Phase-controlled two-dimensional Ag intercalation at the graphene/SiC interface

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Intercalation at the graphene/SiC interface offers a controlled route to stabilize atomically thin phases while maintaining a clean, encapsulated environment: the SiC substrate provides structural templating, and the graphene overlayer acts as an inert cap [1]. In the intercalation method, the defect density of the initial substrate plays an important role. As recently shown using the confinement heteroepitaxy method, Ag intercalation at the graphene/SiC interface can yield two distinct phases: the $Ag_{(1)}$ phase, which adopts a triangular lattice in registry with the SiC substrate, and the denser $Ag_{(2)}$ phase, which exhibits a more complex epitaxial relationship with the substrate [2]. Under ultra-high-vacuum (UHV) conditions, conventional intercalation starting from the zero-layer graphene (ZLG) substrate has been reported to produce $Ag_{(1)}$ [3, 4]. In this study, we demonstrate an in situ UHV preparation route that stabilizes the dense $Ag_{(2)}$ phase at the graphene/SiC interface and compare the structural and electronic properties of $Ag_{(1)}$ and $Ag_{(2)}$ using low-energy electron diffraction (LEED) and angle-resolved photoemission spectroscopy (ARPES).

In UHV, $Ag_{(2)}$ is prepared using the conventional method (deposition of Ag and annealing), starting from a Pb intercalated–quasi-free-standing monolayer graphene (QFMLG) substrate [5, 6, 7], taking advantage of the fact that Pb intercalation can introduce more vacancy and boundary defects. LEED identifies the structural fingerprints of $Ag_{(2)}$: the Ag lattice is rotated by 30° relative to SiC unitcell and forms superstructures with the adjacent layers, including an approximately (6×6) periodicity with respect to graphene and a (5×5) periodicity with respect to SiC (Fig. 1(a)).

High-resolution ARPES shows that $Ag_{(2)}$ is semiconducting and exhibits a more complex valence-band structure than $Ag_{(1)}$, with multiple branches near the $\overline{K}_{SiC}$-point (Fig. 1(b)) and reduced hexagonal warping in constant-energy contours (CECs) (Fig. 1(c)), indicating a weaker imprint of the SiC periodic potential. Density-functional-theory (DFT) calculations based on a structural model for $Ag_{(2)}$ reproduce the main experimental dispersions and show that the redistribution of the valence-band partial charge density plays an important role in the band dispersion of the second Ag phase.

Finally, we show that the intercalated Ag phase tunes the electronic response of the overlying quasi-free-standing graphene. Compared with $Ag_{(1)}$-QFMLG, $Ag_{(2)}$-QFMLG exhibits stronger n-type doping and enhanced electron–plasmon coupling in the graphene layer, consistent with reduced effective screening at the interface. We also observe Dirac-cone replica pockets that directly reflect the formation of the $Ag_{(2)}$/graphene supercell. These results demonstrate defect-enabled access to different confined-2D-Ag phases with distinct electronic structure and phase-dependent proximity coupling to graphene.

References:
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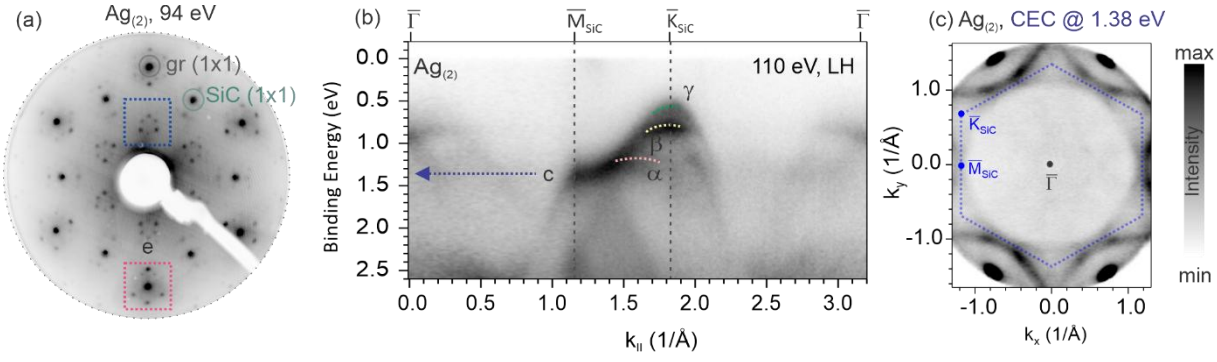

Fig. 1: (a) LEED of $Ag_{(2)}$-QFMLG shows the superstructure spots inside the pink and blue square, (b) ARPES data of $Ag_{(2)}$-QFMLG along $\overline{\Gamma M_{SiC}} \overline{K_{SiC}} \overline{\Gamma}$ direction , (c) CEC of $Ag_{(2)}$ at 1.38 eV binding energy.