Nebula: a Powerful Tool for Low Energy Electrons Simulations

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Why Nebula?

- **What is:** open-source software package with first-principle physical models for electron Monte Carlo simulations developed by L. van Kessel et al. (10.1016/j.softx.2020.100605)

- **Main Goal:** Scanning Electron Microscope (SEM) images simulation

  ✓ Accurate description of low energy (< 1000 keV) electrons behaviour

  ✓ Material file generation tool to keep into account material parameters

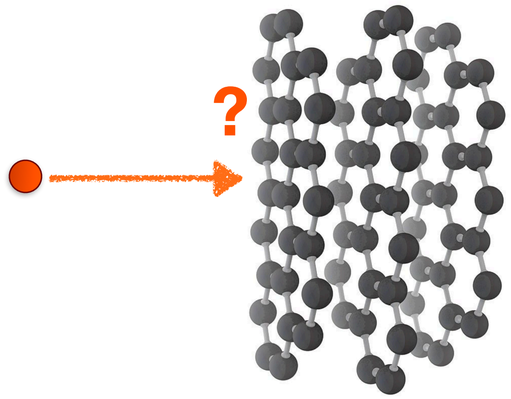
- Used for a variety of different applications in the fields of electron microscopy, electron beam induced deposition, electron beam lithography & electron detection (>15 works)

- Can run on GPUs for fast simulations, or on multi-core CPUs if there is no GPU available

What Can Happen to an Electron?

- Three **types of event**:

1. *Inelastic scattering*
   - Electron energy loss → cross sections with Penn algorithm
   - Secondary electron generation
2. *Elastic scattering*
   - for E < 100 eV: electron–acoustic phonon scattering (Schreiber and Fitting model)
   - for E > 200 eV: Mott scattering → cross section with ELSEPA
3. *Material crossing*
   - Refraction or reflection → transmission coefficient with step function model potential



Ingredients for a Simulation

- Three **types of input**:

1. *Geometry*
   - List of triangles + materials on either side
   - Detector implemented as “special material” ⟷ electron information storage:
     - 100% efficiency
     - no energy broadening
2. *Primary Electrons*
   - List of starting positions, directions, energies & tags
     - stored in a numpy datatype: with distributions → more realistic beams simulation
3. *Material File(s)*

Material File: Two Examples

Silicon

```
name:           silicon
density:        2.3290 g/cm³

elements:
    Si: { count: 1, Z: 14, M: 28.0855 g/mol }

band_structure:
    model:      semiconductor
    valence:    7.27 eV
    band_gap:   1.12 eV
    affinity:   4.05 eV

optical:
    df_file:    elf/df_Si.dat

phonon:
    lattice:    5.430710 Å
    m_dos:      1.08 m_e
    m_eff:      0.26 m_e

    longitudinal:
        alpha:  2.00e-7 m²/s
        c_s:    9130 m/s      # Landolt-Bornstein Vol. III/41A1a 872
        ac_def: 9.2 eV        # Landolt-Bornstein Vol. III/41A1a 648

    transversal:
        alpha:  2.26e-7 m²/s
        c_s:    5842 m/s      # Landolt-Bornstein Vol. III/41A1a 872
        ac_def: 5.0 eV        # Landolt-Bornstein Vol. III/41A1a 648
```

Gold

```
name:               gold
density:            19.30 g/cm³

elements:
    Au: { count: 1, Z: 79, M: 196.97 g/mol }

band_structure:
    model:          metal
    fermi:          5.53 eV
    work_function:  5.38 eV


optical:
    df_file:        elf/df_Au.dat

phonon:
    model:          dual
    lattice:        4.0782 Å


    isotropic:
        resistivity: 2.44e-8 Ω m

    longitudinal:
        c_s:        3240 m/s


    transversal:
        c_s:        1200 m/s
```

