Inhomogeneous Fulde-Ferrell superfluidity in spin-orbit-coupled atomic Fermi gases

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Inhomogeneous superfluidity lies at the heart of many intriguing phenomena in quantum physics. It is believed to play a central role in unconventional organic or heavy-fermion superconductors, chiral quark matter, and neutron star glitches. However, so far even the simplest form of inhomogeneous superfluidity, the Fulde-Ferrell (FF) pairing state with a single center-of-mass momentum, is not conclusively observed due to the intrinsic complexity of any realistic Fermi systems in nature. Here we theoretically predict that the controlled setting of ultracold fermionic atoms with synthetic spin-orbit coupling induced by a two-photon Raman process, demonstrated recently in cold-atom laboratories, provides a promising route to realize the long-sought FF superfluidity. At experimentally accessible low temperatures, the FF superfluid state dominates the phase diagram, in sharp contrast to the conventional case without spin-orbit coupling. We show that the finite center-of-mass momentum carried by Cooper pairs is directly measurable via momentum-resolved radio-frequency spectroscopy. Our work opens the way to direct observation and characterization of inhomogeneous superfluidity.

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Interacting Fermi systems with imbalanced spin populations are quite ubiquitous in nature, with manifestations ranging from solid-state superconductors to astrophysical objects [1]. The spin imbalance disrupts the Bardeen-Cooper-Schrieffer (BCS) mechanism of superconductivity, where fermions of opposite spin and momentum form Cooper pairs. As a result, an exotic superconducting state, characterized by Cooper pairs with a finite center-of-mass momentum and spatially nonuniform order parameter, may occur, as predicted by Fulde and Ferrell nearly 50 years ago [2]. More complicated inhomogeneous pairing states are also possible with the inclusion of more and more momenta for the spatial structure of order parameter, following the idea by Larkin and Ovchinnikov (LO) [3]. These forms of inhomogeneous superfluidity, now referred to as FFLO states, have attracted tremendous theoretical and experimental efforts over the past five decades [4]. Remarkably, to date there is still no conclusive experimental evidence for FFLO superfluidity. In solid-state systems such as the organic superconductor λ-(BETS)$_2$FeCl$_4$ [5] and the heavy fermion superconductor CeCoIn$_5$ [6,7], the experimental difficulty arises from unavoidable disorder effects and orbit-paramagnetic depairings close to the upper critical Zeeman field.

An ultracold atomic Fermi gas has proven to be an ideal tabletop system for the pursuit of FFLO superfluidity [4]. Although it is largely analogous to an electronic superconductor, the high controllability in interactions, spin populations, and purity leads to a number of unique experimental advances [8]. Indeed, following theoretical proposals by Orso [9] and the present authors [10–12], strong experimental evidence for the FFLO pairing has been observed in a one-dimensional spin-imbalanced Fermi gas of $^6$Li atoms [13]. In three dimensions (3D), unfortunately, the FFLO phase occupies only an extremely small volume in parameter space [14,15] and thus is impossible to observe experimentally [16,17].

In this Rapid Communication, we predict that the FF superfluid could be easily observed in a resonantly interacting 3D atomic Fermi gas with synthetic spin-orbit coupling. This system was recently realized experimentally by Wang et al. [18] and and by Cheuk et al. [19], using a two-photon Raman process. Our main result is summarized in Fig. 1. At experimentally accessible temperatures ($T \sim 0.05T_F$) [17] the FF state occupies a major part of the phase diagram. By tuning the strength ($\Omega_R$) and detuning ($\delta$) of two Raman laser beams, the center-of-mass momentum of Cooper pairs $q$ can be as large as the Fermi momentum $k_F$. We propose that such a large FF momentum can be easily measured by momentum-resolved radio-frequency spectroscopy (see Fig. 4) [20]. We note that the emergence of a FF superfluid has also been predicted in a 3D atomic Fermi gas in the presence of two-dimensional (2D) Rashba spin-orbit coupling [21] or 3D isotropic spin-orbit coupling [22], or in a two-dimensional atomic Fermi gas with a two-photon Raman process [23]. These systems are yet to be realized experimentally. We also note that the possibility of the FF superfluidity in our setting has been discussed very recently by Shenoy [24].

