Peer Review File

Manuscript Title: The field-free Josephson diode in a van der Waals heterostructure

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1 (Remarks to the Author):

This paper reports a VDW heterostructure which exhibits Josephson diode behavior. The result is very interesting. The experimental results are solid. However, there are important issues that the authors should address before I can fully judge the value of this paper.

First, the quality of the heterostructure is not carefully examined. It is very important to know the quality of junction. Without this information, it is very difficult to rule out many effects related to disorder or impurities.

Second, the p-breaking is not clear to me. As NbSe2 is quite two dimensional superconductor, the essential physics in the device stems from the two interfaces between NbSe2 and Nb3Br8. The symmetry of the interfaces is not carefully analyzed.

Third, NbSe2 is a multi-band superconductor and is also known to have a CDW order. It will be important to have some measurements about these physics for the junction to determine what is the cause of the diode effect.

Overall, the paper reports an very intriguing observation. I would recommend the paper if the authors can provide detailed measurements to determine the essential origin, or at least, help to understand the experimental results.

Referee #2 (Remarks to the Author):

In this manuscript, Wu and co-authors fabricated Josephson junction built with vdW materials superconductor $NbSe_2$ and semiconductor Nb_3Br_8. They demonstrated the Josephson diode effect at zero magnetic field in the presence of ΔI_c between the positive and negative critical current, and further applied it into a half-wave rectification. The concept of using Josephson diode for rectification devices is not novel enough to warrant the Nature publication. Although the description of the phenomenon seems to be interesting, I found that the explanation of the results is not clear and far from being convincing. Many points need to be addressed and the authors shall provide solid explanations. Below are the questions:

(1) The author ascribes ΔI_c to inversion symmetry breaking of an odd-number interlayer Nb_3Br_8. However, there would be many reasons leading to the asymmetry in I_{c+} and I_{c-}, such as nonequal hetero-interfaces at two $NbSe_2$/Nb_3Br_8 boundaries, especially during the fabrication of vdW heterostructures assisted by the polymer. As far as I can see, it is easy to form asymmetry contact between the $NbSe_2$/Nb_3Br_8/$NbSe_2$ interfaces. And by checking the data in Fig.2a of the paper (Nature Communications 7, 10616 (2016)), we could find that the I_{c-} and I_{c+} at zero field are different. Then a natural question would raise about the novelty of this work.

(2) If the inversion symmetry breaking originates from the odd-number layer structure, then one may expect that the even-number layer devices do not show the ΔI_c behavior. The author should provide such evidence to support their argument, for example, how does ΔI_c change with barrier thickness? As the authors claim, almost all kinds of odd numbers layers of 2D materials servers as the tunneling barrier, then the inversion symmetry would break. So the questions would be whether other odd-number layer vdW materials would give a similar result or not, like graphene or MoS_2.

(3) The Josephson junction seems to have a relatively low critical current density (220A/cm2). so what's the energy gap of the semiconducting Nb_3Br_8? And does it have a magneto-chiral structure? Especially, how do the authors measure the critical current map in figure 4a, what is the magnetic field sweeping direction? What is the typical critical current map of the Josephson device when the magnetic field sweeping back and forth? There're almost no characterizations about the properties of Nb_3Br_8 in this manuscript like the magnetization curves to support that Nb_3Br_8 is non-ferromagnetic or in singlet magnetic ground state, although the authors claim that the properties are similar to Nb_3Cl_8. The author should provide more characterizations of Nb_3Br_8 to support their claims.

(4) The quality of the Fraunhofer pattern they showed in Fig.4a is not high quality, which also seems not so periodic at higher fields. I doubt the interface quality of their device. Besides, there are no parameters such as junction sizes or penetration depths of their superconductors to describe the oscillatory I_c behavior. The author should add some explanations.

(5) Why does the diode effect "turn off" by 35mT or below 2.1μA? The author should make it clear. In Fig.3a, Ic+ seems to go through an anomalous kink at ±35mT but Ic- doesn't. What is the reason?

(6) In figure 4, about the second harmonic signal, there are two peaks in the negative magnetic field while only one peak in the positive magnetic field. What's the reason? Also, what is the behavior of the second harmonic signal at different applied currents?

(7) The author cited Ref.13 to show the differences between their device and bulk superconductor with magnetochiral anisotropy (Also, in a bulk superconductor with magnetochiral anisotropy, ΔIc vs B is sweep direction dependent, flipping sign13……), however, in Ref.13 I haven't find any statements or experimental results about ΔIc vs B.

Referee #3 (Remarks to the Author):

This manuscript reports realization of superconducting diode effect without magnetic field. So far, superconducting nonreciprocal devices are based on magnetochiral anisotropy of the material and inevitably requires magnetic field. However, generally superconducting properties are sensitive to magnetic fields and so applying magnetic field for the diode operation is not preferable in the superconducting circuit applications. Here, the authors used thin layer of Nb3Br8 without inversion symmetry as the tunnel barrier and realized a magnetic-field-free superconducting Josephson diode. They achieved stable half-wave rectification of a square-wave excitation with a very low switching current density, high rectification ratio and high robustness.

The result seems original and quite important. From the viewpoint of the fundamental science, the manuscript opens a new research field of nonreciprocal Josephson effect, and from the application viewpoint, the reported field-free superconducting diode is widely applicable to superconducting electronics. Thus, the results presented are of immediate interest to many people.

The research seems to be done properly, and the data are reliable and clearly presented. Also, the manuscript is clearly written and well organized with sound conclusions. I think the manuscript is worth publishing in Nature.

Author Rebuttals to Initial Comments:

Re: *Nature* submission # 2021-03-05207A-Z, "Realization of the field-free Josephson diode"

Dear referees

We thank the referees for their in-depth examination of our work and their questions/suggestions. Below we respond, point by point, to the referee's questions and we hope that our revised manuscript is found acceptable for publication.

Referee #1 (Remarks to the Author):

This paper reports a VDW heterostructure which exhibits Josephson diode behavior. The result is very interesting. The experimental results are solid. However, there are important issues that the authors should address before I can fully judge the value of this paper.

We thank the referee for the praise of our experimental results, and we hope our replies below can help the referee recommend publication.

First, the quality of the heterostructure is not carefully examined. It is very important to know the quality of junction. Without this information, it is very difficult to rule out many effects related to disorder or impurities.

We thank the referee for this question. To rule out the effects related to extrinsic factors such as disorder or impurities, we fabricated more $NbSe_2/Nb_3Br_8/NbSe_2$ junctions using the same device fabrication techniques described in Methods. For the additional $NbSe_2/Nb_3Br_8/NbSe_2$ junctions, we observed the same field-free Josephson diode (JD) effect in various devices, including devices with different layers such as Device #2 (2-layer Nb_3Br_8) and Device #3 (4-layer Nb_3Br_8) shown in Fig. R1a-b and R1c-d, respectively. In these different devices, it is highly unlikely that the disorder or impurities are identical, yet the field-free JD effect always exists, indicating that the field-free JD effect arises from an intrinsic property of the $NbSe_2/Nb_3Br_8/NbSe_2$ heterostructure instead of an extrinsic effect related to specific disorder or impurities.

Additionally, we also fabricated control junctions ($NbSe_2/NbSe_2$ and $NbSe_2$/few-layer graphene (FLG)/$NbSe_2$ junctions) and both showed the no sign of the field-free JD effect, as shown in Fig. R3 and will be discussed in detail in below. External magnetic field is required in both cases to obtain a ΔI_c, also pointing to the fact that the field-free JD effect is intrinsic to the $NbSe_2/Nb_3Br_8/NbSe_2$ heterostructure and is not arising due to disorder or impurities.

We added the above data and discussion to the Methods and modified the main text with a discussion.

