

# The field-free Josephson diode in a van der Waals heterostructure

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The superconducting analogue to the semiconducting diode, the Josephson diode, has long been sought with multiple avenues to realization being proposed by theorists<sup>1–3</sup>. Showing magnetic-field-free, single-directional superconductivity with Josephson coupling, it would serve as the building block for next-generation superconducting circuit technology. Here we realized the Josephson diode by fabricating an inversion symmetry breaking van der Waals heterostructure of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$ . We demonstrate that even without a magnetic field, the junction can be superconducting with a positive current while being resistive with a negative current. The  $\Delta I_c$  behaviour (the difference between positive and negative critical currents) with magnetic field is symmetric and Josephson coupling is proved through the Fraunhofer pattern. Also, stable half-wave rectification of a square-wave excitation was achieved with a very low switching current density, high rectification ratio and high robustness. This non-reciprocal behaviour strongly violates the known Josephson relations and opens the door to discover new mechanisms and physical phenomena through integration of quantum materials with Josephson junctions, and provides new avenues for superconducting quantum devices.

Reciprocity in charge transport describes the symmetric behaviour of current with voltage; the magnitude of the current generated by a positive voltage is the same as that generated by a negative voltage<sup>4</sup>. Violating reciprocity, that is, non-reciprocal behaviour, forms the basis for numerous important electronic devices such as diodes, a.c./d.c. converters, photodetectors, transistors and so on<sup>5,6</sup>. A well-known example of a non-reciprocal device is the p–n junction, formed by the interface of a p- and n-doped semiconductor, which shows an asymmetric current–voltage characteristic (IVC) without applied magnetic field ( $B$ ) and is widely used in various semiconducting technologies including logic and computation<sup>7</sup>.

So far, in the superconducting regime, field-induced non-reciprocal behaviour has been seen in non-centrosymmetric superconductors<sup>8–10</sup> and two-dimensional electron gases<sup>11</sup>, and multiple mechanisms for this phenomenon have been proposed<sup>9,12,13</sup>. All require an applied magnetic field to achieve simultaneous breaking of inversion symmetry and time reversal symmetry<sup>14–16</sup> so that with sufficient magnetic field strength, the critical current required to destroy the superconducting state in one direction can be different from that in the other direction<sup>8</sup>. Non-reciprocal superconductivity without an applied magnetic field as not yet been observed either in a bulk superconductor or a superconducting junction, such as a Josephson junction (JJ).

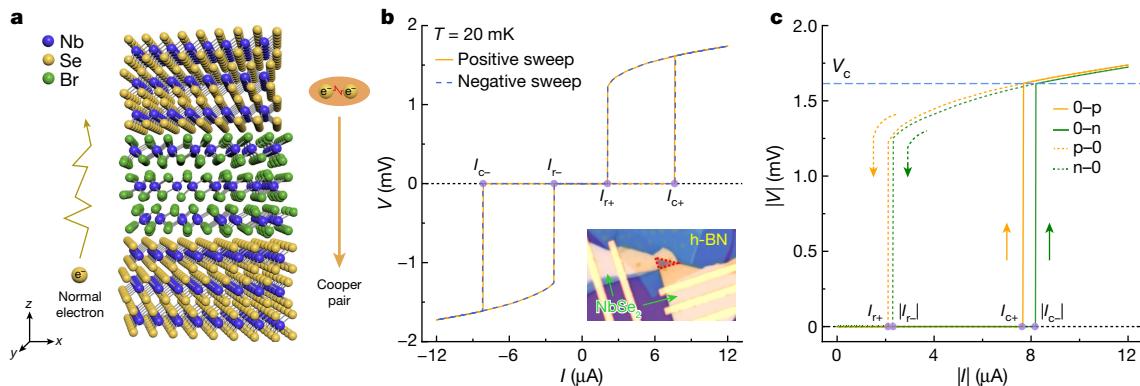
In particular, the JJ is also known to show reciprocal behaviour, as can be seen by the Josephson relations<sup>17,18</sup>. JJs are typically formed by two superconducting electrodes bridged by a non-superconducting barrier composed of classical materials ( $\text{AlO}_x$ ,  $\text{Cu}$  and so on)<sup>19,20</sup>. However,

integrating new quantum materials as the barriers in JJs may lead to anomalous behaviour violating the Josephson relations (such as non-reciprocal behaviour) and is of fundamental and technological interest. Creating a superconducting analogue to the semiconducting diode, a field-free ‘Josephson diode’ (JD), allowing supercurrent in one direction but normal current in the other direction in the absence of an applied magnetic field, is an open challenge.

One approach is to look for non-reciprocal IVCs in superconducting systems that only break inversion but not time reversal symmetry. Recently, JJs with only inversion symmetry breaking were predicted to show non-reciprocal IVCs owing to the formation of a Mott-insulating depletion region at the interface of p-doped and n-doped superconductors<sup>2</sup>. As JJs form the basis of a variety of important technologies<sup>21,22</sup> including superconducting quantum interference devices, superconducting quantum computing bit (qubits) and rapid single flux quantum devices<sup>23–25</sup>, the realization of the JD will have a substantial impact on superconducting electronics.

Here we report the realization of the field-free JD in an inversion symmetry breaking van der Waals heterostructure of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$ . An asymmetric IVC, with sharp superconducting transitions, is seen at a zero magnetic field indicating its field-free JD behaviour. A superconducting half-wave rectification of a square-wave excitation is demonstrated with a large rectification ratio, ultralow switching current density and highly stable performance. Moreover, the  $\Delta I_c$  versus  $B$ , presents a symmetric behaviour, unlike the antisymmetric response seen in previous systems<sup>8</sup>, further proving its

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**Fig. 1 | Schematic and superconductivity of the JD at zero field.** **a**, Schematic of a JD in a vertical JJ architecture, consisting of an  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  sandwich. The junction lacks inversion symmetry along the  $z$  direction, and can show normal conduction in one direction but superconductivity in the other (Cooper pair conduction). **b**,  $V$ - $I$  curves measured by sweeping positive (orange solid line) and sweeping negative (blue dashed line), as defined in the main text.

Inset, a typical device architecture; the red triangle outlines the JJ geometry created by the overlapping flakes. **c**,  $V$ - $I$  curves with positive sweep; the orange solid and dashed lines correspond with the  $0\text{-}p$  and  $p\text{-}0$  branches, respectively, and the green solid and dashed lines correspond with the  $0\text{-}n$  and  $n\text{-}0$  branches, respectively. The purple dots mark the position of  $I_{c+}$ ,  $|I_{c-}|$ ,  $I_r$ , and  $|I_{r-}|$  and the blue dashed line shows the critical voltage  $V_c$ .

field-free nature. The field-free JD effect may lie in the asymmetric tunnelling of supercurrent across the tunnel barrier, and the inversion symmetry breaking of the junction is confirmed by antisymmetric second-harmonic peaks. The effect shown here may be extended to a variety of material systems and architectures, particularly using van der Waals materials, for the advancement of future superconducting electronics and quantum devices. Also, this study shows how the interplay between symmetry breaking, new tunnel barrier materials and superconducting electrodes can lead to new or anomalous Josephson phenomena in JJs, opening the door to a variety of fundamental and applied research directions.

## Experimental data

We fabricated vertical JJs by sandwiching an  $\text{Nb}_3\text{Br}_8$  thin flake with thin flakes of  $\text{NbSe}_2$  as shown in Fig. 1a.  $\text{Nb}_3\text{Br}_8$  is a van der Waals quantum material that crystallizes in the  $\overline{R}\bar{3}m$  space group at low temperature and its unit cell is composed of six layers of Nb-Br edge-sharing octahedra<sup>26,27</sup> with inversion centres lying between adjacent layers (Extended Data Fig. 1a). The inversion symmetry is preserved in even-layered  $\text{Nb}_3\text{Br}_8$  but broken in odd-layered  $\text{Nb}_3\text{Br}_8$ . Additionally,  $\text{Nb}_3\text{Br}_8$  was shown to have a singlet magnetic ground state<sup>27</sup> and moderate band gap like its sister material,  $\text{Nb}_3\text{Cl}_8$ <sup>27–30</sup> (see Methods and Extended Data Fig. 5 for magnetoresistance characterization of the  $\text{Nb}_3\text{Br}_8$  thin flakes). The  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  heterostructures were fabricated by the dry transfer method<sup>31</sup> (Methods) and capped by a top hexagonal boron nitride (h-BN) to avoid degradation, as a typical device in the inset of Fig. 1b shows. Here  $\text{NbSe}_2$  flakes with thickness about 28–37 nm are used as superconducting electrodes, rotated by arbitrary angles with respect to each other, breaking inversion symmetry of the JJ, and a thin flake of  $\text{Nb}_3\text{Br}_8$  with thickness of 2.3 nm (three layers) is used as the tunnel barrier.

