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Fulde-Ferrell-Larkin-Ovchinnikov state in bilayer dipolar systems

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We study the phase diagram of fermionic polar molecules in a bilayer system, with an imbalance of molecular densities of the layers. For the imbalance exceeding a critical value, the system undergoes a transition from the uniform interlayer superfluid to the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state with mostly a stripe structure, and at sufficiently large imbalance a transition from the FFLO to normal phase. Compared to the case of contact interactions, the FFLO regime is enhanced by the long-range character of the interlayer dipolar interaction, which can combine the s-wave and p-wave pairing in the order parameter.

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Introduction. Exotic many-body quantum states in population imbalanced spin-1/2 Fermi systems attract a great deal of interest, to a large extent due to expected nonconventional transport properties. Among these states, the most actively studied is the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase [1,2], in which Cooper pairs have finite momenta and the order parameter shows a lattice structure on top of a uniform background. Theoretical studies of the FFLO phase in condensed matter and in particle physics are lasting for decades [3–9]. The rapid developments in the field of ultracold quantum gases stimulated the studies of this phase in two-component imbalanced Fermi gases [10–19]. However, experimental verification of the existence of the FFLO state is still in progress [13,20–25]. In ultracold gases the search for the FFLO phase is actively pursued for strongly interacting fermions, where one has a crossover from Bardeen-Cooper-Shrieffer (BCS) superfluid to Bose-Einstein condensate of weakly bound molecules [22–24,26–28], and the population imbalance is expected to lead to the FFLO state [14–16].

Recent advances in creating ultracold polar molecules [29,30] interacting with each other via long-range anisotropic dipole-dipole forces open fascinating prospects for many-body physics [31,32]. A variety of novel many-body states was proposed for fermionic dipoles in two dimensions (2D) [33–48], including interlayer superfluids with the BCS-BEC crossover in a bilayer geometry [36,41,43]. Importantly, in 2D the decay of polar molecules due to ultracold chemical reactions [49,50] can be suppressed by orienting the dipoles perpendicularly to the plane, which induces a strong intermolecular repulsion [51–53]. Together with possible experiments with nonreactive polar molecules [54,55], this forms a promising path toward new many-body quantum states.

In this Rapid Communication we predict wide possibilities for creating the FFLO phase of polar molecules in a bilayer geometry, with a finite imbalance of molecular densities of the layers. Cooper pairs are formed by molecules belonging to different layers due to the interlayer dipole-dipole interaction, and the most favored is the FFLO state with a stripe structure of the order parameter. Remarkably, the FFLO regime of this interlayer superfluid is enhanced by the long-range character of the dipolar interaction, which in an imbalanced system may lead to Cooper pairs representing superpositions of contributions of various partial waves. Our work thus opens another direction to investigate novel superfluids of fermionic particles with population imbalance.

Interlayer interaction and order parameters. We consider identical fermionic polar molecules in a bilayer geometry, oriented perpendicularly to the layers by an electric field (see Fig. 1). The interlayer dipole-dipole interaction is partially attractive and it may lead to interlayer superfluid pairing [36,37,41,43], whereas the inelastic decay is suppressed like in a single layer [34]. The inlayer interaction is purely repulsive and it only renormalizes the chemical potential in the regime that we consider [36,43], so that below we omit this interaction. We assume that molecular densities in the layers are different from each other, but in each layer the Fermi energy greatly exceeds the binding energy of interlayer dimers (bound states of dipoles belonging to different layers [36,41,56]). Thus, the only effect of the interlayer dipole-dipole interaction will be the fermionic superfluid pairing, which we consider in the weakly interacting BCS regime.

