
Supplementary information

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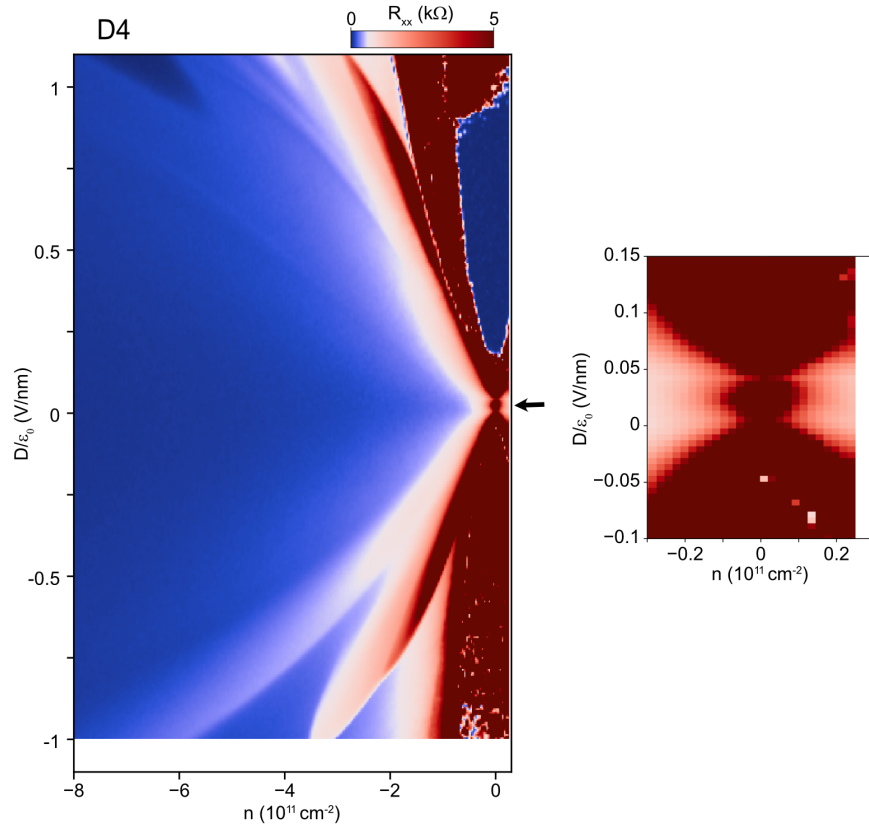
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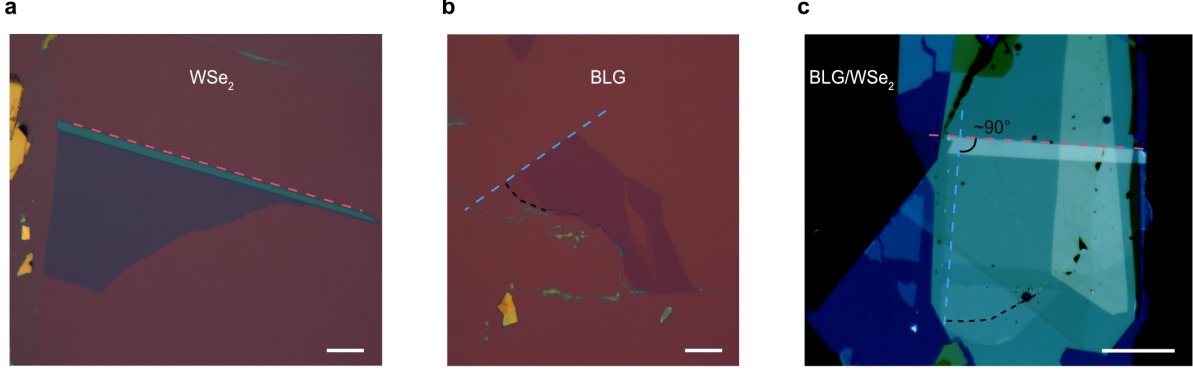
Enhanced Superconductivity in Spin-Orbit Proximitized Bilayer Graphene

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Supplementary Figures



SI Fig. 1 | n - D phase diagram of device D4 including electron doping. R_{xx} versus doping density n and displacement field D including electron doping for device D4. A node-like resistive feature appears around $n = 0$ and $D = 0$ (marked by a black arrow; zoom-in data on the right), showing a small offset in D field ($D/\epsilon_0 \approx 0.025 \pm 0.005$ V/nm).



SI Fig. 2 | Alignment between BLG and WSe₂ for D1. **a–c**, optical image of WSe₂ (**a**), BLG (**b**) and alignment of BLG-WSe₂ (**c**) for device D1. Coloured dashed lines indicate the possible crystal edges with zigzag or armchair orientation. Black dashed lines in **b** and **c** indicate one edge of BLG. The scale bar in each panel corresponds to 20 μm .

Theoretical Analysis

1 Continuum model band structure of bilayer graphene

We consider the low-energy continuum model commonly used to describe Bernal-stacked bilayer graphene (BLG)^{S1}, under a perpendicular displacement field D which generates a potential difference $u = -d_{\perp}D/\epsilon_{\text{BLG}}$ between the top and bottom layers. Here $d_{\perp} = 0.33$ nm is the interlayer distance and $\epsilon_{\text{BLG}} \sim 4.3$ is the relative permittivity of bilayer graphene. A continuum approximation of the band structure returns a Hamiltonian of the form

$$H_0 = \sum_{\xi=\pm} \sum_{\mathbf{k}} \psi_{\xi}^{\dagger}(\mathbf{k}) h_{0,\xi}(\mathbf{k}) \psi_{\xi}(\mathbf{k}), \quad h_{0,\xi}(\mathbf{k}) = \begin{pmatrix} u/2 & v_0 \Pi^{\dagger} & -v_4 \Pi^{\dagger} & -v_3 \Pi \\ v_0 \Pi & \Delta' + u/2 & \gamma_1 & -v_4 \Pi^{\dagger} \\ -v_4 \Pi & \gamma_1 & \Delta' - u/2 & v_0 \Pi^{\dagger} \\ -v_3 \Pi^{\dagger} & -v_4 \Pi & v_0 \Pi & -u/2 \end{pmatrix} \quad (1)$$

where $\Pi = (\xi k_x + i k_y)$ and $v_i \equiv \frac{\sqrt{3}a}{2} \gamma_i$. Here, $\xi = \pm 1$ indicates the valley that has been expanded about: $\mathbf{K}, \mathbf{K}' = (\xi 4\pi/3a, 0)$ with $a = 0.246$ nm the lattice constant of monolayer graphene. The 4×4 matrix $h_{\xi}(\mathbf{k})$ is expressed in the sublattice/layer basis corresponding to creation/annihilation operators of the form $\psi_{\xi}(\mathbf{k}) = (\psi_{\xi,A1}(\mathbf{k}), \psi_{\xi,B1}(\mathbf{k}), \psi_{\xi,A2}(\mathbf{k}), \psi_{\xi,B2}(\mathbf{k}))^T$, where A/B indicate the sublattice, 1, 2 indicate the layer, and the momentum $\mathbf{k}$ is measured relative to $\mathbf{K}_{\xi}$ (indices denoting the spin degrees of freedom have been suppressed). It will sometimes be convenient below to express the Hamiltonian in terms of the spinors $\psi(\mathbf{k}) = (\psi_{+}(\mathbf{k}), \psi_{-}(\mathbf{k}))^T$.

The common values quoted for the five parameters entering the continuum model in Eq. (1) are

$\gamma_0 = 2.61$ eV (intralayer nearest-neighbor tunneling), $\gamma_1 = 361$ meV (leading interlayer tunneling), $\gamma_3 = 283$ meV (also known as trigonal warping term), $\gamma_4 = 138$ meV, and $\Delta' = 15$ meV (potential difference between dimer and non-dimer sites)^{S2}.

A TMD monolayer adjacent to the graphene, such as is the case here with WSe₂, is known to induce SOC via virtual tunnelling^{S3–S5}:

$$H_{\text{SOC}} = \sum_{\xi=\pm} \sum_{\mathbf{k}} \psi_{\xi}^{\dagger}(\mathbf{k}) h_{\text{SOC},\xi} \psi_{\xi}(\mathbf{k}), \quad h_{\text{SOC},\xi}(\mathbf{k}) = \mathcal{P}_1 \left[\frac{\lambda_I}{2} \xi s^z + \frac{\lambda_R}{2} (\xi \sigma^x s^y - \sigma^y s^x) \right], \quad (2)$$

where the Pauli matrices σ^i and s^i , $i = x, y, z$, respectively act on sublattice and spin degrees of freedom. The operator $\mathcal{P}_1$ projects onto the top graphene sheet, i.e., only the sites A1 and B1: $\mathcal{P}_1 = \text{diag}(\mathbb{1}_{2 \times 2}, \mathbb{0}_{2 \times 2})$ in the layer/sublattice basis used to express $h_{0,\xi}(\mathbf{k})$ in (1). The parameters λ_I and λ_R quantify the strength of the Ising (also called “valley-Zeeman”) and Rashba SOC. Ab initio-type numerics and experimental estimates find a range of values $\lambda_I \sim 0 - 5$ meV and $\lambda_R \sim 0 - 15$ meV for the SOC parameters^{S3,S4,S6–S11}, which are also predicted to be strongly twist-angle dependent^{S8–S10}.

In the absence of SOC and an applied displacement field with $v_3 = v_4 = 0$, two bands touch quadratically at charge neutrality. Two remaining bands are at significantly higher and lower energies; their wavefunction are dominated by the “dimer sites,” i.e., the A2 and B1 which sit immediately on top of one another in the bilayer and hybridize strongly through the onsite tunnelling parameter γ_1 . Trigonal warping introduced by the v_3, v_4 associated hoppings in Eq. (1) splits the quadratic band touching at charge neutrality into four distinct Dirac cones separated by van Hove singularities (VHS): one Dirac cone remains at $\mathbf{k} = 0$, while the other three are located at C_3 -related momenta slightly away from the Dirac point. Turning on a displacement field D , a gap opens at charge neutrality and the VHSs move apart in energy. Further, by flattening the band bottom, the applied D field also amplifies divergence of the DOS close to the VHS. The low-energy states near $\mathbf{K}$ and $\mathbf{K}'$ become strongly layer- and sublattice-polarized; e.g., on A1 sites for the valence band and B2 sites for the conduction band, or vice versa for the other sign of D . That is, the low-energy wavefunctions near charge neutrality and under a large D field are strongly localized on the “non-dimer sites” of BLG.

