doi:10.1017/S1431927613011598
Microsc. Microanal. 19 (Suppl 2), 2013

Direct Observation of the Optical Response of Twisted Bilayer Graphene by Electron Energy Loss Spectroscopy.

Leonardo Basile[1,2], Wu Zhou[2], Juan Salafranca[3,2] and Juan-Carlos Idrobo[2]

[1.] Departamento de Física, Escuela Politécnica Nacional, Quito, Ecuador.
[2.] Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, USA.
[3.] Departamento de Física Aplicada III, Universidad Complutense de Madrid, Spain.

Recently, it has been observed through conductivity measurements and Raman spectroscopy that twisted bilayer graphene presents anomalies in its optical response [1,2]. In particular, the optical absorption of bilayer graphene shows peaks at certain misorientation angles between the graphene layers [1], Similarly, wide-field Raman images show unexpected features related to misoriented domains of the graphene layers when compared to single layer graphene domains [2].

In this study, we have measured the optical response of twisted bilayer graphene using electron energy-loss spectroscopy (EELS) in an aberration-corrected scanning transmission electron microscope (STEM). The experiments were performed with a Nion UltraSTEM™ 100, equipped with a cold field emission electron source, a corrector of third and fifth order aberrations, and a Gatan Enfina spectrometer [3]. The microscope was operated at 60 kV acceleration voltage to avoid knock-on damage of the graphene lattice. A semi-convergence angle of 30 mrad, and 54 to 200 mrad semi-collection angles were used to obtain the medium angle annular dark field (MAADF) images. The spectra were collected with an energy resolution of ~350 meV and with an energy dispersion of 50 meV/pixel.

The left panel in Figure 1 shows a set of experimental MAADF images of twisted bilayer graphene obtained at different misorientation angles. The Moiré patterns formed by the interference between the graphene hexagonal honeycomb lattices are clearly observed. The respective simulated MAADF images, using the QSTEM simulation package [4], clearly reproduce the observed Moiré patterns. The MAADF image simulations were obtained by using commensurate unit cells with the smallest number of atoms [5].

The right panel in Figure 1 shows the respective EEL spectra acquired at different misorientation angles. A new absorption peak emerges at about 2.3 eV at misorientation angle between the graphene layers of ~14^{o}. The new absorption peak shifts towards the infrared (ultraviolet) region of the spectra as the misorientation angle decreases (increases). The results indicate that band gap tuning could be achieved by controlling the misorientation angle in twisted bilayer graphene.

References:

[1] Y Wang *et al*, ACS Nano **4** (2010), p. 4074.
[2] RW Havener *et al*, Nano Letters **12** (2012), p. 3162.
[3] OL Krivanek *et al,* Ultramicroscopy **108** (2008), p. 179.
[4] CT Koch in "Determination of core structure periodicity and point defect density along dislocations", PhD Thesis 2002, (Arizona State University, Phoenix), p. 1
[5] S Shallcross *et al,* Physical Review B **81** (2010), p. 1.

[6] This research was supported by the National Secretariat of Higher Education, Science, Technology and Innovation of Ecuador (SENESCYT) (LB), a Wigner Fellowship through the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U. S. Department of Energy (WZ); National Science Foundation through grant No. DMR-0938330 (WZ), ERC starting Investigator Award, grant #239739 STEMOX, and Juan de la Cierva program JCI-2011-09428 (MICINN-Spain) (JS), and Oak Ridge National Laboratory's Shared Research Equipment (ShaRE) User Program (JCI), which is sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy.

