Colossal quantum transport anisotropy in nanoporous graphenes at room temperature

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During the last 15 years it has been shown that bottom-up on-surface synthesis is a powerful technique to realize carbon nanostructures with atomic precision.[1] Good examples are graphene nanoribbons (GNRs) which are semiconducting nano-tapes of graphene acquiring increasing attention for carbon nanoelectronics and quantum technologies.[2,3] Nanoporous graphenes (NPGs) made as 2D arrays of laterally bonded GNRs[4] are also very appealing in that regard, as the coupling between neighboring GNRs may be tuned via chemical design. Concretely, it has been shown that laterally connecting the GNRs with phenyl rings in meta-position leads to destructive quantum interference (QI), thus electronically decoupling the GNRs which become isolated 1D nanowires.[5] Such type of novel phenylated NPGs have recently been experimentally fabricated.[6] However, though QI has been shown to persist at room temperature in single-molecule devices,[7] a priori it is not clear whether QI-effects as embedded within carbon nanostructures also survive at 300 K, which is an essential step towards their technological exploitation. In this work, we have combined molecular dynamics (MD) simulations with quantum transport calculations to evaluate how transport properties of different NPGs, including the meta-NPG (Fig. 1a), are affected by thermal vibrations at 300 K. We build tight-binding (TB) Hamiltonian models that capture the effect of bond vibrations on the NPG electronic structure. Our results show that NPGs transport anisotropy, defined from the conductivities as $A = \sigma_y/\sigma_x$ (Fig. 1b), survives thermal fluctuations. However, only meta-NPG reaches colossal quantum transport anisotropy, due to QI. Our work thus demonstrates the power of QI-engineering to control charge current flow in carbon nanostructures at room temperature, which is an essential step towards its application in real devices. Though the qualitative picture of our results should not change, a closer quantitative agreement with experiments could be reached by using suitably AI-generated inter-atomic potentials and TB models.[8,9] Concretely, our current models completely neglects multiple conformational effects beyond bond vibrations (e.g. phenyl ring rotation, bending, GNR torsion, etc). However, the structural flexibility of NPGs, or any carbon nanostructure, makes it a challenging task to properly include, by hand, all relevant terms affecting the materials electronics. In that regard, the possible use of AI to automatically create sophisticated accurate models, capturing all relevant conformational effects, is also discussed.

References

[1] Grill, L. *et al. Nat. Nanotechnol.* 2, (2007) 687–691.

[2] Cai, J. *et al. Nature* 466, (2010) 470–3.

[3] Wang, H. *et al. Nature Reviews Physics* 3, (2021) 791–802.

[4] Moreno, C. *et al. Science* 360, (2018) 199–203.

[5] Calogero, G., Alcón, I., Papior, N., Jauho, A.-P. & Brandbyge, M. *J. Am. Chem. Soc.* 141, (2019) 13081–13088.

[6] Moreno, C. *et al. J. Am. Chem. Soc.* 145, (2023) 8995.

[7] Liu, S. X., Ismael, A. K., Al-Jobory, A. & Lambert, C. J. *Acc. Chem. Res.* 56, (2023) 322–331.

[8] Deringer, V. L. *et al. Chemical Reviews* **121**, 10073–10141 (2021)

[9] Wang, Z. *et al. npj Comput. Mater.* **7**, 1–10 (2021)

Figures

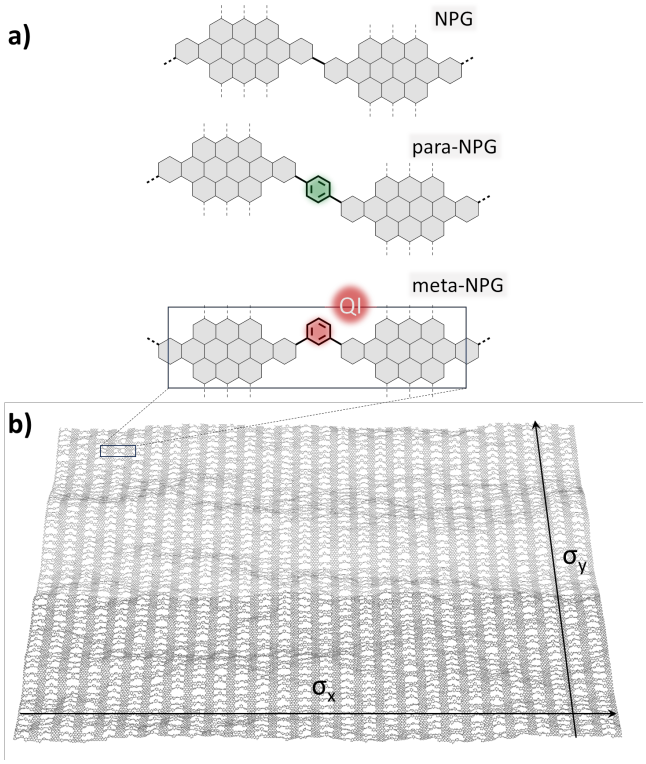

Figure 1. a) Chemical outline of inter-ribbon connections within the original NPG and the phenyl-bridged para- and meta-NPGs, the latter hosting QI. **b)** Thermally activated 50x50 nm^2 meta-NPG sample at 300 K, indicating the relevant in-plane transport directions (x, y).