

Lifshitz transitions and isospin polarization in twist-decoupled monolayer-bilayer graphene

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Bernal-stacked bilayer graphene (BLG) hosts correlated electronic phases tied to low-energy Lifshitz transitions at saddle points in its valence band. To access this regime, ultralow charge disorder and control over a vertical electric field are simultaneously required. Here, we employ a twist-decoupled monolayer (MLG) to bias a proximal BLG in the absence of an external displacement field (D). We thereby reveal threefold degenerate quantum Hall states at $D = 0$, with multiple transitions driven by doping, magnetic and electric field. Spontaneous broken symmetry in the vicinity of the valence band edge is signaled by the emergence of quantum oscillations with anomalous frequencies and large quasiparticle mass. These results indicate that electronic interactions in BLG are preserved in presence of an atomically close MLG, while showcasing the potential of CVD-grown graphene multilayers for the exploration of correlated phases of matter.

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Introduction. Crystalline graphene multilayers have become an increasingly popular platform for the investigation of electronic interactions in two dimensions. Thus far, reported correlated phases include spontaneous gaps [1–3], ferromagnets [4,5], Chern insulators [6], integer [7] and fractional [8,9] quantum anomalous Hall insulators, and superconductors [10], often connected with each other via electrostatic gating [11]. A key advantage of this class of systems is to bypass the issues related to structural disorder of moiré graphene [12], the archetypical correlated two-dimensional (2D) family [13]. However, trilayer and thicker crystalline graphene possess the required flat-band features only in the case of rhombohedral (ABC) stacking, a metastable configuration that needs *ad hoc* strategies to avoid structural relaxation [4,14,15]. Bernal-stacked (AB) bilayer graphene (BLG) samples, conversely, can be handled via conventional van der Waals techniques [16], while displaying exotic properties analogous to their thicker counterparts [17–22]. Nonetheless, the large density

of states in BLG is confined to a narrow energy range (~ 1 meV, approximately a factor of ten smaller than in ABC trilayer [23]), implying that devices with ultralow electrostatic disorder are necessary [24]. In addition, control over a vertical displacement field (D) is needed to enhance saddle-point van Hove singularities (vHs) [25] associated to single-particle gap opening induced by layer polarization [26,27]. A different route to gapping BLG in absence of a D field is offered by large-angle twisting either with another BLG [28] or a graphene monolayer (MLG) [29,30]. This approach takes advantage of electronic decoupling due to momentum mismatch [31–33], combined with proximity-induced energy shifts resulting in layer-polarized BLG subsystems. An energy shift of 18 meV was recently measured in twisted monolayer-bilayer graphene (TMBG) encapsulated in hexagonal boron nitride (hBN) [29], corresponding to the effect of a sizeable external D field (~ 0.15 V/nm, as confirmed by Jiang *et al.* [30]). Therefore, TMBG has the potential to offer D -field-free access to the valence band of biased BLG. In addition, proximal graphene layers result in considerable screening effects [34], which might influence the correlated phase diagram with respect to that of freestanding BLG.

In this Letter, we present low-temperature (magneto)transport experiments on a low-disorder TMBG dual-gated device, in which the built-in asymmetry gaps the BLG subsystem. We observe quantum oscillations associated with a threefold valence band edge at $D = 0$. The evolution of the oscillations under different external fields indicates tunable Lifshitz transitions (LTs) [35] across a saddle-point

