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Superfluidity and Dimerization in a Multilayered System of Fermionic Polar Molecules

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We consider a layered system of fermionic molecules with permanent dipole moments aligned perpendicular to the layers by an external field. The dipole interactions between fermions in adjacent layers are attractive and induce interlayer pairing. Because of the competition for pairing among adjacent layers, the mean-field ground state of the layered system is a dimerized superfluid, with pairing only between every other layer. We construct an effective Ising-XY lattice model that describes the interplay between dimerization and superfluid phase fluctuations. In addition to the dimerized superfluid ground state, and high-temperature normal state, at intermediate temperature, we find an unusual dimerized “pseudogap” state with only short-range phase coherence. We propose light-scattering experiments to detect dimerization.

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The long-range and anisotropic nature of dipole-dipole interactions offers new opportunities for ultracold polar molecules, beyond what is possible for cold-atom systems with only short-range, isotropic contact interactions [1]. A variety of exotic many-body states, including $p_x + ip_y$ fermionic superfluids [2] and nematic non-Fermi liquids [3], are predicted to occur in cold dipolar systems. Additionally, polar molecules could provide a robust toolbox for engineering novel lattice-spin Hamiltonians [4] or hybrid devices for quantum information processing [5]. Recent progress towards trapping and cooling atoms and molecules with permanent electric or magnetic dipole moments has opened the door to exploring these exotic states of matter experimentally [6]. In order to prevent the system from collapsing due to the attractive head-to-tail part of the dipolar interaction [7], it has been proposed [8,9] to create stacks of dipolar particles confined to a set of parallel planes.

In this Letter, we consider a stack of two-dimensional layers of polar fermions whose dipole moments \vec{D} are aligned along the stacking direction (z axis) by an external field (see Fig. 1). The dipole interaction $V_d = \frac{D^2}{r^3}(1 - 3\frac{z^2}{r^2})$ is purely repulsive between fermions in the same layer, and partially attractive (for $r < \sqrt{3}z$) between fermions in different layers. The attractive interlayer component of the dipole interaction induces BCS pairing between layers, with adjacent layers competing for pairing. We demonstrate that competition between adjacent layers favors dimerization, with pairing only between even or odd pairs of layers (Fig. 1).

We find three distinct phases: a high-temperature disordered phase, a fully ordered phase characterized by a dimerized pairing amplitude and a quasi-long-range ordered (QLRO) pairing phase in each layer (Fig. 1), and a dimerized “pseudogap” phase with only short-range superfluid correlations. The latter phase is particularly

interesting, since it can only be characterized by a composite *four-fermion* dimerization order parameter. Therefore, this phase does not admit a mean-field (Hartree-Fock) description. This is analogous to spin nematics [10] and charge $4e$ superconductors [11], which are both phases of strongly interacting fermions that can only be characterized by composite order parameters.

Fermionic pairing in a layered system.—The action for an N -layer system in terms of fermionic fields ψ is

$$S = \sum_{z=1}^N \sum_{\mathbf{k}} \psi_{z,\mathbf{k}}^\dagger (\partial_\tau + \epsilon_{\mathbf{k}} - \mu) \psi_{z,\mathbf{k}} - \sum_{z,z'=1}^N \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} \psi_{z,\mathbf{k}'}^\dagger \psi_{z',\mathbf{q}-\mathbf{k}'}^\dagger V_{|\mathbf{k}-\mathbf{k}'|}^{(z,z')} \psi_{z',\mathbf{q}-\mathbf{k}} \psi_{z,\mathbf{k}}, \quad (1)$$

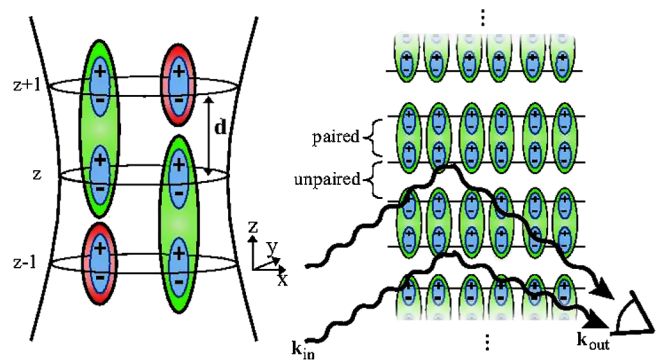


FIG. 1 (color online). Schematic representation of the competition for pairing among adjacent pairs of layers, including the depiction of the optical confinement beam which creates the stack of 2D sheets (left diagram) and the illustration of one of the two equivalent dimerized pairing ground states for a many-layered system (right diagram). The wavy lines illustrate the proposed light-scattering detection scheme discussed below in the text.

where z and z' are (integer) layer labels, and $\psi_{z,\mathbf{k}}^\dagger(\tau)$ creates a fermion with in-plane momentum \mathbf{k} and imaginary time τ in layer z . (The τ labels have been suppressed above.) $V_q^{(z,z')}$ is the dipolar interaction between layers z and z' , Fourier transformed with respect to the in-plane separation: for example, $V_q^{(z,z\pm 1)} = -D^2 q e^{-qd}$.

