Electronic Stopping Power of Diamond at Small Ion Velocities: Integrating the Linear Dielectric Function

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18th Werner Brandt Workshop, June 4, 1998

Abstract

We calculate the wave-vector and frequency-dependent dielectric matrix of bulk crystals by use of first-principles, all-electron Kohn-Sham states in the integral of the irreducible polarizability in the random phase approximation. From this we determine the macroscopic "head" element, the $(\mathbf{0},\mathbf{0})$-element, of the inverse matrix (the energy loss function), and integrate over energy and momentum transfers to obtain the electronic energy loss of protons at low velocities. Numerical results are given for diamond targets.

Supported by grant DAA-H04-95-1-0326 from the U.S. Army Research Office

1 Electronic Stopping

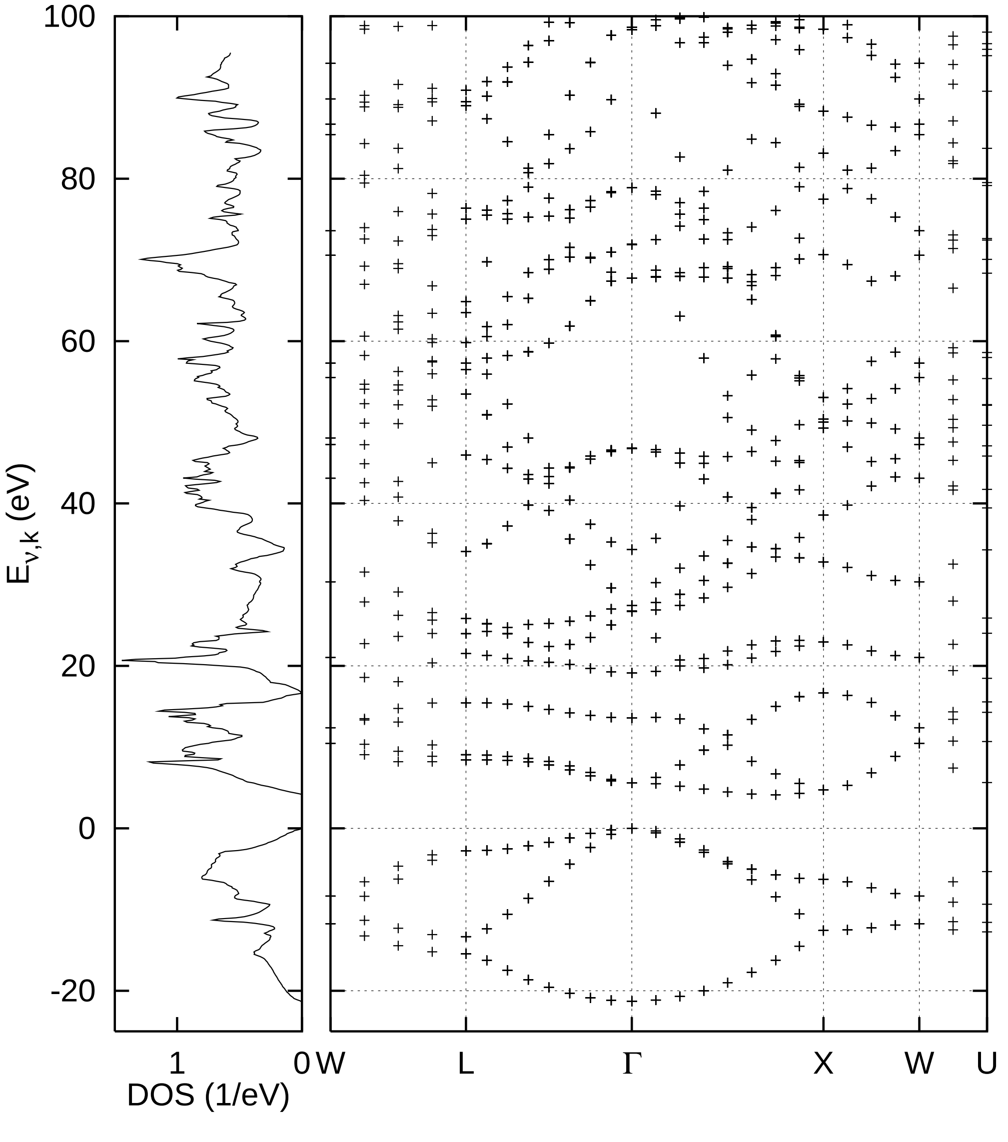
- Look at the potential $Z_1 e/(4\pi\epsilon_0 r)$ of the (bare) ion as an "external" charge that polarizes the target electrons
  - Theory for crystals: decompose the "external" field into plane waves and scatter them independently. Dielectric matrix
$$\epsilon_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega) = \delta_{\mathbf{G},\mathbf{G}'} - \frac{e^2}{\epsilon_0|\mathbf{q}+\mathbf{G}|^2}\Pi(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}',\omega).$$
    In the Random Phase Approximation (RPA) to the irreducible polarizability
$$\Pi(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}',\omega) = \sum_{\nu,\nu'}\int_{\mathrm{BZ}}\frac{d^3k}{(2\pi)^3}\frac{M^*_{\mathbf{G}}M_{\mathbf{G}'}(f_{\nu\mathbf{k}}-f_{\nu'\mathbf{k}+\mathbf{q}})}{\hbar\omega+i\eta+E_{\nu\mathbf{k}}-E_{\nu'\mathbf{k}+\mathbf{q}}}.$$
    Matrix elements
$$M_{\mathbf{G}} \equiv \langle\nu'\mathbf{k}+\mathbf{q}|e^{i(\mathbf{G}+\mathbf{q})\cdot\mathbf{r}}|\nu\mathbf{k}\rangle.$$
  - Kohn-Sham states $|\nu\mathbf{k}\rangle$ and energies $E_{\nu\mathbf{k}}$ of the GTOFF program are functions of bands ν and Bloch vectors $\mathbf{k}$.
- Compute the electric field of the polarization charges at the current position of the ion. The energy transfer to this field ("random" stopping):
$$\frac{dE}{dx} = \frac{(Z_1 e)^2}{4\pi^3\epsilon_0 v}\int d^3q\frac{\mathbf{v}\cdot\mathbf{q}}{q^2}\int_0^\infty d\omega\,\Im\epsilon^{-1}_{\mathbf{00}}(\mathbf{q},\omega)\delta(\omega-\mathbf{q}\cdot\mathbf{v})$$