Material File: Two Examples

Silicon

```
name:           silicon
density:        2.3290 g/cm³

elements:
    Si: { count: 1, Z: 14, M: 28.0855 g/mol }

band_structure:
    model:      semiconductor
    valence:    7.27 eV
    band_gap:   1.12 eV
    affinity:   4.05 eV

optical:
    df_file:    elf/df_Si.d

phonon:
    lattice:    5.430710 Å
    m_dos:      1.08 m_e
    m_eff:      0.26 m_e

    longitudinal:
        alpha:  2.00e-7 m²/
        c_s:    9130 m/s
        ac_def: 9.2 eV

    transversal:
        alpha:  2.26e-7 m²/s
        c_s:    5842 m/s     # Landolt-Bornstein Vol. III/41A1a 872
        ac_def: 5.0 eV       # Landolt-Bornstein Vol. III/41A1a 648
```

Gold

```
name:               gold
density:            19.30 g/cm³

elements:
    Au: { count: 1, Z: 79, M: 196.97 g/mol }

band_structure:
    model:          metal
    fermi:          5.53 eV
    work_function:  5.38 eV

    transversal:
        c_s:        1200 m/s
```

- Generic Parameters (`name`, `density`)
- Element list: for each element
  - `count` = # atoms per unit cell
  - `Z` = atomic number
  - `M` = average atomic mass

Material File: Two Examples

Silic

Band structure:

- Metals: single band = conduction band
- Semiconductors & insulators: valence and conduction bands, band gap in between

```
band_structure:
    model:      semiconductor
    valence:    7.27 eV
    band_gap:   1.12 eV
    affinity:   4.05 eV
```

```
band_structure:
    model:          metal
    fermi:          5.53 eV
    work_function:  5.38 eV
```

- `model`: semiconductor, insulator or metal

- `valence` (band width)
- `band_gap`
- `affinity` = bottom of conduction bands wrt vacuum

- `fermi` = Fermi level respect to bottom of conduction band
- `work_function`

Material File: Two Examples

Silic Phonon:

- `lattice` = material's lattice constant
- `m_dos` & `m_eff` = electron density-of-states mass & effective mass
- `alpha` = band bending of phonon dispersion relation near Brillouin zone edge
- `c_s` = sound speed
- `ac_def` = acoustic deformation potential (or `resistivity` for metals)
  - ➡ if not known for each mode: `isotropic` values

```
phonon:
    lattice:    5.430710 Å
    m_dos:      1.08 m_e
    m_eff:      0.26 m_e

    longitudinal:
        alpha:  2.00e-7 m²/s
        c_s:    9130 m/s      # Landolt-Bornstein Vol. III/41A1a 872
        ac_def: 9.2 eV        # Landolt-Bornstein Vol. III/41A1a 648

    transversal:
        alpha:  2.26e-7 m²/s
        c_s:    5842 m/s      # Landolt-Bornstein Vol. III/41A1a 872
        ac_def: 5.0 eV        # Landolt-Bornstein Vol. III/41A1a 648
```

```
phonon:
    model:          dual
    lattice:        4.0782 Å


    isotropic:
        resistivity: 2.44e-8 Ω m

    longitudinal:
        c_s:        3240 m/s


    transversal:
        c_s:        1200 m/s
```

Material File: Two Examples

Silicon

```
name:           silicon
density:        2.3290 g/cm³

elements:
    Si: { count: 1, Z: 14, M: 28.0855 g/mol }

band_structure:
    model:      semiconductor
    valence:    7.27 eV
    band_gap:   1.12 eV
    affinity:   4.05 eV

optical:
    df_file:    elf/df_Si.dat

phonon:
    lattice:    5.430710 Å
    m_dos:      1.08 m_e
    m_eff:      0.26 m_e

    longitudinal:
        alpha:  2.00e-7 m²/s
        c_s:    9130 m/s      # Landolt-Bornstein Vol. III/41A1a 872
        ac_def: 9.2 eV        # Landolt-Bornstein Vol. III/41A1a 648

    transversal:
        alpha:  2.26e-7 m²/s
        c_s:    5842 m/s      # Landolt-Bornstein Vol. III/41A1a 87
        ac_def: 5.0 eV        # Landolt-Bornstein Vol. III/41A1a 648
```

