Artificial Spin Ice of Liquid Crystal Skyrmions

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A push to generate exotic, emergent phenomena in the collective behavior of simple, interacting, artificial building blocks proved successful using nano magnets, confined micro-colloids, or superconducting vortices as platforms to realize so-called artificial spin ices. Here we demonstrate that liquid crystals are a new, particularly versatile platform for spin ice physics. We show that interacting skyrmions in liquid crystals can be confined in binary traps, where their frustrated interaction reproduces the ice-rule. Besides their importance as materials in industry, liquid crystals allow for real-time, real-space characterization but also direct, dynamic control not only on the spin-ice ensemble, but on the very geometric structure of the system, something generally impossible in more traditional material platforms.

Artificial spin ices [1,10] are arrays of interacting, frustrated, binary variables arranged along the edges of a lattice, and obeying some version of the ice rule [11,12] at the vertices, which only leads to various forms of constrained disorder. Typically they can be characterized at the constituent level, often in real time, real space [13,14], and designed for a wide variety of unusual emergent behaviors [10] often not found in natural materials [15,16].

Their seminal [1,2] and to this day most explored [8–10] realizations are via lithographically fabricated, magnetic nanoislands. Nonetheless, the set of ideas behind these materials, such as collective forms of frustration, topological charges in general, or magnetic monopoles in particular, extend beyond magnetism. Indeed it has been exported to other platforms, such as superconductors [5–7,17], confined colloids [4,14,18], magnetic skyrmions [19], and elastic metamaterials [20].

In this work we propose and numerically demonstrate Liquid Crystals (LC) as a new, timely platform for spin ice physics [21]. By confining liquid crystal skyrmions in binary traps with two preferential positions at the ends [3] we can recreate pseudo-Ising spin variables. Then their frustrated mutual repulsion leads to the ice rule [3,22].

Nematic liquid crystals (LC) are made of elongated molecules, typically organic, which can access phases of liquid-disordered distribution of their centers of mass yet with an alignment of their principal axis along a local director \( \hat{n}(\vec{x}) \). Their nematicity can be captured by a traceless tensor \( Q_{\alpha\beta} = S (3\hat{n}_\alpha \hat{n}_\beta - \delta_{\alpha\beta}) / 2 \), \( S \) being the so-called scalar order parameter quantifying the orientational order. An LC cell consists of a chiral nematic LC confined between two parallel surfaces. Its energetics is successfully described via a phenomenological free energy density

\[
 f = \frac{1}{2} a \text{Tr} (Q^2) + \frac{1}{3} b \text{Tr} (Q^3) + \frac{1}{4} c \left[ \text{Tr} (Q^2) \right]^2 \\
+ \frac{1}{2} L (\partial_\gamma Q_{\alpha\beta}) (\partial_\gamma Q_{\alpha\beta}) - \frac{4\pi}{p} L c_{\alpha\beta} Q_{\alpha\rho} \partial_\gamma Q_{\beta\rho} \\
- \left[ K (\delta(z) + \delta(z - N_z)) + E^2 \Delta \epsilon \right] Q_{zz}. \tag{1}
\]

The first line is the Landau–de Gennes thermal term describing a nematic to isotropic second order phase transition in temperature (parameters \( a, b, \) and \( c \) are chosen to ensure a reasonable value for \( S \), see Supp. Mat.). In the second line elastic energies penalize the gradient of \( Q \) and favor a twist with cholesteric pitch \( p \). The last term reflects the homeotropic surface anchoring of strength \( K \) at the boundaries \( (z = 0, N_z) \) and the coupling to the electric field \( E \) applied in the \( z \) direction [23]. \( \Delta \epsilon \) is the dielectric anisotropy of the LC favoring easy-axis (along \( z \)) or easy-plane (perpendicular to \( z \)) alignment depending on its sign.

The free energy in Eq. (1) admits particle-like, metastable solutions called skyrmions, that were indeed found experimentally [21,24]. Fig. 1a, shows the mid-plane of one such full skyrmion, where the polar angle of the director \( \hat{n} \) rotates by 180° from its center to periphery leading to a topological charge \( \frac{2\pi}{\pi} \int \int \text{d}x \text{d}y \hat{n} \cdot (\partial_x \hat{n} \times \partial_y \hat{n}) = 1 \), as the mapping of the directors to the surface of a sphere covers the surface once.