The layer- and sublattice polarization of the low-energy wavefunctions near the $\mathbf{K}, \mathbf{K}'$ points has important consequences for SOC induced by the TMD. Indeed, Rashba SOC does not act effectively in the low-energy theory because it is off-diagonal in the sublattice degree of freedom. It therefore induces a splitting only at second order in degenerate perturbation theory, with $\lambda_R^{\text{eff}} \sim (\lambda_R^2 v_0 k)^2 / (\gamma_1^2 u)$ with u the interlayer potential^{S12} (neglecting effects of further perturbations such as trigonal warping, which we discuss below). By contrast, the Ising SOC acts effectively in the subspace of sublattice- and layer-polarized wavefunctions.

The normalized frequencies one expects from quantum oscillations for the non-interacting theory are shown in Extended Data Fig. 6a as a function of hole doping for $D/\epsilon_0 = 1$ V/nm. The red lines correspond to the spin-orbit-free case, whereas the blue lines are computed in the presence of SOC; we additionally plot the Fermi surfaces for the spin-orbit coupled band structure in the insets at a few representative fillings. The Ising coupling is set to the experimentally extracted value $\lambda_I = 0.7$ meV, whereas for the Rashba coupling we select $\lambda_R = 3$ meV, which is within the upper bound consistent with the experimental resolution. For the filling range shown, five different sets of Fermi surface topologies are present for the case with SOC. For $|n| < 0.5 \times 10^{11} \text{ cm}^{-2}$, an $\text{FP}(6)_+$ state is realized in which each valley contributed three equally sized (but offset in momentum space) Fermi surfaces. Assuming $\lambda_I > 0$, given that the in-plane mixing by Rashba is relatively small, the states that make up these pockets largely have valley and spin quantum numbers $K', \uparrow$ and $K, \downarrow$. With further doping, another six equally sized Fermi surfaces appear corresponding to the spin degrees of freedom pushed down in energy by SOC ($K, \uparrow$ and $K', \downarrow$). This $\text{FP}(6,6)_+$ state persists with doping until $|n| \sim 7.4 \times 10^{11} \text{ cm}^{-2}$, at which point the system reaches a van Hove singularity and thus the Fermi surface structure changes. The three majority pockets merge, resulting in two degenerate hole pockets with a small electron pocket at their centre. With further doping the electron-like pocket of the majority Fermi surfaces vanishes ($|n| \sim 9 \times 10^{11} \text{ cm}^{-2}$). Subsequently, the minority pockets reach the same VHS ($|n| \sim 9.8 \times 10^{11} \text{ cm}^{-2}$) leading to the formation of the small electron pocket.

A comparison of Extended Data Fig. 6a with the quantum oscillations data in Fig. 3c makes it clear that the non-interacting theory is insufficient. In particular, SOC does explicitly “polarize” the band—they are energetically split—so that the $\text{FP}(6,6)_+$ phase is realized; this state persists up to $|n| \sim 7.4 \times 10^{11} \text{ cm}^{-2}$ at $D = 1$ V/nm. By contrast, in the experiment the $\text{FP}(6,6)_+$ phase terminates at around $|n| \sim 5 \times 10^{11} \text{ cm}^{-2}$, where it is replaced by the $\text{FP}(2,2)_+$ state. Hence, while the Ising and Rashba parameters (obtained from measurements at zero D and doping) do arguably polarize the bands in the non-interacting limit, the resulting splitting is not large enough to account for the observed phase diagram.

2 Interactions

The resistance data as a function of displacement field and doping clearly demonstrate that the non-interacting band structure implied by $H_0 + H_{\text{SOC}}$ in the previous section cannot fully describe the system. Instead, given the large density of states close to charge neutrality in the presence of large displacement fields, a series of polarized phases are observed, which are naturally explained as a consequence of the Coulomb interaction.

The Coulomb interaction is given by

$$H_C = \frac{1}{2A} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} V(\mathbf{q}) \psi_{\alpha}^{\dagger}(\mathbf{k}) \psi_{\beta}^{\dagger}(\mathbf{k}') \psi_{\beta}(\mathbf{k}' - \mathbf{q}) \psi_{\alpha}(\mathbf{k} + \mathbf{q}). \quad (3)$$

Here the indices α and β sum over valley, layer, spin, and sublattice degrees of freedom and $A = A_{\text{u.c.}} N_{\text{site}}$ is the total area of the sample, with $A_{\text{u.c.}}$ denoting the unit cell area, $A_{\text{u.c.}} = \sqrt{3}a^2/2$, and N_{site} denoting the total number of sites. The unscreened Coulomb potential is $V(\mathbf{q}) = e^2/(2\epsilon_r\epsilon_0|\mathbf{q}|)$, where $\epsilon_r \sim 4.3$ is the dielectric constant for hBN-screened graphene. Instead of considering this model, we look at a far simpler model in which the interaction is fully local: $V(\mathbf{q}) = A_{\text{u.c.}} U_C$. We can roughly estimate

$$U_C \sim \frac{1}{A_{\text{u.c.}}} \frac{e^2}{4\pi\epsilon_r\epsilon_0} d, \quad (4)$$

where we have substituted $2\pi/q_F \sim d$ with d the inter-particle spacing. A density of $n = -5 \times 10^{11} \text{ cm}^{-2}$ roughly translates to an inter-particle spacing of $d = 15 \text{ nm}$, which in turn implies $U_C \sim 100 \text{ eV}$. This estimate should be taken as an upper bound since it does not include the effects of screening. Accordingly, more reasonable results are obtained by allowed the effective Coulomb interaction strength to take smaller values. In particular, we often select $U_C = 35 \text{ eV}$ in accord with earlier calculations of Bernal stacked systems^{S13, S14}.

We emphasize that U_C should *not* be thought of as the setting the “energy scale” of the problem. Instead, the interactions naturally scale with the density. In particular, if we let ν_f denote the number of electrons per unit cell, $\nu_f = A_{\text{u.c.}} \cdot n = \sqrt{3}a^2/(2d^2)$, then the energy per electron is

$$\epsilon_C = \frac{U_C}{\text{electron}} \sim \frac{e^2}{4\pi\epsilon_r\epsilon_0} \frac{1}{d}, \quad (5)$$

which is precisely what we would have found with a real space description. In this case, we find $\epsilon_C \sim 20 \text{ meV}$ for $d = 15 \text{ nm}$.

Even with the long-range Coulomb form, $V(\mathbf{q}) \propto 1/|\mathbf{q}|$, the interaction presented in Eq. (3) is not fully general. Instead, it was derived by taking the zero momentum portion of the density. In effect, the density can be expanded in terms of the continuum model operators as $\rho(\mathbf{r}) \sim \rho_{++}(\mathbf{r}) + \rho_{--}(\mathbf{r}) + e^{-i\mathbf{K}\cdot\mathbf{r}}\rho_{-+}(\mathbf{r}) + e^{i\mathbf{K}\cdot\mathbf{r}}\rho_{+-}(\mathbf{r})$ where $\rho_{\xi\xi'}(\mathbf{r}) = \psi_{\xi}^{\dagger}(\mathbf{r})\psi_{\xi'}(\mathbf{r})$ with $\psi_{\xi}(\mathbf{r})$ the real space version of the annihilation operator defined in SI, section 1. Equation (3) only includes ρ_{++} and ρ_{--} , which accounts for the long-range part of the Coulomb interaction. The Hund’s term includes the remaining two piece of the density carrying momentum $\mathbf{K}$, $\mathbf{K}'$ and thus necessarily has a minimum momentum transfer of $\mathbf{K}$. Its magnitude can therefore be characterized by $V(\mathbf{K}) = e^2/(4\pi\epsilon_r\epsilon_0|\mathbf{K}|)$. Translating this scale into the relevant energy scale like in (5), we find

$$\epsilon_H \sim \frac{e^2}{4\pi\epsilon_r\epsilon_0} \frac{1}{a} \sim \frac{d}{a} \epsilon_C, \quad (6)$$

where factors of order unity have been neglected.

3 Symmetries

We begin by discussing the flavour symmetries of the Hamiltonian in the absence of SOC. It immediately follows that the system is invariant under the usual $SU(2)_s$ spin rotation symmetry: $\psi(\mathbf{k}) \rightarrow e^{i\theta \mathbf{n} \cdot \mathbf{s}/2} \psi(\mathbf{k})$, where $\mathbf{n}$ is an arbitrary unit 3-vector. The system similarly preserves the familiar $U(1)_c$ phase rotation symmetry associated with charge conservation, $\psi(\mathbf{k}) \rightarrow e^{i\theta} \psi(\mathbf{k})$. These two standard symmetries are further augmented in bilayer graphene by the preservation of particle number individually within each valley, which follows from the so-called $U(1)_v$ valley symmetry; its action takes the form $\psi(\mathbf{k}) \rightarrow e^{i\theta \tau^z} \psi(\mathbf{k})$, where τ^z is a Pauli matrix acting on the valley indices of $\psi(\mathbf{k})$. In essence, the $U(1)_v$ valley symmetry is a manifestation of the low energy scales at work: extrinsic scattering between states originating from valley $\mathbf{K}$ to those originating from valley $\mathbf{K}'$ are necessarily short range and thus precluded by the high quality of the sample.

Further inspection of the Hamiltonian $H_0 + H_C$ reveals that the physical symmetry group, $U(1)_c \times U(1)_v \times SU(2)_s$, is in fact a subgroup of a much larger *effective* symmetry operative at the dominant energy scales of the system. In particular, the Hamiltonian is invariant under independent *spin* rotations within each valley: $\psi(\mathbf{k}) \rightarrow (\mathcal{P}_+ e^{i\theta_+ \mathbf{n}_+ \cdot \mathbf{s}/2} + \mathcal{P}_- e^{i\theta_- \mathbf{n}_- \cdot \mathbf{s}/2}) \psi(\mathbf{k})$, where $\mathbf{n}_\pm$ are unit 3-vectors and $\mathcal{P}_\pm$ project onto the valley $\mathbf{K}$ and $\mathbf{K}'$. Together with the two $U(1)$ symmetry groups, the result is the existence of a $U(2)_K \times U(2)_{K'} \cong U(1)_c \times U(1)_v \times SU(2)_K \times SU(2)_{K'}$ flavour symmetry. Importantly, this result implies that any degeneracies encoded by the $U(2)_K \times U(2)_{K'}$ effective symmetry are split only at the scale of the Hund's coupling, $\epsilon_H \sim (a/d)\epsilon_c$.

