2	6.487	18.088	221.2	55.9
M_1	1.909	−0.788	9.003	6.7
M_2	1.153	−1.189	4.849	2.9

TABLE II. Orbital weight factors w_k [Eq. (27)] and mean excitation energies I_k for Al calculated from renormalized $l_k'(0)$ values in Eq. (35) which yield $I=163$ eV.

Shell (k)	w_k	$l_k'(0)$	I_k (eV)	% of $\ln I$
K	1.773	8.181	1373.0	19.3
L_1	1.677	4.397	187.2	13.3
L_2	6.487	18.088	221.2	52.9
M_1	1.909	1.304	26.95	9.5
M_2	1.153	0.313	17.86	5.0

We have assumed that the term by which $L(0)$ should be incremented, i.e.,

$$Z_2 \ln \frac{I_{\text{expt}}}{I_{\text{Inokuti}}} ,$$

is added to the valence orbital terms proportionally to the orbital weight factors.

Tables II and III give the renormalized mean excitation energies as calculated from w_k and w_k' in Eqs. (27) and (29), respectively. Comparison of Tables II and III shows that the increase of w_{2p} relative to n_{2p} is more pronounced when we use the original "intuitive" suggestion for w_k' of Livingston and Bethe[5] (Table III) rather than the formula which has a velocity dependence consistent with the use of the Born approximation[22] (Table II).

B. Shell corrections for aluminum

Using the I_k values of Tables I–III in Eq. (15) and using Eqs. (6) and (11), yields the curves for shell corrections versus incident-particle energy displayed in Figs. 1–4. In Fig. 1 we display the calculated C/Z_2 at three levels of approximation. We also include the calculation of Bonderup[4] which treats the target as an electron gas with the appropriate density. The experimental result is that of Andersen *et al.*[27] in which the Barkas corrections and the Bloch corrections, i.e., L_1 and L_2 in Eq. (2), have been subtracted out. It can thus be compared to the present calculation which also does not include these terms. Both curve A [weight factors computed from Eq. (27)] and curve B [w_k' from Eq. (29)] agree well with the experimental C/Z_2 from Andersen *et al.*[27] Thus based on the present calculation, we can form no preference for the weight factors of Ref. 5 or 22. Both choices seem to yield acceptable shell corrections for aluminum. Also we see from Fig. 1 that C/Z_2 obtained by using a common mean excitation energy in $L^{(0)}(v)$ for all atomic shells [Eq. (9)] is much too large. The fact that the single-I shell correction is too large is not caused by the use of an incorrect I value. This can be seen from Fig. 2 where we have displayed the total single-I shell correction for three rather different values of the total mean excitation energy. C/Z_2 is almost independent of I in the single-I approximation. There is only a slight increase in the peak value as I decreases.

All calculations in Fig. 1 are performed with orbital mean excitation energies which give a total I of 163 eV, the "experimental" value for I.[28] Using the unnormalized orbital mean excitation energies in Table I we obtain nearly the same shell corrections as curve B in Fig. 1. The changes are smaller than 0.5%. The reason for the insensitivity of the shell correction to the renormalization effect can be seen from Table IV. The whole M shell contributes merely 1–3% to the total C/Z_2 and only the M-shell corrections are altered when using the orbital mean excitation energies of Table I rather than

TABLE III. Orbital weight factors w_k' [Eq. (29)] and mean excitation energies I_k for Al calculated from renormalized values of $l_k'(0)$ [Eq. (35)] which yield $I=163$ eV.

