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# Layered chiral active matter: beyond odd elasticity

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In equilibrium liquid crystals, chirality leads to a variety of spectacular three-dimensional structures, but chiral and achiral phases with the same broken continuous symmetries have identical long-time, large-scale dynamics. In this paper, starting from *active model H\**, the general hydrodynamics of a pseudoscalar in a momentum-conserving fluid, we demonstrate that chirality qualitatively modifies the dynamics of layered liquid crystals in active systems in both two and three dimensions due to an active “odder” elasticity. In three dimensions, we demonstrate that the hydrodynamics of active cholesterics differs fundamentally from smectic-A liquid crystals, unlike their equilibrium counterpart. This distinction can be used to engineer a columnar array of vortices, with anti-ferromagnetic vorticity alignment, that can be switched on and off by external strain. A two-dimensional chiral layered state – an array of lines on an incompressible, free-standing film of chiral active fluid with a preferred normal direction – is generically unstable. However, this instability can be tuned in easily realisable experimental settings, when the film is either on a substrate or in an ambient fluid.

Chiral molecules form a spectacular range of liquid-crystalline phases [1–3] at thermal equilibrium, of which the best known is the cholesteric, with a helical structure in which the molecular orientation, described by a headless unit vector called the director  $\hat{n}$ , spontaneously twists at a uniform rate  $q_0$  along the pitch axis [3]. This uniform periodic modulation does not break translational invariance: unlike in a density wave, all surfaces of constant phase are equivalent, and an arbitrary translation along the pitch axis can be compensated by a rotation about it. Nevertheless, at scales much larger than  $1/q_0$ , the mechanics of a cholesteric is precisely the same as that of a smectic A which has an achiral one-dimensional *density* modulation. That is, microscopic chirality leads to a one-dimensional periodic structure, but the asymptotic long-wavelength elasticity and hydrodynamics of this structure show no signature of chirality [4–6]. In this Letter we show that this equivalence does not carry over to active cholesteric and smectic A phases [7, 8] when chirality couples non-trivially to the active drive. Thanks to the presence of a mix of solid- and liquid-like directions, we predict an effect odder than odd elasticity [9], in the form of a linear elastic force even at zero strain in directions in which there is no displacement field.

Recall that active systems are materials with a sustained supply of free energy, and hence broken detailed balance, at the scale of its constituents. This microscale drive manifests itself macroscopically as nonequilibrium currents and forces [10–19]. Continuum hydrodynamic theories of fluid [20], liquid-crystalline and crystalline phases [7, 8, 14, 16, 21–23] of active matter have been constructed including extensions with chiral asymmetry [9, 24–32]. In this Letter, we construct theories of layered active chiral systems. Our hydrodynamic theory starts from an active model  $H^*$  – the chiral and active

variant of model H [33] that describes the coupled dynamics of a conserved scalar order parameter and a conserved momentum density field – and applies generically to active variants of any one-dimensional spatially modulated chiral state, not only cholesterics in the traditional sense. A two-dimensional active model  $H^*$  also leads to the theory of an active two-dimensional chiral smectic, which could arise if three-dimensionally chiral particles were restricted to a thin film with a distinguished normal direction.

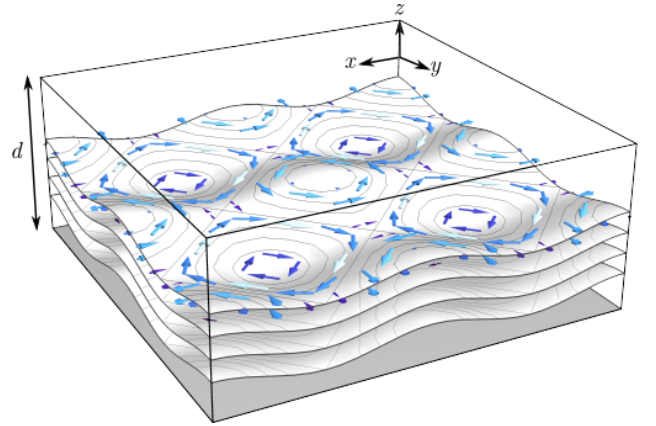


FIG. 1. The spontaneous vortex-lattice state: the chiral active force density (governed by  $z_c$ ) generates counterrotating circulatory flows, with vorticity  $\Omega_z$  obeying  $\eta \nabla^2 \Omega_z = -z_c \nabla^2 \nabla_z^2 u$ , around the undulations  $u$  of the active Helfrich-Hurault instability. Smectic layers (light grey) are shown only for the bottom half of the cell for clarity; blue arrows show the active flows only on the topmost visible layer. The bottom confining surface is shown in grey and the amplitude of the undulation varies as  $\sin(\pi z/d)$ , vanishing on both top and bottom surfaces.

