Modeling of inelastic interactions of fast charged particles in condensed matter

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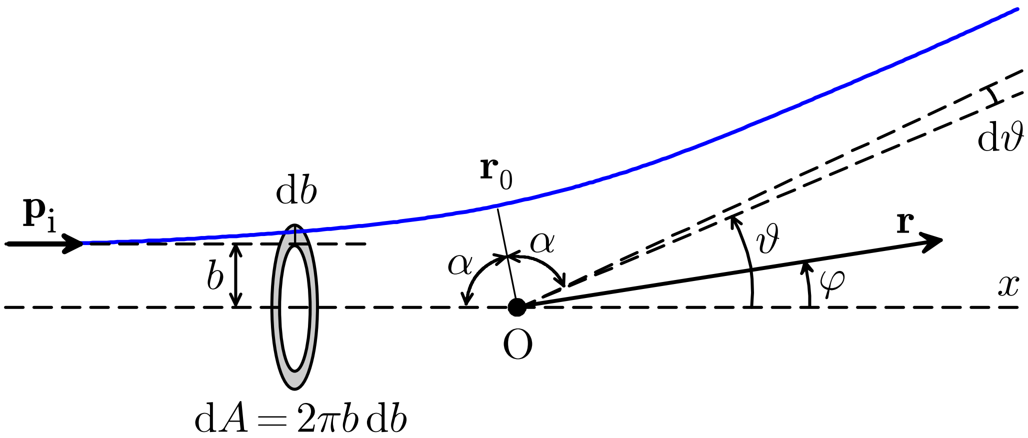
Stopping theory (historical perspective)

- **Rutherford (1911)** Scattering by a Coulomb potential $Z_1 Z_2 e/r$
  Projectile particles: mass M_1 and charge $Z_1 e$



$$\tan(\theta/2) = \frac{Z_1 Z_2 e^2}{\mathrm{m_e} v^2\, b}$$

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \left(\frac{Z_1 Z_2 e^2}{M_1 v^2}\right)^2 \frac{1}{\sin^4(\theta/2)}$$

Rutherford, E. (1911), "LXXIX. The scattering of α and β particles by matter and the structure of the atom," Phil. Mag. S. 6 21:125, 669-688.

- **Thomson (1912)** Collisions of charged particles with free electrons at rest

$$W(b) = W_{\max} \frac{(Z_1 e^2/\mathrm{m_e} v^2)^2}{b^2 + (Z_1 e^2/\mathrm{m_e} v^2)^2} \qquad W_{\max} = \frac{2\mathrm{m_e} v^2}{(1 + \mathrm{m_e}/M_1)^2}$$

$$\frac{\mathrm{d}\sigma_1}{\mathrm{d}W} = \frac{2\pi (Z_1 e^2)^2}{M_2 v^2} \frac{1}{W^2}$$

Thomson, J. J. (1912), "XLII. Ionization by moving electrified particles," Phil. Mag. Series 6 23, 449-457.

Stopping theory (historical perspective)

❑ **Bohr (1913)** Classical stopping theory (only electrons contribute)

Homogeneous material of "atomic number" Z_2 with $\mathcal{N} = \dfrac{N_A \rho}{A_w}$ atoms per unit volume

- Close collisions ($b < a$) treated as classical binary collisions

$$\left[-\frac{\mathrm{d}E}{\mathrm{d}z}\right]_{b<a} = \frac{4\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} Z_2 \ln\left(a \frac{\mathrm{m_e} v^2}{|Z_1|\, e^2}\right)$$

- Distant interactions ($b > a$) electrons respond as classical oscillators with characteristic (angular) frequency ω . The oscillator strength $\mathrm{d}f(\omega)/\mathrm{d}\omega$ is defined as the number of oscillators (electrons) per unit frequency

$$\int_0^\infty \frac{\mathrm{d}f(\omega)}{\mathrm{d}\omega}\,\mathrm{d}\omega = Z_2 \qquad f \text{ sum rule}$$

$$\left[-\frac{\mathrm{d}E}{\mathrm{d}z}\right]_{b>a} = \frac{4\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} Z_2 \left\{\ln\left(\frac{1.123\, v}{a\overline{\omega}}\right) + \frac{1}{2}\ln\left(\frac{1}{1-\beta^2}\right) - \frac{1}{2}\beta^2\right\}$$

$$Z_2 \ln\overline{\omega} = \int_0^\infty \ln(\omega)\,\frac{\mathrm{d}f(\omega)}{\mathrm{d}\omega}\,\mathrm{d}\omega$$

$$\boxed{S \equiv -\frac{\mathrm{d}E}{\mathrm{d}s} = \frac{4\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} Z_2 \left[\ln\left(\frac{1.123\,\mathrm{m_e} v^3}{|Z_1|\, e^2\, \overline{\omega}}\right) + \frac{1}{2}\ln\left(\frac{1}{1-\beta^2}\right) - \frac{1}{2}\beta^2\right]}$$

Bohr, N. (1913), "On the theory of the decrease of velocity of moving electrified particles on passing through matter," Phil. Mag. 26, 1–25.

Stopping theory (historical perspective)

- **Lindhard (1954)** Classical dielectric theory

The material is characterized by its complex **dielectric functions** (DF)

longitudinal $\epsilon^{(\mathrm{L})}(q,\omega)$ and transverse $\epsilon^{(\mathrm{T})}(q,\omega)$; q = wave number, ω = ang. frequency

$$\epsilon^{(\mathrm{L})}(0,\omega) = \epsilon^{(\mathrm{T})}(0,\omega) = \epsilon(\omega)$$ Optical dielectric function (ODF)

DFs available only for a degenerate electron gas (Lindhard, Mermin), complicated analytical expressions

The DFs satisfy various sum rules (implied by the causality principle)

$$\int_0^\infty \omega \, \mathrm{Im}\left[\epsilon^{(\mathrm{L})}(q,\omega)\right] \mathrm{d}\omega = \frac{\pi}{2}\,\omega_\mathrm{p}^2, \qquad \omega_\mathrm{p} = \sqrt{4\pi\mathcal{N}Z_2e^2/\mathrm{m_e}}$$

Kramers-Kronig relation: $$\mathrm{Re}\left[\frac{1}{\epsilon^{(\mathrm{L,T})}(q,\omega)}\right] = 1 - \frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{\omega'}{\omega'^2-\omega^2}\,\mathrm{Im}\left[\frac{1}{\epsilon^{(\mathrm{L,T})}(q,\omega)}\right] \mathrm{d}\omega'$$

The swift charged projectile "polarizes" the medium, creating an induced electric field that acts back on the projectile (stopping force)

$$S = \frac{2(Z_1e)^2}{\pi v^2}\int_0^\infty \omega\,\mathrm{d}\omega \int_{\omega/v}^\infty \frac{\mathrm{d}q}{q}\left[\mathrm{Im}\left(\frac{-1}{\epsilon^{(\mathrm{L})}(q,\omega)}\right)\right.$$
$$\left. +\beta^2\left(1-\frac{\omega^2}{\beta^2c^2q^2}\right)\mathrm{Im}\left(\frac{1}{1-(\omega/cq)^2\,\epsilon^{(\mathrm{T})}(q,\omega)}\right)\right]$$

Lindhard, J. (1954), "On the properties of a gas of charged particles," Dan. Mat. Fys. Medd. 28, 1–57.

Stopping theory (historical perspective)

- **Fermi (1940), Sternheimer (1952)** Density (polarization) effect

In the case of a rarefied material, $\epsilon_1^{(\mathrm{L,T})} \simeq 1$, $\epsilon_2^{(\mathrm{L,T})} \simeq 0$ and

$$S_{\mathrm{unpol}} = \frac{2(Z_1 e)^2}{\pi v^2} \int_0^\infty \omega \, \mathrm{d}\omega \int_{\omega/v}^\infty \frac{\mathrm{d}q}{q} \left[\mathrm{Im} \left(\frac{-1}{\epsilon^{(\mathrm{L})}(q,\omega)} \right) \right.$$

$$\left. + \left(\beta^2 - \frac{\omega^2}{c^2 q^2} \right) \frac{(cq/\omega)^2}{\left[(cq/\omega)^2 - 1\right]^2} \mathrm{Im} \left(\frac{-1}{\epsilon^{(\mathrm{T})}(q,\omega)} \right) \right]$$

The difference is the Fermi density- or polarization-effect correction

$$(\Delta S)_{\mathrm{pol}} \equiv S - S_{\mathrm{unpol}} = \frac{2\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} \, Z_2 \, \delta_{\mathrm{F}}$$

Naturally included in the dielectric formalism

NB: Atomic first principles calculations provide the equivalent to S_{unpol}, that is, aggregation effects should be considered separately

Fermi, E. (1940), “The ionization loss of energy in gases and in condensed materials,” Phys. Rev. 57, 485-493.
Sternheimer, R. M. (1952), “The density effect for the ionization loss in various materials,” Phys. Rev. 88, 851-859.