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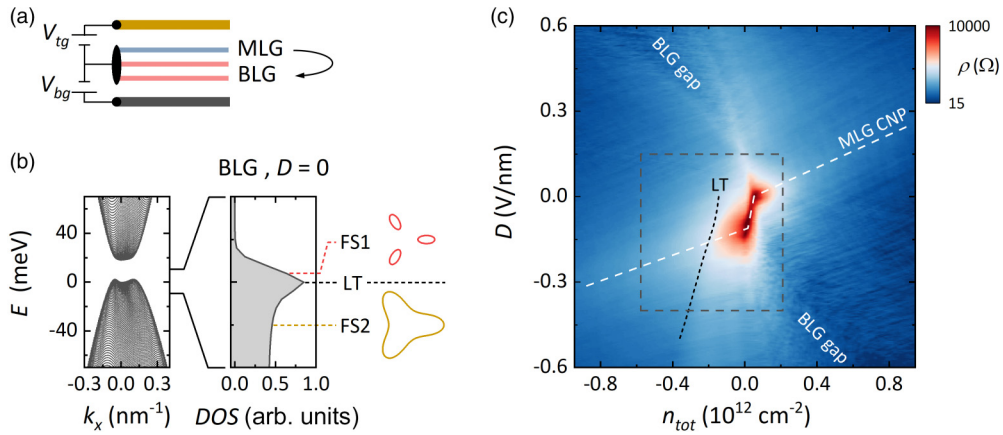


FIG. 1. Accessing the valence band of biased BLG within TMBG. (a) Sketch of the studied device with gating configuration. Yellow: Au gate; dark gray: graphite gate; blue: MLG; red: BLG. (b) Left: low-energy BLG bands within TMBG at $D = 0$ as a function of k_x relative to the K point. Center: density of states in the top part of the BLG valence band ($-10 \text{ meV} < E < +10 \text{ meV}$). Right: evolution of the Fermi surface across the LT. FS1 (red) is shown at $E = 1 \text{ meV}$, FS2 (yellow) is shown at $E = -5 \text{ meV}$. (c) Resistivity as a function of gate-induced charge density and displacement field, measured at $T = 0.4 \text{ K}$ and $B = 0 \text{ T}$. A log color scale is used. The black dashed line is the calculated n-D position of the BLG LT. The white dashed line is a guide to the eye for the MLG CNP. The dashed gray rectangle indicates the n-D ranges adopted in the following parts of the manuscript.

vHs, consistent with the single-particle band structure of biased BLG. We further reveal oscillations beyond the noninteracting picture, signaling broken-symmetry phases stabilized by Coulomb interaction. We discuss these findings in the context of previous studies on freestanding BLG. Our results establish CVD-grown large-angle twisted graphene as a platform for many-body electron physics.

Results and discussion. Figure 1(a) shows the device configuration. An hBN-encapsulated CVD-grown TMBG crystal, composed of a MLG (blue) twisted by 30° on top of BLG (red), is controlled by a top (Au, yellow) and a bottom (graphite, dark gray) gate electrode. The combined action of the gate voltages (V_{bg} and V_{tg}) induces a total charge density $n_{tot} = 1/e \cdot (C_{bg}V_{bg} + C_{tg}V_{tg})$ and a displacement field $D/\epsilon_0 = (C_{bg}V_{bg} - C_{tg}V_{tg})/2$ [31], where C_{bg} and C_{tg} are the capacitances per unit area of the two gates. The sample, fabricated according to the methods reported in Ref. [29] and referred to as device D2 therein, is the TMBG with lowest disorder among those at disposal at the time of performing this experiment. The presence of MLG results in a band gap in the BLG subsystem even at $D = 0$, as shown in the calculations of Fig. 1(b), left (details on the model are reported in Supplemental Material (SM) [36]). Close to the valence band maximum, a saddle point is visible, leading to a vHs in the density of states [DOS, Fig. 1(b), center]. The BLG vHs is associated with a ferromagnetic instability [25], which drives a cascade of isospin (valley and spin) phase transitions in freestanding BLG [20,23,37]. In a single-particle picture [38,39], the vHs is signaled by a characteristic change in the topology of Fermi surface (FS), that is a LT [35]. As shown in Fig. 1(b), right, at larger hole doping, the topology of the Fermi surface evolves from being three disconnected pockets (FS1) into a single hole pocket (FS2), each being fourfold degenerate in spin and valley.

