

Self-charged polar nematic monopoles and hybrid topological states: intertwining and domain integration

Corresponding Author: Professor Satoshi AYA

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I have read the paper by Zhou et al. The authors demonstrate that confinement asymmetry can allow for engineering solitons in fluid ferroelectrics.

Overall I find the subject and results interesting to a degree that I could be sympathetic towards publication provided some issues are cleared up.

The authors talk about “multibody topological superstructures” this is quite confusing. One has a fluid electric with collective degrees of freedom that give rise to an effective winding number. As such I find this term hard to justify.

The authors rely on a mean field solution, under what assumptions are these valid ?

Accordingly more details on the numerical processes should be added as it is hard to pinpoint how this performed

With respect to the above, for a meanfield solution it seems that one get an effective model that can also be viewed as gauge theory see the papers

P. E. Lammert, D. S. Rokhsar, and J. Toner, Topology and Nematic Ordering, Phys. Rev. Lett. 70, 1650 (1993).

P. E. Lammert, D. S. Rokhsar, and J. Toner, Topology and Nematic Ordering. I. A Gauge Theory, Phys. Rev. E 52, 1778 (1995).

J. Nissinen, K. Liu, R.-J. Slager, K. Wu, and J. Zaanen, Classification of Point-Group-Symmetric Orientational Ordering Tensors, Phys. Rev. E 94, 022701 (2016)

Ke Liu, Jaakko Nissinen, Robert-Jan Slager, Kai Wu, Jan Zaanen, Generalized Liquid Crystals: Giant Fluctuations and the Vestigial Chiral Order of I , O , and T Matter, Phys. Rev. X 6, 041025

There has been recent work on defining merons for polar structures especially on moire systems, including their polarization for subdomains and local markers, see

- D Bennett, G Chaudhary, RJ Slager, E Bousquet, P Ghosez, Nature Communications 14 (1), 1629

- D Bennett, WJ Jankowski, G Chaudhary, E Kaxiras, RJ Slager, Physical Review Research 5 (3), 033216

that moreover also have been linked to optical responses see

-A Agarwal, WJ Jankowski, D Bennett, RJ Slager, npj 2D Materials and Applications 9 (1), 43

These works clearly fit the general scope of introduction and ought to be mentioned.

The text would profit from putting the main equation as now it is not self contained without the additional parts.

Reviewer #2

(Remarks to the Author)

This paper reports an experimental study of topological solitons in fluid ferroelectrics discovered in recent years. The authors

explicitly demonstrate the existence of hybrid topological solitons and explore fusion between elementary topological solitons. While the experimental results are interesting and the theoretical explanation is, in broad terms, reasonable to me. However, the novelty and importance of their contribution are not clearly conveyed, like why should we care about these topological patterns of director fields they discovered and why the fusion between topological solitons can be done in their scheme but not others. I will have more detailed questions regarding these points below. Minorly, the organization and presentation of the manuscript are poor, which will hinder its accessibility to a broader audience. A major revision is therefore necessary to clearly convey the key results and their significance. Below, I list my comments and questions point by point.

1 The authors claim that their work aims to construct “higher-order topological structures.” However, they do not clearly define what is meant by this term. Does it refer to topological solitons with higher skyrmion number, or to some other type of topological invariant? This does not appear to be the case, since all of the solitons studied ultimately carry a skyrmion number $Q=\pm 1/2$. A clear and precise definition of “higher-order topological structures” is therefore necessary.

2 What is the size of the solitons relative to the overall system? Are they highly localized?

3 Fig 1 is particularly unclear:

(a) In a and b, what are the upper and lower parts respectively? The surface and bulk patterns of director fields? If so, why is the winding in x-y plane opposite at the surface and in the bulk? What are the colored lines there? Preimages of director field with fixed θ and ϕ ? Why in b there is only one bottom surface shown in the lower-left subfigure?

(b) In c and d, is it possible to put the scale bar in the figure? (note there is also a typo: scare scale, there are lots of similar typos throughout the manuscript which I will not list one by one). The corresponding text defining L_M/H and R_M/H is also unclear and confusing. What exactly is meant by the screw and twisting sense? What defines the chirality, is it the combination of twisting and screw? Is it true that the screw sense, rather than the overall chirality, is important when considering the intertwining of multiple bodies?

(c) In e and f, what do the rectangles on the right side correspond to? There is no v and v_i labelled in the figure. Again, the colored lines are not defined.

(d) The 3D color maps are also confusing. Why is a 3D plot needed even if the ϕ and θ are simply encoded using different color maps?

4 In Fig.1 b, the author illustrated how curved boundaries can lead to the hybrid topological solitons, and then in the text they claimed these hybrid topological solitons are realized in experiments by tuning the cooling rate. They should specify the relationship between the cooling rate and the presence of curved boundaries there instead of postponing it later.

5 In what sense are the hybrid topological solitons different from merons? Topologically, they have the same skyrmion number, which indicates they can be smoothly deformed into each other. If the cooling rate can be smoothly changed, will they observe a smooth deformation from meron to the hybrid topological solitons? If they are smoothly connected, what makes hybrid topological solitons particularly interesting? – We know that the non-topological details of the director fields are not universal but fine-tuned and sample dependent, why would we care about these details? If, on the other hand, they are not smoothly connected, is there a new topological number capturing the difference?

6 When two merons fuse, the overall skyrmion number appears to change from 1 (0) to $\pm 1/2$ locally within the localization length of the two merons. Does this indicate there will be a change of skyrmion number elsewhere in the system, or the skyrmion number is not conserved in the system?

7 The authors mentioned disclinations multiple times, but what exactly are they? Can these be defined more rigorously in terms of holonomy of the director fields?

8 Fig.2-5 suffer from the same issues as Fig.1. All elements in the figures should be clearly defined and properly explained in the corresponding captions.

Reviewer #3

(Remarks to the Author)

The manuscript Self-charged polar nematic monopoles and hybrid topological states: intertwining and domain integration by Yu Zou et al, submitted to Nature Communications, investigates the formation and collective dynamics of polar topological solitons in a ferroelectric nematic liquid crystal RM-OC2 and variant. The authors introduce a novel asymmetric confinement strategy—comprising a glass substrate and an open air interface—to break the symmetry of the anchoring conditions. This approach leads to the discovery of two distinct classes of topological excitations: chiral polar merons or monopoles and substantially more complex hybrid states featuring a nested skyrmion-toron core.

By combining Polarized Light Microscopy (PLM), Fluorescence Confocal Polarizing Microscopy (FCPM), and Second-Harmonic Generation Interferometry (SHG-I) with mean-field simulations, the authors demonstrate that these quasi-particles carry intrinsic self-charge (bound charges arising from the divergence of polarization). Furthermore, the manuscript details the intertwining rules of these solitons, showing that their interactions repulsion vs. attraction are governed by their relative screw sense. Finally, a flow-assisted seeding mechanism is proposed to engineer large-scale, programmable arrays of these monopoles, paving the way for potential applications in reconfigurable polar devices.

The manuscript addresses a timely and significant topic in soft matter physics: the control and diversification of topological solitons in polar fluids. The discovery of the hybrid skyrmion-toron state enabled by asymmetric confinement is a novel contribution that expands the current zoo of topological particles in ferroelectric nematics. The experimental characterization is rigorous, the use of FCPM to reconstruct the 3D director field offers compelling visual evidence of the proposed structures.

The concept of domain integration via thermal flow seeding is particularly impressive, as it bridges the gap between fundamental topological physics and scalable material engineering. The manuscript fits well within the scope of Nature Communications. However, there are specific concerns regarding the stability of the hybrid states and the characterization of the self-charge that require clarification to fully support the authors' claims.

1. Quantification and Screening of "Self-Charge"

The title and abstract emphasize the self-charged nature of the monopoles. While the theoretical basis ($\rho = -\nabla \cdot \mathbf{P}$) is sound, the manuscript would benefit from a more critical discussion on ionic screening.

In fluid systems, free ions typically screen bound charges over the Debye length. The authors mention in the main text that charges are partially screened by ions naturally contained in the air.

Can the authors provide an estimate of the effective net charge of these monopoles compared to the theoretical bound charge? Have the authors performed any electrophoresis experiments (tracking particle velocity under a DC field) to confirm that the monopoles indeed respond as charged particles over long distances, or are the interactions primarily dipolar/elastic? Clarifying this distinction is crucial for the claim of self-charged monopoles.

2. Thermodynamic Stability of Hybrid States (L_H/R_H)

The authors note that hybrid states appear under slow cooling, while monopoles appear under faster cooling.

This suggests that the hybrid state might be the thermodynamic ground state under asymmetric confinement, or conversely, a kinetic trap dependent on the nucleation pathway.

Could the authors clarify the long-term stability of the hybrid states? If an L_H domain is maintained at a constant temperature for an extended period (e.g., 24 hours), does it relax into the monopole state (L_M)? This information is vital for the proposed applications in "large-scale engineering," which requires temporal stability.

3. Robustness of the Air Interface

The asymmetric confinement relies on an open air interface.

While scientifically elegant, open interfaces are susceptible to environmental perturbations (humidity, dust, airflow, evaporation of volatile components).

For the device applications suggested in the conclusion, how robust is this interface? The authors should briefly discuss whether this topological effect can be replicated using a solid "capping" material (a superstrate) that mimics the degenerate planar anchoring of air, which would allow for encapsulated, durable devices.

4. Energy Scales of Intertwining

The interaction between merons is described as a competition between elastic and electrostatic forces.

It would be insightful if the authors could provide a rough comparison of the order of magnitude for the elastic interaction energy vs. the electrostatic interaction energy at the separation distances observed in Figures 2 and 3. This would help readers understand which force is the dominant driver of the self-assembly.

5. Minor Comments

Terminology (Chirality vs. Screw Sense): The distinction between molecular chirality (dopant handedness) and the topological screw sense of the meron is generally clear, but can be confusing in certain passages. I suggest adding a small schematic or table in the Extended Data clearly defining the sign conventions used for Q , screw sense, and molecular chirality to aid the general reader.

Figure 1f (FCPM): The reconstruction of the hybrid state is excellent. However, please ensure the caption or labels clearly indicate the physical scale of the "defect-core-proxy" relative to the optical resolution.

