Calculating Electron Energy Loss Spectroscopy from First Principles

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Electron Energy Loss Spectroscopy (EELS)

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\[ \Delta E_{\text{loss}} = - \int F_{\text{ind}} \cdot dr \]
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\Delta \tilde{E}_{\text{loss}}(q, \omega) \sim -\Im \left[ \varepsilon_m^{-1}(q, \omega) \right]
\]

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(6,4) Single-walled Carbon Nanotube
(a) Band Structure
(b) Density of States

(6,5) Single-walled Carbon Nanotube
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SWNT Van Hove Singularities

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SWNT Excitations

(a) Band Structure

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Calculating EELS from First Principles
Density Functional Theory (DFT) PBE Calculation (GPAW)
Tight Binding (TB) Calculation with $G_0 W_0$ Parameters
Tight Binding (TB) Calculation with LDA Parameters

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Tight Binding (TB) Calculation with LDA Parameters

Density Functional Theory PBE and Tight-Binding LDA Calculations are in good agreement.
Differences between DFT-PBE and TB-LDA are attributable to curvature effects.
Using TB $G_0 W_0$ parameters yields a stretching of the DOS by $\sim 10\%$.

This is attributable to both self-interaction errors in DFT, and the quasi-particle excitation corrections of $G_0 W_0$.

However, $G_0 W_0$ calculations are $O(N^4)$.

Other sources of error in DFT-RPA EELS calculations may be bigger than 10%.
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Loss Function $\mathcal{S} \left[ \varepsilon_{m}^{-1}(q, \omega) \right]$
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Will the energy loss spectra depend on the electron beam energy?
Two-Fluid 2D Hydrodynamic Model

Single Fluid $n_0$

$\pi$ Fluid $n_\pi = n_0/4$

$\sigma$ Fluid $n_\sigma = 3n_0/4$

Plasmon Energies

1. Single-Fluid (SF)
2. Two-Fluid Thomas-Fermi Kinetic Energy (TF)
3. Dirac Exchange Energy (D)
4. Restoring Force ($\omega_\sigma \approx 16\ eV$) (R)

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The low energy \(\pi\) peaks and higher \(\sigma+\pi\) peaks arise from electrostatic repulsion between the \(\pi\) and \(\sigma+\pi\) orthogonal orbitals, and the 1:3 ratio of their densities.
Collaborators

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- Thomas Pichler (University of Vienna)
- Christian Kramberger (University of Vienna)
- Paola Ayala (University of Vienna)
- Zoran L. Mišković (University of Waterloo)
- Silvina Segui (Comisión Nacional de Energía Atómica)