Kinematics of inelastic collisions

Projectile: mass M_1 and charge Z_1
kinetic energy E and momentum $\mathbf{p}$

$$\beta = \frac{v}{c} = \frac{\sqrt{E(E+2M_1c^2)}}{E+M_1c^2}$$

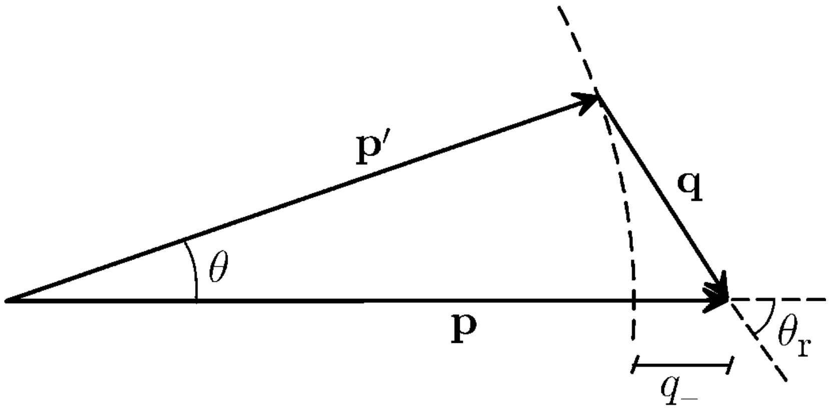
Relativistic kinematics:

$$\gamma = \frac{1}{\sqrt{1-\beta^2}} = \frac{E+M_1c^2}{M_1c^2}$$

$$E(E+2M_1c^2) = c^2p^2$$

Effect of individual collisions *on the projectile*:

- **Energy loss**: $W = E - E'$
- momentum transfer: $\mathbf{q} = \mathbf{p} - \mathbf{p}'$
- **Angular deflection**: $\cos\theta$



Fano (1963) instead of the scattering angle uses the *recoil energy* Q

$$Q(Q+2\mathrm{m_e}c^2) = c^2q^2 = c^2\left(p^2 + p'^2 - 2pp'\cos\theta\right) \qquad Q_{\mathrm{n.r.}} = \frac{q^2}{2\mathrm{m_e}}$$

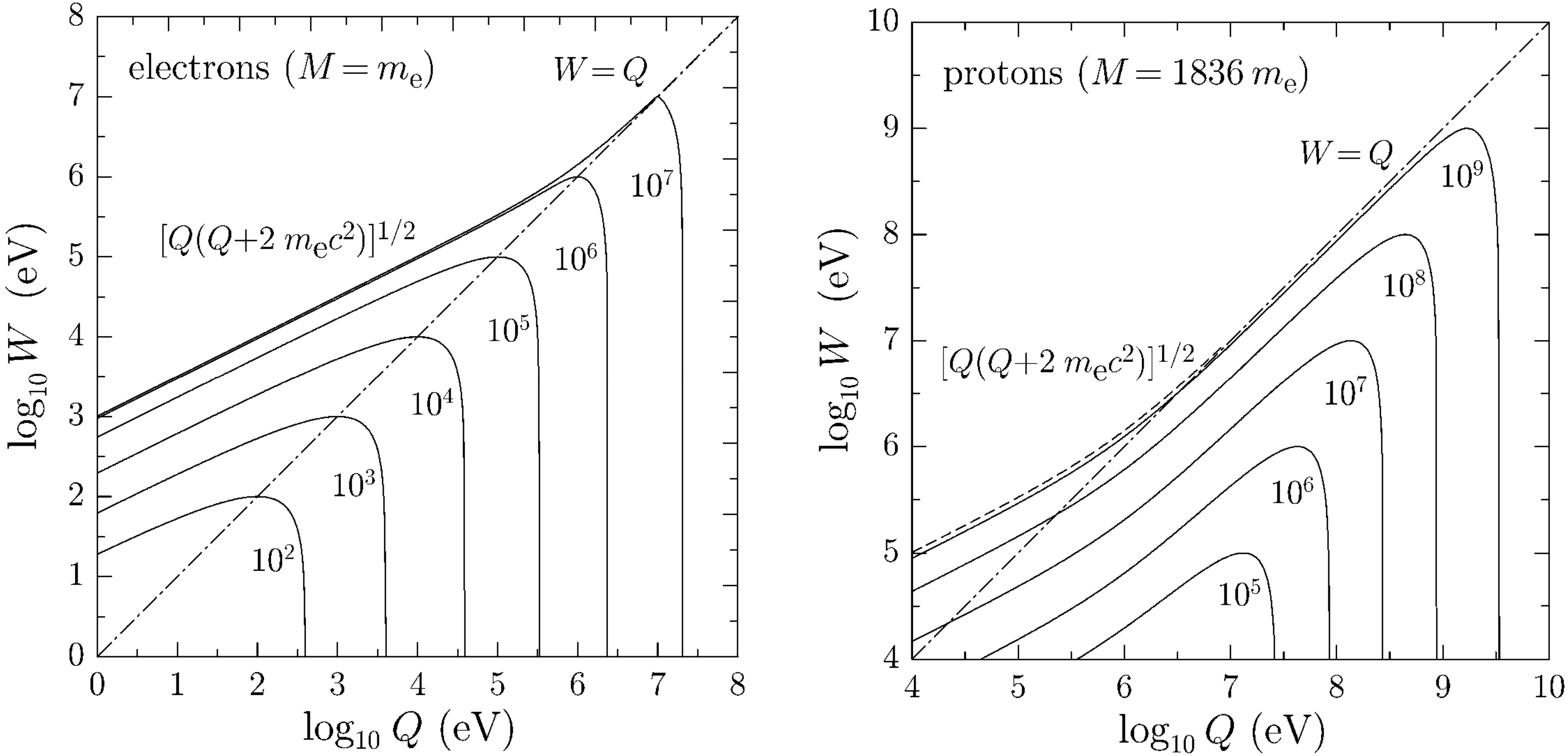
which can take values in the interval

$$Q_\pm = \sqrt{\left[\sqrt{E(E+2M_1c^2)} \pm \sqrt{(E-W)(E-W+2M_1c^2)}\right]^2 + \mathrm{m_e^2}c^4} - \mathrm{m_e}c^2$$

Kinematics of inelastic collisions

For a given Q, the energy loss may take values from 0 to

$$W_{\mathrm{m}}(Q) = E + M_1 c^2 - \sqrt{\left[\sqrt{E(E+2M_1c^2)} - \sqrt{Q(Q+2\mathrm{m_e}c^2)}\right]^2 + M_1^2 c^4}$$



For small Q: $$W_{\mathrm{m}}(Q) \simeq \beta\sqrt{Q(Q+2\mathrm{m_e}c^2)}, \qquad W \ll E$$

Stopping theory (historical perspective)

- **Bethe (1932), Fano (1963)** Plane-wave Born approximation for collisions with atoms
  First-order perturbation calculation, projectile plane waves. Atomic DCS

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}W\,\mathrm{d}Q} = \frac{2\pi Z_1^2 e^4}{\mathrm{m_e} v^2}\left[\frac{2\mathrm{m_e}c^2}{WQ(Q+2\mathrm{m_e}c^2)}\left\{\frac{(2E-W+2\mathrm{m_e}c^2)^2 - Q(Q+2\mathrm{m_e}c^2)}{4\,(E+\mathrm{m_e}c^2)^2}\right\}\frac{\mathrm{d}f(Q,W)}{\mathrm{d}W}\right.$$
$$\left. + \frac{2\mathrm{m_e}c^2 W}{[Q(Q+2\mathrm{m_e}c^2)-W^2]^2}\left(\beta^2\sin^2\theta_\mathrm{r} + \left\{\frac{Q(Q+2\mathrm{m_e}c^2)-W^2}{2(E+\mathrm{m_e}c^2)^2}\right\}\right)\frac{\mathrm{d}g(Q,W)}{\mathrm{d}W}\right]$$

where θ_r is the recoil angle (between $\mathbf{q}$ and $\mathbf{p}$)

$$\cos\theta_\mathrm{r} = \frac{W}{\beta(cq)}\left(1+\frac{(cq)^2-W^2}{2W(E+\mathrm{m_e}c^2)}\right)$$

- Longitudinal Generalized Oscillator Strength (GOS). Sum of contributions of subshell GOSs

$$\frac{\mathrm{d}f_a(Q,W)}{\mathrm{d}W} = \frac{W2(Q+\mathrm{m_e}c^2)}{Q(Q+2\mathrm{m_e}c^2)}\frac{k_b}{(E_{n_a\kappa_a}+W)\pi}\sum_{m_a}\sum_{\kappa_b,m_b}\left|\left\langle\psi_{E_b\kappa_b m_b}\middle|\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r})\middle|\psi_{n_a\kappa_a m_a}\right\rangle\right|^2$$

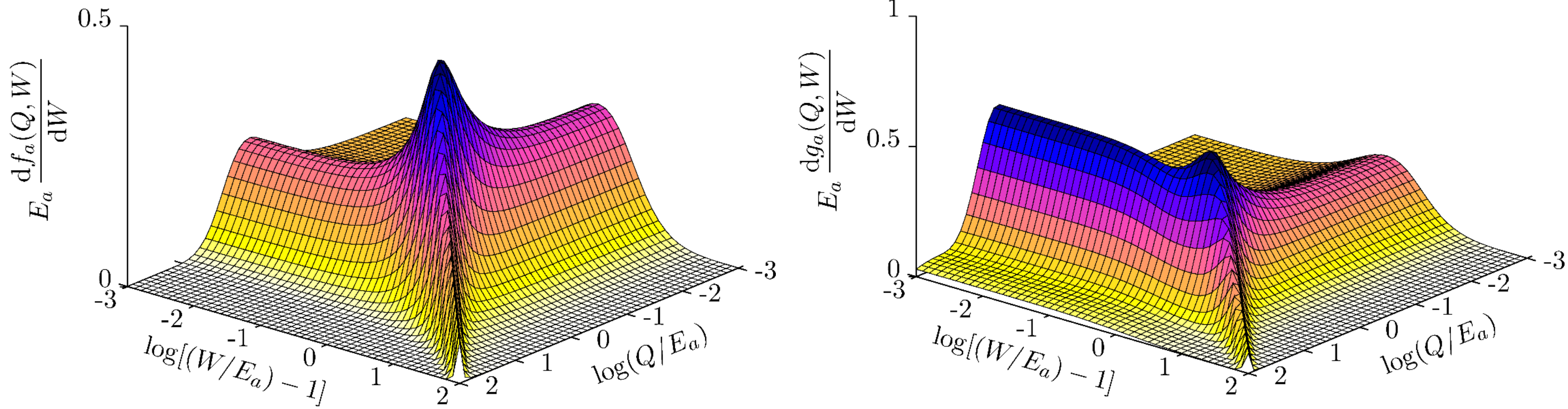
- Transverse Generalized Oscillator Strength (TGOS)

$$\frac{\mathrm{d}g_a(Q,W)}{\mathrm{d}W} = \frac{2(Q+\mathrm{m_e}c^2)}{W}\frac{k_b}{(E_{n_a\kappa_a}+W)\pi}\sum_{m_a}\sum_{\kappa_b,m_b}\left|\left\langle\psi_{E_b\kappa_b m_b}\middle|\widetilde{\alpha}_x\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r})\middle|\psi_{n_a\kappa_a m_a}\right\rangle\right|^2$$