References: The citation of flexoelectric contributions is appropriate, but the authors might consider citing recent works on ferroelectric nematic drops or tactoids where surface charges also play a role, to better contextualize their work within the broader field.

My recommendation is publish with Major Revisions.

The manuscript represents a substantial advance in the field. If the authors can convincingly address the points regarding charge screening and long-term stability, the paper will be an excellent addition to Nature Communications.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I thank the authors for their response. I am satisfied with the changes and can endorse publication.

Reviewer #2

(Remarks to the Author)

The authors have addressed all of my questions, and the quality of the manuscript has been significantly improved. I now recommend it for publication.

Reviewer #3

(Remarks to the Author)

I would like to thank the authors for their detailed responses and the significant effort put into the revised manuscript. After carefully reviewing the rebuttal and the new version of the text, I find that the authors have successfully addressed the concerns raised in my initial report.

I recommend the publication/acceptance of the manuscript in its current form.

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Point-to-Point Reply to the Reviewers

In the following, the statement of the reviewer and our answers/discussions appear in *black* and *blue* fonts, respectively.

To Reviewer 1:

I have read the paper by Zou et al. The authors demonstrate that confinement asymmetry can allow for engineering solitons in fluid ferroelectrics. Overall, I find the subject and results interesting to a degree that I could be sympathetic towards publication provided some issues are cleared up.

-> We thank you for reviewing our manuscript. We agree that several aspects of the manuscript would benefit from further clarification. In the revised version, we have carefully addressed all the points you raised.

1) The authors talk about “multibody topological superstructures” this is quite confusing. One has a fluid electric with collective degrees of freedom that give rise to an effective winding number. As such, I find this term hard to justify.

-> Thank you for your comment. In the original manuscript, we used the term “superstructure” in two related contexts. First, it described the interaction of monopoles that creates complex intertwining configurations. Second, it referred to our ability to arrange these topological quasi-particles into ordered arrays using optical and geometric controls. However, as you pointed out, the term “superstructure” is more commonly associated with hierarchical organization at larger length scales, making its use in this context ambiguous. To avoid confusion and enhance clarity, we have revised the manuscript as follows.

Page 1, Line 30

(before) “The unique mechanism leads to the formation of multibody topological superstructures by intertwining their polar flux.”

(after) “The unique mechanism leads to the formation of multibody topological configurations by intertwining their polar flux.”

Page 3, Line 76

(before) “The study consists of two primary objectives: the discovery of hybrid topological states and the engineering of topological particles into superstructures.”

(after) “The study consists of two primary objectives: the discovery of hybrid topological states and the engineering of topological particles into solitonic arrays.”

Page 3, Line 85

(before) “We show that the dynamic self-assembly process can be employed to engineer large-scale polar solitonic superstructures with controlled polar and chiral orders of the particles.”

(after) “We show that the dynamic self-assembly process can be employed to engineer large-scale polar solitonic arrays with controlled polar and chiral orders of the particles.”

Page 7, Line 200

(before) “Next, we show that the topological solitons serve as elementary quasi-particles for assembling into nontrivial topological superstructures.”

(after) “Next, we show that the topological solitons serve as elementary quasi-particles for assembling into nontrivial topological configurations.”

Page 12, Line 319

(before) “Such programmable, complex domain structures offer one route toward large-scale control of polar superstructures in polar materials.”

(after) “Such programmable, complex domain structures offer one route toward large-scale control of polar solitonic networks in polar materials.”

2) The authors rely on a mean field solution, under what assumptions are these valid? Accordingly, more details on the numerical processes should be added as it is hard to pinpoint how this performed.

-> Thank you for your comment. Our theoretical framework is based on a continuum Landau-type free-energy functional, which relies on minimizing the energy of mesoscale polar textures. Continuum mean-field descriptions like this are commonly used to model micron-scale liquid crystal textures, where there is a clear distinction between molecular and structural length scales, allowing for a coarse-grained approach. In our system, the characteristic size of the observed structures is greater than c.a. 1 μm , significantly larger than the molecular scale of less than 10 nm. Furthermore, our simulations focus on stable or metastable configurations on the micrometer scale, rather than focusing on the details of defect cores or critical fluctuations of molecules near phase transitions. To enhance clarity and reproducibility for our readers, we have significantly expanded the description of the numerical simulations. The detailed implementation has been relocated from the Methods section to the Supplementary Information (Supplementary Methods 1 and 2). We made revisions in the main text as below.

Page 18, Line 475

(before) “The numerical simulation of the polarization field of the N_F LC is based on the Landau-type free energy functional, which includes the following free energy terms:”

(after) “Our theoretical framework employs a continuum Landau-type free energy functional to describe the energy minimization of mesoscale polar textures. In conventional nematics, the orientation order has head-to-tail symmetry, which can be derived from the gauge theory at the molecular scale ($< 10\text{ nm}$). Such head-to-tail symmetry reflects the apolar nature of the director $\mathbf{n}$ at the mesoscale ($\sim 100\text{ nm}$)⁵⁴⁻⁵⁷. At the mesoscale or coarse-grained level, these descriptions reduce to an effective mean-field formulation, where the free energy is expanded in terms of symmetry-allowed invariants (e.g., $\mathbf{n}$, $\mathbf{P}$). Since the structures we observed are significantly larger than molecular scales, and our focus is on (meta)stable configurations rather than critical fluctuations, a continuum description is advantageous. In the N_F phase, the head-to-tail symmetry is broken, and the corresponding continuum theory should incorporate polar terms not present in conventional nematics. The free-energy density is:”

(sentence added) “The numerical simulations were carried out in MATLAB using custom finite-difference codes. Polar fields were obtained through an iterative relaxation process of the Euler–Lagrange equations for both the director and order-parameter fields. The electrostatic potential was determined by solving the Poisson equation in Fourier space, taking the prescribed confinement geometry into account. For the simulations involving optothermal induction, additional hydrodynamic coupling was included. More details about the numerical implementation, such as grid discretization, mask construction, initialization procedures, and iterative schemes, can be found in the Supplementary Information.”

3) With respect to the above, for a mean field solution it seems that one gets an effective model that can also be viewed as gauge theory see the papers:

- 1 P. E. Lammert, D. S. Rokhsar, and J. Toner, Topology and Nematic Ordering, *Phys. Rev. Lett.* **70**, 1650 (1993).
- 2 P. E. Lammert, D. S. Rokhsar, and J. Toner, Topology and Nematic Ordering. I. A Gauge Theory, *Phys. Rev. E* **52**, 1778 (1995).
- 3 {Nissinen, 2016 #52}4 Ke Liu, Jaakko Nissinen, Robert-Jan Slager, Kai Wu, Jan Zaanen, Generalized Liquid Crystals: Giant Fluctuations and the Vestigial Chiral Order of $\mathbb{P}^1$, and $\mathbb{P}^1$ Matter, *Phys. Rev. X* **6**, 041025.

There has been recent work on defining merons for polar structures especially on moiré systems, including their polarization for subdomains and local markers, see

- 1 D Bennett, G Chaudhary, RJ Slager, E Bousquet, P Ghosez, *Nature Communications* **14** (1), 1629
- 2 D Bennett, WJ Jankowski, G Chaudhary, E Kaxiras, RJ Slager, *Physical Review Research* **5** (3), 033216

that moreover also have been linked to optical responses see

- 3 A Agarwal, WJ Jankowski, D Bennett, RJ Slager, *npj 2D Materials and Applications* **9** (1), 43

These works clearly fit the general scope of introduction and ought to be mentioned.

-> Thanks for your comment. In particular, Lammert *et al.* showed that the head–tail symmetry of nematics can be described as a local $\mathbb{Z}_2$ gauge invariance, while Nissinen *et al.* generalized this perspective to a broad class of point-group symmetries within a unified gauge-theoretical approach. While these approaches describe orientational symmetry and topological states in terms of local gauge invariance, our study adopts a continuum mean-field description based on an effective free-energy functional, which can be regarded as a coarse-grained limit of such formulations. Our focus is on mesoscale polarization–topology coupling in the N_F phase, where spontaneous polarization $\mathbf{P}$ generates bound charge ($\rho = -\nabla \cdot \mathbf{P}$) and long-range electrostatic interactions. The mean-field framework enables a direct comparison between elastic and electrostatic energy contributions and allows quantitative reconstruction of POM and FCPM textures for comparison with experiments. Following your suggestion, we have included these references in the Introduction to better situate our work within the broader theoretical context.

We also acknowledge the recent pioneering studies on polar topological textures in moiré systems. Bennett *et al.* and Jankowski *et al.* reported the formation mechanisms and theoretical characterization of polar meron networks in moiré superlattices, while Agarwal *et al.* demonstrated that such polarization textures can generate shift photocurrent vortices, providing a nonlinear optical route for probing topological structures. These studies highlight the broader universality of polar topological physics across different length scales and material platforms. Following your suggestion, we have incorporated these references into the Introduction and added brief discussion to better contextualize our work within this rapidly developing field.

Page 2, Line 55

(sentence added) “The exploration of these polar topologies has recently extended to moiré superlattices, where polar meron networks have been identified and characterized through quantitative subdomain markers^{17, 18}. Crucially, these topological states have been linked to nonlinear optical signatures like shift photocurrent vortices, providing a sophisticated means of optical detection¹⁹.”

Page 18, Line 475

(before) “The numerical simulation of the polarization field of the N_F LC is based on the Landau-type free energy functional, which includes the following free energy terms:”

(after) “Our theoretical framework employs a continuum Landau-type free energy functional to describe the energy minimization of mesoscale polar textures. In conventional nematics, the orientation order has head-to-tail symmetry, which can be derived from the gauge theory at the molecular scale (< 10 nm). Such head-to-tail symmetry reflects the apolar nature of the director $\mathbf{n}$ at the mesoscale (~ 100 nm)⁵⁴⁻⁵⁷. At the mesoscale or coarse-grained level, these descriptions reduce to an effective mean-field formulation, where the free energy is expanded in terms of symmetry-allowed invariants (e.g., $\mathbf{n}$, $\mathbf{P}$). Since the structures we observed are significantly larger than molecular scales, and our focus is on (meta)stable configurations rather than critical fluctuations, a continuum description is advantageous. In the N_F phase, the head-to-tail symmetry is broken, and the corresponding continuum theory should incorporate polar terms not present in conventional nematics. The free-energy density is:”

To Reviewer 2:

This paper reports an experimental study of topological solitons in fluid ferroelectrics discovered in recent years. The authors explicitly demonstrate the existence of hybrid topological solitons and explore fusion between elementary topological solitons. While the experimental results are interesting and the theoretical explanation is, in broad terms, reasonable to me. However, the novelty and importance of their contribution are not clearly conveyed, like why should we care about these topological patterns of director fields they discovered and why the fusion between topological solitons can be done in their scheme but not others. I will have more detailed questions regarding these points below. Minorly, the organization and presentation of the manuscript are poor, which will hinder its accessibility to a broader audience. A major revision is therefore necessary to clearly convey the key results and their significance. Below, I list my comments and questions point by point.