Shell (k)	w_k'	$l_k'(0)$	I_k (eV)	% of $\ln I$
K	1.546	8.181	2704.0	18.5
L_1	1.354	4.397	349.7	12.0
L_2	6.973	18.088	182.1	54.8
M_1	1.818	1.304	27.89	9.1
M_2	1.305	0.313	17.30	5.6

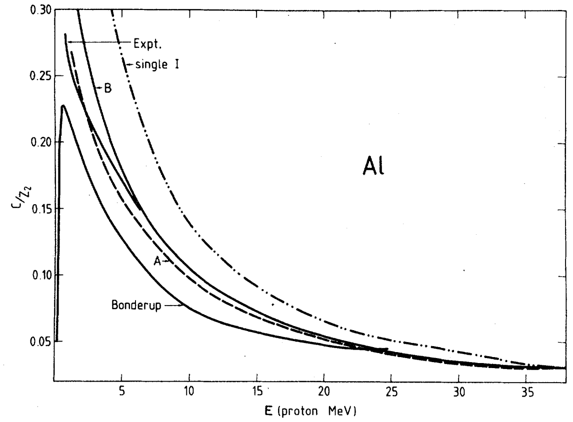

FIG. 1. Total shell correction for Al ($I=163$ eV) as a function of the projectile velocity. Curve A: this work with w_k' and I_k from Table III. Curve B: this work with w_k and I_k from Table II. Expt.: Andersen *et al.* (Ref. 27). Single-I: All $I_k=163$ eV and $w_k=n_k$. Bonderup: Ref. 4.

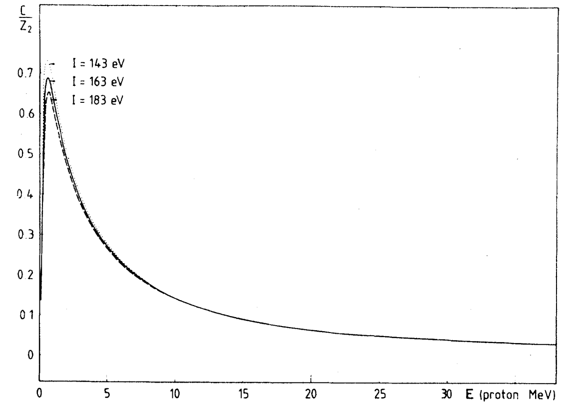

FIG. 2. Total shell correction for Al in the single-I approximation as a function of the projectile velocity calculated for three single-I values of the mean excitation energy $I=143$, 163, and 183 eV.

those of Table II. Thus, the shell correction is predominantly determined by the sum of the K- and L-shell corrections. Table IV shows that the L shell gives the dominant contribution for small projectile velocities (90% for $v=6$ a.u.), while the K-shell corrections do not fall off as fast as the L-shell corrections, being about 66% of C/Z_2 for $v=40$ a.u.

Using the formalism of Bethe[1] and screened hydrogenic orbitals for the target electrons, Walske[2,3] calculated the K- and L-shell corrections for a range of atoms. We compare our calculation with his results in Figs. 3 and 4 for the K and L shell, respectively. For small projectile velocities the Walske K-shell corrections lie between the shell correction obtained with weight factors in Eq. (27) (curve B) and with w_k' of Eq. (29) (curve A), whereas curve B seems to be closer to Walske's results for larger velocities. However, it appears from Fig. 3 as if the Walske shell corrections may be too large for projectile energies 22 MeV and the agreement between the Walske shell correction and curve B in this energy range thus does not tell us whether w_k or w_k' is the most appropriate choice of weight factors. Walske's[2] shell corrections correspond to a K-shell mean excitation energy of about 2300 eV [$\lambda\sim 1.05$ in Eq. (28)], i.e., in between the I_K values of Tables II and III. Thus, Fig. 3 shows that the magnitude of the K-shell correction is mainly a function of I_K; C_K is larger for smaller I_K, with the single-I

TABLE IV. Orbital shell corrections[a] for Al.

v (a.u.)[b]	$\frac{1}{2}M_pv^2$ (MeV)[c]	$\frac{C_{1s}}{Z_2}$	$\frac{C_{2s}}{Z_2}$	$\frac{C_{2p}}{Z_2}$	$\frac{C_{3s}}{Z_2}$	$\frac{C_{3p}}{Z_2}$	$\frac{C}{Z_2}$
6	0.8993	0.023 86	0.043 18	0.290 76	0.007 21	0.004 12	0.369 13
8	1.5988	0.073 57	0.037 70	0.194 33	0.005 30	0.002 25	0.313 16
10	2.4981	0.095 62	0.032 43	0.126 61	0.004 02	0.001 34	0.260 03
12	3.5972	0.100 46	0.026 60	0.085 50	0.003 02	0.000 87	0.216 45
16	6.3951	0.086 57	0.016 79	0.044 77	0.001 71	0.000 45	0.150 28
20	9.9923	0.065 83	0.010 66	0.027 22	0.001 04	0.000 27	0.105 01
24	14.3889	0.048 64	0.007 07	0.018 31	0.000 67	0.000 18	0.074 88
32	25.5803	0.027 41	0.003 61	0.009 97	0.000 34	0.000 10	0.041 42
40	39.9693	0.016 84	0.002 14	0.006 29	0.000 20	0.000 06	0.025 53

