nature research

Peer Review File

Manuscript Title: Topological superconductivity in a van der Waals heterostructure

Editorial Notes:

Redactions – Mention of other journals

This document only contains reviewer comments, rebuttal and decision letters for versions considered at *Nature*. Mentions of the other journal have been redacted.

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Ref #1

In this paper the authors grow islands of ferromagnet (CrBr3) on bulk superconducting NbSe2. They observe a peak centered near the Fermi energy at the edges of the island and attribute this peak to the existence of Majorana edge modes.

B. Similar work was published on the Fe/Re(0001) system (Palacio-Morales, A. et al. Atomic-scale interface engineering of Majorana edge modes in a 2D magnet-superconductor hybrid system. Sci. Adv. 5, eaav6600 (2019)). However, there since the Fe islands are metallic, proximity effect induces unconventional superconductivity in the Fe islands which is a sensible model.

C and E. The paper has many issues both in terms of physics and interpretation as well as in terms of transparency. My comments are below.

1) Fig. 1 b and c are extremely misleading. The authors are using bulk NbSe2 on which CrBr3 is grown. NbSe2 is a bulk superconductor. So the picture shown on Fig 1 does not apply at all to this system. If they were to apply, the bands shown would be bulk bands. The surface FM island cannot affect the bulk NbSe2 bands and open up a gap as shown in the figure.

2)Later in the paper the authors propose a different mechanism (not the one shown in Fig1 a and c). They propose that the FM island generates shiba states in NbSe2 which then form a band. The band then supposedly gaps out (the reason for this is not at all clear) and the edge states live inside this Shiba state gap. There are multiple problems with this scenario. There is no mechanism to gap out the Shiba states. And Shiba state bands would not in general behave like regular bands. Moreover, what role does spin-orbit play in all this?
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3) The calculations are poorly done to say the least. There is no clear understanding or explanation of how one induces non-trivial topology in an existing BULK superconductor simply by putting a FM on it. The normal scenario would be that you induce SC correlations in a ferromagnet which might then effectively behave like a p+ip superconductor. This reverse scenario makes no sense.

4) Some of the authors have already written theoretical paper on this same topic (arXiv:1905.01063v2, published in the New Journal of Physics, Volume 22, January 2020). I find it very surprising that there is no mention of this paper in the current paper.

5) To make their calculation work, the authors make many assumptions which are not justified in the

paper. For example, they require a renormalization of the chemical potential resulting from the contact with the magnetic material. However, there is no proof that this is indeed happening. The authors should have clearly been to see this in their spectroscopy. 100meV is a huge shift of the local chemical potential.

Second, most of their calculations are done on monolayer NbSe2 which is a completely different system from bulk NbSe2 in terms of band structure. There is no possible justification for treating a bulk material like a monolayer.

Third, the authors completely throw out the Se-derived bands near the Gamma point! One cannot selectively ignore bands in a calculation!

6) The experimental data is also not convincing. There are many reasons to see a peak in the density of states near a magnetic island. These could simply be Shiba states for example.

7) There are other huge problems with the authors interpretation. If these are indeed chiral edge modes, they MUST be continuous along the boundary. Instead the edge clearly show breaks i.e., areas where they disappear (behaving like localized shiba states in fact). In fact, one of the biggest reasons for the interest in Majorana states is that they are robust and should simple go around impurities This does not happen in the data. The authors try to explain these gaps in the text, but their explanation is not convincing. There is no reason for these states to disappear due to some kind of interference.

8) 1D Majorana states should show a flat density of states not a peak. Why do the authors expect to see a peak?

9) The paper lacks clarity both in the presentation as well as in the connection between theory and experiment.

Overall, there are numerous issues with this manuscript both on the experimental and on the theoretical front. I do not recommend publication.

I would also urge the authors to substantially revise the paper as well as their interpretation before submitting to another journal.

Ref # 2

The authors reported the growth of CrBr3 islands on superconducting NbSe2 and the observation of the one-dimensional Majorana edge modes using low-temperature STM and STS. Since both the fabrication of topological superconductors by hetero-structures, and the observation Majorana mode by STM/STS have been reported in previous papers, I can't see any significance of this work to gurantee the manuscript publish in Nature.

Below are my detial comments:
1. What's the use of the reported 1D Majorana edge modes?
2. In Fig. 3a), I can't see edge mode is becomes more delocalized at energies closer to the topological gap edge.
3. Is there the minimum size of the island to see the Majaorana edge mode?
4. Also in Fig. 3a), I see the zero bias peaks shift a little bit at some places, what's the reason?
5. In Fig. 3c), the mapping of the edge mode is not continuous, in contradiction to the one in Fig.3j).
6. Reference#25 is not on a proximitized topological insulator.
7. FeTe0.55Se0.45 has been proven to be a topological superconductor, so the sentence "it is presently unclear if topological superconductivity, which has been suggested as a key ingredient for topological quantum computing, exists at all in any naturally occurring material1" is not correct.

Ref # 3

The paper presents very interesting experimental evidence of Majorana edge states at the perimeter of ferromagnetic CrBr3 islands on superconducting NbSe2 substrates. Both the phenomenon and the system itself are remarkable, and I feel the work is worth publishing in Nature or another high-profile journal. However, I do think there are a few critical points that were not carefully accounted for in the paper, which are detailed below:

1. The authors have made a decent effort in developing a quantitative model to describe the supposedly topological superconductivity of the heterostructure. However, in my opinion the model is still oversimplified and should not be directly used to explain the experimental findings. The model is based on DFT calculations of a monolayer CrBr3 on a monolayer NbSe2, and the authors assumed that the only band that is relevant to the topological superconductivity comes from the isolated band crossing the Fermi energy in monolayer NbSe2. The finite thickness of the NbSe2 substrate is completely ignored in the model. In the other systems that host Majorana fermions, their low-energy space has well-defined low dimensionality: In semiconductor nanowires it is the single 1D quantum well state that hosts the topological superconductivity; in Fe atomic chains on Pb substrates it is the 1D Shiba band that is relevant. In contrast, the 2D d-band in a monolayer NbSe2 is not isolated from the other layers in a thick NbSe2 substrate. Although the authors made some arguments on the 2D nature of bulk NbSe2 bands, they are not sufficient to justify such a radical simplification (not to mention that they also argued that the Ising spin-orbit coupling harmful to the topological superconductivity is vanishing because of the finite thickness of NbSe2). To give one example, if they did a model for bilayer or trilayer NbSe2 using the same approach, I would expect the topological phase diagram to be completely different.

2. The oversimplified model has another troubling consequence which is worth mentioning separately--the decay length of the Majorana edge states. In an ideal 2D p-wave superconductor model the decay length of the edge states is the ratio between Fermi velocity and the p-wave gap. If the authors set the parameters of their model using the experimental values, the decay length would be at least 10^2 of the lattice constant, contradicting with the narrow width (~2nm) of the edge modes observed by STM. Their simulation in Fig. 3 was done with a much exaggerated superconducting gap (20 times larger), which is only mentioned in the supplementary information. Such a disagreement may also be addressed by abandoning the monolayer NbSe2 assumption and focusing on a different low-energy space. A similar situation in the case of Fe chain on Pb was discussed in e.g. PRL 114, 106801 (2015) and PRB 90, 235433 (2014). It is also worth noting that in order to get topological superconductivity from the monolayer NbSe2 model a certain degree of fine tuning is needed, while experimentally the edge states are surprisingly ubiquitous as mentioned in the main text. It is possible that the thick substrate relaxes the condition for the edge modes to appear.

3. The size of the Rashba spin-orbit coupling was assumed to be very large (60 meV) in the model, but I did not find how the authors justify this value. They may at least estimate the Rashba SOC from their DFT calculations.

4. The subgap peaks at 0.41 meV are not explained. These should be due to weakly dispersive Shiba bands. Understanding their origin in a more quantitative way should help to build a better model of the superconductivity at the interface.

5. A few typos: (1) H_{SOC} in Eq. (1) in the SI misses a spin Pauli matrix vector in the first term. (2) V_S exists in Eq. (1) of SI but does not appear in Eqs. (2-5)

Author Rebuttals to Initial Comments:

REFEREE #1:

Overall referee comments:

A. In this paper the authors grow islands of ferromagnet (CrBr3) on bulk superconducting NbSe2. They observe a peak centered near the Fermi energy at the edges of the island and attribute this peak to the existence of Majorana edge modes.

B. Similar work was published on the Fe/Re(0001) system (Palacio-Morales, A. et al. Atomic-scale interface engineering of Majorana edge modes in a 2D magnet-superconductor hybrid system. Sci. Adv. 5, eaav6600 (2019)). However, there since the Fe islands are metallic, proximity effect induces unconventional superconductivity in the Fe islands which is a sensible model.

C and E. The paper has many issues both in terms of physics and interpretation as well as in terms of transparency. My comments are below.

Author reply: As shown in detail below, the criticism above is either misplaced or can be refuted as a whole. In fact, the statement B above is clearly a result of a confusion regarding the well-established requirements of topological superconductivity in magnetic-superconducting heterostructures that have been discussed extensively in the literature.

Referee comment 1. Fig. 1 b and c are extremely misleading. The authors are using bulk NbSe2 on which CrBr3 is grown. NbSe2 is a bulk superconductor. So the picture shown on Fig 1 does not apply at all to this system. If they were to apply, the bands shown would be bulk bands. The surface FM island cannot affect the bulk NbSe2 bands and open up a gap as shown in the figure.

Author reply: Unfortunately, there seems to be a misconception that topological su-perconductivity could not be induced on a bulk superconductor. On the contrary, this is a universally accepted mechanism in designer structures combining superconductors with magnets. The band schematics in Figs. 1b and c are, contrary to what the referee maintains above, a faithful schematic description of topological superconductivity induced by a lattice of magnetic moments on top of a *bulk* superconductor. The particle-hole symmetric bands schematically illustrated in the figure are the subgap bands localized in the vicinity of the contact with the magnetic moments. In particular, there is absolutely no controversy that an array of magnetic impurities can (and will) induce a topological superconducting region on a surface of a bulk superconductor.

In relation to this comment and comment B above, the metallic nature of the magnetic ma-terial or array is *completely irrelevant* for achieving the topological phase. The fact that bulk superconductor can be made topological by patterning of magnetic impurities without any contributing electronic states has been discussed in a large body of literature starting from the pioneering works in 1D magnetic impurity chains on a superconductor (see for example

Pientka et al. PRB 88, 155420 (2013), Nadj-Perge et al. PRB 88, 020407(R) (2013)), and in 2D by the pioneering works by one of us (Ref. [28], Röntynen and Ojanen PRL 114, 236803 (2015)) and by the Princeton group (Ref. [29], Li et al. Nat. Comm. 7, 12297 (2016)). These works (and the large body of literature citing them) establish conclusively that even magnetic moments that behave effectively classically (without contributing electronic orbitals) give rise to local in-gap Shiba states and will modify the surface region of a bulk superconductor to become topological. The key point is that a magnetic impurity will always lead to an in-gap Shiba state (irrespective of the existence of additional electronic orbitals) localized around the impurity. An array of impurities will lead to subgap electronic bands localized on the surface. These surface bands are the entities that will become topological. Thus, a magnetic insulator will give rise to a well-defined topological surface band. This is not in any way controversial in the community.

If the magnet is metallic, it will lead to an additional modification of the electronic properties of the structure as analyzed in detail in 1D by Peng et al. (PRL 114, 106801 (2015)). However, even in that case the spectral weight of the topological state mostly comes from the substrate Shiba states (otherwise the localization length would be orders of magnitude larger which is contradicted by experiments). In summary, there is absolutely no need for the magnet to be metallic to achieve the topological phase and the contrary claims of the referee are simply incorrect.

Referee comment 2. Later in the paper the authors propose a different mechanism (not the one shown in Fig1 a and c). They propose that the FM island generates shiba states in NbSe2 which then form a band. The band then supposedly gaps out (the reason for this is not at all clear) and the edge states live inside this Shiba state gap. There are multiple problems with this scenario. There is no mechanism to gap out the Shiba states. And Shiba state bands would not in general behave like regular bands. Moreover, what role does spin-orbit play in all this?

