Self-assembly and electrostriction of arrays and chains of hopfion particles in chiral liquid crystals

Paul J. Ackerman\textsuperscript{1,2}, Jao van de Lagemaat\textsuperscript{1,3,4} & Ivan I. Smalyukh\textsuperscript{1,2,4,5}

Some of the most exotic condensed matter phases, such as twist grain boundary and blue phases in liquid crystals and Abrikosov phases in superconductors, contain arrays of topological defects in their ground state. Comprised of a triangular lattice of double-twist tubes of magnetization, the so-called ‘A-phase’ in chiral magnets is an example of a thermodynamically stable phase with topologically nontrivial solitonic field configurations referred to as two-dimensional skyrmions, or baby-skyrmions. Here we report that three-dimensional skyrmions in the form of double-twist tori called ‘hopfions’, or ‘torons’ when accompanied by additional self-compensating defects, self-assemble into periodic arrays and linear chains that exhibit electrostriction. In confined chiral nematic liquid crystals, this self-assembly is similar to that of liquid crystal colloids and originates from long-range elastic interactions between particle-like skyrmionic torus knots of molecular alignment field, which can be tuned from isotropic repulsive to weakly or highly anisotropic attractive by low-voltage electric fields.
Topological defects\textsuperscript{1–8} are observed during symmetry-breaking condensed-matter phase transitions\textsuperscript{1} and as a result of flow\textsuperscript{9}, application of fields\textsuperscript{1}, temperature changes\textsuperscript{1–3} and patterning of light\textsuperscript{10}, but typically annihilate and cannot be controlled after completion of these transient processes\textsuperscript{1–39}. Rather unexpectedly, it was found recently that self-compensating topological defect pairs in active matter not only annihilate but also can be spontaneously generated, exhibiting dynamics similar to that of active particles\textsuperscript{11,12}. Topological defects can form stable periodic arrays when mediating formation of thermodynamically stable vortex phases, such as cholesteric blue phases and twist grain boundary phases in liquid crystals (LCs), the A-phase of chiral magnets\textsuperscript{1} and Abrikosov phases of superconductors\textsuperscript{1–3}. Control and generation of defects in LCs by colloids and vortex laser beams allowed for obtaining individual stable line and point defects as well as twisted solitons such as torons and localized structures resembling the mathematical Hopf fibration\textsuperscript{5–8,13–19} and even Turing patterns and linked vortices in classical liquids\textsuperscript{9,21}, spin solitons resemble skyrmionic field configurations associated with exhibiting dynamics similar to that of active particles\textsuperscript{11,12}.

Nematic LCs are three-dimensional (3D) fluids comprised of anisotropic molecules with no positional order, but with a ground state having a spatially uniform molecular long-axis orientation called the ‘director’\textsuperscript{1}. In a chiral nematic LC (CNLC), the ground-state director is twisting at a constant rate along a ‘helical axis’, with the distance over which \( \mathbf{n}(r) \) rotates by a \( 2\pi \)-dubbed ‘pitch’ \( \rho \). The CNLC ground-state twist can be suppressed by applying fields or by specially treated surfaces of confining substrates that couple to \( \mathbf{n}(r) \), rendering it uniform and thus frustrated with respect to the twist preference. This frustration is often relieved locally through spontaneous or laser-guided formation of various cholesteric translationally invariant linear or axially symmetric solitonic structures that locally embed director twist into the unwound confined CNLCs\textsuperscript{5–7,13–15,20}. These 2D and 3D twisted solitons resemble skyrmionic field configurations associated with Turing patterns and linked vortices in classical liquids\textsuperscript{9,21}, spin textures in quantum Hall effect systems\textsuperscript{22}, double-twisted building blocks of LC blue phases\textsuperscript{23,24} and ground states in chiral magnets\textsuperscript{4,25}.

The so-called ‘double-twist cylinder’ or ‘double-twist tube’, in which \( \mathbf{n}(r) \) is parallel to the cylinder axis at its centre and exhibits a 2D radial twist to form a barber pole-like pattern on the cylinder surface (Fig. 1a), is the basis of a 2D skyrmion that can be obtained as an isolated topological object in a confinement-unwound CNLC\textsuperscript{26,27}, and is also a building block of the cholesteric blue phases with 3D crystalline arrays of such tubes\textsuperscript{1}. The director \( \mathbf{n}(r) \) coils around the tube with periodicity defined by the distance along the tube axis over which \( \mathbf{n}(r) \) winds around it once (Fig. 1a). When a fragment of such a tube of length corresponding to a single winding of \( \mathbf{n}(r) \) is looped on itself, the director field lines of the ensuing 3D skyrmion in the form of a double-twist torus become closed circles linked with each other (Fig. 1a, top right)\textsuperscript{28}, with single windings around the circle in the interior of the torus \( (P = 1) \) and also around its axis of rotational symmetry \( (Q = 1) \) (Fig. 1b)\textsuperscript{28,29}. The linking of circles of \( \mathbf{n}(r) \)-loops in this 3D skyrmionic structure resembles that of ‘fibres’ in the famous mathematical Hopf fibration\textsuperscript{5,30,31}.

