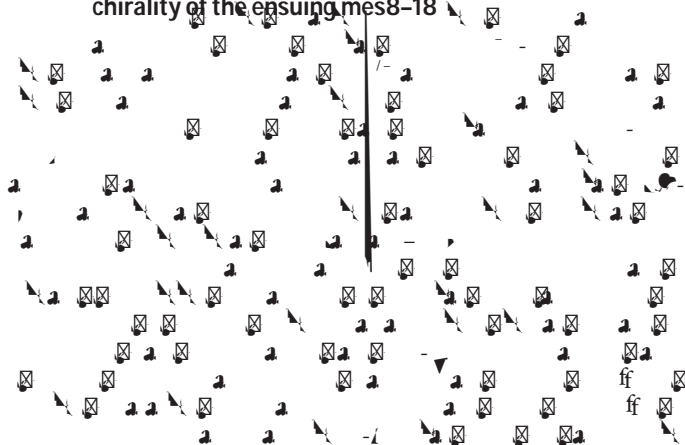


Chiral liquid crystal colloids

Ye Yuan^{1†}, Angel Martinez^{1†}, Bohdan Senyuk¹, Mykola Tasinkevych^{2,3,4} and Ivan I. Smalyukh^{1,5,6*}

Colloidal particles disturb the alignment of molecules into a non-chiral liquid crystal (LC) is that they induce chirality of the ensuing mesophase.



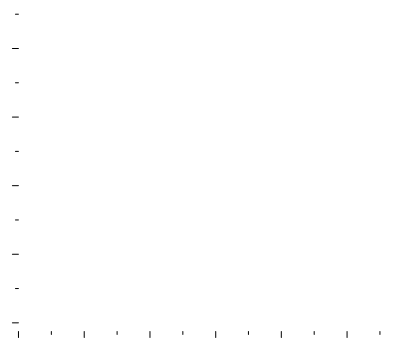
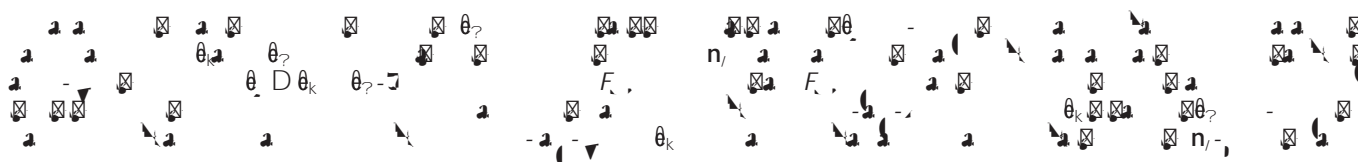


Figure 1 | Microsprings in a nematic liquid crystal. **a,b**, Right- and left-handed colloidal springs, respectively, imaged with the help of 3PEF-PM while surrounded by an isotropic medium (immersion oil). **c,d**, Self-diffusion of the particles oriented with the spring axis roughly parallel (**c**) or perpendicular (**d**) to \mathbf{n}_0 . The insets show optical bright-field micrographs at the corresponding orientations with respect to \mathbf{n}_0 . Solid lines are Gaussian fits to experimental data allowing one to calculate diffusion coefficients¹² along and normal to \mathbf{n}_0 ($D_{\parallel} = 2.9 \times 10^{-3} \mu\text{m}^2 \text{s}^{-1}$, $D_{\perp} = 0.93 \times 10^{-3} \mu\text{m}^2 \text{s}^{-1}$ in **c** and $D_{\parallel} = 3.0 \times 10^{-3} \mu\text{m}^2 \text{s}^{-1}$, $D_{\perp} = 2.7 \times 10^{-3} \mu\text{m}^2 \text{s}^{-1}$ in **d**). **e–j**, Optical micrographs of right- (**e–g**) and left-handed (**h–j**) colloidal microsprings in 5CB imaged using bright-field (**e,h**) and polarizing microscopy without (**f,i**) and with (**g,j**) an additional phase retardation plate (with the slow axis marked by the yellow double arrow) inserted between the crossed polarizers (white double arrows). **k–n**, Detailed analysis of $\mathbf{n}(\mathbf{r})$ -distortions induced in the LC by colloidal springs probed for the same particle using polarizing microscopy (**k**) and 3PEF-PM (**l–n**), with **l** and **n** showing details of two individual in-plane 3PEF-PM optical slices obtained for two different linear 3PEF-PM polarizations (marked by the green and red double arrows, respectively) and **m** showing the corresponding 3D perspective view of the particle reconstructed on the basis of superposition of many such slices. Scale bars are $5 \mu\text{m}$.



