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Layer-selective spin-orbit coupling and strong correlation in bilayer graphene

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Layer-selective spin-orbit coupling and strong correlation in bilayer graphene

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Supplementary material for this article is available [online](#)

Abstract

Spin-orbit coupling (SOC) and electron-electron interaction can mutually influence each other and give rise to a plethora of intriguing phenomena in condensed matter systems. In pristine bilayer graphene (BLG), which has weak SOC, intrinsic Lifshitz transitions and concomitant van-Hove singularities lead to the emergence of many-body correlated phases. Layer-selective SOC can be proximity induced by adding a layer of tungsten diselenide (WSe₂) on its one side. By applying an electric displacement field, the system can be tuned across a spectrum wherein electronic correlation, SOC, or a combination of both dominates. Our investigations reveal an intricate phase diagram of proximity-induced SOC-selective BLG. Not only does this phase diagram include those correlated phases reminiscent of SOC-free doped BLG, but it also hosts unique SOC-induced states allowing a compelling measurement of valley *g*-factor and a correlated insulator at charge neutrality, thereby showcasing the remarkable tunability of the interplay between interaction and SOC in WSe₂ enriched BLG.

1. Main

Various distinct strongly interacting states often compete for the ground state of a disorder-free two-dimensional (2D) electron system under electron-electron interactions. Recently, the seemingly well-explored family of few-layer graphene has gained revived interest [1–10]. For example, in the simplest AB-stacked bilayer graphene (BLG), a plethora of many-body states has been revealed [2, 4, 5, 10]. Here, electric displacement field-controlled inversion symmetry breaking further flattens the bands yet making the low-density saddle points experimentally accessible. Consequently, Stoner ferromagnetic states [2, 4, 5, 10], correlated (semi-) insulating and metallic states [4, 5], and superconducting states [2] have been identified close to the field-controlled van-Hove

singularities. The detailed nature and mechanisms of these states are under active investigations. For example, it has been shown that proximity-induced spin-orbit coupling (SOC) of Ising (valley-Zeeman) type allows the observation of superconducting states in a much broader parameter space [11, 12], leading to the question of whether SOC is responsible for suppressing interacting states that compete with superconductivity [13], or whether superconductivity is enhanced by SOC [12]. To date which effect (e.g. SOC, Coulomb interaction, saddle points) favors which ground state has still been an open question both from the theoretical [13] and experimental [11, 12] sides.

While previous investigations have focused on the impact of proximity-induced SOC on the superconductivity of BLG [11, 12], impacts of such a SOC on

the rest of its correlated phase diagram remains unexplored. Here, we systematically disentangle the intricate interplay between electronic correlations and SOC in spin-orbit proximitized BLG, aiming to elucidate the entire phase diagram. To this end we prioritize the competition between SOC and Coulomb interaction in their impacts on those BLG ground states enriched by the displacement field-controlled saddle points yet outside of the superconducting regime.

A typical device scheme is shown in figure 1(a), where a monolayer of WSe₂ is placed below a BLG flake. The BLG/WSe₂ heterostructures are contacted by two-terminal graphite flakes and encapsulated in hexagonal boron nitride (hBN). Additional graphite top and bottom gates are incorporated to provide electrostatic control and to ensure a high device quality. Optical microscope images of the two encapsulated devices investigated in this study are displayed in figure 1(b) and supplementary figure S1.