Our central finding is that active cholesterics possess a chiral stress corresponding to a non-existent component of the strain tensor that yields a force density tangent to contours of constant mean curvature of the layers. As a result of this odder than odd elasticity, the undulatory instability created by active stresses [7, 8] leads to spontaneous vortical flow arranged in a two-dimensional array with vorticity aligned along the pitch axis and alternating in sign in the plane (Fig. 1). This vortex-lattice state can be switched on or off by means of an externally imposed uniaxial stress. Lastly, a two-dimensional active cholesteric is unstable with an activity threshold that goes to zero for an infinite system. This tilted-varicose instability (Fig. 2) is however not inevitable, as we discuss later in the paper.

We now show how we obtain these results. A pattern-formation framework [34] offers a foolproof approach to the construction of the hydrodynamic equations for active cholesterics, equivalent to the traditional route [7, 8] starting with the equations of motion for an orientation field and eliminating the fast degrees of freedom, but here circumventing the need to identify and remove the non-hydrodynamic mode. Accordingly, we begin by extending [35, 36] to define *active model H\**: the coupled dynamics of a pseudoscalar density  $\psi$  governed by a conservation law  $\partial_t \psi = -\nabla \cdot \mathbf{J}$  and a momentum density  $\rho \mathbf{v}$  whose dynamics in the Stokesian regime is governed by  $\nabla \cdot \boldsymbol{\sigma} = 0$ , with current  $\mathbf{J} = \psi \mathbf{v} - M \nabla \mu + \mathbf{J}_a + \mathbf{J}_c$  and stress tensor  $\boldsymbol{\sigma} = -\eta[\nabla \mathbf{v} + (\nabla \mathbf{v})^T] + \boldsymbol{\sigma}_\psi + p \mathbf{I} - \boldsymbol{\sigma}_c - \boldsymbol{\sigma}_a$ , where subscripts  $a$  and  $c$  denote active achiral and chiral contributions respectively. Here  $M$  is a mobility,  $\mu = \delta F / \delta \psi$  is a chemical potential expressed in terms of a free-energy functional  $F[\psi]$ ,  $\eta$  is a viscosity, the passive force density  $-\nabla \cdot \boldsymbol{\sigma}_\psi = -\nabla \mu$  is the Onsager counterpart to  $\psi \mathbf{v}$  [37], and the pressure  $p$  imposes overall incompressibility  $\nabla \cdot \mathbf{v} = 0$ .  $\mathbf{J}_a = \lambda_1 \psi \nabla \psi \nabla^2 \psi + \lambda_2 \psi \nabla (\nabla \psi)^2$  as familiar from active models B and H [35, 36, 38–41]. In what follows we ignore the chiral currents  $\mathbf{J}_c$ , whose effects on the dynamics of layered states arise at sub-leading order in wavenumber [42]. The achiral active stress [35, 36], in both two and three dimensions, is  $\boldsymbol{\sigma}_a = \zeta_H \nabla \psi \nabla \psi$  while the chiral active stress is

$$\begin{aligned} (\sigma_c)_{ij} &= \bar{z}_c \partial_l (\epsilon_{ijk} \partial_k \psi \partial_l \psi), \quad d = 3; \\ \boldsymbol{\sigma}_c &= \bar{\zeta}_c \left[ \boldsymbol{\varepsilon} : \nabla \psi \nabla \psi - \frac{1}{2} \boldsymbol{\varepsilon} (\nabla \psi)^2 \right], \quad d = 2, \end{aligned} \quad (1)$$

where  $d = 2$  corresponds to a thin film of 3D chiral material with a distinguished normal taken to be along  $\mathbf{N} \equiv +\hat{\mathbf{y}}$ , thus inheriting uniquely the two-dimensional antisymmetric tensor  $\boldsymbol{\varepsilon}$  with components  $\varepsilon_{ij} = \epsilon_{ijk} N_k$ . Though here written as an antisymmetric stress  $(\sigma_c)_{ij}$  can be given in an equivalent symmetric form and is allowed in momentum-conserving systems [31, 42, 49, 50].