[a] Using the orbital I_k's in Table II, i.e., yielding $I=163$ eV.
[b] In units of $e^2/\hbar$,
[c] Proton MeV.

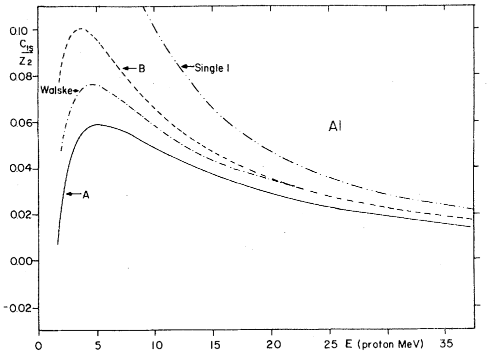

FIG. 3. K-shell correction for Al as a function of the projectile velocity. Curve A: this work with $I_K = 2704$ eV (Table III). Curve B: this work with $I_K = 1373$ eV (Table II). Walske: Ref. 2. Single-I: $I = 163$ eV.

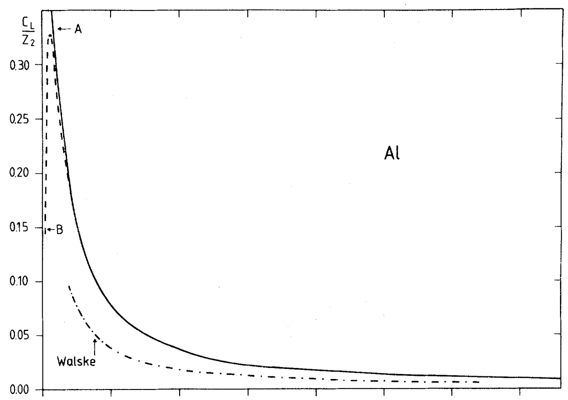

FIG. 4. L-shell correction for Al (L_1+L_2) as a function of the projectile velocity. Curve A: this work with I_k and w_k' from Table III. Curve B: this work with I_k and w_k from Table II. Walske: Ref. 3.

($I = 163$ eV) case producing a value which is far too large. Since the K shell gives the dominant contribution to the shell correction in the single-I case the large difference between curve B and the single-I curve in Fig. 3 has the same origin as the difference between the same two curves in Fig. 1. This is also supported by the fact that if we had included the single-I curve in Fig. 4, it would have coincided with curve A.

Figure 4 shows that the Walske[3] L-shell corrections are substantially smaller than those obtained in both of the present calculations, indicating that the screened hydrogenic approximation does not work as well for the L shell as for the K shell. Noticing the difference in scale between Figs. 3 and 4 this means that the total Walske shell correction is smaller than ours (the contribution to C/Z_2 from the M shell is only a few percent; see Table IV and Ref. 10). The total Walske shell correction is also smaller than that calculated by Bonderup.[4] This is a consistent trend over the periodic system as seen, e.g., in the analysis of stopping powers by Andersen *et al.*[30]

Finally we investigated the validity of the asymptotic expansion of C/Z_2 for large projectile velocities, i.e., Eq. (24). Since we have almost the same convergence for all individual shell corrections, we have given the results for the total shell correction in Table V. The rest correction, defined as C/Z_2 minus the two leading expansion terms in Eq. (24),

TABLE V. The expansion of the total[a] shell correction C/Z_2 of aluminum in terms of moments of $\langle v_2^2 \rangle$ and $\langle v_2^4 \rangle$ [cf. Eq. (24)].