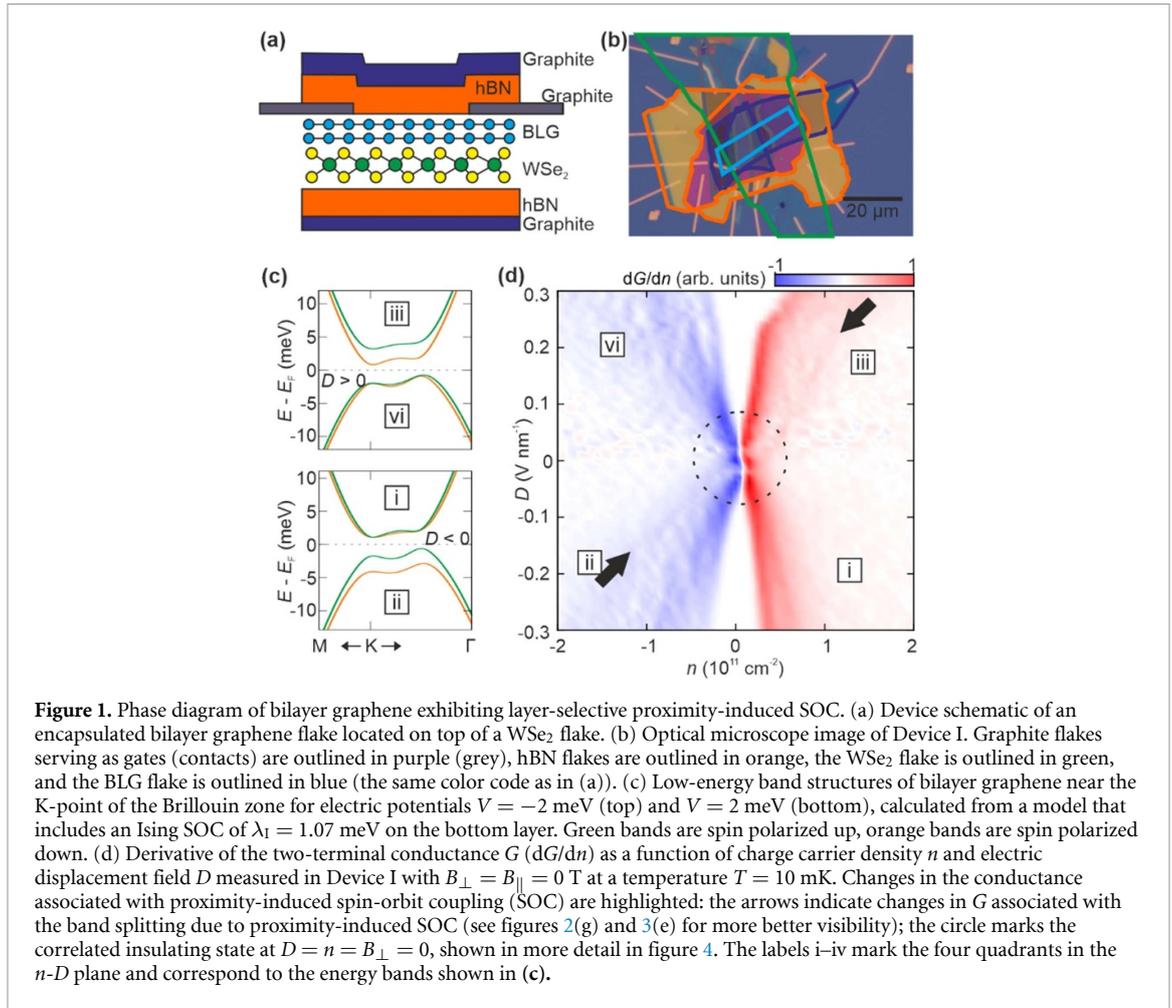
The dual-gate structure enables independent tuning of the charge carrier density (n) and the electric displacement field (D). In such a device configuration, Ising SOC (valley-Zeeman type) [14] is expected to only be introduced to the bottom graphene layer [12, 15, 16], which can be confirmed by our band structure calculations (figure 1(c)). The proximity-induced SOC results in out-of-plane spin-valley locking of the Ising SOC (valley-Zeeman type), and relatively large (meV) spin-splitting of the low-energy bands of BLG at the K and K' points with no overall layer asymmetry [16]. Note that the influence of Rashba SOC on the low-energy band structure is minimal (see supplementary figure S2). As a result, it is neglected in the present study. At $D > 0$ and $n > 0$ (or $D < 0$ and $n < 0$), electronic states near the Fermi energy are polarized to the layer adjacent to the WSe₂ [12, 15, 16], exhibiting the proximity spin-orbit physics. Conversely, the energy bands for $D > 0$ and $n < 0$ (or $D < 0$ and $n > 0$) are only weakly affected by the presence of WSe₂ (figure 1(c)). This D -dependent spin-valley-locked, spin-split band structure has a direct consequence for the charge transport at small n and D as shown in figure 1(d), where the occupation of the high-energy spin-split conduction and valence bands, as marked with arrows, are featured by increased dG/dn with conductance G (see supplementary figure S3 for a complete phase diagram of both devices investigated for the present study) [15]. While these transitions are more clearly resolved in the derivative map shown in figure 1(d), they are also present—albeit less prominently—in the raw conductance data (see, for example, figure 3(e) which will be discussed later). The strength of the Ising SOC λ_I can be determined directly by mapping the magnetic field dependence of particular Landau-Levels (LL),

e.g. the $\nu = 3$ state, to be $\lambda_I = (1 \pm 0.1)$ meV (see methods and supplementary figure S4), which is consistent with previous results [11, 12, 15, 17, 18] and DFT calculations [14, 16].