A Swift-Hohenberg free-energy functional  $F$  [42, 51] allows model H\* to describe the dynamics of spatially

modulated states  $\psi = \psi_0 + \psi_1$  where  $\psi_1$ , with zero spatial average, represents a modulation with wavelength  $2\pi/q_s$  about a uniform background  $\psi_0$ . We consider the dynamics about a steady state with a one-dimensional spatial modulation,  $\psi_1 = \psi_1^0 (e^{i\phi} + e^{-i\phi})$ , with  $\phi = q_s(z - u)$ , describing a periodic array of parallel lines or planes of constant phase, in  $d = 2$  or 3 respectively, with normal along  $\hat{\mathbf{z}}$ , with small fluctuations  $u(\mathbf{r}, t)$ . We begin with  $d = 3$ . Defining the scaled phase-gradient vector, which is parallel to the normal of the fluctuating layers, as  $\mathbf{n} = \nabla \phi / q_s = \hat{\mathbf{z}} - \nabla u(x, y, z, t)$  we obtain the dynamical equation of the displacement field of the layers from their mean positions [42]:

$$\partial_t u = \mathbf{v} \cdot \mathbf{n} + \Lambda_1 \mathbf{n} \cdot \nabla E + \Lambda_2 \nabla \cdot \mathbf{n} (1 - 2E) - \Gamma_u \frac{\delta F[u]}{\delta u}, \quad (2)$$

where  $\Lambda_1 = -2\psi_1^{0^2} q_s^2 (\lambda_1 + \lambda_2)$  and  $\Lambda_2 = 2\psi_1^{0^2} q_s^2 \lambda_2$  are active, achiral permeative terms, the final term is passive permeation with  $\Gamma_u = -M q_s^2$ , and  $E = \partial_z u - (1/2)(\nabla u)^2$  is the covariant strain. Finally,  $F[u] = (1/2) \int [B E^2 + K (\nabla^2 u)^2]$  is the rotation-invariant free energy [3, 52] which would have controlled the relaxational dynamics of the cholesteric state in the absence of activity with  $B$  being the layer-compression modulus and  $K$  being the bending rigidity of the layers which can be expressed in terms of the coefficients in the Swift-Hohenberg free energy [42]. Force balance for our system takes the form

$$\eta \nabla^2 v_i = n_i \frac{\delta F[u]}{\delta u} + \partial_i p + \partial_j [\zeta w_{ij} + z_c \partial_l (\epsilon_{ijk} w_{kl})] \quad (3)$$

where  $w_{ij}$  encodes the active stresses from (1) and preceding. To linear order in displacements, with  $\perp$  denoting directions transverse to  $\hat{\mathbf{z}}$ ,  $\mathbf{w}_{z\perp} = \mathbf{w}_{\perp z} = \nabla_{\perp} u$ ,  $w_{z\bar{z}} = 2\partial_z u$  and all other components are 0,  $\zeta = \psi_1^{0^2} q_s^2 \zeta_H$ ,  $z_c = \psi_1^{0^2} q_s^2 \bar{z}_c$  and the pressure  $p$  enforces three-dimensional incompressibility  $\nabla \cdot \mathbf{v} = 0$ . The term proportional to  $z_c$  in (3) is the chiral active force density. It is the curl of the vector  $\partial_l w_{kl}$  and is therefore divergence-free. Expressed as a vector this chiral active force density is  $-z_c \hat{\mathbf{z}} \times \nabla_{\perp} (\nabla_{\perp}^2 u + \partial_z^2 u)$  and is directed (primarily) tangentially to contours of constant mean curvature of the layer undulation, driving the vortical flow shown in Fig. 1. Like odd elasticity, the  $z_c$ -term in (3) is a parity-breaking stress in response to layer displacements. In-plane gradients of layer mean curvature create responses in the perpendicular in-plane direction like an “odd” Laplace pressure gradient [53]. Unlike the odd elastic force density of two-dimensional chiral active solids [9], which arises from an antisymmetry in the linear relation between stress and strain, this cholesteric chiral force density (3) arises even when the strain  $E = 0$ .

It might seem that  $z_c$  does not affect the hydrodynamics of the layered state as it appears at a higher order in gradients than the achiral active force. Indeed, it does not affect the linear dynamics of the displacement field

at all: the eigenfrequency for displacement fluctuations to leading order in wavenumber, obtained by projecting (3) transverse to the wavevector and solving for the velocity field is  $\omega = -(i/\eta q^4)(Bq_z^2 q_\perp^2 - \zeta q_\perp^2 q^2)$ . Therefore, as noted in [7, 8], the dynamics of the displacement field of cholesterics and smectics are indeed equivalent, with the layered state having long-range order in three dimensions for  $\zeta < 0$  (unlike their equilibrium counterparts which only have quasi-long-range order in three dimensions) and being unstable for  $\zeta > 0$ . However, the hydrodynamics of active cholesterics differs crucially from that of smectics through the effect of the chiral active force on the velocity field *in the plane of the layers*.

