Time-Dependent Density Functional Theory Calculations on Graphene Nanoflakes: Optical Properties and Electron Energy Loss Spectroscopy

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Abstract

The unique properties of graphene[1], such as large conductivity, high mechanical strength, high thermal stability, tunable optical properties[2], have led to an intensive study since it was first isolated in 2004. It has become one of the most promising materials for applications such as optical signal processing and quantum information. Part of this interest stems from the possibility to control the properties of graphene by means of chemical and physical modifications. Apart from extended graphene structures, also graphene quantum dots, antidots, nanoribbons and moebius strips have attracted the interest of the scientific community.

In this work we focused our attention on the optical properties of hydrogen saturated and chemically modified graphene nanoflakes. We investigated in detail how the optical properties depend on the geometry and the size of the flakes. We performed our calculation with an efficient Time-Dependent Density Functional Theory code[3] based on the use of basis set of localized functions (atomic orbitals). In conjunction with the ab-initio SIESTA package[4] this allows to study flakes containing more than a thousand atoms.

We have chosen hydrogen-saturated hexagonal and rectangular planar flakes to characterize size and shape dependence of optical absorption.

Moreover, we found that chemically functionalized edges affect the absorption spectrum. We saturated the carbon flakes with oxygen, fluorine and hydroxyl groups. The obtained results open the way for tuning optical properties by changing the edge functionalization.

We also explored the spatial distribution of the density change induced by an external electric field with a given frequency.

In order to further characterize the optical excitations an alternative approach has been used. Casida's approach[5] allows us to get information about the nature of the single optical excitations and can help to characterize plasmonic peaks in the optical spectrum.

Finally, we developed a code for calculating electron energy loss spectroscopy (EELS) for finite systems. These calculations will allow us to analyze further the electronic excitations of nanostructures. In particular, we focused our attention on nanoflakes of graphene.

The results improve our understanding of graphene properties that could be useful for applications in the field of optical signal processing and quantum information.

References

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