

Switchable geometric frustration in an artificial-spin-ice–superconductor heterosystem

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Geometric frustration emerges when local interaction energies in an ordered lattice structure cannot be simultaneously minimized, resulting in a large number of degenerate states. The numerous degenerate configurations may lead to practical applications in microelectronics¹, such as data storage, memory and logic². However, it is difficult to achieve very high degeneracy, especially in a two-dimensional system^{3,4}. Here, we showcase in situ controllable geometric frustration with high degeneracy in a two-dimensional flux-quantum system. We create this in a superconducting thin film placed underneath a reconfigurable artificial-spin-ice structure⁵. The tunable magnetic charges in the artificial-spin-ice strongly interact with the flux quanta in the superconductor, enabling switching between frustrated and crystallized flux quanta states. The different states have measurable effects on the superconducting critical current profile, which can be reconfigured by precise selection of the spin-ice magnetic state through the application of an external magnetic field. We demonstrate the applicability of these effects by realizing a reprogrammable flux quanta diode. The tailoring of the energy landscape of interacting ‘particles’ using artificial-spin-ices provides a new paradigm for the design of geometric frustration, which could illuminate a path to control new functionalities in other material systems, such as magnetic skyrmions⁶, electrons and holes in two-dimensional materials^{7,8}, and topological insulators⁹, as well as colloids in soft materials^{10–13}.

Flux quanta in a superconductor are quantized magnetic flux bundles, each containing a single flux quantum sustained by circulating superconducting currents. Their motion determines the entire electromagnetic behaviour of a superconductor¹⁴. Controlling the distribution and motion of flux quanta is important for enhancing the superconductor's current-carrying capacity and developing innovative superconducting electronic devices¹⁵. Previous research demonstrates that the flux-quantum system can be a unique platform to investigate geometric frustration^{16–19}, with certain clear advantages over other widely investigated systems, such as artificial-spin-ices (ASIs) of nanoscale bar magnets^{2,3,5,9,20–27}, colloidal ices of microscopic magnetic particles^{10,11} and buckled non-magnetic colloidal monolayers¹². In the ice-analogue-of-flux-quanta system, a pair of closely placed pinning potentials, such as nanopatterned holes, in a superconducting film provide two possible locations for a flux quantum, mimicking the binary degrees of freedom of a ‘spin’ in an ASI system^{16–19}. Both theoretical¹⁷ and experimental^{17,18} studies

have shown that strong interactions between flux quanta could drive the system into the ground state more readily. Analogous to colloidal ices^{10–13}, flux-quantum ices are ‘particle’-based ices exhibiting rich phenomena that are absent in ASI; for example, ice-rule fragility, topological charge transfer and glassiness^{28,29}. However, as is the case for the two-dimensional (2D) ASI^{22–27}, the ground-state configurations of the flux-quantum ice systems are all characterized by twofold degenerate long-range ordering^{17–19}, and so far, a highly degenerate state has not been realized in such systems.

Extensive degeneracy in the ASI system, comprising square lattices of nanoscale bar magnets, has been achieved recently by vertically separating two sublattices of orthogonally orientated bar magnets³, thereby creating a three-dimensional structure. To realize a highly degenerate geometric frustration in a 2D flux-quantum system, we impose a magnetically charged landscape onto a superconductor. This landscape is derived from a recently introduced tri-axial ASI structure consisting of three sublattices of nanomagnetic bars in a square lattice (Supplementary Fig. 1a)⁵, and is very effective in controlling the position of flux quanta in a superconducting thin film (Fig. 1a). The tri-axial ASI produces three types (types I, II and III; Fig. 1c–e) of magnetic-charge order out of eight possible charge configurations (Supplementary Fig. 1b–i). The magnetic-charge configurations can be conveniently obtained by applying an in-plane magnetic field (see Methods). The ground-state type I magnetic-charge order consists of two sublattices of plaquettes (M and N in Fig. 1b)—one rotated by 90° from the other. This is similar to the arrangement of the orthogonally orientated bar magnets in the three-dimensional ASI³, but in a planar 2D structure. Depending on the polarity of the applied magnetic field (that is, pointing in or out of the superconducting film), a magnetic charge can act as an attractive or repulsive potential barrier for the flux quanta (Fig. 1a)³⁰. The type I magnetic charges in each plaquette (Fig. 1b) produce an ice-like ‘two attractive, two repulsive’ potential centre for the flux quanta, and enable the realization of a true 2D flux-quantum system that possesses geometric frustration with extensive degeneracy. More significantly, our ASI–superconductor structure allows us to switch the geometric frustration on and off in situ in a unique way that cannot be realized in a pure ASI or superconductor structure (as demonstrated below).