The vortical flow caused by the chiral active force can be used to control and create a vortex lattice state [54] with a well defined lattice constant in an active cholesteric system. This hinges on a mapping between an *externally* imposed stress and an internal and active *achiral* stress. An external stress can be imposed via a free energy term  $F_{ext}[u] = \int \sigma_0 E$ , which gives rise to a force  $-\sigma_0 \nabla \cdot (\mathbf{w} + E\mathbf{I})$  [42]. As a consequence an achiral active stress acts identically to an *external* stress with  $\sigma_0 = \zeta$  up to an isotropic piece which can be absorbed into the pressure in an incompressible system and the instability of an *active* layered state for  $\zeta > 0$  maps onto the Helfrich-Hurault instability of a passive layered state under dilative stress [55, 56]. In the externally-stressed instability a square lattice undulated pattern  $u = u_0 \cos q_p x \cos q_p y$  (an egg-crate-like structure) is realised [57, 58] and because of this mapping, the same pattern should be realised beyond the achiral active instability as well. Due to the chiral active force  $\propto z_c$ , the egg-crate-like undulation leads to an in-plane vorticity  $\Omega_z \propto (z_c q_p^2 u_0 / \eta) \cos q_p x \cos q_p y$ , arising spontaneously from the active instability. This is the vortex lattice depicted in Fig. 1.

The correspondence between an external stress and the active achiral stress allows for a quantitative measurement of the activity. The critical threshold for a layered state of finite extent  $d$  in the  $z$  direction is  $\zeta + \sigma_0 = (2\pi/d)\sqrt{BK}$ , with the instability setting in at wavevector  $q_p \approx (\pi^2 B/4d^2 K)^{1/4}$ . When  $\zeta$  is negative the layered state is stable to the activity and a dilative external stress,  $\sigma_0 > 0$ , can be applied till the Helfrich-Hurault instability sets in [59]. This measures the active stress:  $\zeta = (2\pi/d)\sqrt{BK} - \sigma_0^{cr}$ . Conversely, for  $\zeta > 0$  the smallest  $|\sigma_0|$  that suppresses the spontaneous Helfrich-Hurault instability,  $\sigma_0^{cr}$  yields the active stress:  $\zeta = |\sigma_0^{cr}| + (2\pi/d)\sqrt{BK}$  from which we can calculate the strength of the achiral active stress using the knowledge of the thickness of the sample, compressibility and bending modulus. The magnitude of the vorticity is  $\propto z_c/\eta$ . The chiral active force should scale as  $z_c \sim \zeta \ell$  where  $\ell$  is the length of an elementary active unit (both  $\zeta$  and  $z_c$  are also likely to be functions of the concentration of the active units). Therefore, in principle, we can

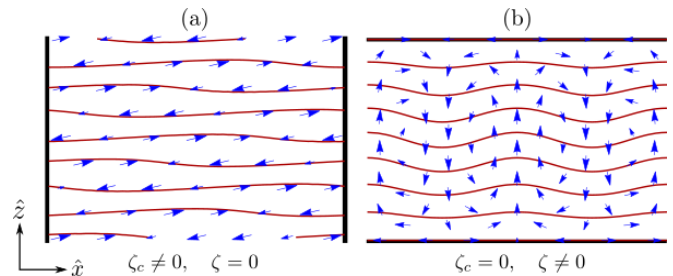


FIG. 2. Active instabilities in two-dimensional layered states. (a) Instability arising from the chiral active force ( $\zeta_c$ , here positive). The layers are indicated by dark red lines, while the linear flow field at instability overlaid (blue arrows). The thick black lines indicate confining walls, which create a finite threshold for the instability. (b) Instability arising from the achiral active force ( $\zeta$ ) for comparison; same stylings.

estimate both the chiral and achiral active stress if an active cholesteric is prepared using living liquid crystals [60], which can be engineered, for instance, by suffusing a *passive* cholesteric with bacteria. The wavelength of the passive Helfrich-Hurault instability [55, 58] can be controlled by tuning the anchoring strength at the boundaries [58, 61, 62], which can be used to engineer a vortex array with a desired lattice constant.

