Peer Review File

Manuscript Title: Magnetic memory and spontaneous vortices in a van der Waals superconductor

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

The authors study a layered structure made of alternating superconducting-Mott insulator layers, known as 4Hb-TaS2. This material has recently risen as a potential candidate for unconventional superconductivity. In the superconducting state below Tc, the system shows a spontaneous vortex phase. The arrangement of those vortexes is shown to be controllable in the normal state, strongly suggesting that the system shows a hidden magnetic ordering above the superconducting Tc. This remarkable behavior is rationalized as stemming from the interplay of a potential chiral spin liquid state of the Mott insulator, coupled to a chiral superconducting state of the superconducting layers.

First of all, I would like to emphasize that I find the results of the authors greatly interesting and that they provide a remarkable finding in the physics of 4Hb-TaS2. Their manuscript is very well written, and their results are compatible with the previous results in 4Hb-TaS2. In particular, the observation of hysteresis in the spontaneous vortex phase of this material represents a remarkable finding for the field of van der Waals materials and unconventional superconductivity. I strongly believe that their results provide a pioneering experiment in the field of van der Waals quantum materials. Therefore, for the reasons stated above, I strongly recommend their manuscript for publication in Nature.

Apart from my positive recommendation for publication in Nature, I would like to mention two points that the authors may optionally consider addressing in their revised manuscript.

My first suggestion is related to the interpretation of their results in terms of a coupling between a chiral spin liquid and a chiral superconducting state. First of all, I would like to emphasize that such a picture would be, of course, highly suggestive and consistent with preliminary results in the field. Nevertheless, as the authors well know, assessing the potential spin-liquid nature of some of the Mott layers is a greatly open challenge, and even knowing the potential spin-liquid nature of a Mott monolayer remains an open problem. Assessing also the chiral superconducting nature of the superconducting layers is a nearly remarkable challenge. Therefore, while their picture would be consistent with their observed results, assessing the validity of their assumptions would, of course, require further future work. Given those points, I would suggest to the authors that they reflect those uncertainties in their discussion, for example, by adding after “For both cases, further work is required to provide a complete microscopic description of the effect.” a statement similar to “We finally note that uniquely assessing the potential chiral spin liquid and chiral superconducting nature of the layers remains a remarkably open problem, and demonstrating the picture above will require further future work.”

My second suggestion is related to potential alternative mechanisms. I want to note that the authors already nicely explain several mechanisms they discard during their discussion, and thus this suggestion only extends an already satisfactory discussion. In particular, I wanted to ask the authors if potential magnetoelastic effects in the system could be discarded as a mechanism for their observations. While I am fully aware that assessing this is a remarkable challenge, I would suggest that at this stage, it would be enough that the authors add one statement about whether magnetoelasticity can be discarded or if it could be considered as a future candidate for the phenomenology they observe.

To summarize, I believe that the current manuscript puts forward a remarkable finding in van der Waal materials, demonstrating the emergence of hidden order in 4Hb-TaS2. Therefore, given all the points above, I strongly recommend the publication of their manuscript in Nature.

Referee #2 (Remarks to the Author):

In the manuscript by Persky et al., experiments are described on stacks of superconducting and Mott-insulating (potentially spin-liquid) materials. These stacks take the form of Van der Waals heterostructures, denoted as 4Hb-TaS2, in which alternatingly superconducting '2H' TaS2 and Mott-insulator '1T' TaS2 layers are stacked. These have different crystallographic properties resulting in their different electronic transport characteristics. The superconductor phase has a critical temperature of 0.7K.

The key finding of the manuscript is that when these stacks are cycled in an applied magnetic field, from below 0.7K to a temperature below a treshold temperature of 3.6K, and subsequently cooled down to below 0.7 in zero applied field, a residual magnetization is observed from a measured amount of Abrikosov vortices penetrating through the superconducting layers. These measurements are performed by Scanning SQUID Microscopy (SSM).

This magnetization is absent when the materials is cooled down in zero field from a temperature above 3.6K. The remnant field observed by Scanning SQUID Microscopy depends on the peak temperature in the above-mentioned temperature cycle and on the magnetic field applied during the field-cooled stage of the measurement procedure.

As no traces of ferromagnetism are discerned by SSM in the heterostructures, the magnetic memory effects displayed by the system are presented as the discovery of a new form of hidden magnetism in the heterostructure in the temperature range below 3.6K.

The manuscript is interesting and the measurements are original and well described. They also nicely demonstrate the power of Scanning SQUID Microscopy for such investigations.

I am left however with a number of questions about the presented work and conclusions, before I can recommend on the publication of the work in Nature.

Can the authors completely rule out artifacts arising from magnetized materials near the sample or from flux trapping in the SQUID chip? It would be good if the authors could show that in pure 2H-TaS2 superconductors, or similar representative control samples, the effects do not occur. Then it would be clear that they arise exclusively because of the presence of and/or coupling with 1T-TaS2.

Similarly, it would be good if the authors could show the results of experiments in which first a field is applied in the temperature range 0.7K - 3.5K (thus above the superconducting transition), then the field is taken away and subsequently the sample is cooled down to below the superconducting transition temperature. Does it then also show the remnant magnetization? If yes, it means that the cause of the 'hidden magnetism' is not related to superconductivity. If no, it would show that the superconductivity has a direct role to play in it. In either cases, the generation of the Abrikosov vortices from a hidden magnetism in the 1T layer could still involve a coupling via a chiral superconductor phase.

Is there a decay over time of the hidden magnetism? This could be probed by varying the time between switching off the magnetic field and starting the cooldown, which was now held at 3 minutes according to the information provided in the Methods section.

Referee #3 (Remarks to the Author):

Persky et al. used the scanning SQUID microscopy to uncover the magnetism hidden in the superconductor 4Hb-TaS2. which hosted superconductivity and chiral spin liquid (CSL) due to the overlaying crystal structures of 2H-TaS2 and 1T-TaS2. They reported the observation of spontaneous vortices in zero field cooling, following some field cooling and heating sequences. They also showed a magnetic-like hysteresis following a specific measurement sequence. Considering there is no long-range magnetic order in the normal state of this system, authors claim that the time reversal symmetry is broken due to the coupling between superconductivity and CSL. The material system and the claimed physics is indeed interesting and the technique is appropriate. However, I do not find the evidence convincing enough for me to recommend publication. The following are my major concerns.

1. The zero-field cooling is not strictly zero. We can still see a vortex in Figure 1.e #1, which was obtained after ZFC. This fact greatly weakens the claim of ‘spontaneous vortex’ in the ‘zero-field’.

2. The evidence for time-reversal symmetry breaking (TRSB) is not consistent with the interpretation. The authors argue that the superconductivity together with CSL leads to TRSB. If so, the memory of the chirality of TRSB should be erased above the superconducting Tc. However, the hidden magnetism has a higher critical temperature of 3.6 K than the superconducting Tc of 2.7 K.

3. The relatively close T* and Tc raises some flags.
a) In this work, the superconducting Tc of the sample is determined by bulk transport measurement

(Extended Data Figure 1a and b). As the authors discussed themselves, it is possible there may exist disconnected superconducting islands even above 2.7 K, (or even different calibrations of the thermometers in different setups), which may contribute to the spontaneous vortices. The authors need to show superconductivity is indeed totally absent for all the temperatures in the field cooling sequences.
b) The quantum spin liquid phase persists to 200 K in 1T-TaS2 (Ref. 23). And there was no other energy scale in this system except for superconducting pairing that can reconcile with the T* reported here. The use of 'hidden' does not help to clarify the picture of what interaction is driving the chiral order.
c) Since the SQUID probe is still superconducting at T*, it is also conceivable that it may affect the field cooling. Even though the authors argue they have pulled the probe away during cooling, evidence is needed to show it does not affect the cooling for sure. Measuring control superconducting samples with similar Tc (2H-TaS2 will be ideal) is definitely something worth doing.