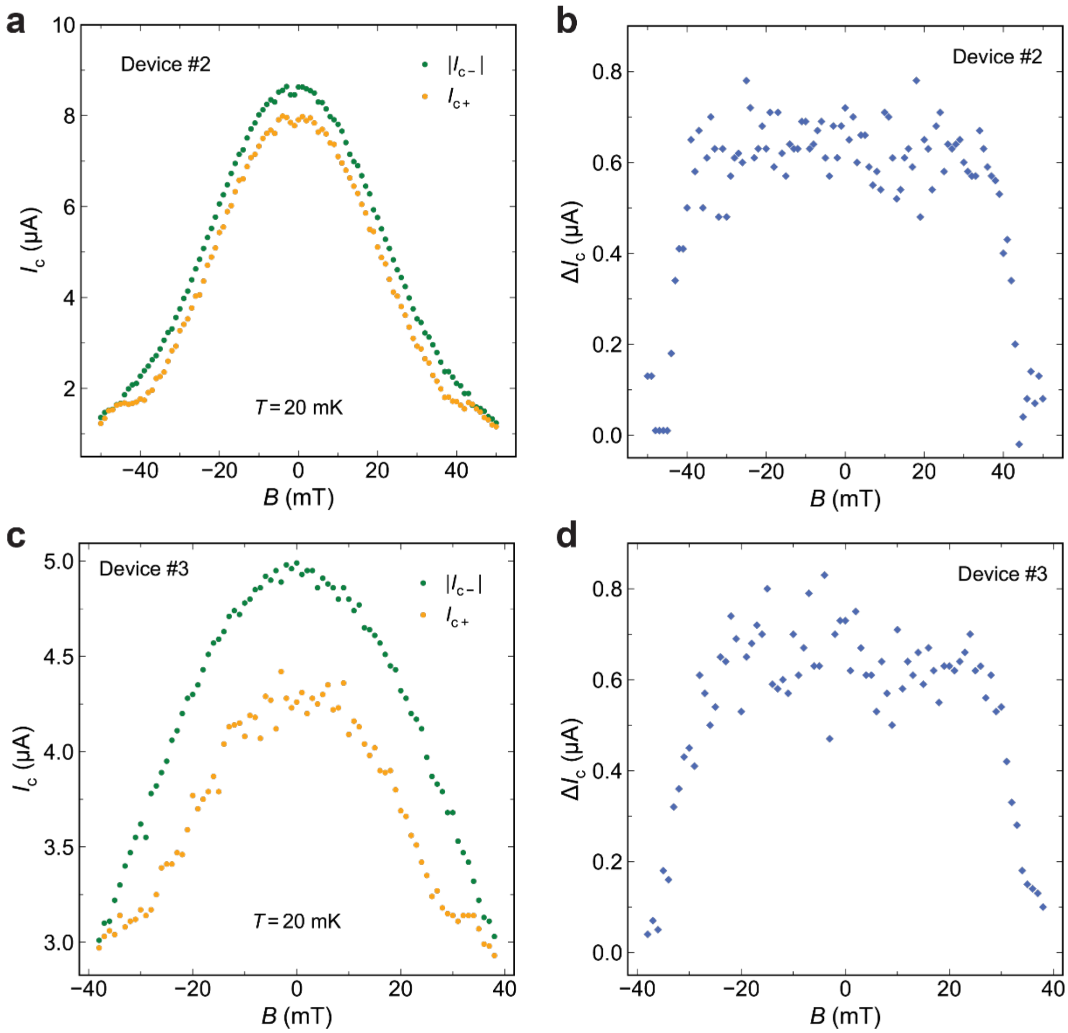

Fig. R1 | **a**, I_{c+} and $|I_{c-}|$ as a function of magnetic field of $NbSe_2$/2-layer Nb_3Br_8/$NbSe_2$ (Device #2). **b**, ΔI_c as a function of magnetic field of Device #2. **c**, I_{c+} and $|I_{c-}|$ as a function of magnetic field of $NbSe_2$/4-layer Nb_3Br_8/$NbSe_2$ (Device #3). **d**, ΔI_c as a function of magnetic field of Device #3.

Second, the p-breaking is not clear to me. As NbSe2 is quite two dimensional superconductor, the essential physics in the device stems from the two interfaces between NbSe2 and Nb3Br8. The symmetry of the interfaces is not carefully analyzed.

We thank the referee for this important question. We agree with the referee that the interface of $NbSe_2$ and Nb_3Br_8 is important and can influence the symmetry of the junction. As we mentioned in the main text, since the top $NbSe_2$ and bottom $NbSe_2$ have been assembled with arbitrary angles relatively to each other in the junction, the top $NbSe_2$/Nb_3Br_8 interface and the bottom Nb_3Br_8/$NbSe_2$ interface are different, which leads to inversion symmetry being broken for the whole vertical junction. In addition, Nb_3Br_8 belongs to space group $R\overline{3}m$, and has inversion centers residing in between the pairs of adjacent layers (we added Extended Data Figure 1(a) to show the crystal structure), meaning that odd-layered Nb_3Br_8 intrinsically breaks inversion symmetry and even-layered Nb_3Br_8 preserves inversion symmetry. For Device #1 (device in main text) which consists of an odd-layered flake of Nb_3Br_8, both the Nb_3Br_8 itself *and* the interface of $NbSe_2$/Nb_3Br_8 contribute to inversion symmetry breaking in the JJ.

Now, as prompted by the referee, we fabricated *even-layered* $NbSe_2/Nb_3Br_8/NbSe_2$ junctions as well, and *also observed* the same field-free JD effect. The results for JJ of a 4-layer Nb_3Br_8 device (Device #3) is shown in Fig. R1 as an example. Also, antisymmetric peaks in the second harmonic signal confirms the inversion symmetry breaking of the junction (Fig. R2). Since the even-layer Nb_3Sb_8 itself preserves inversion, the inversion symmetry breaking of the junction must be induced by the $NbSe_2/Nb_3Br_8$ interfaces. These results indicate that the interfaces of $NbSe_2/Nb_3Br_8/NbSe_2$ junction play the dominant role for inversion symmetry breaking in the junction. We added these important results into the Methods, and also modified the main text of the manuscript with the above discussion.

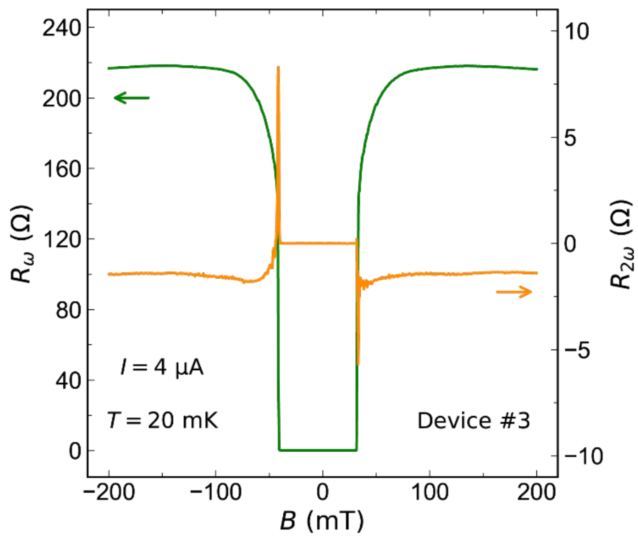

Fig. R2 | Second harmonic resistances ($R_{2\omega}$) of a JJ with 4-layer Nb_3Br_8 (Device #3).

Third, NbSe2 is a multi-band superconductor and is also known to have a CDW order. It will be important to have some measurements about these physics for the junction to determine what is the cause of the diode effect.

We thank the referee for this very good question. To study the influence of $NbSe_2$ and the origin of diode effect, we fabricated more types of junctions with different barriers using the same device fabrication methods as the $NbSe_2/Nb_3Br_8/NbSe_2$ junctions (described in Methods in main text), including creating $NbSe_2/NbSe_2$ junctions and $NbSe_2$/few-layer graphene (FLG)/$NbSe_2$ junctions. In both the $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions, the top and bottom interfaces have arbitrary angles and inversion symmetry is broken just like in the $NbSe_2/Nb_3Br_8/NbSe_2$ junctions.

Figure R3 shows the results of a typical $NbSe_2/NbSe_2$ junction (Device #4); the three steps on the *V-I* curve correspond to the I_c of the junction, the top, and the bottom $NbSe_2$ electrodes, respectively (as labelled in Fig. R3a). To probe the diode effect, we measured the *V-I* curves at various magnetic fields, and extracted the positive and negative critical currents (I_{c+} and $|I_{c-}|$) and plotted them as a function of magnetic field as shown in Fig R3b. A *field-induced* ΔI_c was observed and is shown in Fig R3c; a clear antisymmetric feature under positive and negative magnetic field is visible, and the ΔI_c crosses zero at near-zero magnetic field. A very similar field-dependent, antisymmetric feature of the ΔI_c vs B curve is also observed in our $NbSe_2$/FLG/$NbSe_2$ junction, as shown in Fig R3d. These results illustrate that while a *field-induced* diode effect can arise in these inversion symmetry broken heterostructures of $NbSe_2$, without Nb_3Br_8, a *field-free* JD effect does not. Thus, while $NbSe_2$ is indeed a multiband SC with CDW order, it is not the $NbSe_2$ which

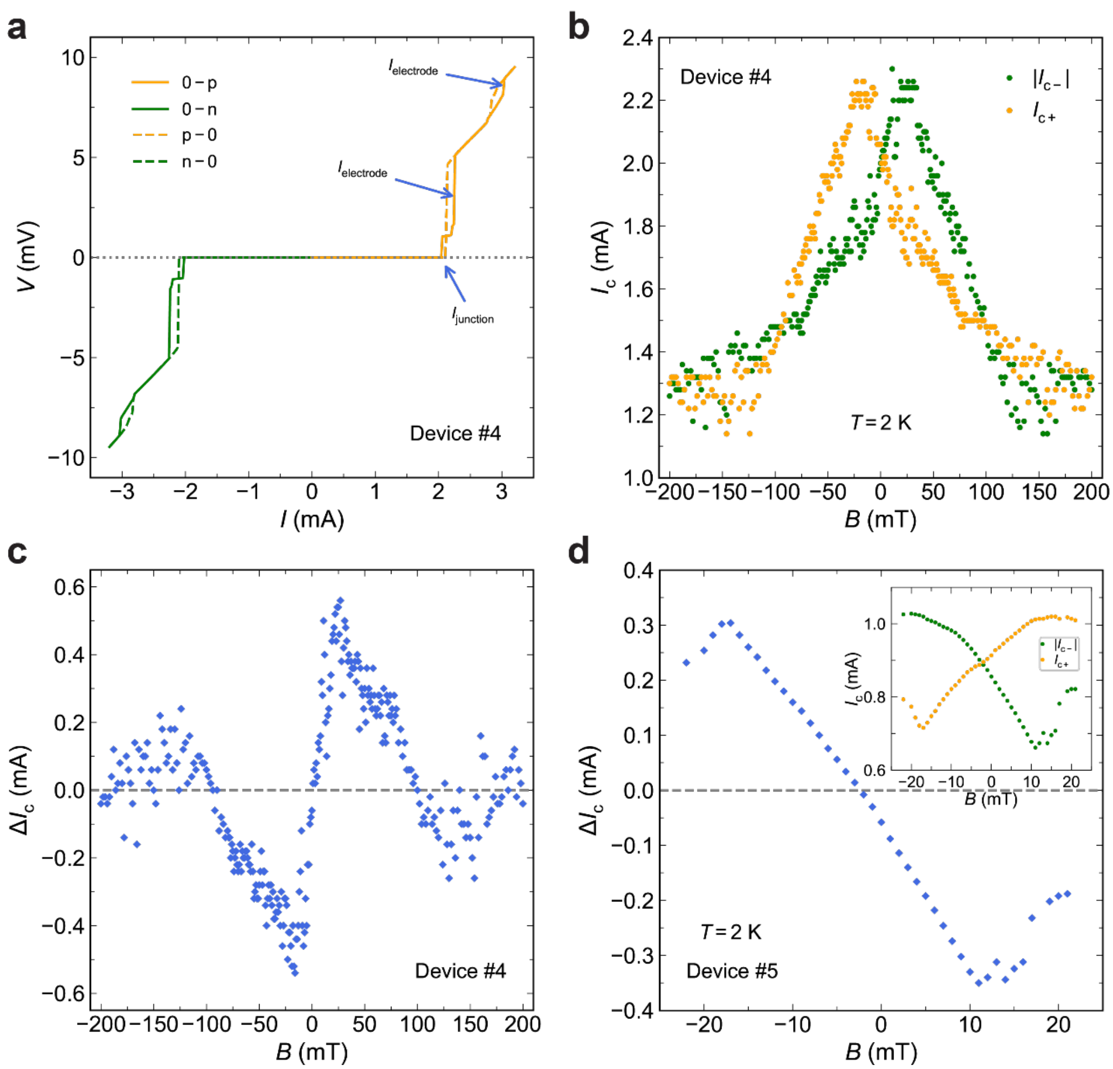

Fig. R3 | **a**, *V-I* curve of $NbSe_2/NbSe_2$ junction (Device #4) measured at 2 K and 0 T. **b**, I_{c+} and $|I_{c-}|$ as a function of magnetic field of Device #4 at 2 K. **c**, ΔI_c as a function of magnetic field of Device 4. **d**, ΔI_c as a function of magnetic field of $NbSe_2$/FLG/$NbSe_2$ junction (Device #5), inset shows the corresponding I_{c+} and $|I_{c-}|$

is giving rise to the *field-free* effect but the heterostructure with the Nb_3Br_8 as the barrier. The main text and Methods were modified to include these new results and conclusions.

