Letter

Fractional skyrme lines in ferroelectric barium titanate

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We predict a topological defect in perfectly screened ferroelectric barium titanate which we call a skyrme line. These are linelike objects characterized by skyrmionic topological charge. As well as configurations with integer topological charge, the charge density can split into well-localized parts carrying a localized fraction of topological charge. We show that under certain conditions the fractional skyrme lines are stable. We discuss a mechanism to create fractional topological charge objects and investigate their stability.

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Skyrmions are topologically nontrivial defects originally proposed as a model of nuclei by Skyrme [1], though now various versions of skyrmions are more commonly studied in ferromagnets and other materials. They exist due to the topology of the system and their topological stability makes them promising candidates for various applications, with the prototypical application being high-density memory storage [2].

Recently, ferroelectric materials were shown to be a host for nontrivial physics associated with topological defects. The most studied are lattices of polar vortices, predicted in Refs. [3,4] and evidenced in Ref. [5]. This motivated the theoretic [6] and experimental [7] searches for lattices with skyrmionic charge. The individual units of this lattice are sometimes called (polar) skyrmion "bubbles" [8,9]. A bubble is a localized region of the material where the polarization has an opposite direction to the background. Evidence of localized skyrmion bubbles have been theoretically predicted in $BaTiO_3$ [10] and strained $PbTiO_3$ [11]. Polar skyrmions and vortices have novel features such as chirality [12] and local negative permittivity [13,14]. Vortexlike Ising lines have also been simulated numerically [15].

In this Letter, we show that another type of defect in ferroelectrics is possible, which we coin a ferroelectric skyrme line. These share many properties with skyrmions, such as their topology and chirality, and can be thought of as skyrmions localized on ferroelectric domain walls. We report the existence of many nontrivial skyrme lines in barium titanate, despite not finding any stable skyrmions in our model. Configurations where skyrme topological charge is confined to a domain wall have attracted interest for a long time both in mathematical physics [16–18] and recently in other physical systems such as magnetic systems [19–21] and superconductors [22,23].

The especially interesting property of these defects is that they exhibit fractionalization of topological charge. Traditionally topological charge is supposed to be an integer, represented as an integral over some topological charge density. However, recently fractionalization, where the topological charge density is split into several stable configurations of localized fractions, has become of interest in a variety of models. Fractional topological defects were searched for in various systems and a fractional vortex was reported recently in superconductors [24], where the interesting aspect of the fractionalization is that it arises from the vorticity in the fields which are not order parameters in the strict Landau theory sense [25], and hence represent effects beyond the conventional symmetry and topology classification. Fractional skyrmions have been seen in condensed matter systems [21,26,27] and mathematical physics [28]. The concept also applies to topological defects in higher dimensions [29,30]. In these examples, fractional defects exist as part of a larger object such as a lattice or integer-charged defect. Importantly, we report that ferroelectrics allow topological line defects with unique fractional skyrme charge, which are themselves stable.

We study a Ginzburg-Landau-Devonshire model of barium titanate in the rhombohedral phase ($T < 201$ K). We focus on a specific model with no electrostatic energy contributions. The model can be written in terms of a polarization vector $\boldsymbol{P} = (P_1, P_2, P_3)$ and a symmetric strain tensor u_{ij} which can be conveniently bundled into a 6-vector $e = (u_{11}, u_{22}, u_{33}, u_{23}, u_{13}, u_{12})$. The free-energy density is given by

$$\mathcal{F} = \tfrac{1}{2} G_{abcd}\partial_a P_b \partial_c P_d + V(P) + \tfrac{1}{2} C_{\alpha\beta} e_\alpha e_\beta - q_{\alpha bc} e_\alpha P_b P_c. \tag{1}$$

The parameters are detailed in the Supplemental Material [31]. The potential $V(P)$ in the rhombohedral phase has eight ground states which point in the directions of cube vertices: $\boldsymbol{P} \propto (1, 1, 1)$, etc. The strain tensor satisfies an addition compatibility constraint, which ensures there are no holes in the

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material. We have developed a method to minimize the free energy while preserving the compatibility constraint. While minimizing for P, we project the energy-minimizing strain $e_\alpha = C^{-1}_{\alpha\beta} q_{\beta cd} P_c P_d$ onto a complete set of compatible functions. Since we can express the strain part of the free energy as an inner product, this is guaranteed to be the unique compatible energy-minimizing strain. More details can be found in the Supplemental Material [31]. We ignore the electrostatic energy for simplicity and so our model is of perfectly screened barium titanate.