-> We thank you for reviewing our manuscript. We agree that several aspects of the manuscript would benefit from further clarification. We agree that the novelty and broader significance of our results were not sufficiently emphasized in the original manuscript. In the revised version, we have placed greater emphasis on the novelty and broader significance of our results. We have substantially clarified (i) the conceptual advancement of identifying hybrid polar topological solitons in a fluid ferroelectric, (ii) the physical mechanism that enables controlled fusion through electrostatic–elastic coupling, and (iii) the distinctions between our platform and previously studied systems. Additionally, we have reorganized the manuscript and improved the presentation to enhance clarity and accessibility.

1) The authors claim that their work aims to construct “higher-order topological structures.” However, they do not clearly define what is meant by this term. Does it refer to topological solitons with higher skyrmion number, or to some other type of topological invariant? This does not appear to be the case, since all of the solitons studied ultimately carry a skyrmion number $Q=\pm 1/2$. A clear and precise definition of “higher-order topological structures” is therefore necessary.

-> Thank you for pointing out this ambiguity. We agree that the term “higher-order topological structures” was not sufficiently precise, as also questioned by the reviewer 1, and may lead to confusion regarding the underlying topological invariants. In the original manuscript, the terms “higher-order” as well as “superstructure” were used in two related contexts. First, it described the interaction of monopoles that creates complex intertwining configurations. Second, it referred to our ability to arrange these topological quasi-particles into ordered arrays using optical and geometric controls. However, as you pointed out, the terms are more commonly associated with higher topological invariants and hierarchical organizations, making their use in this context ambiguous. To avoid confusion and enhance clarity, we have revised the manuscript as follows.

Page 1, Line 28

(before) “We discover nonclassical electric monopoles and hybrid topological states, including

chiral meron and anti-meron variants and their higher-order hybridization with a nested skyrmion core.”

(after) “We discover nonclassical electric monopoles and hybrid topological states, including chiral meron and anti-meron variants and their composite hybridization with a nested skyrmion core.”

Page 2, Line 52

(before) “Although ground-state elementary solitons have been well explored, how to combine and stabilize multiple topological orders into a higher-order topological soliton, thus generating new, distinct, and novel phases, remains unclear.”

(after) “Although ground-state elementary solitons have been well explored, how to combine and stabilize multiple topological textures into organized solitonic assemblies, thus generating new, distinct, and novel phases, remains unclear.”

Page 3, Line 65

(before) “A few previous observations, though not yet designable and controllable, suggest that assemblies of some polar solitons to higher-order structures may trigger nonclassical and exotic phenomena.”

(after) “A few previous observations, though not yet designable and controllable, suggest that assemblies of some polar solitons to integrated topological architectures may trigger nonclassical and exotic phenomena.”

Page 3, Line 74

(before) “In this work, we challenge to construct high-order topological structures.”

(after) “In this work, we challenge to construct integrated topological architectures.”

Page 3, Line 83

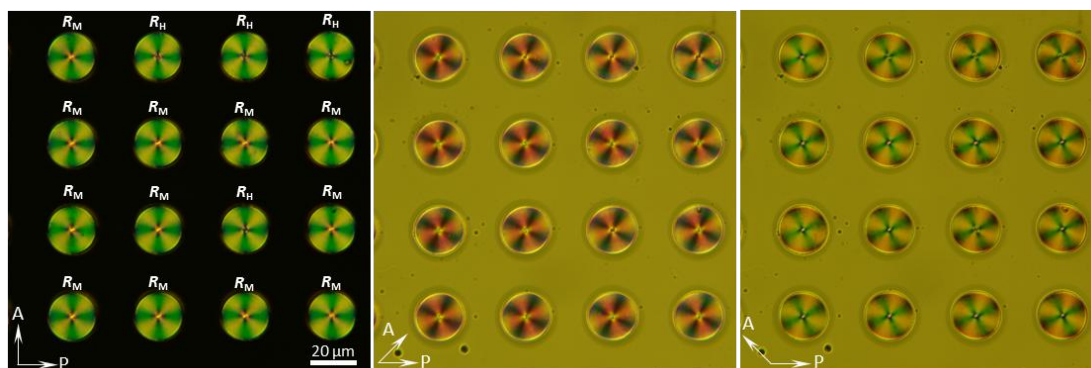
(before) “Polar orientational interactions between these particles drive unique higher-order assemblies, resulting in novel topological enchantments like bi- or triplet-merons and kinked loops.”

(after) “Polar orientational interactions between these particles drive unique domain-structural assemblies, resulting in novel topological configurations like bi- or triplet-merons and kinked loops.”

2) What is the size of the solitons relative to the overall system? Are they highly localized?

-> We thank you for your important question. In our experiments, each meron or hybrid soliton corresponds to 3D polar configuration formed within a single patterned circular pit. The characteristic size of a soliton is therefore set by the micro-confinement, with a typical diameter of 25–30 μm , matching the size of an individual pit. The solitons are highly localized in position at the pit scale. By pre-patterning the glass substrate with arrays of circular pits, we spatially confine the nucleation and growth of the N_F phase from the isotropic state, ensuring that each soliton is deterministically positioned within a designated cavity (please see Supplementary Fig. 1d below). This hierarchy—where individual mesoscale solitons are localized into a macroscale

array—is central to our "topological engineering" approach, allowing for the creation of stable and designable collective configurations that are absent in unconstrained bulk systems.



Supplementary Fig. 1d. At an intermediate cooling rate, twisted meron (labelled R_M) and the hybrid (labelled R_H) grow concurrently within a micro-patterned array.

3) Fig 1 is particularly unclear:

(a) In a and b, what are the upper and lower parts respectively? The surface and bulk patterns of director fields? If so, why is the winding in x-y plane opposite at the surface and in the bulk? What are the colored lines there? Preimages of director field with fixed θ and ϕ ? Why in b there is only one bottom surface shown in the lower-left subfigure?

-> We thank you for the careful reading of Fig. 1 and for pointing out several points that required clarification.

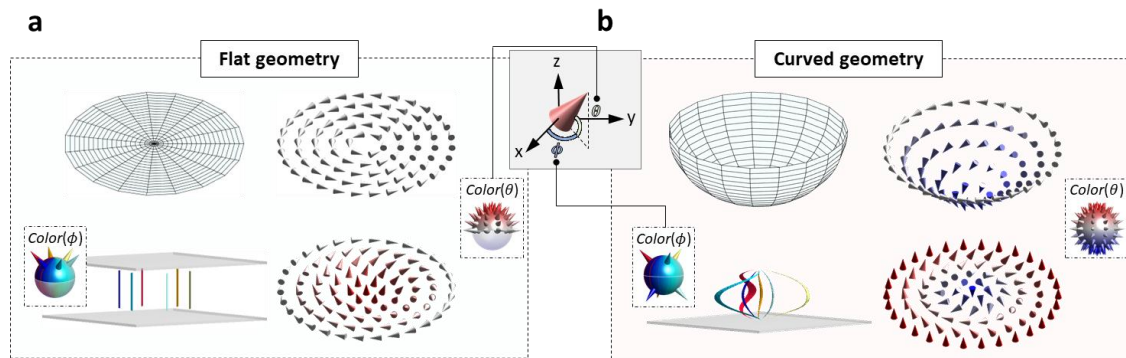
In panels (a) and (b), the upper parts display surface geometry and director fields, while the lower parts show the 3D confinement and the bulk polarization field (at middle plane).

As you correctly noted, the apparent reversal in the director field's winding between the surface and bulk was a schematic error. We have revised the figure to ensure the winding direction is consistent throughout the sample.

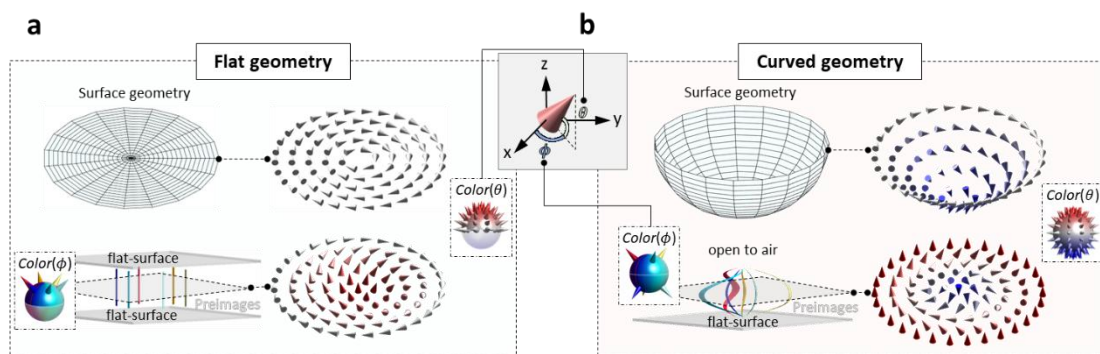
The colored lines represent preimages of the director at fixed angles (θ, ϕ) , where θ is the elevation angle and ϕ is the azimuth angle. In panel (a), six preimages are shown for $\theta = 45^\circ$ with ϕ varying from 0° to 300° in 60° steps. In panel (b), ϕ follows the same sequence, while θ alternates between 45° and -45° . These explanations have now been added to the figure caption.

In panel (b), only one bottom surface is depicted because the N_F bubble is confined within a circular pit that is open at the top. While the bottom interface is flat against the substrate, the top surface is curved and exposed to air. This schematic illustrates the resulting asymmetric confinement geometry.

