

Strain-induced reconstruction in two-dimensional silver intercalated between graphene and SiC

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The realization of air-stable two-dimensional (2D) metals confined at the interface between graphene and SiC has attracted significant attention owing to their distinct properties compared to their bulk counterparts. In this work, we identify two distinct structural phases in a monolayer silver intercalated between graphene and SiC: (i) a moiré pattern arising from the lattice mismatch between graphene and monolayer silver and (ii) a striped reconstruction formed through the interaction with the underlying SiC. Using cryogenic scanning tunneling microscopy combined with density functional theory, we elucidate the origin of the silver striped reconstruction. Driven by the competition between the silver-SiC and the silver-silver interactions within a monolayer, the silver atoms form a one-dimensional Frenkel-Kontorova domain, in which an $(n + 1) \times \sqrt{3}$ silver supercell accommodates an $n \times \sqrt{3}$ supercell ($n = 20$) of the underlying SiC, which partially relieves tensile strain, otherwise imposed by ideal epitaxy. The strain-induced reconstruction creates a long-range modulation of 2D silver electronic density, giving rise to an electronic state at ~ 0.75 eV above the Fermi level in the transition regions between different silver-SiC registries. These findings uncover the important interplay between the structural and electronic properties of intercalated 2D metals and the underlying SiC substrate, providing new insights into substrate-mediated reconstruction and electronic modulation at confined interfaces.

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I. INTRODUCTION

Two-dimensional (2D) metals can give rise to intriguing physical behaviors and constitute a promising material platform for nanoelectronics [1–3]. Such a system can be successfully realized by metal intercalation under graphene on different substrates. Spin-orbit gap opening in the Dirac point of graphene has been achieved, for instance, by intercalation of Pb between graphene and Pt(111) [4], while Au intercalation under graphene on Ni(111) leads to a substantial increase in the spin-orbit splitting of graphene π states [5]. Similarly, a pronounced Dzyaloshinskii-Moriya interaction induced by the Rashba effect has been observed in an intercalated graphene/Co/Ru(0001) system [6]. Moreover, intercalated metal layers themselves can host intriguing

phenomena, as significantly enhancing magnetic anisotropy in Co layers intercalated at the graphene/Ir(111) [7,8] interface.

Notably, 2D metals can be stabilized at the interface between epitaxial graphene and semiconducting SiC substrate, enabling continuous, wafer-scale metal layers [9], and exhibit a wide range of unusual physical properties [1–3,10]. This capability arises from the growth of an initial epitaxial graphene layer that covers the entire SiC substrate, followed by the postmetal intercalation [11]. Intercalated metal structures at the graphene/SiC interface are found to adopt a triangular lattice, resembling the atomic arrangement of the SiC-terminated layers through metal-Si bonding, while remaining a weak van der Waals interaction with the graphene overlayer, with partial charge transfer [1,2,10,12].

The structure and electronic properties of metals intercalated at the graphene/SiC interface, similar to the graphene/metal/metal system [13–15], are strongly influenced by their interaction with the underlying SiC substrate. Although the intercalation picture has been revealed in understanding the formation mechanism of 2D metal layers at the graphene/SiC interface, experimental investigations have found more complex atomic registries between the metals and SiC, leading to nontriangular lattice structures in the intercalated metal layers [16–18]. Scanning tunneling microscopy

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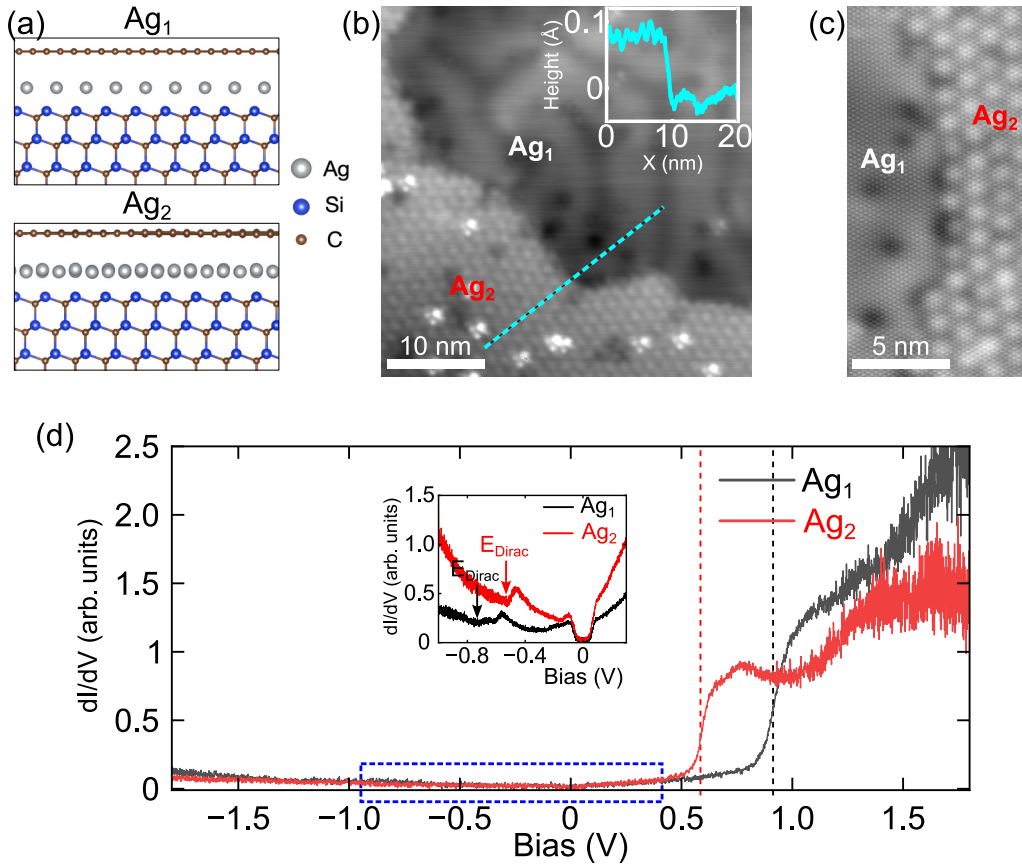


