Relation between microscopic interactions and macroscopic properties in ferroics

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The driving force in materials to spontaneously form states with magnetic or electric order is of fundamental importance for basic research and device technology. The macroscopic prop-2 erties and functionalities of these ferroics depend on the size, distribution and morphology 3 of domains, that is, of regions across which such uniform order is maintained¹. Typically, 4 extrinsic factors like strain profiles, grain size or annealing procedures control the size and 5 shape of the domains²⁻⁵, whereas intrinsic parameters are often difficult to extract due to the 6 complexity of a processed material. Here, we achieve this separation by building artificial 7 crystals of planar nanomagnets that are coupled by well-defined, tunable, and competing 8 magnetic interactions⁶⁻⁹. Aside from analysing the domain configurations, we uncover fun-9 damental intrinsic correlations between the microscopic interactions establishing magneti-10 cally compensated order and the macroscopic manifestations of these interactions in basic 11 physical properties. Experiment and simulations reveal how competing interactions can be 12 exploited to control ferroic hallmark properties such as the size and morphology of domains, 13 topological properties of domain walls, or their thermal mobility. 14

¹⁵ Domain formation is key to the functional properties of ferroics. In ferromagnets, which until now ¹⁶ represent the class of ferroics technologically most relevant, domain formation is mainly a result of ¹⁷ magnetostatic-energy minimisation^{2, 10}. Currently, however, compensated types of magnetic order ¹⁸ such as antiferromagnetism and ferrotoroidicity are gaining attention with a view to an advanced ¹⁹ memory or spintronic technology¹¹⁻¹⁴. Due to their zero net magnetisation, domain formation is

no longer a consequence of the demagnetising field, which means that less understood, yet fun-20 damental factors of equal importance come to the fore. While thermodynamics can make general 21 macroscopic predictions about domain formation, it does not reveal the underlying microscopic 22 mechanisms for this formation. Therefore, the link between the microscopic interactions and the 23 macroscopic physical properties remains largely unclear. Unfortunately, extrinsic effects such as 24 the strain distribution, magnetoelastic interactions or annealing protocols^{4,5,15} compete with the 25 sought-after intrinsic effects, making the separation of the latter from the former challenging. For 26 the identification and control of the intrinsic coupling mechanisms, a magnetically compensated 27 system with tunable competing microscopic interactions that dominate over the extrinsic contribu-28 tions is therefore required. 29

This we accomplish by fabricating artificial two-dimensional arrays of nanomagnets that establish 30 a magnetically compensated ferroic order¹⁶. The transfer from atomic to sub-micrometre length 31 scales requires the replacement of the quantum-mechanical exchange interaction by the classi-32 cal magnetic-dipole interaction as the basis for the ferroic order¹⁷⁻¹⁹. This approach to scaling 33 up yields key advantages such as direct experimental access to the magnetic state, and the possi-34 bility to tailor and implement spin-spin interactions with a degree of control that natural materi-35 als cannot offer. Such artificial spin systems have already proven their ability to answer general 36 questions about fundamental magnetic properties, including magnetic correlations²⁰, frustration²¹, 37 emergent magnetic monopoles^{22,23}, thermal fluctuations^{24,25}, phase transitions²⁶ and relaxation 38 behaviour^{27,28}. Most of these systems are artificial spin ices without macroscopically distinguish-39 able order-parameter-related domain states and parametrised by a single microscopic interaction. 40 The few nanomagnetic systems utilising magnet arrangements with multiple and potentially com-41 peting couplings^{9,29,30} do not exhibit a ferroic order parameter either. Recent work, however, has 42 shown that the so-called toroidal square array exhibits a magnetically compensated long-range-43 ordered ground state characterised by the toroidisation as a primary ferroic order parameter (see 44 Methods) and distinct domain states¹⁶. 45

Here we make use of the toroidal square array comprised of parallel and orthogonal pairs of nanomagnets, with each magnet carrying an in-plane magnetic moment **m** along the long axis as shown in Fig. 1a. Due to the anisotropy of the magnetic-dipole coupling, such an arrangement promotes two interactions J_{\perp} and J_{\parallel} between orthogonal and parallel nearest neighbours, respectively (see Methods and Fig. 1). These two interactions can be seen as classical magnetic-dipole-based analogues to antisymmetric and symmetric exchange interactions as fundamental ingredients determining the magnetic order in materials.

