

Topological Boundary Constraints in Artificial Colloidal Ice

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The effect of boundaries and how these can be used to influence the bulk behavior in geometrically frustrated systems are both long-standing puzzles, often relegated to a secondary role. Here, we use numerical simulations and “proof of concept” experiments to demonstrate that boundaries can be engineered to control the bulk behavior in a colloidal artificial ice. We show that an antiferromagnetic frontier forces the system to rapidly reach the ground state (GS), as opposed to the commonly implemented open or periodic boundary conditions. We also show that strategically placing defects at the corners generates novel bistable states, or topological strings, which result from competing GS regions in the bulk. Our results could be generalized to other frustrated micro- and nanostructures where boundary conditions may be engineered with lithographic techniques.

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In the thermodynamic limit, the bulk properties of a statistical ensemble are no longer influenced by its boundaries. However, in frustrated spin systems, the boundaries can induce configurations that propagate far into the bulk [1,2]. Among several examples of frustrated systems in nature, the most representative one is spin ice [3–5], which can be considered the magnetic “analog” of the water ice [6]. Artificial spin ice systems (ASIs) based on lithographic engineering recently emerged as a versatile experimental platform to investigate geometric frustration effects [7]. An ASI is composed by a lattice of nanoscale ferromagnetic islands, arranged to induce frustration [8–10]. In contrast to natural magnets, an ASI allows one to directly visualize the spin arrangement, a feature that has been used to investigate the effect of disorder [11–13], thermalization [14,15], and degeneracy in many geometries [16–20]. Alternative realizations include arrays of nanowires [21], patterned superconductors [22,23], macroscopic magnets [24], Skyrmions in liquid crystals [25,26], superconducting qubits [27], and colloidal particles in bistable potentials [28].

In such systems, the presence of disorder or a finite temperature often prevents them from reaching the ground state (GS), and, instead, they fall to a metastable state containing defects in the form of charged vertices. These defects can be characterized by a topological charge Q and have a topological nature, since they can be destroyed only when annihilating with other defects of opposite charge. In the GS, the vertices satisfy the ice rule that prescribes a minimization of the local charge, $Q = 0$. While much attention has been placed on how temperature or external fields drive a system toward its GS, the role of boundaries in finite systems has been often overlooked. This is of especial importance when dealing with interacting

magnetic systems where the interaction energies are governed by long-range dipolar forces.

Here, we show how boundaries can be engineered to control the bulk behavior and the formation of topological states such as point defects and topological domain walls spanning the bulk. We demonstrate this concept with an artificial colloidal ice, a system that recently emerged as a microscale soft-matter analog to ASI [29]. Colloidal ice consists of an ensemble of paramagnetic colloids two-dimensionally (2D) confined by gravity in topographic double wells, where the particles may sit in two stable positions and an external magnetic field \mathbf{B} induces repulsive dipolar interactions [Fig. 1(a)]. One can assign a vector (analogous to a spin) to each well such that it points toward the vertex’s center (spin in) or away from it (spin out); see Fig. 1. When arranged in a square lattice, one can classify six possible vertex types, each of them with an associated topological charge $Q = 2n - c_N$, where n is the number of particles in and c_N the lattice coordination number. For the square, it is $c_N = 4$. Thus, vertices of type III and type IV have $Q = 0$ and fulfill the ice rule, and type III gives rise to the GS. Topological defects are charged vertices with $Q \neq 0$ or closed loops of type-IV vertices [Fig. 1(e)].

