# Predicted $Z_2$ Structure and Gas-Solid Difference in Low-Velocity Stopping Power of Light Ions

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Atomic stopping cross sections for low-velocity light ions have been evaluated for the atomic numbers $1 \leq Z_2 \leq 36$ on the basis of the kinetic theory of electronic stopping. Valence electrons make the main contribution. A pronounced $Z_2$ structure in stopping cross sections is found, with minima for noble gases where our estimates agree well with experimental results and with the Lindhard-Scharff formula. Pronounced maxima are predicted for alkalis, where large gas-solid differences are to be expected.

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It is well documented both experimentally and theoretically that the stopping power of ions in matter shows a nonmonotonic dependence on the atomic numbers of the target ($Z_2$) and the penetrating ion ($Z_1$). For swift light ions, experimentally observed $Z_2$ structure[1,2] can be ascribed mainly to a nonmonotonic $Z_2$ dependence of the mean excitation energy[3,4] which enters into the Bethe formula.[5] Pronounced oscillatory structure has been found in the $Z_1$ dependence of low-velocity stopping powers.[6,7] Structure in the $Z_2$ dependence has also been reported from measurements at intermediate[8] and low[9,10] light-ion velocities.

The present note concerns the electronic stopping of light ions at low velocities ($v < v_0 = e^2/\hbar$) where the stopping power is approximately proportional to velocity.[11] Theoretical estimates have been based either on the stopping of a point charge in a free-electron gas,[12] extended to colliding atoms by means of Thomas-Fermi arguments,[13] or on geometric considerations of the quasi-free-electron flux between two colliding atoms.[14] The theoretical literature on incorporation of shell structure into either model is extensive.[15]

Atomic structure enters into both models through the *electron density* distribution possibly differentiated into the densities for individual shells. *Binding energies* enter only indirectly to the extent that they are correlated with the local electron density. This is somewhat surprising in view of the important role played by binding forces in Bohr's[16] and Bethe's theory.[5]

Recently, one of us[17] made an attempt to incorporate atomic binding forces into a kinetic theory of stopping by a method, described below, which was designed to reproduce standard results in the high-velocity limit but turned out also to have potential at low velocities. In brief, the well-known[18] binary-encounter stopping cross section $S(v)$ for a heavy projectile colliding with free electrons with a velocity spectrum $f(v')d^3v'$,

$$S(v) = (m/v)\int d^3v' f(v')\vec{\mathrm{v}}\cdot(\vec{\mathrm{v}}-\vec{\mathrm{v}}')\,|\vec{\mathrm{v}}-\vec{\mathrm{v}}'|\,\sigma^{(1)}(|\vec{\mathrm{v}}-\vec{\mathrm{v}}'|), \tag{1}$$

where $\sigma^{(1)}(v) = \int d\sigma(v,\theta)(1-\cos\theta)$ is a transport cross section and $\theta$ is the c.m. system scattering angle, has the limiting form

$$S_0(v) = mv^2\sigma^{(1)}(v) \tag{2}$$

at high projectile velocities. Elimination of $\sigma^{(1)}$ from (1) and (2) yields a transformation of $S_0$ into $S$ which is an identity in the case of binary elastic encounters between free particles. The relation was then employed for the case of nonnegligible binding forces by insertion of the Bethe formula or its Fermi-gas equivalent for $S_0$. At moderate and high velocity, this scheme turned out to provide valid expansions for shell corrections[17] both in the Bethe scheme and for the free-electron gas.[19] Shell corrections evaluated for individual shells turned out[20] to agree well with literature values.[21] The overall agreement with experimental stopping powers was found to be very good.[20]

Now, the low-velocity limit of Eq. (1) reads

$$S(v) = mv\int_0^\infty 4\pi v'^2 dv' f(v')v'\left[\tfrac{4}{3}\sigma^{(1)}(v') + \tfrac{1}{3}v'\,d\sigma^{(1)}(v')/dv'\right], \tag{3}$$

for the leading term of an expansion in $v$. Elimination of $\sigma^{(1)}$ by means of Eq. (2) yields[17]

$$S(v)=v\int_0^\infty 4\pi v'^2 dv' f(v')[\tfrac{2}{3}S_0(v')+\tfrac{1}{3}dS_0(v')/dv'] \quad (4)$$

for small $v$. When applied to the Fermi gas, this expression was found[17] to reproduce the standard low-velocity stopping cross section for that system[19] to within a factor of 2 over a wide range of electron densities. This provides considerable confidence for applying the same procedure also to atoms, i.e., to insert the Bethe stopping cross section differentiated into individual shells,[22]

$$S_0(v)=\frac{4\pi e_1^2 e^2}{mv^2} Z_2 \sum_k w_k \theta(2mv^2-I_k)\ln\frac{2mv^2}{I_k}, \quad (5)$$

into Eq. (4). The result is[17]

$$S(v)=(4\pi e_1^2 e^2/m)Z_2\times\tfrac{2}{3}v\textstyle\sum_k \omega_k \langle v'^{-3}\rangle_k, \quad (6)$$

where $e_1=Z_1 e$ and

$$\langle v'^{-3}\rangle_k=\int_{(I_k/2m)^{1/2}}^\infty 4\pi v'^2 dv' f_k(v')v'^{-3}. \quad (7)$$

