Heavy Ion Electronic Stopping in Solids (Dielectric Theory)

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Contents

1 Fundamental: Point charge stopping in the jellium

1.1 Dielectric mechanism of electronic energy loss, Lindhard-Winther theory

The ion *is* a charge cloud $\rho_m(r) = Z_1 e\delta(r) + \rho_e(r)$ with velocity $\vec{v}$, ion atomic number Z_1, and charge $Qe = \int \rho_m(r) d^3r$. The target *is* a polarizable homogeneous medium with dielectric function $\epsilon(\vec{k}, \omega)$.

$\Rightarrow$ Maxwell's theory defines the polarization charges

$\Rightarrow$ The electronic energy loss *is* the Coulomb Force between the polarization charges and the ion
(or) the Joule's heat of the ion electric current in the field of the polarization charges.

$$
\begin{aligned}
\frac{dE}{dt} &= \vec{v}\int \rho_m(r)\vec{E}_{\text{ind}} d^3r \\
\frac{dE}{dx} &= \frac{1}{2\pi^2\epsilon_0 v^2}\int_0^\infty \frac{dk}{k}\left|\rho_m(k)\right|^2 \int_0^{kv} \omega \text{Im}\left[\frac{1}{\epsilon(k,\omega)} - 1\right] d\omega
\end{aligned}
$$

$\uparrow$

target properties

$\uparrow$

energy loss

ion form factor

function

$\rho_m(k) = \int e^{i\vec{k}\vec{r}}\rho_m(r) d^3r$

Im$\chi^R(k,\omega) \sim$

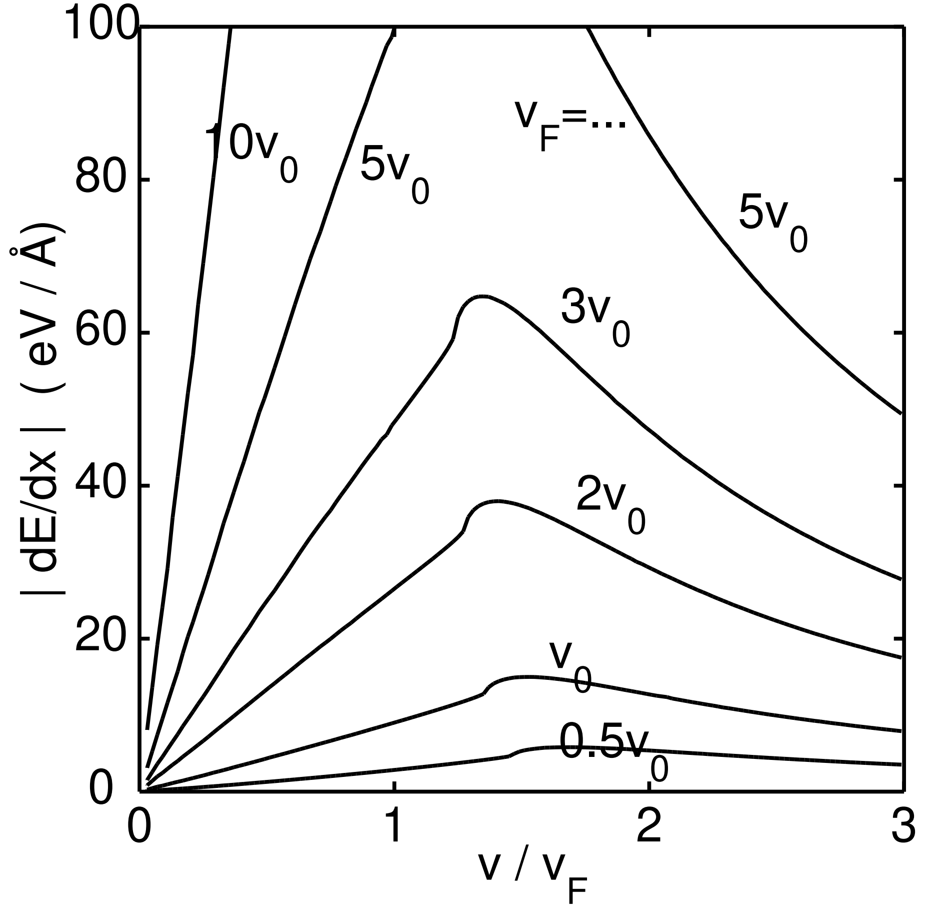
retarded, odd part

of $\chi^R(k,t)$

Bare ions [$\rho_m(k) = Z_1 e$] within linear response obey $|dE/dx| \sim Z_1^2$.

