Linear absorption of single-layer graphene deposited on quartz: density-functional tight binding vs optical measurements

Andrés I. Bertoni[1], Kristyna Gazdova[2], Jan Hrabovsky[2], Cristián G. Sánchez[1], Inam Mirza[2], Thibault J.-Y. Derrien[2,*], and Nadezhda M. Bulgakova[2]

[1] Instituto Interdisciplinario de Ciencias Básicas (ICB-CONICET), Universidad Nacional de Cuyo, Padre Jorge Contreras 1300, Mendoza 5502, Argentina
[2] HiLASE Centre, FZU Institute of Physics, Za Radnici 828, 252 41 Dolni Brezany, Czech Republic
[*]Corresponding author email: derrien@fzu.cz

Fine-tuning of applications based on 2D materials requires advanced characterization of their optical and electronic properties in conditions that are compatible with industry standards. While the related studies can be realized in free-standing conditions or at low temperatures [1–3], identifying the contribution of the substrate in ambient conditions of laser processing is in high demand for the development of applications [4–6]. Moreover, the currently available optical measurements show some dependence on the chosen method [7, 8]. In such circumstances, *ab initio* modeling appears as an important path to gaining knowledge for the development of applications [9, 10].

In this work, we used the density functional tight-binding (DFTB) formalism [11, 12] to study the optical response of a flake of graphene deposited on a sample of quartz. After optimizing the separation between flake and substrate, the effect of lattice temperature on the optical response of the graphene flake was estimated using an ensemble average of absorption spectra corresponding to multiple configurations extracted from a quantum force-field (Born-Oppenheimer) molecular dynamics, with a Nosé-Hoover thermostat. Each absorption spectrum was calculated from the linear response to a Dirac-delta electric field perturbation at the initial time, within the dipolar approximation. The computational results were compared with experimental measurements of transmission and reflection spectra, from 200 nm up to 2.5 μm, using a commercial monolayer graphene sample deposited on fused silica.

Beyond the role of the shape and size of the flake in its optical response [13], it was found that its deposition on a quartz substrate can affect its Fermi energy and linear absorption spectra. Our computational protocol is a step forward in simulating challenging systems under realistic conditions while providing intuitive insights for the design of nanoelectronic applications based on usage of 2D materials.

References

[1] S. A. Sato, H. Hirori, Y. Sanari, Y. Kanemitsu, A. Rubio, *Phys. Rev. B.* ***103***, L041408 (2021).
[2] I. Gierz *et al., J. Phys. Condens. Matter.* **27**, 164204 (2015).
[3] A. H. Castro Neto et al., *Rev. Mod. Phys.* ***81***, 109–162 (2009).
[4] K. A. Drogowska-Horna *et al., Nano Res.* ***13***, 2332–2339 (2020).
[5] M. Kasischke, S. Maragkaki, S. Volz, A. Ostendorf, E. L. Gurevich, *Appl. Surf. Sci.* ***445***, 197–203 (2018).
[6] F. P. Bonafé *et al., J. Chem. Theory Comput.* ***16***, 4454–4469 (2020).
[7] B. Song *et al., Appl. Surf. Sci.* ***439***, 1079–1087 (2018).
[8] K. K. Tikuišis *et al., Phys. Rev. Mater.* **7**, 044201 (2023).
[9] T. J.-Y. Derrien *et al., Phys. Rev. B.* ***104***, L241201 (2021).
[10] Gindl, Adam *et al., Submitt. Nat Photonics* (2023) (available at https://arxiv.org/abs/2310.07254).
[11] B. Hourahine *et al., J. Chem. Phys.* ***152***, 124101 (2020).
[12] A. I. Bertoni, T. J.-Y. Derrien, C. G. Sanchez, in *Density Functional Theory: Fundamental Theory, Key Methods, and Applications* (Elsevier, A. Kuznetsov.).
[13] C. Mansilla Wettstein, F. P. Bonafé, M. B. Oviedo, C. G. Sánchez, *J. Chem. Phys.* ***144***, 224305 (2016).