Visualizing delocalized correlated electronic states in twisted double bilayer graphene

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The discovery of interaction-driven insulating and superconducting phases in moiré van der Waals heterostructures has sparked considerable interest in understanding the novel correlated physics of these systems. While a significant number of studies have focused on twisted bilayer graphene, correlated insulating states and a superconductivity-like transition up to 12 K have been reported in recent transport measurements of twisted double bilayer graphene. Here we present a scanning tunneling microscopy and spectroscopy study of gate-tunable twisted double bilayer graphene devices. We observe splitting of the van Hove singularity peak by ~20 meV at half-filling of the conduction flat band, with a corresponding reduction of the local density of states at the Fermi level. By mapping the tunneling differential conductance we show that this correlated system exhibits energetically split states that are spatially delocalized throughout the different regions in the moiré unit cell, inconsistent with order originating solely from onsite Coulomb repulsion within strongly-localized orbitals. We have performed self-consistent Hartree-Fock calculations that suggest exchange-driven spontaneous symmetry breaking in the degenerate conduction flat band is the origin of the observed correlated state. Our results provide new insight into the nature of electron-electron interactions in twisted double bilayer graphene and related moiré systems.

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the stacking of atomically-thin van der Waals (vdW) materials provides an elegant platform for studying correlated electronic states. Moiré superlattices formed by lattice misalignment between adjacent vdW sheets can create narrow mini-bands with width comparable to or even smaller than the Coulomb interaction energy, leading to the emergence of various correlated phases. Experimental signatures of interaction-driven electronic states in moiré vdW stacks were first observed in magic-angle twisted bilayer graphene (tBLG), where the coexistence of insulating and superconducting phases resembles the phase diagram of high temperature cuprate superconductors. More exotic phases, such as orbital ferromagnets and correlated Chern insulators, were later reported in other moiré vdW systems. Transport measurements on twisted double bilayer graphene (tDBLG), which consists of two sheets of Bernal-stacked bilayer graphene with a small rotational misalignment, have shown new correlated features such as enhancement of a correlation gap under external magnetic field. When the CFB is half-filled we observe an interaction-driven reduction of LDOS at the Fermi level, consistent with an emergent correlated insulating phase interaction-driven reduction of LDOS at the Fermi level, corresponding to a local twist angle of 0.18°, but the moiré pattern is not perfectly three-fold symmetric due to external strain. Variation of the moiré wavelength in different directions allows us to estimate a strain of ~0.2% in this sample. As seen in Fig. 1c we observe three regions that have different apparent heights within each moiré unit cell. We identify these as the three different possible stacking regions of tDBLG: ABBC, ABCA, and ABAB (sketched in Fig. 1d; see Supplementary Note 1).

We characterized the electronic structure of our tDBLG samples by performing dI/dV spectroscopy. Before each set of measurements our STM tips were calibrated against the Cu(111) Shockley surface state to ensure that they were free of artifacts known to arise from loosely bound adsorbates and clusters. dI/dV spectra for tDBLG devices measured over large bias ranges using calibrated tips always exhibited a strong enhancement of the tunneling signal for |V_{bias}| > 60 mV (Supplementary Fig. 1), a familiar graphene effect known to arise due to phonon-mediated inelastic tunneling. The signal from the elastic tunneling channel is typically weak compared to the inelastic signal. Here we focus primarily on the elastic signal within the bias range ~60 mV ≤ V_{bias} ≤ 60 mV, both because correlation effects are expected to be strongest near V_{bias} = 0 mV (the Fermi level) and to avoid inelastic broadening effects.

**Results**

Our tDBLG samples were fabricated using a tear-and-stack technique with a hexagonal boron nitride (hBN) substrate and deposited onto a SiO2/Si wafer (Methods). Figure 1a shows a sketch of the device scheme and Fig. 1b shows an optical microscope picture of a typical sample. The presence of hBN and SiO2 dielectric layers allows us to apply a voltage V_G to the Si back-gate to change the carrier density n and the vertical electric field E in the tDBLG stack. The tDBLG samples were annealed in ultra-high vacuum before being loaded into the STM system at T = 4.7 K for measurement (Methods section). Figure 1c shows a representative STM topographic image (the inset shows a zoom-in image with graphene lattice). The moiré wavelength of ~13 nm corresponds to a local twist angle of θ = 0.18°, but the moiré pattern is not perfectly three-fold symmetric due to external strain. Variation of the moiré wavelength in different directions allows us to estimate a strain of ~0.2% in this sample. As seen in Fig. 1c we observe three regions that have different apparent heights within each moiré unit cell. We identify these as the three different possible stacking regions of tDBLG: ABBC, ABCA, and ABAB (sketched in Fig. 1d; see Supplementary Note 1).

