

Supplementary Information:
**Spectroscopic evidence for a first-order transition to a possible
orbital Fulde-Ferrell-Larkin-Ovchinnikov state**

Zongzheng Cao^{1#}, Menghan Liao^{2,3#*}, Hongyi Yan^{4#}, Yuying Zhu³, Liguozhang³, Kenji Watanabe⁵, Takashi Taniguchi⁶, Alberto F. Morpurgo²,
Haiwen Liu^{4*}, Qi-Kun Xue^{1,3,7} and Ding Zhang^{1,3,8*}

¹ State Key Laboratory of Low Dimensional Quantum Physics and Department of Physics, Tsinghua University, Beijing 100084, China

² Department of Quantum Matter Physics, University of Geneva, Geneva 1211, Switzerland

³ Beijing Academy of Quantum Information Sciences, Beijing 100193, China

⁴ Center for Advanced Quantum Studies, Department of Physics, Beijing Normal University, Beijing 100875, China

⁵ Research Center for Electronic and Optical Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

⁶ Research Center for Materials Nanoarchitectonics, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

⁷ Southern University of Science and Technology, Shenzhen 518055, China

⁸ Hefei National Laboratory, Hefei 230088, China.

Contents

Supplementary Note 1 Discussion of competing orders

Supplementary Note 2 Superconducting gap and its temperature dependence
(Supplementary Fig. 1)

Supplementary Note 3 Extended data of sample S1, S3-S5 (Supplementary Figs. 2-9)

Supplementary Note 4 Influence of sample quality on the sharpness of phase transition (Supplementary Fig. 10)

Supplementary Note 5 Phase transition in the tunneling spectroscopy of 2H-NbS₂
(Supplementary Figs. 11-12)

Supplementary Note 6 Extended data on the thickness dependence and comparison between NbSe₂ and FeSe (Supplementary Figs. 13-14)

Supplementary Note 1. Discussion of competing orders

Recently, a magnetic field induced phase transition between two superconducting phases was observed in CeRh_2As_2 ¹. The crystal structure of CeRh_2As_2 is unique that the two Ce sublayers are inversion partners. The Rashba spin-orbital couplings (SOC) in the two sublayers have opposite signs, producing a “staggered” (layer-alternating) SOC field. Such staggered Rashba SOC allows for both the even-parity and the odd-parity superconducting states. As a magnetic field is applied along the *c*-axis, the energy of the even-parity phase increases due to the Pauli effect. At a certain critical field, it becomes energetically more favorable for the system to switch to the Pauli-insensitive odd-parity phase via a first-order transition. Such a mechanism for the first-order transition is not suitable for 2H-NbSe_2 for the following reasons: (1) In CeRh_2As_2 , the transition field is set by the magnitude of the staggered Rashba SOC and should not vary with the sample thickness. By contrast, in 2H-NbSe_2 we observe a pronounced thickness dependence of B_0 ; (2) The odd-parity superconducting state in CeRh_2As_2 is fragile against impurity scattering and requires the orbital pair-breaking effect to be strongly suppressed. By contrast, 2H-NbSe_2 exhibits strong orbital effects under an in-plane magnetic field, and it is precisely these orbital effects that drive the system into the orb-FFLO state. (3) The crystalline structure of 2H-NbSe_2 does not break the sublayer inversion symmetry such that there exists no staggered Rashba SOC.

In fact, FFLO-like phases, such as helical and stripe phases, were predicted to occur for systems hosting the standard Rashba SOC². However, we point several differences between this proposal and our experimental system. (1) Rashba SOC is absent in a freestanding monolayer of 2H-NbSe_2 as the Nb layer constitutes the mirror symmetry plane. In reality, a thin flake on the substrate may experience Rashba SOC since the top and bottom surfaces may face different materials. However, this effect should get progressively suppressed in thick flakes, such as the ones we used (20-30 nm). (2) The critical field of the first-order transition in 2H-NbSe_2 shows a pronounced dependence on layer thickness and is quantitatively consistent with our orb-FFLO theory. By contrast, the previous theory² primarily treats a monolayer system.

Whether or not the nonuniform stripe (FFLO-like) and helical phases exist in multilayer systems was unaddressed there. (3) The theory² predicts two second-order phase transitions in the high-temperature regime: BCS to helical phase and helical phase to stripe phase. In 2H-NbSe₂, however, we observe only one first-order phase transition, which persists from very low temperature to a temperature close to T_c .

In a similar fashion, we can exclude the scenario of odd-parity pair density wave state (PDW)³, because the theoretical model there considered a bilayer/trilayer transition metal dichalcogenide (TMDC) with 2Hb stacking. This stacking is relevant for intercalated 2H-MoS₂, 2H-WS₂, etc. For 2H-NbSe₂, its 2Ha stacking prevents the system from hosting the essential ingredient for the proposed PDW state. Still, we remark that the Josephson-vortex state analyzed in the reference³ for bilayers is closely related to the orb-FFLO state. In fact, it reduces to the orb-FFLO state when the parameter δ in the model becomes zero. Our work generalizes the orb-FFLO concept to a multilayer situation, where the interplay of orbital effects and interlayer coupling across many layers quantitatively explains the observed thickness dependence, which is not included in the bilayer model of the reference.

The spin-triplet superconductivity scenario was also proposed to occur in a Ising superconductor such as monolayer of NbSe₂⁴. Two channels of spin-triplet pairing η_y and η_z were considered. The emergence of η_y was predicted to be continuous, in sharp contrast to the first-order transition seen in our experiment. For the η_z channel, a sudden jump was predicted but it occurred only when T is below the transition temperature of the spin-triplet component in the z-channel. Furthermore, this jump was predicted to occur at a high magnetic field close to B_{c2} where the singlet component reaches zero. These predictions are distinctly different from our experimental observations. For instance, we obtain the phase transition at an in-plane magnetic field of 1.0 T for the 30-nm thick NbSe₂ at 1.6 K, whereas the corresponding B_{c2} is nearly one order of magnitude higher. Furthermore, the first-order phase transition in our experiment extends from $T \ll T_c$ all the way to the point very close

to T_c . This is distinctly different from the theoretically expected sudden occurrence of a first-order transition as T drops substantially from T_c .

To summarize, while the alternatives can share ingredients with the orb-FFLO framework, their specific, testable predictions—regarding the thickness dependence, the temperature evolution, the nature of the phase transition, and material specificity/disorder sensitivity—do not align with our data. By contrast, the orb-FFLO scenario quantitatively captures the observed phase boundary and its systematic dependence on thickness, making it the most self-consistent explanation at present.

