Nanoelectronic Properties and Applications of Chemically Modified Graphene

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The outstanding electronic transport properties of graphene, including its superlative charge carrier mobilities, have been established on pristine samples in idealized conditions. However, for nanoelectronic applications, graphene needs to be interfaced with other materials in a manner that either preserves its intrinsic properties or modifies its properties in a manner that enhances functionality [1]. Towards these ends, this talk focuses on chemical functionalization strategies for interfacing graphene with other materials and for tailoring its electronic properties. For example, several noncovalent chemistries have been demonstrated and characterized at the molecular scale with ultra-high vacuum (UHV) scanning tunneling microscopy (STM) including 3,4,9,10-perylenetetracarboxylic dianhydride (PTCDA) [2-4] and 10.12 pentacosadiynoic acid (PCDA) [5]. PTCDA is shown to be an effective atomic layer deposition (ALD) seeding layer for high-k dielectrics (e.g., Al₂O₃ and HfO₂) such that the PTCDA monolayer remains intact as a well-defined passivating layer at the graphene-dielectric interface following ALD [6-8]. On the other hand, PCDA forms one-dimensionally ordered self-assembled monolayers on graphene. The PCDA can then be stabilized by ultraviolet photopolymerization, yielding one-dimensional polymers with sub-2 nm widths that are in registry with the underlying graphene lattice [3]. These ordered cross-linked PCDA-based polymers are promising templates for one-dimensional ALD-grown oxide nanostructures [9]. Beyond noncovalent self-assembled monolayers, this talk will also explore covalent modification schemes for graphene based on free radical chemistries [10]. In particular, atomic oxygen has been established as an effective method for homogeneously functionalizing graphene with epoxide groups in UHV [11]. Importantly, this covalent functionalization method is fully reversible under mild thermal annealing conditions (~ 260°C) as shown by atomic resolution UHV STM imaging. In addition to chemically doping graphene, epoxidation yields local modification of the graphene bandstructure and provides multiple pathways for further chemical functionalization [12].

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