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respectively. Increasing the gate voltage causes the sample to become more electron-doped and induces the CFB peak to slowly shift downward in energy. The VFB peak, on the other hand, more rapidly moves toward lower energy and away from the CFB for higher gate voltage. For \( V_G \geq 45 \text{ V} \) the VFB signal shifts out of our measurement bias range \( |V_{\text{bias}}| \leq 60 \text{ mV} \) and only the CFB peak can be observed.

The energetic separation between the CFB and VFB peaks at finite gate voltages allows us to measure the spatial profile of each flat band feature individually. Figure 3a, for example, compares point spectra obtained for the CFB state in the ABBC (blue), ABCA (purple), and ABAB (orange) stacking regions at a gate voltage of \( V_G = 60 \text{ V} \). The CFB peak at \( V_{\text{bias}} = -20 \text{ mV} \) appears in all three regions with nearly equal intensity. In the ABBC region another peak appears at a higher energy of \( V_{\text{bias}} = 6 \text{ mV} \) that has a larger width than the CFB peak. We refer to this feature as the “remote conduction band” (RCB). To better visualize the spatial distributions of the CFB and RCB wavefunctions, we obtained \( \text{d}/\text{dV} \) spectroscopy grids over an area containing several moiré unit cells. Figure 3b, c show \( \text{d}/\text{dV} \) maps obtained at the CFB peak energy (\( V_{\text{bias}} = -20 \text{ mV} \)) and the RCB peak energy (\( V_{\text{bias}} = 6 \text{ mV} \)) plotted using the same color scale. We observe that the CFB signal is highly delocalized with only slight amplitude variation within the moiré unit cell (Fig. 3b). The RCB signal, on the other hand, is more strongly modulated and has its highest amplitude in the ABBC region (Fig. 3c). This is further demonstrated by histograms of the two maps plotted in Fig. 3d, e. The delocalized CFB has shallow corrugation, leading to a narrower \( \text{d}/\text{dV} \) intensity histogram with a smaller maximum/minimum ratio (Fig. 3d) compared to the more corrugated (i.e., localized) RCB state which has a broader \( \text{d}/\text{dV} \) intensity histogram with more weight in the intensity troughs (Fig. 3e). Point spectroscopy and \( \text{d}/\text{dV} \) mapping performed at \( V_G = -60 \text{ V} \) allows the VFB peak to be analyzed in the same way since this gate voltage places it in our “elastic window”. The VFB state is observed to be similarly delocalized compared to a “remote valence band” (RVB) peak that is localized in the ABBC region analogous to the RCB peak (Supplementary Fig. 2).

To further understand the electronic structure of tDBLG, we methodically tuned the doping level of the system by varying the gate voltage and traced the evolution of \( \text{d}/\text{dV} \) spectroscopic features. Figure 4d–f shows density plots of \( \text{d}/\text{dV} \) spectra measured as a function of gate voltage over the range \(-60 \text{ V} < V_G < 60 \text{ V} \) for all three stacking regions (ABBC, ABCA, and ABAB). The relation between the gate voltage \( V_G \) and the filling factor \( \nu \) (the average number of electrons or holes per moiré unit cell) is found to be \( \Delta \nu / \Delta V_G = 0.09 \text{ V}^{-1} \) based on the calculated capacitance of the dielectric layers (Methods). The separation of CFB and VFB peaks is clearly observed in the ABBC region for \( 5 \text{ V} < V_G < 35 \text{ V} \) (Fig. 4f). At \( V_G = 45 \text{ V} \) (\( \nu = 4 \)) the CFB state is fully filled and the RCB peak appears in the \( \text{d}/\text{dV} \) spectra, as shown in Fig. 4a–c. Further increasing the gate voltage quickly alters the chemical potential and causes the CFB peak to rapidly drop from the Fermi level as the RCB state begins to be populated with electrons. A similar rapid change occurs at \( V_G = -45 \text{ V} \) (\( \nu = -4 \)) as the VFB state becomes fully depleted and the RVB state begins to be populated by holes at lower gate voltages (Fig. 4g–i).

