

Supplementary information for

Field-Resilient Supercurrent Diode in a Multiferroic Josephson Junction

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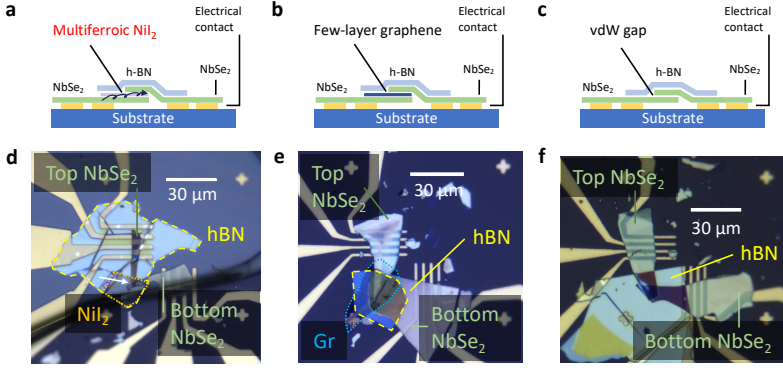
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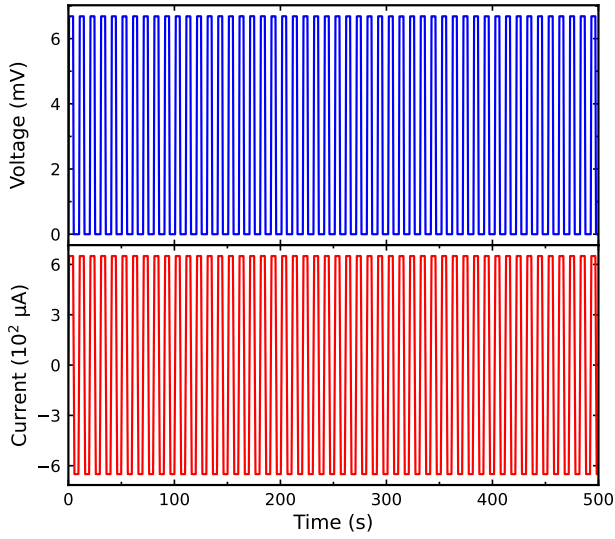
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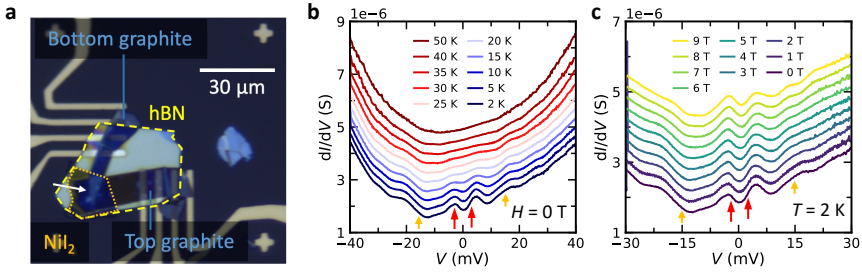
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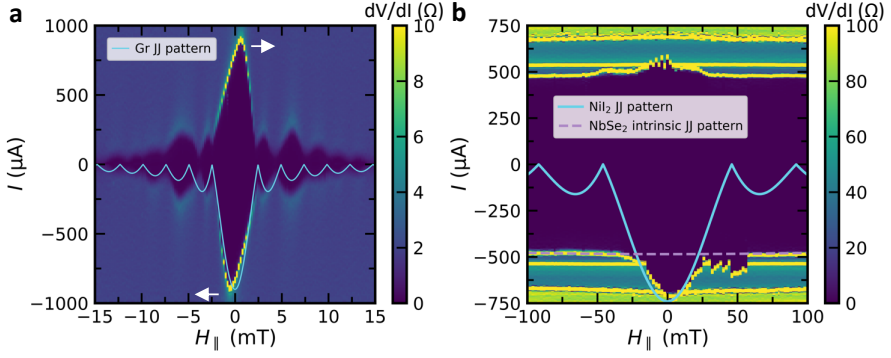
Supplementary Fig. 1 Schematics and optical images of the reported superconducting devices. a-c, Side views of NiI_2 JJ, Gr JJ, and $\text{NbSe}_2/\text{NbSe}_2$ devices. **d-f,** Optical images of NiI_2 JJ, Gr JJ, and $\text{NbSe}_2/\text{NbSe}_2$ devices. The junction area of NiI_2 JJ with a lateral dimension of $2\text{--}3\ \mu\text{m}$ is pointed out by the white arrow. The thickness of the following flakes was measured by AFM: NiI_2 in panel d: $2.8(2)\ \text{nm}$ (4ML); few-layer graphene in panel e: $4.0(2)\ \text{nm}$. The thickness of the following flakes is estimated from the thickness of the flakes with similar color contrast measured by AFM: top NbSe_2 in panel d: $\sim 20\ \text{nm}$; bottom NbSe_2 in panel d: $30\text{--}40\ \text{nm}$; top NbSe_2 flake in panel e: $\sim 20\ \text{nm}$; bottom flake in panel e: $60\text{--}70\ \text{nm}$; both NbSe_2 flakes in panel f: $\sim 30\ \text{nm}$. Multiple colors can sometimes be seen in a single flake, and the thickness above provided the best estimation of the thickness near the junction area.