The introduction of SOC naturally reduces both the effective and physical symmetry groups. The Ising term by itself ($\lambda_R = 0$) reduces the $SU(2)_s$ to $U(1)_z$, the group generating rotations about the spin- z axis, and the remaining physical symmetry group is thus $U(1)_c \times U(1)_v \times U(1)_z$. The large effective symmetry group relevant to scales larger than ϵ_H is similarly diminished by the restriction that only spin- z rotations in either valley leave Hamiltonian unmodified. The result is an effective symmetry group composed of four different $U(1)$ rotations: $U(1)_c \times U(1)_v \times U(1)_{z,K} \times U(1)_{z,K'}$, where $U(1)_{z,K^{(\prime)}}$ rotates the spin of the valley $\mathbf{K}^{(\prime)}$ fermions about the z -axis.

When Rashba spin-orbit coupling is present, with or without Ising SOC, all global, continuous spin rotations are absent. Both the physical and effective flavour symmetry groups are pared down to $U(1)_c \times U(1)_v$. The small upper bound for the Rashba coupling λ_R imposed by experiment leads us to largely neglect its symmetry breaking effect.

The Hamiltonian also possesses a number of discrete symmetries, the most important of which is time reversal symmetry (TRS):

$$\mathcal{T} : \quad \psi(\mathbf{k}) \rightarrow i\tau^x s^y \psi(-\mathbf{k}), \quad i \rightarrow -i. \quad (7)$$

Time reversal remains a good symmetry of the system both with and without spin-orbit coupling.

4 Mean field approximation

We study the interacting theory using mean field theory. In particular, for each filling ν , where ν is the number of carriers per unit cell as measured relative to charge neutrality, we find the Slater determinant ground state $|\phi_\nu\rangle$ that minimize the mean-field ground state energy $E_{\text{MF}}^{(\nu)}[\phi] = \langle\phi_\nu| (H_0 + H_{\text{SOC}} + H_C) |\phi_\nu\rangle$. This procedure is essentially equivalent to replacing the interacting Hamiltonian with the one-particle mean field Hamiltonian

$$H_{\text{MF}}^{(\nu)} = \sum_{\mathbf{k}} \psi^\dagger(\mathbf{k}) h_{\text{MF}} \psi(\mathbf{k}), \quad h_{\text{MF}}^{(\nu)} = -\frac{U_C}{N_{\text{site}}} \sum_{\mathbf{q}} \left[P^{(\nu)}(\mathbf{q}) - \text{tr}(P^{(\nu)}(\mathbf{q})) \mathbb{1} \right], \quad (8)$$

where the projector $P(\mathbf{k})$ is given by

$$P_{\alpha\beta}^{(\nu)}(\mathbf{q}) = \langle \psi_\beta^\dagger(\mathbf{q}) \psi_\alpha(\mathbf{q}) \rangle_\nu = \langle \psi_\beta^\dagger(\mathbf{q}) \psi_\alpha(\mathbf{q}) \rangle - \langle \psi_\beta^\dagger(\mathbf{q}) \psi_\alpha(\mathbf{q}) \rangle_{\text{CNP}}. \quad (9)$$

Here, the values of the correlation functions $\langle \cdot \rangle$ are in turn obtained by diagonalizing $H_0 + H_{\text{SOC}} + H_{\text{MF}}^{(\nu)}$, and the subscript ‘‘CNP’’ indicates that the expectation value is being taken with respect to the charge neutrality point. Self-consistency is attained when the mean field term $H_{\text{MF}}^{(\nu)}$ used to calculate $P^{(\nu)}$ is in turn defined via Eq. (8). It can be shown that the ground state of this self-consistent Hamiltonian is a local minimum of the mean field energy functional $E_{\text{MF}}^{(\nu)}$.

We solve for $P^{(\nu)}$ through the following procedure. We select an initial value $H_{\text{MF}}^{\text{init.}}$ and then iterate between Eqs. (8) and (9) until self-consistency is reached. Crucially, states possessing less symmetry than the initial Hamiltonian $H_0 + H_{\text{SOC}} + H_{\text{MF}}^{\text{init.}}$ are inaccessible. For instance, if the initial mean field Hamiltonian is invariant under the $U(1)_v$ symmetry, the final wavefunction $|\phi_\nu\rangle$ (and the corresponding $P^{(\nu)}$) must also be invariant under the $U(1)_v$ symmetry and hence so must $H_{\text{MF}}^{(\nu)}$. As noted, the symmetries allow us to separate the problem into those that preserve the $U(1)_v$ and those that break it.

In principle, the result should be the minimal energy state that respects the same symmetries as $H_{\text{MF}}^{\text{init.}}$ and the non-interacting terms, $H_0 + H_{\text{SOC}}$. However, in practice, the algorithm sketched above sometimes finds itself trapped in local minima, unable to attain the true ground state within that symmetry class. This happenstance is particularly common when there are many nearly degenerate ground states, which, as we describe in the following section, is the case here. We have not rigorously explored the phase diagram to ensure that all of the solutions presented below represent true symmetry-class ground states largely because the simplicity of the model makes it more appropriate for a qualitative study of trends, as opposed to a quantitative one. There is therefore little reason to ignore low energy states in favour of what, according to this model, is the ‘‘true’’ ground state. In fact, the phenomenological arguments we make below in SI, section 7 imply that a different ground state is realized than suggested by our simulations.

We are primarily interested in what happens upon hole doping the system in the presence of a positive displacement field, and we therefore specialize to this scenario; our discussion can readily

be translated to the case with opposite D -field sign as well as with electron doping. We further note that provided the D -induced gap at charge neutrality is sufficiently large, we do not expect H_C to induce significant mixing between the four sets of (effectively) degenerate spin-valley bands defined by $H_0 + H_{\text{SOC}}$, allowing us to restrict our focus entirely to the active bands of interest.

While the mean field Hamiltonian $h_{\text{MF}}^{(\nu)}$ is independent of momentum, it nevertheless acts on the original 16 degrees of freedom as opposed to the four bands of interest. It is convenient to distill the resulting Hamiltonian to the only degrees of freedom that remain upon projecting to the bands of interest. In particular, instead of directly discussing $h_{\text{MF}}^{(\nu)}$, we focus instead on

$$h_{\text{MF}}^{(\nu)'} = \sum_{\substack{a,i=0,x,y,z \\ (a,i) \neq (0,0)}} t_{ai} \tau^a s^i, \quad t_{ai} = \frac{1}{4} \text{tr}(h_{\text{MF}}^{(\nu)} \tau^a s^i), \quad (10)$$

where we do not include the constant shift of the chemical potential. Notably, $h_{\text{MF}}^{(\nu)'}$ is still a 16×16 matrix, but with any dependence on either the layer or sublattice removed. It follows that $h_{\text{MF}}^{(\nu)'}$ does not account for some of the spatial dependence that results when one projects onto the four hole bands close to charge neutrality. These effects, while present in our numerics, are largely irrelevant for the purpose of understanding the resulting phases.

5 Polarized phases

We are most interested here in the spontaneous breaking of the spin-valley symmetries, resulting in the polarized states seen in experiment. The propensity for this type of symmetry breaking follows from the large density of states induced by the displacement field. Interactions make having a large density of states at the Fermi energy energetically costly. At the expense of the kinetic energy, the DOS at the Fermi energy and its associated energy cost may be lowered by breaking the flavour symmetry and alternately increasing and decreasing the filling of certain flavours. The advantage of this process is roughly encapsulated in the Stoner criterion, which states that polarization occurs when $V\rho \geq 1$, where V is the interaction scale and ρ the density of states.

At the mean field level, the polarized phases are characterized by the (simplified) mean field Hamiltonian $h_{\text{MF}}^{(\nu)'}$ of Eq. (10). We begin by addressing the phases in the absence of SOC where the effective $U(2)_K \times U(2)_{K'}$ symmetry remains a good approximation. In the simplest case, only a single t_{ai} in Eq. (10) is non-zero:

$$h_{\text{MF}}^{(\nu)'} = t_{ai} \tau^a s^i. \quad (11)$$

This mean field term pushes two flavours up and two flavours down in energy, resulting in a set of minority and a set of majority Fermi pockets. In what follows, this type of phase is denoted “singly polarized.” Such singly polarized phases are not limited by mean field Hamiltonians of the form Eq. (11), but are more generally induced by any $h_{\text{MF}}^{(\nu)'}$ given by a sum of *anticommuting* matrices $\tau^a s^i$.

We group the singly polarized phases resulting from Eq. (11) into two broad categories. First are the “simple” polarized states that preserve the $U(1)_v$ valley symmetry, implying that the mean field Hamiltonian associated with such states satisfies $[\tau^z, h_{\text{MF}}^{(\nu)'}] = 0$:

$$\tilde{h}_{\text{MF}}^{(\nu)} \propto \tau^a s^i \quad \text{where} \quad \tau^a s^i \in \{s^{x,y,z}, \tau^z s^{0,x,y,z}\}. \quad (12)$$

Notably, the $h_{\text{MF}}^{(\nu)'}$ above commutes with the non-interacting Hamiltonian $h_0(\mathbf{k})$, and it follows that its primary effect is to generate a relative shift of the band energies. The action of $U(2)_K \times U(2)_{K'}$ rotates the orders leading to spin polarization (SP) ($h_{\text{MF}}^{(\nu)'} \propto s^{x,y,z}$) and spin-valley polarization (SVP) ($h_{\text{MF}}^{(\nu)'} \propto \tau^z s^{x,y,z}$) into each other, hence these states must have the same energy (with respect to the Hamiltonian under consideration currently). Similarly, the action of $U(2)_K \times U(2)_{K'}$ may also rotate $h_{\text{MF}}^{(\nu)'}$ to a linear combination of these order parameters, for instance to induce spin polarization along an arbitrary direction. By contrast, the valley polarized (VP) state characterized by $h_{\text{MF}}^{(\nu)'} \propto \tau^z$ does not break $U(2)_K \times U(2)_{K'}$ (although it does break time reversal), and it is therefore not necessarily degenerate with the SP and SVP states. However, the density-density form of the Coulomb interaction renders this distinction meaningless and prevents the system from distinguishing whether valley K or K' is filled on average. Additional interaction terms—say arising from short-range interactions—will split this accidental degeneracy. For instance, the phonon interaction^{S15} takes the form $\sim \int_{\mathbf{r}} (\psi^\dagger \tau^z \psi)^2$ and thus both preserves the $U(2)_K \times U(2)_{K'}$ symmetry while clearly distinguishing between VP and SP/SVP states. (The Hund’s term whose inclusion does decrease the effective symmetry group also distinguishes these two sets of states.)