Probably the most significant feature in Fig. 4d–f, however, is that the CFB peak splits into two branches labeled CFB+ and CFB− (with a corresponding dip at the Fermi level) for \( V_G = 22.5 \text{ V} \) (corresponding to \( \nu \approx 2 \) and \( E = 0.12 \text{ V/\text{nm}} \)), as highlighted by the dashed black boxes. We observed consistent CFB peak splitting at \( \nu = 2 \) in several devices, with the local twist angle \( \theta \) ranging from 1.05° to 1.17° (see Supplementary Note 2 and Supplementary Fig. 3). The dependence of the splitting on \( \nu \) and \( E \) confirms that it corresponds to the \( \nu = 2 \) correlated phase reported in transport measurements16–20 (see Supplementary Note 3 and Supplementary Fig. 4). The evolution of the CFB with
filling factor can be seen even better in Fig. 5a which shows dI/dV spectra obtained in the ABBC region over the range 1 < ν < 3 (11.5 V < V_G < 33.5 V). A single peak is seen for filling factors away from ν = 2 (e.g. for V_G < 15 V and V_G > 30 V), but a dip feature is clearly resolved at the Fermi level over the range 1.65 ≤ ν ≤ 2.35 (19 V ≤ V_G ≤ 26 V). By fitting the dI/dV signal with the sum of two Lorentzians we are able to extract the magnitude of the energy-splitting δ as a function of filling factor in the different regions of the moiré unit cell (Fig. 5b; see Supplementary Note 4 and Supplementary Fig. 5 for details). The splitting in the ABBC region reaches its maximum value of δ_max = 18.9 ± 1.2 meV at ν = 2 and then decreases to 13 ± 1 meV at ν ≈ 1.7 and ν = 2.3 (Fig. 5b blue dots). Beyond this doping range a smaller splitting may still occur, but cannot be determined due to the width of the CFB peak. dI/dV spectra measured in the ABCA and ABAB regions display similar trends with δ_max(ABCA) = 20.0 ± 1.2 meV and δ_max(ABAB) = 20.4 ± 1.3 meV (Fig. 5b purple and orange dots; also see Supplementary Fig. 6). The spatial dependence of the splitting is further illustrated by Fig. 5c which shows dI/dV spectra measured at ν = 2 along a line cut through the entire moiré unit cell. The CFB+ and CFB− peaks and the dip feature at the Fermi level persist throughout the entire moiré unit cell and do not appear to depend strongly on local stacking order.

Our experimental observations can be understood through comparison to a continuum theoretical model of tDBLG1,27,31 that includes an added interlayer potential difference and a manually adjusted chemical potential to account for gate-induced variation in E-field and carrier density (Methods section). Figure 2e shows the band structure calculated at E = 0 (corresponding to V_G = 0 V) along the high symmetry directions of the moiré Brillouin zone. Two narrow and partially overlapping mini-bands can be seen near the Fermi level (green and pink curves) that are isolated in energy from other bands. Their van Hove singularities result in the CFB and VFB peaks in the calculated LDOS curves of Fig. 2b. For higher values of E-field and n-type doping (corresponding to a positively increasing gate voltage) the conduction and valence flat bands both move downward relative to the Fermi level and their energy separation becomes larger (Fig. 2c, d). The corresponding gate-dependent theoretical LDOS curves in Fig. 2b reasonably reproduce the experimental spectra of Fig. 2a, thus establishing the gate-induced E-field as the origin of the observed separation of the CFB and VFB peaks.

The spatial delocalization/localization behavior of the CFB/RCB states is also nicely reproduced by the theoretical LDOS calculated at an E-field and chemical potential corresponding to the experimental gate voltage V_G = 60 V (Fig. 3f–j). Since STS is most sensitive to the electronic states at the sample surface, we plot the calculated LDOS only at the topmost graphene layer to best compare with our measurements (the corresponding behavior when the LDOS interior to the layers is accounted for is discussed in Supplementary Note 5). The spectral density of the CFB peak at ℰ = −13 meV and the RCB peak at ℰ = 2.8 meV in the theoretical LDOS of Fig. 3f correspond well to their...
of CFB and VFB features for the delocalization/localization behavior exhibited by the VFB/RVB electronic features, as shown in Supplementary Fig. 2. This limitation is evident when we compare the experimental LDOS values seen both in theory (Fig. 3b) and experiment (Fig. 3g), however, is seen in the theoretical LDOS map at the RCB peak energy (Fig. 3h) also agrees with the corresponding experimental density plot for three stacking regions (Fig. 3i) and experiment (Fig. 3d). Some discrepancy between results of Lee et al.27, under these conditions the system can lower its total energy through spontaneous symmetry breaking around \( \nu = 2 \), with the precise nature of the broken symmetry depending on the detailed band structure and the screening parameters \( \epsilon_{\text{eff}} \) and \( d_0 \). When screening is weak and the electron–electron interaction strength is much greater than the flat band width then the ground state of the system favors isospin polarization (ISP) (it can be either spin-polarized, valley-polarized, or spin-valley-locked, all of which are degenerate in energy within our model). On the other hand, when electron-electron interactions are weak then the development of inter-valley coherence (IVC) is favored.

