Semi-empirical phonon calculations for graphene on different substrates

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Introduction

Graphene has some interesting properties:

- high charge-carrier mobility
- the ballistic electron transport at room temperature
- (...)

Practical applications: interaction with a substrate

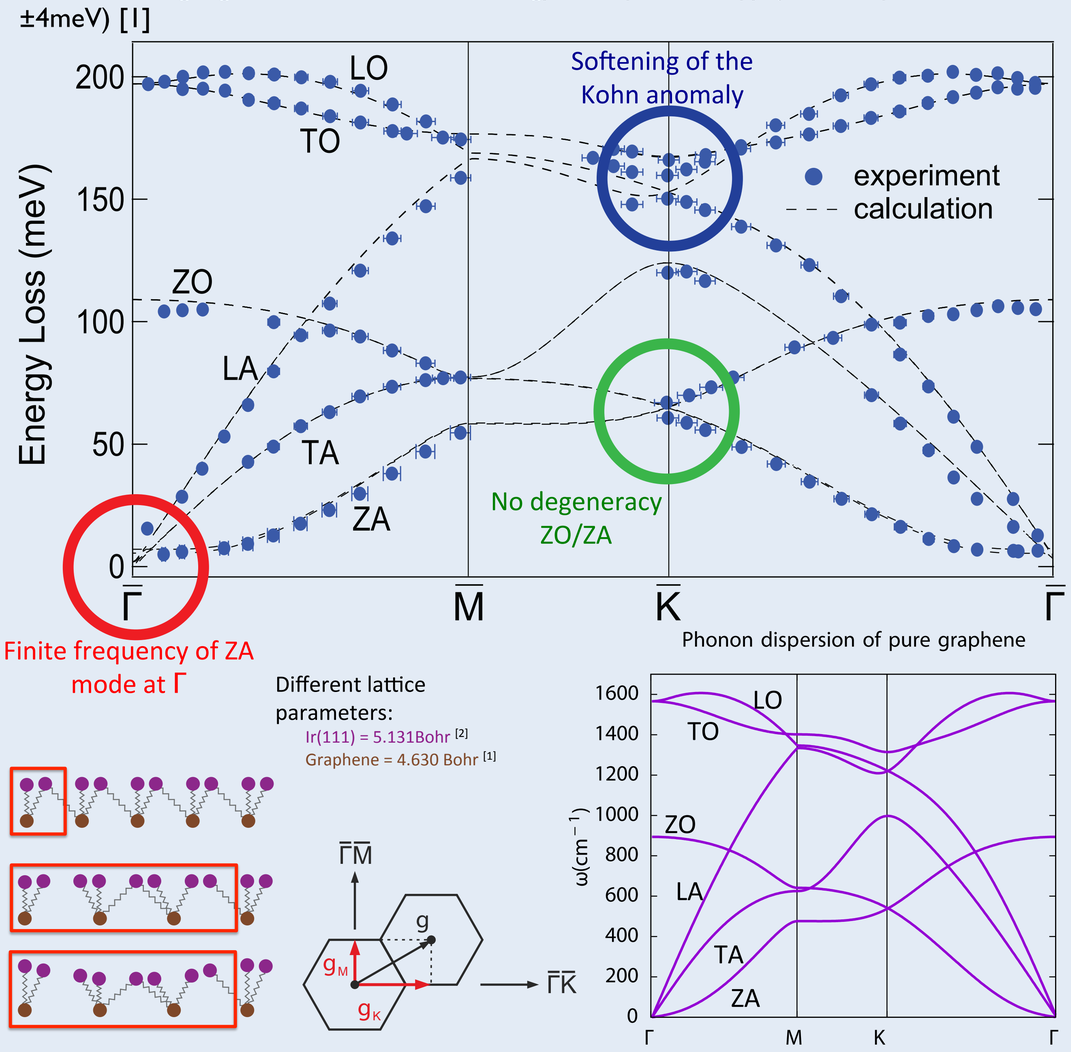
- Ir(111) and Pt(111) good candidates, large separation and weak interaction

Aim of the project:

- Create a force-constant model of graphene on Ir(111)
- Implement the model in a code
- Predict the phonon-dispersion
- Explain the characteristics of the phonon dispersion

Experimental Phonon dispersion of graphene on Ir(111)

Obtained using high resolution electron energy loss spectroscopy (HREELS precision of ±4meV) [1]

