

when $T^* \sim 0$ suggests that it is associated with proximity to the putative nematic quantum phase transition.

P-substituted BaFe_2As_2 is the least disordered of all of the known 122-type iron pnictide families, as evidenced by the fact that quantum oscillations can be observed across the phase diagram (44, 45). The deviation from Curie-Weiss behavior for optimally doped compositions of all other dopants in BaFe_2As_2 suggests that disorder plays an important role in the quantum critical regime. All forms of quenched disorder produce locally anisotropic effective strains, which thus couple to the orientation of the nematic order; this is known as random-field Ising model yields several generically expected effects of random-field disorder, including suppression of the nematic susceptibility below mean-field expectations for a clean system, and, for the case of a quantum phase transition, the enhanced sensitivity of quantum critical phenomena to disorder (36).

The temperature dependence of the nematic susceptibility can also be extracted from measurements of the elastic moduli (20). The two measurements (elastoresistance and elastic moduli) are in broad agreement—for example, in terms of the Curie-Weiss T dependence of χ_N for underdoped compositions of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, and in terms of the deviation from Curie-Weiss behavior near optimal doping. However, there is an important distinction in the relative magnitude of the measured quantities as a function of doping. In particular, the normalized lattice softening $(c_{66}^0 - c_{66})/c_{66}^0$ extracted in (20) monotonically decreases in magnitude and extends over a smaller window of temperature as the Co concentration x is increased. In contrast, the quantity $|2m_{66} - 2m_{66}^0| = c\chi_N$ initially increases with x , peaking for slightly underdoped compositions where $x \sim 0.05$. The apparent enhancement of the elastoresistance $2m_{66}$ over the softening of the elastic modulus for compositions near optimal doping is potentially related to renormalization of the quasiparticle effective mass in the quantum critical regime, as has been observed in P-substituted BaFe_2As_2 (29). Such a mass renormalization is an expected consequence of nematic quantum critical fluctuations (40). It is precisely these low-energy quasiparticles, which are responsible for the large elastoresistivity, that are also involved in the eventual superconductivity.

REFERENCES AND NOTES

- E. Fradkin, S. A. Kivelson, M. J. Lawler, J. P. Eisenstein, A. P. Mackenzie, *Annu. Rev. Condens. Matter Phys.* **1**, 153–178 (2010).
- Y. Ando, K. Segawa, S. Komiya, A. N. Lavrov, *Phys. Rev. Lett.* **88**, 137005 (2002).
- C. Howald, H. Eisaki, N. Kaneko, M. Greven, A. Kapitulnik, *Phys. Rev. B* **67**, 014533 (2003).
- V. Hinkov *et al.*, *Science* **319**, 597–600 (2008).
- R. Daou *et al.*, *Nature* **463**, 519–522 (2010).
- S. A. Kivelson *et al.*, *Rev. Mod. Phys.* **75**, 1201–1241 (2003).
- T. M. Chuang *et al.*, *Science* **327**, 181–184 (2010).
- J. H. Chu *et al.*, *Science* **329**, 824–826 (2010).
- M. A. Tanatar *et al.*, *Phys. Rev. B* **81**, 184508 (2010).
- A. Dusza *et al.*, *Euro. Phys. Lett.* **93**, 37002 (2011).
- M. Yi *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **108**, 6878–6883 (2011).
- X. Lu *et al.*, *Science* **345**, 657–660 (2014).
- J.-H. Chu, H.-H. Kuo, J. G. Analytis, I. R. Fisher, *Science* **337**, 710–712 (2012).
- H.-H. Kuo, M. C. Shapiro, S. C. Riggs, I. R. Fisher, *Phys. Rev. B* **88**, 085113 (2013).
- H.-H. Kuo, I. R. Fisher, *Phys. Rev. Lett.* **112**, 227001 (2014).
- J. Gallais *et al.*, *Phys. Rev. Lett.* **111**, 267001 (2013).
- V. K. Thorsmølle *et al.*, *Phys. Rev. B* **93**, 054515 (2016).
- F. Kretzschmar *et al.*, *Nat. Phys.* (2016).
- M. Yoshizawa *et al.*, *J. Phys. Soc. Jpn.* **81**, 024604 (2012).
- A. E. Böhmer *et al.*, *Phys. Rev. Lett.* **112**, 047001 (2014).
- J. Chang *et al.*, *Nat. Phys.* **8**, 871–876 (2012).
- G. Ghiringhelli *et al.*, *Science* **337**, 821–825 (2012).
- E. Blackburn *et al.*, *Phys. Rev. Lett.* **110**, 137004 (2013).
- F. M. Tranquada, B. J. Sternlieb, J. D. Axe, Y. Nakamura, S. Uchida, *Nature* **375**, 561–563 (1995).
- T. Wu *et al.*, *Nature* **477**, 191–194 (2011).
- L. Nie, G. Tarjus, S. A. Kivelson, *Proc. Natl. Acad. Sci. U.S.A.* **111**, 7980–7985 (2014).
- S. A. Kivelson, E. Fradkin, V. J. Emery, *Nature* **393**, 550–553 (1998).
- B. J. Ramshaw *et al.*, *Science* **348**, 317 (2015).
- J. G. Analytis *et al.*, *Nat. Phys.* **10**, 194–197 (2014).
- K. Fujita *et al.*, *Science* **344**, 612–616 (2014).
- M. A. Metlitski, D. F. Mross, S. Sachdev, T. Senthil, *Phys. Rev. B* **91**, 115111 (2015).
- S. Lederer, Y. Schattner, E. Berg, S. A. Kivelson, *Phys. Rev. Lett.* **114**, 097001 (2015).
- T. A. Maier, D. J. Scalapino, <http://arxiv.org/abs/1405.5238> (2014).
- There are several different ways to define a nematic order parameter in terms of equilibrium electronic correlation functions. Correspondingly, there is ambiguity in defining the nematic susceptibility. Thus, the constants c and c' depend not only on microscopic details but also on the precise microscopic definition given to the nematic order parameter.
- Typical strains using this method are on the order of 10^{-3} to 10^{-4} .
- Materials and methods are available as supplementary materials on Science Online.
- As described in (36), samples that have in-plane dimensions that are larger than $\sim 700 \times 700 \mu\text{m}$ do not exhibit a size dependence of the strain transmission for typical thicknesses. Although most of the samples used in this study are in this
- size range, the P-substituted iron arsenide samples have in-plane dimensions of $\sim 300 \times 300 \mu\text{m}$, resulting in a smaller strain transmission. This does not affect the measured Weiss temperature (36).
- A. Tamai *et al.*, *Phys. Rev. Lett.* **104**, 097002 (2010).
- A. Pourret *et al.*, *Phys. Rev. B* **83**, 020504(R) (2011).
- A. L. Fitzpatrick, S. Kachru, J. Kaplan, S. Raghu, *Phys. Rev. B* **88**, 125116 (2013).
- S. A. Hartnoll, R. Mahajan, M. Punk, S. Sachdev, *Phys. Rev. B* **89**, 155130 (2014).
- Preliminary confirmation that the nematic susceptibility at criticality diverges as $1/T$ has been obtained from determinantal Monte Carlo calculations of a two-dimensional metallic nematic quantum critical system (43).
- Y. Shattner, S. Lederer, S. A. Kivelson, E. Berg, <http://arxiv.org/abs/1511.03282> (2015).
- J. G. Analytis, J.-H. Chu, R. D. McDonald, S. C. Riggs, I. R. Fisher, *Phys. Rev. Lett.* **105**, 207004 (2010).
- H. Shishido *et al.*, *Phys. Rev. Lett.* **104**, 057008 (2010).