Similar construction through looping different 2D skyrmions results in double-twist tori with multiple windings around the two axes and the mutually interlinked \((P,Q)\) torus knots in \( \mathbf{n}(r) \), resembling a more general class of mathematical Seifert fibrations\textsuperscript{28–31}, which include the Hopf fibration in a limited case of \( P = Q = 1 \) (ref. 5). An example of a 3D skyrmion formed by a double-twist torus with linked \((P,Q) = (2,3)\) trefoil knots is shown in the bottom-right of Fig. 1a, with two representative mutually linked knots of \( \mathbf{n}(r) \)-loops shown in red and green colours. The 3D skyrmions with such \((P,Q)\) knots are characterized by a non-zero Hopf charge \( C_H = PQ \) and are commonly called ‘hopfions’\textsuperscript{29}.

In this work, we demonstrate that these topologically protected skyrmionic particles self-assemble into various crystalline arrays and linear chains. The facile frequency-dependent electric response of CNLCs enables large-quantity generation, guided self-assembly and electrostriction of dense and sparse periodic arrays and linear chains of skyrmions, which are often accompanied by various other singular and nonsingular topological defects. Being topologically protected field configurations, with mutually linked different torus knots of looped molecular alignment field lines, these particle-like solitons self-assemble similar to LC colloids and electrostatic dipoles\textsuperscript{8}. Interactions between the skyrmions are electrically switched between repulsive and attractive, mediating self-assembly of crystalline arrays and chains with tunable inter-particle spacing. The exquisite control of topologically nontrivial structures is achieved at voltages of the order of 1 V, potentially enabling mesostructured soft-matter composites with tunable optical properties and a host of new technological applications.