The Lindhard-Winther theory is based on the linear susceptibility χ^R in RPA. The effective interaction between target electrons is

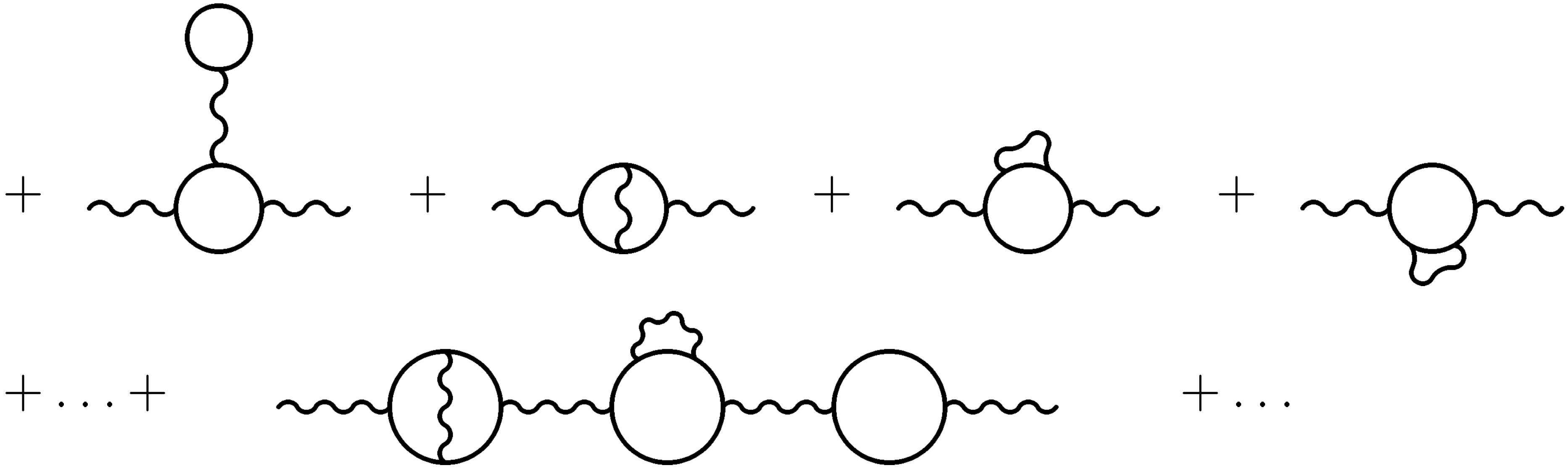
$+ \ldots$



The Lindhard dielectric function is equivalent to the RPA and the non-interacting (zeroth order) polarization

