

## Hexatically Ordered Superfluids

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We develop a theory for a novel state of <sup>4</sup>He films that possesses off-diagonal order (as in the superfluid state) as well as hexatic or bond orientational order. Within our description, both the hexatic and superfluid transitions are still of the Kosterlitz-Thouless type, but the superfluid stiffness is sensitive to the hexatic transition. We briefly discuss the possible relevance of this work to recent experiments on submonolayer helium films.

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In the early 1970s a substantial amount of theoretical work was done on the topic of supersolid helium [1–11]. Such a system would possess both the quantum coherence, characterized by off-diagonal long-range order in the density matrix [12], and crystalline order, characterized by long-range order in the density at some finite wave vector, or “diagonal” long-range order in the density matrix. Progress was hampered by a lack of convincing experimental evidence that such a state existed in nature. In this Letter we investigate the properties of two-dimensional helium films which have off-diagonal order as well as hexatic order (characterized by algebraic long-range order in the orientation of directions to nearest-neighbor atoms) [13]. We call such a system a “superhexatic,” to distinguish it from the superfluid state which possesses no such orientational order.

Superhexatic systems are interesting for several reasons. First, the hexatic constraints required on the many-body wave function are *prima facie* much weaker than the crystalline ones of a supersolid [2], which should make it easier for the quantum and spatial order to coexist. Second, superhexatics can be mapped to a variety of other systems of physical interest such as the two-dimensional XY magnet on a random lattice, vortex lines in superconductors [9,10], or a set of two-dimensional Coulomb charges in quantum gravity [14]. In such models there are two different species of charge (for example, point masses and Coulomb charges), each with its own dynamics and each influencing the other. Third, there is some evidence that superhexatics may have been seen in experiment. Recent third sound measurements of submonolayer films of helium on hydrogen and deuterium substrates demonstrate *two* independent Kosterlitz-Thouless (KT) transitions: the standard superfluid transition near 1 K and a second one near 0.5 K [15,16]. As we shall discuss below, the lower transition may be the freezing of the superfluid to form a superhexatic as the temperature is lowered.

We shall show below that couplings between off-diagonal and hexatic order can exist, and that they need not destroy the superfluid or hexatic transitions. These couplings do allow for the hexatic transition to be seen

in the superfluid stiffness. Finally, we make a brief comparison of this work to experiment.

A qualitative [17–20] and quantitative [21] theory of the superfluid phase transition can be built up from the theory of “ring exchanges.” In this approach the partition function is calculated using standard path-integral techniques, in which the system is evolved for an imaginary “time”  $\hbar\beta \equiv \hbar/kT$ , where  $T$  is the temperature. The indistinguishability of the bosons allows for contributions to the partition function in which helium atoms have exchanged positions. The superfluid transition occurs when it becomes entropically favorable for large “rings” of bosons to permute their positions, establishing phase coherence across the system [19].

Hexatic order can affect this process in at least two ways. First, the additional stiffness of the system might reduce the amplitude for helium atoms to exchange position [2,22], thereby drastically lowering the superfluid density. Second, there is a subtle topological effect. The hexatic-fluid transition can be viewed as a disclination-unbinding transition [23–25], where a perfect disclination is a topological defect in the bond orientational order. These point disclinations can interact with the point charges in the superfluid order. Such a vortex-disclination interaction appears naturally in the context of an XY model with constant couplings embedded in a fluctuating geometry. Consider, by way of example, a lattice of helium containing a single disclination. The theory of ring exchanges of the atoms can be mapped to a Landau-Ginzburg theory for the superfluid. The energy depends only upon gradients of the phase, and can be related to two-dimensional electrostatics [24,26] wherein vortices play the role of Coulomb charges. The effect of the disclination can then be viewed as distorting the plane containing the charges into either a cone for a negative disclination or a saddle for a positive disclination [27]. Using conformal mapping one can solve the electrostatics problem for a single charge on a cone (saddle) and find that it is repelled (attracted) to the disclination independent of the sign of the charge, and that the energy of interaction depends logarithmically on their separation.