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SUPPLEMENTARY MATERIALS

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ARTIFICIAL SPIN ICE

Rewritable artificial magnetic charge ice

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Artificial ices enable the study of geometrical frustration by design and through direct observation. However, it has proven difficult to achieve tailored long-range ordering of their diverse configurations, limiting both fundamental and applied research directions. We designed an artificial spin structure that produces a magnetic charge ice with tunable long-range ordering of eight different configurations. We also developed a technique to precisely manipulate the local magnetic charge states and demonstrate write-read-erase multifunctionality at room temperature. This globally reconfigurable and locally writable magnetic charge ice could provide a setting for designing magnetic monopole defects, tailoring magnonics, and controlling the properties of other two-dimensional materials.

Artificial ices are structures in which the constituents obey analogs of the “two-in, two-out” Pauling’s ice rule that determines the proton positional ordering in water ice. These structures provide a material-by-design approach to physical properties and functionalities (1–27). Artificial ice can be created from ferromagnetic islands and connected wires (3, 5), topological components such as superconducting vortices (24–26), and nonmagnetic colloidal particles (27).

Among them, artificial spin ice is the most investigated system (1–21) that was first demonstrated in a square lattice of elongated interacting ferromagnetic nanoislands (7). In this case, the ice rule corresponds to two spins pointing inward and two pointing outward at the vertex of a square lattice. Extensive experiments have been conducted using various thermal (6–8) and magnetization approaches (9–17) to obtain ordered states of the spin ice. Long-range ordering was realized in the diagonally

polarized magnetic state through the magnetization method (11, 14). For the nominal ground state, sizable domains and crystallites were obtained in as-grown samples (11), and larger domains were obtained in samples heated above their Curie temperatures ($\sim 500^\circ\text{C}$ for permalloy island arrays) (6). At room temperature, long-range ordering of the ground state has been achieved only in ultrathin (3-nm-thick) samples via thermal relaxation (8). Other spin and charge configurations have only been observed locally and at crystallite boundaries (5, 6). The difficulty in creating tailored multiple ordered states limits the experimental investigation of the spin and magnetic charge dynamics that can emerge from or between the ordered states (6, 10–13), especially for a thermally stable (athermal) sample at room temperature. This also hinders the potential applications of artificial ice for data storage, memory, and logic devices (3, 4) or as a medium for reconfigurable magnonics (28).