Figure 2 | Chirality-dictated alignment of microsprings in a nematic liquid crystal. **a**, Orientation-dependent free energy cost of particle-induced elastic distortions versus the angle between the spring axis $\hat{\theta}_k$ (see **b–e** and Supplementary Fig. 1 for the definition of the orthonormal reference frame attached to the particle) and n_0 for different tilt directions of $\hat{\theta}_k$, and for the right- (circles) and left-handed (squares) particles. Open (solid) symbols correspond to the anticlockwise rotation of $\hat{\theta}_k$ about $\hat{\theta}_3(\hat{\theta}_\gamma)$, as shown in **e**. The insets depict free energy variations near the stable and metastable orientation states. **f–i**, Director structures around right-handed springs (**f,h**) at energy-minimizing orientations with $\Delta = 4^\circ$ (**f**) (blue solid circles in **a**) and with $\Delta = 86^\circ$ (**h**) (blue solid circles in **a**), and left-handed springs (**g,i**) with $\Delta = 0^\circ$ (**g**) (red open squares in **a**) and with $\Delta = 90^\circ$ (**i**) (red open squares in **a**) for the metastable (**g**) and stable (**h,i**) particle orientations. **j–m**, Detailed director structures and defects corresponding to a colloidal spring shown in **g**. **n–t**, Detailed director structures and defects corresponding to a spring shown in **i**. Director structures are shown with rods depicting local orientations of $n(r)$ both at LC–particle interfaces and in the bulk, as well as with colour-coded patterns of azimuthal orientations of $n(r)$ when projected from particle surfaces to the plane orthogonal to n_0 , according to the colour scheme in the insets of **g** and **t**, as viewed from different perspectives.



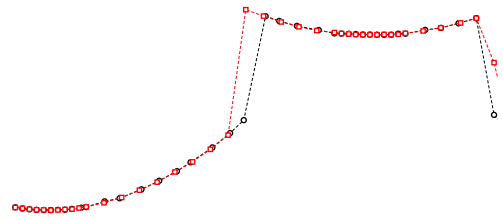
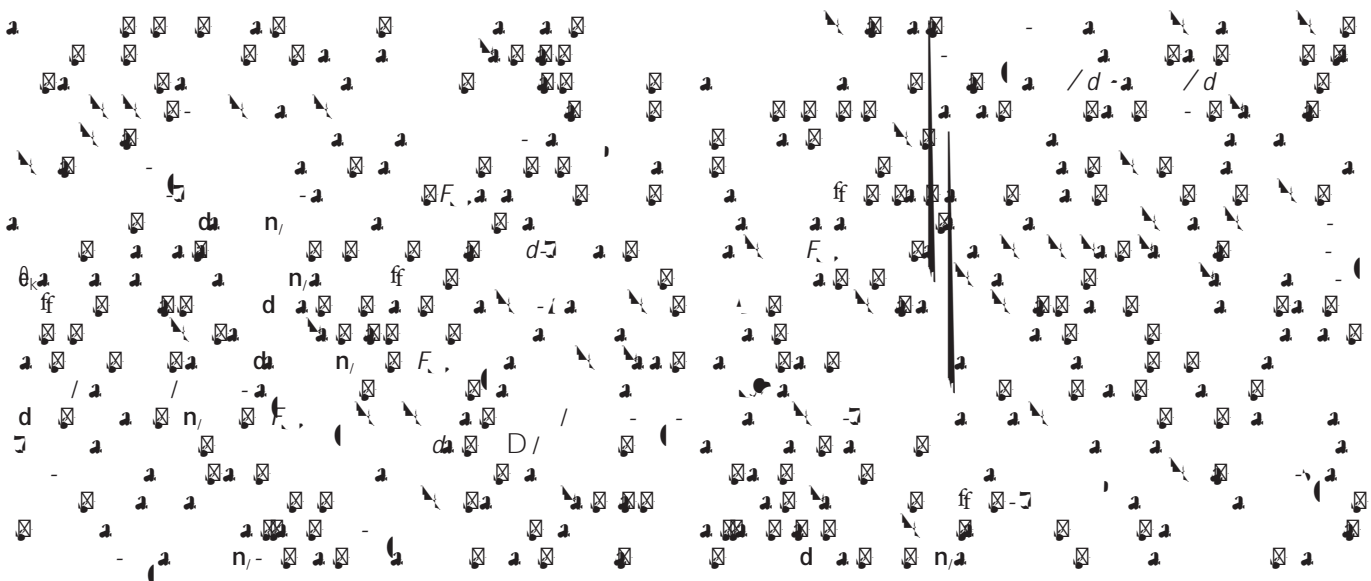


