Live Soap: Stability, Order, and Fluctuations in Apolar Active Smectics

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We construct a hydrodynamic theory of noisy, apolar active smectics in bulk suspension or on a substrate. Unlike purely orientationally ordered active fluids, active apolar smectics can be dynamically stable in Stokesian bulk suspensions. Smectic order in these systems is quasilong ranged in dimension $d = 2$ and long ranged in $d = 3$. We predict reentrant Kosterlitz-Thouless melting to an active nematic in our simplest model in $d = 2$, a nonzero second-sound speed parallel to the layers in bulk suspensions, and that there are no giant number fluctuations in either case. We also briefly discuss possible instabilities in these systems.

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Equilibrium condensed matter physics owes its richness largely to the profusion and complexity of phases of equilibrium matter: crystals, nematics [1], partially translationally ordered systems like smectics *A* [1] and discotics [1], and hybrids like the hexatic *B* [2], to name a few. Beyond the equilibrium domain, in particular, in systems of "active particles" [3], the number of possibilities increases [4], but very few of these have actually been studied; nearly all past work has focused on active particles in a state of orientational order [4–6]. Thus, in understanding active matter, we are roughly where we would be in understanding equilibrium systems if we knew only the nematic liquid crystal.

The few active matter phases that *have* been thoroughly studied exhibit very different fluctuation [7–11] and flow [12–17] behavior from their equilibrium counterparts. They can order in spatial dimensions in which their equilibrium analogs cannot [7] and, paradoxically, they exhibit far larger density fluctuations [7–11] than *any* equilibrium system. Do *translationally* ordered active systems [14,18] exhibit similar phenomena? This Letter provides a partial answer for active systems with spontaneously broken translation invariance in one direction—active smectics. Specifically, we consider apolar systems of particles with their mean orientation axis along the layer normal (i.e., smectics *A* [1]), with active stresses [15] pulling in or pushing out, i.e., contractile or extensile, along that axis.

This work is a major step in the exploration of the varieties of active order: It probes whether dramatic differences between active and equilibrium liquid crystals are unique to orientational order; it is timely because slight modifications of models of ordering in biological systems yield layered phases [19]; and, finally and most importantly, it is relevant to the many observed striped nonequilibrium steady states, including the Rayleigh-Bénard problem [20,21], systems of shaken rods [22], dense collections of rod-shaped bacteria [23] or biological macromolecules, and chemical reaction-diffusion systems [24].

In this Letter, we report our results for two of the many possible models with this spatial symmetry; results for three others will be presented elsewhere [25]. Our first and simplest model treats the dynamics of stripes, ignoring all conserved quantities, and applies to convection roll patterns [21,26] and spontaneously layered phases of self-driven apolar entities, reproducing or dying while in motion [27], on a substrate which serves as a momentum sink. The second is *bulk* layered systems in a background fluid with both number and momentum conservation, which we treat in both the Stokesian (i.e., viscosity-dominated) limit and at large length scales where acceleration dominates over viscosity.

The following is our principal result: Over a finite range of parameter space, apolar active smectic order is dynamically stable and long ranged in the presence of noise in dimension $d = 3$ and quasilong ranged in $d = 2$, in contrast to equilibrium smectics [1,28]. The dynamical stability of Stokesian apolar active smectics is in sharp contrast to the generic instability of bulk active *orientationally* ordered phases in that limit [12]. These conclusions about stability reinforce and extend the findings of Ramaswamy and Simha [18]. Our theory therefore offers the first known examples of smectic long-range order in a physically accessible dimension, a two-dimensional smectic that is stable against dislocations, and a mechanically stable Stokesian ordered phase of active matter, with important implications for experiments. We show further that, unlike their orientationally ordered counterparts [4,5,7,10], apolar active smectics have finite concentration fluctuations, and that bulk apolar active smectics have a nonzero "second-sound" mode in the plane of the layers, in contrast to equilibrium smectics [1]. Finally, we find that apolar active smectics with no conservation laws undergo a transition to an active nematic as the concentration of active particles is varied. In two dimensions, "reentrance" [29] *necessarily* occurs: The active smectic phase is flanked at large and at small concentrations by the active nematic. Both transitions are in the Kosterlitz-Thouless universality class [30].