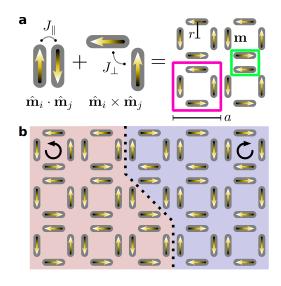


Figure 1: Model of a ferroic crystal with zero net magnetisation and two competing microscopic interactions. a, Two-dimensional arrangement of magnetic moments ($\mathbf{m} = |\mathbf{m}| \hat{\mathbf{m}}$, yellow arrows), coupled via magnetic-dipole interactions that are classical analogues of antisymmetric (J_{\perp}) and symmetric (J_{\parallel}) exchange interactions. Note that the the coupling constants $(J_{\perp}, J_{\parallel})$ are each represented by one of the two geometric units (r, a) parametrising the structure of the system (see Methods). Outlines: Subunits formed by four nanomagnets (octupoles, magenta frame) and two nanomagnets (quadrupoles, green frame) are promoted by J_{\perp} and J_{\parallel} , respectively. **b**, Ferroic order of the two-dimensional array with domain states that are distinguished by the uniform handedness of the unit cells outlined in magenta in (a): clockwise (blue shading) and counter-clockwise (red shading). A domain wall is indicated by the black dotted line.

⁵³ A possible periodic arrangement resulting from the combination of J_{\perp} and J_{\parallel} is shown in Fig. 1b. ⁵⁴ It is one of the simplest systems that allows us to study the consequences of the competition ⁵⁵ between two intrinsic microscopic interactions for the macroscopic properties of a ferroic state ⁵⁶ with zero net magnetisation. For focusing on the most fundamental consequences, we refrain from ⁵⁷ adding further ingredients, such as additional magnetic-anisotropy contributions, to our model. ⁵⁸ The particular spin arrangement in Fig. 1 can be effectively modelled with a Hamiltonian derived ⁵⁹ from a point-dipole approximation (see Methods), namely

$$\mathcal{H} = J_{\perp} \sum_{\langle i,j \rangle_{\perp}} \hat{\mathbf{D}}_{ij} \cdot (\hat{\mathbf{m}}_i \times \hat{\mathbf{m}}_j) + J_{\parallel} \sum_{\langle i,j \rangle_{\parallel}} \hat{\mathbf{m}}_i \cdot \hat{\mathbf{m}}_j \quad , \tag{1}$$

where $\hat{\mathbf{m}}_{i,j}$ are magnetic-moment unit vectors of neighbouring nanomagnets at lattice sites i, j

and $\hat{\mathbf{D}}_{ij}$ is the classical representation of the Dzyaloshinskii-Moriya vector. We can then explore how the relation between J_{\perp} and J_{\parallel} in Eqn. (1) controls characteristic macroscopic observables of ferroic order such as the domain size or domain-wall morphology.