(before)



(after)



Page 23, Line 565, Caption of Figs. 1a and b

(before) “**a**, In a conventional flat cell with planar anchoring, the N_F LC adopts an in-plane circular flux at both surfaces, giving rise to a meron-like configuration in the bulk. In the absence of chirality, preimages with different polar orientations map onto straight lines in S^2 space. **b**, When one allows a curved interface to confine N_F LCs, the surface polarization is allowed to geometrically tilt out of plane. This curvature-enabled boundary condition promotes the formation of a skyrmion- or toron-like configuration.”

(after) “**a**, In a flat cell with planar anchoring (left), circular in-plane polar flux at the surfaces (upper right) gives rise to a meron configuration in the N_F bulk (lower right). In the absence of chirality, the polarization preimages map onto straight lines (lower left) on the S^2 manifold (sphere colored by ϕ). Six representative preimages are shown as colored lines at $\theta = \pi/3$, with ϕ varying from 0 to $5\pi/3$ in steps of $\pi/3$. **b**, In a top-open confinement with an LC–air interface (left), the curved boundary promotes out-of-plane tilting of the polarization (upper right), stabilizing skyrmion- or toron-like configurations in the N_F bulk (lower right). The corresponding polarization preimages map onto curved trajectories (lower left) on the S^2 manifold. Here, θ alternates between $\pm\pi/3$ with ϕ varying from 0 to $5\pi/3$ in steps of $\pi/3$.”

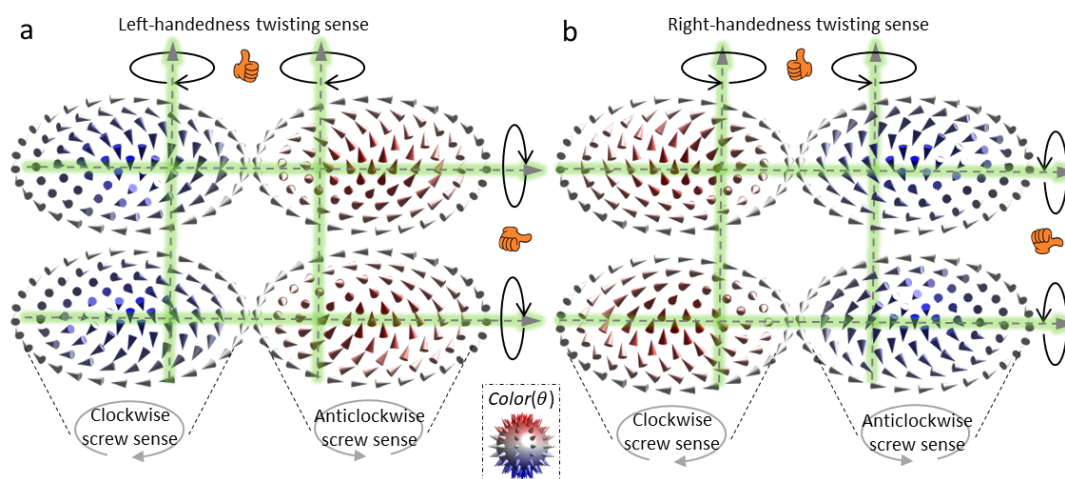
(b) In c and d, is it possible to put the scale bar in the figure? (note there is also a typo: scare & scale, there are lots of similar typos throughout the manuscript which I will not list one by one). The corresponding text defining $L_{M/H}$ and $R_{M/H}$ is also unclear and confusing. What exactly is meant by the screw and twisting sense? What defines the chirality, is it the

combination of twisting and screw? Is it true that the screw sense, rather than the overall chirality, is important when considering the intertwining of multiple bodies?

-> In the revised figure, we have added the length of the scale bars to panels c and d. We also corrected the typographical error (“scare” → “scale”). We have also carefully proofread the manuscript to remove similar typos.

The twisting sense refers to the handedness (left- or right-handed) of how directors rotate in space along a given axis. The twisting sense of the polar fields is dictated by the system's chirality, controlled here by doping N_F materials with S811 (left-handed) or R811 (right-handed) chiral agents.

The screw sense describes the clockwise or anticlockwise rotation of directors within the xy-plane. For achiral N_F droplets, both configurations are energetically equivalent and appear with equal probability during nucleation (see figure below). To improve clarity, we also added a *Supplementary Tables 2* that explicitly define the twisting sense, the screw sense, and other topology-related concepts.



Supplementary Fig. 1. a-b, Schematics illustrating twisting sense and screw sense, as well as how they are coupled in a polar meron.

It is the screw sense, rather than the global chirality, that governs the intertwining of multiple bodies. Specifically, it determines whether the polarization vectors of two approaching solitons are syn-parallel or anti-parallel at their interface: the former enables smooth merging, while the latter leads to the formation of a domain wall.

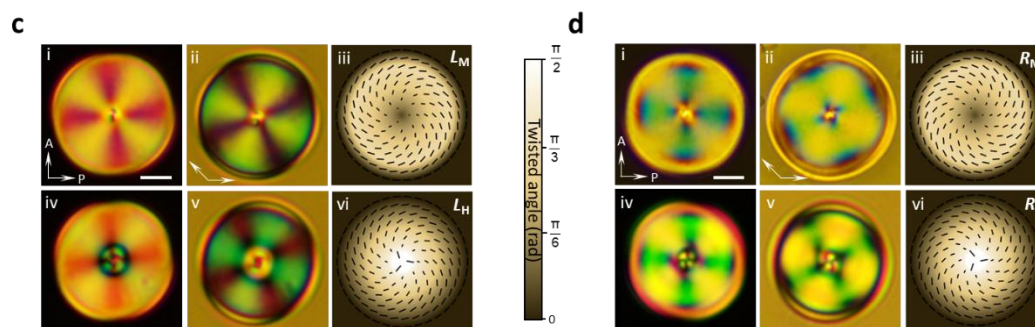
In our experiments, the global twist sense is fixed by the chiral dopant. For instance, in Figs. 2 and 3 (doped with S811), the polarization field is left-handed. However, the screw sense during meron nucleation can be either clockwise or anticlockwise. Clockwise rotation produces a meron with upward central polar flux (skyrmion number $+1/2$), whereas anticlockwise rotation yields downward flux (skyrmion number $-1/2$). Therefore, while the twist sense is fixed, the screw sense remains a local degree of freedom that determines the sign of the meron's skyrmion number and ultimately controls the outcome of soliton intertwining.

(before) “Decrossing the analyzer in the anticlockwise (clockwise) manner leads to the extinction of L_M and L_H (R_M and R_H) bubbles, enabling the visualization of the opposite twisting sense of topology along the depth z -direction. It is essential to note that the twisting sense couples to the screw sense of meron. The screw sense here refers to the direction of in-plane rotation of polar flux, where the screw axis along the depth direction is either upward or downward in the meron's core.”

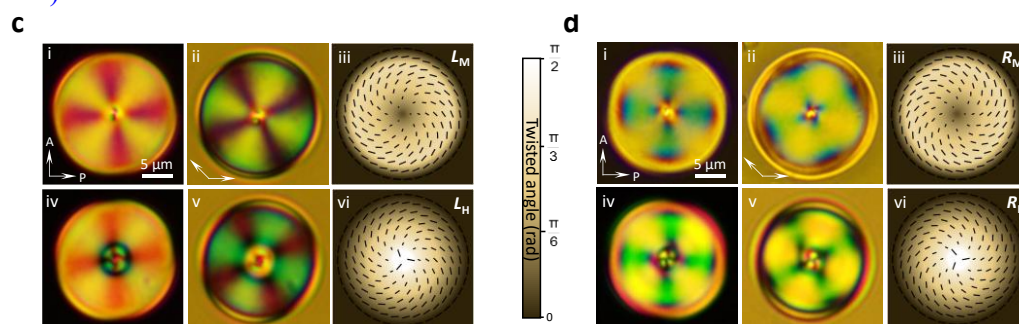
(after) “Decrossing the analyzer in the anticlockwise (clockwise) manner leads to the extinction of L_M and L_H (R_M and R_H) bubbles, enabling the visualization of the opposite twisting sense - the helical handedness of the polarization field along the depth z -direction. It is essential to note that the twisting sense couples to the screw sense of meron, which defines the direction of in-plane rotation of polar flux. In a polar meron, the polarization $\mathbf{P}$ gradually tilts from in-plane at the boundary to out-of-plane at the core, where the polar flux points either upward or downward along the z -direction.”

Page 23, Fig. 1. c and d

(before)



(after)



(c) In e and f, what do the rectangles on the right side correspond to? There is no v and vi labelled in the figure. Again, the colored lines are not defined.

-> The dashed rectangles in panels e and f are used only as visual guides to help relate the spatial region of the polarization field reconstruction to the corresponding preimage structures shown below. In particular, the large rectangle indicates the outer region of the structure, while the smaller rectangle highlights the inner core region. Regarding the labeling, panels e and f only contain subpanels i–iv, and there are no subpanels v or vi associated with these figures. Finally, as noted above, the colored curves represent preimages of the polarization field. We

have added a brief explanation of this visualization method in the figure caption.

Page23, Line 579, Caption of Fig. 1.

(sentence added) “The colored curves represent preimages of distinct polarization directions on S^2 : in (e), θ is fixed at $\pi/4$ with ϕ varying from 0 to $5\pi/3$ in steps of $\pi/3$; in (f), θ alternates between $\pm\pi/4$ with ϕ varying from 0 to $5\pi/3$ in steps of $\pi/3$. Dashed rectangles indicate regions used for correlating reconstructed polar fields with S^2 preimage trajectories. The inset in f (iii) shows magnified bubble core, whose characteristic radius r_d , is $\sim 1-2 \mu\text{m}$, well above the submicron lateral resolution of the FCPM ($\text{NA} = 1.4$). In all panels, cones represent the polarization vectors, with color indicating the elevation angle θ .”

(d) The 3D color maps are also confusing. Why is a 3D plot needed even if the ϕ and θ are simply encoded using different color maps?

-> We have used two color maps to better visualize the topological nature. While color (θ) is used to visualize the polar field itself, color (ϕ) is used to distinguish preimages on the S^2 manifold. By doing so, the variations in color under color (θ) mapping directly reflect how the polarization vector twists in space. The 3D representation of these preimages enables a clear visualization of how different trajectories are distributed and linked in space, thereby revealing the topology of the structure. For example, in Fig. 2d below, by color-coding preimages from the upper (S^2 northern hemisphere; orange, sky blue, gray-white) and lower (S^2 southern hemisphere; cyan, purple, yellow) hemispheres separately, the sign of the local skyrmion number ($Q = \pm 1/2$) becomes immediately identifiable.