**Results**

**Facile generation and structure of skyrmionic particles.** Skyrmions with different torus knots of \( \mathbf{n}(r) \) can be generated in confinement-unwound LC cells on an individual basis by focused beams or colloidal inclusions\textsuperscript{5–7}, as in the example of a laser-generated torus shown in Fig. 1e. In addition to linked unknots \((P,Q) = (1,1)\), trefoil and pentafold torus knots of \( \mathbf{n}(r) \) also can be observed within the double-twist-torus part of the toron structures. This structural behaviour is highly dependent on the details of toron generation, sample thickness over pitch ratio \( dp / \rho \), lateral confinement and other factors that have been detailed previously\textsuperscript{5–7,13–15} as well as will be explored further elsewhere, but, importantly for the present work, all of these skyrmionic configurations exhibit particle-like behaviour. Such skyrmionic configurations are stable long after generation, but can be electrically controlled by applying voltages \( U = (1–4) \) V at 1 kHz (Fig. 1e–h). Consistent with the fact that the used LCs have negative dielectric anisotropy \( \Delta \varepsilon \), the difference between dielectric constants measured for electric field along and perpendicular to \( \mathbf{n} \), the toron structure first laterally expands with increasing \( U \) (Fig. 1e,f), which is expected as its interior has in-plane...
Figure 1 | Structure and topology of 3D skyrmionic particles. (a) Double-twist cylinder of \( n(r) \) and its use in the construction of tori with linked \((P,Q)=(1,1)\) unknots (top right) and various torus knots, such as the \((P,Q)=(3,2)\) trefoil knot (bottom right); the green and red \( n(r) \) lines are used for eye guiding to depict linked unknots and knots of \( n(r) \) loops. (b) A double-twist torus formed by a double-twist cylinder looped on itself is a 3D skyrmion-dubbed ‘hopfion’. (c) When accompanied by two self-compensating hyperbolic point defects of opposite hedgehog charges \( \pm 1 \) (shown in red and green colours), the hopfion forms a toron. (d) \( n(r) \) structure of the toron. (e–h) Polarizing optical micrographs of a toron between two crossed polarizers (shown by white double arrows) at \( U \) marked on the images. The toron is surrounded by a uniform unwound vertical \( n(r) \) at low \( U \), but becomes embedded in the TIC above \( U \approx 2.1 \) V threshold voltage, which is slightly dependent on \( d/p \). The insets in \( g \) and \( h \) were obtained for illumination settings chosen to highlight the dipolar structure of \( c(x,y) \) and the umbilic defect. (i) 3PEF-PM image of a polymerized CNLC sample revealing \( c(x,y) \) of the toron-umbilic pair. The far-field orientations of the midplane \( n(r) \) and \( c(x,y) \) is denoted by a blue double arrow. (j) Schematic of \( n(r) \) in the vertical cross-section of an axially symmetric toron, with the plane of images (k) shown in blue colour. (k) Experimentally reconstructed colour-coded pattern of azimuthal orientation of \( n(r) \) in the XY cross-section in the midplane of a toron at \( U = 0 \) co-located with the numerically simulated \( n(r) \) shown using cylinders with red-blue-coloured ends; the colour scheme is shown in the inset. (l) Computer-simulated \( n(r) \) shown using cylinders and its azimuthal orientation depicted using colours for a toron embedded in TIC in the cell midplane. The white and yellow circles with ‘±’ depict signs of winding numbers of the ±1 defects in \( c(x,y) \). (m) The corresponding qualitative schematic of the \( c(x,y) \). (n,o) \( n(r) \) of the topological particles shown in the vertical cross-sections marked on micrographs (e,h) at corresponding (n) \( U = 0 \) and (o) \( U = 3.1 \) V. Green- and red-filled circles depict hyperbolic point defects with hedgehog charges ±1. The two ±1 singular points in \( c(x,y) \) shown in (n) are nonsingular in \( n(r) \) and correspond to the parts of the cell midplane with vertical \( n(r) \) marked by white and yellow circles.
orientation of \( \mathbf{n} \) favoured by the free-energy term describing its coupling to the field. However, as \( U \) increases further, prompting a transition of the initially vertical director \( \mathbf{n} \) to in-plane \( \mathbf{n}(\mathbf{r}) \) with twist across the cell thickness accompanied by bend/splay distortions\(^{20}\), the effective lateral size of the toron starts to shrink and the birefringent dipolar-like texture around it becomes visible between crossed polarizers (Fig. 1f–h). This is consistent with depth-resolved three-photon excitation fluorescence polarimetry (3PEF-PM)\(^{38,39}\) (Fig. 1i,k) and numerical modelling of such structures at various \( U \) (Fig. 1k–o).

To understand the physical underpinnings, it is instructive to consider the midplane of a cell passing through the equatorial plane of a toron (marked by a blue disc in the simplified structural model shown in Fig. 1j). Figure 1k depicts an experimentally reconstructed pattern of azimuthal orientation of \( \mathbf{n}(\mathbf{r}) \) in this plane at \( U=0 \), overlaid with the corresponding numerically simulated \( \mathbf{n}(\mathbf{r}) \), which we show using cylinders with blue/red ends. The \( \mathbf{n}(\mathbf{r}) \)-tilt direction at large \( U \), which can be described by a 2D vector field \( c(x,y) \) describing the projection of the \( \mathbf{n}(\mathbf{r}) \) to the cell midplane and is concentric at the periphery of a toron, must match the uniform far-field \( c(x,y) \) (Fig. 1l,m). This is achieved by introducing a defect in \( c(x,y) \) with a winding number \(-1\), known as ‘umbilic’\(^{1}\), that compensates for the \(+1\) defect in \( c(x,y) \) in the toron centre. The \( c(x,y) \)-periodic hexagonal arrangements of \( \mathbf{n}(\mathbf{r}) \) is shown in Fig. 1m, with the dipole marked by a green arrow.

In CNLCs doped with ionic surfactants\(^{40}\), large quantities of torons can be generated through the relaxation of the frustrated-state-confined CNLC from hydrodynamic instability (Supplementary Fig. 1). Upon turning off the field, the CNLC relaxes from the disordered hydrodynamic state to a ground-state configuration with varied density of torons that self-organize into periodic hexagonal arrays (Fig. 2a), consistent with our numerical findings that arrays of torons correspond to the ground state of confined CNLCs at cell thickness \( d \) to pitch ratio \( d/p \approx 1 \) (ref. 5).