FIG. 1. (a) Schematic illustration of distinct Ag phases intercalated between graphene and SiC. Upper and lower panels represent the structures of Ag₁ and Ag₂, respectively. Note that the Ag₂ lattice is 30° rotated with respect to that of the SiC; therefore, it appears more condensed in the sideview atomic scheme (lower panel). The graphene layer is sketched only for illustration. (b) Representative STM image (−1.3 V, 100 pA) of the two Ag phases. Ag₁ shows long stripes formed by Ag atoms below graphene. Ag₂ exhibits a bright-dot superstructure. The irregular bright and dark spots on both Ag regions are attributed to vacancies or impurity defects within the Ag monolayer. Inset: line profile crossing the boundary between Ag₁ and Ag₂, revealing a height difference of ~10 pm. (c) High-resolution STM image (−0.1 V, 100 pA) of an area where two Ag phases intersect. At low bias, graphene coexists and covers on top of both intercalated Ag phases. (d) Representative dI/dV spectra measured on Ag₁ (black curve) and Ag₂ (red curve) reveal a sharp onset (marked by vertical dashed lines) in the conduction region at ~0.9 and ~0.6 V for Ag₁ and Ag₂, respectively. Inset: a zoomed dI/dV spectra at low bias range indicated by the blue dashed box revealing the dominant DOS of graphene with the Dirac points (E_{Dirac}) indicated by arrows at −0.73 eV (Ag₁) and −0.52 eV (Ag₂), respectively, and a phonon-induced inelastic gap (~134 mV) centered around the zero energy at low temperature.

(STM) often identifies the moiré pattern arising as a result of the lattice mismatch between the graphene overlayer and intercalated metals on SiC [1,16,19,20]; however, detailed insights into the atomic structure and electronic properties of intercalated metals governed by interaction with the underlying SiC substrate remain largely unexplored.

Among metallic elements, silver (Ag) intercalation forms distinct monolayered structures at the graphene/SiC interface [21,22], behaving as a semiconductor [10,12,23]. In Fig. 1(a), we illustrate two different Ag structural phases, named Ag₁ and Ag₂, coexisting in a monolayer upon intercalation in our samples prepared using the protocols described by Jain *et al.* [22]. It is important to note that the growth of each Ag phase can be selectively controlled by controlling the initial graphene type (either starting from zero-layer or monolayer graphene) [22]. In this work, the starting substrate consists of predominant zero-layer graphene (ZLG) (~90%) with ~10% epitaxial graphene monolayer overgrowth at the terrace edges

as shown in Fig. S1 of the Supplemental Material [24]. Therefore, upon intercalation, we obtained ~90% Ag₂ and ~10% Ag₁, presented in Fig. S2 of the Supplemental Material [24].

Ag₁ has a roughly 1:1 epitaxial match with the topmost SiC, where the Ag atoms are in registry with SiC. The Ag₂ phase has a more compact atomic packing density and its hexagonal unit cell vector is 30° rotated with respect to that of the SiC. In this work, we investigate how the registry of Ag atoms with the underlying SiC modulates the structures and electronic properties of the monolayer Ag in the Ag₁ phase, giving rise to long striped reconstruction patterns. The reconstructed linear structure is considered as a Frenkel-Kontorova domain [25], resulting from the competition between the preferred in-plane Ag-Ag interaction (2.88 Å) of an Ag(111) and the out-of-plane Ag-SiC interaction (where Ag-Ag bonding length in this scenario is the same as that of SiC, 3.10 Å). This reconstruction consists of two regions where Ag atoms have two registries with the topmost SiC layer. The transition

region between them exhibits an electronic state at ~ 0.75 eV above the Fermi level, providing strong evidence of the modulation in the 2D Ag electronic density of states induced by the atomic reconstruction. Our results provide an in-depth understanding of the structures and electronic properties of 2D Ag at the graphene/SiC interface and constitute an important topic in controlling the properties of 2D metal layers at the atomic level [26].

II. METHODS

A. Sample preparation

ZLG was grown by thermal decomposition of semi-insulating 6H-SiC(0001) (Coherent Corp). ZLG consists of $\sim 30\%$ carbon atoms, covalently bonded to the underlying SiC, which promotes intercalation without further treatment to generate defects [10]. SiC substrates were annealed at 1673 K for 30 min in 10% H_2/Ar (700 Torr), followed by annealing at 1873 K for 30 min in pure Ar (700 Torr) to produce a predominantly ZLG surface with $\sim 10\%$ monolayer graphene overgrowth. Silver intercalation was performed in a quartz-tube furnace (Thermo Scientific Lindberg/Blue M Mini-Mite) by placing high-purity Ag powder (99.99%, Sigma-Aldrich) beneath the ZLG/SiC sample in an alumina crucible. The system was heated to 1173 K in flowing Ar (500 Torr) for 1 h and cooled to room temperature, yielding a predominantly quasifreestanding monolayer graphene/Ag/SiC heterostructure.