A similar calculation shows that the energy of a vortex and a dislocation varies inversely with the distance between them.

It is also possible to demonstrate couplings between the hexatic and superfluid order using a microscopic many-body approach [28], but here we will consider only a phenomenological Landau-Ginzburg model. The free energy of a superhexatic may be written as a sum of the elastic energy, the superfluid energy, and an interaction energy. The elastic term is given by [27]

$$E_{el} = \int d\mathbf{r} \left\{ \frac{1}{4\tilde{\mu}(1+\tilde{\nu})} |\nabla^2 \chi(\mathbf{r})|^2 + i\eta(\mathbf{r})\chi(\mathbf{r}) + E_b \mathbf{b}(\mathbf{r})^2 + E_\Theta \Theta(\mathbf{r})^2 \right\}, \quad (1)$$

where  $\tilde{\mu}$  and  $\tilde{\nu}$  are elastic constants,  $\chi(\mathbf{r})$  is the “gauge field” of the stress tensor,  $\sigma_{ij}$ , so that  $\sigma_{ij}(\mathbf{r}) = \epsilon_{ik}\epsilon_{jl}\partial_k\partial_l\chi(\mathbf{r})$ , and  $\mathbf{b}(\mathbf{r})$  and  $\Theta(\mathbf{r})$  are the dislocation (Burgers vector) and disclination densities with core en-

ergies  $E_b$  and  $E_\Theta$ , respectively [29]. These defect densities can be combined into a single scalar, the incompatibility,  $\eta(\mathbf{r}) \equiv \Theta(\mathbf{r}) + \hat{\mathbf{z}} \cdot \nabla \times \mathbf{b}(\mathbf{r})$ , which acts as a source term for the field  $\chi(\mathbf{r})$ , where  $\hat{\mathbf{z}}$  is a unit vector normal to the plane of the film. We assume that we are well above the melting transition of the solid, and treat the dislocation density as a continuous field [23], ignoring the discrete nature of the dislocations. We next write the superfluid energy in terms of a fictitious electrostatic potential  $\phi(\mathbf{r})$ , related to the phase of the superfluid order parameter,  $\theta(\mathbf{r})$ , by  $\nabla\phi(\mathbf{r}) = -\hat{\mathbf{z}} \times \nabla\theta(\mathbf{r})$ . Using this potential, the superfluid energy may be written in a fashion similar to the elastic energy:

$$E_{sf} = \int d\mathbf{r} \left\{ \frac{1}{4\pi\rho_s} |\nabla\phi(\mathbf{r})|^2 + i\nu(\mathbf{r})\phi(\mathbf{r}) + E_\nu\nu(\mathbf{r})^2 \right\}, \quad (2)$$

where  $\nu(\mathbf{r})$  is the density of point vortex “charges” for the field  $\phi(\mathbf{r})$ .

The leading order interaction between the two fields may be written as

$$E_{int} = \int d\mathbf{r} \left\{ i\gamma_{jk\ell m} \partial_j \partial_k \chi(\mathbf{r}) \partial_\ell \phi(\mathbf{r}) \partial_m \phi(\mathbf{r}) + \lambda_{jk\ell m} b_j(\mathbf{r}) b_k(\mathbf{r}) \partial_\ell \phi(\mathbf{r}) \partial_m \phi(\mathbf{r}) + E_{\Theta\nu} \Theta(\mathbf{r}) \nu^2(\mathbf{r}) \right\}. \quad (3)$$

