Distinct nodal and nematic superconducting phases in the 2D Ising superconductor NbSe$_2$

Chang-woo Cho$^1$, Jian Lyu$^{1,2}$, Tianyi Han$^1$, Cheuk Yin Ng$^1$, Yuxiang Gao$^1$, Gaomin Li$^2$, Mingyuan Huang$^2$, Ning Wang$^1$, Jörg Schmalian$^3$, and Rolf Lortz$^{1,*}$

$^1$Department of Physics, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong.

$^2$Department of Physics, Southern University of Science and Technology, 1088 Xueyuan Road, Nanshan District, Shenzhen, Guangdong Province, China.

$^3$Institute for Theory of Condensed Matter and Institute for Solid State Physics, Karlsruhe Institute of Technology, Karlsruhe, Germany.

Superconducting transition metal dichalcogenides like 2H-NbSe$_2$ in their two-dimensional form (2D) exhibit a special form of Ising superconductivity in which the quasiparticle spins are firmly pinned in the direction perpendicular to the basal plane. This enables them to withstand exceptionally high magnetic fields far beyond the Pauli limit for superconductivity. We use field-angle-resolved magnetoresistance experiments for magnetic fields strictly rotated in the basal plane to investigate the dependence of the upper critical field ($H_{c2}$) on the orientation of the field in the plane. The field angle dependence of $H_{c2}$ directly reflects the symmetry of the superconducting order parameter. We observe a transformation from a six-fold nodal symmetry near $H_{c2}(T)$ to a two-fold nodeless symmetry at lower temperatures. While the first phase agrees with theoretical predictions of a nodal topological superconducting phase, the observation of a second distinct superconducting phase with nodeless two-fold symmetry is unexpected and contradicts the crystalline symmetry. It may therefore be another example of an unconventional nematic superconducting phase besides doped superconducting Bi$_2$Se$_3$, and we demonstrate that in NbSe$_2$ such a nematic state can indeed arise from the presence of several competing superconducting channels.

Two-dimensional (2D) superconductors provide a rich playground for observing novel unconventional [1-8] and ultimately topological superconducting phases in materials with strongly coupled spin and orbital degrees of freedom [9,10]. The transition metal dichalcogenide (TMDC) 2H-NbSe$_2$ is a textbook model for a strongly anisotropic layered superconductor with a bulk critical temperature of 7.2 K. With the recent interest in two-dimensional materials, it takes the lead as an intrinsic superconductor, which remains superconducting even when exfoliated to monolayers [7,11]. The monolayer has a honeycomb lattice structure similar to that of graphene, but with a broken A-B sub-lattice symmetry and broken inversion symmetry. The presence of heavy transition metal atoms in the structure causes a strong spin-orbit coupling (SOC) [12-14]. In addition, the electrons have their momentum confined in the plane and experience in-plane electric fields from a broken in-plane mirror symmetry. This leads to a SOC field of the form $S(k)\sigma_z$, which is able to pin the electron spins to the out-of-plane direction, where $S(k)$ is a function that depends on the lattice structure and $\sigma_z$ is the z-component of the Pauli matrix [9,15,16]. To distinguish this effect from the in-plane type of SOC (Rashba SOC), which results from a broken out-of-plane mirror symmetry and would pin the electron spins within the 2D planes in the form of $(k_x\sigma_y - k_y\sigma_x)$, this out-of-plane pinning effect is called Ising SOC [4].

Ising SOC in TMDC materials not only has a very strong strength in the order of hundreds of meV, but it also causes many unusual physical properties. One of these is that Ising SOC requires opposite signs for electrons with opposite momentum. Therefore, electrons at opposite $k$-points experience opposite effective Zeeman fields in form of the spin-valley locking effect [17-19]. The

*Corresponding author: lortz@ust.hk
Fermi surface of the monolayer NbSe$_2$ consists of pockets at the $\Gamma$ and the $\pm$K points. Around each point, the Fermi surface is spin split by the Ising SOC. The split is much larger at the $\pm$K points than at the $\Gamma$ point [7]. Due to this strong pinning of the electron spins in the direction outside the plane, the Cooper pairs can be formed in very strong magnetic fields applied parallel to the layer, far beyond the BCS Pauli limit. This special type of superconducting pairing is called ‘Ising superconductivity’ [4].