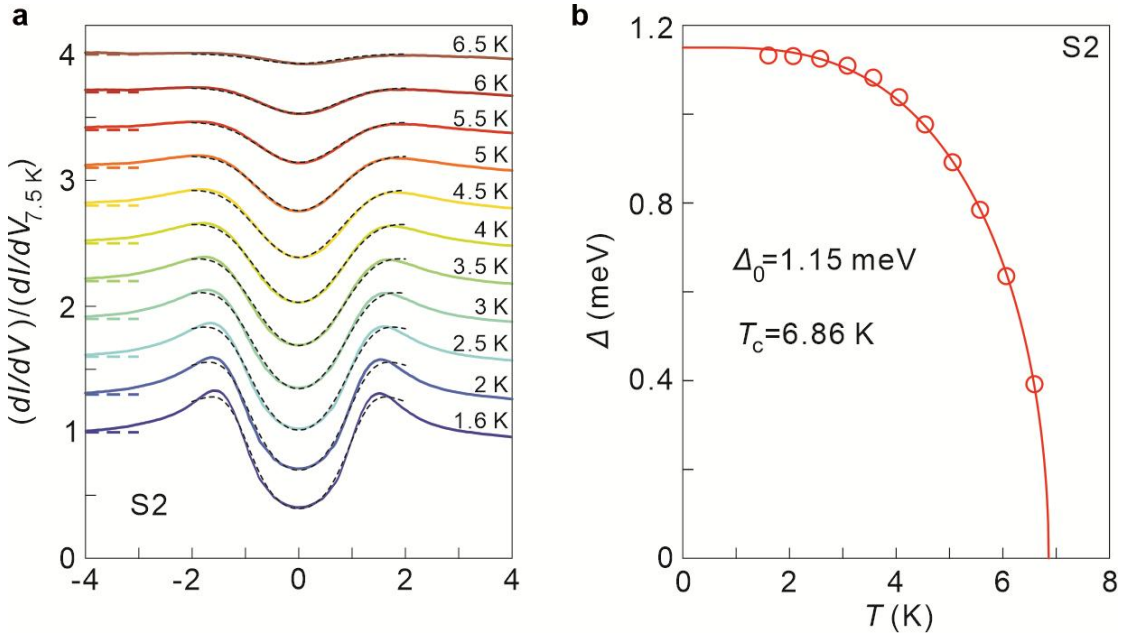
Supplementary Note 2. Superconducting gap and its temperature dependence

We extract the superconducting gap of sample S2 by fitting the normalized tunneling spectra with the Dynes formula⁵. Supplementary Figure 1a shows our data and the theoretical fitting. We employ the formula for the material with a single gap although NbSe₂ is a multi-gap system. This protocol is valid because the two-gap behavior is only prominent in the tunneling spectra at ultralow temperatures⁶. In the temperature regime of our measurements, the tunneling spectra are dominated by a single gap.

Supplementary Figure 1b shows the extracted superconducting gap as a function of temperature. We employ the gap function from the BCS theory to fit the data:

$$\Delta(T) = \Delta_0 \tanh(1.74\sqrt{T_c/T - 1}),$$

where T_c is the critical temperature, and Δ_0 is the superconducting gap at $T=0$ K. The extracted Δ_0 is 1.15 meV, and T_c is 6.86 K. Therefore, $2\Delta/k_B T_c = 3.9$. This value is in agreement with previous studies⁶⁻⁸.



Supplementary Figure 1 Temperature dependence of the tunneling spectrum. a Normalized tunneling spectra of sample S2 at different temperatures. Dashed curves are from theoretical fitting. Curves are vertically offset for clarity. **b** Superconducting gap of sample S2 as a function of temperature. The solid curve is the gap function from the BCS theory.

Supplementary Note 3 Extended data of sample S1, S3-S5

Supplementary Figure 2 collects data from sample S1 (NbSe₂: 13 nm). Supplementary Figure 2a is the optical image of the sample. The resistance measurements (Supplementary Fig. 2b) indicates that the upper critical fields start to deviate from the 2D Ginzburg-Landau equation at around 10.5 T. Supplementary Figure 2c shows the superconducting gap of S1 obtained from the tunneling spectroscopy. Supplementary Figure 2d shows the zero-bias tunneling conductance (ZBTC) as a function of in-plane magnetic field up to 12 T and down to 0.38 K. We observe no indication of phase transition in the tunneling measurements. From the data on S2 to S4, we observe that B_o is usually higher than B^* . Therefore, the absence of kink/jump in ZBTC of S2 up to 12 T probably indicates that $B_o > 12$ T. Besides, this thin sample may experience stronger disorder effect which suppresses the formation of the orb-FFLO state. We present further evidence for the disorder effect in the next note.

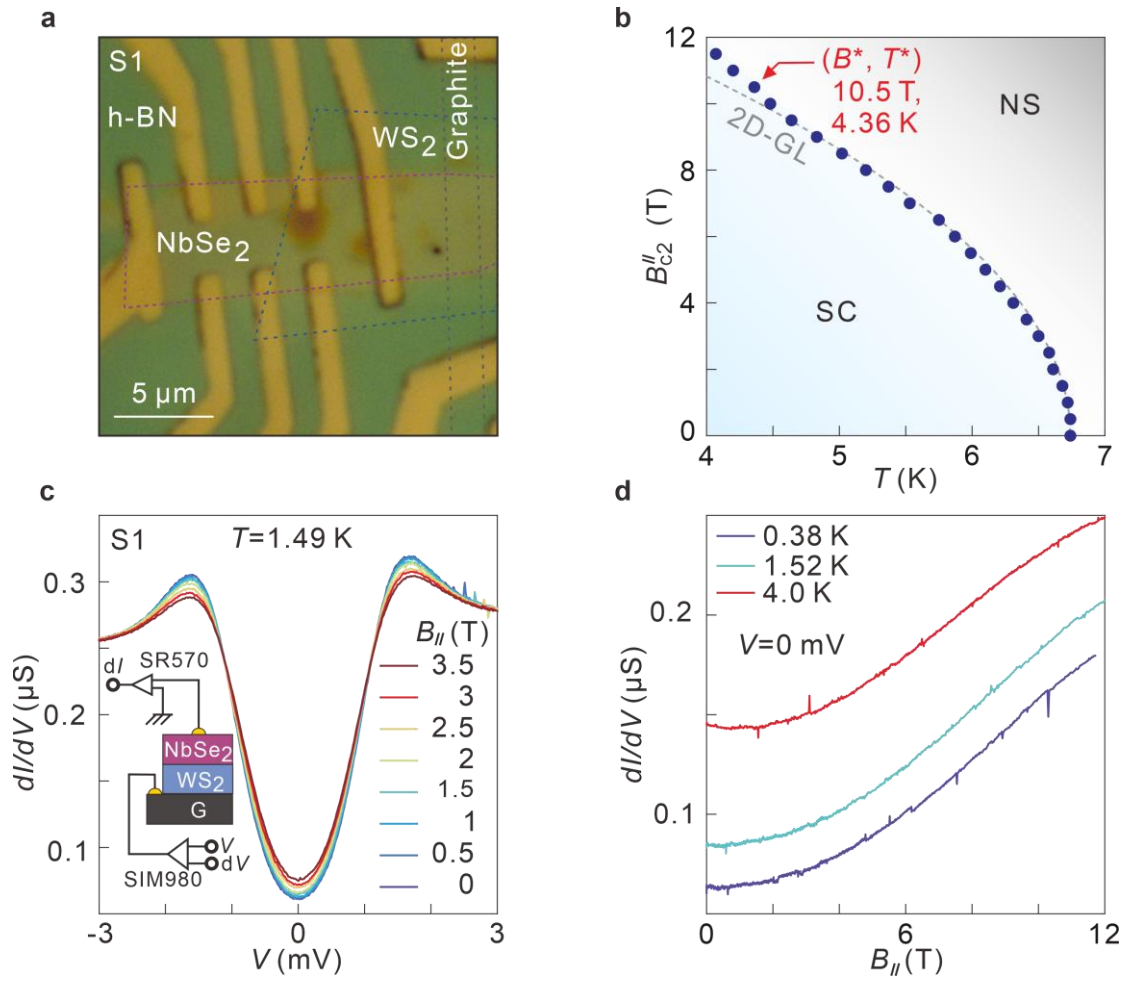
Apart from S1 mentioned above and S2 discussed in the main text, we present data from samples S3 to S5 in Supplementary Figs. 3 to 9. These results confirm the good reproducibility of our major claims.