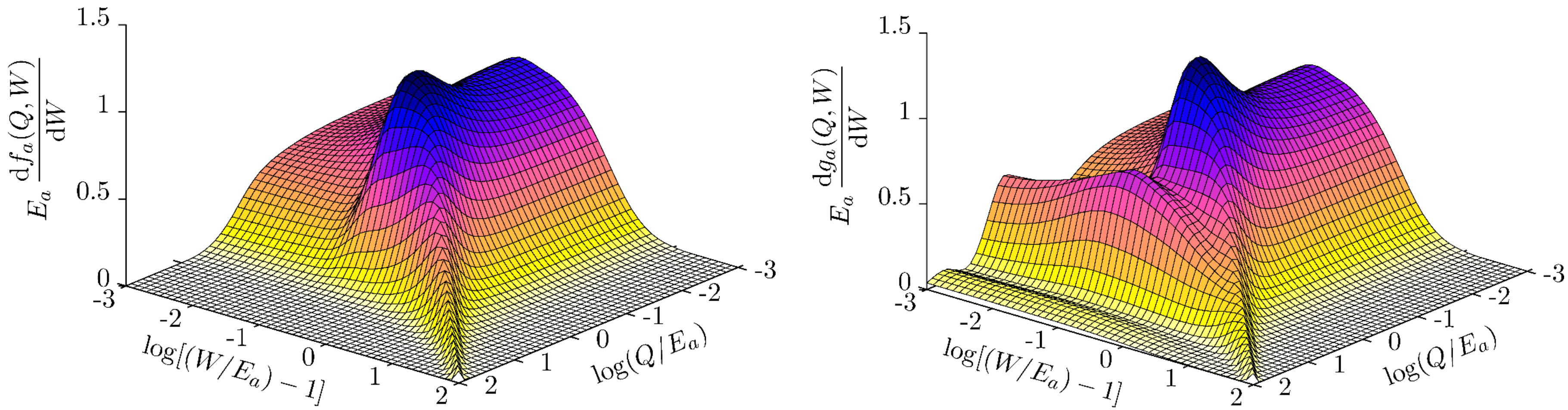
Bethe, H. A. (1932), "Bremsformel für Elektronen relativistischer Geschwindigkeit," Z. Physik 76, 293-299.
Fano, U. (1963), "Penetration of protons, alpha particles and mesons," Ann. Rev. Nucl. Sci. 13, 1-66.

Calculated GOSs

Ne, K shell



Ag, M1 shell



Bote, D. and F. Salvat (2008), "Calculations of inner-shell ionization by electron impact with the distorted-wave and plane-wave Born approximations," Phys. Rev. A 77, 042701.

Properties of the atomic GOS

- Bethe sum rule

$$\int_0^\infty \frac{\mathrm{d}f(Q,W)}{\mathrm{d}W}\mathrm{d}W = Z_2[1+\Delta(Q)]$$

The relativistic departure $\Delta(Q)$ is ~10% for the K shell of heavy elements and much smaller for outer subshells

- Optical oscillator strength

$$\frac{\mathrm{d}f(Q=0,W)}{\mathrm{d}W} = \frac{\mathrm{d}g(Q=0,W)}{\mathrm{d}W} \equiv \frac{\mathrm{d}f(W)}{\mathrm{d}W}$$

- Relationship with the atomic photoeffect (dipole approximation)

$$\frac{\mathrm{d}f_a(Q=0,W)}{\mathrm{d}W} = \frac{\mathrm{m_e}c}{2\pi^2 e^2 \hbar}\,\sigma_a^{\mathrm{ph,dip}}(W)$$

and

$$\frac{\mathrm{d}g_a(Q_{\mathrm{ph}},W)}{\mathrm{d}W} = \frac{\sqrt{\mathrm{m_e^2}c^4+W^2}}{2\pi^2 e^2 c\hbar}\,\sigma_a^{\mathrm{ph}}(W)$$

where

$$Q_{\mathrm{ph}} = \sqrt{\mathrm{m_e^2}c^4+W^2} - \mathrm{m_e}c^2$$

is the recoil energy of the photon line

Macroscopic quantities

- Energy-loss DCS: $$\frac{\mathrm{d}\sigma}{\mathrm{d}W} = \int_{Q_-}^{Q_+} \frac{\mathrm{d}^2\sigma}{\mathrm{d}W\,\mathrm{d}Q}\,\mathrm{d}Q$$
- Total cross section: $$\sigma = \int_0^{W_{\max}} \frac{\mathrm{d}\sigma}{\mathrm{d}W}\,\mathrm{d}W$$
- Stopping cross section: $$\sigma_{\mathrm{st}} = \int_0^{W_{\max}} W\,\frac{\mathrm{d}\sigma}{\mathrm{d}W}\,\mathrm{d}W$$

❑ Consider a material (gas) with $\mathcal{N}$ atoms per unit volume

- Double-differential inverse mean free path: $$\mu(Q,W) = \mathcal{N}\,\frac{\mathrm{d}^2\sigma}{\mathrm{d}W\,\mathrm{d}Q}$$
- Energy-loss DIMFP: $$\mu(W) = \int_{Q_-}^{Q_+} \mu(Q,W)\mathrm{d}Q = \mathcal{N}\,\frac{\mathrm{d}\sigma}{\mathrm{d}W}$$
- IMFP: $$\mu = \int_0^{W_{\max}} \mu(W)\mathrm{d}W = \mathcal{N}\,\sigma = \lambda^{-1}$$
- Stopping power: $$S = \int_0^{W_{\max}} W\mu(W)\mathrm{d}W = \mathcal{N}\,\sigma_{\mathrm{st}}$$

Semiclassical approximation

- Consider the stopping power obtained from the dielectric formalism and introduce the interpretation:

$\hbar q \to q$ momentum transfer, and $\hbar\omega \to W$ energy loss

in individual interactions

- Introduce the variables $Q(Q+2\mathrm{m_e}c^2) = (cq)^2$ and W and write the stopping power as

$$S = \frac{2(Z_1 e)^2}{\pi \hbar^2 v^2} \int_0^{W_{\max}} \mathrm{d}W\, W \int_{Q_-}^{Q_+} \mathrm{d}Q\, (Q+\mathrm{m_e}c^2) \left[\frac{1}{Q(Q+2\mathrm{m_e}c^2)} \,\mathrm{Im}\left(\frac{-1}{\epsilon^{(\mathrm{L})}}\right) \right.$$

$$\left. + \left(\beta^2 - \frac{W^2}{Q(Q+2\mathrm{m_e}c^2)}\right) \frac{W^2[(\epsilon_1^{(\mathrm{T})})^2 + (\epsilon_2^{(\mathrm{T})})^2]}{[Q(Q+2\mathrm{m_e}c^2) - W^2\,\epsilon_1^{(\mathrm{T})}]^2 + W^4(\epsilon_2^{(\mathrm{T})})^2} \,\mathrm{Im}\left(\frac{-1}{\epsilon^{(\mathrm{T})}}\right) \right]$$

where $\epsilon^{(\mathrm{L,T})} = \epsilon_1^{(\mathrm{L,T})} + \mathrm{i}\,\epsilon_2^{(\mathrm{L,T})}$ and

$$W_\mathrm{m}(Q) = \beta\sqrt{Q(Q+2\mathrm{m_e}c^2)} = qv$$

is the maximum allowed energy loss (for collisions with $W \ll E$)

- Compare with

$$S_\mathrm{unpol} = \mathcal{N} \int_0^\infty \mathrm{d}Q \int_0^{W_\mathrm{m}(Q)} \mathrm{d}W\, W \,\frac{\mathrm{d}^2\sigma_\mathrm{class}}{\mathrm{d}Q\,\mathrm{d}W}$$

and identify the atomic "semiclassical" DCS

Semiclassical approximation

- and identify the semiclassical DCS:

$$\boxed{\begin{aligned}\frac{\mathrm{d}^2\sigma_{\rm class}}{\mathrm{d}W\,\mathrm{d}Q} &= \frac{1}{\mathcal{N}}\,\frac{2Z_1^2e^2}{\pi\hbar^2v^2}\,(Q+\mathrm{m_e}c^2)\left[\frac{1}{Q(Q+2\mathrm{m_e}c^2)}\,\mathrm{Im}\left(\frac{-1}{\epsilon^{\rm L}(Q,W)}\right)\right.\\ &\left.+\left(\beta^2-\frac{W^2}{Q(Q+2\mathrm{m_e}c^2)}\right)\frac{W^2[(\epsilon_1^{\rm T})^2+(\epsilon_2^{\rm T})^2]}{[Q(Q+2\mathrm{m_e}c^2)-W^2\,\epsilon_1^{\rm T}]^2+W^4(\epsilon_2^{\rm T})^2}\,\mathrm{Im}\left(\frac{-1}{\epsilon^{\rm T}(Q,W)}\right)\right]\end{aligned}}$$

