Realizing Fulde-Ferrell Superfluids via a Dark-State Control of Feshbach Resonances

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We propose that the long-sought Fulde-Ferrell superfluidity with nonzero momentum pairing can be realized in ultracold two-component Fermi gases of $^{40}$K or $^6$Li atoms by optically tuning their magnetic Feshbach resonances via the creation of a closed-channel dark state with a Doppler-shifted Stark effect. In this scheme, two counterpropagating optical fields are applied to couple two molecular states in the closed channel to an excited molecular state, leading to a significant violation of Galilean invariance in the dark-state regime and hence to the possibility of Fulde-Ferrell superfluidity. We develop a field theoretical formulation for both two-body and many-body problems and predict that the Fulde-Ferrell state has remarkable properties, such as anisotropic single-particle dispersion relation, suppressed superfluid density at zero temperature, anisotropic sound velocity, and rotomic collective mode. The latter two features can be experimentally probed using Bragg spectroscopy, providing a smoking-gun proof of Fulde-Ferrell superfluidity.

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Introduction.—The application of magnetic Feshbach resonance (MFR) in Fermi gases of alkali-metal atoms [1] opens a new paradigm to study strongly correlated many-body phenomena [2,3]. The crossover from Bardeen-Cooper-Schrieffer (BCS) superfluid to Bose-Einstein condensate (BEC) [4–9] in atomic Fermi gases has been experimentally explored in great detail [10–15], leading to a number of new concepts such as unitary Fermi superfluid and universal equation of state [15–17].

Finite-momentum pairing superfluidity, or the so-called Fulde-Ferrell-Larkin-Ovchinikov (FFLO) state [18,19], is another intriguing phenomenon addressed using ultracold Fermi gases [20–25]. It has been studied and pursued for over a half-century in both condensed matter physics and nuclear physics [26,27]. Yet, its existence remains elusive. In three-dimensional free space, the conventional scenario of spin-population imbalance leads to a rather narrow window for FFLO in atomic Fermi gases [22,23]. It was proposed that the stability regime for FFLO can be significantly enhanced via engineering single-particle properties [28], using optical lattice [29–36] or spin-orbit coupling [37–45]. While it was theoretically predicted that in the presence of spin-orbit coupling and an in-plane Zeeman field, the Fulde-Ferrell (FF) superfluid state is energetically favored in a large parameter space [38–41], the heating problem in realizing spin-orbit coupled FF superfluids at low temperature has not yet been solved experimentally [46].

In this Letter, we propose that the FF superfluidity can be realized without spin-population imbalance, via engineering interatomic interaction. The new scenario is based on the recent ground-breaking demonstration of a dark-state optical control of MFRs [47] and its innovative extension to allow a center-of-mass (c.m.) momentum $q$-dependent interatomic interaction [48]. As shown in Fig. 1, the dark-state optical control of the MFR uses two ground molecular states $|g_1\rangle$ and $|g_2\rangle$ that couples to an excited molecular state $|e\rangle$ by two optical fields of frequencies $\omega_1$ and $\omega_2$, wave vectors $k_1$ and $k_2$, and Rabi frequencies $\Omega_1$ and $\Omega_2$, respectively [47–50]. The MFR is induced by the hyperfine coupling between the atomic pair state and the molecular state $|g_1\rangle$ [51–53]. In the dark-state regime, the resulting Stark shift $\Sigma_1$ in the state $|g_1\rangle$ is attributed to the Doppler effect [48], i.e., $\Sigma_1 \sim (\Omega_1/\Omega_2)^2 q \cdot (k_1 - k_2)$. Hence, if the two optical fields propagate along opposite directions (i.e., $k_1 = -k_2 = k_R e_z$), the violation of Galilean invariance becomes significant when $(\Omega_1/\Omega_2)^2 \gg 1$, and may lead to interesting many-body consequences.

FIG. 1. Level scheme for the dark-state control of MFR. The ground molecular state $|g_1\rangle$, responsible for the MFR, is shifted by two optical fields (green lines). The small Doppler effect due to $k_1 \neq k_2$ can be greatly amplified in the dark-state regime by a factor of $(\Omega_1/\Omega_2)^2$.

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One of the key observations in this Letter is that the zero-momentum pairing state has a nonzero current \( j \propto k_1 - k_2 \) carried by the condensate and suffers from severe instability. The true ground state of the system therefore falls toward a FF state so that the currents carried by the condensate and the fermionic quasiparticles cancel each other precisely. This compensation mechanism is equally important for reducing the Doppler effect in the two-photon detuning and keeps the system in the dark-state regime. As a result, optical loss and heating become negligible.