We begin with the simplest case, dealt with briefly in Ref. [18]: active elements whose number and momentum are *not* conserved, spontaneously condensed into a unidirectional fore-aft symmetric periodic structure, i.e., a smectic *A*, with mean layer normal $\hat{\mathbf{n}}_0$ along $\hat{\mathbf{z}}$ (Fig. 1). This model applies to Rayleigh-Bénard stripes in a thin fluid layer [20,21]. The only hydrodynamic field in this case is the layer displacement u, whose long-wavelength dynamics, retaining terms permitted by symmetry [31], to leading order in gradients and u, reads

$$\partial_t u = \tilde{B}\partial_z^2 u + D\nabla_\perp^2 u - \tilde{K}\nabla_\perp^4 u + f^u, \qquad (1)$$

where f^u is a Gaussian, zero-mean spatiotemporally white noise with variance 2Δ. The term with coefficient D [32] is forbidden by rotation invariance of the free energy in an *equilibrium* smectic without an aligning field but permitted here because rotation invariance *at the level of the equation of motion*, which is all one can demand in an active system, does not rule it out [34]. It implies that the local vectorial asymmetry of a curved layer leads to directed motion, as this is a driven system.

Symmetry does not fix the sign of D. A negative D leads to an undulation instability [35]. The spatial Fourier components $u(\mathbf{q}, t)$ for small wave vectors $\mathbf{q} = (\mathbf{q}_\perp, q_z)$ in the stable steady state of (1) for positive D can readily be shown to have variance $\langle |u(\mathbf{q}, t)|^2\rangle = \Delta/(\tilde{B}q_z^2 + Dq_\perp^2)$. The real-space variance $\langle [u(\mathbf{r}, t)]^2\rangle = \int_{\mathbf{q}} \langle |u(\mathbf{q}, t)|^2\rangle$ is thus finite in $d = 3$, corresponding to long-range smectic order, and $\sim \log L$ in $d = 2$ for system size L, corresponding to quasi-long-range order [18]. This establishes our principal result for the simplest case.

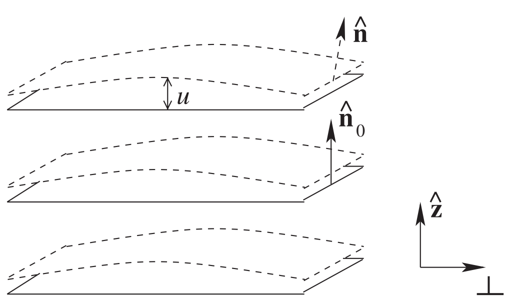

FIG. 1. Schematic representation of a smectic *A*. The solid and dotted lines represent layers in the reference and a perturbed state, respectively. The mean layer normal $\hat{\mathbf{n}}_0$ and local layer normal $\hat{\mathbf{n}}$, along with the $\hat{\mathbf{z}}$ and $\perp$ axes, are shown, as is the layer displacement field u specifying the displacement of perturbed layers along $\hat{\mathbf{z}}$.

Ignoring the subdominant $\tilde{K}$ term, Eq. (1), suitably rescaled, also describes the relaxational dynamics of an equilibrium *XY* model with a stiffness/temperature ratio $\kappa \equiv \tilde{B}^{(3-d)/2}D^{(d-1)/2}a^2/(2\pi^2\Delta)$. It then follows from well-known results on the $d = 2$ *XY* model [30] that topological defects (i.e., dislocations) in an active smectic in dimension $d = 2$ unbind, driving the system into the active nematic phase, when $\kappa = 2/\pi$, i.e., when $2\pi^2\Delta/a^2(\tilde{B}D)^{1/2} = \pi/2$. This locus is plotted in the Δ-D plane in Fig. 2(a).

We expect that the purely active quantity $D \propto c_0$, where c_0 is the concentration of active particles, and that the noise strength Δ gets an active contribution $\propto c_0$, and a c_0-independent thermal part $\propto k_BT$. Hence, varying c_0 maps out a straight line with positive intercept on the Δ axis in the Δ-D plane, as illustrated in Fig. 2(a). As is clear from that figure, this experimental locus can only enter the active smectic region by crossing the active smectic to active nematic phase boundary twice. This implies our conclusion that reentrance is inevitable in two dimensions for these systems.