The interlayer dipole-dipole interaction potential and its Fourier transform are given by

$$V(\mathbf{r}) = d^2(r^2 - 2\lambda^2)/(r^2+\lambda^2)^{5/2}, \tag{1}$$

$$V_{\mathbf{k}\mathbf{k}'} = \int d\mathbf{r} V(\mathbf{r}) e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}} = -2\pi d^2 \kappa e^{-\kappa\lambda}, \tag{2}$$

where $\kappa \equiv |\mathbf{k}-\mathbf{k}'|$, d is the effective dipole moment of a molecule, λ is the distance between the two layers, and $\mathbf{r}$ is the in-plane separation between two dipoles. The pairing potential can be expanded in a series in angular momenta $V_{\mathbf{k}\mathbf{k}'} = \sum_l V_l(k,k') e^{i(\phi-\phi')l}$. The leading part of the scattering

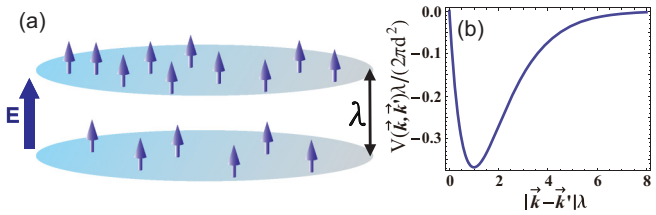

FIG. 1. (a) The bilayer system with population imbalance of polar molecules (see text). (b) The interlayer dipolar interaction in the k space in units of $2\pi d^2/\lambda$ versus $|\mathbf{k}-\mathbf{k}'|$, as shown in Eq. (2).

(interaction) amplitude can be obtained in the Born approximation. It is important that in contrast to contact interactions, the dipolar (interlayer) interaction amplitude with $l > 0$, and especially the p-wave amplitude, can be comparable with the s-wave amplitude.

After omitting the inlayer interaction the system maps onto spin-1/2 fermions with intercomponent dipolar interaction. The Hamiltonian reads $H = H_0 + H_I$, where $H_0 = \sum_{\mathbf{k},\sigma} \xi_{\mathbf{k},\sigma} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}$ is the kinetic energy, and

$$H_I = \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} V_{\mathbf{k}\mathbf{k}'} c^\dagger_{-\mathbf{k}'+\mathbf{q}/2,\uparrow} c^\dagger_{\mathbf{k}'+\mathbf{q}/2,\downarrow} c_{\mathbf{k}+\mathbf{q}/2,\downarrow} c_{-\mathbf{k}+\mathbf{q}/2,\uparrow} \quad (3)$$

is the interaction energy. Here $c_{\mathbf{k}\sigma}$ are fermionic field operators, $\sigma = \uparrow, \downarrow$, stands for the layer index, $\xi_{\mathbf{k}\sigma} = k^2/2m - \mu_\sigma$ (hereinafter we put $\hbar = 1$), and $\mu_\sigma = \mu \pm h$ are chemical potentials of the layers. When the densities in the layers are not equal to each other, there is a difference in the Fermi momenta of the two (pseudo)spin states, $\delta k_F \equiv |k_{F,\uparrow} - k_{F,\downarrow}| \neq 0$, and the effective magnetic field $h \neq 0$.

Relying on Eq. (3) we define the pairing gap function as $\Delta_{\mathbf{k}\mathbf{Q}} \equiv \sum_{\mathbf{k}'} V_{\mathbf{k}\mathbf{k}'} \langle c_{\mathbf{k}'+\mathbf{Q}/2,\downarrow} c_{-\mathbf{k}'+\mathbf{Q}/2,\uparrow} \rangle$, where $\mathbf{k}'$ is the relative momentum of two paired fermions, and $\mathbf{Q}$ is their center-of-mass (CM) momentum. In the coordinate space, the order parameter is then consisting of pairing wavefunctions for several CM momenta:

$$\Delta_{\mathbf{k}}(\mathbf{R}) = \sum_{n=1}^{N_Q} \Delta_{\mathbf{k}\mathbf{Q}_n} e^{i\mathbf{Q}_n\cdot\mathbf{R}}, \quad (4)$$