B. STM/STS measurements

STM/STS were performed on a Createc cryogenic STM operating at 5 K under ultrahigh vacuum ($\sim 10^{-10}$ mbar). Electrochemically etched W tips were used for measurements, cleaned by Ne ion sputtering and electron beam annealing, and calibrated on Ag surfaces. STM images were obtained in constant-current mode. Scanning tunneling spectroscopy (STS) were performed using a lock-in technique with a sinusoidal modulation (peak to peak of 5 mV) at a frequency of 675 Hz to directly measure differential conductance dI/dV spectra. The intercalated Ag samples were transported in air, then annealed at ~ 423 K under UHV before transferring into a cold STM head at 5 K.

C. Theoretical calculations

All DFT calculations are done by VASP [27–30] at the PBE level [31] with cutoff energy at 450 eV, electronic self-consistent convergence at 2.0×10^{-8} eV/atom or lower, and residual force convergence at 0.01 eV/Å on atoms with D3 van der Waals correction [32]. The k -point samplings for self-consistent calculations (and non-self-consistent calculations of DOS) are Γ -centered $13 \times 13 \times 1$ ($51 \times 51 \times 1$) for 1×1 SiC unit cells, MP sampling [33] $1 \times 8 \times 1$ ($2 \times 16 \times 1$) for 1D supercells, and Γ -only (Γ -centered $5 \times 5 \times 1$) for the Ag_2 supercell. The cell dimension in the z direction is 20 Å. Two layers of SiC are included in all systems, terminated with H atoms at the opposite site of the Ag layer. The STM simulation is based on the local density of states (DOS) using the Tersoff-Hamann method [34]. The structure visualization

and 2D slices of the partial charge density are made with VESTA [35]. The partial charge density is created with the VASPBANDUNFOLDING code [36].

III. RESULTS AND DISCUSSION

Figure 1(b) shows a representative STM image taken at -1.3 V, which reveals two Ag structural phases separated by a well-defined boundary: herringbone-like stripes formed in Ag_1 , while a triangular superlattice in Ag_2 . The latter is only ~ 10 pm higher than the former (see the inset), which is substantially smaller than the thickness of a single Ag layer (~ 3 Å, Fig. S3, Supplemental Material [24]) [37]. The observed height difference between Ag_1 and Ag_2 [see line profile in the inset of Fig. 1(b)] likely arises from the fact that the Ag_1 phase preferentially forms under an initially grown monolayer graphene on SiC, where intercalation results in a bilayer graphene on top. Whereas the Ag_2 phase forms at the ZLG/SiC interface, where roughly 30% of the carbon atoms in the ZLG remain covalently bonded to the topmost SiC layer [38], intercalation at this interface yields monolayer graphene above the Ag_2 phase [22]. Note that the height difference observed in Fig. 1(b) is mainly bias dependent and does not represent a true topographic variation between Ag_1 and Ag_2 . Instead, it arises from the differences in the electronic density of state, as evident in Fig. 1(c) where both Ag phases show nearly identical apparent heights.

At a low sample bias, atomic resolution of graphene is observed in both Ag phases with a low corrugation of ~ 10 pm [Fig. 1(c)]. The typical $(6\sqrt{3} \times 6\sqrt{3})R30^\circ$ superstructure originating from the reconstruction in ZLG [39–41], which is typically observed in the STM images of initial monolayer graphene on SiC, is absent, providing additional evidence for successful Ag intercalation. Note that, at bias voltages beyond approximately ± 0.5 V about the Fermi level, the honeycomb lattice is not resolved, indicating that the topographical contrast in Fig. 1(b) reflects structural characteristics mainly derived from the intercalated Ag. This suggests that the Ag electronic states tunnel through the graphene and extend towards the vacuum and strongly overlap with the tip wave function at a large sample bias. Random dark and bright defects, depending on the imaging conditions, are tentatively attributed to vacancies or unknown foreign impurities in the Ag monolayer [42–45].

Differential conductance (dI/dV) spectra were acquired to probe the local DOS of each Ag phase [Fig. 1(d)]. At a small bias, graphene DOS are evident, consistent with the topographic images where only the honeycomb lattice is resolved. The ~ 134 mV gap centered around the Fermi level [$V = 0$, inset in Fig. 1(d)] originates from the phonon-induced inelastic tunneling in graphene observed at low temperature [46]. The minima at -0.73 eV and -0.52 eV observed in Ag_1 and Ag_2 , respectively, correspond to the Dirac points (E_{Dirac}), revealing an n -type doping induced by the charge transfer from the Ag, in agreement with the previous work [10,23]. These observations indicate a relatively weak Ag-graphene coupling upon intercalation since the graphene properties remain essentially unchanged [10].