To simulate colloidal ice, we perform Brownian dynamics, carefully parametrized to mimic the experiments [28]. We consider a 2D array of double wells, each filled by one paramagnetic colloid of diameter $d = 10.3 \mu\text{m}$ and magnetic volume susceptibility $\chi = 0.048$. The overdamped equation of motion for one colloid at position \mathbf{r}_i is

$$\gamma \frac{d\mathbf{r}_i}{dt} = \mathbf{F}_i^T + \mathbf{F}_i^{\text{dd}} + \boldsymbol{\eta}, \quad (1)$$

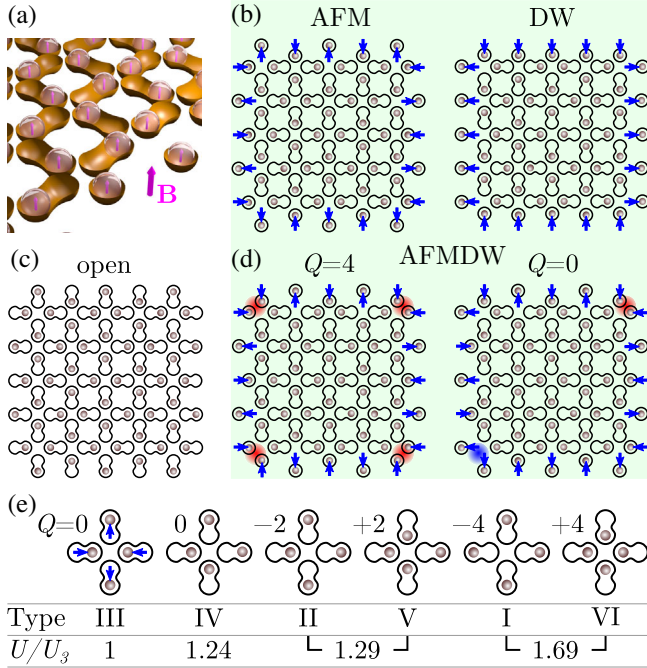


FIG. 1. (a)–(e) Different schematics showing (a) the double-well geometry with paramagnetic colloids and the method of fixing the boundaries by using a single well; (b),(c) the three basic types of boundary conditions, antiferromagnetic (AFM), domain wall (DW), and (c) open boundaries; and (d) AFMDW boundaries designed with defects which can carry a net charge ($Q = 4$, left) or can be neutral ($Q = 0$, right). The shaded region indicates boundaries which topologically protect the enclosed charge. (e) Different vertex types with their effective normalized energetic weight (bottom) and topological charge (side). Vertices are ordered by increasing energetic ratio respect to type III. The associated spins are shown on the type-III vertex.

where $\gamma = 0.032 \text{ pN s } \mu\text{m}^{-1}$ and \mathbf{F}_i^T is the force from the double well which is modeled as a bistable harmonic potential; more details are in the Supplemental Material [30]. The dipolar force acting on particle i due to the neighboring colloids is $\mathbf{F}_i^{\text{dd}} = (3\mu_0/4\pi) \sum_{j \neq i} (\mathbf{m}^2 \hat{\mathbf{r}}_{ij} / |\mathbf{r}_{ij}|^4)$, where $\mathbf{m} = \chi V \mathbf{B} / (\mu_0)$ is the dipole moment induced by the external field \mathbf{B} , $\mu_0 = 4\pi \times 10^{-7} \text{ H/m}$, and $\hat{\mathbf{r}}_{ij} = (\mathbf{r}_i - \mathbf{r}_j) / |\mathbf{r}_i - \mathbf{r}_j|$. To consider long-range dipolar interactions between the particles, we apply a large cutoff of $200 \mu\text{m}$. Finally, $\boldsymbol{\eta}$ represents a random force due to thermal fluctuation, with zero mean, $\langle \boldsymbol{\eta} \rangle = 0$ and delta correlated, $\langle \boldsymbol{\eta}(t) \boldsymbol{\eta}(t') \rangle = 2k_B T \gamma \delta(t - t')$, with a temperature $T = 300 \text{ K}$. Simulations are performed for different system sizes, ranging from $L = 3$ to $L = 40$, where L is the number of vertices along the side. We increase B linearly up to $B = 25 \text{ mT}$, at a rate 0.03125 mT/s . For $B = 25 \text{ mT}$, the pair potential between two particles in the closest (farthest) place of two double wells is $U = 1176k_B T$ ($U = 115k_B T$). Our strategy to fix the boundary consists in placing a particle in a single harmonic well at a location such that it corresponds to a spin pointing in or out.