Gold

```
name:               gold
density:            19.30 g/cm³

elements:
    Au: { count: 1, Z: 79, M: 196.97 g/mol }

band_structure:
    model:          metal
    fermi:          5.53 eV
    work_function:  5.38 eV

optical:
    df_file:        elf/df_Au.dat

phonon:
    model:          dual
    lattice:        4.0782 Å
                    c_s:        3240 m/s

    transversal:
        c_s:        1200 m/s
```

Optical:

- `df_file` = optical data to represent probability of electron energy loss in a material

Energy Loss Function from Optical File

- Optical data to represent probability of electron energy loss

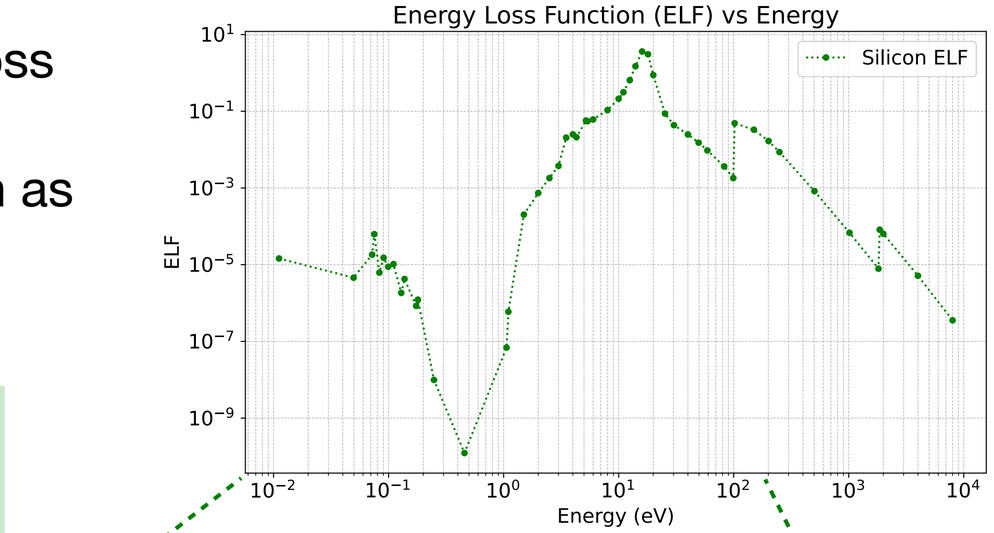
  - dielectric function ϵ to compute Energy Loss Function as

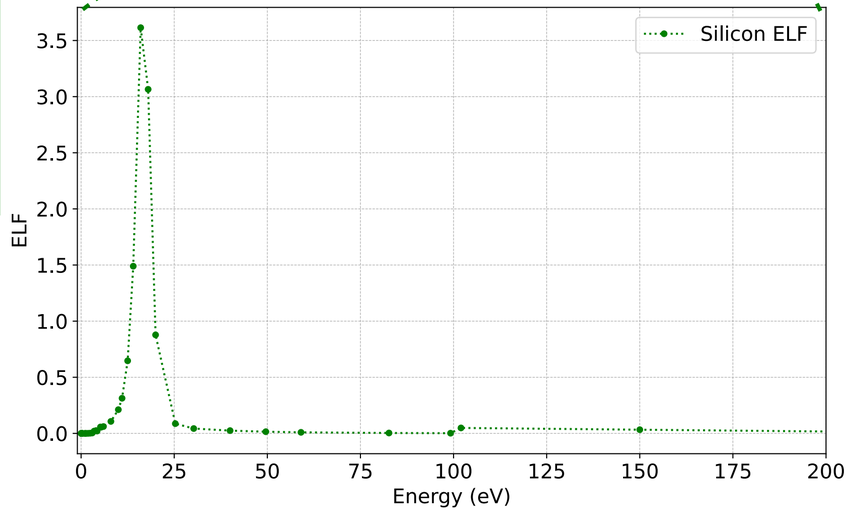
$$ELF = Im\left(-\frac{1}{\epsilon}\right)$$

HOW IS TREATED

- q dependent ELF expanded from q=0 one in terms of Lindhard function
- Single pole approx. of Lindard function used (Penn algorithm)