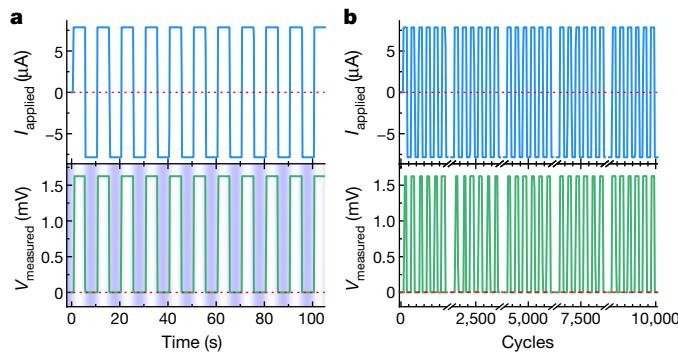
When cooling the junction to 20 mK in zero field, a sudden drop to zero resistance at  $T_c = 6.6$  K is observed, as shown in Extended Data Fig. 1b. Figure 1b investigates the voltage versus current ( $V$ - $I$ ) behaviour, measured by sweeping the d.c. current at 20 mK (see  $V$ - $I$  curves at different temperatures in Extended Data Fig. 1c). We emphasize that the measurement of the  $V$ - $I$  curves contains four branches; sweeping the current from zero to positive ( $0\text{-}p$ ), from positive back to zero ( $p\text{-}0$ ), from zero to negative ( $0\text{-}n$ ) and from negative back to zero ( $n\text{-}0$ ). These four branches can be measured in the above order (defined as the positive sweep), or in reverse order,  $0\text{-}n$ ,  $n\text{-}0$ ,  $0\text{-}p$  and  $p\text{-}0$  (defined as the negative sweep). Figure 1b shows  $V$ - $I$  curves of the positive sweep and

the negative sweep; both of them show hysteresis, as expected, arising from the capacitance induced different critical currents when breaking ( $I_c$ ) compared with returning ( $I_r$ ) to the superconducting state<sup>1</sup>. Note that the properties measured here all come from the JJ itself as the maximum current applied here is much smaller than the critical current of thick  $\text{NbSe}_2$  (a few millamps)<sup>32</sup>. The two curves lie precisely on top of each other meaning that the positive versus negative sweep order does not affect the  $V$ - $I$  response. However, the  $I_c$  and  $I_r$  in the positive current regime (labelled  $I_{c+}$  and  $I_{r+}$ ) can be compared with those in the negative current regime (labelled  $|I_{c-}|$  and  $|I_{r-}|$ ). For reciprocal transport, as is expected for a conventional JJ without magnetic field, there should be no difference between  $I_{c+}$  versus  $|I_{c-}|$  or  $I_{r+}$  versus  $|I_{r-}|$ .

Figure 1c shows the absolute values of  $I_{c+}$  and  $I_{r+}$  as well as  $I_{c-}$  and  $I_{r-}$  of the positive sweep. Immediately evident is that  $I_{c+}$  and  $I_{r+}$  are not equal to their negative current counterparts, with a  $\Delta I_c$  ( $\Delta I_c = |I_{c-}| - I_{c+}$ ) roughly 0.5  $\mu\text{A}$  and a smaller  $\Delta I_r$  roughly 0.1  $\mu\text{A}$ , indicating the non-reciprocal  $V$ - $I$  response in our JJ. It is important to note that these differences are intrinsic properties of the JJ rather than an extrinsic Joule heating effect (see discussion in the Methods). The different absolute values of  $I_{c+}$  and  $I_{c-}$  combined with the sharp superconducting transition indicate that when the applied current is in between  $I_{c+}$  and  $|I_{c-}|$ , the junction would show superconducting behaviour with a negative current but normal conducting behaviour with a positive current.

On the basis of this idea, we demonstrated half-wave rectification at zero field, as shown in Fig. 2a. Because the  $I_{c+}$  and  $|I_{c-}|$  being 7.61 and 8.14  $\mu\text{A}$ , respectively, we applied a square-wave excitation (Fig. 2a, top panel) with an amplitude of 7.9  $\mu\text{A}$  at a frequency of 0.1 Hz. As shown in the bottom panel of Fig. 2a, the junction remains in the superconducting state with the negative current (purple area) while switching to the normal state during the positive current (white area). The measured voltage in the superconducting state is smaller than  $4 \times 10^{-7}$  V (our detection limit), whereas the normal state junction voltage is 1.6 mV, meaning the diode rectification ratio demonstrated here is at least around 10<sup>4</sup>. In addition, the switching current density and switching power are  $2.2 \times 10^2$   $\text{A cm}^{-2}$  and 12.3 nW, respectively, which are two and four orders of magnitude smaller than in the bulk superconducting diode<sup>8</sup>.

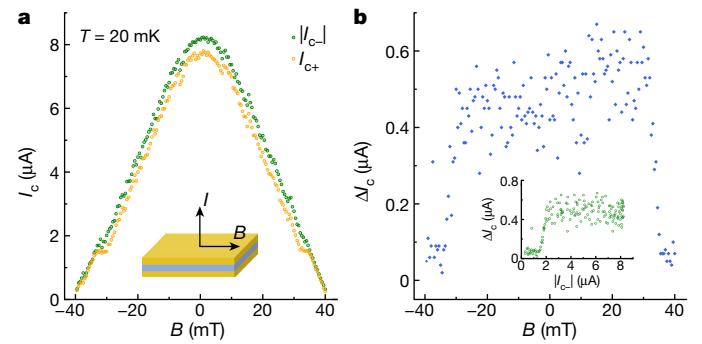
We further probed the durability of the JD and robustness of half-wave rectification, as shown in Fig. 2b by conducting 10,000 continuous cycles with an applied square-wave excitation with an amplitude of 7.9  $\mu\text{A}$  at 0.5 Hz and zero field. We note that the ‘on’ and ‘off’ state voltages stayed constant during the 10,000 cycles, and the device remains stable after that, indicating the high stability of the JD. Importantly, the JD effect is not just an ultralow temperature property; according



**Fig. 2 | Half-wave rectification and durability test of the JD at 20 mK and zero field.** **a**, The top panel shows the applied square-wave excitation with an amplitude of 7.9  $\mu$ A (in between  $I_{c+}$  and  $|I_{c-}|$ ) and frequency of 0.1 Hz. The bottom panel is the coincidentally measured junction voltage. The purple shaded region denotes the superconducting state in which the voltage is zero during negative current bias and the white region denotes the normal state in which the voltage is  $V_c$  during positive current bias. The red dotted line is the zero line. **b**, The top panel is the applied square-wave excitation with an amplitude of 7.9  $\mu$ A and frequency of 0.5 Hz over 10,000 cycles. The bottom panel is the coincidentally measured junction voltage over those cycles, showing no degradation in the response. The red dashed lines are zero lines.

to the asymmetric  $V$ - $I$  curves observed at 0.9 and 3.86 K (Extended Data Fig. 2), an ideal half-wave rectification was also realized at 0.9 K whereas the half-wave rectification at 3.86 K had some punch-through error, likely due to thermal fluctuations, as shown in Extended Data Fig. 2b, d, respectively. In addition, the large hysteresis at ultralow temperatures indicates the junction is in the strongly underdamped regime. However, by 3.86 K, the p-0 (n-0) almost coincides with the 0-p (0-n) branch, indicating the junction changed to the overdamped regime (Methods), enabling possible operation frequencies of the order of the superconducting switching speed (THz)<sup>24,33</sup>.

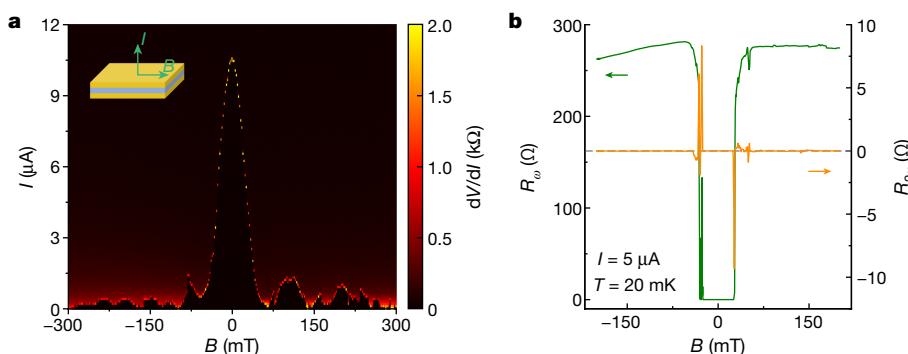
To further confirm the field-free nature of the JD effect, we measured  $V$ - $I$  curves containing 0-p and 0-n branches with an in-plane magnetic field (inset of Fig. 3a) swept from 40 to -40 mT with 0.5 mT steps. As before, the  $I_{c+}$  and  $|I_{c-}|$  was extracted and plotted in Fig. 3a ( $V$ - $I$  curves with magnetic fields shown in Extended Data Fig. 3). Both  $I_{c+}$  and  $|I_{c-}|$  decrease with increasing magnetic field, as expected for a JJ. Below 35 mT,  $\Delta I_c$  emerges, as the  $|I_{c-}|$  increases faster than  $I_{c+}$  as  $B$  goes to zero. Note that  $|I_{c-}|$  stays larger than  $I_{c+}$  regardless of the direction of the magnetic field. Figure 3b shows the  $\Delta I_c$  plotted as a function of  $B$ ; the  $\Delta I_c$  is sustained at zero field and its magnitude is independent of  $B$  in the low-field region. The inset of Fig. 3b shows the variation of  $\Delta I_c$  with decreasing  $|I_{c-}|$ , and that  $|I_{c-}|$  can be up to 25% larger than  $I_{c+}$ . The field-dependent  $\Delta I_c$  behaviour here differs from the previously mentioned field-induced superconducting diode effect in which the  $\Delta I_c$  shows an antisymmetric field dependence<sup>8</sup>. Also, in bulk superconductors with field-induced superconducting diode effects,  $\Delta I_c$  versus  $B$



**Fig. 3 | Magnetic field dependence of  $I_c$  and  $\Delta I_c$ .** **a**,  $I_{c+}$  (orange dots) and  $|I_{c-}|$  (green dots) obtained from the 0-p and 0-n branches of the positive sweep as a function of applied magnetic field. The magnetic field was swept from positive to negative. The diode effect ‘turns off’ by  $\pm 35$  mT. Inset, a schematic of the measurement geometry; the orange and blue layers represent  $\text{NbSe}_2$  and  $\text{Nb}_3\text{Br}_8$ , respectively. **b**,  $\Delta I_c$  as a function of magnetic field.  $\Delta I_c$  is roughly field invariant until 35 mT with the diode behaviour robust in zero field. Inset,  $\Delta I_c$  as a function of  $|I_{c-}|$  from modulation of magnetic field with the diode effect ‘turning off’ below 2.1  $\mu$ A.

has been sweep direction dependent, flipping sign<sup>8</sup>, whereas in the field-free JD demonstrated here,  $\Delta I_c$  versus  $B$  shows no dependence on sweep direction (Extended Data Fig. 4).