4. In Figure 1, the authors show spontaneous vortices in the bulk region. If the global TRS is indeed broken, I believe there should also exist spontaneous current near the sample edges. The lack of this evidence leads to other interpretations due to extrinsic effects, such as impurities.

Overall, I think the authors are reporting an interesting observation of an interesting material. However, scientific rigorousness is missing throughout the current manuscript to prevent the authors from drawing a firm conclusion.

Referee #1 (Remarks to the Author):

The authors study a layered structure made of alternating superconducting-Mott insulator layers, known as 4Hb-TaS2. This material has recently risen as a potential candidate for unconventional superconductivity. In the superconducting state below Tc, the system shows a spontaneous vortex phase. The arrangement of those vortexes is shown to be controllable in the normal state, strongly suggesting that the system shows a hidden magnetic ordering above the superconducting Tc. This remarkable behavior is rationalized as stemming from the interplay of a potential chiral spin liquid state of the Mott insulator, coupled to a chiral superconducting state of the superconducting layers.

First of all, I would like to emphasize that I find the results of the authors greatly interesting and that they provide a remarkable finding in the physics of 4Hb-TaS2. Their manuscript is very well written, and their results are compatible with the previous results in 4Hb-TaS2. In particular, the observation of hysteresis in the spontaneous vortex phase of this material represents a remarkable finding for the field of van der Waals materials and unconventional superconductivity. I strongly believe that their results provide a pioneering experiment in the field of van der Waals quantum materials. Therefore, for the reasons stated above, I strongly recommend their manuscript for publication in Nature.

Apart from my positive recommendation for publication in Nature, I would like to mention two points that the authors may optionally consider addressing in their revised manuscript.

We thank the reviewer for the positive view of the manuscript and for the constructive comments.

My first suggestion is related to the interpretation of their results in terms of a coupling between a chiral spin liquid and a chiral superconducting state. First of all, I would like to emphasize that such a picture would be, of course, highly suggestive and consistent with preliminary results in the field. Nevertheless, as the authors well know, assessing the potential spin-liquid nature of some of the Mott layers is a greatly open challenge, and even knowing the potential spin-liquid nature of a Mott monolayer remains an open problem. Assessing also the chiral superconducting nature of the superconducting layers is a nearly remarkable challenge. Therefore, while their picture would be consistent with their observed results, assessing the validity of their assumptions would, of course, require further future work. Given those points, I would suggest to the authors that they reflect those uncertainties in their discussion, for example, by adding after “For both cases, further work is required to provide a complete microscopic description of the effect.” a statement similar to “We finally note that uniquely assessing the potential chiral spin liquid and chiral superconducting nature of the layers remains a remarkably open problem, and demonstrating the picture above will require further future work.”

We completely agree that the proposed scenario is a suggestion, which we hope will inspire further experimental work probing the spin liquid and chiral insulator natures.

We added the following sentence to the discussion:

We finally note that the proposed chiral spin liquid and chiral superconducting natures of the 1T and 1H layers await direct confirmation.

My second suggestion is related to potential alternative mechanisms. I want to note that the authors already nicely explain several mechanisms they discard during their discussion, and thus this suggestion only extends an already satisfactory discussion. In particular, I wanted to ask the authors if potential magnetoelastic effects in the system could be discarded as a mechanism for their observations. While I am fully aware that assessing this is a remarkable challenge, I would suggest that at this stage, it would be enough that the authors add one statement about whether magnetoelasticity can be discarded or if it could be considered as a future candidate for the phenomenology they observe.

We thank the reviewer for this suggestion. Through the magneto-elastic effect, magnetic memory can be stored in an elastic deformation of the crystal, which can in turn induce magnetization along a certain axis. We note, however, that coupling between elastic and magnetic degrees of freedom is only possible when TRS is broken. Thus, even if the magneto-elastic effect is relevant, it does not account for why TRS is broken to begin with. Experimentally testing this is unfortunately beyond the scope of the current work, but we are developing tools to apply local and global strain to the sample and hope to study this aspect in the future.

We added the following text to the manuscript:

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.

To summarize, I believe that the current manuscript puts forward a remarkable finding in van der Waal materials, demonstrating the emergence of hidden order in 4Hb-TaS2. Therefore, given all the points above, I strongly recommend the publication of their manuscript in Nature.

Referee #2 (Remarks to the Author):

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The key finding of the manuscript is that when these stacks are cycled in an applied magnetic field, from below 0.7K to a temperature below a threshold temperature of 3.6K, and subsequently cooled down to below 0.7 in zero applied field, a residual magnetization is observed from a measured amount of Abrikosov vortices penetrating through the superconducting layers. These measurements are performed by Scanning SQUID Microscopy (SSM).

This magnetization is absent when the materials is cooled down in zero field from a temperature above 3.6K. The remnant field observed by Scanning SQUID Microscopy depends on the peak temperature in the above-mentioned temperature cycle and on the magnetic field applied during the field-cooled stage of the measurement procedure.

As no traces of ferromagnetism are discerned by SSM in the heterostructures, the magnetic memory effects displayed by the system are presented as the discovery of a new form of hidden magnetism in the heterostructure in the temperature range below 3.6K.

The manuscript is interesting and the measurements are original and well described. They also nicely demonstrate the power of Scanning SQUID Microscopy for such investigations.

I am left however with a number of questions about the presented work and conclusions, before I can recommend on the publication of the work in Nature.

We thank the reviewer for the positive view of the work and for the constructive suggestions for further experiments. We performed these measurements and expanded the data presented in the manuscript and extended data figures accordingly.

Can the authors completely rule out artifacts arising from magnetized materials near the sample or from flux trapping in the SQUID chip? It would be good if the authors could show that in pure 2H-TaS2 superconductors, or similar representative control samples, the effects do not occur. Then it would be clear that they arise exclusively because of the presence of and/or coupling with 1T-TaS2.

We performed a control experiment that removes this concern. Unfortunately, we cannot study 2H-TaS_2 as its critical temperature is too low (4Hb has T_c of 2.7 K while 2H has T_c of 0.7 K. The latter is lower than the base temperature of our system). We performed a control experiment on a thick flake of $NbSe_2$. Even though $NbSe_2$ has a higher T_c, its Meissner response is a direct probe of magnetic fields near the flake. The Meissner currents at the edges of the flake generate magnetic fields proportional to the external field, which we can directly measure with our SQUID.

A magnetic memory effect would therefore appear as a spontaneous Meissner current following a field training protocol. To test this, we replicated the field-training process of Figure 1 by cooling down the $NbSe_2$ flake from 4.2 K to 1.7 K with an external field of 0.25 Oe. We then imaged the Meissner response at 1.7 K before and after removing the external field. The zero field images at both 1.7 K and 4.2 K reveal no Meissner response, proving that no additional magnetic fields were present in the vicinity of the flake following the "training" process. This excludes effects stemming from the SQUID setup (the sensor itself or any magnetic elements).

Changes made to the manuscript:

- We added a paragraph to the Methods section explaining the control experiment.
- We show the data from the $NbSe_2$ flake in Extended Data Figure 3.