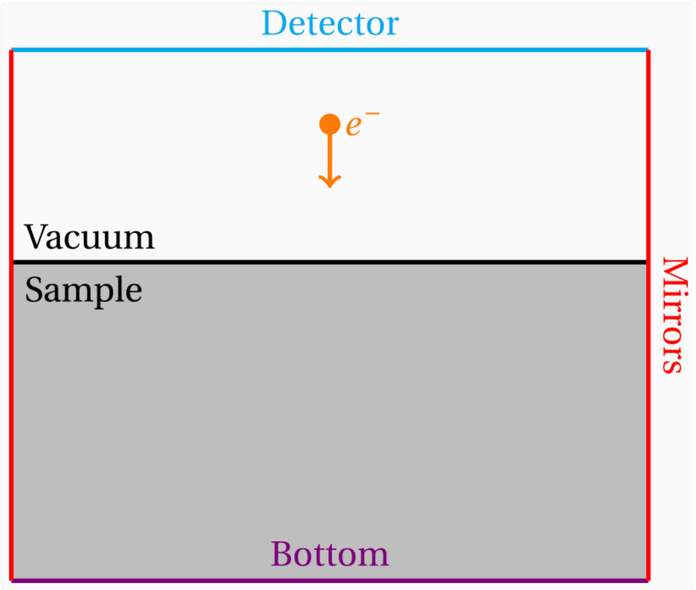
  - Can also take as input ELF tabulated over very large range of energies (e.g. Si optical file)





