

Mott states proximitized to a relativistic electron gas in epitaxial graphene

Chitran Ghosal¹, Siheon Ryee^{2,*}, Zamin Mamiyev¹, Maria-Elisabeth Federl⁵, Niklas Witt^{2,3,4},
Tim Wehling^{2,3} and Christoph Tegenkamp^{1,†}

¹*Solid Surface Analysis, Institute of Physics, Chemnitz University of Technology, Reichenhainer Strasse 70, 09126 Chemnitz, Germany*

²*I. Institute of Theoretical Physics, University of Hamburg, Notkestrasse 9, 22607 Hamburg, Germany*

³*The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany*

⁴*Institute for Theoretical Physics and Astrophysics, University of Würzburg, Am Hubland, 97074 Würzburg, Germany*

⁵*Department for Experimental and Applied Physics, University of Regensburg, Universitätsstraße 31, 93053 Regensburg, Germany*



(Received 6 December 2024; revised 25 May 2025; accepted 28 May 2025; published 12 June 2025)

Graphene, renowned for its exceptional electronic and optical properties as a robust two-dimensional (2D) material, traditionally lacks electronic correlation effects. Proximity coupling offers a promising method to endow quantum materials with novel properties. In this study, we realized a 2D heterostructure by intercalating Sn into zero layer graphene on SiC(0001), allowing us to explore the coupling between a correlated 2D electron gas and a Dirac metal. This results in the stabilization of Sn- $\sqrt{3}$ superlattice structures at the interface, which reveal Mott-Hubbard bands, in qualitative agreement with both experimental observations and theoretical predictions.

DOI: [10.1103/vk9s-zzcw](https://doi.org/10.1103/vk9s-zzcw)

I. INTRODUCTION

Strong electronic correlation effects are prevalent in low-dimensional systems due to reduced Coulomb screening and the low kinetic energy of charge carriers. Thereby, two-dimensional (2D) materials play a special role in this context, as different stacking sequences may come along with new quantum states [1–7].

A central concept in stacked systems is moiré structures, which have demonstrated superconducting, correlated insulating, and topological behavior, such as in twisted bilayer graphene [2,3]. This controlled symmetry breaking and formation of flatbands can be extended to multilayer systems, including rotated graphene bilayer systems [8]. Transition metal dichalcogenides (TMDs), inherently three-layer systems, also fit into this category. For example, the preserved inversion symmetry in 1T-TaS₂ results in an insulating charge density wave system, while the inversion-broken 2H-TaS₂ 2D layer exhibits Ising superconductivity [9,10]. The Fermi-surface nesting in these systems is highly sensitive to doping. Instead of an antiferromagnetic ground state, quantum fluctuations prevent magnetic order, leading to spin-liquid behavior [10]. Additionally, stacks of TMDs, such as 4Hb-TaS₂ and heterogeneous Ta-dichalcogenide bilayers, display a delicate interplay of correlated insulator, charge transfer, and heavy fermion physics [11–13] leading to signatures of exotic quan-

tum states like chiral superconductivity [14]. Furthermore, interlayer correlation effects and charge transfer are critical for the superconductivity observed in bilayer nickelates [15,16].

The overarching theme of these correlated states is driven by Mott physics. The physics of proximity coupling in these systems can be successfully explained using extended Hubbard and heavy fermion models, which describe insulating Mott- and Wigner-Mott systems and consider the roles of spin and charge order as well as band filling factors [5,17–20].

The interfaces of the aforementioned systems play a crucial role in realizing modulated periodic potentials. Epitaxial growth is a well-established method for achieving extremely clean and atomically well-defined interfaces. For example, epitaxial graphene can be grown on large scales and is suitable for quantum metrology [21–23]. Moreover, the sp^2 -hybridized sheet of C atoms on SiC(0001) is ideal for subsequent intercalation. By using various elements for intercalation between the buffer layer (BL) and the SiC surface, the 2D carbon layer is lifted, revealing the honeycomb structure and the Dirac cone. Intercalation has been used to control interface charges, enabling charge-neutral, p -type, or n -type graphene (e.g., H [24], Ge [25], Au [26], S [27], Pb [28]). Recently, extreme doping schemes with lanthanides, such as Gd, Yb, and Tb, have garnered research interest due to their ability to enforce correlation effects in monolayer graphene. These effects include band renormalization [29], Lifshitz transitions [30], and Kohn-Luttinger superconductivity [31].

Among these, group-IV elements adsorbed on semiconducting surfaces have attracted attention for their strong spin-orbit coupling and correlated electronic behavior. Phenomena such as the Mott insulating state, chiral superconductivity, and topological edge states have been observed on these surfaces [32–38]. Submonolayer coverages of Sn on SiC(0001) have been shown to host a robust Mott state [39]. Thus, the intercalation of Sn into BL/SiC structures is a promising prototype

*Contact author: sryee@physnet.uni-hamburg.de

†Contact author: christoph.tegenkamp@physik.tu-chemnitz.de

system for bringing a correlated state into proximity with a highly mobile electron gas. Recent studies using plasmon spectroscopy have examined Sn intercalation in greater detail. A fully saturated interface layer results in quasifree and charge-neutral graphene, while Sn submonolayer structures produce *n*-type doped graphene [40,41].

In this paper, we demonstrate that the proximity coupling of an electronically correlated 2D electron gas to Dirac electrons in graphene reveals signatures of both a Dirac metal and strongly correlated Hubbard bands. Using scanning tunneling microscopy and spectroscopy, supplemented by electron diffraction and electron energy loss spectroscopy, we identify the Mott-Hubbard bands from the interface layer and observe weak renormalization effects on the Dirac states. Through dynamical mean-field theory (DMFT) calculations, we reveal that besides the hybridization and Coulomb interaction also the charge transfer plays an important role for electronic states that emerge in such systems.

II. MATERIALS AND METHODS

A. Phase preparation and analysis

The experiments were performed in various ultrahigh vacuum (UHV) systems, all operating at base pressures of 5×10^{-9} Pa. Samples were transferred between systems without breaking the vacuum using a UHV suitcase. The Sn intercalation process and interface structures were controlled and studied with a spot profile analysis (SPA) low-energy electron diffraction (LEED) system. Plasmonic and interband excitations were measured using high-resolution electron energy loss spectroscopy (EELS) LEED, providing high-energy and -momentum resolution, typically around 25 meV and $0.001 \text{ \AA}^{-1}$ [42,43]. Scanning tunneling microscopy (STM) experiments were performed at 4 K using W tips, and scanning tunneling spectroscopy (STS) was conducted with a lock-in technique (4 mV, 800 Hz). The STS data were fitted using Gaussian functions and a parabolic background. In the case of noninteracting Sn layers on graphene, one would expect a linear background, resembling the density of states of graphene. However, to account for phonon-assisted tunneling, doping effects, and hybridization with the tin layer, a parabolic background proved to be a more suitable and stable model for fitting the spectra. The Hubbard band positions were described using Gaussian functions. Since the states exhibit substructure, consistent with our calculations, each Hubbard band was modeled using two Gaussian functions. The average position of each Hubbard band was determined by intensity-weighted peak positions.

BL graphene samples were grown epitaxially by Si sublimation on the Si face of 6H-SiC(0001) substrates as described in detail in Ref. [21]. For the intercalation experiments, Sn was evaporated at room temperature from a Mo crucible at a deposition rate of 0.15 monolayer/min, followed by several annealing steps until LEED reveals $\sqrt{3} \times \sqrt{3}$ spots as a hallmark for a Mott phase on SiC(0001) [39]. After intercalation, the Sn-interface layer is covered by a single layer of graphene. The Sn coverage was checked using a quartz microbalance and calibrated via the α -Sn reconstruction on

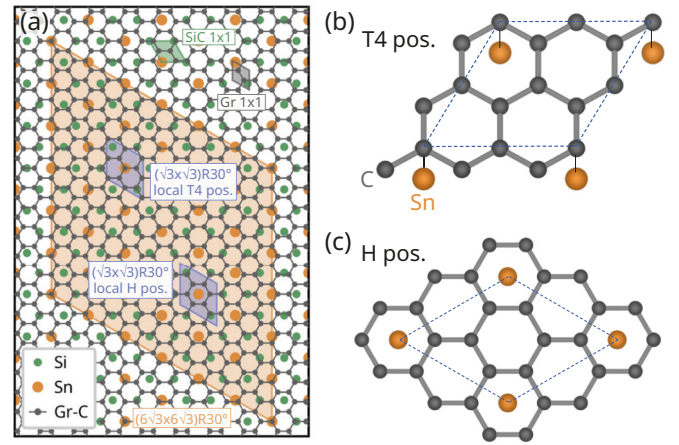


FIG. 1. (a) Illustration of the full $6\sqrt{3} \times 6\sqrt{3}$ supercell reconstruction of graphene on SiC(0001) and $\sqrt{3} \times \sqrt{3}$ supercells capturing the Sn superstructure with different local alignments of approximate 2×2 graphene sheets with respect to SiC. The unit cells of graphene and SiC are also shown. For the SiC substrate, only the topmost Si atom is depicted. (b), (c) Crystal structure of the approximate $\sqrt{3} \times \sqrt{3}$ cell model for graphene/Sn in the stacking configuration of Sn being in the T4 position or in the center of the hexagon (H). The unit cell is highlighted with blue dashed lines and the SiC substrate is not shown.

Si(111) as outlined in Ref. [40]. Temperatures were measured using an infrared pyrometer (Impac IGA).

B. Electronic structure calculations

To describe the electronic structure of graphene in proximity to a Sn layer with $(\sqrt{3} \times \sqrt{3})$ symmetry on the SiC substrate, we perform density functional (DFT) calculations of the layered heterostructure. We employ the Vienna Ab Initio Software Package [44–46] within the generalized gradient approximation of Perdew, Burke, and Ernzerhof [47] and the projector augmented wave [48,49] basis sets. We use a $12 \times 12 \times 1$ *k* mesh, a plane wave cutoff of 400 eV, and DFT-D3 van der Waals corrections [50] in our calculations. We relaxed the distance between the Sn layer and graphene using the conjugate gradient algorithm for ionic relaxation until all force components are smaller than $0.005 \text{ eV \AA}^{-1}$. The data of the DFT calculations are made available in Ref. [51].