We consider four different situations: two fixed boundary conditions, namely, antiferromagnetic (AFM) and domain wall (DW), illustrated in Fig. 1(b). In AFM boundaries, colloids are placed alternately pointing in and out. However, flipping a subset of the colloids in an AFM boundary can create defects that are topologically constrained to the inner region, as illustrated in Fig. 1(d). This is the basis of the Gauss' law analog introduced in Ref. [27] for a qubit system. As constructed, the AFM state has a neutral charge at the boundaries. This charge neutrality is broken when a spin is changed from out to in and two defects are created on the AFM state. With this strategy, we introduce in Fig. 1(d) the antiferromagnetic domain wall (AFMDW), where we mix AFM boundaries with DW corners. This configuration produces different behavior with system size L . With L even, two corners point in, two point out, and the charge is $Q = 0$. Instead, with L odd, the four corners point either in or out, and a total charge $Q = \pm 4$ is locked inside the bulk. Furthermore, we also ran simulations with periodic boundaries, that are similar to previous simulations on particle-based ice [31,32], and with open boundaries, which represent the experiments with no fixed particles [Fig. 1(c)].

To show how borders can be manipulated in experiments, we realize a square colloidal ice with antiferromagnetic domain walls. The system setup has been described in Ref. [33]. Here, we modify the boundaries of an isotropic lattice by adding nonmagnetic silica particles to the corresponding double wells. The silica particles induce local jamming, fixing the paramagnetic particle to a stable location, as shown below.

We start by showing in Fig. 2(a) how the four different boundary conditions influence the bulk behavior in terms of the fractions of type-III vertices (top) and of the average vertex charge (bottom). Both the open and DW frontiers show very similar trends, failing to reach the GS for all sizes. For these type of boundaries, the system accumulates charged defects at the boundaries, which are all negative for open boundaries and positive (negative) for inward-pointing (outward-pointing) spins in the DW. Only open boundaries allow the appearance of a net nonzero topological charge, which converges to a size-dependent negative value at high field, as shown in the bottom in Fig. 2(a). This effect can be appreciated also from the time evolution of the system in Fig. 2(b) and in Video S1 [30]. Above $B = 16.6 \text{ mT}$, all the borders exhibit particles displaced toward the outer region (spins out), a radial polarization effect predicted in Ref. [34]. Such an effect arises since the analogy between spin and colloidal ice is broken near the boundaries due to the repulsive interactions between the particles, while in ASIs nanoislands interact due to in-plane dipolar forces. In contrast, periodic, AFM, and DW satisfy the conservation of topological charge Q for all field values and system sizes. As shown in Fig. 2(b) and Videos S2 and S3 [30], we find that a system with periodic or DW