In addition to the dramatically enhanced $H_{c2}$ in the plane, it has been predicted that Ising superconductivity in the monolayer NbSe$_2$ may feature a topological superconducting phase with six pairs of point nodes on the lines connecting the $\Gamma$ and the M points in the Brillouin zone [10,20,21]. These nodes are protected by an anti-unitary time-reversal like symmetry and are connected by Majorana arcs, similar to the Weyl points in Weyl semimetals being connected by surface Fermi arcs [22-24]. However, relevant experimental gap symmetry studies have rarely been performed to date [25,26] and have not been reported for NbSe$_2$ monolayers. In this Letter, we investigate the gap symmetry of different monolayer and few-layer devices of NbSe$_2$ using field-angle-resolved magnetoresistance measurements with fields applied strictly parallel to the basal plane. The angular dependence of $H_{c2}$ for different orientations of the field with respect to the crystalline directions has been demonstrated to reflect directly the superconducting gap symmetry [27,28]. We find experimental evidence that an in-plane magnetic field can drive the material into two distinct superconducting phases occurring in different temperature and magnetic field regimes. This may include the predicted six-fold nodal superconducting phase [10]. In addition, we observe that at slightly lower temperatures and fields, the nodes open and instead a superconducting phase occurs, which spontaneously breaks the trifold crystalline symmetry in the plane in the form of a two-fold superconducting gap symmetry. The latter appears to be similar to the nematic superconducting phase observed in doped Bi$_2$Se$_3$ [29], albeit with significant differences where a primary single-component order parameter competes with a sub-leading unconventional two-component order parameter.

Results

Three different transport devices have been used in this work. In the main article, we focus on Sample #1. The electrical resistance of Sample #1 in zero magnetic field is shown in Figure 1a. It shows a metallic behavior with a sharp superconducting transition at $T_c = 4.4$ K, defined by the center of the transition (0.5 $R_N$). Although the $T_c$ value is slightly higher than reported for monolayers [7], our local measurements of micro Raman spectroscopy confirm that this device represents a pure monolayer (Figure 1b & c). Data of Sample #2 is included in the Supplementary Information. The device contains monolayer and bilayer regions, as shown by the slightly broadened two-stage superconducting transition corresponding to the critical temperatures of a monolayer and a bilayer [7], respectively (Figure 1d). Data taken within the upper critical field transition corresponding to the monolayer region agree perfectly with the results of Sample #1, thus confirming the reproducibility. Sample #3, which is included in the Supplementary Information, contains different layer thicknesses that lead to a cascade of superconducting transitions due to the different critical temperatures of the regions with different layer numbers (bilayer, tri-layer and bulk). The latter allows us to investigate the thickness dependence of the observed effects.
Figure 1 | Device characterization for the field-angle magnetoresistance experiment. (a) Zero-field electrical resistance of Sample #1. $T_c = 4.4$ K is defined as $0.5 R_N$. (b) An optical image of the device illustrating that the area under investigation in the transport experiment represents a monolayer. (c) Local Raman spectra of the Sample #1 at room temperature at different positions on the sample as marked in (b). For clarity, the spectra were shifted vertically.

In Fig. 2a, we plot resistance data measured in an applied magnetic field of 12 T for Sample #1 for many different orientations of the field in the plane. The inset shows resistance data for a fixed orientation ($192^\circ$) in various applied fields. A significant angular variation is observed. We marked characteristic temperatures at which the resistance reaches a certain value near the lower onset, the midpoint and the upper onset of the superconducting transition through stars. Fig. 2b-d shows these characteristic temperatures as a function of the angular orientation of the field in the plane. Only the filled circles represent measurement data. The open circles have been derived from the same data, but with an angle of $180^\circ$ added to illustrate the full angular dependence. For a few selected angles, we have tested that the reversal of the field leads to the same critical temperature and thus excludes a broken time reversal symmetry. At the upper onset, just before the sample enters the normal state, the angular dependence shows a pronounced six-fold variation with sharp spike-like maxima at $60^\circ$, $120^\circ$ and $180^\circ$, superimposed by a weaker two-fold variation. At lower temperatures (midpoint and lower transition onset), the spikes disappear and the data show a pronounced two-fold symmetry.