A skyrmion is a texture that exists and is stable due to the topology of the system. Usually, this topology is due to the structure of the field. In magnetic materials, the fundamental field is the magnetization, a field which takes values on the sphere S^2. Maps from S^2 to a plane with fixed boundary condition have nontrivial topology through the second homotopy group $\pi_2(S^2)$. It is this topology which makes the magnetic skyrmion stable. In ferroelectrics the order parameter field is the polarization $\boldsymbol{P} \in \mathbb{R}^3$, which has trivial topology. So a naive symmetry-based analysis would suggest that the field structure cannot support skyrmions. However, the point $\boldsymbol{P} = \boldsymbol{0}$ has very high energy and there is an energy cost for a configuration to contain this point. This is why Bloch walls (which do not contain $\boldsymbol{P} = \boldsymbol{0}$) are often energetically favored over Ising walls (which contain $\boldsymbol{P} = \boldsymbol{0}$) in low-temperature barium titanate [32]. If $\boldsymbol{P}$ is never zero, the field can be thought of as $\boldsymbol{P} \in \mathbb{R}^3 \setminus \{0\}$, which has nontrivial topology and can support skyrmions since $\pi_2(\mathbb{R}^3 \setminus \{0\}) = \mathbb{Z}$. Note that the topological argument involving homotopy groups, which are the basis for skyrmion stability in magnetic materials and the skyrme lines here, only strictly hold in the continuum limit.

We find in barium titanate a stable object which carries skyrmionic topological charge, which we call a skyrme line. These can be viewed as an "unwrapped skyrmion" as shown in Fig. 1. The configurations are similar to domain walls but have a nontrivial structure along the wall. The skyrme lines lie on planes, embedded in $\mathbb{R}^3$. The free energy depends on the orientation of the two-dimensional (2D) plane in the three-dimensional (3D) material. We consider the plane spanned by two orthogonal vectors $\boldsymbol{s}$ and $\boldsymbol{r}$. First, consider a domain wall that connects two antipodal ground states $\pm\boldsymbol{P}^V$ along a direction $\boldsymbol{s}$; the material's extent in the $\boldsymbol{s}$ direction, L_s, should be much larger than the width of the domain wall. We take periodic boundary conditions in the r direction for simplicity and discuss more realistic boundary conditions in the next section. Overall, the boundary conditions are then

$$\boldsymbol{P}(\pm L_s, r) = \pm\boldsymbol{P}^V, \quad \boldsymbol{P}(s, L_r) = \boldsymbol{P}(s, -L_r). \tag{2}$$

The skyrme line seen in Fig. 1 respects these boundary conditions. It is constructed using the initial configuration

$$P^{\mathrm{sk}}_a(s, r; r_0) = |P^V| R_{ab} \begin{pmatrix} \cos(N_1 r + r_0)\sin[f(s)] \\ \sin(N_1 r + r_0)\sin[f(s)] \\ \cos[f(s)] \end{pmatrix}_b , \tag{3}$$

where R_{ab} is the rotation matrix taking (0,0,1) to the boundary ground state, $(-1, -1, -1)$ in this case, and $f(-L_s) = 0$, $f(L_s) = N_2\pi$. The topological charge is equal to $N = N_1 N_2$ and we have taken $N_1 = N_2 = 1$. Equation (3) is the unwrapped form of the standard "hedgehog" baby skyrmion

