Letter

Mechanism for π phase shifts in Little-Parks experiments: Application to $4Hb$-TaS_2 and to $2H$-TaS_2 intercalated with chiral molecules

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Recently, unusual π phase shifts in Little-Parks experiments performed on two systems derived from the layered superconductor $2H$-TaS_2 were reported. These systems share the common feature that additional layers have been inserted between the $1H$-TaS_2 layers. In both cases, the π phase shift has been interpreted as evidence for the emergence of exotic superconductivity in the $1H$ layers. Here, we propose an alternative explanation assuming that superconductivity in the individual $1H$ layers is of conventional s-wave nature derived from the parent $2H$-TaS_2. We show that a negative Josephson coupling between otherwise decoupled neighboring $1H$ layers can explain the observations. Furthermore, we find that the negative coupling can arise naturally assuming a tunneling barrier containing paramagnetic impurities. An important ingredient is the suppression of non-spin-flip tunneling due to spin-momentum locking of Ising type in a single $1H$ layer together with the inversion symmetry of the double layer. In the exotic superconductivity scenario, it is challenging to explain why the critical temperature is almost the same as in the parent material and, in the $4Hb$ case, the superconductivity's robustness to disorder. Both are nonissues in our picture, which also exposes the common features that are special in these two systems.

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Introduction. A π phase shift in Little-Parks experiments is usually taken as a strong indication for unconventional superconductivity [1–3]. A recent experiment observing such a shift involves the intercalation of a chiral molecule between $1H$-TaS_2 layers [4], while an earlier experiment involves the compound $4Hb$-TaS_2 which consists of alternating layers of $1T$ and $1H$ forms of TaS_2 [5] [see Fig. 1(c)]. In both cases, the samples were cut out of single crystals and it was proposed that the π phase shifts can be explained by the emergence of a superconducting state with multiple degenerate order parameters, resulting in a chiral superconducting state at low temperature, instead of the conventional s-wave pairing that is believed to describe the pristine $2H$-TaS_2 [6–8]. In this paper, we propose an alternative explanation of the π phase shift that does not require the postulation of a different pairing mechanism.

Our motivation to seek a more conventional mechanism is based on the following observations: First, in both experiments the superconducting transition temperature T_c is hardly changed compared to other experiments in $2H$-TaS_2 [7–9]. Second, the robustness of the superconducting state against disorder speaks against an unconventional pairing mechanism in the $4Hb$ case, where data is available [10]. The issue is even more severe in the molecular-intercalation case: we do not expect any significant charge transfer from the molecules to the TaS_2 layers and the effect of the molecules on the electronic structures of the metallic layers should be minimal. Thus, the emergence of an entirely different pairing state should be considered a great surprise. Finally, it is aesthetically more pleasing to find a common mechanism for both systems sharing a common parent superconductor.

In the Little-Parks experiments, the superconducting material is fabricated into a ring and the resistivity is measured as a function of the magnetic flux piercing the ring very close to the transition temperature. While the basic idea is that T_c is modulated periodically as a function of the magnetic flux, in the actual experiment the dependence of the resistivity is measured in the transition region. Importantly, for conventional superconductors, the resistivity is minimal at zero field. The π phase-shift effect refers to cases where the resistivity is maximal at zero field, indicating that T_c is increased and the free energy lowered by the magnetic flux [11]. Such a behavior is usually taken as evidence for unconventional superconductivity and is only expected to happen in polycrystalline samples [1,2] or rings with weak links [12,13].

The $J < 0$ scenario. We begin by stating our basic hypothesis: As seen in Fig. 1(c), in both examples, there are two $1H$ layers per unit cell stacked in an AB pattern. We assume that these layers are largely decoupled and inherit the conventional (intralayer) superconductivity of the pristine TaS_2 [9]. Importantly, we argue that in the two systems, where the π shift was observed, the Josephson coupling J between neighboring layers has a negative sign. Namely, $E_J = -J\cos(\phi_l - \phi_{l+1})$ with ϕ_l the phase of the order parameter in layer l, and thus $J < 0$ favors a π phase difference between the layers. In what follows, we show that the combination of negative Josephson coupling between individual $1H$ layers and lattice defects offers an explanation of the Little-Parks experiment without requiring a novel superconducting order parameter.

