



Crystalline Order in Superfluid ^3He Films

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We predict an inhomogeneous phase of superfluid ^3He films in which translational symmetry is spontaneously broken in the plane of the film. This phase is energetically favored over a range of film thicknesses, $D_{c_2}(T) < D < D_{c_1}(T)$, separating distinct homogeneous superfluid phases. The instability at the critical film thickness, $D_{c_2} \approx 9\xi(T)$, is a single-mode instability generating striped phase order in the film. Numerical calculations of the order parameter and free energy indicate a second-order instability to a periodic lattice of degenerate B -like phases separated by domain walls at $D_{c_1} \approx 13\xi(T)$.

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The phases of superfluid ^3He provide a beautiful example of spontaneously broken symmetry in condensed matter physics, exhibiting properties common to superconductors, nematic liquid crystals, and antiferromagnets. Many of the unique physical properties of superfluid ^3He , including the spectrum of low-energy excitations, are connected to the breaking of orbital and spin rotation symmetries in combination with global gauge symmetry that is associated with superfluidity and superconductivity. In spite of the complex order that develops, the bulk A and B phases of ^3He are translationally invariant. Indeed, translational symmetry is generally assumed to hold even in reduced dimensions, e.g., superfluid films [1–4].

NMR measurements on relatively thick (μm) films show evidence of an A - to B -like transition predicted within the context of Ginzburg-Landau (GL) theory [5]. However, unexplained anomalies in film flow [6] and third sound experiments [7] suggest that our current theoretical understanding of the phases of superfluid ^3He films is insufficient. One of the intriguing questions raised by these experiments is whether or not there may be qualitatively new phases stabilized in reduced dimensions [8].

Here we report the theoretical prediction of a phase of superfluid ^3He exhibiting *spontaneously* broken translational symmetry. This phase is shown theoretically to be the stable ground state of a superfluid ^3He film, with the broken translational symmetry occurring in the plane of the film. The mechanism responsible for this phase is competition between surface depairing and domain-wall formation between degenerate ground states, and is generic to ^3He confined in at least one spatial dimension [9].

The superfluid phases of ^3He are Bardeen-Cooper-Schrieffer (BCS) condensates of orbital p -wave ($L = 1$) Cooper pairs formed from quasiparticles with zero total momentum ($+\mathbf{p}$, $-\mathbf{p}$) near the Fermi surface in spin-triplet ($S = 1$) states. In terms of the basis of triplet states the order parameter is given by

$$\Delta = \Delta_+(\hat{\mathbf{p}})|\uparrow\uparrow\rangle + \Delta_-(\hat{\mathbf{p}})|\downarrow\downarrow\rangle + \Delta_0(\hat{\mathbf{p}})\frac{1}{\sqrt{2}}|\uparrow\downarrow + \downarrow\uparrow\rangle, \quad (1)$$

where $\Delta_m(\hat{\mathbf{p}}) = \sum_{i=x,y,z} A_{mi}\hat{\mathbf{p}}_i$ for $m = 0, \pm 1$. There are

two bulk phases of superfluid ^3He in zero field. For a narrow temperature range near T_c at high pressures, $p > p_c = 21$ bar, ^3He condenses into the A phase with an order parameter of the form, $\Delta_+ = \Delta_- = 0$ and $\Delta_0 = \Delta(T) \times (\hat{\mathbf{p}}_x + i\hat{\mathbf{p}}_y)$. This phase exhibits antiferromagnetic spin correlations, and an orbital state that breaks time-inversion symmetry, i.e., a condensate of pairs with orbital angular momentum $+\hbar$. The B phase, which is the stable state over most of the phase diagram in zero magnetic field, is a superposition of all three triplet spin states and all three orbital states, with $\Delta_+ = \Delta(T)(\hat{\mathbf{p}}_x - i\hat{\mathbf{p}}_y)/\sqrt{2}$, $\Delta_- = \Delta(T)(\hat{\mathbf{p}}_x + i\hat{\mathbf{p}}_y)/\sqrt{2}$, $\Delta_0 = \Delta(T)\hat{\mathbf{p}}_z$. This state describes a condensate of spin-triplet, p -wave pairs in a state with total angular momentum $J = 0$. There is a continuous manifold of B -phase states related by a relative rotation of the spin and orbital coordinate axes. Surface and nuclear dipolar energies resolve most, but not all, of the degeneracy. In addition to the bulk A and B phases, the planar (P) phase is a possible ground state for thin films of ^3He . The P phase is a version of the B phase with $\Delta_0 = 0$. Alternatively, the P phase is an equal amplitude superposition of degenerate, time-reversed A -phase orbital states with opposite angular momenta. As a result the P phase is degenerate with the A phase in the weak-coupling BCS theory, but preserves time-inversion symmetry.