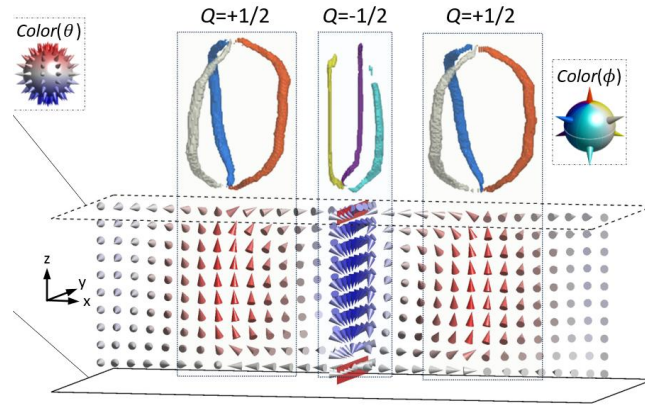


Fig. 2. d, The curves represent preimages of distinct polarization directions on S^2 : three from the northern hemisphere [$\theta = \pi/4$; $\phi = -\pi/6$ (gray-white), $\phi = \pi/2$ (orange-red), and $\phi = 7\pi/6$ (sky-blue)] and three from the southern hemisphere [$\theta = -\pi/4$; $\phi = \pi/6$ (yellow), $\phi = 5\pi/6$ (purple), and $\phi = 3\pi/2$ (cyan)].

4) In Fig.1 b, the author illustrated how curved boundaries can lead to the hybrid topological solitons, and then in the text they claimed these hybrid topological solitons are realized in experiments by tuning the cooling rate. They should specify the relationship between the cooling rate and the presence of curved boundaries there instead of postponing it later.

-> Thanks for this suggestion. We agree that the relationship between the cooling rate and the resulting boundary geometry should be established early in the manuscript: Curved surfaces naturally facilitate out-of-plane polarization tilting—a prerequisite for stabilizing the hybrid state with skyrmion- or toron-like cores. In our patterned confinement with an open top surface, N_F bubbles can nucleate on either flat or curved boundaries depending on the cooling rate: fast cooling leads to bulk nucleation and relatively flat interfaces, whereas slow cooling promotes nucleation near the LC–air interface, resulting in hemispherical droplets with curved boundaries. Crucially, flat closed cells fail to support these hybrids under any cooling rates. As suggested, we now clarify the relationship between cooling rate and boundary geometry early in the manuscript.

Page 5, Line 122

(sentence added) “The topological diversity stems from the cooling-rate-dependent nucleation pathway. The nucleation geometry of the N_F bubbles changes from cylindrical to hemispherical as the cooling rate decreases. While the cylindrical confinement with flat boundaries favors the axisymmetric formation of monopoles, the hemispherical confinement introduces geometric asymmetry that facilitates the stabilization of hybrid topological solitons (see **Discussion**).”

5) In what sense are the hybrid topological solitons different from merons? Topologically, they have the same skyrmion number, which indicates they can be smoothly deformed into each other. If the cooling rate can be smoothly changed, will they observe a smooth deformation from meron to the hybrid topological solitons? If they are smoothly connected, what makes hybrid topological solitons particularly interesting? – We know that the non-topological details of the director fields are not universal but fine-tuned and sample dependent, why would we care about these details? If, on the other hand, they are not smoothly connected, is there a new topological number capturing the difference?

-> Thank you for your question. We agree that merons and hybrid solitons belong to the same topological category, as they both have the same skyrmion number ($Q = \pm 1/2$). However, in our system, these two configurations correspond to distinct metastable minima in the free-energy landscape (see Supplementary Fig. 4). Converting a hybrid soliton into a meron involves unwinding and reversing the polar field, which requires overcoming significant polarization-gradient and electrostatic energy barriers. Our numerical simulations demonstrate that this energy barrier is much larger than thermal fluctuations, preventing spontaneous transformation between the two states, even when the cooling rate is varied smoothly. To clarify this point, we have included the calculated time evolution of free energy during this transition in the Supplementary Information. The significance of the hybrid soliton, therefore, lies not in introducing a new topological invariant, but in demonstrating that ferroelectric liquid systems can host multiple energetically (meta)stable polarization textures. This degeneracy reflects the complex interplay among elasticity, chirality, and electrostatics in polar liquid crystals, and provides additional structural degrees of freedom for engineering solitonic states in confined geometries. Of course, such a potential of topological switching between multiple metastable states may offer an essential platform for exploring switchable topological optics and memory.

Page 5, Line 122

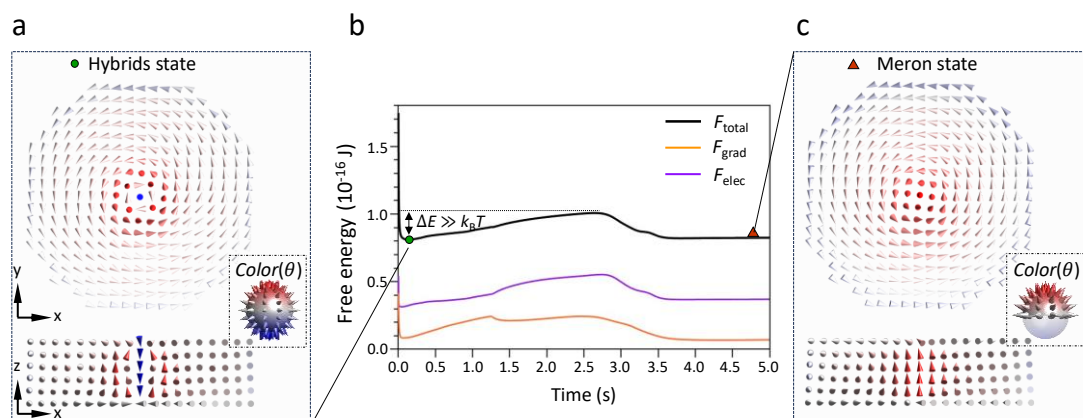
(sentence added) “The topological diversity stems from the cooling-rate-dependent nucleation pathway. The nucleation geometry of the N_F bubbles changes from cylindrical to hemispherical as the cooling rate decreases. While the cylindrical confinement with flat boundaries favors the axisymmetric formation of monopoles, the hemispherical confinement introduces geometric asymmetry that facilitates the stabilization of hybrid topological solitons (see **Discussion**). Once they are formed, both monopole and hybrid states remain stable, persisting for at least several days at room temperature without structural switching and degradation.”

Page 7, Line 183

(sentence added) “Notably, the skyrmion-toron core is energetically stable. Transformation between the hybrid topology and the twisted meron must overcome energy barriers caused by polarization gradients and electrostatic interactions (see Supplementary Fig. 4).”

Supplementary information, Supplementary Fig. 4

(Figure added)

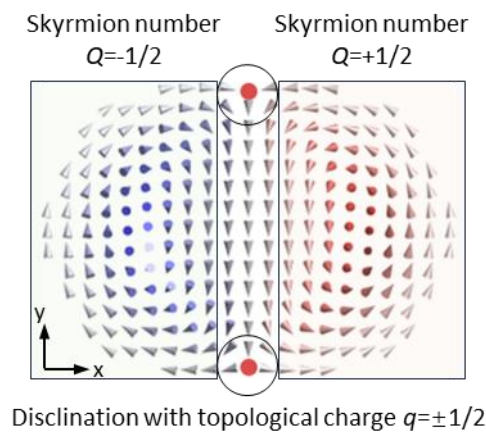


Supplementary Fig. 4. Simulated transformation between hybrid and meron solitons. **a**, Reconstructed polarization field of the hybrid soliton obtained from numerical simulation. Polarization vectors are colored by elevation angle θ . **b**, Evolution of the free energy during the simulated transformation from the hybrid state to a meron state. The total free energy F_{total} (black), polarization gradient energy F_{grad} (orange), and electrostatic energy F_{elec} (purple) are shown. The transformation requires overcoming an energy barrier $\Delta E \gg k_B T$, indicating that the two configurations are separated by a substantial energetic barrier despite being topologically connected. **c**, Final polarization configuration corresponding to the meron state after relaxation. Please refer to the *supplementary Table I* for parameter settings for simulation. In all panels, cones represent the polarization vectors, with color indicating the elevation angle θ .

6) When two merons fuse, the overall skyrmion number appears to change from 1 (0) to $\pm 1/2$ locally within the localization length of the two merons. Does this indicate there will be a change of skyrmion number elsewhere in the system, or the skyrmion number is not conserved

in the system?

-> The skyrmion number is not strictly conserved locally during the fusion process. During this fusion event, the two merons initially reside in separate confined regions with skyrmion numbers $Q = +1/2$ (anticlockwise polar flux in the xy plane) and $Q = -1/2$ (clockwise polar flux in the xy plane). Upon contact, they merge into a single confinement where the boundary polarization becomes discontinuous as seen in the figure below. These discontinuities manifest as disclination lines at the interface between the two solitons (see figure below). Local conservation of the skyrmion number breaks down here, as topological protection applies only when boundary conditions remain static and continuous.



In this intermediate intertwined state, the total skyrmion number of the merged region vanishes ($Q = +1/2 + (-1/2)$), while the defect lines carry the corresponding topological charge ($\pm 1/2$). As the system further relaxes, one meron is expelled toward the boundary and annihilates together with the associated disclination arcs. After this defect-mediated process, the system recovers a single-meron configuration without disclinations, with a final skyrmion number of $Q = \pm 1/2$.

7) The authors mentioned disclinations multiple times, but what exactly are they? Can these be defined more rigorously in terms of holonomy of the director fields?

-> Thank you for pointing this out. In our manuscript, disclinations refer to line defects in the polarization (director) field where the polarization orientation becomes singular. More rigorously, they can be defined through the rotation of the director field along a closed loop encircling the defect. Specifically, when the director field is transported around such a loop, the total rotation angle satisfies $\Delta\theta = 2\pi s$, where s denotes the disclination strength. In our system, the defects appearing at the interface of intertwining merons correspond to half-integer disclinations ($s = \pm 1/2$), which is consistent with the standard classification of defects in nematic-like orientational fields. We agree that this point should be stated more clearly and have therefore added a brief clarification in the revised manuscript.