2 Challenge

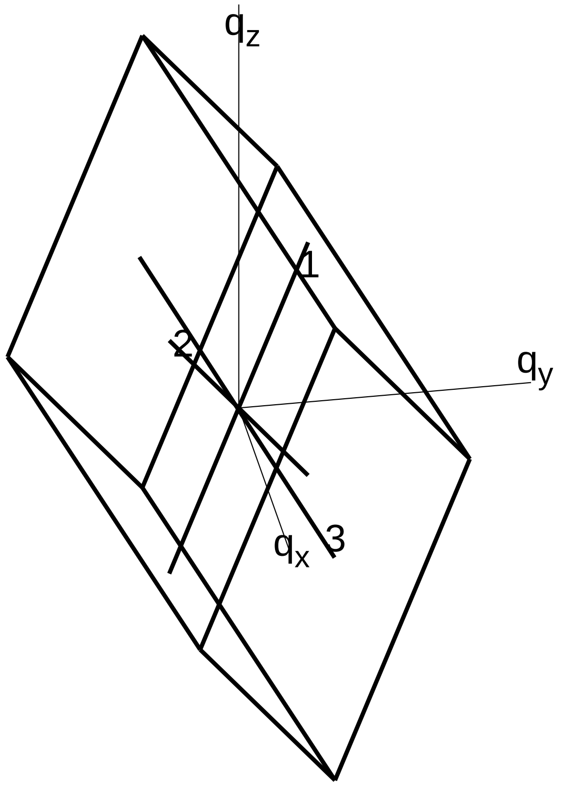
- Long-range Coulomb potential of the ion: Compute $\Pi(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}',\omega)$ for virtually all $\mathbf{q}$
- Include a broad range of energies $\hbar\omega$ and typically dozens of band pairs (ν,ν')
- Compute $\epsilon_{\mathbf{G},\mathbf{G}'}$ for virtually all row- and column indexes and build the inverse (local field effects)

…with finite computing resources.