First, equilibrium Monte-Carlo simulations are used to calculate the phase diagram in Fig. 2 as a 64 function of J_{\perp}/J_{\parallel} using simulated annealing, starting from high temperatures and then decreas-65 ing the temperature to well below the ordering temperature T_c of the array. In the upper part of 66 Fig. 2a,b, where $J_{\perp} \gg J_{\parallel}$, the dominance of J_{\perp} promotes the alignment of four magnetic moments, 67 as shown in Fig. 1a, to form a magnetic flux-closure state that mimics the magnetic-field config-68 uration of an octupole. The octupole population density that saturates well above $T_{\rm c}$ implies that 69 thermal fluctuations of single-nanomagnet dipoles are replaced by fluctuations of four-nanomagnet 70 octupoles. The interaction between octupoles drives the phase transition to long-range order, now 71 defined by the weaker coupling parameter J_{\parallel} . Likewise, in the lower part of Fig. 2a,b, where 72 $J_{\perp} \ll J_{\parallel}$, the ordering process is dominated by J_{\parallel} such that pairs of neighbouring parallel moments 73 (see Fig. 1a) form fluctuating two-nanomagnet quadrupoles above T_c. Their long-range ordering is 74 then driven by the weaker coupling parameter J_{\perp} . In the centre part of Fig. 2, where $J_{\perp} \approx J_{\parallel}$, the 75 nanomagnets continue to fluctuate individually as the temperature is reduced to $T_{\rm c}$ with the phase 76 transition being characterised by a simultaneous increase of octupole and quadrupole density near 77 $T_{\rm c}$, see Fig. 2b. Hence, systems with different J_{\perp}/J_{\parallel} ratios differ in their short-range order above 78 $T_{\rm c}$ and thus in their pathways towards long-range order on reducing the temperature. Note that all 79 three pathways lead to the same long-range ordered ground state, so that the macroscopic physical 80 properties of the system are not affected by symmetry changes, but exclusively by the interplay of 81 the two microscopic coupling parameters J_{\perp} and J_{\parallel} . 82

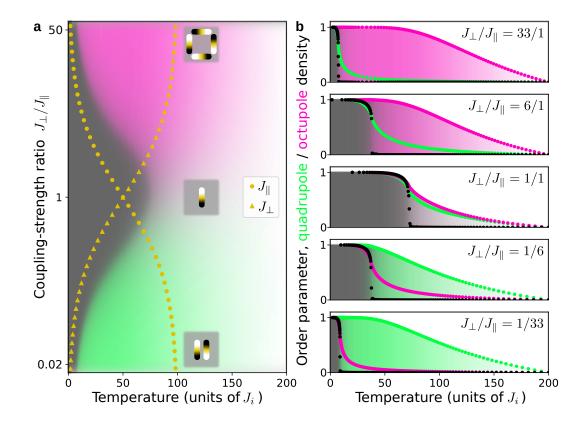


Figure 2: Phase diagram revealing the correlation between short-range and long-range order. a, MC-simulated phase diagram of the model Hamiltonian in Eqn. (1) for different couplingstrength ratios J_{\perp}/J_{\parallel} . The two varying coupling strengths are represented by yellow triangles and circles, respectively. The regions dominated by the octupole- and quadrupole-subunit formation (see text) are shaded magenta and green, respectively. Grey shading indicates the long-rangeordered ground state. b, Temperature dependent short- and long-range order for five exemplary choices of J_{\perp}/J_{\parallel} . The multipole-population densities in (a) and (b) are reflected by the intensity of the colour shading.

To study the impact of the short-range order on the formation of domains, we employ kinetic 83 Monte-Carlo (kMC) simulations and perform a temperature quench from $T \gg T_c$ to $T \ll T_c$. 84 Under identical cooling conditions, we obtain the non-equilibrium multi-domain configurations 85 shown in Fig. 3a for five values of J_{\perp}/J_{\parallel} corresponding to the five values shown in Fig. 2b. The 86 domain size reaches a maximum for $J_\perp \approx J_\parallel$ and decreases continuously with an increasing 87 imbalance of J_{\perp} and J_{\parallel} . While the relationship between the ordering temperature and the domain 88 size might be concluded from a thermodynamics point of view, here we see explicitly how the 89 competition between the exchange coupling constants, an intrinsic microscopic factor, controls the 90

⁹¹ domain size, which is a macroscopic property of a magnetically compensated ordered state.