The orbital weight factors $w_k$ are normalized to $\sum w_k=1$. $I_k$ is the mean excitation energy of the $k$th shell, and $f_k(v')$ is the velocity distribution of electrons in the $k$th shell $[\int_0^\infty 4\pi v'^2 f_k(v')dv'=1]$. The step function $\theta$ in Eq. (5) serves to exclude nonphysical contributions to the stopping power.

It is seen that atomic binding forces enter directly into the stopping formula (6) through insertion of the Bethe formula Eq. (5). Their influence is substantial.

Equation (7) does not incorporate deviations from the first Born approximation, which may be sizable in view of the importance of low relative velocities $[v'\gtrsim(I_k/2m)^{1/2}]$ in the integral in Eq. (7). Moreover, as it stands, Eq. (6) disregards screening of the projectile charge. We do not wish to deny that both effects may be the cause of noticeable uncertainties in the estimates presented below. However, they hardly overshadow the pronounced $Z_2$ variations found.

Equations (6) and (7) have been evaluated for $1\leq Z_2\leq 36$ by means of orbital weights $w_k$ and mean excitation energies $I_k$ taken from Refs. 4 and 23. Orbital velocity distributions $f_k(v')$ have been obtained according to the scheme described by Oddershede and Sabin[24] which is based on numerical atomic Hartree-Fock wave functions.[25]

Table I shows that the magnitude of the essential material parameter, the term $\sum_k w_k\langle v'^{-3}\rangle_k$, is determined almost exclusively by the contribution from the most loosely bound electrons. From this, it follows that high stopping cross sections are to be expected for materials with low average orbital velocities and low mean excitation energies in the valence shell, in particular, alkali-metal and alkaline-earth atoms. Figure 1 predicts the stopping cross section of an alkali atom to exceed that of the adjacent noble-gas atom by more than an order of magnitude.

In view of the dominant contribution of the outermost shell, it is evident that our present results, which are based on atomic wave functions, are applicable only to *atomic* gases and vapors. To illustrate this point, note that for atomic Na, the

TABLE I. Contribution of inner shells to low-velocity stopping cross section, Eq. (6).

| Element | Shells | Relative contribution (%) |
|---|---|---|
| Li | 1s | 0.9 |
| Ne | 1s | 0.5 |
| Na | 1s-2p | 2.1 |
| Ar | 1s-2p | 1.7 |
| K | 1s-3p | 4.1 |
| Kr | 1s-3d | 2.3 |

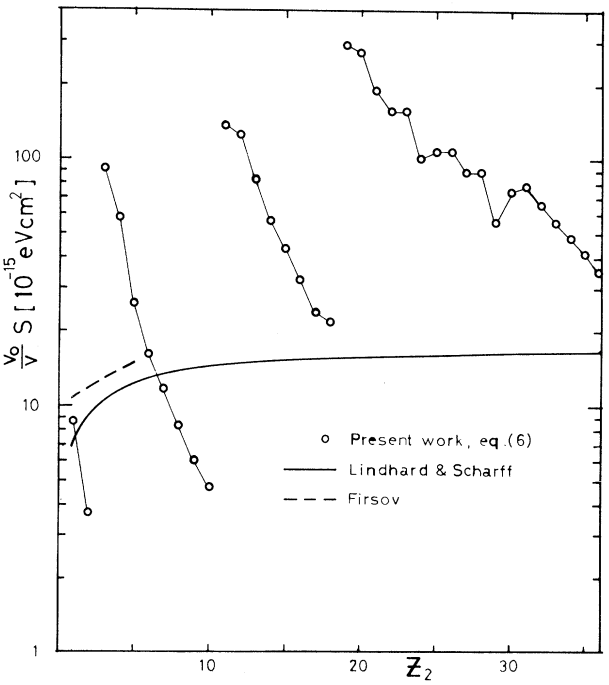


FIG. 1. Atomic stopping cross sections for bare protons in the low-velocity limit, evaluated from Eq. (6), compared with Lindhard-Scharff (Ref. 12) and Firsov (Ref. 13) predictions.

value of $I$ for the 3s electron is 2.462 eV, while for metallic Na the equivalent quantity, the plasma frequency $\omega_p$ for conduction electrons, is estimated to be $\hbar\omega_p \simeq 5.9$ eV. Moreover, the velocity of a 3s electron in atomic Na is given by $\langle v'^2\rangle_{3s}^{1/2} = 0.223 v_0$, while the equivalent quantity in metallic Na is $(3v_F^2/5)^{1/2} \simeq 0.37 v_0$, $v_F$ being the Fermi velocity. Thus, regardless of the details, one expects a considerably higher stopping power in sodium vapor than in metallic Na.

