# Numerical simulation of electron holography response in folded graphene sheets

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The analysis of the response in transmission electron microscopy, and in particular in electron holography depends ultimately on the electromagnetic field experienced by the electron beam within the sample. However dealing with nanostructures it is not always clear how to relate quantitatively the observed response to the average value of the inner potential integrated across the sample thickness. The Density Functional Theory (DFT) approach provides a set of numerical tools to compute various properties of bulk and nanoscopic materials; specifically, the set of Kohn-Sham equations yields a self-consistent and complete estimate of the inner potential experienced by the electrons. By retaining a sufficient number of basis elements, one can quantify the inner potential and density fields with a sub-angstrom resolution.

However the actual possibility of carrying out an analysis of this type is limited by three factors:
(a) in a plane-wave setting the infinitely extended directions are easily treated but for the spatially limited ones a supercell scheme is needed;
(b) one has to adopt a suitable transferable form of the exchange-correlation terms and associated pseudopotential;
(c) the total number of atoms in a unit supercell is necessarily limited by computational resources, ranging from hundreds to a few thousands with current high-performance parellel machines.

In this contribution we perform a systematic study of aspects related to (a) and (b), on flat multilayer graphene sheets with up to ten layers, in different stacking configurations [1], employing different proposals of van der Waals exchange-correlation schemes [2,3], to check how the binding energies, interlayer distances, vacuum fields and integrated profiles compare to experimental data. In these cases the approach is fully of DFT nature, meaning that first the nanostructures are relaxed towards minum energy configurations and then the self-consistent fields are post-processed to give the desired response. The resulting dependence on the layers separation and number provides an estimate of a surface- and an interlayer term to be taken as references values in holographic experiments, especially close to folded edges where the layers distance locally increases. Moreover it is seen that the dependence on the relative orientation is weak, probably below the current experimental reach.

As far as point (c) is concerned, we have extended and tested the method above using an effective two-stage strategy. When the number of atoms is prohibitively large for a full DFT relaxation, the optimal configurations are first determined by means of a molecular dynamics approach using suitable force fields that yield practically the same shapes as with DFT for samples where the data are available. The larger structures we have are interested in [4] are both multilayer folded graphene membranes and large squeezed chiral nanotubes that can be viewed also as folded graphene sheets with chiral folding axis. Once the optimal configuration is available from the first stage the inner charge and potential field can be extracted fully at the quantum level in a final self-consistent "snapshot", allowing for a quantitative numerical counterpart to be compared with microscopy findings.
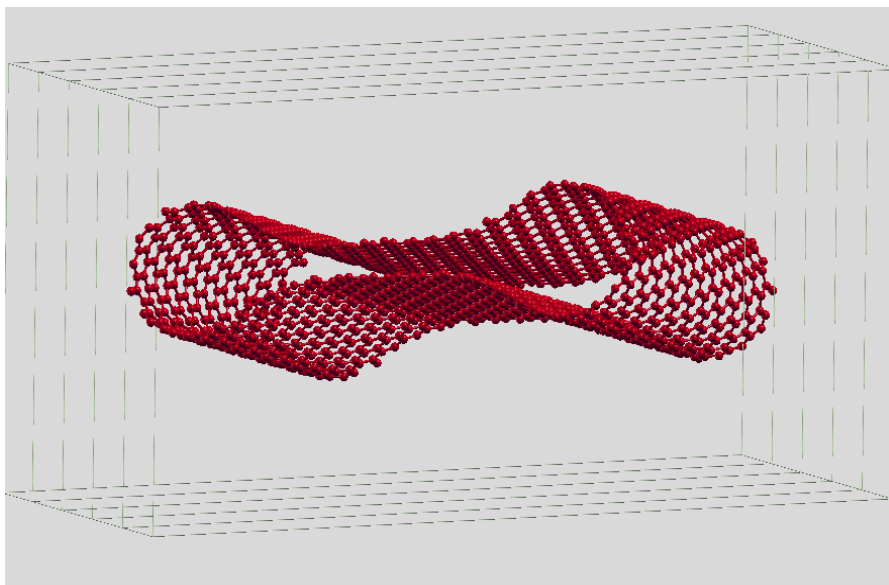


Figure 1: Graphene folded edge simulated locally as a squeezed chiral nanotube

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