In $d = 3$, as well, the transition to a nematic for this model is in the *XY* universality class. However, equilibrium smectic order at $D = 0$ exists at low enough T, so the phase boundary ends at $\Delta_c > 0$ on the Δ axis, but its slope at $D = 0$ diverges. To see this, note that, when approaching the transition from $D = 0$, D becomes significant in (1) when $D/\xi_\perp^2 \sim \tilde{K}/\xi_\perp^4$, i.e., $\xi_\perp \sim \sqrt{\tilde{K}/D}$, where $\xi_\perp \propto |\Delta - \Delta_c|^{-\nu_\perp}$ is the equilibrium in-plane correlation length for smectic order [36] at $D = 0$, implying a positive shift $CD^{1/2\nu_\perp}$ in Δ's critical value, where C is a constant. Theory [36] and experiment [37] find $1/2\nu_\perp < 1$, so the phase boundary in Fig. 2(b) has infinite slope as $D \rightarrow 0$, as illustrated in the figure. That $\Delta_c(D = 0) > 0$ means that reentrance is not inevitable; see locus (3) in Fig. 2(b).

We next consider active smectics with a constant particle number, suspended in an incompressible fluid. The conserved momentum density $\mathbf{g}$, active-particle concentration c, and broken symmetry displacement field u are now the slow variables. The particle + fluid mass density $\rho = \rho_0 =$ constant and $\nabla \cdot \mathbf{v} = 0$, where $\mathbf{v} \equiv \mathbf{g}/\rho$ is the velocity field.

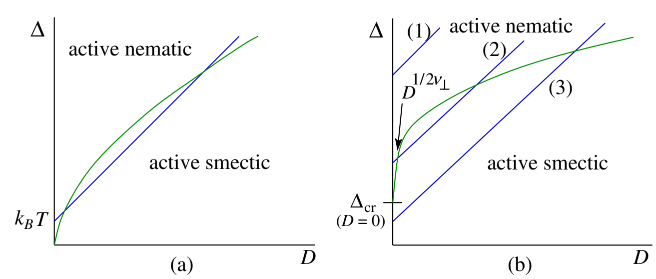

FIG. 2 (color online). Phase diagram of active smectics, in the activity D noise strength Δ plane, for (a) $d = 2$ and (b) $d = 3$. The straight lines indicate approximate loci mapped out in this parameter space by varying the concentration c_0 of active particles with other parameters fixed.

Conservation of total momentum reads $\partial_t \mathbf{g} = -\nabla \cdot \boldsymbol{\sigma}$, with linearized stress tensor

$$\boldsymbol{\sigma} = p\mathbf{I} - \boldsymbol{\eta}(\nabla\mathbf{v} + \nabla\mathbf{v}^T) + \boldsymbol{\sigma}^{(el)} + \boldsymbol{\sigma}^a + \boldsymbol{\sigma}^N, \quad (2)$$

with p the fluid pressure, $\boldsymbol{\eta}$ the viscosity tensor, and the elastic force density $-\nabla \cdot \boldsymbol{\sigma}^{(el)} = -\mathbf{n}\delta F/\delta u$, with

$$F = \frac{1}{2}\int d^d x[B(\partial_z u)^2 + K(\nabla_\perp^2 u)^2 + A(\delta c)^2 + 2C\delta c \partial_z u]. \quad (3)$$

Here, B and K are the layer compression and bend moduli, respectively; A is the osmotic modulus; and C is a cross coupling. The active stress [5,12] $\boldsymbol{\sigma}^a = -Wc\hat{\mathbf{n}}\,\hat{\mathbf{n}}$, where $\hat{\mathbf{n}} \equiv (\hat{\mathbf{z}} - \nabla u)/|\hat{\mathbf{z}} - \nabla u|$ is the local normal to the smectic layers; negative and positive activity W per particle correspond, respectively, to extensile and contractile stresses; and $\boldsymbol{\sigma}^N$ is noise.