In order to analyze the field-angular variations of the characteristic fields in more details, we used fitting functions in form of a combination of the theoretical expectation of a nematic two-fold symmetry (first term in Eq. 1 & 2 [30]) with a simulation of a nodal (Eq. 1) or nodeless (Eq. 2) 6-fold symmetry.

$$H_{c2}(\phi) = \frac{A_2}{\sqrt{\cos^2(\phi + \phi_2) + \Gamma^2 \sin^2(\phi + \phi_2)}} + A_6 \cos(3\phi + \phi_6) \quad (1)$$

$$H_{c2}(\phi) = \frac{A_2}{\sqrt{\cos^2(\phi + \phi_2) + \Gamma^2 \sin^2(\phi + \phi_2)}} + A_6 \cos(6\phi + \phi_6) \quad (2)$$

Here $\Gamma$ is the anisotropy parameter, $A_i$ and $\phi_i$ ($i = 2, 6$) indicate the gap amplitude and phase of the fitting function according to the different symmetries, respectively. The fitting functions are included as the additional lines in Fig. 2b, c & d. It is important to note that a nodal six-fold gap function can only be used to describe the fairly sharp spike-like maxima of the data in Fig. 2b (representing the upper onset of $T_c$ in a field of 12 T). In addition, a two-fold variation must be considered, although its amplitude is weaker than at lower temperatures. At lower temperatures (Fig. 2, c, d) the spikes disappear, which most likely means that the superconducting order parameter becomes fully gapped. The sharp, six-fold spike-like structure is replaced by a fully

\[ H_{c2}(\phi) = \frac{A_2}{\sqrt{\cos^2(\phi + \phi_2) + \Gamma^2 \sin^2(\phi + \phi_2)}} + A_6 \cos(6\phi + \phi_6) \]
gapped, pronounced two-fold symmetry together with a weaker sinusoidal nodeless six-fold variation at lower temperatures.

Figure 2 | Field-angle-dependence of the resistance of monolayer NbSe$_2$. (a) Resistance of Sample #1 in a magnetic field of 12 T for different field orientations in the plane defined by the angle $\phi$. The data were shifted vertically for clearer presentation, except for the 55° data. The inset shows resistance data in 0, 4, 8 and 12 T for a fixed angle $\phi = 192^\circ$. The stars mark characteristic temperatures at which a certain fixed resistance value is reached at the lower onset, the midpoint and the upper onset of the superconducting transition. (b,c,d) Field-angle dependence of Sample #1 of the three characteristic temperatures marked in (a), which illustrates the field-angle symmetries of the resistance data in three different temperature ranges. Only the filled circles are real data, while the open circles represent the same data with an angle of 180° added to illustrate the full angular dependence. The blue lines represent fitting functions considering a 6-fold nodal symmetry, (b) a 6-fold nodeless symmetry (c,d) and two-fold nematic symmetries (b,c,d). (e,f) Field-angle dependence of the upper critical field $H_{c2}$ obtained from magnetoresistance data of Sample #2 (see Supplementary Information for details) at 3.7 K (e) and 2.3 K (f). The blue lines represent fitting functions considering a 6-fold nodal symmetry (e), a 6-fold nodeless symmetry (f) and a two-fold nematic symmetry (e,f).

Note that a very similar behavior with two distinct field regions, in which there is 6-fold nodal and 2-fold nodeless symmetry, respectively, is observed in our second monolayer device Sample #2, for which the magnetoresistance data are shown in the Supplementary Information (Fig. S0). For this sample, we obtained better results in measuring the magnetoresistance at fixed temperatures as a function of the magnetic field, allowing us to determine the upper critical field $H_{c2}$ for various directions of the field in the basal plane. The resulting field angle dependence of $H_{c2}$ is shown in Fig. 2 e & f. At 3.7 K, the angular dependence of $H_{c2}$ shows very similar peaks with a 6-fold rotational symmetry as the critical temperature of Sample #1 in Fig 2b, while at 2.3 K a pronounced 2-fold symmetry appears, similar to Fig. 2 c & d, demonstrating the reproducibility of our results in two separate devices.

We attribute the six-fold sharp kink-like dependence of the critical field to the theoretically predicted nodal superconducting phase [10], which according to our data appears in a narrow regime near $H_{c2}(T)$ in accordance with the theoretical predictions. However, the two-fold variation of the broad $H_{c2}$ transition at lower fields and temperatures is unexpected. Since a two-fold symmetry breaks the trifold symmetry of the NbSe$_2$ basal plane, this can be seen as a nematic superconductivity similar to the nematic superconducting phase in doped Bi$_2$Se$_3$ [29].
The resistance data for the directions in which the nodal kinks occur show small bumps around 3.6 K, followed by a weaker increase slightly below the onset of the resistive transition (Fig. 3). The reduced slope above causes the spikes in the field-angular dependence shown in Fig. 2b. The bumps may be an indication of a phase transition just before the upper critical field transition or at least an abrupt change of some superconducting characteristics. These bumps are absent in the anti-nodal directions. This is particularly evident in the temperature derivative of the resistance shown in the inset of Fig. 3, where sharp peaks at 3.5 K occur in $dR(T)/dT$ for the nodal directions but not for the anti-nodal direction.