The second category of states breaks the $U(2)_K \times U(2)_{K'}$ to a *diagonal* subgroup through the spontaneous generation of inter-valley tunnelling. These “inter-valley coherent” (IVC) ordered states occur when $[\tau^z, h_{\text{MF}}^{(\nu)'}] \neq 0$:

$$\tilde{h}_{\text{MF}}^{(\nu)} \propto \tau^a s^i \quad \text{where} \quad \tau^a s^i \in \{\tau^x s^{0,x,y,z}, \tau^y s^{0,x,y,z}\}. \quad (13)$$

As with the SP and SVP states, the IVC order parameters may all be mapped to one another through the action of $U(2)_K \times U(2)_{K'}$, meaning that they must be degenerate. Unlike the previous set of states, the IVC mean field Hamiltonian mixes states from different valleys and therefore significantly alters the form of the band structure.

In addition the singly polarized states, the system may also favour breaking more than a single symmetry, resulting in a “multiply polarized” state. In this case, $h_{\text{MF}}^{(\nu)'}$ is a sum of multiple *commuting* $\tau^a s^i$ matrices. An example of such a mean field term is

$$h_{\text{MF}}^{(\nu)'} = t_0 \tau^z + t_1 s^z + t_3 \tau^z s^z. \quad (14)$$

When $|t_1| = |t_2| = |t_3|$, this mean field Hamiltonian pushes one flavour to a higher or lower energy on average, leaving the remaining three degenerate. More commonly, however, we find that the coefficients satisfy $|t_1| > |t_2| \cong |t_3|$. As with the “singly polarized” state above, the multiply polarized states may also be categorized depending on whether they break or preserve $U(1)_v$.

We calculated the self-consistent mean field Hamiltonians and corresponding ground states in the absence of SOC for a variety of parameters. For filling ranges that prefer singly polarized states, we consistently find IVC ordered states to have the lowest energies. In this case, no other symmetries are broken. Similarly, for filling ranges preferring multiply polarized states, IVC order is typically also generated, although this time it must be present alongside another symmetry breaking order. We stress, however, that our model is very crude and is not expected to yield quantitatively accurate results.

We finally turn to the case of primary interest: bilayer graphene with proximity-induced SOC. Given the relative smallness of the effective Rashba coupling in the parameter range of interest, we focus on a system with only Ising SOC; modifications brought by the reintroduction of Rashba are briefly addressed below. In this case, the $U(2)_K \times U(2)_{K'}$ is reduced to a $U(1)_c \times U(1)_v \times U(1)_{z,K} \times U(1)_{z,K'}$, where $U(1)_c$ and $U(1)_v$ denote the charge and valley symmetries while $U(1)_{z,K^{(\prime)}}$ represents spin rotations about the z axis in either valley. It naturally follows that the $SU(2)_s$ spin degeneracies present in the absence of SOC are lifted. In fact, because the Ising SOC term resembles a self-generated “spin-valley order”, $\tau^z s^z$, its presence results in a ‘singly polarized’ state even in the non-interacting limit. (As discussed in SI, section 1 and shown in Extended Data Fig. 6a, the splitting induced by the bare Ising coupling is substantially smaller than what is required to explain the phases seen in experiment.) Clearly, *if* the SP or SVP phases were the preferred ground state without SOC, the mean field Hamiltonian generated in the presence of SOC would have a clear energetic preference for Ising-like SVP polarized states. Indeed, we consistently find that the effective Ising SOC is enhanced by the interactions.

The introduction of Rashba SOC breaks the $SU(2)$ spin symmetries operative in either valley, leaving only a $U(1)_c \times U(1)_v$ flavour symmetry. Although it is included below, it has relatively little qualitative effect on the resulting phase diagrams.

In Extended Data Fig. 6c,e we present the normalized frequencies expected in quantum oscillations and the corresponding Fermi surfaces obtained through simulations performed with $\lambda_I = 0.7$ meV and $\lambda_R = 3$ meV. Before describing these results in detail, we emphasize that c and e were both simulated using a single choice of $H_{MF}^{init.}$. As described in SI, section 4, although all phases represented in Extended Data Fig. 6c,e are *low energy* states, it is possible that our algorithm has not found the *true* ground state of the model. Since the simplicity of the model prevents us from making quantitative predictions based on its behaviour, we do not view this as a particularly significant failing of the simulations. Nevertheless, we have verified by doing multiple runs with different starting positions that the phases given in Extended Data Fig. 6c,e are not flukes of our specific choice of $H_{MF}^{init.}$ but are instead overall representative of the different low energy states present.

The plot in Extended Data Fig. 6c was obtained in the absence of $U(1)_v$ breaking (as explained in SI, section 4, the presence of $U(1)_v$ is enforced by our choice of $H_{MF}^{init.}$). Comparing with Extended Data Fig. 6a, it is clear even for low dopings, in the $FP(6,6)_+$ phase, that the splitting between

the two sets of pockets is larger when SOC is present: the simulation that included interactions finds minority Fermi surfaces that are even smaller relative to the majority pockets than what is seen without interactions. Further doping sees a first order transition at around $|n| \sim 5 \times 10^{11} \text{ cm}^{-2}$ where the ground state discontinuously jumps to an $\text{FP}(1, 3, 6)_+$ state, whose background is coloured yellow. Here, we see that the Fermi surfaces of the two majority flavours differ not only from the two minority flavours, but they also differ from one another. The $\text{FP}(1, 3, 6)_+$ state is therefore multiply polarized: in addition to the Ising polarization, an additional symmetry breaking order was generated (here, a mixture of spin s^z and valley τ^z polarization). When the doping reaches $|n| \sim 5.8 \times 10^{11} \text{ cm}^{-2}$, another first order transition occurs, yielding a singly polarized state with two large and six small Fermi pockets, $\text{FP}(2, 6)_+$ (shown in red). Its development simply follows from a large enhancement of the effective λ_I ; no additional symmetries are broken. Another transition occurs at $|n| \sim 8.8 \times 10^{11} \text{ cm}^{-2}$, into the $\text{FP}(3, 3)_+$ phase, where three of the flavours have large Fermi surfaces and one of the flavours has three small surfaces. Again, this state is multiply polarized, thus requiring additional symmetry breaking.

Comparing the theory simulation of Extended Data Fig. 6 against the experimental data, it is tempting to associate the $\text{FP}(2, 6)_+$ phase found here with the experimentally observed $\text{FP}(2, 2)_+$ phase that serves as a parent to superconductivity, despite the difference in the number of small Fermi pockets. The latter discrepancy may be justified through the subdominant inclusion of rotational symmetry breaking, which could spontaneously reduce the number of filled small pockets from six to two (we address this process in more detail in the next section as well as in Extended Data Fig. 6d,f). However, as mentioned, the large enhancement of λ_I required to obtain this phase is at odds with the observed in-plane magnetic field dependence¹, making this type of Ising-dominated polarized phase an unlikely candidate. The quantum oscillations characterizing the $\text{FP}(3, 3)_+$ phase is also reminiscent of the large-doping regime adjacent to the superconducting $\text{FP}(2, 2)_+$ phase. In particular, the downward sloping frequency around $\sim 1/3$ is also present in Fig. 3c and Extended Data Fig. 8 (Our assertion that the Ising polarized phase $\text{FP}(2, 6)_+$ is unlikely present experimentally does not rule out the experimental relevance of $\text{FP}(3, 3)_+$).

Extended Data Fig. 6e shows the quantum oscillation frequencies and Fermi surfaces for a mean field solution defined with the same parameters as in c, but whose initialization condition allowed IVC order to develop. Setting aside technicalities surrounding the self-consistent mean field procedure, we emphasize that IVC should technically only develop when it is energetically favourable to do so. Unsurprisingly, the solution at low dopings is identical to what is shown in c, with only Ising SOC present. A multiply polarized phase coloured in yellow, $\text{FP}(1, 6)_+$, is attained around $|n| \sim 3.8 \times 10^{11} \text{ cm}^{-2}$, and the Fermi surface shape makes the difference between this solution and the one in c apparent. While the two minority pockets shown in the inset resemble those found in the non-interacting and $U(1)_v$ -preserving cases (Extended Data Fig. 6a,c), the large pocket is quite different—a direct consequence of the inter-valley hybridization. A complicated series of intermediate phases existing only within a narrow filling range follows with additional doping before the

¹One may argue that if Rashba SOC were also enhanced by interactions, the Pauli-limit violation ratio could remain unchanged. However, we see no evidence in our calculations of any Rashba enhancement.

system enters a singly polarized $\text{FP}(2)_+$ phase at $|n| \sim 6 \times 10^{11} \text{ cm}^{-2}$. Two large, star-shaped Fermi surfaces that clearly do not resemble those found in the interaction-free band structure are present, yet again as a direct consequence of IVC order; we colour this region in blue to distinguish it from the red singly polarized phases without IVC order. Despite its singly polarized nature, the IVC order responsible for the star-shaped Fermi surface is generated alongside an enhancement of the Ising order:

$$h_{\text{MF}}^{(\nu)'} = \frac{\Delta_{\text{IVC}}}{2} \tau^x + \frac{\delta \lambda_I}{2} \tau^z s^z. \quad (15)$$

Importantly, the state remains singly polarized because τ^x and $\tau^z s^z$ anticommute: only a single gap is opened by the mean field potential. Further doping leads first to a $\text{U}(1)_v$ -breaking multiply polarized state, then to a $\text{U}(1)_v$ -preserving polarized state analogous to what is realized at the same filling range in Extended Data Fig. 6c, and then finally to another IVC-ordered multiply polarized state.