We now turn to a two-dimensional layered state – an array of lines in the  $x - z$  plane with normals on average along  $\hat{\mathbf{z}}$  – in a chiral, internally driven fluid. As in the three-dimensional cholesteric state, we obtain the coupled dynamics of the displacement and velocity field equations to leading order in gradients:  $\partial_t u = v_z$  and

$$\eta \nabla^2 \mathbf{v} = \hat{\mathbf{z}} \frac{\delta F[u]}{\delta u} + \nabla p + \nabla \cdot (\zeta \mathbf{w} - \zeta_c \boldsymbol{\varepsilon} \cdot \mathbf{w}) \quad (4)$$

where, to linear order in  $u$ ,  $w_{zz} = -w_{xx} = \partial_z u$  and  $w_{zx} = w_{xz} = \partial_x u$ ,  $\zeta = (\psi_1^0 q_s)^2 \zeta_H$  and  $\zeta_c = -(\psi_1^0 q_s)^2 \bar{\zeta}_c$  [42]. In (4), the chiral active force  $\propto \zeta_c$  appears at the *same* order in gradients as the achiral active force, unlike in three-dimensional cholesterics. Further, again unlike in three-dimensional cholesterics, it will be shown to affect the displacement field dynamics at linear order. In fact, the term  $\propto \zeta_c$  is fundamentally distinct from the force  $\propto z_c$  in (3); it is not obtained by averaging a thin  $x - z$  slice of a three-dimensional cholesteric. It leads to a chiral active force *along* the layers in response to both curvature and compression of the layers i.e., a pure  $\hat{\mathbf{z}}$  deformation leads to a force along  $\hat{\mathbf{y}}$ , in a direction determined by the sign of  $\zeta_c$ , which is only possible since the film has a distinguished normal, breaking three-dimensional rotation invariance, and the layered state breaks rotation (and translation) invariance in the plane of the film. This effect is related to the odd elasticity [9] of chiral active solids, but is odder still. A smectic breaks translation invariance only along one direction, so the (linearised) strain is simply  $\begin{bmatrix} 0 & 0 \\ 0 & \partial_z u \end{bmatrix}$ . Ordinary odd elasticity would create a stress  $\propto \begin{bmatrix} 0 & 0 \\ 0 & \partial_z u \end{bmatrix}$  acting along the

layers of the smectic in a direction where there is no elastic mode. Instead, the chiral activity  $\zeta_c$  produces both a *simple* shear stress  $\sigma_{xz} = \sigma_{zx} = -\zeta_c \partial_z u$  in response to strain and also a *pure* shear stress  $\sigma_{xx} = -\sigma_{zz} = -\zeta_c \partial_x u$  in response to tilt. The chiral active stress implies that a localised compression of the layer spacing produces a shear flow parallel to the layers.

We now demonstrate that a periodic array of lines in a two-dimensional film is generically destabilised due to the chiral active force. Eliminating the pressure using the incompressibility constraint in (4), solving for the velocity field and writing the wavevector  $\mathbf{q} \equiv (q_x, q_z) = q(\sin \theta_q, \cos \theta_q)$  where  $\theta_q$  is the angle between the layer normal and the wavevector, we obtain the eigenfrequency to  $\mathcal{O}(q^0)$

$$\omega = -\frac{i}{4\eta} (B \sin^2 2\theta_q - 4\zeta \sin^2 \theta_q - 2\zeta_c \sin 2\theta_q) + \mathcal{O}(q^2). \quad (5)$$

This implies an instability of the layered state for wavevector direction  $\theta_q$  just above (just below) zero for  $\zeta_c > 0$  ( $< 0$ ). This generic chiral instability for either sign of  $\zeta_c$  is distinct from the spontaneous Helfrich-Hurault [3] instability of active smectics or cholesterics, which is achiral, arises for positive  $\zeta$  [7, 8], and grows fastest at  $\theta_q \approx \pi/2$ . Eq. (5) implies that in a system confined at a scale  $d$  along  $\hat{\mathbf{x}}$  so that the smallest  $q_x \sim 1/d$ , the minimum value of the chiral active stress for which the layered state is unstable  $\sim 1/d$ . Further, this instability requires both momentum conservation *and* incompressibility. It is eliminated if the film is supported on a substrate which would add a wavevector-independent damping  $-\Gamma \mathbf{v}$  to (4). The eigenfrequency for the displacement fluctuations then vanishes at small  $q$  as  $\mathcal{O}(q^2)$ , and permeative [3] terms  $\nabla \nabla u$  in the displacement equation, subdominant for a free-standing film, now enter at the same order in gradients. Of these, terms  $\propto \partial_z^2 u$  are crucial, while others can be absorbed into redefinitions of  $\zeta$  and  $\zeta_c$ . The resulting eigenfrequency is