The flux quanta in a superconducting film can be driven into motion by an electric current (Fig. 1a), enabling dynamics studies of geometric frustration through magneto-transport experiments^{18,19}. In Fig. 1f–h, we show colour-coded maps of the magnetic-field and

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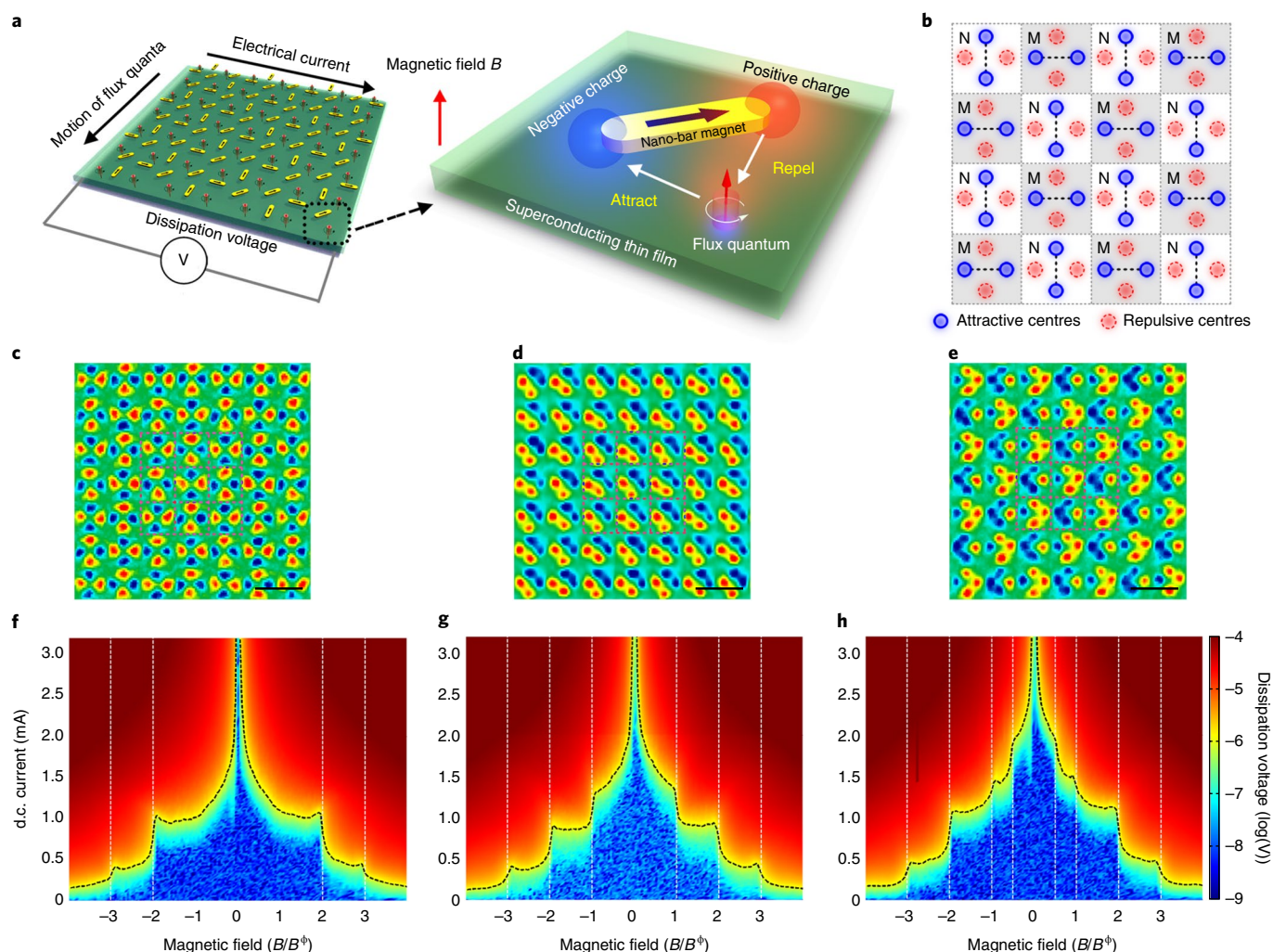