In our calculation we address a simplified structure to account for the main experimental findings. Instead of the rather large supercell associated with $6\sqrt{3} \times 6\sqrt{3}$ reconstruction of graphene on the SiC(0001) substrate [52], we use a smaller $\sqrt{3} \times \sqrt{3}$ cell in our DFT calculations [53] which fully contains the Sn- $(\sqrt{3} \times \sqrt{3})$ triangular lattice reconstruction [39]. As is seen in Fig. 1(a), this $\sqrt{3} \times \sqrt{3}$ supercell approximates local registries of the full $6\sqrt{3} \times 6\sqrt{3}$ supercell with varying stacking configuration of the graphene sheet relative to the SiC surface. In our approximate supercell, graphene is stretched to fit in a 2×2 arrangement on SiC. We consider two limiting cases of the stacking configuration: the Sn atoms are located either right below a C atom [Fig. 1(b), “T4” position] or below the center of the hexagon [Fig. 1(c), “H” position]. They realize the maximal and the minimal hybridization between Sn and C atoms, respectively.

C. Model Hamiltonian

For studying the strongly correlated nature of p_z electrons stemming from Sn- $(\sqrt{3} \times \sqrt{3})$ on SiC(0001) [39], we consider an effective low-energy model with Hamiltonian $\mathcal{H} = H_0 + H_{\text{int}}$ capturing the DFT band structure (see Fig. 8 below). In the kinetic part H_0 of our model, we only consider contributions by Sn and graphene p_z orbitals as

$$H_0 = \sum_{i \neq j, \sigma} (t_{ij}^C c_{i\sigma}^\dagger c_{j\sigma} + t_{ij}^{\text{Sn}} d_{i\sigma}^\dagger d_{j\sigma}) + \sum_i (\epsilon_C n_i^C + \epsilon_{\text{Sn}} n_i^{\text{Sn}}) + \sum_{i\sigma} V_{ij} (c_{i\sigma}^\dagger d_{j\sigma} + \text{H.c.}). \quad (1)$$

The indices i and j refer to the in-plane lattice sites and σ ($\sigma \in \{\uparrow, \downarrow\}$) denotes the electron spin. $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) is the electron creation (annihilation) operator for graphene and $d_{i\sigma}^\dagger$ ($d_{i\sigma}$) for Sn. The corresponding number operators are $n_i^C = \sum_\sigma n_{i\sigma}^C = \sum_\sigma c_{i\sigma}^\dagger c_{i\sigma}$ and $n_i^{\text{Sn}} = \sum_\sigma n_{i\sigma}^{\text{Sn}} = \sum_\sigma d_{i\sigma}^\dagger d_{i\sigma}$, respectively. $t_{ij}^{C/\text{Sn}}$ is the hopping amplitude between sites i and j . We hereafter simply denote the n th neighbor hopping by $t_n^{C/\text{Sn}}$. For C sites (graphene), we use only the nearest-neighbor hopping $t_1^C = -2.8$ eV which is the typical value for the pristine graphene [54]. For Sn sites, we set $t_1^{\text{Sn}} = 27.3$ meV used in Ref. [39] and $t_2^{\text{Sn}} = -0.3881 t_1^{\text{Sn}}$ and $t_3^{\text{Sn}} = 0.1444 t_1^{\text{Sn}}$ for longer-range hopping by referring to the *ab initio* data for Sn- $(\sqrt{3} \times \sqrt{3})$ on Si(111) [34,55]. $\epsilon_{C/\text{Sn}}$ is the on-site energy level with respect to the chemical potential. The last term in Eq. (1) couples graphene and Sn- $(\sqrt{3} \times \sqrt{3})$ via V_{ij} , which accounts for the hybridization between Sn and C atoms. To model V_{ij} , we adopt an approximation used for parametrizing interlayer couplings in the twisted bilayer graphene [56,57], which reads

$$V_{ij} = V_0 \exp\left(-\frac{\sqrt{l_{ij}^2 + l_\perp^2} - l_\perp}{\delta_0}\right) \frac{l_\perp^2}{(l_{ij}^2 + l_\perp^2)}, \quad (2)$$

where V_0 is the hybridization strength between vertically located Sn and C atoms, l_{ij} the in-plane distance between atoms at sites i and j , $l_\perp = 3.45$ Å the DFT-optimized vertical spacing between the Sn layer and graphene, and $\delta_0 = 0.184 a$ the decay length (a is the distance between the neighboring C atoms in graphene). Thus we have only a single free parameter V_0 . For simplicity, we truncate V_{ij} for $l_{ij} > a$ since V_{ij} decays exponentially with l_{ij} . We use V_0 and the crystal field $\Delta \equiv \epsilon_{\text{Sn}} - \epsilon_C$ as our control variables.

To account for correlation effects, we use the Hubbard interaction for H_{int} which reads

$$H_{\text{int}} = U \sum_i (n_{i\uparrow}^d - \langle n_{i\uparrow}^d \rangle_0) (n_{i\downarrow}^d - \langle n_{i\downarrow}^d \rangle_0). \quad (3)$$

Here, U denotes the local Coulomb (Hubbard) interaction for electrons sitting on the same Sn site, and $\langle n_{i\sigma}^d \rangle_0$ is the electron occupation in the noninteracting case ($U = 0$). The above expression for H_{int} implies that our H_0 already includes the static mean-field self-energy à la the DFT, and thus H_{int} accounts for only dynamical correlations.

D. DMFT calculations

The Hamiltonian $\mathcal{H}$ is solved using the DMFT to address nonperturbatively local quantum fluctuations [58]. A numeri-

cally exact hybridization-expansion continuous-time quantum Monte Carlo algorithm is used as an impurity solver [59,60]. We restrict ourselves to paramagnetic phases without any spatial symmetry breaking, and we set the temperature to $T = 0.005$ eV $\simeq 58$ K. We use $U = 1.2$ eV as a representative value considering the previous studies on Sn surface systems [39]. To calculate the momentum-dependent spectral functions $A(\mathbf{k}, \omega)$ ($\mathbf{k}$, crystal momentum; ω , real frequency) and the momentum-integrated spectral function $A(\omega)$, we analytically continue the DMFT local self-energy $\Sigma(i\omega_n)$ (ω_n : fermionic Matsubara frequency) defined on the Matsubara frequency axis to the real frequency axis by employing the maximum entropy method [61,62]. The chemical potential is adjusted during the DMFT self-consistency to ensure the average electron occupation $\langle n \rangle$ of $\langle n \rangle = 9$ per unit cell, with eight electrons from the eight carbon atoms and one electron from the Sn atom per unit cell.

III. RESULTS AND DISCUSSION

A. Structure of the EG/Sn interface: SPA-LEED and STM

The intercalation process of Sn and formation of the $\sqrt{3} \times \sqrt{3}$ phase were monitored by SPA-LEED and STM. In Fig. 2 we show a sequence of diffraction images before and after intercalation as well as partial deintercalation of Sn. Starting with the BL/SiC(0001), the LEED pattern reveals all characteristic spots of the $6\sqrt{3} \times 6\sqrt{3}$ reconstruction of the carbon honeycomb structure with the SiC(0001) surface [52,63]. By intercalation of Sn the BL is transformed into a quasifree monolayer graphene. We adsorbed 5 ML of Sn on BL/SiC(0001) at room temperature and annealed the film subsequently at 1070 K for 15 min [40]. The LEED pattern in Fig. 2(b) revealed the same symmetry, but shows in addition the so-called bell shape background at all graphene integer spots, which is a hallmark for the formation of free-standing graphene [64]. In agreement with previous measurements and also the STM data, this phase refers to an intercalated Sn-(1 × 1) phase. Although crystalline Sn phases are formed only locally, the dangling bonds of the SiC substrate are on average saturated coming along with charge neutral graphene, e.g., seen in angle-resolved photoemission spectroscopy measurements [65]. Since the formation of a Sn- $(\sqrt{3} \times \sqrt{3})$ phase by direct intercalation of 1/3 of a monolayer of Sn did not succeed so far, we started out with the fully saturated interface phase. Annealing the surface to 1320 K for 5 min, referring to one annealing step, came along with the emergence of $(\sqrt{3} \times \sqrt{3})$ reflexes in the SPA-LEED pattern, highlighted in the inset of Fig. 2(c). Since this phase is formed by deintercalation, the domains are limited in size but can be enlarged by subsequent annealing cycles, i.e., the full width at half maximum (FWHM) of the $\sqrt{3}$ reflexes decreases while normalized integral intensity increases as shown in Fig. 2(e). From the analysis of the FWHM of the $\sqrt{3}$ spot at an out-of-phase scattering condition the domain sizes vary between 20 and 80 nm [66].