where $\mathbf{R}$ is the CM position of the Cooper pair, $\mathbf{Q}_n$ is the CM momentum involved in the order parameter, and N_Q is the total number of considered $\mathbf{Q}_n$. Below we consider several symmetries of the order parameter: uniform superfluid ($N_Q = 1$ and $\mathbf{Q}_1 = 0$), plane-wave FFLO ($N_Q = 1$ and $\mathbf{Q}_1 = q\hat{x}$, where $\hat{x}$ is a unit vector in the x direction), stripe FFLO ($N_Q = 2$ and $\mathbf{Q}_{1,2} = \pm q\hat{x}$), and triangular state ($N_Q = 3$ and three $\mathbf{Q}_n$ vectors have the same amplitude, with $2\pi/3$ difference in their orientation). Close to the FFLO-normal phase boundary we also consider square and hexagonal FFLO structures.

The finite-temperature normal and anomalous Green's functions $G_{\sigma\sigma'}$ and $F^\dagger_{\sigma\sigma'}$ are found from the Gor'kov equations [57,58] (see Supplemental Material for details [59]):

$$G_{\sigma\sigma'}(\mathbf{k}_1,\mathbf{k}_2;i\omega_n) = \delta_{\sigma\sigma'}\delta_{\mathbf{k}_1,\mathbf{k}_2}\left(i\omega_n - \xi_{\mathbf{k}_1\sigma} - \sum_{m=1}^{N_Q} \frac{\Delta_{\mathbf{k}_1,\mathbf{Q}_m}\Delta^\dagger_{\mathbf{k}_2-\mathbf{Q}_m,\mathbf{Q}_m}}{i\omega_n + \xi_{\mathbf{k}_1-\mathbf{Q}_m,\sigma}}\right)^{-1}; \quad (5)$$

$$F^\dagger_{\sigma\sigma'}(\mathbf{k}_1,\mathbf{k}_2;i\omega_n) = \frac{-\sum_{m=1}^{N_Q} \Delta^\dagger_{\mathbf{k}_1,\mathbf{Q}_m}\delta_{\mathbf{k}_1+\mathbf{Q}_m,\mathbf{k}_2}}{i\omega_n + \xi_{-\mathbf{k}_1\sigma}} \times G_{\sigma'\sigma'}(\mathbf{k}_2,\mathbf{k}_2;i\omega_n)(1-\delta_{\sigma\sigma'}), \quad (6)$$

where $\omega_n = (2n+1)\pi T$ are Matsubara frequencies, and T is the temperature. The gap equation can then be obtained self-consistently:

$$\Delta^*_{\mathbf{k}\mathbf{Q}} = -T\sum_{n,\mathbf{k}'} V_{\mathbf{k}\mathbf{k}'} F^\dagger_{\uparrow\downarrow}\left(\mathbf{k}' - \frac{\mathbf{Q}}{2}, \mathbf{k}' + \frac{\mathbf{Q}}{2}; \omega_n\right). \quad (7)$$

We thus identify the Gor'kov equations for the Green's functions and the gap equations for $\Delta^*_{\mathbf{k}\mathbf{Q}}$ and $\Delta_{\mathbf{k}\mathbf{Q}}$ as self-consistent Gor'kov equations.

The plane-wave, stripe, triangular, square, and hexagonal phases break the rotational symmetry and have an anisotropic gap in the momentum space. In principle, such gap function can be measured using Bragg spectroscopy by exciting particles with a finite momentum.

In the following, we first solve the self-consistent Gor'kov equations by assuming a certain gap function symmetry (uniform, plane-wave, stripe, or triangular) and then determine the phase diagram by comparing the obtained free energies of these candidates. The derivation of Eqs. (5)–(9) and a detailed presentation of the numerical procedure of obtaining the phase diagrams at zero and finite temperatures are contained in the Supplemental Material.