Note that as yet, no one has reported the *field-induced* diode effect *either* in $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions and that is also a new result. This shows that there is ample room for exciting future work in this field. Recently, field-induced superconducting diode effects in superconductors were proposed to come from magnetochiral anisotropy, finite momentum superconductivity or nonreciprocal Landau critical momentum (*Sci. Adv.* **3**, e1602390 (2017); arXiv:2106.01909v1 (2021); arXiv:2106.03326v1 (2021)). The effects we observe in the $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions may be explained by these theoretical proposals and that is an area of investigation that is now underway. A clear understanding of the underlying physical origin in these kinds of heterostructures requires more theoretical and experimental investigations and the field is still in its infancy.

As yet, we do not have a full understanding of its fundamental physical origin in our $NbSe_2/Nb_3Br_8/NbSe_2$ junctions. It is a quite new observation and much more theoretical study is required, which is beyond our current expertise. Very recently, a theoretical work (arXiv:2106.10276 (2021)) predicted a new kind of insulator named an "obstructed atomic insulator", which has centers of charge not localized on the atoms, and Nb_3Br_8 is in fact one of these predicted materials. It is unclear whether that property may influence the Josephson tunneling but it is an interesting direction for future theoretical and experimental work as well. We also added this speculation to the main text as well.

Overall, the paper reports an very intriguing observation. I would recommend the paper if the authors can provide detailed measurements to determine the essential origin, or at least, help to understand the experimental results.

We thank again for the referee's comment and praise of our results. With the many new experiments shown above, we can rule out extrinsic factors and the influence of the $NbSe_2$ electrode as the driving forces of the field-free JD effect, and we demonstrate that the inversion symmetry breaking is indeed dominated by the interfaces. It's very difficult to have a full understanding of the physical origin of field-free diode effect at this stage, but we believe we have narrowed the primary suspects significantly and our work will drive a large amount of future work in this regard. We believe this work is a breakthrough for the exploration of fundamental superconducting and Josephson phenomena, and can help the development of superconducting electronics in the future. We hope that with these new experiments and data, our manuscript is found acceptable for publication.

Referee #2 (Remarks to the Author):

In this manuscript, Wu and co-authors fabricated Josephson junction built with vdW materials superconductor NbSe2 and semiconductor Nb3Br8. They demonstrated the Josephson diode effect at zero magnetic field in the presence of ΔIc between the positive and negative critical current, and further applied it into a half-wave rectification. The concept of using Josephson diode for rectification devices is not novel enough to warrant the Nature publication. Although the description of the phenomenon seems to be interesting, I found that the explanation of the results is not clear and far from being convincing. Many points need to be addressed and the authors shall provide solid explanations. Below are the questions:

We thank the referee for these comments and recognizing that our experiments interesting. We clarify, that the main concept of our work, as we explain in the introduction, is the never before seen *field-free* Josephson diode (JD) effect. Previous examples of a superconducting diode effect require the presence of magnetic field. Our JD result shows nonreciprocal behavior without magnetic field which has no *known* mechanism, violates the known Josephson relations, and opens the door for novel physical phenomena through the integration of quantum materials with Josephson junctions (JJs). Based on this field-free JD effect, we further demonstrated superconducting half-wave rectification for the first time with low switching current density, high rectification ratio and highly stable performance. Just a difference of critical current is not sufficient to realize diodic behavior; it also needs a very sharp and stable transition between superconducting and resistive states. Use of the JD for rectification devices is a demonstration of the field-free JD effect and an interesting avenue of future work as well.

(1) The author ascribes ΔIc to inversion symmetry breaking of an odd-number interlayer Nb3Br8. However, there would be many reasons leading to the asymmetry in Ic+ and Ic-, such as nonequal hetero-interfaces at two NbSe2/Nb3Br8 boundaries, especially during the fabrication of vdW heterostructures assisted by the polymer. As far as I can see, it is easy to form asymmetry contact between the NbSe2/Nb3Br8/NbSe2 interfaces. And by checking the data in Fig.2a of the paper (Nature Communications 7, 10616 (2016)), we could find that the Ic- and Ic+ at zero field are different. Then a natural question would raise about the novelty of this work.

We thank the referee for the comment about interfacial symmetry breaking, which we agree with completely, and had included in the main text. We did not ascribe the field-free JD effect to inversion symmetry breaking *only* arising from odd-layered Nb_3Br_8; we also discussed, as the referee agrees, that the different interfaces of $NbSe_2$/Nb_3Br_8 can also contribute to inversion symmetry breaking, since the top and bottom $NbSe_2$ can have an arbitrary angles (from the main text (submitted version) "*There are two reasons for this; first is the intrinsic lack of inversion symmetry of the 3-layer thick Nb_3Br_8, and second is the geometry of the vertical junction where the top and bottom $NbSe_2$ electrodes are misaligned (rotation) with the Nb_3Br_8 layer.*")

Also, to be rigorous, we investigated $NbSe_2$/$NbSe_2$ junctions as well, using the same device fabrication methods described in main text and measured *V-I* curves at different magnetic fields. The extracted I_{c+} and I_{c-} and ΔI_c ($\Delta I_c = |I_{c-}| - I_{c+}$) vs. magnetic field for a typical device are shown in Fig. R4a and Fig. R4b, respectively. The ΔI_c vs B curve shows a clear *antisymmetric* behavior, with ΔI_c crossing zero at near-zero magnetic field. This shows that $NbSe_2$/$NbSe_2$ heterostructures exhibit a *field-induced* diode effect, not *field-free,* which is analogous to the magnetic field induced superconducting diode effect in ref. (*Nature* **584**, 373-376 (2020)). The field-free JD effect, as only observed so far in our $NbSe_2$/Nb_3Br_8/$NbSe_2$ Josephson junction, is clearly demonstrated by the *symmetric* ΔI_c vs B plots shown in Fig. 3 in main text. We have included these results in Methods and modified the main text with the above discussion.

It is important to note that an individual *V-I* curve at nominal "zero" magnetic field is not sufficient to prove a field-free effect; a map of ΔI_c vs B is necessary. Sensitive magnetic measurements always have to account for some degree of trapped magnetism in the coil magnets, making it likely to observe a non-zero ΔI_c at "zero field". This may be the reason for the anomaly in the *V-I* curve of the $NbSe_2/NbSe_2$ junction in ref. (*Nat. Commun.* **7**, 10616 (2016)) which the referee refers to as well as in ref. (*Nature* **584**, 373-376 (2020) Fig. 2c). However the authors of ref. (*Nature* **584**, 373-376 (2020)) appropriately do not emphasize their "zero-field" data as they measure an antisymmetric ΔI_c vs B dependence and correspondingly discuss their field-induced superconducting diode effect. For a field-free diode effect, a symmetric dependence of ΔI_c vs B can demonstrate, as we showed here for the first time.

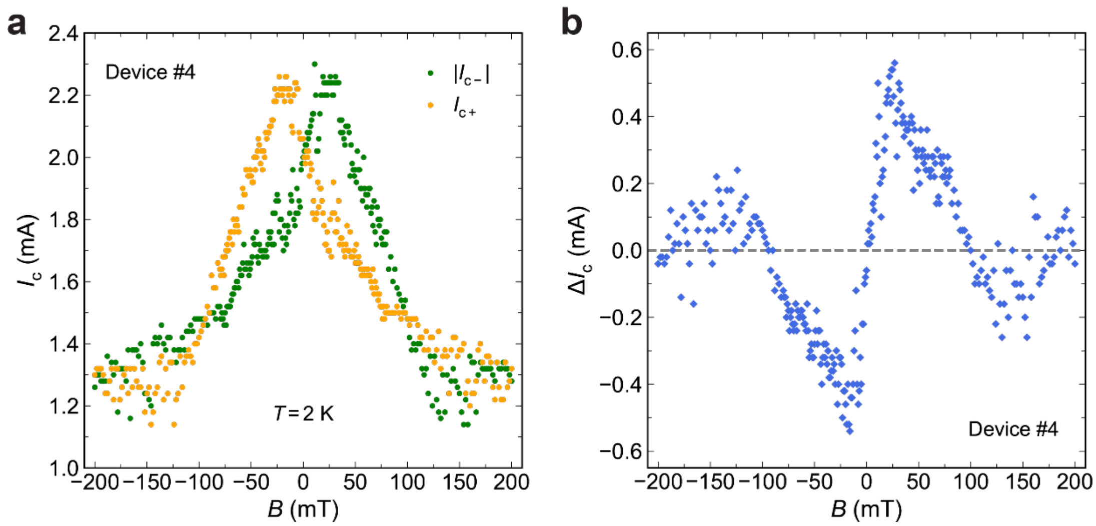

Fig. R4 | **a**, I_{c+} and $|I_{c-}|$ as a function of magnetic field of a typical $NbSe_2/NbSe_2$ junction at 2 K. **b**, ΔI_c as a function of magnetic field of the $NbSe_2/NbSe_2$ junction.

