

# Chiral active hexatics: Giant number fluctuations, waves and destruction of order

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Active materials, composed of internally driven particles, have been shown to have properties that are qualitatively distinct matter at thermal equilibrium. However, most spectacular departures from equilibrium phase behaviour were thought to be confined to systems with polar or nematic asymmetry. In this paper we show that such departures are also displayed in more symmetric phases such as hexatics if in addition the constituent particles have chiral asymmetry. We show that chiral active hexatics whose rotation rate does not depend on density, have giant number fluctuations. If the rotation-rate depends on density, the giant number fluctuations are suppressed due to a novel orientation-density sound mode with a linear dispersion which propagates even in the overdamped limit. However, we demonstrate that beyond a finite but large lengthscale, a chirality and activity-induced relevant nonlinearity invalidates the predictions of the linear theory and destroys the hexatic order. In addition, we show that activity modifies the interactions between defects in the active chiral hexatic phase, making them non-mutual. Finally, to demonstrate the generality of a chiral active hexatic phase we show that it results from the melting of chiral active crystals in finite systems.

Active matter is driven out of equilibrium by a continuous supply of energy at the scale of constituent particles which couples with the underlying asymmetry of the system leading to macroscopic forces and currents [1–3]. Active hydrodynamics [4, 5], a continuum theory for such nonequilibrium states, have been constructed for multiple active liquid-crystalline phases and have, more recently, been extended to include chiral asymmetry [6–10]. This extension has important experimental implications – materials such as actin or microtubule filaments [11] or even cells that are microscopic constituents of multiple frequently-studied biological systems are themselves chiral. The interaction between chirality and activity, especially in two dimensions, leads to several surprising features including the suppression of the generic instability [1, 12] in orientationally-ordered active fluids [13], odd viscosity waves in isotropic chiral fluids [14] and waves in overdamped chiral active solids [15].

In this paper we consider large scale properties of *hexatic* chiral active systems in two dimensions. Chiral hexatic ordering can arise upon dislocation unbinding of chiral active solids [15] just as passive hexatic ordered phases result from the solid phase. Active Brownian particles that are chiral can be engineered and should form such phases, just as achiral active Brownian particles form hexatic phases [16]. Further, hexatic correlations are routinely observed in cell-layers and tissues [17, 18] and since it is known that multiple cells are chiral [19] they should be described by our theory. A chiral phase with local hexagonal correlations has also been observed in spermatozoa at a planar interface [20] and bacteria has been shown to organise into a chiral hexagonal crystal [21]. Furthermore, hexagonal organisation of chiral microscopic units is common even at the subcellular level – in clathrin coats for instance – and our theory will form

the basis for the study of topological defects in this system which may, in the future, lead to an understanding of topology transition in this system [22]. Beyond hexatics, the hydrodynamic theory we construct also describes *all*  $n$ -atic chiral active phases, for  $n > 2$  and therefore, our results are valid for *all* of them, including tetratics. Finally, as we will discuss at the end of the paper, our theory may be more widely applicable even in systems in which microscopic constituents are not themselves chiral.

Much of the interesting phase behaviour in active systems arise from the interaction of the nonequilibrium drive with dipolar or quadrupolar asymmetry. This interaction leads to “sound modes” with linear dispersion in *overdamped* polar active systems [23] and giant number fluctuations in polar and nematic active systems [1, 23–25]. However, it was believed that more symmetric phases will not display such spectacular departures from equilibrium behaviour [16, 25]. This is broadly correct for *achiral* hexatic phases. However, we demonstrate that this is *not* correct for *chiral* hexatic phases. We show that the active hexatic phase has anomalous number fluctuations – the root-mean-squared number fluctuations  $\sqrt{\delta N^2}$  in a region containing on average  $N$  particles scaling as  $N$  instead of  $\sqrt{N}$  as in equilibrium – when the global rotation rate is either 0 or constant [13]. However, a *density-dependent* rotation rate changes the picture significantly: the giant number fluctuations are suppressed and the coupled density-orientational fluctuations lead to either a wave with a linear dispersion relation or an instability with a linear growth rate at small wavevectors even in the overdamped limit. However, the interplay of activity and chirality also yields a *relevant* nonlinearity, in the renormalisation-group sense, leading to the destruction of quasi-long-range-ordered (QLRO) hexatic state in two dimensions and implying that the earlier predictions