3 Diamond



KS band-structure $E_{\nu,\mathbf{k}}$ of diamond using $16 \times 16 \times 16$ **k**-points in the BZ (213 in the IBZ). TOV at 0 eV. The direct gap at Γ is 5.59 eV, the indirect gap 4.13 eV. The two bands with four K electrons at -263 eV are not shown.



The smaller values of $S(v)$ in the following Fig. 1 refer to an integration over a parallelepiped in $\mathbf{q}$-space as shown. The larger values of $S(v)$ refer to an integration over a larger and more symmetrical volume in $\mathbf{q}$-space by applying

$$\epsilon_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega) = e^{iO(\mathbf{G}-\mathbf{G}')\cdot\mathbf{w}}\epsilon_{O\mathbf{G},O\mathbf{G}'}(O\mathbf{q},\omega),$$

with $\{O|\mathbf{w}\}$ any of the 24 point operations of the point group. The difference yields an estimate of the error that stems from the truncation inherent in the numerical approximation over the integral over d^3q.

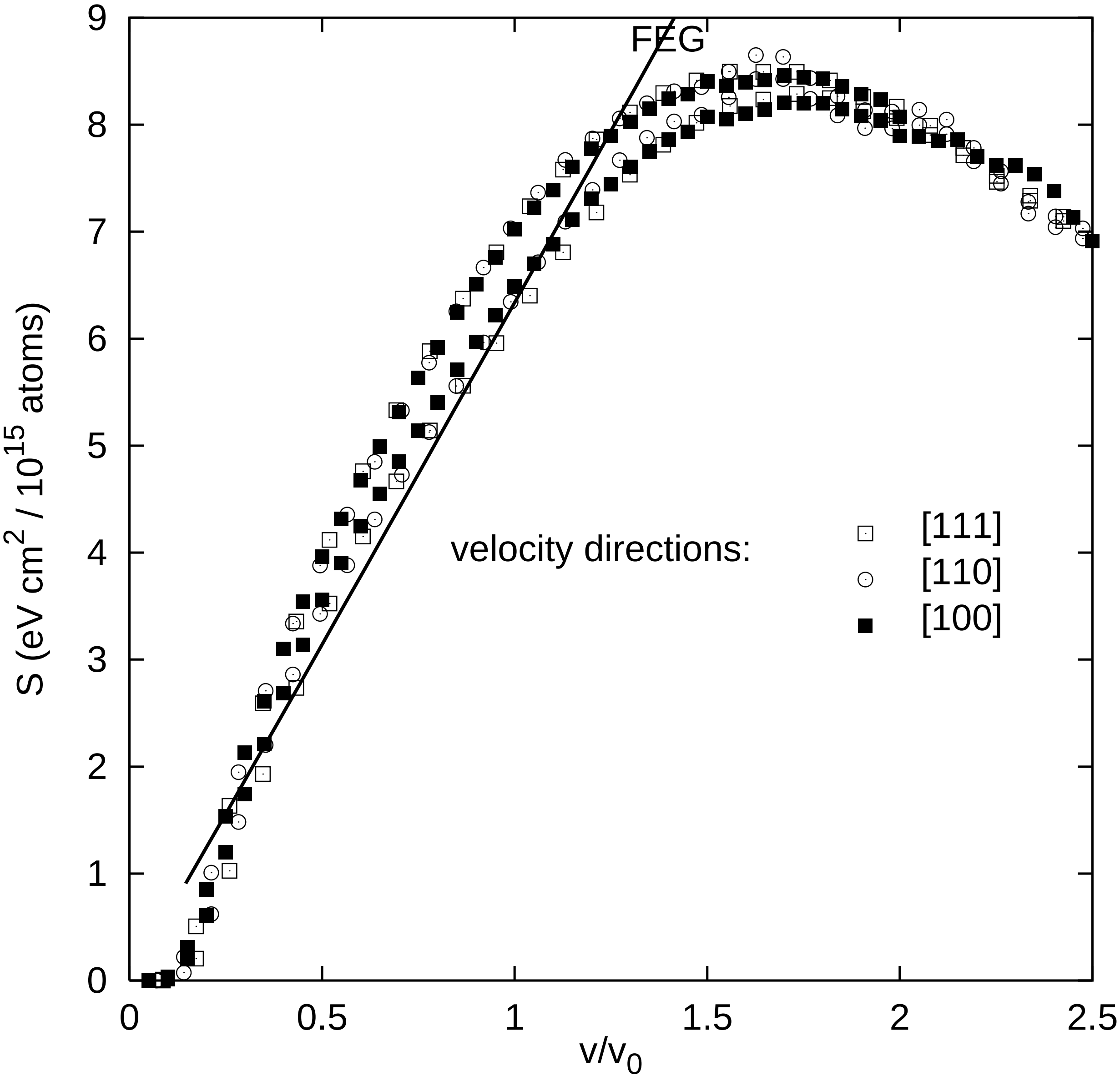


Figure 1: Electronic stopping cross section of diamond. The stopping power was integrated on a $30\times30\times30$ mesh in **q**-space (parallelepiped $6.1\times6.1\times6.1a_0^{-3}$) and on a mesh with 72 points on the ω-axis ($0\ldots103$ eV). The lowest 28 bands were included in the sum over band-pairs. The line refers to the free electron gas with a density equivalent to 4 electrons per diamond atom.