Supplementary Figure 3a is the optical image of sample S3 (21 nm). Supplementary Figure 3b,c present the magneto-resistance around the superconducting transition and tunneling spectra of sample S3. We note that this sample has almost the same thickness of sample S2. Indeed, Supplementary Figure 4 shows that sample S3 hosts very similar evolution of the first-order phase transition in ZBTC as the temperature decreases. The phase boundary also occurs around 6 T. In Supplementary Note 3, we provide a close comparison between S2 and S3, showing that the phase transition sensitively depends on the disorder effect.

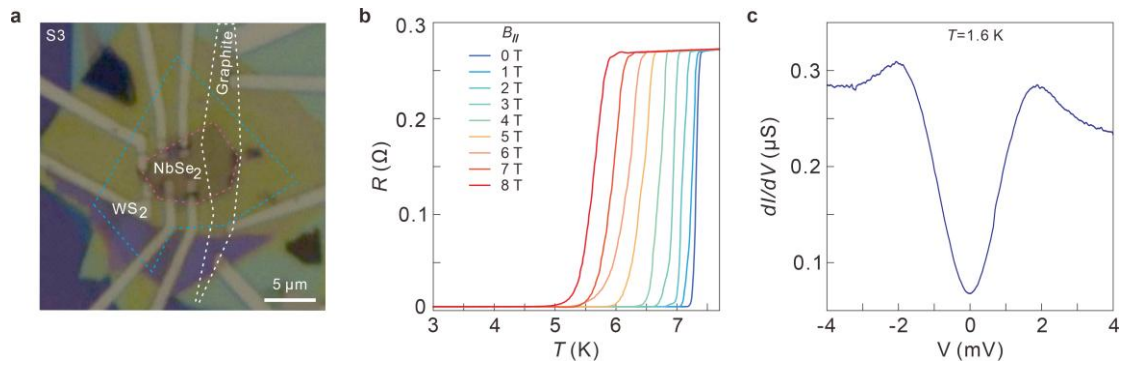
Supplementary Figure 5 collects the magneto-resistance and tunneling spectra of sample S4 (25 nm) (Supplementary Fig. 5a is the optical image of the sample). This

sample was measured first in a helium-4 cryostat for the data shown in Supplementary Fig. 5b,c and in a dilution refrigerator for the data in Supplementary Fig. 5d-f. In this sample, we reproduce the sudden enhancement of the superconducting gap at an in-plane magnetic field much smaller than the upper critical field. The phase boundary now moves to about 5 T. Supplementary Figure 6 further demonstrates the temperature evolution of the zero-bias tunneling conductance (ZBTC) of sample S4 as a function of $B_{\parallel}$. Clearly, a kink develops at a field well below the gap closing field at temperatures below 6.6 K. At lower temperatures (below 4.8 K), the kink becomes more pronounced and shows hysteresis. Supplementary Figure 7 shows the tilt-field dependence of ZBTC. It confirms again that the first-order phase transition occurs only within a narrow range of tilt angles around 0° . We also measure ZBTC as a function of $B_{\parallel}$ at various temperatures. The critical field B_o of the transition slightly decreases with increasing temperature, if comparing the local peak at 22 mK and that at 1.3 K.

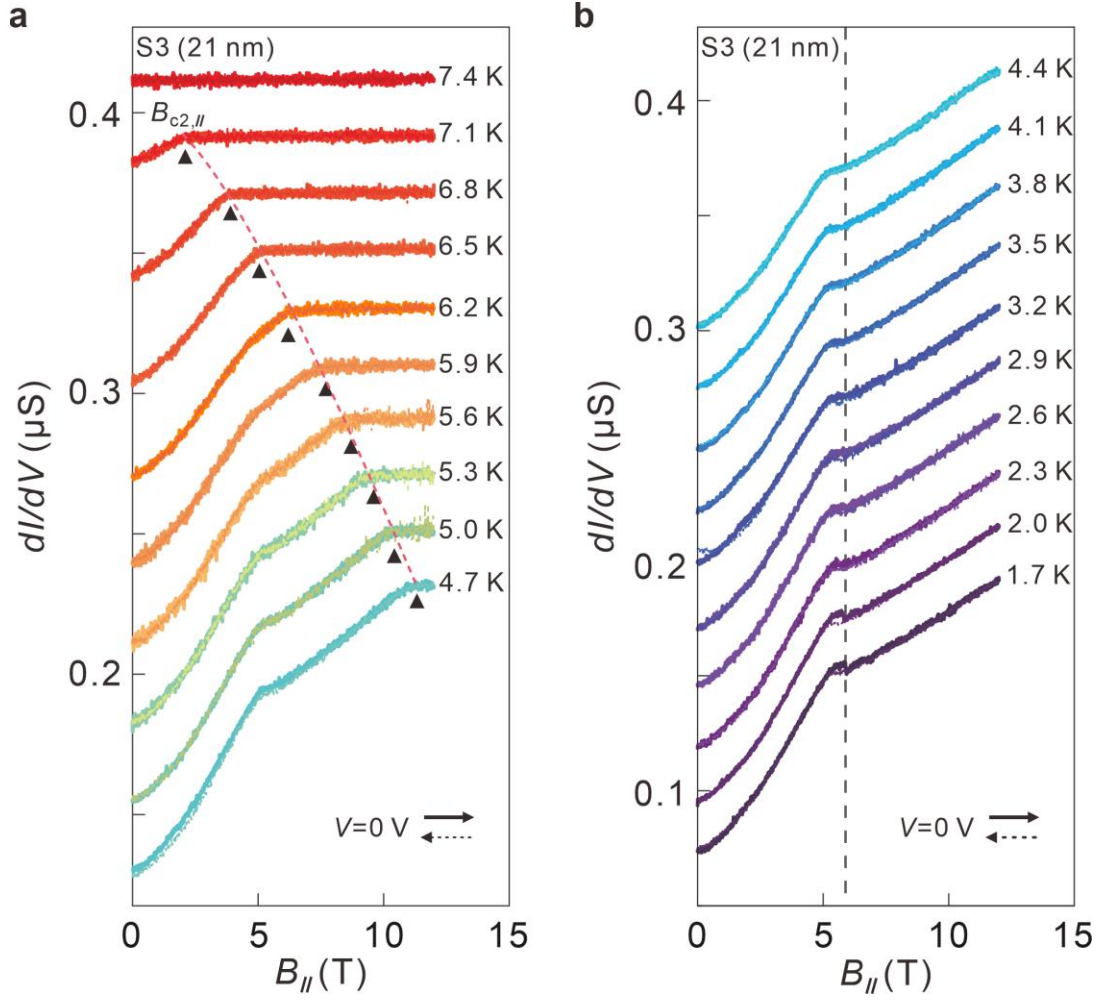
Supplementary Figure 8 and 9 present the data from sample S5 (30 nm). Essentially, we reproduce all the features observed in samples S2-S4 in this sample too.