Field theory.—The MFR can be described by the atommolecule theory \([9,51–53]\), of which the Lagrangian density is given by \( \mathcal{L}_\text{MFR} = \mathcal{L}_A + \mathcal{L}_M + \mathcal{L}_\text{AM} \), with

\[
\mathcal{L}_A = \sum_{\sigma = \uparrow, \downarrow} \psi_\sigma^\dagger \hat{K}_\sigma \psi_\sigma - u_0 \psi_{\uparrow}^\dagger \psi_{\uparrow} \psi_{\downarrow} \psi_{\downarrow}^\dagger,
\]

\[
\mathcal{L}_M = \phi_\sigma^\dagger (\hat{K}_B - \nu_0) \phi_\sigma,
\]

\[
\mathcal{L}_\text{AM} = -g_0 (\phi_\uparrow \psi_{\downarrow} \psi_{\downarrow}^\dagger + \phi_\downarrow \psi_{\uparrow} \psi_{\uparrow}^\dagger).
\]

Here, \( \psi_\sigma \) \((\sigma = \uparrow, \downarrow)\) denotes the open-channel fermions and \( \phi_\sigma \) denotes the closed-channel molecular state \(|g_1\rangle\). We use the notations \( \hat{K}_F = i \partial_t + \nabla^2 / (2m) \) and \( \hat{K}_B = i \partial_t + \nabla^2 / (4m) \), with \( t \) being the time and \( m \) being the atom mass. The units \( h = k_B = 1 \) will be used throughout. The bare couplings \( u_0 \) and \( g_0 \) as well as the bare magnetic detuning \( \nu_0 \) should be renormalized in terms of the background scattering length \( a_{bg} \), resonance width \( \Delta B \), and detuning \( \Delta \mu(B - B_0) \), in the forms of \( u = 4\pi a_{bg} / m \), \( g = \sqrt{\Delta \mu \Delta B} \), and \( \nu = \Delta \mu(B - B_0) \) \([51–54]\). In the presence of optical fields, we add a new molecular part

\[
\mathcal{L}'_M = \phi_\sigma^\dagger (\hat{K}_B - E_2) \phi_\sigma + \phi_e^\dagger \left( \hat{K}_B - E_e + i \frac{\gamma_e}{2} \right) \phi_e - \sum_{i=1,2} \left( \Omega_i \frac{\phi_i^\dagger \phi_e e^{i \theta_i(t)} + \Omega_i^* \phi_e^\dagger \phi_i e^{-i \theta_i(t)} }{2} \right),
\]

where \( \phi_\sigma \) and \( \phi_e \) denote the states \(|g_2\rangle \) and \(|e\rangle \) with energies \( E_2 \) and \( E_e \), respectively, \( \theta_i(t) = k_i \cdot r - \omega_it \) and \( \gamma_e \) is the decay rate of the excited molecular state \(|e\rangle \). The last term in Eq. (2) describes the one-body Raman transitions between the molecular states.

The phase factors \( \theta_i(t) \) can be eliminated by defining two new molecular fields, \( \phi_1 = \phi_\sigma e^{-i \theta_1} \) and \( \phi_2 = \phi_\sigma e^{-i \theta_2} \). By setting \( \phi_1 = 1 \), we obtain a compact form \( \mathcal{L}'_M + \mathcal{L}'_{AM} = \Phi^\dagger M \Phi d^2r dt \), where \( \Phi = (\phi_1, \phi_2, \phi_e)^T \), and the inverse propagator matrix in momentum space reads

\[
M(q_0, q) = \begin{pmatrix}
I_1(q_0, q) & 0 & -\Omega_1^*/2 \\
0 & I_2(q_0, q) & -\Omega_2^*/2 \\
-\Omega_1^*/2 & -\Omega_2^*/2 & I_e(q_0, q)
\end{pmatrix}.
\]

with diagonal elements \( I_1(q_0, q) = Z - \nu_0 \) and

\[
I_2(q_0, q) = Z - \frac{q \cdot (k_1 - k_2)}{2m} - \frac{(k_1 - k_2)^2}{4m} + \delta,
\]

\[
I_e(q_0, q) = Z - \frac{q \cdot k_1}{2m} - \frac{k_1^2}{4m} + \Delta_e + i \frac{\gamma_e}{2}.
\]

Here, \( \Delta_e = \omega_1 - E_e \) is the one-photon detuning, \( \delta = (\omega_1 - \omega_2 - E_2) \) is the two-photon detuning, and \( Z = q_0 - q^2/(4m) \) is a Galilean invariant combination, with \( q_0 \) and \( q \) being the c.m. energy and momentum of two incident atoms. The Rabi frequencies and the detunings are experimentally tunable \([47,57]\).