To prove Josephson coupling of the junction, the  $dV/dI$  mapping as a function of bias current and magnetic field was measured and is shown in Fig. 4a. As expected, the characteristic single slit Fraunhofer pattern from the Josephson effect is observed (see Methods and Extended Data Fig. 9 for further measurements and discussion). The critical current of the JJ decreases rapidly with increasing magnetic field, consistent with the  $I_c$  decreasing in Fig. 3a. To further confirm the inversion symmetry breaking in the junction, the field-dependent first- and second-harmonic resistances were measured with an a.c. current with an amplitude of 5  $\mu$ A and frequency of 7.919 Hz, as shown in Fig. 4b. The sharp drop of the first harmonic resistance in roughly  $\pm 30$  mT shows the onset of superconductivity, whereas the two antisymmetric peaks appear in the second-harmonic resistance at the same fields indicating that inversion symmetry in the junction must be broken<sup>8-10</sup>. There can be two contributions to inversion symmetry breaking here; first is the intrinsic lack of inversion symmetry of the three-layer  $\text{Nb}_3\text{Br}_8$ , and second is the geometry of the vertical junction in which the top and bottom  $\text{NbSe}_2$  electrodes are arbitrarily rotated relative to each other and the  $\text{Nb}_3\text{Br}_8$  layer. Hence we also fabricated and measured four-layer  $\text{Nb}_3\text{Br}_8$  devices, which show the same symmetric  $\Delta I_c$  versus  $B$  behaviour and field-free JD effect as the three-layer  $\text{Nb}_3\text{Br}_8$  JJ as well as antisymmetric peaks in the second-harmonic resistance measurements (see details in the Methods and Extended Data Fig. 6). These results indicate that the asymmetric  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8$  interfaces in the junction play an essential role for inversion symmetry breaking and the field-free JD effect.



**Fig. 4 | Critical current map and second-harmonic resistance of the JD.** **a**, Colour map of  $dV/dI$  as a function of applied magnetic field and applied current, showing the single slit Fraunhofer pattern from the Josephson effect of a JJ. **b**, First-harmonic (green line) and second-harmonic (orange line) resistances measured by applying an a.c. current of 5  $\mu$ A at 20 mK. The antisymmetric peaks in the second harmonic correspond with the critical field, showcasing the non-reciprocal nature of the superconductivity.

## Discussion

We discuss the possible origin of  $\Delta I_c$  and the diode behaviour. Fundamentally, to obtain a  $\Delta I_c$  and  $\Delta I_r$ , an asymmetric  $V$ - $I$  curve is required, that is, the  $I_+$  behaviour needs to be distinct from the  $I_-$  behaviour. From the phenomenological resistively and capacitively shunted junction (RCSJ) model of a JJ<sup>17</sup>, the total current through the junction  $I$  is determined by  $I = I_F + I_{Cap} + I_R + I_J$ , where  $I_F$  is the fluctuation current of the noise channel,  $I_{Cap}$  is the current of the capacitive channel,  $I_R$  is the extra resistive channel arising from finite temperature quasiparticle excitation and  $I_J$  is the Josephson current. As  $I_F$ ,  $I_{Cap}$  and  $I_R$  have negligible influence on the critical current (see the Methods for details), we speculate that the critical current through the JJ should be governed by the Josephson current  $I_J$ , ( $I_J = I_c \sin \varphi$ , where  $\varphi$  is phase difference of two superconductors). Because  $\Delta I_c$  is suppressed with  $B$  and  $I_{c+}$  ( $I_{c-}$ ) are symmetric with  $B$ , and the current is parallel to the symmetry breaking direction while perpendicular to magnetic field direction in our junction, magnetochiral anisotropy can be ruled out as the driving mechanism here. Also to exclude the NbSe<sub>2</sub> electrodes as the driving factor, we fabricated JJs of arbitrarily rotated NbSe<sub>2</sub>/NbSe<sub>2</sub> and NbSe<sub>2</sub>/few-layer graphene (FLG)/NbSe<sub>2</sub> junctions<sup>34,35</sup>, and found they both show a field-induced superconducting diode effect with antisymmetric  $\Delta I_c$  versus  $B$  characteristics (Methods and Extended Data Figs. 7 and 8). Therefore, in the Nb<sub>3</sub>Br<sub>8</sub>JJs,  $\Delta I_c$  must be induced by asymmetric Josephson tunnelling at a zero field.

Recently, Kitamura et al. discussed non-reciprocal Landau–Zener tunnelling in inversion symmetry breaking materials with polarization<sup>36</sup>, explaining that the effect in the normal state originates from direction dependent modulation of the electron tunnelling probability across a tunnel barrier because of a shift in wave-packet position. Alternatively, Hu et al. proposed a JD based on an interface of p- and n-doped superconductors<sup>2</sup> that breaks inversion symmetry and forms a polarized Mott-insulating region that can be suppressed by voltage in one direction and elongated in the other direction, resulting in diodic behaviour.

In the situation of NbSe<sub>2</sub>/Nb<sub>3</sub>Br<sub>8</sub>/NbSe<sub>2</sub>JJs, Nb<sub>3</sub>Br<sub>8</sub> was recently predicted to be an obstructed atomic insulator (OAI) with Wannier charge centres symmetrically pinned at the unoccupied inversion centres in between two Br-Nb-Br layers<sup>37,38</sup> (Methods and Extended Data Fig. 10). The negative charge centres are separated from the positive ones along the *c* axis of the crystal, and combined with the asymmetric NbSe<sub>2</sub>/Nb<sub>3</sub>Br<sub>8</sub> interfaces, an out-of-plane (OOP) polarization can occur<sup>38</sup>. In analogy to the previously described theoretical works of polarized systems, we propose that a polarization in the NbSe<sub>2</sub>/Nb<sub>3</sub>Br<sub>8</sub>/NbSe<sub>2</sub>JJs may induce asymmetric Josephson tunnelling and lead to the field-free JD effect; further theoretical and experimental study is necessary to fully explain the mechanism.

Also, future optimization of the JD architecture through tuning the geometry or type of barrier can result in more ideal diodic behaviour. Creating further quantum material JJs, such as those demonstrated here, can uncover emergent phenomena leading to new or anomalous Josephson phenomena through the effect of the intrinsic properties of the barrier. Future work combining materials possessing properties such as topological states, ferroelectricity, magnetoelectricity, non-collinear magnetism, OAI and emergent properties from twisted heterostructures, with superconducting electrodes may allow the realization of new Josephson phenomena. Thus the opportunities for creating superconducting electronics using quantum materials are vast and just beginning.

## Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41586-022-04504-8>.

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## Methods

### Synthesis of $\text{Nb}_3\text{Br}_8$ crystal

$\text{Nb}_3\text{Br}_8$  crystals used in this work were grown through chemical vapour transport. Stoichiometric mixtures of niobium powder (Alfa, 99.99%) and  $\text{NbBr}_5$  (Strem, 99.9%) with a total mass of 1.5 g were ground together and added to a 14 mm inner diameter (with a 2 mm thick wall) fused silica tube in a glovebox and handled using standard air free techniques. Next, 20 mg of  $\text{NH}_4\text{Br}$  was included as a transport agent. The tubes were then sealed air free at a length of approximately 30 cm, about 5 cm longer than the first two zones of a three-zone furnace. A three-zone furnace was used with a temperature gradient of 840, 785 and 795 °C with all but the last few centimetres of the tube between the first two zones. This discouraged the formation of large intergrown clumps of crystals at the end of the tube. The furnace was held at temperature for 3–5 days before being cooled to room temperature over 7 h.

### Fabrications of the JJ device

We fabricated the  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  JJ in a glove box with an inert environment to avoid oxidation and decay. The bottom  $\text{NbSe}_2$  flake was directly exfoliated from an  $\text{NbSe}_2$  single crystal (HQ graphene) to a  $\text{SiO}_2/\text{Si}$  wafer, then  $\text{Nb}_3\text{Br}_8$  thin film and top  $\text{NbSe}_2$  flake were exfoliated and successively transferred layer by layer by using the polydimethylsiloxane assistant dry transfer method<sup>31</sup>. A h-BN (HQ graphene) layer was finally transferred on the top to protect the JJ from degradation in the atmosphere. Then Ti (3 nm)/Au (50 nm) electrodes were deposited on the top and bottom  $\text{NbSe}_2$  flakes for transport measurement.

### Transport measurements

The transport properties of JJ device were measured in a BlueFors dilution refrigerator with a base temperature of 20 mK. A Keithley 6221 AC/DC Current Source Meter was used to inject both d.c. current and square-wave excitation, and the d.c. voltage was measured using a Keithley 2182a Nanovoltmeter. The a.c. measurements were performed by injecting a.c. current with a frequency of  $\omega$  (7.919 Hz) using a Zurich MFLI amplifier, and the second-harmonic signals were obtained from the out-of-phase  $2\omega$  component of the a.c. voltage. A Fraunhofer pattern was obtained by measuring the differential resistance ( $dV/dI$ ) versus / curve under different magnetic fields using a lock-in amplifier (Zurich MFLI) with a d.c. bias (Keithley 2636).

### Excluding the Joule heating effect

In Fig. 1b, we measured positive and negative sweep  $V$ – $I$  curves with the same sweep rates. These two curves lie on top of each other, indicating that the  $V$ – $I$  curves and associated  $I_{c+}$  and  $I_{c-}$  are not dependent on the two sweep directions. As shown in Fig. 1c, the  $V$ – $I$  is asymmetric and  $I_{c+}$  and  $I_{c-}$  have prominently different values. In general, the Joule heating effect is proportional to the heating time and the  $I_{c+}$  (or  $I_{c-}$ ) obtained in positive and negative sweeps undergo different heating times, which can cause different local temperatures in sample. If the asymmetric  $V$ – $I$  was induced by the Joule heating effect, the values of  $I_{c+}$  and  $I_{c-}$  would switch for the positive and negative sweeps: that is, the effect would be sweep direction dependent. However, the  $V$ – $I$  curves of positive and negative sweeps are the same, strongly indicating that the different  $I_{c+}$  and  $I_{c-}$  obtained indeed are intrinsic to the device rather than induced extrinsically by the Joule heating effect.