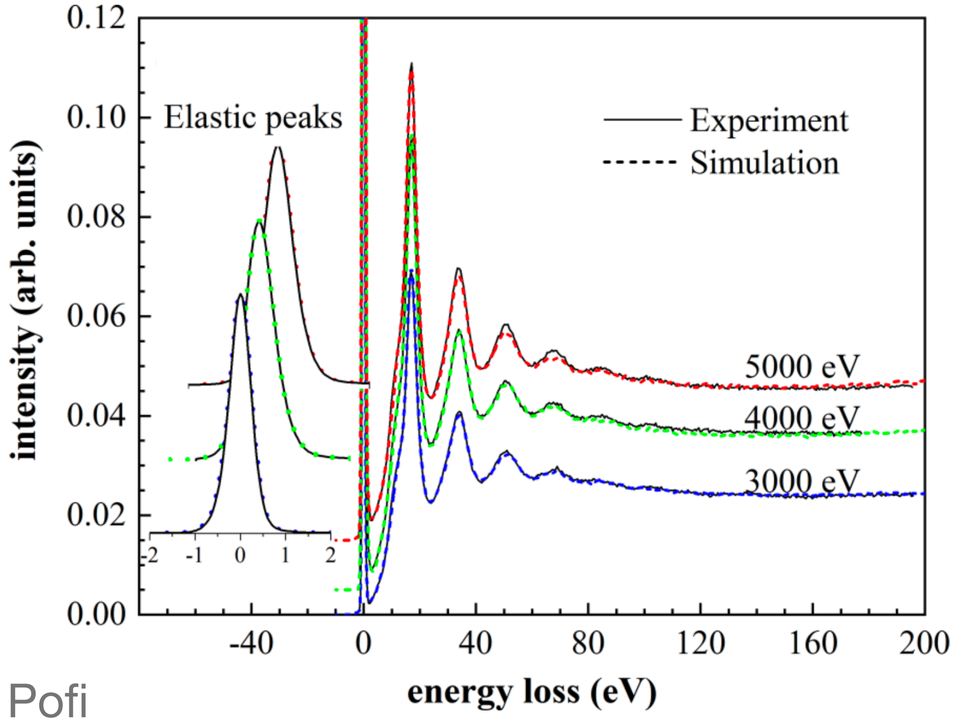
A First Trial: Bulk Silicon REELS

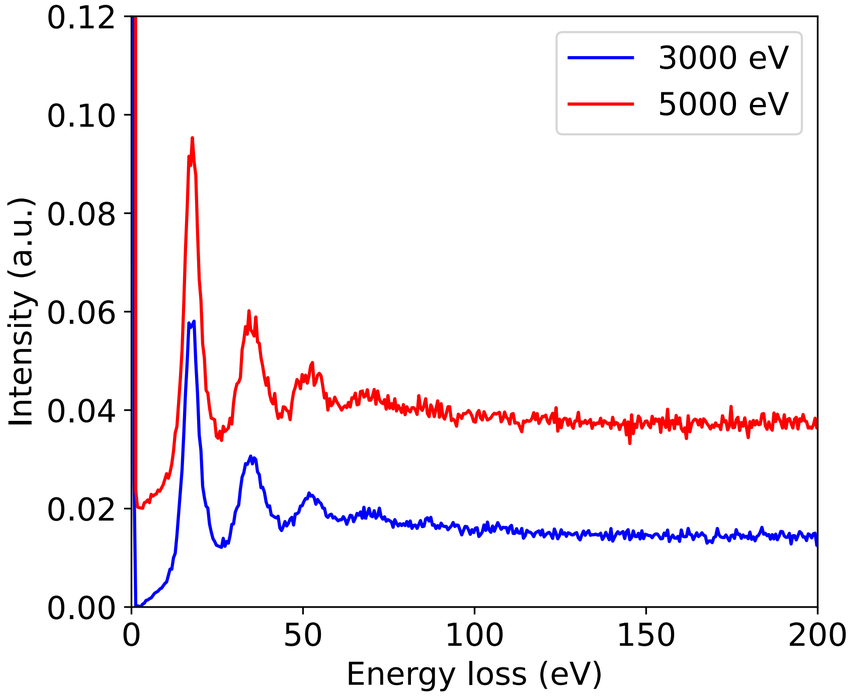
- Aims:
  - Try simulation tool with a material already available on it → Reflection Electron Energy Loss Spectroscopy of Silicon
  - Compare results with experimental ones from literature
- Geometry:
  - 1 μm x 1 μm x 10 μm Silicon sample
  - Detector at 10 nm distance from sample surface, // to it
  - Terminator surface on bottom, mirror surfaces on sides
- Primary Electrons:
  - 3000 eV & 5000 eV monochromatic electrons
  - 10 millions, emitted perpendicular to surface at 5 nm distance



Good Agreement with Experimental Results

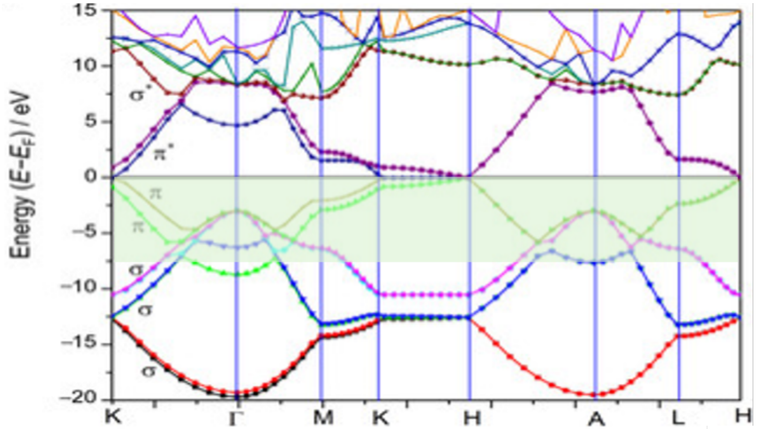
- Comparison with REELS Si spectra from L. H. Yang et al., 10.1103/PhysRevB.100.245209
- Good agreement with theoretical & measured bulk plasmon peaks energies (~17 eV & multiples)
- Slightly lower relative peaks intensities (e.g. 2nd/1st peak @ 3000 eV: 1/2 vs 2/3 experimental)





