Electronic and transport properties of unbalanced sublattice Nitrogen-doping in Graphene

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Abstract

The incorporation of foreign atoms into the carbon honeycomb lattice has been widely investigated in order to modify the electronic and chemical properties of carbon-based materials [1,2]. In contrast with conventional materials, the effect of foreign atoms in a 2D material, such as graphene, is expected to depend significantly on the position and surrounding of each atom due to the quantum confinement of the electrons [2]. Recent scanning tunneling microscopy and spectroscopy studies of nitrogen doped graphene have revealed how the incorporation of this foreign atom into the sp^2 lattice occurs. Joucken and coworkers showed that the exposure of graphene to a nitrogen plasma flux after synthesis leads to a homogeneous distribution of substitutional atoms [3]. However, when a nitrogen source is introduced during the CVD growth of graphene, the nitrogen incorporation exhibits a preferential accommodation within one of the two triangular sublattice that compose the honeycomb lattice [4,5,6]. This wayward incorporation of nitrogen atoms into graphene is not hitherto understood [5]. Nevertheless, the consequences of this peculiar atom arrangement on the electronic and transport properties of graphene are addressed in this work.

Electronic structure and transport properties of nitrogen-doped graphene with a single sublattice preference are investigated using both first-principles techniques and a real-space Kubo-Greenwood approach [7]. Such a break of the sublattice symmetry leads to the appearance of a true band gap in graphene electronic spectrum. A band gap opening due to an ordered superlattice of dopants has already been discussed [8,9]. However, such a periodic doping configuration is rather difficult to envisage experimentally. In this work, we demonstrate the robustness of the band gap opening for the case of a random distribution of dopants in the same sublattice. In addition, a natural spatial separation of both types of charge carriers at the band edge is observed, leading to a highly asymmetric electronic transport. For such N-doped graphene systems, the carriers at the conduction band edge present outstanding transport properties with long mean free paths, high conductivities and mobilities. This phenomenon is explained by a non-diffusive regime, and originates from a low scattering rate. The fact that corresponding electrons reside mainly in the unaltered sublattice explains such low scattering rate.

The presence of a true band gap along with the persistence of carriers traveling in an unperturbed sublattice suggest the use of such doped graphene in GFET applications, where a high I_{ON}/I_{OFF} ratio is needed. The present simulations should encourage more investigation and specific measurements on N-doped graphene samples where such an unbalanced sublattice doping has been observed.

References

[1] P. Ayala, R. Arenal, A. Loiseau, A. Rubio, and T. Pichler, Rev. Mod. Phys. **82** (2010) 1843.
[2] M. Terrones, A. Filho, A. Rao. Doped Carbon Nanotubes: Synthesis, Characterization and Applications in Carbon Nanotubes Springer (2008) 531.
[3] F. Joucken, Y. Tison, J. Lagoute, et al. Phys. Rev. B **85** (2012) 161408(R).
[4] L. Zhao, R. He, K.T. Rim et al., Science **333** (2011) 999.
[5] A. Zabet-Khosousi, L. Zhao, L. Palova et. al., JACS **136** (2014) 1391.
[6] R. Lv, Q. Li, A.R. Botello-Mendez et al., Nature Scientific Reports **2** (2012) 586.
[7] A. Lherbier, A. R. Botello-Mendez, J.-C. Charlier, Nano Lett.**13** (2013), 1446.
[8] R. Martinazzo, S. Casolo, and G.F. Tantardini, Phys. Rev. B **81** (2010) 245420.
[9] S. Casolo, R. Martinazzo, and G.F. Tantardini, J. Phys. Chem. C **115** (2011) 3250.

