Emerging Chirality in Artificial Spin Ice

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Artificial spin ice, made up of planar nanostructured arrays of simple ferromagnetic bars, is a playground for rich physics associated with the spin alignment of the bars and spin texture associated with the magnetic frustration at the bar vertices. The phase diagram is exotic, showing magnetic monopole-like defects and liquid and solid phases of spins arranged in loop states with predicted chiral order. We show that magnetotransport measurements in connected honeycomb structures yield the onset of an anomalous Hall signal at 50K. The temperature scale can be attributed to the long-range dipolar ice phase. The topological Hall signal arises because chiral loops form at the sample edges, indicating a generic route to exotic states via nanoarray edge structure.

Frustrated magnets have complex spin structures. Spin ices are a class of frustrated magnets where the spin disorder resembles the proton disorder in water ice (1-3). In addition to naturally occurring compounds, artificial spin-ice nanostructures have been built with single-domain ferromagnetic islands, or bars, that act as macrospins with dipolar interactions. These form local order by short-range frustration (4-6). We study honeycomb structures, sometimes called kagome spin ice (7), in which the consideration of longer range correlations has led to the prediction of exotic phases (6, 8) that have specific spin texture (i.e. characteristic features in the specific bar spin arrangements).

A uniformly magnetised bar can be considered as a dumbbell of magnetic charge (9). In artificial spin ice, if the dumbbell length l is smaller than the lattice spacing a (Fig 1A) then the uneven charge distribution around the vertex (Fig 1B) gives rise to a vertex dipole moment (VDM) as shown in Fig 1C. In honeycomb structures all possible low-energy vertex configurations (a-f in Fig 1D) have finite magnetic charge and VDM. Theoretical analysis of the system yields (6, 8) a rich phase diagram (Fig 1E). The macrospin correlations are zero in the gas-like Ising paramagnet, nearest neighbour only in the liquid-like short-range spin ice (Ice I), near neighbour in the distinct liquid-like (long-range) spin ice phase (Ice II) and infinite in the fully ordered ground state. The Ice I to Ice II phase transition is chiral (6), at the transition a number of mobile closed loops of each chirality form. Further spin flips in the Ice II system conserve the populations of clockwise and anticlockwise loops. However the phases have proved inaccessible experimentally because the system remains trapped in a metastable state and the groundstate is hidden (10-13).

Spin on the conduction electrons follows the local magnetization direction, and chirality in the spin texture imparts to them a phase called the Berry phase. This mechanism produces an anomalous Hall signal called the topological Hall effect (14-17). It is an exquisitely sensitive probe of the spin chiral order parameter (18) requiring a net imbalance in either total chirality or its spatial distribution. Connected honeycombs naturally lend themselves to interrogation by such methods. Here we search for Hall and magnetoresistance (19) signatures of topological effects by exploring the transport response to swept magnetic fields. The results are most clearly observed using parallel Hall geometry, where the switching transition states of the array in the forward quadrant of the magnetization loop are measurably different from any state realized in the backward quadrant.

Our electrically-continuous cobalt honeycomb covers an area $100\mu\text{m} \times 100\mu\text{m}$ square, and is composed of bars $1\mu\text{m}$ long, 100 nm wide and 20nm high. The four-point technique for measuring magnetotransport in large area arrays (20, 21) is described in the methods section. The raw magnetotransport data is shown in Fig. 2 for five different relative orientations of current, voltage and field and honeycomb as indicated in the schematics. The data were taken at 100K in Figs. 2A-E, 2K in Figs. 2F-I and 25K for the standard Hall in Fig. 2J. The magnetoresistance and Hall components of the magnetotransport were separated (22) by virtue of their symmetry with respect to the applied field. The reversible and irreversible components (respectively the sum and the difference between the forward and backward field sweeps) of the parallel Hall resistance are shown in Fig. 3A and 3B.

All the transport phenomena observed at 100K (and above) are straightforward consequences of the lattice geometry and the standard transport effects of ferromagnetic metals. The 100K MR data in Figs 2A-C is similar to a previous permalloy honeycomb (19) (with two inequivalent current directions, “armchairs” (Fig. 2A, F) and “zigzags” (Fig. 2B, G)). The vertices are complex magnetic structures containing a vertex domain wall (VDW), the magnetization of which can be either parallel or perpendicular to the current. The irreversible magnetoresistance at 100K is simply the anisotropic magnetoresistance (AMR) of ferromagnetic metals, and the magnitude is determined by the number of vertices whose VDW magnetization is perpendicular to the current direction (the low resistance state). The maximum magnitude of the effect ($\Delta R=0.1\Omega$) complements recent measurements on single perpendicular transverse domain walls in a single nanobar ($\Delta R=0.23\Omega$) (23) which is expected as the maximum possible AMR in the array would be one perpendicular VDW per nanobar, and the array is square.