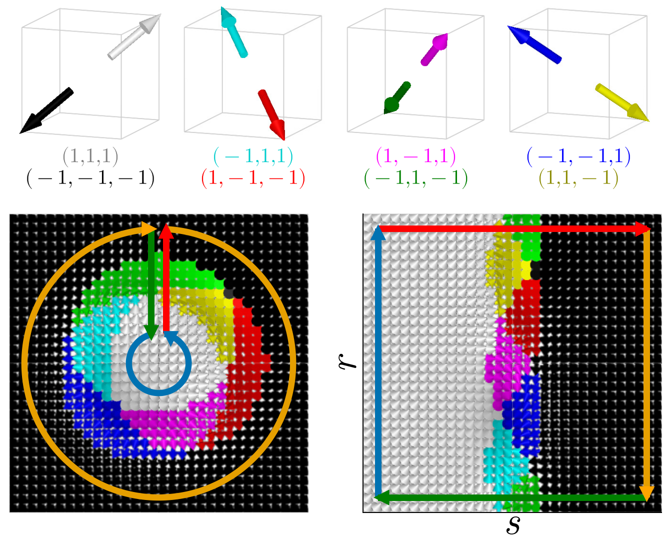

FIG. 1. Plots of the polarization vector $\boldsymbol{P}$ for a skyrmion (left) and skyrme line (right). We find that ordinary skyrmions are unstable in barium titanate, but that skyrme lines can be stable. One can think of a skyrme line as an "unwrapped" skyrmion: The long arrows demonstrate how a skyrmion is mapped onto a skyrme line. The short arrows, representing $\boldsymbol{P}$, are colored white, red, green, blue, teal, pink, yellow, and black when their nearest ground state is P^V times (1,1,1), $(-1, 1, 1)$, $(1, -1, 1)$, $(1, 1, -1)$, $(1, -1, -1)$, $(-1, 1, -1)$, $(1, -1, 1)$, and $(-1, -1, -1)$ respectively, visually represented in the top subfigure.

[33]. To have periodic boundary conditions r must be a multiple of π/L_r. The parameter r_0 allows us to shift the skyrmion along the r axis.

When the point $\boldsymbol{P} = \boldsymbol{0}$ does not appear in a configuration we can construct the normalized polarization vector $\hat{\boldsymbol{P}}$ and use it to calculate a topological charge, usually known as the skyrme charge:

$$N = \frac{1}{4\pi} \int \hat{\boldsymbol{P}} \cdot \partial_s \hat{\boldsymbol{P}} \times \partial_r \hat{\boldsymbol{P}}_c \, ds \, dr. \tag{4}$$

The skyrme line in Fig. 1 has charge $N = 1$. The charge is conserved provided that $\boldsymbol{P}$ is never zero. If this does happen, the charge becomes undefined and the skyrme line collapses into a regular domain wall.

To show stability, we numerically relax the skyrme line (3) using a gradient flow (see Supplemental Material [31]). We know that the allowed domain walls depend on the orientation of the wall in the material, and so we expect that the skyrme line stability depends on the plane orientation $(\boldsymbol{s}, \boldsymbol{r})$. We search over various plane orientations and find various stable configurations, including skyrme lines. One such configuration is plotted in Fig. 2, with $\boldsymbol{s} = 1/\sqrt{3}(1, 1, 1)$ and $\boldsymbol{r} = 1/\sqrt{2}(0, 1, -1)$. Note that the three contours $P_1 = 0$, $P_2 = 0$, and $P_3 = 0$ in Fig. 2 never touch. Their intersection would correspond to the point $\boldsymbol{P} = \boldsymbol{0}$, which has very high energy. One can only "unknot" the contours by passing through the point. This energy barrier generates an outward pressure on the skyrme line. Conversely, the gradient energy $G_{abcd}\partial_a P_b \partial_c P_d$ is minimized when the line collapses into a simple domain wall and so encourages the line to shrink. The balance between these two forces stabilizes the skyrme line.

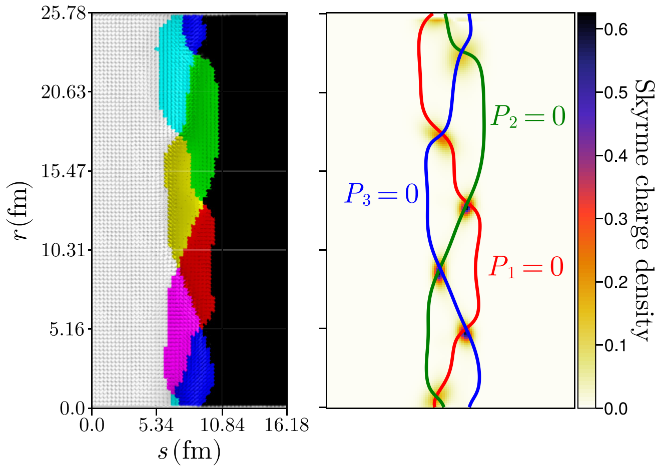

FIG. 2. A numerically generated skyrme line plotted twice. We plot the polarization vector $\boldsymbol{P}$ colored to reflect the closest vacuum (left) and the contours $P_i = 0$ overlaid with the topological charge density (right). The charge is fractionalized: Its density is mostly concentrated on the contour intersections.

