Silicon passivation of zigzag graphene edge enabling robust spin-polarized nanogap quantum transport

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INTRODUCTION

A major motivation for the research on graphene stems from the ferromagnetically-aligned spin-polarized states appearing at the zigzag graphene edges (ZGEs) in view of spintronic applications [1]. However, in spite of the significant progress made in both top-down and bottom-up synthesis methods, the reliable preparation of magnetic ZGE states in ambient conditions remains a formidable challenge [2]. The preferable condition for the existence of spin-polarized ZGE states is the preservation of sp^2-type edge C atoms, which liberates the π electrons from p_z orbitals of carbons to populate the edge states. Unfortunately, however, the sp^3-type hydrogenated ZGE C configurations are thermodynamically favored over their sp^2 counterparts across a wide range of thermodynamic conditions, representing a serious bottleneck in realizing ZGE-based spintronics.

Meanwhile, because Si is an essential element in the existing semiconductor industry, there has been significant interest in elucidating the nature of interactions between Si or its native oxide, SiO_2, and C nanomaterials in the context of electronic and energy device applications. Particularly, we note recent experiments reported that Si atoms can effectively passivate the edge of graphene nanopores and prevent the healing of nanopores upon the supply of extra C atoms [3]. In related works, an extended linear array of Si atoms along the ZGE was observed by transmission electron microscopy. Given the feasibility of Si-based graphene edge stabilization provided in these experiments, whether the Si edge passivation preserves the ferromagnetic (FM) ZGE states would be an intriguing question. In particular, notable achievements were recently made toward graphene edge-based tunnel junctions [4], bringing us closer to the realization of atomically thin sensors including DNA sequencers with single-molecule resolution.

COMPUTATIONAL DETAILS

Spin-polarized DFT calculations were performed using the SIESTA code [5] within the Perdew-Burke-Ernzerhof parameterization of generalized gradient approximation. Double ζ-plus-polarization-level numerical atomic orbital basis sets were employed together with the Troullier-Martins type norm-conserving nonlocal pseudopotentials. The mesh cutoff of 200 Ry for the real-space integration, and the $1 \times 1 \times 64$ (32 and 20) $\vec{k}$-points were sampled for the two (four and six) primitive ZGNR unit-cell models. The atomic geometries were optimized until the total residual forces are converged below 0.02 eV/ Å.

For the quantum transport calculation, we used the DFT-based MGF method implemented within the TranSIESTA code [6]. The surface green functions, g_s, for the semi-infinite electrode regions were extracted from separate DFT calculation for the ideal planar graphene structure with 16 $\vec{k}$-point sampling along the infinite direction (y-axis), and 20 $\vec{k}$-points sampling along the charge transport direction.

REFERENCES

[1] Han, W *et al.* Nat. Nanotechnol. 9 (10), 794-807. (2014)
[2] Magda, G. Z *et al.* Nature 514 (7524), 608-611 (2014)
[3] Lee, J *et al.* Proc. Natl. Acad. Sci. U. S. A111 (21), 7522-7526. (2014)
[4] Bellunato, A *et al*., Nano Lett. 18 (4), 2505-2510. (2018)
[5] José, M. S *et al*., J. Phys. Condens. Matter, 14 (11), 2745. (2002)
[6 Brandbyge, M *et al*, Phys. Rev. B: 65 (16), 165401. (2002)