Figure 3 | Chirality-dictated alignment of single helices in a nematic liquid crystal. **a–f**, Optical micrographs of left-handed microhelices obtained using bright-field (**a,d**) and polarizing microscopy without (**b,e**) and with (**c,f**) a phase retardation plate. **g,h**, Self-diffusion of particles oriented with $\hat{\theta}_k$ roughly orthogonal (**g**) and parallel (**h**) to n_0 . The insets show optical bright-field micrographs at the corresponding orientations with respect to n_0 ($D_k = D_\gamma = 2.2 \times 10^3 \mu\text{m}^2 \text{s}^{-1}$ in **g** and $D_k = 2.7 \times 10^3 \mu\text{m}^2 \text{s}^{-1}$, $D_\gamma = 0.68 \times 10^3 \mu\text{m}^2 \text{s}^{-1}$ in **h**). **i**, Orientation-dependent free energy cost of elastic distortions induced by particles versus the angle between $\hat{\theta}_k$ and n_0 for different orientations of the tilt direction of $\hat{\theta}_k$, for left- (squares) and right-handed (circles) helices. Open (solid) symbols correspond to anticlockwise rotation of $\hat{\theta}_k$ about $\hat{\theta}_3$ ($\hat{\theta}_\gamma$), as shown in Fig. 2b–e. The insets depict free energy variations near the stable and metastable orientation states. **j–m**, Director structures around right-handed helices at $\Delta = 8^\circ$ (**j**) (blue solid circles in **i**) and at $\Delta = 82^\circ$ (**l**) (blue solid circles in **i**) and left-handed helices at $\Delta = 0^\circ$ (**k**) (red open squares in **i**) and at $\Delta = 90^\circ$ (**m**) (red open squares in **i**), which are stable (**j,k**) and metastable (**l,m**) particle orientations. The director field is shown with the help of rods and colour-coded azimuthal orientations of $n(r)$ on particle surfaces relative to n_0 according to the colour scheme shown in the inset of **k**. Scale bars are $5 \mu\text{m}$.



Figure 4 | Chirality-dependent pair interactions of colloidal springs. **a**, Time-colour-coded trajectory of attraction of like-handed microsprings initially separated along n_0 . The bottom inset shows colour scale of elapsed time counted from the moment of releasing particles from laser traps (t_{\min}) until the moment when they approach each other at t_{\max} ($t_{\max} - t_{\min} \approx 27$ s), as shown in the bright-field micrographs in the inset. The final self-assembled colloidal structure for the same particles at $t \approx 35$ s $> t_{\max}$ is also shown in the inset in a slightly enlarged micrograph and emerges as a result of additional rotation of the colloidal springs around their axes. **b**, Separation distance d versus time corresponding to **a**, with the distance dependence of the corresponding interaction force shown in the inset. **c**, Time-colour-coded trajectory of repulsion of like-handed microsprings initially separated so that the separation vector is orthogonal to n_0 . **d**, Time-colour-coded trajectory of repulsion of opposite-handed microsprings initially separated along n_0 . The insets show the corresponding optical micrographs of the interacting particles. **e**, Separation distance versus time corresponding to **d**; the distance dependence of the corresponding interaction force is shown in the inset. **f**, Time-coded trajectory of attraction of opposite-handed microsprings initially separated so that the separation vector is orthogonal to n_0 . The red curves in **b,e**, are the best fits of the experimental data with $d(t) \approx d_0 (n - t)^{1/n}$ where $n \approx 5$ for dipole-dipole interaction²¹; the fitting coefficients are $d_0 \approx 25 \mu\text{m}$, $D \approx 1.3 \times 10^5 \mu\text{m}^5 \text{s}^{-1}$ in **b** and $d_0 \approx 29 \mu\text{m}$, $D \approx 0.46 \times 10^5 \mu\text{m}^5 \text{s}^{-1}$ in **e**. Solid red lines in the insets of **b** and **e** are the best linear fits with $21 \ln F \approx (n - 1) \ln d + \text{const}$. Scale bars are $5 \mu\text{m}$.