The hydrodynamic instability plays the role of generating torons in the entire sample similar to how focused laser beams do this locally\(^{5-7}\), that is, it allows the system to relieve frustration imposed by boundary conditions via formation of toron arrays (Fig. 2a,b). When generated at varying densities, toron particles form Wigner-crystal-like hexagonal lattices due to isotropic repulsive interactions (Fig. 2a,b; Supplementary Fig. 2). The 2D crystallites with hexagonal arrangements of torons are hundreds of micrometres or even millimetres in size, 100–1,000 times larger than individual torons (Fig. 2a). Uncontrolled orientation of crystallographic axes of the crystallites upon their formation through relaxation from the hydrodynamic instability causes grain boundaries (Fig. 2a).

**Electric switching and self-assembly.** Application of electric fields above a threshold voltage of \( U \approx 2.1 \) V at 1 kHz leads to a transition of the unwound homeotropic \( \mathbf{n}(\mathbf{r}) \) to the so-called ‘translationally invariant configuration’ (TIC) of \( \mathbf{n}(\mathbf{r}) \)\(^{20}\), with a voltage-dependent twist and bend-splay distortions across the cell thickness (Fig. 2e,f). With increasing \( U \), similar to individual topological particles (Fig. 1e–h), lateral dimensions of torons within the arrays first expand and then shrink while inducing the umbilical defects in \( c(x,y) \). This leads to the reversal of interactions from repulsive to attractive, self-assembly of torons into hexagonal lattices of voltage-dependent periodicity, and finally to the formation of chains of toron–umbilical dipoles (Fig. 2c,d; Supplementary Fig. 2), as we summarize in Fig. 3. The dipolar chains (Figs 2d and 3c) resemble the ones formed by electrostatic dipoles and elastic dipoles in nematic LC colloids\(^{15}\), except that our topological defect dipoles always have the same orientation orthogonal to \( \mathbf{c} \) because of the polar nature of \( c(x,y) \) (Fig. 2d). Similar to the case of nematic colloids, both repulsive and attractive interactions between the topological particles occur to minimize the free energy as the corresponding director configurations transform in response to fields. Finally, at \( U > 5 \) V, dipolar defect chains become fully unstable due to the strong coupling between the electric field and \( \mathbf{n}(\mathbf{r}) \) in the LC with negative dielectric anisotropy, which discontinuously destroys the topological particles. Annihilation of torons and umbilics, which includes mutual destruction of the hopfion-like double-twist torus structure and umbilic, and annihilation of oppositely charged hyperbolic point defects, eventually lead to a uniform TIC. Within the stability range of the skyrmionic particles, the inter-particle spacing changes markedly by a factor of 2–3 with varying \( U \) (Fig. 3a), yielding a giant electrostriction of 2D hexagonal and linear chain assemblies that shrink by over 50% at 0.5 \( \mu \)m\(^{-1}\). The voltage-tunable self-assembly of particle-like ‘mobile’ torons is markedly different from that of torons pinned to the confining surfaces (Fig. 2g) optically generated at high laser powers above 70 mW. In the latter case, upon increasing \( U \) above the threshold for inducing TIC, the torons remain pinned to the spatial locations of their generation despite the formation of toron–umbilical dipoles (Fig. 2g), until disappearing at \( \sim 5 \) V.

**Dynamics and characterization of pair interactions.** Skyrmionic topological particles and their self-assemblies undergo Brownian motion (Fig. 4), which we characterize using videomicroscopy by probing their displacements during time intervals \( \tau = 200 \) ms over 18.3 min. This highly overdamped motion (Reynolds number \( \ll 1 \)) is direction independent when a toron is embedded in the unwound CNLC, but becomes slightly anisotropic in TIC at \( U = (2.1-5) \) V. The direction-averaged half-width \( \Delta \) of histograms of displacements described by Gaussian distributions (Fig. 4d) yield voltage-dependent diffusivity\(^{1} \) of the topological particles within \( D = A^2/\tau = (1.1-1.6) \times 10^{-3} \text{ m}^2 \text{ s}^{-1} \), which was found to be independent of frequency within the 1–10 kHz used to electrically guide self-assembly. Using this experimental diffusion constant, we find the effective viscous drag coefficients from the Einstein relation \( \zeta = k_BT/DF = (2.6-3.7) \times 10^{-6} \text{ N s m}^{-1} \), where \( k_B = 1.38 \times 10^{-23} \text{ K}^{-1} \) is Boltzmann’s constant and \( T \) is temperature\(^{12}\). The forces arising from minimization of free energy pull the toron–umbilical dipoles to an equilibrium distance (Fig. 4a–d) and lead to \( U \)-dependent histograms of their relative centre-to-centre separations \( P(D_{\text{r}}) \) away from the equilibrium. The relative pair-interaction free energy \( F - F_0 \) near the equilibrium state with free energy \( F_0 \) (Fig. 4e) is then computed from this experimental histogram by inverting the Boltzmann relation, \( P(D_{\text{r}}) \propto \exp (F - F_0)/k_BT \).