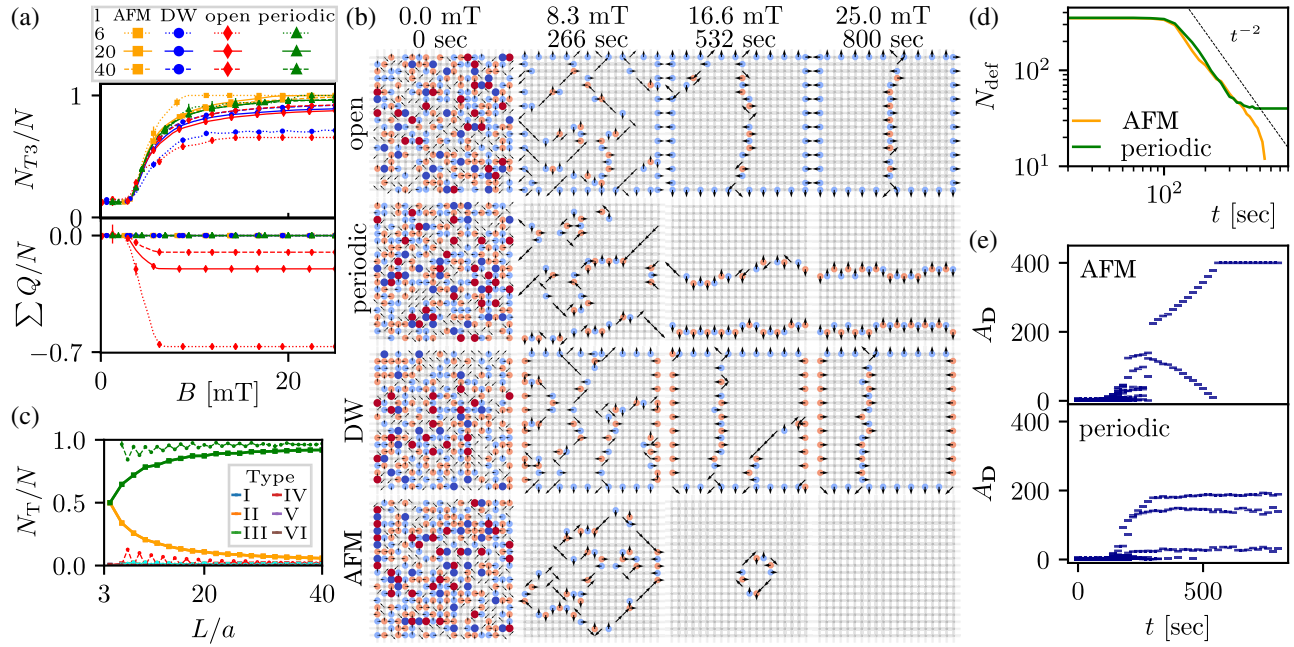


FIG. 2. (a) Fraction of type-III vertices (top) and average vertex charge (bottom) for different system sizes (numbers) and type of boundaries (symbols). (b) Maps of defects for different types of boundaries. The colored circles indicate charges, while colorless arrows indicate type-IV defects. The system width is $L = 20$ vertices. (c) Fraction of vertices at the maximum field ($B = 25$ mT) versus system size, for open (solid lines) and periodic (dotted lines) boundaries. (d) Number of defects N_{def} vs time for the AFM and periodic case; the line denotes t^{-2} scaling. (e) Type-III domain area A_D vs time for AFM (top) and periodic (bottom).

boundaries induces the formation of system-spanning domain walls, not allowed by the AFM (Video S4 [30]). These defect lines are very difficult to erase by increasing B further, as they require the simultaneous flipping of large GS regions in the bulk. In a system with periodic boundaries, the parity of the domain walls is also topologically protected: When the boundaries are of even size ($L \in 2\mathbb{Z}^+$), defect lines can appear only in pairs. In contrast, for odd values of L , at least one defect line is always present. This effect appears also in Fig. 2(c), where the periodic boundaries exhibit a zigzag trend: Odd lengths have an excess of type-IV vertices, which become less relevant as the boundary to bulk ratio becomes smaller. In contrast, we found that AFM boundaries can equilibrate to the GS faster and at lower fields, since they restrict the phase space as predicted in Ref. [1]. The kinetics of the defects is analyzed in Fig. 2(d) for AFM boundaries and compared to the periodic case. Both display coarsening dynamics with a power law scaling. This behavior can be also appreciated from the time evolution of the type-III domains in Fig. 2(e). Initially, both systems create similar domain structures, but, while a system with periodic boundaries falls to a metastable state with several smaller domains, the AFM creates a single loop of defects that continuously shrink, giving rise to the GS.