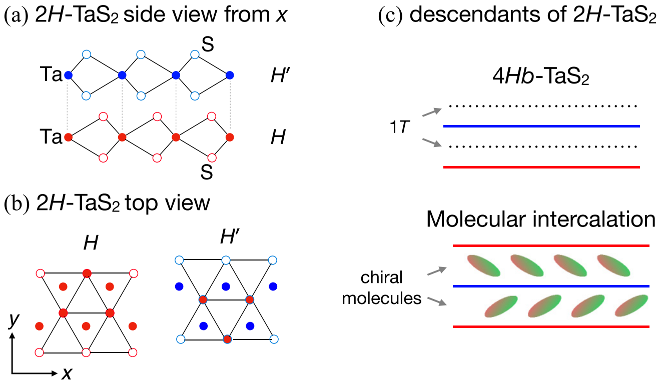

FIG. 1. (a) Side view of the crystal structure of 2H-TaS$_2$. The full circles are Ta atoms while empty circles are S. The unit cell consists of two layers that are inversion partners, denoted by H (red) and H' (blue). (b) Top view of the two types of H layers. (c) Types of descendants of 2H-TaS$_2$ considered here. Top: 4Hb-TaS$_2$, where monolayers of 1T-TaS$_2$ separate the H and H' layers. In 1T, the S layer above Ta is rotated by 60° relative to the layer below. Bottom: chiral molecules intercalated between the 1H layers.

For illustration, let us first consider a ring made of a single crystal, except for a single lattice defect, a screw dislocation, that pierces the center of the ring as shown in Fig. 2(a). As one completes a circuit around the ring, one ends up on another layer. The (global) pair phase is given by a slowly varying function multiplied by a sign, $\phi(\mathbf{r}) = (-1)^l \phi(r, \varphi)$, which switches from one layer to the next due to the negative Josephson coupling. Here, r is the radial coordinate and φ the in-plane angle. The screw dislocation therefore creates a phase mismatch of π. In order to smoothly connect the layers after going around once, the phase $\phi(r, \varphi)$ must wind by π costing kinetic energy. In contrast, applying a magnetic field yielding a half flux quantum through the ring requires no such phase winding and the associated energy cost [14]. This produces the π phase shifts in the Little-Parks experiment as shown in Fig. 2(b).

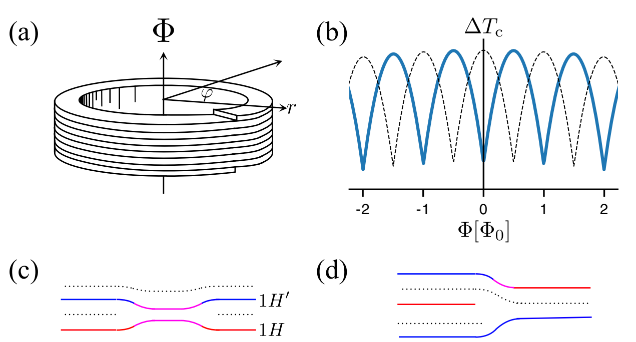

FIG. 2. (a) A superconducting ring made of a layered material, which encloses a single screw dislocation ($N = 1$). Due to the screw defect, a circuit around the ring is equivalent to a single-layer translation. To accommodate the screw, the superconducting order parameter develops a π phase shift when the layers are coupled with a negative Josephson coupling. (b) The Little-Parks oscillations of a screw-dislocation sample showing a π phase shift (solid line) compared to the standard case (dashed line). (c) A stacking fault with a dislocation of a single layer (a missing region of 1T layer). This locally reduces the negative Josephson coupling effect. (d) A missing double layer, which necessarily incurs a π phase shift on the 1H layers either above or below the dislocation line.

In general, we can expect that the screw dislocation will be frozen in during the deposition of the thin film. Furthermore, it is likely that the winding number N is not unity, but can be even or odd. Odd N favors a π phase shift while even N favors zero phase shift. There is a small complication in the systems under consideration because each unit cell contains two 1H layers, labeled H and H' in Fig. 1. Therefore, N odd technically corresponds to a half-integer screw dislocation, which will necessarily induce a domain-wall boundary between the H and H' layer. We will assume that the domain wall energy is small and not sufficient to form a bias between the integer and half-integer screw dislocations.