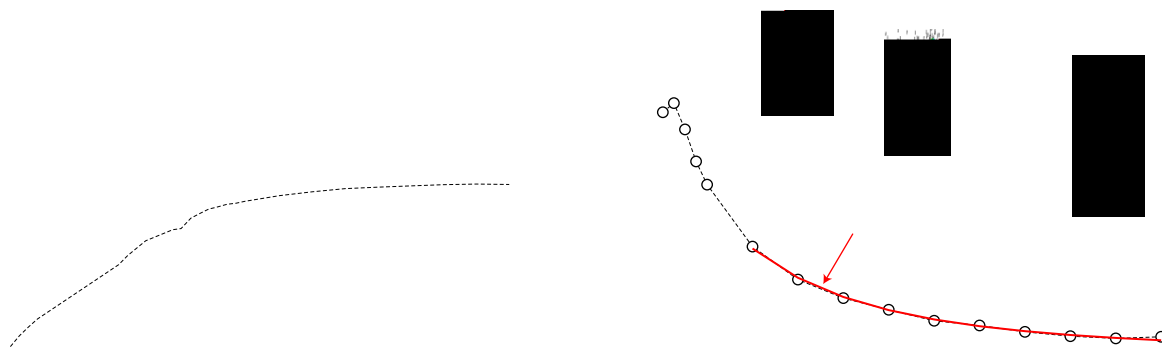


Figure 5

Figure 6 | Twisting of chiral particles relative to each other. **a**, Numerically calculated Landau–de Gennes free energy as a function of the angle γ between the axes of like-handed springs, for the case when these spring axes rotate about the particles' \hat{e}_γ vectors in opposite directions and by equal amounts, as is shown schematically in **e**. The spring centre-to-centre vector \mathbf{d} is fixed with $d \approx 1.625R$ and with the angle θ between \mathbf{d} and \mathbf{n}_0 at 90° . **b**, Free energy as a function of d at $\gamma \approx 180^\circ$, all the other parameters are the same as in **a**. The insets in **a, b** show the director configurations around the springs with the help of rods and colour-coded azimuthal orientations, with respect to \mathbf{n}_0 , of $\mathbf{n}(\mathbf{r})$ on particle surfaces. **c, d**, The case of like- (circles) and opposite-handed (squares) springs, depicting the free energy variations with γ (particles' axes rotate about \hat{e}_γ) or γ_k (particles' axes rotate about \hat{e}_3) at $d \approx 1.625R$ and $\theta \approx 90^\circ$, near the stable (**d**) and metastable (**c**) orientation states. Open (solid) symbols correspond to the case when the spring axes \hat{e}_k rotate about particles' aligned \hat{e}_γ vectors, see **e** (\hat{e}_3 vectors, see **f**) in opposite directions by equal amounts.



θ is the angle of the interface between the two phases. The effective surface energy γ_{eff} is defined as the energy per unit area of the interface, which is given by $\gamma_{\text{eff}} = \gamma_{12} \cos \theta$, where γ_{12} is the interfacial energy between the two phases. The effective surface energy γ_{eff} is a function of the interfacial energy γ_{12} and the contact angle θ . The effective surface energy γ_{eff} is a function of the interfacial energy γ_{12} and the contact angle θ . The effective surface energy γ_{eff} is a function of the interfacial energy γ_{12} and the contact angle θ .

Conclusions

We have shown that the effective surface energy γ_{eff} is a function of the interfacial energy γ_{12} and the contact angle θ . The effective surface energy γ_{eff} is a function of the interfacial energy γ_{12} and the contact angle θ . The effective surface energy γ_{eff} is a function of the interfacial energy γ_{12} and the contact angle θ .

Methods