The different correlated ISP and IVC scenarios for tDBLG lead to very similar results for basic STM observables such as wave-function maps and energy-dependent LDOS spectra. Nevertheless, we find some quantitative indications for ISP behavior over IVC behavior due to the size of the energy-splitting that we observe experimentally at \( \nu = 2 \), and so we focus on the tDBLG ISP solution in what follows (ISP versus IVC results are directly consistent Hartree-Fock approximation (Methods section)32. We assume that the Coulomb interaction, which is screened by the graphene, the hBN/SiO\(_2\) substrate, and the metallic STM tip, takes the single-plane-screened form \( V(q) = \frac{e^2}{\pi \epsilon_0 \epsilon_{\text{eff}}}[1 - \exp(-2qd_0)] \) where \( \epsilon_{\text{eff}} \) is the effective dielectric constant and \( d_0 \) is the effective macro tip-sample separation (these are treated as fitting parameters, see Supplementary Note 7). Consistent with the theoretical results of Lee et al.27, under these conditions the system can lower its total energy through spontaneous symmetry breaking around \( \nu = 2 \), with the precise nature of the broken symmetry depending on the detailed band structure and the screening parameters \( \epsilon_{\text{eff}} \) and \( d_0 \). When screening is weak and the electron–electron interaction strength is much greater than the flat band width then the ground state of the system favors isospin polarization (ISP) (it can be either spin-polarized, valley-polarized, or spin-valley-locked, all of which are degenerate in energy within our model). On the other hand, when electron-electron interactions are weak then the development of inter-valley coherence (IVC) is favored.

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![Fig. 4 Gate-dependent \( \text{d}/\text{dV} \) spectroscopy for three different stacking regions. a–c \( \text{d}/\text{dV} \) spectra for three stacking regions for \( V_G = 45 \) V (\( \nu = 4 \)). d–f Gate-dependent \( \text{d}/\text{dV} \) density plot for three stacking regions over gate-voltage range \(-60 < V_G < 60 \) V. The vertical black dashed line denotes zero bias (the Fermi level). The black dashed box highlights correlation-driven splitting of the CFB peak near gate, \( V_G = -45 \) V (\( \nu = -4 \)). Spectroscopy parameters: modulation voltage \( V_{\text{mod}} = 1 \text{ mV} \); initial \( V_{\text{bias}} = -100 \text{ mV} \), \( I_0 = 0.5 \text{ nA} \). CFB conduction flat band, VFB valence flat band, RCB remote conduction band, RVB remote valence band.]

![Image of Gate-dependent \( \text{d}/\text{dV} \) spectroscopy for three different stacking regions.](https://example.com/gate-dependent.png)
compared in Supplementary Note 8). Figure 5d, e shows the ISP Hartree-Fock band structure of tDBLG for $ν = 2$ for the best fit parameters $ε_{\text{eff}} = 14$ and $d_e = 50$ nm. For these parameters the two isospin sub-bands of the CFB are split by an average of 19 meV across the moiré Brillouin zone, in good agreement with our experimental results (Fig. 5a, b). For other bands outside of the CFB and away from the chemical potential the isospin-splitting becomes less significant ($< 5$ meV) and the single-particle band structure is approximately preserved. To better compare the Hartree-Fock calculations with our STS measurements we plot the theoretical isospin-splitting averaged over the mini-Brillouin zone as a function of the filling factor for $1.5 < ν < 2.5$. Calculated Hartree-Fock DOS for $ν = 2$ at different positions across the moiré unit cell corresponding to white dashed line in c. CFB conduction flat band, VFB valence flat band.