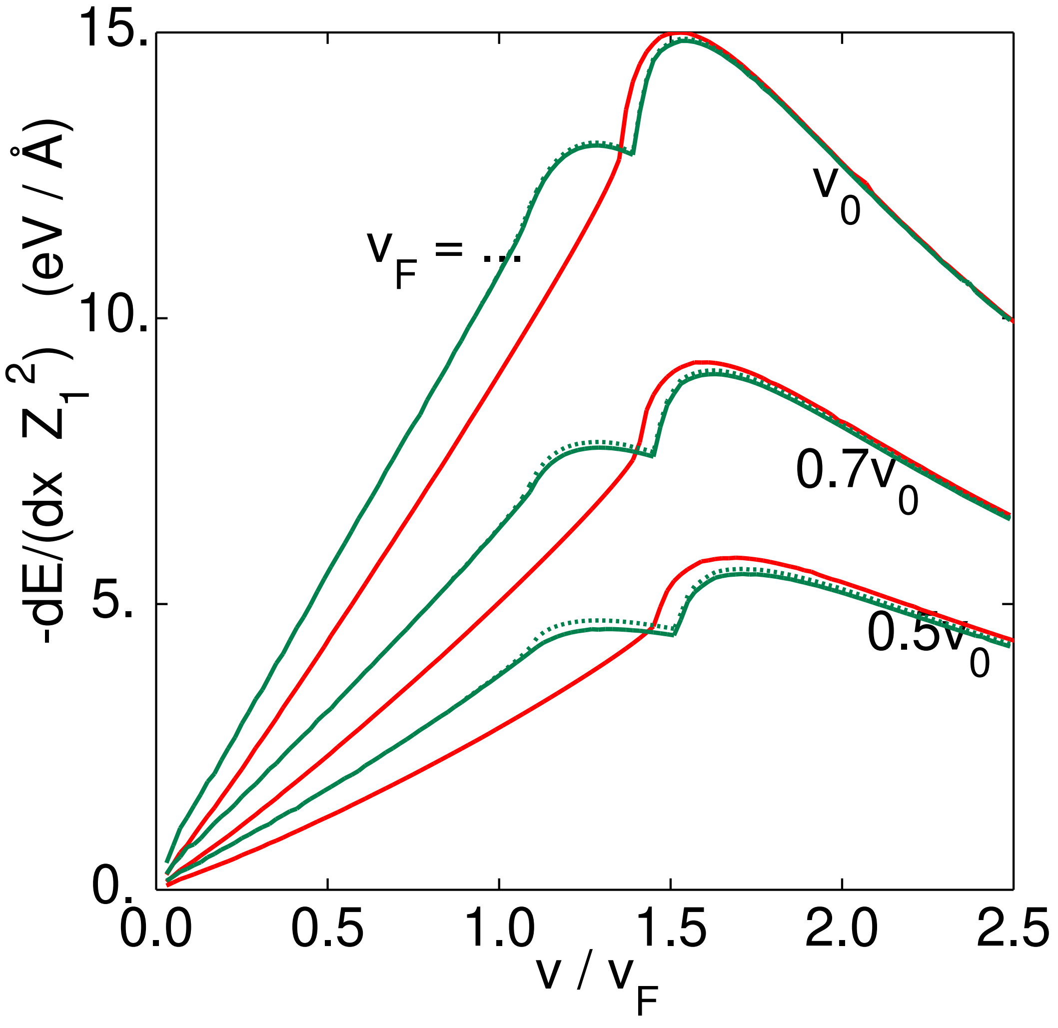
1.2 Energy loss functions "beyond" RPA

As a wishful extension to electronic systems with medium density (to compete with DFT results, but incorporate the "full" dynamics of the excitation) the first order polarization diagrams with one interaction line are added:



$$\epsilon(k,\omega) = 1 - U_0(k)\left(\Pi^{(0)}(k,\omega) + \Pi^{(1)}(k,\omega) + \ldots\right)$$

Finally build Im$\dfrac{1}{\epsilon(k,\omega)}$ and integrate . . .



Result: in the region $dE/dx \propto v$ the increase is as expected; its magnitude is comparable to the additional contribution from the nonlinear susceptibility (Z_1^3-effects, higher order Born terms).

New: Above the velocity, where plasmon excitation starts, dE/dx is diminished, which is not expected from the extrapolation of static local field corrections.

2 Heavy (partially ionized) ions and effective charges

2.1 Ionization degree vs. effective charge

Heavy ions with intermediate velocities are not fully stripped, but their ionization degree is

$$0 < \quad q < \quad 1$$

slow, atomic $\uparrow$ $\uparrow$ swift, highly stripped

q is the essential part of the ion form factor. It depends on v, Z_1, and to some degree on Z_2. The "effective charge theory" keeps the ansatz of linear response

$$\frac{dE}{dx} = (\gamma Z_1)^2 \frac{dE}{dx}_{|Z_1=1} .$$

The effective charge fraction γ is meant to contain the dependence on ion velocity, ion internal charge distribution (and the target).

The ion charge Qe is not observed directly, because the ion stopping effectively is between the stopping of point charges Qe and Z_1e.

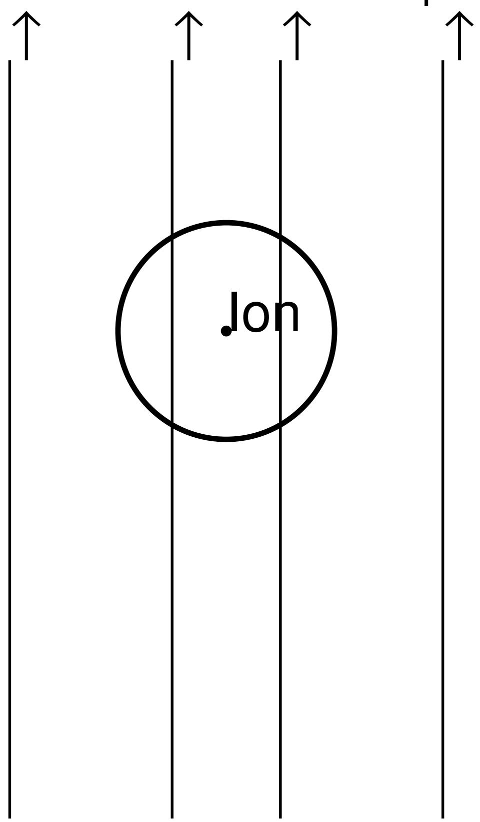
$$q < \gamma < 1$$

q: "hidden", input to theory

γ: experimental, should be the integrated theory

close, hard collisions: potential ≈ Z_1e

distant, soft collisions: potential ≈ Qe



2.2 Stripping criteria

The theme is to “explain” the ionization degree by looking at the energies (kinetic, binding, . . .) of electrons which are bound to the ion on the threshold to be stripped. The idea is elementary and old (Lamb 1940).