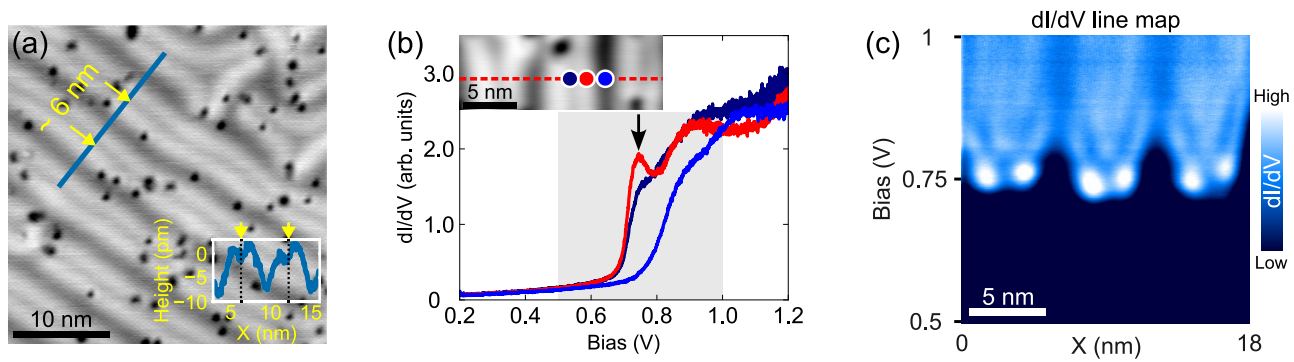


FIG. 2. (a) STM image (1.5 V, 100 pA) of a reconstructed Ag_1 area, displaying long stripes with a periodicity of ~ 6 nm, as shown in the line profile in the lower inset. (b) dI/dV spectra acquired at different locations across the stripes indicated by colored dots in the STM image (1 V, 100 pA) shown in the inset (upper left). A sharp peak (indicated by the black arrow) is observed when measured at the transition (bright line) between thin and thick dark lines. The shaded rectangle highlights the energy range where the spatial dI/dV map in (c) is taken. (c) dI/dV map (acquired at a constant-tip height) along the red dashed line in the STM image of the inset in (b). The electronic state corresponding to the energy of the peak indicated by the black arrow in (b) appears as bright features and disperses into the thick and thin dark stripes. The peak located at ~ 0.75 eV in (b) is identified as the bright-dot features in the dI/dV map.

Over a wider bias range, the dI/dV spectra reveal a sharp onset marked by the vertical dashed lines in the unoccupied region at ~ 0.9 and ~ 0.6 V for Ag_1 and Ag_2 , respectively, as shown in Fig. 1(d). This onset is absent in the DOS of pristine graphene [47] or SiC [47,48] and should be attributed to the electronic structures of the new interface formed by 2D Ag, similar to others reported for intercalated metals, such as indium [49]. Note that the intercalated Ag monolayer behaves as a semiconductor with an indirect gap of ~ 1 eV, demonstrated by angle-resolved photoelectron emission spectroscopy (ARPES), in which the valence band maximum (VBM) is located at -0.59 V [10] or -0.45 V [12,23] and conduction band minimum (CBM) at 0.56 eV [23]. Therefore, we attribute the observed onsets in Fig. 1(d) to the CBM of the semiconducting 2D Ag. We note that the VBM of the intercalated Ag was not detectable, which might likely be governed by the tunnel matrix elements for states close to the valence band edge [50,51] or by other effects that remain unclear.

Particular attention is focused on the striped structures observed in Ag_1 . This structure differs from the triangular pattern previously found for indium intercalated at the graphene/SiC interface, where the patterns originate from the strain-induced lattice mismatch between two intercalated metal layers [44]. Long striped features have previously been observed in Au [52], Bi [13], Li [14], and Eu [15] at the graphene/Ir(111) interface, and more recently in Pb [16] and Ga [18] intercalated between graphene and SiC. Therefore, atomic-scale investigation of these structures in intercalated metals at the graphene/SiC interface would provide important insight into how their structures are modulated by the SiC substrate.

The STM image of the Ag_1 phase in Fig. 2(a) shows well-defined dark and bright stripes, periodically spaced by approximately 6 nm (see the inset line profile). The apparent height corrugation between the dark and bright lines is measured to be only ~ 8 – 10 pm, which is much smaller than the thickness of an Ag monolayer; see Fig. S3 of the Supplemental Material [24]. Notably, these stripes are visible and

exhibit a nearly inverted contrast between the occupied- and unoccupied-state dI/dV maps; see Fig. S4 of the Supplemental Material [24]. At a negative bias, the bright stripes are uniform and equally separated by thick dark lines (~ 6 nm). In contrast, at a positive bias, each thick dark line transforms into a broad bright stripe with a thin dark line at its center.

These observed stripes in the intercalated Ag appear quite similar to the herringbone reconstruction in Au(111), which is characterized by reconstruction of alternating hexagonal close-packed (hcp) and face-centered cubic (fcc) regions at the topmost layer and is observed at low temperature [53–56]. The observation suggests that the intercalated Ag has been deformed or reconstructed, giving rise to a long-range modulation in both topography and DOS, which will be discussed in detail below.

dI/dV spectra acquired on different sites across the stripes are plotted in Fig. 2(b). The spectrum measured at the center of the bright stripe (red curve) between the thin and thick dark lines reveals a peak at ~ 0.75 V above the Fermi level. This peak disappears when measuring at the thin and thick dark lines (navy and blue curves, respectively); see more details in Fig. S5 of the Supplemental Material [24]. This behavior is best seen in the spatial dI/dV map shown in Fig. 2(c), obtained over the bias range indicated by the shaded rectangle, taken along the red dashed line indicated in Fig. 2(b). Remarkably, the peak observed in the red dI/dV spectrum corresponds to bright features located at the center of the bright stripe [Fig. 2(c)], which is confined and disperses into the conduction band at higher biases, forming characteristic interference fringes. This observation provides strong evidence of modulated electron density of states induced by the reconstructed superstructure in the intercalated Ag monolayer [54].