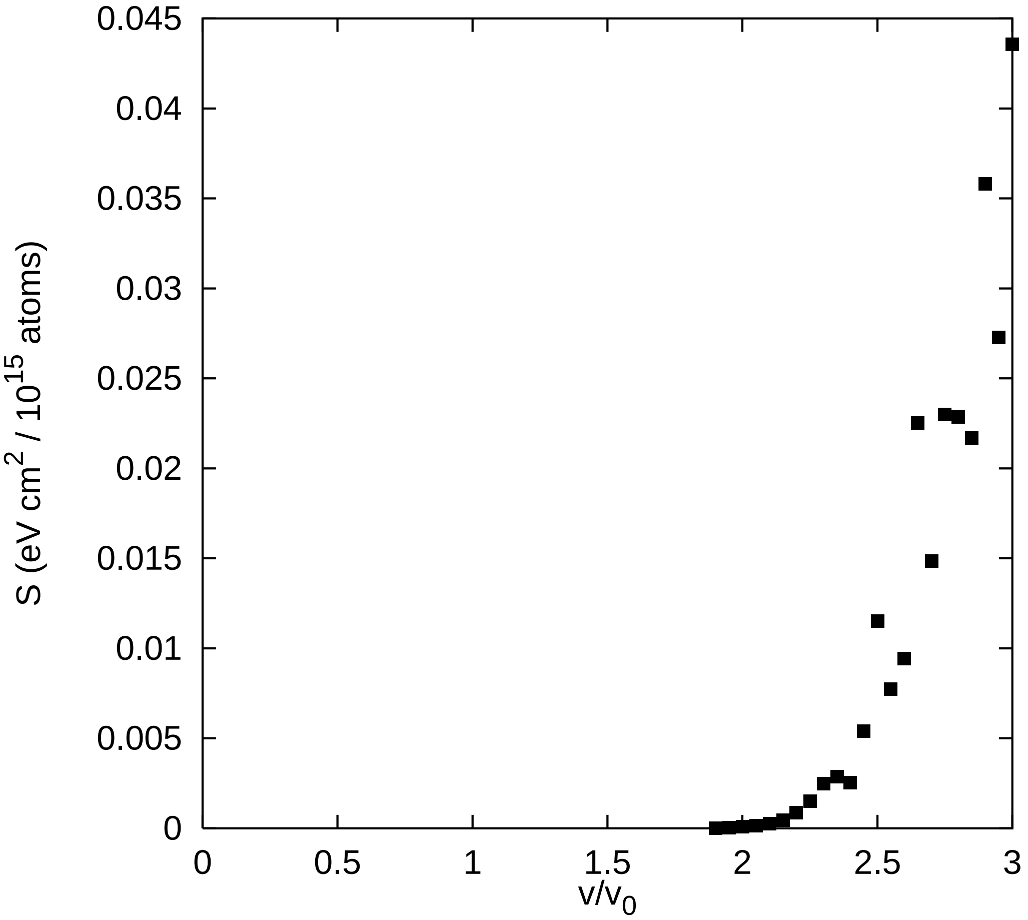


Figure 2:

Electronic stopping cross section of K-shell electrons. The stopping power was integrated on a $30 \times 30 \times 30$ mesh in $\mathbf{q}$-space (parallelepiped $6.1 \times 6.1 \times 6.1 a_0^{-3}$) and on a mesh with 126 points on the ω-axis ($0 \ldots 343$ eV). Excitations of K-electrons to the lowest unoccupied 28 bands were included in the sum over band-pairs.

Threshold velocity: $\frac{1}{2} v_0 \sqrt{\hbar\omega_g / E_0}$, with $v_0 \ldots$ Bohr velocity, $E_0 \ldots 1$ rydberg, $\hbar\omega_g \ldots$ excitation gap.

4 What is missing?

- Non-Linearities, $S \propto Z_1^{\geq 3}$
- dynamic charge fraction of the $p \leftrightarrow$H–system

