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Supplementary information

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Supplementary Information: Superconductivity in metallic twisted bilayer graphene stabilized by WSe_2

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I Theoretical Modeling: Spin-orbit coupling in twisted bilayer graphene

Monolayer graphene with induced spin-orbit coupling: We begin by describing the induced spin-orbit interaction felt by monolayer graphene adjacent to a transition metal dichalcogenide (TMD). In the absence of spin-orbit coupling, the low-energy Hamiltonian for the monolayer is

$$H_0 = -\hbar v_0 \int_{\boldsymbol{k}} \Psi^\dagger(\boldsymbol{k})\Big(k_x\sigma^x\tau^z + k_y\sigma^y\Big)\Psi(\boldsymbol{k}), \tag{1}$$

where $\Psi(\boldsymbol{k})$ is an eight component spinor with sublattice, valley, and spin indices. The Pauli matrices $\sigma^{x,y,z}$ act on sublattice indices of the spinor, while $\tau^{x,y,z}$ act on the valley indices. The Fermi velocity is $v_0 = \sqrt{3}ta/(2\hbar) \sim 10^6$ m/s where $t = 2.61\,\text{eV}$ is the nearest-neighbour tunnelling between carbon atoms and $a = 0.246\,\text{nm}$ is the lattice constant[55]. The proximate TMD may induce a combination of Ising, Rashba, and Kane-Mele spin-orbit terms, which may be included by taking $H_0 \to H_0 + H_{\text{SO}}$, where[56,57]

$$H_{\text{SO}} = \int_{\boldsymbol{k}} \Psi^\dagger(\boldsymbol{k})\left(\frac{\lambda_I}{2}\tau^z s^z + \frac{\lambda_R}{2}\left(\tau^z\sigma^x s^y - \sigma^y s^x\right) + \frac{\lambda_{\text{KM}}}{2}\tau^z\sigma^z s^z\right)\Psi(\boldsymbol{k}). \tag{2}$$

Here, $s^{x,y,z}$ act on the spin indices. The parameters λ_I, λ_R , and λ_{KM} respectively quantify the strength of the Ising, Rashba, and Kane-Mele[58] spin orbit couplings. Their values, as extracted from density functional theory calculations[56,59] and various experiments[60–64], vary widely: $\lambda_I \sim 1 - 5\,\text{meV}$, and $\lambda_R \sim 1 - 15\,\text{meV}$. While DFT does not predict a substantial λ_{KM}, recent experiments[65] attributed Kane-Mele SOI as the dominant source of spin relaxation close to the CNP.

We note that a mass term, $\sim m\sigma^z/2$, may also be generated due to inversion symmetry breaking by the TMD. However, DFT predicts reasonably small values[56,59] for the mass relative to Ising and Rashba SOI. Given this expectation, and our focus on the effects of SOI, we hereafter neglect this contribution.

Twisted bilayer graphene without spin-orbit coupling: We briefly turn away from the question of spin orbit to provide a short overview of the continuum model[66,67] of twisted bilayer graphene with twist angle θ. It suffices to specialise to states proximate to the $+\boldsymbol{K}$ valley (*i.e.*, $\tau^z = +1$). We let ψ_t (ψ_b) represent the electron annihilation operator of top (bottom) layer. The model Hamiltonian may be expressed as

$$H_{\text{cont}} = H_t + H_b + H_{\text{tun}}. \tag{3}$$

The first two terms on the left-hand side respectively denote the Dirac Hamiltonian of the top and bottom layers in the absence of interlayer tunnelling:

$$H_{t/b} = \int_{\boldsymbol{k}} \psi^\dagger_{t/b}(\boldsymbol{k}) h_{t/b}(\boldsymbol{k}) \psi_{t/b}(\boldsymbol{k}), \tag{4}$$

where

$$h_t(\boldsymbol{k}) = -\hbar v_0 e^{i\theta\sigma^z/4} \boldsymbol{k}\cdot\boldsymbol{\sigma} e^{-i\theta\sigma^z/4}, \qquad h_b(\boldsymbol{k}) = -\hbar v_0 e^{-i\theta\sigma^z/4} \boldsymbol{k}\cdot\boldsymbol{\sigma} e^{i\theta\sigma^z/4}. \tag{5}$$