The orientations of the reconstruction stripes are well aligned with the zigzag directions of the graphene lattice, which is determined from high-resolution STM images [see the Y-shape black lines in Fig. 3(a)], where both the reconstruction lines and graphene atomic lattice are clearly visible (upper-right inset). It is important to note that graphene layers interact relatively weakly with the monolayer Ag and do not

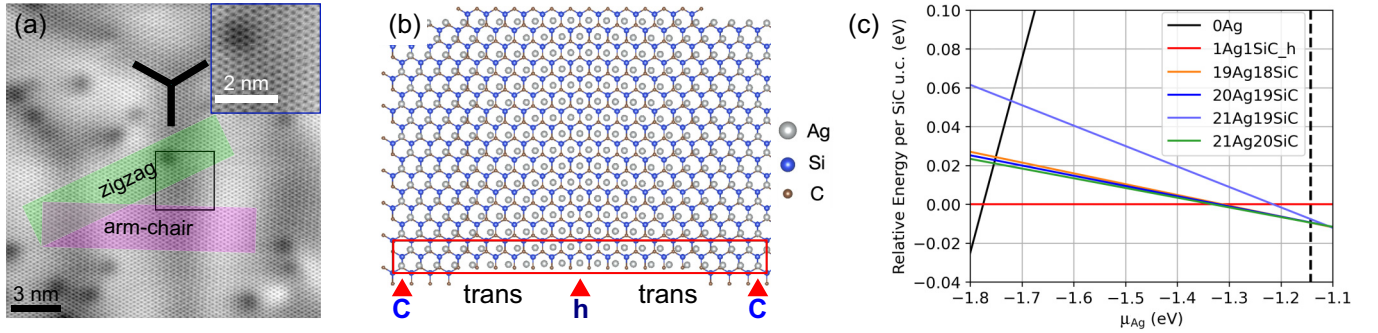


FIG. 3. (a) High-resolution STM image (0.05 V, 100 pA) of an Ag_1 area with reconstruction stripes. Upper right inset: atomic resolution STM image (0.05 V, 100 pA) taken at the same bias voltage from the central black square, revealing the honeycomb atomic corrugation of the graphene cap. Light green and purple rectangles indicate the zigzag and armchair directions of the graphene, respectively. Note that Ag intercalation only lifts the ZLG; hence the graphene layers are not rotated with respect to the underlying SiC. Thus the system has three symmetrically equivalent directions in which the linear domains were formed, indicated by the Y-shape black lines. (b) Top view of the relaxed 21Ag20SiC structure: $21 \times \sqrt{3}$ Ag atoms are commensurate with a $20 \times \sqrt{3}$ SiC supercell (long red rectangle), resulting in a transition region (trans) between the carbon (C) and hollow (h) site registries. Only the topmost SiC layer is visualized in this structure. (c) The free energy densities for structures without graphene of different ratios between Ag and SiC. The vertical dashed line is the chemical potential of bulk Ag. “0Ag” indicates no Ag intercalation. “h” for “1Ag1SiC” denotes the hollow site registry, which has the lowest energy for this ratio. The 1D structures can be more stable than the simple 1:1 ratio structure.

undergo rotation relative to the SiC underneath upon metal intercalation [10,57–59]. This allows the crystalline orientation of the underlying SiC to be determined as aligned with the armchair directions of the graphene (along the purple rectangle) [41,60], as further confirmed by low-energy electron diffraction (LEED) measurements [22]. The width of the stripes is ~ 6 nm [Fig. 2(a)], which corresponds to ~ 20 SiC unit cells (6.19 nm).

Coincidence lattice accommodation, in which 21 Ag spacings match 20 SiC spacings, is confirmed by density functional theory (DFT) calculations, elucidating that the structure of the reconstruction lines formed in the Ag_1 is induced by a competition between the in-plane Ag-Ag and the out-of-plane Ag-Si bonding interactions. Considering that the Ag-Ag bond length in a closed-packed fcc (111) plane is 2.88 Å, an ideal epitaxial Ag layer on SiC (with SiC lattice constant of 3.10 Å) would lead to a tensile strain along the Ag atomic plane. This strain could be partially relieved through the formation of a one-dimensional (1D) structure by increasing the areal density along one direction (armchair of graphene which is perpendicular to the stripes, to be consistent with the experiments).

One example supercell describing such a 1D structure is presented as the red rectangular supercell in Fig. 3(b), where 42 Ag atoms fit in a $20 \times \sqrt{3}$ SiC supercell. Inside the supercell, Ag atoms gradually change between C- and h-registries where Ag atoms are on top of the C sites and hollow sites of the topmost SiC, respectively. This model is consistent with the observation that the Ag_1 phase contains both C- and h-registries [22,61]. In the top view of Fig. 3(b), this registry change corresponds to a shift in the y position and the periodicity of this shift defines the width of the 1D structure. Figure S6 (see Supplemental Material [24]) shows the Ag-Ag interatomic distance as a function of the x position in the supercell. The largest interatomic distance occurs in the transition region, implying that Ag atoms in this region have

higher energies and tend to be attracted by C- and h-registry regions. Notably, in an ideal epitaxial picture, the C- and h-registries correspond to the lowest energies [22] and a similar energy profile is expected in a near 1:1 Ag-to-SiC ratio, consistent with our observation. Another possible supercell with a higher energy, containing the Si- and h-registries, is discussed in Fig. S7 of the Supplemental Material [24]. Note that the supercell can be formed in three equivalent orientations due to the threefold symmetry in SiC, in good agreement with the experimental finding shown as black lines in Fig. 3(a).

This reconstruction in monolayer Ag again shows a visual similarity to the well-known herringbone reconstruction on the Au(111) surface, once mentioned above. However, these two structures differ in their physical origins. In Au(111), the herringbone reconstruction is characterized by a $(22 \times \sqrt{3})$ supercell, in which the topmost Au atoms are compressed along the $[1\bar{1}0]$ direction, forming characteristic stripes with alternating fcc and hcp stacking regions. Notably, the Au(111) reconstruction exhibits an electronic state located at -0.52 eV below the Fermi level which is derived from the Au surface state electrons and induced by the modulation in the electronic density of states imposed by the reconstructed superstructure [54]. This observation shows very similar tendency with the electronic state observed in the dI/dV spectrum in Fig. 2(b). However, unlike the near-free electron behavior described by the extended Kronig-Penney model for the Au(111) surface [54], the Ag-SiC system is semiconducting and further investigation is required to develop a model that fully explains the observed electronic state at 0.75 eV.