The coupling tensors  $\gamma_{ijk\ell}$  and  $\lambda_{ijk\ell}$  are required to be symmetric in order to preserve rotational invariance. For the purposes of this calculation we consider only the trace (diagonal) contributions,

$$E_{int}^{(0)} = \int d\mathbf{r} \left\{ i\gamma_0 \nabla^2 \chi(\mathbf{r}) |\nabla\phi(\mathbf{r})|^2 + \lambda_0 \mathbf{b}(\mathbf{r})^2 |\nabla\phi(\mathbf{r})|^2 + E_{\Theta\nu} \Theta(\mathbf{r}) \nu^2(\mathbf{r}) \right\}; \quad (4)$$

this is the lowest order interaction one can write for the two fields. Time-reversal invariance places a powerful constraint on the theory: any interaction requires an even number of powers of  $\nabla\phi$ ; there is no similar requirement on  $\nabla\chi(\mathbf{r})$ . The first term in Eq. (4) represents the coupling of the stress tensor to the superflow. In the absence of dislocations it generates the logarithmic interactions between vortices and disclinations mentioned earlier. The second term represents the possible variation of the local superfluid density (stiffness) with the local dislocation density. The final term represents the interaction between the cores of disclinations and vortices, and can be shown to be generated from the first term in Eq. (4).

The partition function of the superhexatic system may be written as

$$Z = \sum_{\Theta(\mathbf{r}), \nu(\mathbf{r})} \int \mathcal{D}\phi[\mathbf{r}] \mathcal{D}\chi[\mathbf{r}] \mathcal{D}\mathbf{b}[\mathbf{r}] e^{-\beta(E_{el} + E_{sf} + E_{int}^{(0)})}. \quad (5)$$

We simplify the partition function in two steps. First, we integrate out the Gaussian dislocation vector field,  $\mathbf{b}(\mathbf{r})$ , treating  $\lambda_0$  as a small parameter in the resulting action. This introduces a  $|\nabla\chi(\mathbf{r})|^2$  term in the action so that the interaction of bare disclinations is reduced from  $r^2 \ln r$  in the absence of dislocations to a logarithmic one due to partial screening by the dislocations [23]. Second, we will work in the small fugacity limit, so that we may limit the contributions of the sums over vortex and disclination charge to 0 and  $\pm 1$ , turning the problem into that of two coupled sine-Gordon systems [26]. After some algebra we obtain

$$Z = \int \mathcal{D}\phi[\mathbf{r}] \mathcal{D}\chi[\mathbf{r}] \exp \left( -\beta \int d^2r \left\{ \frac{1}{4\mu(1+\nu)} |\nabla^2 \chi(\mathbf{r})|^2 + \frac{1}{2E_b} |\nabla\chi(\mathbf{r})|^2 + \frac{1}{4\pi\rho_s} |\nabla\phi(\mathbf{r})|^2 + i\gamma_0 \nabla^2 \chi(\mathbf{r}) |\nabla\phi(\mathbf{r})|^2 - \frac{\lambda_0}{E_b} |\nabla\chi(\mathbf{r})|^2 |\nabla\phi(\mathbf{r})|^2 + g_1 \cos \beta\phi(\mathbf{r}) + g_2 \cos \beta\chi(\mathbf{r}) + g_3 \cos \beta\chi(\mathbf{r}) \cos \beta\phi(\mathbf{r}) + i g_4 \sin \beta\chi(\mathbf{r}) \cos \beta\phi(\mathbf{r}) \right\} \right). \quad (6)$$

The constants  $g_i$  can be simply related to the original core energies:  $g_1$  and  $g_2$  are the fugacities of the vortices and disclinations,  $g_3$  represents the added energy required to place a vortex on a disclination, and  $g_4$  reflects whether or not the energy cost depends upon the

sign of the disclination.

