Twisted bilayer graphene: phonon dispersion of microscopic rainbows

Jessica Campos-Delgado[1], Luiz Gustavo Cançado[2], Gerardo Algara-Siller[3], Ado Jorio[2], Carlos A. Achete[4], Ute Kaiser[3], Jean-Pierre Raskin[1]

[1] ICTEAM, Université catholique de Louvain, 1348 Louvain-la-Neuve, Belgium
[2] Departamento de Física, Universidade Federal de Minas Gerais, Belo Horizonte, MG, 30123-970, Brazil
[3] Ulm University, Group of electron microscopy for materials science, Ulm, 89081, Germany
[4] Divisão de Metrologia de Materiais, INMETRO, Xerém, RJ, 25250-020, Brazil

jessica.campos@uclouvain.be

Abstract

In this work we study naturally produced bilayer graphene presenting different rotational angles. The production of the samples is carried out by low pressure CVD of methane using copper film as catalyst. The grown graphene layer is mostly a monolayer film with distributed bilayer and multi-layer graphene patches formed at the nucleation centers.

The rotational angle between layers has proven to influence the electrical properties of bi-layer graphene [1,2,3] and generates superlattice structures known as Moiré patterns [4].

We demonstrate that different colorations appear visually for certain misorientation angles of bilayer graphene when graphene is transferred on an optimal SiO_2 thickness of 100 nm stacked on top of a Si substrate, as exemplified in Figure 1A. In particular, Raman spectroscopy constitutes a powerful tool to probe bilayer graphene since the differences in orientation are sensed through the intensity of the G band [5,6] and the positions of the R bands [4]. We have studied these spectra of colorations combining Raman spectroscopy and high resolution transmission electron microscopy. Our investigations reveal that angles in the range of 9°-11° can be attributed to blue colorations, yellow colorations appear for rotational angles between 11° and 13° and finally pink-reddish colorations are present for angles of 13° up to 15° [7].

Moreover, we demonstrate that these superlattices provide θ-dependent $\boldsymbol{q}$ wavevectors that activate phonons in the interior of the Brillouin zone. We show this superlattice-induced Raman scattering can be used to probe the phonon dispersion in twisted bi-layer graphene [8]. We have found features in the 100-900 cm^{-1} region (see Figure 1B) that are successfully attributed to phonon branches of graphene (ZA, TA, LA, ZO phonons) and to layer breathing vibrations (ZO' phonons).

References

[1] J.M.B. Lopes dos Santos, N.M.R. Peres, A.H. Castro Neto. Graphene bilayer with a twist: electronic structure. Phys. Rev. Lett. 99, 256802, 2007.

[2] G. Li, A. Luican, J.M.B. Lopes dos Santos, A.H. Castro Neto, A. Reina, J. Kong, E.Y. Andrei. Observation of Van Hove singularities in twisted graphene layers. Nature Physics 6, 109-113, 2010.

[3] A. Luican, G. Li, A. Reina, J. Kong, R.R. Nair, K.S. Novoselov, A.K. Geim, E.Y. Andrei. Single-layer behavior and its breakdown in twisted graphene layers. Phys. Rev. Lett. 106, 126802, 2011.

[4] V. Carozo, C.M. Almedia, E.H.M. Ferreira, L.G. Cançado, C.A. Achete, A. Jorio. Raman signature of graphene superlattices. Nano Lett. 11, 4527-4534, 2011.

[5] R. Havener, H. Zhuang, L. Brown, R. G. Hennig, J. Park. Angle-resolved Raman imaging of interlayer rotations and interactions in twisted bilayer graphene. Nano Lett. 12, 3162-3167, 2012.
[6] K. Kim, S. Coh, L.Z. Tan, W. Regan, J.M. Yuk, E. Chatterjee, M.F. Crommie, M. L. Cohen, S.G. Louie, A. Zettl. Raman spectroscopy study of rotated double-layer graphene: misorientation-angle dependence of electronic structure. Phys. Rev. Lett. 108, 246103, 2012.
[7] J. Campos-Delgado, G. Algara-Siller, U. Kaiser, J.-P. Raskin. Twisted bi-layer graphene: microscopic rainbows (*Submitted in 2013*).
[8] J. Campos-Delgado, L.G. Cançado, C.A. Achete, A. Jorio, J.-P. Raskin. Raman-scattering study of the phonon dispersion in twisted bi-layer graphene. arXiv:1301.3795 [cond-mat.mes-hall].

Figures

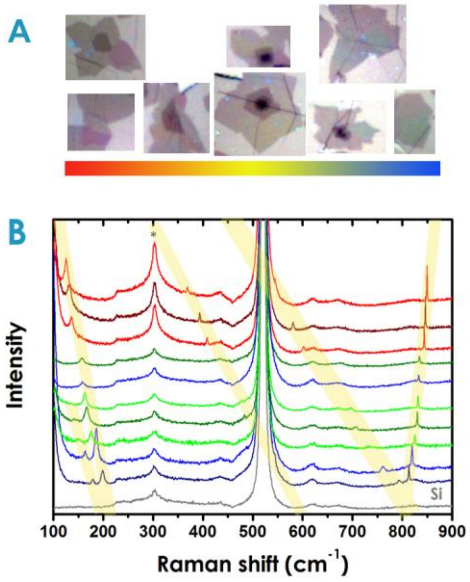

Figure 1. A) Optical micrograph of bilayer graphene transferred to Si/SiO_2 substrate (100 nm silicon dioxide thickness). Pink-, red-, yellow- and blue- colored areas can be identified. A color scale is included as guide to the colors pictured. B) Raman spectroscopy of bilayer graphene, new families of features (highlighted in yellow) can be identified. Curve coloring corresponds to E_{laser} used: blue shades correspond to spectra recorded with E_{laser}=2.54 eV, green shades E_{laser}=2.41 eV and red shades E_{laser}=1.96 eV. Exception for the bottom (gray) spectrum (E_{laser}=2.41 eV), which corresponds to Si substrate reference signal.