Figures

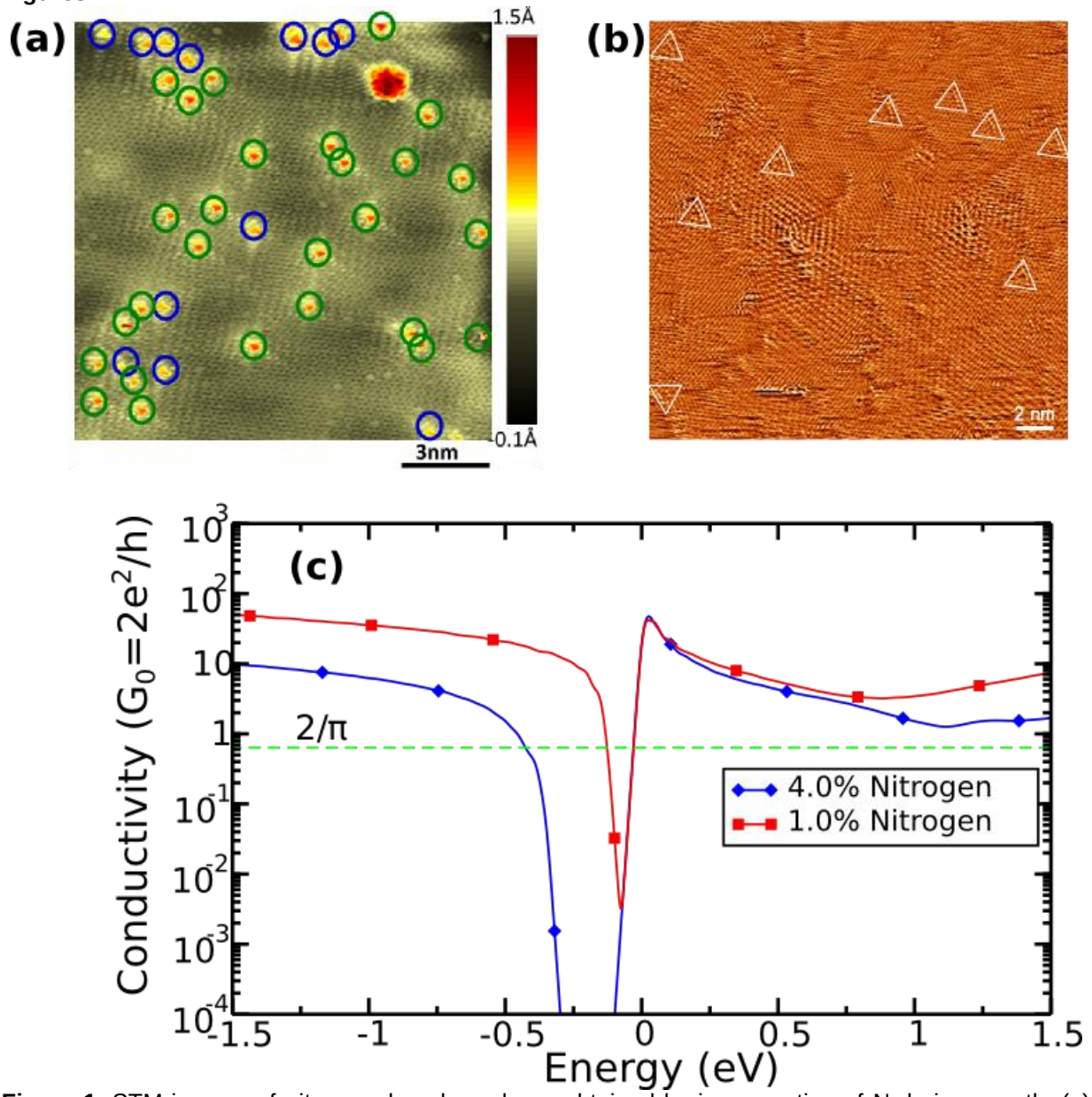

Figure 1. STM images of nitrogen doped graphene obtained by incorporation of N during growth: (a) single substitution [4], and (b) double substitution [6]. (c) Calculated semiclassical conductivities in graphene for various concentrations of N dopants randomly distributed in one sublattice [7].