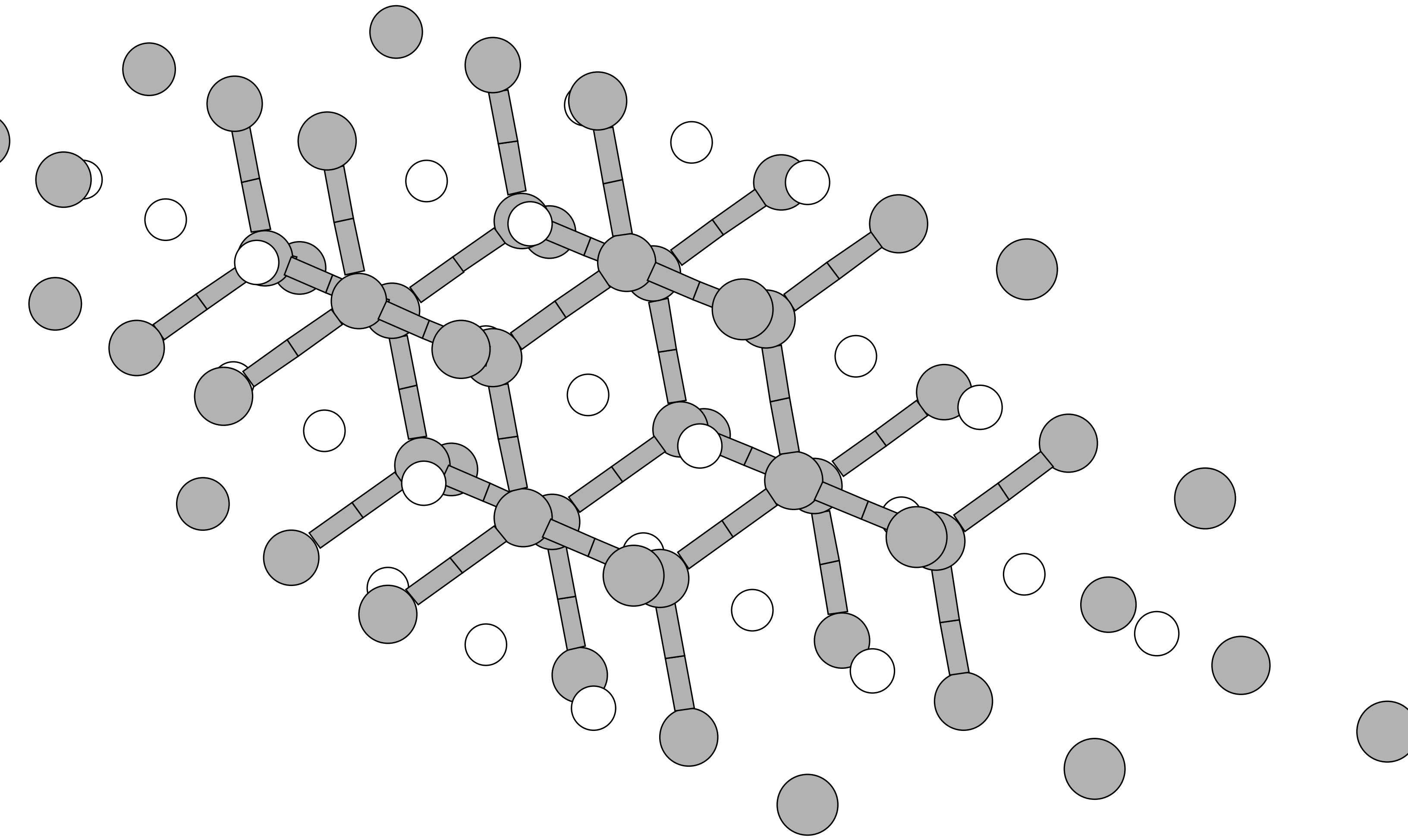


Figure 3: An artificial material created by loading each primitive unit cell of diamond with one hydrogen atom, placed at the inversion center on the rotation axis of order 3 along [111]. Formula unit C_2H. C and C–C bonds are grey, H are white.