To determine the phase stability of the 1D structures, we calculate and compare the free energy density of different structures, referenced to the 1:1 ratio epitaxial Ag-SiC [presented in Fig. 3(c)], using the following formula:

$$\frac{F}{A} = \frac{E - [n_{\text{SiC}}E_{1:1} + (n_{\text{Ag}} - n_{\text{SiC}})\mu_{\text{Ag}}]}{n_{\text{SiC}}},$$

where E is the energy of the structure, n_{Ag} is the number of Ag atoms, n_{SiC} is the number of SiC unit cells in the ab plane, μ_{Ag} is the chemical potential of Ag, and $E_{1:1}$ is the energy of the structure with a 1:1 ratio between Ag and SiC. The 1:1 ratio has three registries (h, C, Si), and we choose the h registry since it has the lowest energy [22]. The graphene cap was not included since it cannot fit in without an artificial 9% strain. The 20:18 and 22:20 (Ag:SiC) ratios were not calculated because they correspond to the $9 \times \sqrt{3}$ and $10 \times \sqrt{3}$ SiC supercells, respectively, which are only half the size of the experimentally observed stripe width. For a given chemical potential of Ag, the structure with the lowest free energy density is most thermodynamically favored in the 21Ag20SiC structure, shown as the green line in Fig. 3(c). We found that, on the Ag-rich side within the chemical potential window where Ag can intercalate, the structures with one more ($1 \times \sqrt{3}$)-Ag unit cell are more stable than an exact 1:1 structure, although the overall energy differences remain small over the entire window. Because these 1D supercells contain additional Ag atoms with no graphene, the Fermi level is therefore located in the conduction band; see Fig. S8 of the Supplemental Material [24]. However, in the experiments with the graphene cap, charge transfer to the graphene shifts the Fermi level back to the band gap, as evidenced by optical and ARPES measurements [22]. This effect further stabilizes the 1D structures, hence enlarging the chemical potential range, as compared to the calculation, where the 1D structures remain most stable. Among the calculated configurations, the free energy density of the 21:19 structure is consistently higher than that of other 1D structures. This is attributed to the presence of extra $2 \times \sqrt{3}$ Ag unit cells, adding a similar Ag areal density as the 20:18 or 22:20 structures and thus a similar higher free energy density as those that do not show in the experiments. It should be noted that these calculations assume an Ag monolayer, which in our experiments occurs only under confinement by the graphene cap; otherwise, Ag would form agglomerated particles; see Fig. S9 of the Supplemental Material [24]. Consequently, although the graphene cap is not explicitly included in the computational model, it is essential for both the intercalation and reconstruction.

STM simulation based on the Tersoff-Hamann approach [34] gives a further correspondence between the model and experiments. Due to a non-negligible matrix element effect at negative biases, as presumed above, direct comparison between STM images and the calculated DOS, which assumed a simplified form of the tunneling matrix element for STM simulation, in the valence bands might be unreliable. In the conduction band, dI/dV maps are challenging to compare directly since the bright/dark line positions are highly sensitive to the applied bias, as indicated by the fringe features in Fig. 2(c). Instead, we focus on the constant-height images at a relatively large positive bias (>0.75 V), where the characteristic double bright line appears [Fig. 2(a) and Fig. 4(a)]. This feature is relatively insensitive to the bias choice (as long as it is ~ 1 V) since the strong dI/dV signal at ~ 0.75 eV significantly contributes to these constant-tip height images. Comparing the constant-tip height STM current image at 1.0 eV [Fig. 4(a)] with the calculated partial charge density [Fig. 4(b)] reveals that the thin and thick dark lines correspond to h- and C-registered Ag, respectively, and the

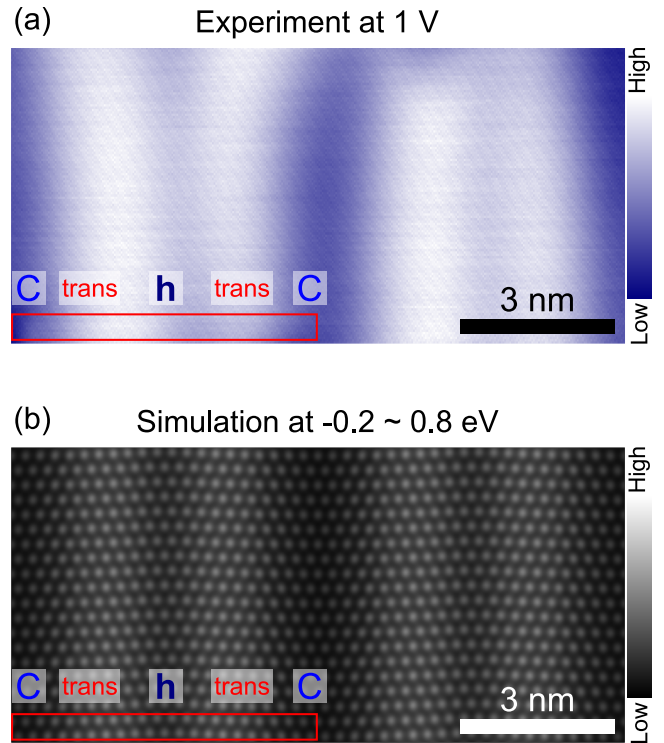


FIG. 4. (a) Experimental current STM image taken with a constant-tip height at unoccupied-state sample bias (1 V, 100 pA) across the stripes. (b) 21Ag20SiC partial charge density maps simulated 1.6 Å above Ag atoms of states between -0.2 and $+0.8$ eV relative to the calculated Fermi level. The long red rectangle shows the computational unit cell which has the same reconstruction periodicity (~ 6 nm) indicated by the same red rectangle in (a).