Similarly, it would be good if the authors could show the results of experiments in which first a field is applied in the temperature range 0.7K - 3.5K (thus above the superconducting transition), then the field is taken away and subsequently the sample is cooled down to below the superconducting transition temperature. Does it then also show the remnant magnetization? If yes, it means that the cause of the 'hidden magnetism' is not related to superconductivity. If no, it would show that the superconductivity has a direct role to play in it. In either cases, the generation of the Abrikosov vortices from a hidden magnetism in the 1T layer could still involve a coupling via a chiral superconductor phase.

We performed these experiments and added the results to Figure 2. we cooled down the sample from 4.2 K to 3 K in the presence of a magnetic field, then turned the field off, and then cooled down to 1.7 K. In this procedure, the magnetic field was never turned on when the sample was in the superconducting state. This procedure resulted in a spontaneous vortex phase, which indeed suggests that the hidden magnetism is not directly caused by superconductivity. As the referee suggested, the coupling could still involve chiral superconductivity.

Changes made:

- We included these results in Figure 2c-e.
- We added a description of these experiments to the main text:

To verify that the magnetic memory is generated in the normal state, we cooled down the sample from 4.2 K to 3 K in the presence of a magnetic field, then turned the field off, and then further cooled the sample to 1.7 K (Figure 2c). In this procedure, the magnetic field was never turned on when the sample was in the superconducting state. This procedure generated the same spontaneous vortex density as the FC-ZFC procedures described above (Figure 2d,e), demonstrating that the magnetic memory is generated independently of superconductivity.

Is there a decay over time of the hidden magnetism? This could be probed by varying the time between switching off the magnetic field and starting the cooldown, which was now held at 3 minutes according to the information provided in the Methods section.

We now report on this measurement in Extended Data Figure 2. We generated spontaneous vortices and then warmed up the sample above T_c, to 2.8 K. We kept the sample at this temperature for 3, 6 or 12 minutes and then cooled down and imaged the vortices again. The images showed a spontaneous vortex phase whose vortex density did not depend on the time spent above T_c. The vortices decorated different locations after each cycle, which is expected after cycling the temperature above T_c.

These time scales are far larger than any electronic timescale in this system which further excludes superconducting fluctuations as a possible mechanism. We note that an ordinary spin glass state, which could produce memory lasting for long times, must have a net magnetization which varies slowly compared to the typical scan times (~10 minutes). Thus, such spin glass state would generate DC magnetic signals detectable by both SQUID and muon spin relaxation. The absence of these signals serves as evidence to exclude this possibility. It is still possible to envision a “hidden” glassy state where the magnetic memory decays over long times (>> minutes), but it unfortunately does not shed light on the hidden nature of this state. We hope that the work motivates further research aimed at characterizing the hidden properties of this phase.

Changes made:

- We included these results in extended data figure 2.
- We added the following text to the manuscript:

We have also verified that the magnetic memory does not decay over time in the normal state by training the sample and then keeping it at 3 K without an external field for various amounts of time (Extended Data Figure 2a). The spontaneous vortex density obtained after a subsequent ZFC to 1.7 K did not decay over time, for up to 12 minutes (Extended Data Figure 2b-d). These time scales are much larger than the electronic time scales of the system, further exclude superconducting fluctuations in the normal state as a mechanism to preserve magnetic memory.

Referee #3 (Remarks to the Author):

Persky et al. used the scanning SQUID microscopy to uncover the magnetism hidden in the superconductor 4Hb-TaS2. which hosted superconductivity and chiral spin liquid (CSL) due to the overlaying crystal structures of 2H-TaS2 and 1T-TaS2. They reported the observation of spontaneous vortices in zero field cooling, following some field cooling and heating sequences. They also showed a magnetic-like hysteresis following a specific measurement sequence. Considering there is no long-range magnetic order in the normal state of this system, authors claim that the time reversal symmetry is broken due to the coupling between superconductivity and CSL. The material system and the claimed physics is indeed interesting and the technique is appropriate. However, I do not find the evidence convincing enough for me to recommend publication. The following are my major concerns.

We thank the reviewer for his/her constructive comments. We hope that the revisions and additional experiments will convince the referee that the evidence provided is solid.

1. The zero-field cooling is not strictly zero. We can still see a vortex in Figure 1.e #1, which was obtained after ZFC. This fact greatly weakens the claim of ‘spontaneous vortex’ in the ‘zero-field’.

We used several layers of μ-metal magnetic shields to eliminate earth’s magnetic field (~0.5 G). Naturally, the field cancelation is not perfect and a field of ~5-10 mG remains in the system (a factor of 1000 suppression). We further reduce this by active cancelation using a copper coil, and we consistently achieve fields ~3 mG in our system, as characterized by dozens of superconducting samples that we studied over the years. This field cancelation is comparable to that available in state-of-the-art magnetic sensing facilities and achieving better performance is an extremely challenging task. This 3 mG ambient field is the “zero” of our system, and as accepted in our field, our Zero-field-cooling experiments were performed in its presence.

Scan #1 of Figure 1e indeed shows a single vortex, consistent with the 3 mG field that we are unable to cancel further. The subsequent scans (#3,4,…) of figure 1e were taken with the same 3 mG field but show a vortex density corresponding to fields ~ 100 mG. This cannot be accounted for by the 3 mG field that previously generated only a single vortex and we therefore refer to the excess vortices as spontaneous.

Changes made:

- We added the following text to the Methods section:

Earth's magnetic field was reduced by including two low-temperature μ-metal (A4K, Amuneal) cylindrical shields with diameters of 96 mm and 100 mm around the microscope and coil, achieving fields as small as 5-10 mG near the sample. Further cancelation was achieved by applying a small current through the copper coil, reaching fields ~ 3 mG. "Zero field" measurements were performed under these conditions.

2. The evidence for time-reversal symmetry breaking (TRSB) is not consistent with the interpretation. The authors argue that the superconductivity together with CSL leads to TRSB. If so, the memory of the chirality of TRSB should be erased above the superconducting Tc. However, the hidden magnetism has a higher critical temperature of 3.6 K than the superconducting Tc of 2.7 K.

The reviewer correctly pointed out that if TRSB were related to superconductivity, the magnetic memory would be erased at T_c. The observation that the memory persists above T_c, while superconductivity does not, forces us to consider a different phase that breaks TRS up to 3.6 K, regardless of superconductivity. This is precisely one of the main points of our paper. We propose a CSL as a suitable phase since it breaks TRS, thus storing magnetic memory, without producing detectable magnetic fields. Note that this phase is independent of superconductivity.

To further demonstrate that TRSB is not related to superconductivity (also see response to reviewer #2), we turned the magnetic field off already in the normal state (between 2.7 K and 3.6 K) and then cooled down further below T_c. This procedure still resulted in spontaneous vortices, suggesting that the magnetic state is not related to superconductivity. We further verified that the spontaneous vortex density is insensitive to the amount of time spent in the normal state (with the external field turned off), for times up to 12 minutes.

Changes made:

- We included the results of the two experiments in Figure 2c-e and in Extended Data Figure 2.

3. The relatively close T* and Tc raises some flags.

a) In this work, the superconducting Tc of the sample is determined by bulk transport measurement (Extended Data Figure 1a and b). As the authors discussed themselves, it is possible there may exist disconnected superconducting islands even above 2.7 K, (or even different calibrations of the thermometers in different setups), which may contribute to the spontaneous vortices. The authors need to show superconductivity is indeed totally absent for all the temperatures in the field cooling sequences.