This is the main novelty of this work as it has never been either seen before and does not have a known explanation. Such an abnormal phenomenon in a Josephson junction hints at unexplained Josephson behavior from incorporation of novel, quantum material tunnel barriers. In addition, the superconducting rectification realized at zero field in our work has low switching current density and high rectification ratio and high robustness, all these features are important for their potential application in superconducting electronics. Therefore, we think the novelty and breadth of this work will appeal to the readership of *Nature*.

(2) If the inversion symmetry breaking originates from the odd-number layer structure, then one may expect that the even-number layer devices do not show the ΔIc behavior. The author should provide such evidence to support their argument, for example, how does ΔIc change with barrier thickness? As the authors claim, almost all kinds of odd numbers layers of 2D materials servers as the tunneling barrier, then the inversion symmetry would break. So the questions would be whether other odd-number layer vdW materials would give a similar result or not, like graphene or MoS2.

We thank the referee for this question. As mentioned in the reply to the previous question, we had stated in the manuscript that the inversion symmetry breaking could originate both from the odd-layer Nb_3Br_8 as well as the asymmetric $Nb_3Br_8/NbSe_2$ interfaces. In accordance with the referee's question and to further study the origin of symmetry breaking, we further made new heterostructures of *even-layered* Nb_3Br_8 as well. For clarity, an even number of layers intrinsically preserves inversion symmetry in Nb_3Br_8.

Fig. R5 is the result of a JJ made by a 4-layered Nb_3Br_8 (Device #3), which shows the same symmetric ΔI_c vs B curve as the odd-layered $NbSe_2/Nb_3Br_8/NbSe_2$ junctions of the original manuscript. Also, the second harmonic resistance measurement shows the same antisymmetric behavior. These results unambiguously demonstrate that the interface of $NbSe_2/Nb_3Br_8$ plays a critical role for breaking inversion symmetry and creating the field-free JD effect. We have updated the main text to make this clear and added these results to the Methods.

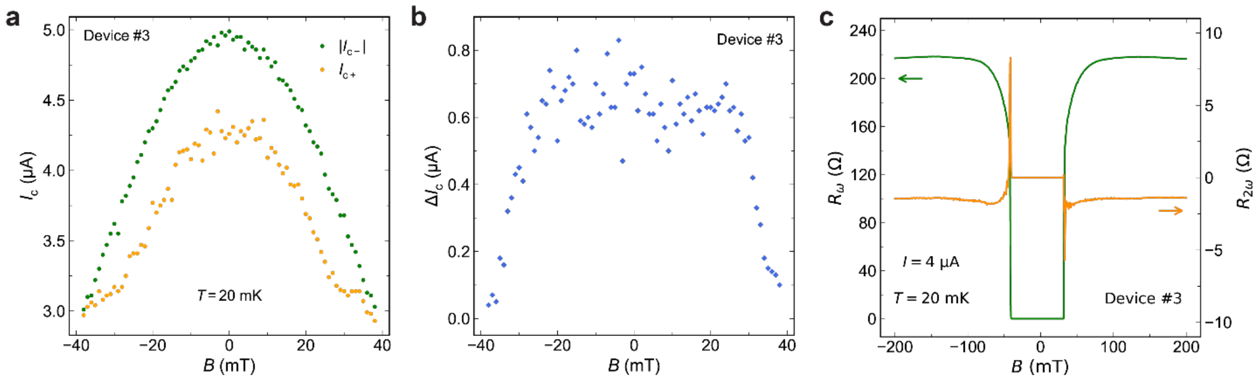

Fig. R5 | a, I_{c+} and $|I_{c-}|$ as a function of magnetic field of $NbSe_2$/4-layer $Nb_3Br_8/NbSe_2$ Josephson junction (Device #3). **b**, ΔI_c as a function of magnetic field of Device #3. **c**, Second harmonic resistances ($R_{2\omega}$) of the Device #3.

Additionally, as the referee suggested, we also fabricated $NbSe_2$/few-layer graphene (FLG)/$NbSe_2$ JJ (Device #5) with the same technique described in main text and measured its *V-I* curves at different magnetic fields. The extracted ΔI_c vs B curve shows again a clear *antisymmetric* pattern that cross zero at near-zero magnetic field (Fig. R6), similar to the behavior in $NbSe_2/NbSe_2$ junctions discussed previously and ref. (*Nature* **584**, 373-376 (2020)), indicating the presence of a *field-induced* diode effect, (which is also not yet reported).

Recently, *field-induced* superconducting diode effects in nonreciprocal superconductors were proposed to come from magnetochiral anisotropy, finite momentum superconductivity or nonreciprocal

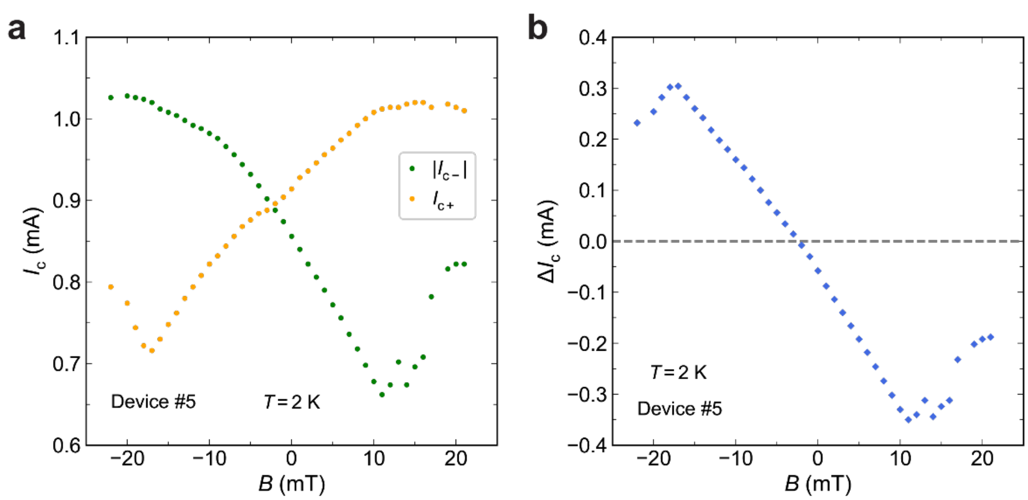

Fig. R6 | a, I_{c+} and $|I_{c-}|$ as a function of magnetic field of $NbSe_2$/FLG/$NbSe_2$ Josephson junction (Device #5). **b**, ΔI_c as a function of magnetic field of Device #5.

Landau critical momentum (*Sci. Adv.* **3**, e1602390 (2017); arXiv:2106.01909v1 (2021); arXiv:2106.03326v1 (2021)). The effects we observe in the $NbSe_2$/$NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions may be explained by these theoretical proposals and that is an area of investigation that is now underway. A clear understanding of the underlying physical origin in these kinds of heterostructures requires more theoretical and experimental investigations and the field is still in its infancy.

As yet, we do not have a full understanding of its fundamental physical origin in our $NbSe_2$/Nb_3Br_8/$NbSe_2$ junctions. It is a quite new observation and much more theoretical study is required, which is beyond our expertise. Very recently, a theoretical work (arXiv:2106.10276 (2021)) predicted a new kind of insulator named an "obstructed atomic insulator", which has a center of charge not localized on the atoms, and Nb_3Br_8 is in fact one of these predicted materials. It is unclear whether that property may influence the Josephson tunneling but it is an interesting direction for future theoretical and experimental work as well. We also added this speculation to the main text as well.

We completely agree with the referee's good suggestions that it's very valuable to study Josephson junctions with different barrier materials, and it is an important direction for future research based on our findings. This will help to understand the influence of the barrier and its properties on the Josephson effects, like the novel field-free effect in the $NbSe_2$/Nb_3Br_8/$NbSe_2$ junction, and also explore atypical superconducting properties with quantum material barriers.

(3) The Josephson junction seems to have a relatively low critical current density (220A/cm2). so what's the energy gap of the semiconducting Nb3Br8? And does it have a magneto-chiral structure? Especially, how do the authors measure the critical current map in figure 4a, what is the magnetic field sweeping direction? What is the typical critical current map of the Josephson device when the magnetic field sweeping back and forth? There're almost no characterizations about the properties of Nb3Br8 in this manuscript like the magnetization curves to support that Nb3Br8 is non-ferromagnetic or in singlet magnetic ground state, although the authors claim that the properties are similar to Nb3Cl8. The author should provide more characterizations of Nb3Br8 to support their claims.

We thank the referee for these questions. Since there are many questions wrapped together here, we will address them one at a time.

First, the band gap as measured via optical absorption in ref. (*ACS Nano* **13**, 9457-9463 (2019)), is ~1.75 eV. This reference was included as ref. 27 in main text.

Second, there is no evidence for Nb_3Br_8 having magneto-chiral effect. Regarding to magnetochiral anisotropy, there are two types of mangetochiral anisotropy resistances. One is described with the form: $R = R_0[1+\gamma(\boldsymbol{P}\times\boldsymbol{B})\cdot\boldsymbol{I}]$, where $\boldsymbol{P}$ is the vector to represent the direction and magnitude of the inversion symmetry breaking, $\boldsymbol{B}$ is the magnetic field and $\boldsymbol{I}$ is the current. The other is described by the formular: $R = R_0[1+\gamma(\boldsymbol{B}\cdot\boldsymbol{I})]$, which is also known as electrical magentochiral anisotropy or the electrical magnetochiral effect as explained by Nagaosa et al. in ref. (*Nat. Commun.* **9**, 3740 (2018)). In our device geometry, when considering the direction of these three components, $\boldsymbol{P}$ is out-of-plane, $\boldsymbol{B}$ is in-plane, and $\boldsymbol{I}$ is out-of-plane. Therefore, both types of magnetochiral anisotropy effects should have no contribution to our measurement. Moreover, the mangetochiral anisotropy can only lead to a field-induced superconducting diode effect, however, we observed the field-free JD effect, which cannot originate from mangetochiral anisotropy.