Here we consider ^3He films of uniform thickness, D , bound to a solid substrate. The liquid-vapor interface is assumed to be perfectly reflecting and atomically smooth. Thus, we consider $p \rightarrow 0$ bar. This is also the weak-coupling limit for superfluid ^3He , as indicated by the heat capacity jump $\Delta C/C_N \rightarrow 1.43$ for $p \rightarrow 0$ bar [11]. Substrates may provide scattering between specular and diffuse limits depending on the degree of roughness.

Scattering of ^3He quasiparticles off the free surface and substrate suppresses the orbital $\hat{\mathbf{p}}_z$ component of the order parameter (for either specular or diffuse scattering) in films less than about $1 \mu\text{m}$ thick. We choose the x and y axes in the plane of the film and the z axis perpendicular to the film, and use the boundary conditions described in Ref. [12]. In such thin films the A phase or the P phase is stable; in the weak-coupling limit these states are degenerate.

erate even with strong pairbreaking from diffuse scattering. In thin films the excitation spectrum is typically dominated by gapless excitations. The effect of this spectrum on strong-coupling energies, combined with relative importance of gradient energies for low-temperature, low-pressure thin films means that we cannot infer the relative stability of phases from what is known about strong-coupling energies in bulk ^3He .

Microscopic calculations show that as the film thickness increases surface scattering is unable to completely suppress the $\hat{\mathbf{p}}_z$ component of the order parameter. Results by several groups predict that equilibrium phase for film thickness, $D \gtrsim D_c(T) \sim 10\xi(T)$, is the deformed B phase described by the order parameter, $\mathbf{\Delta}_B = (\Delta_{\parallel}\hat{\mathbf{p}}_x, \Delta_{\parallel}\hat{\mathbf{p}}_y, \Delta_z\hat{\mathbf{p}}_z)$ [13], with $\Delta_z = \Delta_{\perp}(T)\sin(\pi z/D)$ [4,12,14]. The transition is first or second order at a critical film thickness, $D_c(T)$, depending on whether the low-temperature phase is the A phase or the P phase. The phase boundary, taken from our earlier calculation [12], is shown in the left panel of Fig. 1 in terms of the critical wave vector, $Q_z(T) = \pi/D_c(T)$. The inset emphasizes the reentrance ($A \leftarrow B \leftarrow A$) for $T \lesssim 0.42T_c$ near the critical line, which suggests that a lower energy state at low temperatures, in the vicinity of the critical line, may be achieved by an inhomogeneous phase that incorporates features of both phases. This is the case, but as we show below the structure of inhomogeneous

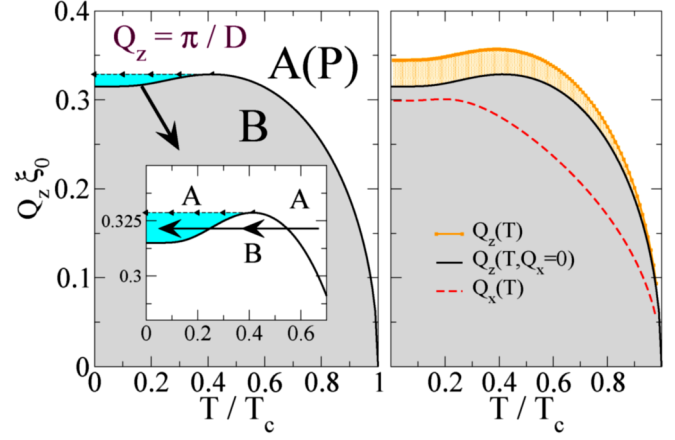


FIG. 1 (color online). Left: Q_z vs T phase diagram showing reentrance for homogeneous phases. Right: instability onsets for higher $Q_z(T)$ (orange line with boxes) for an inhomogeneous phase with in-plane wave vector $Q_x(T)$ (red dashed curve). Note that $\xi_0 = \hbar v_f / 2\pi T_c = 77$ nm at $p = 0$ bar.

phase is more complex than any of the homogeneous phases and evolves over a range of film thickness.