### Magnetoresistance characterization of $\text{Nb}_3\text{Br}_8$ thin flake

The  $\text{Nb}_3\text{Br}_8$  single crystal has previously been shown to have a phase transition from a high temperature ferromagnetic state to a magnetic singlet ground state at low temperatures ( $\beta$  phase)<sup>27</sup>. Here we use transport measurements to investigate the magnetic property of a  $\text{Nb}_3\text{Br}_8$  thin flake. First, we measured sweep-up (magnetic field sweep from negative to positive) and sweep-down (magnetic field sweep from positive to negative) magnetoresistances of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  JJ

(device 1, device in main text) with a temperature above  $T_c$  of  $\text{NbSe}_2$  superconducting electrodes. Extended Data Fig. 5a, b shows the magnetoresistances of device 1 at 10 K with in-plane and OOP magnetic fields up to 7 T, respectively. The overlapping sweep-up and sweep-down magnetoresistances curves reveal no hysteresis or reversion between these curves, indicating no ferromagnetic signal in the  $\text{Nb}_3\text{Br}_8$  thin flakes<sup>39,40</sup>. Moreover, we also replaced  $\text{NbSe}_2$  superconducting electrodes with FLG to fabricate FLG/ $\text{Nb}_3\text{Br}_8$ /FLG devices and measured their magnetoresistances with an OOP magnetic field. Extended Data Fig. 5c shows the results of one of the FLG/ $\text{Nb}_3\text{Br}_8$ /FLG devices (device 6) measured at 2 K. Similar to the results in device 1, the magnetoresistances overlap well and no hysteresis or reversion signals can be observed in the curves, which is also consistent with the observation of the magnetic singlet ground state<sup>27</sup>. It is worth noting that the oscillations in FLG/ $\text{Nb}_3\text{Br}_8$ /FLG device are induced by the quantum oscillation of FLG.

### Field-free JD effect in a $\text{NbSe}_2$ /four-layer $\text{Nb}_3\text{Br}_8/\text{NbSe}_2$ JJ (device 3)

A  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  JJ with a four-layer  $\text{Nb}_3\text{Br}_8$  as the tunnel barrier (device 3) was fabricated using the same technique as device 1 (device in main text). We measured  $V$ – $I$  curves at different magnetic fields at 20 mK, and extracted  $I_{c+}$  and  $|I_{c-}|$  from 0–p and 0–n branches, respectively, and plotted them as a function of magnetic field as shown in Extended Data Fig. 6a. The  $I_{c+}$  and  $|I_{c-}|$  show similar tendencies and behaviours to device 1. Extended Data Fig. 6b shows the field-dependent calculated  $\Delta I_c$ . The non-zero  $\Delta I_c$  can be clearly observed at zero field and low-field region, and it decreases to almost zero at relative higher fields, which is similar to the phenomenon in device 1. Therefore, the JJ with four-layer  $\text{Nb}_3\text{Br}_8$  also has a field-free JD effect. To confirm the inversion symmetry breaking, we also performed the second-harmonic resistances ( $R_{2\omega}$ ) measurements at 20 mK with an applied current of 4  $\mu\text{A}$ , as shown in Extended Data Fig. 6c. The two antisymmetric  $R_{2\omega}$  peaks corresponding with the superconducting transition of  $R_\omega$  reveal the inversion symmetry breaking nature of the JJ<sup>9,15</sup>. Because the four-layer  $\text{Nb}_3\text{Br}_8$  preserves inversion symmetry, as shown in Extended Data Fig. 1a, the inversion symmetry breaking of device 3 originates from the different interfaces of the top and bottom  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8$ .

### Magnetic-field-induced superconducting diode effects in $\text{NbSe}_2/\text{NbSe}_2$ (devices 4 and 7) and $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$ (device 5) junctions

We also fabricated other junctions as control experiments to narrow the suspects of the field-free JD effect mechanism. First, we considered the influence of the superconducting electrode  $\text{NbSe}_2$ , which is a type II and multi-gapped superconductor. A previous study reported non-reciprocal transport in atomically thin  $\text{NbSe}_2$  films that broke inversion symmetry, which disappeared in bulk  $\text{NbSe}_2$  crystal<sup>10</sup>. In all our devices, we used thick  $\text{NbSe}_2$  flakes (>20 nm) as superconducting electrodes, which have similar properties to bulk  $\text{NbSe}_2$ . We also fabricated a  $\text{NbSe}_2/\text{NbSe}_2$  junction (device 4) to explore its property. Extended Data Fig. 7a shows the  $V$ – $I$  curve measured with the positive sweep (a sequence of 0–p, p–0, 0–n and n–0 branches) at 2 K and 0 T. As shown in Extended Data Fig. 7a, there are three transition steps in the  $V$ – $I$  curve, which correspond with the critical currents of junction, bottom and top  $\text{NbSe}_2$  electrodes, respectively. There is no obvious difference between  $I_{c+}$  and  $I_{c-}$  of the junction. The field dependence of critical currents was further measured; we extracted the  $I_{c+}$  and  $|I_{c-}|$  of the junction from each  $V$ – $I$  curve measured at different magnetic fields and plotted them as a function of magnetic field as shown in Extended Data Fig. 7b. The  $I_{c+}$  and  $|I_{c-}|$  seemed to have a mirror symmetry with each other.  $\Delta I_c$  was also calculated and is shown in Extended Data Fig. 7c. The antisymmetric  $\Delta I_c$  curve indicates that a  $\text{NbSe}_2/\text{NbSe}_2$  junction can only show a field-induced superconducting diode effect, rather than a field-free JD effect observed in  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  junctions. Second, to further confirm the influence of barrier materials, we also fabricated a  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junction (device 5) as a control experiment. The  $I_{c+}$

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and  $|I_{c-}|$  was extracted as before and shown in the inset of Extended Data Fig. 7d.  $\Delta I_c$  was also calculated and plotted as a function of magnetic field in Extended Data Fig. 7d. Similar to the results from the  $\text{NbSe}_2/\text{NbSe}_2$  junction device (device 4), the  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junction also shows the field-induced superconducting diode effect.

Previous studies have shown that a self-field effect in tunnel junctions with a large critical current density can result in a skewed Fraunhofer pattern, which may contribute to the field-induced superconducting diode effect in these junctions<sup>41,42</sup>. We also fabricated a  $\text{NbSe}_2/\text{NbSe}_2$  control device (device 7) with low critical current ( $I_c \approx 120 \mu\text{A}$ ) and a correspondingly lower critical current density ( $J_c \approx 18.5 \mu\text{A} \mu\text{m}^{-2}$ ) than device 4,  $J_c \approx 66 \mu\text{A} \mu\text{m}^{-2}$ . Extended Data Fig. 8 shows the  $I_c$  and corresponding  $\Delta I_c$  as a function of applied magnetic field. The  $I_{c+}$  and  $|I_{c-}|$  versus  $B$  have a mirror symmetry with each other, and the obvious antisymmetric feature of  $\Delta I_c$  versus  $B$  curve also confirms the field-induced superconducting diode effect in device 7, similar to device 4.

In addition, recent studies on planar  $\text{Nb}/\text{WTe}_3/\text{NbJJs}$ <sup>43</sup> and the indium arsenide two-dimensional electron gas system<sup>11</sup>, in which the self-field effect does not exist, also reported a mirrored  $I_{c+}$  ( $|I_{c-}|$ ) versus  $B$  curves, similar to the results from these  $\text{NbSe}_2/\text{NbSe}_2$  and  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junctions. So far, the field-induced superconducting diode effect has been proposed to arise from effects such as magnetochiral anisotropy<sup>9</sup>, finite momentum superconductivity<sup>12</sup> and non-reciprocal Landau critical momentum<sup>13</sup>. More investigation is needed to understand the origin of field-induced diode effect in  $\text{NbSe}_2/\text{NbSe}_2$  and  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junctions.

In summary, both  $\text{NbSe}_2/\text{NbSe}_2$  and  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junctions show the field-induced superconducting diode effect, similar to the behaviour of the  $[\text{Nb}/\text{V}/\text{Ta}]_n$  superlattice<sup>8</sup>. These observations in  $\text{NbSe}_2/\text{NbSe}_2$  and  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junctions indicate that the field-free JD effect does not originate from the superconducting property of  $\text{NbSe}_2$ . Also, even though the interfaces of these junctions also break the inversion symmetry, the field-free JD effect does not occur. These results, along with the observations in  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  devices (devices 1 and 3), indicate that the field-free JD effect originates from asymmetric Josephson tunnelling induced by the  $\text{Nb}_3\text{Br}_8$  barrier and the associated  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8$  interfaces in the junction.

## Basic analysis of the JJ (device 1) and discussion of Fraunhofer patterns (devices 1 and 2)

The large hysteresis between 0-p (0-n) and p-0 (n-0) branches on the  $V/I$ -curves (Fig. 1b) indicates the JJ (device 1) lies in the underdamped region. On the basis of the critical current ( $I_c$ ) and return current ( $I_r$ ) of  $V/I$ -curve measured at 20 mK, the Stewart-McCumber parameter ( $\beta_c \approx \left(\frac{4I_c}{mI_r}\right)$ )  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  JJ is 21, which is much larger than 1, further confirming that the JJ is in underdamped region<sup>17,18</sup>. On the basis of the equation  $\beta_c = \frac{2e}{h} I_c R_N^2 C$  (where  $C$  is the capacitance and  $R_N$  is normal state resistance of junction)<sup>17,18</sup>, the junction capacitance calculated is  $2.0 \times 10^{-14} \text{ F}$ , corresponding with specific capacitance ( $C/A$ , where  $A$  is the junction area and is roughly  $3.68 \mu\text{m}^2$  in device 1) of  $0.54 \mu\text{F} \text{ cm}^{-2}$ .