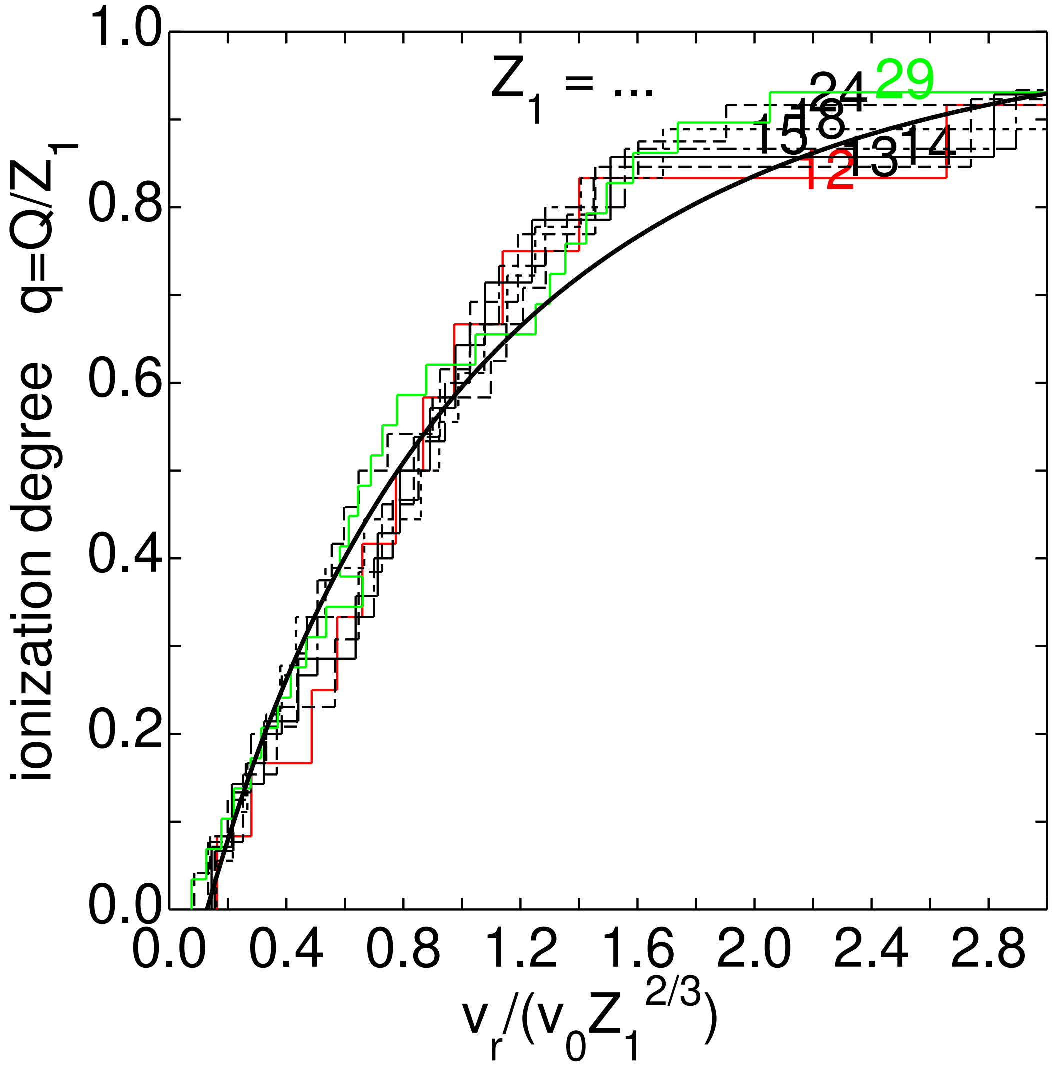
The motivation is to replace the velocity criterion, which argues that electrons with orbital velocities comparable with the ion velocity are stripped, and became the standard interpretation of effective charge theory.

(The virial theorem is a shaky bridge connecting orbital velocities with binding energies, though.)