The topological charge deserves careful study. We observe what can be interpreted as topological charge fractionalization in this system. Namely, the topological charge density is equally concentrated at the six points, where two of the three contours $P_i = 0$ intersect. At these intersections, two of the $\boldsymbol{P}$ components are zero and so the polarization points along a Cartesian axis. Now consider a loop around an intersection, which is in target (or $\boldsymbol{P}$) space and encircles an axis. We call the orientation of this loop the chirality of the intersection. If the direction is anticlockwise, the chirality is positive and vice versa. Each positive chirality intersection contributes $+1/6$ to the charge and each negative chirality loop $-1/6$. In Fig. 2 the skyrmion contains six positive chirality intersections, and so has charge $N = 1$. More formal arguments, using different language, were recently made for a magnetic system in Ref. [34]. The intersections are extrema of the potential and so each charge $1/6$ skyrmion has the same mathematical structure as their "non-Abelian vortices," which are topologically stable due to an energetics-motivated puncturing of the target manifold.

We have seen that the topological charge fractionalizes into sixths and will now show that we can construct fractional skyrme lines by adjusting the boundary conditions of the system. We again construct a domain wall connecting two ground states $\pm\boldsymbol{P}^V$. In barium titanate, there are several energy-degenerate domain walls that connect the ground states. We suppose that one type of domain wall $P_{W+}(s)$ is present at one side of the material, L_R, and another $P_{W-}(s)$ at the other side, $-L_R$. We expect this situation to occur when the system is annealing. Since there is no energetic reason for one wall to be preferred, both will form in different regions and our system describes what will happen between these regions. The boundary conditions are now