Resistivity (ρ) measurements at low temperature ($T = 0.4 \text{ K}$) and zero magnetic field, presented in Fig. 1(c), give a first indication of whether this scenario and the associated

many-body physics may be detected in our CVD-grown TMBG. Based on previous works [29,30], we identify the feature at highest ρ (dark red color) as the MLG charge neutrality point (CNP) crossing the BLG band gap. Outside of the BLG gap, the MLG CNP acquires a smaller slope due to screening by the BLG DOS, following the trajectory depicted in Fig. 1(c) (white dashed line), and confirmed by measurements of the MLG $N = 0$ Landau level (0-LL) at finite magnetic field (discussed later). The BLG gap collapses at $D \sim 0.15 \text{ V/nm}$ [29,30]. On the left side of the gap (i.e., in the BLG valence band) we observe a region with moderate ρ (light red to white color) that fans out of the gap closing point and expands toward negative displacement field. ρ shows a steep increase upon entering this region (see Fig. S1 [36])—which aligns with the position of the BLG LT obtained from our TMBG model [dashed black line in Fig. 1(c)]—and a series of peaks reminiscent of freestanding BLG under comparable displacement fields [20,37] (see Fig. S2 [36]). Such features, previously unreported in large-angle TMBG, define the n - D region where we focus for the remainder of our study [dashed gray rectangle in Fig. 1(c)].

We now address the response of the device to a finite magnetic field. At $D = 0$, we measure the LL fan shown in Fig. 2(a). The contribution of MLG LLs is restricted to the high-conductivity region (red color) at low magnetic field ($B < 0.3 \text{ T}$), while MLG keeps at $\nu_{MLG} = -2$ throughout the majority of the map. The pinning of ν_{MLG} is evidenced by the straight trajectories of BLG LLs, following $n_{tot} h/eB$. We can thus focus on quantum Hall (QH) features deriving from the BLG valence band, with associated filling factor and Hall conductivity quantified as $\nu_{BLG} = \nu_{tot} - \nu_{MLG}$ and $\sigma_{xy}^{BLG} = \sigma_{xy} - \sigma_{xy}^{MLG}$, respectively. The $\nu_{BLG} = 0$ QH state extends to vanishing B due to the gap of layer-polarized BLG. In the valence band, we observe different QH sequences. At moderate magnetic field [see red dashed line in Fig. 2(a)] we observe threefold degenerate QH states ($\nu_{BLG} = -3, -6$), which evolve into fourfold degenerate ones upon hole doping.