As in Eq. (1), v_0 is the Fermi velocity of graphene. The layers tunnel through

$$H_{\text{tun}} = \sum_{\ell=1,2,3} \int_{\boldsymbol{k}} \psi^\dagger_t(\boldsymbol{k}) T_\ell \psi_b(\boldsymbol{k}+\boldsymbol{q}_\ell) + h.c., \tag{6}$$

where

$$\boldsymbol{q}_\ell = k_\theta \left(-\sin\left[\frac{2\pi}{3}(\ell-1)\right]\hat{\boldsymbol{x}} + \cos\left[\frac{2\pi}{3}(\ell-1)\right]\hat{\boldsymbol{y}} \right), \qquad k_\theta = \frac{4\pi}{3a} 2\sin(\theta/2), \tag{7}$$

and

$$T_\ell = w_0 + w_1 \left(e^{-2\pi(\ell-1)i/3}\sigma^+ + e^{2\pi i(\ell-1)/3}\sigma^- \right). \tag{8}$$

Twisted bilayer graphene with induced spin-orbit coupling: Suppose now that a TMD resides adjacent to one of the graphene monolayers that compose twisted bilayer graphene. We assume (in correspondence to exprimental realization) that the TMD touches the top layer. The primary modification to the continuum model presented in Eq. (3) then occurs in H_t:

$$H_t = \int_{\boldsymbol{k}} \psi_t^\dagger(\boldsymbol{k}) \left(h_t(\boldsymbol{k}) + h_{t,\mathrm{SO}}\right) \psi_t(\boldsymbol{k}). \tag{9}$$

Here, $h_{t,\mathrm{SO}}$ represents the appropriately rotated projection of H_{SO} (Eq. (2)) onto the $+\boldsymbol{K}$ valley:

$$h_{t,\mathrm{SO}} = e^{i\theta\sigma^z/4}\left(\frac{\lambda_I}{2}s^z + \frac{\lambda_R}{2}\left(\sigma^x s^y - \sigma^y s^x\right) + \frac{\lambda_{\mathrm{KM}}}{2}\sigma^z s^z\right)e^{-i\theta\sigma^z/4}. \tag{10}$$

Given the interfacial nature of the induced spin-orbit effect, we do not expect substantial modifications to the Hamiltonian governing the bottom monolayer (when the TMD is only placed on one side).

II Landau-level spectrum near charge neutrality

We begin by very briefly outlining prior theoretical treatments of the Landau-level spectrum in BN-encapsulated TBG—for which SOI is not expected to play a role. Specializing to the experiments presented in this paper, we then demonstrate that SOI is not only generically expected to induce further splittings, but that the simplest model of TBG with SOI can return the observed LL spectrum (Fig. 3) for a reasonable set of parameters. Despite their importance, interactions are neglected in the discussion below for the sake of simplicity.

Boron nitride encapsulated TBG: The Landau-level spectrum observed near the CNP in TBG encapsulated in BN sufficiently far from the magic angle is usually 8-fold symmetric[68,69]. This result is in agreement with the band structure one obtains from the continuum model. In the absence of both SOI and a magnetic field, the system possesses two Dirac cones per $\boldsymbol{K}$-valley per spin at the $+\boldsymbol{\kappa}$ and $-\boldsymbol{\kappa}$ points in the moiré Brillouin zone—thus implying eight degenerate Fermi surfaces upon either electron or hole doping (see Fig. 4 and Extended Data Fig. 10). Equation (1) serves as a good low-energy description of the system close to the CNP provided a few changes are made: (i) the Pauli matrices $\sigma^{x,y,z}$ now act on the band indices of the operators Ψ ($\tau^{x,y,z}$ and $s^{x,y,z}$ still act on the $\boldsymbol{K}$ and spin flavour indices, respectively); (ii) there is an additional flavour index given by the moiré valley, $\pm\boldsymbol{\kappa}$; and (iii) the Fermi velocity is substantially renormalized from its value in monolayer graphene.

Experimentally, however, this simple band-structure picture appears insufficient close to the magic angle: the 8-fold degeneracy is lifted to a 4-fold degeneracy[70,71]. The latter degeneracy is instead obtained when a *single* Fermi surface is present per spin per ($\boldsymbol{K}$-)valley upon doping away from the CNP. Such a scenario has been shown to occur for reasonable parameters close to the magic angle, where the band structure is very sensitive to small changes in the angle and tunnelling parameters (w_0, w_1)[72]. Alternatively, in the presence of strain, the Dirac cones are no longer pinned to $\pm\boldsymbol{\kappa}$ and move closer together—again resulting in a single Fermi surface per spin and $\boldsymbol{K}$-valley upon doping away from charge neutrality[73,74]. Each Fermi surface yields a sequence $\pm 1, \pm 2, \pm 3, \ldots$, which becomes $\pm 4, \pm 8, \pm 12, \ldots$ upon accounting for the spin and valley degrees of freedom. The definitive explanation for the observed reduction in degeneracy, however, remains unclear at present.