As the equilibrium inter-particle separation decreases with increasing \( U \) upon formation of chains (Fig. 3a), the spring constant \( k \) describing Hookean-like behaviour near the equilibrium \( F - F_0 = kD_{\text{r}}^2 \) increases with voltage within \( k = 0.08-0.83 \text{ pN \mu m}^{-1} \) (Fig. 4e).

To uncover physical underpinnings behind self-assembly of skyrmionic particles, we use holographic laser tweezers\(^{28}\) capable of trapping and manipulating them at different \( U \). Topological particles can be placed at desired initial positions corresponding to different inter-particle-separation vector orientations with respect to the far-field \( \mathbf{c} \) (Fig. 5). Once released from the traps, topological particles repel or attract, depending on the initial conditions and \( U \), allowing us to probe both pair interactions (Fig. 5) and many-body interactions (Supplementary Fig. 3), which we study using videomicroscopy. As voltage increases, the interactions change from isotropic repulsive (Fig. 5a,b) to isotropic and then weakly anisotropic attractive (Fig. 5c,d), and...
finally to strongly anisotropic dipolar-like (Fig. 5e,f), with the parallel toron–umbilical dipoles attracting when their centre-to-centre separation vector is orthogonal to the far-field \( \mathbf{c} \) (Fig. 5f; Supplementary Movie 1) and repelling when it is parallel to the far-field \( \mathbf{c} \) (Fig. 5f; Supplementary Movie 2). Remarkably, this marked change occurs as \( U \) is varied within a narrow range of \((0–3.5) \) V, corresponding to the self-assembly of larger- and smaller-period hexagonal lattices and dipolar chains of toron–umbilical dipoles aligned orthogonal to the far-field \( \mathbf{c} \) (Figs 2 and 3). From the topological particle position versus time data, we calculate their velocities that range within \( \frac{dr}{dt} = (0–3) \) \( \mu \)m s\(^{-1}\). Neglecting inertia effects, we estimate the inter-particle elasticity-mediated forces from their balance with the effective viscous drag force \( -\zeta \frac{dr}{dt} = (0–10) \) pN, consistent with their origin as the values of elastic constants are of the order of \( 10 \) pN (Supplementary Table 1).

**Comparison of experiments and numerical modelling.** Our experimental findings are fully reproduced by numerical
modelling based on the director relaxation method\textsuperscript{5,29} of minimizing the CNLC free energy at strong perpendicular boundary conditions on confining plates\textsuperscript{1}. Since the half-integer defect lines in $n(r)$ do not occur in the structures studied in this work, this approach is ideally suitable for 3D modelling of the skyrmionic particles as it accounts for all elastic constants while also allowing for simulations of relatively large sample sizes ranging from micrometres to tens of micrometres (computer simulations accounting for elastic anisotropy based on the Q-tensor approach for such large samples would be rather slow)\textsuperscript{5,29}. Taking experimental material and geometric parameters, we obtain the equilibrium 3D $n(r)$ configurations of torons and toron–umbilic dipoles embedded in the TIC at various voltages (Figs 1k,l,n,o and 2e,f; Supplementary Fig. 4) closely matching their experimental counterparts. Modelling of elastic pair interactions of these particles also closely reproduces experimental results. For example, Fig. 4b,c shows the experimentally reconstructed (from 3PEF-PM images) and corresponding computer-simulated colour-coded $n(r)$ configurations in the equatorial plane of the toron–umbilic dipole passing through the cell midplane; the simulated $n(r)$ is additionally represented using cylinders with coloured ends overlaid on the top of the colour-coded texture (Fig. 4c).

This agreement between modelling and experiments supports our explanation of self-assembly of skyrmionic particles through the electrostatic and nematic colloidal analogies\textsuperscript{8}.

Discussion

On the basis of both numerical modelling and experiments, tunable elastic interactions between skyrmionic topological particles can be qualitatively explained as follows. At no applied fields, the $n(r)$ structure of a toron embedded into a uniform far-field director (Fig. 1d) has quadrupolar symmetry. Therefore, the torons interact repulsively as elastic quadrupoles (Fig. 4a,b), although these interactions in lateral directions are screened by confinement of the CNLC into a cell with strong boundary conditions. As the lateral spatial extent of $n(r)$ distortions smoothly increases with $U$ at voltages below the realignment threshold away from torons, the strength of their quadrupole moments and lateral distance range of repulsive interactions increase too. At the realignment threshold, a symmetry-breaking structural transition occurs, transforming the initial quadrupolar toron into an elastic toron–umbilical dipole with an in-plane orientation of the dipole moment orthogonal to the far-field c. As the strength of the elastic dipole moment gradually increases with
in angles between the inter-particle separation vectors and the far-field.