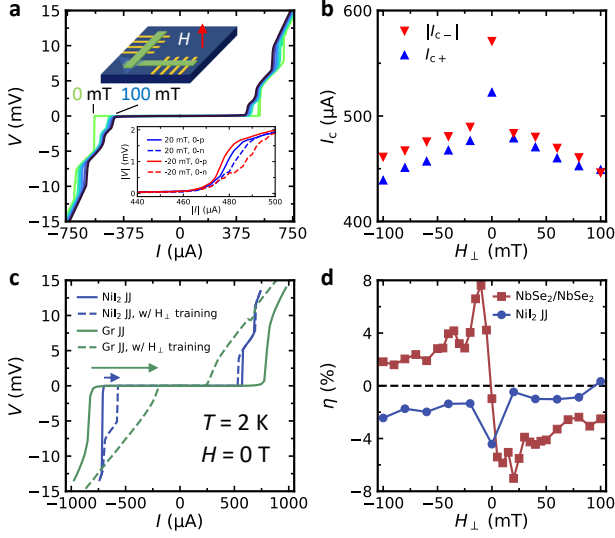
Supplementary Fig. 2 Robust supercurrent switching in the NiI_2 device. Demonstration of supercurrent switching with $I_{\text{bias}} = \pm 650 \mu\text{A}$ used in Fig. 1d of the main text for 50 cycles.



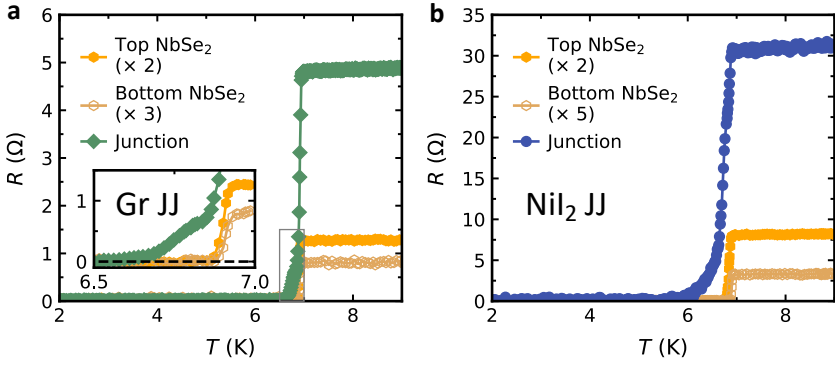
Supplementary Fig. 3 Inelastic electron tunneling experiments on the graphite/NiI₂/graphite tunnel junction. **a**, The optical image of the graphite/NiI₂/graphite tunnel junction device. The junction area is pointed out by the white arrow. The thickness of the NiI₂ flake at the junction is 10.5 nm as measured by AFM. The thickness of the top and bottom graphite flakes is about 20 nm and 40 nm based on the AFM results of other flakes of similar color contrast. **b**, Tunneling conductance v.s. dc voltage bias measured at different temperatures at zero field. The data taken at temperatures other than 2 K are shifted vertically for clarity. The red arrows ($\sim \pm 3$ -5 meV) and orange arrows ($\sim \pm 15$ meV) represent a symmetric step-like increase of tunneling conductance, potentially enhanced by collective excitations [1]. The inferred excitation energies agree with the electromagnons and magnons energies in NiI₂ as detected by Raman spectroscopy and Terahertz spectroscopy [2, 3]. The onset temperature of collective excitations agrees with the ordering temperature of NiI₂ in the thin layer limit, supporting their magnetic origin [2]. **c**, Tunneling conductance v.s. dc voltage bias measured at different in-plane magnetic fields. The data taken at fields other than 0 T are shifted vertically for clarity. The features representative of collective excitations in NiI₂ persist all the way to $H_{\parallel} = 9$ T, suggesting that the multiferroic order in thin-layer NiI₂ survives to high magnetic fields as in the bulk limit [4].