Fig. 1 | Reconfigurable potential landscapes and flux-quantum motion controlled by ASI. **a**, Heterostructure consisting of a reconfigurable ASI (interacting nanoscale bar magnets) on top of a superconducting thin film. The dashed arrow points to a magnified image delineating the interaction between a flux quantum (pointing up) in the superconducting layer and the magnetic dipole charges. The attractive and repulsive interactions will be reversed for a flux quantum pointing down. **b**, Two sublattices of type I magnetic-charge order. The red and blue circles represent positive and negative magnetic charges, respectively. **c–e**, MFM images of magnetic-charge orders of types I (**c**), II (**d**) and III (**e**), obtained from the fabricated ASI pattern (Supplementary Fig. 1a) of the heterostructure. The dashed lines show the 3×3 square lattice of the charges. Scale bars, $1 \mu\text{m}$. **f–h**, Colour maps of the magnetic-field and current-dependent voltages associated with magnetic-charge orders of types I (**f**), II (**g**) and III (**h**). The black dashed curves indicate the superconducting critical currents defined by a voltage criterion of $1 \mu\text{V}$. The magnetic-field unit $B^\Phi = 40 \text{ Oe}$ corresponds to a flux density of one flux quantum per square plaquette.

current-dependent dissipation voltage induced by the motion of flux quanta. Supplementary Fig. 2 shows maps associated with all eight possible magnetic-charge configurations. The superconducting critical current (black dashed line), defined as the maximum current a superconductor can carry without energy dissipation, exhibits clear kinks at certain magnetic fields, as indicated by the vertical white lines in Fig. 1f–h. These are typical features of field-matching effects^{18,19,31} originating from collective energy minimization when the flux quanta form a crystallized lattice commensurate with an ordered lattice of pinning potentials. Here, the flux-quantum density m is determined by the magnetic field B through the relationship $m = B/\Phi_0$, where $\Phi_0 = 2.07 \times 10^{-15} \text{ Wb}$ is the magnetic flux carried by a single flux quantum. The first matching field is $B/B^\Phi = 1$, where $B^\Phi = 40 \text{ Oe}$ corresponds to a flux-quantum density of one flux quantum per plaquette for our sample. Note that while kinks are observed at all integers under the type II and III charge configurations, the matching effect is missing at $B/B^\Phi = 1$ under the type I charge configuration, indicating a disordered arrangement of flux quanta.

To obtain further insight into the flux-quantum matching effect and determine the distribution of flux quanta, we carried out extensive molecular dynamics simulations (see Methods). Since the observed flux-quantum matching effect is determined by magnetic-charge patterns (see Supplementary Fig. 3), we can simplify our magnetic/superconducting system with a model based on magnetic charges interacting directly with flux quanta. The simulated critical current curves (Supplementary Fig. 4) reproduce all the matching features shown in our experiments. Furthermore, the simulated flux-quantum distributions (Fig. 2 and Supplementary Fig. 5) confirm that the flux quanta crystallize into ordered states at all the magnetic fields where the kinks are present, while they show no long-range order under the type I charge configuration at $B/B^\Phi = 1$. Figure 2 also shows that all the flux quanta sit at the positions of attractive potential centres (blue circles) under type I charge configurations. For an arbitrary pair of attractive potential centres, we denote the energetically favourable flux-quantum filling configuration ‘one filled, one empty’ with a twofold degeneracy