We designed an artificial spin structure in which we can conveniently obtain multiple long-range orderings of a magnetic charge ice lattice at room temperature. In a typical square spin ice (Fig. 1A), the single-domain magnetic islands are considered to be macro-Ising spins (11). Each macrospin can be replaced with a dumbbell of magnetic charges, one positive and one negative (3, 4, 29) (Fig. 1B). If we break the connections between the pairs of magnetic charges with opposite signs (Fig. 1C), we can design a different pattern of connections (Fig. 1D). Figure 1E shows the resulting artificial spin structure explicitly. The structure consists of a square lattice of plaquettes (labeled M and N) containing ferromagnetic nanoislands with three orientations (horizontal, vertical, and diagonal). The magnetic charge distribution is indicated by the calculated stray field distribution (Fig. 1F), which leads to a magnetic charge ice with the charge ice rule of two negative and two positive charges within each square plaquette.

The magnetic charge distribution in Fig. 1F is exactly the same as that of the ground state of a square spin ice structure (fig. S1). However, in contrast to the square spin ice, which has spins on four sides of the square plaquette and all oriented toward the plaquette center or vertex (Fig. 1A), the structure in Fig. 1E places one spin within the plaquette while removing two from the sides, breaking the fourfold symmetry of the square lattice. Moreover, the three spins associated with each plaquette do not meet at a vertex. Our design contains two types of plaquettes, one rotated by

180° from the other (denoted as M and N in Fig. 1E). Each plaquette (M or N) consists of three islands, each with two degrees of freedom coming from spin, resulting in a total multiplicity of eight configurations of magnetic charges. (See figs. S1 and S2 for detailed comparisons of the spin and charge configurations between the standard square spin ice and our design.) The eight ordered charge configurations (Fig. 1G) that we predicted are

separated into three groups on the basis of their energies (see the calculated energies in fig. S3): a twofold degenerate type I ground state (I_1 and I_2), a twofold degenerate excited type II state (II_1 and II_2), and a fourfold degenerate excited type III state (III_1 , III_2 , III_3 , and III_4) (Fig. 1G).

Because of the large energy barrier for the spins, the spin system in Fig. 1E is athermal at room temperature. An external applied magnetic