Third, the critical current map in Fig 4a in manuscript was obtained by measuring the differential resistance (dV/dI) vs I curve under different magnetic fields using a lock-in amplifier with a DC bias; we added this description in Methods. The Fraunhofer pattern in Fig. 4a is measured sweeping the magnetic

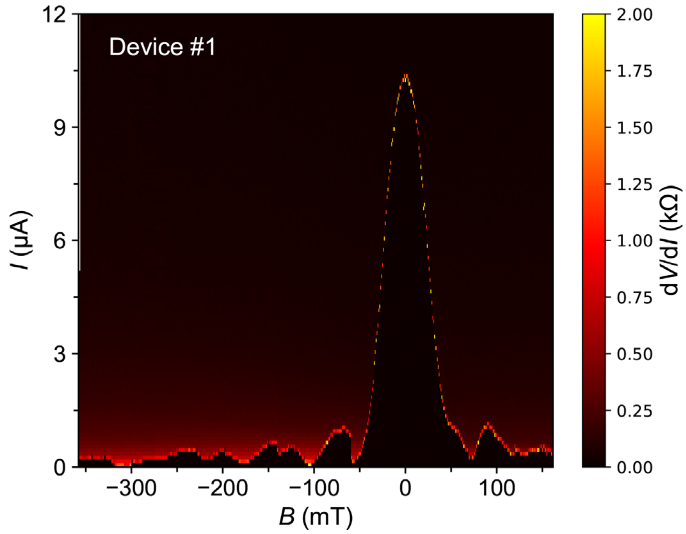

Fig. R7 | Fraunhofer pattern of Device #1 with sweep-down magnetic field

field from negative to positive ("sweep-up"). Fig R7 shows the Fraunhofer pattern measured with sweep-down, which is similar with the sweep-up one. This has been added to the Methods.

Fourth, regarding the magnetic behavior of Nb_3Br_8, in ref. (*ACS Nano* **13**, 9457-9463 (2019)) they measure the magnetization vs. temperature of an Nb_3Br_8 single crystal, and found it to have a singlet magnetic ground state.

Also, we also performed several transport measurements to look for signs of magnetization in Nb_3Br_8 thin flakes. First, we measured the magnetoresistance, with both out of plane (OOP) and in plane (IP) magnetic field, of Device #1 (from the main text) above the T_c of $NbSe_2$ as shown in Fig. R8a and R8b. The magnetoresistances are completely overlapped regardless of field sweep direction, indicating that there is no ferromagnetic texture in Nb_3Br_8 thin flakes.

In addition, we also utilized FLG (few-layer graphene) as electrodes and fabricated FLG/Nb_3Br_8/FLG heterostructures (Device #6) to measure the transport behaviors of Nb_3Br_8 thin flakes. The magnetoresistance, with different magnetic field sweeping direction at different temperatures are shown in Fig. R8c and R8d. Again the curves are completely overlapped, showing that there no hysteresis in the magnetoresistances with sweep up and down curves. Therefore, Nb_3Br_8 thin flakes appear to be non-ferromagnetic, which is consistent with the reported singlet magnetic ground state at low temperature in ref. (*ACS Nano* **13**, 9457-9463 (2019)). We have added the above discussion to the Methods.

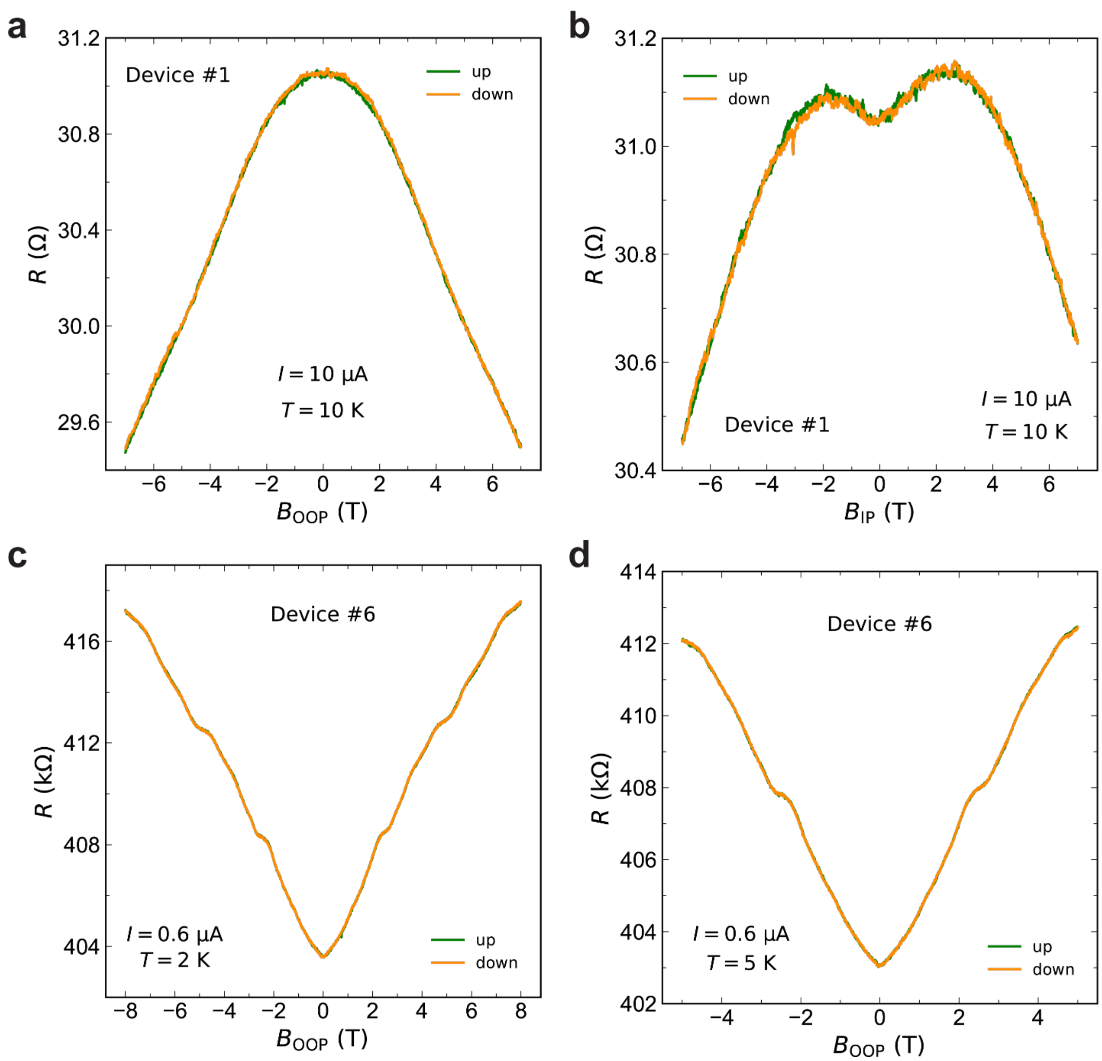

Fig. R8 | a, magnetoresistances of Device #1 (device in manuscript) with out-of-plane magnetic field at 10 K (above T_c of $NbSe_2$). **b**, magnetoresistances of Device #1 with in-plane magnetic field at 10 K. **c**, magnetoresistance of FLG/Nb_3Br_8/FLG with out-of-plane magnetic field at 2 K. **d**, magnetoresistance of FLG/Nb_3Br_8/FLG with out-of-plane magnetic field at 5 K.

(4) The quality of the Fraunhofer pattern they showed in Fig.4a is not high quality, which also seems not so periodic at higher fields. I doubt the interface quality of their device. Besides, there are no parameters such as junction sizes or penetration depths of their superconductors to describe the oscillatory Ic behavior. The author should add some explanations.

We thank the referee for this question. While the sweep-down Fraunhofer pattern, shown in Fig. R7, has a better periodicity, the Fraunhofer patterns are indeed not so periodic at higher field. However this is a common case as seen in many other 2-dimensional tunnel Josephson junctions, including the $NbSe_2$/$NbSe_2$ junctions the referee mentioned earlier (*Nat. Commun.* **7**, 10616 (2016)) as well as graphene vertical JJs (*Nano Lett.* **17**, 6125-6130(2017)). In these vertical junctions with very thin barriers, the Fraunhofer pattern can be influenced by many effects, such as magnetic vortices in the superconducting electrodes and the self-field effect (*Nano Lett.* **17**, 6125-6130(2017)), which can make it difficult to get an ideal, highly periodic Fraunhofer pattern.

In addition, from the theory, the ideal Fraunhofer pattern requires a uniform current profile (rectangular junction area shape and magnetic field parallel with either side of the rectangle). As studied in Ref. (*J. Appl. Phys.* **103**, 07C707 (2008)), the Fraunhofer pattern is not so periodic in junctions with, for example, a triangular shape like Device #1 in main text (see Fig. 2a and 2b in ref. (*J. Appl. Phys.* **103**, 07C707 (2008))). The junction shape and magnetic field direction can lead to a non-ideal Fraunhofer pattern, even in a high quality junction with high quality interfaces. Importantly, we observed the field-free JD effect in *many devices* indicating the quality of our interfaces is not an issue.

According to the referee's suggestion, we characterized more junction parameters by analyzing the Fraunhofer pattern in Fig. R7 at low magnetic field. In a JJ, the London penetration depth (λ) of $NbSe_2$ can be obtained from $\phi_0 = \Delta BW(d + 2\lambda)$, where ϕ_0 is the magnetic flux quantum, ΔB is the period of the oscillation in the Fraunhofer, W is the width of the junction perpendicular to the field direction, and d is the barrier thickness. From the Fraunhofer pattern, the oscillation period is ~59 mT and with $W \approx 3.5$ μm, and $d \approx 2.3\ nm$, λ is calculated to be 3.85nm. We added this information to the Methods.

(5) Why does the diode effect "turn off" by 35mT or below 2.1μA? The author should make it clear. In Fig.3a, Ic+ seems to go through an anomalous kink at ±35mT but Ic- doesn't. What is the reason?