We verify this insight experimentally by growing a series of two-dimensional arrays composed of 92 sub-micrometre-sized single-domain ferromagnetic building blocks with the magnetisation point-93 ing along the long axis as schematically shown in Fig. 1 (see Methods). We tune J_{\perp} and J_{\parallel} by 94 varying the lateral spacing between the magnets while conserving the size of the four-magnet unit 95 cell (see Fig. 3b). Although the arrays are thermally inactive at room temperature, they are in a 96 superparamagnetic state during growth before their increasing thickness suppresses fluctuations so 97 that the magnetic configuration freezes 31,32 . The resulting domain configurations are imaged by 98 magnetic force microscopy (see Methods). 99

The agreement between the simulated and the measured domain configurations in Figs. 3a and 3c, 100 in terms of size and morphology, is remarkable. Note that, for $J_{\perp} \gg J_{\parallel}$, domain walls align 101 horizontally and vertically between strongly-coupled four-nanomagnet octupoles (left panel in 102 Fig. 3d,e). In a similar way, for $J_{\perp} \ll J_{\parallel}$, domain walls run diagonally between strongly-coupled 103 two-nanomagnet quadrupoles (right panel in Fig. 3d,e). For $J_{\perp} \approx J_{\parallel}$ (centre panel in Fig. 3d,e), 104 a combination of both wall types is observed. Along with the preferential alignment of a domain 105 wall, the handedness of the magnetisation within a wall can be tuned from equal $(J_{\perp} \gg J_{\parallel})$ to 106 opposite $(J_{\perp} \ll J_{\parallel})$ with respect to the magnetic handedness of the enclosed domain as shown in 107 Fig. 3d. A particularly interesting case occurs for $J_{\perp} \approx J_{\parallel}$, where the direction of magnetisation 108 along the wall alternates due to the competition between the two domain-wall types. Here the 109 walls as such can be regarded as one-dimensional ferromagnetic multi-domain entities with head-110 to-head or tail-to-tail meeting points of magnetic moments, similar to so-called Bloch points, also 111 described as emergent magnetic charges^{22,23,29,33} as indicated in the centre panel of Fig. 3e. 112

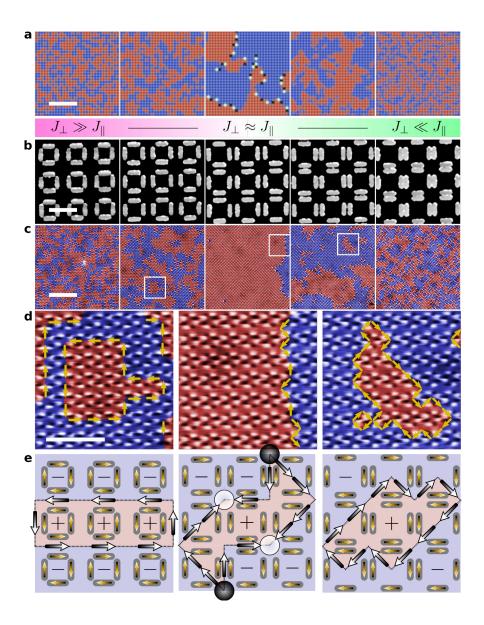


Figure 3: **Correlation between short-range order and domain formation.** a, MC-simulated multi-domain states obtained by a temperature quench (see Methods) for $J_{\perp}/J_{\parallel} = [33/1, 6/1, 1/1, 1/6, 1/33]$ (from left to right). Scalebar: 10 µm. b, Atomic-force-microscopy images of permalloy nanoarrays on silicon. The geometric variation across the series reflects the variation of J_{\perp} and J_{\parallel} as defined in Eqs. 5 and 6. Scalebar: 1 µm. c, Magnetic-force-microscopy images of the corresponding as-grown domain patterns with opposite domain states in blue and red, see Fig. 1. Scalebar: 10 µm. d, Magnified view of the regions outlined by white frames in (c). The local direction of magnetisation within the domain walls is shown with yellow arrows. Scalebar: 3 µm. e, Schematics of the domain-wall configurations for $J_{\perp} \gg J_{\parallel}$ (left), $J_{\perp} \approx J_{\parallel}$ (centre) and $J_{\perp} \ll J_{\parallel}$ (right). For $J_{\perp} \approx J_{\parallel}$, head-to-head and tail-to-tail meeting points of magnetic moments result in emergent magnetic charges (black- and white-shaded discs).

