Superlattices of finite interlayer-bonded domains in twisted bilayer graphene – structural, electronic, and some thermodynamic properties

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In this talk, we will present a comprehensive computational analysis of the atomic and electronic structure of 2D carbon nanostructures formed due to interlayer covalent sp^3 C-C bonding in twisted bilayer graphene (TBG) as a result of controlled chemical functionalization (hydrogenation or fluorination). Depending on the twist angle (from 0 to 30 degrees) and local stacking of layers, these nanostructures consist of superlattices of finite domains of either diamond-like (diamane) nanocrystals [1,2] or caged fullerene-like configurations [2,3] embedded within the bilayer. These superlattices have the same periodicity as that of the Moiré pattern corresponding to the rotational layer stacking in TBG. According to density functional theory (DFT) calculations, the electronic behavior of these sp^2/sp^3 hybrid configurations ranges from semi-metallic, characterized by linear dispersion around the K point in the first Brillouin zone (Dirac cones), to semi-conducting/insulating with electronic band gaps ranging from a few meV to ~4 eV; this electronic character depends on the symmetry and periodicity of the superlattices, the density and spatial distribution of the interlayer C-C bonds, and on the type of chemisorbed species (H or F atoms). Using DFT calculations and classical molecular dynamics simulations, we also demonstrate that the thermal expansion coefficient of these materials range from negative to positive values at room temperature and, most importantly, causes it to exhibit zero thermal expansion (ZTE) behavior in a wide range of temperatures [4]. We show that by controlling the density of interlayer bonds, it is possible to tune not only the sign of the thermal expansion of the system, but also the range of temperatures on which the ZTE behavior is observed. We also show that these superlattices of diamane finite domains can also be naturally created in multi-walled carbon nanotubes [5]. The rich and diverse combination of physical properties exhibited by this class of 2D materials makes them very attractive for enabling several technologies in nanoelectronics and miniaturized devices.

References:

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