We thank the referee for this question. However we don't know the exact reason why the JD effect turns off there, or rather turns on below it where I_{c+} and I_{c-} diverge. This field-free JD effect violates the known Josephson junction theories, and theoretical understanding of its origin is a fertile of future work that is, unfortunately, beyond our current expertise. As speculated in the manuscript, the JD effect may arise from asymmetric tunneling with positive and negative currents, which can be further modulated by magnetic flux from an external magnetic field.

(6) In figure 4, about the second harmonic signal, there are two peaks in the negative magnetic field while only one peak in the positive magnetic field. What's the reason? Also, what is the behavior of the second harmonic signal at different applied currents?

We thank the referee for this question. In the second harmonic resistance ($R_{2\omega}$) measurements of Device #1, the $R_{2\omega}$ peaks occur corresponding with the superconducting transition. Since the superconductivity of a JJ can be modulated by the Josephson coupling through magnetic flux and vortex formation in the electrodes, it's common to have multiple superconducting transitions which do not have to be identical for positive and negative field. In Fig R9a, $R_{2\omega}$ at an applied current of 3 μA of Device #1 is shown; with small applied currents the superconducting state can be broken with magnetic field smaller than the upper critical field (H_c), and $R_{2\omega}$ peaks occur corresponding with these transitions. However, the two antisymmetric $R_{2\omega}$

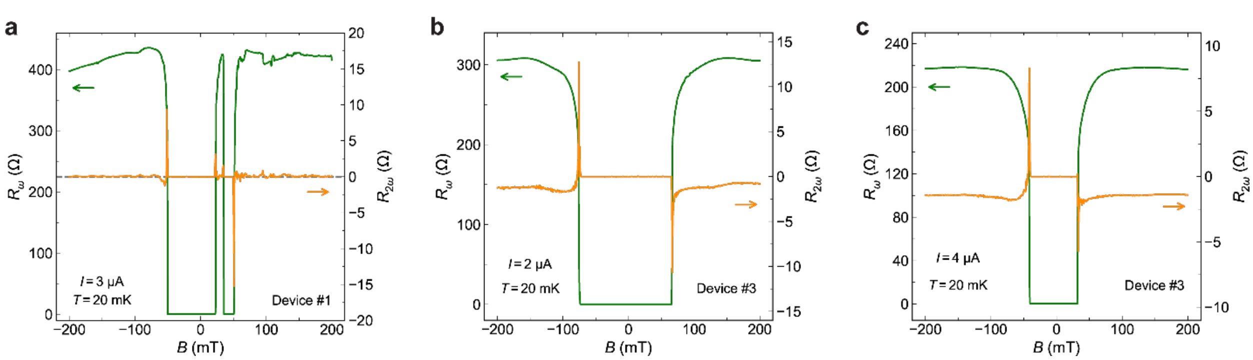

Fig. R9 | a, Second harmonic resistances ($R_{2\omega}$) of Device #1 with an applied current of 3 μA. **b**, $R_{2\omega}$ of $NbSe_2$/4L-Nb_3Br_8/$NbSe_2$ JJ with an applied current of 2μA. **c**, $R_{2\omega}$ of $NbSe_2$/4L-Nb_3Br_8/$NbSe_2$ JJ (Device #3) with an applied current of 4μA.

peaks are still clearly observed corresponding with the superconducting transitions at H_c, which demonstrate the expected inversion symmetry breaking of the $NbSe_2/Nb_3Br_8/NbSe_2$ Josephson junction. In addition, in Device #3 with 4-layer Nb_3Br_8 as the tunnel barrier, antisymmetric $R_{2\omega}$ peaks can also be clearly observed as shown in Fig R9b and R9c for different applied currents as well. It's worth mentioning that the antisymmetric $R_{2\omega}$ peaks in these devices refer to the inversion symmetry breaking of the whole junction, in this case dominated by the interfaces. We have included the new $R_{2\omega}$ results for Device #3 in Methods and added a discussion of the even-layered devices in the main text.

(7) The author cited Ref.13 to show the differences between their device and bulk superconductor with magnetochiral anisotropy (Also, in a bulk superconductor with magnetochiral anisotropy, ΔIc vs B is sweep direction dependent, flipping sign13……), however, in Ref.13 I haven't find any statements or experimental results about ΔIc vs B.

We thank the referee for pointing out this typo. We accidentally cited a wrong reference here; it should be *Nature* **584**, 373-376 (2020) (ref. 8 in manuscript). In this reference, their ΔI_c vs B is antisymmetric, similar with the results in our $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ devices shown in Fig R4 and R6. This is completely different with our *field-free* JD result, in which ΔI_c is symmetric with magnetic field and remains nonzero at zero field. We have rectified this mistake in the manuscript.

In summary, the field-free JD effect is a new discovery in Josephson junctions, and is completely different from the field-induced superconducting diode effect which was observed in ref. (*Nature* **584**, 373-376 (2020)), and our $NbSe_2/NbSe_2$, $NbSe_2$/FLG/$NbSe_2$ junctions. Since the underlying fundamental mechanism of this exotic phenomenon is still unclear at this stage, it requires more theoretical and experimental work to fully understand. We hope that with these new experiments and data, our manuscript is found acceptable for publication.

Referee #3 (Remarks to the Author):

This manuscript reports realization of superconducting diode effect without magnetic field. So far, superconducting nonreciprocal devices are based on magnetochiral anisotropy of the material and inevitably requires magnetic field. However, generally superconducting properties are sensitive to magnetic fields and so applying magnetic field for the diode operation is not preferable in the superconducting circuit applications. Here, the authors used thin layer of Nb3Br8 without inversion symmetry as the tunnel barrier and realized a magnetic-field-free superconducting Josephson diode. They achieved stable half-wave rectification of a square-wave excitation with a very low switching current density, high rectification ratio and high robustness.

The result seems original and quite important. From the viewpoint of the fundamental science, the manuscript opens a new research field of nonreciprocal Josephson effect, and from the application viewpoint, the reported field-free superconducting diode is widely applicable to superconducting electronics. Thus, the results presented are of immediate interest to many people.

The research seems to be done properly, and the data are reliable and clearly presented. Also, the manuscript is clearly written and well organized with sound conclusions. I think the manuscript is worth publishing in Nature.

We thank the referee for the highly praise of our work and recommendation for publication.

Reviewer Reports on the Second Revision:

Referee #1 (Remarks to the Author):

The paper has been improved. I do think that the paper can be published.

Referee #2 (Remarks to the Author):

I would like to thank the authors for their answers to my questions. They have performed some new experiments. Parts of the doubts have been addressed in their rebuttal with additional experiments mainly on the NbSe2/even-Nb3Br8/NbSe2, NbSe2/NbSe2, and NbSe2/FLG/NbSe2 Josephson junctions (JJs), however, there are still some issues unsettled which I believe may not be conducive to the significance of this work. Below are my concerns:

(1) I understand the novelty of a field-free diode JJ from their interpretations and admire the half-wave rectification results and durability test as shown in Fig.2. But, to the standard of publication in Nature, coherent discussions or theoretical studies on the regime are needed for the in-depth research, which is inadequate or even unclear in this manuscript and also admitted by the authors themselves. Some answers are not comprehensive or satisfactory like the reason for 'turn off' of the diode effect in question 5.

(2) I don't exactly understand the role of using Nb3Br8 as the interlayer material, as it is an 'unusual' semiconductor with a singlet magnetic ground state, so what are the differences compared with the known vdW semiconductors or insulators like MoS2 or hBN to this structure? Also from their even-layer Nb3Br8-JJ data, the Josephson diode effect is more likely to come from the interfaces than the symmetry of the barrier itself. I wonder this point would inevitably harm their work's uniqueness.

(3) The quality of the data they provided is not good enough for Nature as I'm concerned. For instance, the several 'broken points' of Ic in Fig.4 and Fig.R7. I fully understand the difficulty in realizing an ideal Fraunhofer pattern especially with 2D films or vdW materials, but I could see that the quality of Fraunhofer patterns in the previous NbSe2/NbSe¬¬2 JJ paper (ref. 1) is better than theirs, published in Nature Communications.

(4) 'Field-induced' Josephson diode effect evidenced by NbSe2/NbSe2 and NbSe2/FLG/NbSe2 JJs results is probably attributed to the self-field effects from large Ic densities, leading to a skewed shape of Fraunhofer pattern. For the field modulation of Nb3Br8-JJ seems to be symmetric, I think that the authors should provide a controlled experiment excluding this factor as stronger proofs, in

which the Fraunhofer pattern is more symmetric and the Ic is also reduced to a similar order as that of Nb3Br8-JJ.

(5) The London penetration depth λ the authors obtained is about 4 nm, while in former reports λ for NbSe2 is around 100-300 nm (refs. 2-4). I think the authors should give some explanations.

Refs.

1. Yabuki, N. et al. Supercurrent in van der Waals Josephson junction. Nat. Commun. 7, 10616 (2016).

2. de Trey, P. & Gygax, S. Anisotropy of the Ginzburg-Landau Parameter κ in NbSe2. J Low Temp Phys 11, 421–434 (1973).

3. Le, L. P. et al. Magnetic penetration depth in layered compound NbSe2 measured by muon spin relaxation. Phys. C 185–189, 2715–2716 (1991).

4. Fletcher, J. D. et al. Penetration Depth Study of Superconducting Gap Structure of 2H − NbSe2. Phys. Rev. Lett. 98, 057003 (2007).

Referee #3 (Remarks to the Author):

The paper is clearly written and the response to the referee comments is satisfactory. I think the paper is now ready for publication.

Author Rebuttals to Second Revision:

Re: *Nature* submission # 2021-03-05207B, "Realization of the field-free Josephson diode"

Dear referees

We thank the referees for their in-depth examination of our work and their questions/suggestions. Below we respond, point by point, to the referee's questions and we hope that our revised manuscript is found acceptable for publication.

Referee #1 (Remarks to the Author):

The paper has been improved. I do think that the paper can be published.

We thank the referee for the recommendation for publication.