We now use the kMC-simulated multi-domain configurations to predict further relations between 113 the microscopic coupling strength and macroscopic ferroic properties such as the density of emer-114 gent magnetic charges, which are known to affect the domain-wall mobility², and the associated 115 spin-spin autocorrelation time τ (see Methods). As shown in Fig. 4, a significant number of emer-116 gent magnetic charges are obtained around $J_{\perp} \approx J_{\parallel}$. Our next step is therefore to see if, depending 117 on J_{\perp}/J_{\parallel} , the path via which the system enters the ferroic phase (see Fig. 2) has an impact on the 118 thermal domain-wall mobility, which would have direct consequences for the material's coercivity 119 and remanence. To parametrise this domain-wall mobility, we use kMC simulations to calculate τ 120 at 0.95 $T_{\rm c}$ with the result shown in Fig. 4. The corresponding spin-relaxation rate τ^{-1} closely fol-121 lows the emergent-magnetic-charge density. For $J_{\perp} \gg J_{\parallel}$ and $J_{\perp} \ll J_{\parallel}$, a collective switching of 122 the short-range-ordered octupolar or quadrupolar subunits is required. The high energy barrier for 123 this process results in a decreased thermal mobility of the domain wall. For $J_{\perp} \approx J_{\parallel}$, however, the 124 lack of short-range order above T_c results in comparable excitation energies for both microscopic 125 interactions and thus in an increased density of emergent magnetic charges and a high domain-wall 126 mobility. Hence, both spatial and temporal macroscopic measures are closely linked to the com-127 peting short-range orders so that the coupling-strength ratio can be used to control them. Note that 128 the J_{\perp}/J_{\parallel} dependencies in Fig. 4 are asymmetric with the peaks shifted towards $J_{\perp} < J_{\parallel}$. This 129 reflects the different switching-energy barriers of the octupolar and quadrupolar subunits due to 130 the different number of magnetic moments involved. 131

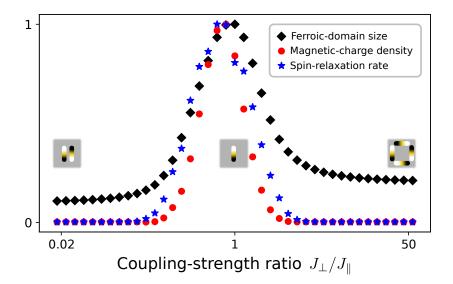


Figure 4: Macroscopic physical properties determined by the microscopic interactions. The average domain size (here parametrised as the mean distance between domain walls), the emergent-magnetic-charge density and the spin-relaxation rate τ^{-1} as a function of J_{\perp}/J_{\parallel} . All plots are normalised with respect to their maximum value. The asymmetry and shift of the peaks with respect to $J_{\perp} = J_{\parallel}$ is caused by the different energy barrier for switching the quadrupolar and octupolar short-range-ordered subunits (see text).

In conclusion, we have demonstrated, with excellent agreement between simulation and experi-132 ment, how the competition between intrinsic microscopic interactions can be used to control the 133 macroscopic properties of a ferroic state. In our simple artificial ferroic system with compensated 134 magnetic order, the competition of two short-range interactions determines hallmark properties of 135 the ferroic state, such as the morphology and size of the domains, and the structure and mobility of 136 the domain walls. Nanomagnetic arrays constitute a versatile platform to regulate the ferroic state 137 because coupling strengths can be introduced and manipulated at will and the resulting magnetic 138 configurations can be experimentally accessed with high spatial resolution. This important insight 139 into the relationship between microscopic and macroscopic aspects in ordered systems with no 140 net magnetisation is relevant given the current interest in antiferromagnetic spintronics as route 141 towards faster and more robust digital memory as well as for neuromorphic computing¹²⁻¹⁴. 142