only applies for systems below a critical size. We then demonstrate that the behaviour of both single topological defects *and* interaction between defects are modified by chirality and activity. A single defect spontaneously rotates and its angular far-field is modified due to the interplay of activity and chirality. The force between two oppositely-charged defects is also modified and becomes non-mutual – the magnitude of the force exerted by a positively charged defect on a negatively charged one is different from that exerted by the negatively charged defect on the positive one, which leads to a directed motion of a defect-pair on a substrate. This modification of the defect-interaction may lead to defect-separation and destruction of the ordered phase, but the non-mutual interactions also opens up the possibility of more exotic behaviour such as defect charge-separation. Finally, we will discuss how an active chiral hexatic emerges from a chiral solid and relate the phenomenological coefficients in the chiral hexatic to those in the active solid.

We first discuss non-rotating hexatics on a substrate. The local density of the particles is described by  $\rho(\mathbf{x}, t)$  which obeys a continuity equation  $\partial_t \rho = -\nabla \cdot (\rho \mathbf{v})$  where  $\mathbf{v}(\mathbf{x}, t)$  is their velocity field. The six-fold-symmetric phase is characterised by the complex order parameter  $\Psi = \psi e^{i\theta(\mathbf{x}, t)}$  where, for hexatic phases formed of circular particles,  $\theta(\mathbf{x}, t)$  denotes the orientation of the line joining two neighbouring particles with respect to an arbitrary but fixed axis (see Fig. 1). It can also describe the orientation of more complex elementary units such as the hexagonal structures formed by clathrin triskelia [22]. The small angular deviations about the perfectly ordered state is described by

$$\partial_t \theta = \frac{1}{2} \epsilon_{ij} \partial_i v_j + \gamma_c \nabla \cdot \mathbf{v} - \Gamma_6 \frac{\delta F}{\delta \theta} + \xi_\theta(\mathbf{x}, t) \quad (1)$$

where  $F = \int d\mathbf{r} K(\nabla \theta)^2/2 + f(\rho)$  is the free energy with  $f(\rho)$  being a function of density,  $\Gamma_6$  is a dissipative kinetic coefficient,  $\xi_\theta(\mathbf{x}, t)$  is a Gaussian white noise of strength  $\Delta_\theta$  and

$$\epsilon = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} \quad (2)$$

is the Levi-Civita tensor. The first term on the R.H.S. of (1) denotes the precession of the angular distortion in a local vorticity field [26]. The second, with a chiral but *passive* coefficient  $\gamma_c$ , leads to a chiral precession due to a local isotropic compression or dilation. The third term controls the relaxation of the bond-angle order parameter to its equilibrium value in the absence of activity and flow. Finally, the equation for the velocity field is

$$\rho \partial_t \mathbf{v} = -\Gamma \mathbf{v} - \gamma_c K \nabla \nabla^2 \theta - \frac{K}{2} (\hat{z} \times \nabla) \nabla^2 \theta - \rho \nabla \frac{\delta F}{\delta \rho} - \zeta \hat{z} \times \nabla \theta - \zeta_c \hat{z} \times (\hat{z} \times \nabla) \theta + \Gamma_c \hat{z} \times \mathbf{v} + \boldsymbol{\xi}_v \quad (3)$$

The terms in the first line of (3) are passive forces, with the first being the usual frictions and the others being

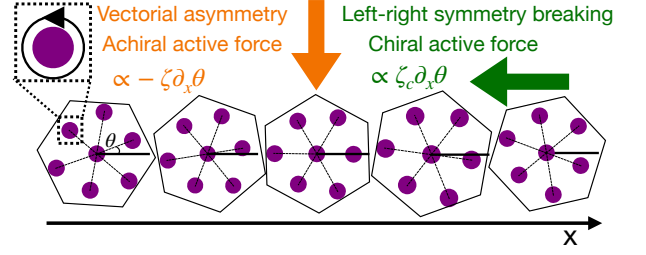


FIG. 1: Illustration of the angle-dependent active forces. For a distortion of the hexatic along  $\hat{x}$ , the achiral active force, denoted by the green arrow, is directed along the natural vectorial asymmetry direction. However, the direction of the force  $\propto \zeta_c$  does not correspond to any natural vectorial asymmetry and requires chiral or left-right asymmetry. Inset, we show the microscopic chirality of a single particle, with the handedness being denoted by a circular arrow, which leads to this asymmetry.