- In the case of a low-density gas,

$$\begin{aligned}\frac{\mathrm{d}^2\sigma_{\rm class}}{\mathrm{d}W\,\mathrm{d}Q} &= \frac{1}{\mathcal{N}}\,\frac{2Z_1^2e^2}{\pi\hbar^2v^2}\,(Q+\mathrm{m_e}c^2)\left[\frac{1}{Q(Q+2\mathrm{m_e}c^2)}\,\mathrm{Im}\left(\frac{-1}{\epsilon^{\rm L}(Q,W)}\right)\right.\\ &\left.+\left(\beta^2-\frac{W^2}{Q(Q+2\mathrm{m_e}c^2)}\right)\frac{W^2}{[Q(Q+2\mathrm{m_e}c^2)-W^2]^2}\,\mathrm{Im}\left(\frac{-1}{\epsilon^{\rm T}(Q,W)}\right)\right]\end{aligned}$$

- To be compared with the atomic PWBA result (for $W \ll E$)

$$\begin{aligned}\frac{\mathrm{d}^2\sigma}{\mathrm{d}W\,\mathrm{d}Q} &= \frac{2\pi Z_1^2e^4}{\mathrm{m_e}v^2}\left[\frac{2\mathrm{m_e}c^2}{WQ(Q+2\mathrm{m_e}c^2)}\,\frac{\mathrm{d}f(Q,W)}{\mathrm{d}W}\right.\\ &\left.+\left(\beta^2-\frac{W^2}{Q(Q+2\mathrm{m_e}c^2)}\right)\frac{2\mathrm{m_e}c^2W}{[Q(Q+2\mathrm{m_e}c^2)-W^2]^2}\,\frac{\mathrm{d}g(Q,W)}{\mathrm{d}W}\right]\end{aligned}$$

Semiclassical approximation

- We conclude that the two formulations are equivalent (linear response theories), and

$$\boxed{\begin{aligned} \frac{\mathrm{d}f(Q,W)}{\mathrm{d}W} &\equiv W\left(1+\frac{Q}{\mathrm{m_e}c^2}\right)\frac{2Z_2}{\pi\Omega_\mathrm{p}^2}\,\mathrm{Im}\left(\frac{-1}{\epsilon^\mathrm{L}(Q,W)}\right) \\ \frac{\mathrm{d}g(Q,W)}{\mathrm{d}W} &\equiv W\left(1+\frac{Q}{\mathrm{m_e}c^2}\right)\frac{2Z_2}{\pi\Omega_\mathrm{p}^2}\,\mathrm{Im}\left(\frac{-1}{\epsilon^\mathrm{T}(Q,W)}\right) \end{aligned}}$$

where $\Omega_\mathrm{p} = \sqrt{4\pi\mathcal{N}Z_2\frac{\hbar^2e^2}{\mathrm{m_e}}}$ is the plasma resonance energy of the material

- ❑ The semiclassical approximation provides the best methodology available for describing inelastic collisions of charged particles.

- ❑ In the case of electrons, the DCS must be modified to account for exchange effects. A practical solution is provided by the Ochkur approximation (non-relativistic)

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}W\,\mathrm{d}Q} = \frac{\mathrm{d}^2\sigma_\mathrm{class}}{\mathrm{d}W\,\mathrm{d}Q}\,F_\mathrm{Ochkur}(Q,W)$$

$$F_\mathrm{Ochkur}(Q,W) = 1+\left(\frac{Q}{K_a+E-W}\right)^2 - \frac{Q}{K_a+E-W}$$

K_a = kinetic energy of the target electron

Ochkur, V.I. (1964) “The Born-Oppenheimer method in the theory of atomic collisions”, Soviet Phys. JETP 18, 503-508.

Modeling the DF of materials

- First principles calculations are only feasible for inner subshells of atoms
- Models based on empirical optical information (assumed to be reliable!)
- We consider the inverse DFs,

$$\eta^{(\mathrm{L,T})}(Q,W) \equiv \frac{1}{\epsilon^{(\mathrm{L,T}))}(Q,W)} = \eta_1^{(\mathrm{L,T})}(Q,W) - \mathrm{i}\eta_2^{(\mathrm{L,T})}(Q,W)$$

because the imaginary part (~GOS) is additive, and satisfy the Kramers-Kronig relations

$$\eta_1(Q,W) = 1 - \frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{W'\,\eta_2(Q,W')}{W'^2-W^2}\mathrm{d}W'$$

$$\eta_2(Q,W) = \frac{2W}{\pi}\mathcal{P}\int_0^\infty \frac{\eta_1(Q,W')-1}{W'^2-W^2}\mathrm{d}W'$$

- Sum rules:
  - f-sum: $$\int_0^\infty W\,\eta_2(Q,W)\,\mathrm{d}W = \frac{\pi}{2}\Omega_\mathrm{p}^2$$
  - perfect-screening sum: $$\eta_1(0) + \frac{2}{\pi}\int_0^\infty \frac{\eta_2(W)}{W}\,\mathrm{d}W = 1$$

Modeling the DF of materials

- Low-frequency excitations (up to ~100 eV): optical DF as a linear combination of Mermin optical DFs (same form as a classical damped oscillator)

$$\eta_2(W_J, s_J; W) = \frac{s_J W_J^2 W}{(W_J^2 - W^2)^2 + s_J^2 W^2}$$

- We use a large set of "oscillators" with predefined resonance frequencies and damping constants:

$$\eta_2(W) = \sum_{J=1}^{N_o} F_J \eta_2(W_J, s_J; W)$$

and determine the "oscillator strengths" F_J from a least-squares fit (occasionally, we may have negative strengths)

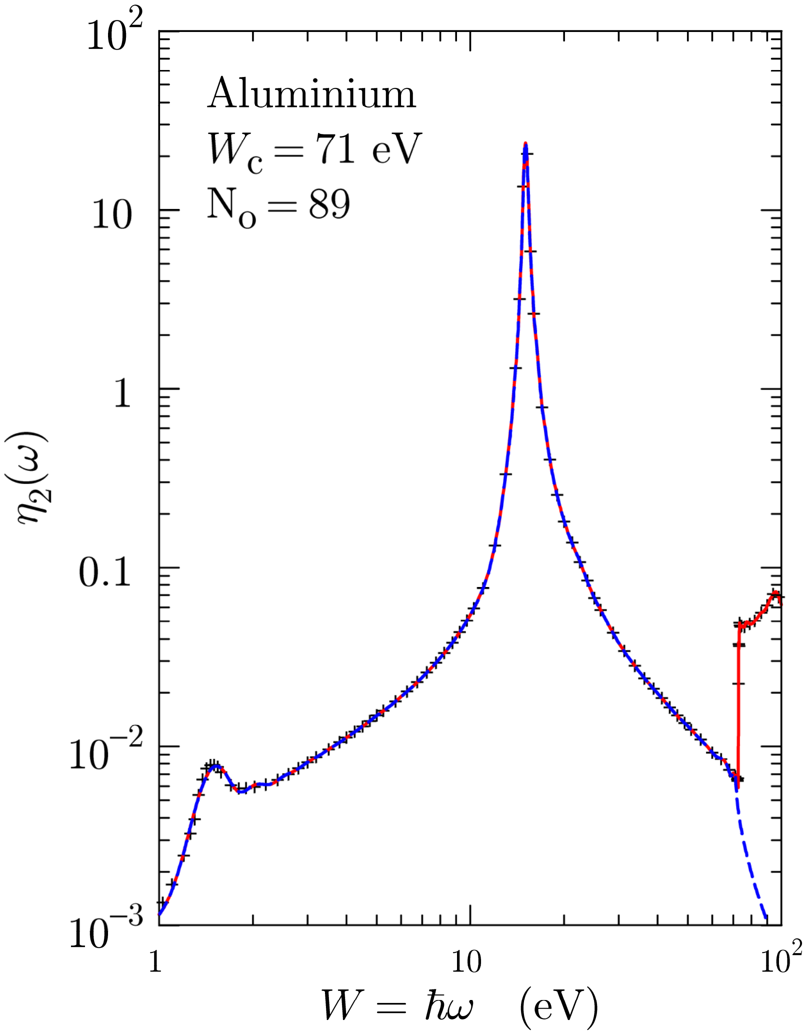
- The Mermin DF has a transverse part (with the same optical DF)