We included additional characterization of the superconducting transition to Extended Data Figure 1. Using the scanning SQUID as a susceptometer, we *locally* measured the diamagnetic response of the sample. This careful mapping showed no traces of superfluid islands above T_c of 2.7K. The 4Hb-TaS_2 samples we studied present a homogenous landscape of the superconductivity. We now include additional data from these measurements in Extended data figure 1.

We note that scanning SQUID susceptometery is extremely sensitive to small traces of superconductivity. We can detect Pearl lengths as large as ~10 mm and we previously used this technique to detect spatial and temporal fluctuations above the transport T_c of 2D superconductors [Nat. Phys. **14**, 1205–1210 (2018)]. Here, local susceptibility maps taken at 2.8 K (above the measured bulk T_c) show no signal within our sensitivity. The absence of signal is homogeneous over large area of the sample (millimeter size). These data clearly demonstrate that superconductivity is completely absent already at 2.8 K. Our thermal cycles to temperatures between 2.8 K and 3.6 K therefore make the sample completely normal and exclude traces of superconductivity as an explanation to our data.

We also note that a recent STM study of 4Hb-TaS_2 (Ref. 34) measured the superconducting gap as a function of temperature, yielding T_c = 2.6 K. The sample they studied originated from the same large crystal. The agreement between Tc measured on a nanometric scale (STM), micron-100's of microns (SQUID) and globally (μSR, global magnetometry, resistance and specific heat) on samples from the same batch further demonstrates that these samples are highly homogeneous.

Changes made:

- We added the following discussion to the main text:

All samples showed a sharp transition around 2.7 K, and at 2.8 K the diamagnetic response disappeared, leaving no traces within our sensitivity. Furthermore, vortices appearing in the DC magnetometry images below Tc disappeared at 2.8 K. Combined, these data show that the entire field of view was normal at 2.8 K (Extended Data Figure 1b). ~~and at 2.8 K the entire scan area was no longer superconducting.~~ Scanning tunneling microscopy measurements[34], Specific heat measurements[6], as well as global magnetometry and resistivity measurements (Extended Data Figure 1d,e) also show a sharp superconducting transition at 2.7 K.

- We added additional data to Extended Data Figure 1, including susceptibility images showing no response at 2.8 K, and the disappearance of vortices at 2.8 K.

b) The quantum spin liquid phase persists to 200 K in 1T-TaS2 (Ref. 23). And there was no other energy scale in this system except for superconducting pairing that can reconcile with the T* reported here.

Thank you for bringing this up. Indeed, the spin liquid state observed in bulk 1T-TaS_2 persists to 200 K. Furthermore, STM and ARPES (Refs. 6, 34) show that the basic CDW motif in the 1T layers in 4Hb is similar to the one found in bulk 1T. However, there are differences between bulk 1T and the 1T layers within 4Hb which provide two mechanisms explaining the emergent temperature scale, T*, we report.

- In 4Hb there is charge transfer between the 1T and 1H layers, due to the work function difference between the layers (Ref. 36). This moves the 1T layers from half filling and is expected to change the Mott physics. A large charge transfer could destroy the spin liquid state altogether and force us to consider a chiral metal state. As we note in the discussion, our experiment does not distinguish between such a chiral metallic state and a chiral spin liquid, and it is a possible phase of the system.
- The spin liquid in the 1T layers may survive if the charge transfer is small. In this case, we note that the experimental observations in 1T-TaS_2 point to a gapless spin liquid with a spinon Fermi surface (Refs. 22-26, 28), which is different than the gapped chiral spin liquid that we proposed in 4Hb-TaS_2. The gapped chiral state can arise as a low temperature instability of the gapless state at some emergent scale (like superconductivity). In this scenario it is natural that the magnetic memory transition is much smaller than J.
- The difference between 2.7 K and 3.6 K is still significant (the latter being larger by 33%) because there are no signatures of superconducting fluctuations above T_c both in local and global probes. Thus, we can safely rule out any superconducting origin of T*.

We believe that these distinctions can reconcile the different temperature scales between the onset of the gapless SL in 1T and the onset of the gapped CSL we propose.

Changes made:

- We added the following discussions:

We note that the spin liquid phase proposed in bulk 1T-TaS_2 is different from the spin liquid proposed here. Namely, the bulk 1T spin liquid persists up to 200 K [23] and is likely gapless[22–24,26,28]. The CSL we propose is gapped and the chirality onsets at ~3.6 K. These differences could be driven by the interactions between the 1T and 1H layers, present in the 4Hb structure[27,28]. For example, it is possible that 4Hb hosts a gapless spin liquid at high temperatures, but it undergoes a gapping transition at a lower temperature. The resulting gapped CSL~~ Such CSL state~~ accounts for our observations in the normal state.

Other chiral states residing in the 1T layer could be consistent with the data if they generate a very weak magnetization. For example, charge transfer between the 1T and 1H layers[34] could result in ~~the 1T layers may also be in~~ a chiral metallic state[33] residing on the 1T layers.

The use of 'hidden' does not help to clarify the picture of what interaction is driving the chiral order.

The referee is completely right; we do not know what drives the chiral order. Our results provide evidence that a chiral order exists. We believe this evidence is sufficiently interesting, unambiguous, and important to report, even if their microscopic origin is still unclear. Such microscopic understanding requires further theoretical and experimental studies, sensitive to the microscopic structure. This is beyond the scope of the current work, and we hope that our results will inspire follow-up experimental and theory work that would resolve this question.

c) Since the SQUID probe is still superconducting at T*, it is also conceivable that it may affect the field cooling. Even though the authors argue they have pulled the probe away during cooling, evidence is needed to show it does not affect the cooling for sure. Measuring control superconducting samples with similar Tc (2H-TaS2 will be ideal) is definitely something worth doing.

First, we note that the SQUID is thermally isolated from the sample, and its temperature does not change when the sample temperature is varied between 1.7 K to 4.2 K. Therefore, it cannot account for the observed effect which onsets when the sample temperature is 3.6 K.

To further remove this concern (also in response to Reviewer #2), we performed a control experiment. Unfortunately, we cannot use 2H-TaS_2 since its critical temperature (0.7 K) is lower than the base temperature of our system. Instead, we used a flake of $NbSe_2$, utilizing its Meissner response as a probe for magnetic fields in the vicinity of the sample. Had the observed effects been caused by an extrinsic element (such as the SQUID or magnetic parts in the setup), a "spontaneous" Meissner current would have appeared in the flake, to screen the fields generated by the extrinsic element.

We tested this by replicating the field-training protocol of Figure 1 on the $NbSe_2$ flake. We then imaged the Meissner response at 1.7 K before and after removing the external training field. The zero field images at both 1.7 K and 4.2 K reveal no Meissner response, suggesting that no additional magnetic fields were created in the vicinity of the flake following the "training" process. This excludes effects stemming from the SQUID setup (the sensor itself or any magnetic elements).

Changes made:

- We added a paragraph to the Methods section, describing the control experiment.
- We added the null results from this experiment as Extended Data Figure 3.

4. In Figure 1, the authors show spontaneous vortices in the bulk region. If the global TRS is indeed broken, I believe there should also exist spontaneous current near the sample edges.