Results of $E_{\text{HF}|Q+1} - E_{\text{HF}|Q} + \int \rho_e \Phi_{\text{ind}} d^3r = \frac{1}{2}mv^2 \frac{\tilde{N}}{\tilde{N}+1}$,

which means the energy balance of

- ionization energy
- field ionization (Stark term)
- (chemical potential, used to transform $v \longrightarrow v_r$)
- energy transfer by striking target electron — with kinetic correction



3 Training Kaneko's target model for heavy ions

3.1 proton stopping, inner shell excitation gaps

Kaneko's theory establishes dielectric functions for the inner shells of target electrons; only the top valence band and conduction band is described by "free" electrons. "All-shell" theories become essential at medium and high ion velocities, when the excitation of more and more deeply bound target electrons is switched on. They need not insert experimental proton data into the "heavy ion scaling rule."

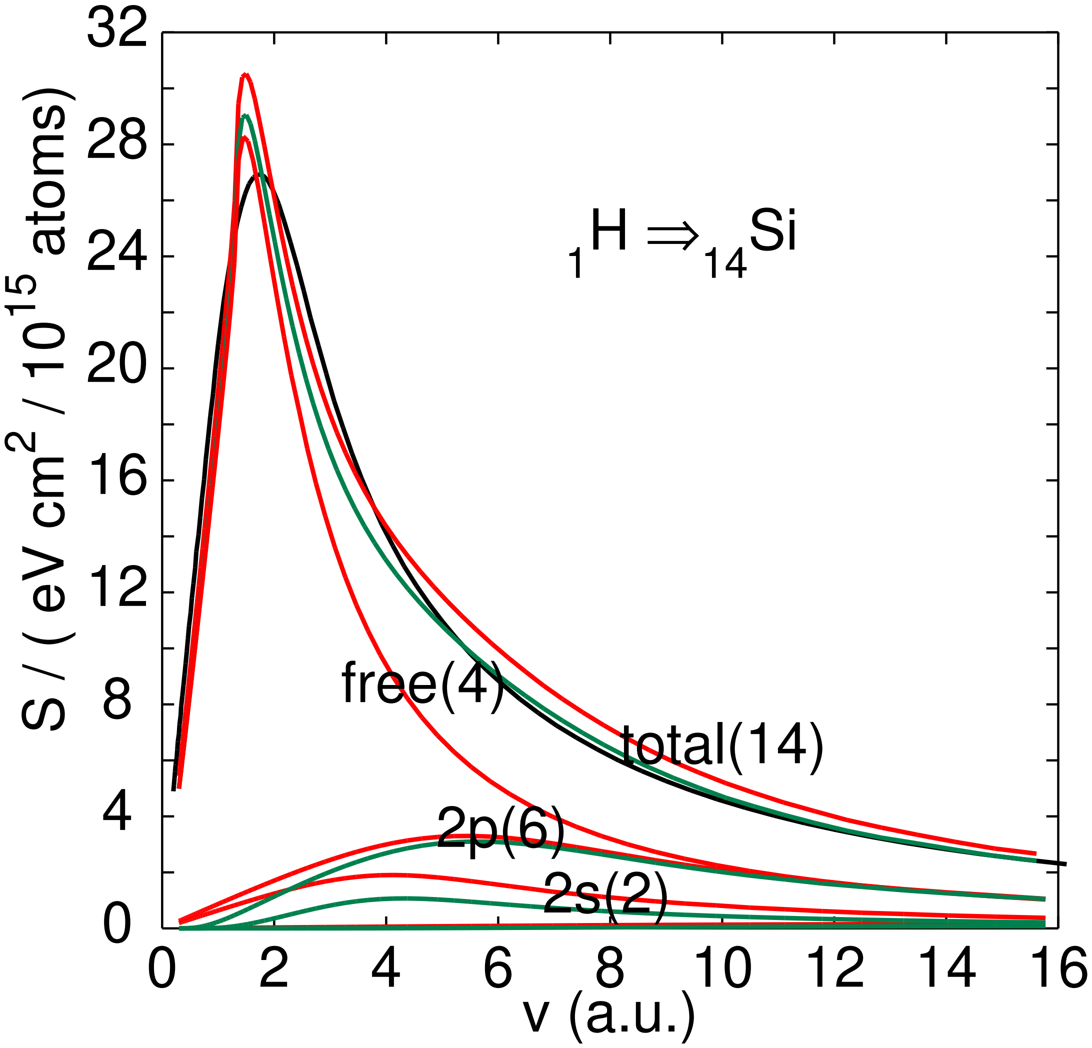
$$\text{Stopping cross section of one target atom: } S = \sum_{\text{bound,free}} S_{\text{shell}}$$

$$\text{Susceptibilities: } \chi^R_{\text{bnd}}(k,\omega) = \frac{e^2}{4\pi^3 \hbar k^2 \epsilon_0} \int d^3p \frac{w(\vec{p}+\vec{k}) - w(\vec{p})}{\omega - \hbar(\vec{p}+\vec{k})^2/(2m) + \hbar\vec{p}^2/(2m) + i\eta}.$$

with occupation probabilities $w(p) \propto \exp\{-(p/\bar{q}_{\text{bnd}})^2\}$ instead of the "Fermi ice block" of the free electron gas.