v (a.u.)[b]	$\frac{1}{2}M_p v^2$ (MeV)[c]	C/Z_2	$\frac{\langle v_2^2 \rangle^{\mathrm{d}}}{v^2}$	$\frac{1}{2}\frac{\langle v_2^2 \rangle^{\mathrm{c}}}{v^4}$	Rest correction[f]
10	2.4981	0.260 03	0.346 46	0.982 79	−1.069 22
12	3.5972	0.216 45	0.240 60	0.473 95	−0.498 11
16	6.3951	0.150 28	0.135 34	0.149 96	−0.135 02
20	9.9923	0.105 01	0.086 62	0.061 42	−0.043 03
24	14.3889	0.074 88	0.060 15	0.029 62	−0.014 89
32	25.5803	0.041 42	0.033 83	0.009 37	−0.001 78
40	30.9693	0.025 53	0.021 61	0.003 80	−0.000 04

[a] Using the I_k values in Table II, i.e., with $I = 163$ eV.

[b] In units of $e^2/\hbar$.

[c] Proton MeV.

[d] $\langle v_2^2 \rangle = 34.646$ a.u.

[e] $\langle v_2^4 \rangle = 1.9656 \times 10^5$ a.u.

[f] The rest correction is defined as $C/Z_2 - \frac{\langle v_2^2 \rangle}{v^2} - \frac{1}{2}\frac{\langle v_2^4 \rangle}{v^4}$.

TABLE VI. Orbital weight factors w_k [Eq. (27)] and mean excitation energies I_k for Ar yielding $I=194$ eV.

Shell (k)	w_k	$I'_k(0)$	I_k (eV)	% of ln$I$
K	1.705	8.462	1947.0	13.6
L_1	1.691	5.666	388.1	10.6
L_2	6.447	24.107	572.2	43.2
M_1	1.337	1.886	55.78	5.7
M_2	6.820	7.713	42.15	26.9

is always negative, which means that the two-term expansion always lies above C/Z_2, while the leading term in the expansion, proportional to v^{-2}, lies above C/Z_2 for small v but below it for larger ($\geq$5-MeV) projectile velocities. Table V also shows that the series expansion converges rather well; above 25 MeV the rest correction is less than 5% of the total shell correction.

C. Results for argon

We also performed a calculation on Ar using the orbital weight factors of Eqs. (27) and (29). The weight factors obtained from Eq. (27) and the corresponding mean excitation energies are given in Table VI, and they are calculated from the oscillator strength moments of Inokuti[24] renormalized according to Eq. (35) to yield $I=194$ eV (Ref. 28) instead of the value of 175 eV found by Dehmer *et al.*[12,24] However, according to the discussion concerning Fig. 1, this renormalization has nearly no effect on the computed shell corrections.

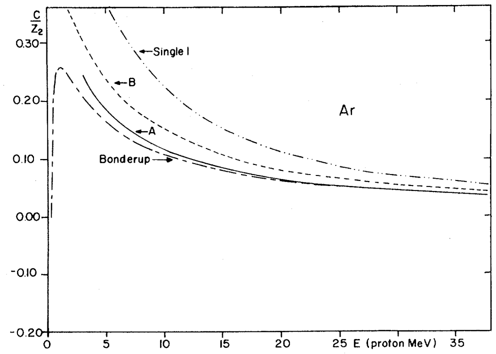

FIG. 5. Total shell correction for Ar as a function of the projectile velocity with $I=194$ eV. Curve A: w_k and I_k from Table VI. Curve B: w'_k calculated from Eq. (29). Single-I: All $I_k=194$ eV and $w_k=n_k$. Bonderup: Ref. 4.