The singular point defects opposite hedgehog charges, which could be replaced by loops of assembled into various voltage-tunable periodic arrays and these defects of different topological classes while being nonsingular in.

The different classes of topological defects co-existing with each other.

The quadrupolar repulsive at low $U$, the elastic interactions gradually transform from isotropic quadrupolar repulsive at low $U$ to strongly anisotropic dipolar-like attractive at relatively high $U$, giving the origin to different voltage-dependent self-assemblies. Beyond this qualitative picture, quantitative understanding of interactions between skyrmionic particles requires accounting for detailed contributions of twist and dielectric terms associated with the complex 3D structure of the toron–umbilical field configurations at different fields, as well as the short-range interaction effects that cannot be described through the electrostatic analogy, further contributing to the richness and complexity of interactions in cholesteric systems that recently attracted a great deal of attention.

Another interesting feature of our system is the diversity of different classes of topological defects co-existing with each other. The $\pi_3 \left( RP^2 \right)$ hopfions in the central part of the toron are accompanied by self-compensating $\pi_1 \left( RP^2 \right)$ point defects of opposite hedgehog charges, which could be replaced by loops of $\pi_1 \left( RP^2 \right)$ line defects when generating torons by optical vortices, and also by nonsingular in $n(r)$ umbilical defects, which are singular point defects $\pi_1 \left( S^1 \right)$ in the 2D $c(x,y)$ field. The stability of these defects of different topological classes while being assembled into various voltage-tunable periodic arrays and chains may allow for using our system in probing details of their structure and interactions, which may provide new insights into the topological nature and properties of similar skyrmionic field configurations predicted to exist in other physical systems.

From the standpoint of view of practical applications, because of the contrast of the effective refractive index between the twisted and non-twisted regions of periodic arrays, the self-assembled structures of skyrmionic particles can be readily used as electrically reconfigurable diffraction gratings (Supplementary Fig. 5) and optical vortex generators. Giant electrostriction, which we find to be even stronger than that observed in LC colloids, light sensitivity, which could be further enhanced by doped LC with dyes and using other types of generating light and entrapment of metal and semiconductor nanoparticles within these topological particles that we demonstrated recently can be used in combination to form composites with new tunable properties emerging from controlling mesoscopic order of the nanoparticles.

To conclude, we have described facile generation and voltage-tunable self-assembly of 3D skyrmions, along with different types of singular and nonsingular defects, into periodic arrays and linear chains emerging from reconfigurable elasticity-mediated

$\text{Figure 5 | Pair interactions of topological particles.}$ (a) Colour-coded centre-to-centre separation vs time trajectories for two particles released with initial separation vectors at 0, 45 and 90 degrees with respect to the far-field c demonstrating isotropic inter-particle repulsion at $U=0$. (b) The corresponding colour-coded-time trajectories of one toron of the pair with respect to the position of the second. White double arrows show orientations of crossed polarizers. (c-f) Centre-to-centre separation and trajectories similar to the ones shown in a and b, but at (c,d) $U=2.7 \text{ V}$ and (e,f) $U=3.1 \text{ V}$; the initial angles between the inter-particle separation vectors and the far-field c are 0, 22, 45 and 90 degrees, as marked next to the trajectories. Red arrows in d and f denote the far-field c and the green arrows denote the orientation of the toron-umbilical dipoles.
interactions. Furthermore, the inter-skyrmion separation in the self-organized structures was tuned by varying applied voltages up to 5 V, giving rise to strong electrostriction. This behaviour bridges markedly different forms of observation of condensed matter defects, ranging from active LCs to thermodynamically stable phases with periodic vortex lattices. The exquisite control over matter defects, ranging from active LCs to thermodynamically parallel and perpendicular to electric fields. With tangential anchoring.

References

1. Chalik, P. M. & Lubensky, T. C. Principles of Condensed Matter Physics (Cambridge Univ. Press, 2000).

2. Alexander, G. P., Chen, B. G., Matsutomo, E. A. & Kamien, R. D. Colloquium: disclination loops, point defects, and all that in nematic liquid crystals. Rev. Mod. Phys. 84, 497–514 (2012).