Proton stopping "test" data are in good agreement with experimental data.

A (first) correction with respect to the excitation gaps of inner shell target electrons removes a general residual 5 – 10 % surplus from the theory at high velocities (and some artificial inner shell plasmons).



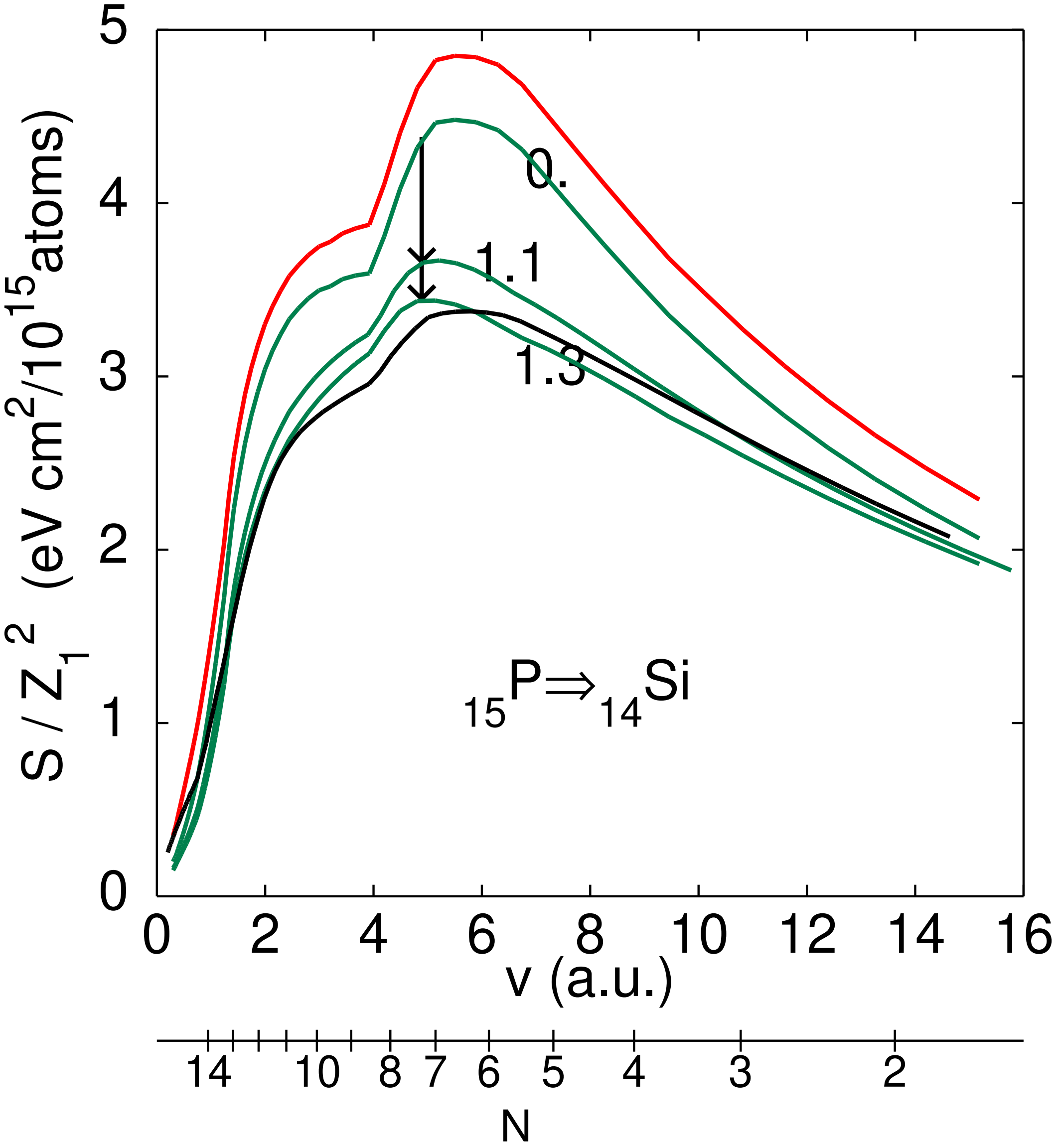
3.2 More refinements: "dead sphere", non-Gaussian momentum distributions . . .