Page 9, Line 238

(sentence added) “This disclination represents a line defect in the polarization field, characterized by a $\pm 1/2$ winding of the polar flux around the defect core (Fig. 3c).”

8) Fig.2-5 suffer from the same issues as Fig.1. All elements in the figures should be clearly defined and properly explained in the corresponding captions.

-> We apologize for the lack of clarity in the previous figure descriptions. Following your suggestion, we have carefully revised the captions of Figs. 2–5 to ensure that all graphical elements, symbols, and color encodings are clearly defined and explained. We have also reviewed the figures throughout the manuscript to remove similar ambiguities and to ensure that the captions are self-contained.

To Reviewer 3:

The manuscript "Self-charged polar nematic monopoles and hybrid topological states: intertwining and domain integration" by Yu Zou et al, submitted to Nature Communications, investigates the formation and collective dynamics of polar topological solitons in a ferroelectric nematic liquid crystal RM-OC2 and variant. The authors introduce a novel asymmetric confinement strategy—comprising a glass substrate and an open-air interface—to break the symmetry of the anchoring conditions. This approach leads to the discovery of two distinct classes of topological excitations: chiral polar merons or monopoles and substantially more complex hybrid states featuring a nested skyrmion-toron core. By combining Polarized Light Microscopy (PLM), Fluorescence Confocal Polarizing Microscopy (FCPM), and Second-Harmonic Generation Interferometry (SHG-I) with mean-field simulations, the authors demonstrate that these quasi-particles carry intrinsic self-charge (bound charges arising from the divergence of polarization). Furthermore, the manuscript details the intertwining rules of these solitons, showing that their interactions (repulsion vs. attraction) are governed by their relative screw sense. Finally, a flow-assisted seeding mechanism is proposed to engineer large-scale, programmable arrays of these monopoles, paving the way for potential applications in reconfigurable polar devices. The manuscript addresses a timely and significant topic in soft matter physics: the control and diversification of topological solitons in polar fluids. The discovery of the hybrid skyrmion-toron state enabled by asymmetric confinement is a novel contribution that expands the current zoo of topological particles in ferroelectric nematics. The experimental characterization is rigorous, the use of FCPM to reconstruct the 3D director field offers compelling visual evidence of the proposed structures. The concept of domain integration via thermal flow seeding is particularly impressive, as it bridges the gap between fundamental topological physics and scalable material engineering. The manuscript fits well within the scope of Nature Communications. However, there are specific concerns regarding the stability of the hybrid states and the characterization of the self-charge that require clarification to fully support the authors' claims.

-> We thank you for reviewing our manuscript. You have raised important questions regarding the stability of the hybrid states and the characterization of the self-charge associated with the polar solitons. These points are very helpful for clarifying the presentation of our results. In the revised manuscript and the reply below, we provide additional explanations and clarifications to address these concerns.

1) Quantification and Screening of "Self-Charge". The title and abstract emphasize the self-charged nature of the monopoles. While the theoretical basis ($\rho = -\beta(\nabla \cdot \mathbf{P})$) is sound, the manuscript would benefit from a more critical discussion on ionic screening. In fluid systems, free ions typically screen bound charges over the Debye length. The authors mention in the main text that charges are partially screened by ions naturally contained in the air. Can the authors provide an estimate of the effective net charge of these monopoles compared to the theoretical bound charge?

-> Thank you for raising this important point regarding ionic screening. For RM-OC₂, the spontaneous polarization is on the order of $P_0 \sim 10^{-2} \text{ C m}^{-2}$. In the absence of screening this

polarization would generate depolarizing fields of magnitude $E \sim P_0/\epsilon$, corresponding to $E \sim 10^6 - 10^7 \text{ V m}^{-1}$. The associated electrostatic energy density (P_0^2/ϵ) would therefore be on the order of $10^5 - 10^6 \text{ J m}^{-3}$, which exceeds the typical nematic elastic energy density by three to four orders of magnitude. However, our numerical simulations and experimental observations show that electrostatic and elastic energies compete at comparable levels in determining the solitonic structures. This implies that the electrostatic contribution must be strongly reduced by ionic screening ($P_{\text{eff}} = \beta P_0$). From this energetic comparison, we estimate that the effective screening factor satisfies approximately $\beta \approx 10^{-3}$, meaning that only one-thousandth of the bound charge remains unscreened. This estimate is also consistent with prior studies indicating that partial ionic screening is a generic feature of N_F systems (please refer to [Zavvou E et.al., *Soft Matter* **18**, 8804-8812 (2022)] and [Máthé MT et.al., *Physical Review E* **105**, L052701 (2022)]). We have clarified this point in the revised manuscript.

Page 13, Line 345

(sentence added) “Such screening is expected to reduce the effective bound charge to approximately 10^{-3} of its nominal value (see the estimation in Methods), thereby allowing electrostatic and other free energies to compete on comparable scales.”

Page 19, Line 509

(before) “The value range of β in the simulation is 0.001 to 0.01.”

(after) “Following the order-of-magnitude estimate used in the Ref 31, we note that in the unscreened N_F state, the spontaneous polarization $|\mathbf{P}| \sim 10^{-2} \text{ C m}^{-2}$ would generate depolarizing fields of order $E \sim P/\epsilon \approx 10^6 - 10^7 \text{ V m}^{-1}$ with the dielectric constant $\epsilon \sim 10^{-9} - 10^{-8} \text{ F m}^{-1}$, corresponding to electrostatic energy densities ($\sim \frac{1}{2} \epsilon E^2$) of $10^5 - 10^6 \text{ J m}^{-3}$. This value is several orders of magnitude larger than typical nematic elastic energies. The observation that electrostatic and elastic contributions compete in our simulations therefore implies strong ionic screening ($E_{\text{eff}} = \beta E$), corresponding to an effective screening factor $\beta \sim 10^{-3}$. This value is also comparable in magnitude to previous estimates inferred from independent experiments, such as electro-optic measurements³⁴ and measurements of the effective splay elastic constant K_{11} ²⁴, supporting the physical reasonableness of the present treatment.”

2) Have the authors performed any electrophoresis experiments (tracking particle velocity under a DC field) to confirm that the monopoles indeed respond as charged particles over long distances, or are the interactions primarily dipolar/elastic? Clarifying this distinction is crucial for the claim of self-charged monopoles.

-> Thank you for the suggestion. First, we clarify that, while these solitons are stable in confinement, as in our experiments, they cannot be embedded in a homeotropic or planar far-field background at this stage. Therefore, we have tried two complementary experiments as follows. In the first experiment, an in-plane DC electric field was applied to the N_F bubble in the current confinement. We observed that both meron and hybrid soliton cores translate in a direction perpendicular to the applied field while preserving their internal structure. This

behavior reflects the strong polarization–field coupling characteristic of N_F states rather than conventional electrophoretic motion. Because the coupling term $-\mathbf{P} \cdot \mathbf{E}$ strongly favors polarization reorientation, the system can reduce its electrostatic energy primarily through rearrangement of the surrounding polarization texture instead of translating the solitonic cores as charged particles.

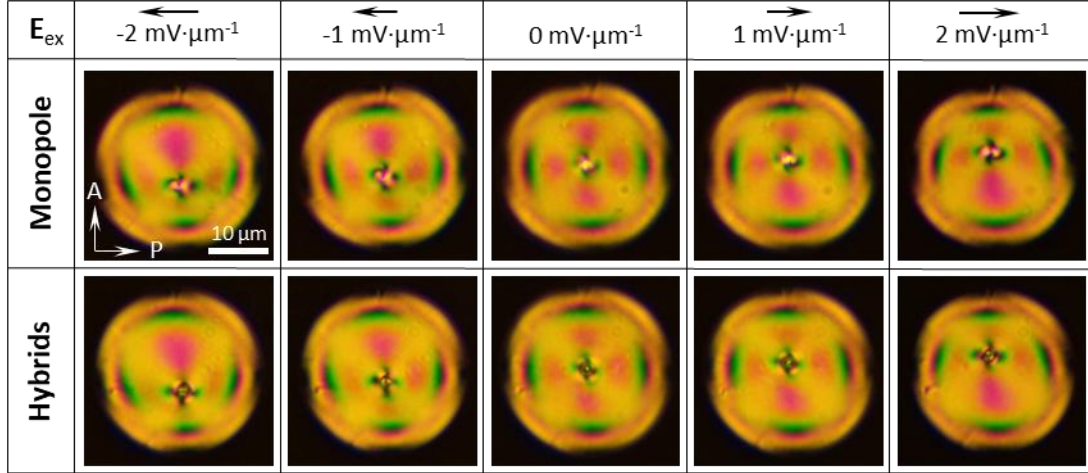


Figure. Field-driven translation of topological solitons in a confined N_F bubble. Time-lapse POM images showing the response of soliton cores under an in-plane DC electric field. Both meron and hybrid soliton cores exhibit a steady translational motion perpendicular to the applied field E_{ex} , as indicated by the black arrow, while their internal topological structures remain intact throughout the process. Scale bar, $10 \mu\text{m}$.

In the second experiment, we have tried a tricky thing: while these solitons cannot be embedded in a homeotropic or planar far-field background, we found that they can be temporarily stabilized by floating in an isotropic liquid background during the isotropic- HN^* phase transition (please see Figure below). This makes us possible to apply a DC field to the movable solitons. We observe a field-dependent crossover: At low fields ($< 0.1 \text{ mV} \cdot \mu\text{m}^{-1}$), the motion of solitons' core remains predominantly perpendicular to the field, consistent with polarization reorientation. There is no electrophoresis of the quasi-particles in this range. However, at higher fields ($\geq 1 \text{ mV} \cdot \mu\text{m}^{-1}$), the core of the soliton is pushed toward the boundary, and the entire soliton exhibits a notable drift in the direction of the electric field, resembling electrophoretic motion. This observation supports the self-charged nature of the solitons.