Figure 3 | Difference in selected resistance data of Sample #1 for fields applied along the nodal and the anti-nodal direction in a field of 12 T. Additional bumps appear around 3.6 K for magnetic fields applied along nodal directions (120° and 185°), which are absent for fields applied along anti-nodal directions (100°). The inset shows the corresponding temperature derivative of the resistance. The bumps in the resistance for the nodal directions cause sharp peaks at 3.6 K.

Data from Sample #3 containing contributions from different layer thicknesses are presented in the Supplementary Information. The lowest resistance jump corresponds to a bilayer region in the sample (Fig. S1). The field-angle dependence of this bilayer is significantly weaker than that of Samples #1 and #2 and is dominated by a weak six-fold variation (Fig. S2). The variation is too weak to distinguish a nodal from a node-less symmetry. At the center of the partial superconducting transition at 4.4 K, an even weaker anisotropic structure is formed, which we attribute to the superconducting transition of the trilayer region in the device. Partial superconducting transitions, which occur at even higher temperatures and are assigned to thicker sample regions, show an almost isotropic behavior, as is expected for the bulk limit. Sample #3 thus shows that the observed strong distinct anisotropies are exclusively found in NbSe$_2$ monolayers, while multilayers have much weaker anisotropies.

Discussion
We observe the spikes of the 6-fold rotational symmetry in the field angle dependence of the magnetoresistance only within the field or temperature range near the resistive upper critical field transition. At the points where zero resistance is gradually approached at lower temperatures (Fig. 2c & d) the kinks are absent and the field angle dependence is dominated by a pronounced two-fold symmetry. This suggest that there may be a region near the $H_{c2}$ boundary dominated by a six-
fold nodal superconducting gap symmetry. This is consistent with the theoretical predictions in Ref. [10], where a six-fold nodal topological superconducting phase is induced by high magnetic fields applied parallel to the basal plane of monolayer NbSe$_2$. The nodes represent 6 pairs of nodes on the two $\Gamma$ pockets along the three $\Gamma$-$M$ lines in the Brillouin zone, which are protected by the time-reversal-like symmetry [21]. Ising SOC is absent along these directions, so that the Zeeman energy can exceed the pairing energy of the spin-singlet Cooper pairs, thus closing the gap and forming point nodes [10]. In a recent theoretical work [21] it has been shown that there are two instabilities in singlet and triplet interaction channels leading to a crystalline topological superconducting phase, which involves the nodal phase described in Ref. 10. Such a nodal crystalline topological superconducting phase has particle-hole symmetry and an anti-unitary time reversal-like symmetry, the latter being a composition of time-reversal symmetry and a reflection with respect to the $xy$-plane [21]. The point nodes in the predicted topological superconducting phase should be connected by Majorana flat bands [10], this stimulates more direct spectroscopic experiments to detect them and thus to verify the possible topological nature of this phase.

![Diagram](image)

**Figure 4 | Assumed magnetic field vs. temperature phase diagram (a).** The horizontal line marks the Pauli limit $H_P$. The star marks the regions in which we observe evidence of a six-fold nodal (red) or two-fold node-less order parameter symmetry, respectively. b Schematic representations of the monolayer structure of NbSe$_2$. c Field-angle-dependent upper critical field measurement in the plane with respect to the crystalline structure, defining the field orientation in the plane with respect to the $x$-axis by the angle $\psi$. d Temperature dependence of the primary and secondary order parameter attributed to the nematic superconducting state (see text for details).

In Fig. 4 we summarize our results in the form of an $H$-$T$ phase diagram for the monolayer NbSe$_2$, which is inspired by Ref. 10. The stars indicate the regions in which we observe evidence for the phase with six-fold nodal gap symmetry. An additional node-less phase with dominant two-fold rotational symmetry is observed instead at lower fields and temperatures. The latter phase is unexpected. While we do not have sufficient data in high magnetic fields beyond 12 T and our transport data do not provide any information in the zero resistance range at low fields and low temperatures, the exact phase diagram remains unclear. However, our data can be plausibly explained by a fully gapped nematic superconducting phase, which occurs in a regime below a high-field nodal region in the phase diagram.
Nematic superconductivity means that a rotational symmetry breaking occurs, which is closely related to the onset of superconductivity. In the following we present a scenario, which naturally accounts for the observations presented in Fig. 2, and discuss the details of the phase diagram (Fig. 4a). It implies that there are several competing superconducting channels in NbSe$_2$: a dominant single-component (likely $s$-wave) pairing state and a sub-leading, unconventional two-component order parameter, which leads to nematicity because of the symmetry-permitted nonlinear coupling between both pairing states (Fig. 4d).