The Fraunhofer pattern in Fig. 4a is measured with a sweep-up magnetic field and Extended Data Fig. 9a shows the Fraunhofer pattern measured with a sweep-down magnetic field, which is similar to the sweep-up one. Both Fraunhofer patterns are not so periodic at higher field, which is a common case as seen in many other two-dimensional tunnel JJs, including  $\text{NbSe}_2/\text{NbSe}_2$  junctions<sup>34</sup> as well as  $\text{NbSe}_2/\text{graphene}/\text{NbSe}_2$  junctions<sup>35</sup>. 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<sup>35</sup>, 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). Therefore, the junction shape and magnetic field direction can also lead to a non-ideal Fraunhofer pattern, even in a high-quality junction with high-quality interfaces<sup>44</sup>.

Here we also characterized more junction parameters by analysing the Fraunhofer pattern in Extended Data Fig. 9a at a low magnetic field. In a JJ, the London penetration depth ( $\lambda$ ) of  $\text{NbSe}_2$  can be obtained from  $\phi_0 = \Delta B W (d + 2\lambda)$  where  $\phi_0$  is the magnetic flux quantum,  $\Delta B$  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 roughly 59 mT and with  $W \approx 3.5 \mu\text{m}$  and  $d \approx 2.3 \text{ nm}$ ,  $\lambda$  is calculated to be 3.85 nm.

In addition, to further demonstrate the  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  heterostructure is indeed a JJ, we fabricated another  $\text{NbSe}_2/2\text{-layer } \text{Nb}_3\text{Br}_8/\text{NbSe}_2$  heterostructure device (device 2), and obtained the expected high-quality Fraunhofer pattern, as shown in Extended Data Fig. 9b. Moreover, the field-free JD effect is also observed in this junction, as shown in Extended Data Fig. 9c and its inset. The  $|I_{c+}|$  and  $|I_{c-}|$  versus  $B$  curves shown in the inset of Extended Data Fig. 9c show a similar tendency and behaviour to device 1, and the extracted  $\Delta I_c$  shows non-zero values at zero field and in the low-field region, and decreases to around zero at higher fields similar to device 1.

## Discussion on RCSJ mode of JD

Here we discuss field-free JD behaviour based on the phenomenological RCSJ model of a JJ<sup>17</sup>; the total current through the junction  $I$  is determined by  $I = I_F + I_{\text{Cap}} + I_R + I_J$ , where  $I_F$  is the fluctuation current of the noise channel,  $I_{\text{Cap}}$  is the current of the capacitive channel,  $I_R$  is the extra resistive channel arising from finite temperature quasiparticle excitation and  $I_J$  is the Josephson current. In d.c. measurement, the thermal noise current ( $I_T$ ) dominates the fluctuation current, which can be calculated according to equation  $I_T = (2e/h)k_B T$  (ref. <sup>17</sup>), where  $h$  is reduced Planck constant,  $k_B$  is Boltzmann constant,  $e$  is elementary charge and  $T$  is the temperature. At 20 mK,  $I_T$  is about 0.8 nA, far less than  $\Delta I_c$ , and so will be disregarded as a cause of the  $V/I$  asymmetry. In principle, the other three components could have asymmetric  $V/I$  responses if inversion/time reversal symmetry is broken. Below, we examine each of those possibilities in the context of our JJ.

Regarding the capacitive channel, as mentioned earlier, Misaki et al.<sup>1</sup> proposed that in an inversion symmetry broken JJ, an asymmetric charging energy of capacitance can lead to a  $\Delta I_c$ , without an applied magnetic field, which was observed in our JJ device (Fig. 1c). However, this model did not include an expectation of a  $\Delta I_c$ , which is one of the main requirements (the other being sharp transitions) for realizing half-wave rectification. As  $I_J$  corresponds with the return to the superconducting state, applying a square-wave current excitation with an amplitude of  $I_{r+} + \Delta I_r/2$ , for example, would not result in rectification as both ends of the wave lie in the superconducting regime.

Considering the extra resistive channel ( $I_R = V/R_N$ ) from quasiparticle excitations, its contribution to the diode effect would stem from asymmetry in  $R_N$  and be significant if the resistance in the superconducting state was not zero. However, because the voltage in the superconducting state is zero in our device, the contribution of  $I_R$  to the critical current can be neglected. Therefore, the critical current through the JJ is governed by the Josephson current  $I_J$ , ( $I_J = I_c \sin \varphi$ ,  $I_c = V/R_N$  and  $\varphi$  is phase difference of two superconductors). The difference of positive and negative critical currents should originate from the asymmetric Josephson tunnelling in our JJ.

## The OAI phase of $\text{Nb}_3\text{Br}_8$

The crystal structure of  $\text{Nb}_3\text{Br}_8$  has the symmetries of the space group 166 ( $R\bar{3}m$ ), as shown in Extended Data Fig. 1a, in which Nb and Br atoms occupy the Wyckoff positions 6c and 18h, respectively. The electronic band structure and the band representation analysis of  $\text{Nb}_3\text{Br}_8$  have been studied<sup>38,45</sup>, and the symmetry properties of its valence bands can be characterized by the multiplicities of irreducible representations at all the high-symmetry momenta, which is referred to as the symmetry data vector,

$$B = (m(\bar{\Gamma}_4\bar{\Gamma}_5), m(\bar{\Gamma}_6\bar{\Gamma}_7), m(\bar{\Gamma}_8), m(\bar{\Gamma}_9), m(\bar{\Gamma}_3\bar{\Gamma}_4), m(\bar{\Gamma}_5\bar{\Gamma}_6), \dots, m(\bar{\Gamma}_9))^T \quad (1)$$
$$= (15, 15, 33, 32, 48, 47, 47, 48, 15, 15, 32, 33)^T$$

where  $m(\rho)$  represents the multiplicity of the irreducible representations  $\rho$  formed by the occupied Bloch bands at the corresponding high-symmetry momentum. From topological quantum chemistry<sup>46</sup>, the band representation, which are characterized by the above symmetry data vector, can be expressed as a linear combination of several elementary band representations (which are the band representations induced from atomic orbitals) with non-negative-integer coefficients. Hence,  $\text{Nb}_3\text{Br}_8$  is a topologically trivial insulator.

A topologically trivial insulator can be further classified as an atomic insulator or an OAI. If the band representation of a topological insulator cannot be induced from only the atomic orbitals at atom occupying sites, there must be some atomic orbitals (or Wannier functions) centred at the empty sites (that is, the Wyckoff positions that are not occupied by any atoms) and this topologically trivial insulator is referred to as OAI. The OAI can be indicated by the non-zero integer real space invariants at the empty sites and all the OAI on the Topological materials database (<https://www.topologicalquantumchemistry.org>)<sup>46</sup> have been exhausted by performing a high-throughput calculations<sup>38</sup>, in which  $\text{Nb}_3\text{Br}_8$  is identified as an OAI material indicated by a non-zero real space invariants at the empty Wyckoff position 3b, whose definition is

$$\delta(b) = \frac{1}{2}[-m(\bar{\Gamma}_6\bar{\Gamma}_7) + m(\bar{\Gamma}_9) + m(\bar{\Gamma}_5\bar{\Gamma}_6) - m(\bar{\Gamma}_5\bar{\Gamma}_6) + m(\bar{\Gamma}_6\bar{\Gamma}_7) - m(\bar{\Gamma}_9)]. \quad (2)$$

By substituting the symmetry data vector in equation (1) in to equation (2), we have  $\delta(b) = -1$ , which means there must have a Wannier function centred at 3b. As the positions of 3b is not occupied by any atom,  $\delta(b) = -1$  indicates that  $\text{Nb}_3\text{Br}_8$  is an OAI and the Wyckoff position 3b is referred to as the obstructed Wannier charge centre.

To demonstrate the OAI phase of  $\text{Nb}_3\text{Br}_8$ , we have also calculated the electron density distribution in the lattice of  $\text{Nb}_3\text{Br}_8$ . As the results show in Extended Data Fig. 10, the electron charge density is symmetrically centred at the Wyckoff position 3b (the inversion centre between two  $\text{Nb}_3\text{Br}_8$  layers), indicating the special separation of positive and negative charge centres along the *c* axis of the crystal. In the heterostructures of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$ , the asymmetric top and bottom interfaces break inversion symmetry of the whole junction along *c* axis. The distribution of charge centres between layers is easy to be influenced by this symmetry breaking, which could result in the charge polarization in the whole junction along *z* direction and hence show an OOP polarization. As discussed in the main text, in analogy to previous theoretical investigations of the polarized system<sup>2,36</sup>, we propose that a polarization in the  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  JJs may induce asymmetric Josephson

tunnelling and lead to the field-free JD effect; further theoretical and experimental study is necessary to fully explain the mechanism.

## Data availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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**Author contributions** H.W. and M.N.A. conceived and designed the study. C.P. grew the samples. H.W. and Y.W. fabricated the devices. H.W., Y.W., P.K.S. and U.F. performed the transport measurements. H.W. and Y.W. carried out the data analysis. Y.X. provided theoretical support and discussion. Y.-J.Z. and S.S.P.P. provided facility and instrument support. T.M. and M.N.A. are the principal investigators. All authors contributed to the preparation of the manuscript.