Author reply: Contrary to what the referee claims, we are *not proposing a different mechanism* than illustrated in Fig. 1 and there is no discrepancy in our description of the topological phase and Figure 1. It seems that the main confusion of the referee can be traced back to the misunderstanding expressed in the comment 1 above. The physical picture we use is simple and fully in-line with the standard picture of topological superconductivity in magnetic-superconducting heterostructures. In particular, the big picture relies only on well-established results that are universally accepted by the experts in the field.

As explained in the response to Referee comment 1, magnetic impurities/lattices on an s-wave superconductor will always give rise to subgap states/bands in the vicinity of the impurities (irrespective of any extra electronic states in the impurities). This is fundamental and universally accepted fact and the basic principle of topological state engineering in superconductors, a field in which one us (T.O.) is a well-known pioneer. The magnetization-induced subgap states/bands, often called Shiba states/bands, are formed in particle-hole

space and have topological or trivial character in exact analogy to electronic bands in an insulator. In the studied system, the subgap (Shiba) band (illustrated in Fig. 1c) results from the electronic bands in $NbSe_2$ in the presence of Rashba spin-orbit coupling, proximity-induced magnetism (Fig. 1b) and superconductivity. Since the pioneering papers cited above (Refs. [28,29], PRL 114, 236803 (2015) and Nat. Comm. 7, 12297 (2016)), it has been generally known that ferromagnetic order with Rashba coupling will generally result in a gapped Shiba band with topological character. There are no "multiple problems of this scenario" and this is backed up by a considerable body of literature.

In the so-called dense impurity limit where magnetic moments cover densely the superconducting surface (for example, see the analysis in page 3 in Nat. Comm. 7, 12297 (2016) by the groups of Bernevig and Yazdani), which is relevant for our experimental system, the system can be modelled effectively by adding a local Zeeman field to every lattice site. In this case, the subgap band is simply understood in terms of the normal state dispersion with a Zeeman splitting and effective p-wave pairing between the particle-hole bands. Mathematically the model of magnetic moments on a superconductor with Rashba spin-orbit coupling is analogous to a lattice version of the famous (but yet unrealized) magnet-2DEG-superconductor sandwich structure proposed in Ref. [36] (Phys. Rev. Lett. 104, 040502 (2010)) although the physical origin is quite different. The Bogoliubov-de Gennes equation describing both these systems has analogous structure even though in the sandwich model the source of superconductivity and Rashba-coupled electron gas come from physically separate entities. However, a single superconducting element with Rashba coupling would suffice.

In summary, contrary to what the referee maintains, we use the "standard model" of topological superconductivity in magnetic-SC heterostructures and it is based on universally accepted general principles. The real case-specific novelty is that instead of treating a toy square lattice model (as in Ref. [29], Nat. Comm. 7, 12297 (2016)) and several papers afterwards), we have a more complex lattice symmetry and employ a band structure which has been obtained from the DFT spectrum after a tight-binding regularization. This gives new quantitative details (e.g. the topological phase can arise from states around different high-symmetry points within the Brillouin zone), but does not change the physics qualitatively.

In contrast to what the referee implies above, the role of the Rashba coupling (and other parameters) as well as the appearance and the magnitudes of the spectral gaps in the subgap (Shiba) bands are extensively discussed and analytically treated in Sec. 2 of the Supplementary Information. While the full details can be found there, as a brief summary of the detailed analysis, *without the Rashba SOC the system becomes topologically trivial or gapless*.

Referee comment 3. The calculations are poorly done to say the least. There is no clear understanding or explanation of how one induces non-trivial topology in an existing BULK superconductor simply by putting a FM on it. The normal scenario would be that you induce SC correlations in a ferromagnet which might then effectively behave like a p+ip superconductor. This reverse scenario makes no sense.

Author reply: Unfortunately, this comment again reiterates the referee's demonstrably wrong impression regarding the feasibility of obtaining topological superconductivity by a ferromagnet in contact with a bulk superconductor. As pointed out above, the pioneering papers (and the hundreds of works citing them) make it evident that the referee's opinion is in direct conflict with the universally accepted state of affairs in the field. The magnet induces subgap states in the superconductor that form the topological bands in the presence of Rashba spin-orbit coupling.

The appearance of the topological superconductivity is shown in detail in Sec. 2 of the Supplementary Information. In a nutshell, the subgap bands are always gapped in the presence of ferromagnetic order and Rashba SOC. This follows from the generic symmetries of the system and does not even require the knowledge of the details of the band structure. The details of the topological phase diagram, characterized by Chern numbers, then follow from more specific band-structure features, although the generic features are sufficient in identifying the Chern numbers of the nontrivial states.

Referee comment 4. Some of the authors have already written theoretical paper on this same topic (arXiv:1905.01063v2, published in the New Journal of Physics, Volume 22, January 2020). I find it very surprising that there is no mention of this paper in the current paper.

Author reply: The above paper studies the possibility of realizing a *gapless* (or nodal) topological state which does not have chiral edge modes, spectral gap and is not classified by Chern numbers. Also, the nodal state would require an in-plane magnetization in contrast to the experimental realization ($CrBr_3$ has out-of-plane magnetization). Thus, the completely different nature of the nodal state and the fundamentally different physical requirements for its appearance makes the paper only tangentially relevant (at best) for the present work. In general, we try to avoid self-citation unless our earlier work is directly relevant for the present work.

Referee comment 5. To make their calculation work, the authors make many assumptions which are not justified in the paper. For example, they require a renormalization of the chemical potential resulting from the contact with the magnetic material. However, there is no proof that this is indeed happening. The authors should have clearly been to see this in their spectroscopy. 100meV is a huge shift of the local chemical potential.

Second, most of their calculations are done on monolayer NbSe2 which is a completely different system from bulk NbSe2 in terms of band structure. There is no possible justification for treating a bulk material like a monolayer.

Third, the authors completely throw out the Se-derived bands near the Gamma point! One cannot selectively ignore bands in a calculation!

Author reply: Regarding the renormalization of the chemical potential, this effect will definitely take place when the surface of the $NbSe_2$ is brought to a contact with the 2D magnet (it would be unreasonable to expect that the effective contact potential would vanish). While the shift is substantial, it is difficult to probe experimentally. Tunneling probability to the $NbSe_2$ bulk bands is substantially reduced on the $CrBr_3$ islands and it is difficult to reliably identify how much they shift. To the best of our knowledge, this shift also cannot be reliably estimated from *ab initio* methods and thus serves as a free parameter. The overall magnitude of 100 meV is reasonable and, while the appearance of the topological phase requires a degree of the shift, it does not need to be fine tuned.

Regarding the effective 2D modelling, it is not correct to state that "There is no possible justification for treating a bulk material like a monolayer". In fact, a convincing justification for 2D modelling already existed before our work: Ménard et al. (Nat. Phys. 11, 1013 (2015)) have studied the effect of single magnetic impurities in $NbSe_2$. In this work, the experimental results show unambiguously that the resulting Shiba states have 2D nature and do not exhibit any characteristics of 3D substrate. The fact that the experimentally measured spatial profile of subgap Shiba states shown in the Fig. 7 in the Supplementary of the above mentioned paper matches essentially perfectly the prediction of theoretical modelling based on 2D approximation proves conclusively that the effective planar treatment is justified when studying the magnetization-induced subgap physics. So while it is, of course, an approximation to treat the system as two-dimensional, there is already an experimental proof that it is an extremely good starting point. Also, in that work, the Shiba states are modelled on a 2D lattice by using an effective tight-binding model extracted from a 3D DFT calculation, which is similar approach that we have taken. So contrary to what the referee claims, there is excellent motivation to treat the system as we have done.

Regarding the third point, the Se orbitals are "not thrown away", the DFT band structure calculations include all the orbitals. The calculated band structure is then used to obtain an effective tight-binding model which reproduces the most relevant features of the quasi-2D spectrum near the Fermi energy. It is true that the TB model is 2D in nature and thus does not contain all the information of the bulk bands. However, as found out in the previous and present work, the experimental observations of the magnetic subgap physics in the layered system are excellently in line with the effective 2D modelling. As always, the ultimate justification of the theoretical models come from comparison to experiments. As explained above, the previous work of magnetic impurities on $NbSe_2$ has already proven that the effective 2D TB model after fitting the full electronic structure leads to a quantitative match of experimental results and theoretical predictions. Therefore it is the natural starting point of our work, which recovers excellent qualitative match between theory and experiments. In fact, the excellent agreement between our theory and experiments is remarkable considering that the full microscopic treatment of the structure (2D magnet+superconductivity+interface physics+full 3D bulk) is well beyond of all presently known computational and theoretical methods.

Action: We have included a reference to the work of Ménard et al. (Nat. Phys. 11, 1013 (2015)) in the revised manuscript to motivate our treatment of the $NbSe_2$ as two-dimensional.

Referee comment 6. The experimental data is also not convincing. There are many reasons to see a peak in the density of states near a magnetic island. These could simply be Shiba states for example.

Author reply: Stating "The experimental data is also not convincing." is not very helpful without detailed criticism and we strongly refute any claim that our experimental data is not state-of-the-art: we demonstrate an extremely high-quality experimental realization of a $CrBr_3$–$NbSe_2$ vdW heterostructure and characterize it extensively using low temperature STM at $T = 350$ mK.

While it is possible to have Shiba states at the Fermi energy, it would require a "magic" coincidence of having suitable free spins with the correct exchange coupling to the $NbSe_2$ substrate to have them at the Fermi energy on all the island edges. As the exchange coupling between isolated impurities and the substrate depends heavily on e.g. their adsorption site, this is extremely unlikely. In addition $CrBr_3$ is expected to be a simple ferromagnet and the well-defined edges are not expected to have additional magnetic moments or weird spin-textures. Taken together, this renders the suggestion of the referee very unlikely.

Moreover, our experimental results show a minigap between the Shiba bands on the $CrBr_3$ islands, the states we observe at the edges exist inside this minigap. This is an important point and further indication that the edge states are indeed topological edge modes. This observation also contradicts the explanation in terms of isolated Shiba states proposed by the referee.

Action: We explicitly point out in the revised manuscript that the states we observe at the edges of the $CrBr_3$ islands exist inside the minigap between the Shiba bands and that this serves as a further indication that the edge states are indeed topological edge modes.

Referee comment 7. There are other huge problems with the authors interpretation. If these are indeed chiral edge modes, they MUST be continuous along the boundary. Instead the edge clearly show breaks i.e., areas where they disappear (behaving like localized Shiba states in fact). In fact, one of the biggest reasons for the interest in Majorana states is that they are robust and should simple go around impurities This does not happen in the data. The authors try to explain these gaps in the text, but their explanation is not convincing. There is no reason for these states to disappear due to some kind of interference.

Author reply: There are no alleged "huge problems of interpretation". The chiral edge states are continuous along the edge. As explained in the manuscript, the reason why they appear cutoff is that there are modulations of the intensity of the edge modes due to the irregular edge geometry around the corners of the $CrBr_3$ islands. We have in fact shown

in our theoretical calculations how the edge irregularities lead to this effect: near sharp edge features, the edge mode can have strong enhancement or suppression of intensity. While the wavefunction is not exactly zero in the suppressed areas, with finite experimental resolution, the signal is below visibility and it creates an effect of "cutoff" modes while the edge mode is perfectly continuous. The fact that the periodic edge mode modulation on smooth edges matches the moire´ periodicity of the LDOS everywhere below the magnetic islands is concrete evidence for the explained scenario. Together with the fact the edge modes exist for all subgap energies, this constitutes a compelling argument for the topological origin of the edge modes.

As already state above, our experimental results show a minigap between the Shiba bands on the $CrBr_3$ islands, and the states we observe at the edges exist inside this minigap. This is an important point and further indication that the edge states are indeed topological edge modes. The modulation of the chiral edge mode does not affect its topological nature or quantized transport, as is evident from the studies of transport in highly irregular and even amorphous topological systems (see for example Pöyhönen et al. Nat. Comm. 9, 2103 (2018)).