143 Methods

Effective Model. For our model, we parametrise the magnetic dipole-dipole interaction in the 144 framework of magnetic exchange interactions with the purpose to retain the link to real materials 145 where the exchange interactions dominate the magnetic behaviour. Note that we aim here to use 146 the simplest possible system that is sufficient to simulate ferroic order without a net magnetisa-147 tion and that permits us to relate intrinsic microscopic interactions to macroscopic phenomena of 148 ferroic states such as domain and domain-wall formation, and the associated basic physical prop-149 erties. Therefore, we do not consider magnetic-dipole interactions beyond nearest-neighbours and 150 higher-order multipole interactions because these would merely yield quantitative changes without 151 significant qualitative differences in the results. 152

This leads us to the array shown in Fig. 1. For this particular design, we choose the unit cell outlined by a magenta square and define its center as the origin. From here, we identify two relevant distances: r, the distance from the center of the unit cell to the nearest nanomagnet and a, the lattice constant. We represent each nanomagnet as a point dipole \mathbf{m}_i at lattice site i of equal magnetisation $m \equiv |\mathbf{m}_i|$ pointing along the long axis. The interaction energy E_D with its jth neighbouring magnetic dipole \mathbf{m}_j is given by

$$E_{\rm D} = \frac{\mu_0}{4\pi} \left(\frac{(\mathbf{m}_i \cdot \mathbf{m}_j)}{|\mathbf{r}_{ij}|^3} - \frac{3 \left(\mathbf{m}_i \cdot \mathbf{r}_{ij} \right) (\mathbf{m}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5} \right) \quad , \tag{2}$$

where $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ is the distance between dipoles at positions $\mathbf{r}_{i,j}$. We first evaluate Eqn. (2) for orthogonal nearest neighbours, where the first term becomes zero because of $\mathbf{m}_i \perp \mathbf{m}_j$. Using $\mathbf{m}_i \perp \mathbf{r}_i$ and $\mathbf{m}_i \parallel \mathbf{r}_j$ yields

$$E_{\perp} = \frac{3\mu_0}{4\pi |\mathbf{r}_{ij}|^5} ((\mathbf{m}_i \cdot \mathbf{r}_j)(\mathbf{m}_j \cdot \mathbf{r}_i) + (\mathbf{m}_i \cdot \mathbf{r}_i)(\mathbf{m}_j \cdot \mathbf{r}_j)) \quad , \tag{3}$$

where the second product in the parenthesis is zero. By subtracting this product twice and using
the Binet-Cauchy identity, Eqn. (3) can be rewritten as a vector product:

$$E_{\perp} = \frac{3\mu_0}{4\pi |\mathbf{r}_{ij}|^5} ((\mathbf{m}_i \times \mathbf{m}_j) \cdot (\mathbf{r}_i \times \mathbf{r}_j)) \quad .$$
(4)

Using $|\mathbf{r}_i| = |\mathbf{r}_j| = r$, $|\mathbf{r}_{ij}| = \sqrt{2}r$, $|\mathbf{m}_i| = m$, and defining $\hat{\mathbf{D}}_{ij} = \hat{\mathbf{r}}_i \times \hat{\mathbf{r}}_j$ with $\hat{\mathbf{r}}_{i,j}$ and $\hat{\mathbf{m}}_{i,j}$ as unit vectors, gives

$$E_{\perp} = J_{\perp} \,\hat{\mathbf{D}}_{ij} \cdot (\hat{\mathbf{m}}_i \times \hat{\mathbf{m}}_j) \quad \text{with} \quad J_{\perp} \equiv \frac{3\mu_0 m^2}{16\pi\sqrt{2}r^3} \quad , \tag{5}$$