bright line corresponds to the transition region. Note that, due to the absence of graphene in the calculation, the calculated Fermi level is in the conduction band (Fig. S8, Supplemental Material [24]). Thus the energy window chosen for the STM simulation at 1.0 eV from the gap center is $[-0.2, 0.8]$ eV. Further simulation for $[0.0, 1.0]$ eV energy window shows a similar pattern, confirming the bias insensitivity of the double bright lines (Fig. S10, Supplemental Material [24]).

Figure S11 in the Supplemental Material [24] shows the calculated partial charge density in the unoccupied states with an energy window of 0.1 eV. The results are in qualitative agreement with Fig. 2(c): the partial charge density shows a maximum in the transition region in the $[0.7-0.8]$ eV energy window, while the intensity is lower at lower energies and the fringe patterns emerge at higher energies. Because the calculation does not include the energy correction for the unoccupied states (which would shift the peak to a higher energy) [62] and the charge transfer to the graphene (which would numerically correspond the peak to a 0.2 eV lower energy, consistent with the Fermi level shift) in this calculation, the numerical agreement in the peak energy is only coincident. Nevertheless, the reproduction of the overall features in Figs. 2(b) and 2(c) supports that the structural model is correct. Moreover, in Fig. S12 of the Supplemental Material [24] we present the simulated STM image with the graphene cap, cognizant of the strain (9% along the armchair and 0.7%

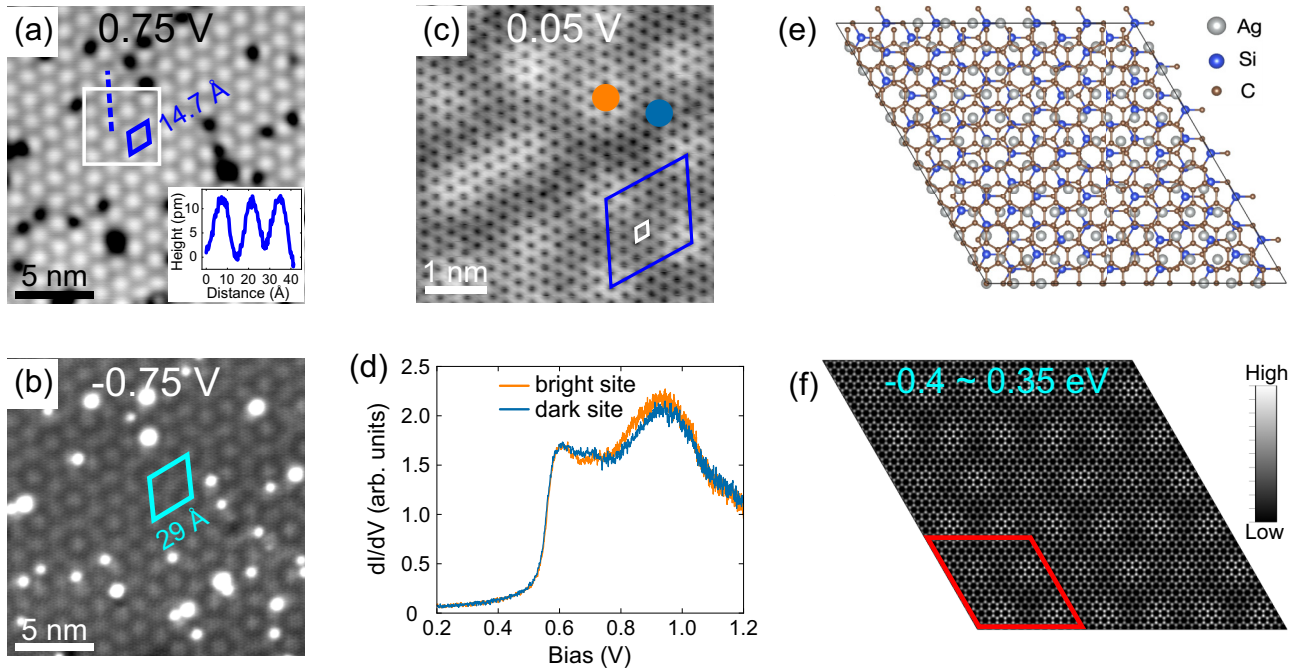


FIG. 5. (a) STM image taken at 0.75 V (100 pA) on Ag_2 displaying well-defined bright spots as a moiré pattern with a lattice constant of 14.7 Å (blue rhombus) formed by the lattice mismatch between the intercalated Ag and the graphene cap. Inset: line profile along the blue dashed line indicating the periodicity of the superstructure. (b) STM image (−0.75 V, 100 pA) of the same area in (a), revealing a new superstructure with a doubled periodicity of the supercell (~ 29 Å) in (a), which likely arises from the interference between the 13×13 graphene and $6\sqrt{3} \times 6\sqrt{3}$ SiC supercells, as described in (e). (c) Zoomed-in low-bias STM image (0.05 V, 100 pA) displays both graphene atomic resolution and the supercell, allowing an accurate measure of supercell size in (a) (blue rhombus), which is roughly equal to six times the graphene unit cell (small white rhombus) with the same orientation. (d) dI/dV spectra taken right above the bright (orange curve) and dark (dark-cyan curve) sites of the superstructure, as indicated by orange and dark-cyan dots in (c). (e) Top view of the relaxed Ag_2 phase with 13×13 graphene, 11×11 Ag, and $6\sqrt{3} \times 6\sqrt{3}$ SiC unit cells. (f) The partial charge density of the Ag_2 STM simulated between −0.4 eV and 0.35 eV at 0.74 Å above the graphene layer of states within the energy range given relative to the calculated Fermi level. The red rhombus shows the simulation supercell shown in (e). The bright spots correspond to the regions where Ag atoms are around the hollow site of the graphene.

along the zigzag directions) to fit it into the supercell, to test the effect of the graphene cap on the STM simulation. It again shows similar features as in Fig. 4(b), further confirming that the line structures observed in experiments originate from the Ag-SiC superstructure and can be detected by STM through the graphene layer.