**Competing interests** The authors declare no competing interests.

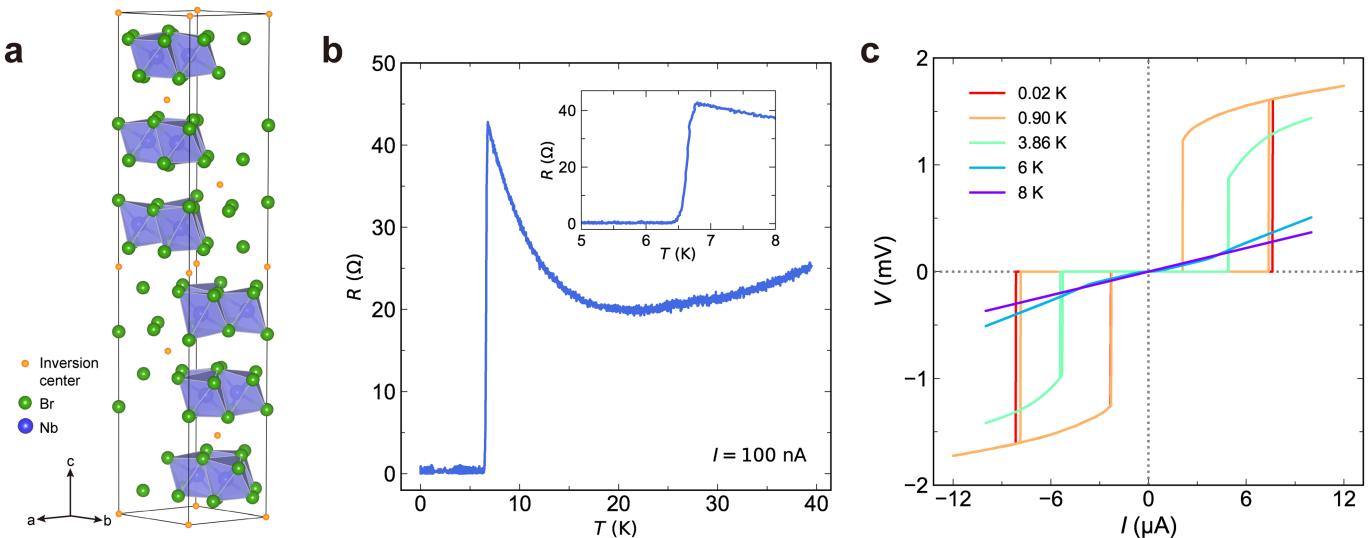
## Additional information

**Supplementary information** The online version contains supplementary material available at <https://doi.org/10.1038/s41586-022-04504-8>.

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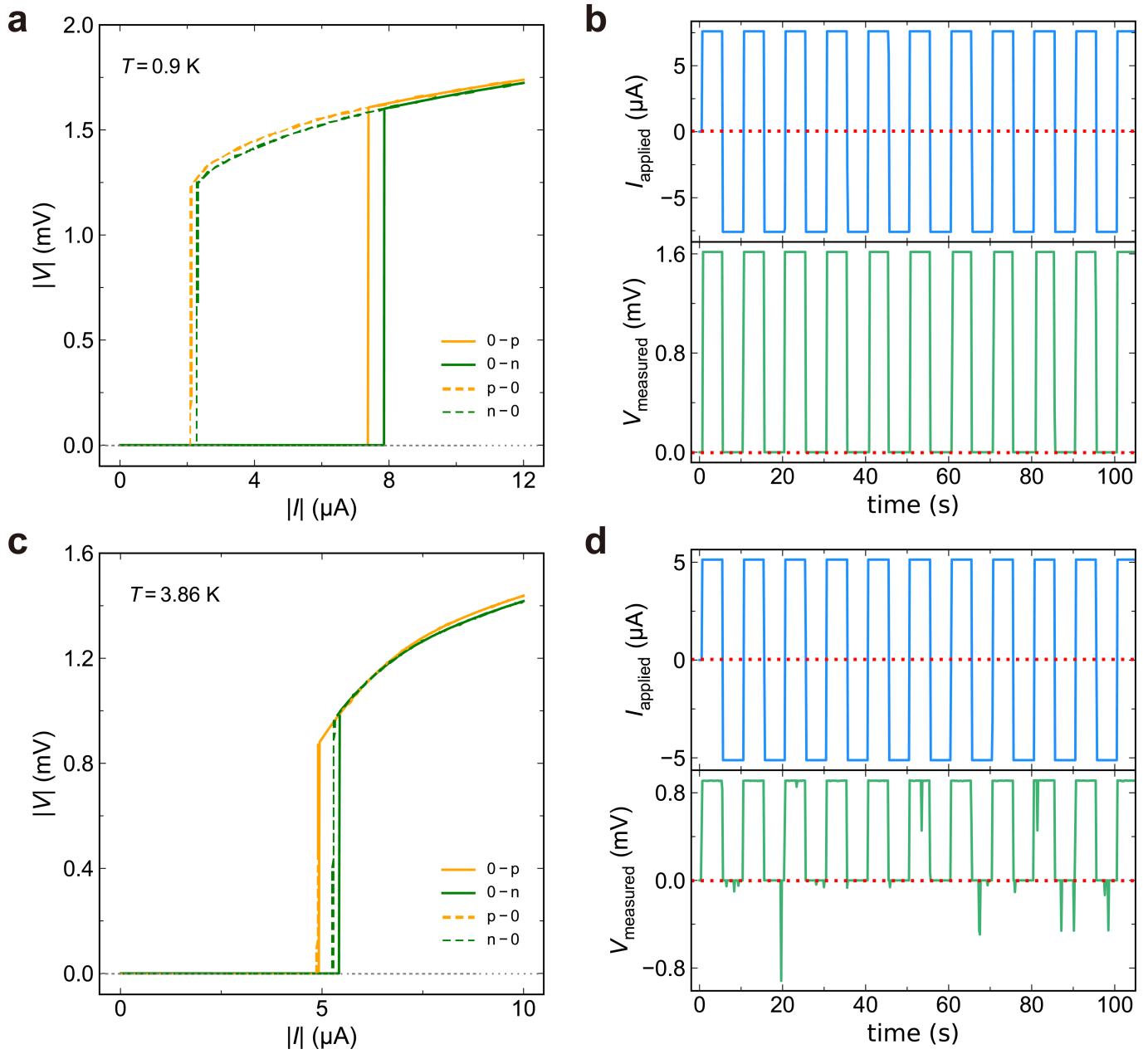
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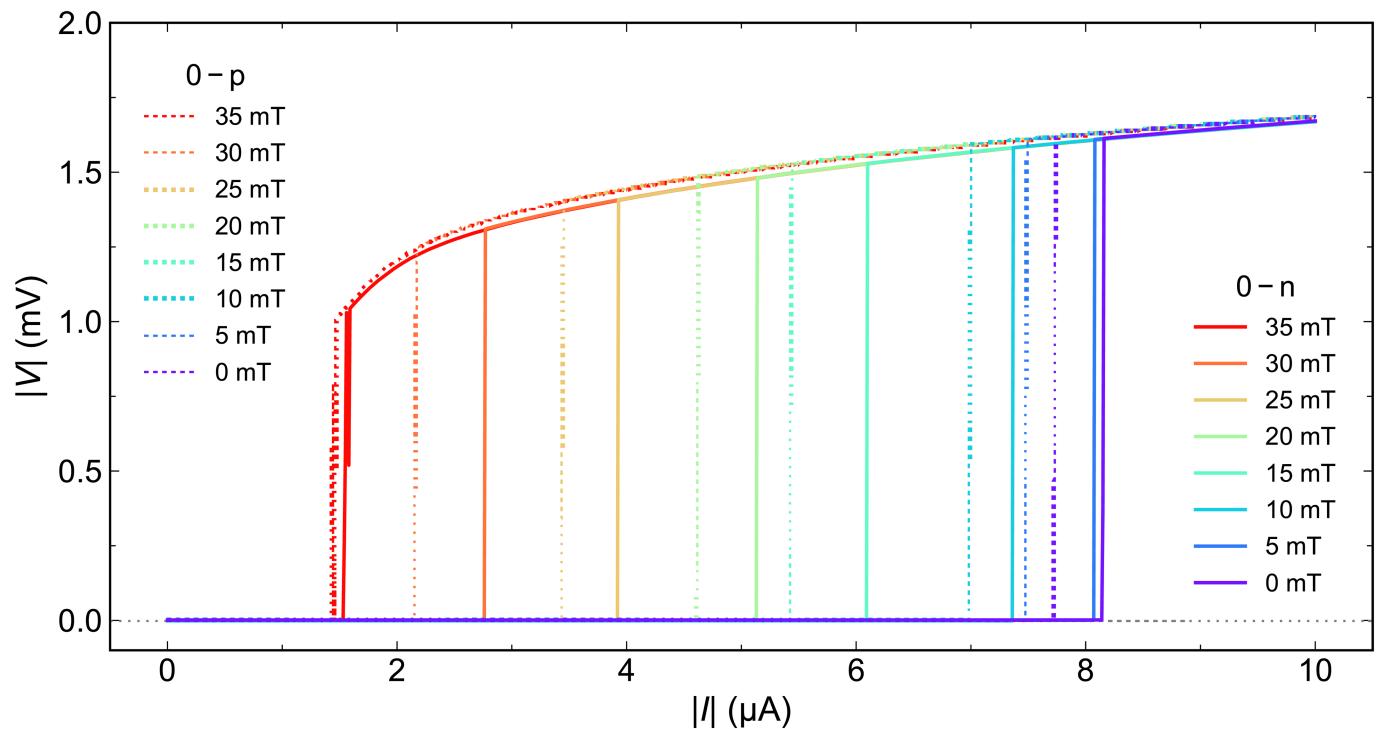
**Extended Data Fig. 1 | Temperature dependent resistance and  $V$ - $I$  curves of the Josephson diode.** **a**,  $\text{Nb}_3\text{Br}_8$ : crystal structure and inversion center locations. **b**, Resistance versus temperature measured using an a.c. current of

100 nA. Inset shows the enlarged plot near the superconducting transition of 6.6 K. **c**,  $V$ - $I$ -curves with positive sweep measured at different temperatures showing nonlinear behavior appearing below  $T_c$ .

