Understanding local electronic properties in epitaxial graphene heterostructures by scanning tunneling microscopy and spectroscopy

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Despite long-standing efforts to understand the interface between epitaxial graphene and the Si-terminated silicon carbide (0001) substrate, the exact atomic-scale structure, its complex bonding configurations, and the resulting electronic properties of the first epitaxial graphene carbon layer (C_{buffer}) remain open problems. In the first part of the talk, we will present how to control the local structure of the epitaxial graphene-SiC interface with a low-temperature scanning tunneling microscope (STM) [1]. We will show that the covalent bonds between C_{buffer} and the SiC substrate may be reversibly broken and restored using the polarity of the electric field from the STM tip, generating protrusions at the interface of slightly higher amplitude. This buried interface manipulation enabled us to pattern epitaxial graphene with lateral precision, reaching the scale of a single 1.8 nm unit cell of the graphene-SiC interface (6×6)SiC moiré lattice. We propose applying this local patterning of the epitaxial graphene heterostructures at the bottom interface to synthesize metastable interface configurations by controlling the location of the nanoprotrusions. The decoupled interface could be locally arranged in ordered superstructures, creating a new 2D template with a long-range periodic potential for the Dirac electrons in graphene to tune its electronic properties. This strategy will be similar to the templates currently used in the artificially arranged atomic or molecular adsorbates.

However, the STM manipulation experiments also provide direct evidence of silicon vacancies at the topmost reconstructed SiC(0001) layers [2]. Bias-voltage- and epitaxial graphene thickness-dependent characterization of the collective C_{buffer}-SiC interface showed that 'Si' vacancy sites exhibit stable, non-switching behavior under STM electric fields. Moreover, vacancies introduce localized electronic states below the Fermi level. Our experimental results directly elucidate the atomic-scale interface and the influence of atomic and bonding configurations on the local density of states for graphene of different thicknesses on SiC(0001).

In the second part of the talk, we will present the methodology for identifying the intrinsic electronic density of states of unoccupied bands above the vacuum level as a function of pristine graphene thickness and intercalated metal phases under graphene, using high-bias scanning tunneling spectroscopy. The low-temperature STM operating at high sample bias voltages up to ~40 V was used to study the interaction between graphene interface states within the unoccupied electronic band regime and the high-bias field emission resonances formed within the triangular potential in the tip-sample gap. The systems studied include C_{buffer}, monolayer, bilayer, and trilayer graphene prepared on a SiC(0001) surface, as well as intercalated phases of gadolinium and dysprosium. With increasing tunneling current for the same tip-sample voltage, the potential well becomes progressively steeper and narrower, and the confined energy levels of the resonances shift. The alignment between the resonance's energy and the intrinsic interface electronic states is a unique way to map the unoccupied bands in the different types of heterostructures and selectively study the characteristic interface states.

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References:

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