Figure 1 includes the stopping cross section obtained from the Lindhard-Scharff formula,[12] which is known to provide reasonable estimates of the stopping in solid as well as atomic and molecular gas targets. This prediction, which does not incorporate shell-structure effects, is intermediate between our predicted maximum and minimum values for $Z_2 \leq 10$, while for $Z_2 \geq 11$, our predicted values are systematically higher.

Table II presents a comparison with experimental data on noble-gas targets (as compiled in Besenbacher *et al.*[26]). It is evident that our predicted stopping cross sections agree with the measured values about as well or slightly better than those evaluated from Ref. 12. The maximum discrepancy occurs for neon where our result is too low by ~ 40%—possibly due to lack of a correlation correction both in $I_k$ and the atomic velocity distributions entering Eq. (1)—while the Lindhard-Scharff prediction is too high by ~75%.

TABLE II. Stopping cross sections for protons in gases ($10^{-15}$ eV cm$^2$/atom). In the fourth column are data evaluated by the procedure described in Ref. 20. Deviations from the low-velocity limit, Eq. (6), are noticeable at 40 keV and are found to be either positive or negative.

| Gas | Theoretical Ref. 12 | Theoretical Eq. (6) | Theoretical Eq. (1) | Experimental |
|---|---|---|---|---|
| | | Proton energy 10 keV | | |
| He | 5.8 | 2.3 | 2.7 | 3.0 [a] |
| Ar | 9.9 | 13.9 | 13.0 | 16.5 [a,b] |
| Kr | 10.5 | 22 | 21 | 17 [a] |
| | | Proton energy 40 keV | | |
| He | 11.6 | 4.5 | 5.1 | 5.6–6.7 [a,c,d,e] |
| Ne | 18.1 | 6.0 | 6.3 | 10.2–10.6 [c] |
| Ar | 20 | 28 | 22 | 26–32 [a,b,c,d,f] |
| Kr | 21 | 44 | 34 | 29–36 [a,c,d] |

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[c] Ref. 26.
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In order to avoid spurious results due to the low-velocity expansion, Eq. (6), all stopping cross sections have also been evaluated directly from Eq. (1) according to the procedure described in Ref. 20 and by Sabin and Oddershede,[27] and the results have been included in Table II. Only minor deviations of either sign are observed for the noble gases in the range of energies in question. However, major deviations have been found especially for alkali-metal and alkaline-earth vapors where the maximum of the curve of stopping power versus energy is predicted to shift toward much lower energies.[28] The latter phenomenon is presumably related to a recently reported oscillatory effect.[29]

By comparison with electron-gas calculations,[17] one expects the present predictions to overestimate stopping cross sections for low mean excitation energies ($I_k \lesssim 3$ eV). While this may be the cause of a systematic error in Fig. 1 in the region of $Z_2 \gtrsim 19$, it cannot wipe out the pronounced variations in the regions $3 \leq Z_2 \leq 10$ and $11 \leq Z_2 \leq 18$.

Both the dominance of outer-shell electrons on the stopping cross section and the pronounced deviations of our predicted stopping cross sections from the Lindhard-Scharff values for elements in the first columns of the periodic table indicate the existence of pronounced gas-solid differences in those stopping cross sections. The literature on this topic is extensive. While noticeable differences have been found,[30] many investigations seem to have dealt with systems where large differences could not be expected, either because of a minor contribution of the valence shell to the stopping power as is the case at high velocity[23] or because of small differences in the electron structure between the gaseous and condensed state, as is the case for noble-gas targets,[31] or both. Sizable gas-solid differences in low-velocity stopping have been found recently for $N_2$ targets.[32]

The present predictions are expected to apply primarily for low-$Z$ ions, i.e., hydrogen and helium isotopes and, perhaps, lithium. Additional structure must be expected for heavier projectiles originating from electron promotion[33,34] which is not included in the present treatment.

In summary, we expect (i) large variations —peak-to-peak ratios exceeding one order of magntiude—in the $Z_2$ dependence of low-velocity

stopping cross sections measured on atomic gases and vapors; (ii) large gas-solid differences in low-velocity stopping powers for elements in the first columns of the periodic table; (iii) stopping cross sections far in excess of the Lindhard-Scharff estimate for alkali-metal and alkaline-earth vapors.

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