Near the phase transition line (superfluid–normal state), where the order parameter is small, we may use the Ginzburg-Landau free-energy functional $F = F_2 + F_4 + \cdots$, with

$$F_2 = \sum \Delta_{\mathbf{k}\mathbf{q}}(V^{-1})_{\mathbf{k}\mathbf{k}'}\Delta^*_{\mathbf{k}'\mathbf{q}} - T\sum |\Delta_{\mathbf{k}\mathbf{q}}|^2 G_+\left(\mathbf{k}+\frac{\mathbf{q}}{2},\omega_n\right) \times G_-\left(-\mathbf{k}+\frac{\mathbf{q}}{2},-\omega_n\right), \quad (8)$$

$$F_4 = \frac{T}{2}\sum_{\{\mathbf{q}_j\},\mathbf{k}n} \Delta^*_{\mathbf{k}\mathbf{q}_1}\Delta^*_{\mathbf{k}\mathbf{q}_2}\Delta_{\mathbf{k}\mathbf{q}_3}\Delta_{\mathbf{k}\mathbf{q}_4} G_+(\mathbf{k}+\mathbf{q}_1,\omega_n) \times G_-(-\mathbf{k}+\mathbf{q}_3-\mathbf{q}_1,-\omega_n)G_+(\mathbf{k}+\mathbf{q}_4,\omega_n) \times G_-(-\mathbf{k},-\omega_n)\delta_{\mathbf{q}_1+\mathbf{q}_2,\mathbf{q}_3+\mathbf{q}_4}, \quad (9)$$

where the Green's functions of the normal state are

$$G_\pm(\mathbf{k},\omega_n) = \frac{1}{i\omega_n - \xi_{\mathbf{k}} \mp h}, \quad (10)$$

and V^{-1} is the inverse matrix of $V_{\mathbf{k}\mathbf{k}'}$. The term F_4 is necessary to find the minimum energy configuration, whereas F_2 determines the tricritical point.

Zero temperature. At zero field ($h = 0$) the ground state is a uniform superfluid with the order parameter on the Fermi surface $\Delta_0(k_F) \equiv \Delta_0$, where $k_F = \sqrt{2m\mu}$ is the Fermi momentum at $h = 0$. For a given interaction strength the FFLO phase emerges at a critical value h_{c1}, and our calculations show that this is a stripe phase. For sufficiently large field h_{c2} the ground state becomes normal. Note that the stripe FFLO phase is clearly the ground state at h that is lower than h_{c2} by a few percent (see Supplemental Material [59]). The dependence of h_{c1}, h_{c2} on the parameter $k_F\lambda$ displayed in Fig. 2 shows that for $k_F\lambda > 1$ the FFLO region is significantly wider than in the case of contact interaction, where the

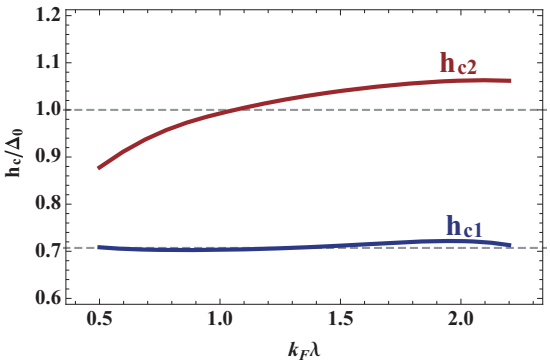

FIG. 2. Critical fields h_{c1} and h_{c2} at $T = 0$ in units of Δ_0 versus the parameter $k_F\lambda$ at $k_F r_* = 0.7$, where $r_* = md^2$ is the dipole-dipole distance. The dashed lines show h_{c1} and h_{c2} for the case of contact interaction.

FFLO phase emerges for $\Delta_0 > h > \Delta_0/\sqrt{2} \approx 0.707\Delta_0$. Here we arrive at a very important point. For $k_F\lambda < 1.055$ the p-wave interaction on the Fermi surface is repulsive and interlayer Cooper pairs only contain the contribution of an attractive s-wave interaction. However, if $k_F\lambda > 1.055$, then the p-wave interaction $V_1(k_F,k_F)$ also becomes attractive and Copper pairs are already composed of both s-wave and p-wave contributions. This makes the modulus of the order parameter larger and requires a higher field h_{c2} to destroy superfluidity and get to the normal state (see Supplemental Material).