The shell corrections are plotted in Fig. 5 together with C/Z_2 calculated by Bonderup[4] and C/Z_2 in the approximation which uses a common I in all $L_k^{(0)}(v)$, i.e., Eq. (9). The relation between the curves is similar to that found for Al in Fig. 1. The only experimental determination of C/Z_2 versus v is that of Besenbacher *et al.*[31] However, these measurements are carried out only in the energy range 100–800 keV which corresponds to such low projectile velocities that it is difficult to compare with the present calculation. Furthermore, Besenbacher *et al.*[31] have not determined the Barkas and Bloch corrections separately. These terms are thus included in the experimental C/Z_2 but not in our calculated values. Besenbacher *et al.*[31] made an empirical estimate of L_1 and L_2, and by subtracting those terms they arrive at an experimental curve for C/Z_2 vs v which is rather close to the calculation of Bonderup.[4] However, the uncertainties in the Barkas and Bloch terms are so great that the agreement with Bonderup's calculation may be fortuitous.

IV. SUMMARY AND DISCUSSION

Using the theory of Sigmund,[11] which relates the stopping cross section of moving scatterers to that of scatterers at rest, $S^{(0)}(v)$, we have calculated shell corrections to electronic stopping powers for Al and Ar. The velocity distribution of the scatterers is calculated from Froese Fischer nonrelativistic atomic wave function at the Hartree-Fock level of approximation. We have shown that by using an $S^{(0)}(v)$ function composed of orbital mean excitation energies which summed and weighted according to Eq. (4) give the experimental[28] mean excitation energy, we are obtaining a total shell correction which is substantially smaller than that obtained when we use a common mean excitation energy (equal to the experimental mean excitation energy[28]) for all the shells. Furthermore, the shell corrections calculated from orbital mean excitation energies are in good agreement with both the calculation of Bonderup[4] and the measurements of Andersen *et al.*[27] for Al.

The orbital mean excitation energies are large for the K shell (small stopping ability) and small for M shell (large stopping ability). The orbital weight factors and mean excitation energies are computed from the Hartee-Slater calculations of Inokuti and co-workers.[12–14,24]

One of the key assumptions in Sigmund's derivation of $L(v)$ in terms of $L^{(0)}(v)$ is that the stopping of the projectile is accomplished by collisions with target electrons with a velocity distribution $f(v_2)$. By using a different mean excitation energy for each orbital velocity distribution we take into account that the binding energy differs from shell to shell. In the single-I approximation we do not consider the shell structure of the atom which probably is the reason for the unrealistically large shell correction in the latter approximation.

At sufficiently high projectile velocities the shell correction is given by the asymptotic expansion in Eq. (24) and is thus independent of the mean excitation energy. However, the velocity at which this is true is much larger than the velocities normally attained in stopping-power measurements. Also, our calculations show that even at a proton energy of 35 MeV we still do not obtain quite the same shell correction in the single-I and in the orbital approximations.

We will end the discussion with a few remarks on the implication of the present calculation on the experimental data analysis as, e.g., pursued by Andersen *et al.*[27] From the experiments one obtains the stopping number $L(v)$ and Andersen *et al.* then add the *total* shell correction as computed by Bonderup[4] or Walske[2,3] to $L(v)$ in order to obtain the logarithmic term in the Bethe formula [cf. Eq. (2) without Barkas and Bloch corrections]. If the computed shell correction did indeed include all velocity-dependent terms in $L(v)$ except for $\ln(mv^2)$, the inferred value for I would be a constant as a function of v. This is of course not the case. However, if I as computed by adding the shell corrections of both Bonderup[4] and Walske[2,3] seems to converge towards the same value for large projectile velocity, Andersen *et al.*[27] conclude that this value of I represents the experimental mean excitation energy. Thus, in the data analysis only the total shell correction is needed. Even though we compute the total stopping number by adding up orbital contributions which are computed from different orbital mean excitation energies, we still obtain the total shell correction from the relation

$$C/Z_2=\ln(2mv^2/I)-L(v)\ . \qquad (36)$$

This follows trivially from Eqs. (4)–(8) for $2mv^2>I_k$. Since Eq. (36) is just the definition used for the total shell correction in the data analysis of Andersen *et al.*[27] and of Besenbacher *et al.*,[31] we conclude that the evaluation of $L(v)$ from orbital mean excitation energies does not deviate from the conventional definition of C/Z_2 nor does it influence the standard procedure used for extracting experimental mean excitation energies from measurements of stopping cross sections.

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