By solving the BCS gap equation, $\Delta_{z,\mathbf{k}} = -\sum_{\mathbf{k}'} \times V_{\mathbf{k}-\mathbf{k}'}^{(z,z+1)} \langle \psi_{z+1,-\mathbf{k}'} \psi_{z,\mathbf{k}'} \rangle$, we find that the attractive inter-layer interactions induce fermionic pairing between adjacent layers z and $z \pm 1$ (see Ref. [12] for details). The interaction between next-nearest layers and beyond is small, and will be neglected throughout most of this Letter. To decouple the four-fermion interaction term, we introduce Hubbard-Stratonovich (H-S) fields $\Delta_z(\mathbf{r}_\perp)$ associated with the pairing order parameters (where \mathbf{r}_\perp is the in-plane coordinate), and integrate out the fermionic degrees of freedom [12]. Expanding the resulting fermionic determinant to quartic order (valid in the vicinity of the phase transition where $|\Delta|$ is small) yields the following Ginzburg-Landau (GL) free energy:

$$F = \sum_z \int d^2r (\kappa |\nabla_\perp \Delta_z|^2 + r |\Delta_z|^2 + u |\Delta_z|^4 + 2u |\Delta_z|^2 |\Delta_{z+1}|^2), \quad (2)$$

where ∇_\perp denotes the gradient restricted to the xy plane. The GL coefficients are given by $\kappa = \frac{7\zeta(3)}{32\pi^3} \frac{\varepsilon_F}{T^2}$, $r = \nu t$, and $u = \frac{1.7}{32} \frac{\nu}{T^3}$, where ζ is the Riemann zeta function, $t = (T - T_c)/T$ is the reduced temperature, ε_F is the Fermi energy, and ν is the two-dimensional density of states (for details we refer the reader to the Ref. [12]).

An important feature of this free energy is that the H-S expansion does not generate $|\partial_z \Delta|^2$ terms, but only terms of the form $|\partial_z \Delta|^2$. The absence of $|\partial_z \Delta|^2$ terms is not an artifact of the H-S expansion; rather, it is guaranteed by particle number conservation for each layer individually. Particle conservation for each layer stems from the absence of interlayer tunneling, and formally corresponds to N_{Layers} independent $U(1)$ phase rotation symmetries, $\psi_z \rightarrow e^{i\theta_z/2} \psi_z$, of fermion fields ψ_z in layer z . In contrast to other quasi-two-dimensional systems, such as superconducting thin films where the behavior of the system tends towards three dimensional as the film thickness is increased, two-dimensional Berezinskii-Kosterlitz-Thouless (BKT) physics remains important even for a large number of layers.

Mean-field ground state.—The $2u|\Delta_i|^2|\Delta_{i+1}|^2$ term in (2) indicates that adjacent pairs of layers compete with each other for pairing. For $N_{\text{Layers}} > 3$, the mean-field theory predicts that it is energetically favorable for the system to spontaneously dimerize into one of two equivalent configurations, where Δ vanishes between every other layer: $|\Delta_j| = \frac{1}{2}[1 \pm (-1)^j] \Delta_0$ (see Fig. 1). The situation for $N_{\text{Layers}} = 3$ is more subtle, and we defer its discussion.

Effective lattice model for many-layer systems.—The above mean-field analysis suggests that the relevant

degrees of freedom for a many-layer dipolar system are Ising-like dimerization between even or odd layers, and two-dimensional XY -like phase fluctuations of the inter-layer pairing order parameters. In order to describe phase transitions in this system, we coarse-grain the GL theory (in-plane) over length scales below the GL coherence length $\xi_{\text{GL}} \equiv (\kappa/|r|)^{1/2}$, and obtain the following effective lattice model [13],

$$F = \sum_z \left\{ K_z \sum_i \sigma_{z,i} \sigma_{z+1,i} - K_\perp \sum_{\langle ij \rangle} \sigma_{z,i} \sigma_{z,j} - \sum_{\langle ij \rangle} J(\sigma_{z,i}, \sigma_{z,j}) \times [\cos(\theta_{z,i} - \theta_{z,j}) - 1] \right\}, \quad (3)$$

of Ising variables $\sigma_{i,z} \in \{\pm 1\}$ coupled to XY phase variables $\theta_{z,i} = \arg \Delta_z(\vec{r}_i) \in [0, 2\pi]$, where z labels physical layers, i labels lattice sites in the xy plane, and $J(\sigma_{z,i}, \sigma_{z,j}) \equiv J_0(1 + \sigma_{z,i})(1 + \sigma_{z,j})/4$.