Finite-temperature phase diagram: At finite temperatures the equilibrium phases are determined by comparing the free energies of the uniform superfluid, FFLO, and the normal state. In Fig. 3 the phase diagram is presented in terms of $T/T_{0,s}$ and h/Δ_0, where $T_{0,s}$ is the transition temperature at $h = 0$ (provided by the s-wave pairing). Note that the two-dimensional superfluid-to-normal transition is of Berezinskii-Kosterlitz-Thouless (BKT) type, but for weakly interacting fermions the superfluid transition temperature is very close to that calculated in the BCS mean-field approach [60,61]. With increasing temperature, the critical field h_{c1} for the transition from the uniform superfluid to FFLO phase decreases. So does the imbalance for the transition from the FFLO to the normal state. The three phases (SF, FFLO, and normal) merge at the tricritical point T^*. For $k_F\lambda < 1$ we have $T^* \approx 0.56T_{0,s}$ like in the case of contact interactions. However, for $k_F\lambda > 1$ the p-wave pairing contribution to the order parameter comes into play and the ratio $T^*/T_{0,s}$ increases. Already for $k_f\lambda = 2.2$ we obtain $T^* \simeq 0.62T_{0,s}$ and a somewhat wider FFLO region than in the case of contact interactions (see Fig. 3).

The FFLO phase is mostly the Larkin-Ovchinnikov stripe state. In the temperature interval from $0.02T_{0,s}$ to $0.3T_{0,s}$ the stripe phase has the lowest free energy for h lower than h_{c2} by more than 5%. For h closer to h_{c2} our numerical calculations based on the Ginzburg-Landau functional show that the equilibrium state is a triangular FFLO (it is also recovered from the self-consistent Gor'kov equations at h close to $0.95h_{c2}$), which with decreasing temperature becomes a square and then hexagonal FFLO (see Supplemental Material). This sequence of FFLO states is similar to that observed for contact interactions [8,12,62].

The structure of the phase diagram is similar to that in the case of contact interactions [2,5,62]. However, in our case the FFLO region significantly depends on the parameters, so that $T^*/T_{0,s}$ and h^*/Δ_0 are not universal numbers as they are for contact interactions where $T^* \approx 0.56T_{0,s}$ and the corresponding critical imbalance is $h^* \approx 0.6\Delta_0$ [5,62].

The tricritical temperature can be determined analytically (see Supplemental Material for details):

$$-\ln\frac{T_{0,s}}{T_{0p}} + \mathrm{Re}\left[\Psi\left(\frac{1}{2} + i\frac{h^*}{2\pi T^*}\right) - \Psi\left(\frac{1}{2}\right)\right] = 0; \quad (11)$$

$$-\mathrm{Re}\left[\Psi''\left(\frac{1}{2} + i\frac{h^*}{2\pi T^*}\right)\right] = \frac{[\mathrm{Im}\Psi'(1/2 + ih^*/2\pi T^*)]^2}{\ln(T_{0,s}/T_{0,p})}, \quad (12)$$

where $T_{0,p}$ is the critical temperature of superfluid transition for the p-wave pairing at $h = 0$, and Ψ is the digamma function. In the limit $T_{0,p} \to 0$, Eqs (11) and (12) give the known result for the contact interaction, which is specified in the previous paragraph. These equations also reproduce the numerical results of Fig. 3. The maximum tricritical temperature $T^* \to T_{0,s}$ is reached in the limit $k_F\lambda \gg 1$, where $T_{0,p}$ is close to $T_{0,s}$. However, the critical temperature $T_{0,s}$ decreases with increasing $k_F\lambda$. For $k_F\lambda \simeq 2$, where one can