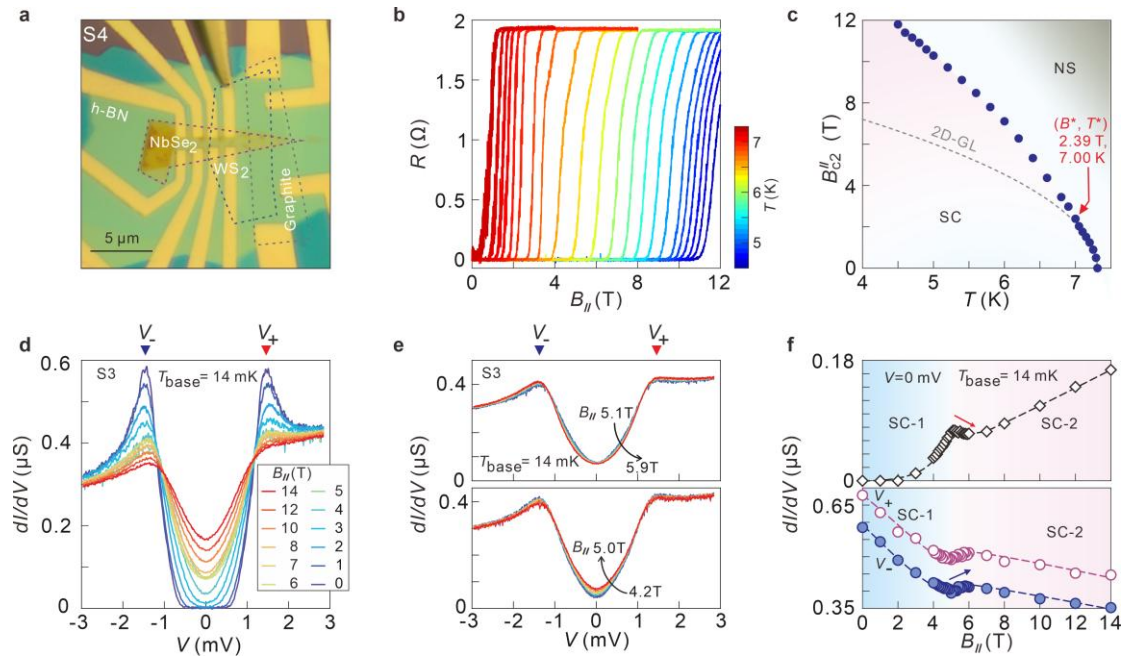
Supplementary Figure 2 Extended data of sample S1. **a** Optical image of sample S1. **b** Temperature dependence of the in-plane upper critical magnetic field ($B_{c2}^{\parallel}$) of sample S1. Dashed curve is from the theoretical fitting by using the 2D G-L formula. **c** Tunneling spectra under different $B_{\parallel}$. Inset illustrates the measurement set-up, with G represents the graphite layer. DC and AC voltages are added together by using the analog summing amplifier (Stanford Research SIM980). SR570 represents the current amplifier. **d** ZBTC as a function of $B_{\parallel}$ at different temperatures.



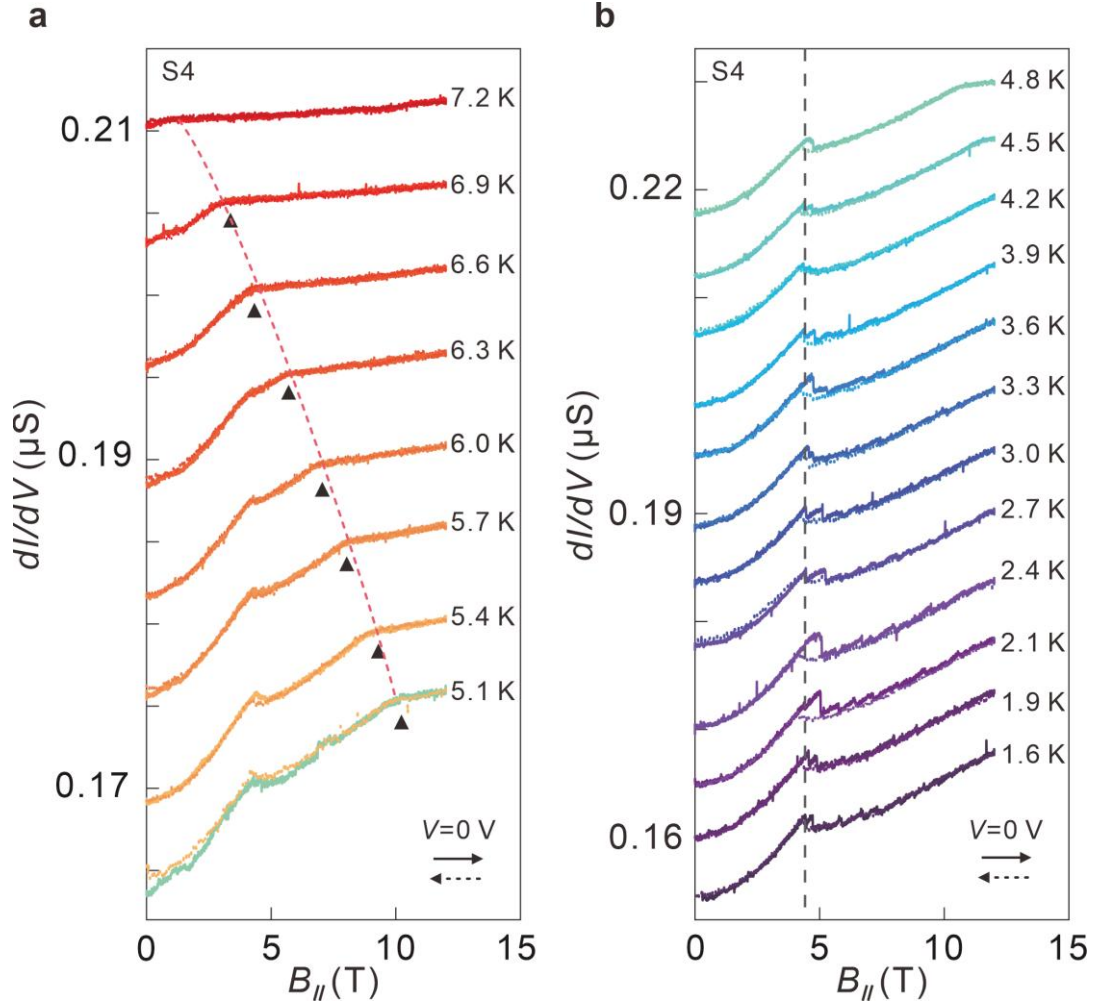
Supplementary Figure 3 Basic characterization of sample S3. **a** Optical image of sample S3 (21 nm). Dashed polygons indicate the flakes of NbSe₂, WS₂ and thin graphite. **b** Temperature dependent resistance of sample S3 under different in-plane magnetic fields ($B_{||}$). **c** Tunneling spectra of sample S3 at 1.6 K at zero magnetic field.