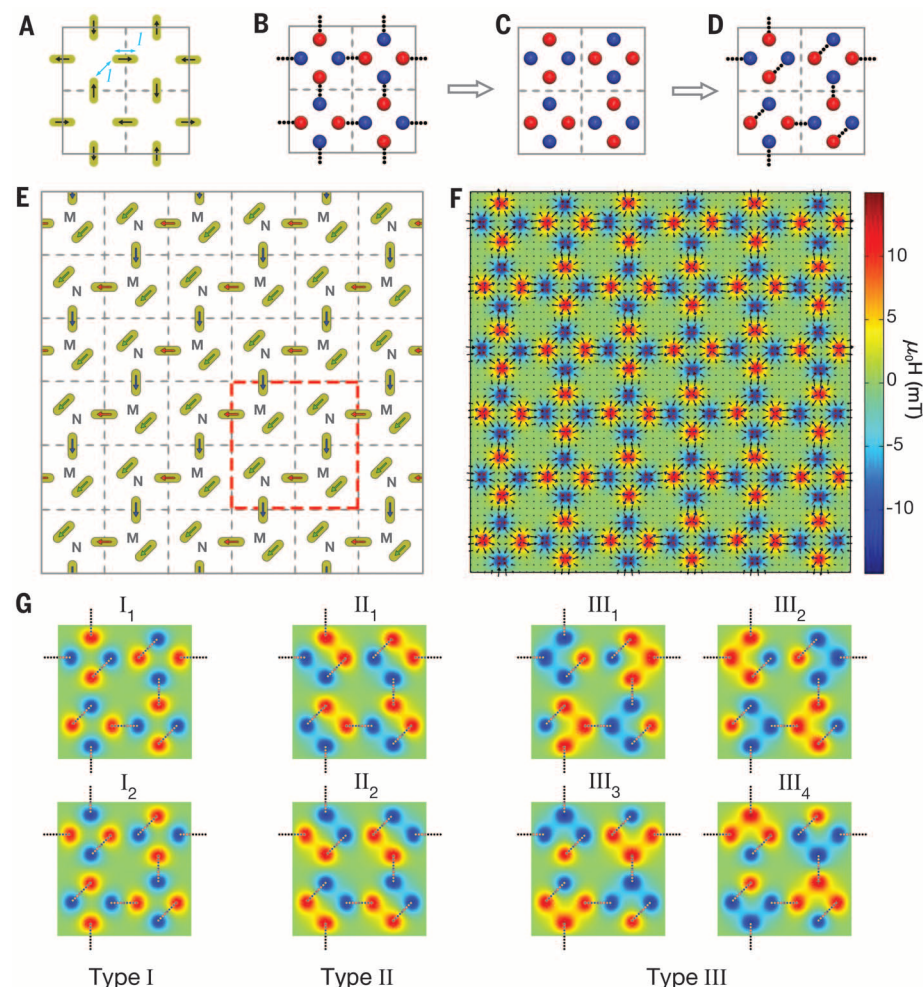


Fig. 1. Design of magnetic charge ices. (A) Typical artificial square spin ice in which the length of the magnetic nanoisland equals the separation of the ends of the nearest islands. Each island is an Ising spin (black arrows). The spin configuration of the lowest-energy ground state is shown. (B) Distribution of magnetic charges corresponding to the square spin ice shown in (A). Pairing of the positive (red) and negative (blue) charges is indicated by black dotted lines. (C) Distribution of magnetic charges with charge connections removed. (D) Redesign of the connections of paired charges with positive and negative polarities. (E) Design of the magnetic nanostructure based on (D). The arrows indicate the spin configuration of the ground state and are color coded for the three subsets of islands placed in three different orientations (horizontal, vertical, and diagonal). Two types of plaquettes are marked with M and N. (F) Calculated magnetic stray field associated with the spin configuration of the structure in (A) for islands with dimensions of 300 nm by 80 nm by 25 nm. Arrows indicate the local field directions. Color is encoded by the out-of-plane component of the field (red, out of the plane; blue, into the plane). The red and blue spots represent the positive and negative magnetic charges, respectively. μ_0 , vacuum permeability; H , magnetic field. (G) Calculated magnetic stray fields of a 2-by-2 square plaquette, highlighted by the red frame in (E), for the eight possible spin- and charge-ordered configurations, separated into three charge types. Dotted lines denote the orientation of the islands containing the pair of magnetic charges (negative charge, blue; positive charge, red). To compare the magnetic simulations with the experimental magnetic force microscopy images, the stray field was calculated at a plane 100 nm above the surface of the sample.

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field can be used to overcome the energy barrier. For a given island, the minimal magnetic field required to flip its spin moment varies with the angle between the applied field and the island

(fig. S4). This enables separate control of the spin moments of each differently oriented island. In a square spin ice, there are two orientations of the islands, and the spins can only be aligned in di-

agonal directions by an applied magnetic field, enabling solely the type II phases with long-range ordering (*II*, *II'*). Our design (Fig. 1E) provides an additional freedom for aligning the spins: There

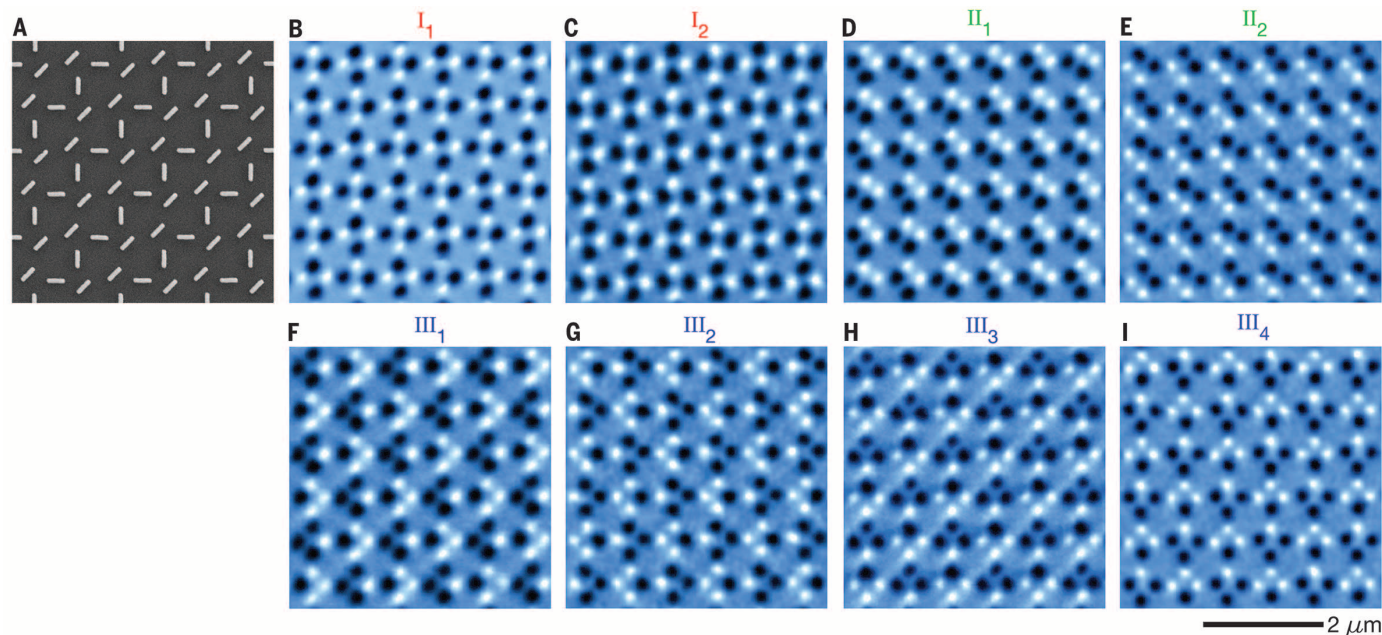


Fig. 2. Realization of magnetic charge ices. (A) Scanning electron microscopy image of permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) magnetic islands (300 nm long, 80 nm wide, and 25 nm thick). (B to I) Magnetic force microscopy images of the various ordered states corresponding to all of the configurations in Fig. 1G. (B and C) Twofold degenerate type I ground states: I_1 (B) and I_2 (C). (D and E) Twofold degenerate excited type II states: II_1 (D) and II_2 (E). (F to I) Fourfold degenerate excited type III states: III_1 (F), III_2 (G), III_3 (H), and III_4 (I). The lift height of the MFM scanning is 100 nm.