which is mathematically equivalent to an antisymmetric exchange interaction with J_{\perp} as the coupling strength. For the case of parallel nearest neighbours, the second term in E_D becomes zero because $\mathbf{m}_i \perp \mathbf{r}_{ij}$. Using $|\mathbf{r}_{ij}| = 2(a - r) \equiv d$ yields

$$E_{\parallel} = J_{\parallel} \hat{\mathbf{m}}_i \cdot \hat{\mathbf{m}}_j \quad \text{with} \quad J_{\parallel} \equiv \frac{\mu_0 m^2}{4\pi d^3} \quad ,$$
 (6)

which is mathematically equivalent to a symmetric exchange interaction with J_{\parallel} as the coupling transformation strength.

- Hence, the two coupling constants $(J_{\perp}, J_{\parallel})$, whose competition determines the physical properties
- of the ferroic system, are each represented by one of the two geometric units (r, d) parametrising
- the structure of the system.

Monte-Carlo simulations. We use a lattice of $N = 100 \times 100$ unit cells with periodic boundary conditions and represent the magnetisation of each nanomagnet as a bidirectional classical spin, coupled to its neighbours with interaction strengths J_{\perp} and J_{\parallel} , as detailed above. We define temperature in units of the coupling strengths and therefore set $k_B = 1$. We tune J_{\perp} and J_{\parallel} while keeping the sum $J_{\perp} + J_{\parallel}$ constant, which we achieve by setting

$$J_{\perp} = J_0(1 - \tanh\left(\delta\right)) \quad \text{and} \tag{7}$$

$$J_{\parallel} = J_0(1 + \tanh(\delta)) \quad , \tag{8}$$

where the amplitude J_0 represents an arbitrary scaling factor and and δ determines the ratio between the interactions. Choosing $J_0 = 50$ and $\delta \in \{-2.0, -1.9, ..., 1.9, 2.0\}$ we obtain the 41 coupling strengths used in Fig. 2, which cover all three different pathways to the ground state (see main text). In order to distinguish between the three pathways, we introduce the octupole population density O and the quadrupole population density Q as follows

$$O = \left\langle \frac{1}{4N} \sum_{u=1}^{N} \left| \sum_{j=1}^{4} \hat{\mathbf{r}}_{j} \times \hat{\mathbf{m}}_{u,j} \right| \right\rangle \quad \text{and}$$
(9)

$$Q = \left\langle \frac{1}{16N} \sum_{u=1}^{N} \sum_{j=1}^{4} \left| \hat{\mathbf{m}}_{u,j} - \hat{\mathbf{m}}_{u,j}^{\parallel} \right| \right\rangle \quad , \tag{10}$$

where $\langle ... \rangle$ indicates the statistical average, \mathbf{r}_j denotes the position vector of the *j*-th nanomagnet within a unit cell, while the index *u* runs over all unit cells. Furthermore, $\mathbf{m}_{u,j}^{\parallel}$ indicates the magnetisation of the parallel nearest neighbour within the adjacent unit cell of the nanomagnet $\mathbf{m}_{u,j}$. The amplitude of the order parameter of the system¹⁶ is defined as

$$\mathcal{T} = \langle \frac{1}{4N} | \sum_{u=1}^{N} \sum_{j=1}^{4} \hat{\mathbf{r}}_{j} \times \hat{\mathbf{m}}_{u,j} | \rangle \quad .$$
(11)

The equilibrium MC simulations displayed in Fig. 2 are obtained using the Metropolis algorithm 179 with simulated annealing. For each ratio of J_{\perp}/J_{\parallel} , we start at $T = 4J_0$ and decrease the tem-180 perature as described below. At each temperature, we perform 10^3 Metropolis sweeps to reach 181 thermal equilibrium, and then perform another 10^4 sweeps to average the observables Q, O and 182 \mathcal{T} . We calculate the specific heat C_v and decrease the temperature in steps of $\Delta T = 2 \cdot T / \sqrt{C_v}$, 183 which allows us to increase the density of data points where the observables vary most. We repeat 184 this procedure until we reach the ground state characterised by $\mathcal{T} = 1$. The four-spin octupolar 185 and two-spin quadrupolar subunits at $J_{\perp} \gg J_{\parallel}$ and $J_{\perp} \ll J_{\parallel}$, respectively, cost significant energy 186 to break up, so that we introduce collective flips of the magnetisation of these subunits into our 187 simulation. This allows the system to efficiently find its ground state with decreasing temperature. 188