We have not looked for spontaneous edge currents in 4Hb-TaS_2. In the superconducting state, there are always Meissner currents at the edge of the sample, and for a large (~1-2 mm) single crystal with an undefined geometry, these currents may be large, even for minute external fields. Therefore, quantitatively analyzing these signals and separating the spontaneous response due to global TRSB from the Meissner currents is challenging, and we examined the spontaneous vortices instead. We further note that edge currents are not a universal feature of chiral superconductors. Several theoretical works [Phys. Rev. B **90**, 224519 (2014); Phys. Rev. B **97**, 064510 (2018)] suggest that the response depends on the exact chiral order parameter and boundary conditions. Previous experimental works on candidate chiral superconductors such as Sr_2RuO_4 failed to find such edge currents [Phys. Rev. B **81**, 214501 (2010)], and it is not obvious that the situation here is different. Not finding such spontaneous edge currents therefore does not exclude chiral superconductivity or global TRS breaking.

Changes made:

- We added the following sentence to the discussion:

We finally note that the proposed chiral spin liquid and chiral superconducting natures of the 1T and 1H layers await direct confirmation.

The lack of this evidence leads to other interpretations due to extrinsic effects, such as impurities.

Isolated magnetic impurities typically suppress superconductivity through pair-breaking and cause the superconducting state to be inhomogeneous [Sov. Phys. JETP **12**, 1243–1253 (1961)]. In our case, there are no indications of dirty superconductivity in global (heat-capacity or resistivity) and local (the magnetic susceptibility maps) measurements.

Moreover, for magnetic impurities to generate magnetic memory, they must order ferromagnetically. The normal state data impose a strict limit on the moment density (distance between moments > 40 nm), which makes ferromagnetic ordering highly unlikely, for two reasons:

1. The moments are dilute compared to the electron gas and their RKKY interactions are therefore random in sign, leading to glass-like behavior rather than ferromagnetism.
2. In the dilute regime, Kondo screening, which is memory free, is typically dominant over magnetic ordering.

Changes made:

- We added the following text:

The large inter-moment distance suggests that the interactions are dominated by Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions. However, the moments are dilute compared to the electron gas, suggesting that the RKKY interactions are random in sign. This type of interactions leads to glass-like behavior, rather than ferromagnetism. In addition, for such a dilute moment concentration, Kondo screening, which is memory-free, is expected to be dominant.

Overall, I think the authors are reporting an interesting observation of an interesting material. However, scientific rigorousness is missing throughout the current manuscript to prevent the authors from drawing a firm conclusion.

We are happy that the referee finds the results interesting. We hope that, with the additional experiments and explanations, the paper is now sufficiently convincing and can be published in Nature.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

I have read in detail the response of the authors to my comments. The authors included changes in their manuscript following those points, fully addressing my suggestions. I believe that their experiment provides a highly inspiring and remarkable demonstration of the interplay between superconducting vortices and potential quantum spin liquid behavior, a topic of outstanding interest for the community of quantum materials. Given all the points above, I strongly recommend the manuscript for publication in Nature.

Furthermore, I have read in detail the comments of Referees #2 and #3, together with the responses of the authors and the revised version of their manuscript. I believe that the authors have successfully addressed all the criticisms of the Referees and included positive modifications in their manuscript following their suggestions. In particular, I would like to note that, while several of the suggestions of Referee #3 would be interesting to consider in-depth, addressing those points in detail would clearly go beyond the scope of the present manuscript. I believe that the authors have presented a very solid picture of their experiment, and therefore I strongly recommend publication of their manuscript in Nature in its current form.

Referee #2 (Remarks to the Author):

With the additional experiments and textual amendments to the manuscript the authors have made it more convincing that the effects measured are intrinsic to the 4Hb-TaS2 system, rather than arising from artefacts. The presentation of the study is also more balanced, as the authors better indicate now that various aspects need further study.

To my opinion, the study is of sufficent interest for publication in Nature and the manuscript provides a clear and well written description. I have a few remarks left that I would like the authors to address before definitely recommending publication.

1) I have some difficulties with the expression 'hidden magnetism'. The authors use this term primarily as no (ferro-)magnetism is detectable with their Scanning SQUID Microscope. This technique however has its limitations, for example in spatial resolution and with regard to field orientation. In plane magnetic textures are for example difficult to pick up with an SSM. Perhaps with other techniques, the 'hidden magnetism' could be well observable.

To put 'Hidden magnetism' so prominently, even as the starting words of the title, to my opinion suggests more than what the paper can claim. This impression is even amplified by the statement 'This discovery of a new form of hidden magnetism ..', suggesting that the 4Hb-TaS2 exhibits a form of magnetism not known until now.

I deem it appropriate to replace 'Hidden magnetism' in title and throughout the manuscript by 'Magnetic memory' or 'Magnetic memory effect', as this is ultimately what the paper is about.

2) While the authors state, likely for simplicity, that the material under study is 4Hb-TaS2, it is clear from the Methods section that it is actually TaS_1.99Se_0.01. While this may seem unimportant at this moment in time, it may perhaps turn to be relevant when the understanding of the magnetic memory effect develops further. I suggest that the authors make a clearer statement on the sample composition in the main text.

Referee #3 (Remarks to the Author):

The revised manuscript by Persky et al. provided some additional data to support the authors' claim. In particular, Extended Data Figure 1 is helpful to rule out superconductivity above 2.7 K. However, other revisions do not qualitatively change the lack of scientific rigor of the previous manuscript. Since the claimed chiral spin liquid and chiral superconductivity are largely speculative and unexpected, the experiments have to leave no room for doubt. I find the revised version still short of this standard and therefore I do not recommend its publication.

The control experiment on NbSe2 in Extended Data Figure 3 is a good example of the problem with the manuscript. NbSe2 is a poor choice as the control sample here because its Tc is above T*. It is still in the superconducting state at 4.2 K and any remnant field at this temperature will be screened out. Repeating the same temperature cycle as on 4Hb-TaS2 therefore did not mean the equivalent field history for the control sample. Control sample with lower Tc is needed as I have suggested in the first report.

I think the authors are responsible for removing any doubt for a big claim (or any scientific claim) to publish in Nature (or in any journal). Finding ways to measure 2H-TaS2 and show data on statistically significant number of 4Hb samples will go a long way.

Some further comments regarding the authors' responses:
1. How was the remnant field determined? Was there more data on other samples than the one in Fig. 1e? Would the claimed chiral superconductivity affect the perceived 'zero field'?
2. The argument about the thermal isolation between the SQUID and the sample does not answer my question about the SQUID. The flux repulsion from the SQUID in its superconducting state may affect the zero field and field coolings.
3. The response about the absence of spontaneous edge current is unsatisfactory. First, the authors did not modify the manuscript to discuss the edge at all. Second, using Sr2RuO4 as an example is misleading. The absence of edge current in Sr2RuO4 is actually against its candidacy for chiral superconductivity, rather than supporting a chiral superconductor without edge current. Recent findings of experimental artifacts in the previous NMR data (e.g., Ishida et al., J. Phys. Soc. Jpn. 89,034712 (2020)), which had been widely regarded as evidence for its chiral superconductivity,

further showed the importance of seeing spontaneous edge current. Third, the authors claim their ‘zero field’ is pretty small even by SQUID standards. The images at zero field on NbSe2 did not show Meissner current. Why would the Meissner current on 4Hb more significant under such ‘zero field’ than the spontaneous edge current, which should be pretty big according to Ref.8 in the supplementary information?