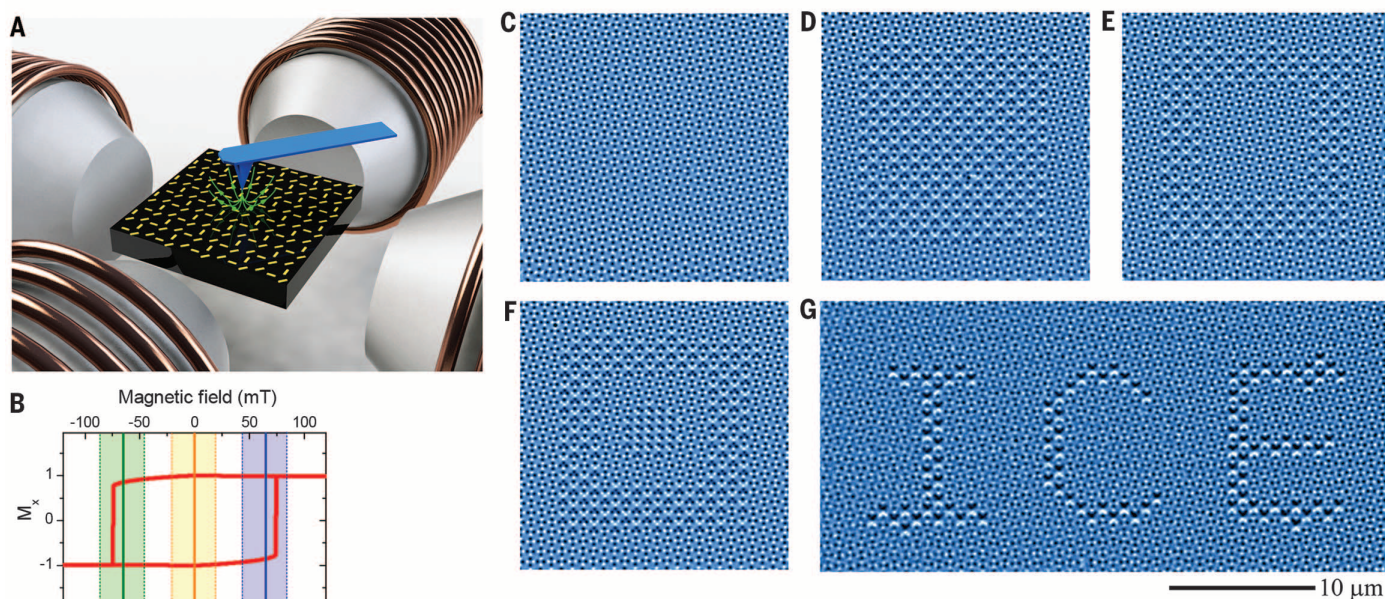


Fig. 3. Rewritable magnetic charge ices. (A) Sketch of the experimental setup: an MFM equipped with a 2D vector magnet. The 2D solenoid magnet provides magnetic fields in any desired orientation in the sample plane. The vertically magnetized MFM probe generates a stray magnetic field (green arrows) with in-plane components at the tip. (B) Magnetization loop of a single magnetic island, with an illustration of the write, erase, and read functions. M_x , magnetization along the island. (C to G) Magnetic force microscopy images of the patterned magnetic charge ice at the same area of the sample. (C) The initial state is a type I_1 state. (D) A square area of a type III_3 state was written in the center of (C). (E) A smaller square region of type III_3 order was erased back to a type I_1 state from (D). (F) A round region of type II_2 order was written onto the freshly erased area from (E). (G) "ICE" letters of type III_4 states were scribed on a type I_1 background state.

are three sets of islands oriented in the horizontal, vertical, and diagonal directions. More importantly, for each of the predicted ordered states shown in Fig. 1G, the spin moments of each of the three oriented islands (horizontal, vertical, and diagonal) are all magnetized in the same direction. This enables the creation of long-range ordering for all of the predicted charge configurations by tuning the in-plane external magnetic field angle and amplitude. We calculated the field-angle dependence of the moment-flipping curves for the three sets of islands (fig. S4C) and designed an effective magnetization protocol to realize the various ordered charge states (30).

To experimentally demonstrate the magnetic charge ordering, we fabricated arrays of permalloy ($\text{Ni}_{0.8}\text{Fe}_{0.2}$) nanoislands (300 nm long, 80 nm wide, and 25 nm thick) onto a Si/SiO_2 substrate (30), according to the design in Fig. 1E. A scanning electron microscopy image of the sample is presented in Fig. 2A. To control and visualize the charge configurations, we used a customized magnetic force microscope (MFM) equipped with a two-dimensional (2D) vector magnet. Figure 3A shows a schematic drawing of our experimental setup. The 2D electromagnetic solenoid magnet provides in-plane magnetic fields in any desired orientation, enabling us to accurately tune the field angle and amplitude.