Referee #2 (Remarks to the Author):

I would like to thank the authors for their answers to my questions. They have performed some new experiments. Parts of the doubts have been addressed in their rebuttal with additional experiments mainly on the NbSe2/even-Nb3Br8/NbSe2, NbSe2/NbSe2, and NbSe2/FLG/NbSe2 Josephson junctions (JJs), however, there are still some issues unsettled which I believe may not be conducive to the significance of this work. Below are my concerns:

(1) I understand the novelty of a field-free diode JJ from their interpretations and admire the half-wave rectification results and durability test as shown in Fig .2. But, to the standard of publication in Nature, coherent discussions or theoretical studies on the regime are needed for the in-depth research, which is inadequate or even unclear in this manuscript and also admitted by the authors themselves. Some answers are not comprehensive or satisfactory like the reason for 'turn off' of the diode effect in question 5.

We thank the referee for the comment. We appreciate the referee's understanding of the novelty of the field-free Josephson diode effect, as well as the meaning of the rectification results.

For the theoretical studies, although the field-free diode effect is not fully understood at this time, we have discussed the possible origin based on many experiments, analysis and contemporary models. As detailed in the manuscript, according to the resistively capacitance shunted junction (RCSJ) model of Josephson junctions and other recent nonreciprocal transport theory work, the origin of a difference in I_{c+} and I_{c-} may arise from: different Josephson currents induced by asymmetric Josephson tunnelling, Joule heating, capacitance, or fluctuation noise current. We went through a systematic elimination of the latter three effects.

In the 1st reply to the referee, we performed additional control experiments requested by the referee, including $NbSe_2$/even-layer Nb_3Br_8/$NbSe_2$, $NbSe_2$/$NbSe_2$ and $NbSe_2$/few-layer graphene (FLG)/$NbSe_2$, and magnetoresistance characterization of Nb_3Br_8 thin flakes, and we can confirm that the asymmetric $Nb_3Br_8/NbSe_2$ interfaces along with the intrinsic property of Nb_3Br_8 play an essential role for the asymmetric tunnelling, and are driving the field-free Josephson diode effect.

Previous theoretical work with semiconductors proposed that $\mathcal{P}$-breaking systems with *polarization* can induce asymmetric Landau-Zener tunnelling, which can lead to the nonreciprocal transport behavior (*Commun. Phys.* **3**, 63 (2020)). While another theoretical work (*Phys. Rev. Lett.* **99**, 067004 (2007)) proposed that Josephson junctions formed by mating p- and n-doped superconductors, could form a self-organized Mott insulator depletion region with *polarization*, again leading to the nonreciprocal transport behavior. In our situation of $NbSe_2/Nb_3Br_8/NbSe_2$ Josephson junctions, Nb_3Br_8 is predicted as an obstructed atomic insulator (OAI), with Wannier charge centers symmetrically pinned at the unoccupied inversion centers in between two Br-Nb-Br layers (arXiv: 2106.10276 (2021)) which was theorized to result in a net *polarization* if inversion symmetry is broken (arXiv: 2111.02433v1 (2021)). As requested by the referee, we enlisted the help of theoretical collaborators, we have performed calculation of the charge distribution in Nb_3Br_8, with results shown in Fig. R1, showing the charge center pinning at the unoccupied inversion centers. These charge centers combined with the asymmetric interfaces may result in a polarization along the z-axis of our junction, and our observed nonreciprocal behavior, and is an important direction of future work.

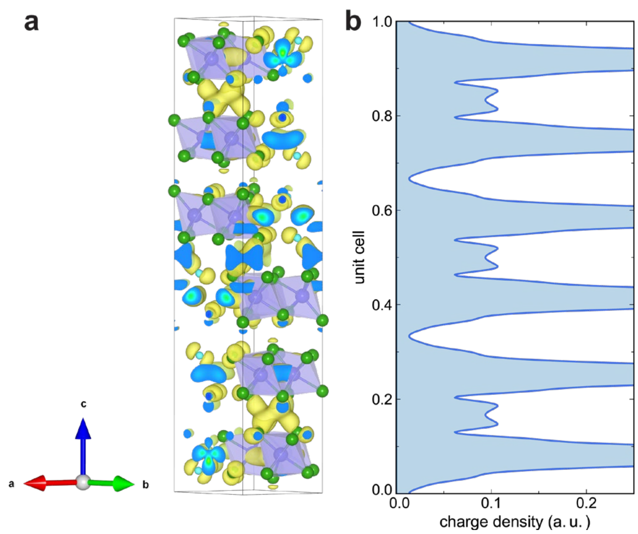

Fig. R1 | **a**, charge density distribution in a Nb_3Br_8 unit cell, the yellow/blue lobes indicate the charge density. **b**, charge density distribution as a function of unit cell location. Note the charge density does not drop to near 0 in every other van der Waals gap.

We have modified the main text to improve the clarity of the above discussion, and added the theoretical calculation of charge centers and associated discussion in Methods.

For the "turn off" of the diode effect, since the applied magnetic field can modulate the Josephson coupling of the junctions, and the diode effect disappears when the critical current becomes small in all the devices, we speculate that the magnetic field effect on the Josephson coupling may cause the shut off of the diode effect. We expect that many experimental and theoretical investigations in future works such as in junctions of many different barriers and superconducting electrodes can help clarify the picture, but that is beyond the scope of this work, whose focus is the first observation of the field-free Josephson diode effect.

For new discoveries, it is extremely difficult to be fully explained in a single paper and typically requires effort from the community which will drive a lot of future work. For example, the mechanism of *field-induced* superconducting diode effect discovered in $[Nb/V/Ta]_n$ superlattices, published in Nature (*Nature* **584**, 373 (2020)), is *also* still not clear. The authors also admitted that "…*Our theoretical understanding of the nonreciprocal charge transport and critical current is as yet far from complete*". Since then, there are several theoretical papers (arXiv: 2106.01909v2 (2021), arXiv: 2106.03326 (2021)) trying to explain the field-induced superconducting diode effect, and different explanations have been proposed but the field is far from complete.

In this work, the breakthroughs of the field-free Josephson diode effect and realization of robust rectification at zero field are novel and open the door for exploring new exotic properties based on different barriers and interfaces in Josephson junction structures, which have been overlooked in previous research. Although the field-free diode effect is not fully explained here, the different control experiments and analysis in this work have narrowed down the possibilities of field-free Josephson diode effect and will help direct future research in fully elucidating the mechanisms.

We hope with this further discussion the referee finds publication in Nature acceptable.

(2) I don't exactly understand the role of using Nb3Br8 as the interlayer material, as it is an 'unusual' semiconductor with a singlet magnetic ground state, so what are the differences compared with the known vdW semiconductors or insulators like MoS2 or hBN to this structure? Also from their even-layer Nb3Br8-JJ data, the Josephson diode effect is more likely to come from the interfaces than the symmetry of the barrier itself. I wonder this point would inevitably harm their work's uniqueness.

We thank the referee for the comment. As we mentioned in the previous manuscript and in the 1st reply to referees, Nb_3Br_8 has been predicted to be an obstructed atomic insulator (OAI), and has Wannier charge centers symmetrically pinned at the unoccupied (by atom) inversion centers in between two Br-Nb-Br layers (arXiv: 2106.10276 (2021)) and is expected to host a polarization when breaking inversion symmetry. Enlisting the help of theoretical collaborators, we have performed calculation of the charge distribution in Nb_3Br_8, with results shown in Fig. R1, showing the charge center pinning at those inversion centers. We added the theoretical calculation of obstructed atomic insulator and charge centers in Nb_3Br_8 and discussion of possible mechanism of field-free Josephson diode effect in Methods.

Some other vdW barriers like h-BN, is trivial insulator and not in the OAI phase, while others, like MoS_2, is also OAIs, but with charge centers localized at the Wyckoff positions that are coplanar with the MoS_2 layer (arXiv: 2111.02433v1 (2021)), differing from Nb_3Br_8. Future work on more OAIs of differing types is a strongly suggested direction of investigation, which we mention in the revised manuscript. Also, we agree, and explain in the manuscript, that the field-free Josephson diode effect is observed in both even and odd-layered Nb_3Br_8-based Josephson junction, indicating that the asymmetric top and bottom $Nb_3Br_8/NbSe_2$ interfaces play an important role for the origin of this effect. However, only asymmetric interfaces are not enough for exhibiting field-free Josephson diode effects, as is demonstrated by the control experiments we performed for the 1st reply in $NbSe_2$/few-layer graphene/$NbSe_2$ and $NbSe_2/NbSe_2$ junctions, which also have interface induced symmetry breaking. Taken together these studies show that the origin of field-free diode effect has a close relation to both the barrier property as well as the asymmetric interfaces.

Our work's uniqueness is not harmed by these facts. This is the *first demonstration of the field-free Josephson diode effect ever; regardless of material type or interface type*. We believe that the field-free Josephson diode effect will be seen in many more heterostructures with other barrier materials and superconductors, now that it has been demonstrated in this work. And through material optimization, the Josephson diode will have a significant impact on superconducting electronics and future computing technologies.

(3) The quality of the data they provided is not good enough for Nature as I'm concerned. For instance, the several 'broken points' of Ic in Fig.4 and Fig.R7. I fully understand the difficulty in realizing an ideal Fraunhofer pattern especially with 2D films or vdW materials, but I could see that the quality of Fraunhofer patterns in the previous NbSe2/NbSe¬¬2 JJ paper (ref. 1) is better than theirs, published in Nature Communications.

We thank the referee for the comment. Indeed Fraunhofer patterns are difficult and time consuming to measure, but we spent the time to address the referee's concern and the result of a new $NbSe_2/Nb_3Br_8/NbSe_2$ Josephson junction device (Device #2) is presented in Fig. R2, showing a high quality Fraunhofer pattern compared with the previous Fraunhofer pattern. We also observed the field-free Josephson diode effect in this device, characterized in Fig. R3, and it is completely consistent with the results in the main text. The results of this device are also included in Methods.