Unlike the SOC-free model, where IVC order was always found to have the lowest energy, the addition of Ising SOC has made the SVP phase competitive against the IVC—even when IVC order was allowed to develop, there is a least one region in Extended Data Fig. 6e where the Ising SVP state is preferred. In fact, the effective Ising coupling is substantially enhanced both in the singly polarized state without IVC order in Extended Data Fig. 6c and with IVC order in Extended Data Fig. 6e—more than is strictly compatible with the in-plane field measurements of the superconducting state. However, importantly, the latter state has spontaneously broken an additional symmetry relative to the Ising SOC-induced SVP order, establishing it as a distinct phase. Within this IVC-ordered state, we expect the relative magnitudes of the effective Ising SOC term and the IVC order Δ_{IVC} is a matter of details—precisely the quantitative information our model is unable to provide reliably.

6 Nematicity

In addition to the internal flavour symmetries of the continuum model, the system also possess a C_3 symmetry that rotates the system by 120° . This transformation acts on the spinors ψ_ξ as

$$C_3 : \quad \psi_\xi(\mathbf{k}) \rightarrow e^{i2\pi\xi\sigma^z/3} \psi_\xi(R_3\mathbf{k}), \quad R_3 = \begin{pmatrix} -1/2 & \sqrt{3}/2 \\ -\sqrt{3}/2 & -1/2 \end{pmatrix}. \quad (16)$$

As alluded to in the discussion of Extended Data Fig. 6c, the experimental observation of the $\text{FP}(2, 2)_+$ state is consistent with a spontaneous breaking of this rotational symmetry, i.e., nematicity. The development of nematic order had been predicted in this system through a momentum-condensation-like Pomeranchuk instabilities^{S16–S18}. Reference S17 argues that the development of the nematic order is subdominant to the polarizing energy scale. That is, the internal flavour symmetries are first broken in the manner described in the previous section, and the electrons subsequently choose to occupy one out of the three small pockets instead of occupying all three

pockets equally. The momentum space dependence of the Coulomb interaction, which our simulations ignore, plays a crucial role in the derivation of this effect, and our model is therefore unable to self-consistently prefer the formation of the nematic order.

To compensate for the lack of spontaneous nematic order, we instead explicitly break the C_3 symmetry by modifying the non-interacting portion of the Hamiltonian. In particular, we replace $h_{0,\xi}(\mathbf{k})$ in Eq. (1) with $h_{0,\xi}(\mathbf{k}) + \delta h_{\text{nem}}$ where

$$\delta h_{\text{nem}} = \begin{pmatrix} 0 & 0 & \alpha_1 + \alpha_2 & \alpha_3 \\ 0 & 0 & 0 & -\alpha_1 + \alpha_2 \\ \alpha_1 + \alpha_2 & 0 & 0 & 0 \\ \alpha_3 & -\alpha_1 + \alpha_2 & 0 & 0 \end{pmatrix}. \quad (17)$$

We otherwise implement the identical procedure to the one described in the previous section, with the results shown in Extended Data Fig. 6d and f.

The simulations responsible for Extended Data Fig. 6d are the analogue to those of Extended Data Fig. 6c in that the $U(1)_v$ symmetry was not allowed to break spontaneously. Unsurprisingly, the explicit breaking of the C_3 symmetry makes the resulting quantum oscillation frequencies and Fermi surface structures more complicated than those of shown in Extended Data Fig. 6c. Again, the low doping regime is characterized by an enhancement of the effective Ising SOC compared to the non-interacting theory, resulting in the $\text{FP}(2, 2, 4)_+$ phase shown. Around $|n| \sim 3.2 \times 10^{11} \text{ cm}^{-2}$, the system transitions to a multiply polarized phase $\text{FP}(1, 1, 2, 2)_+$. This phase undergoes a Lifshitz transition that does not change the polarizing order at $|n| \sim 5 \times 10^{11} \text{ cm}^{-2}$, after which it evolves continuously into a singly ordered $\text{FP}(2, 2)_+$ phase at $|n| \sim 5.5 \times 10^{11} \text{ cm}^{-2}$. This $\text{FP}(2, 2)_+$ phase and its higher doping partner $\text{FP}(2, 2, 4)_+$ phase are the analogues of the $\text{FP}(2, 6)_+$ phase in Extended Data Fig. 6c in accordance with the discussion of the previous section: these singly polarized states do not arise out of an interaction-induced spontaneous breaking of a symmetry, but simply out of the enhancement of the Ising SOC induced SVP order (the enhancement, however, is so large relative to the scale of the Ising SOC parameter appearing in the non-interacting Hamiltonian that it is still reasonable to identify $\text{FP}(2, 2)_+$ and $\text{FP}(2, 2, 4)_+$ as polarized states). As in that section, however, we are forced to conclude that the $\text{FP}(2, 2)_+$ phase here is not compatible with the in-plane field measurements since such an extreme increase in the effective Ising SOC would imply a far greater Pauli-limit violation than observed experimentally.

The plot in Extended Data Fig. 6f illustrates a set of solutions in which the ground state was allowed to develop IVC order. The low doping regime is identical to d, but the multiply polarized phase the system transitions into around $|n| \sim 3.2 \times 10^{11} \text{ cm}^{-2}$ differs: the star-like shape of the Fermi surface in the $\text{FP}(1, 2)_+$ phase clearly indicates that IVC order is present. After some minor changes in the Fermi surface topology, a first order transition to a singly polarized phase, $\widehat{\text{FP}}(2, 2)_+$, occurs at around $|n| \sim 4.6 \times 10^{11} \text{ cm}^{-2}$. This phase once more follows from the enhancement of the effective Ising coupling as opposed to the spontaneous breaking of an additional symmetry, and is thus partner to the $\text{FP}(2, 2)_+$ phase in d. We note that the $\widehat{\text{FP}}(2, 2)_+$ in f appears

at lower fillings than the same phase appears in d. Since all phases realized in d can also be realized in f, we would normally expect any phase lacking IVC in f to be represented in d at those same filling. This discrepancy is related to the discussion of SI, section 4 on how the algorithm may find local minima instead of true minima when many states with similar energies are present. The existence of this near-degenerate manifold of mean field states is apparent upon further doping, which sees the simulation alternate between $U(1)_v$ symmetric (coloured red) and $U(1)_v$ breaking (coloured blue) singly polarized ground states multiple times, up until $|n| \sim 8 \times 10^{11} \text{ cm}^{-2}$ where the system becomes multiply polarized. Importantly, from within the IVC ordered $\text{FP}(2)_+$ state, a first order transition to a different IVC-ordered state possessing two large and two small Fermi surfaces occurs at $|n| \sim 5.5 \times 10^{11} \text{ cm}^{-2}$, reminiscent of the state that gives rise to superconductivity in the experiment.

With regards to the singly polarized states, we make no claims that our model strictly prefers one of these options in the density range shown relative to the other. The primary takeaway message from this discussion is that with nematicity, IVC-ordered $\text{FP}(2, 2)_+$ *can be* realized in the system.

7 Ising-SOC-mediated ground state selection

These experiments prompt important questions regarding the role of WSe_2 and the concomitant spin-orbit coupling in promoting superconductivity: what does the SOC change so that bilayer graphene is able to superconduct at zero field? The persistence of superconductivity across the entirety of the $\text{FP}(2, 2)_+$ phase suggests the realization of this ground state as the key to the development of superconductivity. We propose that in the absence of SOC, interactions favour a ground state that is inhospitable to zero-field superconductivity; with the addition of SOC, a distinct ground state amenable to superconductivity is selected instead. Given the relatively small effect of Rashba spin orbit on the band structure, we completely ignore its influence on the interacting ground state selection for this discussion, focusing instead on the effects of Ising SOC.

We first recall that Ising SOC itself splits the flavour degeneracy, resulting in a non-interacting singly polarized SVP phase even at the level of the band structure. We therefore infer that *if* the SOC-free $\text{FP}(2, 2)_+$ ground state was an SVP polarized state, the addition of Ising SOC would not alter the ground state. An SVP SOC-free ground state is therefore unlikely.

A natural next proposition is that the $\text{FP}(2, 2)_+$ phase seen experimentally in this paper *is* precisely an Ising SVP state: the Coulomb interaction serves to increase the magnitude of SOC-induced band splitting, but otherwise induces no spontaneous symmetry breaking. The numerically obtained phases shaded in red in Extended Data Fig. 6c-d are all examples of such states. Importantly, these states result from a large enhancement of the Ising coupling by interactions and are thus seemingly only consistent with a correspondingly enhanced Pauli-limit violation. While the PVR is reasonably large in the low doping regime of the superconductor, at large dopings, the Pauli limit is barely violated at all. This large variation in behaviour across the superconducting dome may be

generally interpreted in two ways:

1. The nature of the interacting ground state $\text{FP}(2, 2)_+$ remains largely unchanged as a function of doping. The change in PVR instead follows from relatively small band structure effects compared to the interaction scale, such as Rashba SOC or the orbital coupling of an in-plane magnetic field. The specifics of this mechanism are discussed in more detail in the subsequent section.
2. The nature of the interacting ground state changes substantially as a function of a doping.

These two possibilities are of course not mutually exclusive nor even strictly distinct. They nevertheless provide a useful framework for organizing the energy scales and their implications in what follows.

Taking the perspective of scenario (1), we conclude that the $\text{FP}(2, 2)_+$ is incompatible with an Ising-induced SVP ground state. We are thus left with a scenario in which Ising SOC selects a non-SVP phase that in turn hosts superconductivity. Similarly, the same reasoning used to reject the SVP states removes the SP states as potential ground states—the Pauli-limit violation of an SP state would be even larger than expected for an SVP state. Assuming the full $U(2)_K \times U(2)_{K'}$ symmetry, the discussion in SI, section 5 leaves two remaining classes of singly polarized ground states: the VP state ($h_{\text{MF}}^{(\nu)'} \propto \tau^z$) and states with IVC order ($h_{\text{MF}}^{(\nu)'} \propto \tau^{x,y} s^\mu$, $\mu = 0, x, y, z$), the latter set of which may be treated on equal footing at the level of the $U(2)_K \times U(2)_{K'}$ symmetric theory. Notably, the VP state is clearly hostile to the development of superconductivity. Above, we described how time reversal imposes the requirement that the (SOC-free, symmetry-unbroken) band structure energies satisfy $\epsilon_K(\mathbf{k}) = \epsilon_{K'}(-\mathbf{k})$; such resonance conditions constitute a strong prerequisite to the formation of superconductivity. Breaking time reversal symmetry and polarizing the bands according to valley thus precludes the possibility of superconductivity except in certain exotic theoretical scenarios. Conversely, the IVC ordered states present no obvious impediment to superconductivity.