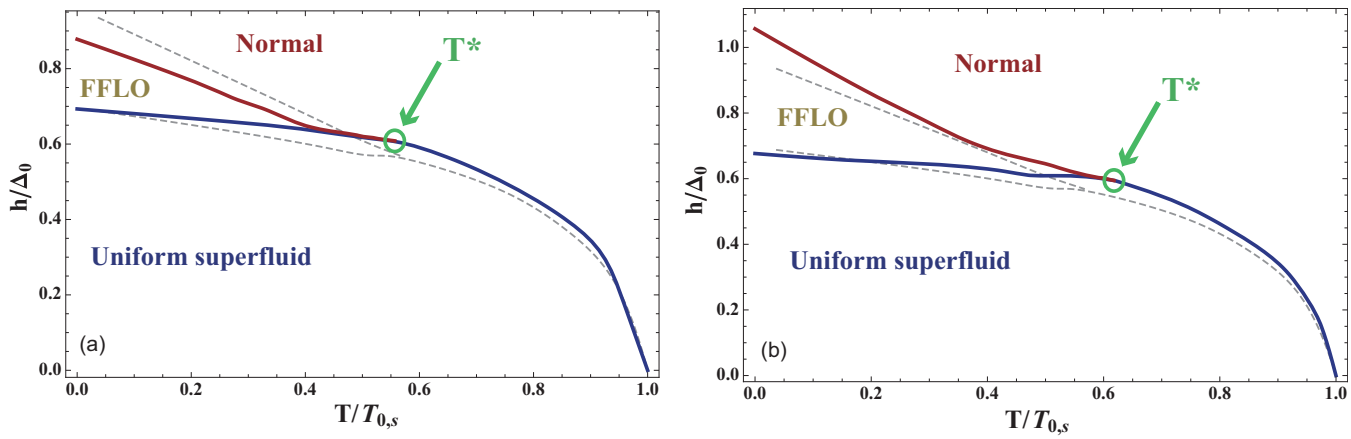

FIG. 3. (a) Finite-temperature phase diagram in terms of $T/T_{0,s}$ and the imbalance h/Δ_0. In (a) $k_F\lambda = 0.5$ and $r_* = \lambda/2$, and in (b) $k_F\lambda = 2.2$ and $r_* = \lambda$. The dashed curves are the phase boundaries for the case of contact interaction.

still hope to achieve $T_{0,s}$ on the level of nanokelvins (see below), we obtain $T^* \approx 0.62 T_{0,s}$.

The lattice wave vector $|\mathbf{Q}_m|$ behaves as $Q \sim \sqrt{T^* - T}$ near the tricritical point and vanishes at $T = T^*$ (see Supplemental Material).

As is already stated above, the equilibrium FFLO phase in our case is the stripe state. Like in the case of contact interactions [62,63], the transition from the uniform superfluid to FFLO state is of the first order, whereas the transition from the FFLO to normal phase is of the second order. At 2D densities $\sim 10^9$ cm^{-2} the Fermi energy for weakly reactive NaLi molecules or nonreactive NaK molecules is $\epsilon_F \sim 1$ μK, and for the interlayer spacing 200 nm we have $k_F\lambda \simeq 2$. Then, on approach to the strongly interacting regime with $k_F r_*$ slightly exceeding unity, the superfluid transition temperature will be up to 10 nK. For magnetic atoms and even for molecules with a large magnetic dipole moment, e.g., Dy_2, in the same conditions, however, the superfluid transition temperature is below 1 nK at densities approaching 10^9 cm^{-2}. Larger values of $k_F\lambda$ and, hence, higher ratios $T^*/T_{0,s}$ would require $k_F r_*$ significantly larger than unity, so that the inlayer interaction becomes very strong driving the system far away from the dilute regime. This case is beyond the scope of the present Rapid Communication.

Conclusions. We used both a theoretical field approach based on the Gor'kov equations and the theory of phase transitions based on the Ginzburg-Landau free-energy functional to study possible FFLO phases in a bilayer system of fermionic polar molecules with a finite imbalance of molecular densities of the layers.

Our work demonstrates the importance of the long-range character of the dipole-dipole interaction, which can combine the s-wave and p-wave pairing in the order parameter and enhance the FFLO regime. The observation of this FFLO state is feasible for nonreactive NaK molecules or weakly reactive NaLi molecules at temperatures ~ 10 nK.

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