Referee #1 (Remarks to the Author):

I have read in detail the response of the authors to my comments. The authors included changes in their manuscript following those points, fully addressing my suggestions. I believe that their experiment provides a highly inspiring and remarkable demonstration of the interplay between superconducting vortices and potential quantum spin liquid behavior, a topic of outstanding interest for the community of quantum materials. Given all the points above, I strongly recommend the manuscript for publication in Nature.

Furthermore, I have read in detail the comments of Referees #2 and #3, together with the responses of the authors and the revised version of their manuscript. I believe that the authors have successfully addressed all the criticisms of the Referees and included positive modifications in their manuscript following their suggestions. In particular, I would like to note that, while several of the suggestions of Referee #3 would be interesting to consider in-depth, addressing those points in detail would clearly go beyond the scope of the present manuscript. I believe that the authors have presented a very solid picture of their experiment, and therefore I strongly recommend publication of their manuscript in Nature in its current form.

We thank the reviewer for his/her positive view of the work.

Referee #2 (Remarks to the Author):

With the additional experiments and textual amendments to the manuscript the authors have made it more convincing that the effects measured are intrinsic to the 4Hb-TaS2 system, rather than arising from artefacts. The presentation of the study is also more balanced, as the authors better indicate now that various aspects need further study.

To my opinion, the study is of sufficent interest for publication in Nature and the manuscript provides a clear and well written description. I have a few remarks left that I would like the authors to address before definitely recommending publication.

We thank the reviewer for the positive comments and for the constructive comments. We have revised the manuscript accordingly:

1) I have some difficulties with the expression 'hidden magnetism'. The authors use this term primarily as no (ferro-)magnetism is detectable with their Scanning SQUID Microscope. This technique however has its limitations, for example in spatial resolution and with regard to field orientation. In plane magnetic textures are for example difficult to pick up with an SSM. Perhaps with other techniques, the 'hidden magnetism' could be well observable.

To put 'Hidden magnetism' so prominently, even as the starting words of the title, to my opinion suggests more than what the paper can claim. This impression is even amplified by the statement 'This discovery of a new form of hidden magnetism ..', suggesting that the 4Hb-TaS2 exhibits a form of magnetism not known until now.

I deem it appropriate to replace 'Hidden magnetism' in title and throughout the manuscript by 'Magnetic memory' or 'Magnetic memory effect', as this is ultimately what the paper is about.

We removed the term "hidden magnetism" from various places throughout the manuscript, including the title, abstract, "Here, we investigate…" paragraph, and the discussion.

2) While the authors state, likely for simplicity, that the material under study is 4Hb-TaS2, it is clear from the Methods section that it is actually TaS_1.99Se_0.01. While this may seem unimportant at this moment in time, it may perhaps turn to be relevant when the understanding of the magnetic memory effect develops further. I suggest that the authors make a clearer statement on the sample composition in the main text.

We added a clear statement about this in the main text:

We studied two ~~4Hb-TaS_2~~ 4Hb-$TaS_{1.99}Se_{0.01}$ single crystals. The small Se concentration significantly improves the quality of single crystals (Methods).

Referee #3 (Remarks to the Author):

The revised manuscript by Persky et al. provided some additional data to support the authors' claim. In particular, Extended Data Figure 1 is helpful to rule out superconductivity above 2.7 K. However, other revisions do not qualitatively change the lack of scientific rigor of the previous manuscript. Since the claimed chiral spin liquid and chiral superconductivity are largely speculative and unexpected, the experiments have to leave no room for doubt. I find the revised version still short of this standard and therefore I do not recommend its publication.

We thank the reviewer for his/her comments. We hope that the revisions clarify our statements convince that they are indeed strongly supported by the data.

We emphasize that the main claim is the observation of the unusual magnetic phase, not the interpretation in terms of a chiral spin liquid or chiral superconductivity.

This claim is a direct result of the large discrepancy between the observed spontaneous vortex density and normal state magnetization. The experimental evidence leaves no room for doubt that there is unconventional magnetism in the system, and that it couples to superconductivity in an unusual way. Furthermore, as explained in detail below, the control experiment proves that the effect is intrinsic to the material and does not stem from the measurement system.

We completely agree that the proposed chiral spin liquid and chiral superconductor phases are largely speculative. We make no attempt to prove these statements since conclusive proof would require multiple experimental probes combined with a complete microscopic theory. Both efforts are beyond the scope of the current work. We only bring up the CSL and CSC as possibilities, to be explored by future work. We have revised the manuscript to further emphasize the speculative nature of the interpretation and to suggest specific future experiments that could (dis)prove these ideas.

Changes made:

1) In the fourth paragraph (starting with "Here, we investigate…"), we explicitly stated that we cannot resolve the order parameters of the normal and superconducting states:

Although we cannot resolve the order parameter of the superconducting and the TRSB phases, ~~We~~ We propose a CSL in the 1T layers as a candidate TRSB phase.

2) We revised the discussion paragraph describing the possible CSC to better reflect the uncertainty regarding the theoretical interpretation. The new paragraph reads:

While a CSL state is consistent with our results, there is no direct evidence for this scenario in our data. Furthermore, the mechanism through which a CSL induces spontaneous vorticity is unclear. In Supplementary Note 1 we discuss a possible scenario, where the superconducting phase is itself chiral. A chiral superconductor (CSC) generates an internal magnetization which may generate spontaneous vortices[46]. In this scenario, the chirality of the CSL acts as an ordering field which aligns the domains of the CSC. Supplementary Notes 2 and 3 explain why the data provided here cannot decisively prove whether the superconducting state is chiral and suggest alternative coupling mechanisms that do not require a CSC. Resolving the order parameters of both the normal and superconducting states requires additional experimental work. For example, Sagnac interferometry could provide additional information on the TRSB[60] in both the normal and superconducting states. The superconducting order parameter could be probed by Josephson interferometry.

3) We added Supplementary Notes 2 and 3 which (also in response to the following comments) explain why we cannot resolve the superconducting order parameter from our measurements, and discuss briefly other possible mechanisms.

The control experiment on NbSe2 in Extended Data Figure 3 is a good example of the problem with the manuscript. NbSe2 is a poor choice as the control sample here because its Tc is above T*. It is still in the superconducting state at 4.2 K and any remnant field at this temperature will be screened out. Repeating the same temperature cycle as on 4Hb-TaS2 therefore did not mean the equivalent field history for the control sample. Control sample with lower Tc is needed as I have suggested in the first report.

We used $NbSe_2$ as a probe for external magnetic sources in the system, which might generate magnetic fields below 3.6 K. There are two ways to use a superconductor as a probe for such field. One is to cool it down through Tc in the presence of the field and measure the vortex density. The other way is to apply the field already in the superconducting state and measure the Meissner currents generated to screen the field. In this case we do not need vortices and the critical temperature of the superconductor is immaterial.

To test whether a part of the system can be field trained below 4 K, it is essential to repeat the training protocol used on the TaS_2 sample. The absence of "spontaneous" Meissner currents in NbSe2 following this process proves that there was no external field to be screened. Thus, the spontaneous vortices observed in 4Hb-TaS2 must come from an intrinsic effect.

<u>Changes made:</u>

1. We added the following text to the Methods section describing the control experiment:

If a something in the system can be trained by field cooling through 3.6 K, the flake would generate Meissner currents to screen the fields generated by the trained field source. Thus, measuring the Meissner response from the flake after a training protocol probes field sources in our setup.

I think the authors are responsible for removing any doubt for a big claim (or any scientific claim) to publish in Nature (or in any journal).