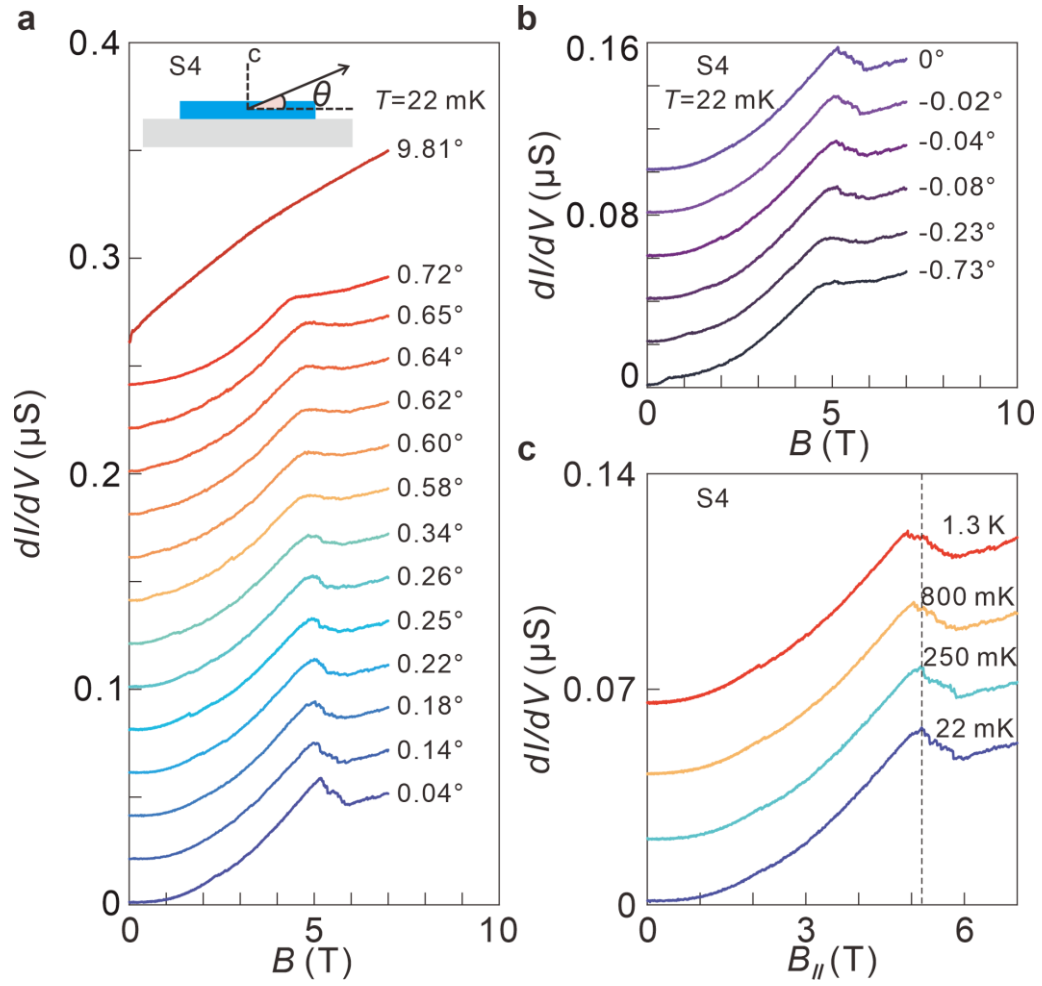
Supplementary Figure 4 Temperature dependence of zero-bias tunneling conductance (ZBTC) in sample S3. a, b ZBTC as a function of $B_{||}$ for sample S3 at a series of temperature points. Data are from sample S3. Solid and dotted curves are obtained from positive and negative sweeps. Curves are vertically offset for clarity. Black arrows connected by a dashed curve in panel **a** indicate the critical magnetic fields where the gap closes. Dashed line in panel **b** marks the first-order phase transition from the uniform Ising superconducting state to the orbital FFLO state based on the data at 1.7 K.



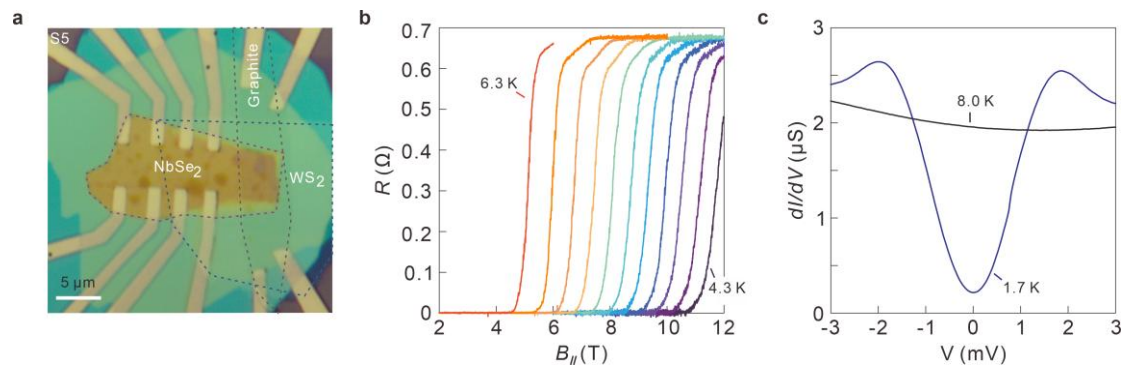
Supplementary Figure 5 Tunneling spectroscopy of sample S4. **a** Optical image of sample S4 (25 nm). **b** In-plane magnetic field ($B_{||}$) dependent resistance of sample S4 at different temperatures. **c** Temperature dependence of the in-plane upper critical magnetic field ($B_{c2}^{||}$) of sample S4. Filled symbols represent the critical fields where the resistance becomes $50\%R_n$ at fixed temperatures. Dashed curve is a theoretical fit to the data around the zero-field transition temperature by using the 2D Ginzburg-Landau (2D G-L) formula. Color shades correspond to the superconducting state (SC). Gray shade indicates the normal state (NS). Red arrow indicates the crossover point. **d, e** Tunneling spectra of sample S4 under different $B_{||}$ at a base temperature of 14 mK. The blue and red triangles mark the coherence peak positions. $B_{||}$ increases in a step of 0.1 T from 4.2 T to 5.0 T (5.1 T to 5.9 T) in the lower (upper) panel of (E). **f** Tunneling conductance at zero bias (top) or at the positions of the superconducting coherent peaks (bottom). Curves are guide to the eye.



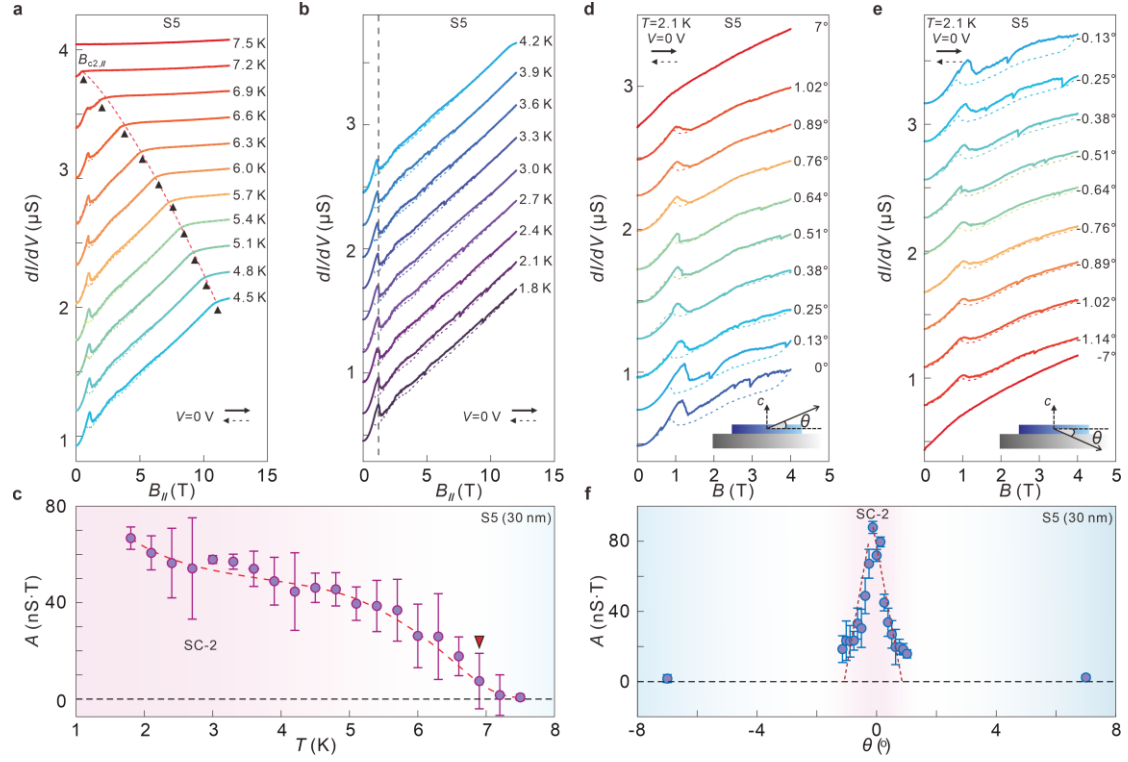
Supplementary Figure 6 Temperature dependence of zero-bias tunneling conductance (ZBTC) in sample S4. *a,b* ZBTC as a function of $B_{||}$ at various temperatures. Data are from sample S4. Solid and dotted curves are from positive and negative sweeps. Curves are vertically offset for clarity. Triangles in **a** (connected by a dashed curve) indicate the gap closing fields. Dashed line in **b** marks the first-order phase transition from the uniform Ising superconducting state to the orbital FFLO state based on the data at 1.6 K.