Two-body problem.—The off-shell \( T \) matrix for atom-molecule scattering is exactly given by the bubble summation, \( T_{2b}(q_0, q) = [U^{-1}(q_0, q) - \Pi(q_0, q)]^{-1} \). Here, \( U(q_0, q) = u_0 + q_0^2 D_1(q_0, q) \) is an energy- and momentum-dependent interaction vertex, with \( D_1(q_0, q) \) being the propagator of the molecular state \(|g_1\rangle \). With optical fields, \( D_1(q_0, q) = \frac{1}{\Omega_1^2(q_0, q) - \Omega_1^2} \) is given by the 11-component of \( M^{-1}(q_0, q) \), where the self-energy or the so-called Stark shift reads

\[
\Sigma_1(q_0, q) = \frac{\Omega_1^2}{4} \left[ I_e(q_0, q) - \frac{\Omega_2^2}{4I_2(q_0, q)} \right]^{-1}.
\]

The two-atom bubble function \( \Pi(q_0, q) \) is given by \( \Pi(q_0, q) = \sum_{\rho} (Z + i0^+) \epsilon_\rho^{-1} \) with \( \epsilon_\rho = p^2/(2m) \), and is to be replaced by \( \Pi_r(q_0, q) = m/(4\pi) \sqrt{-m(Z + i0^+)} \) after renormalization. More explicitly, in terms of the renormalized quantities, the \( T \) matrix \( T_{2b}(q_0, q) \) takes the form \( T_{2b}(q_0, q) = [U_R^{\dagger}(q_0, q) - \Pi_r(q_0, q)]^{-1} \), where the effective coupling reads \([54]\)

\[
U_R(q_0, q) = u + \frac{g^2}{Z - \nu - \Sigma_1(q_0, q)},
\]

which fully characterizes the interatomic interaction in the presence of optical fields.

For the optical control of MFRs in atomic gases of \(^6\text{Li}\) and \(^{40}\text{K}\), the Doppler effect term \( \nu = k_R \cdot e \) is of the order of the recoil energy, \( E_R = k_R^2/(2m) \approx 2\pi \times 10 \text{ kHz} \), and is usually neglected, in comparison with the decay rate and Rabi frequencies \( \gamma_e, \Omega_{1,2} \approx 2\pi \times 10 \text{ MHz} \). However, in the dark-state regime with \( \delta = 0 \) (i.e., \( I_e \ll \Omega_2^2/I_2 \)) and a large ratio \( \Omega_1/\Omega_2 \), it could be greatly enhanced, leading to a Stark shift as large as \( 10^{-2} \Delta \mu \Delta B \). This gives rise to a c.m. momentum-dependent interaction \([48]\) and hence a strong violation of Galilean invariance. Throughout the work, we assume \( k_1 = k_R e_1 = -k_2 \), with \( k_R = 8.138 \times 10^6 \text{ m}^{-1} \), and focus on the case of \(^{40}\text{K}\) atoms near the broad resonance at \( B_0 = 202.02 \text{ G} \) with...
\(a_{bg} = 174a_0, \ \Delta B = 7.04 \text{ G}, \ \text{and} \ \Delta \mu = 2\mu_B \) [58]. We consider the typical values \(\Delta_c = -2\pi \times 500 \text{ MHz}, \ \gamma_c = 2\pi \times 6 \text{ MHz}, \ \delta = 0, \ \Omega_1 = 2\pi \times 120 \text{ MHz}, \ \text{and} \ \Omega_2 = 2\pi \times 20 \text{ MHz}, \) unless specified elsewhere [57]. We also take a typical atom density \(n = 1.82 \times 10^{13} \text{ cm}^{-3}, \) corresponding to a Fermi momentum \(k_F = (3\pi^2n)^{1/3} = k_R \) [57].