In a nematic superconductor, the onset of superconductivity not only breaks the global $U(1)$ symmetry of the pairing state, but also a discrete rotational symmetry of the crystalline lattice, a symmetry breaking that can even occur above the actual superconducting phase transition [31,32,33]. In order for such a nematic state to exist, the superconducting order parameter must transform according to a two-dimensional irreducible representation of the point group. The point group of monolayer NbSe$_2$ is $D_{3h}$ if the layer exists in free space. On a substrate, the horizontal mirror plane disappears as an element of symmetry and the resulting group becomes $C_{3v}$. In $C_{3v}$ there is a two-dimensional irreducible representation $E$ with leading polynomials $(k_x, k_y)$, or $(k_x^2 - k_y^2, k_xk_y)$. A superconductor that orders according to this $E$ representation would then be characterized by

$$\Delta_{ab}(k) = \psi_1(D_{x^2-y^2}(k) + d_x(k) \cdot \sigma)i\sigma_y + \psi_2(D_{xy}(k) + d_y(k) \cdot \sigma)i\sigma_y$$

(3)

Here $\Delta_{ab}(k) = \langle c_{ka}c_{-kb} \rangle$ describes the Cooper pair with crystal momentum $k$ and spin $\alpha, \beta$. $D_{x^2-y^2}(k) \propto \cos k_x - \cos k_y$ and $D_{xy}(k) \propto \sin k_x \sin k_y$ describe singlet pairing amplitudes while the $d_x, d_y \langle k \rangle$ describe the triplet component, which is allowed given the broken inversion symmetry at the interface. The pairing is then characterized by the two-component order parameter $\psi = (\psi_1, \psi_2)$. A nematic state has the helical form $(\psi_1, \psi_2) \propto (\cos \theta, \sin \theta)$. An immediate concern of such a pairing state is that one would expect pairing states in bulk NbSe$_2$ that naturally merge with the $E$ representation on the surface. After all, the bulk and surface transition temperatures are rather comparable. These pairing states would then be either of $E_{2g}$ or $E_{1u}$ symmetry with the bulk point group $D_{6h}$. There seems to be no evidence for such behavior in the bulk material. Another, arguably more fundamental objection against primary nematic superconductivity in monolayer NbSe$_2$ follows from the analysis of Ref. 34, where monolayers with broken inversion symmetry were considered in the limit where the spin splitting due to the inversion symmetry breaking is larger than the superconducting gap, a condition which is clearly satisfied in our case. It has been shown that superconductivity with higher-dimensional irreducible representations, such as $E$, always breaks time reversal symmetry, i.e. $(\psi_1, \psi_2) \propto (1, \pm i)$, instead of being nematic.

A natural explanation for the observed behavior is that the primary superconducting order parameter $\varphi$ is a single-component degree of freedom. However, in addition to this primary superconducting order parameter, pairing in the $E$-symmetry channel, i.e. with the pairing wave function given in Eq. 3 with the two-component order parameter $\psi = (\psi_1, \psi_2)$, is a close contender. Then, the symmetry properties at the interface allow a phase transition $T_{nem} < T_c$ at zero field (Fig. 4a), where $\psi$ becomes finite and enters a nematic state (Fig. 4d). This nematic order is a direct consequence of the nonlinear coupling between the two almost degenerated order parameters. The most conservative choice would be $s$-pairing in the $A_1$ symmetry. Then a coupling of the type

$$f_{int} = \frac{g}{4}[\varphi^*(\psi_1|\psi_1|^2 - \psi_2\psi_1^*\psi_2 - 2\psi_1|\psi_2|^2) + h.c.]$$