The transition from B phase to the P phase is second order on the critical line, $D_c(T)$. Thus, we first look for a second-order instability to an inhomogeneous phase that preempts the P - B transition. Our starting point is the weak-coupling gap equation,

$$\frac{1}{3} \ln\left(\frac{T}{T_c}\right) \mathbf{\Delta}^{(\pm)}(\hat{\mathbf{p}}, \mathbf{R}) = \int \frac{d\Omega_{\hat{\mathbf{p}}'}}{4\pi} (\hat{\mathbf{p}} \cdot \hat{\mathbf{p}}') T \sum_m \left[\mathbf{f}^{(\pm)}(\hat{\mathbf{p}}', \mathbf{R}; \varepsilon_m) - \pi \frac{\mathbf{\Delta}^{(\pm)}(\hat{\mathbf{p}}', \mathbf{R})}{|\varepsilon_m|} \right], \quad (2)$$

where $\mathbf{\Delta}^{(\pm)}(\hat{\mathbf{p}}, \mathbf{R})$ are the real (+) and imaginary (-) parts of the order parameter, and $\mathbf{f}^{(\pm)}(\hat{\mathbf{p}}, \mathbf{R}; \varepsilon_m) = [\mathbf{f}(\hat{\mathbf{p}}, \mathbf{R}; \varepsilon_m) \pm \mathbf{f}(\hat{\mathbf{p}}, \mathbf{R}; -\varepsilon_m)^*] / 2$ are the corresponding pair propagators in the Matsubara formulation for equilibrium Fermi superfluids [12,15]. These objects satisfy second-order mode equations with the order parameter providing source terms,

$$\frac{1}{4} (\mathbf{v}_f \cdot \nabla)^2 \mathbf{f}^{(+)} - \omega_m^2 \mathbf{f}^{(+)} = -\pi \omega_m [\mathbf{\Delta}^{(+)} + \mathbf{\delta}^{(+)}] + \frac{\pi}{\omega_m} [\mathbf{\Delta}^{(+)}(\mathbf{\Delta}^{(+)} \cdot \mathbf{\delta}^{(+)} + \mathbf{\Delta}^{(-)}(\mathbf{\Delta}^{(+)} \cdot \mathbf{\delta}^{(-)}) - \mathbf{\Delta}^{(-)} \times (\mathbf{\Delta}^{(-)} \times \mathbf{\delta}^{(+)} + \mathbf{\Delta}^{(+)} \times (\mathbf{\Delta}^{(-)} \times \mathbf{\delta}^{(-)})], \quad (3)$$

$$\frac{1}{4} (\mathbf{v}_f \cdot \nabla)^2 \mathbf{f}^{(-)} - \omega_m^2 \mathbf{f}^{(-)} = -\pi \omega_m [\mathbf{\Delta}^{(-)} + \mathbf{\delta}^{(-)}] + \frac{\pi}{\omega_m} [\mathbf{\Delta}^{(-)}(\mathbf{\Delta}^{(-)} \cdot \mathbf{\delta}^{(-)} + \mathbf{\Delta}^{(+)}(\mathbf{\Delta}^{(-)} \cdot \mathbf{\delta}^{(+)} - \mathbf{\Delta}^{(+)} \times (\mathbf{\Delta}^{(+)} \times \mathbf{\delta}^{(-)} + \mathbf{\Delta}^{(-)} \times (\mathbf{\Delta}^{(+)} \times \mathbf{\delta}^{(+)})]. \quad (4)$$

Note that $\mathbf{v}_f = v_f \hat{\mathbf{p}}$ is the Fermi velocity, $\varepsilon_m = (2m+1)\pi T$ is the Matsubara energy, and $\omega_m^2 \equiv \varepsilon_m^2 + |\mathbf{\Delta}^{(+)}|^2 + |\mathbf{\Delta}^{(-)}|^2$, where $\mathbf{\Delta}^{(\pm)}$ is the order parameter of the unperturbed, translationally invariant phase. Lastly, $\mathbf{\delta}^{(\pm)}$ is the first-order correction we seek to find. These equations are valid up to first order in order parameter corrections, and in their derivation we assumed that the gradient terms of $\mathbf{f}^{(\pm)}$ in strongly confined space are of the same order as $\mathbf{f}^{(\pm)}$ themselves.