The total DOS of the C- and h-registries and the transition region (Fig. S13 of the Supplemental Material [24]) show features similar to those observed in the STS spectra in Fig. 2(b), but do not allow direct one-to-one mappings between peaks without shifting energies. This indicates strong interactions between local environments and the 1D superstructure must be considered as a whole system to reproduce local DOS features comparable to those observed in STM images.

Next, we show that the superstructure observed in the Ag_2 phase, which coexists in a monolayer with the Ag_1 , has a completely different origin. This Ag_2 phase arises from the moiré pattern induced by the lattice mismatch between Ag and graphene/SiC, as observed in other intercalated metals [1,16,19,20]. Figure 5(a) shows a well-defined superstructure of bright dots with a constant of ~ 14.7 Å as indicated by the blue rhombus. The zoomed STM image (taken at 0.05 V) from the white rectangle in Fig. 5(a), which is shown in Fig. 5(c), reveals that the superstructure periodicity is equal to roughly

six-unit cells of graphene with the same alignments. The origin of this superstructure can be explained by comparison with the STM simulation discussed below, as the moiré pattern formed by the lattice mismatch between graphene and the Ag layer underneath despite the individual intercalated Ag atoms are not resolved [1,16,19,20]. Moreover, the dI/dV spectra measured at the bright and dark sites of the superstructure [Fig. 5(d)] exhibit no discernible difference in the density of states.

Interestingly, at −0.75 V, a new superstructure is identified in the same scanning area of Fig. 5(a) with a periodicity about twice that of the original periodicity (~ 29 Å), as marked by a cyan rhombus and shown in Fig. 5(b). This supercell presumably originates from the commensuration between the quasi-free-standing graphene (13×13) and the SiC ($6\sqrt{3} \times 6\sqrt{3}$) (~ 32 Å), demonstrated as the large supercell in Fig. 5(e).

The origin of the superstructure observed in the Ag_2 phase can be explained by matching a $(3\sqrt{3} \times 3\sqrt{3})R30^\circ$ Ag_2 lattice with a lattice constant of ~ 2.98 Å (that is $3\sqrt{3} \times 2.98 = 15.48$ Å) over a (5×5) SiC (15.4 Å). This indicates that the Ag-Ag bonding length in Ag_2 is close to that of the bulk Ag(111) (2.88 Å), in good agreement with the LEED measurements [22]. This epitaxy is referred to as the (27:25)

structure, considering the Ag:SiC atomic ratio. The total and Ag₂-graphene partial DOS are presented in Fig. S14 of the Supplemental Material [24].

The partial charge density STM simulation of Ag₂ requires a graphene layer on top, as the moiré superstructure is the result of commensurability between the Ag layer and the graphene. The smallest supercell with relatively small strain for the graphene layers ($\sim 1\%$) and Ag layer ($\sim 2\%$) is shown in Fig. 5(e) with 13×13 graphene, 11×11 Ag, and $6\sqrt{3} \times 6\sqrt{3}$ SiC unit cells. Similar to the reasons discussed for the Ag₁ STM simulation, we focus on a conduction band energy window in Ag₂ from the band gap center to 0.75 eV above it (see Fig. S14 of the Supplemental Material [24]) and thus the $[-0.4, 0.35]$ eV window in [Fig. 5(f)] relative to the calculated Fermi level. By comparing the STM simulation to the atomic structure, we found that the bright moiré pattern dots originate from the local structures where Ag atoms reside at the hollow sites of the graphene. Due to the periodic boundary condition of the (13×13) graphene unit cells, the moiré pattern lattice constant is 6.5 times the graphene lattice size [Fig. 5(f)]. It is therefore deducible that, in real systems without such an artificial strain, the distance between bright features should be closer to six times the graphene unit cell, consistent with the experiment.

IV. CONCLUSIONS

To summarize, we have investigated the structural and electronic properties of different Ag structural phases intercalated between graphene and SiC, focusing in particular on the origin of the striped reconstruction observed in the confined silver monolayer. This long-range reconstruction is formed through strain-induced effects driven by the competition

between Ag-Ag and Ag-SiC interactions. The resulting tensile strain in the epitaxial Ag layer is partially relaxed through the rearrangement of Ag atoms in a one-dimensional $21 \times \sqrt{3}$ supercell, realized as one-dimensional Frenkel-Kontorova domains. This structure reconstruction gives rise to an electronic state at ~ 0.75 eV above the Fermi level, highlighting potential modulation in the density of states induced by the coupling between Ag and SiC substrate at the confined graphene/SiC interface. Our findings provide direct experimental and theoretical insight into the complex behavior of intercalated metals, revealing how the interplay between the metal-metal and metal-SiC interactions can be exploited to modulate the structural and electronic properties, therefore enabling possible atomic-level control of these 2D quantum materials.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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