Let us now consider the low temperature effects (Figs. 2F-J). New features emerge in all configurations but are seen most clearly in parallel Hall geometry (Fig. 2I) because the current and field are nominally parallel, eliminating conventional Hall effects. At 100K (Fig 2D) there is zero signal with Hall symmetry, only mixed-in AMR. However at 2K a peak appears in the field range associated with magnetic switching (Fig. 4A). The peak feature is hysteretic (irreversible), and has Hall symmetry. We have observed the onset of a parallel Hall signal in a honeycomb array fabricated from a thin cobalt film from a different source (21), so these are not the results of a specific sample; no parallel Hall component is observed in unpatterned films (24). The symmetrised reversible and irreversible parallel Hall components are plotted vs. field at selected

temperatures in Figs 3A and 3B respectively. The two components have opposite sign, and different magnitude, but their thermal evolution is strikingly similar, the maximum value of each tends to zero at around 50K (Fig 3B, inset). As there is no out-of-plane field, no out-of-plane magnetization and no angle between the field and average current direction, the clear handedness in the transverse deflection of the carriers is direct evidence of spin-chiral asymmetry emerging below about 50K. The hysteretic peak effect mid-transition bears a strong similarity to that observed in the itinerant spin-liquid pyrochlore $\text{Pr}_2\text{Ir}_2\text{O}_7$ when the field is applied along the [111] (kagome ice) direction (18).

To investigate the source of the topological Hall signal, we performed zero temperature micromagnetic simulations using the Object Oriented Micromagnetic Framework (OOMMF) code (21, 25) The simulated arrays were 6*5 hexagons with the same bar dimensions and corrugated edges as the sample (Fig S2). The field was swept from magnetic saturation in the +ve x-direction to negative saturation, and then back to positive saturation in 10mT steps. The simulated MH loop is compared to experiment in Fig 4A and captures the switching fields well. The micromagnetic configurations showed a tendency to form loop states with substantial vector spin chirality at the edges during switching (Figs. 4B-E). The net asymmetry across the whole array is investigated by counting loops at each field point and then defining both a total loop chirality and a spatial loop chirality, shown in Figs. 4F and 4G, respectively. Ferromagnetic switching is a stochastic process so we repeated the full loop with identical input parameters. The randomness in both quantities is much more significant in the small simulated arrays than the experiment. However, whereas the total chirality (Fig 4F) fluctuates randomly around zero, the spatial chirality gives the field asymmetric peak effect (Fig 4G) observed in the

2K parallel Hall resistance data (ΔR , Fig 4H). Magnetic switching occurs by nucleation and propagation of domain walls. Nucleation is favoured at the left and right sample edges. The domain walls tend to propagate along the field direction, rather than moving to the top and bottom edges of the sample, so these edge bars are often the last to switch. This results in clockwise and anticlockwise loops tend to form at the top and bottom edges, and the handedness is inverted in the opposite sweep direction. The sign of the topological Hall effect is such that the loops observed in the backward sweep direction (4d) (C_{bottom} and A_{top}) push carriers near the edges back toward the centre and those in the forward sweep (4e) (C_{top} and A_{bottom}) drive the conduction electrons further toward the edge. The simulation, without finite temperature or structural imperfections simulates magnetic switching sharper than the data.

Micromagnetic simulations offer an explanation of the chiral asymmetry at base temperature, but not of the thermal evolution of the topological Hall components below 50K (Fig 3B, inset). The cobalt Curie temperature, 1400K, is far removed, suggesting the scale is ice related, with the chiral (6) phase transition from Ice I to Ice II a promising candidate. Using the simulated magnetic charge distribution we estimate the VDM of our structure, and so obtain an Ice II transition temperature (6) of approximately 50K (21). Constrained dynamics in artificial spin ice (10, 11, 26) prevent the phase transition, without suppressing its expression in the topological Hall signal, via the irreversible selection of chiral intermediates in the switching process. Recent predictions indicate that constrained dynamics may in fact help support the topological Hall asymmetry (26) The VDM is dependent on bar material (4, 27) dimension and separation in disconnected kagome structures (13, 28) enabling the temperature scale associated with these

topological states to be tuned. We expect our result to be relevant for controlled investigation of other emerging magnetic degrees of freedom such as monopoles (4, 9, 27) loops (6) and topological edge (29, 30) states. We have shown that field-driven dynamics allow thermal scales to be accessed experimentally in a system far from thermal equilibrium, an important step towards application of magnetic nanostructures as model frustrated systems and potential devices based on selection of states, such as neural networks (31).