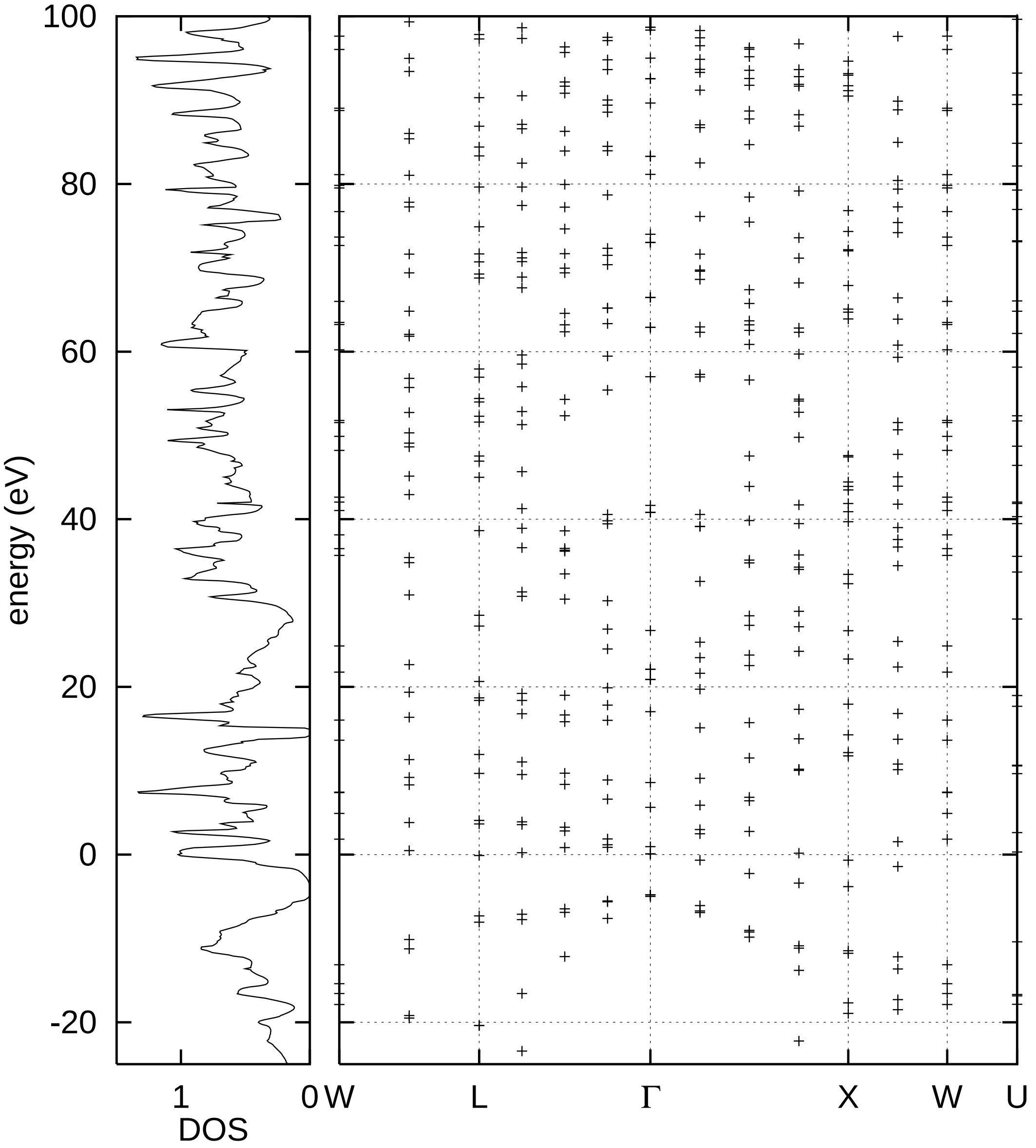