We start by considering a 3D spin-orbit-coupled spin-1/2 Fermi gas of $^6$Li or $^{40}$K atoms near a broad Feshbach resonance [18,19]. Experimentally, the synthetic spin-orbit coupling is induced using two counterpropagating Raman laser beams (i.e., along the $z$ direction) that couple the two different spin states, following the same scenario as in the National Institute of Standards and Technology experiment for a $^{87}$Rb Bose-Einstein condensate (BEC) [25]. The Raman process can be described by ($\Omega_R/2) \int dx [\Psi_+^2(x) e^{2ikRx} \Psi_-(x) + \text{H.c.}]$, where $\Psi_\pm(x)$ is the creation field operator for atoms in the spin state $\sigma$, $\Omega_R$ is the coupling strength of Raman beams, and $k_R = 2\pi/\lambda_R$ is the recoil wave vector determined by the wavelength $\lambda_R$ of the two beams. Thus, during the two-photon Raman process, a momentum of $2hk_R$ is imparted to an atom while its spin is changed from $|\downarrow\rangle$ to $|\uparrow\rangle$. This creates a correlation between the spin and orbital motion, which can be seen more clearly by taking a gauge transformation, $\Psi_i(x) = e^{i\lambda_R z} \Psi_i(x)$.
The color shows the magnitude of the center-of-mass momentum of a Fermi gas at a broad Feshbach resonance and at finite detunings. For a given temperature, via first-order (dashed line) and second-order (solid line) transitions at low and high $\Omega_R$, respectively. The FF superfluid can either be gapped or gapless, as separated by the dotted-dashed line. The color shows the magnitude of the center-of-mass momentum of Cooper pairs for the FF superfluid, $q/k_F$. The FF superfluid reduces to a BCS superfluid as $q \to 0$ when $\delta = 0$. The inset shows the critical temperature at $\Omega_R = 2E_F$.

\[ \Psi_\downarrow(x) = e^{-ik_Fz^2}\psi_\downarrow(x). \]

Near a Feshbach resonance, the system may therefore be described by a single-channel model Hamiltonian $\hat{H} = \int dx [\hat{H}_0 + \hat{H}_{\text{int}}]$, where the single-particle part

\[ \hat{H}_0 = \begin{bmatrix} \hat{\xi}_k + \lambda \hat{k}_z + \delta / 2 & \Omega_R / 2 \\ \Omega_R / 2 & -\hat{\xi}_k - \lambda \hat{k}_z - \delta / 2 \end{bmatrix} \begin{bmatrix} \psi_\downarrow \\ \psi_\uparrow \end{bmatrix}, \]

and $\hat{H}_{\text{int}} = U_0 \hat{\psi}_\downarrow \hat{\psi}_\downarrow \hat{\psi}_\uparrow \hat{\psi}_\uparrow(x)$ is the interaction Hamiltonian that describes the contact interaction between two spin states with strength $U_0$. The interaction strength should be expressed in terms of the $s$-wave scattering length $a_s$, i.e., $1/U_0 = m/(4\pi\hbar^2a_s) - 1/V \sum_\mathbf{k} m/(\hbar^2k^2)$, which can be tuned precisely by sweeping an external magnetic field around the Feshbach resonance [8]. Here $V$ is the volume of the system. In the single-particle Hamiltonian (2), $\hat{k}_z = -\delta \hat{\xi}_k \equiv -\hbar^2v^2/(2m) - \mu$ after dropping a constant recoil energy, and $\delta$ is the two-photon detuning from the Raman resonance. For convenience, we have defined a spin-orbit-coupling constant $\lambda = \hbar^2k_F/m$. However, for our Raman setting it should be noted that the strength of spin-orbit coupling is determined by both the constant $\lambda$ and Raman coupling strength $\Omega_R$. In the Shanxi experiment with $^{40}$K atoms [18], the Fermi wave vector is about $k_F \approx 1.6k_F$ and the coupling strength is $\Omega_R \approx 1.5E_R \approx 0.6E_F$, in units of the recoil energy $E_R \equiv \hbar^2k_F^2/(2m)$ or Fermi energy $E_F \equiv \hbar^2k_F^2/(2m)$.

For the model Hamiltonian (1), most previous theoretical studies focused on the case with $\delta = 0$ and considered the standard BCS superfluid [26,27]. In this work, we are interested in the FF pairing state with a single valued center-of-mass momentum. Our investigation is motivated by the interesting finding that the two-body bound state at $\delta \neq 0$ always acquires a finite center-of-mass momentum along the $z$ axis [28]. At the many-body level, we therefore anticipate that at finite detunings a FF superfluid could be more favorable than a BCS superfluid [24].