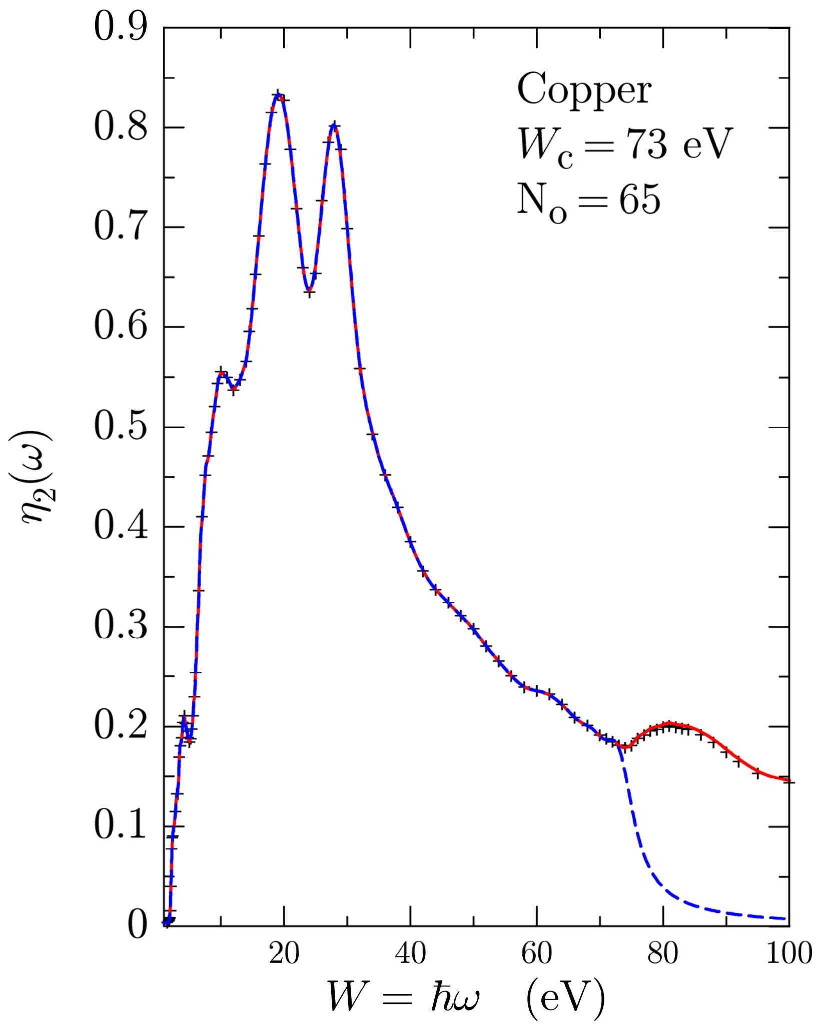
- The full DF is obtained by replacing the optical terms by the full Mermin forms:

$$\eta_2^{(\mathrm{L,T})}(Q, W) = \sum_{J=1}^{N_o} F_J \eta^{(\mathrm{L,T})}(W_J, s_J; Q, W)$$

- Provides a very accurate reproduction of empirical optical functions

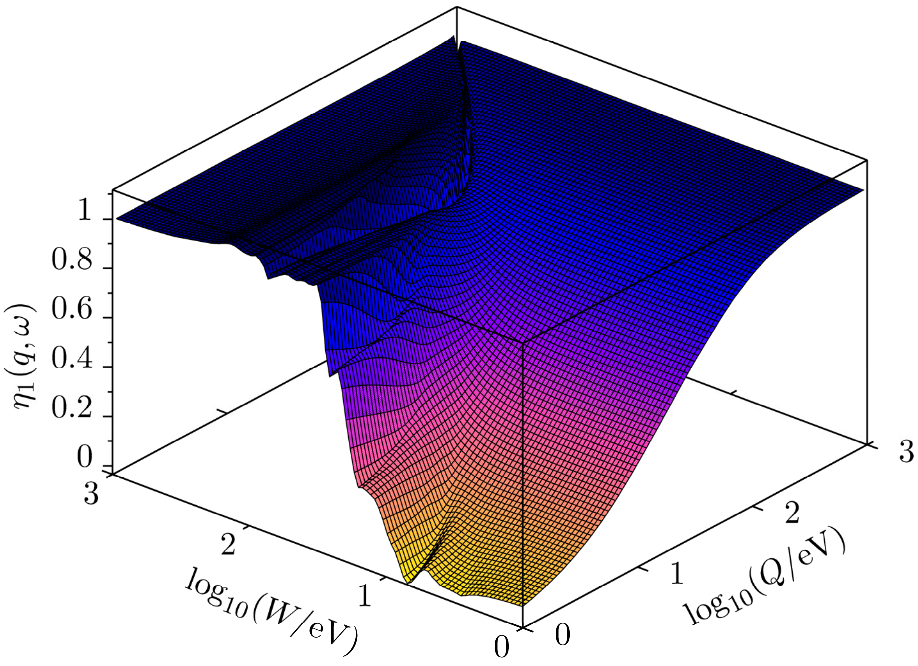
Modeling the DF of materials

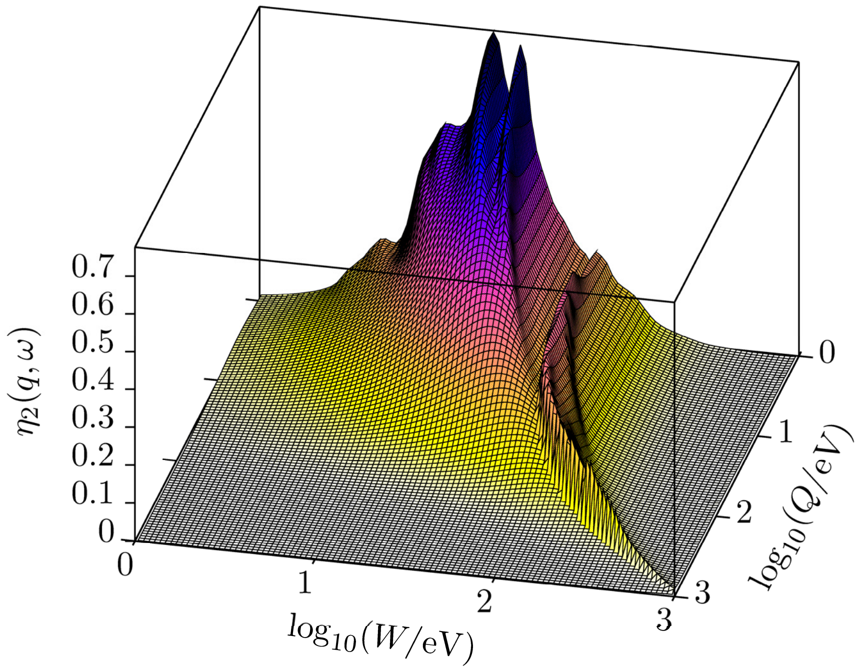


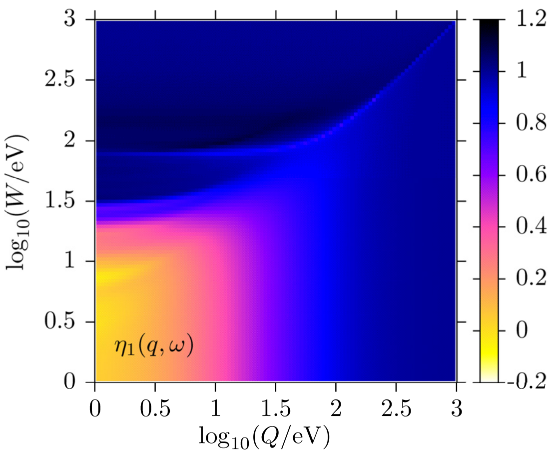


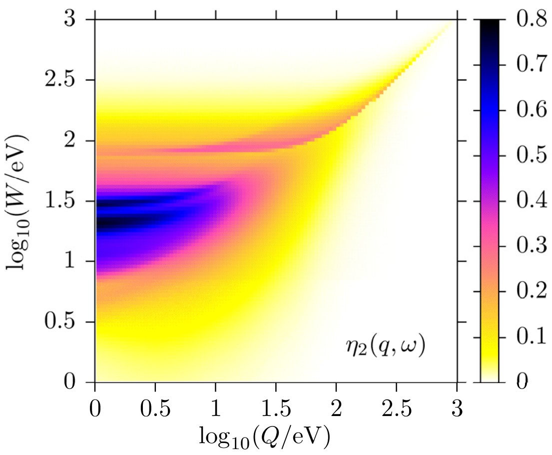
Palik, E. D. (editor) (1985), Handbook of Optical Constants of Solids (Academic Press, San Diego, CA).