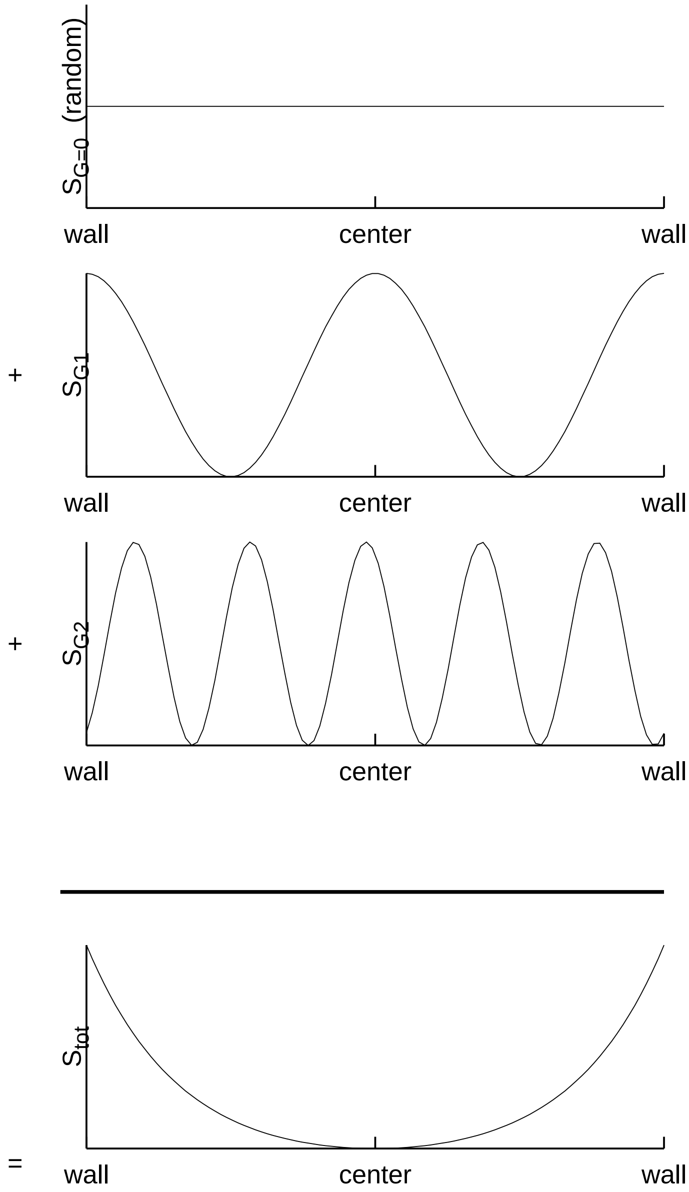