In Fig. 3(a) we show a typical large scale STM image after the Sn monolayer intercalation and partial deintercalation. As is obvious, the surface is inhomogeneous and mainly three different phases can be identified. Depending on the bias voltage, in Figs. 3(b) and 3(c) the graphene

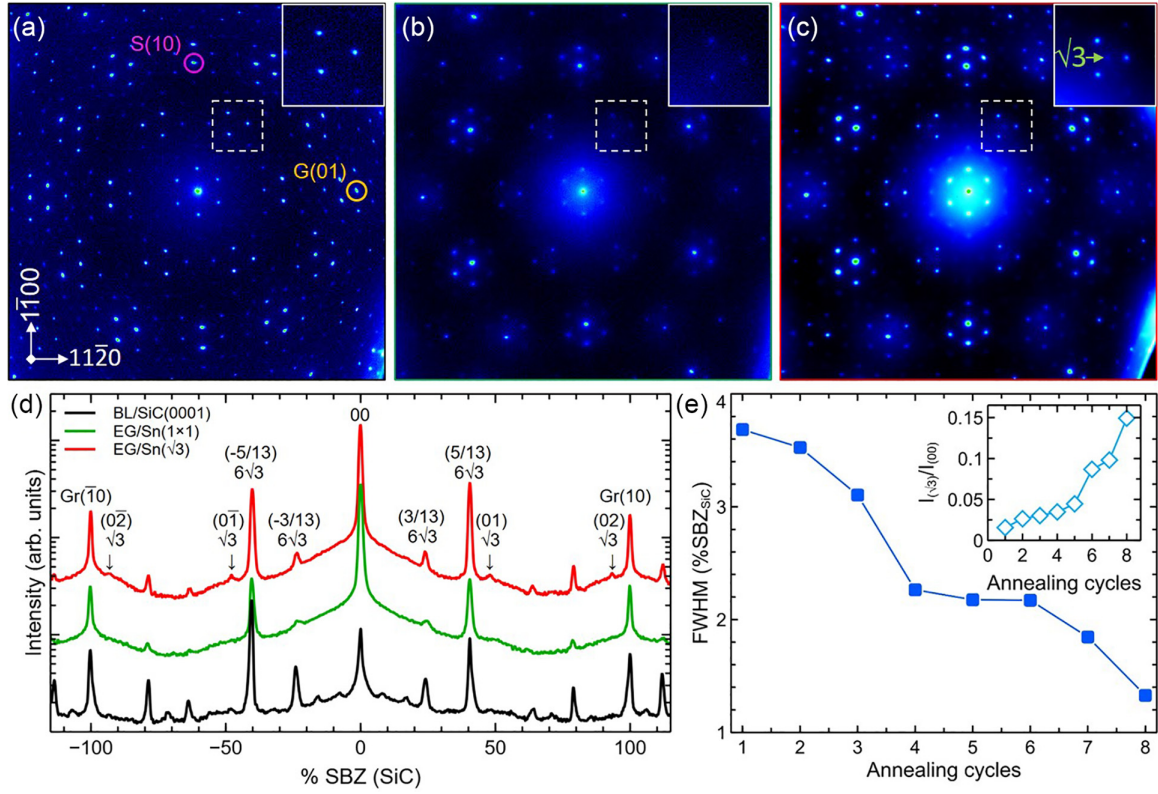


FIG. 2. (a) SPA-LEED image of the buffer layer (BL) before intercalation of Sn. (b) SPA-LEED image after intercalation of a full monolayer of Sn. (c) LEED image after deintercalation of Sn revealing $(\sqrt{3} \times \sqrt{3})$ reflexes. All LEED images were taken at $E = 150$ eV and 300 K. (d) Line scans of all three phases taken along the $[11\bar{2}0]$ direction. (e) FWHM and normalized integral intensity (inset) of a $(\sqrt{3} \times \sqrt{3})$ spot after different cycles of annealing. The profiles were fitted with Voigt profiles including a linear background.

lattice and atomic structure of the interface are shown. This phase refers to the fully intercalated interface structure and shows partially a Sn-(1 $\times$ 1) reconstruction. The proximity of these crystalline Sn islands to the graphene layer induces the

intervalley scattering, thus the graphene shows a $(\sqrt{3} \times \sqrt{3})$ -reconstruction [67]. In Fig. 3(d) we show an apparently more or less fully deintercalated area. Irrespective of the STM bias

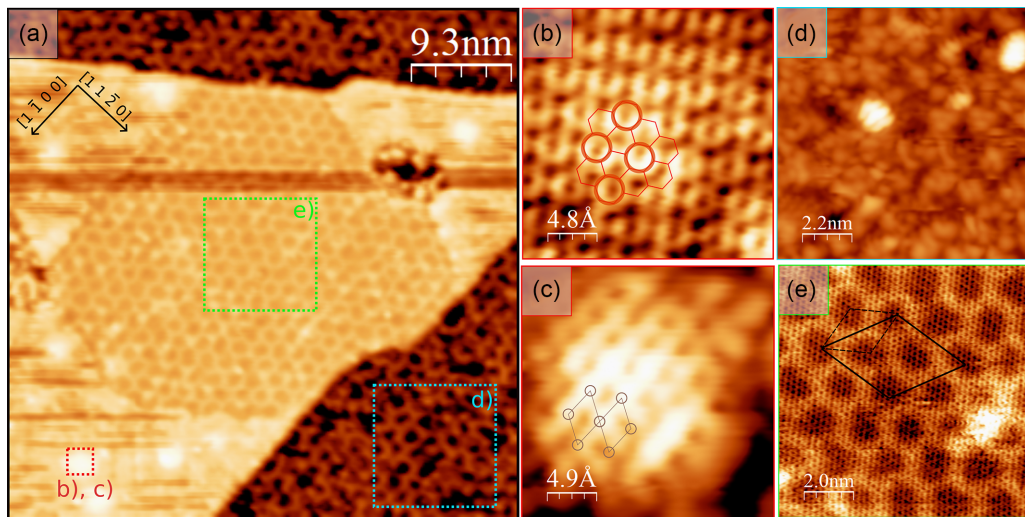


FIG. 3. (a) Large scale STM image (+2.0 V, 0.5 nA) showing three different phases, i.e., Sn-(1 $\times$ 1) [red square, zoom in shown in (b) and (c)], disordered BL [blue square, (d)] and Sn- $\sqrt{3}$ [green square, (e)]. (b), (c) Atomically resolved STM of the Sn-(1 $\times$ 1) phase at (b) 100 mV, 25 pA and (c) 1 V, 350 pA showing the graphene lattice (hexagons), the $\sqrt{3} \times \sqrt{3}$ reconstruction (circles), and the (1 $\times$ 1) interface structure, respectively. (d) Disordered phase measured at +1.5 V, 500 pA. (e) Sn- $\sqrt{3}$ phase ($V = +0.5$ V, 500 pA) showing both the graphene lattice and a (6×6) periodicity (dashed rhombus). The solid rhombus marks the $(6\sqrt{3} \times 6\sqrt{3})$ unit cell.

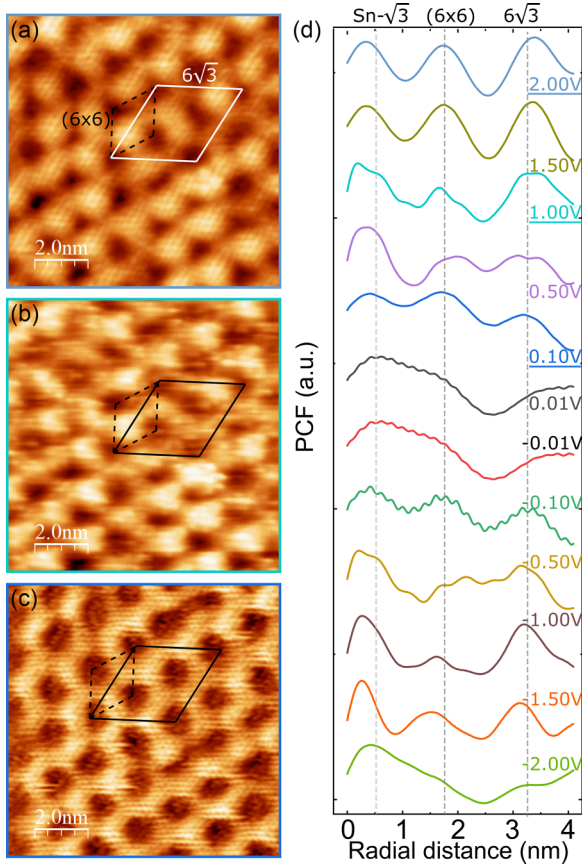


FIG. 4. (a)–(c) The STM area phase of the green box in Fig. 3 at different bias conditions: (a) +2 V, (b) +1 V, and (c) +0.1 V. (d) Pair correlation function (PCF) calculated from STM images taken at different bias voltages as indicated. The solid vertical lines show the distances of the main symmetries. At -0.5 - and $+1$ -V bias voltages, distances belonging to $\sqrt{3}a_{\text{SiC}} = 5.3$ Å appear most clearly. The dashed and solid rhombuses denote the (6×6) and $(6\sqrt{3} \times 6\sqrt{3})$ unit cells, respectively.

voltages, we were not able to resolve any periodic lattice structures, thus this phase is reminiscent of a disordered BL. The most important new phase for the further discussion is shown in Fig. 3(e). Besides the graphene lattice, the (6×6) symmetry of the former BL is seen. We will show in the following that this phase contains locally $\text{Sn-}\sqrt{3}$ phases, which are responsible for the additional LEED reflexes seen in Fig. 2(c). In contrast to the phase shown in Fig. 2(b), the graphene itself does not reveal any reconstruction [67]. Apart from the defect in Fig. 2(e), the unperturbed part of this phase is obviously not enabling intervalley scattering within the graphene layer. Under these experimental conditions, we observed only three distinct phases, in full agreement with the LEED patterns. At the very least, the formation of other ordered phases can be ruled out.

In Fig. 4 we show details of the phase which we associate with the $\text{Sn-}\sqrt{3}$ phase. The images taken at different bias voltages clearly show different contrasts, i.e., various states at the interface are involved in the tunneling process. While at +2 V mainly the $\text{SiC}(1 \times 1)$ lattice with the (6×6) symmetry is obvious, at small voltages close to the Fermi

energy the graphene lattice is seen [Fig. 4(c)]. At 1 V a high contrast is achieved where the protrusion apparently follows the former bonds and reveals the (6×6) superperiodicity.