Longitudinal DFs of cooper

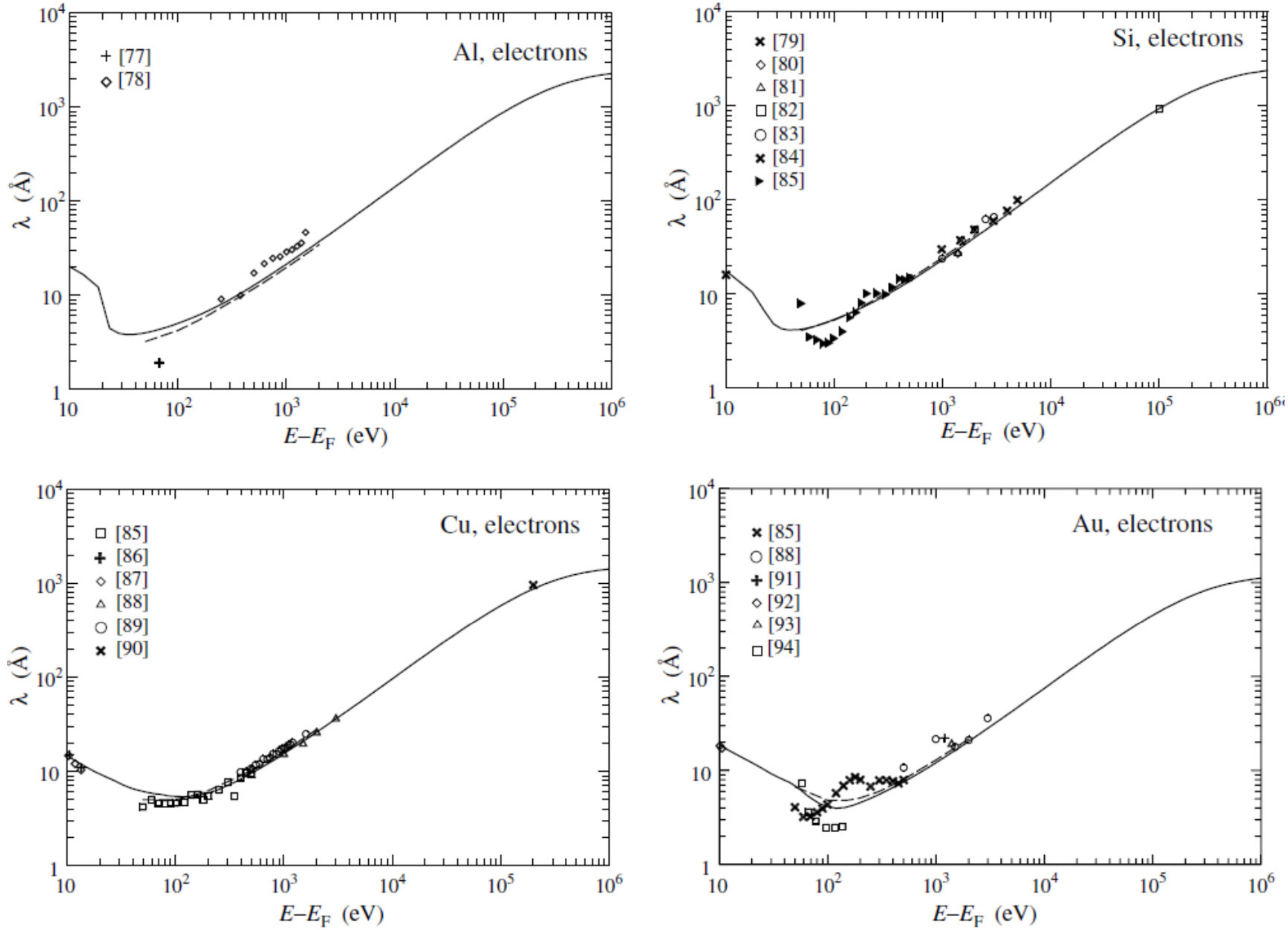






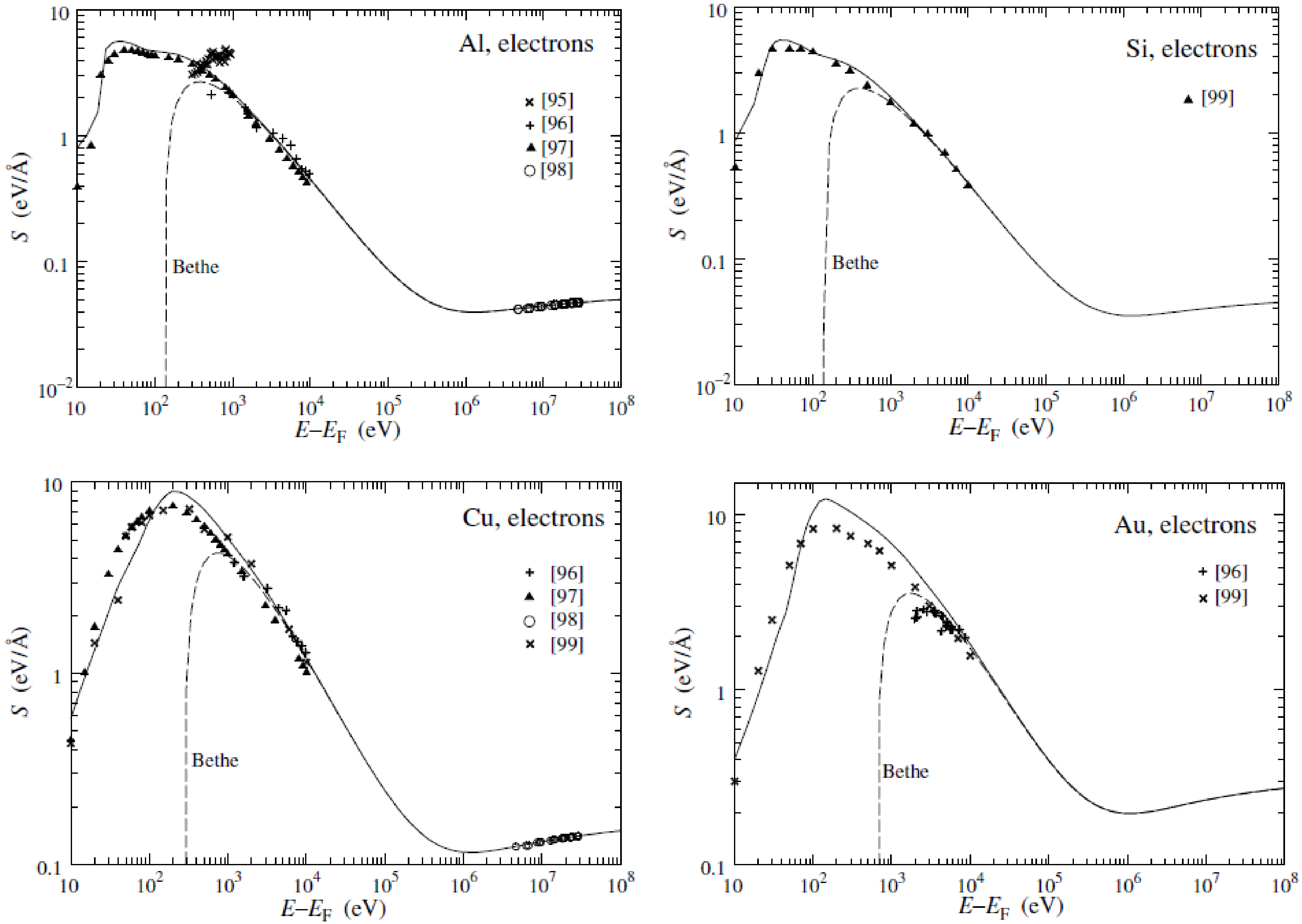


Comparison with experiments



Fernández-Varea, J. M., F. Salvat, M. Dingfelder, and D. Liljequist (2005), "A relativistic optical-data model for inelastic scattering of electrons and positrons in condensed matter," Nucl. Instrum. Meth. B 229, 187–218.

Comparison with experiments



Fernández-Varea, J. M., F. Salvat, M. Dingfelder, and D. Liljequist (2005), “A relativistic optical-data model for inelastic scattering of electrons and positrons in condensed matter,” Nucl. Instrum. Meth. B 229, 187–218.

Beyond the PWBA: distorted waves

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Calculations of inner-shell ionization by electron impact with the distorted-wave and plane-wave Born approximations

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A method is described for computing total cross sections for the ionization of inner shells of atoms and positive ions by impact of electrons and positrons with arbitrary energies. The method combines the relativistic plane-wave Born approximation (PWBA) with a semirelativistic version of the distorted-wave Born approximation (DWBA). Formal expressions for the longitudinal and transverse generalized oscillator strengths (GOSs) of closed shells are derived. Tables of GOSs for K shells and for L and M subshells of neutral atoms have been calculated for a discrete grid of energy losses and recoil energies. A suitable interpolation scheme allows the easy evaluation of PWBA ionization cross sections from these GOS tables. The difference between the total ionization cross sections that result from the DWBA and the PWBA (considering the longitudinal interaction only) has been calculated numerically for projectiles with kinetic energies up to 16 times the ionization energy of the active shell. In this energy range, ionization cross sections with the accuracy of a distorted-wave calculation are obtained by simply adding this difference to the cross section resulting from the conventional PWBA. For higher energies, the cross section is obtained by multiplying the PWBA cross section by an energy-dependent scaling factor that is determined by a single fitted parameter. Numerical results are shown to agree with experimental data, when these are available.