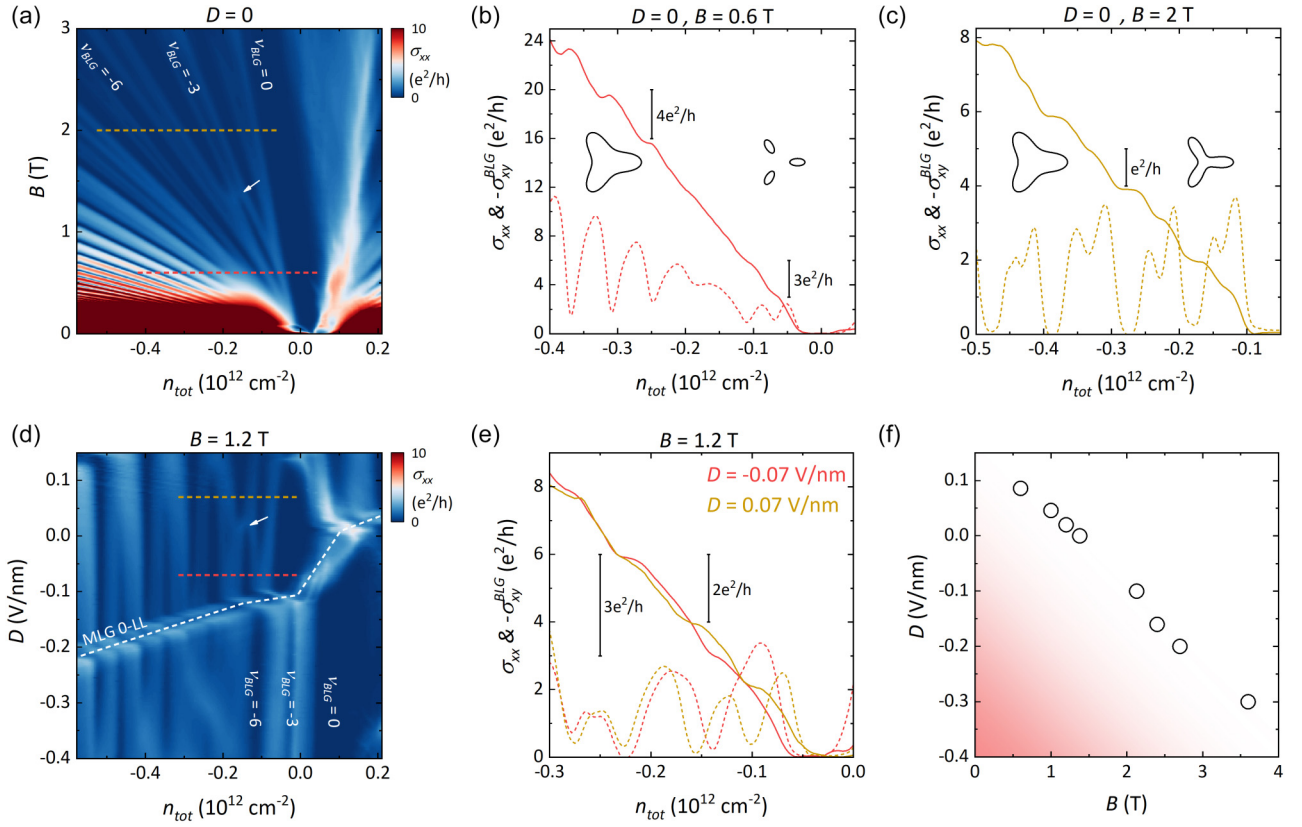


FIG. 2. Tunable BLG Lifshitz transitions in TMBG. (a) Longitudinal conductivity as a function of magnetic field and carrier density, measured at $D = 0$. The red and yellow dashed lines locate data plotted in (b) and (c), respectively. The white arrow indicates a LL crossing close to the BLG valence band edge. (b) Longitudinal (dashed line) and Hall (continuous line) conductivity, measured along the red dashed line in (a) ($B = 0.6$ T). The evolution of the FS induced by hole doping is sketched. (c) Same as (b), measured along the yellow dashed line in (a) ($B = 2$ T). (d) Longitudinal conductivity as a function of displacement field and carrier density, measured at $B = 1.2$ T. The red and yellow dashed lines locate data plotted with the same colors in (e). The white arrow indicates the same LL crossing individuated in (a). (e) Longitudinal (dashed line) and Hall (continuous line) conductivity, measured along the red and yellow dashed lines in (d) ($D = -0.07$ V/nm and $D = 0.07$ V/nm, respectively). (f) Position of the LL crossings as a function of magnetic and displacement field. FS1 is observable only on the bottom-left side of the diagram (indicated by the red shading). For better visibility, σ_{xx} traces are multiplied by a factor $\times 2$, $\times 10$, $\times 4$ in (b), (c), (e), respectively.

Line cuts of longitudinal conductivity (σ_{xx}) and σ_{xy}^{BLG} , shown in Fig. 2(b), confirm the change in degeneracy, which marks the underlying doping-induced LT from FS1 to FS2 at zero field. According to single-particle calculations [38,39] FS1 should give a first QH state at $\nu_{BLG} = -6$, due to the presence of three pockets and spin degeneracy (the valley degeneracy is lifted in the LL spectrum of gapped BLG). $\nu_{BLG} = -3$ is stabilized by exchange interaction and shows comparable robustness to $\nu_{BLG} = -6$, as also reported for BLG in Ref. [40]. By increasing the magnetic field, we observe a LL crossing [white arrow in Fig. 2(a)] that alters the QH sequence. As shown in Fig. 2(c), above this LL crossing we observe all integer QH states, with even (spin-unpolarized) states more robust than odd (spin-polarized) ones. Hence, irrespective of the doping level, QH states characteristic of a single hole pocket are observed [see sketch in Fig. 2(c)]. This transition was first identified in Ref. [38] and attributed to electronlike LLs in the BLG valence band that do not mix with holelike ones. We note that the same change in the QH sequence could be also driven by magnetic breakdown [40], considering that

the k -space separation of the pockets in FS1 is comparable to the inverse of the magnetic length ($\sim 0.06 \text{ nm}^{-1}$ at $B = 2$ T).