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Fig. 1: Magnetic vertex configurations and equilibrium phase diagram for artificial spin ice honeycombs. The background (A-D) is a scanning electron micrograph of a small section of the array. (A) The magnetic charge $\pm q = \pm m/l$ dumbbell representation of a magnetic moment m of length l on a honeycomb of lattice parameter a . Deviations from ideal Ising behaviour are characterised by $\varepsilon = 1 - l/a$. (B) Charges on neighbouring dumbbells reside on the circumference of a circle of diameter εa . This is equivalent to (C) a point charge $Q = \sum q$ and a vertex dipole moment (VDM) of magnitude $\varepsilon a \Delta q / 2$ colocated at the vertex centre. ($\Delta q = q_{\max} - q_{\min}$ at the vertex). In connected arrays the local magnetic structure at the vertex minimises the VDM (21). (D) The eight possible configurations of

magnetic moments (black arrows) at a kagome spin ice vertex. Vertices a-f obey the ice rule and have $Q=\pm q$; $VDM=\epsilon a q$ (purple arrows) and spin chirality $\Omega=-1/3$; vertices g and h are ice rule defects with $Q=\pm 3q$; $VDM=0$ and $\Omega=+1(2I)$. (E) The predicted phase diagram for artificial spin ice honeycombs (6, 8) (only the Ice I phase is observed in zero-field. There are four distinct phases: the gas-like Ising paramagnet, the liquid-like short range spin ice (Ice I) phase, the long-range spin ice (Ice II) phase where the near order extends to second nearest neighbour macrospins, giving favoured vertex pairs [ab], [cd] and [ef], and the long range ordered macrospin solid state.

Fig 2: Raw Magnetotransport data. Resistance vs. field with positive (filled squares) and negative (open circles) sweep direction. Data was taken at 100K (A-E) and 2K (F-I) and 25K in J. Schematics of measurement geometries: (K) longitudinal (armchair) MR geometry in A, F; (L) longitudinal (zigzag) MR geometry in B, G; (M) transverse MR geometry in C, H; (N) parallel Hall geometry in D, I and (O) standard Hall geometry (E, J).

Fig. 3: Topological Hall signal and possible sources of spin chirality in the kagome spin ice structure. (A) Reversible Hall symmetry component and (B) irreversible Hall symmetry component of the parallel Hall resistance (R_{PH}) vs. applied field at selected temperatures from 2K to 75K. Inset (B): Temperature dependence of the saturation reversible and peak irreversible parallel Hall resistance.

Fig 4: Spatial Separation of Chirality: (A) Normalised magnetization of honeycomb at 100K vs field swept in the negative direction (black solid squares) and positive direction (red open circles), as determined from longitudinal AMR measurements. Solid lines show the negative (black) and positive (red) MH curves calculated by OOMMF (25) for 6×5 arrays. (B-E) OOMMF simulations at selected point in the hysteresis loop indicated by the straight arrows to Fig. 4A. The magnetization direction of each bar is given by the colour-wheel (inset 4B), clockwise and anticlockwise loops in the magnetization are indicated by blue and red circular arrows respectively. (F) total loop chirality ($\Omega_{total} = A - C$) and (G) spatial loop chirality [$\Omega_{spatial} = (A_{bottom} - A_{top}) - (C_{bottom} - C_{top})$] where C and A are

the number of clockwise and anticlockwise loops respectively at each field in two nominally identical OOMMF simulations. Up sweeps (dashed) have been offset slightly from the down sweeps (solid lines) for clarity. (H) change in 2K parallel Hall resistance ($\Delta R = R_H - R_H(0T)$) vs field in the negative sweep direction (black squares) and positive sweep direction (red circles). The topological Hall effect of the loops observed in (D) (C_{bottom} and A_{top}) pushes carriers near the edges back toward the centre (decreasing R_H) and loops shown in (E) (C_{top} and A_{bottom}) drive carriers into the edge (increasing R_H).