For the P state we can fix the overall phase so that $\mathbf{\Delta}$ is real. We then have $\mathbf{\Delta}^{(-)} = 0$ and $\mathbf{\Delta}^{(+)} = \Delta_{\parallel}(z)(\hat{\mathbf{p}}_x, \hat{\mathbf{p}}_y, 0)$. The instability to an inhomogeneous phase is then a single-

mode instability for pairs with zero spin projection along \mathbf{z} [16]. The eigenfunction for the instability has the form

$$\delta_z(\hat{\mathbf{p}}, \mathbf{R}) = \sum_{j=x,y,z} a_{z,j}(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{R}} \hat{\mathbf{p}}_j. \quad (5)$$

For a single-mode instability in the plane of the film we choose $\mathbf{Q} = (Q_x, 0, Q_z)$. The resulting solution for the Fourier component of the $S_z = 0$ pair propagator is

$$f_z^{(+)}(\hat{\mathbf{p}}, \mathbf{Q}) = \pi \omega_m(\hat{\mathbf{p}}) \frac{\sum_i a_{z,i}(\mathbf{Q}) \hat{\mathbf{p}}_i}{\frac{1}{4} (\mathbf{v}_f \cdot \mathbf{Q})^2 + \omega_m^2(\hat{\mathbf{p}})}. \quad (6)$$

The boundary conditions on the pair propagator at the free surface ($z = D$) and substrate ($z = 0$) yield a set of eigenvector equations for the unstable mode. For specular scattering by the substrate,

$$\mathbf{f}^{(\pm)}(x, z = 0, \hat{\mathbf{p}}) = \mathbf{f}^{(\pm)}(x, z = 0, \hat{\mathbf{p}}) \quad (7)$$

for any x and $\hat{\mathbf{p}}$, with $\hat{\mathbf{p}} = \hat{\mathbf{p}} - 2\hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \hat{\mathbf{p}})$, and similarly for the free surface. These boundary conditions reduce to connections between the Fourier components of the order parameter, $a_{z,x}(Q_x, -Q_z) = +a_{z,x}(Q_x, Q_z)$, $a_{z,y}(Q_x, -Q_z) = +a_{z,y}(Q_x, Q_z)$, $a_{z,z}(Q_x, -Q_z) = -a_{z,z}(Q_x, Q_z)$, and fixes the wave vector $Q_z = \pi/D$ in terms of the film thickness D at the instability. These results and the gap equation generate the eigenvalue equations for the mode amplitudes, $a_{z,i}(\mathbf{Q})$,

$$\ln(T/T_c)a_{z,i} - \sum_{j=x,y,z} I_{ij}a_{z,j} = 0, \quad i = x, y, z, \quad (8)$$

$$I_{ij} = 6\pi T \sum_{m=0}^{\infty} \int \frac{d\Omega \hat{\mathbf{p}}}{4\pi} \hat{\mathbf{p}}_i \hat{\mathbf{p}}_j \left(\frac{\omega_m}{\frac{1}{4}(\mathbf{v}_f \cdot \mathbf{Q})^2 + \omega_m^2} - \frac{1}{\epsilon_m} \right).$$

Translational symmetry is unbroken along the y axis in which case $I_{xy} = I_{yz} = 0$. The mode amplitudes separate into linearly independent blocks: a 2D ($a_{z,x}, a_{z,z}$) and a 1D ($a_{z,y}$) block. A nontrivial solution to Eq. (8) exists if $[\ln(T/T_c) - I_{yy}] = 0$ or $[\ln(T/T_c) - I_{xx}][\ln(T/T_c) - I_{zz}] - I_{xz}^2 = 0$. The eigenvalue equation for the 1D mode amplitude has a maximum unstable wave vector only for the transition to the homogeneous phase, $Q_z = \pi/D_c(T)$, $Q_x = 0$. However, the eigenvalue equation for the 2D block gives an unstable mode $Q_z(Q_x, T)$ that preempts the homogeneous transition. The maximum value of $Q_z(Q_x, T)$ as a function of Q_x for each temperature determines the *lower* critical film thickness, $D_{c2}(T) < D_c(T)$, for the transition to an inhomogeneous film with broken translational symmetry in the plane of the film. The critical wave vector, $Q_z(T)$, and the locus of values of $Q_x(T)$ are shown in the right panel of Fig. 1.