3. Mosseri, R. Geometrical frustration and defects in condensed matter systems. C. R. Chimie 11, 192–197 (2008).

4. Muhlbauer, S. et al. Skyrmion lattice in a chiral magnet. Science 323, 915–919 (2009).

5. Smalyukh, I. I., Lanskaya, C., Clark, N. & Trivedi, R. Three-dimensional structure and tunability of skyrmions in magnetic toroidal nanowires. Nat. Phys. 11, 139–145 (2015).

6. Chen, R. G., Ackerman, P. J., Alexander, G. P., Kamien, R. D. & Smalyukh, I. J. Generating the Hopf fibration experimentally in nematic liquid crystals. Phys. Rev. Lett. 110, 237801 (2013).

7. Pandey, M. B. et al. Self-assembly of skyrmion-dressed chiral nematic colloids with tangential anchoring. Phys. Rev. E 86, 060502(R) (2013).

8. Poulin, P., Stark, H., Lubensky, T. C. & Weitz, D. A. Novel colloidal liquid crystal toroids with tunable skyrmion states in liquid crystals. Science 275, 1770–1773 (1997).

9. Kleckner, D. M. & Irvine, W. T. Creation and dynamics of knotted vortices. Nat. Phys. 9, 253–258 (2013).

10. Denis, M. R., King, R. P., Jack, B., O’Holleran, K. & Padgett, M. J. Isolated skyrmion states in liquid crystals decorated by plasmonic nanoparticles. Science 338, 431–434 (2012).

11. Giomi, L., Bowick, M. J., Ma, X. & Marchetti, M. C. Defect annihilation and proliferation in active nematics. Phys. Rev. E 110, 228101 (2013).

12. Ackerman, P. J., Qi, Z. & Smalyukh, I. I. Optical generation of crystalline, quasicrystalline, and arbitrary arrays of torons in confined cholesteric liquid crystals for patterning of optical vortices in laser beams. Phys. Rev. E 86, 021703 (2012).

13. Ackerman, P. J. et al. Laser-directed hierarchical assembly of liquid crystal defects and control of optical phase singularities. Sci. Rep. 2, 414 (2012).

14. Frauenheim, T., Chen, D. T. N., DeCamp, S. J., Heymann, M. & Dogic, Z. A semi-analytical approach for self-assembling photonic crystals from skyrmions. Nat. Commun. 4, 2821 (2013).

15. Leonov, A. O., Dragunov, I. E., Röder, U. K., Bogdanov, A. N. & Pfleiderer, C. Spontaneous skyrmion ground states in magnetic nanowires. Nature 442, 79–83 (2006).

16. Tkalec, U., Ravnik, M., Čopar, S., Žumer, S. & Mušičev, I. Defects and control of optical phase singularities. Science 334, 62–65 (2011).

17. Semyuk, B. et al. Topological colloids. Nature 493, 200–205 (2013).

18. Nelson, D. R. Toward a tetrahedral chemistry of colloids. Nano Lett. 2, 1125–1129 (2002).

19. Oswald, P., Baudry, J. & Pirkle, S. Dynamic and static properties of cholesteric fingers in electric field. Phys. Rep. 337, 67–96 (2000).

20. Cross, M. C. & Hohenberg, P. C. Pattern formation outside of equilibrium. Rev. Mod. Phys. 65, 852–1112 (1993).

21. Sondhi, S. L. K., Kivelson, S. A. & Rezayi, E. H. Skyrnobons and the crossover from the fractional quantum Hall effect at small Zeeman energies. Phys. Rev. B 47, 16149–16146 (1993).

22. Wright, D. C. & Mermin, N. D. Cholesteric crystals—the blue phases. Rev. Mod. Phys. 61, 385–432 (1989).

23. Fukuda, J. & Žumer, S. Quasi-two-dimensional Skyrmion lattices in a chiral nematic liquid crystal. Nat. Commun. 2, 246 (2011).

24. Robler, U. K., Bogdanov, A. N. & Pfeifferer, C. Spontaneous skyrmion ground states in magnetic materials. Nature 442, 797–801 (2006).

25. Ackerman, P. J., Trivedi, R. P., Semyuk, B., van de Lagemaat, J. & Smalyukh, I. I. Two-dimensional skyrmions and other solitonic structures in confinement-frustrated chiral nematics. Phys. Rev. E 90, 012505 (2014).

26. Gurevich, A. O., Drak, S. I., E, R. & Bogdanov, A. N. Theory of skyrmions in liquid crystals. Phys. Rev. E 90, 042502 (2014).