2. Correlated phases in the vicinity of tunable van-Hove singularities

We start our analysis in the conduction band at large D fields, where, in the absence of SOC, a cascade of correlated phases was found as low-temperature ground states (figures 1–3) [5]. Since BLG is only proximitized on one side by WSe₂, we can investigate the presence and absence of SOC in the same sample, by selectively addressing the layer-polarized energy levels. We first re-investigate the conduction band without SOC ($D < 0, n > 0$) in both non-proximitized BLG (figures 2(a)–(c)) and in proximitized BLG when electron transport is taking place in the top layer where the effects of SOC are weak (figures 2(d)–(f)). Consistent with previous works [5, 10], three phases can be identified at large $|D|$ close to the band edge: a spin- and valley-polarized (quasi-) insulating phase (1x), a spin-polarized (quasi-) insulating phase (2x), and a fully spin- and valley-degenerate metallic phase (4x) (figures 2(a)–(f)). The (quasi-) insulating character of the 1x and 2x phases was discussed in [5]. The labels '1x,' '2x,' and '4x' reflect the different spin and valley degeneracies of the system which are determined by examining the behavior of the phase boundaries with respect to the applied out-of-plane and in-plane magnetic field (B_{\perp} and B_{\parallel}) [5, 10]. Applying a finite B_{\parallel} (figures 2(c) and (f)), which only couples to the spin degree of freedom, does not shift the phase boundary between the 1x and 2x phases, indicating similar spin polarizations of both phases. By contrast, the phase boundary between the 2x and 4x phases shifts towards higher electron densities, indicating spin polarizations in the 1x and 2x phases but not in the 4x phase. Applying B_{\perp} , which couples to the valley and out-of-plane spin degrees of freedom via the orbital and spin Zeeman effects, affects both phase boundaries (figures 2(b) and (e)), indicating a change in valley polarization between the 1x and 2x phases, i.e. we can identify the 1x phase to be valley polarized while the 2x and 4x phases are valley unpolarized. This method allows to identify the degeneracies even at $B_{\perp} = 0$ where no quantum Hall oscillations are present. Remarkably, these characteristics are consistent in both non-proximitized BLG (figures 2(a)–(c)) and in proximitized BLG when electron transport is taking place in the top layer where the effects of SOC are weak (figures 2(d)–(f)).

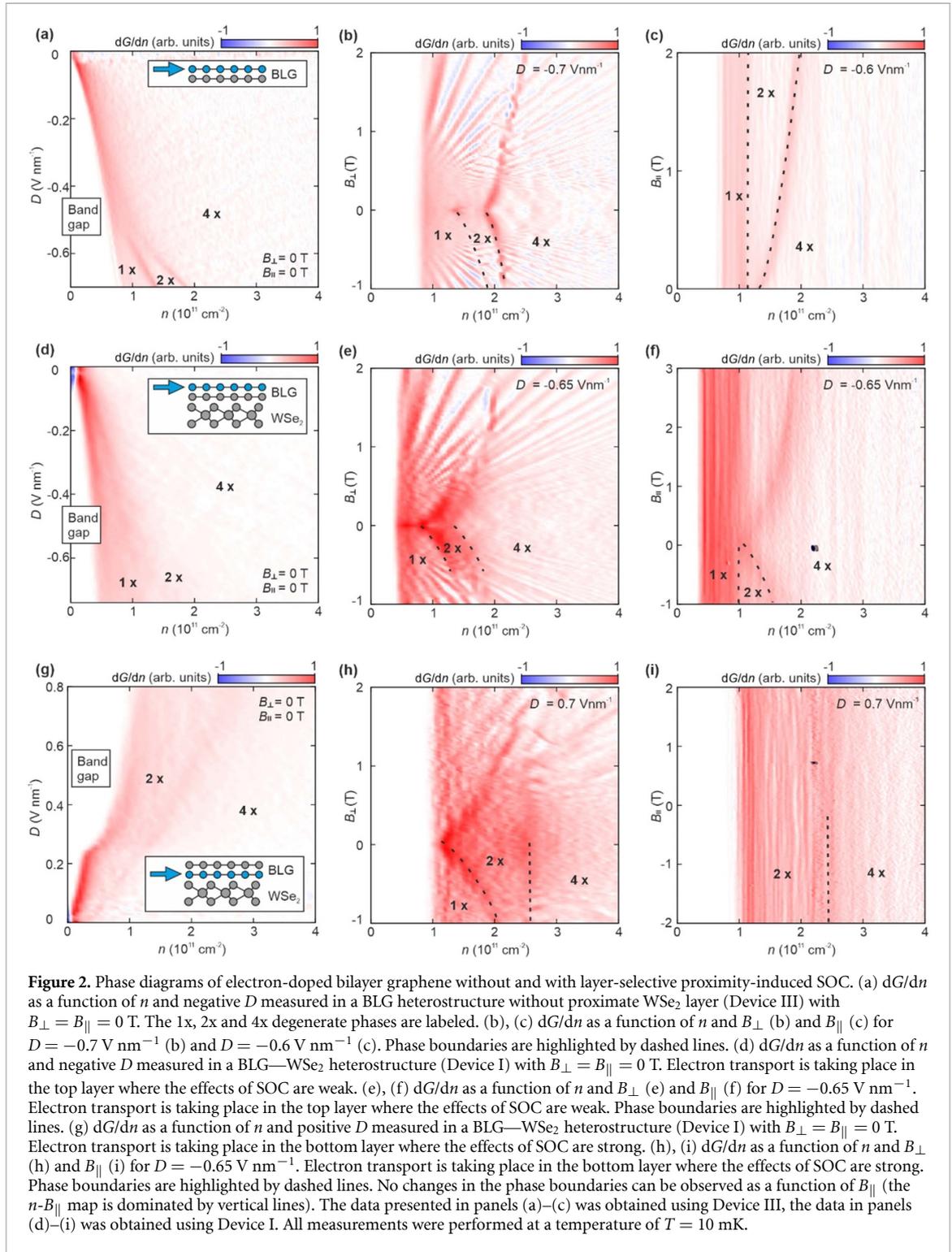
A drastically different phase diagram appears in the same parameter space ($|D|$ and n) but for the