Looking for Graphite Parameters

- Middle Step: REELS of bulk graphite simulation
- Material file for *q in plane* graphite (thanks to Valentina):




```
band_structure:
    model:          metal
    fermi:          7.5 eV
    work_function:  4.7 eV
```

well established in literature

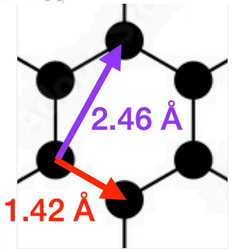
distance between Fermi level & bottom of π band

```
phonon:
    lattice:    2.461 Å   #or 1.42
    m_dos:      1.0 m_e
    m_eff:      0.02 m_e

    longitudinal:
        alpha:  3e-7 m²/s
        c_s:    22160 m/s
        ac_def: 6.8 eV
        #or 4.5 or 7.8

    transversal:
        alpha:  4e-7 m²/s
        c_s:    14660 m/s
        ac_def: 6.8 eV
```

m_{eff} really small (found ~1/18 m_e)

m_{dos} to be defined

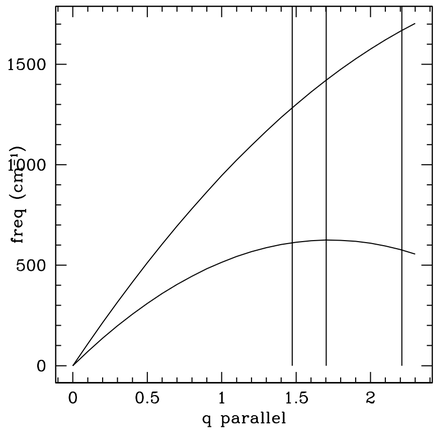
lattice constant but crystal symmetry not given, maybe to be replaced with C-C distance



value for graphene from 10.1103/PhysRevB.85.165440

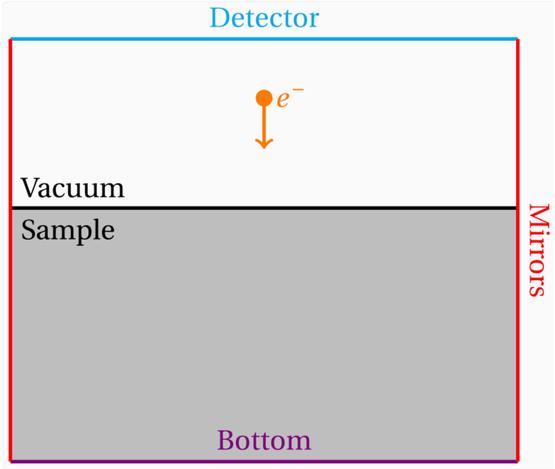
fitted to reproduce zone boundary acoustic branch bending