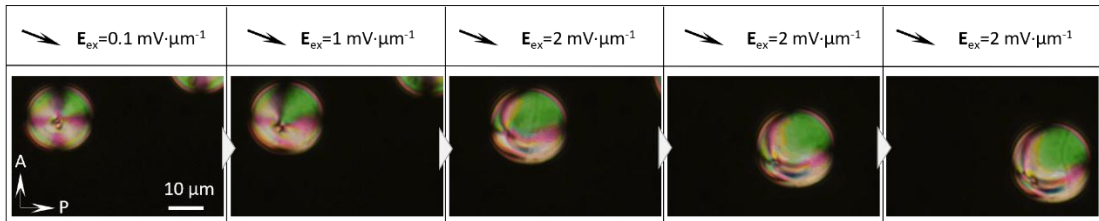


Figure. Time-lapse POM images show that the N_F bubble undergoes directional motion accompanied by internal reorganization of the polar texture. The displacement occurs predominantly along the applied field E_{ex} , as indicated by the black arrow, while the overall droplet integrity is maintained. The scale bar is $10 \mu\text{m}$.

To further support this point, we conducted a preliminary measurement of the drift velocity under various in-plane DC fields, as illustrated in following Figure. The result demonstrates a clear field-dependent behavior with a threshold, providing an initial demonstration of the relationship between velocity and field strength.

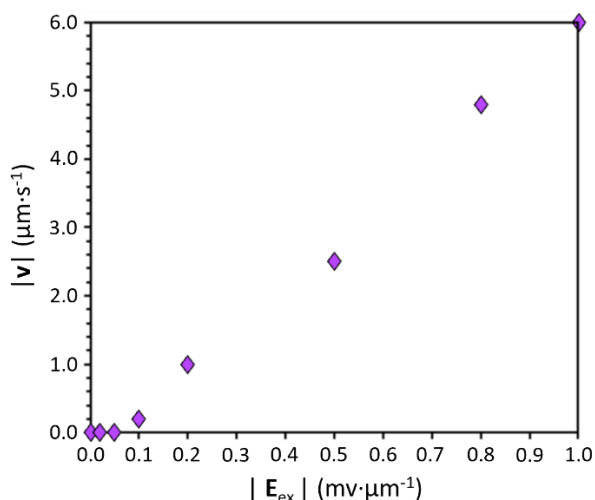


Figure. Bubble velocity $|v|$ as a function of the applied in-plane electric field $|E_{\text{ex}}|$. The purple rhombuses represent the experimental data obtained directly from the measurements.

Yet, it is obvious that many aspects of the dynamics differ from traditional electrophoresis. First, the electrophoresis-like phenomenon only occurs when a specific field threshold is reached. Second, the initiation of solitonic motion is complex due to the reorganization of the polar field induced by the electric field both before and during the electrophoresis process. These findings suggest that charge accumulation due to the bound charge likely drives the observed motion; however, a thorough understanding of the electrophoresis phenomenon is beyond the scope of this work. In the revised manuscript, we have clarified the definition of self-charge and made revisions as below.

Page 12, Line 321

(before) “The discovered topologies are self-charged, being electrically processible fundamental quasi-particles. Their production is kinetically controlled through distinct pathways. In the following, we discuss how these polar quasi-particles are developed.”

(after) “The discovered topologies are self-charged, being electrically processible fundamental quasi-particles. Their 'self-charged' nature arises from the topology, which requires a nonzero polarization divergence ($\nabla \cdot \mathbf{P} \neq 0$), leading to intrinsic bound charges (Figure 4). These solitons can be regarded as self-charged entities whose formation and integration are governed by the interplay between electrostatics and confinement. Below, we first examine the kinetic pathways that drive the emergence of these polar quasi-particles.”

3) Thermodynamic Stability of Hybrid States ($L_{\text{H}}/R_{\text{H}}$). The authors note that hybrid states appear under slow cooling, while monopoles appear under faster cooling. This suggests that the hybrid state might be the thermodynamic ground state under asymmetric confinement, or conversely, a kinetic trap dependent on the nucleation pathway. Could the authors clarify the

long-term stability of the hybrid states? If an L_H domain is maintained at a constant temperature for an extended period (e.g., 24 hours), does it relax into the monopole state (L_M)? This information is vital for the proposed applications in "large-scale engineering," which requires temporal stability

-> We thank the reviewer for this insightful question. The emergence of hybrid versus monopole states is primarily governed by the nucleation pathway. Slow cooling promotes nucleation at the LC–air interface, where boundary curvature stabilizes hybrid states; conversely, fast cooling triggers bulk nucleation, favoring monopole states.

Our numerical simulations (Supplementary Fig. 4) confirm that monopole and hybrid states correspond to distinct metastable minima in the free-energy landscape. Converting a hybrid soliton into a meron involves unwinding and reversing the polar field, which requires overcoming significant polarization-gradient and electrostatic energy barriers. These barriers far exceed thermal fluctuations, thereby preventing spontaneous transformation between the two states, even when the cooling rate is varied smoothly.

Our experimental results further show that the hybrid states are long-term stable at constant temperature. In particular, a hybrid (L_H) domain does not relax into the monopole state (L_M) over at least 24 h; more generally, we observe no detectable inter-state transformation even after several days (> 3 days). We have clarified this point in the revised manuscript.

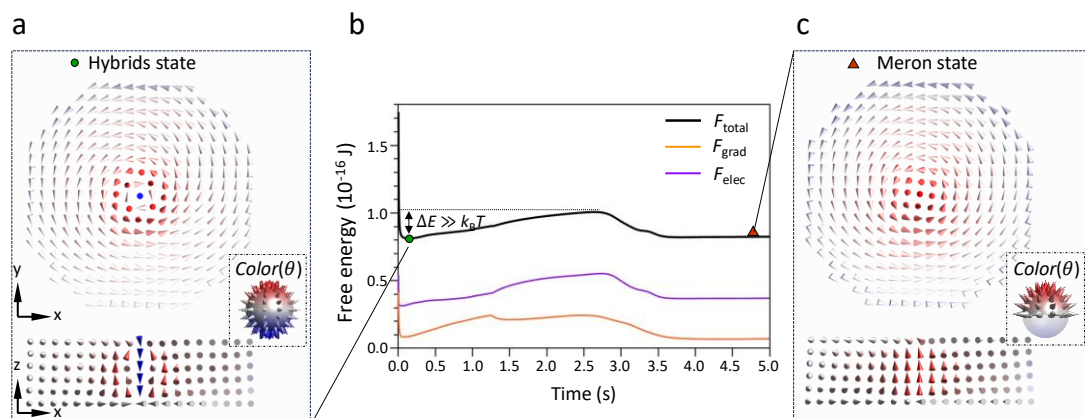
Page 5, Line 122

(sentence added) “The topological diversity stems from the cooling-rate-dependent nucleation pathway. The nucleation geometry of the N_F bubbles changes from cylindrical to hemispherical as the cooling rate decreases. While the cylindrical confinement with flat boundaries favors the axisymmetric formation of monopoles, the hemispherical confinement introduces geometric asymmetry that facilitates the stabilization of hybrid topological solitons (see **Discussion**). Once they are formed, both monopole and hybrid states remain stable, persisting for at least several days at room temperature without structural switching and degradation.”

Page 7, Line 183

(sentence added) “Notably, the skyrmion-toron core is energetically stable. Transformation between the hybrid topology and the twisted meron must overcome energy barriers caused by polarization gradients and electrostatic interactions (see Supplementary Fig. 4).”

Supplementary information, Supplementary Fig. 4
(Figure added)



Supplementary Fig. 4. Simulated transformation between hybrid and meron solitons. **a**, Reconstructed polarization field of the hybrid soliton obtained from numerical simulation. Polarization vectors are colored by elevation angle θ . **b**, Evolution of the free energy during the simulated transformation from the hybrid state to a meron state. The total free energy F_{total} (black), polarization gradient energy F_{grad} (orange), and electrostatic energy F_{elec} (purple) are shown. The transformation requires overcoming an energy barrier $\Delta E \gg k_B T$, indicating that the two configurations are separated by a substantial energetic barrier despite being topologically connected. **c**, Final polarization configuration corresponding to the meron state after relaxation. Please refer to the *supplementary Table I* for parameter settings for simulation. In all panels, cones represent the polarization vectors, with color indicating the elevation angle θ .

4) Robustness of the Air Interface. The asymmetric confinement relies on an open-air interface. While scientifically elegant, open interfaces are susceptible to environmental perturbations (humidity, dust, airflow, evaporation of volatile components). For the device applications suggested in the conclusion, how robust is this interface? The authors should briefly discuss whether this topological effect can be replicated using a solid "capping" material (a superstrate) that mimics the degenerate planar anchoring of air, which would allow for encapsulated, durable devices.

-> We agree that open-air interfaces may, in principle, be susceptible to environmental perturbations such as humidity or dust. In practice, we have placed a glass coverslip coated with a thin glycerol layer on the upper surface. During this process, both the meron and hybrid solitons remain stable and preserve their internal structures without noticeable changes. This not only eliminates environmental perturbations but also allows an electric field to be applied perpendicular to the LC cell, making it ideal for optoelectric and optoelectronic device applications. These observations indicate that the associated solitonic structures are remarkably robust. More generally, the key mechanism underlying the observed states is the asymmetric confinement geometry that allows polarization tilting near the upper boundary. In principle, similar conditions could be realized using a solid substrate that provides weak or degenerate planar anchoring, suggesting that the effect should be compatible with encapsulated device architectures. We have included the relevant discussion in the revised manuscript.

Page 5, Line 122

(sentence added) “The topological diversity stems from the cooling-rate-dependent nucleation pathway. The nucleation geometry of the N_F bubbles changes from cylindrical to hemispherical as the cooling rate decreases. While the cylindrical confinement with flat boundaries favors the axisymmetric formation of monopoles, the hemispherical confinement introduces geometric asymmetry that facilitates the stabilization of hybrid topological solitons (see **Discussion**). Once they are formed, both monopole and hybrid states remain stable, persisting for at least several days at room temperature without structural switching and degradation.”

Page 15, Line 366

(sentence added) “It is important to note that both monopole and hybrid states are (meta)stable states. The selection of the topological states, controlled by the cooling rate and the resulting confinement geometry, provides a mechanism of adjusting boundary or anchoring asymmetry to control topological characteristics. For instance, instead of using the air interface, we could employ a solid "capping" material, such as a coverslip or a glycerol layer, to achieve both meron and hybrid topologies. This finding highlights their resilience to environmental perturbations and their potential for manipulation under various electric field conditions.”

