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Supplementary information

Magnetic memory and spontaneous vortices in a van der Waals superconductor

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sSupplementary Information for
Magnetic memory and spontaneous vortices in a van der Waals superconductor

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Supplementary Note 1. Ginzburg Landau theory for coupling a chiral superconductor to a chiral spin liquid

Here we provide a phenomenological Ginzburg-Landau theory that describes how a chiral spin liquid (CSL) induces vortices in a chiral superconductor (CSC).

The first element in the theory is the chiral state residing on the T layers, which is captured by the free energy density

(1)

$$f_{SL} = a_P(T - T_*)|\mathbf{P}|^2 + b_P|\mathbf{P}|^4 - \mu\,\mathbf{P}\cdot\mathbf{B}\,,$$

where $\mathbf{P} = \mathbf{S}_1 \cdot (\mathbf{S}_2 \times \mathbf{S}_3)\hat{\mathbf{z}}$ is the chiral order parameter, $\mathbf{S}_1, \mathbf{S}_2$ and $\mathbf{S}_3$ are the three spin operators on the plaquette of the triangular lattice of stars-of-David and T_* is the ordering temperature. The last term, which is proportional to μ, is the coupling of the chiral order parameter to the magnetic field $\mathbf{B}$. Following Ref. [1] we have

$$\mu = \frac{\pi A_\Delta}{18\Phi_0}\frac{W^3}{U^2}\,,$$

where $A_\Delta = 13a^2 \sin\pi/3$ is the area of the super unit-cell, a = 3.38 Å, Φ_0 is the flux quantum, W ≈ 50-100 meV is the width of the flat miniband[2], and U ≈ 200 meV is the on-site interaction strength. The magnetic field generated by a finite chirality, $\langle \mathbf{P} \rangle \neq 0$ (which we assume to be of order one), is then given by $|\mathbf{B}_P| = 4\pi\mu|\mathbf{P}| \approx 13$ pT. Thus, the generated field is expected to be much below the sensitivity of our probe.

Next, we consider a chiral superconducting state (CSC) since it allows a direct coupling between the spin chirality and superconductivity. Such coupling is necessary to account for the discrepancy between the spontaneous vortex density and normal state magnetization observed in the experiment. While a CSC is possible in 4Hb-TaS_2[3,4], our experimental data cannot decisively confirm its existence (see Supplementary Note 2). Furthermore, since bulk 2H-TaS_2 is an s-wave superconductor[5], it is necessary to include a single-component order parameter in the theory[6]. This s-wave order parameter, Δ, is described by the free-energy density

$$f_\Delta = a_\Delta \left(T - T_c^{(1)}\right) |\Delta|^2 + b_\Delta |\Delta|^4 + \frac{1}{2m^*} |\mathbf{D}\Delta|^2, \tag{2}$$

where $T_c^{(1)}$ is the s-wave transition temperature, $\mathbf{D} = -i\boldsymbol{\nabla} - 2e\mathbf{A}$ is the gauge invariant derivative and $\mathbf{A}$ is the vector potential related to the magnetic field through $\mathbf{B} = \boldsymbol{\nabla} \times \mathbf{A}$. To support chiral superconductivity, we consider a second order parameter which belongs to one of the two-component representations of the trigonal point group symmetry D3 and is described by the free-energy density[6–9]

$$f_\eta = a_\eta \left(T - T_c^{(2)}\right) |\boldsymbol{\eta}|^2 + b_\eta |\boldsymbol{\eta}|^4 + c_\eta |\boldsymbol{\eta}^* \times \boldsymbol{\eta}|^2 + \kappa_1 \left[|\mathbf{D}\eta_x|^2 + \left|\mathbf{D}\eta_y\right|^2\right] + \kappa_2 |\mathbf{D} \cdot \boldsymbol{\eta}|^2 + \kappa_3 |\mathbf{D} \times \boldsymbol{\eta}|^2, \tag{3}$$

where $\boldsymbol{\eta} = (\eta_x, \eta_y)$ is a vector of the two components of the order parameter. For $c_\eta < 0$ the system prefers a chiral ground state with $i\langle \boldsymbol{\eta}^* \times \boldsymbol{\eta} \rangle \neq 0$. Notice that here we have assumed symmetry permits $\kappa_2 \neq -\kappa_3$.

Integrating the last two terms by parts leads to the following contribution to the free-energy

$$i\gamma\, (\boldsymbol{\eta}^* \times \boldsymbol{\eta}) \cdot \mathbf{B},$$

where $\gamma = 2e(\kappa_3 - \kappa_2)$. Evidently, this term remains finite in the uniform chiral state. Therefore, a uniform chiral state generates a magnetic field in the z direction

$$\mathbf{B}_\eta = 4\pi\gamma \langle i\boldsymbol{\eta}^* \times \boldsymbol{\eta} \rangle. \tag{4}$$

It is important to note that without an external ordering field the SC chiral order parameter $i\boldsymbol{\eta}^* \times \boldsymbol{\eta}$ is expected to be break into domains of opposite chirality[10], $\boldsymbol{\eta}_\pm = \eta_0(1, \pm i)$. The net magnetic field is then an average over these domains.