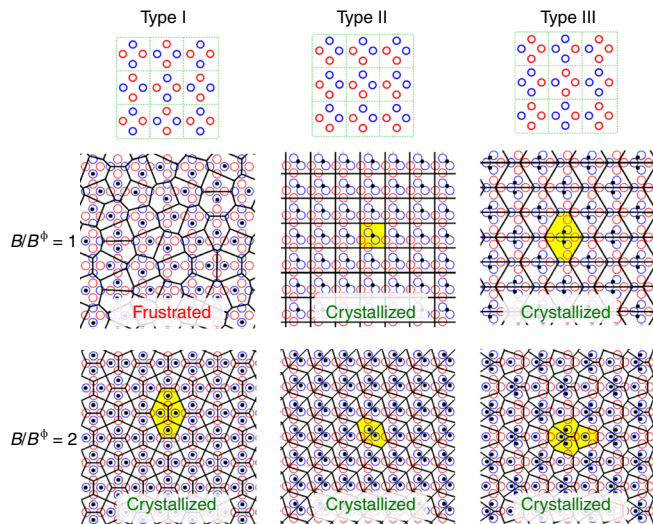


Fig. 2 | Switchable geometric frustration and crystallization. Simulated distributions of flux quanta under type I, II and III magnetic-charge orders and various flux-quantum densities. Solid black lines constitute Voronoi diagrams elucidating the flux-quantum ordering. One repeating unit structure in each of the crystallized flux-quantum lattices is highlighted in yellow. The disordered flux quanta under the type I magnetic-charge configuration at $B/B^\Phi = 1$ originate from geometric frustration with very high degeneracy.

as a ‘favourable pair’ in Fig. 3a, and the ‘two-empty’ configuration with a higher flux-quantum–magnetic-charge interaction energy and the ‘two-filled’ configuration with a higher inter-flux-quantum

interaction energy as ‘frustrated pairs’. Our simulation for $B/B^\Phi = 1$ indicates that the favourable pairs of nearest-neighbour attractive centres (α pairs defined in Fig. 3b) within each plaquette are systematically ubiquitous with population up to 80%. When favourable α pairs form, there is no way to simultaneously satisfy all the β pairs in the favourable configuration (Fig. 3b), resulting in geometric frustration between the nearest-neighbour M–N plaquettes and a twofold degeneracy within each plaquette.

In addition to the frustrated local interactions, the breakdown of long-range order is required to obtain very high degeneracy in a geometrically frustrated system²¹. In Fig. 3c–e, we compare the populations of favourable and frustrated pairwise configurations (see Fig. 3b for a definition of α , β and γ pairs) with those of the non-interacting flux quanta obtained from the magnetic-field-dependent population rate formula listed in Fig. 3a. The results show that the ‘one-filled, one-empty’ configuration is strongly favoured over a wide range of magnetic fields centred to $B/B^\Phi = 1$ for the α and β pairs (Fig. 3c,d, respectively). In contrast, for the γ pairs, the favourable and frustrated pairwise populations nearly overlap with those of the non-interactive pairs over the entire magnetic-field range, indicating uncorrelated γ pairs with random filling. We found that the repulsive potential centres (red circles in Figs. 2 and 3b) play an important role in breaking down the γ pairwise correlation, as indicated by the appearance of highly favourable γ pairs and the emergence of long-range ordered flux quanta when the repulsive potential centres are removed (see Supplementary Fig. 6). Thus, the frustrated local correlations and the breakdown of long-range order, respectively governed by attractive and repulsive potential centres, lead to a high degeneracy of 2^n -fold (with n being the number of square plaquettes) in the entire 2D square lattice.

Since the flux-quantum density at $B/B^\Phi = 1$ corresponds to one flux quantum per plaquette, equal numbers of the two different frustrated plaquettes (‘two-filled’ and ‘two-empty’ α pairs) must emerge