Referee comment 8. 1D Majorana states should show a flat density of states not a peak. Why do the authors expect to see a peak?

Author reply: The claim of the referee that the 1D Marojana edge modes should show a flat density of states is incorrect. This scenario is correct only when when the edge modes have exactly constant slope for all subgap energies. However, this happens very rarely (if ever) in more realistic models. The existence of subgap edge modes is guaranteed by topology, but the dispersion of the edge modes, which determines the specific LDOS features, are completely non-universal and model specific. Also, the peaked LDOS we see and reproduce in theoretical calculations has weight in all subgap energies, but more weight near midgap than towards gap edge. So the LDOS pattern can have arbitrary shape as long as it is non-zero for all subgap energies. In fact, it is very strong evidence supporting our interpretation that our theoretical model reproduces quite precisely the shape of the subgap LDOS.

Referee comment 9. The paper lacks clarity both in the presentation as well as in the connection between theory and experiment.

Author reply: We have completely refuted all the specific remarks of the referee above. In the absence of specific criticism on "clarity both in the presentation as well as in the connection between theory and experiment" it is difficult to offer counter arguments. As demonstrated in multiple cases above, the referee perceives a disagreement with the univer-sally accepted physical picture (established in extensive literature as discussed above) and our approach. We can only once more reiterate that is does not exist and point the referee to the literature cited above.

REFEREE #2**:**

Overall referee comments:

The authors reported the growth of CrBr3 islands on superconducting NbSe2 and the observation of the one-dimensional Majorana edge modes using low-temperature STM and STS. Since both the fabrication of topological superconductors by hetero-structures, and the observation Majorana mode by STM/STS have been reported in previous papers, I can't see any significance of this work to gurantee the manuscript publish in Nature.

Author reply: Importantly, there is no comprehensive demonstration of 2D topological superconductivity to date and we demonstrate significant advances past the state-of-the-art in our manuscript. In particular, we go beyond the previous reports:

- Ménard et al. Two-dimensional topological superconductivity in Pb/Co/Si(111). Nat. Commun. 8, 2040 (2017): The sample geometry (buried cobalt islands under lead monolayer on Si(111) is poorly defined and allows only crude modelling without any material-specific inputs or predictions. No possible (even conceivable) extensions to constructing real devices.

- Palacio-Morales et al. Atomic-scale interface engineering of Majorana edge modes in a 2D magnet-superconductor hybrid system. Sci. Adv. 5, eaav6600 (2019): iron mono-layer decoupled by an oxygen interlayer from a superconducting Re substrate. While there is clear disorder in the system (on the atomic scale), the calculated phase diagram indicates that very small changes of e.g. the chemical potential should alter the topological phase (with strongly different Chern numbers). It is not at all clear how the enhanced LDOS at the island edges should be understood considering this. Similar problems with extending this work to devices as above.

- Wang et al. Evidence for dispersing 1D Majorana channels in an iron-based superconductor. Science 367, 104 (2020): Observed bound states at (half-unit cell shift) domain walls are proposed to be a realization of the so-called "π-phase shift domain wall" in the theoretical proposal of Fu and Kane (Phys. Rev. Lett. 100, 096407 (2008)). This is possible, but certainly not proved beyond reasonable doubt. Again, the system lacks tuneability and device applications required very significant further work.

Our work presents two major breakthroughs: fabrication of epitaxially coupled 2D magnet-superconductor structure and demonstration of 2D chiral topological superconductivity through direct observation of the chiral Majorana edge modes. Together, these achievements clearly justify our work to be considered in Nature. Furthermore, the near-ideal artificial magnetic superconductor demonstrated in our work provides a great versatility for future applications.

Referee comment 1. What's the use of the reported 1D Majorana edge modes?

Author reply: The chiral Majorana edge modes represent highly-sought after and fundamentally new type of state of matter with a number of exotic properties. They have been proposed as a platform to probe non-abelian statistics of Majorana zero modes and they act as ideal 1D thermal guides. Moreover, the chiral Majorana states indicate that the system supporting them are topological superconductors, which can also support Majorana bound states on dislocations and vortices. Majorana bound states are the extremely heavily researched building blocks for topological qubits and quantum computing.

Referee comment 2. In Fig. 3a), I can't see edge mode is becomes more delocalized at energies closer to the topological gap edge.

Author reply: The referee is correct and we have revised our manuscript here. As discussed in more detail in the response to comment 2 by Referee #3, this somewhat counter-intuitive observation reproduced accurately by our theoretical model. The peculiar feature of the edge modes include their unusually strong spatial localization and their coexistence with the bulk states in a substantial energy window as seen in the strip spectra in Fig. 2d. This is also clearly seen in the measured and calculated maps in Figs. 3e,f,l,m. The spectrum in Fig. 2d shows that the edge modes survive above the bulk gap at and above the topological gap at 0.3Δ. Thus, no substantial delocalization take place at energies comparable to the topological gap. The edge modes broaden and merge to bulk bands only at relatively high energies where the bulk contribution is already strong.

Action: The discussion around this point has been modified in the revised manuscript (lower half of p. 8 in the revised manuscript).

Referee comment 3. Is there the minimum size of the island to see the Majorana edge mode?

Author reply: Based on theoretical considerations, to observe the modes for all subgap energies, the linear dimension of the islands should be much larger than the edge mode penetration depth. As is evident from the experimental data and calculations, this condition is clearly fulfilled in all our work.

Action: We have added a comment on this point in the revised manuscript.

Referee comment 4. Also in Fig. 3a), I see the zero bias peaks shift a little bit at some places, what's the reason?

Author reply: The sugbap LDOS for chiral Majorana modes is non-universal in the sense that the exact LDOS as a function of energy does not follow from topology but is model-specific feature. The topology only dictates that the LDOS must be non-vanishing for all

subgap energies but the exact edge mode dispersion is a model-specific property. Thus, there is no universal explanation for the general shape of the LDOS and the detailed features such as the slight shift of the LDOS maximum when crossing the edge. However, remarkably, our theoretical model reproduces this behaviour as can be seen from the simulated LDOS curves. We speculate that the origin of the shift is due to the drop in the local chemical potential (from the magnetic island) which would give rise to relative shift of the particle and hole like spectral density in a single Shiba state. While the effect is small, this robust feature lends additional support to our theoretical modelling.

Referee comment 5. In Fig. 3c), the mapping of the edge mode is not continuous, in contradiction to the one in Fig.3j).

Author reply: Looking at the STM topography image shown in Fig. 3b, the right edge of the $CrBr_3$ island has atomic scale step at distances roughly matching the moire´ period. These steps cause suppression of the edge mode intensity, similarly to the sharp island corners. The theoretical calculation also shows how these modulations happen at sharp edge features: near the tip of the island the calculated edge mode intensity is strongly suppressed (while the edge mode is fully continuous).

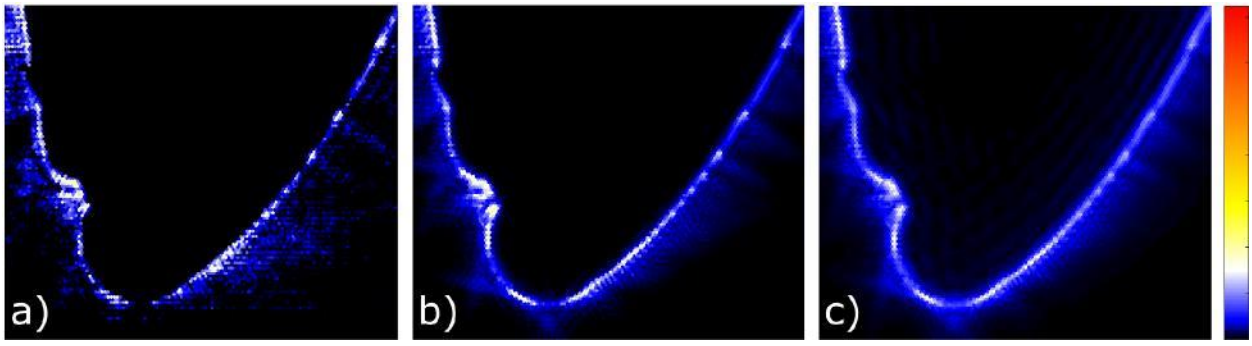

FIG. R1. The midgap LDOS maps ($E = 0$) for different values Δ: a) corresponds to physical value $\Delta = 1.25$ meV while b) and c) correspond to exaggerated values $\Delta = 10$ meV and $\Delta = 20$ meV respectively. The parameters are $t_1 = 40$ meV, $t_2 = 132$ meV, $t_3 = 12$ meV, $\alpha_R = 60$ meV and chemical potential in the bulk was $\mu_B = -111.2$ meV. Chemical potential and the Zeeman splitting on the island was $\mu_I = -260$ meV and the $V_Z = 85$ meV. The calculation has been carried out on a lattice with 10^5 sites.

We have carried out new calculations with smaller Δ in response to the comment 2 from Referee #3 (details are given there). Figure R1 shows calculated LDOS maps at $E = 0$ for three different values of Δ and it can be seen that the edge mode develops intensity variations at the sharp corners, but also at the single-site sized steps on the right edge of the magnetic island. These intensity variations are stronger the smaller the superconducting gap is and nicely reproduce intensity variations seen in the experiment for a realistic value of Δ.

Action: Fig. R1 is included in the revised version of the Supplementary Information.

Referee comment 6. Reference #25 is not on a proximitized topological insulator.

Author reply / action: We thank the referee for pointing out this typographical error which has been corrected in the revised manuscript.

Referee comment 7. FeTe0.55Se0.45 has been proven to be a topological superconductor, so the sentence "it is presently unclear if topological superconductivity, which has been sug-gested as a key ingredient for topological quantum computing, exists at all in any naturally occurring material1" is not correct.

Author reply: The $FeTe_{1-x}Se_x$ family are candidates for 3D topological superconductivity where helical (as opposed to chiral) Majorana modes could appear in domain walls or hinge states (signalling higher-order topology). While the results of the $FeTe_{1-x}Se_x$ family are certainly encouraging, there is a number of unresolved issues (as the recent commentaries https://www.condmatjclub.org/uploads/2019/06/JCCM_June_2019_02.pdf and https://www.condmatjclub.org/uploads/2018/12/JCCM_December_2018_03.pdf explain). Furthermore, the first alleged signatures of dispersive Majorana modes on these materials was published as recently as Jan 2020 (in Science, Ref. [15]). As pointed out in the commentary above, there is still room for reasonable doubt regarding the topological state. As a final remark, irrespective of the final conclusion on $FeTe_{1-x}Se_x$ family, the topological state (if present) is topologically distinct from the studied 2D chiral superconductors characterized by Chern numbers. The designer platform we have demonstrated represents a breakthrough since it offers direct route towards electronic devices and demonstrates how long-sought after exotic states of matter can be designed and fabricated in the laboratory.

REFEREE #3:

Overall referee comments:

The paper presents very interesting experimental evidence of Majorana edge states at the perimeter of ferromagnetic CrBr3 islands on superconducting NbSe2 substrates. Both the phenomenon and the system itself are remarkable, and I feel the work is worth publishing in Nature or another high-profile journal. However, I do think there are a few critical points that were not carefully accounted for in the paper, which are detailed below:

Author reply: We thank the referee for his/her positive assessment of our manuscript and the recommendation to publish it in Nature. We reply to the specific points in detail in the following.

