Correct handling of vacuum and slab thickness in ab-initio 2D materials

Lorenzo Sponza
François Ducastelle
Laboratoire d'Etude des Microstructures (LEM), CNRS – ONERA, Université Paris Saclay, 29 Avenue de la Division Leclerc, Châtillon, France
lorenzo.sponza@onera.fr

In most ab-initio simulation packages, the system is implicitly assumed periodic in the three Cartesian directions. But this is not the case for isolated nano-objects like 2D sheets. The approach consisting in including some vacuum inside the simulation cell (supercell approach) has *(i)* limited feasibility because memory consumption and *(ii)* poor accuracy, especially for excited-states, because of fictitious and unavoidable long-range interactions. The most used way around consists in cutting by hand the Coulomb interaction between the replicas of the system (Coulomb truncation) [1, 2]. We show that this technique solves just a part of the problem, since an issue related to the normalization of the polarizability remains. As a result, even with the Coulomb truncation, the 2D polarizability depends on the amount of vacuum included in the simulation, and is therefore arbitrary to some extent.

We present a methodology, derived within the tight-binding formalism, which generalises recent works by Nazarov, Latini and Giorgetti [3, 4, 5] and results in a simple set of post-processing recipes that can be applied to any ab-initio calculation. As a proof of their validity, we show results on hexagonal boron nitride that do not depend on the size of the cell, nor on the method used at the ab-initio level (supercell or Coulomb truncation).

From the 2D polarizability, it is essential to go to the 2D dielectric function to make GW or BSE calculations, and to compare with experiments. We show that working in the exact 2D limit, leads to severe divergencies at finite momentum, that are cured only if a non-vanishing thickness is taken into account. As the thickness of a 2D material is quite a vague concept, we introduce a rigorous method to compute from the 2D polarizability an effective thickness, which is the relevant quantity for all screening processes.

References

[1] C. A. Rozzi, et al., Phys. Rev. B, 73 (2006), 205191
[2] S. Ismail-Beigi, Phys. Rev. B, 73 (2006), 233103
[3] V. U. Nazarov, New J. Phys., 17 (2015), 073018
[4] S. Latini, T. Olsen, and K. S. Thygesen, Phys. Rev. B, 92 (2015), 245123
[5] N. Tancogne-Dejean, C. Giorgetti, and V. Véniard, Phys. Rev. B, 92 (2015), 245308

Figures

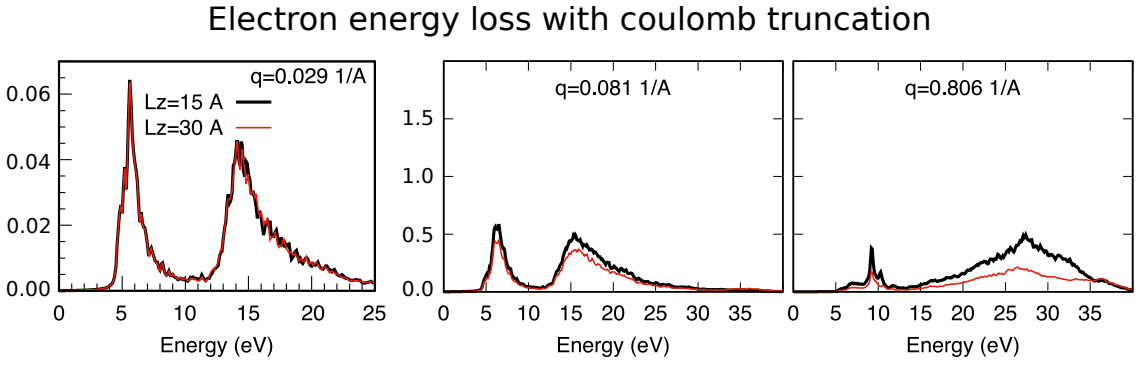

Figure 1: Dependence on the cell-size (Lz) of electron energy loss spectra computed with the Coulomb truncation for increasing momentum q (given in inverse Angstrom, 1/A).