Landau levels for TBG with SOI: Spin-orbit coupling breaks the $\mathrm{SU}(2)$ spin symmetry and provides another mechanism for reducing the Landau-level degeneracy. Starting from the scenarios highlighted in the previous section, the usual $\pm 4, \pm 8, \pm 12, \ldots$ sequence observed near the magic angle would then naturally further split down to $\pm 2, \pm 4, \pm 6, \ldots$ in the presence of SOI. We stress, however, that effects such as strain that can return fourfold degenerate Fermi surfaces close to charge neutrality are not included in the band structures shown in Fig. 4 and Extended Data Fig. 10. In these figures, two Fermi surfaces enclosing $\boldsymbol{\kappa}$ per spin per $\boldsymbol{K}$-valley are present at charge neutrality, as in the scenario discussed above for twisted bilayer graphene away from the magic angle. We show here that the experimentally observed twofold symmetry of Landau levels may nevertheless be obtained directly in the limit that the low-energy theory is described by eight Dirac cones with SOI. That is, SOI can break the the eighfold degeneracy down to a twofold degeneracy without have to take any other effects (e.g., strain) into account.

We first assume that we are close enough to charge neutrality that the Dirac description of TBG remains valid. As outlined in the previous section, an appropriately repurposed Eq. (1) provides a good starting point. SOI may then be included in a manner directly analogous to the approach we took above with monolayer graphene (*i.e.*, H_{SO} defined in Eq. (2)). We note, however, that the spin-orbit parameters (λ_I, λ_R, λ_{KM}) are not prohibited from taking different values at the $\pm\boldsymbol{\kappa}$ Dirac cones (the parameters at $\pm\boldsymbol{K}$ are related by time reversal). Nevertheless, we assume for simplicity that any differences in the effective parameters between $\boldsymbol{\kappa}$-valleys is unimportant.

Without loss of generality, we therefore begin by considering the Dirac cone at $+\boldsymbol{\kappa}$ with valley flavour $+\boldsymbol{K}$ (which, according to the assumption made above, is equally valid for the Dirac cones at $-\boldsymbol{\kappa}$ with valley flavour $-\boldsymbol{K}$). The first-quantized Hamiltonian takes the form

$$H = -\hbar v_F(i\partial_x\sigma^x + i\partial_y\sigma^y) + \frac{\tilde{\lambda}_I}{2}s^z + \frac{\tilde{\lambda}_R}{2}(\sigma^x s^y - \sigma^y s^x) + \frac{\tilde{\lambda}_{\mathrm{KM}}}{2}\sigma^z s^z. \tag{11}$$

This effective description may be derived in perturbation theory in powers of the interlayer tunnelling ($w_{1/2}/(\hbar v_0 k_\theta)$, to be precise) in a manner analogous to Ref. 67's original derivation of the magic angle. We have added a tilde to the SOI parameters as compared with previous sections to emphasise that $\tilde{\lambda}_I$, $\tilde{\lambda}_R$, and $\tilde{\lambda}_{\rm KM}$ describe the *effective* SOI relevant to the moiré Dirac cones, and that they are not expected to be the same as the SOI induced by the TMD on monolayer graphene. We have again neglected a possible mass term.

The magnetic field may now be included in a straightforward manner by taking $-i\boldsymbol{\partial} \to -i\boldsymbol{\partial} + e\boldsymbol{A}$, where e is the electronic charge and $\boldsymbol{A}$ is the vector potential corresponding to a magnetic field of strength B (the Zeeman splitting is negligible at the energy scales considered). We work in the Landau gauge, setting $\boldsymbol{A} = B(-y, 0)$ and write the wavefunction as $\Phi(x,y) = e^{ikx}\phi(y)$, $\phi(y) = (\phi_{1\uparrow}(y), \phi_{1\downarrow}(y), \phi_{2\uparrow}(y), \phi_{2\downarrow}(y))^T$ where $1, 2$ denote the band indices (acted on by $\sigma^{x,y,z}$) and $\uparrow, \downarrow$ denote the spin (acted on by $s^{x,y,z}$). The eigenvalue equation may be expressed as[55]

$$E\phi(y) = \begin{pmatrix} \tilde{\lambda}_+/2 & 0 & \hbar\omega_c A & 0 \\ 0 & -\tilde{\lambda}_+/2 & i\tilde{\lambda}_R & \hbar\omega_c A \\ \hbar\omega_c A^\dagger & -i\tilde{\lambda}_R & -\tilde{\lambda}_-/2 & 0 \\ 0 & \hbar\omega_c A^\dagger & 0 & \tilde{\lambda}_-/2 \end{pmatrix} \phi(y) \tag{12}$$