5) Energy Scales of Intertwining. The interaction between merons is described as a competition between elastic and electrostatic forces. It would be insightful if the authors could provide a rough comparison of the order of magnitude for the elastic interaction energy vs. the electrostatic interaction energy at the separation distances observed in Figures 2 and 3. This would help readers understand which force is the dominant driver of the self-assembly.

-> We thank the reviewer for this insightful suggestion. We have performed additional numerical calculations for two N_F droplets ($R = 3 \mu\text{m}$) as a function of their separation. Our results show that the electrostatic energy consistently outweighs the elastic energy. As the separation decreases from $10 \mu\text{m}$ to contact, the elastic energy remains nearly constant, whereas the electrostatic energy density decreases by $\sim 10^{-16}\text{J}$ for two merons with opposite screw senses and increases by a similar magnitude for a pair with the same screw sense. This sign-dependent behavior confirms that polarization-induced bound charges dominate the interaction at these scales.

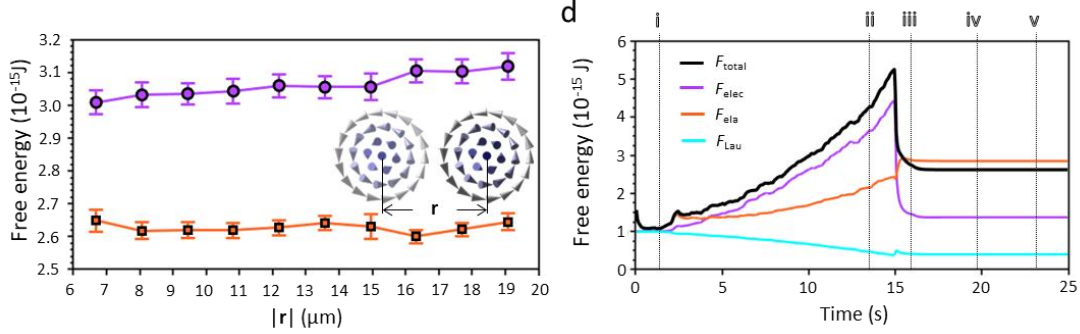
During intertwining, the observed structures are accompanied by a decrease in electrostatic energy that is approximately one order of magnitude greater than the concurrent increase in elastic energy. We discussed this in detail in Supplementary Discussions 1 and 2 and included the corresponding results in Supplementary Figs. 5 and 6.

Page 8, Line 209

(sentence added) “Calculations of the energy landscape for meron pairs show that the elastic contribution is nearly independent of separation. In contrast, the electrostatic energy changes by $\sim 10^{-16} \text{ J}$ as the merons approach contact, decreasing for the opposite-screw-sense pairs and increasing for the same-screw-sense pairs (Supplementary Figs. 5 and 6). These results indicate that electrostatic coupling provides a robust long-range route to control polar soliton

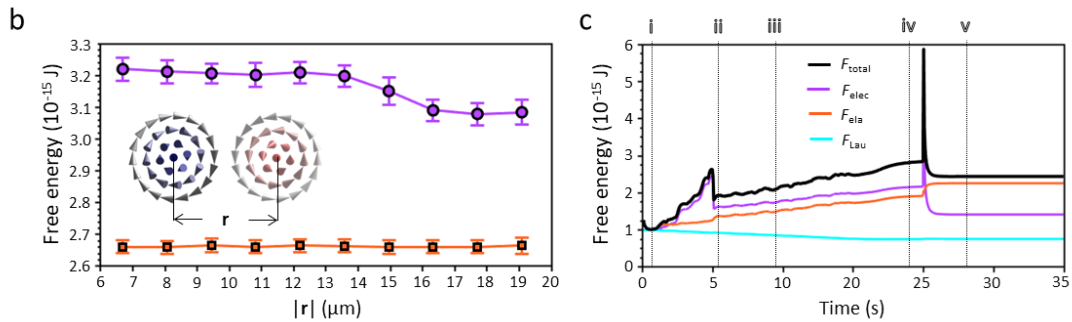
dynamics, distinguishing N_F systems from conventional elastic-dominated nematics.”

Supplementary information, Supplementary Fig. 5
(Figure added)



Supplementary Fig. 5. c, Separation-dependent free-energy landscape for a pair of merons with the same screw senses prior to contact. The electrostatic (purple circles) and elastic (orange squares) energy components are plotted as a function of the center-to-center distance $|\mathbf{r}|$, with the soliton radius fixed at $R = 3 \mu\text{m}$. Each data point represents a time-averaged value over a finite simulation window, and the error bars indicate the corresponding variance arising from the continuous motion of the solitons. **d**, Time evolution of various free energies during the dynamic interaction process shown in (b). In this case, the merons simultaneously approach each other while their radii evolve in time, mimicking experimental conditions. The dashed vertical lines (i–v) mark the characteristic stages of the interaction sequence corresponding to the snapshots in (b).

Supplementary information, Supplementary Fig. 6
(Figure added)



Supplementary Fig. 6. b, Separation-dependent free-energy landscape for a pair of merons with the opposite screw senses prior to contact. The electrostatic (purple circles) and elastic (orange squares) energy components are plotted as a function of the center-to-center distance $|\mathbf{r}|$, with the soliton radius fixed at $R = 3 \mu\text{m}$. Each data point represents a time-averaged value over a finite simulation window, and the error bars indicate the corresponding variance arising from the continuous motion of the solitons. **d**, Time evolution of various free energies during the dynamic interaction process shown in (a). In this case, the merons simultaneously approach each other while their radii evolve in time, mimicking experimental conditions. The dashed vertical lines (i–v) mark the characteristic stages of the interaction sequence corresponding to the snapshots in (a).

1) Minor Comments.

a) Terminology (Chirality vs. Screw Sense): The distinction between molecular chirality (dopant handedness) and the topological screw sense of the meron is generally clear, but can be confusing in certain passages. I suggest adding a small schematic or table in the Extended Data clearly defining the sign conventions used for Q , screw sense, and molecular chirality to aid the general reader.

-> We thank the reviewer for this suggestion. Molecular chirality determines the twisting sense of the polar field, i.e., the handedness (left- or right-handed) of local helix. In contrast, the screw sense describes the clockwise or counterclockwise rotation of the director within the xy -plane. For achiral N_F droplets, these two screw-sense configurations are energetically equivalent and can emerge with equal probability during nucleation. To enhance clarity, we have added *Supplementary Table II*, which outlines the terminology and conventions used throughout the manuscript, making them easier for a broader audience to understand.

Supplementary Information, *Supplementary Table II*
(table added)

Supplementary Table II | Summary of Terminology and Topological Concepts

Term	Definition	Remarks
<i>Twisting Sense</i>	The handedness of how the polarization $\mathbf{P}$ rotates in space along a given axis (Supplementary Fig. 1).	Determined by the type of chiral agent: R811 for right-handed, S811 for left-handed.
<i>Screw Sense</i>	The rotation direction of the polarization $\mathbf{P}$ in the xy -plane (Supplementary Fig. 1).	Classified into clockwise and anti-clockwise rotations.
<i>Soliton</i>	A localized, topologically protected deformation of the polarization/director field that maintains its structure due to energetic and/or topological constraints.	Including merons, skyrmions, toron and their hybrid structures
<i>Meron</i>	A half-skyrmion structure typically features a core with vertical polarization and a surrounding in-plane rotation.	Its polarization field covers half of the S^2 manifold sphere.
<i>Skyrmion</i>	A topological soliton in which the polarization field provides a full mapping from real space to the S^2 manifold sphere.	Can be viewed as the tight union of two Merons, which is continuous and singularity-free in space.
<i>Toron</i>	A localized 3D soliton consisting of a twisted skyrmion-like core terminated by point defects (or defect loops).	Typically confined in finite geometry and appearing as a "spinning wheel" texture.
<i>Disclination</i>	A topological line defect in the polarization/director field where the orientational order becomes singular or ill-defined.	Typically characterized by a quantized winding of the field around the defect core.
<i>Skyrmion Number Q</i>	A topological invariant quantifying	Determines topological

	how many times the polarization field wraps the unit sphere. $Q = \frac{1}{4\pi} \int \mathbf{n} \cdot (\partial_x \mathbf{n} \times \partial_y \mathbf{n}) dx dy$	class; meron: $\pm 1/2$, skyrmion: ± 1. Conserved under continuous deformation.
<i>Preimage on S^2</i>	The set of spatial points that map to a given orientation on the unit sphere (order-parameter space).	Useful for visualizing topology.

b) Figure 1f (FCPM): The reconstruction of the hybrid state is excellent. However, please ensure the caption or labels clearly indicate the physical scale of the "defect-core-proxy" relative to the optical resolution.

-> We thank you for this helpful comment. In the FCPM reconstruction shown in Fig. 1f, the characteristic radius of the defect-core-proxy is on the order of 1~2 μm . This length scale is significantly larger than the optical resolution of our confocal setup (submicron lateral resolution for the NA = 1.4 objective), indicating that the observed structure is not limited by instrumental resolution. To avoid possible confusion, we have revised the figure caption to clearly indicate the physical scale of the defect-core-proxy and its relation to the optical resolution.

Page 24, Line 583, Caption of Fig. 1

(sentence added) "The inset in f (iii) shows magnified bubble core, whose characteristic radius r_d , is ~1–2 μm , well above the submicron lateral resolution of the FCPM (NA = 1.4)."

c) References: The citation of flexoelectric contributions is appropriate, but the authors might consider citing recent works on ferroelectric nematic drops or tactoids where surface charges also play a role, to better contextualize their work within the broader field.

-> We thank you for this helpful suggestion. We agree that recent studies on ferroelectric nematic droplets and tactoids, where surface-bound charges play an important role, provide valuable context for understanding electrostatic effects in confined polar nematic systems. Following the reviewer's recommendation, we have added the following two appropriate references in the revised manuscript:

1. Xu Z, *et al.* Interface-Engineered Polar Topological Domains in Ferroelectric Nematic Liquid Crystals. *Advanced Materials* **37**, 2501395 (2025).
2. Máthé MT, Buka Á, Jákli A, Salamon P. Ferroelectric nematic liquid crystal thermomotor. *Physical Review E* **105**, L052701 (2022).

Page 3 Line 70

(sentence added) "Recent studies have revealed diverse polar structures in N_F systems—such as interface-engineered domains and active droplet-like inclusions—where surface charges profoundly influence interfacial structures and dynamics^{32, 33}."