Referee comment 1. The authors have made a decent effort in developing a quantitative model to describe the supposedly topological superconductivity of the heterostructure. However, in my opinion the model is still oversimplified and should not be directly used to explain the experimental findings. The model is based on DFT calculations of a monolayer CrBr3 on a monolayer NbSe2, and the authors assumed that the only band that is relevant to the topological superconductivity comes from the isolated band crossing the Fermi energy in monolayer NbSe2. The finite thickness of the NbSe2 substrate is completely ignored in the model. In the other systems that host Majorana fermions, their low-energy space has well-defined low dimensionality: In semiconductor nanowires it is the single 1D quantum well state that hosts the topological superconductivity; in Fe atomic chains on Pb substrates it is the 1D Shiba band that is relevant. In contrast, the 2D d-band in a monolayer NbSe2 is not isolated from the other layers in a thick NbSe2 substrate. Although the authors made some arguments on the 2D nature of bulk NbSe2 bands, they are not sufficient to justify such a radical simplification (not to mention that they also argued that the Ising spin-orbit coupling harmful to the topological superconductivity is vanishing because of the finite thickness of NbSe2). To give one example, if they did a model for bilayer or trilayer NbSe2 using the same approach, I would expect the topological phase diagram to be completely different.

Author reply: To summarize our theoretical scheme: we perform DFT calculations to obtain the band structure of monolayer $NbSe_2$ – $CrBr_3$ heterostructure and then project the obtained electronic structure to an effective 2D spin-split tight-binding model. Our theoretical scheme is essentially similar to what was used in a paper by Ménard et al. (Nat. Phys. 11, 1013 (2015)) to model the subgap states of magnetic impurities in $NbSe_2$. That work demonstrated beyond doubt that the experimentally measured Shiba states have effectively 2D nature even though the layered structure is three-dimensional. As demon-strated in Fig. 7 in the Supplementary Information of the above work, the layers in the bulk system are so decoupled that the subgap Shiba states are definitely inconsistent with 3D substrate but excellently reproduced by the effective 2D modelling. This agreement is so striking that the authors conclude that "This behavior justifies a posteriori our 2D ap-

proximation of the $NbSe_2$ band structure" (Supplement, page 7). This provides concrete evidence that the layers in bulk $NbSe_2$ are sufficiently decoupled that effectively 2D approach is justified in studying subgap structure of localized magnetic structures.

In our work, we discover similar striking agreement with the effective 2D modelling. In addition to the universal signatures of topological superconductivity, our theoretical model reproduces a number of experimentally observed system-specific characteristics including 1) the correct edge mode penetration depth which is orders of magnitude smaller than naive estimates (discussed below), 2) the specific form of the subgap local density of states, which depends on system-specific dispersion of the topological edge modes, and 3) a non-generic coexistence of the topological edge modes and bulk states in a substantial energy window (clearly seen in the predicted edge dispersion in Fig. 2d and seen in the measured and simulated LDOS maps in Fig. 3). This provides extraordinary evidence for our model and conclusions. In the light of the remarkable agreement with our theory and experimental results, characterizing our model as "oversimplified" is not appropriate. In the light of previous work and our present work, we conclude that the effective 2D modelling with realistic band parameters is the most natural starting point for the theoretical analysis.

Of course the true material is indeed 3D and this is reflected, for example, in the higher superconducting T_c than in mono- and few-layer systems. However, a full microscopic (or ab initio) treatment the epitaxial interface of the 2D magnet and the bulk $NbSe_2$ in the presence of superconductivity, spin-orbit interactions and 3D effects is well beyond all present theoretical and computational techniques. Thus, the models for these complex systems are ultimately assessed by comparison to experiments. Our 2D model is in-line with previous work and already reproduces the experimental observations down to system-specific details. It would be counter-productive to add further complexity into the model.

Action: We have added a reference to the work by Ménard et al. and explained the motivation of using a 2D model for the $NbSe_2$ in the revised manuscript and highlighted the comparison between theory and experiment (starting at the bottom of p. 8 in the revised manuscript.

Referee comment 2. The oversimplified model has another troubling consequence which is worth mentioning separately–the decay length of the Majorana edge states. In an ideal 2D p-wave superconductor model the decay length of the edge states is the ratio between Fermi velocity and the p-wave gap. If the authors set the parameters of their model using the experimental values, the decay length would be at least 10^2 of the lattice constant, contradicting with the narrow width ($\sim$ 2 nm) of the edge modes observed by STM. Their simulation in Fig. 3 was done with a much exaggerated superconducting gap (20 times larger), which is only mentioned in the supplementary information. Such a disagreement may also be addressed by abandoning the monolayer NbSe2 assumption and focusing on a different low-energy space. A similar situation in the case of Fe chain on Pb was discussed in e.g. PRL 114, 106801 (2015) and PRB 90, 235433 (2014). It is also worth noting that in

order to get topological superconductivity from the monolayer NbSe2 model a certain degree of fine tuning is needed, while experimentally the edge states are surprisingly ubiquitous as mentioned in the main text. It is possible that the thick substrate relaxes the condition for the edge modes to appear.

Author reply: We thank the referee for raising this important point. The referee is correct in noting that the numerical real-space simulations were carried out with an exaggerated superconducting gap (as previously explained in the Supplementary Information). This was done to restrict the computation to smaller lattices because accurate calculation of the subgap spectral features require lattices with linear dimension $L >> t/\mathrm{Li}$, where t is the relevant band width $t \sim 100$ meV and $\mathrm{Li} \sim 1$ meV. Therefore it is common to employ artificial parameters (e.g. in Ref. [14] the bandwidth was reduced by two order of magnitude!). With realistic separation of the energy scales, the exact diagonalization becomes computationally very expensive even with optimized sparse matrix algorithms. We did already check the Li-scaling of different observables to confirm that our exaggerated Li calculations were qualitatively justified.

However, to clear any doubts and address the concerns of the referee, in the revised version of our manuscript include large scale numerical calculations to show that the realistic value of Li (while in the limits of exact diagonalization) does not modify our essential conclusions (results shown in Fig. R2). This explicitly shows that our model predicts the very narrow edge mode signal in agreement with the experiment. We stress that, contrary to the speculations of the referee, the narrow edge mode width does not have anything to do with the alleged shortcomings of the 2D modelling. On the contrary, it provides additional compelling evidence that our model is faithful description of the experimental situation.

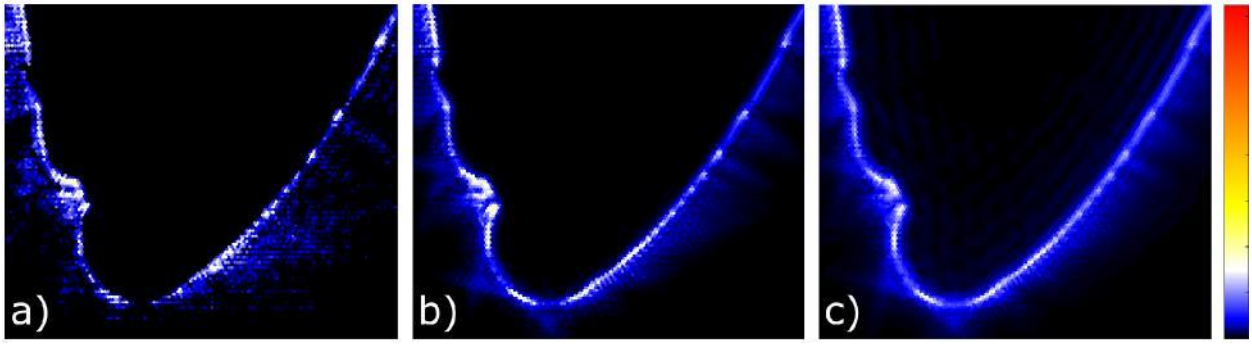

FIG. R2. The midgap LDOS maps ($E = 0$) for different values Δ: a) corresponds to physical value $\Delta = 1.25$ meV while b) and c) correspond to exaggerated values $\Delta = 10$ meV and $\Delta = 20$ meV respectively. The parameters are $t_1 = 40$ meV, $t_2 = 132$ meV, $t_3 = 12$ meV, $\alpha_R = 60$ meV and chemical potential in the bulk was $\mu_B = -111.2$ meV. Chemical potential and the Zeeman splitting on the island was $\mu_I = -260$ meV and the $V_Z = 85$ meV. The calculation has been carried out on a lattice with 10^5 sites.

The standard estimate for the edge mode penetration depth $\xi = v_F/\mathrm{Li}$ invoked by the referee

fails here. In fact, the effective determined by the narrow bright strip localized at the edge is largely insensitive to the underlying $NbSe_2$ pairing gap Δ as explicitly shown by our new calculation. In particular, by reducing the gap in the calculations from 20 meV to a realistic value 1.25 meV does not lead to 20 times larger edge penetration depth as implied by the referee. This counter-intuitive fact can be understood by considering a dilute case where magnetic moments are placed on the substrate so that their separation is larger than the Fermi wavelength. In this regime one can derive an effective subgap Bogoliubov-de Gennes Hamiltonian for a 1D chain (Pientka et al. PRB 88, 155420 (2013)) or a 2D ferromagnetic lattice with Rashba coupling (Ro¨ntynen and Ojanen, PRL 114, 236803 (2015)). The effective BdG Hamiltonian describing the subgap physics is such that the "particle-hole block" and the "pairing block" are both proportional to Δ of the substrate. Thus, estimating from the effective Hamiltonian shows that Δ is in fact cancelled (though it is crucial that it remains finite).

This phenomenon is essentially the same which is called "Fermi velocity renormalization" by Peng et al. PRL 114, 106801 (2015). That work physically explains why in the original Princeton experiment with 1D Fe chain on Pb substrate, the edge mode localization was orders of magnitude smaller than the naive estimate (result similar to our work in 2D) with the added feature that the role of the electronic d-orbitals in the magnetic atoms was included. Since the spectral weight from the d-states is expected to be very small when the atoms are strongly hybridized with the substrate, this case is practically the same as the pure Shiba case (for lucid exploration of these matters see the talk by Leonid Glazman https://www.youtube.com/watch?v=scA-PeTm9KM).

The picture remains essentially valid when making the magnetic lattice denser (as also demonstrated by the Princeton experiment which is not in the dilute limit). We observe that, in analogy to the 1D case, in addition to the bright narrow edge signal insensitive to Δ, there exists a strongly suppressed tail of which decay reflects Δ. However, this contribution is so weak compared to the narrow bright edge signal that it is below experimental resolution.

In summary, it is well-established theoretically and experimentally that the naive estimate for

yields orders of magnitude wrong answer for the penetration depth of the topological states in magnetic structures and our theoretical model indeed reproduces the previously observed insensitivity of effective on bulk Δ. This mechanism is perfectly operational independently of the dimension of the substrate. As discussed in our reply to the first comment, our model reproduces convincingly various non-generic aspects of the experimental results without signatures of being "oversimplified" due to 2D description. Our work, together with the previous work on single impurities (Nat. Phys. 11, 1013 (2015)), shows that the effective 2D modelling is an excellent starting point in studying the subgap physics of superconducting $NbSe_2$ with low-dimensional magnetic structures.

Action: We have performed large scale numerical calculations in lattices with over 10^5 lattice sites. While reproducing all the previous calculations in the full energy range with

the realistic value of Δ would require extensive computational resources and sophisticated numerics beyond exact diagonalization, we have calculated the midgap LDOS maps for Δ = 20, 10 and 1.25 meV (realistic value). These results clearly show that the edge mode signal is roughly independent of Δ and in excellent agreement with the experimentally observed value. These results are included in the SI of the revised manuscript. We have also confirmed that the predicted subgap LDOS shape is insensitive to Δ as long as the linear size of the lattice is sufficiently large ($L >> t/\Delta$).

Referee comment 3. The size of the Rashba spin-orbit coupling was assumed to be very large (60 meV) in the model, but I did not find how the authors justify this value. They may at least estimate the Rashba SOC from their DFT calculations.

Author reply: We thank the referee for the suggestion. Firstly we point out that we switched our first principles code from Quantum Espresso to VASP. We found that VASP has much fewer restrictions on spin polarization while using van-der Waals functions and is also more suitable for calculations considering magnetization and spin-orbit coupling simultaneously.