In order to obtain a deeper insight into the underlying symmetries, we calculated the radial pair correlation functions from STM images taken at the same area but for different tunneling voltages ranging from +2 to -2 V, shown in Fig. 4(d). As shown, various distances belonging to different symmetries of the interface are found. Besides the distances belonging to the (6×6) and $(6\sqrt{3} \times 6\sqrt{3})$ symmetries, in particular for small bias voltages, the graphene lattice becomes visible (e.g. see the high frequency component at -0.1 V). Distances referring to the $\sqrt{3}$ symmetry show up only for two distinct voltages, namely -500 and $+1000$ mV. For these tunneling conditions also the second order of the $\sqrt{3}$ distance is visible. Compared to other intercalated systems, e.g. Pb or In [28,68], resolving the Sn structure with STM by locking to defined states of the interface layer is more difficult. This finding might already indicate more pronounced hybridization of Sn with graphene. In a recent theoretical study, it was shown that for group IV elements, the hybridization strength decreases with increasing atomic number (Z). Nevertheless, while Pb exhibits almost no interaction with epitaxial graphene, a moderate interaction of the Sn p orbitals with graphene has been reported [69]. We will show in the following that these states are related to Sn, thus the Sn forms locally a $\sqrt{3} \times \sqrt{3}$ reconstruction, which was shown on $\text{SiC}(0001)$ to host Mott states [39].

Apparently, the adsorption sites of Sn atoms in the diluted phase are determined by the previous bonding sites of the buffer layer. While the saturation of these former BL bonds is sufficient to form freestanding graphene, the Sn coverage is insufficient to saturate all dangling bonds of the SiC surface. Consequently, this diluted phase inevitably should result in a n -type doped graphene layer, which indeed was seen in previous EELS measurements [41]. The starting point for the formation of the $\text{Sn-}\sqrt{3}$ phase is the intercalated Sn monolayer. As the Sn density is gradually reduced by heating, the resulting diluted phase exhibits n -type doping of graphene. However, the continuous dilution requires desorption, which occurs most efficiently at larger defects, such as step edges. Consequently, the formation of an intercalated $\text{Sn-}\sqrt{3}$ phase is limited by interface diffusion. The intercalated monolayer phase appears to act as a reservoir. This complex interplay between diffusion in reduced dimensions and the desorption seems to explain the emergence of the three observed phases.

B. Electronic structure: EELS and STS

The aforementioned phases were further analyzed by means of electron energy loss spectroscopy. As explained above, the $\text{Sn-}\sqrt{3}$ phase can currently only be synthesized through a subsequent desorption of the intercalated Sn monolayer. This process results in a mixture of the intercalated Sn monolayer phase, the fully deintercalated phase, and the $\text{Sn-}\sqrt{3}$ phase, as confirmed by STM measurements in the previous section. As we will show, EELS, also as an integrating method, is a powerful tool for identifying the topic of interest here. Figure 5 displays EEL spectra of epitaxial graphene with a nominally intercalated $\text{Sn-}\sqrt{3}$ phase.

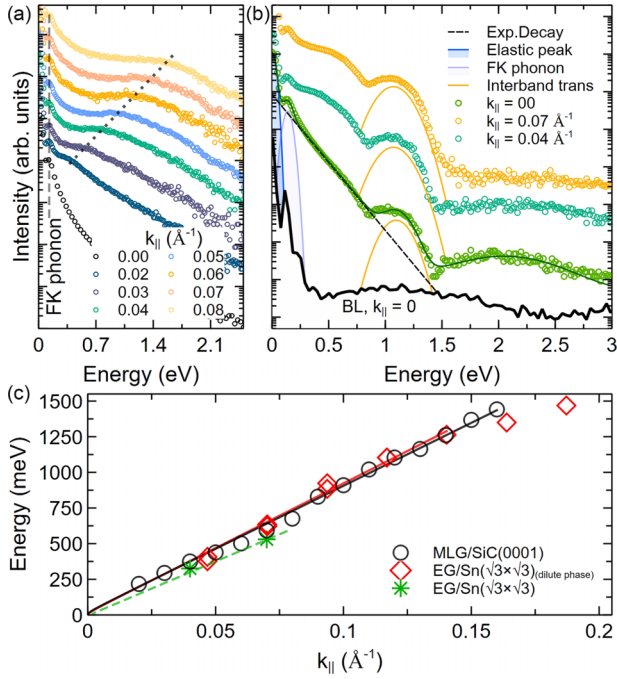


FIG. 5. EEL spectra of the Sn- $\sqrt{3}$ phase after different steps of annealing. (a) Loss spectra taken after the first appearance of $\sqrt{3}$ reflexes in LEED, showing the dispersing graphene sheet plasmon loss [41]. (b) Loss spectra after various heating cycles revealing besides the graphene plasmon an intense but nondispersing loss-peak at around 1.2 eV. For reference, also the loss spectrum of the BL is shown. (c) Sheet plasmon dispersions of the graphene plasmons deduced from the spectra in (a) (red diamonds) and (b) (green stars). For reference, also the dispersion for the sheet plasmon in monolayer graphene (MLG, black circles) is shown. The lines denote linear regressions. The experiments were taken at 300 K with a primary electron energy of 20 eV.

Figure 5(a) reveals a sheet plasmon loss peak that disperses with increasing the momentum transfer. The analysis of this collective mode indicates that this plasmon feature corresponds to n -type doped graphene, with a doping level of approximately $n = 7.8 \times 10^{12} \text{ cm}^{-2}$ carriers, shifting the Fermi level around $E_F = 330 \text{ meV}$ above the Dirac point [41]. The dispersion is plotted in Fig. 5(c)

Upon further heating cycles of the Sn- $\sqrt{3}$ phase the LEED intensity of the $\sqrt{3}$ reflexes increases [see Fig. 2(e)] and the loss spectrum undergoes significant changes, as shown in Fig. 5(b). The fully intercalated Sn monolayer phase is metallic, and the graphene remains charge neutral, exhibiting no plasmonic excitations [41]. In contrast, the fully deintercalated phase, which largely resembles a disordered BL, shows no losses in the spectral region of interest. As a result, EELS is selectively sensitive to the Sn- $\sqrt{3}$ phase. Besides the sheet plasmon, visible at lower loss energies, the spectrum shows a distinct nondispersing loss peak at 1.2 eV. The high loss intensity of this peak suggests also a high combined density of states, e.g., as for Mott-Hubbard bands. We will show below that this loss energy fits nicely to the electronic gap found in STS measurements and DMFT calculations. The fitting of the spectra reveals that the loss peak is astonishingly broad (FWHM=0.4 eV). For reference, we also present the

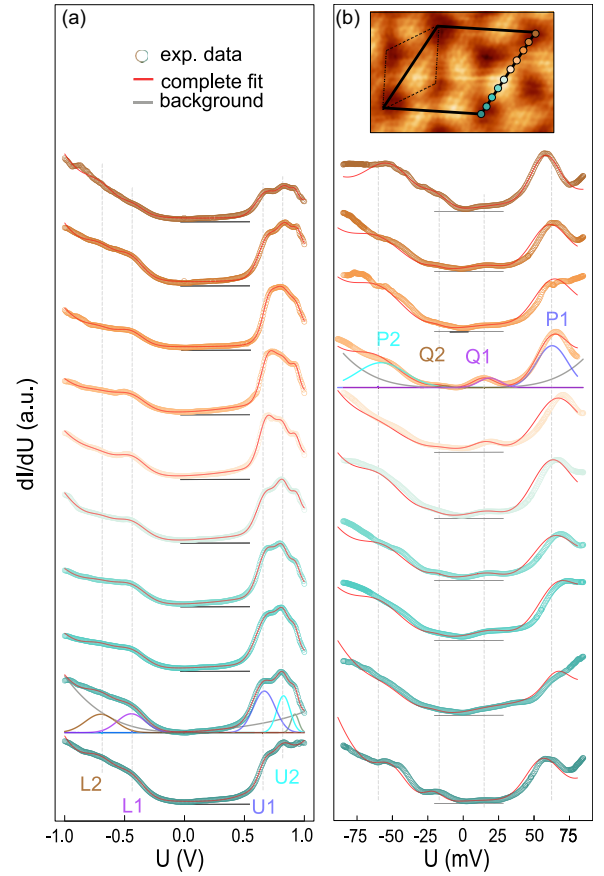


FIG. 6. High resolution STS spectra taken along the $6\sqrt{3}$ unit cell shown in the inset. (a) Voltage range from -1 to $+1$ V (set point $+2$ V, 500 pA). The color code refers to the positions shown in the inset. (b) Same sequence as in (a), but with a voltage range from -0.1 to $+0.1$ V (set point $+0.1$ V, 500 pA). The detailed fit model, consisting of five (left) and four (right) Gaussian functions and a background, allows for a consistent fitting for all spectra for the two regimes shown exemplarily.

loss spectrum of a BL, demonstrating that while the Sn- $\sqrt{3}$ phase shares the same symmetry as an ordinary BL, it exhibits distinct electronic properties. As mentioned, there is also the graphene plasmon at lower loss energies visible. The remaining dispersing nature of this state is highlighted through spectral fitting and its dispersion is plotted also in Fig. 5(c). From the similarity of both we conclude that freestanding graphene is indeed present and that also the doping has not severely changed. Therefore, in agreement with the evolution of the $\sqrt{3}$ intensity shown in Fig. 2(e), also EELS shows that the Sn- $\sqrt{3}$ domains become gradually larger by subsequent annealing.