The field-driven QH transitions observed at $D = 0$ can be further tuned by the D field, as exemplified by data at $B = 1.2$ T shown in Fig. 2(d). In this map, BLG QH states disperse vertically, unless shifting by $\nu_{tot} = +4$ when crossing the MLG 0-LL. We detect again a transition close to the valence band edge, positioned at $D \sim 0$: for $D < 0$ (red dashed line) we observe threefold QH states, while only twofold states are visible at $D > 0$ (yellow line).

The corresponding conductivity line traces are shown in Fig. 2(e). The phenomenology is similar to the B -driven transition discussed above, and consistent with previous observations in biased freestanding BLG [38–40]. We note that odd states are absent for $D > 0$, while $\nu_{BLG} = -3$ is present for $D < 0$ at the same magnetic field, suggesting a different stabilization mechanism of spin-polarized states on the two sides of the QH transition. A B - D phase diagram is shown in Fig. 2(f), where we plot the position of the transition from additional measurements either at fixed B or D (shown in

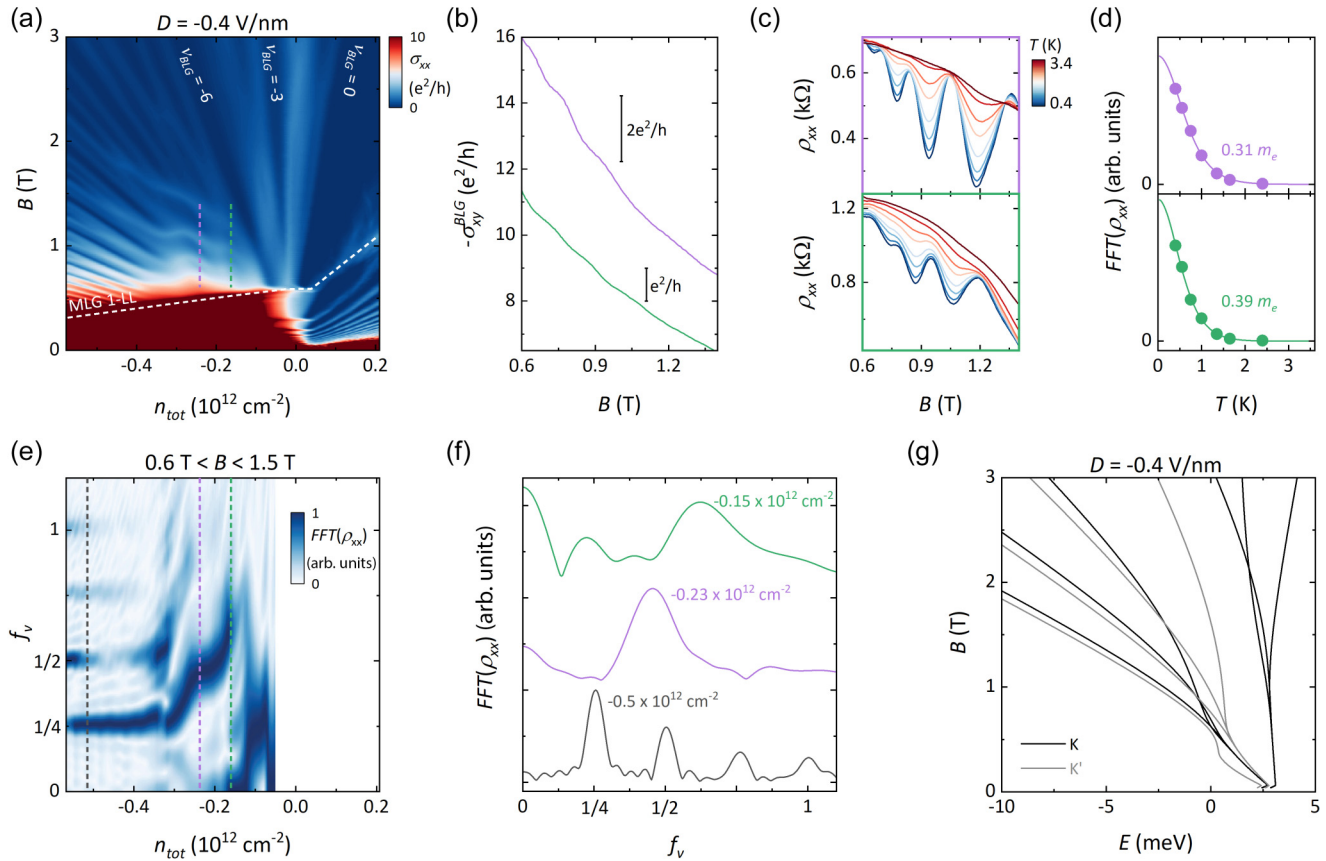


FIG. 3. Signatures of spontaneous isospin polarization of BLG holes. (a) Longitudinal conductivity as a function of magnetic field and carrier density, measured at $D = -0.4 \text{ V/nm}$. The pink and green dashed lines identify data plotted in (b) and (c). (b) Hall conductivity measured along the dashed lines in (a). (c) Longitudinal resistivity at selected temperatures, measured along the dashed pink (upper) and green (lower) lines in (a). (d) Amplitude of the FFT peak as a function of temperature (dots), corresponding to the resistivity oscillations in (c). The continuous lines are fits to the data using the Lifshitz-Kosevich formula. (e) FFT power spectral density as a function of carrier density and frequency, calculated from data in (a). The dashed lines correspond to individual spectra plotted in (f). (f) FFT spectra at selected carrier density. (g) Single-particle calculations of BLG LLs in biased TMBG. Black (gray) lines correspond to levels from the K (K') valley. Each level has a twofold spin degeneracy.

Fig. S3 and Fig. S4, respectively [36]). The magnetic field required for the transition increases with $|D|$ for $D < 0$, while it reduces for $D > 0$, showing a trend toward vanishing at the gap closing point ($D = 0.15 \text{ V/nm}$, where we measure a conventional fourfold LL fan, shown in Fig. S4 [36]).