We now explore the behavior when boundaries are fixed in the AFMDW state. Figure 3 shows a system with $Q = -4$, where, if the energy of the type-IV vertices were

similar to that of type III, the charge would be contained in a single type-I vertex, with four lines of type IV connecting it to the corners. However, due to line tension, as the applied field increases, it becomes more stable to break up the excess of charges and distribute them along two lines connecting the four corners. This leads to a symmetry breaking, where the system must choose whether to arrange the two connecting lines horizontally [state 0 in the left of Fig. 3(a) and Video S5 [30]] or vertically (state 1 in the right of Fig. 3(a) and Video S6 [30]). We quantify this bistability using the order parameter $\Phi = \langle |\mathbf{s} \cdot \hat{\mathbf{e}}_x| - |\mathbf{s} \cdot \hat{\mathbf{e}}_y| \rangle$, where \mathbf{s} is a sum over the vectors associated to charged vertices and $\langle \dots \rangle$ is an average calculated over all vertices. By definition, Φ acquires a positive (negative) value for defects arranged in the state 0 (state 1) [Fig. 3(b)]. As shown in Fig. 3(c), we observe a bifurcation starting from $B \sim 5.6$ mT. The process of choosing one of these two states develops via a coarsening of small type-III domains and consequent reduction of the highly charged defects until three main domains are formed at $B \sim 9.4$ mT. From here, the rest of the process consists of pulling, through line tension, the defect line toward the edges.

Another type of AFMDW boundary condition can be imposed by introducing only two defects in opposite corners. This constraint creates two equal and incompatible type-III regions that meet along the diagonal and are separated by a string of type-IV vertices. The corresponding evolution from a disordered state is shown in Figs. 4(a) and 4(b) and in

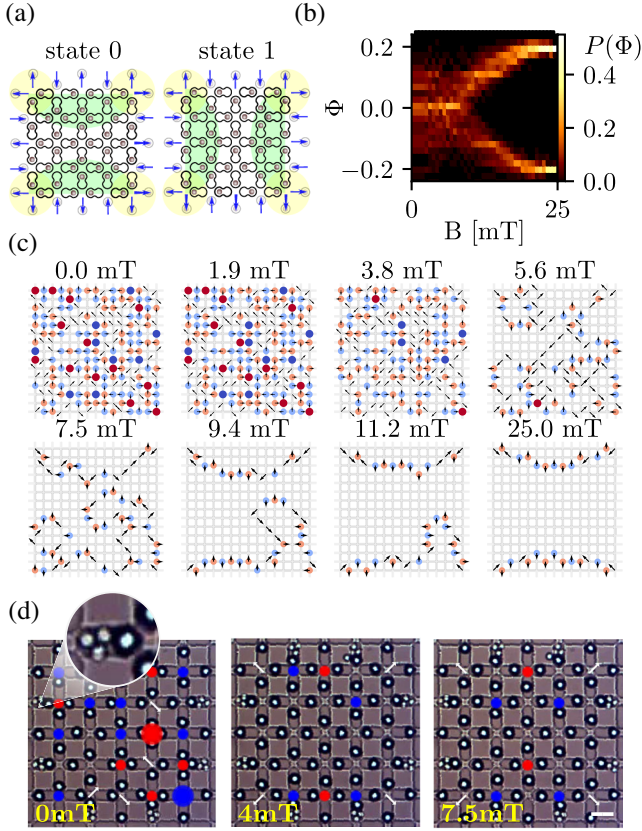


FIG. 3. (a) Schematics showing two states that appear after application of the field when using the AFMDW configuration. Here, topological charges can be connected either horizontally (right) or vertically (left). (b) Bifurcation of the order parameter observed for $N = 100$ simulations. (c) Color map showing the field-induced symmetry breaking where the system chooses the state 0. (d) Experimental observation of the first type of defect pattern; the second one is in Supplemental Material [30]. The enlargement shows the trap jammed by silica particles. The scale bar is $20 \mu\text{m}$, and red (blue) dots indicate positive (negative) Q .