The as-grown (fig. S5) and demagnetized (fig. S6) samples show mixed charge states of all eight charge configurations at both the M and N plaquettes. Results of statistical analysis for the demagnetized samples show that the charge-neutral configurations (type I) are strongly favored and the collective interaction can be enhanced by reducing the charge separation (fig. S6). Using the designed magnetization protocol (fig. S7), we successfully obtained all eight configurations of magnetic charge ordering, as shown by the magnetic force microscopy images in Fig. 2, B to I. Each of these ordered states possesses long-range ordering and can be reproduced over the entire patterned sample area (80 μm by 80 μm). For type I and II ordered states, there is no net charge in each plaquette, and the magnetic charge follows the “two-positive, two-negative” charge ice rule. In the type III ordered states, each plaquette with magnetic charges following the “three-positive (or negative), one-negative (or positive)” charge ice rule has an effective magnetic charge of two, with opposite signs on the M and N plaquettes. This distribution of magnetic charges in the square lattice of the type III state resembles the distribution of electrical charges in ionic compounds, such as $\text{Mg}^{2+}\text{O}^{2-}$. Thus, the type III states (Fig. 2, F to I) resemble an ionic crystal with magnetic charges (31), where we can associate $\text{M}^{2+}\text{N}^{2-}$ with the type III_2 and III_4 states and $\text{M}^{2-}\text{N}^{2+}$ with the type III_1 and III_3 states. In principle, all of these magnetic charge distributions could also exist in a square spin ice structure (figs. S1E and S2). In fact, our micromagnetic simulation result indicates that the square spin ice and our engineered spin structure not only produce the same magnetic charge arrangements but also have the same excitation energies for the type

I, II, and III configurations (fig. S3). However, with the square spin ice arrangement, long-range ordering of type I states has only been realized in a thermally relaxed sample, and that of the type III states has not yet been experimentally realized because of the spin arrangement of the islands.

The reconfigurable ordered magnetic charges can be used, for example, as templates to form other artificial ices, such as superconducting vortex ices (25, 26), by introducing icelike pinning potentials for superconducting vortices. These charges can also be applied to couple with other electronic materials, such as 2D electron gas (32, 33) and graphene (34), by producing reconfigurable periodically distributed field potentials. For applications such as data storage, memory, and logic devices (3, 35, 36), however, local control of the magnetic charge states is desired. Toward this end, we developed a 2D magnetic field-assisted magnetic force microscopy patterning technique, which allows us to conveniently manipulate the local charge configurations.

As illustrated in Fig. 3A, the magnetic tip of an MFM generates an in-plane component of stray magnetic fields near the tip. The interaction of the MFM tip's stray field, ΔH^m , with a single ferromagnetic island can be tuned by adjusting the height of the MFM tip from the sample, which is 100 nm in this experiment. To locally switch the spin states of an island, we apply an in-plane magnetic field H^{ap} slightly below the ferromagnetic island's spin-moment-flipping field H^f (Fig. 3B). At this field value, the spin states of the entire sample will not be altered because $H^{\text{ap}} < H^f$. When the MFM probe scans over an island, the total magnetic field on that island will change in the range $H^{\text{ap}} \pm \Delta H^m$. We adjust H^{ap} and ΔH^m to satisfy the condition $H^{\text{ap}} < H^f < (H^{\text{ap}} + \Delta H^m)$ (light blue region in Fig. 3B). In this case, the spin of the underlying island flips when the MFM tip scans over it, providing a “write” function. A subsequent applied magnetic field in the opposite direction with $-(H^{\text{ap}} + \Delta H^m) < -H^f < -(H^{\text{ap}} - \Delta H^m)$ (green region in Fig. 3B) will switch the spin back, implementing the “erase” function (Fig. 3B). When the applied field is zero, the stray field provided by the MFM probe (yellow region in Fig. 3B) is too small to flip any islands, and this works as the “read” mode. Because the value of H^f depends on the angle between the applied field and the island, similar to the global control of the charge ordering, we can locally manipulate the charge states into any desired configuration.

We demonstrate the experimental realization of the write, read, and erase functions in Fig. 3, C to F. We first prepare the entire sample in the type I ground state (Fig. 3C) by applying and zeroing an in-plane magnetic field of 90 mT along the diagonal direction. We then write a square area of type III ordered state in the center (Fig. 3D) and subsequently erase a smaller square region in the central area by switching the type III state back into the type I state (Fig. 3E). Finally, we write a type II state into a small circular area inside the type III square (Fig. 3F). We can also

write letters, as presented in Fig. 3G where the word “ICE” is scribed with type III order on a type I background. Such magnetic charge patterning can easily be realized by programming the 2D magnet to turn on or off and to switch the field directions during the magnetic force microscopy scanning, resembling the patterning process applied in electron beam lithography and in optical lithography using a laser pattern generator. See fig. S8 for more patterns of magnetic charge arrays. These rewritable magnetic charge patterns could be transferred to other materials, for example, through magnetolithography (37).