$$\omega = -\frac{iq^2}{4\Gamma} (B \sin^2 2\theta_q + \mathcal{B} \cos^2 \theta_q - 4\zeta \sin^2 \theta_q - 2\zeta_c \sin 2\theta_q) \quad (6)$$

where  $\mathcal{B}$  is the coefficient of the  $\partial_z^2 u$  permeative term multiplied by the friction coefficient, and the instability now occurs only if  $|\zeta_c| > (B/2) + \mathcal{B}/2 - \zeta$ , for directions  $\theta_q \approx \pi/4$ . Compressibility, as in a film bounded by bulk fluid, at large enough scales [63], is also stabilizing [64]. A detailed solution [42] of the tangent-plane velocity in this case leads to the eigenfrequency for the displacement field

$$\omega = -\frac{i|q|}{4\eta} [(B \cos^2 \theta_q - \zeta)(1 + \sin^2 \theta_q) - 12\zeta_c \sin 2\theta_q] \quad (7)$$

Equation (7) yields an instability if  $|\zeta_c| > [(B/2) - \zeta]/8$ , for  $\theta_q \approx \pi/4$  irrespective of the sign of  $\zeta_c$ . We expand on this in [42]. While a free-standing film with a generically

unstable chiral layered state may be difficult to access experimentally, films supported on a substrate or immersed in a bulk fluid can be engineered and the instability as in (6) and (7) may be observed.

In this paper we have developed the hydrodynamic theory of active chiral, layered states in two and three dimensions and demonstrated that the combination of internal drive and broken-parity qualitatively modifies the dynamics and stability of these phases unlike in their equilibrium counterparts. This difference derives from the “odd” elasticity of phases with a mixture of solid and fluid directions. By adopting the framework of a scalar field theory, active model H\* offers a fool-proof method for obtaining the correct hydrodynamic description [65, 67]. This general approach can be utilised beyond active cholesterics to construct the hydrodynamic theory of any spatially modulated active chiral state such as active chiral solid or columnar phases [68] and may present the simplest framework to understand the dynamics of and (possibly non-reciprocal) interactions between dislocations in these phases [69]. Further, the one-dimensional spatially modulated states discussed in this paper appear due to *microphase* separation in active model H\*; the dynamics of simple phase separation in this model [70] is also likely to be distinct from either passive model H or achiral but active model H.

We conclude with a brief discussion of proposals for experimental realisations, applications and possible extensions. A three-dimensional active or living cholesteric can be constructed by releasing swimming bacteria into passive biocompatible cholesteric liquid crystals, yielding a system which should display the vortex-lattice state we predict. Similarly, introducing *passive* chiral particles in an active but achiral fluid also leads to the chiral active forces discussed in our work, allowing for the realisation of a wide range of artificial active cholesteric materials. Furthermore, multiple biological systems display cholesteric organisation, the most spectacular of which is DNA in chromatin [72], which *in vivo* may be affected by DNA polymerases leading to chiral active forces of the form that we describe here.

In addition to free surfaces or interfaces of three-dimensional materials, there are numerous possibilities for realising a two-dimensional cholesteric phase. For instance, via the melting of a chiral version [9] of anisotropic active solids [23] along one direction in analogy with the emergence of (achiral) smectic phases due to an anisotropic dislocation-mediated melting of two-dimensional crystals in which dislocations unbind along one direction [73–78] (see [69] for a description of dislocations in chiral active solids). Two-dimensional layered states are also observed in active nematic fluids both in experiments on motor-microtubule gels [79–81] and simulations [82] and since these gels are known to be chiral [83], the physics we describe for two-dimensional, chiral layered states may be observable there. Chirality has

been shown to be important in epithelial cell layers [30] and a density modulated phase in these systems will lead to another realisation of two dimensional cholesterics. Non-mutual, two-species Cahn-Hilliard models [84, 85] also spontaneously form banded phases and chiral variants of these models would lead to two-dimensional chiral layered states. Finally, two-dimensional smectic phases in parity-broken systems are also possible in two-dimensional electron gases [86] and, when they are irradiated by microwave radiation [87], may have a dynamics equivalent to the one described here. Therefore, there are abundant possibilities for creating two- and three-dimensional active cholesteric states.

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