WSe₂ proximitized conduction band ($D > 0$, $n > 0$, figures 2(g)–(i)). Consistent with other works [19–21], we do observe signatures of a spin/valley polarized phase, which we attribute to the SOC-induced splitting of the originally four-fold degenerate band into two sets of two-fold degenerate spin-valley-locked bands. While the transition between the two sets of SOC-split bands shown in figure 1(d) extends to large D (figure 2(g)), in the investigated range up to 0.8 V nm, no additional conductance features indicating additional spin or valley polarization appear at $D > 0$ and $B = 0$, unlike on the non-WSe₂-proximitized side. It appears that in the presence of SOC, the formation of interaction-driven spin and/or valley polarized Stoner phases near the conduction band edge of D field-gapped BLG is suppressed. This picture is also consistent with the magnetic field data. Applying B_{\perp} induces a valley splitting in the low-energy spin-valley-locked band due to the valley-Zeeman effect. This splitting shifts to higher energies with increasing B_{\perp} and manifests as a peak in dG/dn that is marked with a dashed line in figure 2(h) and is shifting to higher n for increasing B_{\perp} . At a critical magnetic field B_C (supplementary figure S5), the upper band of the low-energy spin-valley-locked band (K valley, spin down and the lower

band of the high-energy spin-valley locked band (K' valley, spin down) intersect. These bands share the same spin but have opposite valley indices. This crossing point allows us to determine the D -dependent valley g -factor, which quantifies the strength of the valley magnetic moment [22]. Using $\lambda_I = \frac{1}{2} \mu_B g_V B_C$ with Bohr magneton μ_B and $B_C = 1.9$ T, we find g_V to be 22 at $D = 0.7$ Vnm⁻¹, which aligns well with previous results obtained in BLG quantum dots [23–25]. In the B_{\parallel} map up to 2 T we do not observe any change of the phase boundaries, since in this regime the spin Zeeman energy $E_z \ll \lambda_I$ [16].

All our observations point to the weakening or absence of Stoner ferromagnetic phases when the electrons are polarized to the layer adjacent to WSe₂. Theoretically, the Ising SOC of out-of-plane spin quantization, together with the WSe₂-enhanced screening effect, is expected to disfavor those correlated phases with in-plane spin orientations as previously identified in the absence of SOC [4, 5]. We note that WSe₂ does have a larger dielectric constant than hBN [26]. Given that monolayer WSe₂ is used in our devices, the short-range part of Coulomb interaction is more significantly screened than the long-range Coulomb tail. Nevertheless,—even though the effect will likely be small—the WSe₂-enhanced screening



may disfavor Stoner and more elaborate correlated phases [27–29]. The suppression of the Stoner phases on the WSe_2 polarized layer might be further enhanced by greater disorder at the WSe_2 /BLG interface compared to the BLG/hBN interface (see methods for details on the sample fabrication).