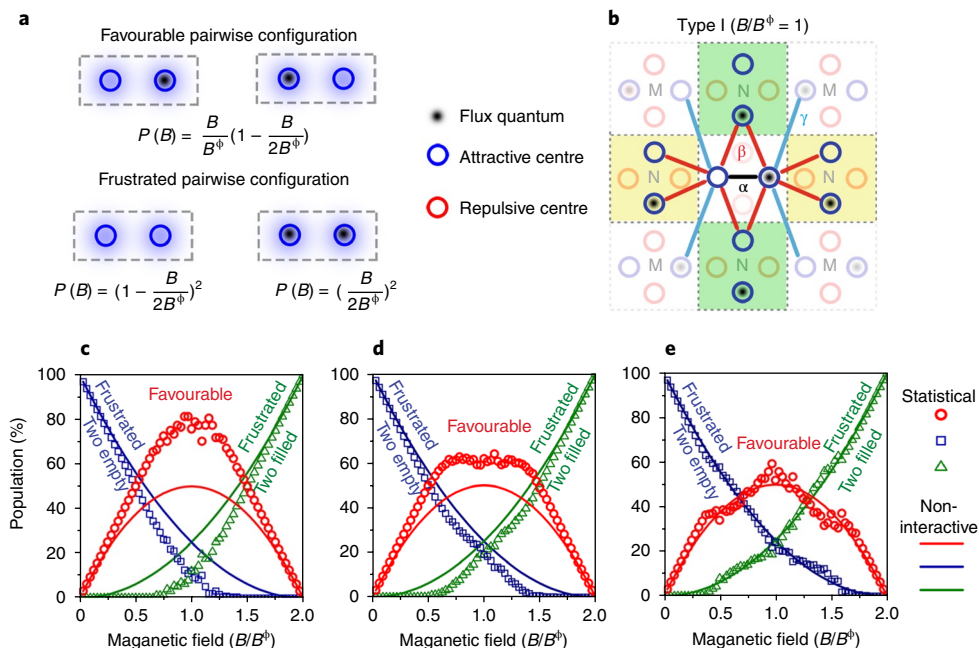


Fig. 3 | Geometrically frustrated local interactions and breakdown of long-range order. **a**, Flux-quantum filling configurations for a pair of attractive potential centres. The magnetic-field-dependent population rate $P(B)$ of each configuration for non-interacting flux quanta is listed below each configuration, where $B^\Phi = 40$ Oe corresponds to one flux quantum per plaquette (each plaquette contains two attractive potential centres). **b**, Illustration of α , β and γ pairs of attractive centres under the type I charge order. Switching the flux quantum between the two attractive centres in an M (or N) plaquette does not change the interaction energy with the two green N (or M) plaquettes, and cannot simultaneously minimize the energy with the two yellow N (or M) plaquettes. This leads to a twofold degeneracy in each plaquette. **c–e**, Magnetic-field-dependent populations (open symbols) of pairwise flux-quantum filling configurations for the α (**c**), β (**d**) and γ (**e**) pairs. The solid lines represent populations for non-interacting flux quanta calculated from the formula listed in **a**.