passive couplings to the orientation and density fields. The first three terms in the second line are active while the last is a Gaussian white noise of strength  $\Delta_v$ . The first active term, with the coefficient  $\zeta$ , is an achiral force equivalent to the one discussed in [25] which can only be present in systems which do not conserve angular momentum [27]. The second, with a coefficient  $\zeta_c$  is explicitly chiral, with the handedness being encoded in the sign of  $\zeta_c$  (see Fig. 1). However, this can be reexpressed as  $\zeta_c \nabla \theta$  and is not a source of any internal angular momentum density. The final term is an *active chiral* friction and has the same form as a Lorentz force on a charged particle in a plane in the presence of a magnetic field perpendicular to the plane.

We eliminate  $\mathbf{v}$  using (3) in the overdamped limit to obtain coupled hydrodynamic equations for the density and angle fields in the Fourier space:

$$\partial_t \theta = -q^2 \left[ \left( \Gamma_6 K + \frac{\Gamma_c \zeta_c - \Gamma \zeta + 2\gamma_c (\Gamma \zeta_c + \zeta \Gamma_c)}{2(\Gamma^2 + \Gamma_c^2)} \right) \theta - \frac{\Gamma_c + 2\Gamma \gamma_c}{2(\Gamma^2 + \Gamma_c^2)} \rho \right] + \xi_\theta \quad (4)$$

$$\partial_t \rho = -q^2 \rho_0 \left[ -\frac{\Gamma \zeta_c + \zeta \Gamma_c}{\Gamma^2 + \Gamma_c^2} \theta + \frac{\Gamma}{\Gamma^2 + \Gamma_c^2} \rho \right] + \xi_\rho. \quad (5)$$

where  $\xi_\rho$  is a conserving Gaussian white noise inherited from (3) and has the correlation  $q^2 \Delta_v / (\Gamma^2 + \Gamma_c^2)$ . In achiral but active hexatics, the  $\theta$  and  $\rho$  equations would have been linearly decoupled at this order in wavevectors. The coupling to the angle field in (5), which is purely chiral, yields a mass density current  $\propto i\mathbf{q}\theta$  which in the steady-state must be balanced by diffusive current  $\propto i\mathbf{q}\rho$  [28]. Therefore, density fluctuations in chiral

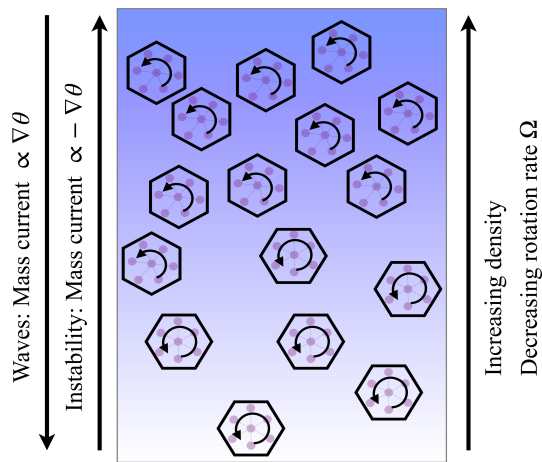


FIG. 2: Mechanism of a wave with a linear dispersion or an instability with growth-rate  $\sim q$  in hexatics with density-dependent rotation rates. The density gradient (denoted by the colour gradient) leads to faster particle rotation in the less-dense region and therefore, to an orientation gradient. When the orientation gradient leads to a current from the dilute to the dense region, the ordered homogeneous state is destabilised. The propagating wave is obtained when the angular gradient leads to a particle current from the denser to the more dilute regions.

active hexatics must scale as orientational fluctuations just as in active nematics. The orientational fluctuations are Goldstone modes of broken rotational symmetry and  $\lim_{q \rightarrow 0} \langle |\theta|^2 \rangle \sim 1/q^2$  [29] and as we explicitly show in [30]; this implies that the static structure factor of density fluctuations is  $\lim_{q \rightarrow 0} \langle |\delta\rho|^2 \rangle \sim 1/q^2$ . This is in contrast to passive systems and achiral active hexatics in which it goes to a constant as  $q \rightarrow 0$ . The number fluctuations are, therefore, giant – a region containing on average  $N$  particles must have R.M.S. fluctuations  $\sqrt{\delta N^2} \sim N$  – which was hitherto believed to require polar or nematic asymmetry [16, 25]. This demonstrates that *all*  $n$ -atic systems, including tetratics and hexatics should display giant number fluctuations if they are chiral.