High resolution dI/dV spectra taken along the $6\sqrt{3}$ unit cell in the range from -1 to $+1$ V are shown in Fig. 6(a). Most obvious are the peaks at -0.5 and around $+0.7$ V. The size of this gap fits nicely to the energy loss of 1.2 eV found in EELS [see Fig. 5(b)]. Each of the peaks consists of subpeaks, respectively, as deduced from the fitting of the data. The subpeak structure is for example shown for one spectrum. Moreover, the substructure of the initial and final states seems to be responsible for the large peak width of

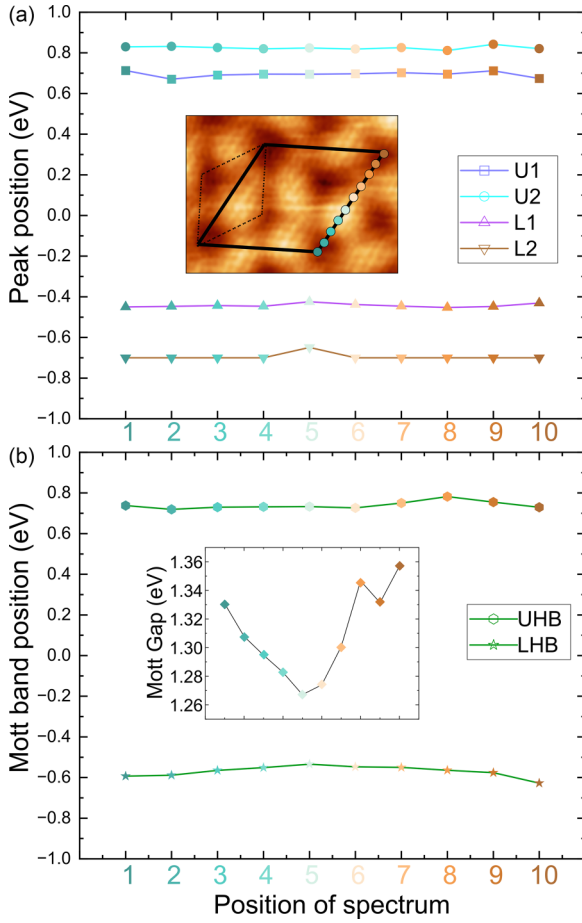


FIG. 7. Evaluation of the STS spectra shown in Fig. 6. (a) Peak positions of the lower and upper Hubbard band peaks, U1, U2, L1, and L2, across the unit cell as marked in the inset for the peaks discussed in context of Fig. 6(a). (b) Averaged peak positions of the U1, U2, L1, and L2 states. The inset shows the difference, i.e., the variation of the Mott gap across the unit cell.

around 0.3 eV. The detailed analysis of the peaks by our fitting routine revealed that the peak at around -0.5 eV is split into two peaks located at -430 meV (L1) and -680 meV (L2). Also the unoccupied state at $+0.7$ eV reveals substructures U1 and U2. In view of the DMFT calculations presented hereafter, these states seem to resemble the lower Hubbard band (LHB) and upper Hubbard band (UHB). As obvious, the energetic positions of the peaks and their intensity show some slight variation along the axis of the $(6\sqrt{3} \times 6\sqrt{3})$ unit cell. Details will be discussed in the context of Fig. 7(a). The STS measurements revealed splitting within the Hubbard bands, which is consistent with the DMFT calculations (see below).

In Fig. 6(b) high resolution STS spectra around the Fermi level are shown. Details were again deduced from fitting the data, as exemplarily for one spectrum. Within this energy window mainly four peaks are seen. The outer ones (P1, P2) at around ± 60 meV define a gaplike feature around the Fermi level arising from a suppression of electronic tunneling to graphene states near E_F . At around 60 meV, an enhancement of electronic tunneling sets in due to a phonon-mediated inelastic channel [70]. The presence of this gap is a hallmark for

freestanding and defect-free graphene. Moreover, unlike the $\text{Sn}(1 \times 1)$ patches, we found also no Kekulé distortion in the $\text{Sn}-\sqrt{3}$ phases, thus we can assume here a comparably weak interaction between the graphene and Sn interface layer.

Moreover, the sequence of spectra shows peaks close to E_F at $Q2 = -18$ meV and $Q1 = 14$ meV. Most likely, these spectroscopic features hint towards signatures of a correlated metal due to the interplay between hybridization between the Sn layer and graphene, which can be partly reproduced by DMFT, however, by assuming a stronger hybridization or charge transfer.

We have conducted a detailed analysis of the Sn states. Figure 7(a) shows peak positions of the L1, L2, U1, and U2 states along the unit cell, as indicated in the inset. Including also the different intensities for each of the peaks and each of the positions, we determined the average position of the lower and upper Hubbard band, shown in Fig. 7(b). The bands and correspondingly the Mott gap, shown in the inset, vary across the unit cell. In context of the theoretical results, presented hereafter, T4 and H positions of the Sn atom with respect to the graphene lattice (see Fig. 1) give rise to different hybridization strengths, and thus to a variation of the Hubbard bands. The overall variation is on the order of 0.1 eV and explains the comparably large peak width seen in EELS (see Fig. 5) and agrees reasonably with our theoretical findings.

C. Interplay of hybridization, electronic correlation, and doping: DFT and DMFT

To understand qualitatively how the low-energy electronic structure of graphene is affected by the hybridization with Sn, we used a simplified $\sqrt{3} \times \sqrt{3}$ supercell with different graphene alignments (T4 and H positions) shown in Fig. 1 accounting for the experimental findings discussed in the context of Figs. 2–4.

The corresponding band structures from DFT are shown in Figs. 8(a) and 8(b) with fatbands of Sn and graphene p_z orbitals. Both stacking configurations show a flat band at the Fermi level made up by Sn- p_z orbitals coexisting with electron doped Dirac bands of graphene. Such a situation is reminiscent of the typical band structures of heavy fermion materials including f -electron compounds [71,72], magic-angle twisted graphene multilayers [7,73,74], and also transition metal dichalcogenide multilayer structures [13]. Close to the crossing of the two kinds of bands around the K point, we find hybridization-induced avoided crossings in the T4 structure, which are largely suppressed in the H structure. This stacking-induced modulation of the hybridization arises from the following selection rule: the graphene states at K transform according to the E_1 and E_2 representations under C_{6v} symmetry with respect to the hexagon centers, whereas Sn p_z orbitals belong to the A_1 representation [75]. This kind of selection rule does not apply in the T4 position.

Figures 8(c) and 8(d) present the band structures obtained from solving the tight-binding Hamiltonian H_0 [Eq. (1)] for $\Delta = 0.5$ eV and two representative hybridization strengths of $V_0 = 0.7$ and 1.3 eV neglecting correlation effects. The aforementioned selection rule manifests also here: when comparing the band structures with $V_0 = 0.7$ and 1.3 eV in the T4 structure, we see an enhancement of the corresponding

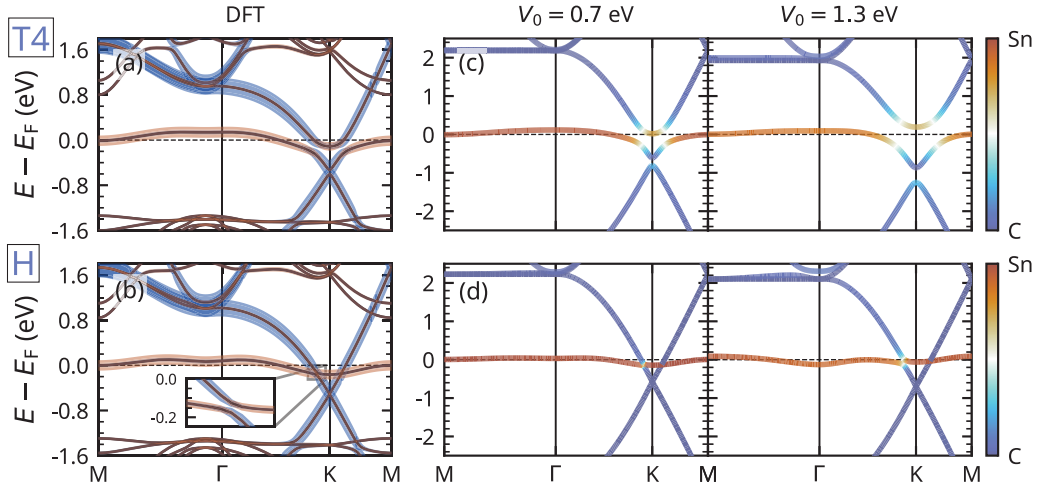


FIG. 8. Electronic structure from DFT with fatband highlighting of the orbital weight from Sn (orange) and graphene (blue) p_z orbitals for the (a) T4 and (b) H cell (see Fig. 1). The inset in (b) shows a closeup of the hybridization gap along the Γ K direction. (c), (d) Model band structure from Eq. (1) at $V_0 = 0.7$ and 1.3 eV without local Coulomb interaction ($U = 0$). The color bar indicates the spectral weight of orbital $m \in \{p_z^{\text{Sn}}, p_z^{\text{C}}\}$ in the lowest-lying noninteracting band.

hybridization gaps, while band structures for the H structure are almost insensitive to this change in V_0 . In comparison, the tight-binding bands shown in Figs. 8(c) and 8(d) closely replicate the corresponding DFT bands around the Fermi level for each structure presented in Figs. 8(a) and 8(b). Thus, $V_0 = 0.7$ and 1.3 eV should fall into a reasonable range of V_0 . We note that higher-energy bands from the insulating SiC substrate are excluded in the low-energy model because their exact positions are unclear due to the band gap problem of DFT, and they have negligible hybridization with other bands.

D. DMFT results

We now investigate how the Hubbard interaction modifies the electronic excitation spectra. Figures 9(a)–9(d) show the momentum-dependent spectral function $A(\mathbf{k}, \omega)$ and the momentum-integrated spectral function $A(\omega)$, which can be measured by photoemission or scanning tunneling spectroscopy, respectively.

The most notable and robust feature is the emergence of the UHB and the LHB from the flat Sn- p_z derived band for both T4 and H structures. This shows that regardless of the model details and correspondingly the local configuration of graphene, the Sn p_z orbitals are in a regime of strong Mott-Hubbard type correlations.

While strong correlations in the Sn p_z states are generic here, the stacking configurations (namely, T4 vs H), the hybridization strength V_0 , and the energy offset Δ can strongly affect the nature of electronic states in the Sn-graphene hybrid system. Due to suppressed hybridization near the Dirac point, the Hubbard bands are very sharp and closely resemble an atomic limit for the H structures at $\Delta = 0$ [lower panels in Figs. 9(a) and 9(b)]. In the T4 states, hybridization broadens the Hubbard bands [upper panels in Figs. 9(a) and 9(b)], but they robustly emerge even at values of V_0 larger than those suggested by the DFT calculations.