The picture becomes more complex on the hole-doped side of BLG, where trigonal warping is more

pronounced [30] and Lifshitz transitions with concomitant van-Hove singularities give rise to a cascade of correlated insulating and metallic phases of both Stoner and non-Stoner types [4]. These phases were initially observed in pristine BLG (see also figures 3(a), (b)) and can also be identified similarly in the BLG/ WSe_2 devices for $D > 0$ and $n < 0$, where charge transport occurs in the layer non-adjacent to

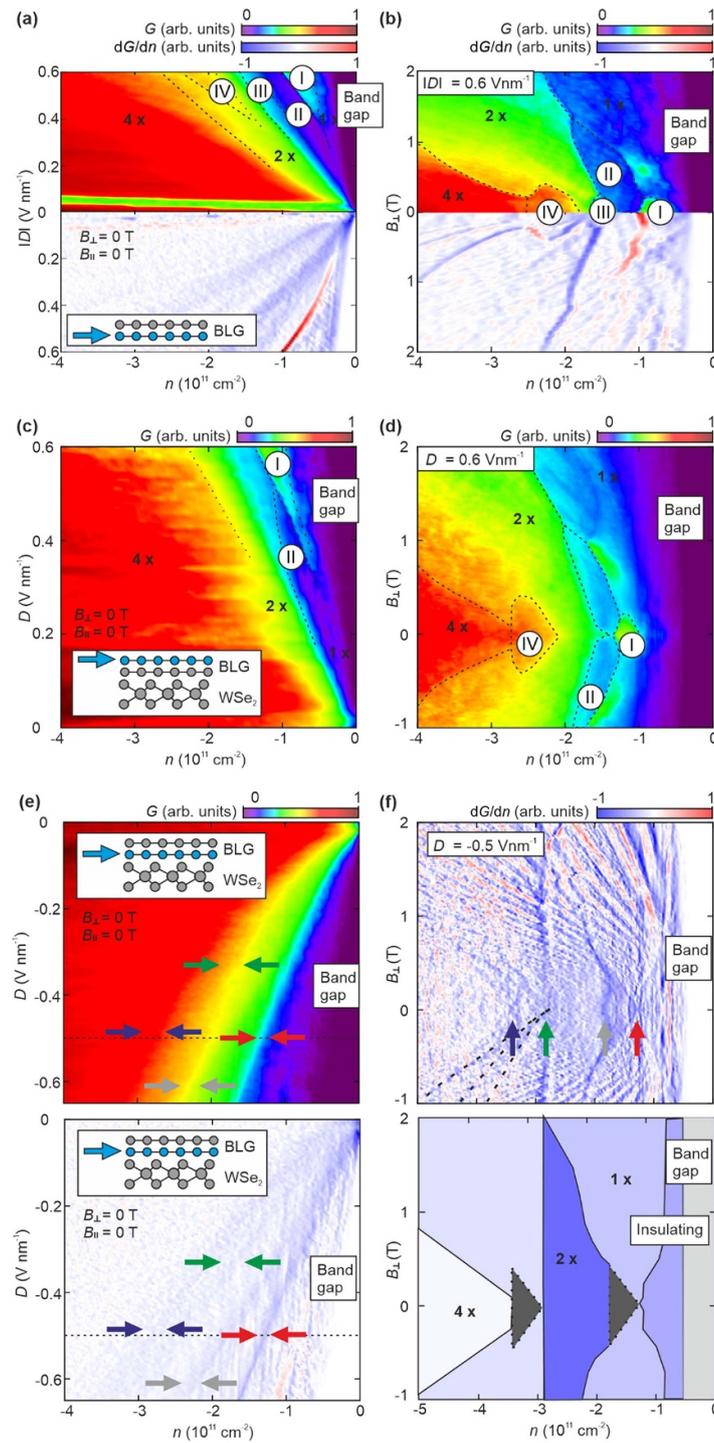


Figure 3. Phase diagrams of hole-doped bilayer graphene without and with layer-selective proximity-induced SOC. (a) Conductance G (top panel) and derivative of the conductance dG/dn (bottom panel) as a function of the charge carrier density n and electric displacement field $|D|$ measured in a BLG heterostructure without proximate WSe_2 layer (Device III) with $B_{\perp} = B_{\parallel} = 0$ T. Electron transport is taking place in the top layer where the effects of SOC are weak. Phase boundaries are highlighted by dashed lines. The 1x, 2x and 4x degenerate phases are labeled. Interaction-induced phases are labeled according to Seiler *et al* [4]. Where phases I and IV correspond to correlated metals of non-Stoner type and phases II and III correspond to correlated insulating phases consistent with a Wigner-Hall crystal (phase II) and a trivial Wigner crystal (phase III). (b) G (top panel) and dG/dn (bottom panel) as a function of n and B_{\perp} at $|D| = 0.6 \text{ Vnm}^{-1}$. (c) G as a function of n and positive D measured in a BLG— WSe_2 heterostructure (Device II) with $B_{\perp} = B_{\parallel} = 0$ T. Electron transport is taking place in the top layer where the effects of SOC are weak. Phase boundaries are highlighted by dashed lines. The interaction-induced phases are labeled. (d) G as a function of n and B_{\perp} at $D = 0.6 \text{ Vnm}^{-1}$. (e) G (top panel) and dG/dn (bottom panel) as a function of n and negative D with $B_{\perp} = B_{\parallel} = 0$ T. Electron transport is taking place in the bottom layer where the effects of SOC are strong. Phase boundaries are highlighted by arrows. The interaction induced phases I–IV are not present. (f) Top panel: dG/dn as a function of n and B_{\perp} for $D = -0.5 \text{ Vnm}^{-1}$. Phase boundaries are highlighted by arrows. Quantum Hall states arising from the second energy band are traced by dashed lines for negative B_{\perp} . Bottom panel: schematic of the different phases induced by SOC. The one-fold, two-fold, and four-fold degenerate phases are labeled as 1x, 2x, and 4x, respectively. Additional phases in the low-density regime of the 2x and 4x degenerate bands are shaded in gray. Furthermore, an insulating phase emerges near the band edge. The data presented in panels (a), (b) was obtained using Device III, the data in panels (c)–(f) was obtained using Device II. All measurements were performed at a temperature of $T = 10$ mK.