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Supporting Online Material

The Temperature Scale from multipole analysis:

Let us now consider the temperature scale. It is immediately apparent that the structure undergoes some sort of transition at 50K, that can have nothing at all to do with the far higher bulk cobalt Curie temperature ($T_C = 1394\text{K}$). This 50K transition is a property of the nanostructured lattice, not of the unprocessed films. Intriguingly a thermodynamic phase transition between two different ice phases (Ice I – Ice II) was recently predicted(6) with a critical temperature around 50K. The exact phase transition temperature is governed by a parameter ($\varepsilon = 1-l/a$) where a is the vertex separation and l is the coherently magnetised bar length. ε reflects how perfectly the structure mirrors the Ising model, or the smearing of the magnetic charge. An approximation for ε can be obtained geometrically assuming the magnetic charge spreads over the entire vertex, to $w/2\sqrt{3}$ from the vertex centre, giving $\varepsilon_{app} = w/a\sqrt{3} = 0.058$. In fact OOMMF simulations of even an ice-rules vertex suggests the distortion from perfect Ising behaviour spreads twice this far, to $w/\sqrt{3}$ from the vertex centre, giving $\varepsilon_{upper} = 2w/a\sqrt{3} = 0.116$, as illustrated by the larger triangle in Supplementary Fig S1. Note that the net magnetization within the distorted region is antialigned with the vertex dipole moment illustrated in Fig 1B-D. In a nanowire of width w , a lower bound for ε can be approximated from the domain wall thickness t_{dw} where $t_{dw} \sim w(32) = 100\text{nm}$ giving $\varepsilon_{lower} = (t_{dw}/a) \sim 0.1$. In fact in terms of the vertex dipole interaction, the vertex dipole length $l_{vd} \sim w/\sqrt{3} \sim 0.058a$ as shown in Supplementary Fig S1 is the appropriate length (defined as εa in Ref (6)). This illustrates the difficulty of defining ε in this system: where opposite charges meet they are able to

get very close, maximizing the Coulombic attraction, whereas when like charges meet, the tractable nature of the “Ising” spins allows very significant spreading, minimising the Coulombic repulsion. An Ice-Ice transition temperature of 50K ($=0.035T_C$) was predicted for $\varepsilon=0.055$, suggesting the simple geometric model is appropriate. However some consideration must be given as to whether such thermodynamic calculations are appropriate in our system. In zero-field it does not thermalise at all, even at room temperature(4) because of the large coercive fields of the bars. Here we are concerned not with the zero-field behaviour, but with what happens at the point of switching when the applied field approaches the coercive field. At that point thermodynamics play a key role in determining which bar switches during domain wall propagation, and so influences the nature of the field-driven mid-transition states.

Fig S1: The Ice-rule vertex: The “two-in one-out” and “one-in two-out” in-plane structures result in stray fields at the vertices pointing up (light contrast) and down (dark contrast) respectively out of the plane. After saturation in an applied field the MFM pattern there is (almost) perfect order of alternating 1-in-2-out and 2-in-1-out vertices,(4) which corresponds to the unique FM kagome ice structure which is then stable if the field is removed.

Calculating the Vector Spin Chirality

The vector spin chirality for any given pair of spins is $\Omega_v = \mathbf{s}_i \times \mathbf{s}_j$. The handedness at a given vertex, defined as the difference between cyclic and anticyclic permutations(33) of the three spins $\Omega = [(\mathbf{s}_1 \times \mathbf{s}_2 + \mathbf{s}_2 \times \mathbf{s}_3 + \mathbf{s}_3 \times \mathbf{s}_1) - (\mathbf{s}_1 \times \mathbf{s}_3 + \mathbf{s}_3 \times \mathbf{s}_2 + \mathbf{s}_2 \times \mathbf{s}_1)]/3$. As the in-plane angle is always 120° each term is $\pm \sqrt{3}s^2\hat{u}/2$ (where $\hat{u}$ is the unit vector normal to the top surface) and the eight possible vertex configurations (Fig. 1D) have $\Omega = C\sqrt{3}s^2\hat{u}$, $C = -1/3$ in (vertex a-f), and $C = +1$ in (vertex g, h) (34).