The delocalized spatial distribution of the CFB wavefunctions and the persistent LDOS reduction at $ν = 2$ across the entire moiré unit cell point to a spatially extended correlated state in tDBLG. This result has significant implications for our understanding of electronic correlations in tDBLG and related moiré systems. Metal-insulator transitions in strongly interacting systems often arise from Coulomb repulsion between localized electrons, as exemplified by the single-orbital Hubbard model, and the resulting Mott insulators are usually anti-ferromagnetic due to super-exchange. In the case of tBLG, the localization of low-energy electronic states in the AA stacking region is a promising candidate for the observed correlated state in tDBLG.
specific ground state is likely determined by small effects not accounted for in our model, such as defects and inter-valley Coulomb scattering (see Supplementary Note 8). Transport studies have also resulted in some ambiguity regarding this point, since evidence has been provided that supports both spin-polarized\(^{17-19}\) and valley-polarized\(^{16}\) ground states. Nevertheless, in all of these theoretical scenarios the correlated state is the result of non-local exchange interactions and the electron orbitals are spatially delocalized. Future STM studies involving spin-polarized STM, edge-state detection, and/or quasiparticle interference could potentially provide definitive evidence regarding the symmetry of the ground state and would have significant implications for the pairing channel of any proximate superconductivity\(^{27}\).

In conclusion, combined STM/STS measurements reveal a correlated electronic state in tDBLG that is induced by electron–electron interactions when the conduction flat band is separated from the valence flat band and is half-filled. In contrast to tBLG, the flat band wavefunctions in tDBLG are delocalized in space and correlation-driven Fermi-level LDOS reduction is observed everywhere inside the moiré unit cell. A Hartree-Fock analysis of the interacting continuum, Hamilomon shows good agreement with our experimental results and highlights the importance of non-local exchange interactions.

**Methods**

**Sample preparation.** Samples were prepared using the “flip-chip” method\(^{10}\) followed by a forming-gas anneal\(^{41,42}\). Electrical contacts were made by evaporating Cr (5 nm)/Au (50 nm) through a silicon nitride shadow-mask onto the heterostructure. The resulting continuum model is modeled using a four-band tight-binding model with Hartree-Fock Hamiltonian shown in Table S9 and Supplementary Fig. 13.

**Estimation of carrier density and electric field.** The relation between the gate voltage \(V_G\), the carrier density \(n\), and the vertical electric field \(E\) was estimated by modeling the back-gate configuration in Fig. 1a as a parallel plate capacitor. Therefore

\[
\begin{align*}
\rho & = \frac{\varepsilon_1 \varepsilon_0 V_G}{d_0} \\
E & = \frac{\varepsilon_1 V_G}{2d_0}
\end{align*}
\]

where \(d_0 = 310 \text{ nm}\) is the thickness of the dielectric layers (hBN and SiO\(_2\)), \(\varepsilon_0\) is the vacuum permittivity, \(\varepsilon_1 = 3.5\) is the average perpendicular dielectric constant, and \(\varepsilon\) is the elementary charge. The filling factor \(\nu\) and the carrier density \(n\) are further related by

\[
\nu = \frac{\sqrt{3}}{2} n_{BLG}
\]

where \(n_{BLG}\) is the moiré wavelength. Tip-induced gating effects were not observed in our spectroscopic measurements and thus are not included in the above estimation (see Supplementary Note 9 and Supplementary Fig. 13).

**Continuum model and single-particle calculations.** Our band-structure calculations follow those of Lee et al.\(^{27}\), who are based on the Bistritzer-MacDonald continuum approach to moiré structures\(^{1}\). The Bernal-stacked bilayer graphene is modeled using a four-band tight-binding model with \(t_0 = -2.61, t_1 = 0.361, t_2 = 0.283, t_3 = 0.138, \beta = 0.015 \text{ eV}\). One bilayer is then rotated by angle \(\theta\) and the two bilayers hybridize only through their proximate layers, with intra-sublattice strength \(w_0 = 0.075 \text{ eV}\) and inter-sublattice strength \(w_1 = 0.1 \text{ eV}\) in the notation of Lee et al.\(^{27}\). We note that in our convention \(\theta = 0\) corresponds to the structure obtained when a single sheet of BLG is torn in half, one half translated without rotation, and the two halves stacked with ABBC-alignment. The resulting continuum model is truncated by keeping all states within a radius of 5 mini-Brillouin zones (BZs) of the mini-BZ F-point. The effect of the gate-induced vertical \(E\)-field is modeled by a constant energy difference \(U/3\) between two adjacent layers. To relate \(U\) to the applied gate voltage \(V_G\) and the physical \(E\)-field estimated above, we assume an interlayer spacing of \(d = 0.33 \text{ nm}\) and model the tDBLG as a uniform dielectric with \(\varepsilon_1 = 6.5\), giving \(U/3E = 0.15 \text{ nm}\) similar to that reported in He et al.\(^{20}\).