The above scenario of a nearly perfect single crystal is, of course, unrealistic. Indeed, it is known that transition metal dichalcogenides (TMDs) are prone to developing stacking faults. We can include these faults by decorating the screw dislocation with additional edge dislocation lines that lie in the plane. We show some examples in Fig. 2. However, the main effect of these additional dislocations will be a reduction of the energy splitting between zero and π phase winding. For a ring with a general structural defect, we expect the phase $\phi(\mathbf{r})$ will adjust in a slowly varying way to minimize the kinetic energy. Inserting a half flux quantum through the ring again leads to a readjustment of the overall phase. However, due to the built-in frustration arising from the sign change between layers, we can expect that in general the state with zero flux may have a free energy larger or smaller than the state with a half flux quantum, depending on the detailed defect structure. Note that a key prediction of this picture is that on average about half the samples will show a π phase shift, while the other half show no phase shift. This is indeed consistent with the reports on 4Hb-TaS$_2$ [5].

It should be pointed out that our picture is quite general and does not rely on the fact that the unit cell of 2H-TaS$_2$ contains two superconducting layers. However, as we will see in the following, the specific symmetry of the 2H system, with its two inversion-broken layers connected by inversion symmetry [15], greatly enhances the chances of finding this effect.

For the remainder of this work, we provide explanations for the origin of the postulated negative Josephson coupling between neighboring 1H layers. We base our discussion on an early paper by Kulik [16], who considered the presence of a spin-flip tunneling amplitude t_{sf} due to paramagnetic impurities in the tunneling barrier in addition to the standard spin-independent amplitude t_n. He found that the Josephson coupling has the form

$$J \propto (|t_n|^2 - |t_{sf}|^2). \tag{1}$$

While Kulik was interested mostly in the reduction of the Josephson coupling in a tunneling junction, this coupling can in principle change sign, if the spin-flip term dominates [13]. In this paper, we will extend Kulik's theory to the case where the superconductors have strong spin-orbit coupling (SOC), and apply it to the special case of 1H layers. We emphasize

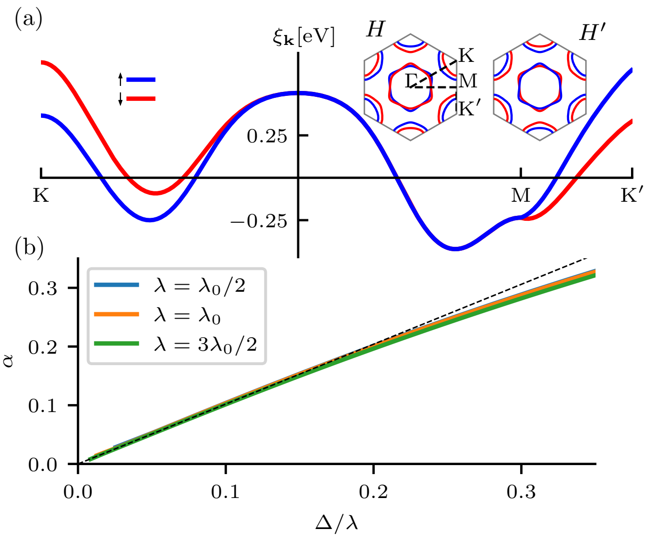

FIG. 3. (a) Typical 1*H*-TMD dispersion with the expected spin-splitting due to the Ising SOC (here $\lambda_0 \approx 0.07$ eV) [18,19]. The dispersion for the H' layer is identical except that the spins are interchanged, as illustrated by the Fermi surfaces of the two 1*H* layers (H and H') in 2*H*-TaS_2 and its descendants, such as 4*Hb*. As a result, momentum conserved interlayer tunneling for states near the Fermi surface is allowed only with spin-flip (except along Γ to M). The resulting suppression of spin-conserving tunneling, denoted by α in Eq. (6), is shown in (b) and is $\propto \Delta/\lambda$ (dashed line).

that in this mechanism, spin flip has to be due to scattering from magnetic impurities in the junction area, while spin flip from SOC preserves TRS and is insufficient, contrary to what was stated in Kulik's paper [14]. This is consistent with more recent papers that focus on the magnetic-impurity case [12,13]. Even more recently, Spivak and Kivelson have emphasized the role of strong correlations, which goes beyond the effective tunneling Hamiltonian approach [17].

General formalism. With the individual 1*H* layers largely decoupled, we consider the coupling of the layers within a tunneling approach. For this purpose, we consider in the following only two 1*H* layers, which we denote T (top) and B (bottom). In order to discuss Kulik's mechanism in more detail and extend it to the specific case of 1*H* layers, we need to examine the effect of symmetries on the tunneling process as well as present some details of the band structure. We begin by reviewing the electronic structure of the 2*H* layer, which is the same in both cases.