(4)

is symmetry allowed and induces a nematic state which is either $\psi^{(1)} = \psi_0(1,0)$, $\psi^{(2)} = \psi_0\left(-\frac{1}{2}, \frac{\sqrt{3}}{2}\right)$, or $\psi^{(3)} = \psi_0\left(-\frac{1}{2}, -\frac{\sqrt{3}}{2}\right)$ with amplitude $\psi_0 \propto \varphi$ below a first order transition at
A similar behavior occurs if $\varphi$ is odd under vertical mirror reflections, i.e. an $A_2$ order parameter, which yields instead

$$f_{\text{int}} = \frac{g}{4} [\varphi^*(\psi_2 \psi_2)^2 - \psi_1 \psi_2^* \psi_1 - 2\psi_1 \psi_2^* \psi_2] + h. c. \]$$

with similar nematic order. These nematic states occur for a sufficiently large coupling constant $g$, despite the fact that the secondary order parameter $\psi$ prefers, on its own, a chiral state with time-reversal symmetry breaking [34]. Interestingly, a coupling term like Eq. 4 has recently been discussed in the context of nematic superconductivity in twisted bilayer graphene [35]. If there are two almost degenerated order parameters that transform simultaneously under $A_1$ and $E$, an in-plane magnetic field plays an important role in mixing these two states at any temperature below $T_c$ (Fig. 4d). This is a consequence of the coupling $\propto i(\varphi^* B_x \psi_1 + \varphi^* B_y \psi_2 - h. c. )$. It allows to induce the nematic state anywhere below $T_c$. Only directly at the transition temperature the nematic effects are absent, which explains the observed six-fold symmetry, which is shown in Fig. 2. This magnetic field coupling term is even under time reversal, is gauge invariant and compatible with all point symmetries. Within a microscopic theory, the physics underlying this coupling was recently discussed in Ref. 36.

To summarize, the observed two-fold symmetry below $T_c$ reveals that this material is characterized by a primary order parameter, likely of $s$-wave nature, and by a close competitor that transforms according to a non-trivial symmetry and has two components. The nonlinear coupling between the two order parameters induces a nematic state at $T_{\text{nem}} < T_c$ and pins a two-fold symmetry axis in the system. An in-plane magnetic field induces nematic order at temperatures even above $T_{\text{nem}}$, all the way up to $T_c$.

A natural question is whether the near degeneracy is also present in bulk samples and if so, why it does not induce a nematic state as well. One answer to the second question might be that the near degenerate pairing state in bulk NbSe$_2$ is in fact a $p$-wave triplet state with $E_{1u}$ symmetry. For such a state no coupling of the type given in Eq. 4 is symmetry allowed and therefore no induced nematicity is expected. Hence, one of the implications of our findings is that bulk NbSe$_2$ should have a dominant $s$-wave order parameter with a potential sub-leading $p$-wave triplet channel, a prediction that could be probed by optical excitation of the sub-leading order in analogy to the Bardasis Schrieffer mechanism [35].

**Conclusions**

To conclude, our field-angle-resolved magnetoresistance measurements reveal a pronounced in-plane anisotropy in monolayer 2H-NbSe$_2$ when a magnetic field is applied strictly to the layer. Such an anisotropy is directly linked to the superconducting order parameter [27,28]. Close to the onset of the transition, just before the sample becomes normal conducting, a sharp six-fold nodal symmetry occurs, which agrees perfectly with the theoretical predictions of a six-fold crystalline nodal topological superconducting phase in the vicinity of the in-plane upper critical field of an Ising superconductor [10,20,21]. At lower fields but still within the broad resistive upper critical field transition the nodes open and the order parameter anisotropy becomes dominated by a strong 2-fold gapped symmetry, which is at odds with the tri-fold crystalline symmetry. This appears to be similar to the nematic superconducting state known from doped Bi$_2$Se$_3$ [29], and we have shown that such a nematic state can be created by the presence of several competing superconducting channels with a leading single-component pairing state and a sub-leading unconventional two-component order parameter with nonlinear coupling between both pairing states. These exotic superconducting phases are caused by the strongly coupled spin-orbital degrees of freedom, complex inter and intra-band interactions [21], and the broken inversion symmetry.

After completion of this work we became aware of a preprint that reported a similar two-fold
breaking of rotational symmetry in few-layer NbSe$_2$ [37], which further confirms our experimental results. In this work, the ability to induce nematic order with an in-plane magnetic field even above $T_{\text{nem}}$ was also discussed.

Methods
Sample Preparation
The ultra-thin 2H-NbSe$_2$ samples on a silicon substrate were fabricated using the standard exfoliation method. The sample was first exfoliated onto PMMA and transferred directly onto pre-patterned 10 nm/50 nm Ti/Au terminals on SiO$_2$/Si substrate defined by electron-beam lithography and e-beam evaporation. Few-layer NbSe$_2$ is known to degrade rapidly in air, thus to protect our samples, the PMMA layer used in transfer was kept as a capping layer, and we carried out all experiments in high vacuum. Between the experiments, the sample was short-circuited using an electronic switch to protect the sample from charging effects.