The key signature of spontaneously broken translational symmetry in the xy plane is the appearance of the order parameter amplitudes, $a_{z,x}(Q_x, Q_z) \exp(iQ_x x) \cos(Q_z z)$ and $a_{z,z}(Q_x, Q_z) \exp(iQ_x x) \sin(Q_z z)$. These amplitudes are shown in the left panel of Fig. 2 for $T = 0.5T_c$ and $D = 9.3\xi_0 \lesssim D_c(T)$. Note that the full solution for the order parameter above the lower critical thickness also shows very small oscillatory amplitudes for the in-plane spin components, e.g., $a_{x,z}$.

All modes with in-plane unstable wave vectors \mathbf{Q}_{xy} such that $|\mathbf{Q}_{xy}| = Q_x$ are degenerate. In the absence of an external bias to select the direction of the unstable mode, the instability may propagate in any direction in the plane of the film. For $D > D_{c2}$ the spatial structure of the order parameter that is realized is determined from the minimum free energy. This phase may exhibit one-dimensional, stripe-phase order, or possibly a two-dimensional structure defined by two noncollinear wave vectors, e.g., a triangular

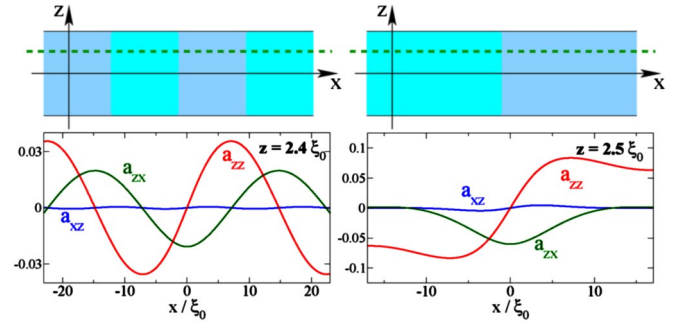


FIG. 2 (color online). Order parameter (in units of $2\pi T_c$) for the stripe phase at $T = 0.5T_c$ along the film for $z \approx 2.5\xi_0$. Left: $D_{c2} < D = 9.3\xi_0 \lesssim D_c$. Right: $D_c < D = 10\xi_0 < D_{c1}$.

lattice. A comparison of the possible minimum energy configurations of the inhomogeneous phase has not been carried out. Here we focus on the structure of the one-dimensional stripe phase.

The broken symmetry phase persists for film thickness well above the original critical line, $D_c(T)$, for the homogeneous A - B transition. However, for $D > D_{c2}(T)$ the gap equation includes nonlinear driving terms that couple modes with different wave vectors. The ground state is periodic, but the structure is nonsinusoidal. The right panel of Fig. 2 shows the order parameter amplitudes for a film with $D = 10\xi_0 > D_c(T)$. The basic structure of this phase is indicated by the amplitude $a_{z,z}$, which has developed a solitonlike structure separating “domains” of degenerate B -like phases: e.g., $\Delta_B^< = (\Delta_{\parallel}\hat{\mathbf{p}}_x, \Delta_{\parallel}\hat{\mathbf{p}}_y, -\Delta_{\perp}\hat{\mathbf{p}}_z)$ and $\Delta_B^> = (\Delta_{\parallel}\hat{\mathbf{p}}_x, \Delta_{\parallel}\hat{\mathbf{p}}_y, +\Delta_{\perp}\hat{\mathbf{p}}_z)$. Also, centered on the soliton is a non- B -like phase, represented by $a_{z,x}$, bound to the domain wall.

This basic structure also provides a clue to the underlying mechanism stabilizing the inhomogeneous phase; it is the competition between the energy associated with surface pairbreaking and the energy cost of a domain wall separating two degenerate B -like phases [17]. Consider the two trajectories (labeled 1 and 2) shown in Fig. 3. The left panel shows a homogenous B phase, while the right panel shows two degenerate B -like phases corresponding to amplitudes $-\Delta_{\perp}$ left of a domain wall and $+\Delta_{\perp}$ to the right.