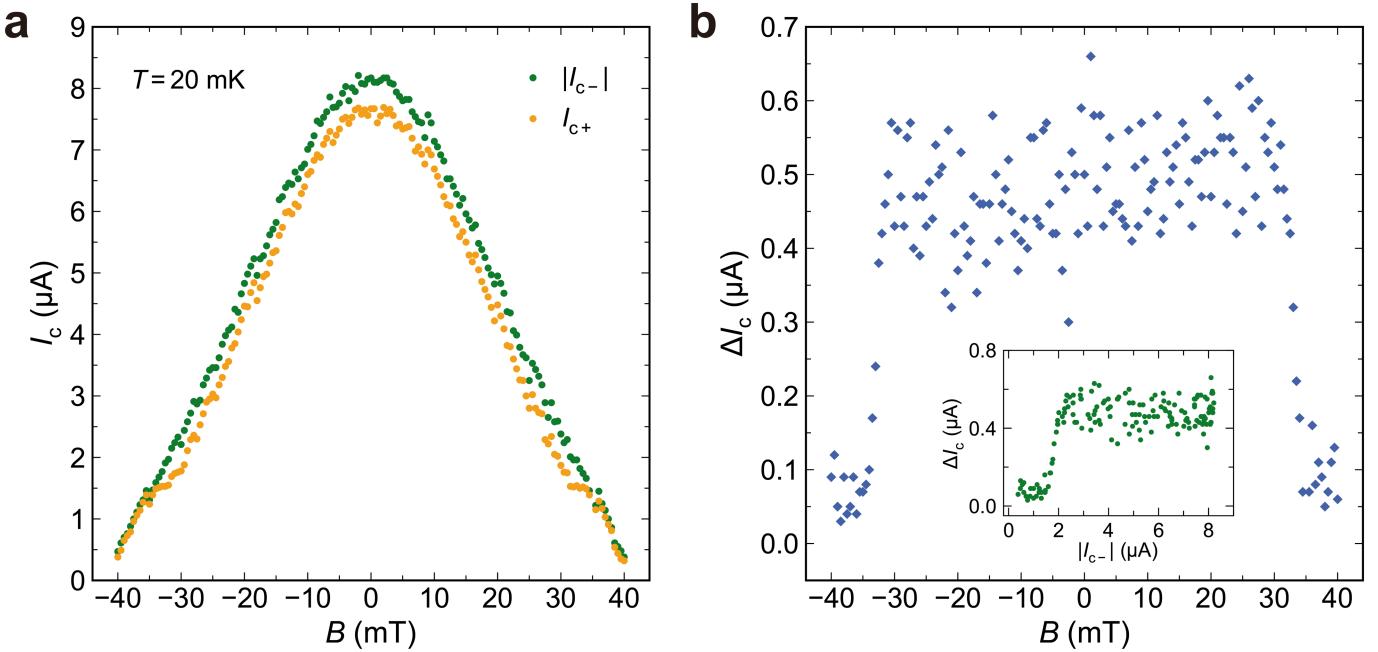


**Extended Data Fig. 2 |  $V$ - $I$  curves and half-wave rectification at different temperatures.** **a**, Positive sweep  $V$ - $I$  curve measured at 0.9 K. Both  $\Delta I_c$  and  $\Delta I_r$  are visible. **b**, Half-wave rectification measured at 0.9 K with an applied current of  $7.6 \mu$ A at 0.1 Hz. The red dotted lines are the zero lines, showing that junction is in the superconducting state with negative current and switches to the

normal state with positive current. **c**, Positive sweep  $V$ - $I$  curve measured at 3.86 K.  $\Delta I_c$  is still visible, while the hysteresis is almost completely suppressed. **d**, Half-wave rectification measured at 3.86 K with an applied current of  $5.14 \mu$ A at 0.1 Hz. The red dotted lines are the zero lines. Imperfect rectification is evident with some punch-through error, probably due to thermal fluctuation.

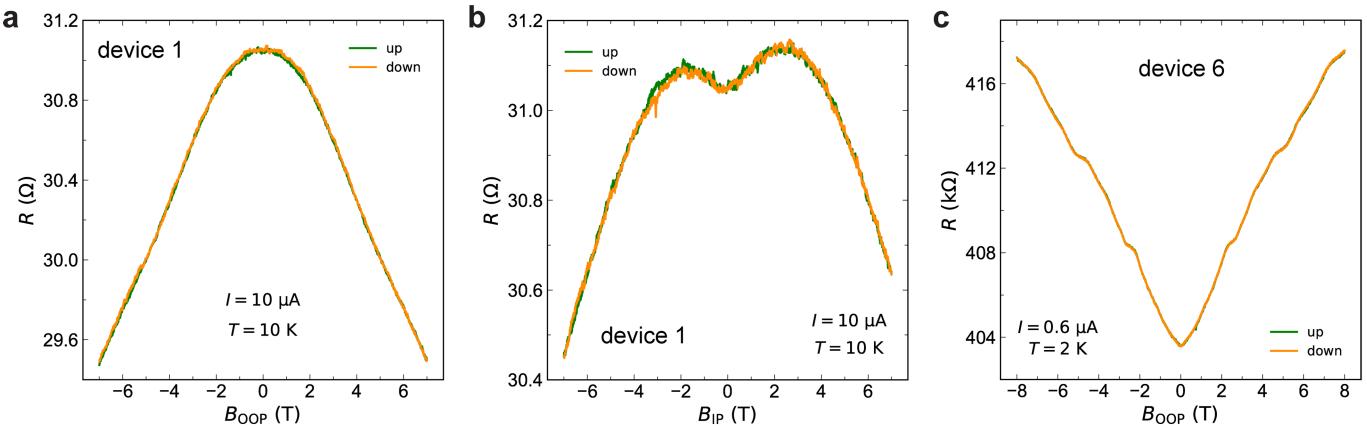


**Extended Data Fig. 3 |  $V$ - $I$  curves with 0-p and 0-n branches measured at different magnetic fields.** Solid lines are 0-n branches (where  $I_c$  was extracted) and dotted lines are 0-p branches (where  $I_{c+}$  was extracted) corresponding to Fig. 3.  $\Delta I_c$  almost ‘turns off’ at 35 mT.

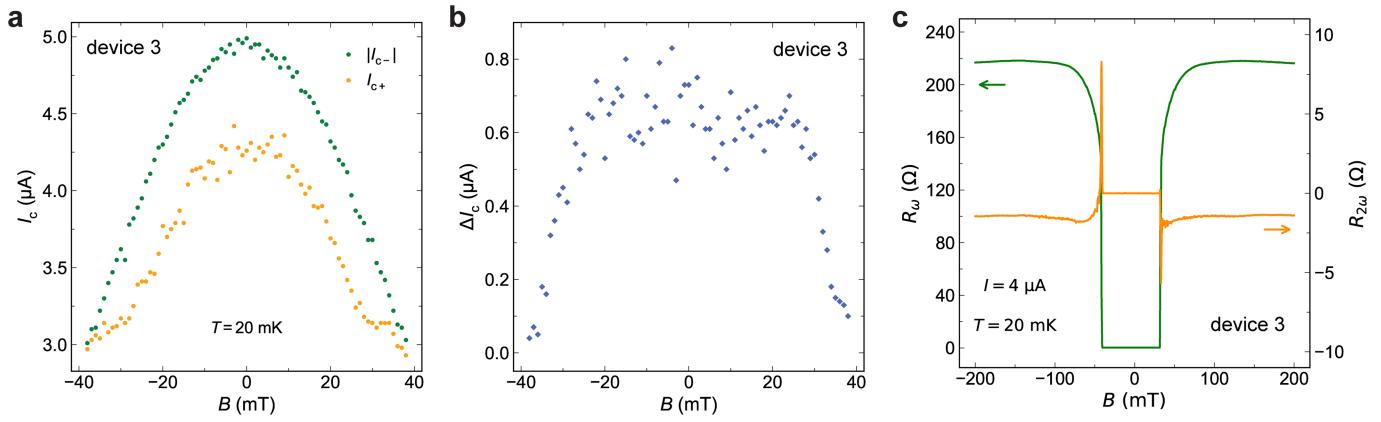


**Extended Data Fig. 4 | Sweep-up magnetic field dependence of  $I_c$  and  $\Delta I_c$ .** **a**,  $I_{c+}$  (orange dots) and  $|I_{c-}|$  (green dots) obtained from the 0-p and 0-n branches of the positive sweep as a function of applied magnetic field. The in-plane magnetic field was swept from negative to positive. **b**,  $\Delta I_c$  as a function of

magnetic field. Inset shows  $\Delta I_c$  as a function of  $|I_{c-}|$  from modulation of magnetic field with the diode effect ‘turning off’ below  $2.1 \mu\text{A}$ . These sweep-up results are nearly identical with the sweep-down results shown in Fig. 3.

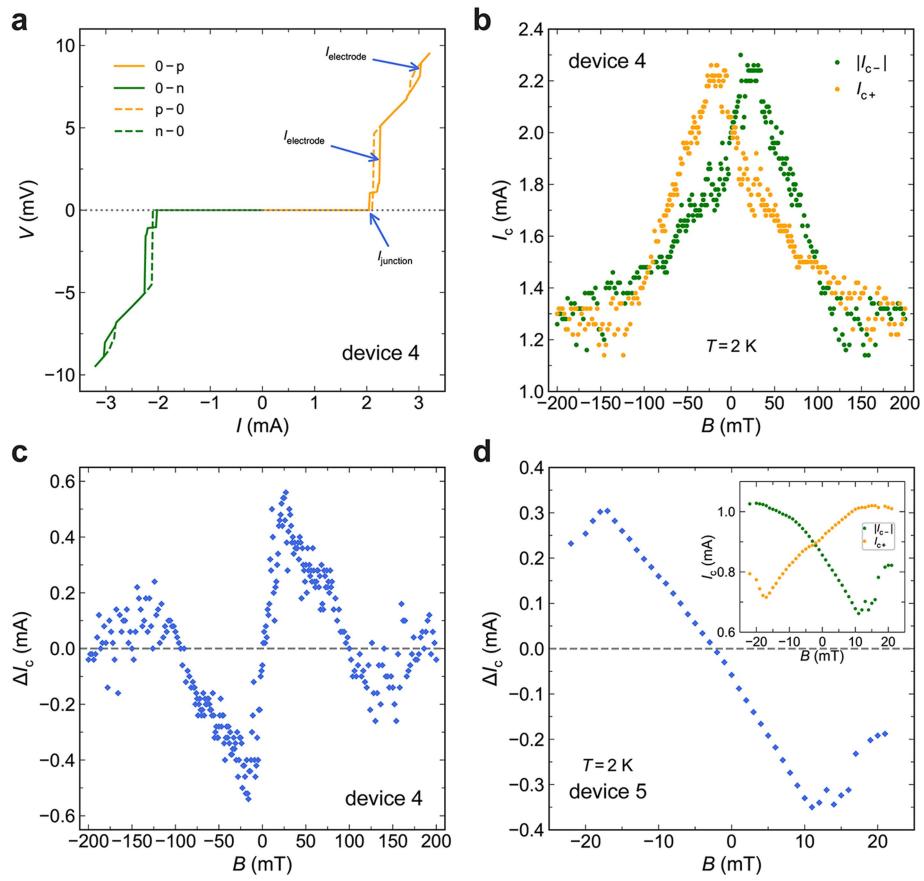


**Extended Data Fig. 5 | Magnetoresistance of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  and  $\text{FLG}/\text{Nb}_3\text{Br}_8/\text{FLG}$  heterostructures.** **a**, Sweep-up and sweep-down magnetoresistances of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  (device 1) with OOP magnetic field at 10 K (above  $T_c$  of  $\text{NbSe}_2$ ). **b**, Sweep-up and sweep-down magnetoresistances of device 1 with in-plane magnetic field at 10 K. **c**, Sweep-up and sweep-down magnetoresistance of  $\text{FLG}/\text{Nb}_3\text{Br}_8/\text{FLG}$  (device 6) with OOP magnetic field at 2 K.