We have added the Supplementary Figures 3a-f, showing the band structures obtained considering spin-orbit coupling (SOC), as well as the bands with and without SOC obtained through an unfolding procedure. The unfolded bands reveal that, besides the spin splitting, the bands are discontinuous near the M point, clearly indicating that the Nb atoms in the heterostructure are no longer identical, thus lifting this degeneracy with the same order of magnitude as the observed spin splittings. As can be seen in Supplementary Figure 1, in all stacking configurations considered, one Nb atom is always located in a different site relative to the $CrBr_3$ layer, resulting in different couplings between the Nb atom and the $CrBr_3$ layer in the same heterostructure. To circumvent this problem, it would require the inclusion of SOC effects in large supercell structures where all Nb atoms couple equally to the $CrBr_3$ layer, or at least large enough to average the coupling from each Nb atom, which is beyond any current computational capability.

As the band structures shown in Supplementary Figures 3d and 3e were obtained considering all the effects simultaneously (induced magnetization + SOC + different coupling between each Nb atom with the $CrBr_3$ layer), many states are observed crossing the spin polarized band gap at the Γ point, mainly due to the non equivalency between the Nb atoms. However, a spin inversion is observed in the vicinity of the M point in the unfolded band structure, compatible with the spin inversion in the Rashba effect. As the spin polarized band gap is kept when SOC is taken into account, we conclude that the Rashba effect is of the same order of magnitude as the spin splitting due to the induced magnetization.

Finally, our theoretical model, which correctly reproduces the essential observed experimental features also predicts that for the observed magnitude of the topological gap, the Rashba constant and magnetization have the same order of magnitude. This consideration is also consistent that the Rashba coupling is several tens of meV.

Action: We have performed additional band structures calculations considering SOC and an unfolding procedure was used in an attempt to estimate the Rashba coupling. These results were added to our revised manuscript in Supplementary Figure 3. An additional discussion about the SOC effects and the order of magnitude of the Rashba coupling have beeb added to pages 6 and 7 of the Supplementary Material.

Referee comment 4. The subgap peaks at 0.41 meV are not explained. These should be due to weakly dispersive Shiba bands. Understanding their origin in a more quantitative way should help to build a better model of the superconductivity at the interface.

Author reply / action: The referee is correct here and we have changed the wording the manuscript to make this clearer.

Referee comment 5. A few typos: (1) H_{SOC} in Eq. (1) in the SI misses a spin Pauli matrix vector in the first term. (2) V_S exists in Eq. (1) of SI but does not appear in Eqs. (2-5)

Author reply / action: We thank the referee for pointing out these typographical errors, which have been corrected in the revised manuscript. V_S term is included in E_t, defined as $E_t(k) = \epsilon(k) - \mu + V_S$ after Eq. (3).

Reviewer Reports on the First Revision:

Ref # 1

This paper focuses on the observation of propagating Majorana states on the edges of magnetic CrBr3 islands grown on bulk NbSe2. The experimental proof is the measurement of a sharp peak in the density of states on the edges of the islands. They simulate the STM results by solving a tight-binding Hamiltonian on a triangular lattice with periodic boundary conditions.

I have read the referees rebuttal to my previous comments. One big problem with the paper is that the authors need to make many assumptions to justify their claim but provide little to no evidence to support these assumptions. Many of my previous comments have not been addressed in this version.

The paper does not meet the standards of Nature either in novelty (there are already other papers claiming propagating Majorana modes along edges) or robustness.

My detailed comments are below.
1) The figure below on the left shows a spin-split states for NbSe2 (as obtained from the paper). According to the authors, to make their model work, the Fermi energy must move down by 100-200meV such that it intersects one of the spin split bands at the M point of the Brilluion zone. For the simple band structure shown in Fig 2a and Fermi level shift, the authors calculate that the Chern number must be 3. However, the actual band structure calculated is much more complicated. The authors calculate the band structure including band folding for the heterostructure in the supplement. Now, when the Fermi energy moves down, it intersects multiple bands and there is no clean regime where only one spin is involved. It is not even simple to calculate the number of band crossings. And one must remember that this is a model for a magnetic system which involves many assumptions about energy scales. It is not at all clear that they are in the topological regime. (Note: finding a peak in the density of states theoretically does not mean these are Majorana modes).
2) A critical feature of their construction is that they require a ~100meV-150meV Fermi level shift in NbSe2 which I would stress is very large for a metal. The authors however provide no proof of the Fermi energy shift. A calculation saying this is plausible is not sufficient and is certainly not proof. ARPES data on a monolayer (or close to that) of CrBr3 on NbSe2 would show this shift in the bands. Also, the authors are incorrect in saying that this cannot be measured by STM. STM spectra obtained over higher energies would show this shift if it exists. The authors would find a feature in the density of states at higher energies and track it across the island. This feature should shift as the potential changes. This is a simple check. But the authors provide no such data. This is a very large assumption to make without experimental proof.
3) An important aspect of topological states is that they should be continuous along the boundary. The authors attribute the multiple gaps in seeing these states to a resolution issue. However, the peak at zero bias is district and large. This peak should be visible in their data even if it is broadened. In their theory for example, the state is suppressed but never disappears. It is therefore extremely strange that there are areas all along the boundary (for every boundary measured) where this peak just cannot be seen. This is not a matter of detail. This one of the requirements to prove that the states are topological. The lack of continuity cannot be explained away and is not consistent with a topological edge mode.
4) The density of states for propagating states should be linear around zero (even according to their calculations). This cannot give a peak in the density of states. A peak typically represents a bound state or perhaps many bound states closely spaced.
5) The spatial extent of the edge modes should increase with increasing energy, eventually merging into the bulk bands. The states seen experimentally do not broaden with increasing energy (Fig. 3 c,d,e and f). All that seems to happen is that at higher energies one begins to see the Shiba bands. This is not consistent with topological states.

6) The authors use an artificially large gap of 20meV (approximately 20 times larger than the actual gap) to simulate the STM data. The other energy scales in the problem are similar (~50-100meV). These are all within an order of magnitude. The use of the large gap in simulations is therefore not justified.
7)The authors should make it clear whether they are in the regime of Fig.1b (reference 35 and 36) or in the regime of dominant magnetization talked about in terms of Shiba states (references 28-30). The hierarchy of energy scales for these two regimes is different. Both scenarios cannot apply.
To summarize: the authors claims rest on a host of plausibility arguments with very little experimental proof. Moreover, the data they have does not match what is expected for a Majorana state in three critical aspects (a) the density of states, (b) the continuity along the boundary and (c) the spatial spread with energy.

Ref # 2

The authors addressed my questions properly. I would like to recommend it publish in Nature, although I still concern a little about the significance of the work.

Ref # 3

The new calculations in the revised manuscript have substantiated the main conclusions and have addressed some of my concerns mentioned in the previous report. However, I have a couple of questions regarding their new results and hope the authors could address them before I can make my recommendation.

(1) The boundary of the magnetic island is defined by a thickness in which the Zeeman field smoothly drops to zero. In 1D such a smooth confinement potential may lead to quasi-Majorana modes even when the bulk is topologically trivial [see e.g. PRB 86, 100503 (2012)]. Can the authors provide more numerical evidence on whether the zero-energy features are really Majoranas by e.g. showing the dependence of the decay length on the parameter l used on page 15 of the SI, and showing whether the zero-energy features at the boundary disappear in the topologically trivial regime?

(2) The authors tried to estimate the Rashba SOC using DFT but did not quite succeed due to practical limitations. While this is understandable, the new supplementary Figure 3 raises some concerns. Comparing the top panels of figure b and e one can see that the intrinsic spin-orbit splitting of NbSe2 is on the order of 10^2 meV. Since the Rashba-type SOC comes from mirror-symmetry breaking due to the presence of CrBr3 as a 2nd-order effect, and the coupling between the two layers is rather weak, it is hard to believe that it could be as large as 60 meV as claimed by the authors. Although it is hard to directly obtain the Rashba SOC from the DFT results, the authors can e.g. compare the DFT and model results of the spin splitting along Gamma-M before and after switching on the spin-orbit coupling.

Author Rebuttals to First Revision:

REFEREE #1:

Overall referee comments:

This paper focuses on the observation of propagating Majorana states on the edges of magnetic CrBr3 islands grown on bulk NbSe2. The experimental proof is the measurement of a sharp peak in the density of states on the edges of the islands. They simulate the STM results by solving a tight-binding Hamiltonian on a triangular lattice with periodic boundary conditions.

I have read the referees rebuttal to my previous comments. One big problem with the paper is that the authors need to make many assumptions to justify their claim but provide little to no evidence to support these assumptions. Many of my previous comments have not been addressed in this version.

The paper does not meet the standards of Nature either in novelty (there are already other papers claiming propagating Majorana modes along edges) or robustness.

Author reply: We thank the referee for their time on the review process. Naturally, we disagree in the strongest possible terms with the above characterization of our work. In contrast to the views of Referee #1, Referee #2 is recommending our paper to be published in Nature and Referee #3 wrote already in the first round that "I feel the work is worth publishing Nature or another high-profile journal".

The comment "many of my previous comments have not been addressed in this version" is incorrect - we provided an extraordinarily detailed 7 page refutation of all the criticism. It is remarkable that the central criticism of Referee #1 in the previous round of reviews, namely that topological superconductivity cannot, *even in principle*, be induced on a topologically trivial bulk superconductor by contact with an insulating magnetic material, is no longer addressed at all. We outlined (with several key references) that the claim by the reviewer is factually incorrect. Instead of acknowledging our reply, Referee #1 now concentrates on a handful of other issues. This seems to imply that Referee #1 now agrees with us on this point.

The essential aspects of comments 4.-6. below by Referee #1 were already addressed in our previous response to Referees #2 and #3. This lead to a number of improvements to the resubmitted manuscript, and Referees #2 and #3 now indicate that they are satisfied with our replies. However, Referee #1 is not addressing our last round response to these issues.

Below, we reply in detail to all the additional concerns of Referee #1.

Referee comment 1. The figure below on the left shows a spin-split states for NbSe2 (as obtained from the paper). According to the authors, to make their model work, the Fermi energy must move down by 100-200meV such that it intersects one of the spin split bands

at the M point of the Brilluion zone. For the simple band structure shown in Fig 2a and Fermi level shift, the authors calculate that the Chern number must be 3. However, the actual band structure calculated is much more complicated. The authors calculate the band structure including band folding for the heterostructure in the supplement. Now, when the Fermi energy moves down, it intersects multiple bands and there is no clean regime where only one spin is involved. It is not even simple to calculate the number of band crossings. And one must remember that this is a model for a magnetic system which involves many assumptions about energy scales. It is not at all clear that they are in the topological regime. (Note: finding a peak in the density of states theoretically does not mean these are Majorana modes).

Author reply: As we stressed in our previous replies to all the referees, our theoretical approach has been previously employed in explaining the subgap magnetic states with quantitative agreement with experimental results (Ref. 37). Also, our model explains various independent experimental observations, e.g. 1) the correct edge mode penetration depth, 2) the specific form of the subgap local density of states, which depends on system-specific dispersion of the topological edge modes, and 3) a non-generic coexistence of the topological edge modes and bulk states in a substantial energy window.

Regarding the complexity of the folded bulk electronic band structure, this is essentially irrelevant for the modelling of the subgap magnetic states localized at the top layer. Indeed, compared to a monolayer, the bulk $NbSe_2$ has a number of additional bands dispersing perpendicular to layers. However, the layers are so efficiently decoupled that the additional bands with 3D character do not affect the subgap magnetic states localized near the top layer. The irrefutable proof of this is the pioneering work by Menard et al. (Ref. 37), where the Shiba states in bulk $NbSe_2$ are observed to have definite two-dimensional character. If the additional 3D bands (that we and Ref. [37] leave out from the theoretical model) had a measurable effect on the magnetic subgap states, the Shiba states in Ref. [37] would not have the observed 2D shape ($1/\sqrt{\rho}$ decay) but exhibit 3D characteristic ($1/\rho$ decay). As the pioneering work [37] and our own work shows, the effective 2D modelling (neglecting the 3D bands) captures all the essential features of the subgap magnetic states. Thus, the fact that our system is indeed in the topological regime is determined by the effective 2D model. Topological superconductivity with finite Chern numbers is always effectively 2D phenomenon by definition. It still takes place in 3D world as long as the relevant physics is effectively two-dimensional as in the studied system. Adding further complexity to the model is not reasonable as it already successfully explains all the observed features.