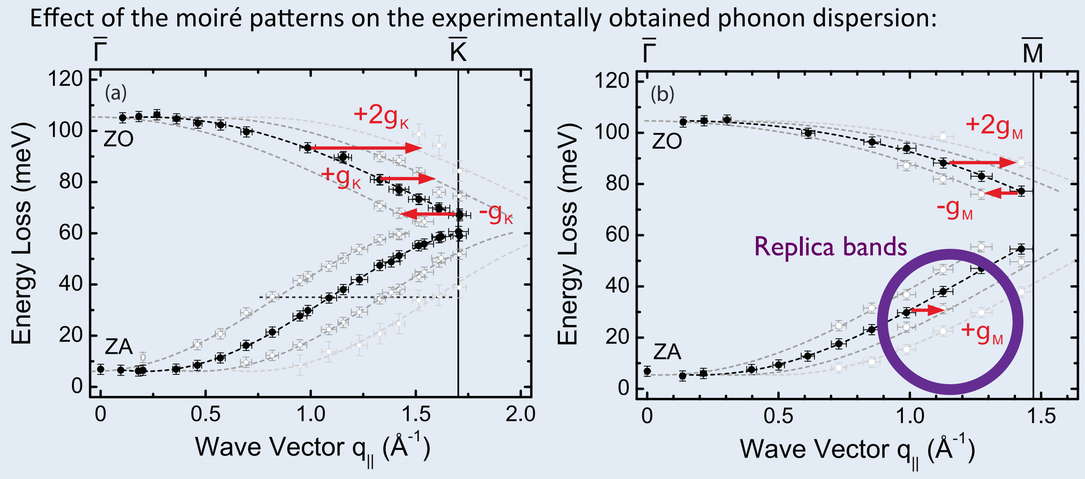


Finite frequency of ZA mode at Γ

Different lattice parameters:
Ir(111) = 5.131Bohr [2]
Graphene = 4.630 Bohr [1]

Phonon dispersion of pure graphene

Effect of the moiré patterns on the experimentally obtained phonon dispersion:



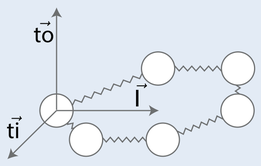
Force constant model

Force constant matrix of the interaction (local coordinates) of two atoms, depends on:

- Types of atoms interacting;
- Distance between them (using cubic splines).

$$C_n = \begin{pmatrix} \phi_n^l & \xi & 0 \\ \xi & \phi_n^{ti} & 0 \\ 0 & 0 & \phi_n^{to} \end{pmatrix}$$

off diagonal elements set to zero



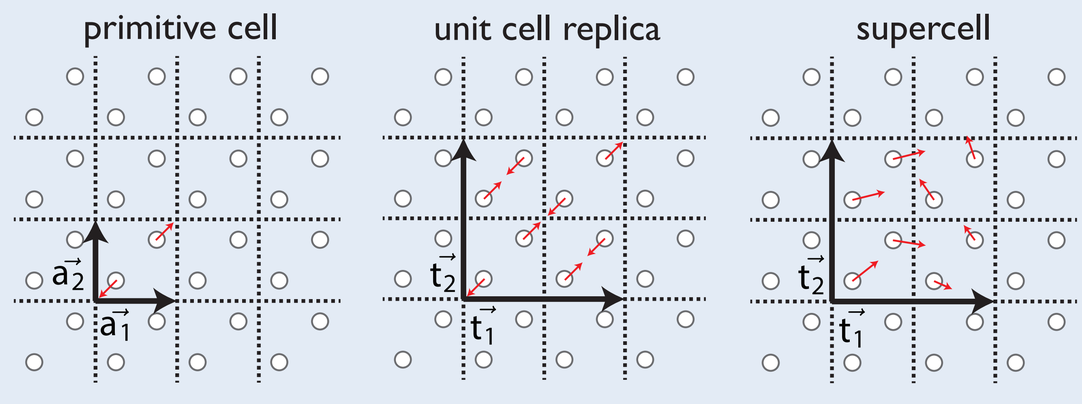
Transform to global coordinates: $C_n' = R^T C_n R$

Build the dynamical matrix: $D_{ij}(\mathbf{k}) = \frac{1}{\sqrt{m_i m_j}} \sum_s C_{ij}^s \exp(i\mathbf{k}\cdot\mathbf{r}_s)$

Calculate the phonon frequencies: $\det\left|D_{ij}(\mathbf{k}) - \omega^2(\mathbf{k})\right| = 0$

Force constants for pure graphene from the literature[3]