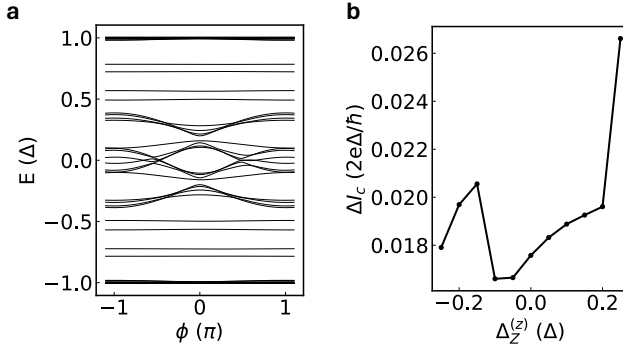
Supplementary Fig. 4 Fraunhofer interference patterns of Gr JJ and NiI₂ JJ. **a**, Critical current mapping of the Gr JJ at $T = 2$ K. The white arrows point out the tilt of the central maxima towards opposite directions due to the self-field induced by the device's cross-junction geometry (see Fig. 1 of the main text and Supplementary Fig. 1) [5–7]. The supercurrent flowing through the top and bottom NbSe₂ flakes generates current-dependent magnetic field (H_{self}) at the few-layer graphene weak link in addition to the externally applied magnetic field. In this case, the maximum of I_c appears at a finite magnetic field (instead of zero field) with which H_{self} is offset, causing the shift of maxima and the tilt of the interference pattern. The tilted pattern is directly linked to the anti-symmetric component of SDE in Gr JJ and NiI₂ JJ shown in Fig. 2 of the main text. Other than the tilt, the field dependence of the critical current agrees with the Fraunhofer interference pattern as expected from a dc Josephson effect, $I_c(H) = I_c(0)|\sin(\pi\Phi/\Phi_0)/(\pi\Phi/\Phi_0)| = I_c(0)|\text{sinc}(\Phi/\Phi_0)|$, where Φ_0 is the magnetic flux quantum, and Φ is the magnetic flux going through the JJ [8]. Typically, Φ is calculated as $H_{\parallel} \times (d + 2\lambda) \times W$, where d is the separation between the two superconducting electrodes, λ is the London penetration depth of the superconductor, and W is the lateral width of the junction. Here, we use $\Phi = H_{\parallel} \times d_{\text{eff}} \times W$ to model the interference patterns in our vdW JJs. With W determined from the optical image of the device, the simulated curve (cyan line) showing a good agreement is generated with $d_{\text{eff}} = 40$ nm, similar to other vdW JJs reported in the literature [9–11]. We also note that if the flux focusing effect [12] plays a role, a slightly modified expression $I_c(H) = I_c(0)|\text{sinc}(\Gamma\Phi/\Phi_0)|$ with $\Gamma = 3$ and $d_{\text{eff}} = 14$ nm can also lead to a decent agreement between the observed and simulated patterns. **b**, Critical current mapping of the NiI₂ JJ at $T = 2$ K. At low fields, a typical interference pattern ($I_c(0)|\text{sinc}(\Phi/\Phi_0)|$) as expected from the NiI₂ JJ was observed (cyan solid line, $W = 3$ μm and $d_{\text{eff}} = 15$ nm). At high fields, the pattern was truncated by the other interference pattern caused the interlayer Josephson coupling between each layer within NbSe₂ (purple dashed line). The coexistence of both types of interference patterns was also observed in artificially engineered multilayer JJ [13] and twisted Bi₂Sr₂CaCu₂O_{8+x}/Bi₂Sr₂CaCu₂O_{8+x} vdW JJ [14] as a result of two JJs in series. In general, in vdW JJs, the Josephson coupling may be established between 1) the vdW gaps within each superconducting flake, and 2) the weak link between the top and bottom flakes. We note that in our work, such a behavior is present in the NiI₂ JJ but not in the Gr JJ, most likely due to the difference of Josephson penetration depths λ_J (~ 10 μm for the NiI₂ JJ and ~ 400 μm for the Gr JJ) compared to the junction's lateral width w (about several μm) [15]. The huge difference between the λ_J of Gr JJ and w makes the Josephson coupling across the few-layer graphene the dominant one, while the proximity between λ_J of NiI₂ JJ and w makes it less dominant and potentially comparable with the intrinsic Josephson coupling within NbSe₂ flakes, creating a two-JJs-in-series scenario and the coexistence of two interference patterns.