The resulting equation of motion for $\mathbf{v}$, linearized in $\mathbf{v}$, u, and $\delta c = c - c_0$, with c_0 the mean concentration, reads

$$\rho_0 \partial_t \mathbf{v} = -\nabla p + \hat{\mathbf{z}}[B\partial_z^2 u - K\nabla_\perp^4 u + (C + W)\partial_z \delta c] - Wc_0(\hat{\mathbf{z}}\nabla_\perp^2 u + \partial_z \nabla_\perp u) + \nabla \cdot (\boldsymbol{\eta}\nabla\mathbf{v}) + \mathbf{f}^v, \quad (4)$$

where $\mathbf{f}^v = \nabla \cdot \boldsymbol{\sigma}^N$ is a momentum-conserving noise and $\langle \sigma_{ij}^N(\mathbf{0}, 0)\sigma_{kl}^N(\mathbf{r}, t)\rangle = 2\Delta_{ijkl}\delta(\mathbf{r})\delta(t)$. For simplicity, and free from fluctuation-dissipation constraints [38], we will take $\Delta_{ijkl} = \Delta_v(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ and $\eta_{ijkl} = \eta\delta_{jk}\delta_{il}$. The linearized hydrodynamic equation of motion for u is

$$\partial_t u = v_z + \tilde{B}\partial_z^2 u + D\nabla_\perp^2 u - \tilde{K}\nabla_\perp^4 u + \tilde{C}\partial_z \delta c + f^u, \quad (5)$$

where the noise f^u has statistics as in (1) [39].

Number conservation implies that $\partial_t c = -\nabla \cdot \mathbf{J}^c$. Gradient expanding the current $\mathbf{J}$ subject to the symmetry constraints (rotational and translational invariance) gives

$$\mathbf{J}_c = -\hat{\mathbf{z}}[(A_z - W^c)\partial_z \delta c + W^c c_0 \nabla_\perp^2 u + C_{zz}\partial_z^2 u] - \nabla_\perp[A_\perp \delta c + (C_{\perp z} + W^c c_0)\partial_z u] + \boldsymbol{f}^c, \quad (6)$$

where the Gaussian noise $\boldsymbol{f}^c = (\boldsymbol{f}_\perp^c, f_z^c)$ has variance $2\Delta_\perp^c$ and $2\Delta_z^c$ transverse to and along z, respectively. In (6), we have included an active current [9] $W^c\nabla \cdot (c\hat{\mathbf{n}}\,\hat{\mathbf{n}})$, where W^c is a phenomenological coefficient. In an *equilibrium* two-component smectic, the constraints $W^c = 0$ and $C_{\perp z}/C_{zz} = A_\perp/A_z = \Delta_\perp^c/\Delta_z^c$ would apply. For simplicity, we take $\Delta_\perp^c = \Delta_z^c \equiv \Delta^c$, $C_{\perp z} = C_{zz} = E$, and $A_\perp = A_z \equiv G$.

We solve (4) in the Stokesian limit $\rho_0 \partial_t \mathrm{v} \ll \eta\nabla^2 \mathrm{v}$ and insert the resulting $\mathbf{v}$ into (5). The spatial Fourier transforms of $\Phi \equiv -\partial_z u$ and δc obey

$$\partial_t \Phi_{\mathbf{q}} = -M_{\mathbf{q}}\{[Bq_z^2 + Wc_0(q_z^2 - q_\perp^2) + Kq_\perp^4]\Phi_{\mathbf{q}} - (C + W)q_z^2 \delta c_{\mathbf{q}}\} + [\partial_t \Phi_{\mathbf{q}}]_P - iq_z\left(f_{\mathbf{q}}^u + \frac{P_{zj,\mathbf{q}} f_{j,\mathbf{q}}^v}{\eta q^2}\right), \quad (7)$$

$$\partial_t \delta c_{\mathbf{q}} = (Eq^2 + 2W^c c_0 q_\perp^2)\Phi_{\mathbf{q}} - (Gq^2 - W^c q_z^2)\delta c_{\mathbf{q}} - i\mathbf{q} \cdot \boldsymbol{f}_{\mathbf{q}}^c, \quad (8)$$

where $M_{\mathbf{q}} \equiv q_\perp^2/\eta q^4$, $P_{zj,\mathbf{q}} = \delta_{zj} - q_z q_j/q^2$, and $[\partial_t \Phi_{\mathbf{q}}]_P$ summarizes the "permeative" $\tilde{B}$, $\tilde{K}$, $\tilde{C}$, and D terms from (5), which are of higher order in wave number than those shown explicitly in (7).