Let  $T_\Theta$  and  $T_\nu$  be the hexatic and superfluid transition temperatures, respectively, for the uncoupled models ( $T_\nu > T_\Theta$ ). Elementary power counting indicates

that all of the couplings between the two order parameters allowed by time-reversal symmetry are irrelevant (in the renormalization-group sense) at the Gaussian fixed point,  $T_\Theta$ . A more rigorous calculation using momentum shell cutoff renormalization [30] upholds this conclusion near the line of Gaussian fixed points, even in the regime  $T_\Theta < T < T_\nu$ , where the disclination fugacity  $g_2$  is perturbatively relevant [28]. This result is quite important: it shows that weak coupling of the two models will not destroy the KT nature of either the vortex-unbinding or disclination-unbinding transition. It also allows us to conclude that there will not be a discontinuous jump in the superfluid stiffness when the hexatic transition occurs.

However, this does not prove that the superfluid stiffness is wholly insensitive to the presence of the hexatic order. For example, when we pass through the hexatic transition we expect a change in  $\langle |\nabla\chi|^2 \rangle$  proportional to the integral of the bump in the specific heat. Such a bump is a nonuniversal feature of the transition in that its width and magnitude depend upon detailed features of the system [31]. If we have a coupling of the form  $|\nabla\chi|^2 |\nabla\varphi|^2$ , then we would expect a change in the superfluid stiffness proportional to this integrated bump. For a suitable choice of parameters this can be fairly sharp. However, the lowest-order perturbative renormalization-group analysis discussed above is only valid near the transition, and cannot demonstrate such a feature.

As a proof of principle, we have simulated the superhexatic transition numerically using a Monte Carlo analysis. If  $\theta_i(\mathbf{r})$  is the angle to nearest neighbor  $i$  of the site at  $\mathbf{r}$ , we can define a hexatic order parameter  $\psi_6(\mathbf{r}) = \sum_{\text{neighbors}} e^{i6\theta_i(\mathbf{r})} \equiv |\psi_6(\mathbf{r})| e^{i6\theta_6(\mathbf{r})}$ , so that  $\theta_6(\mathbf{r})$  plays a role similar to that of the superfluid phase  $\theta(\mathbf{r})$ ; its gradient is at right angles to the gradient of its corresponding gauge field,  $\nabla\chi(\mathbf{r})$ , introduced above. The energy can then be written in terms of  $\nabla\theta$  and  $\nabla\theta_6$ . This gradient model can be approximated using the cosine of a phase difference on a lattice, since they are in the same universality class. We thus simulate the problem as two coupled XY models on a discrete lattice:

$$E = - \sum_{(i,j)} k_1 \cos(\theta(r_i) - \theta(r_j)) + k_2 \cos(\theta_6(r_i) - \theta_6(r_j)) \\ + s [\cos(\theta(r_i) - \theta(r_j)) - 1] [\cos(\theta_6(r_i) - \theta_6(r_j)) - 1]. \quad (7)$$

For a suitable choice of parameters we can obtain a change in the superfluid stiffness as a function of temperature near  $T_\Theta$ , as shown in Fig. 1. This size of this jump should depend upon the disclination fugacity, which cannot be varied in this simple approach.

We conclude with a brief comparison of this work with recent experiments by Chen and Mochel [15,16] as well as other theoretical approaches to the same experiments [32,33]. Measurements of third sound velocities in submonolayer  $^4\text{He}$  films on hydrogen and deuterium sub-