We next focus on the behavior at large negative displacement field ($D = -0.4 \text{ V/nm}$), where the vHs is enhanced, shifts toward higher hole density, and measurements in magnetic field are less restricted by the transitions discussed in the previous section. Comparison with analogous data from freestanding BLG [20,23,37] can be made by considering the D field acting on BLG as the sum of the gate induced one (see Fig. S2 [36]) and the one due to energy shifts in hBN-encapsulated TMBG, resulting in $\sim -0.55 \text{ V/nm}$. Figure 3(a) shows a rich LL fan diagram. MLG electron LLs are progressively filled in the lower part of the plot (up to $\sim 0.6 \text{ T}$), implying that $v_{MLG} = +2$ should be considered at higher fields. Therefore, the slope of BLG QH states shifts by a factor $+4$ with respect to the fan diagram measured at $D = 0$ [Fig. 2(a)]. Close to the valence band edge, QH states at $v_{BLG} = 0, -3, -6$ can be again identified. $v_{BLG} = -3$ extends uninterrupted over the whole magnetic

field range considered, as expected from the positioning on the phase diagram in Fig. 2(f). Nonetheless, we observe a series of crossings at partial integer fillings ($v_{BLG} = -1, -2$ and $v_{BLG} = -4, -5$, see also Fig. S5 [36]). According to Ref. [37] these structures arise from LLs crossings for the non-degenerate case, thus indicating isospin polarization in FS1. Further crossings and nonmonotonous behavior are visible at higher hole density, qualitatively similar to observations in Ref. [37], eventually leading to the standard fourfold QH states associated to FS2. We focus on the intermediate doping range ($-0.3 \times 10^{12} \text{ cm}^{-2} < n_{tot} < -0.1 \times 10^{12} \text{ cm}^{-2}$) in the vicinity of the vHs, where we identify two separate sets of oscillations [pink and green dashed lines in Fig. 3(a)], which were not detected at $D = 0$. The corresponding Hall conductivities [Fig. 3(b)] show approximately double and single quantized steps, consistent with an approximate double and single spacing in filling factor (see Fig. S5 [36]), respectively. The oscillations are quickly suppressed by temperatures of a few Kelvin [Fig. 3(c)], following a Lifshitz-Kosevich-type dependence [41] (see fits in Fig. 3(d) and SM for analysis details [36]). The extracted effective mass m^* is found to be within $0.3\text{--}0.4$ electron mass (m_e) for both oscillations.