We now consider the chiral hexatics which rotate spontaneously – this is a purely chiral nonequilibrium phase in which the argument of the order parameter  $\Psi(\mathbf{x}, t) = \psi e^{i(\theta - \Omega t)}$  rotates spontaneously [31]. We must, therefore add  $\Omega$  to the R.H.S. of (1), where

$$\Omega(\rho) = \Omega(\rho)|_{\rho=\rho_0} + \left. \frac{\partial \Omega}{\partial \rho} \right|_{\rho=\rho_0} \delta\rho = \Omega_0 + \Omega_1 \delta\rho. \quad (6)$$

The constant rotation rate can be eliminated by simply transforming to a rotating reference frame with  $\theta \rightarrow \theta - \Omega_0 t$  implying that the results discussed for a non-rotating active chiral hexatic also apply to its steadily rotating counterpart. However, a density-dependent rotation rate fundamentally modifies the phase behaviour. The lowest

order in wavevector equation for the angle field in a frame rotating at  $\Omega_0$  is  $\partial_t \theta = \Omega_1 \delta\rho + \mathcal{O}(q^2)$  while the density dynamics is still described by (5). This yields a *linear* dispersion  $\omega_{\pm} = \pm c q$  where

$$c = \sqrt{-\frac{\rho_0 \Omega_1 (\Gamma_c \zeta + \Gamma_c \zeta_c)}{\Gamma^2 + \Gamma_c^2}}. \quad (7)$$

When  $\Omega_1 (\zeta_c \Gamma + \Gamma_c \zeta) < 0$ , i.e. either when the rotation rate slows down with increasing density and a gradient of the angle field leads to a mass current in the direction of the gradient or when the rotation rate increases with density and an angular gradient leads to a mass current in the opposite direction, this leads to a propagating density-orientation wave with a *linear dispersion*. Such a linear sound-wave-like mode in an overdamped system is only possible in an active system and was, in addition, thought to require polarity [1, 23, 28]. Here, however, they arise due to *chiral* asymmetry. This chiral active current also reduces the number fluctuations which now obeys the law of large number:  $\lim_{q \rightarrow 0} \langle |\delta\rho|^2 \rangle \sim \text{const.}$  [30]. Heuristically, since  $\omega \sim q$  and  $-i\omega\delta\rho \sim -q^2\theta$ ,  $\delta\rho \sim -iq\theta$  and therefore,  $\langle |\delta\rho|^2 \rangle \sim q^2 \langle |\theta|^2 \rangle \sim q^2 (1/q^2)$ . Eq. (7) further implies that when  $\Omega_1 (\zeta_c \Gamma + \Gamma_c \zeta) > 0$ , the homogeneous hexatic phase is *unstable* with a growth rate of fluctuations  $\propto q$  (see Fig. 2). If  $\Omega_1 > 0$  and  $(\zeta_c \Gamma + \Gamma_c \zeta) > 0$ , an angular gradient leads to a mass current *in the direction of the gradient* leading to an increase in density. This higher density leads to a local increase of the rotation rate reinforcing the angular gradient and leading to an instability of the homogeneous hexatic state, possibly towards a patterned structure.

We have till now considered only linear deviations away from the steady-state. We now check whether nonlinear terms affect the conclusions reached using the linear theory. For this, we first consider a ‘‘Malthusian’’ hexatic [32] i.e. one in which the density is not globally conserved, but locally held fixed. The nonlinearity with fewest gradients and fields in the equation for angular fluctuations  $\sim (\nabla\theta)^2$  which arises from the advective nonlinearity  $\mathbf{v} \cdot \nabla\theta$  in the equation for angular fluctuations, since  $\mathbf{v}$  has a chiral contribution  $\propto \nabla\theta$ . The nonlinear equation of motion for the angle field of a noisy Malthusian hexatic, upon eliminating the velocity field is