All out-of-equilibrium simulations (Fig. 3a and Fig. 4) are performed using a kinetic Monte-Carlo (kMC) algorithm with the so-called *n*-fold way introduced by Bortz, Karlos and Lebowitz³⁴. When generating the multi-domain patterns shown in Fig. 3a, we start at $T = 4J_0$ and rapidly decrease the temperature below T_c in order to prevent the system from reaching equilibrium. We extract the resulting domain configuration, the emergent-magnetic-charge density and the average distance between domain walls with the results shown in Fig. 4, where all quantities are averaged over 10^3 independent runs. To calculate the autocorrelation time τ , defined as

$$\tau = \int_0^\infty \frac{G(t)}{G(0)} dt \quad , \tag{12}$$

we create an equilibrium state at $0.95 T_c$. This relative temperature, in contrast to an absolute temperature, results in a comparable thermal excitation for all systems. We use kMC to allow this equilibrium state to evolve and calculate the spin-autocorrelation function $G(t) = \langle \hat{\mathbf{m}}_i(0) \cdot \hat{\mathbf{m}}_i(t) \rangle$. We extract G(t) after each time step and determine its average over the entire system. Finally, we determine the average of τ over 100 runs.

Sample fabrication. Using electron-beam lithography, the different two-dimensional arrays of 201 nanomagnets were patterned on a single silicon-(100) substrate. Subsequently, a 12-nm-thick 202 permalloy (Ni₈₁Fe₁₉) film was deposited simultaneously on all patterns by electron-beam evap-203 oration with a growth rate of 3 Å/min and capped with a 4-nm-thick film of gold to prevent ox-204 idation. Finally, unwanted material was removed in an ultrasonic-assisted lift-off process. All 205 patterned arrays measure $55 \times 55 \,\mu\text{m}^2$. Four stadium-shaped nanomagnets with lateral dimensions 206 $450 \times 150 \text{ nm}^2$ form the artificial unit cell of our square lattice with $a = 1 \,\mu\text{m}$ lattice period, see 207 Fig. 1. The nanomagnet aspect ratio of 3:1 and the choice of permalloy as a soft magnetic mate-208 rial ensure the formation of single-domain macrospins with magnetic moments pointing parallel 209 (or antiparallel) to the nanomagnet's long edge. To tune the strength of the pairwise interactions 210

 J_{\perp} and J_{\parallel} , the structural parameter r is modified to vary the spacing between parallel neighbouring nanomagnets from 32 nm up to 257 nm and, accordingly, between perpendicular neighbouring magnets from 216 nm down to 57 nm.

Micromagnetic imaging. Magnetic imaging was performed using an NT-MDT NTEGRA magnetic force microscope in semi-contact mode with a lift height of 45 nm. We probed the magnetic configurations by scanning the nanomagnetic arrays with a Nanosensors PPP-LM-MFMR magnetic tip. Raster scans with a 10 μ m/s tip velocity along the fast axis and line-to-line spacings of 30 nm were applied for scanning areas of $30 \times 30 \mu$ m² within the array.

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- **Data availability** The data that support the Figures and other findings of this study can be found here (doi:10.3929/ethz-b-000429489).
- Code availability The program codes that support the Figures and other findings of this study can
 be found here (doi:10.3929/ethz-b-000429490). Additional data and information is available from
 the corresponding authors upon reasonable request.

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³⁰⁵ Competing Interests The authors declare that they have no competing financial interests.

306 Additional Information

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