In reality, the SOC-free theory is not invariant under the full $U(2)_K \times U(2)_{K'}$, but instead under $U(1)_c \times U(1)_v \times \text{SU}(2)$. Working from the perspective of the physical $U(1)_c \times U(1)_v \times \text{SU}(2)$ symmetry, the IVC ground state can be grouped into two categories: IVC singlets that break only the $U(1)_v$ symmetry and IVC triplet states that additionally spontaneously break the spin symmetry. The former state is represented by mean field Hamiltonians composed of matrices τ^x and τ^y , whereas the latter triplets case follows from presence of matrices $\tau^x s^{x,y,z}$ and $\tau^y s^{x,y,z}$. Again, the large variation in Pauli violation ratio as a function of filling and our working assumption that the interacting ground state remains largely unmodified across the $\text{FP}(2, 2)_+$ phase (temporarily) disqualifies the triplet orders as viable candidates, leaving the IVC singlet polarized state as the proposed superconducting parent state.

On these phenomenological grounds, provided scenario (1) holds, we conclude that the addition of SOC increases the energy of the VP $\text{FP}(2, 2)_+$ state relative to an IVC singlet ordered state, establishing the latter as the new, SOC-mediated ground state. A simple schematic of the energy

levels as a function of λ_I is shown in Fig. 4f.

Theoretically, the above line of reasoning can be supported on an intuitive level. We start by making some natural assumptions regarding the nature of the interacting theory in this doping regime. We suppose first that interactions dominate the energy scales of the problem and that these interactions necessarily favour the formation of a singly polarized state. As addressed at the end of SI, section 5, the IVC singlet order parameters, e.g., τ^x , anticommutes with the Ising term, $\tau^z s^z$. Hence, a mean field Hamiltonian $h_{\text{MF}}^{(\nu)'} = \Delta_{\text{IVC}} \tau^x / 2$ still results in a singly polarized state even when $h_{\text{SOC}} = \lambda_I \tau^z s^z$ is included. Together, they induce an energetic separation $\sqrt{\Delta_{\text{IVC}}^2 + \lambda_I^2}$ between the flavours. Treating λ_I as a perturbation to the IVC ground state and expanding in $\lambda_I / \Delta_{\text{IVC}}$, it's clear that the change in ground state energy induced by Ising SOC will be suppressed by a factor of IVC order $\Delta E_{\text{MF}}^{\text{IVC}} \sim \lambda_I^2 / \Delta_{\text{IVC}}$. By contrast, the VP mean field Hamiltonian $h_{\text{MF}}^{(\nu)'} = \Delta_{\text{VP}} \tau^z / 2$ commutes with the Ising SOC Hamiltonian $h_{\text{SOC}} = \lambda_I \tau^z s^z$. The introduction of SOC then splits the energy of the hitherto twofold spin degenerate energies in either valley: $\epsilon_{K(\nu)} \rightarrow \epsilon_{K(\nu)} \pm \lambda_I / 2$. Not only is Δ_{VP} unable to suppress the change in mean field ground state energy, but, unlike for the IVC case, the Ising SOC has destroyed the singly polarized nature of the ground state. We infer then that the change in mean field energy brought by Ising SOC will be larger and more positive for the VP state than it will be for the IVC state².

As a proof of concept, we numerically evaluate the effect of an Ising SOC perturbation of the VP and IVC ground state energies. Naturally, we focus on a density regime where singly polarized states are preferred. The first step is then to self-consistently solve for mean field Hamiltonians with VP and IVC order for a system without SOC in the manner described in SI, section 4, obtaining $H_{\text{MF}}^{(\nu), \text{VP}}$ and $H_{\text{MF}}^{(\nu), \text{IVC}}$. The mean field ground states $|\phi_{\nu}^{\text{VP/IVC}}\rangle$ are in turn employed to calculate the SOC-free mean field energies, $E_{\text{MF}}^{\text{VP/IVC}, \nu}(\lambda_I = 0) = \langle \phi_{\nu}^{\text{VP/IVC}} | H_0 + H_C | \phi_{\nu}^{\text{VP/IVC}} \rangle$. We subsequently introduce Ising SOC by adding $H_{\text{SOC}}[\lambda_I]$ to the mean field Hamiltonian, $H_0 + H_{\text{MF}}^{(\nu), \text{VP/IVC}} \rightarrow H_0 + H_{\text{MF}}^{(\nu), \text{VP/IVC}} + H_{\text{SOC}}[\lambda_I]$ and solving for the mean field ground state $|\phi_{\nu}^{\text{VP/IVC}}(\lambda_I)\rangle$. Importantly, the terms $H_{\text{MF}}^{(\nu), \text{VP/IVC}}$ are identical to those obtained for the $\lambda_I = 0$ calculation; this calculation is no longer self-consistent. Finally, the Ising-perturbed energy is then calculated in the same fashion: $E_{\text{MF}}^{\text{VP/IVC}, \nu}(\lambda_I) = \langle \phi_{\nu}^{\text{VP/IVC}}(\lambda_I) | H_0 + H_C + H_{\text{SOC}}[\lambda_I] | \phi_{\nu}^{\text{VP/IVC}}(\lambda_I) \rangle$. We find, as expected, that the addition of SOC *increases* the mean energy for both cases $\Delta E_{\text{MF}}^{\text{VP/IVC}, \nu}(\lambda_I) = E_{\text{MF}}^{\text{VP/IVC}, \nu}(\lambda_I) - E_{\text{MF}}^{\text{VP/IVC}, \nu}(0) > 0$. In Extended Data Fig. 6b the difference between the change in mean field energies between the VP and IVC polarized ground states is plotted as a function λ_I for several fillings. In accordance with our intuition, we find $\Delta E_{\text{MF}}^{\text{IVC}, \nu}(\lambda_I) - \Delta E_{\text{MF}}^{\text{VP}, \nu}(\lambda_I) < 0$, meaning that the energy of the VP ground state increases more with the addition of λ_I than the IVC ground state does³.

²Crucial to this line of reasoning is the assumption that interactions dominate the problem; the argument given above could otherwise be turned around to argue in favour of Ising SOC abetting the development of a VP state. In particular, the ease by which the VP ground state accommodates the addition of Ising SOC makes it more agreeable to the non-interacting Hamiltonian h_{SOC} compared to the IVC ground state.

³As mentioned above, however, our model does find that the IVC ground state is consistently lower energy than

Although much of the logic used above will follow through, scenario (2) is somewhat more subtle. First, although no longer applicable across the full $\text{FP}(2, 2)_+$ phase, the conclusions we reached for scenario (1) *do* still hold when restricting to the high doping regime. Namely, at the large doping end of the superconducting dome, the experiments still point to a scenario in which the addition of Ising SOC pushes the system away from a VP normal state towards an IVC singlet normal state. Further, since an evolving SOC-mediated $\text{FP}(2, 2)_+$ ground state need not imply an evolving SOC-free ground state, we continue in our assumption that a VP state is realized prior to the addition of SOC. The primary distinction between scenarios (1) and (2) is therefore that we can reject neither the SVP nor the IVC triplet phases as candidate ground states at low dopings on the basis of the small PVR at large dopings. Among the potential IVC triplet states, the reasoning provided above for why Ising would prefer an IVC singlet over the VP state similarly selects the z -component IVC triplet, represented by $\tau^{x,y} s^z$, as the most likely candidate. We are therefore left with a possible mixed mean field Hamiltonian of the form

$$h_{\text{MF}}^{(\nu)'} = \frac{\Delta_{\text{IVC}}^{\text{singlet}}(\nu)}{2} \tau^x + \frac{\Delta_{\text{IVC}}^{\text{triplet}}(\nu)}{2} \tau^y s^z + \frac{\delta\lambda_I(\nu)}{2} \tau^z s^z, \quad (18)$$

where we have explicitly indicated the functional dependence of the mean field order parameters on the filling ν . Note that the IVC orders τ^x and $\tau^y s^z$ were chosen such that they anticommute, guaranteeing that (18) describes a singly polarized phase.

We can make some arguments towards the functional form of the mean field parameters $\Delta_{\text{IVC}}^{\text{singlet}}(\nu)$, $\Delta_{\text{IVC}}^{\text{triplet}}(\nu)$, and $\delta\lambda_I(\nu)$. First, even at the very edge of the underdoped superconductor, where the PVR is at its largest, a mean field Hamiltonian in which the IVC single component completely vanishes, $\Delta_{\text{IVC}}^{\text{singlet}}(\nu_{\text{low dopings}}) = 0$, remains unlikely. For instance, in the simulations of Extended Data Fig. 6c,d, IVC order is prohibited and the singly polarized phases (shaded red) are characterized entirely by an interaction-induced increase $\delta\lambda_I \sim 3.5$ meV of the effective Ising SOC. Such an extreme enhancement of the effective Ising would in turn imply a PVR of order ~ 20 . It's therefore likely that $|\Delta_{\text{IVC}}^{\text{singlet}}(\nu)| \geq \sqrt{[\Delta_{\text{IVC}}^{\text{triplet}}(\nu)]^2 + [\lambda_I + \delta\lambda_I(\nu)]^2}$ throughout the $\text{FP}(2, 2)_+$ phase.

We can similarly discuss the relative magnitudes of $\delta\lambda_I$ and $\Delta_{\text{IVC}}^{\text{triplet}}$. On the one hand, the SOC term h_{SOC} very clearly prefers the development of an Ising SVP phase, suggesting that $\delta\lambda_I$ may be the next-largest contribution to $h_{\text{MF}}^{(\nu)'}$ after $\Delta_{\text{IVC}}^{\text{singlet}}$. Such an interplay between the interaction-induced IVC singlet and Ising orders is in fact seen in the numerics presented in Extended Data Fig. 6c,d: $\Delta_{\text{IVC}}^{\text{singlet}}(\nu)$ increases with filling whereas $\delta\lambda_I(\nu)$ decreases⁴. Conversely, the symmetry

the VP ground state.