The individual 1*H* layer lacks inversion as well as C_2 symmetry around the z axis and, as a result, has a strong (Ising) SOC. The energy band can be labeled by momentum $\mathbf{k}$ and spin s quantized in the z direction. The dispersion for a given band (e.g., in the top layer) then reads $\xi^{\mathrm{T}}_{\mathbf{k}s} = \varepsilon_{\mathbf{k}} + s\lambda f_{\mathbf{k}}$, with $s = \pm$ for up and down spin, $\varepsilon_{\mathbf{k}} = \varepsilon_{-\mathbf{k}}$, and $f_{\mathbf{k}} = -f_{-\mathbf{k}}$. The SOC in these materials is extremely large $\lambda \sim 100$ meV [20]. We see in Fig. 3(a) that this leads to a large splitting between the spin up and down bands for generic $\mathbf{k}$. Importantly, note that $\lambda \mapsto -\lambda$ when going from a H layer to the H' layer, such that $\xi^{\mathrm{T}}_{\mathbf{k}s} = \xi^{\mathrm{B}}_{\mathbf{k}\bar{s}}$ with $\bar{s}$ the opposite spin. To discuss Josephson coupling, we introduce two (complex) spin-singlet s-wave superconducting order parameters in each layer, $\Delta^l_s = s|\Delta^l|e^{i\phi_l}$ with $l = \mathrm{T, B}$ the layer index.

For the Josephson coupling, we follow Kulik [16], who computed the Josephson coupling between two superconductors as the change in energy to second order in perturbation theory for a tunneling Hamiltonian,

$$\mathcal{H}_{\mathrm{tun}} = \sum_{\mathbf{k},\mathbf{p}} \sum_{ss'} \left[T^{ss'}_{\mathbf{kp}}\, c^{\dagger}_{\mathbf{k},\mathrm{T},s} c_{\mathbf{p},\mathrm{B},s'} + \mathrm{H.c.}\right], \tag{2}$$

where $T^{ss'}_{\mathbf{kp}} = t^{\mathrm{n}}_{\mathbf{kp}}\sigma^0_{ss'} + t^{\mathrm{sf}}_{\mathbf{kp}}\sigma^x_{ss'}$ describes both spin-independent and spin-dependent tunneling, σ^0 is the identity matrix, and σ^x is a Pauli matrix. Note that due to the origin of the spin-dependent Hamiltonian, we find $(t^{\mathrm{sf}}_{\mathbf{kp}})^* = t^{\mathrm{sf}}_{-\mathbf{k}-\mathbf{p}}$. This form breaks time-reversal symmetry, which requires that $(t^{\mathrm{sf}}_{\mathbf{kp}})^* = -t^{\mathrm{sf}}_{-\mathbf{k}-\mathbf{p}}$. In contrast to Ref. [16], we include the spin index to label the states, as this is crucial for our discussion. The spin-independent and spin-dependent corrections read

$$\Delta E_{\mathrm{n}} = -\sum_{\mathbf{k},\mathbf{p}} \sum_{s} \left|t^{\mathrm{n}}_{\mathbf{kp}}\right|^2 \frac{\left|u^{\mathrm{T}}_{\mathbf{k}s} v^{\mathrm{B}}_{\mathbf{p}s} + v^{\mathrm{T}}_{\mathbf{k}s} u^{\mathrm{B}}_{\mathbf{p}s}\right|^2}{E^{\mathrm{T}}_{\mathbf{k}s} + E^{\mathrm{B}}_{\mathbf{p}s}}, \tag{3}$$

$$\Delta E_{\mathrm{sf}} = -\sum_{\mathbf{k},\mathbf{p}} \sum_{s} \left|t^{\mathrm{sf}}_{\mathbf{kp}}\right|^2 \frac{\left|u^{\mathrm{T}}_{\mathbf{k}s} v^{\mathrm{B}}_{\mathbf{p}\bar{s}} + v^{\mathrm{T}}_{\mathbf{k}s} u^{\mathrm{B}}_{\mathbf{p}\bar{s}}\right|^2}{E^{\mathrm{T}}_{\mathbf{k}s} + E^{\mathrm{B}}_{\mathbf{p}\bar{s}}}, \tag{4}$$