The superconductor does not sustain the field in Eq. (4) when it is non-zero. If $B_\eta < H_{c1}$ it will screen the magnetic field by means of Meissner currents on domain or sample boundaries. On the other hand, for $B_\eta > H_{c1}$ one expects the magnetic field to penetrate in the form of vortices. Such vortices are not generated by an external field and may appear even when an external field is absent, if the domain structure is imbalanced and has a net magnetization. Ng and Varma[11] have discussed this scenario in the context of ferromagnetic materials that become superconducting. The situation here is similar[12], where the SC chiral order parameter takes the role of the magnetization of the ferromagnet.

The final element in the model is the coupling between the three fields $\mathbf{P}, \Delta$ and $\boldsymbol{\eta}$. By symmetry, the allowed couplings are

$$f_{coupling} = \lambda_{\Delta\eta}|\Delta|^2|\boldsymbol{\eta}^* \times \boldsymbol{\eta}|^2 + \lambda_{\Delta P}|\Delta|^2|\mathbf{P}|^2 + \lambda_{P\eta}(i\boldsymbol{\eta}^* \times \boldsymbol{\eta}) \cdot \mathbf{P} \tag{5}$$

Note that the third term in Eq. (5) is linear in $\mathbf{P}$ and is thus the only term that allows the domain structure embedded in the chiral spin-liquid to be transmitted to the superconducting state as described below.

We may consider two scenarios to explain the spontaneous vortex state:

- *The superconducting order parameter is chiral* – In this case the coupling to the chiral order parameter $\langle\mathbf{P}\rangle$ acts as an effective magnetic field for the SC chiral order, $i\langle\boldsymbol{\eta}^* \times \boldsymbol{\eta}\rangle = \frac{\lambda_{P\eta}}{2c_\eta}\langle\mathbf{P}\rangle$. If $c_\eta \propto T - T_c^{(2)}$ is divergent at the transition point (equivalent to assuming that there a single transition into the chiral superconducting state without a vestigial order), the chiral spin-liquid acts as an ordering field for the SC chiral order parameter. In turn, the CSC generates a spontaneous internal magnetization proportional to $\langle\mathbf{P}\rangle$, $\mathbf{B}_\eta = 4\pi\gamma\lambda_{P\eta}/c_\eta\langle\mathbf{P}\rangle$. If $B_\eta > H_{c1}$ this leads to a spontaneous vortex state.
- *The superconducting state is s-wave* – In this case there is no direct coupling of the chiral order parameter $\mathbf{P}$ to the superconducting one Δ. However, we may still assume that c_η is small and that $\lambda_{\Delta\eta} < 0$, which implies that the chiral order is soft, and that the chiral and s-wave orders are cooperative and not competing. In this case, when the superconducting order parameter condenses ($\langle\Delta\rangle \neq 0$), the susceptibility of $i\langle\boldsymbol{\eta}^* \times \boldsymbol{\eta}\rangle$ is shifted, $2/c_\eta \to 2/(c_\eta - \lambda_{\Delta\eta}\langle|\Delta|^2\rangle)$. In this way the superconducting transition into the s-wave state can also push the TRSB order parameter to develop a finite expectation value $i\langle\boldsymbol{\eta}^* \times \boldsymbol{\eta}\rangle$, or enhance it if it existed prior to the transition. This resembles the vestigial phase predicted to exist above the transition temperature of nematic and chiral superconductors[13], where the order parameter itself $\boldsymbol{\eta}$ does not condense, while its composite does.

Note that the order parameter $\boldsymbol{P}$ can represent TRSB states other than a chiral spin liquid, provided that they produce a weak magnetic field (support small μ). For example, charge transfer between the 1T and 1H layers[4] could result in a chiral metallic state[14] residing on the 1T layers. This state is expected to generate a magnetization significantly smaller than 1 μ_B per site.

Supplementary Note 2. Absence of direct evidence for chiral superconductivity

Our data cannot directly resolve the symmetry of the superconducting order parameter. Edge currents are an expected signature of chiral superconductors[7,10] and they are expected to generate a magnetic field response that may be detected with scanning SQUID microscopy. However, careful analysis of these currents is required before conclusions are drawn from SQUID experiments. For example, the presence of spontaneous currents is not a universal feature of chiral superconductors. They are expected in p-wave but not necessarily in d- and f-wave superconductors[15]. Even in p-wave superconductor, the magnitude of the currents is determined by the boundary conditions and could be small if the surface is rough[9]. Finally, even if considerable edge currents are present, they generate a magnetic field that is screened from the superconductor by counter propagating Meissner currents. The overall magnetic field generated by these contributions is expected to cancel when studied on length scales larger than the penetration depth[9].