We are completely aware that a peak in LDOS does not necessarily indicate a Majorana state. However, all our findings are consistent with a topological Majorana mode, but not with anything else known to date. In this respect, our work is at least on the same level as previous high-impact publications on topological superconductivity and substantially more convincing than any previous work on chiral 2D topological superconductivity.

Referee comment 2. A critical feature of their construction is that they require a 100meV-150meV Fermi level shift in NbSe2 which I would stress is very large for a metal. The authors however provide no proof of the Fermi energy shift. A calculation saying this is plausible is not sufficient and is certainly not proof. ARPES data on a monolayer (or close to that) of CrBr3 on NbSe2 would show this shift in the bands. Also, the authors are incorrect in saying that this cannot be measured by STM. STM spectra obtained over higher energies would show this shift if it exists. The authors would find a feature in the density of states at higher energies and track it across the island. This feature should shift as the potential changes. This is a simple check. But the authors provide no such data. This is a very large assumption to make without experimental proof.

Author reply: The suggestion of the referee on carrying out ARPES experiments on monolayer $CrBr_3$ on $NbSe_2$ is an important topic for further study, but clearly outside the scope of the present manuscript. We do not have access to ARPES in our lab and the required energy resolution would in any case necessitate low-temperature experiments with very good energy resolution (synchrotron-light source or laser-ARPES), which is a very specialized experiment on its own right. These types of experiments would only be available on a time scale > 6 months, which rules them out in the present case.

It is not easy to accurately measure the electronic bands of $NbSe_2$ through the insulating $CrBr_3$ layer. This problem is further compounded by the fact that STS is most sensitive to the states close to the Γ point and not the M point which would be more relevant in this case. This phenomenon arises from nonuniform sampling of the states with different in-plane momenta [1, 2].

Referee comment 3. An important aspect of topological states is that they should be continuous along the boundary. The authors attribute the multiple gaps in seeing these states to a resolution issue. However, the peak at zero bias is district and large. This peak should be visible in their data even if it is broadened. In their theory for example, the state is suppressed but never disappears. It is therefore extremely strange that there are areas all along the boundary (for every boundary measured) where this peak just cannot be seen. This is not a matter of detail. This one of the requirements to prove that the states are topological. The lack of continuity cannot be explained away and is not consistent with a topological edge mode.

Author reply: As already discussed extensively on our last round response to Referee #2, the edge state signal is completely consistent with our theory of topological edge modes. As the large scale simulations in the Supplementary Fig. 6 show, the LDOS signal on irregular edge such as the one in the experiment exhibits so strong intensity variations (in LDOS) that it appears ”cut-off”. Since we know for certain that the simulation result corresponds to a topological edge mode, there is no question whether it can look effectively ”cut-off” in places. This effect is obviously even stronger in an experimental situation since the experiment has limited sensitivity, a fact that leads to even stronger changes in the ”visibility” of the edge

mode. In contrast to what the referee insists, the LDOS topological edge modes can look very irregular and granular. Still, the edge mode in reality is fully continuous and supports quantized responses.

Referee comment 4. The density of states for propagating states should be linear around zero (even according to their calculations). This cannot give a peak in the density of states. A peak typically represents a bound state or perhaps many bound states closely spaced.

Author reply: The claim that, for all topological superconductors, edge mode dispersion should be linear, giving rise to a constant density of states is incorrect. The referee is probably referring to the calculations shown in Fig. 2d, which show the band structure calculated in a ribbon geometry. However, there are important factors that invalidate the point of the referee. The LDOS, unlike the DOS, is determined by a local shape of the wave functions near the edge. Thus, contrary to what the referee implies, there is no way to simply "see" from the band structure that the LDOS should be "linear around zero". Additionally, the irregular shape of the edge, as in the experiment and calculations, lead to strong intensity variations along the edge. This has been established beyond any doubt in our calculations, where we know for the fact that the system is in the topological state with Chern number 3. In general, making generic claims regarding theoretical calculations without references to technical details is not helpful.

Referee comment 5. The spatial extent of the edge modes should increase with increasing energy, eventually merging into the bulk bands. The states seen experimentally do not broaden with increasing energy (Fig. 3 c,d,e and f). All that seems to happen is that at higher energies one begins to see the Shiba bands. This is not consistent with topological states.

Author reply: The above statement of the referee is incorrect and again, this point was already discussed in the reply to the comments of the other referees in the previous round (Referee #2, comment 2 and Referee #3, comment 2). The broadening is not observed even at energies comparable to the topological gap, and this is a real feature also arising in the simulations (where it is certain that we are dealing with a topological edge mode). This is related to other system-specific features of the edge modes such as the anomalously narrow penetration depth discussed in the response to Referee #3 in the last round. As explained already then, these features are fingerprints that strongly support the validity of our theoretical model and the interpretation of our experimental results.

Referee comment 6. The authors use an artificially large gap of 20meV (approximately 20 times larger than the actual gap) to simulate the STM data. The other energy scales in the problem are similar (50-100meV). These are all within an order of magnitude. The use of the large gap in simulations is therefore not justified.

Author reply: This issue was already discussed in detail in the previous round in response to Referee #3 (comment 2) and the referee seems to have missed that and the Supplementary Fig. 6 in the revised manuscript addressing this issue. Referee #3 indicated that this was solved in their opinion.

As discussed earlier in detail, it has been discovered in a number of previous works that the edge mode penetration depth of magnetic systems is largely insensitive to the underlying magnitude of the gap. Thus, the edge mode LDOS is essentially similar for the exaggerated gap. Furthermore, we provided large scale simulation results, which explicitly prove this point (down to realistic gap sizes), see Supplementary Fig. 6a.

Referee comment 7. The authors should make it clear whether they are in the regime of Fig.1b (reference 35 and 36) or in the regime of dominant magnetization talked about in terms of Shiba states (references 28-30). The hierarchy of energy scales for these two regimes is different. Both scenarios cannot apply.

Author reply: Our system most closely resembles the dense-impurity situation originally studied in Ref. [29] as is evident in the theoretical treatment in the Supplementary Informa-tion. However, the generic conditions of the topological superconductivity (in-plane Rashba coupling, out-of plane magnetization and σ-wave superconductivity) are the same for these models, so the pictorial description in Fig. 1b applies.

Referee summary comment. The authors claims rest on a host of plausibility arguments with very little experimental proof. Moreover, the data they have does not match what is expected for a Majorana state in three critical aspects (a) the density of states, (b) the continuity along the boundary and (c) the spatial spread with energy.

Author reply: The central criticism of Referee #1 in the previous round of reviews, namely that topological superconductivity cannot, *even in principle*, be induced on a topologically trivial bulk superconductor by contact with an insulating magnetic material, is no longer mentioned at all. We outlined (with several key references) that the claim by the reviewer is factually incorrect. Instead of acknowledging our reply, Referee #1 has switched their previous position described by ”not possible even in principle” to ”data does not match” and invoked a number of issues which we settled in our response to the other referees to their satisfaction in the previous round.

The answers to these ”new” concerns - energy and spatial dependence of the edge mode LDOS - were already provided in the replies to Referees #2 and #3 in the previous round of reviewing. We reply to these concerns again above. All these ”issues” are fully consistent with our theoretical model describing a topological superconductor with Chern number 3.

REFEREE #2**:**

Overall referee comments:

The authors addressed my questions properly. I would like to recommend it publish in

Nature, although I still concern a little about the significance of the work.

Author reply: We thank the referee for their recommendation to publish the manuscript in Nature. To reiterate the central novel aspects of our work: 2D topological superconductivity has not been conclusively demonstrated to date, the previous reports have serious shortcomings (as outlined in our reply below) and our work goes significantly beyond the state-of-the-art in terms of both further opportunities offered by the type of the samples we use (vdW heterostructures) and the quantitative level of theoretical understanding of the experimental results. In addition to the paradigmatic recipe for 2D topological superconductivity that we demonstrate here (superconductivity, out-of-plane ferromagnetism and Rashba spin-orbit coupling), there are other proposals for topological superconductivity including the recent report (Science 367, 104 (2020)) on the observation of bound states at (half-unit cell shift) domain walls in $FeTe_{0.55}Se_{0.45}$. This system is controversial and certainly not proved to be topological "beyond reasonable doubt". Our results are the first convincing demonstration of 2D topological superconductivity.

REFEREE #3:

Overall referee comments:

The new calculations in the revised manuscript have substantiated the main conclusions and have addressed some of my concerns mentioned in the previous report. However, I have a couple of questions regarding their new results and hope the authors could address them before I can make my recommendation.

Author reply: We thank the referee for their efforts and constructive criticism which has improved the manuscript. Below we have addressed the remaining two inquiries below. In the light of the positive assessment already in the previous review round, we are optimistic about the final recommendation of the referee.

Referee comment 1. The boundary of the magnetic island is defined by a thickness in which the Zeeman field smoothly drops to zero. In 1D such a smooth confinement potential may lead to quasi-Majorana modes even when the bulk is topologically trivial [see e.g. PRB 86, 100503 (2012)]. Can the authors provide more numerical evidence on whether the zero-energy features are really Majoranas by e.g. showing the dependence of the decay length on the parameter l used on page 15 of the SI, and showing whether the zero-energy features at the boundary disappear in the topologically trivial regime?

Author reply: The effect of smoothed magnetic confinement can be definitely ruled out as the source of the edge signal. Already in the previous version of resubmitted manuscript we included the large-lattice LDOS simulations where the magnetization abruptly dropped to zero from the bulk value, see Supplementary Fig. 6. This proves that the edge signal in the topological state is practically the same for a sharp edge and smoothed edge. To rule out the option that the edge signal could be generated in a trivial state due to different magnetic profiles, in Fig. R1 we have included the LDOS maps corresponding to a trivial state for a sharp and smoothed magnetic edge. The figure clearly shows that no spurious edge signals in the trivial state are generated by different prescriptions and that a trivial state cannot be confused with a topological state. Thus, the nature of the magnetic edge (sharp vs. smooth) has no consequences on our conclusions.

Referee comment 2. The authors tried to estimate the Rashba SOC using DFT but did not quite succeed due to practical limitations. While this is understandable, the new supplementary Figure 3 raises some concerns. Comparing the top panels of figure b and e one can see that the intrinsic spin-orbit splitting of NbSe2 is on the order of 10^2 meV. Since the Rashba-type SOC comes from mirror-symmetry breaking due to the presence of CrBr3 as a 2nd-order effect, and the coupling between the two layers is rather weak, it is hard to believe that it could be as large as 60 meV as claimed by the authors. Although it is hard to directly obtain the Rashba SOC from the DFT results, the authors can e.g. compare the

FIG. R1. Left: Midgap LDOS for a trivial C = 0 state for a sharply-defined magnetic edge and uniform chemical potential μ = □111.2 meV over the sample. Middle: The same as on the left but for a smoothed magnetic edge with l = 2.2a. Right: Midgap LDOS for topological C = 3 state for smoothed edge l = 2.2a and chemical potential μ_B = □111.2 meV on the bulk and μ_I = □260 meV on the island. In all the figures, the other parameters are t_1 = 40 meV, t_2 = 132 meV, t_3 = 12 meV, α_R = 60 meV, Δ = 20 meV and V_Z = 85 meV.

DFT and model results of the spin splitting along Gamma-M before and after switching on the spin-orbit coupling.