Supplementary Figure 7 Tilt-field and temperature dependences of zero-bias tunneling conductance (ZBTC) in sample S4. a,b ZBTC as a function of $B_{||}$ at a fixed base temperature of 22 mK with different tilting angles. Data are from sample S4 with positive sweeps of the magnetic field. Inset illustrates the direction of the magnetic field (black arrow) relative to the in-plane axis. **c** ZBTC as a function of $B_{||}$ at different temperatures. Dashed line marks the critical field for the first order phase transition based on the data at 22 mK.



Supplementary Figure 8 Basic characterization of sample S5. a Optical image of sample S5 (30 nm). **b** In-plane magnetic field ($B_{||}$) dependent resistance of sample S5 at different temperatures. **c** Tunneling spectra of sample S5 at 1.7 K and 8.0 K at zero magnetic field.



Supplementary Figure 9 Temperature and tilt-field dependences of zero-bias tunneling conductance (ZBTC) in sample S5. *a, b* ZBTC as a function of $B_{||}$ at different temperatures. Data are from sample S5 (30 nm). Solid and dotted curves are positive and negative sweeps. Black arrows in panel *a* indicate the critical magnetic fields where the gap closes. Dashed line in panel *b* marks the phase transition from the uniform Ising superconducting state to the orb-FFLO state based on the data at 1.8 K. *c* Hysteresis area around the phase transition as a function of temperature. We evaluate the area encircled by the hysteresis loop of ZBTC from 0.5 to 1.5 T. Error bar represents the measurement noise. To evaluate the noise, we first sum up the difference between the positive and negative sweeps from 0 to 0.5 T at a fixed temperature. This value is then rescaled by taking into account the different ranges used for this summation and that for estimating the hysteresis loop area. Red arrow indicates the onset temperature for the hysteresis. *d, e* ZBTC as a function of the total magnetic field at a fixed temperature for different tilting angles (schematically shown in the insets). Data are from sample S5. Solid and dotted curves are from positive and negative sweeps. *f* Tilting dependence of the hysteretic region in ZBTC around 1 T (from 0.5 to 1.5 T). Error bar represents the measurement noise, which is evaluated by the same method described in the caption of panel *c*.

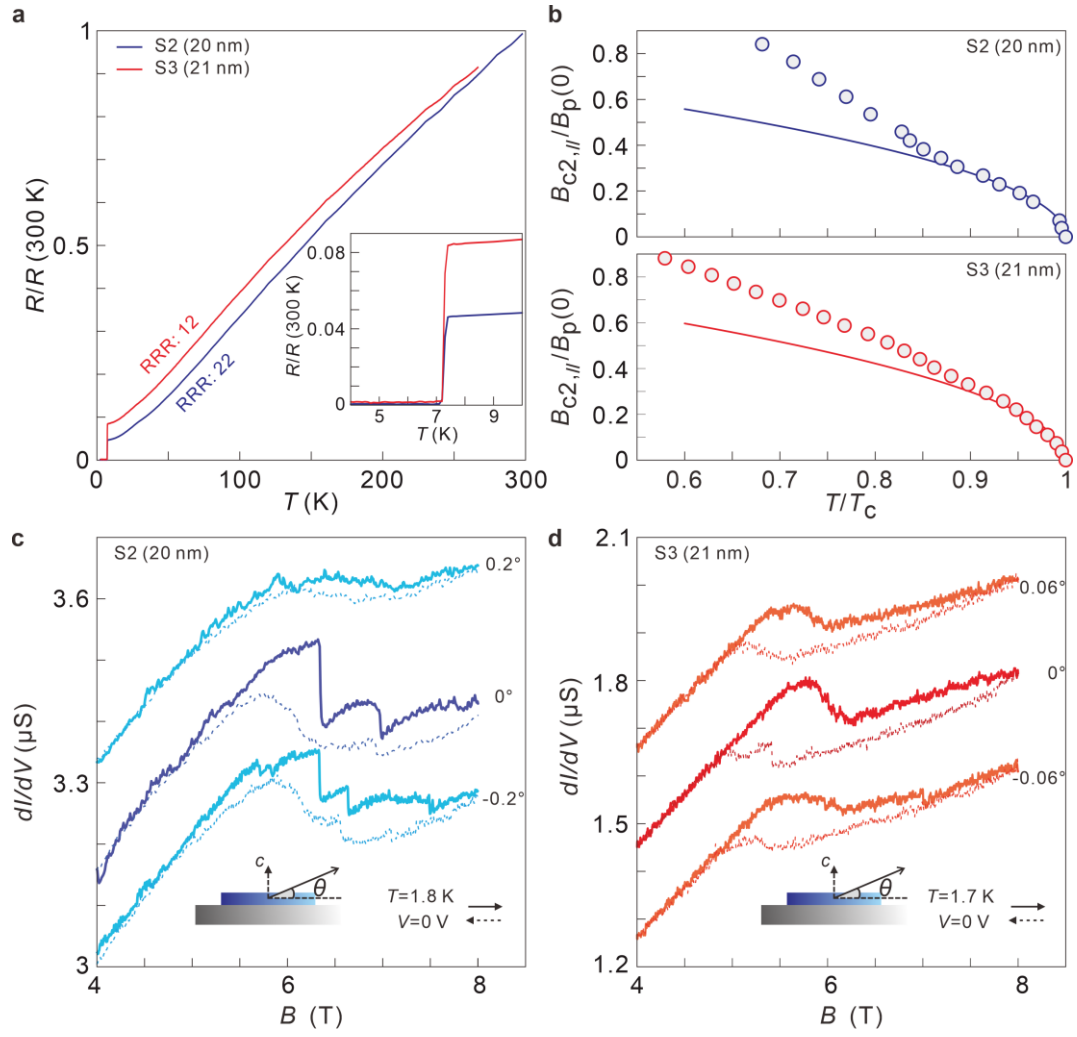
Supplementary Note 4 Influence of sample quality on the sharpness of phase transition

The orb-FFLO state, similar to the Zeeman-type FFLO state, is expected to be sensitive to disorder effect. Here, we compare the obtained phase transition data in samples S2 and S3. These two samples have comparable thicknesses (20 nm and 21 nm). Their superconducting transition temperatures (Supplementary Fig. 10a) and their phase boundaries are nearly the same (Supplementary Fig. 3). However, the residual resistivity ratio (RRR) of sample S2 is 22 while it drops to 12 in sample S3.

Indeed, sample S2 exhibits prominent up-turn in the in-plane upper critical magnetic field (Supplementary Fig. 10b top), whereas the up-turn becomes less discernible in S3. A similar correlation between up-turn in $B_{c2,\parallel}(T)$ and RRR has been reported in the previous study on the orb-FFLO state by resistance measurements⁹.