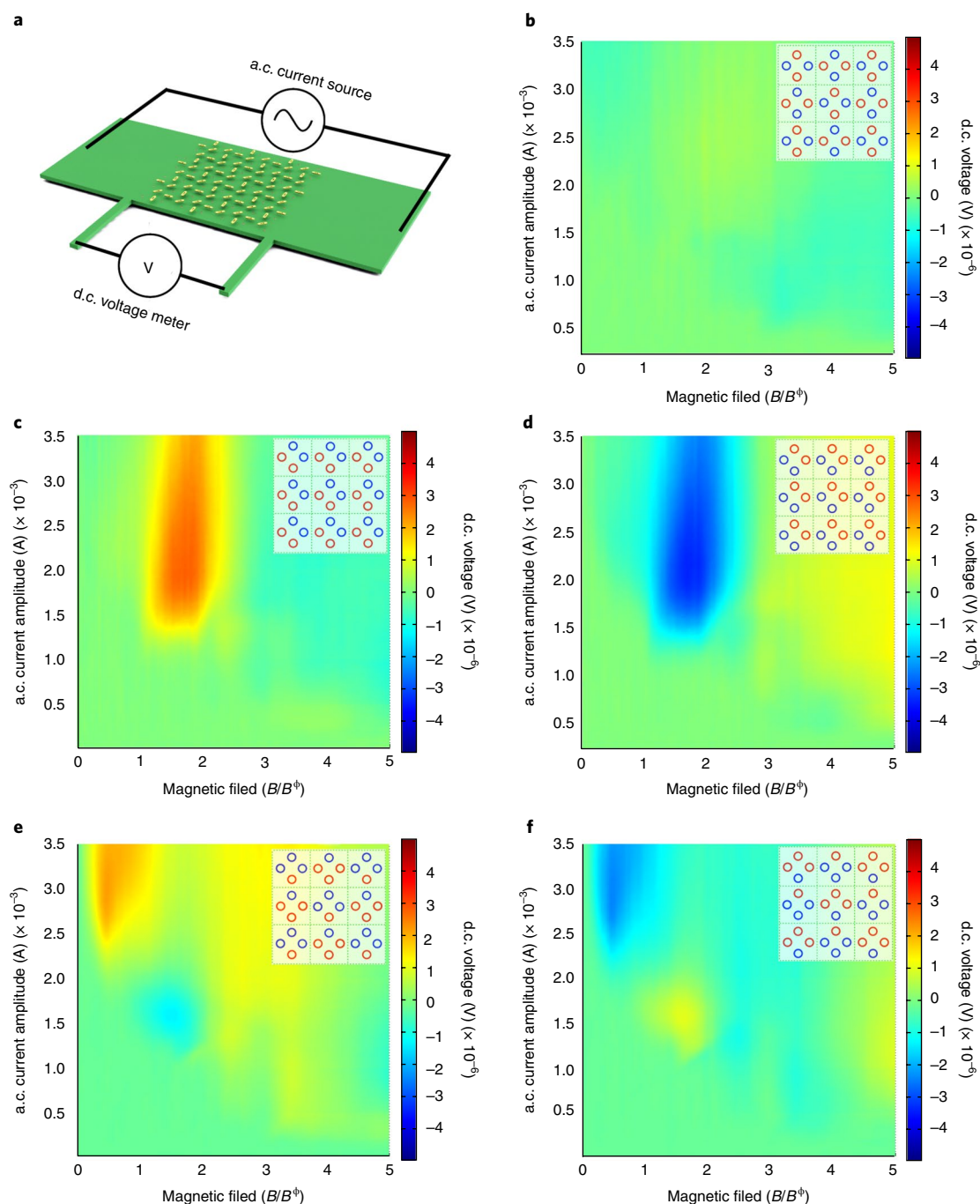


Fig. 4 | Reprogrammable flux-quantum rectifier. **a**, Schematic of the experimental set-up with a.c. current drive and d.c. voltage detection. **b–f**, Colour maps of the d.c. voltage response associated with rectification as a function of the a.c. current amplitude and magnetic field under the type I (**b**), II (**c** and **d**) and III (**e** and **f**) magnetic-charge orders shown in the insets. The magnetic charges in **d** and **f** are reversed (positive to negative and vice versa) compared with those in **c** and **e**, respectively.

simultaneously. These frustrated plaquettes can be assigned a positive ('two-filled' α pair) or negative ('two-empty' α pair) net flux-quantum value with respect to the background of one flux quantum per plaquette, resembling the simultaneous emergence of positive and negative magnetic monopole pairs connected by Dirac strings in a magnetic spin-ice system³². Therefore, 'Dirac strings' connecting pairs of 'two-filled' and 'two-empty' frustrated plaquettes can also be designed in our flux-quantum system, as demonstrated in Supplementary Fig. 7. Though the emergence of frustrated plaquettes (frustrated α pairs) reduces the degenerate manifold, the

location of these defects provides a new degree of freedom, thus inducing massively degenerate states for excited defects.

The combination of ASI and superconductor not only enables the realization of geometric frustration with very high degeneracy, but it also allows reprogrammability of frustration and transport properties that cannot be achieved in either a pure ASI or a superconductor. Figure 2a indicates two ways of switching flux quanta between geometrically frustrated and crystallized states: (1) by tuning flux-quantum density by magnetic field; and (2) by switching the potential landscape by reconfiguring the magnetic-charge

orders. The first method enables easily controlled defect formation and tunable geometric frustration by the external magnetic field (Supplementary Fig. 8), which is not accessible in ASI. This method is straightforward in flux-quantum systems. Though it can also be applied to a nanopatterned superconductor, the attractive pinning potential in nanopatterned superconductors is determined by the lithographic pattern and cannot be changed once fabricated. In contrast, the magnetic pinning with both attractive and repulsive potentials in the ASI-superconductor is highly tunable, leading to a second method of magnetically switching the potential landscapes with reconfigurable magnetic-charge orders. This method is unique to our ASI-superconductor heterostructure. It allows in situ tuning of the transportation properties that can be used to develop reprogrammable superconducting electronic devices, as demonstrated below. Moreover, the method of using an ASI template is not limited to flux quanta. It could be applied to other magnetically interacting particle systems, such as magnetic skyrmions⁶ and magnetic colloids^{10,11}.