27. Ronada, A. F. & Trueba, J. L. Electromagnetic knots. Phys. Lett. A 202, 337–342 (1995).

8 NATURE COMMUNICATIONS | DOI: 10.1038/ncomms7012 | www.nature.com/naturecommunications
29. Kobayashi, M. & Nitta, M. Torus knots as Hopfions. Phys. Lett. B 728, 314–318 (2014).
30. Battye, R. A. & Sutcliffe, P. M. Knots as stable soliton solutions in a three-dimensional classical field theory. Phys. Rev. Lett. 81, 4798–4801 (1998).
31. Kobayashi, M. & Nitta, M. Winding Hopfions on $\mathbb{R}^2 \times S^1$. Nuclear Phys. B 876, 605–618 (2013).
32. Humar, M. et al. Electrically tunable diffraction of light from 2D nematic colloidal crystals. Euro. Phys. J. E Soft Matter 27, 73–79 (2008).
33. Loussert, C. & Brasselet, E. Multiple chiral topological states in liquid crystals from unstructured light beams. Appl. Phys. Lett. 104, 051911 (2014).
34. Liu, Y.-K., Zhang, C. & Yang, S.-J. 3D skyrmion and knot in two-component Bose–Einstein condensates. Eur. Phys. J. E Soft Matter 25, 335–341 (2008).
35. Cooper, N. R. 'Smoke rings' in ferromagnets. Phys. Rev. Lett. 100, 190403 (2008).
36. Sutcliffe, P. Vortex rings in ferromagnets. Phys. Rev. Lett. 82, 1554–1557 (1999).
37. Borisov, A. B. & Rybakov, F. N. Dynamical toroidal hopfions in a ferromagnet with easy axis anisotropy. JETP Lett. 90, 544–547 (2009).
38. Lee, T., Trivedi, R. P. & Smalyukh, I. I. Multimodal nonlinear optical polarizing microscopy of long-range molecular order in liquid crystals. Opt. Lett. 35, 3447–3449 (2010).
39. Trivedi, R. T., Lee, T., Bertness, K. A. & Smalyukh, I. I. Three dimensional optical manipulation and structural imaging of soft materials by use of laser tweezers and multimodal nonlinear microscopy. Opt. Express 18, 27658–27669 (2010).
40. Haas, W. E. L. & Adams, J. E. Electrically variable diffraction in spherulitic liquid crystals. Appl. Phys. Lett. 25, 263–264 (1974).
41. Stratford, K., Cates, M., Henrich, O., Lintuvuori, J. & Marenduzzo, D. Self-assembly of colloid-cholesteric composites provides a possible route to switchable optical materials. Nat. Commun. 5, 3954–3954 (2014).
42. Winfree, A. T. Persistent tangled vortex rings in generic excitable media. Nature 371, 233–236 (1994).
43. Batty, R. A. & Sutcliffe, P. M. Knots as stable soliton solutions in a three-dimensional classical field theory. Phys. Rev. Lett. 81, 4798–4801 (1998).
44. Kobayashi, M. & Nitta, M. Winding Hopfions on $\mathbb{R}^2 \times S^1$. Nuclear Phys. B 876, 605–618 (2013).
45. Humar, M. et al. Electrically tunable diffraction of light from 2D nematic colloidal crystals. Euro. Phys. J. E Soft Matter 27, 73–79 (2008).
46. Nyv. Ackerman, P. J. et al. Self-assembly and electrostriction of arrays and chains of hopfion particles in chiral liquid crystals. Nat. Commun. 5, 544–547 (2014).
47. Loussert, C. & Brasselet, E. Multiple chiral topological states in liquid crystals from unstructured light beams. Appl. Phys. Lett. 104, 051911 (2014).
48. Anderson, J. E., Watson, P. E. & Bos, P. J. LC3D: Liquid Crystal Display 3-D Director Simulator Software and Technology Guide (Artech House, 2001).
49. Yeh, P. & Gu, C. Optics of Liquid Crystal Displays (John Wiley & Sons Inc., 1999).
50. Born, M. & Wolf, E. Principles of Optics (Pergamon, 1975).

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Author contributions
P.J.A. and I.I.S. performed experimental work. P.J.A., J.v.d.L. and I.I.S. analysed experimental results. P.J.A. did numerical modelling. P.J.A. and I.I.S. reconstructed director fields and defect structures. J.v.d.L. and I.I.S. provided funding. P.J.A. and I.I.S. wrote the manuscript. I.I.S. conceived and designed the project.

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