$$\partial_t \theta = \frac{\lambda}{2} (\nabla\theta)^2 + \bar{K} \nabla^2 \theta + \xi_\theta \quad (8)$$

where  $\lambda = -2\Gamma\zeta_c + \zeta\Gamma_c/(\Gamma^2 + \Gamma_c^2)$  and

$$\bar{K} = \left( \Gamma_6 K + \frac{\Gamma_c \zeta_c - \Gamma \zeta + 2\gamma_c (\Gamma \zeta_c + \zeta \Gamma_c)}{2(\Gamma^2 + \Gamma_c^2)} \right) \quad (9)$$

in terms of the previously introduced variables. Eq. (8) has the same form as the KPZ equation with the only distinction being that  $\theta$  is a compact variable. From the studies of the KPZ equation, it is known that the nonlinearity with the coefficient  $\lambda$  is *marginally relevant* in two

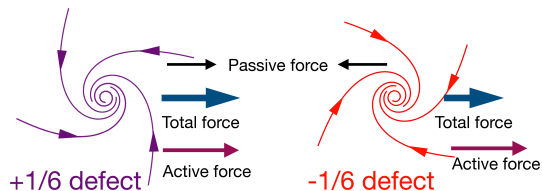


FIG. 3: Non-mutual interaction between  $\pm 1/6$  defects (we have schematically shown  $\nabla\theta$  for each defect; see [30]). The passive interaction between the defect-pair is attractive and mutual, but the active force may be non-mutual. Therefore, a  $+1/6$  defect may be attracted to a  $-1/6$  defect while repelling the  $-1/6$  defect leading to the motion of the centre-of-symmetry of the  $\pm 1/6$  pair.

dimensions i.e., it grows larger upon renormalisation, invalidating the linear theory at thermodynamic scales. In the surface growth problem, this yields the “rough state” at large scales with an algebraic scaling of height fluctuations. In the context of the hexatic phase this implies that the hexatic state loses even algebraic order beyond the scale  $L_* \sim e^{16\pi\bar{K}^3/\Delta\theta\lambda^2}$  [33, 34]. While this calculation is for a Malthusian hexatic system, a coupling to the density field cannot reduce the angular fluctuations. The lowest order nonlinearity in the angle field equation that couples density and angular fluctuations has the form  $\nabla\delta\rho \cdot \nabla\theta$ . Since the linear static structure factor of the density fluctuations is either as large as angular fluctuations (when the spontaneous rotation-rate doesn’t depend on density) or smaller (for density-dependent rotation rate) this nonlinearity is either as relevant as  $(\nabla\theta)^2$  or less relevant than it. In either case, the conclusion that even algebraic order is destroyed due to the  $(\nabla\theta)^2$  nonlinearity cannot be modified by the extra nonlinearities coupling density and angular fluctuations.

Further, we have, till now, only considered smooth fluctuations of the angle field. While we have already demonstrated that the chiral hexatic only has short-range order, topological defects, which in a hexatic predominantly have charges  $\pm 1/6$ , can still have significant impact on the phase behaviour. We show that chirality leads to a spontaneous rotation of a single defect with an angular speed  $\propto \zeta_c$  even when a defect-free hexatic does not rotate [30]. The  $\lambda$  term in (8) also leads to a qualitative modification of the far field structure of the angle-field due to a defect – unlike in achiral hexatics, in which the angular far-field is independent of the distance from the defect core, in chiral active hexatics the angle field is an explicit function of the distance from the core-radius. This is similar to defect structures in compact KPZ equation [33] and spiral waves in complex Ginzburg-Landau equation. Furthermore, activity also qualitatively modifies the interaction between two defects. In particular, we show in [30] that the inter-

actions between defects are *non-mutual* – the strength of attractive or repulsive force exerted on a  $+1/6$  defect by a  $-1/6$  defect is different from the force exerted by a  $+1/6$  defect on a  $-1/6$  – implying that though a single  $+1/6$  or  $-1/6$  defect doesn’t self propel, a  $\pm 1/6$  pair, maintained at a fixed separation, does (see Fig. 3). Furthermore, [33] demonstrated that the two-defect interaction potential *in the absence of non-mutual interaction* changes sign for  $L_d \sim e^{2\bar{K}/\lambda}$  implying that defects unbind beyond this scale. In chiral active hexatics, this is complicated by the non-mutuality of interaction but we find that the *sign* of the interaction between a  $\pm 1/6$  pair changes beyond a critical distance within a perturbative treatment to  $\mathcal{O}(\lambda^2)$ . However, non-mutual interactions between defects complicates the many-body physics, and opens up possibilities for novel behaviours, such as charge-separation, which we will explore in a future publication.