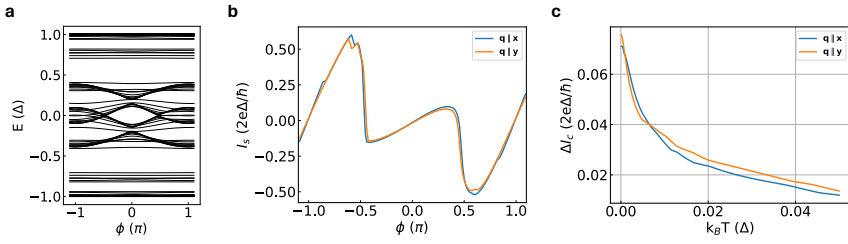
Supplementary Fig. 5 Out-of-plane magnetic field dependence of supercurrent diode effect in the NiI_2 JJ. **a**, $V - I$ characteristic of the NiI_2 JJ with $0 \text{ mT} < H_\perp < 100 \text{ mT}$, with a 20 mT field increment at $T = 2$ K. Inset: Representative switching curves at $H_\perp = \pm 20 \text{ mT}$ showing negative supercurrent diode effect in both cases. **b**, Critical current I_{c+} and $|I_{c-}|$ as a function of $H_\perp$. At zero field, it is noted that the ΔI_c as deduced from panel b is smaller than the corresponding zero-field value from Fig. 2b of the main text, due to the reduction of critical current when the measurement is taken after an out-of-plane field history. **c**, $V - I$ characteristic of NiI_2 JJ and Gr JJ with and without an $H_\perp$ training history. The short blue and long green arrows show the reduction of I_{c-} after an $H_\perp$ training for NiI_2 JJ (-100 mT) and Gr JJ (-10 mT), respectively. The reduction is likely due to trapped Abrikosov vortices induced by $H_\perp$ in the NbSe_2 electrodes [?], different from the Josephson vortices induced by $H_\parallel$. **d**, Comparison of η as a function of $H_\perp$ between the $\text{NbSe}_2/\text{NbSe}_2$ junction and the NiI_2 JJ. The $\text{NbSe}_2/\text{NbSe}_2$ homostructure can be viewed as a superconductor with enhanced inversion symmetry breaking by stacking two NbSe_2 flakes. It shows an SDE with anti-symmetric out-of-plane field dependence, as expected from superconductors without inversion symmetry [17]. The NiI_2 JJ defies this general trend and shows a predominantly symmetric field dependence with a persistent negative η .



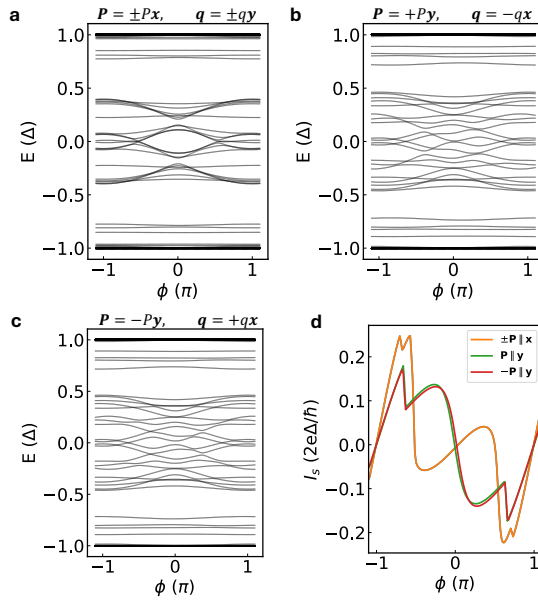
Supplementary Fig. 6 Temperature dependence of the resistance across the junction and flakes. **a**, Four-terminal resistance of the top NbSe₂ flake, the bottom NbSe₂ flake, and across the junction in the Gr JJ. The inset shows the resistance near the superconducting transition of NbSe₂ (marked by the gray square). **b**, Four-terminal resistance of top NbSe₂ flake, bottom NbSe₂ flake, and across the junction in the NiI₂ JJ. In both JJs, the resistance of both top and bottom NbSe₂ flakes sharply drops to zero at the transition temperature ~ 7 K [18], while the junction resistance drops to zero at a lower temperature, consistent with the typical behavior of JJs [19, 20]. It is noted that the resistance of both JJs decreases monotonically as the temperature approaches the transition temperature, suggesting the high quality of our device compared to a non-homogeneous superconductor which would show an anomalous peak above the transition temperature [21].