We believe that the control experiment removes the concern regarding extrinsic field sources in our setup. It therefore shows that the 4Hb-TaS_2 data is robust and supports the conclusion regarding the presence of an unusual magnetism.

Finding ways to measure 2H-TaS2 and show data on statistically significant number of 4Hb samples will go a long way.

The T_c of 2H-TaS_2 is unfortunately lower than the base temperature of our setup, so we cannot access its superconducting state with scanning SQUID. Using a different setup to study 2H-TaS2 could be beneficial, but it would not rule out the artefacts that the referee had proposed.

The results presented in the manuscript are from two single crystals of 4Hb-TaS_2 grown in two batches almost a year apart. Both samples displayed the magnetic memory effect and did not display magnetization above T_c.

Pieces of one of these two samples were also characterized by ARPES, specific heat, resistivity, μSR and thermal conductivity (Ref. 6), and by scanning tunneling microscopy (Ref. 34).

Some further comments regarding the authors' responses:

1. How was the remnant field determined?

The remanent field in our system was determined by counting vortices and dividing by the area after zero field cooling in dozens of superconducting materials.

Was there more data on other samples than the one in Fig. 1e?

Yes. Figure 2c-e and Extended Data Figure 2 are from a second 4Hb single crystal that was grown ~ 1 year after the original sample. Both samples displayed spontaneous vortices with magnetic temperatures 3.6 K and 3.8 K.

<u>Changes made:</u>

In the Methods section, we added details about which data were taken from which sample:

Data shown in Figures 1, 2b, 3 and Extended Data Figures 1 and 4 were taken from sample 1, and data shown in Figure 2d and Extended Data Figure 2 were taken from sample 2.

Would the claimed chiral superconductivity affect the perceived 'zero field'?

The "zero field" condition in our system is a reproducible condition that does not depend on the sample under study. This point was confirmed by the $NbSe_2$ control experiment.

A chiral superconductor can generate spontaneous magnetization, which might change the perceived external field in the system. In such a scenario, an untrained sample would generate a net magnetization and therefore spontaneous vortices. Since zero-field cooled samples did not produce spontaneous vortices, we conclude that for the proposed chiral superconductor scenario, the most likely case is that untrained samples are broken into small domains of opposite chirality, such that the net magnetization is zero. This is analogous to many ferromagnets, where observing net magnetization requires training. In this case the proposed chiral superconductivity does not affect the perceived zero field.

2. The argument about the thermal isolation between the SQUID and the sample does not answer my question about the SQUID. The flux repulsion from the SQUID in its superconducting state may affect the zero field and field coolings.

We are confident that this suggested artifact cannot explain our observations, for several reasons:

1. It is true that flux repulsion from the SQUID may affect the field at the sample. However, it cannot lead to the observed temperature dependence where cycling the sample temperature to 3.6 K (keeping the SQUID at a fixed temperature) changes the ZFC.
2. If the SQUID affected the local fields at the sample, the vortex distribution would have a distinct spatial texture determined by the sensor's geometry [Physica C **533**, 114-117 (2017)]. As seen in Extended Data Figure 1b, such textures are not present in our data, because the SQUID was retracted from the sample before cooling.
3. Such distortions would have generated Meissner currents in the $NbSe_2$ flake, which were not observed.

3. The response about the absence of spontaneous edge current is unsatisfactory. First, the authors did not modify the manuscript to discuss the edge at all. Second, using Sr2RuO4 as an example is misleading. The absence of edge current in Sr2RuO4 is actually against its candidacy for chiral superconductivity, rather than supporting a chiral superconductor without edge current. Recent findings of experimental artifacts in the previous NMR data (e.g., Ishida et al., J. Phys. Soc. Jpn. 89,034712 (2020)), which had been widely regarded as evidence for its chiral superconductivity, further showed the importance of seeing spontaneous edge current.

We brought up Sr_2RuO_4 to demonstrate how tricky edge currents in chiral superconductors can be. We agree that absence of edge currents does not support chiral superconductivity. However, although edge currents were not detected, there is a large volume of other experiments that do support chiral superconductivity in Sr_2RuO_4. Recent examples include ultrasound experiments which show that SC is two-component [Nat. Phys. **17**, 194–198 (2021); Nat. Phys. **17**, 199–204 (2021)], and μSR showing a split between TRSB temperature and SC temperature under strain [Nat. Phys. **17**, 748–754 (2021)], a strong indication of chiral superconductivity. Whether Sr_2RuO_4 is a chiral superconductor is still under debate. This demonstrates that absence of edge currents cannot be viewed as definitive evidence against chiral superconductivity.

Changes made:

1. We added Supplementary Note 2 which discusses the problems arising with interpreting edge signals from candidate chiral superconductors. This note includes Sr_2RuO_4 as an example.

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.

Third, the authors claim their ‘zero field’ is pretty small even by SQUID standards. The images at zero field on NbSe2 did not show Meissner current. Why would the Meissner current on 4Hb more significant under such ‘zero field’ than the spontaneous edge current, which should be pretty big according to Ref.8 in the supplementary information?

The difference between $NbSe_2$ and 4Hb-TaS_2 stems from a geometrical effect. A circular loop of radius R with current I generates a magnetic field

$$B = \frac{\mu_0 I}{2R}$$

at the center of the loop. The 4Hb sample was a large single crystal (R ~ 1 mm) and therefore generated large Meissner currents to screen the "zero" field. The NbSe2 flake was smaller (R ~ 30 μm) and therefore required a smaller current to screen the same "zero" field.

For the spontaneous edge currents, we refer to Supplementary Ref. 9 (Ref. 7 in the previous version) and note that it is not at all clear that the spontaneous edge currents should be large, and that in any case they are expected to be canceled by counter-propagating Meissner currents. There shouldn’t be any signal from the spontaneous currents on length scales relevant to the SQUID. The presence of vortices further complicates this interplay. For this reason, we did not investigate the edge currents in depth, and we would not have been able to draw robust conclusions from such data.

Changes made:

1. We added the following text to the Methods section about the control experiment:

Note that the zero-field Meissner response of the flake generates a smaller magnetic signal than the Meissner currents observed after zero field cooling an untrained 4Hb-TaS_2 sample. The difference in signals can be accounted for by the different geometries of the two samples. The Meissner current generated by a superconducting sample is such that it screens the applied external field. For a circular sample of radius R, the Meissner current I_M required to completely screen an external field B_{ext} satisfies

$$B_{ext} = -\frac{\mu_0 I_M}{2R}.$$

Therefore, a large single crystal with R~1 mm generates a larger Meissner response than a small (R ~ 30 µm) flake.

Reviewer Reports on the Second Revision:

Referees' comments:

Referee #2 (Remarks to the Author):

With their new manuscript, the authors have complied largely with my remarks from the previous round of reviewing. I am pleased to see that the term 'hidden magnetism' has been removed, although in the abstract and the main text there are still a few sentences left, that suggest the discovery of a form of magnetism that is not detectable.

I am especially refering to the sentences:

'Strikingly, this phase does not generate detectable magnetic signals.'
and
'Surprisingly, we show that this TRSB order does not produce detectable magnetic signatures in the normal state. '

Please rephrase these sentences to something like:
'Strikingly, this phase does not generate ferromagnetic signals detectable using common techniques.'
and
'Surprisingly, we show that this TRSB order does not produce ferromagnetic signatures in the normal state detectable using common techniques.'