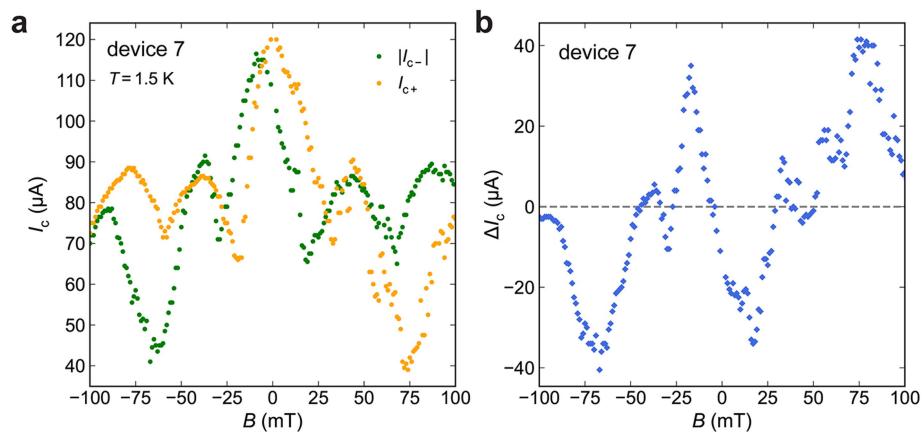
**Extended Data Fig. 6 | Field-free Josephson diode effect of a  $\text{NbSe}_2$ /four-layer  $\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  device.** **a**,  $I_c$ , and  $|I_c|$  as a function of magnetic field at 20 mK in  $\text{NbSe}_2$ /four-layer  $\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  junction (device 3). **b**,  $\Delta I_c$  as a function

of magnetic field of device 3. **c**, First harmonic ( $R_w$ ) and second-harmonic resistances ( $R_{2\omega}$ ) as a function of magnetic field measured at 20 mK with an applied current of 4  $\mu$ A of device 3.

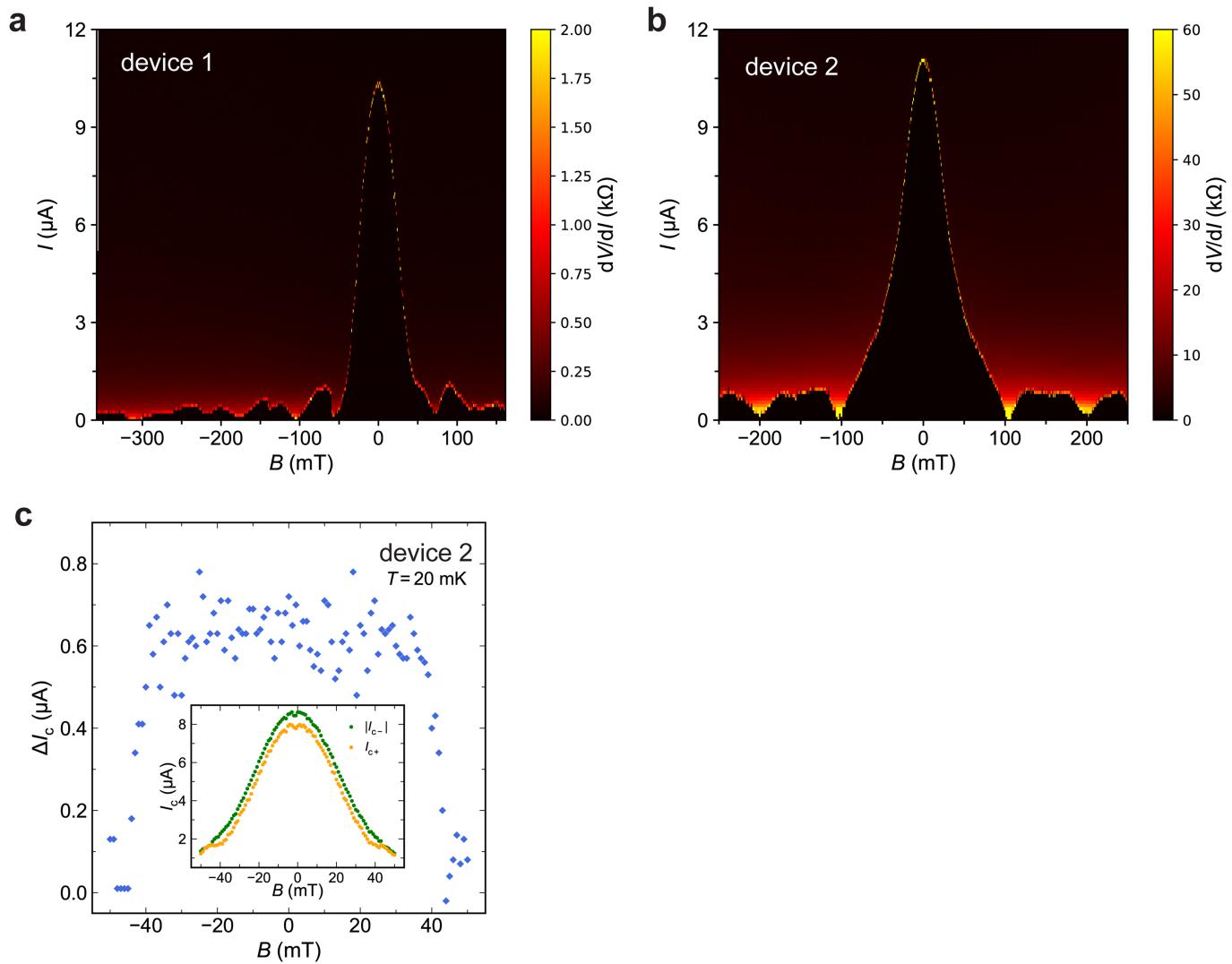


**Extended Data Fig. 7 | Field-induced superconducting diode effect in  $\text{NbSe}_2/\text{NbSe}_2$  and  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  heterostructures.** **a**,  $V$ - $I$  curve of  $\text{NbSe}_2/\text{NbSe}_2$  junction (device 4) measured at 2 K and 0 T. **b**,  $I_c$  and  $|I_c|$  as a function of

magnetic field of device 4 at 2 K. **c**,  $\Delta I_c$  as a function of magnetic field of device 4. **d**,  $\Delta I_c$  as a function of magnetic field of  $\text{NbSe}_2/\text{FLG}/\text{NbSe}_2$  junction (device 5) measured at 2 K, inset shows the corresponding  $I_{c+}$  and  $|I_{c-}|$ .

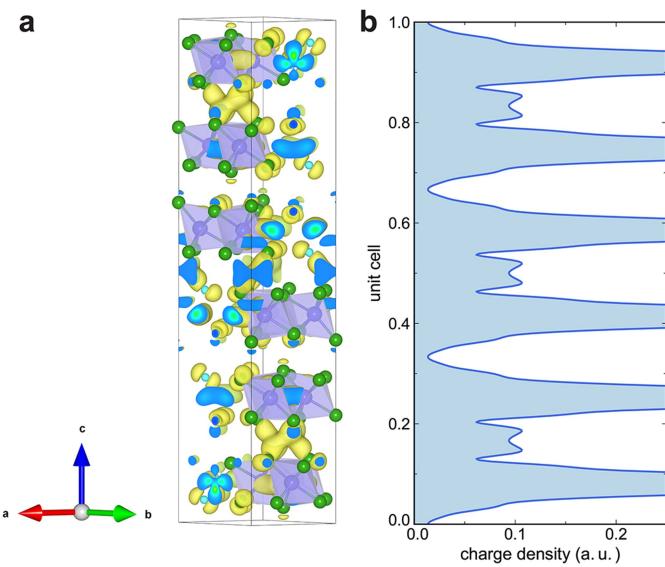


**Extended Data Fig. 8 | Field-induced superconducting diode effect in  $\text{NbSe}_2/\text{NbSe}_2$  heterostructures with a lower critical current. a.**  $I_c$  and  $|I_c|$  as a function of applied magnetic field of  $\text{NbSe}_2/\text{NbSe}_2$  junction (device 7). **b.**  $\Delta I_c$  as a function of magnetic field of device 7.



**Extended Data Fig. 9 | Fraunhofer patterns of  $\text{NbSe}_2/\text{Nb}_3\text{Br}_8/\text{NbSe}_2$  heterostructures.** **a**, Fraunhofer pattern of device 1 with sweep-down magnetic field. **b**, Fraunhofer pattern of device 2. **c**,  $\Delta I_c$  as a function of applied

magnetic field of device 2 measured at  $20\text{ mK}$ , inset shows the corresponding  $I_{c+}$  and  $|I_c|$ .



**Extended Data Fig. 10 | Obstructed atomic insulator property of  $\text{Nb}_3\text{Br}_8$ .**

**a**, charge density distribution in a  $\text{Nb}_3\text{Br}_8$  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 zero in every other van der Waals gap.