With the above parameters, the violation of Galilean invariance is already clearly seen in the dimer bound state, whose energy \(E_d(q)\) is determined by the pole of the \(T\) matrix, i.e., \(T_{q_0}^{-1}[E_d(q), q] = 0 \) [54,59]. Without optical fields, the Galilean invariance ensures that \(E_d(q) = \varepsilon_R + q^2/(4m), \) with \(\varepsilon_R\) being the binding energy.

In the presence of optical fields, the effective interaction \(U_R(q_0, q)\) depends not only on \(Z\) but also on the pair momentum \(q\) itself, which indicates that the Galilean invariance and especially the spatial inversion symmetry are broken. As a consequence, \(E_d(q)\) has a nontrivial \(q\) dependence and the lowest dimer energy locates at \(q \neq 0.\)

In Fig. 2(a), we show the momentum of the dimer bound state, \(q_0 = Q e_c,\) by using a dashed line. We have in general \(Q \neq 0\) at the BEC side of the MFR. The corresponding two-body Stark shift is reported in Fig. 2(b). Its imaginary part (i.e., decay rate) is about \(10^{-5}\gamma_c \sim 2\pi \times 100 \text{ Hz},\) indicating a reasonably long dimer lifetime \(\sim 0.01-0.1 \text{ s} \) [48,54].

**Many-body theory.**—The partition function of the system is given by the imaginary-time formalism \(Z = \int \mathcal{D}[\psi, \psi^*; \Phi, \Phi^*] \exp\left[\int d\xi \left(\mathcal{L}_{\text{MFR}} + \mathcal{L}_M + \mathcal{L}_C\right)\right],\) where \(x = (\mathbf{r}, \tau)\) and the chemical potential \(\mu\) is introduced through the term \(\mathcal{L}_\mu = \mu \sum_{\sigma = 1,2} \psi^\dagger_\sigma \psi_\sigma + 2\mu \sum_{i=1,2} \phi^\dagger_1 \phi_i.\)

To decouple the four-fermion interaction term, we introduce an auxiliary field \(\phi_j(x) = \eta_0 \psi^\dagger_1(x) \psi_1(x),\) perform the Hubbard-Stratonovich transformation, and integrate out the fermions to obtain \(Z = \int \mathcal{D}[\phi_f, \phi^*; \Phi, \Phi^*] \exp (-S_{\text{eff}}),\) with the effective action \([\Delta(x) = \phi_f(x) + \eta_0 \phi_1(x)]:\)

\[
S_{\text{eff}} = -\text{Tr} \ln \left[ \left( \frac{\hat{K}_F + \mu}{\Delta_1(x)} \right) \frac{\Delta(x)}{-\hat{K}_F - \mu} \delta(x - x') \right]
\]

\[
- \int dx \left[ \frac{|\phi_f(x)|^2}{\eta_0} + \Phi^2 M(2\mu - \partial_x - i\nabla)\right].
\]

We evaluate \(Z\) in the mean-field approximation, which amounts to searching for the static saddle-point solution \(\hat{\phi}_f(x) = \hat{\phi}_f(r) (l = 1,2,e,f)\) that minimizes the effective action \(S_{\text{eff}}.\) Motivated by the fact that the dimer ground state has nonzero momentum, we expect that the fermion pairing favors nonzero momentum in the superfluid state. Thus, we take the Fulde-Ferrell ansatz for the saddle-point solution, \(\hat{\phi}_f(r) = C_l e^{\Omega Q r^2},\) where \(Q\) is the pairing momentum. The fermionic part (i.e., the Tr ln term) can be evaluated by performing a phase transformation for the fermion fields, \(\psi_\sigma = \tilde{\psi}_\sigma e^{\Omega Q r^2/2}.\) Using the saddle-point condition \(\partial S_{\text{eff}}/\partial C_l = 0,\) we can express \(C_l\) in terms of \(\Delta = C_f + \eta_0 C_1.\) By further using the renormalized couplings and detuning, the thermodynamic potential at zero temperature reads [54,59]

\[
\Omega = \Omega_q + \sum_k \left( \xi_k - E_k + \frac{|\Delta|^2}{2\varepsilon_k} \right) - \frac{|\Delta|^2}{U_R(2\mu, Q)}.
\]

Here the dispersions are defined as \(\xi_k = \varepsilon_k + Q^2/(8m) - \mu\) and \(E_k = (\xi^2_k + |\Delta|^2)^{1/2}.\) The quasiparticle term \(\Omega_q = \sum_{i=x} \sum_k E_k \Theta(-E_k)\) contributes only when the quasiparticle excitations \(E_k^x = E_k \pm k \cdot Q/(2m)\) are gapless. The last term in the expression is quite meaningful: The condensation energy contains the effective two-body interaction \(U_R(q_0, q)\) evaluated at \((q_0, q) = (2\mu, Q)\).