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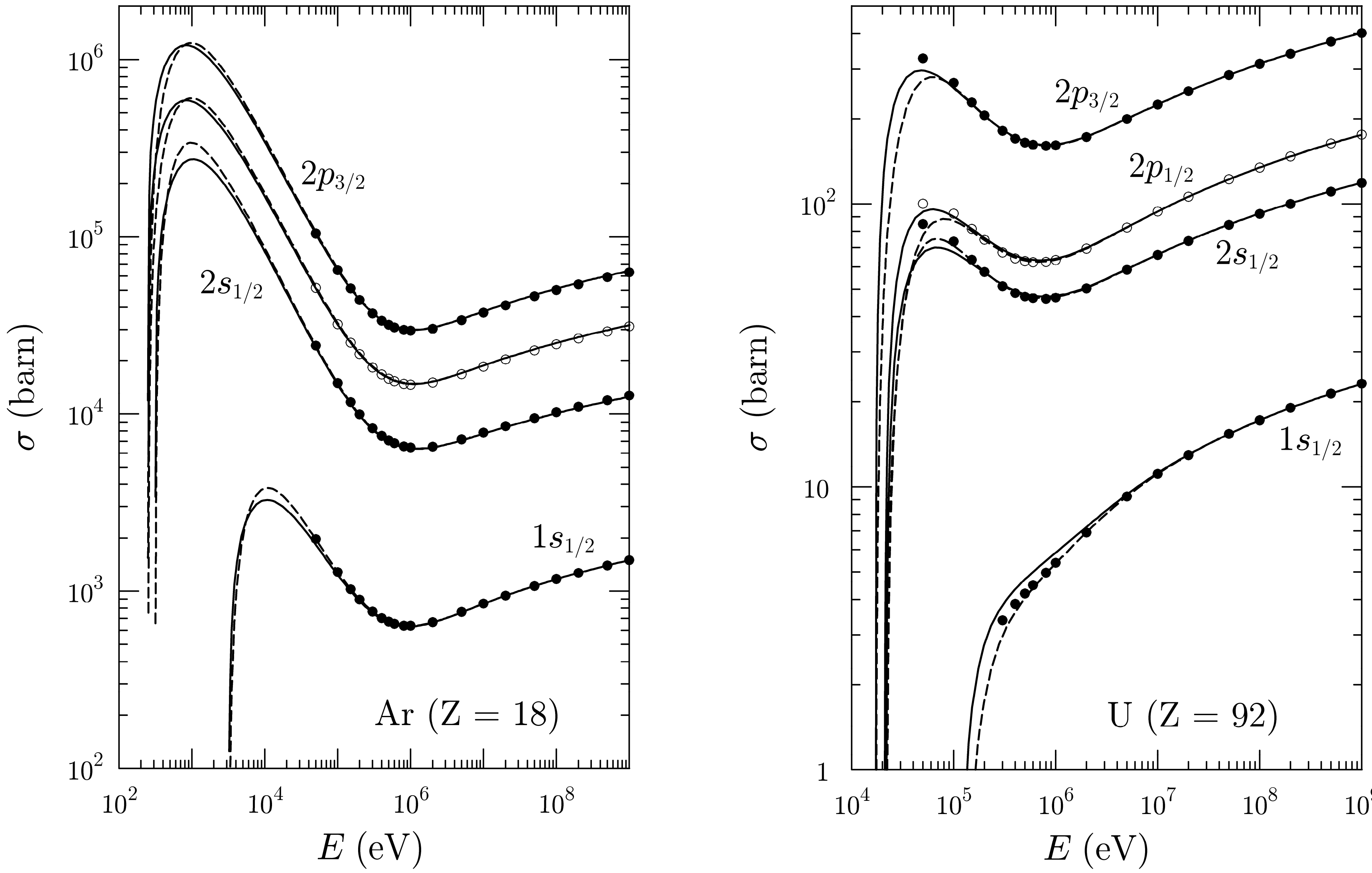
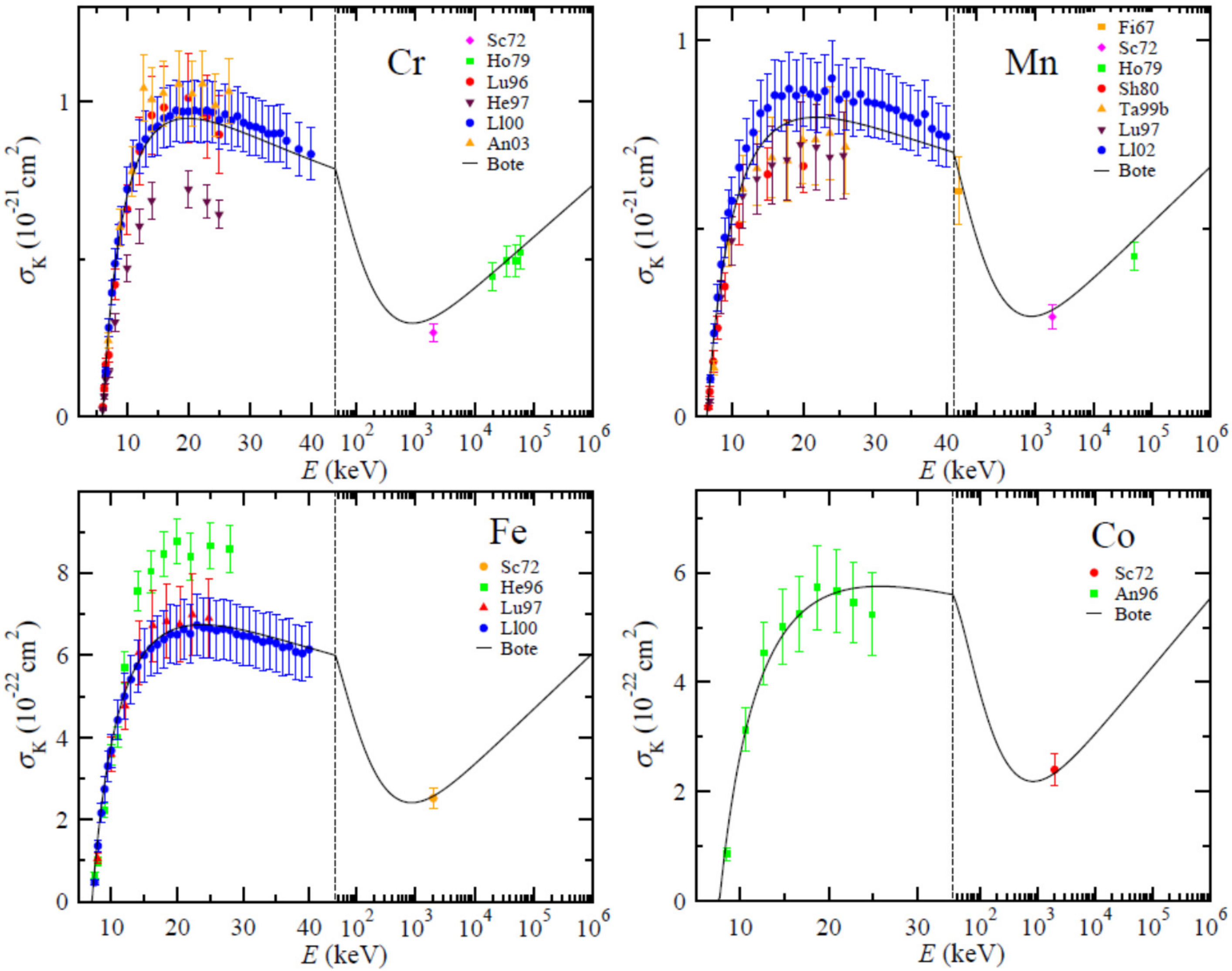
Distorted-wave BA vs. PWBA



Dashed, PWBA; solid, distorted-wave BA

DWBA vs experiment



Llovet, X., C. J. Powell, A. Jablonski, and F. Salvat (2014), “Cross sections for inner-shell ionization by electron impact,” J. Phys. Chem. Ref. Data 43, 013102.

Stopping theory (historical perspective)

❑ **Bethe (1932), Fano (1963)** The stopping power for high-energy particles obtained from the plane-wave Born approximation is given by the (asymptotic) formula

$$S_{\mathrm{Bethe}} = \frac{4\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} Z_2 \left[\ln\left(\frac{2\mathrm{m_e}c^2\beta^2}{I}\right) + \ln\left(\frac{1}{1-\beta^2}\right) - \beta^2 \right]$$

where I is the "mean excitation energy" defined as

$$Z_2 \ln I = \int_0^\infty \ln(W) \frac{\mathrm{d}f(W)}{\mathrm{d}W} \,\mathrm{d}W$$

ICRU Report 37 (1984) Stopping Powers for Electrons and Positrons (ICRU, Bethesda, MD).

❑ **Bloch (1933)** Under certain circumstances, the classical theory is applicable

Validity of the theory: Classical (Bohr) $\eta = \dfrac{|Z_1|\, e^2}{\hbar v} \gg 1$

PWBA (Bethe) $\dfrac{Z_2\, e^2}{\hbar v} \ll 1$

$$S_{\mathrm{Bethe-Bloch}} = \frac{4\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} Z_2 \left[\ln\left(\frac{2\mathrm{m_e}c^2\beta^2}{I}\right) + \ln\left(\frac{1}{1-\beta^2}\right) - \beta^2 + \Delta L_2 \right]$$

with $\Delta L_2 = -\eta^2 \sum_{n=0}^{\infty} \dfrac{1}{n(n^2+\eta^2)}$ gives the correct (classical or quantum perturb.) limits

Bloch F. (1933) "Zur Bremsung rasch bewegter Teilchen beim Durchgang durch Materie," Ann. Phys. (Leip.) 16, 285-320.
Lindhard, J. and A. H. Sørensen (1996), "Relativistic theory of stopping for heavy ions," Phys. Rev. A 53, 2443-2456.