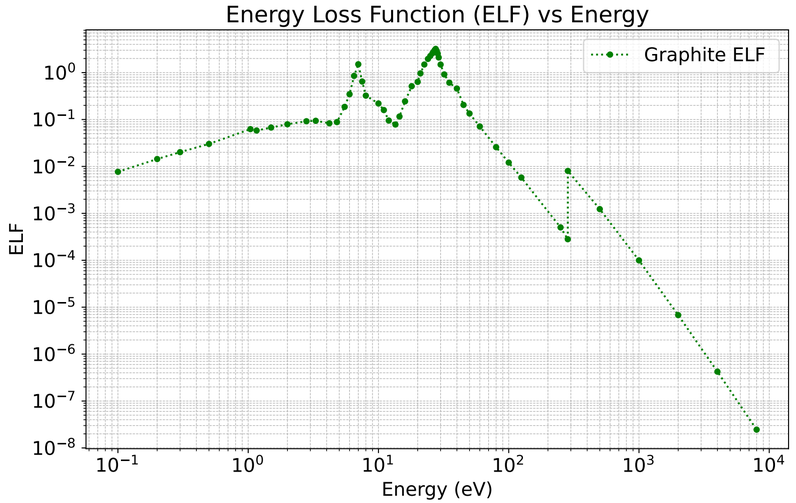
from 10.1103/PhysRevB.75.153408

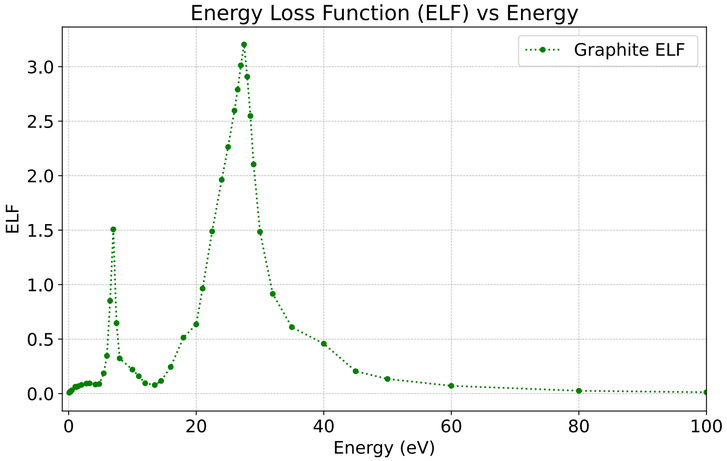


A First Trial: Bulk Silicon REELS

- Geometry:
  - Same of first simulation but with bulk graphite as sample
- Primary Electrons:
  - 1000 eV monochromatic electrons
  - 10 millions, emitted perpendicular to surface at 5 nm distance
- Energy loss function:
  - Already available on Nebula