Suppose $B > CE/D = C^2/A$, so that when activity $W = 0$ the smectic state is stable. Let $|W| > C > 0$; a similar analysis holds for $C < 0$. At small q, where $[\partial_t \Phi_{\mathbf{q}}]_P$ is negligible, it is clear from (7) that negative (i.e., extensile) W can lead to an instability with $\mathbf{q}$ along z, i.e., a modulation in layer spacing. However, the layer compression modulus B always stabilizes this when $(B - |W|c_0) > 0$. Thus, the system is stable for small enough $|W|$, establishing one of our main results.

For contractile active stresses $W > 0$, we see from (7) and (8) that the most unstable modes have $\mathbf{q}$ in the $\perp$ direction, in which neither the layer compression elasticity nor the coupling to the concentration act. Hence, the instability threshold for W vanishes in the limit of large system size, as in Refs. [12,13]. The instability causes splay and self-generated flow, as in active nematics [12,13]. For smectics, this is a spontaneous version of the Helfrich-Hurault [1,35] undulation instability.

The instability that arises in the extensile ($W < 0$) case when $|W| > B$ is interesting. Equations (7) and (8) have the same form as the linear part of the Fitzhugh-Nagumo [40,41] equation, which exhibits sustained oscillations under rather general conditions. We speculate that such oscillations could also occur here, i.e., a breathing smectic. We will explore this in future work [25].

We now turn to the statistics of fluctuations in the bulk Stokesian limit. It is clear by inspection that (7) and (8) will have a mode each with frequency $\sim q^0$ and q^2 at small q, corresponding primarily to stress relaxation and concentration, respectively. We use the clear separation of these time scales to simplify their evaluation from (7) and (8): For $q \ll q_> \equiv B/\sqrt{\eta E(C + W)}$, δc can be shown to be negligible in (7) for the purpose of evaluating the variance of u, and $\Phi_{\mathbf{q}}$ in (8) can be eliminated in favor of $\delta c_{\mathbf{q}}$ by treating $\Phi_{\mathbf{q}}$ as fast in (7). On the other hand, the Stokesian approximation $\rho_0 \partial_t \mathbf{v} \ll \eta q^2 \mathbf{v}$ can be shown to hold if and only if $B/\eta \ll \eta q^2$, which requires $q \ll q_< \equiv \sqrt{B\rho_0}/\eta$. To estimate parameters to see when the ratio $q_>/q_< = \sqrt{B\eta/\rho_0 E(C + W)} \gg 1$ is large, we argue that an order-unity splay of the smectic layers, i.e., $|\partial_z \nabla_\perp u| \sim 1/a$, should give a particle current $\sim O(c_0 v_0)$, where a is the layer spacing and v_0 is the typical propulsion speed. This yields $E \sim c_0 v_0 a$, where a is the smectic layer spacing, assumed comparable to the particle size. Estimating in addition $C \sim W \sim B/c_0$, we find

$$q_>/q_< \sim \sqrt{\eta/\rho_0 v_0 a} = 1/\sqrt{\mathrm{Re}}, \quad (9)$$

where Re $\equiv \rho_0 v_0 a/\eta$ is the Reynolds number of an individual particle. Thus, we see that, provided individual swimmers are in the low Reynolds number limit, our two approximations are valid over a *large* range $q_< \ll q \ll q_>$ and can be brought to bear on (7) and (8). The displacement variance is thereby found to be

$$\langle |u(\mathbf{q}, t)|^2 \rangle = \frac{\Delta_v}{\eta[Bq_z^2 + Wc_0(q_z^2 - q_\perp^2)]}. \tag{10}$$

This scales like $1/q^2$ for all directions of $\mathbf{q}$, precisely as in the simplest model considered earlier. Hence, like that model, the Stokesian apolar active smectic also exhibits smectic translational order that is long ranged in the presence of noise in dimension $d = 3$ and quasilong ranged in $d = 2$. The δc correlator takes the form

$$\langle |\delta c(\mathbf{q}, t)|^2 \rangle = \frac{\Delta^c q^2}{Gq^2 - W^c q_z^2 - (Eq^2 + 2W^c c_0 q_\perp^2)(C + W) q_z^2 / G_q}, \tag{11}$$

where $G_q \equiv Bq_z^2 + Wc_0(q_z^2 - q_\perp^2)$.