WSe₂ (figures 3(c) and (d)). However, the proximity-induced Ising SOC is present for $D < 0$ and $n < 0$ when charge transport occurs in the layer adjacent to WSe₂, and evidently it disfavors these correlated phases (see figures 3(e) and (f) for the data from Device II and supplementary figure S6 for data from Device I). Notably, our findings reveal multiple distinctive features in the density derivative of the conductance, as indicated by arrows in figures 3(e) and (f), which is in sharp contrast to our observations at low D fields (figure 1(d)), at electron doping (figures 2(e) and (f)), and in previous literature [13].

The first feature at lowest n near the band edge separates low and high conductance regimes (red arrows in figures 3(e) and (f)). In the low-conductance regime, the conductance sharply increases with increasing applied current I_{Bias} (supplementary figures S7(a) and (b)), indicating insulating behavior, possibly arising from disorder [31, 32], Wigner crystallization [33], or more exotic phenomena, e.g. induced by exchange interaction between the trigonal warping induced mini valleys [34–36]. A detailed analysis of this regime, however, extends beyond the scope of this present study. This regime is markedly different from that when WSe₂ is absent, where a metallic phase had been observed at the valence band edge [4].

Another marked difference is in the higher density regime, where a cascade of correlated metallic and insulating phases were found [4] in the absence of SOC, whereas in the presence of SOC no clear signs of physics beyond SOC can be identified. These features can be associated with the SOC-induced band splitting and the occupation of the second energy band (green arrow in figures 3(e) and (f)), exhibiting B_{\parallel} - and B_{\perp} -dependences similar to those observed at the electron doped case with induced SOC (supplementary figures S8 and 3(f)).

Additional phase boundaries (blue and gray arrows in figures 3(e) and (f)) emerge within the spin-valley-locked bands, with the corresponding phases shaded gray in the lower panel of figure 3(f) [11, 12]. Since trigonal warping is more pronounced in the valence band [30], these boundaries may separate regions with a different Fermi surface topologies, i.e. where the Fermi surface transitions from being three-fold degenerate to one-fold degenerate. Alternatively, they may mark the transition from a partially isospin-polarized to a fully isospin-polarized regime. This interpretation is supported by the appearance of an additional fan with Landau levels that curve in the n - B_{\perp} space (see e.g. dashed lines in figure 3(f)) and has been attributed to a partially isospin-polarized regime with small minority Fermi pockets [1, 2].