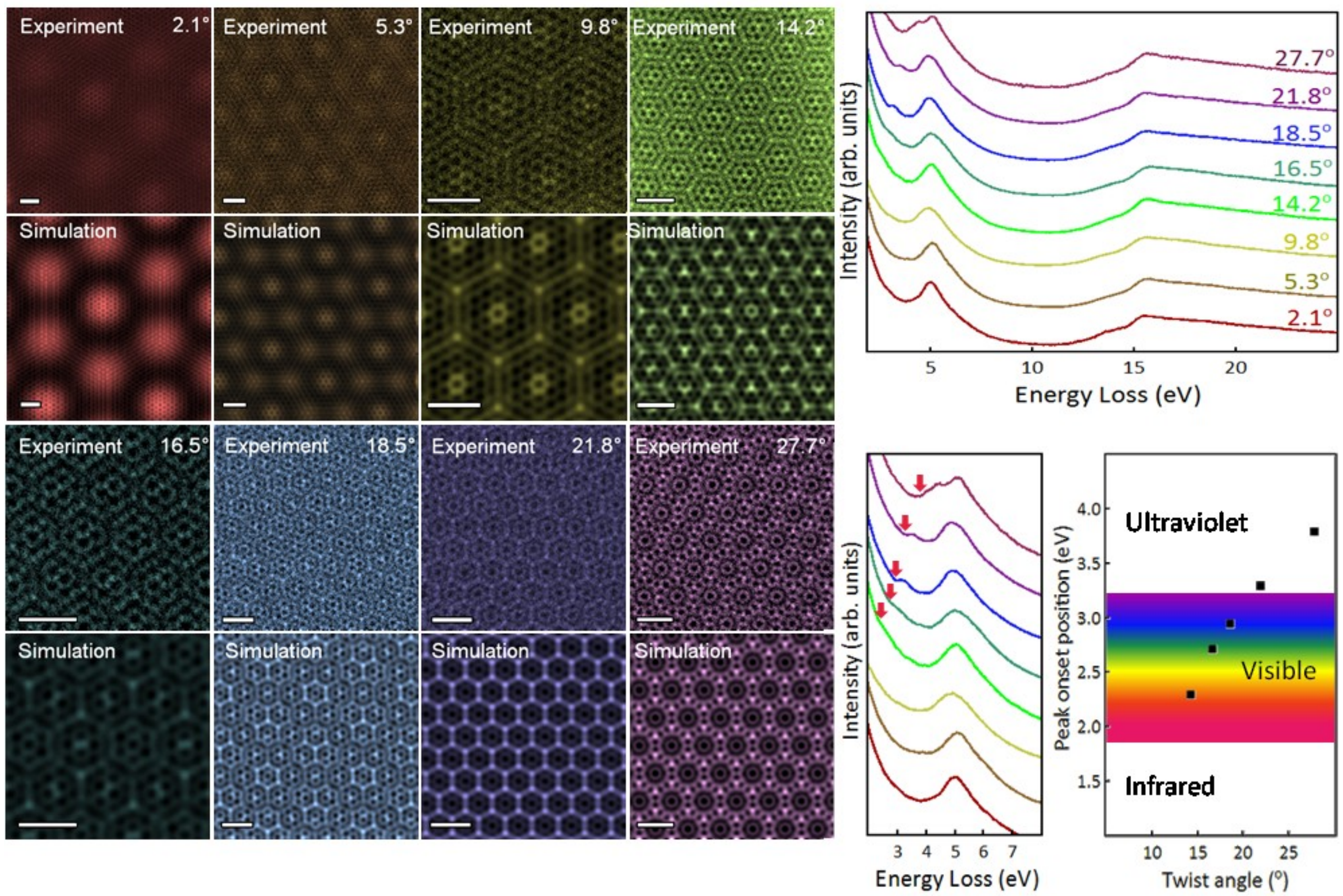

Figure 1. (Left) Experimental MAADF (top panel) and simulated images (bottom panel) of twisted bilayer graphene. Scale bars are 1 nm. (Right, top). EEL spectra obtained from twisted bilayer graphene samples. (Right, bottom) The EEL spectra present an absorption peak emerging at ~14^{o} and shifting to higher energies as a function of misorientation angle between the graphene layers. The red arrows in the spectra highlight the onset of the additional absorption peak and the plot on the right shows its angle dependence.