Author reply: Since the Rashba coupling in the theoretical model includes only first neighbour hopping, its validity is restricted to the vicinity of high symmetry points Γ, K and M where it vanishes (the last being the relevant one for our work). To obtain a more comprehensive description would require inclusion of higher hopping terms, which in turn would require accurate knowledge of the spin splitting over the whole Brillouin zone (not available from DFT). Thus, plotting of the splitting on the full Γ-M line is not meaningful. However, we can motivate the order of magnitude of the Rashba splitting in the vicinity of M point in the theoretical model using the information from the DFT calculation. In the Supplementary Fig. 3c and f we have plotted the DFT results with and without spin-orbit effects (magnetization is present in both cases since this and the Rashba splitting are both induced by the same source). The latter result is also included in Fig. R2 below. It shows that, despite the discontinuous structure of the bands as explained in the Supplementary Information, there exists a magnetization induced gap at the M point. We also see that the ordering of the spin bands are switched there, signalling that the system exhibits a Rashba effect which has a comparable order of magnitude in the vicinity of M point. Thus, while exact values are elusive, we conclude that DFT predicts magnetization and Rashba splitting of tens of meV. In Fig. R2 we have also illustrated the Rashba splitting around the M point in the theoretical model. By fixing the parameter α = 60 meV in the Rashba term, we see that the splitting near the M point is a few tens of meV. (Note: the parameter α = 60 meV in the model does not imply that the Rashba splitting at M point is 60 meV since the Rashba term vanishes there for symmetry reasons.)

To summarize, our calculations and similar available in the literature [3–5] allow us to estimate the order of magnitude of magnetization and Rashba coupling. DFT calculations

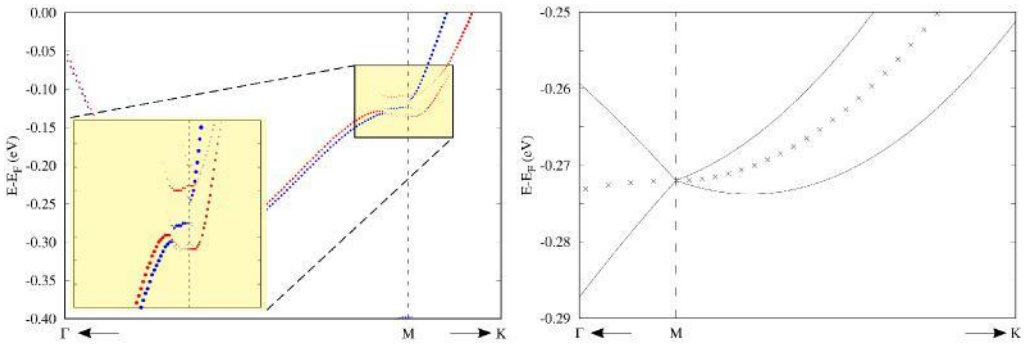

FIG. R2. Left: DFT unfolded band structure obtained considering spin-orbit coupling effects. Blue and red circles indicate the positive and negative projection of the Nb electrons' spin on the quantization axis, respectively. Right: band structure of the tight-binding model including the magnetization. The solid curves correspond to $\alpha = 60$ meV and the dashed curve shows the $\alpha = 0$ case for comparison.

predict that the Rashba splitting is comparable to magnetization which can be 10-100 meV. This is sufficient for the theoretical model. The relative magnitude of the Rashba coupling and the magnetic splitting determines the size of the topological gap which can be extracted from the experiment. The observed topological gap, in agreement with DFT considerations, predicts a comparable orders of magnitude of Rashba and magnetization effects. The precise value of Rashba coupling is not crucial since a moderate relative shift of the Rashba coupling would be compensated by a shift in magnetization (keeping the observed topological gap intact).

[1] Yuanbo Zhang, Victor W Brar, Feng Wang, Caglar Girit, Yossi Yayon, Melissa Panlasigui, Alex Zettl, and Michael F Crommie, "Giant phonon-induced conductance in scanning tunnelling spectroscopy of gate-tunable graphene," Nat. Phys. 4, 627–630 (2008).

[2] Philipp Leicht, Lukas Zielke, Samuel Bouvron, Riko Moroni, Elena Voloshina, Lukas Hammerschmidt, Yuriy S Dedkov, and Mikhail Fonin, "In situ fabrication of quasi-free-standing epitaxial graphene nanoflakes on gold," ACS Nano 8, 3735–3742 (2014).

[3] Huisheng Zhang, Wenjia Yang, Yaohui Ning, and Xiaohong Xu, "Abundant valley-polarized states in two-dimensional ferromagnetic van der Waals heterostructures," Phys. Rev. B 101, 205404 (2020).

[4] Klaus Zollner, Paulo E. Faria Junior, and Jaroslav Fabian, "Proximity exchange effects in $MoSe_2$ and WSe_2 heterostructures with CrI_3: Twist angle, layer, and gate dependence," Phys. Rev. B 100, 085128 (2019).

[5] Jingshan Qi, Xiao Li, Qian Niu, and Ji Feng, "Giant and tunable valley degeneracy splitting in $MoTe_2$," Phys. Rev. B 92, 121403 (2015).

Reviewer Reports on the Second Revision:

Ref # 2

The revised manuscript is much improved.
However, the comment#2 of referee#1 is also very important. I think the authors can provide more evidence to convince readers that there is the Fermi level shift in NbSe2. If there is the Fermi level shift under the CrBr3 island, a band bending should be observed near the island. The authors just need doing STS near the CrBr3 islands, if they can see any evidence for the Fermi level shift in NbSe2, the work will be much more convincing.
The Fermi level shift is the same for both the gamma point and the M point. STS should be sensitive enough to tell out the shift.

Once the authors can provide evidence for the the Fermi level shift in NbSe2, I would like to recommend the paper publish in Nature without any further review.

Ref # 3

The authors have addressed my first question in the previous report. To elaborate on my second question: The spin splitting near M point in the DFT calculation with SOC may also be due to the intrinsic Ising-like SOC Of NbSe2, which is why I suggested to look at the spin splitting along Gamma-M where it should vanish. Although the authors cited Ref. 20 in the SI which suggests the intrinsic spin splitting in 3D NbSe2 near M (kz = 0) is small, their DFT calculations are using a monolayer NbSe2 and the conclusion in that paper may not apply. Could the authors provide more evidence showing that the SOC-induced splitting in Fig. 3f of the SI is not mainly due to the intrinsic Ising SOC of monolayer NbSe2?

Ref # 4

After reviewing this paper after it has gone through several rounds of review and the comments from Reviewers 1, 2 and 3, I am in favour of publication. There is strong experimental evidence for topological superconductivity, and it has all the hallmarks of a Majorana mode. They provide detailed theory and calculations which reproduce the key experimental features. But I do agree with Reviewer 2 on the slight concern of significance, but think it should be either published in Nature or definitely in [REDACTED]. I will leave this to the editor.

I disagree with Reviewer 1 in their claims regarding lack of novelty and robustness. This is an excellent and thorough paper, and very strong evidence for topological superconductivity. They also successfully answered the majority of Reviewer 1 comments to a level that I am satisfied with.

Reviewer 1 however does raise one claim that I do agree with, the shift of NbSe2 bands by 100-150 meV should be explored in more detail. I agree with the authors that an ARPES measurement is beyond the scope of the paper and that an STS measurement probing LDOS is unlikely to clearly resolve these bands at the M point.
However, there are two potential things that could be explored or atleast discussed in the paper.
1. Have they considered a QPI measurement? The islands of CrBr3 in Figure 1 appear large enough that detailed QPI mapping could be carried out. This would perhaps enable measurement of this shift at the M point, if this is not possible can they explain why? They mention that the NbSe2 states are difficult to measure with STS underneath the CrBr3 monolayer, but I would like to see evidence of this across a larger energy range.

2. This shift may actually be explained from a band alignment argument and lack of Fermi level pinning in this type of vdW junction. The work function of 2H-NbSe2 is between 5.6-5.9 eV [Y. Liu, et

al., Sci. Adv. 2, e1600069 (2016) and Shimada et al., Japanese Journal of Applied Physics, 33, 2696 (1994)] whilst, the electron affinity of monolayer CrBr3 is predicted to be 5.1 eV [Zhang et al., Phys. Chem Chem Phys. 21, 17087 (2019)]. This indicates that there should indeed be a shift in the NbSe2 due to spontaneous charge transfer.

Furthermore, in the Liu et al., paper they also demonstrate that there is very weak Fermi level pinning in a van der Waals metal-semiconductor junction, and I assume the NbSe2/CrBr3 junction would be similar. I think it would be useful for the authors to mention this possibility, and ideally a calculation on the magnitude of such a shift based on band alignment.

Whilst, there will be some uncertainty in the work function of NbSe2 in the superconducting state this addition would add plausibility to the assumption of such a large shift in NbSe2 bands as required by the model.

Author Rebuttals to Second Revision:

REFEREE #2**:**

Overall referee comments: The revised manuscript is much improved. However, the comment #2 of referee #1 is also very important. I think the authors can provide more evidence to convince readers that there is the Fermi level shift in NbSe2. If there is the Fermi level shift under the CrBr3 island, a band bending should be observed near the island. The authors just need doing STS near the CrBr3 islands, if they can see any evidence for the Fermi level shift in NbSe2, the work will be much more convincing. The Fermi level shift is the same for both the gamma point and the M point. STS should be sensitive enough to tell out the shift.

Once the authors can provide evidence for the the Fermi level shift in NbSe2, I would like to recommend the paper publish in Nature without any further review.

Author reply: We thank the referee for their positive comments and recommending publication of our manuscript in Nature once there is evidence of a shift of the Fermi level under the $CrBr_3$ islands. We agree with them (and other reviewers) that this is a very important point. Motivated by their comments, we have carried out additional experiments to uncover a possible shift of the $NbSe_2$ bands under $CrBr_3$.

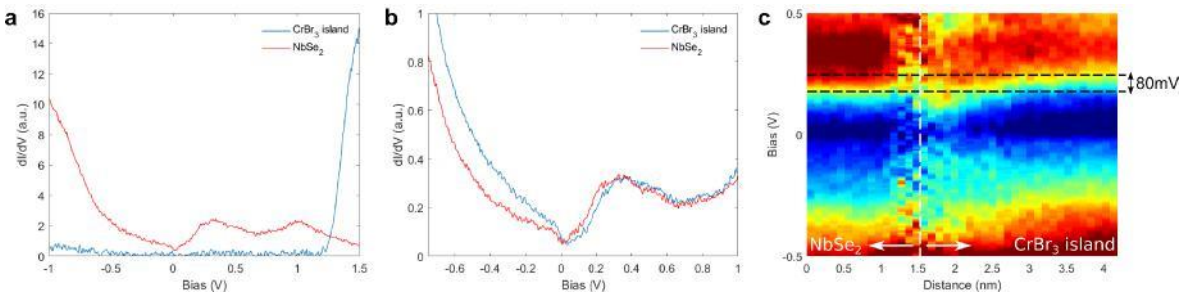

FIG. R1. **Comparison of d*I*/d*V* spectra on bare NbSe2 and on CrBr3. a**, large bias range spectra on $NbSe_2$ (red line) and $CrBr_3$ (blue line). **b,** Spectroscopy within the gap of the $CrBr_3$ on

$NbSe_2$ (red line) and $CrBr_3$ (blue line). **c,** Spectra recorded along a line from $NbSe_2$ to $CrBr_3$ showing the shift of ca. 80 meV.

Figure R1 shows the results of careful spectroscopy experiments focusing on the position of the spectroscopic features arising from the Nb *d*-band, which is the relevant band for superconductivity that we also use as an input in the tight-binding calculations. Fig. R1a shows large bias range overview spectra. While the measurement on bare $NbSe_2$ shows the *d*-band close to zero bias, the spectrum on the $CrBr_3$ island only shows the conduction band onset (at a bias of 1.2 V) and some very faint features at smaller bias. However, focusing on biases closer to zero and increasing the setpoint current sufficiently, allows also resolution of the Nb *d*-band features under the $CrBr_3$. This is shown in Fig. R1b, where there is a clear shift of the spectroscopic features towards higher energy under $CrBr_3$. This evolution is tracked more systematically in Fig. R1c, which shows spectra recorded along a line starting from the bare $NbSe_2$ substrate and ending on the $CrBr_3$ island. There is a clear shift of ca. 80 mV.