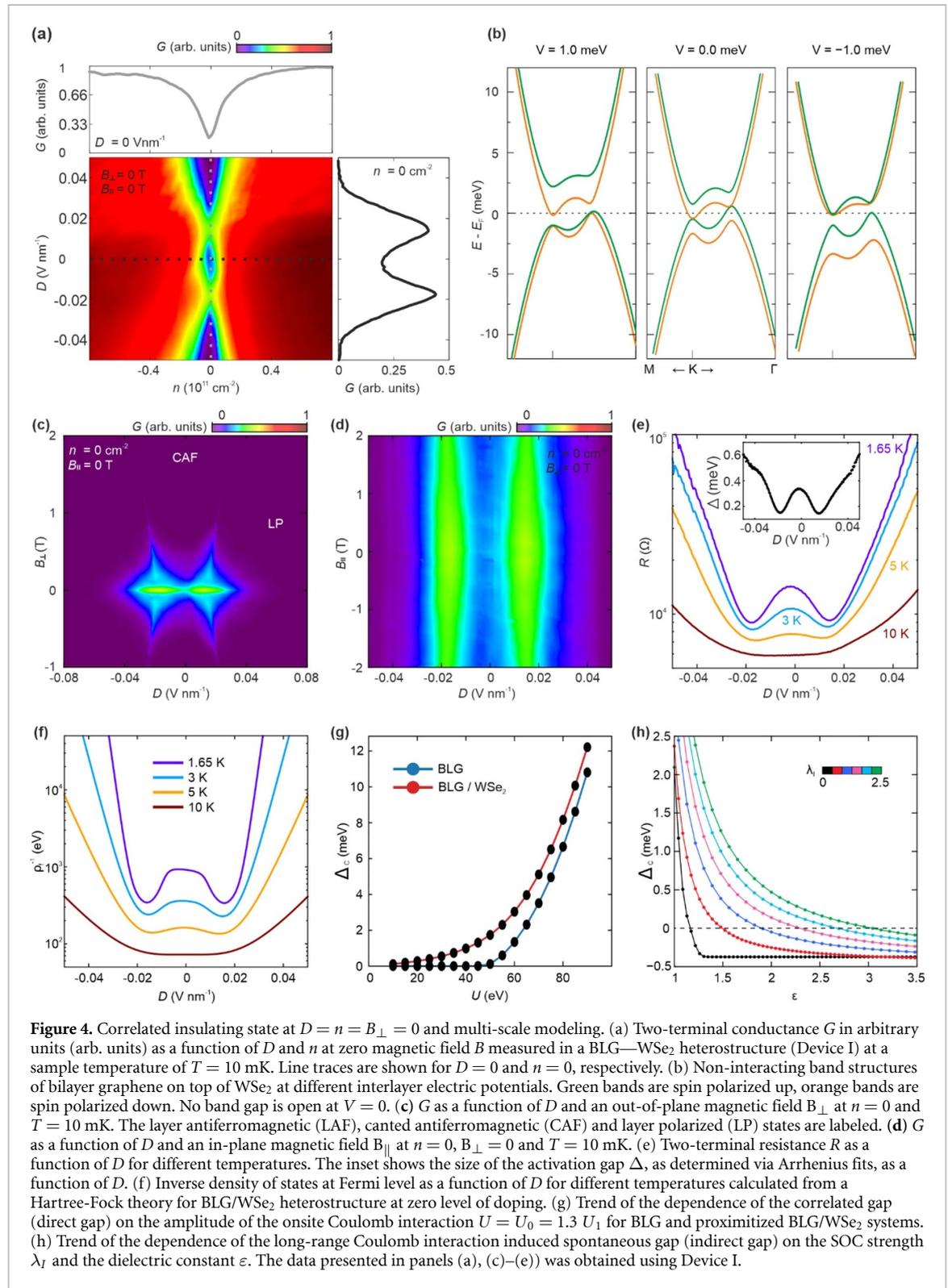
With the exception of the phase at lowest n , the prevalence of mainly SOC-driven states is also supported by the absence of non-linear dI/dV feature

and the lack of strong temperature dependence (supplementary figures S7(a)–(c)). Finally, we note the absence of any indication of superconductivity near these potential phase boundaries [11, 12]. This may be attributed to constraints imposed by the application of higher D -fields due to elevated gate currents and the use of thicker hBN dielectrics in the range of 37 nm to 62 nm compared to two previous studies [11, 12].

3. Transport in WSe₂ proximitized BLG at overall charge neutrality

While at large D fields and finite doping the correlated phases are strongly suppressed when charge carriers are layer-polarized to the WSe₂-BLG interface, the situation is different at charge neutrality when there is no layer polarization ($D = n = 0$). The ground state of undoped BLG has been intensively addressed by multiple works [7, 37–41]; the strength of Coulomb interaction plays a critical role. In the limit of strong interaction, as experimentally realized in suspended BLG, an insulating layer antiferromagnetic (LAF) state has been identified as the ground state [7, 37–41]. In the LAF state, two spin species spontaneously polarize to opposite layers [22]. On the other hand, in hBN encapsulated BLG, interaction is weaker due to the larger dielectric constant ϵ_r and closer distance of screening metal gates (about 150 nm in the suspended samples and typically below 60 nm in hBN encapsulated samples), and the LAF state has not been found. Thus, one may wonder whether the ground state is still correlated or non-interacting given the induced SOC but the weakened Coulomb interaction.