Heavy ion electronic stopping is systematically overestimated by the original theory. The model of a non-polarizable sphere around the ion site explains and removes this discrepancy.

$$\Phi_{\text{ind}}(\vec{r}, t) =$$

$$\frac{1}{4\pi\epsilon_0} \int_{|\vec{r}' - \vec{v}t| > r_c} \frac{\rho_p(\vec{r}', t)}{|\vec{r} - \vec{r}'|} d^3r'.$$

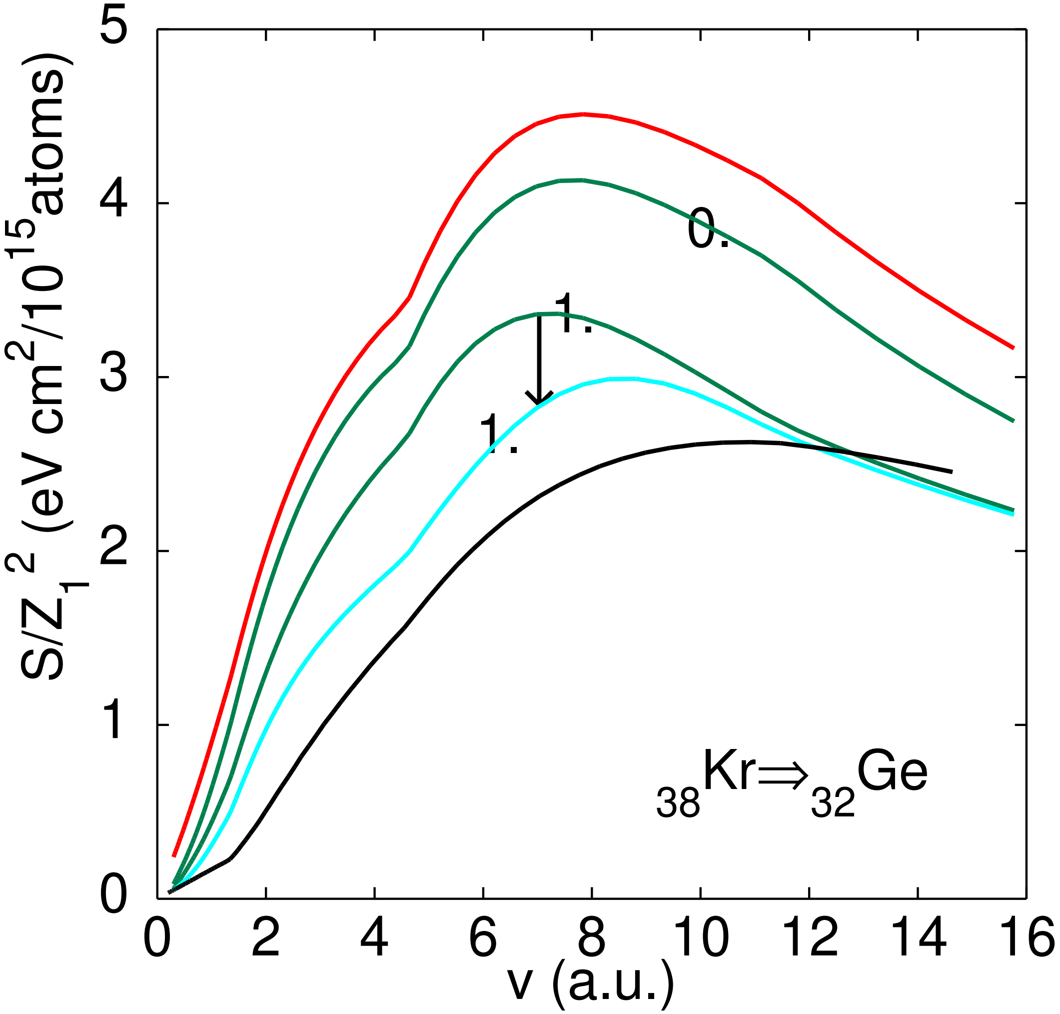
The sphere radius r_c is approximately the ion radius.



The maximum of the stopping power of heavy ions is shifted to higher velocities, if the occupation probabilities are generalized (better adapted to atomic calculations):

$$w(p) \propto p^{2l} \exp\{-(p/\bar{q}_{\mathrm{bnd}})^2\}$$

$l \ldots$ target sub-shell angular momentum quantum number. The overall change is rather small.