Micro Raman Spectroscopy
To determine the thickness of the NbSe$_2$, we performed micro Raman spectroscopy measurements in different regions of the 2D samples. These Raman measurements were carried out with a HeNe 632.8 nm laser at room temperature. The beam was focused to a diameter of 1 um onto the sample by a 50 times magnification objective. Details of the number of layers are defined from their characteristic shear mode frequencies as shown in the main article.

Electrical and Magneto-Resistance Measurements
Electrical resistance and magneto-resistance measurements were performed in an AMI cryostat down to 1.5 K. Magnetic fields were applied strictly parallel to the basal plane of NbSe$_2$ up to 12.5 T. All resistance data was collected by a four-probe AC method with current of 20-100 nA measured with a lock-in technique with an AC modulated current at a frequency of 10-20 Hz. A low-noise band-pass filter was used to achieve sufficient experimental resolution. The measurements were performed at slow field or temperature sweep rates, which provides a long integration time for signal averaging. The ultra-thin NbSe$_2$ sample on a Si substrate was mounted on an Attocube piezo rotator, which allowed us to perform in-plane field-angle-resolved magneto-resistance measurements with high accuracy.

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**Author contributions**

This work was initiated by R.L., the devices were prepared by T.H. under supervision of N.W., C.w.C., J.L., G.L. and M.H. have conducted the Raman characterization of the devices, C.w.C., and J.L. have carried out the field angle resolved magneto-transport measurements with help of C.Y. N. and Y. G., C.w.C., J.L. and R.L. have analyzed the data, J.S. has contributed the theoretical analysis. The manuscript was prepared by C.w.C., J.S. and R.L, all authors were involved in discussions and contributed to the manuscript.

**Competing financial interests**

The authors declare no competing financial interests.
Distinct nodal and nematic superconducting phases in the 2D Ising superconductor NbSe$_2$

Chang-woo Cho$^1$, Jian Lyu$^{1,2}$, Tianyi Han$^1$, Cheuk Yin Ng$^1$, Yuxiang Gao$^1$, Gaomin Li$^2$, Mingyuan Huang$^2$, Ning Wang$^1$, Jörg Schmalian$^3$, and Rolf Lortz$^{1,*}$

$^1$Department of Physics, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong.

$^2$Department of Physics, Southern University of Science and Technology, 1088 Xueyuan Road, Nanshan District, Shenzhen, Guangdong Province, China.

$^3$Institute for Theory of Condensed Matter and Institute for Solid State Physics, Karlsruhe Institute of Technology, Karlsruhe, Germany.

Table S1 provides an overview over the devices used in this study. Sample #1, for which the data is included in the main text, is dominated by a monolayer region with the electrodes arranged so that only this region is probed in the magneto-transport experiments. Sample #2 was found to be a mixture of a mono- and a bilayer, while Sample #3 contains bi-layer, tri-layer and thicker regions, which all show up in form of a cascade of superconducting transitions due to the different critical temperatures of the regions with different layer thickness. The latter allowed us to probe the field-angular dependence of thicker samples.

| Sample | $T_c$ (0.5$R_N$) | Number of layers |
|--------|-----------------|-----------------|
| Sample #1 | 4.4 K | Monolayer |
| Sample #2 | 3.7 K | Mono / bilayer |
| Sample #3 | 4.7 K(2), 5.9 K(3), 7.2 K(Bulk) | Mixture (bilayer + 3-layer + bulk) |

Table S1 | Summary of the 2D NbSe$_2$ devices used in this study. The critical temperature $T_c$ is determined by the midpoint of the resistive transition (0.5$R_N$).

Sample #2 is a mixture of a monolayer and a bilayer as shown by the slightly broadened two-stage superconducting transition corresponding to the critical temperatures of a monolayer and a bilayer [6], respectively (Fig. S1a). The device was particularly small and the local Raman spectra (Fig. S1c in combination with the optical image in Fig. S1b) was not able to provide separate indications of the different regions.

* Corresponding author: lortz@ust.hk
Figure S1 | Device characterization for the field-angle magnetoresistance experiment. (a) Zero-field electrical resistance of Sample #2 exhibiting a characteristic double transition attributed to mixed mono and bilayer behavior (b) Optical image of the device and (e) local Raman spectra at room temperature of Sample #2 recorded at different positions as indicated by the numbers in (b). The Raman spectra provide only evidence of a bilayer, while the double transition in resistance in (a) suggests that the two layers are partially separated or loosely bonded. For clarity, the spectra were shifted vertically.