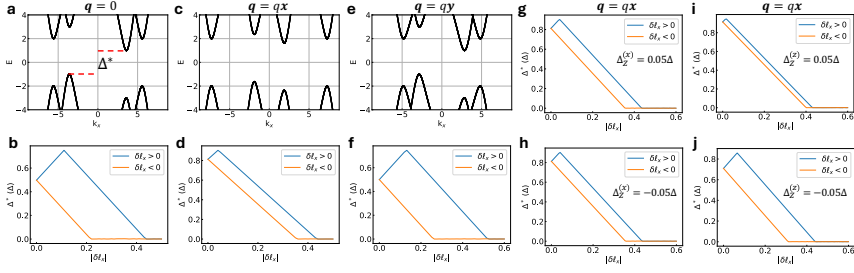
Supplementary Fig. 7 Additional simulations of a helimagnet JJ. **a**, Andreev bound state spectrum of the helimagnet JJ with CPR corresponding to Fig. 4c of the main text with RSOC for $\mathbf{q} \parallel \mathbf{x}$. **b**, ΔI_c as a function of $\Delta_Z^{(z)}$ (external Zeeman spin-splitting along $\mathbf{z}$ in Fig. 4a of the main text) with a mixed even-odd functional dependence. Parameters used in simulations are: $\Delta = 0.4t$, $\mu = 1.57t$, $\alpha_R = 0.004ta$, $J_{exc} = 0.3t$, $U_{barrier} = 4t$, $|\mathbf{q}| = 0.01 \frac{\pi}{a}$, $L_{x,s} = 300a$, $L_{x,n} = 3a$, $L_y = 10a$, and $\xi_{exc} = 5a$ where $t = \frac{\hbar^2}{2m^*a^2}$ and a is the tight binding lattice constant.



Supplementary Fig. 8 Simulation of a wide helimagnet JJ. Simulation of a wide helimagnet JJ with $L_y = 20a$. **a**, Andreev bound state spectrum for $\mathbf{q} \parallel \mathbf{x}$ with RSOC. **b**, Current-phase relationships. and **c**, Temperature dependence of ΔI_c .



Supplementary Fig. 9 Simulations of multiferroic JJs. **a**, Andreev bound state spectra of multiferroic JJs for $\mathbf{P} = \pm P\mathbf{x}$ and **b-c** $\mathbf{P} = \pm P\mathbf{y}$ with $\mathbf{P} \times \mathbf{q} \parallel +\mathbf{z}$ (see Fig. 4a of the main text). **d**, CPRs of multiferroic JJs calculated from their corresponding spectra shown in panel **a-c**.



Supplementary Fig. 10 Single- ℓ helical superconductivity with a helimagnet. Dispersion with $\ell_x = \ell_{x,0}$ and gap dependence on depairing momentum $\delta\ell_x = \ell_{x,0} - \ell_x$, where $\ell_{x,0}$ is the equilibrium momentum, for a **a-b**, helical Rashba superconductor, **c-f**, helimagnet superconductor with Rashba spin-orbit coupling. **g-h**, Spectral gap suppression with depairing momentum for positive (top) and negative (bottom) Zeeman splitting along the x-direction. **i-j**, Spectral gap suppression with depairing momentum for positive (top) and negative (bottom) Zeeman splitting along the z-direction.

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