For the trajectory 1 that reflects from the free surface we have $p_z \rightarrow -p_z$. This sign change is the origin of surface

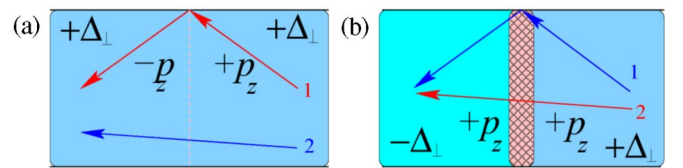


FIG. 3 (color online). (a) Surface reflection (trajectory 1) with $p_z \rightarrow -p_z$ leads to strong pairbreaking for the homogenous B phase. (b) Trajectory 2 is the strong pairbreaking trajectory because of the sign change $\Delta_{\perp} \rightarrow -\Delta_{\perp}$.

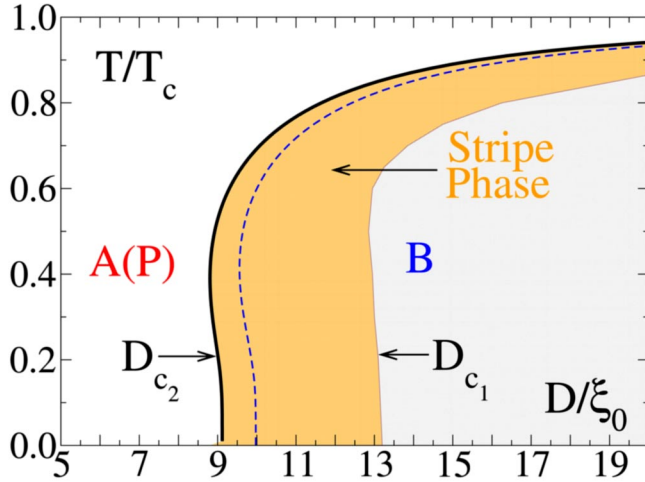


FIG. 4 (color online). $T - D$ -phase diagram for superfluid ^3He films in the weak-coupling limit ($p = 0$ bar). The phase with crystalline order separates translationally invariant $A(P)$ and B phases.

pairbreaking; it leads to the suppression of Δ_{\perp} and the formation of surface Andreev bound states [12]. The energy cost is directly related to the spectrum of surface states. By contrast the shallow trajectory which passes through the dashed plane without intersecting a surface encounters a nearly uniform order parameter resulting in little or no pairbreaking.

Surface pairbreaking can be suppressed locally by compensating the sign change from surface reflection. In particular, for the domain-wall configuration [Fig. 3(b)] the sign change for the scattering trajectory ($p_z \rightarrow -p_z$) is compensated by the sign change associated with the degenerate states on opposite sides of the domain wall ($\pm \Delta_{\perp}$). Thus, over a few coherence lengths near the domain wall surface reflection does not lead to strong pairbreaking, and correspondingly the energy cost of surface scattering is reduced. However, it is not all “savings.” There is an energy cost for the domain wall. The shallow trajectory crossing the domain wall now incurs a sign change. Pairbreaking occurs near the domain wall and a spectrum of Andreev bound states forms on the interface.

For very thick films [$D \gg D_c(T)$] the translationally invariant B phase is favored because the surface pairbreaking energy is small compared with the pairbreaking cost of a domain wall. But, for sufficiently thin films the domain-wall energy is less than the surface pairbreaking energy and the broken symmetry phase is favored. The critical line where one domain wall is favored over the uniform B phase is $D_{c1}(T)$.

For $D < D_{c1}$ multiple domains are favored. Further reduction in the film thickness favors more domain walls until they dissolve into the P phase at the $D_{c2}(T)$, or a first-order transition to the A phase occurs [18]. Numerical calculations of the free energy based on the formalism described in Ref. [12] give a range of film thicknesses of

order $0.75 \mu\text{m} \leq D \leq 1.0 \mu\text{m}$ for $T \leq 0.75T_c$ for an energetically favored stripe phase. The approximate critical line obtained from these calculations is labeled $D_{c1}(T)$ in the T versus D phase diagram shown in Fig. 4.

In conclusion, our calculations predict that films of superfluid ^3He should exhibit an inhomogeneous phase with spontaneously broken translational symmetry in the plane of the film. This phase has no analog in bulk ^3He , and should be identifiable by its anisotropic transport properties, e.g., reduced heat conductivity normal to the stripes. Signatures of the inhomogeneous phase should also be observable as a broadening of the NMR linewidth.

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