Localized state and charge transfer in nitrogen-doped epitaxial graphene

Frederic Joucken[1*], Yann Tison[2], Jerome Lagoute[2], Jacques Dumont[1], **Damien Cabosart**[3*], Bing Zheng[3], Vincent Repain[2], Cyril Chacon[2], Yann Girard[2], Andres Rafael Botello-Mendez[3], Sylvie Rousset[2], Robert Sporken[1], Jean-Christophe Charlier[3], and Luc Henrard[1]

[1] *Research Center in Physics of Matter and Radiation (PMR), University of Namur (FUNDP), 61 Rue de Bruxelles, 5000 Namur, Belgium*
[2] *Laboratoire Materiaux et Phenomenes Quantiques, UMR7162, Universite Paris Diderot Paris 7, Sorbonne Paris Cite, CNRS, UMR 7162 case courrier 7021, 75205 Paris 13, France*
[3] *Universite catholique de Louvain (UCL), Institute of Condensed Matter and Nanosciences (IMCN), 1/6 Place L. Pasteur, 1348 Louvain-la-Neuve, Belgium*
**two presenting authors*

Contact : fjoucken@fundp.ac.be

Nitrogen-doped epitaxial graphene grown on SiC(000-1) was prepared by exposing the surface to an atomic nitrogen flux. The atomic nitrogen was produced by a remote RF plasma source. The exposure results in the creation of nitrogen-related defect sites in the graphene lattice, the concentration of which depends on the exposure time (the settings of the plasma are held constants for each sample).

Using Scanning Tunneling Microscopy (STM) and Spectroscopy (STS), supported by Density Functional Theory (DFT) calculations, the simple substitution of carbon by nitrogen atoms has been identified as the most common doping configuration, as in ref. [1] (although the production method is different). High resolution images reveal a reduction of local charge density on top of the nitrogen atoms, indicating a charge transfer to the neighboring carbon atoms. For the first time, local STS spectra clearly evidenced the energy levels associated with the chemical doping by nitrogen, localized in the conduction band. To highlight this state, the tunneling spectra above the defect and above the graphene have been taken at constant tip's height. This allowed to unambiguously identify the donor-state of the substitutional nitrogen.

The shift of the Dirac point and the n-doping level associated have been estimated, giving a charge transfer of 0.8 electron by dopant atom. However, STS should be completed by a complementary technique (as ARPES) to confirm this point as it is known that the tunneling spectra of graphene around the Fermi level are perturbed by the absence of a phonon-induced channel (ref [2]).

Various other nitrogen-related defects have been observed. The bias dependence of their topographic signatures demonstrates the presence of structural configurations more complex than substitution as well as hole-doping. Additional work is being carried out to acquire the STS signature of those more complex defects in order to identify them confidently.

[1] Liuyan Zhao, Rui He, Kwang Taeg Rim, Theanne Schiros, Keun Soo Kim, Hui Zhou, Christopher Gutiérrez, S. P. Chockalingam, Carlos J. Arguello, Lucia Pálová, Dennis Nordlund, Mark S. Hybertsen, David R. Reichman, Tony F. Heinz, Philip Kim, Aron Pinczuk, George W. Flynn, Abhay N. Pasupathy, "Visualizing Individual Nitrogen Dopants in Monolayer Graphene", Science 333, 999 (2011)

[2] Y. Zhang, V. W. Brar, F. Wang, C. Girit, Y. Yayon, M. Pan-lasigui, A. Zettl, and M. F. Crommie, Giant "phonon-induced conductance in scanning tunneling spectroscopy of Gate-tunable graphene", Nat. Phys. 4, 627 (2008).