In the lattice model, $\sigma_z = +1$ ($\sigma_z = -1$) indicates that layers z and $z + 1$ are paired (unpaired, respectively). The uniformly dimerized ground state of the multilayer system corresponds to antiferromagnetic Ising order along the z axis and ferromagnetic order within the xy plane. The Ising domain walls (DW) correspond to regions where pairing switches between the two equivalent dimerization configurations over a distance of the order of the GL coherence length, either along the z axis or within the xy plane. The coupling constants K_z and K_\perp reflect the energy cost of deforming the magnitude of the pairing order parameter, $|\Delta|$, to form a domain wall along the z axis or in the xy plane, respectively (see Fig. 2).

The coupling $J(\sigma_{z,i}, \sigma_{z,j})$ corresponds to the average superfluid stiffness $\rho \sim \kappa |\Delta|^2$ in the vicinity of the lattice site (i, z) and determines the energy cost of twisting the phase of the order parameter $\theta_{z,i}$ between sites i and j in the same plane. The local stiffness is nonzero wherever $\sigma_{z,i} = +1$, and zero otherwise [12].

The lattice-model couplings (K_z , K_\perp , J_0) can be estimated from the GL model. An in-plane dimerization

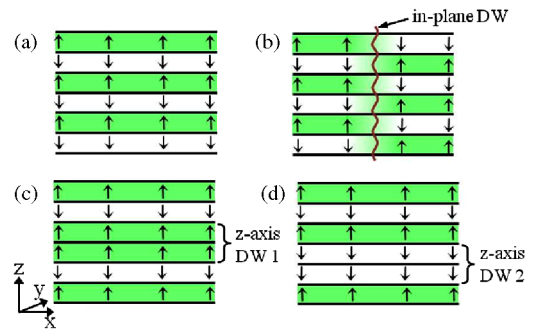


FIG. 2 (color online). Schematic depiction of the fully dimerized phase (a), in-plane Ising DW (b), the z -axis Ising DW with pairing between two adjacent pairs of layers (c), and the z -axis Ising DW with no pairing for two adjacent pairs of layers (d). Green shading between layers indicates pairing.

domain wall along the x direction [Fig. 2(b)] corresponds to pairing configurations of the form $\Delta_z(x) = \frac{\Delta_0}{2}[1 + (-1)^z \alpha(x)]$, where $\Delta_0^2 = \frac{|r|}{2u}$ and $\alpha(x)$ is a function that changes from -1 to $+1$ around $x = 0$ and tends to a constant away from $x = 0$. Minimization of the free energy with respect to $\alpha(x)$ yields $\alpha(x) = \tanh(2x/\ell_{\text{DW}})$, where $\ell_{\text{DW}} \equiv \sqrt{\frac{32\kappa}{3r}}$ [12]. The corresponding free energy cost per unit length is $U_{\text{DW}}^{(\perp)} = \ell_{\text{DW}} \frac{|r|^2}{8u}$.

There are two possible z -axis domain-wall configurations, shown in Figs. 2(c) and 2(d). To determine their free energy cost, we consider a system with periodic boundary conditions along the z axis, and compare the free energy of the ground state to that of the domain-wall configurations. This yields an energy cost per unit area $U_{\text{DW}}^{(z)} = \frac{|r|^2}{8u}$ for both types of domain walls. Setting the lattice spacing equal to ℓ_{DW} , the energetics of in-plane and z -axis domain walls are reproduced by $K_z = 2K_{\perp} = \frac{4\kappa|r|}{3u}$. In order to determine the lattice phase stiffness J_0 , we equate the cost of an infinitesimal phase twist, $\theta_{z,j} = \theta_{z,i} + \delta\theta$, in a fully paired layer ($\sigma_z = 1$) to the corresponding cost in the GL free energy [Eq. (2)]. This gives $J_0 = \frac{\kappa|r|}{u}$.