We now discuss how a chiral hexatic phase may arise, at least in a small enough system (since there is no chiral active hexatic in the thermodynamic limit, at large scales a solid must melt directly to a liquid), from the melting of a chiral solid. In the crystalline phase, which breaks both rotation and translation symmetries, the bond-angle field is slaved to the displacement field, which we denote by  $\mathbf{u}(\mathbf{x}, t)$ , as

$$\epsilon_{ij}\theta = \frac{1}{2}(\partial_i u_j - \partial_j u_i) = W_{ij}^a, \quad (10)$$

where  $\mathbf{W}^a$  is the antisymmetric part of the displacement gradient tensor which denotes a rotation of the crystal structure. Inserting this form of  $\theta$  into the active forces  $-\zeta\hat{z} \times \nabla\theta$  and  $-\zeta_c\hat{z} \times (\hat{z} \times \nabla)\theta$  in (3), we find that these correspond to the active forces  $\zeta\nabla \cdot \mathbf{W}^a$  and  $-\zeta_c\nabla \cdot (\boldsymbol{\epsilon} \cdot \mathbf{W}^a)$  in the solid phase which were ignored in [15]. We therefore construct a complete theory of active chiral solids, including all active forces in [30] and formulate a phenomenological theory of dislocation unbinding which yields the hydrodynamic equations of the chiral active hexatic that we have discussed in this paper. This connects the phenomenological parameters in (1) and (3) with those in the theory of the solid.

We close by discussing the generality of our results and its experimental implications. First, though we considered a chiral hexatic on a substrate, our primary results, giant number fluctuations in a non-rotating hexatic, waves if the rotation rate of the ordered phase depends on the density, destruction of quasi-long-range order due to nonlinearities all remain valid even for a hexatic phase formed by a suspension of active particles in an incompressible momentum conserved fluids with  $\nabla \cdot \mathbf{v} = 0$  [30]. Next, though our results are strictly applicable for an inherently chiral system, they may have relevance for microscopically *achiral* systems, in which chiral symmetry is broken spontaneously. We outline two possibilities for breaking of chiral symmetry in achiral active hexat-

ics. First is a hexatic structure formed by active Brownian particles (ABPs) in which there is an explicit aligning interaction between the polar vectors (which determine the motility direction) of the ABPs [35, 36]. In these systems, while the arrangement of the particles themselves are hexatic, polar vectors form aligned domains whose configuration can lead to a chiral rotation of the hexatic structure with the sense of rotation depending on the detailed configuration of the domains. The second arises in hexatics formed by ABPs in which there is no explicit alignment interaction between the internal polar vectors [16, 35, 37]. Even here, however, the polar vectors have an interaction with the structure which can be non-mutual – the polar vectors may try to align with the structure while the structure anti-aligns with the polar vectors. This may lead to a spontaneous chiral symmetry-broken state in which the structure rotates at a constant rate [38]. Our theory should describe the hydrodynamic fluctuations of both these states. We also discuss the potential implication of the nonlinear destabilisation and non-mutual defect interactions uncovered here for chiral phase with lower angular symmetry such as nematics in the supplement [30]. Our results also have implications for multiple experimental systems such as active, chiral vortex lattice phases in motor-microtubule systems [39] and in fast-moving bacteria [21]. Epithelial tissues also often have a hexagonal structure [17, 18, 40] and intrinsic chirality have been observed in multiple cellular systems [19], raising the possibility that certain tissues may be described as active chiral hexatics and our result for giant number fluctuations for non-rotating systems may be tested there at timescales at which cellular birth and death are unimportant. Recently, cells in an isotropically confined epithelial sheet were shown to all spontaneously rotate in the same sense [41]; the predicted density-orientation wave may be observed in this system. Chiral hexagonally ordered phases have also been observed in simulations [42], [43, 44] and our predictions regarding the hexatic phase should be verifiable there.

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