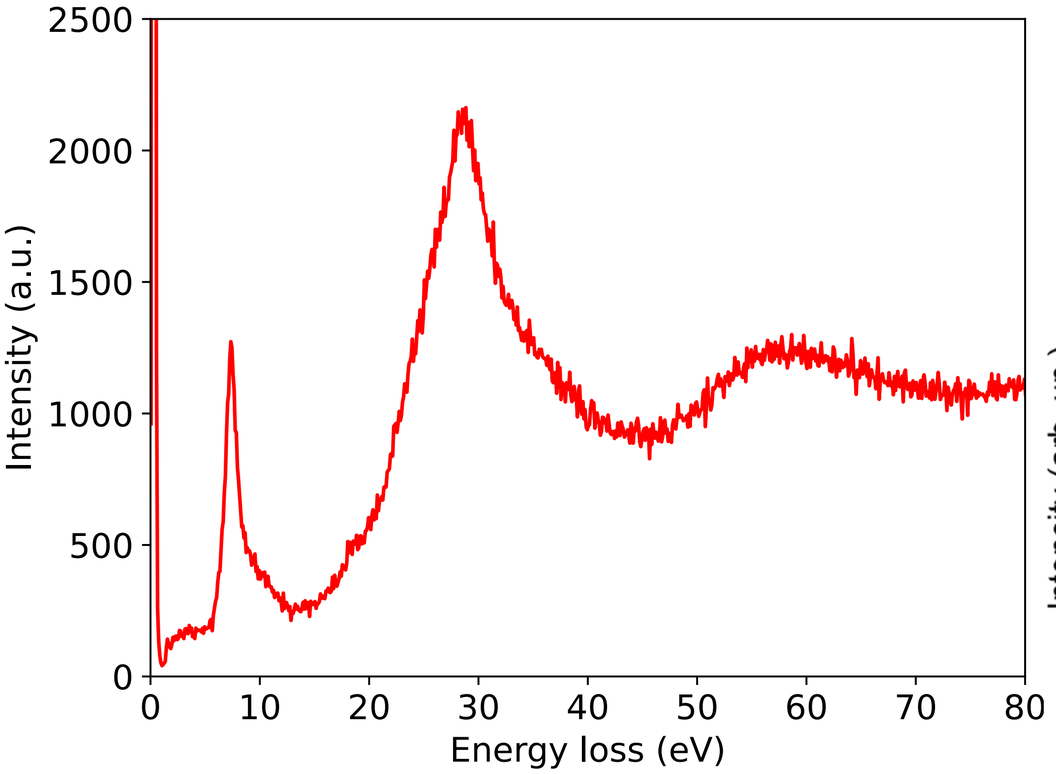


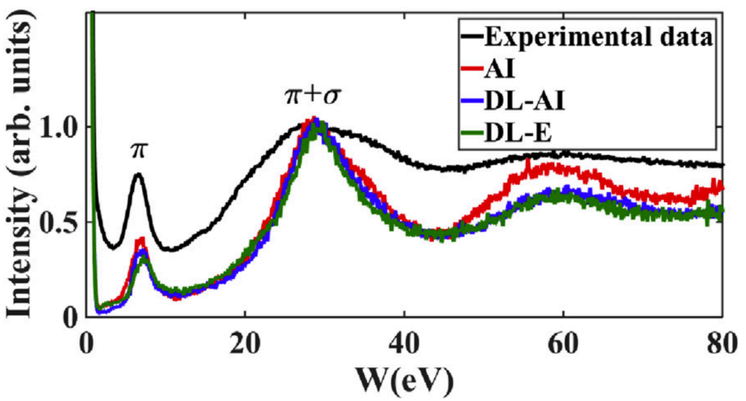
Well Reproduced Energy Loss

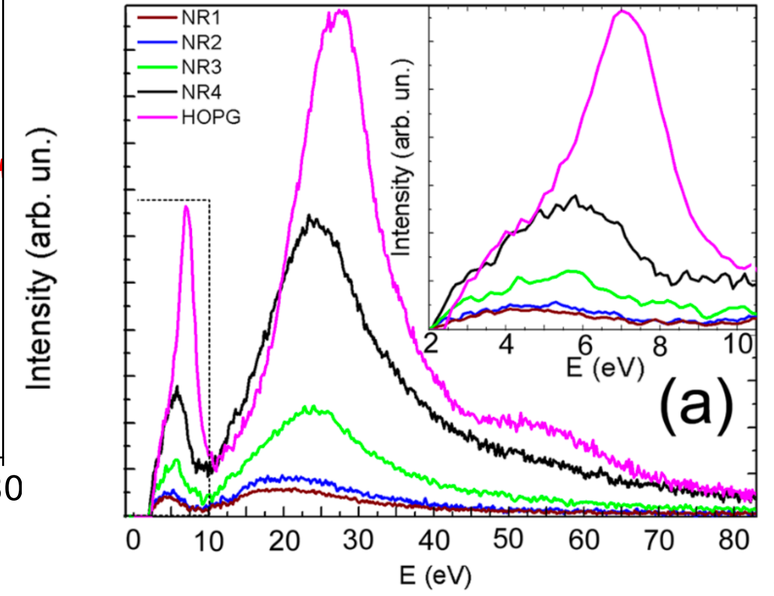
- Simulation results in agreement with measurements from literature
  - Azzolini et al. 10.1016/j.carbon.2017.03.041
  - Persichetti et al. (10.1063/1.3615802)

- π & $\pi + \sigma$ peaks near expected energies (~7 eV, 27 eV)

- two-plasmon excitation at higher energy (~60 eV) also reproduced

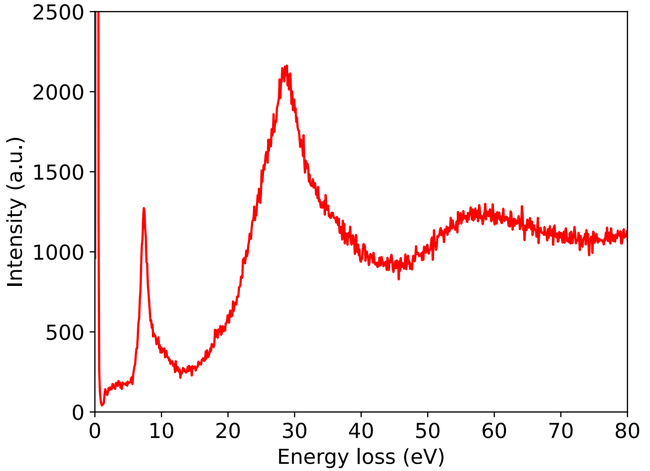






Recap & Next Step

- We found good agreement with literature for bulk graphite REELS
  - Some parameters still to be well understand and better defined
  - Just q-in-plane graphite component for now, should be weighted with q-out-of-plane one



Work ongoing, still lot of improvements to achieve!

- Next steps: improve graphite → simulate graphene → simulate graphane
  - What are the parameters?
  - Possible comparison with experimental energy loss for low primary energy (<1 keV)