Interestingly, the spectra shown in Figs. 9(a) and 9(b) for weak to intermediate hybridization naturally explain several of our experimental findings. The “Mott gaps” (the energy level difference between UHB and LHB) on the order of 1 eV fit to the loss energy in EELS [see Fig. 5(b)] and match the gap between the L1/L2 and U1/U2 states in STS [see Fig. 6(a)], resembling the LHB and UHB, respectively. The Hubbard U for the Sn/SiC system [39] is considerably larger, showing here the importance of screening by the proximitized quasifree monolayer graphene. As already shown, the loss peak is comparatively broad. In addition, the STS measurements reveal a splitting of the Hubbard bands, which is according to the calculated spectra shown in Fig. 9. This can be understood as a hybridization effect, particularly in the T4 configurations [see upper panels in Figs. 9(a) and 9(b)]. The splitting of Hubbard-type bands through hybridization effects is quite common in correlated multiband systems as exemplified by the Zhang-Rice singlet from cuprate high- T_c superconductors [76–78]. To explore the potential for quasiparticle formation in our model and understand its implications for the low-energy peaks of our STS measurements, we examine the case of a larger V_0 of 1.9 eV, which exceeds the DFT-estimated value. Interestingly, quasiparticles begin to emerge at $\omega = 0$ for the T4 structure as shown in the upper panel of Fig. 9(c), whereas the LHB merely touches the Fermi level without a clear signature of a quasiparticle peak for the H structure [lower panel of Fig. 9(c)]. It implies that at temperatures below 58 K, a quasiparticle peak could develop even at a realistic value of V_0 for the T4 structure since the onset temperature of quasiparticle (Kondo) resonance decreases exponentially with decreasing V_0 . Indeed, the spectral function at $V_0 = 1.3$ eV [upper panel of Fig. 9(b)] shows a tantalizing signature of quasiparticle formation at lower temperatures, manifested as an incipient peak near $\omega = 0$. However, reaching the STS temperature of 4 K, at which a signature of a quasiparticle state is captured (see Fig. 6), is challenging in our calculations.

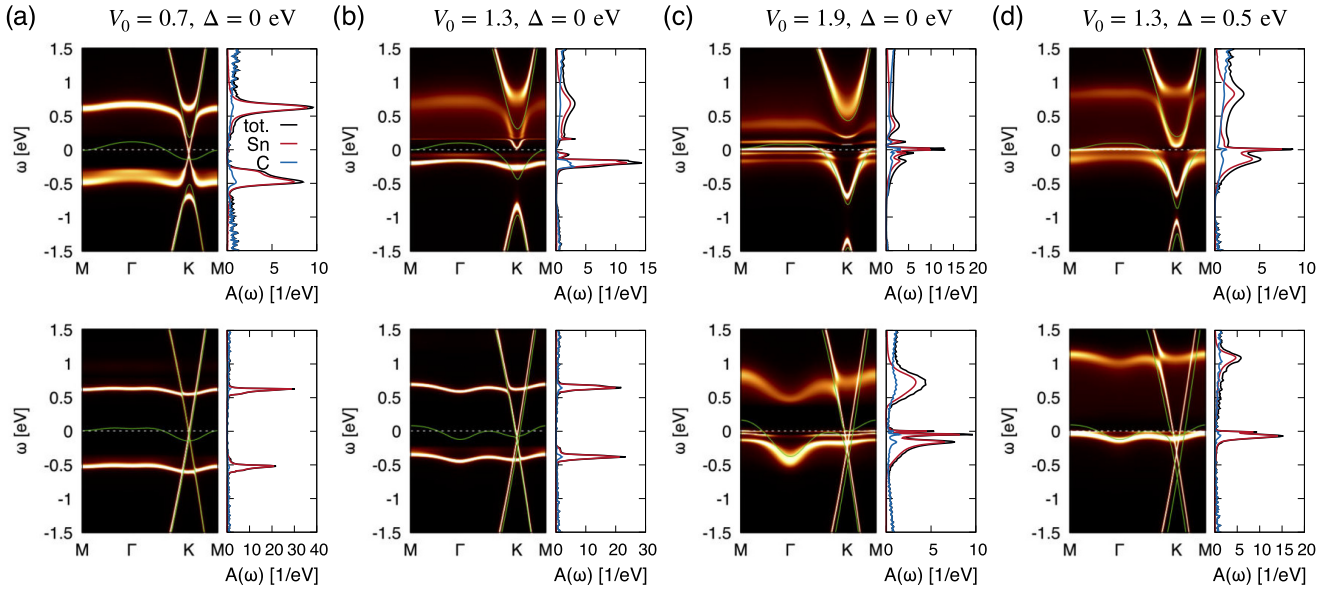


FIG. 9. (a)–(d) The momentum-dependent spectral function $A(\mathbf{k}, \omega)$ (color coded) and the orbital-resolved momentum-integrated spectral function $A(\omega)$ obtained from DMFT for $U = 1.2$ eV for different (a)–(c) hybridizations V_0 and (b), (d) crystal fields Δ . Top and bottom panels show spectra for the T4 and H configuration, respectively, analogous to Fig. 8. The chemical potential is at $\omega = 0$. The corresponding noninteracting bands (green solid lines) are overlaid.

Potential energy offsets directly affect the position of the Hubbard bands, leading from an (almost) particle-hole symmetric situation at $\Delta \rightarrow 0$ [Figs. 9(a) and 9(b)] towards a “mixed valence” case where the lower Hubbard band is pinned to the Fermi level [Fig. 9(d)]. Comparison of calculated spectra to our STS experiments suggests $|\Delta| \ll 0.5$ eV.

One of the appealing aspects of the system at hand is that graphene offers the opportunity of two-side functionalization and correspondingly doping of the system [79]. We thus study the case featuring stronger charge transfer closer [$\Delta = 0.5$ eV, Fig. 9(d)]: From the Hubbard band pinned to the Fermi level, quasiparticle peaks emerge both in the T4 and in the H configuration. Yet, the nature of the emergent state upon charge transfer is very different in each case. In the H structure, the Sn- p_z merely shifts but hybridization remains suppressed as it must be due to symmetry reasons. Thus, we have essentially a doped Mott insulator coexisting with a charge reservoir (the Dirac electrons of graphene). This situation bears a close resemblance to 1T/1H Ta-dichalcogenide bilayers [13] in which charge transfer from uncorrelated electrons (1H layer) to a Mott insulator (1T layer) gives rise to a correlated metal as observed in experiments.

On the other hand, in the T4 case the doped Mott state of the Sn p_z band strongly hybridizes with the graphene Dirac electrons as indicated in the upper panel in Fig. 9(d). This case is thus closer to systems like magic-angle twisted bilayer graphene featuring similar hybridization effects [2,3,73,74].

We note in closing that a variety of electronic orderings can emerge out of Mott-Hubbard states as temperature is lowered (e.g., antiferromagnetic spin ordering in Mott insulators [80]). In effect, it is possible that spin or charge ordering occurs in our system, despite the geometric frustration of the Sn layer complicating the spin-ordering scenario. Here, we can exclude the charge order or struc-

tural reconstruction as we would directly see this in our STM measurements.

IV. SUMMARY AND CONCLUSIONS

The Sn-intercalated graphene system resembles a novel platform of proximitized Dirac and correlated electrons. In the $\sqrt{3}$ patches, Sn p_z electrons exhibit robust correlation effects manifesting as characteristic Hubbard bands in STS and loss peaks in EELS. The system reveals spatially modulated hybridization between the Dirac and correlated electrons. Theoretical modeling and experiment show a reasonable agreement regarding the spectral properties, i.e., the Mott gap and the subpeak structure of the Hubbard bands, and reveal symmetry related selection rules as origin of the modulated hybridization.

Our analysis reveals that besides the hybridization and Coulomb interaction also the charge transfer plays an important role for the electronic state that emerges in these artificial correlated systems. While the former are defined by the specifics of the material, the charge transfer can be controlled in these epitaxial systems quite easily, e.g., by molecular doping with F4-TCNQ. Further experiments are underway to investigate systematically the interplay of doped Mott states in direct spatial proximity to more hybridized heavy fermions.

ACKNOWLEDGMENTS

We gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft through FOR5242 (Grants No. TE386/22-1 and No. WE5342/7-1). The DMFT calculations were done on the supercomputer Lise at NHR@ZIB as part of the NHR infrastructure under Project No. hhp00056.