In addition to the aforementioned applications on coupling with superconducting vortices and other electronic systems, this reconfigurable magnetic charge ice can provide a platform to explore phenomena such as the ground state of a frustrated lattice (6). Combined with other control parameters such as the thickness of the islands (11), temperature (6), or oscillating magnetic field (10), our platform provides a versatile system to study and tailor phase transitions and defect formation (fig. S9). For example, the single spin control enabled by our method allows the creation of magnetic defects such as magnetic monopoles and Dirac strings (16, 17, 20) at any desired location. It also provides a direct technique to program magnetic logic circuits (35, 36). Our strategy to decouple the arrangement of spins and magnetic charges should stimulate further creation and exploration of exotic phases of magnetic charges and their phase transitions and should also foster applications. We also note that Fig. 1E is not the only spin arrangement for achieving the same magnetic charge distribution. In fig. S10, we present several other possible designs of the spin and charge arrangements, including the type IV charge-ordered state in which all positive or negative charges are confined within a single plaquette. Furthermore, it is not necessary to keep the length of all ferromagnetic islands the same, as recently reported in Shakti spin ices (18, 19). Our strategy could also be applied to other artificial spin ices to produce artificial structures with controllable magnetic charge orders, which would provide reconfigurable platforms for magnonic investigations, such as programming spin-wave band structures and designing spin-wave transmission channels (5, 28).

REFERENCES AND NOTES

1. R. F. Wang et al., *Nature* **439**, 303–306 (2006).
2. W. R. Branford, S. Ladak, D. E. Read, K. Zeissler, L. F. Cohen, *Science* **335**, 1597–1600 (2012).
3. L. J. Heyderman, R. L. Stamps, *J. Phys. Condens. Matter* **25**, 363201 (2013).
4. L. J. Heyderman, *Nat. Nanotechnol.* **8**, 705–706 (2013).
5. C. Nisoli, R. Moessner, P. Schiffer, *Rev. Mod. Phys.* **85**, 1473–1490 (2013).
6. S. Zhang et al., *Nature* **500**, 553–557 (2013).
7. C. Nisoli et al., *Phys. Rev. Lett.* **105**, 047205 (2010).
8. A. Farhan et al., *Phys. Rev. Lett.* **111**, 057204 (2013).
9. C. Nisoli et al., *Phys. Rev. Lett.* **98**, 217203 (2007).
10. X. Ke et al., *Phys. Rev. Lett.* **101**, 037205 (2008).
11. Z. Budrikis et al., *Phys. Rev. Lett.* **109**, 037203 (2012).
12. J. P. Morgan, A. Stein, S. Langridge, C. H. Marrows, *Nat. Phys.* **7**, 75–79 (2011).
13. V. Kapaklis et al., *Nat. Nanotechnol.* **9**, 514–519 (2014).

14. A. Remhof *et al.*, *Phys. Rev. B* **77**, 134409 (2008).
15. Z. Budrikis, P. Politi, R. L. Stamps, *Phys. Rev. Lett.* **105**, 017201 (2010).
16. S. Ladak, D. E. Read, G. K. Perkins, L. F. Cohen, W. R. Branford, *Nat. Phys.* **6**, 359–363 (2010).
17. E. Mengotti *et al.*, *Nat. Phys.* **7**, 68–74 (2011).
18. I. Gilbert *et al.*, *Nat. Phys.* **10**, 670–675 (2014).
19. G. W. Chern, M. J. Morrison, C. Nisoli, *Phys. Rev. Lett.* **111**, 177201 (2013).
20. C. Phatak, A. K. Petford-Long, O. Heinonen, M. Tanase, M. De Graef, *Phys. Rev. B* **83**, 174431 (2011).
21. A. Schumann, B. Sothmann, P. Szary, H. Zabel, *Appl. Phys. Lett.* **97**, 022509 (2010).
22. L. Anghinolfi *et al.*, *Nat. Commun.* **6**, 8278 (2015).
23. I. Gilbert *et al.*, *Nat. Phys.* **12**, 162–165 (2015).
24. A. Libál, C. J. Reichhardt, C. Reichhardt, *Phys. Rev. Lett.* **102**, 237004 (2009).
25. M. L. Latimer, G. R. Berdiyorov, Z. L. Xiao, F. M. Peeters, W. K. Kwok, *Phys. Rev. Lett.* **111**, 067001 (2013).
26. J. Trastoy *et al.*, *Nat. Nanotechnol.* **9**, 710–715 (2014).
27. A. Libál, C. Reichhardt, C. J. Reichhardt, *Phys. Rev. Lett.* **97**, 228302 (2006).
28. M. Krawczyk, D. Grundler, *J. Phys. Condens. Matter* **26**, 123202 (2014).
29. C. Castelnovo, R. Moessner, S. L. Sondhi, *Nature* **451**, 42–45 (2008).
30. Supplementary materials are available on Science Online.
31. G. Möller, R. Moessner, *Phys. Rev. B* **80**, 140409 (2009).
32. A. Singha *et al.*, *Science* **332**, 1176–1179 (2011).
33. P. D. Ye *et al.*, *Phys. Rev. Lett.* **74**, 3013–3016 (1995).
34. M. Taillefumier, V. K. Dugaev, B. Canals, C. Lacroix, P. Bruno, *Phys. Rev. B* **84**, 085427 (2011).
35. A. Imre *et al.*, *Science* **311**, 205–208 (2006).
36. I. Amlani *et al.*, *Science* **284**, 289–291 (1999).
37. A. Bardea, R. Naaman, *Small* **5**, 316–319 (2009).