I'd further like to note that Referee 3 has a valid point on the superconducting Tc of NbSe2. Because of it, the test experiment has turned into a quite indirect analysis based on the Meissner response. It would be much more convincing if the text experiment would be more similar to the experiment conducted for the 4B-TaS2 system, thus with a similar superconducting Tc <3.6 K but with a different composition.

From my side I will not uphold the publication of this manuscript based on this aspect alone, in line with my statements in the previous round of reviewing, but I want to express my understanding if referee 3 considers it a critical point for accepting the manuscript for publication.

Finally, I noted a few typographical errors in the manuscript. I take it that these will be resolved in the final version and the editorial process.

Referee #3 (Remarks to the Author):

In this revision and the rebuttal letter, the authors did not provide any additional data. They provided an argument for my main criticism about the control experiment on NbSe2. The argument is not convincing to me for the following reasons.

1. A control experiment, almost by definition, should be carried out in similar manner under similar conditions.
2. The $NbSe_2$ sample is so much smaller than the 4Hb-TaS_2 sample. This makes it much less sensitive to any field sources in the setup. Their argument about the absence of Meissner current in $NbSe_2$ under the perceived 'zero field' showed this point.
3. Cooling through T_c, which they did for 4Hb-TaS_2, requires much less field to generate vortices than applying field in the superconducting state, which they did for the control sample.

Because no additional evidence is provided, my recommendation remains the same.

Referee #2 (Remarks to the Author):

With their new manuscript, the authors have complied largely with my remarks from the previous round of reviewing. I am pleased to see that the term 'hidden magnetism' has been removed, although in the abstract and the main text there are still a few sentences left, that suggest the discovery of a form of magnetism that is not detectable.

I am especially referring to the sentences:

'Strikingly, this phase does not generate detectable magnetic signals.'

And

'Surprisingly, we show that this TRSB order does not produce detectable magnetic signatures in the normal state. '

Please rephrase these sentences to something like:

'Strikingly, this phase does not generate ferromagnetic signals detectable using common techniques.'

And

'Surprisingly, we show that this TRSB order does not produce ferromagnetic signatures in the normal state detectable using common techniques.'

We revised the sentences accordingly.

I'd further like to note that Referee 3 has a valid point on the superconducting Tc of NbSe2. Because of it, the test experiment has turned into a quite indirect analysis based on the Meissner response. It would be much more convincing if the text experiment would be more similar to the experiment conducted for the 4B-TaS2 system, thus with a similar superconducting Tc <3.6 K but with a different composition.

From my side I will not uphold the publication of this manuscript based on this aspect alone, in line with my statements in the previous round of reviewing, but I want to express my understanding if referee 3 considers it a critical point for accepting the manuscript for publication.

We address this point in detail in response to referee #3 below and in the revised Methods section. Briefly, although the $NbSe_2$ experiment does not exactly replicate the conditions of the 4Hb-TaS_2 experiments, it provides a more decisive way to rule out external magnetic sources.

Finally, I noted a few typographical errors in the manuscript. I take it that these will be resolved in the final version and the editorial process.

We corrected several grammatical errors and typos in the text.

Referee #3 (Remarks to the Author):

In this revision and the rebuttal letter, the authors did not provide any additional data. They provided an argument for my main criticism about the control experiment on NbSe2. The argument is not convincing to me for the following reasons.

We thank the reviewer for the additional comments on the $NbSe_2$ experiment. We addressed these comments below by revising the Methods section so that it provides a more complete and critical discussion of this experiment. The key point is that although the $NbSe_2$ experiment does not replicate the conditions of the 4Hb-TaS_2 experiments, it is a suitable probe for ruling out external magnetic field sources as a cause for spontaneous vortices. Thus, it effectively eliminates the main concern in the previous reports, as we explain in detail below.

1. A control experiment, almost by definition, should be carried out in similar manner under similar conditions.

We agree that in this respect, the terminology “control experiment” was wrong. In an ideal control experiment, we would indeed replace 4Hb-TaS_2 with another superconductor with similar T_c and similar dimensions and repeat the vortex counting experiment. However, an adequate control sample must have a critical temperature in the sharp range of 2 K to 3 K (higher than our base temperature but low enough to allow for cycles between T_c to 3.6 K). The superfluid density must be homogeneous, and the superconducting transition must be extremely sharp. Inhomogeneities adversely affect the vortex density and prevent us from drawing strong conclusions. This was the case with several materials that we tried to use as controls, including **[redacted]**. We also tried using **[redacted]** which are more homogeneous, but films with T_c in the appropriate range have extremely large penetration depths, which prevented us from counting vortices. Figure R1 demonstrates these issues in **[redacted]**. The nominal T_c is **[redacted]**. This means that the sample generates or distorts magnetic fields in its "normal" state, and that the results of a control experiment would not be reliable. The inhomogeneity is in sharp contrast to 4Hb-TaS_2 which was highly homogeneous below Tc and did not show any traces of superconductivity above 2.7 K. Finding superconductors with $2 \text{ K} < T_c < 3.2 \text{ K}$ that are sufficiently homogeneous turned out to be surprisingly difficult.

Image and Text Readacted

The $NbSe_2$ experiment is a much better alternative for excluding an extrinsic source for a spontaneous field below T = 3.5 K. The Meissner current generated by a superconductor is directly proportional to the external field and to the size of the superconductor. By using a small flake with a size that fits into our field of view we can quantitatively determine, a-priori, the expected signal due to an extrinsic magnetic memory. Because we use flakes with T_c > 3.5 K, we avoid the risk of having normal regions in the control sample (due to inhomogeneity) that distort field lines. In that sense, this experiment is much cleaner than using a bulk superconductor with unknown geometry or even counting vortices in a field-cooled sample.

Indeed, the lack of spontaneous Meissner currents following the training protocol means there are no trainable field sources in the system. This was the main concern, and the goal of any control experiment. The $NbSe_2$ experiment conclusively proves that the effect in 4Hb-TaS_2 is intrinsic, and it does so better than any control experiment on a large sample.

Changes made:

1) In the Methods section, we included a complete discussion of the possible artifacts and how we excluded them. This discussion also includes artifacts discussed in the previous rounds of review (previously in the main text). We stated that we do not conduct a control experiment that resolves all issues at once but take the approach of resolving one issue at a time.
2) We included an analysis of possible magnetic sources in our system, that could generate a magnetic field of 100 mG near the sample, with gradients smaller than (0.3 mG)/(100 μm), as is required to explain the 4Hb-TaS_2 data. This analysis makes it clear that such extrinsic sources are

highly unlikely, and the $NbSe_2$ complements this argument by directly showing that indeed there are no stray fields near the sample, following a training sequence.

3) We expanded the discussion of the $NbSe_2$ experiment so that it explains clearly the choice of material and size of the flake. This discussion also states the expected trained signal and resulting signal to noise ratios in this measurement.

2. The NbSe2 sample is so much smaller than the 4Hb-TaS2 sample. This makes it much less sensitive to any field sources in the setup. Their argument about the absence of Meissner current in NbSe2 under the perceived ‘zero field’ showed this point.

The signal is indeed smaller. We accounted for this by carefully choosing both the flake size and the applied training fields so that the expected signal would be well above our noise floor, within our scan range, and below the saturation of our electronics.

3. Cooling through Tc, which they did for 4Hb-TaS2, requires much less field to generate vortices than applying field in the superconducting state, which they did for the control sample.

This is true, but we did not intend to induce vortices in the sample. On the contrary, Meissner currents provide a much cleaner proof.