DATA AVAILABILITY

The data of the DFT calculations are made available in Ref. [51]. The other/remaining data [or the experimental and DMFT data] that support the findings of this article are not

publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

-
- [1] H.-W. Guo, Z. Hu, Z.-B. Liu, and J.-G. Tian, Stacking of 2D materials, *Adv. Funct. Mater.* **31**, 2007810 (2021).
- [2] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, Unconventional superconductivity in magic-angle graphene superlattices, *Nature (London)* **556**, 43 (2018).
- [3] Y. Cao, V. Fatemi, A. Demir, S. Fang, S. L. Tomarken, J. Y. Luo, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, R. C. Ashoori, and P. Jarillo-Herrero, Correlated insulator behaviour at half-filling in magic-angle graphene superlattices, *Nature (London)* **556**, 80 (2018).
- [4] Y. Tang, L. Li, T. Li, Y. Xu, S. Liu, K. Barmak, K. Watanabe, T. Taniguchi, A. H. MacDonald, J. Shan, and K. F. Mak, Simulation of Hubbard model physics in WSe₂/WS₂ moiré superlattices, *Nature (London)* **579**, 353 (2020).
- [5] Y. Tang, K. Su, L. Li, Y. Xu, S. Liu, K. Watanabe, T. Taniguchi, J. Hone, C.-M. Jian, C. Xu, K. F. Mak, and J. Shan, Evidence of frustrated magnetic interactions in a Wigner-Mott insulator, *Nat. Nanotechnol.* **18**, 233 (2023).
- [6] K. P. Nuckolls and A. Yazdani, A microscopic perspective on moiré materials, *Nat. Rev. Mater.* **9**, 460 (2024).
- [7] B. A. Bernevig and D. K. Efetov, Twisted bilayer graphene's gallery of phases, *Phys. Today* **77**(4), 38 (2024).
- [8] M. He, Y. Li, J. Cai, Y. Liu, K. Watanabe, T. Taniguchi, X. Xu, and M. Yankowitz, Symmetry breaking in twisted double bilayer graphene, *Nat. Phys.* **17**, 26 (2021).
- [9] J. M. Lu, O. Zheliuk, I. Leermakers, N. F. Q. Yuan, U. Zeitler, K. T. Law, and J. T. Ye, Evidence for two-dimensional Ising superconductivity in gated MoS₂, *Science* **350**, 1353 (2015).
- [10] K. T. Law and P. A. Lee, 1T-TaS₂ as a quantum spin liquid, *Proc. Natl. Acad. Sci. USA* **114**, 6996 (2017).
- [11] W. Ruan, Y. Chen, S. Tang, J. Hwang, H.-Z. Tsai, R. L. Lee, M. Wu, H. Ryu, S. Kahn, F. Liou, C. Jia, A. Aikawa, C. Hwang, F. Wang, Y. Choi, S. G. Louie, P. A. Lee, Z.-X. Shen, S.-K. Mo, and M. F. Crommie, Evidence for quantum spin liquid behaviour in single-layer 1T-TaSe₂ from scanning tunnelling microscopy, *Nat. Phys.* **17**, 1154 (2021).
- [12] V. Vaño, M. Amini, S. C. Ganguli, G. Chen, J. L. Lado, S. Kezilebieke, and P. Liljeroth, Artificial heavy fermions in a van der Waals heterostructure, *Nature (London)* **599**, 582 (2021).
- [13] L. Crippa, H. Bae, P. Wunderlich, I. I. Mazin, B. Yan, G. Sangiovanni, T. Wehling, and R. Valentí, Heavy fermions vs doped Mott physics in heterogeneous Ta-dichalcogenide bilayers, *Nat. Commun.* **15**, 1357 (2024).
- [14] I. Silber, S. Mathimalar, I. Mangel, A. K. Nayak, O. Green, N. Avraham, H. Beidenkopf, I. Feldman, A. Kanigel, A. Klein, M. Goldstein, A. Banerjee, E. Sela, and Y. Dagan, Two-component nematic superconductivity in 4Hb-TaS₂, *Nat. Commun.* **15**, 824 (2024).
- [15] H. Sun, M. Huo, X. Hu, J. Li, Z. Liu, Y. Han, L. Tang, Z. Mao, P. Yang, B. Wang, J. Cheng, D.-X. Yao, G.-M. Zhang, and M. Wang, Signatures of superconductivity near 80 K in a nickelate under high pressure, *Nature (London)* **621**, 493 (2023).
- [16] S. Ryee, N. Witt, and T. O. Wehling, Quenched pair breaking by interlayer correlations as a key to superconductivity in La₃Ni₂O₇, *Phys. Rev. Lett.* **133**, 096002 (2024).
- [17] E. C. Regan, D. Wang, C. Jin, M. I. Bakti Utama, B. Gao, X. Wei, S. Zhao, W. Zhao, Z. Zhang, K. Yumigeta, M. Blei, J. D. Carlström, K. Watanabe, T. Taniguchi, S. Tongay, M. Crommie, A. Zettl, and F. Wang, Mott and generalized Wigner crystal states in WSe₂/WS₂ moiré superlattices, *Nature (London)* **579**, 359 (2020).
- [18] Y. Shimazaki, I. Schwartz, K. Watanabe, T. Taniguchi, M. Kroner, and A. Imamoğlu, Strongly correlated electrons and hybrid excitons in a moiré heterostructure, *Nature (London)* **580**, 472 (2020).
- [19] H. Li, S. Li, E. C. Regan, D. Wang, W. Zhao, S. Kahn, K. Yumigeta, M. Blei, T. Taniguchi, K. Watanabe, S. Tongay, A. Zettl, M. F. Crommie, and F. Wang, Imaging two-dimensional generalized Wigner crystals, *Nature (London)* **597**, 650 (2021).
- [20] A. Ghiotto, E.-M. Shih, G. S. S. G. Pereira, D. A. Rhodes, B. Kim, J. Zang, A. J. Millis, K. Watanabe, T. Taniguchi, J. C. Hone, L. Wang, C. R. Dean, and A. N. Pasupathy, Quantum criticality in twisted transition metal dichalcogenides, *Nature (London)* **597**, 345 (2021).
- [21] K. V. Emtsev, A. Bostwick, K. Horn, J. Jobst, G. L. Kellogg, L. Ley, J. L. McChesney, T. Ohta, S. A. Reshanov, J. Röhr, E. Rotenberg, A. K. Schmid, D. Waldmann, H. B. Weber, and T. Seyller, Towards wafer-size graphene layers by atmospheric pressure graphitization of silicon carbide, *Nat. Mater.* **8**, 203 (2009).
- [22] M. Kruskopf, D. M. Pakdehi, K. Pierz, S. Wundrack, R. Stosch, T. Dziomba, M. Götz, J. Baringhaus, J. Aprojanz, C. Tegenkamp, J. Lidzba, T. Seyller, F. Hohls, F. J. Ahlers, and H. W. Schumacher, Comeback of epitaxial graphene for electronics: Large-area growth of bilayer-free graphene on SiC, *2D Mater.* **3**, 041002 (2016).
- [23] Y. Yin, M. Kruskopf, S. Bauer, T. Tschirner, K. Pierz, F. Hohls, R. J. Haug, and H. W. Schumacher, Quantum Hall resistance standards based on epitaxial graphene with *p*-type conductivity, *Appl. Phys. Lett.* **125**, 064001 (2024).
- [24] C. Riedl, C. Coletti, T. Iwasaki, A. A. Zakharov, and U. Starke, Quasi-free-standing epitaxial graphene on SiC obtained by hydrogen intercalation, *Phys. Rev. Lett.* **103**, 246804 (2009).
- [25] K. V. Emtsev, A. A. Zakharov, C. Coletti, S. Forti, and U. Starke, Ambipolar doping in quasifree epitaxial graphene on SiC(0001) controlled by Ge intercalation, *Phys. Rev. B* **84**, 125423 (2011).
- [26] I. Gierz, T. Suzuki, R. T. Weitz, D. S. Lee, B. Krauss, C. Riedl, U. Starke, H. Höchst, J. H. Smet, C. R. Ast, and K. Kern, Electronic decoupling of an epitaxial graphene monolayer by gold intercalation, *Phys. Rev. B* **81**, 235408 (2010).