⁴We acknowledge that our simulations also find an overly-large mean-field enhancement of the Ising SOC even in the presence of IVC. For the C_3 symmetry simulation shown in Extended Data Fig. 6e, the parameters obtained in the low-doping region of the $\text{FP}(2)_+$ phase, $\nu_{\text{low doping}} \sim -6 \times 10^{11} \text{ cm}^{-2}$, are given by $\delta\lambda_I(\nu_{\text{low doping}}) \sim 3$ meV, $\Delta_{\text{IVC}}^{\text{singlet}}(\nu_{\text{low doping}}) \sim 2$ meV. Towards the large doping end of the IVC ordered region, $\nu_{\text{large doping}} \sim -7 \times 10^{11} \text{ cm}^{-2}$, the relative magnitudes are the IVC and Ising mean field parameters are interchanged: $\delta\lambda_I(\nu_{\text{large doping}}) \sim 2.5$ meV, $\Delta_{\text{IVC}}^{\text{singlet}}(\nu_{\text{large doping}}) \sim 2.7$ meV. Hence, although the numerics demonstrate the correct trends, they again fail to quantitatively account for the experimental observations.

arguments above make it very natural for the IVC singlet and IVC z -triplet orders to have very similar energies. In fact, the degeneracy between the two orders, $\tau^{x,y}$ and $\tau^{x,y}s^z$, is only lifted at the level of the valley Hund's interaction and Rashba energy scale. Indeed the introduction of Ising SOC breaks the large symmetry group $U(2)_K \times U(2)_{K'}$ down to $U(1)_c \times U(1)_v \times U(1)_{K,z} \times U(1)_{K',z}$, under whose action the singlet and z -triplet IVC orders are still able mix.

8 Orbital coupling to in-plane magnetic fields

Magnetic fields oriented in the graphene plane also enter the low-energy theory of BLG through orbital effects, i.e., a renormalization of the hopping terms due to the magnetic flux between the two graphene layers. While this effect vanishes for purely two-dimensional monolayer graphene, in BLG the orbital coupling scales linearly with the finite width d between the layers. The leading-order contribution of this type comes from the renormalization of the intralayer nearest-neighbor hopping term γ_0 , which is the largest energy scale in the problem by an order of magnitude. Choosing a gauge that preserves translation invariance in the plane, $\mathbf{A} = d(B_y, -B_x, 0)$ where $d = 0.33$ nm is the interlayer distance, this leads to orbital contributions on layers 1 and 2 given by^{S19}

$$h_{\text{orbital},1} = v_0 (-\tau_z \sigma_x b_y + \sigma_y b_x) \quad (19)$$

$$h_{\text{orbital},2} = v_0 (\tau_z \sigma_x b_y - \sigma_y b_x) \quad (20)$$

where $b_j = edB_j/2\hbar$ and $j = x, y$. The system thus has an orbital magnetic moment given (to leading order) by

$$\mu_{\text{orbital}} = \frac{v_0 ed}{2\hbar} = \frac{\sqrt{3}\gamma_0 a_0 ed}{4\hbar} \approx 0.14 \frac{\text{meV}}{\text{Tesla}}. \quad (21)$$

This scale nominally yields a stronger coupling than the spin Zeeman term $h_Z = \mu_B \mathbf{B} \cdot \mathbf{s}$ with the Bohr magneton $\mu_B \sim 0.06$ meV/Tesla. However, orbital effects will be suppressed near the $\mathbf{K}, \mathbf{K}'$ points because of the strong sublattice polarization of the low-energy wavefunctions—analogue to the suppression of the “bare” Rashba SOC discussed above.

Projecting down to the 2×2 low-energy subspace spanned by the $A1$ and $B2$ sites enables estimation of the effective orbital coupling, given to leading order as^{S19}

$$h_{\text{orbital}}^\xi(\mathbf{k}) = \frac{2v_0^2}{\gamma_1^2} u(\mathbf{k} \times \mathbf{b})_z \sigma_0 s_0 = \mu_{\text{orbital}}^{\text{eff}} B_x \sigma_0 s_0, \quad (22)$$

where $\mathbf{k}$ is the momentum measured from either of the Dirac points $\mathbf{K}, \mathbf{K}'$. We picked the in-plane field in the x direction and defined the effective orbital magnetic moment as

$$\mu_{\text{orbital}}^{\text{eff}} = -\frac{3a_0 ed \gamma_0^2}{4\hbar \gamma_1^2} u(k_y a_0). \quad (23)$$

For $u \approx -80$ meV (corresponding to positive $D/\epsilon_0 \approx 1$ V/nm) one finds $\mu_{\text{orbital}}^{\text{eff}} \sim 0.4 \frac{\text{meV}}{\text{Tesla}} (k_y a_0)$, which for Fermi momenta satisfying $k_y a_0 = k_F a_0 \sim 0.05$ near the center of the small pockets gives $\mu_{\text{orbital}}^{\text{eff}} \sim 0.02 \frac{\text{meV}}{\text{Tesla}}$, or $\mu_{\text{orbital}}^{\text{eff}} = 0.35 \mu_B$. In the following discussion we will use the dimensionless quantity $g_0 k_F$ to denote the strength of the orbital coupling, defined as $\mu_{\text{orbital}}^{\text{eff}} = g_0 k_F \mu_B$.

9 BCS mean-field analysis in the presence of Zeeman field, SOC terms and orbital coupling

In this section, we describe the model used to investigate the evolution of the Pauli-limit violation ratio in our sample, as well as its analytical solution. We follow the treatment first developed in Ref. S20 to compute the response to a Zeeman field of non-centrosymmetric superconductors with Rashba SOC, later generalized in Refs. S21, S22 to systems with a coexistence of Ising and Rashba SOC. We also incorporate the effect of orbital depairing in a simple model for BLG inspired by Ref. S19 and the discussion in SI, section 8.

We assume that as a result of a symmetry-breaking transition (cascade), the system is in an $\text{FP}(2, 2)_+$ phase with two large and two small hole pockets. We model the small pockets by two electronic bands centered around trigonal-warping loci $\pm \mathbf{T}$ that respectively originate from the K and K' valleys. In other words, we imagine a scenario where exchange interaction effects promote nematic order such that electrons in each valley “flock” from evenly occupying the three small pockets to completely polarizing one pocket^{S17}—and further that the selected pockets in the two valleys are time-reversed partners of each other. We note that the quantum oscillation data do not directly reveal nematicity, but do indicate the presence of only two small Fermi pockets (instead of the 6 pockets predicted by the non-interacting band structure). The following modelling could also apply to a situation where the transition to the $\text{FP}(2, 2)_+$ state is not nematic, provided that time-reversal symmetry $\mathcal{T}$, which relates the two remaining small pockets, is preserved.

Adopting the preceding scenario, we take the normal-state Hamiltonian to be

$$H(\xi \mathbf{T} + \mathbf{k}) = \xi_{\mathbf{k}} + \frac{1}{2} \xi g_I s^z + \frac{1}{2} g_R (\mathbf{s} \times \mathbf{k}) \cdot \mathbf{z} + \mathbf{b} \cdot \mathbf{s} + g_{\text{orb}} (\mathbf{b} \times (\mathbf{k} + \mathbf{k}_0)) \cdot \mathbf{z}, \quad (24)$$

with $\xi = \pm 1$ the valley index, $\mathbf{T}$ the momentum of one of the trigonal-warping loci, and $\mathbf{s} = (s^x, s^y, s^z)$ a vector of Pauli matrices that act on the spin degree of freedom. On the right side $\xi_{\mathbf{k}}$ is the spin-orbit-free normal state band structure, which we linearize near the Fermi surface as $\xi_{\mathbf{k}} \approx v_F(k - k_F)$ (k_F denotes the Fermi momentum measured from the center $\mathbf{T}$ of the pocket). The next two terms incorporate Ising and Rashba SOC with strengths g_I and g_R , respectively. We highlight that the effective model of Eq. (24) features a form of Rashba SOC that is linear in momentum, instead of quadratic^{S12}, due to trigonal warping terms that split the quadratic band touching at zero D field into four Dirac cones. The final two terms incorporate effects of an in-plane magnetic field $B_{\parallel}$, packaged into a vector $\mathbf{b} = (\mu_B B_{\parallel}, 0, 0)$ with μ_B the Bohr magneton: $\mathbf{b} \cdot \mathbf{s}$ is simply the Zeeman energy while g_{orb} captures orbital effects^{S19} of the in-plane field. The momentum shift $\mathbf{k}_0 = \mathbf{T} - \mathbf{K}$ captures the fact that the relevant momenta for orbital effects of in-plane fields are measured with respect to the Dirac points rather than the center $\mathbf{T}$ of the small pockets.

We then consider a local (momentum-independent) spin-singlet pairing term that gives rise to superconductivity with a critical temperature T_c^0 at zero magnetic field. In the presence of an in-plane magnetic field $B_{\parallel}$ the superconductivity is weakened through a combination of spin and

orbital effects, with $T_c < T_c^0$ given by the solution of a self-consistent gap equation, linearized near the second-order transition at T_c where the pairing amplitude $\Delta \rightarrow 0$,^{S20-S24}

$$\ln \left(\frac{T_c}{T_c^0} \right) = \frac{T_c}{2} \sum_{\omega_n} \left(\left\langle \int d\xi_{\mathbf{k}} \text{Tr} \{ s_y G_0(\mathbf{T} + \mathbf{k}, i\omega_n) s_y G_0^*(-\mathbf{T} - \mathbf{k}, i\omega_n) \} \right\rangle_{\text{FS}} - \frac{\pi}{|\omega_n|} \right). \quad (25)$$

Here $\sum_{\omega_n}$ denotes a summation over Matsubara frequencies $i\omega_n$, $\langle \cdots \rangle_{\text{FS}}$ denotes a Fermi surface average and $\text{Tr}\{\cdots\}$ is a trace over spin Pauli matrices, and $G_0(\mathbf{T} + \mathbf{k}, i\omega_n)$ is the normal-state Green's function given by

$$G_0(\mathbf{T} + \mathbf{k}, i\omega_n) = \frac{(i\omega_n - \chi_+) + \mathbf{p}_+ \cdot \mathbf{s}}{(i\omega_n - \chi_+)^2 - \mathbf{p}_+^2}, \quad G_0^*(-\mathbf{T} - \mathbf{k}, i\omega_n) = \frac{(-i\omega_n - \chi_-) + \mathbf{p}_- \cdot \mathbf{s}^*}{(-i\omega_n - \chi_-)^2 - \mathbf{p}_-^2}. \quad (26)$$