with $E^l_{\mathbf{k}s} = \sqrt{(\xi^l_{\mathbf{k}s})^2 + |\Delta^l|^2}$ with the spin- and layer-dependent Bogoliubov transformation functions $u^{\mathrm{T,B}}_{\mathbf{k}s}$ and $v^{\mathrm{T,B}}_{\mathbf{k}s}$. Using $(u^l_{\mathbf{k}s})^* v^l_{\mathbf{k}s} = s|\Delta^l|\exp(i\phi_l)/E^l_{\mathbf{k}s}$, we find for the phase-dependent contributions

$$E_{\mathrm{J}} = -\sum_{\mathbf{k},\mathbf{p}} \sum_{s} \left[\left|t^{\mathrm{n}}_{\mathbf{kp}}\right|^2 \frac{|\Delta^{\mathrm{T}}\Delta^{\mathrm{B}}|}{E^{\mathrm{T}}_{\mathbf{k}s}E^{\mathrm{B}}_{\mathbf{p}s}\left(E^{\mathrm{T}}_{\mathbf{k}s} + E^{\mathrm{B}}_{\mathbf{p}s}\right)} - \left|t^{\mathrm{sf}}_{\mathbf{kp}}\right|^2 \frac{|\Delta^{\mathrm{T}}\Delta^{\mathrm{B}}|}{E^{\mathrm{T}}_{\mathbf{k}s}E^{\mathrm{B}}_{\mathbf{p}\bar{s}}\left(E^{\mathrm{T}}_{\mathbf{k}s} + E^{\mathrm{B}}_{\mathbf{p}\bar{s}}\right)}\right] \cos(\phi_{\mathrm{T}} - \phi_{\mathrm{B}}). \tag{5}$$

In the original discussion [16], momentum is not conserved in the tunneling process. The sum is dominated by contributions close to the original Fermi surface, as there, $|u^*_{\mathbf{k}s}v_{\mathbf{k}s}| = |\Delta|/E_{\mathbf{k}s} \propto 1$ and the energy denominator is the pairing gap. This gives rise to Eq. (1).

We can now ask what happens if we consider the tunneling to be (almost) momentum conserving. In the usual case of tunneling through an oxide barrier, the common assumption is that momentum is not conserved. This is due to strong scattering at the interface and in the oxide barrier itself. In the case of stacked van der Waals materials, the situation is different. The interface between layers is smooth and if the intercalated molecules form an ordered array, there is little in-plane scattering. We note that momentum conservation applies even if hopping between molecular dimers is negligibly small. For simplicity, we will proceed with the extreme case of perfect momentum conservation, considering the tunneling of a state with spin s and momentum $\mathbf{k}$ close to the original Fermi surface from the top layer to the bottom. Since the spin label in the dispersion is flipped between the layers, it is clear from Fig. 3(a) that the final state with spin s in the bottom layer is an excited state with energy given by the splitting between the red and blue bands, which is of order λ. This leads to a large energy denominator and a small coherence factor in the first term in Eq. (5), resulting in an overall reduction by a factor $\Delta/\lambda \ll 1$. In contrast, the spin-flip contribution remains $O(1)$, as the final state can be near the Fermi surface. To summarize, Eq. (1) is replaced by

$$J \propto (\alpha|t_n|^2 - |t_{sf}|^2), \quad (6)$$

where $\alpha \propto \Delta/\lambda \ll 1$. This behavior is confirmed in Fig. 3(b), where we used a momentum-independent tunneling matrix element for both tunneling processes [14]. We now apply this equation to the 4*Hb* and the molecular-intercalation cases.

*Case 1: The 4*Hb *system*. The intermediate 1*T* layer started out as a Mott insulator in a superlattice structure formed out of the "star-of-David" charge density wave [21]. The Mott insulator may be heavily depleted due to charge transfer. In the absence of disorder, momentum is conserved up to reciprocal superlattice vectors in tunneling. While van der Waals layers have negligible interface disorder, the star-of-David order gives a relatively small reciprocal lattice vector. Importantly, it is known that there is a dilute distribution of local moments [22,23], which will give rise to a finite t_{sf}. We assume the impurities to be sufficiently dilute and the superlattice scattering weak so that momentum is still conserved in the tunneling process. Furthermore, the pair-breaking effect of these impurities must not adversely affect the superconductivity in a significant way. It is therefore important to note that the electronic structure of the 1*H* layers is such that normal tunneling is strongly suppressed. A small amount of t_{sf} is sufficient for the second term in Eqs. (5) and (6) to dominate, resulting in the negative Josephson coupling.