In the previous study, it was considered that the absence of up-turn in $B_{c2,\parallel}(T)$ indicates the full suppression of the orb-FFLO state. Here, we demonstrate that the phase transition into the orb-FFLO state still occurs in the sample with reduced quality (Supplementary Fig. 10c,d). The sharpness of the phase transition, however, shows clear correlation with the sample quality. Supplementary Figure 10c shows that sample S2 shows several vertical jumps in ZBTC when sweeping the magnetic field upward, representing clear first-order phase transitions. Also, these jumps survive up to a deviation angle of about 0.2 degree. In sharp contrast to these sharp and robust features, sample S3 exhibits only a smooth transition and it becomes weaker by tilting the angle for a tiny amount of 0.06 degree.

In general, the first-order phase transition from our tunneling spectroscopy becomes less prominent as the sample quality drops. This is consistent with the expectation that the orb-FFLO state requires the superconductor to be sufficiently clean.

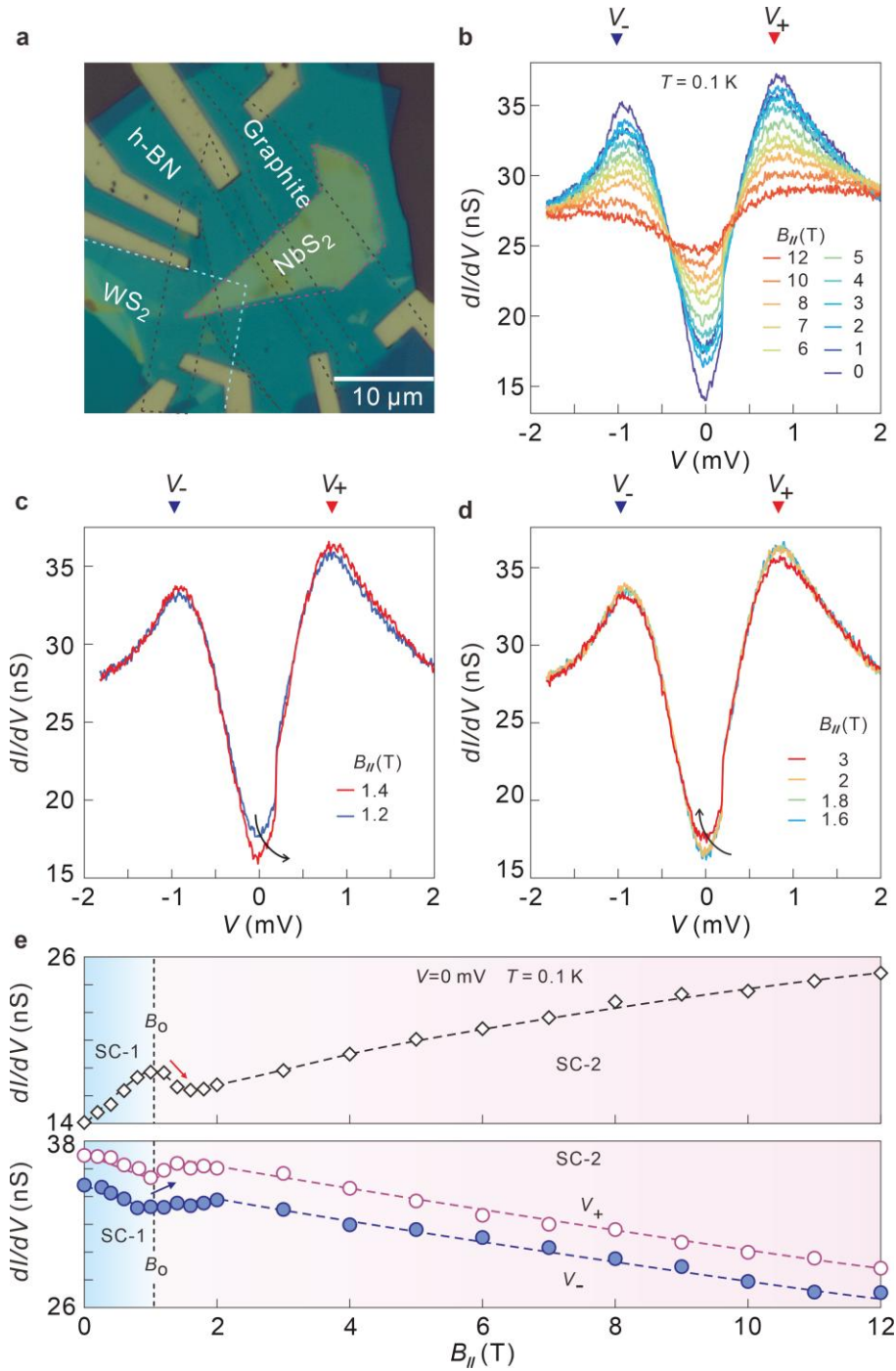


Supplementary Figure 10 Comparison of data from sample S2 and sample S3. a Normalized resistances of samples S2 and S3 as a function of temperature. Inset shows temperature dependent resistances around the superconducting transitions, demonstrating the same transition temperature of the two samples. **b** Temperature dependences of the in-plane upper critical magnetic field of samples S2 and S3. Solid curves show the 2D G-L fitting to the data around T_{c0} . **c, d** ZBTC as a function of the total magnetic field for samples S2 and S3. Tilting angles (indicated by the inset) are close to 0 degree. Curves are offset for clarity.

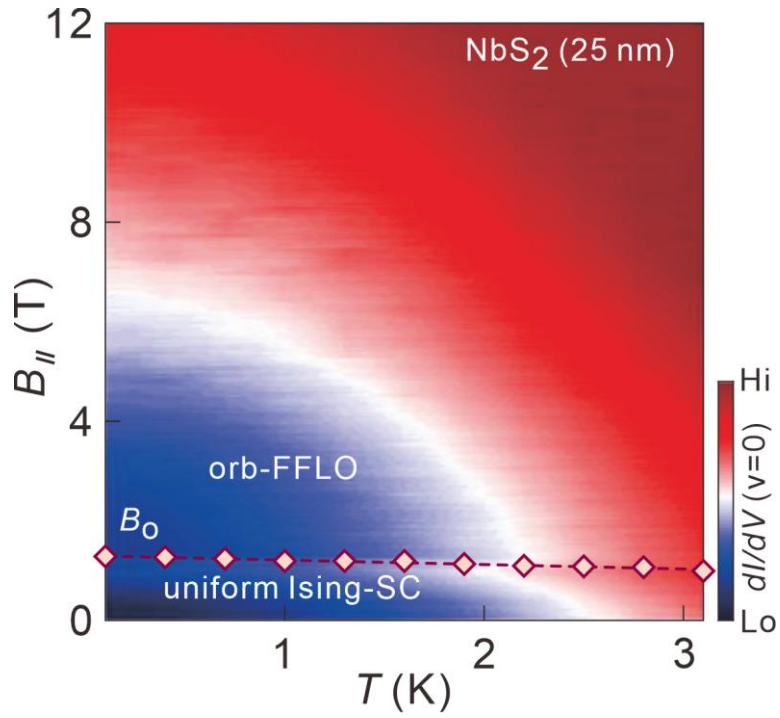
Supplementary Note 5 Phase transition in the tunneling spectroscopy of 2H-NbS₂

We carry out a comparative study on 2H-NbS₂. In contrast to 2H-NbSe₂ hosting charge density wave (CDW), 2H-NbS₂ has been established to have no CDW¹⁰, except for the monolayer case¹¹. Supplementary Fig. 11a shows the optical image of our sample, consisting of a NbS₂/WS₂/few-layer graphene tunnel junction. Here the thickness of NbS₂ is 25 nm. Supplementary Fig. 11b provides the tunneling spectra at 0.1 K. The superconducting gap of about 1 meV is consistent with that reported in previous studies⁹. In a wide magnetic field range, we observe a continuous suppression of the superconducting gap. However, in the small field range, the evolution is non-monotonic. Especially, Supplementary Figure 11c shows that the superconducting gap becomes deeper as the in-plane magnetic field increases from 1.2 T to 1.4 T, similar to that observed in NbSe₂ (Supplementary Fig. 1f for S2, Supplementary Fig. 5e for S4). At higher magnetic fields, the gap shrinks monotonically (Supplementary Fig. 11d). Supplementary Figure 11e presents the extracted differential conductance values at zero bias and at the positions of the two superconducting coherence peaks. It shows consistently the sudden change in the gap at around 1 T.