- [27] S. Wolff, N. Tilgner, F. Speck, P. Schädlich, F. Göhler, and T. Seyller, Quasi-freestanding graphene via sulfur intercalation: Evidence for a transition state, *Adv. Mater. Interfaces* **11**, 2300725 (2024).
- [28] P. Schädlich, C. Ghosal, M. Stettner, B. Matta, S. Wolff, F. Schölzel, P. Richter, M. Hutter, A. Haags, S. Wenzel, Z. Mamiyev, J. Koch, S. Soubatch, P. Rosenzweig, C. Polley, F. S. Tautz, C. Kumpf, K. Küster, U. Starke, T. Seyller, F. C. Bocquet, and C. Tegenkamp, Domain boundary formation within an intercalated Pb monolayer featuring charge-neutral epitaxial graphene, *Adv. Mater. Interfaces* **10**, 2300471 (2023).
- [29] S. Link, S. Forti, A. Stöhr, K. Küster, M. Rösner, D. Hirschmeier, C. Chen, J. Avila, M. C. Asensio, A. A. Zakharov, T. O. Wehling, A. I. Lichtenstein, M. I. Katsnelson, and U. Starke, Introducing strong correlation effects into graphene by gadolinium intercalation, *Phys. Rev. B* **100**, 121407(R) (2019).
- [30] P. Rosenzweig, H. Karakachian, S. Link, K. Küster, and U. Starke, Tuning the doping level of graphene in the vicinity of the van Hove singularity via ytterbium intercalation, *Phys. Rev. B* **100**, 035445 (2019).
- [31] S. A. Herrera, G. Parra-Martinez, P. Rosenzweig, B. Matta, C. M. Polley, K. Küster, U. Starke, F. Guinea, J. A. Silva-Guillén, G. G. Naumis, and P. A. Pantaleón, Topological superconductivity in heavily doped single-layer graphene, *ACS Nano* **18**, 34842 (2024).
- [32] S. Schuwalow, D. Grieger, and F. Lechermann, Realistic modeling of the electronic structure and the effect of correlations for Sn/Si(111) and Sn/Ge(111) surfaces, *Phys. Rev. B* **82**, 035116 (2010).
- [33] P. Hansmann, T. Ayrar, L. Vaugier, P. Werner, and S. Biermann, Long-range coulomb interactions in surface systems: A first-principles description within self-consistently combined GW and dynamical mean-field theory, *Phys. Rev. Lett.* **110**, 166401 (2013).
- [34] M. Jäger, C. Brand, A. P. Weber, M. Fanciulli, J. H. Dil, H. Pfnür, and C. Tegenkamp, α -Sn phase on Si(111): Spin texture of a two-dimensional Mott state, *Phys. Rev. B* **98**, 165422 (2018).
- [35] T. Nakamura, H. Kim, S. Ichinokura, A. Takayama, A. V. Zotov, A. A. Saranin, Y. Hasegawa, and S. Hasegawa, Unconventional superconductivity in the single-atom-layer alloy Si(111)- $\sqrt{3} \times \sqrt{3}$ -(Ti, Pb), *Phys. Rev. B* **98**, 134505 (2018).
- [36] X. Cao, T. Ayrar, Z. Zhong, O. Parcollet, D. Manske, and P. Hansmann, Chiral d -wave superconductivity in a triangular surface lattice mediated by long-range interaction, *Phys. Rev. B* **97**, 155145 (2018).
- [37] X. Wu, F. Ming, T. S. Smith, G. Liu, F. Ye, K. Wang, S. Johnston, and H. H. Weitering, Superconductivity in a hole-doped Mott-insulating triangular adatom layer on a silicon surface, *Phys. Rev. Lett.* **125**, 117001 (2020).
- [38] F. Ming, X. Wu, C. Chen, K. D. Wang, P. Mai, T. A. Maier, J. Stroock, J. W. F. Venderbos, C. González, J. Ortega, S. Johnston, and H. H. Weitering, Evidence for chiral superconductivity on a silicon surface, *Nat. Phys.* **19**, 500 (2023).
- [39] S. Glass, G. Li, F. Adler, J. Aulbach, A. Fleszar, R. Thomale, W. Hanke, R. Claessen, and J. Schäfer, Triangular spin-orbit-coupled lattice with strong Coulomb correlations: Sn atoms on a SiC(0001) substrate, *Phys. Rev. Lett.* **114**, 247602 (2015).
- [40] Z. Mamiyev and C. Tegenkamp, Sn intercalation into the BL/SiC (0001) interface: A detailed SPA-LEED investigation, *Surf. Interfaces* **34**, 102304 (2022).
- [41] Z. Mamiyev and C. Tegenkamp, Exploring graphene-substrate interactions: plasmonic excitation in Sn-intercalated epitaxial graphene, *2D Mater.* **11**, 025013 (2024).
- [42] U. Scheithauer, G. Meyer, and M. Henzler, A new LEED instrument for quantitative spot profile analysis, *Surf. Sci.* **178**, 441 (1986).
- [43] Z. Mamiyev, C. Fink, K. Holtgrewe, H. Pfnür, and S. Sanna, Enforced long-range order in 1D wires by coupling to higher dimensions, *Phys. Rev. Lett.* **126**, 106101 (2021).
- [44] G. Kresse and J. Hafner, *Ab initio* molecular dynamics for liquid metals, *Phys. Rev. B* **47**, 558 (1993).
- [45] G. Kresse and J. Furthmüller, Efficiency of *ab-initio* total energy calculations for metals and semiconductors using a plane-wave basis set, *Comput. Mater. Sci.* **6**, 15 (1996).
- [46] G. Kresse and J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [47] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [48] P. E. Blöchl, Projector augmented-wave method, *Phys. Rev. B* **50**, 17953 (1994).
- [49] G. Kresse and D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Phys. Rev. B* **59**, 1758 (1999).
- [50] S. Grimme, J. Antony, S. Ehrlich, and H. Krieg, A consistent and accurate *ab initio* parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu, *J. Chem. Phys.* **132**, 154104 (2010).
- [51] The DFT calculations are available on the NOMAD repository, doi:10.17172/NOMAD/2025.02.24-1.
- [52] C. Riedl, C. Coletti, and U. Starke, Structural and electronic properties of epitaxial graphene on SiC(0001): A review of growth, characterization, transfer doping and hydrogen intercalation, *J. Phys. D* **43**, 374009 (2010).
- [53] A. Mattausch and O. Pankratov, *Ab initio* study of graphene on SiC, *Phys. Rev. Lett.* **99**, 076802 (2007).
- [54] T. O. Wehling, E. Şaşıoğlu, C. Friedrich, A. I. Lichtenstein, M. I. Katsnelson, and S. Blügel, Strength of effective Coulomb interactions in graphene and graphite, *Phys. Rev. Lett.* **106**, 236805 (2011).
- [55] G. Li, P. Höpfner, J. Schäfer, C. Blumenstein, S. Meyer, A. Bostwick, E. Rotenberg, R. Claessen, and W. Hanke, Magnetic order in a frustrated two-dimensional atom lattice at a semiconductor surface, *Nat. Commun.* **4**, 1620 (2013).
- [56] G. Trambly de Laissardière, D. Mayou, and L. Magaud, Localization of Dirac electrons in rotated graphene bilayers, *Nano Lett.* **10**, 804 (2010).
- [57] P. Moon and M. Koshino, Optical absorption in twisted bilayer graphene, *Phys. Rev. B* **87**, 205404 (2013).
- [58] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions, *Rev. Mod. Phys.* **68**, 13 (1996).
- [59] E. Gull, A. J. Millis, A. I. Lichtenstein, A. N. Rubtsov, M. Troyer, and P. Werner, Continuous-time Monte Carlo methods for quantum impurity models, *Rev. Mod. Phys.* **83**, 349 (2011).

- [60] C. Melnick, P. Sémon, K. Yu, N. D’Imperio, A.-M. Tremblay, and G. Kotliar, Accelerated impurity solver for DMFT and its diagrammatic extensions, *Comput. Phys. Commun.* **267**, 108075 (2021).
- [61] M. Jarrell and J. Gubernatis, Bayesian inference and the analytic continuation of imaginary-time quantum Monte Carlo data, *Phys. Rep.* **269**, 133 (1996).
- [62] D. Bergeron and A.-M. S. Tremblay, Algorithms for optimized maximum entropy and diagnostic tools for analytic continuation, *Phys. Rev. E* **94**, 023303 (2016).
- [63] T. Seyller, A. Bostwick, K. Emtsev, K. Horn, L. Ley, J. McChesney, T. Ohta, J. D. Riley, E. Rotenberg, and F. Speck, Epitaxial graphene: A new material, *Phys. Status Solidi B* **245**, 1436 (2008).
- [64] M. Petrović, F. J. M. zu Heringdorf, M. H. von Hoegen, P. A. Thiel, and M. C. Tringides, Broad background in electron diffraction of 2D materials as a signature of their superior quality, *Nanotechnology* **32**, 505706 (2021).
- [65] M.-E. Federl, N. Witt, B. Yang, N. Hofmann, J. Gradl, L. Weigl, I. Piquero-Zulaica, J. V. Barth, N. Mishra, C. Coletti, T. O. Wehling, and I. Gierz, Non-equilibrium carrier dynamics and band structure of graphene on 2D tin *Phys. Rev. B* **111**, 125158 (2025).
- [66] M. Horn-von Hoegen, Growth of semiconductor layers studied by spot profile analysing low energy electron diffraction, *Z. Kristallogr.* **214**, 684 (1999).
- [67] Z. Mamiyev, N. Balayeva, C. Ghosal, D. R. T. Zahn, and C. Tegenkamp, Confinement induced strain effects in epitaxial graphene, *Carbon* **234**, 120002 (2025).
- [68] C. Schmitt, J. Erhardt, P. Eck, M. Schmitt, K. Lee, P. Keßler, T. Wagner, M. Spring, B. Liu, S. Enzner, M. Kamp, V. Jovic, C. Jozwiak, A. Bostwick, E. Rotenberg, T. Kim, C. Cacho, T.-L. Lee, G. Sangiovanni, S. Moser, and R. Claessen, Achieving environmental stability in an atomically thin quantum spin Hall insulator via graphene intercalation, *Nat. Commun.* **15**, 1486 (2024).
- [69] N. Witt, S. Ryee, L. Klebl, J. Cano, G. Sangiovanni, and T. O. Wehling, Quantum geometry and local moment swapover in correlated graphene heterostructures, [arXiv:2503.03700](https://arxiv.org/abs/2503.03700).
- [70] Y. Zhang, V. W. Brar, F. Wang, C. Girit, Y. Yayon, M. Panlasigui, A. Zettl, and M. F. Crommie, Giant phonon-induced conductance in scanning tunnelling spectroscopy of gate-tunable graphene, *Nat. Phys.* **4**, 627 (2008).
- [71] G. R. Stewart, Heavy-fermion systems, *Rev. Mod. Phys.* **56**, 755 (1984).
- [72] J. G. Checkelsky, B. A. Bernevig, P. Coleman, Q. Si, and S. Paschen, Flat bands, strange metals and the Kondo effect, *Nat. Rev. Mater.* **9**, 509 (2024).
- [73] Z.-D. Song and B. A. Bernevig, Magic-angle twisted bilayer graphene as a topological heavy fermion problem, *Phys. Rev. Lett.* **129**, 047601 (2022).
- [74] G. Rai, L. Crippa, D. Călugăru, H. Hu, F. Paoletti, L. de’ Medici, A. Georges, B. A. Bernevig, R. Valentí, G. Sangiovanni, and T. Wehling, Dynamical correlations and order in magic-angle twisted bilayer graphene, *Phys. Rev. X* **14**, 031045 (2024).
- [75] T. O. Wehling, A. V. Balatsky, M. I. Katsnelson, A. I. Lichtenstein, and A. Rosch, Orbitally controlled Kondo effect of Co adatoms on graphene, *Phys. Rev. B* **81**, 115427 (2010).
- [76] F. C. Zhang and T. M. Rice, Effective Hamiltonian for the superconducting Cu oxides, *Phys. Rev. B* **37**, 3759 (1988).
- [77] C. Weber, K. Haule, and G. Kotliar, Strength of correlations in electron- and hole-doped cuprates, *Nat. Phys.* **6**, 574 (2010).
- [78] P. Werner, R. Sakuma, F. Nilsson, and F. Aryasetiawan, Dynamical screening in La_2CuO_4 , *Phys. Rev. B* **91**, 125142 (2015).
- [79] S. Schumacher, T. O. Wehling, P. Lazić, S. Runte, D. F. Förster, C. Busse, M. Petrović, M. Kralj, S. Blügel, N. Atodiresei, V. Caciuc, and T. Michely, The backside of graphene: Manipulating adsorption by intercalation, *Nano Lett.* **13**, 5013 (2013).
- [80] M. Imada, A. Fujimori, and Y. Tokura, Metal-insulator transitions, *Rev. Mod. Phys.* **70**, 1039 (1998).