We will employ these full skyrmions as trapped particles to realize spin ice in liquid crystals. Other topological solutions appear less suitable for our task. For instance merons, which cover only half of the sphere, usually exist as ground state lattices [22,31], and are accompanied by defects around them. Instead full LC skyrmions can exist individually, and do not appear spontaneously in the background field. They have been realized experimentally and are reported as metastable for months [24], with significant progress in their confinement, and manipulation via a rich variety of control knobs [30,32] including light, electric fields and surface chemistry [24,27]. In particular they can be generated and decayed at will, actuated, and arranged to exhibit a variety of collective

\[
\text{Collective}
\]
FIG. 1. Skyrmions in Liquid Crystals. (a) Mid-layer of a vectorized skyrmion in LC cell with homeotropic anchoring and mapping of directors on to the surface of a sphere (b) External electric field (coupling coefficient $\alpha$) and and surface anchoring (coupling coefficient $K$) can be used in various proportions to sustain long lived skyrmions. Different skyrmion shapes appear depending on $K$ and $\alpha$. (c) 2D skyrmion stabilized in a background field ($\alpha > 0$). Reducing field strength results in increased skyrmion size and deformability near obstacles.

Easy-axis alignment in the vertical direction is employed to stabilize skyrmions in a uniform vertical background and can be achieved by field and surface anchoring, which combine in the last line of the free energy (1), to offer freedom in the realization of long-lived skyrmions (Figure 1b). Reducing the field coupling $\Delta \epsilon E^2$ increases the skyrmion size (Figure 1c). Furthermore when field alone is used and surface anchoring $K$ is zero or very small, we have an approximatively $z$-invariant structure, the so-called 2D skyrmion [24]. In the main text we report on 2D skyrmions, but our findings extend to 3D ones (see Supp. Mat.).

Elsewhere [32], we show that skyrmions are driven towards regions of weak (and repelled by regions of strong) easy-axis alignment, which allows for confinement and trapping. Confinement can also be achieved by light exposure, which increases the helical pitch $p$ of certain liquid crystals and repels the skyrmions away from the exposed region [33]. We exploit these properties to confine skyrmions in binary traps.

We verify numerically that designs in Figs. 2, 3 realize such confinement, without eliminating mutual interaction. The black circles represent repulsive areas due to strong easy-axis alignment. They create a set of traps, aligned along the edges of the lattice, where skyrmions have two preferential positions at the ends. This realizes the familiar binary state of particle-based spin ice [3, 22]. Note that traps are open at the end, except for smaller repulsive dots: because skyrmions interact repulsively through the elastic deformation of the LC medium, ends cannot be closed. The possible configurations of skyrmions in the vertices of the two lattices are shown in Fig. 2. It is known that in these systems [3], as a result of non-local frustration [22] the lowest energy state is an ordered “antiferromagnetic” tessellation of type IV vertices (for a square lattice) or a disordered mix of type II-III for a honeycomb lattice. Both cases obey the ice rule [11, 12]: 2 particles in the vertex and 2 out, for the square geometry and 1-in/2-out or 2-in/1-out for the honeycomb one (Fig. 2).

We simulate the LC systems of Fig. 3 by solving via finite difference method the over-damped dynamic equa-
FIG. 3. Relaxed skyrmion spin ices obey the ice rule. **Top:** Skyrmions initially swollen and then de-swollen, relax to lower energy states. **Bottom:** 2D (very thin) square and hexagonal ice with surface anchoring/field relax to ice-rule obeying states with defects. The square geometry shows an ordered state where domains of ice-rule, type IV vertices are separated by domain walls of ice-rule obeying Type III (blue) and ice-rule violating Type II (yellow) and Type V (white). The hexagonal geometry converges to a disordered manyfold of ice-rule obeying Type II (2-in/1-out) and Type III (1-in/2-out) vertices, with excitations as ice-rule violating Type I (3-in, yellow) or Type IV (3-out, white) vertices. White lines superimposed on the lattice define the vertices.

tions for the system

\[
\frac{\partial Q(r, t)}{\partial t} = -\Gamma \frac{\delta F}{\delta Q(r, t)}
\]

for 2D skyrmions (\(\Gamma\) is a mobility constant) with periodic boundary conditions. The initial state corresponds to 288 skyrmions in the square geometry and 192 in the hexagonal geometry placed randomly in the traps, stabilized by background field. Traps can be realized via either extra field or surface anchoring. This entails about three million finite difference elements to be updated at each time step, which we implement by exploiting the intrinsic parallelism of GPUs (see methods). The number of time steps needed to reach stationary states were \(10^7\) and \(3 \times 10^7\), for the square and hexagonal lattice respectively. After an initial relaxation, we reduce the background field to swell the skyrmions until they occupy almost their entire traps (Fig. 3 top) so as to bring them in close interaction, and then we reduce their size.