For convenience we introduced $\chi_{\pm} = \xi_{\mathbf{k}} \pm g_{\text{orb}}(k_y + k_{0,y})\mu_B B_{\parallel}$ ($k_{0,y}$ is the y -component of $\mathbf{k}_0$) and $\mathbf{p}_{\pm} = (\pm g_R k_y/2 + \mu_B B_{\parallel}, \mp g_R k_x/2, \pm g_I/2)$. Carrying out the $\xi_{\mathbf{k}}$ integral, Fermi surface average, and Matsubara summation yields a final form of the gap equation:

$$\ln \left(\frac{T_c}{T_c^0} \right) + \Phi(\rho_-, \tilde{\chi}_0) + \Phi(\rho_+, \tilde{\chi}_0) - \frac{\tilde{\mathbf{p}}_+ \cdot \tilde{\mathbf{p}}_-}{|\tilde{\mathbf{p}}_+||\tilde{\mathbf{p}}_-|} [\Phi(\rho_-, \tilde{\chi}_0) - \Phi(\rho_+, \tilde{\chi}_0)] = 0, \quad (27)$$

where $\tilde{\chi}_0 = -\tilde{g}_{\text{orb}}\mu_B B_{\parallel}/2\pi T_c$ ($\tilde{g}_{\text{orb}} \equiv g_{\text{orb}}(k_F + k_{0,y})$ denotes a characteristic scale for the orbital depairing), $\tilde{\mathbf{p}}_{\pm} = (\pm g_R k_F/2 + \mu_B B_{\parallel}, \mp g_R k_F/2, \pm g_I/2)$, and $\rho_{\pm} = (|\tilde{\mathbf{p}}_+| \pm |\tilde{\mathbf{p}}_-|)/2\pi T_c$. The function $\Phi(\rho, \tilde{\chi}_0)$ is defined in terms of the digamma function $\psi(z)$ as

$$\Phi(\rho, \tilde{\chi}_0) = \frac{1}{4} \left\{ \text{Re} \left[\psi \left(\frac{1+i\rho}{2} + i\tilde{\chi}_0 \right) - \psi \left(\frac{1}{2} \right) \right] + \text{Re} \left[\psi \left(\frac{1+i\rho}{2} - i\tilde{\chi}_0 \right) - \psi \left(\frac{1}{2} \right) \right] \right\}. \quad (28)$$

In the limit $\tilde{\chi}_0 \rightarrow 0$, i.e., without orbital depairing, Eq. (27) reduces to the form used in Ref. S21. This equation can be solved numerically to obtain the relationship between the critical temperature T_c and the critical in-plane field $B_{c\parallel}$ of the superconductor, given input SOC and orbital coupling parameters. As in the main text we denote the limits of zero-field critical temperature and zero-temperature critical field by T_c^0 and $B_{c\parallel}^0$, respectively.

Extended Data Fig. 11a,b displays the dependence of the Pauli-limit violation ratio on the Ising SOC (g_I), Rashba SOC (g_R) and orbital coupling ($\tilde{g}_{\text{orb}}$) parameters. In the presence of a purely Ising-type SOC ($g_R = \tilde{g}_{\text{orb}} = 0$), the evolution of $B_{c\parallel}$ as a function of T_c and g_I shows the characteristic low-temperature divergence^{S20} of $B_{c\parallel}$, which is due to the inability of the in-plane field—perpendicular to the Ising spin quantization axis—to destroy the resonance condition for spin-singlet pairing between electronic states at $\mathbf{k}$ and $-\mathbf{k}$. In other words, there is always a non-zero density of electronic states available for pairing opposite spin-components (albeit decreasing with $B_{\parallel}$), thus leading to persistent superconductivity⁵ as $T \rightarrow 0$. In contrast, when either Rashba

⁵Our linearized treatment can only capture second-order transitions, and thus neglects a possible first-order transition to a polarized normal state, in analogy with van Vleck paramagnetism. In this framework, the Zeeman energy gain in the normal state scales as $E_z \sim (\mu_B B)^2 / \lambda_I$ to second order in perturbation theory, instead of the standard form $\mu_B B$. This is due to the spin projection being locked in the out-of-plane directions by Ising SOC. Equating to the condensation energy Δ leads to the condition $B_c \sim \sqrt{\Delta \lambda_I} / \mu_B$ or $B_c / B_p \sim \sqrt{\lambda_I / \Delta}$ with the Pauli limiting field $B_p = \Delta / \sqrt{2} \mu_B$, a scaling form often quoted in the study of Ising superconductivity in TMDs^{S21,S22,S25}

SOC⁶ or orbital decoupling is added, the low-temperature divergence of $B_{c\parallel}$ is strongly suppressed, alongside a reduction of the Pauli-limit violation at all temperatures. This suppression occurs because both Rashba and orbital effects lead to a non-trivial deformation of the Fermi pockets under an in-plane field, which destroys the resonance conditions necessary for spin-singlet, zero-momentum pairing (we neglect finite-momentum, FFLO-type pairing channels in our analysis).

10 Modeling of the Pauli-limit violation data

We now describe various efforts at fitting the Pauli-limit violation data from Fig. 4c and Extended Data Fig. 9f to our theoretical model. The difference between these two data sets is that $B_{c\parallel}$ reported in Fig. 4c is obtained by a phenomenological extrapolation to zero temperature, whereas $B_{c\parallel}$ in Extended Data Fig. 9f is measured at a fixed base temperature $T_c \sim 30$ mK.

In the following we fix the value of Ising SOC to $g_I = 0.7$ meV as extracted from quantum Hall measurements, and assume that it remains constant as a function of doping (thus ignoring a potential interaction-induced enhancement of its bare value). We first consider Rashba and orbital effects separately. Model 1 (see Table below) considers only Rashba SOC ($\tilde{g}_{\text{orb}} = 0$), which because of its linear dependence on k_F is expected to scale as $g_R k_F \sim \sqrt{n_{\text{small}}}$ with n_{small} the electronic density in the small pocket. We note in passing that even if the dependence of the Rashba coupling in Eq. (24) was quadratic and thus produced a Rashba SOC that depends linearly on n_{small} , then a qualitatively similar trend of Pauli limit violation on filling would be seen. Due to the difficulty of extracting the size of the small Fermi pockets directly from quantum oscillation data, we take a phenomenological ansatz for the small pocket density. Motivated by the quantum oscillation data for the $\text{FP}(2,2)_+$ phase, we assume that the normalized SdH frequency $f_\nu = n_{\text{small}}/n$ (n is the total electronic density in the system) varies linearly with n in the $\text{FP}(2,2)_+$ phase—that is, we take $f_\nu = an + b$ with constants a, b to be treated as fitting parameters. This assumption leads to the form $g_R k_F \sim \sqrt{an^2 + bn}$ for the energy scale associated with Rashba coupling.

As shown in Extended Data Fig. 11c,d, Model 1 fits the PVR data well. However we find that to account for the strong PVR dependence, the Rashba energy scale $g_R k_F$ must vary strongly over the corresponding doping range. In the current Model 1, this variation occurs through an order-of-magnitude increase in the electronic density in the small pocket, which would require the small pocket density to nearly vanish at the edge of the SC dome. Part of the variation in $g_R k_F$ could also come from a doping dependence of the Rashba SOC parameter g_R itself, either due to interaction-induced renormalization or through band structure effects (e.g., higher-order terms in the low-energy projection that relates the bare λ_R to g_R in the low-energy description, Eq. 24).

⁶In the case of pure Rashba SOC ($g_I = 0$) we recover the $\sqrt{2}$ enhancement of the critical field compared to a spin-degenerate metallic state, first predicted by Gork'ov and Rashba^{S26} based on a microscopic calculation of the in-plane spin susceptibility.

The same conclusion is obtained for the case of only orbital effects ($g_R = 0$, see fits to the data in Extended Data Fig. 11c,d). A model with only an orbital source of depairing (as introduced in the previous section) can similarly account for the suppression of PVR as hole doping is increased. In our fits, motivated by the form of Eq. (24), we assume for simplicity a dependence of $\tilde{g}_{\text{orb}} \sim a\sqrt{n} + b$ where the constant offset qualitatively stems from the finite location of the locus of the trigonally warped pockets with respect to the Dirac point. In Models 2, 3 and 4 (see Table at the end of this section) we assume: filling-dependent $g_R k_F$ and constant orbital coupling $\tilde{g}_{\text{orb}} \sim c$; $g_R k_F = 0$ and filling dependent $\tilde{g}_{\text{orb}}$; and both filling dependent $g_R k_F$ and $\tilde{g}_{\text{orb}}$.

In summary, our theoretical modeling can account for the observed PVR evolution through a strong doping-dependence of either Rashba SOC and/or orbital depairing effects, both of which compete with Ising SOC. This is reminiscent to the phenomenology of Ref. S21, where a doping-dependent PVR was observed and attributed to increasing effects of Rashba SOC with carrier density. However, the required doping dependence appears very large in view of naive band-structure estimates. Consequently, band structure reconstruction mediated by electron interactions in the $\text{FP}(2, 2)_+$ phase must be significant for our scenario to capture the physics responsible for the evolution of Pauli-limit violation—providing a guidance for further theory modelling of superconductivity in BLG that is beyond the scope of this work. A potential resolution of this issue could involve an interaction-induced enhancement of Ising SOC—or the nucleation of another symmetry-broken phase which increases the out-of-plane spin canting, see discussion below Eq. (18)—in a doping dependent manner, particularly near the low hole doping region of the $\text{FP}(2, 2)_+$ phase.

Model #	g_I (meV)	$g_R k_F$ (meV)	$\tilde{g}_{\text{orb}}$ (1)	$T = 30$ mK	$T \rightarrow 0$
1	0.7	$\sqrt{an + bn^2}$	0	$a = -0.212$ $b = 0.0359$	$a = -0.137$ $b = 0.0231$
2	0.7	$\sqrt{an + bn^2}$	c	$a = -0.225$ $b = 0.0382$ $c = 0.0848$	$a = -0.136$ $b = 0.0236$ $c = 0.205$
3	0.7	0	$a\sqrt{n} + b$	$a = 1.81$ $b = -4.25$	$a = 1.79$ $b = -4.24$
4	0.7	$\sqrt{an + bn^2}$	$c\sqrt{n} + d$	$a = -0.259$ $b = 0.0439$ $c = 0.0122$ $d = 0.0872$	$a = -0.134$ $b = 0.0227$ $c = 0.0426$ $d = -0.0796$

Table 1: Models used for the fitting procedure. Here n is the total doping density in units of $\times 10^{11} \text{ cm}^{-2}$. Units of a, b, c, d fitting parameters are chosen to yield correct units of the physical parameters $g_I, g_R k_F, \tilde{g}_{\text{orb}}$. Their filling dependence is shown in Extended Data Fig. 11e,f.

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