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SUPPLEMENTARY MATERIALS

www.sciencemag.org/content/352/6288/962/suppl/DC1
Materials and Methods
Figs. S1 to S10
Reference (38)

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SUPERCONDUCTIVITY

Supercurrent in the quantum Hall regime

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A promising route for creating topological states and excitations is to combine superconductivity and the quantum Hall (QH) effect. Despite this potential, signatures of superconductivity in the QH regime remain scarce, and a superconducting current through a QH weak link has been challenging to observe. We demonstrate the existence of a distinct supercurrent mechanism in encapsulated graphene samples contacted by superconducting electrodes, in magnetic fields as high as 2 tesla. The observation of a supercurrent in the QH regime marks an important step in the quest for exotic topological excitations, such as Majorana fermions and parafermions, which may find applications in fault-tolerant quantum computing.

The interplay of the quantum Hall (QH) effect with superconductivity is expected to result in excitations with nontrivial braiding statistics such as Majorana fermions and non-Abelian Majorana anyons (1–4). When a QH region is contacted by two superconducting electrodes, the gapped QH bulk prevents the flow of a supercurrent (5–11). However, it has been predicted that the supercurrent may still be mediated by QH edge states (12). Because of its chiral nature, a single edge can conduct charge carriers in only one direction, so both edges must be involved in establishing a supercurrent between the two contacts. This situation is fundamentally different from that of the

Josephson junctions made out of two-dimensional (2D) topological insulators, where each edge can support its own supercurrent (13–16). Contrary to the case of topological insulators, the magnetic field in the QH regime breaks time-reversal symmetry, which is essential for s-wave pairing of conventional superconductors. Working in this regime, we nonetheless observe a robust supercurrent, which we attribute to an unconventional form of Andreev bound states circulating along the perimeter of the QH region and involving electron and hole trajectories separated by several micrometers.

We performed transport measurements on four Josephson junctions (J_{1-4}) made of graphene encapsulated in boron nitride and contacted by electrodes made of a molybdenum-rhenium alloy (Fig. 1A) (17), a type II superconductor with a high upper critical field of $H_{c2} = 8$ T. The high quality of these heterostructures allowed us to observe Fabry-Perot oscillations of the junctions' resistance and critical current, indicating that the transmission of charge carriers between the contacts is ballistic (17). The supercurrent is uniformly distributed along the width of the contacts, as evidenced by the regular Fraunhofer pattern (18) measured at small magnetic fields (17). All junc-

tions demonstrate supercurrent in the QH regime (figs. S6 to S12); for consistency, we choose to present data measured on sample J_1 . It has a distance between contacts of length $L = 0.3$ μm and a width of contacts $W = 2.4$ μm (Fig. 1B).

Recently, the observation of a supercurrent through encapsulated graphene in moderate magnetic fields was reported (19); in that setup, the diameter of the cyclotron orbit was larger than but comparable to the length of the junction, $2r_C \geq L$. (Here, $r_C = \hbar k_F / eB$ is the cyclotron radius and k_F is the Fermi wavenumber.) This supercurrent was attributed to Andreev bound states made of closed trajectories connected by several elastic and Andreev reflections, which yield pockets of superconductivity at random values of density and field. We further explore this regime in (17). In the main text, we demonstrate that a very different regime emerges at even larger magnetic fields, when r_C is much smaller than the device dimensions and the mean free path. In this regime, the bulk of the junction is gapped by Landau quantization so that a current may only flow along the edges.

Figure 1, C and D, show the differential resistance of the sample, $R \equiv dV/dI$, plotted versus back-gate voltage, V_G , and magnetic field, B . The resistance is measured in a four-terminal configuration where four MoRe electrodes merge into two contacts on each side of the junction (Fig. 1B). The map in Fig. 1C is measured with an AC excitation current $I_{AC} = 50$ pA applied on top of a large DC current of $I_{DC} = 6$ nA, which suppresses supercurrent and highlights the QH features. As B increases, a fan diagram characteristic of the QH effect in graphene emerges: Resistance plateaus follow contours of constant filling factor $\nu \equiv nh/eB = \pm 2, 6, 10, \dots$ (20). This quantization becomes visible as soon as B exceeds the red parabolic contour $2r_C = L$ because device dimensions prevent the development of the QH effect at lower fields. The dark region under the parabola ($2r_C > L$) indicates a vanishing differential resistance as a supercurrent of tens of nanoamperes may flow in this semiclassical regime (19).

Figure 1D shows $R(V_G, B)$ measured simultaneously with Fig. 1C using exactly the same AC excitation of 50 pA, but without applying a DC current. Notably, pockets of supercurrent extend far into the QH regime. They are visible as dark

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Rewritable artificial magnetic charge ice

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From a bar to a charge, magnetically

Artificial spin ices are arrays of nanoscale bar magnets that can mimic the behavior of naturally occurring “frustrated” magnetic materials. Usually the arrays take the form of a square lattice with the bar magnets perpendicular to its sides. Wang *et al.* “broke up” each bar into a positive and negative magnetic charge. Working backward from an array of these charges, they designed a structure that has bar magnets oriented not only perpendicularly to the sides of the square lattice but also diagonally. Compared to the traditional one, this structure was much more controllable by global and local magnetic fields.

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