MAR14-2013-020300

Abstract for an Invited Paper
for the MAR14 Meeting of
the American Physical Society

Theoretical studies of structure-property relations in graphene-based carbon nanostructures

DIMITRIOS MAROUDAS, University of Massachusetts, Amherst

This presentation focuses on establishing relations between atomic structure, electronic structure, and properties in graphene-based carbon nanostructures through first-principles density functional theory calculations and molecular-dynamics simulations. We have analyzed carbon nanostructure formation from twisted bilayer graphene, upon creation of interlayer covalent C-C bonds due to patterned hydrogenation or fluorination. For small twist angles and twist angles near 30 degrees, interlayer covalent bonding generates superlattices of diamond-like nanocrystals and of fullerene-like configurations, respectively, embedded within the graphene layers. The electronic band gaps of these superlattices can be tuned through selective chemical functionalization and creation of interlayer bonds, and range from a few meV to over 1.2 eV. The mechanical properties of these superstructures also can be precisely tuned by controlling the extent of chemical functionalization. Importantly, the shear modulus is shown to increase monotonically with the fraction of sp^3-hybridized C-C bonds. We have also studied collective interactions of multiple defects such as random distributions of vacancies in single-layer graphene (SLG). We find that a crystalline-to-amorphous structural transition occurs at vacancy concentrations of 5-10% over a broad temperature range. The structure of our defect-induced amorphized graphene is in excellent agreement with experimental observations of SLG exposed to a high electron irradiation dose. Simulations of tensile tests on these irradiated graphene sheets identify trends for the ultimate tensile strength, failure strain, and toughness as a function of vacancy concentration. The vacancy-induced amorphization transition is accompanied by a brittle-to-ductile transition in the failure response of irradiated graphene sheets and even heavily damaged samples exhibit tensile strengths near 30 GPa, in significant excess of those typical of engineering materials.