Work function variations in twisted graphene layers

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Vetting graphene and 2D materials as candidates for advanced electronics requires examination of both intrinsic and extrinsic influences on their material properties, as well as a nuanced characterization of how they respond in different heterogeneous configurations. As the field of 2D crystals expands to include stacked heterosystems, the relative orientation (or twist angle) between layers becomes important. In the simplest case, stacking two graphene layers to form twisted bilayer graphene (TBG) already leads to measureable differences in variables such as interlayer screening[1], optical absorption[2], chiral charge carriers [3], or chemical reactivity [4].

In this talk, we describe the characterization (e.g, imaging, Raman, kelvin probe force microscopy (KFPM), and photoemission electron microscopy (PEEM)) of rotationally faulted graphene. The variable inter-layer interactions in twisted graphene layers leads to a wide range of variable properties, including optical absorption and surface potential (Φ). We find that Φ can vary up to $\Delta \Phi$ =39mV between 'small' ($\theta_s \approx 0^\circ$) and 'large' ($\theta_L > 16^\circ$) twist angles, and between $\Delta \Phi$ =36–129mV for different layer thickness (N=1-4). The PEEM measured work function of 4.4eV for graphene is consistent with doping levels on the order of 10^{12} cm⁻², and we find that Φ scales linearly with Raman G-peak wavenumber shift (slope = 22.2 mV/cm⁻¹) for all layers and twist angles, which is consistent with doping-dependent changes to graphene's Fermi energy in the 'high' doping limit[5]. The results discussed here emphasize that layer orientation is equally important as layer thickness when designing multilayer 2D systems.

Figure 1: Analysis of a rotationally faulted, multilayer graphene film. The same $20x20\mu m^2$ area mapped by (a) optical and (b) KPFM with labeled twist angles (e.g., $\theta_s \approx 0^\circ$, $\theta_L > 16^\circ$) and layer thickness



(e.g., $2\theta_L = 2$ -layers; $3\theta_s = 3$ -layers). The 'pink' and 'yellow' domains in (a) are at special twist angles that preferentially absorb in the visible spectrum. (c) plot showing change of surface potential vs. the raman G-peak shift between different layers and twist angles.

[1] Sanchez-Yamagishi, et al., PRL 108, 076601 (2012).

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