From our multi-scale theoretical investigation of the single-particle bands (figures 4(f) and (g)), consistent with previous reports, a SOC-split but overall gapless band structure is present. An interesting question now is whether the induced Ising SOC and the more screened interaction lead to a correlated insulating state like in the freestanding case or disfavor correlated states as in the case of large D fields discussed above. Figure 4 shows our experimental observations, and a region of suppressed conductance is evident at charge neutrality. This region is unstable against the application of doping or a D field of either sign. These suggest most likely the topologically trivial LAF state as in the freestanding case [7, 37–41], and with increasing B_{\perp} the LAF state evolves into a canted antiferromagnetic (CAF) state (figure 4(c)) [42–44]. Furthermore, this state is not strongly affected by the application of a B_{\parallel} field (figure 4(d)), consistent with two previous studies [41, 44]. In addition, we observe insulating behavior, i.e. decreasing resistance with increasing temperature or increasing current (figure 4(e) and supplementary figure S9). The insulating state is stable below 5 K, and we extract an energy gap of 0.4 meV using Arrhenius



fits (see inset of figure 4(e) and supplementary figure S10). The respective stability of the LAF state in the D , B_{\perp} , B_{\parallel} space is consistent with the observations made in freestanding graphene, i.e. both states are only stable at low D and become suppressed at finite $D \approx \pm 0.015$ V nm⁻¹ (figure 4(a) and supplementary figure S11(a)) and gradually evolve into a CAF at finite B_{\perp} without apparent changes in the conductance (figure 4(c) and supplementary figure S11(b)).

Also the extracted activation gaps are of similar order (see supplementary figure S11(c) for a direct comparison of the gaps). These experimental observations also align with our self-consistent Hartree-Fock calculations addressing both short- and long-range interactions (see methods for details): opening a spontaneous gap in a BLG/WSe₂ heterostructure requires a smaller Coulomb interaction (figures 4(f) and (g)), and the larger the induced SOC the stronger

the spontaneous indirect gap opening (figure 4(h)). We thus believe that a larger activation gap can be achieved when maximizing the SOC, e.g. by minimizing the twist angle between BLG and WSe₂ [45, 46] or by replacing the WSe₂ with WS₂ [46].

It is worth noting that SOC would introduce single-particle gaps in WSe₂/BLG heterostructures in case they are spin-polarized (figure 4(b)). In this case, the gap would, however, not symmetrically close at finite values of D . Furthermore, the extracted energy gap is approximately one order of magnitude larger than these spin gaps (supplementary figure S17). Lastly, our observations are not only at odds with the single-particle band structure of our WSe₂/BLG heterostructure but also at odds with a previous study of WSe₂/BLG/WSe₂ structures [15, 47], where symmetrically induced Ising SOC results in a single-particle Kane-Mele SOC gap that is suppressed by the application of a B_{\parallel} field [15, 47].

4. Conclusions

In summary, asymmetrically introducing WSe₂ to hBN-encapsulated BLG devices provides a unique platform for the distinct exploration of correlated phases and SOC-induced states, respectively, in the presence of large D fields, depending on the signs of n and D . Whereas a cascade of correlated phases emerges, like those devices without WSe₂, when the charge carriers are polarized on the WSe₂-remote layer of BLG, clear signatures of SOC dominant band splitting have been identified allowing the further measurement of a strong valley g -factor, as if the interaction is nearly absent, when the charge carriers are polarized on the WSe₂-proximate layer of BLG. At zero D field for which the BLG is not layer-polarized, surprisingly, the interaction strength in BLG appears to be enhanced by the induced SOC, giving rise to a correlated insulating state with both theoretically and experimentally anticipated features under B_{\perp} , B_{\parallel} , and D fields. Our results have established a rich phase diagram of BLG, paving the way for exploring the interplay between geometry, interaction, and SOC in strongly correlated electrons.

Data availability statement

All data supporting the messages of the manuscript is displayed in the manuscript. The raw data is available from the authors upon request. The data that support the findings of this study are available upon reasonable request from the authors.

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Note from the authors

While analyzing the data we became aware of similar results presented in a manuscript by Masseroni *et al* [48]. It is remarkable that very similar data was obtained by two different groups, using a different TMD on bilayer graphene (WSe₂ in Göttingen and MoS₂ in Zurich).

Author contributions

A.M.S. fabricated Device I and II with help of D.U. and conducted the measurements and data analysis. E.R.G. fabricated and measured Device III. K.W. and T.T. grew the hexagonal boron nitride crystals. Y.Z., K.Z., C.Y., J.F. and F.Z. contributed to the theoretical part. All authors discussed and interpreted the data. R.T.W. supervised the experiments and the analysis. The manuscript was prepared by A.M.S., Y.Z., K.Z., J.F., F.Z. and R.T.W. with input from all authors.

Conflict of interest

Authors declare no competing interests.

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