Figure 4:

KS band-structure $E_{\nu,\mathbf{k}}$ of the artificial C_2H (Fermi level at 0 eV). This conductor would probably exhibit the linear $S(v) \propto v$ behavior at small v.

• Channeling



The dielectric theory of the electronic stopping power generally includes a resolution of the position of the ion ("channeling") by interference, $S(\mathbf{r}) = \sum_{\mathbf{G}\perp\mathbf{v}} S_{\mathbf{G}}(\mathbf{r})$, but only the $\mathbf{G} = \mathbf{0}$ ("random") term has been computed here.

5 Things To Do

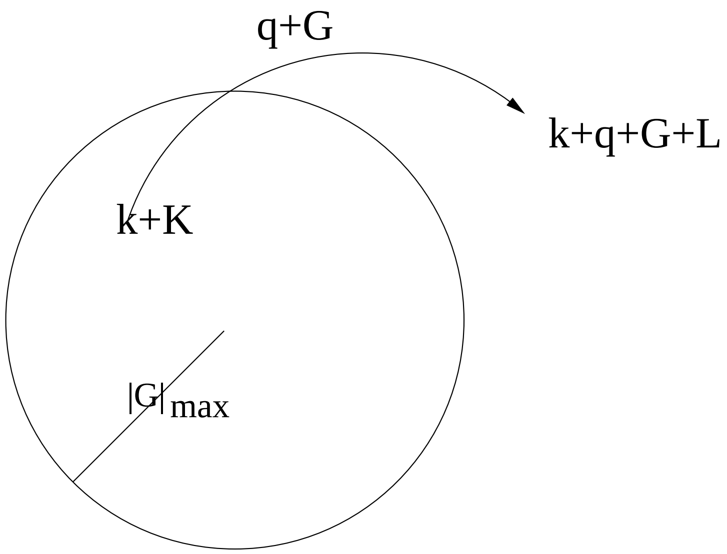
- Here: intermediate plane wave representation

$$M_{\mathbf{G}} = \frac{1}{V_{\mathrm{UC}}} \sum_{\mathbf{K}} u^*_{\nu',\mathbf{k}+\mathbf{q}+\mathbf{G},\mathbf{K}} u_{\nu,\mathbf{k},\mathbf{K}} \quad ,$$

with

$$\langle \mathbf{r}|\nu\mathbf{k}\rangle \equiv \sum_{\mathbf{G}} e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}} u_{\nu,\mathbf{k},\mathbf{G}}$$

that artificially reduces $M_{\mathbf{G}}$ for large $\hbar\mathbf{q}$ because the $u_{\nu,\mathbf{k},\mathbf{G}}$ are tabulated just to a max $|\mathbf{G}|$.



An optional evaluation of matrix elements $M_{\mathbf{G}}$ in real space is wishful to treat localized band states (core electrons) accurately: Fig. 2 is only qualitatively correct.

- The finite basis set of GTO's (s, p, …functions) in GTOFF implies that bands are missing in the region $E_{\nu\mathbf{k}} \gg 0$. The letter to Santa Claus includes
  - Green's function approach that replaces the sum over conduction bands by the construction of "polarization" functions, or
  - Completion of the GTO basis by one more diagonalization of the SCF Hamiltonian with orthogonalized plane waves.