**Hartree-Fock calculations.** Hartree-Fock calculations are done in a k-space analogous to earlier Hartree-Fock studies of tBLG\(^{12,18,39}\) and tDBLG\(^{37}\). Our code is an extension of the tBLG code used in Ref. 3. The Hamiltonian takes the form

\[
H = H_0 + PH_f P - H_{\text{BLG}}^{\text{eff}}
\]

where \(H_0\) is the continuum band Hamiltonian discussed above, \(H_f\) is the real-space Coulomb interaction \(V(q)\), \(P\) denotes projection into some number of moiré bands near charge neutrality, and \(H_{\text{BLG}}^{\text{eff}}\) is a single-particle correction to be discussed shortly. The Coulomb matrix elements for \(H_f\) are evaluated in the basis of the continuum band structure and projected into the 6 bands nearest to the charge neutrality point per valley and spin, for a total of \(4 \times 6 = 24\) bands. When evaluating the Coulomb integrals, we ignore the interlayer spacing \(d\), which is accurate up to corrections of order \(d/\hbar V_f\). We note that while small, the neglected \(d/\hbar V_f\) terms will lead to interlayer screening which modifies the effective \(U_f\) which may be an interesting direction for future work. Since the BLG tight-binding parameters obtained from DFT already contain the effect of renormalization by the Coulomb interaction on the physics of a single bilayer, we follow refs. 32,38 by substituting off the Hartree-Fock Hamiltonian \(H_{\text{BLG}}\) (exchange energy) of two decoupled BLG layers at charge neutrality.

We then consider a Slater-determinant ansatz which is diagonal in the BZ momentum \(k\),

\[
\begin{align*}
|u_i\rangle & = \prod_{k \in \text{BZ}} \prod_{n} \left( \sum_{c} u_{i,c}^{n}(k) |c\rangle \right) |0\rangle \\
\end{align*}
\]

Here \(c\) creates an electron in eigenstate \((k, n)\) of the band structure, while \(|c\rangle\)'s are the set of variational parameters to be optimized. The total occupation \(n_i\) is allowed to vary across the mbz to account for the presence of Fermi surfaces. In addition to translation invariance, we constrain the \(v\)'s to preserve a spin-symmetry about the \(S\) axis (ruling out non-colinear magnetism, which is not expected in this model), but do not enforce the valley-U(1) symmetry, allowing for spontaneous inter-valley coherence as has been argued to occur in tBLG\(^{24}\). Discretizing the model on a \(20 \times 20\) k-grid, the \(v\)'s are iteratively adjusted to minimize the energy \(\langle u | H | u \rangle\), using the optimal damping algorithm to achieve Hartree-Fock self-consistency. Solving for the self-consistent Hartree-Fock Hamiltonian \(H_{\text{HF}}(\nu, U)\) at each filling \(\nu\) and electric field \(U\), we then reconstruct DOS and LDOS curves by diagonalizing \(H_{\text{HF}}\) and converting back to real space using the continuum-model wavefunctions. To account for finite temperature and instrumental broadening effects and avoid spurious spikes due to the numerical discretization of the mini-BZ, the (LD)OS is broadened by convolving with \(\langle \gamma, \gamma' \rangle = \frac{\pi}{2} \text{sech} \left( \frac{2\pi C}{\gamma} \right) \). For finite \(\gamma = 2\text{ meV}\) for DOS and \(4\text{ meV}\) for LDOS. Unless specified, the LDOS is always projected onto the topmost graphene layer to enable comparison with d/dV spectroscopy.

**Data availability**

All other data that support the plots within this paper and the findings of this study are available from the corresponding authors upon reasonable request. Source data are provided with this paper.

**Code availability**

The computer codes that support the plots within this paper and the findings of this study are available from the corresponding authors upon reasonable request.

Received: 24 November 2020; Accepted: 22 March 2021; Published online: 04 May 2021

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