Figure 3 shows snapshots of the final states for the two geometries. Square ice converges to an ordered tessellation of type IV vertices, with two skyrmions close to and two away from each vertex, thus obeying the ice rule. Deviations from type IV correspond to ice-rule obeying Type III, but also to violations of the ice rule in the form of monopoles [37], or Type II and V. Together, these excitations form familiar domain walls among the two possible orientations of “antiferromagnetic” order. Hexagonal ice also converges properly to an ice state. Unlike square ice, it is a disordered mixture of Type II and Type
III, thus obeying the pseudo ice rule (1-in/2-out and 2-in/1-out), together with sparse deviations (Type I, IV) of extra topological charge. Both results are exactly what is expected in particle-based spin ices of corresponding geometries \([3, 5, 22]\). 

Structural parameters control the proximity to the ice-manifold. The geometry is defined by two lengths: \(r\) the radius of the circle of strong easy axis alignment, and \(D\) the length of the edge of the lattice. As shown in Fig. 4, spin ice behavior is obtained for a window of the \(r/D\) ratio: if too large (pink region in figure), the skyrmions cannot move within the trap; if too little (violet region), the positions at the end of the trap will cease to be preferable, the skyrmions will often sit at the center, and the trap will cease to be a binary variable. Transition to this second regime is interesting as it can lead to the realization of a Potts Ice, a still largely unexplored model on which we will report elsewhere. At intermediate value (green region) states closer to the ice-rule are easily obtained.

The distinction between the three regions is fuzzy, also because the final ice manifold maintains memory of its preparation. Indeed Fig. 4 shows that when the skyrmions are not swollen, deviations from the ice rule are larger, and become smaller when we swell them. The process of swelling and de-swelling of the skyrmions helps the system find lower energy states and extends the suitable range for ice configurations. However, we find, repeated swelling past a cycle of 2-3 does not change much the final state, though this might change in physical realizations, due to unavoidable quenched disorder. The upper end of the green zone is assumed to be at the point where the curve gets steeper. Beyond this point, the skyrmions are stuck in the halves of the trap they were initially placed. We determine the corresponding \(r/D\) ratios to be 0.28 and 0.66 for square and hexagonal lattices, respectively. While these two numbers are very different, if we look at another ratio \(w/D\) where \(w\) is the width of the traps we find similar ratios in both cases: for square lattice, \(w = 2(D/2 - r)\) thus \(w/D = 1 - 2r/D = 1 - 2 \times 0.28 = 0.44\); for the hexagonal one, \(w = 2(D\sqrt{3}/2 - r)\) thus \(w/D = \sqrt{3} - 2r/D = \sqrt{3} - 2 \times 0.66 = 0.41\). (These numbers hold for \(K = 100\) and vary slightly with \(K\) as shown in Fig. 4c.)

We have demonstrated numerically that skyrmions in LC can be properly confined to create binary degrees of freedom whose frustrated interaction leads to ice manifolds. We have reported here a numerical proof of principle on the two most common lattices, and extensions to other, more complex \([16, 33, 34]\) geometries are readily possible. As in the case of gravitationally confined colloids \([1, 13]\) thermal fluctuations play no role in the collective ensemble of skyrmions, yet the resulting ice manifold can be controlled by various knobs. In general LCs provide a more malleable medium to explore decimation, ice-rule fragility, doping, and unlike previous platforms can allow dynamic change of structure, for instance for cycling between topologically equivalent geometries of different ice behavior, for memory effects. Unlike trapped colloids, the skyrmions can change size, can be created or destroyed optically, and their mutual interaction can be controlled, from anisotropic to isotropic. Growing abundance of techniques \([42]\) for confining LC defects and recent efforts to exploit collective behavior of LC skyrmions clearly show that they can be employed for actuation in soft robotics, optical applications, and functional materials design. Our proposal to realize spin ice with LC skyrmions is a promising development in this direction which we believe will stimulate experimental efforts.

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