The superfluid ground state is determined by solving the gap equations [54], \(\partial \Omega/\partial \Delta = 0\) and \(\partial \Omega/\partial Q = 0,\) and the number equation, \(n = -\partial \Omega/\partial \mu.\)

**Finite-momentum superfluidity.**—In the conventional FF state with Galilean invariance, the thermodynamic potential is an even function of \(Q\) and has a trivial solution \(Q = 0,\) which indicates that the instability toward FF occurs at the order \(O(Q^2)\) [60,61]. However, here we find that \(Q = 0\) is no longer a trivial solution. Physically, this means that the \(Q = 0\) state has a spontaneously generated current \(j \neq 0\) from the condensate due to the violation of Galilean...
invariance. Explicitly, we find $j \propto k_1 - k_2 = 2k_F e_x$ [54] from $j = 2m\partial \Omega / \partial Q$ evaluated at $Q = 0$. Thus, the instability toward FF occurs at the order $O(Q)$. Therefore, to stabilize the system, the ground state falls to a FF state so that a new current generated by the fermionic quasiparticles, $j' \propto Q$, cancels precisely the current carried by the condensate. This also shows that the pair momentum is along the $z$ direction, $Q = Q e_z$.

On the other hand, in the BEC limit, $\mu$ becomes large and negative and $|\mu| \gg \Delta$. To the leading order in $\Delta/|\mu|$, the gap equation $\partial \Omega / \partial Q = 0$ can be expressed as [54]

$$U_R^{-1}(2\mu, Q) - \Pi_R(2\mu, Q) = 0,$$

(9)

which is exactly the equation determining the dimer energy $E_d(Q) = 2\mu(Q)$ as a function of $Q$. Moreover, using the fact $|\mu| \gg \Delta$, we can show that the other two equations, $\partial \Omega / \partial Q = 0$ and $n = -\partial \Omega / \partial \mu$, give rise to the equation $\partial \mu(Q)/\partial Q = 0$ [54]. Thus, $2\mu$ approaches the lowest energy of the dimer state, and the superfluid ground state is a finite-momentum Bose-Einstein condensation of tightly bound dimers.

Figure 2(a) reports a typical calculation of the FF momentum $Q$ across the MFR (solid circles). We find that unlike the two-body case (dashed line), the FF state with $Q \neq 0$ arises even at the BCS side. It is remarkable that the imaginary part of the many-body Stark shift is very small (i.e., $<10^{-6} \gamma$) at the BCS side [Fig. 2(b)], indicating negligible optical loss and heating effect. This is largely due to the reduced chemical potential, which compensates the Doppler effect in $I_2(2\mu, Q)$ and thereby locks the system in the dark-state regime. Near resonance, the lifetime of the system is estimated to be 100 ms [54].

Numerically we have checked that the FF state is the true minimum of the energy landscape [Fig. 3(a)] and always has lower free energy than the $Q = 0$ state [Fig. 3(b)]. In the BEC limit, the FF momentum approaches the momentum of the ground-state dimer, consistent with the above analysis. Around the MFR, the FF momentum $Q$ reaches a sizable value $Q \sim k_F$, which may lead to visible observational effects in cold atom experiments. Figure 3(c) reports a typical energy spectrum of the collective phonon modes at $Q = 0$, as reported in Fig. 4. Along the $Q$

- $\Omega_\perp = 2k_F$.
- $\Omega_\parallel = 0$.

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direction, the phonon mode splits into two branches with different velocities. At large momentum, one branch merges into the two-particle continuum, leading to an interesting maxon-roton structure. These predictions can be probed by applying the Bragg spectroscopy [63].

Summary.—We have proposed that the dark-state optical control of MFR provides a natural and robust way to realize the FF superfluidity as well as the finite-momentum BEC of dimers. While our calculations are specific for $^{40}$K atoms, our theory and mechanism for FF superfluidity is generic and is applicable to other systems including $^6$Li atoms. The unique advantage of our proposal is that the system is free from optical loss and heating due to the dark-state manipulation. It opens a fascinating way to explore some unique features of Fulde-Ferrell superfluids, in particular, the anisotropic phonon dispersion and emergent roton structure, by using Bragg spectroscopy.

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