Magnetotransport

Three large area cobalt electrical contact pads are connected to each side of the square and these were fabricated in the same lithographic step as the honeycomb. Transport measurements were performed with a four-point technique. The contacts down each of two opposite sides of the square device (see Fig S2) were shorted to form current contacts (note that the two possible directions are not equivalent). For MR measurements, two of the contacts on a third side were used as the voltage probes, for Hall measurements two contacts transverse to the current direction were used as voltage probes. From the temperature dependence of the zero field resistance, we can determine a residual resistivity ratio [RRR = $R(290K) / R(0K)$] of 1.25. The functional form of R vs. T and the RRR are similar to those reported for a 20nm polycrystalline Co film on Si(100)(35) and so we infer that the intrinsic electrical properties of the cobalt were not significantly changed by the processing. Four quadrant longitudinal (armchairs and zigzags), transverse MR and parallel and standard Hall measurements were performed at selected temperatures from 2K to 290K.

Fig S2: SEM micrograph of the full array with electrodes. The stripes in contrast intensity are an artefact of the large area image, and are not observed in higher magnification images.

Reproducibility of the 2K parallel Hall effect in a second sample

To test reproducibility transport of a second cobalt honeycomb nanostructure (Sample 2) was measured. The sputtered cobalt film came from a different source; the nanoarray was fabricated in the same e-beam lithography apparatus, using the same pattern. Sample 2 has a broader magnetic switching transition and a higher coercive field than the one studied in the main report (Sample 1), suggesting that there is stronger pinning from defects in the film. The raw parallel Hall resistance data at 100K (Fig S3A) shows a larger offset resistance and strong mixing of MR effects into the raw Hall data, and again no effect with Hall symmetry. The raw parallel Hall resistance data at 2K (Fig S3B) shows a clear Hall asymmetry and a peak effect is again observed at 2K. The irreversible components of the parallel Hall resistance at 2K are shown for sample 1 (Fig S4A) and sample 2 (Fig S4B). The mixed-in irreversible MR components are shown in (Fig S4C) for sample 1 and (Fig S4D) for sample 2. The sign and shape of all the components are similar in sample 1 and sample 2. The irreversible Hall component is approximately a factor of 4 larger in sample 1, whereas the MR components are of similar magnitude. As the Hall and the mixed-in MR peaks are of similar magnitude at 2K for sample 2, and the raw data is the sum of components in positive field and the difference in negative field, there is large positive peak in positive field and a small positive peak in negative field. In contrast the negative Hall peak in negative field is clearly seen in the raw data in the sample 1 (Fig 2I). The additional “quenched disorder” in sample 2 implies the order in which the bars switch is controlled more by variation in their inherent coercive fields and less by the interbar interactions than in sample 1. We expect this would reduce both the population of loops and the asymmetry in their distribution. Quantitative comparisons of

Hall effects are generally only sensible when using Hall conductivity, which requires exact knowledge of the transport cross section. There would be a very large error in any estimate we could make of the cross sectional area, which may vary strongly between nanostructures if fluctuations in the thickness or edge roughness may be of the order of 1nm and the array thickness is only 18nm.

Fig S3: Raw parallel Hall Resistance vs. Field (as defined in Fig 2) of a second cobalt honeycomb nanostructure at A) 100K and B) 2K.

Figure S4: Irreversible Hall symmetry component of the parallel Hall resistance (R_{PH}) (as defined in Fig 3) vs. applied field at 2K (black squares) and 100K (red open circles) for A) sample 1 and B) sample 2. Irreversible MR symmetry component of the parallel Hall resistance measurement (R_{PH}) (as defined in Fig 3) vs. applied field at 2K (black squares) and 100K (red open circles) for C) sample 1 and D) sample 2.

Modelling

The OOMMF software (25) solves the Landau-Lifshitz equation at each point on a rectangular mesh:

$$\frac{d\mathbf{M}}{dt} = -\gamma(\mathbf{M} \times \mathbf{H}_{eff}) + \alpha(\mathbf{M} \times \frac{d\mathbf{M}}{dt})$$

Where $\mathbf{H}_{eff} = -\nabla_{\mathbf{M}} E$ and E is the free energy which includes terms for the Zeeman, demagnetization, and the anisotropy energies, γ is the gyromagnetic ratio and α is the damping parameter. A cell size of 5nm was used to construct the mesh, and the magnetization was allowed to relax in the absence of an applied magnetic field in order to find a minimum energy configuration. The magnetocrystalline anisotropy of cobalt was assumed to be zero, the exchange stiffness was taken to be $1.3 \times 10^{-11} \text{ Jm}^{-1}$, and the saturation magnetization was assumed to be 1400 emu/cm^3 .