where $\tilde{\lambda}_\pm = (\tilde{\lambda}_{\rm KM} \pm \tilde{\lambda}_I)/2$, E is the energy to be obtained, and $\omega_c = \sqrt{2}v_F/\ell_B$ is the cyclotron frequency, with $\ell_B = \sqrt{\hbar/eB}$ the magnetic length. The operator $A = -\ell_B\partial_y + \ell_B k - y/\ell_B$ is an annihilation operator, while $A^\dagger = \ell_B\partial_y + \ell_B k - y/\ell_B$ is a creation operator. They satisfy $[A, A^\dagger] = 1$. It follows that the functions $\phi_{i\alpha}(y)$ are superpositions of solutions to the $1d$ simple harmonic oscillator. Further, each solution $\phi(y)$ with energy E corresponds to an extensively large set of degenerate states labelled by k. With this information, it is straightforward to show that there are generically four distinct solutions satisfying

$$\begin{aligned} 0 &= E - \tilde{\lambda}_-, \\ 0 &= \left(E + \frac{\tilde{\lambda}_+}{2}\right)\left(E + \frac{\tilde{\lambda}_-}{2}\right)\left(E - \frac{\tilde{\lambda}_-}{2}\right) - \tilde{\lambda}_R^2\left(E - \frac{\tilde{\lambda}_-}{2}\right) - \omega_c^2\left(E + \frac{\tilde{\lambda}_-}{2}\right). \end{aligned} \tag{13}$$

We denote these four solutions by $E^{(+)}_{1,\mu}$, $\mu = 1 - 4$. The remaining energies are obtained as solutions to

$$\begin{aligned} 0 = &\left((n-1)\omega_c^2 - \left(E - \frac{\tilde{\lambda}_+}{2}\right)\left(E + \frac{\tilde{\lambda}_-}{2}\right)\right)\left(n\omega_c^2 - \left(E + \frac{\tilde{\lambda}_+}{2}\right)\left(E - \frac{\tilde{\lambda}_-}{2}\right)\right) \\ &- \lambda_R^2\left(E - \frac{\tilde{\lambda}_+}{2}\right)\left(E - \frac{\tilde{\lambda}_-}{2}\right) \end{aligned} \tag{14}$$

for $n \geq 2$. We similarly label these energies by $E^{(+)}_{n,\mu}$, $\mu = 1 - 4$. It is then straightforward to show that the Landau level spectrum corresponding to a Dirac cone of the opposite chirality ($\tau^z = -1$) may be obtained directly by solving Eqs. (13) and (14) but with $\tilde{\lambda}_I \to -\tilde{\lambda}_I$ (*i.e.* $\tilde{\lambda}_\pm \to \tilde{\lambda}_\mp$). We denote these energies $E^{(-)}_{n,\mu}$.

We present in Extended Data Fig. 9b and Extended Data Fig. 9d solutions to Eqs. (13) and (14) for $n \leq 30$. Since the density of a Dirac cone (without a magnetic field) is proportional to the energy squared, we plot the magnetic field against the energy *squared*. The parameters chosen are $(\tilde{\lambda}_I, \tilde{\lambda}_R, \tilde{\lambda}_{\rm KM}) = (3, 4, 0)\,$meV and $(\tilde{\lambda}_I, \tilde{\lambda}_R, \tilde{\lambda}_{\rm KM}) = (1.5, 2.5, 2)\,$meV with $v_F \approx 10^5$ m/s, as appropriate for $\theta = 0.79°$ and $\theta = 0.87°$.

In the physical sample, the Landau levels are not actually expected to be infinitely degenerate, but rather possess some finite width. We model this effect through a phenomenological broadening of the density of states $D(E, B)$:

$$D(E,B) = 2 \cdot \frac{eB}{2\pi} \sum_{n=1}^{\infty} \sum_{\mu=1}^{4} \sum_{\eta=\pm} \frac{1}{\sqrt{2\pi}\Gamma} \exp\left[-\frac{(E_{n,\mu}^{(\eta)} - E)^2}{2\Gamma^2} \right]. \tag{15}$$

The coefficient out front, $2 \cdot eB/(2\pi)$, gives the degeneracy of each Landau level—$eB/(2\pi)$ times the number of Dirac cones within each of the $\boldsymbol{K}$ valleys. The sum over $\eta = \pm$ then accounts for the two valley flavours $\pm\boldsymbol{K}$. We present colour density plots of this function in Extended Data Fig. 9a and Extended Data Fig. 9c as a function of the magnetic field and E^2 using the same SOI parameters as above with broadenings $\Gamma = 0.22\,$meV and $\Gamma = 0.15\,$meV, respectively. The sum over n is evaluated up to $n = 30$. Notably, this simple model is able to reproduce several key features of the Landau level sequence shown in Fig. 3. For instance, at the higher fields shown, a sequence 2, 4, 6, 8, 10,... is observed. By contrast, at lower fields, we instead find 4, 6, 10, 14,....

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