The ability to tune the symmetry of the flux-quantum lattice in situ (Fig. 2) can be extended to design superconducting devices, such as flux-quantum rectifiers. Such flux-quantum diodes can be realized based on the ratchet motion of flux quanta induced by an asymmetric potential^{33–36}. We applied an a.c. current to the superconducting layer of the heterostructure and measured a d.c. voltage response associated with the directional motion of flux quanta (Fig. 4a). The rectification effect was observed for the type II (Fig. 4c) and type III (Fig. 4e) magnetic-charge configurations and could be turned off with the type I (Fig. 4b) configuration. Supplementary Videos 1–3 clearly show the asymmetric dynamics of flux-quantum motion under the type II and III magnetic-charge orders. We can also reverse the rectification polarity by reversing the magnetic-charge configurations, as shown in Fig. 4d,f for the type II and III configurations, respectively. This reprogrammable spin-ice flux-quantum diode could provide the basis for logic gates for low-power computing.

In addition to designing geometric frustration in various magnetic systems, reconfigurable electronic devices could be envisioned by combining an ASI with functional materials, such as 2D electron gases⁷, graphene⁸, topological insulators⁹, and Dirac and Weyl semimetals. Their electrical conductance, magnetoresistance and (topological) Hall effects could be tuned by controlling the motion of electrons using reconfigurable magnetic potentials. Furthermore, the precise control of the magnetic-charge configuration with a magnetic tip⁵ enables local ‘write and erase’ of geometric frustrated and crystallized patterns, leading to even more exotic properties and enhanced control of functionalities at the nanoscale. The distinguishable features in the transport behaviour shown in Fig. 1f–h and Supplementary Fig. 2 demonstrate that measuring electrical transport on a subfilm is a convenient technique for detecting the spin-charge configurations of magnetic systems and complementing the magnetic imaging techniques. Finally, by replacing the present type I magnetic charges with nanobar magnets or magnetic nanodots (Supplementary Fig. 9), one can create novel geometric frustration that can be investigated by direct imaging and thermal annealing techniques²⁴.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available at <https://doi.org/10.1038/s41565-018-0162-7>.

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Author contributions

Y.-L.W., Z.-L.X., B.J. and W.-K.K. conceived and supervised the project. Y.-L.W., J.X., Z.-L.X. and W.-K.K. designed the experiments. X.M. and B.J. designed the

simulations. Y.-L.W., J.X., R.D., L.E.O. and J.E.P. fabricated samples. Y.-L.W., J.X. and A.S. conducted the measurements. X.M. conducted the simulations. Y.-L.W., X.M. and J.X. analysed the data. Y.-L.W., X.M., J.X., Z.-L.X., B.J. and W.-K.K. co-wrote the manuscript. All authors contributed to the discussion of the data and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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Methods

Sample fabrication. The superconductor in our heterostructure is a highly uniform $\text{Mo}_{0.79}\text{Ge}_{0.21}$ (MoGe) film with weak intrinsic pinning, allowing transport measurements in a wide temperature range^{18,37–41}. A 50-nm-thick superconducting MoGe film was sputtered from an MoGe alloy target onto silicon substrates with an oxide layer. The samples were first patterned into a 50- μm -wide microbridge with voltage leads separated by 50 μm using standard photolithography. Subsequently, an ASI of permalloy ($\text{Ni}_{0.80}\text{Fe}_{0.20}$) bar magnets (each with the dimensions 300 nm $\times$ 80 nm $\times$ 25 nm) was patterned on top of the MoGe film using electron-beam lithography. The detailed fabrication parameters and procedures for patterning the tri-axial ASI have been described previously⁵. The MoGe film has a superconducting transition temperature of 6.9 K, as shown in Supplementary Fig. 10.

Experiments. Magnetic force microscopy (MFM) imaging was conducted in a custom-designed MFM system using a commercial MFM probe (Point Probe Plus Magnetic Force Microscopy; NANOSensors). The detailed imaging parameters have been reported previously⁵. The transport experiments were carried out using a standard four-probe method at a temperature of 5.8 K. The sample was placed in a three-axis superconducting vector magnet, which allowed us to apply a magnetic field in any desired orientation. The magnetic-charge ordering was manipulated by controlling both the orientation and amplitude of the in-plane magnetic field. The detailed protocols have been reported previously⁵. The transport experiments were carried out with only the out-of-plane magnetic field. The in-plane magnetic field was only used to reconfigure the magnetic-charge state and ramped to zero thereafter.