Such values, one order of magnitude larger with respect to measurements on unbiased BLG at similar carrier density [42], are indicative of band flattening at the vHs. Following Refs. [20,23], we consider the fast Fourier transform (FFT) of resistivity data as a function of $1/B$ (see Supplemental Material for technical details [36]), presented as a color map as a function of n_{tot} in Fig. 3(e) [selected line traces are shown in Fig. 3(f)]. We limit this analysis to $0.6 \text{ T} < B < 1.5 \text{ T}$, where the two high-mass oscillations previously discussed are clearly discernible, and avoiding contributions from the MLG LLs. On the y axis, we divide the oscillation frequency f_B by the carrier density and the magnetic flux, obtaining $f_v = f_B/(n_{\text{tot}} h/e)$, which corresponds to the fraction of carriers enclosed by the coherent orbit producing the $1/B$ -periodic resistivity oscillation [4]. At large hole densities (dark gray dashed line), we observe a main peak at $f_v = 1/4$, indicating four Fermi contours with equal carrier population, as expected for FS2. This FFT peak progressively shifts to $f_v > 1/4$ with reducing hole density, consistent with the nucleation of a small electron orbit [4]. Close to $n_{\text{tot}} \sim -0.3 \times 10^{12} \text{ cm}^{-2}$ the FFT spectrum shows discontinuity, followed by a progressive increase in the frequency of the main peak, which saturates close to $f_v = 1/2$. At this point, we encounter the first high-mass oscillation described above (pink dashed line), consistent with the approximate twofold steps in σ_{xy}^{BLG} [Fig. 3(b)]. However, the fact that the peak frequency is always $f_v < 1/2$ indicates the presence of a population of minority carriers with a different spin-valley flavor, in line with partially isospin polarized (PIP) phases previously identified in BLG [20,23]. The same picture of partial isospin polarization applies to the second oscillation (green dashed line), which shows a frequency $1/2 < f_v < 1$. Here, we observe also lower-frequency components in the FFT, which might arise from the minority carriers.

Single-particle calculations for BLG under a comparable D field, shown in Fig. 3(g), predict lifting of the valley degeneracy and a series of LLs crossings. However, the salient features identified experimentally, i.e., partial isospin polarization and the two high-mass oscillations at intermediate densities, are not expected from the BLG LL spectrum (an interacting band model would be likely required, see for

instance Ref. [43]). Despite the proximity of MLG, which has been shown to effectively screen disorder in large-angle twisted bilayer graphene [34], the observed sequence of PIP phases agrees with those reported in Refs. [20,23]. As such, the screening effect of MLG does not appear to suppress or significantly alter the phase diagram of gapped BLG. Our results are in accordance with experiments on magic-angle graphene with a proximal large-angle twisted MLG [44], which show preservation of correlated states from the flat-band subsystem.

Conclusions. In conclusion, we showed that twist-decoupled monolayer-bilayer graphene stack offers a clean and controllable way to access the regime where Bernal bilayer graphene develops strong interaction effects. We uncover tunable changes in Fermi-surface topology and high-mass oscillations in correspondence of partially isospin-polarized states, showing that key features of the correlated physics of BLG are preserved in the presence of a nearby MLG.

These results establish CVD-based TMBG crystal as a scalable platform capable of hosting the rich many-body behavior previously accessible only in the cleanest exfoliated devices, broadening the experimental landscape for studying correlated electrons in multilayer graphene and opening new opportunities for engineered van der Waals systems.

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Data availability. The data that support the findings of this Letter are openly available [45].

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