Case 2: Molecular intercalation. Equation (6) applies equally well to the intercalation of chiral molecules. However, unlike the 4*Hb* case, we do not have sufficient understanding of the molecular system to provide an explicit mechanism for the requisite spin-flip scattering. Perhaps some kind of local moment is trapped near the contact point between the chiral molecule and the TaS_2 layer. We also note that our argument for the necessity of time-reversal breaking to form a π junction [14] only refers to the formulation involving an effective tunneling Hamiltonian. In contrast, the mechanism described by Spivak and Kivelson [17] produces a π junction without TRS breaking in a four-step process via a strongly correlated intermediate state, which is outside of the effective tunneling picture. What we can state is that intercalation into 2*H*-TaS_2 has the special feature of strong suppression of spin-independent tunneling near the Fermi level. As a result, any other tunneling process involving either extrinsic defects or Coulomb correlation may dominate. While we do not have an explanation of why the control sample with achiral molecule does not show the effect, we note that only one achiral molecule, which is quite different from the chiral molecule, was tested. It will be interesting to test other achiral molecules, especially ones with local moments, to see whether they show the π phase shift. Finally, we point out that this system exhibits strong chiral-induced spin selectivity (CISS) which sets in at a finite temperature [24]. While the mechanism is not understood, some form of spin-dependent interaction in the barrier is generally taken as an essential starting point [25].

Discussion. In this paper, we have provided an alternative explanation for the observed π phase shifts in the Little-Parks experiments in two systems, which does not postulate the appearance of a novel and exotic superconducting state. The common theme is that in both cases, the superconductivity resides in the 1*H* planes of the TMD superconductor. Furthermore, the breaking of inversion symmetry in the individual plane leads to spin-momentum locking of Ising type strongly suppressing non-spin-flip tunneling, leaving other spin-dependent processes that may change the sign of the interlayer Josephson coupling. The sign change leads to the appearance of π phase shifts in the Little-Parks experiments about half the time on average. For the 4*Hb* case, we propose that the requisite spin-flip process can come from known paramagnetic impurities in the 1*T* layers.

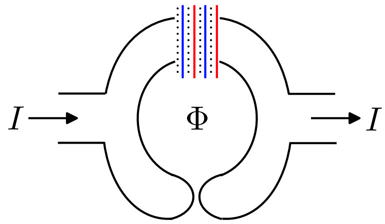

FIG. 4. SQUID geometry to measure the negative interlayer Josephson coupling. For an even number of layers, the SQUID enclosed a π flux

While this paper offers an alternative explanation of the π phase shift to that given in the original paper [5], we note that there have been several reports of intriguing findings in the 4*Hb* system [26–29] that point to exotic pairing, particular of the time-reversal-breaking type. It is worth remarking that in our model, we can expect to find screw dislocations that either penetrate the sample or form dislocation loops in the bulk. These induce π junctions and are unstable towards the formation of spontaneous current loops (clockwise or anticlockwise), which may affect the muon resonance relaxation rate [26]. In order to settle the question definitively, it is a challenge for future experiments to directly detect the presence or absence of the π phase in the Josephson coupling. In a vertical SQUID measurement, see Fig. 4, the interference pattern will exhibit a π shift in half of the samples if there is a negative interlayer Josephson coupling, while it is not expected in the case of a novel two-component order parameter. As a final note, we emphasize that our model has the general advantage of explaining why the superconductivity apparently survives in the dirty limit, as mentioned in the introduction.

Finally, we put our finding in the context of other systems that have been proposed to show similar physics. In particular, it was argued that in almost decoupled layered materials, the even and odd stacking of *s*-wave order parameters is almost degenerate in energy, such that potentially even a weak perturbation, such as a magnetic field, could tip this balance. This has first been proposed for artificial heterostructures [30] and is believed to happen in the Ce-based heavy-fermion compound $CeRh_2As_2$ [31]. Interestingly, the common scheme in these discussions is the local absence of inversion in the layers [15], with SOC driving the decoupling. In our scenario of spin-dependent tunneling between the layers, we argue that this tipping of the balance can happen without any external perturbation such as a magnetic field.

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