Molecular dynamics simulation. We consider a 2D system with periodic boundary conditions in the x and y directions. The sample size is $L \times L$ with $L = 16\lambda$. λ is the London penetration depth, which is chosen as 720 nm for MoGe (ref. 17). The magnetic charges at the bar magnet ends are kept as attractive or repulsive centres for flux quanta. The interaction energy of a flux quantum with a single pinning site is modelled as a Gaussian function, with attractive (repulsive) pinning sites introducing negative (positive) interaction energy to the system.

The dynamics of an individual flux quantum i is determined by the following overdamped equation of motion:

$$\eta \frac{d\vec{R}_i}{dt} = \vec{F}_i^{\text{vv}} + \vec{F}_i^{\text{pa}} + \vec{F}_i^{\text{pt}} + \vec{F}_i^{\text{d}} + \vec{F}_i^{\text{T}}$$

Here, η is the damping constant, which is set equal to 1. The repulsive inter-flux-quantum interaction force is given by $\vec{F}_i^{\text{vv}} = \sum_{j \neq i} F_0 K_1(R_{ij}/\lambda) \hat{R}_{ij}$, where $\vec{R}_i$ is the location of the flux quantum i , K_1 is the modified Bessel function, $R_{ij} = |\vec{R}_i - \vec{R}_j|$, $\hat{R}_{ij} = (\vec{R}_i - \vec{R}_j)/R_{ij}$, $F_0 = \phi_0^2/(2\pi\mu_0\lambda^3)$, ϕ_0 is the flux quantum and μ_0 is the permeability⁴². The pinning force is derived from the gradient of pinning potential energy, with the form

$$\vec{F}_i^{\text{pa}} = - \sum_{k=1}^{N_{\text{pa}}} F_{\text{p}} R_{ik}^{(p_a)} \exp(-R_{ik}^{(p_a)^2}/r_{\text{p}}^2) \hat{R}_{ik}^{(p_a)},$$

where $r_{\text{p}} = 240$ nm is the standard deviation of Gaussian function, N_{pa} is the total number of attractive pinning sites, F_{p} is the pinning coefficient, $\vec{R}_k^{(p_a)}$ is the centre of attractive pinning site k , $R_{ik}^{(p_a)} = |\vec{R}_i - \vec{R}_k^{(p_a)}|$ and $\hat{R}_{ik}^{(p_a)} = (\vec{R}_i - \vec{R}_k^{(p_a)})/R_{ik}^{(p_a)}$. $\vec{F}_i^{\text{pt}}$ has the same form as $\vec{F}_i^{\text{pa}}$, but the opposite sign. $\vec{F}_i^{\text{d}}$ is the Lorentz force when there is a constant current applied, following $\vec{F} = (\vec{J} \times \hat{z})\phi_0 d$ (ref. 28), where $\vec{J}$ is the current density, $\hat{z}$ is the direction of flux-quantum vorticity and d is the thickness of the superconducting film. Thermal forces are modelled as Langevin kicks $\vec{F}_i^{\text{T}}$ satisfying $\langle \vec{F}_i^{\text{T}}(t) \rangle = 0$ and $\langle \vec{F}_i^{\text{T}}(t) \cdot \vec{F}_j^{\text{T}}(t') \rangle = 2\eta k_{\text{B}} T \delta_{ij} \delta(t-t')$, where k_{B} is the Boltzmann constant, T is the temperature and t is time.

We start from a high temperature where flux quanta are in a molten state and then slowly reduce the temperature to 0 K with 130 intermediate temperatures. At each temperature, we let the system evolve 2×10^5 steps, each being $dt = 0.01$ in simulation units. After annealing, we obtain the static flux-quantum configurations as the ground states. To obtain the critical current, we apply and increase the driving force slowly. For each driving force, we measure the time-averaged total flux-quantum velocity in the force direction, $V_x = \frac{1}{N_{\text{v}}} \sum_i^{N_{\text{v}}} \hat{x} \cdot \vec{v}_i$, where N_{v} is the total number of flux quanta, N_{t} is the total number of time steps and v is the velocity of a single flux quantum. V_x corresponds to the dissipation voltage. When V_x exceeds the threshold, which is set at 40 in the simulation, most flux quanta start to move and the non-dissipative superconducting phase breaks down. The critical driving force is defined as the depinning force, which represents the critical current. To speed up our simulation, we used binary search here to find the critical current. Though the true ground states may only be attainable for prohibitively long waiting times, the agreement between our simulation results and experiments indicates validation of our model⁴³.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

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