$$P(\pm L_s, r) = \pm\boldsymbol{P}^V, \tag{5}$$

$$P(s, -L_r) = P_{W-}(s), \;\; P(s, L_r) = P_{W+}(s), \tag{6}$$

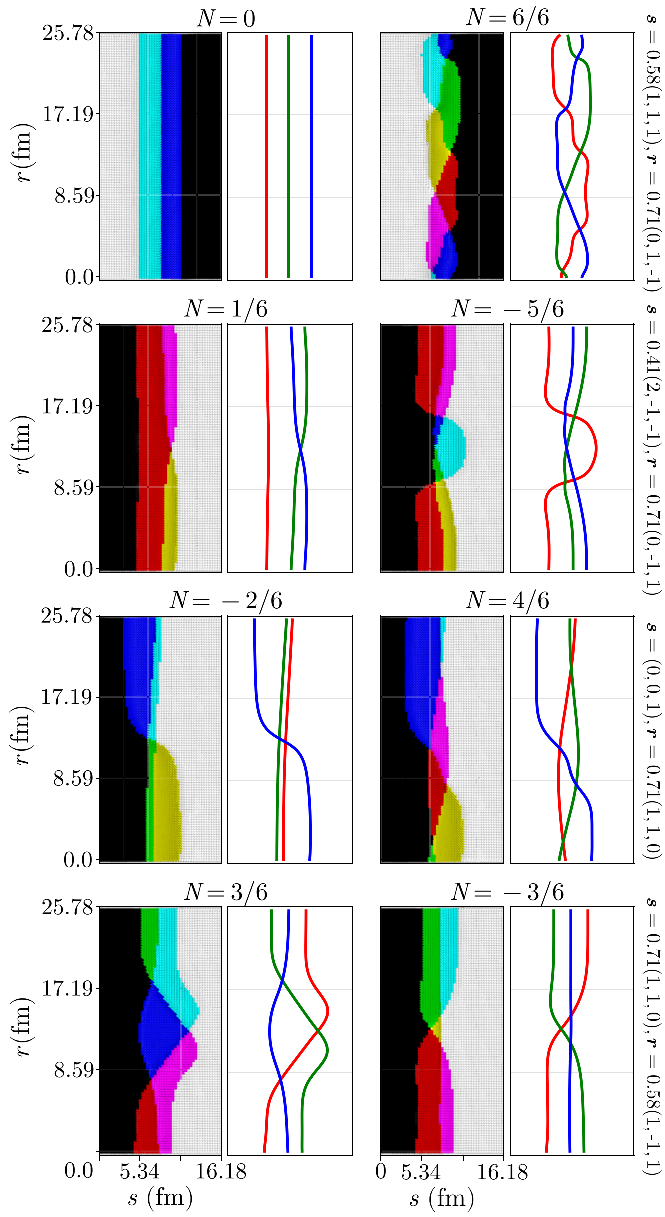

FIG. 3. Fractional skyrme lines with topological charge $n/6$, which are stable in a variety of planes spanned by $\boldsymbol{s}$ and $\boldsymbol{r}$. Skyrme lines with the same boundary data have a relative topological charge 1.

where P_{W+} and P_{W-} are genuine 1D domain wall solutions. These can be found in arbitrary orientations following the methods developed in Ref. [35]. We then generate an initial configuration that satisfies the boundary conditions (5), of the form

$$\boldsymbol{P}(s, r) = \boldsymbol{P}_{W-}(s)g(r) + \boldsymbol{P}_{W+}(s)[1 - g(r)], \tag{7}$$

with $g(L_r) = 0$ and $g(-L_r) = 1$.

We now apply gradient flow to the initial data (7). By using different $P_{W\pm}$ and different $g(r)$s, we can generate a zoo of skyrme lines. Solutions with absolute topological charge $n/6$, $n \in [1, 6]$ are plotted in Fig. 3. All configurations here have fixed boundary conditions. There are four different sets of boundary conditions, and for each we plot two configurations separated by one unit of charge. The fact that both are stable suggests that there is an energy barrier due to their different topological charges. We calculate the topological charge by simply counting the contour intersections, with chirality. Given the order of contours at the top and bottom of the box, the charge is fixed to be some fraction, up to an integer. Hence the fractional charge is due to the structure of the domain wall solutions at each side of the system. We verify this simple counting by calculating the topological charge numerically using (4). For each skyrme line with charge N, there is an energy-degenerate partner with negative skyrmionic charge $-N$. This can be generated by applying a reflection to $\boldsymbol{P}$, across the plane with normal $\boldsymbol{P}^V \times \boldsymbol{s}$: a symmetry of the system.

We can better probe the wall stability by then relaxing the boundary condition to be open. That is, take $\partial_r P(s, \pm L_r) = 0$. When this is done, all the walls in Fig. 3 are stable except the 4/6-charge wall, which ejects two intersections to become a 2/6-charge wall. Overall, we have found stable, localized skyrme lines with various fractional topological charges. As another check of the stability of the skyrme lines, we have applied the simplified string method [36,37] to a skyrme line. This method calculates the energy barrier between the skyrme line and a simple domain wall, and shows that the topological charge becomes ill defined at this barrier. The analysis confirms that the skyrmionic topology is what provides the skyrme line with its stability. More details can be found in the Supplemental Material [31].

The skyrme lines discussed here have some similarities and differences with other objects in the literature. In Ref. [6], the authors found a lattice with skyrmionic charge 1 per unit cell. Like ours, the charge fractionalizes and is concentrated at the intersection of contours $P_i = 0$. Unlike ours, the lattice is only stable due to the presence of a nanowire. Recently, the existence of individual antiskyrmions (similar to the configuration shown in Fig. 1 left) in barium titanate at $T = 0$ K has been reported [10]. Despite searching, we do not find such stable configurations in our simulations. Finally, note that the charge $N = \pm 1/2$ skyrme lines have the same topological structure as merons.