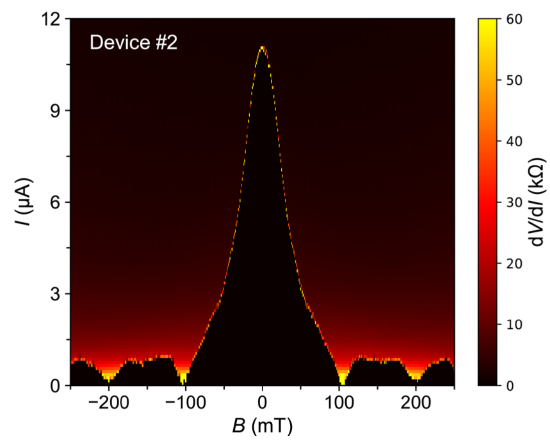

Fig. R2 | Fraunhofer pattern of a $NbSe_2/Nb_3Br_8/NbSe_2$ Josephson junction (Device #2).

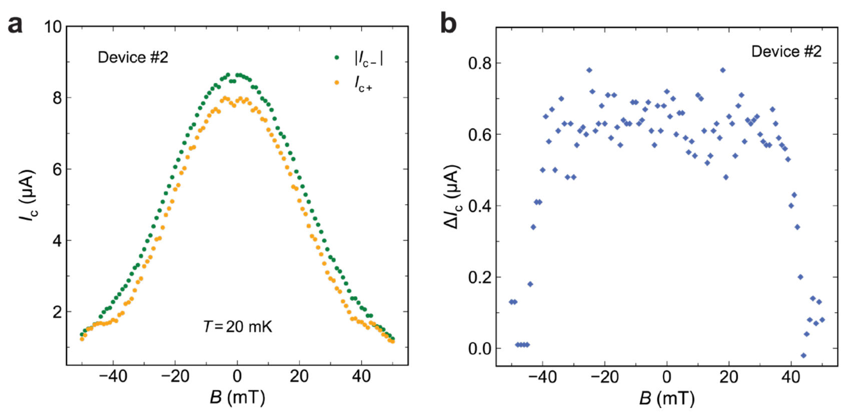

Fig. R3 | **a**, I_{c+} and $|I_{c-}|$ as a function of magnetic field of $NbSe_2/Nb_3Br_8/NbSe_2$ (Device #2). **b**, ΔI_c as a function of magnetic field of Device #2.

(4) 'Field-induced' Josephson diode effect evidenced by NbSe2/NbSe2 and NbSe2/FLG/NbSe2 JJs results is probably attributed to the self-field effects from large Ic densities, leading to a skewed shape of Fraunhofer pattern. For the field modulation of Nb3Br8-JJ seems to be symmetric, I think that the authors should provide a controlled experiment excluding this factor as stronger proofs, in which the Fraunhofer pattern is more symmetric and the Ic is also reduced to a similar order as that of Nb3Br8-JJ.

We thank the referee for the comment and suggestion. It is important to note that no field-free Josephson diode effect was seen in the $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions. Whether the *field-induced* diode effect $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions is actually from a self-field effect due to large critical current density or not, has no bearing on the origin of the *field-free* effect in $NbSe_2/Nb_3Br_8/NbSe_2$.

As the referee requested we fabricated a new $NbSe_2/NbSe_2$ control device (Device #7) with low critical current (I_c ~ 120 μA) and a correspondingly lower critical current density (J_c ~ 18.5 μA/μm^2), which is closer to that in the $NbSe_2/Nb_3Br_8/NbSe_2$ junction (Device #3, J_c ~3.5 μA/μm^2) and lower than the previous control $NbSe_2/NbSe_2$ device (Device #4, J_c ~ 66 μA/μm^2). Fig. R4 shows the I_c and corresponding ΔI_c as a function of applied magnetic field. The I_{c+} and $|I_{c-}|$ vs B have a mirror symmetry with each other, and the obvious antisymmetric feature of ΔI_c vs B curve confirms the presence of the field-induced superconducting diode effect in Device #7, similar to the results of the earlier measured Device #4.

Previous studies on planar Josephson junctions Nb/WTe_2/Nb (*Nano Lett.* **20**, 4228 (2020)) or the InAs 2DEG system (*Nat Nanotechnol*, DOI: 10.1038/s41565-021-01009-9, (2021)), in which the self-field effect doesn't exist, also reported mirrored I_{c+} ($|I_{c-}|$) vs. B curves, similar with the results of these $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions. So far, the *field-induced* superconducting diode effect has been proposed to arise from effects such as magnetochiral anisotropy (*Sci. Adv.* **3**, e1602390 (2017)), finite momentum superconductivity (arXiv: 2106.01909v2 (2021)), and nonreciprocal Landau critical momentum (arXiv: 2106.03326 (2021)). More investigation in future work is needed to understand the origin of the effect in $NbSe_2/NbSe_2$ and $NbSe_2$/FLG/$NbSe_2$ junctions, but that is not the point of this work.

We added the results of this $NbSe_2/NbSe_2$ junction with smaller critical current in Methods alongside the results of previous $NbSe_2/NbSe_2$ junction with large current.

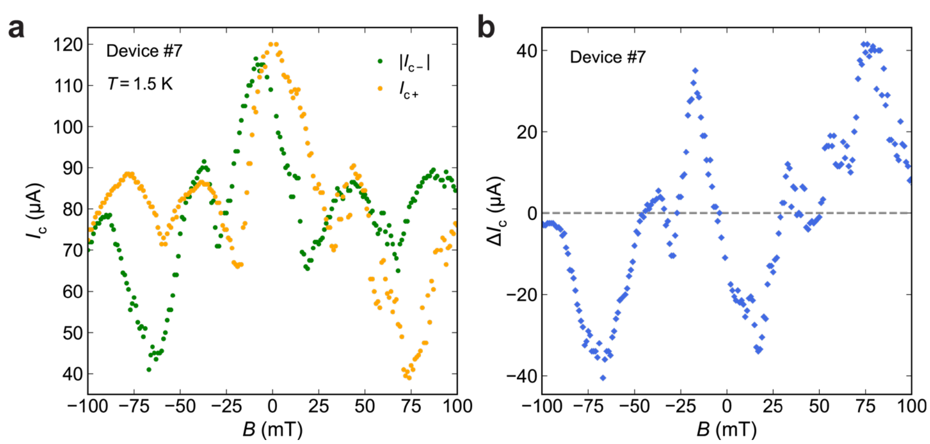

Fig. R4 | a, I_{c+} and $|I_{c-}|$ as a function of applied magnetic field of $NbSe_2/NbSe_2$ junction (Device #7). **b**, ΔI_c as a function of magnetic field of Device #7.

(5) The London penetration depth λ the authors obtained is about 4 nm, while in former reports λ for NbSe2 is around 100-300 nm (refs. 2-4). I think the authors should give some explanations.

Refs.

1. Yabuki, N. et al. Supercurrent in van der Waals Josephson junction. Nat. Commun. 7, 10616 (2016).

2. de Trey, P. & Gygax, S. Anisotropy of the Ginzburg-Landau Parameter κ in NbSe2. J Low Temp Phys 11, 421–434 (1973).

3. Le, L. P. et al. Magnetic penetration depth in layered compound NbSe2 measured by muon spin relaxation. Phys. C 185–189, 2715–2716 (1991).

4. Fletcher, J. D. et al. Penetration Depth Study of Superconducting Gap Structure of 2H − NbSe2. Phys. Rev. Lett. 98, 057003 (2007).

We thank the referee for the comment. The London penetration depth of $NbSe_2$ in our $NbSe_2/Nb_3Br_8/NbSe_2$ Josephson junctions is smaller than that in ref. 2-4, because the experimental condition of $NbSe_2$ is very different with those in ref. 2-4. First, these studies examined $NbSe_2$ single crystals, rather than $NbSe_2$ thin flakes or $NbSe_2$ based heterostructures. Second, $NbSe_2$ is known to have strong anisotropy between in-plane and out-of-plane transport due to its layered nature. However, in Ref. 3, the layered $NbSe_2$ is considered to have an isotropic penetration depth, which ignored the highly anisotropic nature of $NbSe_2$. And in ref. 4, only the in-plane penetration depth (λ~120-160 nm) can be found, which is not comparable to the penetration depth in our vertical Josephson junction.

Actually, in a more recent work, a very small London penetration depth (<5 nm) in bulk $NbSe_2$ at low temperature (~ 1.5 K) (*Nat. Commun.* **9**, 2796 (2018)) was seen. And in $NbSe_2$ based Josephson junctions, the London penetration depths were reported to be around ~25 nm and ~35 nm in $NbSe_2$/graphene/$NbSe_2$ (*Nano Lett.* **17**, 6125 (2017)) and $NbSe_2/Cr_2Ge_2Te_6/NbSe_2$ (*Nat. Commun.* **12**, 6580 (2021)) Josephson junctions, respectively.

Finally, in ref. 1 the referee mentioned in comment 3, the authors also found λ of 5nm in their $NbSe_2/NbSe_2$ Josephson junctions, similar to the λ in our $NbSe_2/NbSe_2$ Josephson junctions (~7 nm) according to Fig. R4. In addition, in another $NbSe_2/Nb_3Br_8/NbSe_2$ junctions (Device #2), the λ is also found to be ~ 4 nm according to Fig. R2, extremely similar to the λ ~ 4nm in $NbSe_2/Nb_3Br_8/NbSe_2$ junctions in main text. Therefore, the London penetration depth of $NbSe_2$ in our junction is very reasonable.

In conclusion, we appreciate the comments of the referee who prompted further experiments which have yielded high quality data and clarity. We have enlisted theoretical support and added calculations on the barrier material, obtained more experimental results and discussion where we re-demonstrated the Josephson junction nature of our heterostructures by providing a high-quality Fraunhofer pattern of a new $NbSe_2/Nb_3Br_8/NbSe_2$ device and confirmed the importance of interface and barrier material through control experiments. We hope with these modifications our work is found acceptable to publish in Nature.

Referee #3 (Remarks to the Author):

The paper is clearly written and the response to the referee comments is satisfactory. I think the paper is now ready for publication.

We thank the referee for the recommendation for publication.

Reviewer Reports on the Third Revision:

Referee #2 (Remarks to the Author):

The authors have addressed my concerns on the origin of the Josephson diode effect from the asymmetric Josephson tunneling, and their data on the Fraunhofer pattern have been improved. I appreciate their efforts and recommend this publication.