The sensitivity of the edge signals to multiple parameters makes it difficult to conclusively interpret experimental results. For example, although scanning SQUID[16] and scanning Hall probe[17] experiments on Sr_2RuO_4 did not detect signals at the edge of an untrained sample, other experimental indications, such as a split superconducting and TRSB transition temperature under strain[18], do support chiral superconductivity. In 4Hb-TaS_2, it is challenging to determine whether the currents observed at the edge of the untrained sample below T_c are a result of Meissner currents due to the small external field still present at the system, or due to spontaneous currents from a CSC state.

Supplementary Note 3. Possible coupling mechanisms between a CSL and an s-wave superconductor

Although a two-component order parameter provides a direct path for coupling between the chiral state and the SC, an indirect mechanism may exist for a single-component SC order parameter. For example, Kondo coupling between itinerant electrons and a chiral spin texture is expected to generate an anomalous Hall effect[19]; an analogous effect may occur in superconductors[20]. Furthermore, a magneto-elastic effect could store magnetic information in elastic deformations of the crystal, providing an alternative coupling mechanism between the hidden chiral state and the superconductor. Further work is required to provide a complete microscopic description of the effect.

References

1. Motrunich, O. I. Orbital magnetic field effects in spin liquid with spinon Fermi sea: Possible application to κ-(ET)2Cu2(CN)3. *Phys. Rev. B* **73**, 155115 (2006).
2. Wang, Y. D. *et al.* Band insulator to Mott insulator transition in 1T-TaS2. *Nat. Commun.* **11**, 4215

(2020).
3. Ribak, A. *et al.* Chiral superconductivity in the alternate stacking compound 4Hb-TaS 2. *Sci. Adv.* **6**, eaax9480 (2020).
4. Nayak, A. K. *et al.* Evidence of topological boundary modes with topological nodal-point superconductivity. *Nat. Phys.* **17**, 1413–1419 (2021).
5. Nagata, S. *et al.* Superconductivity in the layered compound 2H-TaS2. *J. Phys. Chem. Solids* **53**, 1259–1263 (1992).
6. Chirolli, L., de Juan, F. & Guinea, F. Time-reversal and rotation symmetry breaking superconductivity in Dirac materials. *Phys. Rev. B* **95**, 201110 (2017).
7. Sigrist, M. & Ueda, K. Phenomenological theory of unconventional superconductivity. *Rev. Mod. Phys.* **63**, 239–311 (1991).
8. Zyuzin, A. A., Garaud, J. & Babaev, E. Nematic Skyrmions in Odd-Parity Superconductors. *Phys. Rev. Lett.* **119**, 167001 (2017).
9. Etter, S. B., Bouhon, A. & Sigrist, M. Spontaneous surface flux pattern in chiral p-wave superconductors. *Phys. Rev. B* **97**, 64510 (2018).
10. Kallin, C. & Berlinsky, J. Chiral superconductors. *Reports Prog. Phys.* **79**, 54502 (2016).
11. Ng, T. K. & Varma, C. M. Spontaneous Vortex Phase Discovered? *Phys. Rev. Lett.* **78**, 330–333 (1997).
12. Knigavko, A. & Rosenstein, B. Spontaneous vortex state and ferromagnetic behavior of type-II p-wave superconductors. *Phys. Rev. B* **58**, 9354–9364 (1998).
13. Fernandes, R. M., Orth, P. P. & Schmalian, J. Intertwined Vestigial Order in Quantum Materials: Nematicity and Beyond. *Annu. Rev. Condens. Matter Phys.* **10**, 133–154 (2019).
14. Song, X.-Y., Vishwanath, A. & Zhang, Y.-H. Doping the chiral spin liquid: Topological superconductor or chiral metal. *Phys. Rev. B* **103**, 165138 (2021).
15. Huang, W., Taylor, E. & Kallin, C. Vanishing edge currents in non- p-wave topological chiral superconductors. *Phys. Rev. B* **90**, 224519 (2014).
16. Hicks, C. W. *et al.* Limits on superconductivity-related magnetization in Sr2RuO4 and PrOs4Sb12 from scanning SQUID microscopy. *Phys. Rev. B* **81**, 214501 (2010).
17. Kirtley, J. R. *et al.* Upper limit on spontaneous supercurrents in Sr2RuO4. *Phys. Rev. B* **76**, 014526 (2007).
18. Grinenko, V. *et al.* Split superconducting and time-reversal symmetry-breaking transitions in Sr2RuO4 under stress. *Nat. Phys.* **17**, 748–754 (2021).
19. Martin, I. & Batista, C. D. Itinerant Electron-Driven Chiral Magnetic Ordering and Spontaneous Quantum Hall Effect in Triangular Lattice Models. *Phys. Rev. Lett.* **101**, 156402 (2008).
20. Zeytinoğlu, S., İmamoğlu, A. & Huber, S. Tunable Flux Vortices in Two-Dimensional Dirac Superconductors. *Phys. Rev. Lett.* **124**, 207006 (2020).