As is clear from our discussion above, at sufficiently long wavelengths (i.e., for $q < q_<$), the Stokesian approximation must break down. We must then take the acceleration $(\partial_t \mathbf{v})$ into account in Eq. (4). The complete hydrodynamics in this case will be presented elsewhere [25]; here, we will limit ourselves to the two most important results: that the second-sound speed is finite even for propagation *within* the plane of the smectic layers (in which direction this speed vanishes in equilibrium smectics [1]) and that the smectic translational order is long ranged in the presence of noise in dimension $d = 3$ and quasilong ranged in $d = 2$, in contrast to equilibrium smectics, which have only quasi-long-ranged order in $d = 3$ [1] and short-ranged order in $d = 2$ [28].

Fourier transforming (4)–(6) in space and time and defining $\theta_{\mathbf{q}}$ to be the angle between $\mathbf{q}$ and the z axis yields, at the longest wavelengths, a pair of sound modes with frequency $\omega(\mathbf{q}) = \pm c(\theta_{\mathbf{q}}) q - i\Gamma(\theta_{\mathbf{q}}) q^2/2$, with direction-dependent second-sound speed and damping coefficient

$$c(\theta_{\mathbf{q}}) \equiv |\sin(\theta_{\mathbf{q}})| \sqrt{\frac{[B + Wc_0]\cos^2(\theta_{\mathbf{q}}) - Wc_0 \sin^2(\theta_{\mathbf{q}})}{\rho_0}}, \tag{12}$$

$$\Gamma(\theta_{\mathbf{q}}) \equiv \left(\frac{\eta}{\rho_0} + \tilde{B}\cos^2(\theta_{\mathbf{q}}) + D\sin^2(\theta_{\mathbf{q}})\right), \tag{13}$$

respectively. Note that the second-sound speed does not vanish for propagation parallel to the layers ($\theta_{\mathbf{q}} = \pi/2$); instead, it goes to $\sqrt{-Wc_0/\rho}$ (recall that $W < 0$ in the stable regime). Note also that this sound speed *would* vanish in the absence of activity $W = 0$, recovering the well-known [1] result for an equilibrium smectic.

With an extension of the same algebra, we find that the variances of u and δc are

$$\langle |u(\mathbf{q}, t)|^2 \rangle = \frac{[\rho_0^{-1}\Delta_v q^2 + \Delta G_q + (C+W)^2 q_z^2(\Delta_\perp^c q_\perp^2 + \Delta_z^c q_z^2)/G_q]}{G_q \Gamma(\theta_{\mathbf{q}}) q^2 + (C+W) q_z^2 C_q}$$

and

$$\langle |\delta c(\mathbf{q}, t)|^2 \rangle = \frac{(\Delta_\perp^c q_\perp^2 + \Delta_z^c q_z^2)}{(A_z - W^c) q_z^2 + A_\perp q_\perp^2 - (C + W) q_z^2 C_q / G_q},$$

where G_q is defined after (11) and $C_q \equiv C_{zz} q_z^2 + (C_{\perp z} + 2W^c c_0) q_\perp^2$. Since once again the variance $\langle |u(\mathbf{q}, t)|^2 \rangle \propto 1/q^2$ for all directions of $\mathbf{q}$, we again find that translational order is long ranged in $d = 3$ and quasilong ranged in $d = 2$, while the fact that the variance $\langle |\delta c(\mathbf{q}, t)|^2 \rangle$ is finite as $q \to 0$ for all directions of $\mathbf{q}$ again implies that there are no giant number fluctuations.

In conclusion, we have constructed the dynamical equations for active smectics, both in bulk suspensions and in confined systems in contact with a momentum sink. Our theory is generic, applicable to any driven system with spontaneous stripe order and appropriate conservation laws. We show, extending Ref. [18], that noisy active smectic order is long ranged in dimension $d = 3$ and quasilong ranged in $d = 2$ for all dynamical regimes and that active smectic suspensions have a nonzero second-sound speed parallel to the layers. For $d = 2$, we predict a Kosterlitz-Thouless transition from active nematic to active smectic, with a reentrant nematic at low concentration. We show that smectic elasticity suppresses the giant number fluctuations and extensile instabilities that occur in active nematics.

Our results should apply to a wide range of active systems, including horizontal layers of granular matter agitated vertically or fluids heated from below. Extensions to active mesophases in agitated two-dimensional electron gases [42], where Coulomb and magnetic-field effects enter, will be discussed elsewhere [25]. We look forward to detailed experimental tests of our predictions.

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