Video S7 [30]. In the simulations, after $B \sim 5$ mT, the system nucleates two type-III regions which coarsen to the two final domains at $B = 25$ mT. The final straightening process of the topological string results from line tension, as also confirmed by experiments [Fig. 4(b)]. However, we find that this defect line is not always completely stretched, and defects might appear in the form of small distortions connected by a string of type-IV vertices, parallel and pointing along the opposite direction from the main defect line (Fig. S1 [30]). We capture this effect by measuring in Fig. 4(c) the distribution of the number of defects. As the field increases, the system gets rid of all non-type-III vertices, until it reaches a steady state close to the topologically protected minimum number of defects, which is equal to L [dotted white line in Fig. 4(c)]. Nevertheless, the inset shows that such a minimum value is not reached by many of the realizations, since many systems fall to a state with a small number of defects distributed along the domain wall and deviating from the diagonal.

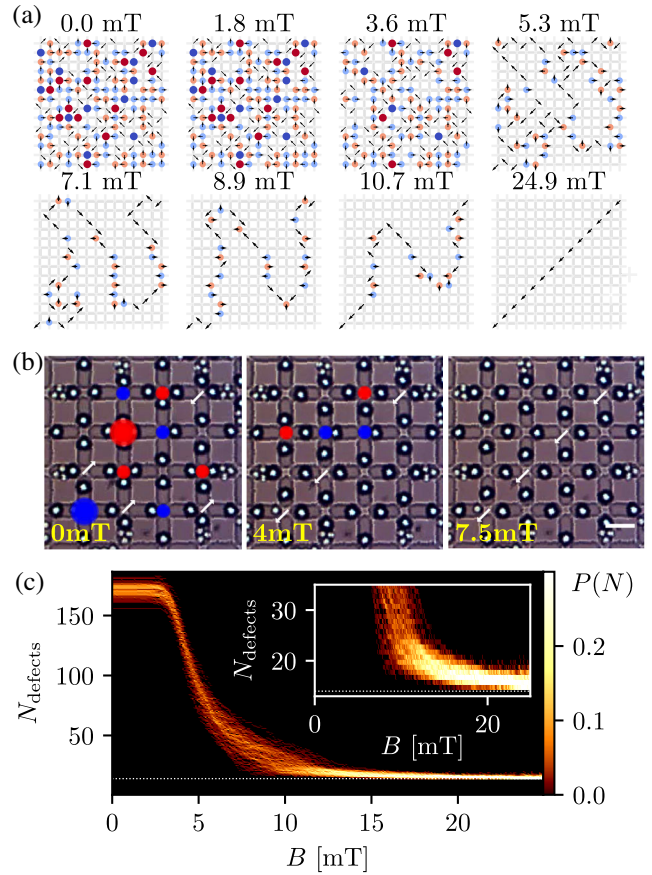


FIG. 4. (a) Color map illustrating the time evolution of the topological line connecting two edge defects in the AFMDW configuration. (b) Experimental realization of the line of defects using the AFMDW configuration. The scale bar is $20 \mu\text{m}$. (c) The probability of finding a specific number of defects in a system, as a function of the field. The dotted line indicates the system's width ($L = 14$). The inset shows an enlargement of the central portion of the curve.

In conclusion, we show how to engineer different boundary conditions to control the bulk behavior in a geometrically frustrated soft-matter system. We demonstrate this concept with an artificial colloidal ice combining numerical simulation and experimental realizations. Topological defects placed at the boundaries propagate inside the bulk, forming bistable states with symmetry breaking or topologically protected strings. The fact that the observed phenomena display a topologically protected nature suggests that they could be observed on other systems such as nanomagnetic artificial ice. This could be tested experimentally, for example, by using lithography to design smaller and compact islands such as ferromagnetic cubes [35], cylinders [36], or disks [37] at the system edges to impose a desired bulk configuration. From the technological perspective, writing or erasing defect lines in the GS region can be used to freeze information into the system by applying a bias during the equilibration process [38].

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