There is some uncertainty on the exact energy position of the Nb *d*-bands at the *M*-point of the Brillouin zone. The best estimates based on DFT-calculations and ARPES experiments on bulk $NbSe_2$ put this feature at ca. 100 meV below the Fermi level (Borisenko et al. Phys. Rev. Lett. 102, 166402 (2009), Ugeda et al. Nat. Phys. 12, 92 (2016)). In this case, our experimentally observed upward shift of 80 meV would bring the *M*-point very close to the Fermi level, which offers further support for our conclusions.

Action: We have added the panels b and c of the above figure to the Extended Data of the revised manuscript and revised the associated discussion in the manuscript to:

"While the *M*-point is at ~270 meV below the Fermi level in our TB model (calculated phase diagram shown in Fig. 2b), it is estimated to be ~100 meV below Fermi level in bulk $NbSe_2$ [38,39].This implies that for a reasonable magnetization of $M_{r'}$<100 meV, we need slight doping to bring the experimental system into the C = 3 state. This is precisely what we observe by following the Nb *d*-bands from bare $NbSe_2$ onto a $CrBr_3$ island. There is an upward shift of the $NbSe_2$ bands of ~80 meV under $CrBr_3$ (Extended Data Fig. 2)."

REFEREE #3**:**

Referee comment: The authors have addressed my first question in the previous report.

To elaborate on my second question: The spin splitting near M point in the DFT calculation with SOC may also be due to the intrinsic Ising-like SOC Of NbSe2, which is why I suggested to look at the spin splitting along Gamma-M where it should vanish. Although the authors cited Ref. 20 in the SI which suggests the intrinsic spin splitting in 3D NbSe2 near M (kz = 0) is small, their DFT calculations are using a monolayer NbSe2 and the conclusion in that paper may not apply. Could the authors provide more evidence showing that the SOC-induced splitting in Fig. 3f of the SI is not mainly due to the intrinsic Ising SOC of monolayer NbSe2?

Author reply: We thank the referee for elaborating the question. The comparison of the band structures along the $\Gamma - M$ line shown in Fig. 3c (without SOC) and Fig. 3f (with SOC) show that the introduction of the SOC results in a shift and inversion of the order of bands when approaching the M point. This restructuring of bands is naturally attributed to the Rashba coupling since the intrinsic SOC in $NbSe_2$ vanishes along the $\Gamma - M$ line due to reflection symmetry. This argument invoking crystal symmetry works for both monolayer and bulk $NbSe_2$. Thus we can conclude that the SOC-induced splitting along the $\Gamma - M$ arises solely from the Rashba effect.

This fact is further elaborated in Fig. R2. The left panel shows that the intrinsic SOC effect of isolated monolayer $NbSe_2$ indeed vanishes at the M point and along the $\Gamma - M$ line.

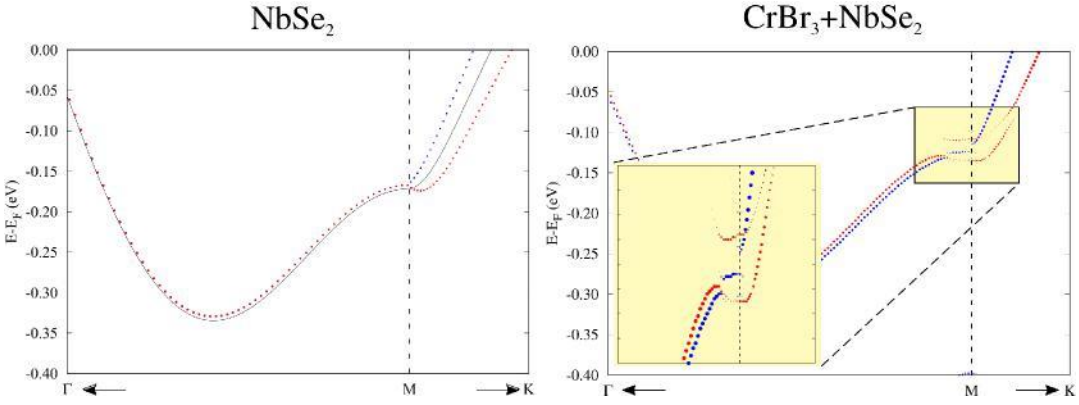

FIG. R2. Left: band structures of the isolated monolayer $NbSe_2$ with and without SOC. The black continuous line corresponds to the band structure without SOC. The dots correspond to the band structures with SOC, where the blue (red) color indicate a positive (negative) projection of the Nb electrons' spin on the quantization axis. Right: unfolded band structure with SOC of the $CrBr_3$-$NbSe_2$ heterostructure, where the blue (red) color indicate a positive (negative) projection of the Nb electrons' spin on the quantization axis.

This is a clear evidence that the effects observed in the band structure of the $CrBr_3$-$NbSe_2$ heterostructure (shown in right panel of Figure R2 and in Supplementary Figure 3f) is due to interactions of the $NbSe_2$ substrate with the magnetic layer, rather than intrinsic Ising-like SOC effects on the $NbSe_2$ itself.

Action: We have also added the reference Phys. Rev. B 96, 155439 (2017) (Ref 9) in SI where it reads: "There is no SOC splitting along $\Gamma - M$ while a large splitting is obtained along the $K - \Gamma$ line in the pristine $NbSe_2$ case[9], pushing the spin-up bands above the spin-down bands." (page 6 of SI).

REFEREE #4:

Overall referee comments: After reviewing this paper after it has gone through several rounds of review and the comments from Reviewers 1, 2 and 3, I am in favour of publication. There is strong experimental evidence for topological superconductivity, and it has all the hallmarks of a Majorana mode. They provide detailed theory and calculations which reproduce the key experimental features. But I do agree with Reviewer 2 on the slight concern of significance, but think it should be either published in Nature or definitely in [REDACTED]. I will leave this to the editor.

I disagree with Reviewer 1 in their claims regarding lack of novelty and robustness. This is an excellent and thorough paper, and very strong evidence for topological superconductivity. They also successfully answered the majority of Reviewer 1 comments to a level that I am satisfied with.

Author reply: We thank the referee for recommending publication of our paper and recog-nizing the quality of the experimental and theoretical results. We will reply to the remaining comments in detail below.

Referee comment 1. Reviewer 1 however does raise one claim that I do agree with, the shift of NbSe2 bands by 100-150 meV should be explored in more detail. I agree with the authors that an ARPES measurement is beyond the scope of the paper and that an STS measurement probing LDOS is unlikely to clearly resolve these bands at the M point.

However, there are two potential things that could be explored or at least discussed in the paper.

Have they considered a QPI measurement? The islands of CrBr3 in Figure 1 appear large enough that detailed QPI mapping could be carried out. This would perhaps enable mea-surement of this shift at the M point, if this is not possible can they explain why? They mention that the NbSe2 states are difficult to measure with STS underneath the CrBr3 monolayer, but I would like to see evidence of this across a larger energy range.

Author reply. We agree with the referee that in principle, QPI measurements could be used to probe the energy positions of the various high-symmetry points within the Brillouin zone. However, there are several experimental factors that render this approach difficult: (1) The QPI signal arises from scattering events that connect states at the energy given by the bias voltage, i.e. the nature of the scatters also matters and the QPI signal is not equal to the so-called joint-density of states (extreme example given on a topological insulator surface, see Roushan et al. Nature 460, 1106 (2009)). (2) The moire´ pattern of the $CrBr_3$ is quite strongly visible in topography images and dI/dV maps also at energies inside the gap of $CrBr_3$. Any possible QPI contrast would be superimposed on this strong background.

The spectroscopy measurement (elaborated in the reply to Referee #2 as a response to the question about the shift of $NbSe_2$ bands) is much simpler than a complete QPI measurement. While it does not directly give the energy of the band at the M-point, it gives the value of the shift (that should be the same across the Brillouin zone) of 80 meV. Together with independent estimates of the energy position at the M-point, this allows us to conclude that the Nb d-band at the M-point is very close to the Fermi level in our experimental system, which is precisely the requirement for pushing the system into the $C = 3$ topological state.

Referee comment 2. This shift may actually be explained from a band alignment argument and lack of Fermi level pinning in this type of vdW junction. The work function of 2H-NbSe2 is between 5.6-5.9 eV [Y. Liu, et al., Sci. Adv. 2, e1600069 (2016) and Shimada et al., Japanese Journal of Applied Physics, 33, 2696 (1994)] whilst, the electron affinity of monolayer CrBr3 is predicted to be 5.1 eV [Zhang et al., Phys. Chem Chem Phys. 21, 17087 (2019)]. This indicates that there should indeed be a shift in the NbSe2 due to spontaneous charge transfer.

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Whilst, there will be some uncertainty in the work function of NbSe2 in the superconducting state this addition would add plausibility to the assumption of such a large shift in NbSe2 bands as required by the model.

Author reply. We thank the referee for pointing out this way of estimating the shift of the $NbSe_2$ bands. As can be seen in Fig. R3, our DFT calculations show that all states between the VBM and CBM of the $CrBr_3$ come from the metallic substrate, which is expected for such a weakly interacting system. Thus, any metal-induced gap states are negligible and could therefore indicate weak Fermi level pinning in such heterostructures, as stated by the referee.

The barrier heights for electrons and holes are $\Phi_e = 0.45$ eV and $\Phi_h = 0.70$ eV, respectively (calculated through equation 1 of Y. Liu, et al.) and agree very well with the Schottky-Mott model, where we get $\Phi_e = 0.5$ eV (calculated through equation 2 of Y. Liu, et al. using the electron affinity of monolayer $CrBr_3$ as 5.1 eV and work function of 2H-$NbSe_2$ as 5.6 eV). Positive values for both Φ_e and Φ_h indicate that there is no spontaneous charge transfer either from or to the metallic substrate. While there is no indication of charge transfer within the limits of DFT, the new spectroscopic results shown in Fig. R1 offer sufficient evidence for the shift of the $NbSe_2$ states in the direction favouring the topological superconducting state.

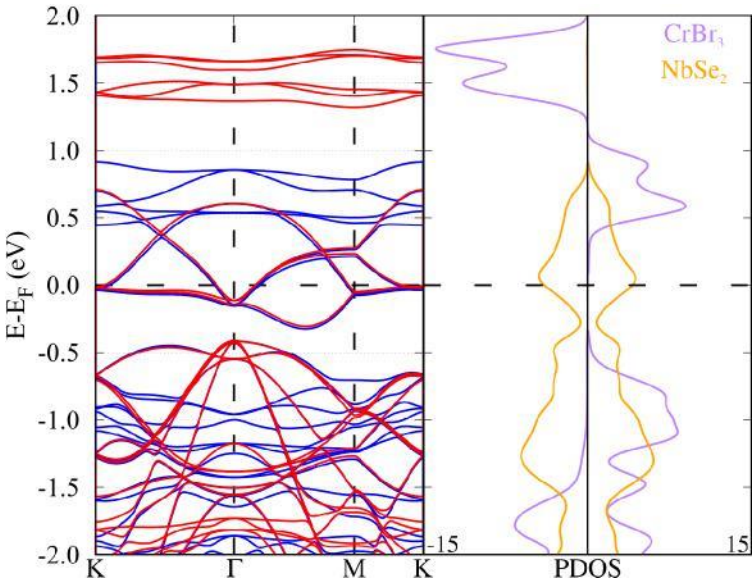

FIG. R3. Band structure and project density of states (PDOS) of the $CrBr_3$-$NbSe_2$ heterostructure. Spin up and down channels are represented by blue and red colors in the band structure, respectively, while in the PDOS they are represented by positive (spin up) and negative (spin down) projections.

Action: We added the following to the discussion of the DFT band structure in the Supplementary Information:

”Note that our DFT calculations show that all states between the VBM and CBM of the $CrBr_3$ come from the metallic substrate, which is expected for such a weakly interacting system. Estimates of the electron and hole barrier heights based on our DFT calculations[16–18] indicate that there is no spontaneous charge transfer either from or to the metallic substrate.”

Reviewer Reports on the Third Revision:

Ref # 3

The authors have fully addressed my questions and concerns. I would like to recommend the paper's publication in Nature.

Ref # 4

The authors have addressed my concerns, and I recommend publication.

Author Rebuttals to Third Revision:

N/A