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Probing Graphene Defect Structures and Local Properties at the Atomic Scale with Gentle STEM

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The potential application of graphene in future nanoelectronics and optoelectronics calls for understanding of the correlation between defect structures and local properties at the atomic scale [1]. Here, we investigate the link between atomic structure, bonding, electronic and optical properties of structural defects using aberration-corrected low-energy, low-dose, scanning transmission electron microscopy (STEM), also known as gentle STEM [2], in combination with density-functional quantum-mechanical calculations. Using a combination of quantitative annular dark-field (ADF) imaging and fast-scan sequential acquisition (FSSA), we show that single dopant atoms can be directly visualized and identified in graphene, and defect dynamics can be studied at the single-atom level. We observe that a Si dopant in graphene can oscillate between three- and four-fold coordinated configurations. The precise atomic configuration determines the local bonding and electronic structure, which are directly identified via atomic resolution electron energy-loss spectroscopy (EELS). Furthermore, with FSSA, the reversible oscillatory motion of a Si6 magic cluster trapped in a graphene nanopore can be directly imaged, following the calculated atomic-scale energy landscape, which provides a promising technique for quantitative study of molecular dynamics at the atomic scale. Atom-by-atom observation of both reversible and irreversible grain boundary migration in graphene will also be presented. The local optical response at various defect sites in graphene is studied by EEL spectrum imaging at the low-loss regime. We observe that a point defect complex acts as an atomic-scale antenna in the petaHertz (10^15 Hz) frequency range, inducing a localized surface plasmon enhancement at the sub-nanometer scale [3]. In addition, a new one-dimensional plasmon enhancement is observed at the open edge of monolayer graphene with a spatial extent of ~ 0.6 nm [4]. Finally, we show that highly localized plasmon modes are generated at a graphene quantum disk (GQD) due to the confinement from the edge of the GQD at all directions [5]. Our results open new possibilities for designing nanoscale optoelectronic devices based on monolayer graphene. References: [1] A. K. Geim, Science 234, 1530 (2009). [2] O. L. Krivanek et al. Ultramicroscopy, 110, 935 (2010). [3] W. Zhou et al. Nature Nanotechnology, 7, 161-165 (2012). [4] W. Zhou et al. Ultramicroscopy, in press (2012). [5] W. Zhou et al. J. Phys.: Condens. Matter, in press (2012). This research was supported by NSF grant No. DMR-0938330 (WZ), DOE grant DE- F002-09ER46554 (MK, STP), Oak Ridge National Laboratory's Shared Research Equipment (ShaRE) User Facility (JCI), which is sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy; and the Office of Basic Energy Sciences, Materials Sciences and Engineering Division, U.S. Department of Energy (JL, ARL, SJP).