The lattice model [Eq. (3)] describes three-dimensional Ising spins coupled to many independent two-dimensional XY layers. For temperatures near or below the Ising transition temperature, the Ising variables have large correlation lengths and hence see an average over many independent layers of XY spins. With this self-averaging property in mind, we decouple the XY and Ising variables in a mean-field factorization,

$$F_{\sigma} = K_z \sum_{\langle zz' \rangle, i} \sigma_{zi} \sigma_{z'i} - K_{\perp}^{(\text{eff})} \sum_{z, \langle ij \rangle} \sigma_{zi} \sigma_{zj} - h \sum_{z, i} \sigma_{zi}$$

$$F_{XY} = - \sum_{\langle ij \rangle} \frac{J_0}{4} [1 + (-1)^z \sigma_0]^2 \cos(\theta_{z,i} - \theta_{z,j}),$$

where $K_{\perp}^{(\text{eff})} = K_{\perp} + \frac{J_0}{4} \frac{(A+B)}{2}$ and $A, B \equiv \langle \cos(\theta_i^{(e/o)} - \theta_j^{(e/o)}) \rangle_{F_{XY}} - 1$ are the averages (with respect to F_{XY}) of the cosine terms in even and odd layers, respectively, $\sigma_0 \equiv \langle \sigma \rangle_{F_{\sigma}}$, and $h = \frac{(A-B)}{2} \frac{J_0}{2}$.

The decoupled Ising model and XY models can then be analyzed separately but self-consistently. A mean-field analysis is adequate for the 3D Ising model. The phase action is treated by a variational self-consistent harmonic approximation (SCHA) [14]. While the SCHA provides a reasonable estimate of the location of the 2D BKT transition, it spuriously predicts a strong first-order transition in which $\langle F_{XY} \rangle_{\text{SCHA}}$ drops abruptly to zero at the XY transition temperature, T_{XY} . At higher temperatures, the SCHA dramatically underestimates the contribution to the energy density from phase fluctuations. In order to avoid this undesirable feature, we supplement the SCHA value for $\langle \cos \Delta_{ij} \theta \rangle_{\text{SCHA}}$ with a high-temperature expansion for $T > T_{XY}$:

$$\langle \cos \Delta_{ij} \theta^{(z)} \rangle = \begin{cases} \langle \cos \Delta_{ij} \theta^{(z)} \rangle_{\text{SCHA}} & T < T_{XY} \\ J(\sigma_0, \sigma_0)/2T & T > T_{XY}. \end{cases} \quad (4)$$

Figure 3 shows the phase diagram predicted by the effective lattice model. The main figure displays the phase diagram where the model parameters are taken from the GL coefficients in (3). The BCS transition temperature T_c^{BCS} is obtained by solving numerically the BCS gap equation for the dipole potential. Whereas the dimerization transition occurs close to the mean-field BCS transition temperature T_c^{BCS} , the BKT transition to the QLRO phase occurs at a lower temperature, leaving an intermediate region with full dimerization but only short-range superfluid correlations.

Recent experiments on 3D clouds of ultracold $^{40}\text{K}^{87}\text{Rb}$ molecules have achieved densities on the order of $n_{3d} = 10^{12} \text{ cm}^{-3}$ and permanent electrical dipole moments of up to 0.566 Debye [6]. If similar densities were achieved in a layered system with layer spacing on the order of 400 nm, the ratio of typical dipole interactions to Fermi energy would be $D^2/(4\pi\epsilon_0 d^3 \epsilon_F) \sim 3$.

While the GL parameters in Eq. (2) provide an initial estimate of the lattice-model coupling constants, in principle, the model coefficients can be renormalized by higher order terms in the GL expansion. The inset shows the phase diagram for generic values of the model parameters K_{\perp} and J_0 with $K_z/K_{\perp} = 2$ (the qualitative features do not depend sensitively on this ratio). An additional feature emerges for generic coefficients: for J sufficiently bigger than K , there is a tricritical point where the BKT and Ising transitions fuse into a weakly first-order phase transition.