In Fig. S2 we plot magnetoresistance data at two different fixed temperatures (2.3 and 3.7 K) for selected field orientations in the plane for Sample #2. It should be noted that Sample #2 is a mixture of monolayer and a bilayer, but the bilayer has a lower anisotropy than the monolayer and thus any anisotropy observed is attributed to the monolayer, which is further confirmed by comparison with the data of Sample #1 (pure monolayer) and #3 (bilayer, trilayer and bulk). At 3.7 K (a) and 2.3 K (b), a significant field-angle variation can be observed in the high magnetic field regime. We derive characteristic fields from both data sets, which refer to the upper critical field \( H_{c2} \), and draw their field-angle dependence in Fig. S2c and d. At a lower temperature of 2.3 K (lower panel), the onset point \( (H_{c0}) \) of the broad upper critical field transition is determined from the zero crossing point of linear fits in high fields where zero resistance is lost (Fig. S2a). At the higher temperature of 3.7 K (top panel), we used another criterion to define the upper critical field \( H_{c2} \), since at this temperature we are already in the middle of the resistive transition of the monolayer where the zero resistance is lost. Thus, \( H_{c2} \) was used to see the angle-dependent variation defined as the field in which 90% of the normal state resistance \( (0.9 \, R_N) \) is restored. A significant amplitude variation is also found for Sample #2 in the field-angle variation with a distinct behavior at the two temperatures. At 3.7 K sharp peaks appear in \( H_{c2}(\phi) \) at 60° and 120° (Fig. S2c). At a lower temperature of 2.3 K, the peaks are replaced by a pronounced two-fold variation (Fig. S2d). This behavior agrees all perfectly with the results of Sample #1 and confirms the reproducibility of our results.

![Figure S2](image_url)
functions considering 6-fold nodal symmetries (c), 6-fold nodeless symmetries (d) and two fold nematic symmetries (c,d).

The characterization of the device Sample #3 is shown in Fig. S3. White lines in the optical image in Fig. S3a illustrate the different regions of different thickness, which were identified with help of the local micro-Raman spectra (Fig. S3c). The main regions between the six contacts is represented by a bi-layer, but also a tri-layer region is extending in between the two upper voltage probes and thus will be probed as a parallel resistance. In addition, there is a thicker region which is marked as ‘bulk’. Fig. S3b shows the zero field resistance of the sample in which a cascade of at least three superconducting transitions is visible. The lowest two steps occur at the characteristic temperatures known from bi-layer and tri-layer NbSe₂, respectively [7], and can be attributed to the corresponding regions marked in the optical image. The upper one occurs at the same temperature as $T_c$ in bulk samples and is attributed to the third region.

Figure S3 | (a) Optical image of Sample #3. Three different areas with different thickness can be seen, marked as ‘2L’ (bilayer), ‘3L’ (trilayer) and ‘Bulk’. (b) Zero field electrical resistance as a function of temperature. Each step corresponds to the superconducting transition of an area of different thickness: bulk, trilayer and bilayer. (c) Raman spectra recorded on the three regions of different thickness, confirming their layer numbers. The background data were recorded on a bare Silicon substrate without NbSe₂.

In Fig. S4 we show the field-angle dependence data for Sample #3. Fig. S4a shows the electric resistance for 4 selected orientations with respect to the in-plane direction of a 12 T magnetic field
applied strictly parallel to the layers. While there is a certain dependence on the in-plane angle, it is obvious that the angular variation is much weaker than for Sample #1 and Sample #2. In Fig. S4a & b we plot the resistance as a function of the in-plane direction of the magnetic field at various characteristic fixed temperatures.

**Figure S4** | (a) Representative field-angle resistance data at a fixed magnetic field of 12 T for Sample #3. (b) Field-angle dependence of the resistance at 12 T in the normal state (6.5 K). A weak six-fold symmetry is visible, which was not obvious for Sample #1 and #2 and is therefore attributed to the bulk NbSe$_2$ region. (c) Detailed field angle dependence of the normalized resistance, where we distinguish three different regimes based on each step-like resistive transition. No significant angular dependence can be observed in the bulk superconducting phase. For the temperature regimes dominated by the superconducting transitions of the tri- and bilayer some weak angular variations occur, which can be fitted with a combination of a two-fold and 6-fold node-less symmetry. However, all data of sample #3 show a significantly lower anisotropy compared to Sample #1 and #2.