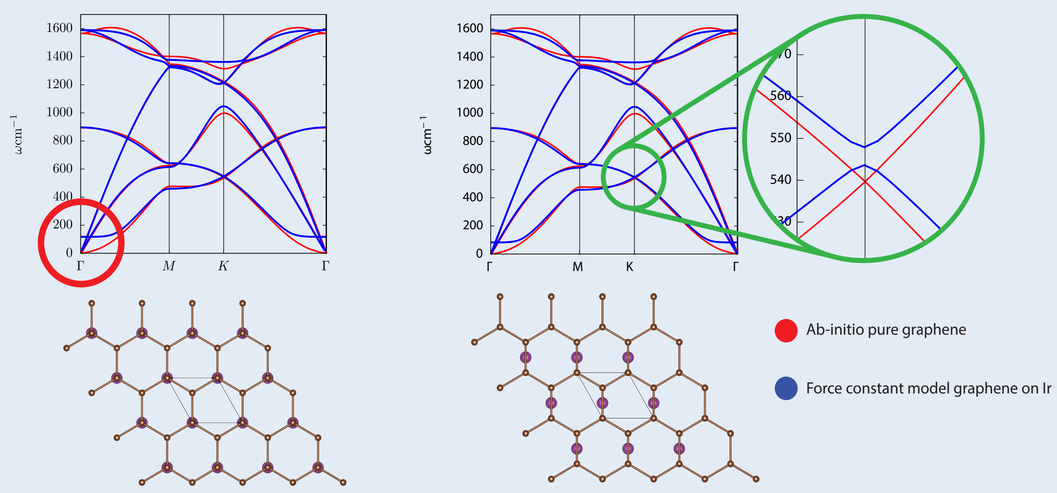
Folding of the phonon dispersion



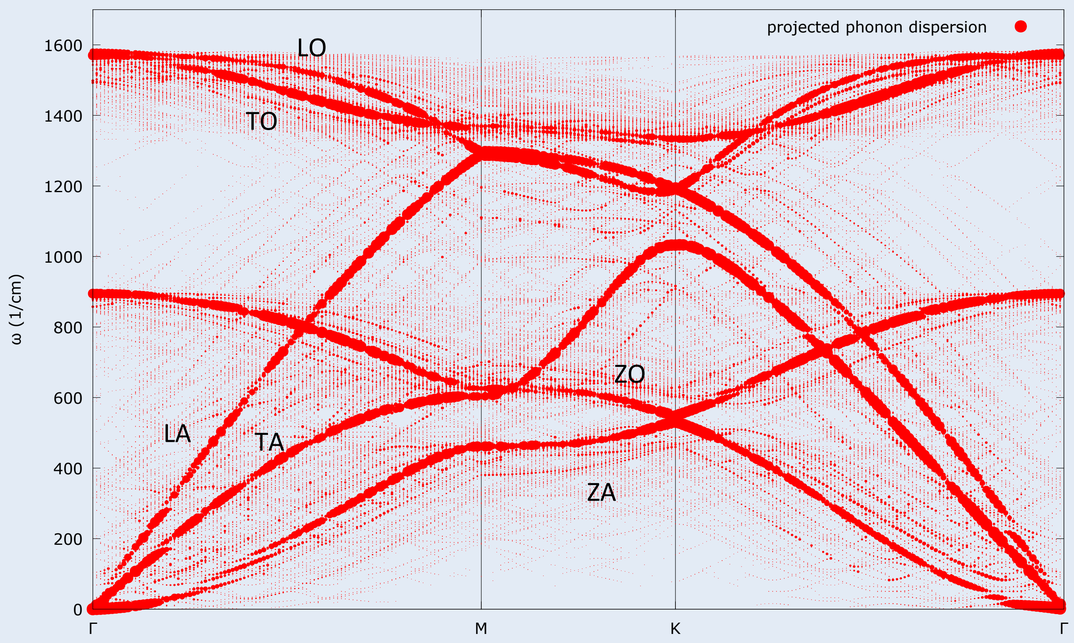
1. Find correspondence between the atoms of the supercell and the unit cell
2. Construct replicas of the unit cell with the size of the supercell
3. Project the phonons of the replica of the primitive cell in the supercell

Results

Unit cell of graphene with one Ir atom:



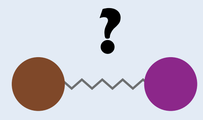
10x10 supercell of graphene on 9x9 Ir(1111) with 443 atoms with corrugation:



- Degeneracy of the ZO/ZA lifted
- First indication of the parallel phonon bands of a supercell

Conclusions

- **What the code can do:**
  - Easily tunable model
  - Fast: 443 atoms 250 kpoints takes 26min on a desktop (serial)
  - Get the force constants from fisrt-principles (ABINIT)
- **Further research:**
  - Quantitative interaction for Ir-C in graphene
  - Study different substrates
  - Study of the bandstructure



References

[1] M. Endlich, A. Molina-Sánchez, L. Wirtz, and J. Kröger, Phys. Rev. B 88, 205403 (2013).
[2] M. Endlich, A. Molina-Sánchez, H. Miranda, L. Wirtz, and J. Kröger (work in progress)
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