The skyrme lines that we found can be created and manipulated using external electric fields. First, one can create a whole skyrme line from a configuration with zero topological charge. The process requires a complicated stencil; we use a four-part stencil. Each part of the electric field stencil had a strength $|E| = 0.3$ MeV/m^2. Each square was 3×6 nm large and point in the direction indicated by the color in Fig. 4. The color scheme is visually represented in Fig. 1. The electric field was applied until the system reached equilibrium (less than 1 unit of τ; see Supplemental Material [31]), then turned off. This creates enough energy to overcome the $\boldsymbol{P} = \boldsymbol{0}$ energy barrier and a skyrme line with topological charge $N = 1$ is created. Note that the stencil used here is a nanoscale object and hence cannot be created with current technology. But this simulation shows that fractional skyrme lines can theoretically be produced by an external field.

Since each part of the previous stencil points in a different direction, engineering such an external field is a challenge. A simpler process, with a simpler stencil, can be seen at the

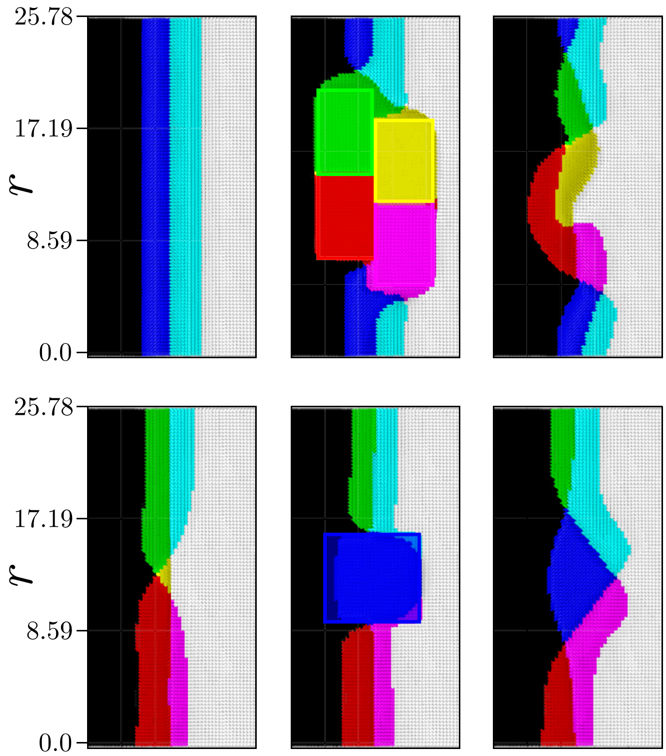

FIG. 4. Making a charge 1 skyrme line (top) and switching an $N = -1/2$ line to an $N = +1/2$ line (right) using external electric fields. The color of each stencil represents the direction of the applied external field.

bottom of Fig. 4. Here, we switch from a $N = +1/2$ line to a $N = -1/2$ line using a one-piece stencil. A 6 nm $\times$ 6 nm electric field of strength 0.3 MeV/m^2 was applied until equilibrium was achieved (less than 1 unit of τ), then turned off. In the process, three positive-chirality contour intersections are turned into three negative-chirality ones.

We have considered another type of topological defect in perfectly screened ferroelectric barium titanate, which we call a skyrme line. Their stability depends on a topological charge which is protected by the high-energy cost of the point $\boldsymbol{P} = \boldsymbol{0}$. We found stable skyrme lines and studied their stability, creation, and switching. A unique feature of ferroelectric skyrme lines is that they can exist with fractional topological charge and these structures should appear naturally in an annealed sample. We found all examples of possible fractional skyrme lines $N = n/6$ in the rhombohedral phase.

In this work, we have ignored the electrostatic energy contribution. Physically, we have modeled perfectly screened barium titanate. This approximation is most reasonable for neutral domain walls, when $\boldsymbol{s} \cdot \boldsymbol{P} = 0$, such as the charge $N = \pm 1/2$ walls studied here. These are the skyrme lines most likely to exist in real barium titanate. This motivates further studies that would include the electrostatic energy. One can add a nonlocal term which is numerically expensive. Including also raises difficult theoretical questions of regularization, especially for nonperiodic boundary conditions [38].

We discussed how to switch domain walls using a very simple mechanism. The robustness and manipulability of these objects suggest that fractional skyrme lines might be useful objects for data storage devices. The $+1/2$ and $-1/2$ skyrmions have different chirality, and this property might be determinable using four-dimensional scanning transmission electron microscopy (4D-STEM) experiments [12].

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