In Supplementary Fig. 12, we provide the phase diagram obtained from the NbS₂ sample (25 nm thick). This phase diagram is in close analogy to that of the NbSe₂ sample S5 (30 nm thick).



Supplementary Figure 11 Tunneling spectroscopy of a NbS₂ sample. **a** Optical image of the NbS₂-WS₂-Graphite tunneling junction. Dashed polygons indicate the regions of NbS₂, WS₂, and thin graphite flakes. The thickness of NbS₂ is 25 nm. **b**, **c**, **d** Tunneling spectra at 0.1 K and under different in-plane magnetic fields ($B_{||}$). Red and blue triangles indicate the positions of two superconducting coherence peaks. Black arrows in panel (C) indicate the enhanced gap features with increasing $B_{||}$. **e** Zero bias tunneling conductance (ZBTC) (top) or at the positions of the superconducting coherent peaks (bottom). Curves are guide to the eye. B_0 marks the critical magnetic field for the transition.

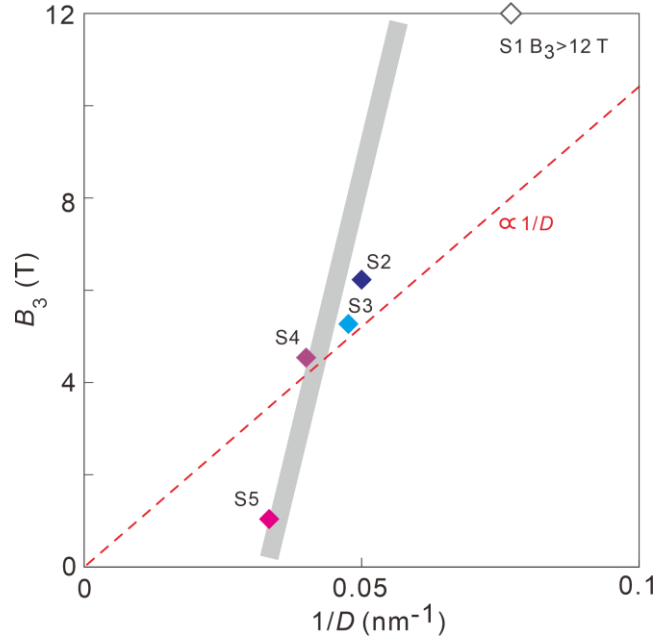


Supplementary Figure 12 Phase diagram of a NbS_2 sample. Color plot of zero-bias tunneling conductance (ZBTC) as a function of in-plane magnetic field and temperature for the NbS_2 sample. Here the data are obtained in the positive field sweeps. Diamonds represent the phase boundary between the uniform Ising superconductor and the orb-FFLO state. Dashed line is a linear fit to the phase transition magnetic fields.

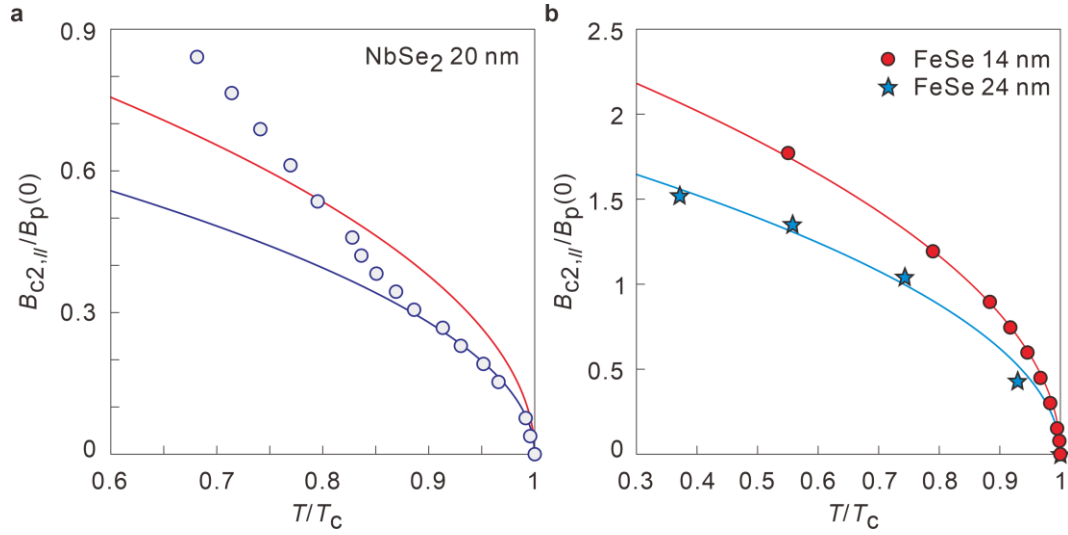
Supplementary Note 6 Extended data on the thickness dependence and comparison between NbSe₂ and FeSe

The vortex entry field B_{en} is inversely proportional to the sample thickness: $B_{en} \propto 1/D$, when the thickness D is much smaller than the penetration depth along the c -axis $\lambda_c \sim 230$ nm (Table I of ref. [12]). This thickness dependence is distinctly different from the thickness dependence of B_o or B_3 (B_o has a slight temperature dependence as shown in Fig. 3 and B_3 is the crossing point of $B_o(T)$ and $B_{c2}(T)$ as shown in Fig. 3a-d). Supplementary Fig. 13 shows that our data points deviate strongly from the $1/D$ dependence. It indicates that our experimentally observed trend disagrees with the expected thickness dependence for B_{en} .

In Supplementary Fig. 14, we draw a comparison between $B_{c2,\parallel}(T)$ measured in NbSe₂ (sample S2) and that measured in FeSe with a comparable thickness. Data points for FeSe flakes are digitized from ref. [13]. No up-turn in the in-plane upper critical field can be discerned for FeSe, despite the fact that FeSe is a well-established multiband superconductor. This comparison suggests that the up-turn in $B_{c2,\parallel}(T)$ is not a universal feature of multiband superconductors.



Supplementary Figure 13 Tricritical field as a function of the inverse of the sample thickness. B_3 as a function of $1/D$. The empty symbol represents Sample S1, which has B_3 that exceeds the maximum magnetic field applied (12 T, as shown in Supplementary Fig. 2). Dashed line indicates $1/D$ dependence that is expected for the entry field of vortices due to the surface barrier effect.



Supplementary Figure 14 In-plane upper critical fields of NbSe₂ in comparison with data of FeSe. **a**, Normalized temperature dependence of the in-plane upper critical fields of sample S2. Blue(red) represents the theoretical fit, by using the 2D G-L formula to fit the data close to T_{c0} (all data points). **b**, Normalized temperature dependence of the in-plane upper critical fields of FeSe. They are taken from ref. [13]. Solid curves are theoretical fits by using the 2D G-L formula.

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