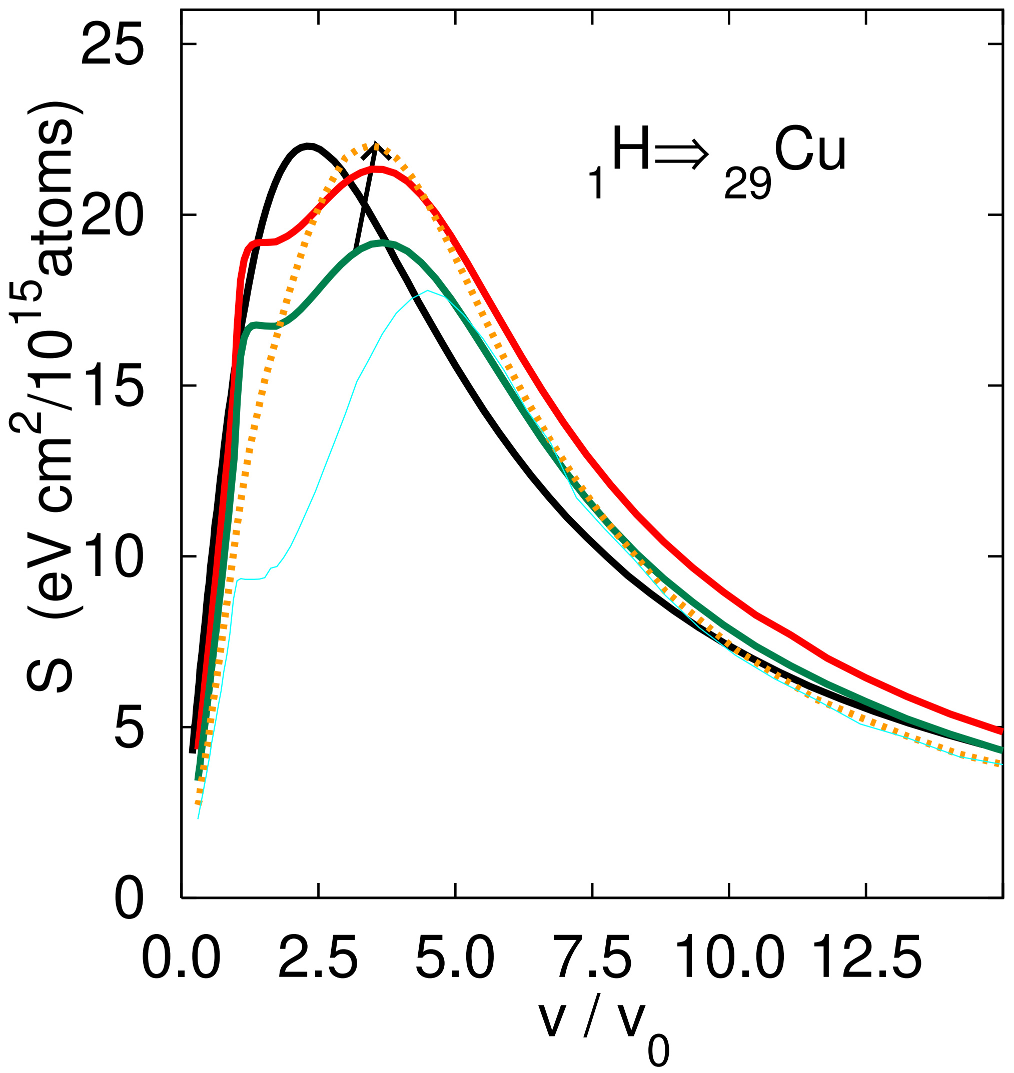


The independence of the shell by shell excitation should be replaced by a mutual (anti)screening of the polarization charges:

$$S \neq \sum_{\text{bound,free}} S_{\text{shell}}$$

▶ $\epsilon(k,\omega) = 1 + \sum \chi_{\text{shell}}(k,\omega).$

In practice, this affects transition metal targets (and light ions) and dissolves the limitation of the description of d-bands in solids.



Summary

We understand how to include results from many-particle theory to extend the free-electron-gas model of targets beyond the non-interacting approximation. It is straight-forward, but laborious. Results for the linear susceptibility must be combined with the non-linear susceptibilities from the literature.

The energy should be the quantity to knit a stripping criterion, not the fuzzy orbital velocities of bound electrons. The task of dressing *moving* bare ions with electrons in a solid has not yet been picked up from the more ab initio point of view.

The composite target model by Kaneko introduces dielectric functions to inner target shells. We may improve some specific details of the theory, but to make good use of it with heavy ions, some local correction of the target polarizability in the presence of voluminous ions is necessary.