Stopping theory (historical perspective)

❑ **Barkas effect (1972)** Differences between stopping powers of particles and antiparticles

Contributions of order Z_1^3 from distant ($b > a$) and close ($b < a$)interactions

- Distant interactions (displacement of electrons from equilibrium position, to 1st order) In a way, similar to the 2nd-order Born approximation

$$\frac{\mathrm{d}\,(\Delta\sigma)_{\mathrm{Bd}}}{\mathrm{d}W} = \frac{2\pi\, Z_1^2 e^4}{\mathrm{m_e} v^2}\, \frac{Z_1 \alpha\beta}{\gamma^2\beta^4\, \mathrm{m_e} c^2}\, \frac{\mathrm{d}f(W)}{\mathrm{d}W} \left[I_1(\xi) + \frac{1}{\gamma^2}\, I_2(\xi) \right]$$

with $\xi_a = \omega a/\gamma v$. The cutoff a is a parameter of the theory: $a = \dfrac{\hbar}{1.781 \mathrm{m_e} v}$ (Lindhard)

$$I_1(\xi_a) = -\int_{\xi_a}^{\infty} \frac{1}{\xi^2}\, K_1(\xi)\, G_1(\xi)\, \mathrm{d}\xi \qquad I_2(\xi_a) = \int_{\xi_a}^{\infty} \frac{1}{\xi^2}\, K_0(\xi)\, G_0(\xi)\, \mathrm{d}\xi,$$

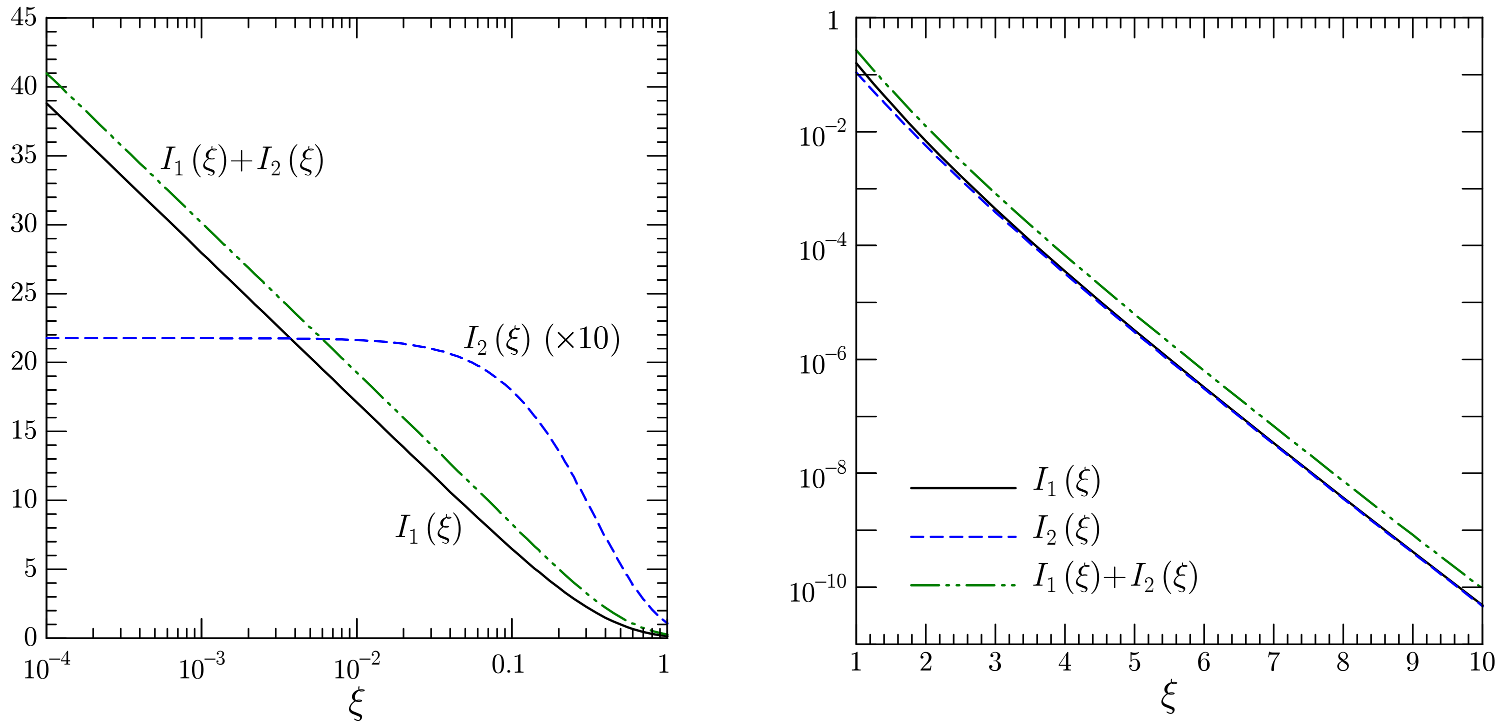
$$G_1(\xi) = \int_{-\infty}^{\infty} \mathrm{d}x\, \frac{\cos(\xi x)}{(1+x^2)^{5/2}} \left[(x^2-2) F_1(\xi,x) - 3x\, F_2(\xi,x) \right]$$

$$G_0(\xi) = \int \mathrm{d}x\, \frac{\sin(\xi x)}{(1+x^2)^{5/2}} \left[3x\, F_1(\xi,x) - (1-2x^2)\, F_2(\xi,x). \right]$$

$$F_1(\xi,x) = \int_{-\infty}^{y} \frac{\sin[\xi(x-y)]}{(1+y^2)^{3/2}}\, \mathrm{d}y \qquad F_2(\xi,y) = \int_{-\infty}^{x} \frac{y\, \sin[\xi(x-y)]}{(1+y^2)^{3/2}}\, \mathrm{d}y$$

Ashley, J., R. H. Ritchie, and W. Brandt (1972), “Z_1^3 effect in the stopping power of matter for charged particles,” Phys. Rev. B 5, 2393-2397.
Jackson, J. and R. L. McCarthy (1972), “Z_1^3 corrections to energy loss and range,” Phys. Rev. B 6, 4131-4141.

Stopping theory (historical perspective)



- Close collisions (McKinley-Feshbach expansion of Mott's DCS replaces Rutherford DCS)

$$\frac{\mathrm{d}\sigma}{\mathrm{d}W} = \frac{2\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \frac{1}{W^2} \left\{ 1 - \beta^2 \frac{W}{W_{\max}} + \pi Z_1 \alpha \beta \left[\left(\frac{W}{W_{\max}} \right)^{1/2} - \frac{W}{W_{\max}} \right] \right\}$$

$$W_{\max} = \frac{2\gamma^2 \beta^2 \, \mathrm{m_e} c^2}{1 + (2\mathrm{m_e}/M_1)\gamma + \mathrm{m_e^2}/M_1^2}$$ maximum energy transfer in a single collision

Stopping power formula

❑ **Corrected Bethe-Bloch formula** (with proper kinematical limits)

$$S = \frac{4\pi Z_1^2 e^4}{\mathrm{m_e} v^2} \mathcal{N} Z_2 \left[L_0 + \Delta L_1 + \Delta L_2\right]$$

- Particles heavier than the electron: $L_0 = \ln\left(\frac{2\mathrm{m_e} c^2 \beta^2}{I}\right) + f(\gamma) - \frac{C}{Z_2} - \delta_{\mathrm{F}}$

$$f(\gamma) = \ln\left(\gamma^2\right) - \beta^2 + \frac{1}{2}\left[\ln R + \beta^2(1-R) - 2\gamma\beta^2 \frac{\mathrm{m_e}}{M_1} R + \gamma^2\beta^4 \frac{\mathrm{m_e^2}}{M_1^2} R^2\right]$$

$$R = \left(1 + \frac{\mathrm{m_e^2}}{M_1^2} + 2\gamma \frac{\mathrm{m_e}}{M_1}\right)^{-1}$$

- Electrons (−) and positrons (+): $L_0 = \frac{1}{2}\left[\ln\left(\frac{E^2}{I^2}\frac{\gamma+1}{2}\right) + f^{(\pm)}(\gamma) - \frac{2C}{Z_2} - \delta_{\mathrm{F}}\right]$

$$f^{(-)}(\gamma) = 1 - \beta^2 - \frac{2\gamma - 1}{\gamma^2}\ln 2 + \frac{1}{8}\left(\frac{\gamma-1}{\gamma}\right)^2$$

$$f^{(+)}(\gamma) = 2\ln 2 - \frac{\beta^2}{12}\left[23 + \frac{14}{\gamma+1} + \frac{10}{(\gamma+1)^2} + \frac{4}{(\gamma+1)^3}\right]$$

$$\ln I = \frac{1}{Z_2}\int_0^\infty \ln(W)\,\frac{\mathrm{d}f(W)}{\mathrm{d}W}\,\mathrm{d}W$$

- Barkas or Z_1^3 correction:

$$\Delta L_1 = \frac{1}{2} Z_1 \alpha \beta \left\{ \pi + \frac{1}{\gamma^2 \beta^4} \frac{1}{Z_2} \frac{1}{\mathrm{m_e} c^2} \int_0^\infty \mathrm{d}W \, \frac{\mathrm{d}f(W)}{\mathrm{d}W} \, W \left[I_1(\xi) + \frac{1}{\gamma^2} I_2(\xi) \right] \right\}$$

- Bloch correction: $$\Delta L_2 = -\eta^2 \sum_{n=0}^{\infty} \frac{1}{n(n^2 + \eta^2)}$$

- Density-effect (or polarization) correction:

$$\delta_{\mathrm{F}} \equiv \frac{1}{Z_2} \int_0^\infty \frac{\mathrm{d}f(W)}{\mathrm{d}W} \ln \left(1 + \frac{L^2}{W^2} \right) \mathrm{d}W - \frac{L^2}{\hbar^2 \Omega_{\mathrm{p}}^2} \left(1 - \beta^2 \right)$$

where L is the positive root of the equation

$$\frac{1}{Z} \hbar^2 \Omega_{\mathrm{p}}^2 \int_0^\infty \frac{1}{W^2 + L^2} \frac{\mathrm{d}f(W)}{\mathrm{d}W} \, \mathrm{d}W = 1 - \beta^2$$

- Shell correction C : Difference between the actual stopping power and the Bethe-Bloch formula.
For electrons and positrons the shell correction is negligible for energies above ~10 keV

- **Summarizing:** Knowledge of the OOS is required for devising realistic DCS models for the simulation of inelastic collisions of low-energy particles, and for describing fine features of its integrals (stopping power and mean-free path)

- **Collaboration:**

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Xavier Llovet (U Barcelona)
Francesc Salvat-Pujol (CERN)

Thanks!