Remote epitaxial frustration

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Remote epitaxy promises to circumvent the lattice and chemical mismatch challenges of conventional epitaxy, to enable low defect density and chemically abrupt heterostructures of dissimilar materials. However, definitive experimental evidence for a true "remote" mechanisms remains elusive because most observations can be explained by alternative pinhole or van der Waals mechanisms, which are often macroscopically indistinguishable from a true "remote" mechanism [1,2]. Here, using GdAuGe films grown on graphene/SiC (0001), we present two long-range signatures of a remote mechanism that cannot be explained by the leading alternatives: (1) a two atomic layer thick disordered interlayer at the GdAuGe/graphene interface and (2) a new 30 degree rotated epitaxial relationship between GdAuGe film and SiC substrate. Density functional theory calculations suggest that these signatures arise from remote epitaxial "frustration,'" i.e. a competition between epitaxy of the GdAuGe film to the screened remote potential of the substrate (φ_{sub}), versus direct epitaxy to graphene (φ_{gr}) and to the long-range graphene-induced surface reconstruction (φ_{rec}). Our results highlight the importance of considering the multiple contributions to the total lattice potential above graphene-covered surfaces, rather than an exclusive focus on φ_{sub}. Moreover, tuning the relative magnitude [3] and periodicities of φ_{gr}, φ_{sub}, and φ_{rec} provides new opportunities to (1) control short- and medium-range ordering of films stabilized at graphene-covered interfaces, towards the discovery of new glasses and quasicrystals, and (2) direct synthesis of rotated moire heterostructures for tuning magnetism and correlated phases.

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Supplementary Pages

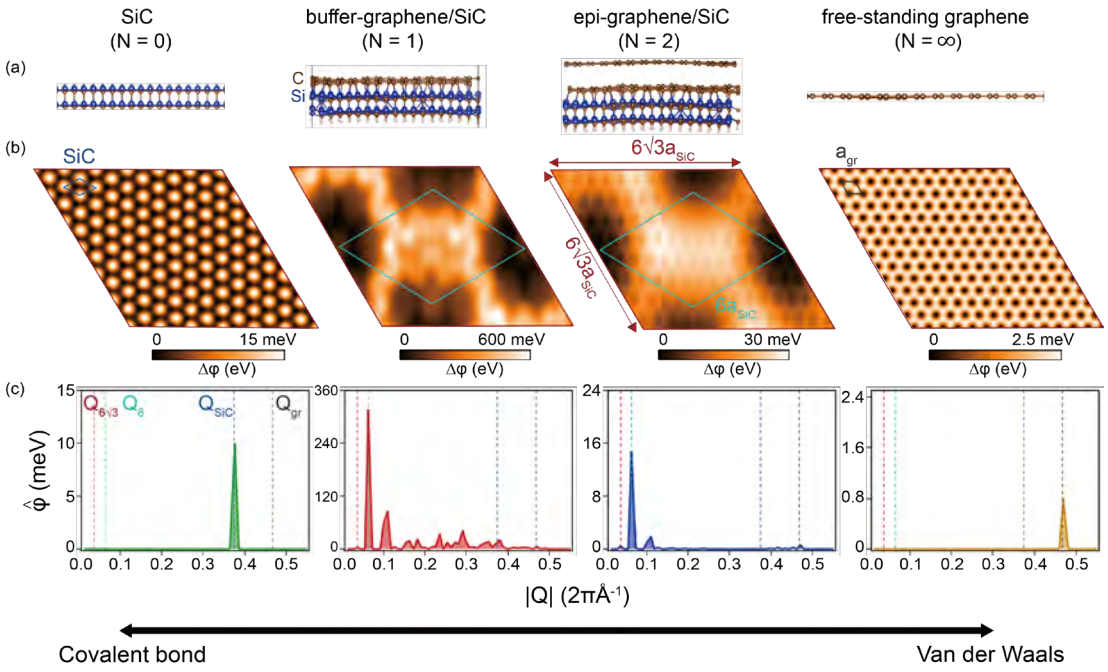

Fig. 1. Concept for remote frustration. (a) Slab models for graphene, epitaxial graphene on 6H-SiC (0001), buffer graphene on SiC, and SiC (0001). (b) Calculated electrostatic potential at 3 Angstrom above each surface. (c) Fourier components of the electrostatic potential, at the reciprocal lattice vectors for graphene (Q_{gr}), the (6x6) reconstruction (Q_{rec}), and SiC (Q_{SiC}).

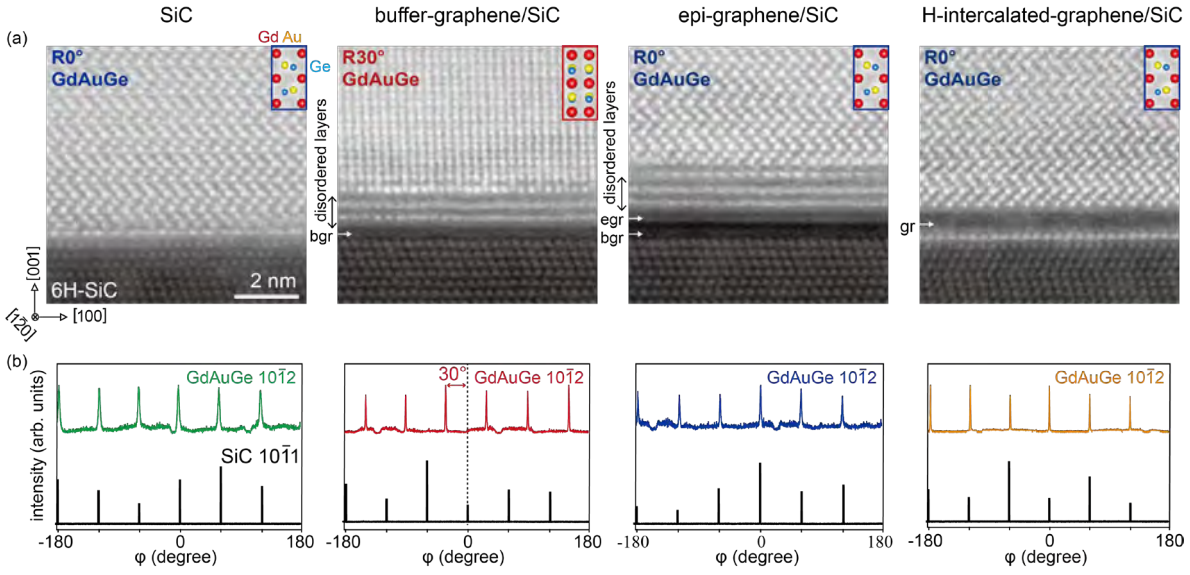

Fig. 2. Remote frustration of GdAuGe on graphene/SiC. (a) Cross-sectional TEM, showing a 2-monolayer-thick disordered layer at the graphene interface, for GdAuGe on epi graphene and on buffer graphene. Gd (red), Au (yellow), Ge (blue). (b) X-ray azimuthal phi scan showing GdAuGe on buffer graphene is rotated in-plane by 30 degrees.