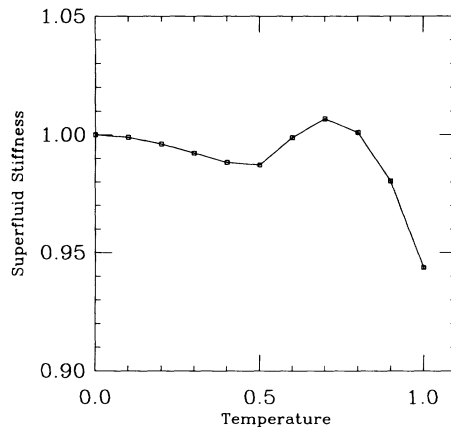


FIG. 1. The superfluid stiffness as a function of dimensionless temperature for a system with a hexatic transition, as determined by a Monte Carlo simulation of the action in Eq. (7) for a  $100 \times 100$  lattice with periodic boundary conditions. The drop in stiffness near  $T = 0.50$  is due to the onset of hexatic order; its size and width are proportional to that of the integral of the specific heat of the hexatic over the transition. The parameters  $k_1 = 1$  and  $k_2 = 2$  were chosen so that the uncoupled hexatic and superfluid transitions occur at 1 and 2, in these dimensionless units. The stiffness is calculated for coupling  $S = 0.5$ . The solid line is a guide for the eye; the error bars are smaller than the symbols.

strates suggest the existence of two KT transitions. The upper transition occurs near 1 K, and is consistent with the standard superfluid transition on conventional substrates. The lower one occurs at roughly 0.5 K, and is signaled by a change in the third sound velocity as the temperature is lowered: for deuterium substrates the velocity drops, whereas for hydrogen the velocity increases. The temperature of this transition scales linearly with the  $^4\text{He}$  density. For films thicker than a single monolayer deposited on hydrogen substrates, an additional third sound mode appears over the temperature range between the two transitions.

If the lower transition is indeed KT, then it seems likely to be a melting transition of either a solid or a hexatic. A superhexatic would have no shear modulus, so that superflow would not be reduced by pinning, which is consistent with experiment. Depending upon the details of the coupling between the hexatic and the superfluid order parameters, it is possible to obtain an increase or decrease in the superfluid stiffness. This change may be sharp, but is not discontinuous, which is also consistent with the experiments.

The superhexatic model cannot explain the additional third sound mode seen in some cases. However, this mode is only seen for coverages greater than one monolayer, and may result from different dynamics in the two layers. A more crucial difficulty is dealing with the effect of the substrate potential on the hexatic transition. In classical systems a hexagonal substrate will lock in the hexatic order, so that no transition occurs [23]. The large zero point motion of both the helium and the hydrogen may

mitigate this effect, but that must be demonstrated by further study.

Instead of considering spatial ordering of the helium atoms, Zhang [32] and Kapitulnik [33] have independently postulated an ordering of the thermally excited vortices. In such a picture the intermediate state would be a vortex-antivortex lattice (VVL) with spontaneously broken time-reversal symmetry. In this picture the upper transition is the melting of the lattice to produce a normal fluid, and the lower transition is the sublimation of the lattice as its constituents disappear. Zhang has shown how the additional third sound branch can be nicely explained in terms of the optical modes of the VVL. However, in such a picture *neither* transition should produce KT behavior for the superfluid. The upper melting transition would produce a universal jump in the shear modulus of the VVL, not in the superfluid density, as is observed experimentally, and as predicted in our model. In addition, there is no reason why the additional branch should only be observed on hydrogen substrates when the coverage exceeds one monolayer.

The two pictures differ in that the superhexatic phase should occur below the new, lower transition, while the VVL is predicted to occur at and above it. By measuring the vortex diffusivity and the onset of nonlinear dissipation as a function of temperature, one should be able to establish which regime exhibits anomalous ordering.

The above analysis of the interaction of spatial and superfluid order can be extended to physical systems with a similar mathematical description. In particular, it can be applied to an array of Josephson junctions that has been deliberately constructed to include a disclination or dislocation. The geometric analysis discussed above would predict a force on vortices that attracts or repels them from the defect center. Such an interaction might be visible in experiments on the ballistic motion of vortices.

In conclusion, we have introduced the notion of a novel phase for  $^4\text{He}$  films called a "superhexatic," which displays both off-diagonal and bond orientational order. We have shown that couplings between the two exist, and that the couplings preserve the KT nature of the two transitions. We have shown that this phase is roughly consistent with experiments, and suggested ways to test if this approach is correct. Studies of the microscopics and dynamics are under way [28].

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