Order parameter and detection.—The dimerized phase breaks translational symmetry in the z direction. It can be characterized by the following *four-fermion* order

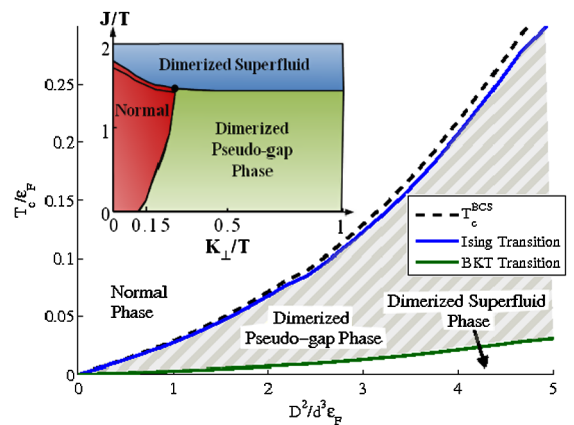


FIG. 3 (color online). The lattice-model phase diagram, calculated using the temperature dependence of the GL coefficients in (3) and plotted in terms of the dipole-interaction strength D^2/d^3 and temperature T , each measured in units of ϵ_F (main figure). The inset shows the phase diagram predicted by the effective lattice model for generic model parameters, with $K_z/K_{\perp} = 2$. The double line indicates the first-order transition.

parameter: $\mathcal{D} = \langle n_{z-1,r} n_{z,r} - n_{z,r} n_{z+1,r} \rangle$, where $n_{z,r} = \psi_{z,r}^\dagger \psi_{z,r}$ is the local fermion density. For finite transverse confinement, in the dimerized phase, every two paired layers shift slightly towards each other. The displacement scales as $\delta z \propto \Omega_z^{-2}$, where Ω_z is the layer-confinement frequency in the z direction. The dimerized phase can be detected by the appearance of new Bragg peaks in elastic light scattering (see Fig. 1) with wave vector $\mathbf{Q} = n\pi\hat{\mathbf{z}}/d$, $n = 1, 3, \dots$, with intensity $\sim \delta z^2$.

In the strong-confinement limit, $\Omega_z \rightarrow \infty$, the particle density does not show any sign of dimerization. However, in this regime, the dimerized phase could still be detected by measuring correlations between the amplitudes of light scattered at different wave vectors: $\langle n_{\mathbf{q}} n_{\mathbf{q}'} \rangle \propto n_0^2 \delta_{\mathbf{q}+\mathbf{q}'} + \delta_{\mathbf{Q}-\mathbf{q}-\mathbf{q}'} \mathcal{D}$, where \mathbf{q} and \mathbf{q}' are two scattering wave vectors and n_0 is a constant.

Three-layer case.—The three-layer system is a special case that requires more careful analysis. If one proceeds as above and includes interactions only between neighboring layers, the system possesses an extra $SU(2)$ symmetry generated by $I^z = \int d^2r (\psi_3^\dagger \psi_3 - \psi_1^\dagger \psi_1)$ and $I^\pm = \int d^2r (\psi_3^\dagger \psi_1 \pm i \psi_1^\dagger \psi_3)$. The $U(1)$ generator $N_2 = \int d^2r \psi_2^\dagger \psi_2$ completes the $SU(2)$ symmetry to $U(2)$. These generators commute with $\mathcal{H} = \mathcal{H}_{\text{kin}} + V_{12} + V_{23}$, where V_{ij} is the interaction between layers i and j . This $U(2)$ symmetry dictates that, to all orders in the GL expansion, the free energy should be a function of $(|\Delta_1|^2 + |\Delta_2|^2)$ only, which does not energetically distinguish dimerization from uniform pairing.

However, intralayer and next-nearest neighbor interactions $\tilde{V} = V_{13} + \sum_{j=1}^3 V_{jj}$ break the $SU(2)$ symmetry of the three-layer system, and generate a quartic term of the form $-|v| |\Delta_1|^2 |\Delta_2|^2$ in the GL free energy. This term is relevant [15] (in the renormalization group sense), and hence, we expect the trilayer system to exhibit uniform pairing with $|\Delta_1| = |\Delta_2|$. In contrast, for $N_{\text{Layers}} > 3$, already the dominant nearest neighbor interactions strongly favor dimerization, and \tilde{V} only produces small subleading corrections.

Discussion.—We expect that the Ising-XY model description of the layered dipolar fermions will be insufficient deep in the BEC regime, where interaction energies are dominant compared to the Fermi energy. For sufficiently strong interactions or sufficiently dense systems, the system will form a Wigner crystal [16]. Another possibility is that the formation of longer chains of three or more dipoles may become important [9]. In a regime where chains of n dipoles are favorable, a many-layered system would undergo n -merization rather than dimerization. Correspondingly, an n -merized phase may undergo an n -state clock-model-type phase transition which generalizes the Ising-type dimerization transition considered above. Furthermore, for even n , bosonic chains could condense into an exotic superfluid of dipolar chains. Such states offer an intriguing chance to examine the

relatively unexplored boundary between few-body interactions and many-body phase transitions, and they deserve further study.

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