By assuming a FF-like order parameter $\Delta(x) = -U_0 \langle \psi_\downarrow(x)\psi_\downarrow(x) = \Delta e^{iqz}$, we consider the mean-field decoupling of the interaction Hamiltonian, $\hat{H}_{\text{int}} \approx -[\Delta(x)\psi_\downarrow(x)\psi_\downarrow(x) + \text{H.c.}] - \Delta^2/U_0$. Then, using a Nambu spinor $\Phi(x) \equiv [\psi_\uparrow, \psi_\downarrow, \psi_\downarrow, \psi_\uparrow]^T$, the total Hamiltonian can be written in a compact form, $\hat{H} = (1/2) \int dx \Phi(x)\hat{H}_{\text{BdG}}\Phi(x) - V\Delta^2/U_0 + \sum_\mathbf{k} \hat{\xi}_k$, where the Bogoliubov Hamiltonian

\[ \hat{H}_{\text{BdG}} = \begin{bmatrix} S^+_k & \Omega_R/2 & 0 & -\Delta(x) \\ \Omega_R/2 & S^-_k & \Delta(x) & 0 \\ 0 & \Delta^*(x) & -S^+_k & -\Omega_R/2 \\ -\Delta^*(x) & 0 & -\Omega_R/2 & -S^-_k \end{bmatrix} \]

and $S^\pm_k \equiv \hat{\xi}_k \pm \lambda \hat{k}_z \pm \delta / 2$. It is straightforward to diagonalize the Bogoliubov Hamiltonian $\hat{H}_{\text{BdG}}\Phi(k) = E_{k\eta}\Phi(k)\Phi(x)$ with quasiparticle wave function $\Phi(k) = 1/\sqrt{\text{det} \left[ \hat{H}_{\text{BdG}} \right]} e^{i\eta k_F\mathbf{r} + i\eta qz}/\sqrt{2} \psi_\downarrow - \lambda e^{i\eta qz} \psi_\downarrow - \Delta / \sqrt{2} \psi_\uparrow$. The mean-field thermodynamic potential $\Omega$ at temperature $T$ is then given by

\[ \frac{\Omega}{V} = \frac{1}{2V} \left[ \sum_{k\eta} (\xi_{k+q/2} + \xi_{k-q/2}) - \sum_{k\eta} E_{k\eta} \right] - \frac{k_BT}{V} \sum_{k\eta} \ln(1 + e^{-E_{k\eta}/k_BT}) - \frac{\Delta^2}{U_0}. \]

Note that the summation over the quasiparticle energy must be restricted to $E_{\eta} \geq 0$ because of an inherent particle-hole symmetry in the Nambu spinor representation [29]. For a given set of parameters (i.e., the temperature $T$, interaction strength $1/k_Fa_s$, etc.), different mean-field phases can be determined using the self-consistent stationary conditions $\partial \Omega/\partial Q = 0$ and $\partial \Omega/\partial q = 0$, as well as the conservation of total atom number, $N = -\partial \Omega/\partial \mu$. At finite temperatures, the ground state has the lowest free energy $F = \Omega + \mu N$.

Without loss of generality, hereafter we consider the resonance case with a divergent scattering length $1/k_Fa_s = 0$ and set $T = 0.05T_F \approx 0.1T_{C,\text{MF}}$, where $T_{C,\text{MF}}$ is the mean-field critical temperature without spin-orbit coupling. According to the typical number of atoms in experiments [18,19], we shall take $k_F = k_F$, corresponding to a dimensionless spin-orbit-coupling constant $\lambda = k_F E_F \approx 2$. The Raman coupling strength $\Omega_R$ and detuning $\delta$ can be easily tuned experimentally. Thus, we focus on the phase diagram as functions of $\Omega_R$ and $\delta$.

In general, for any set of parameters there are three competing ground states that are stable against phase separation (i.e., $\partial^2 \Omega/\partial \Delta^2 \geq 0$), as shown in Fig. 2(a): normal gas ($\Delta = 0$),...
BCS superfluid ($\Delta \neq 0$ and $q \neq 0$), and FF superfluid ($\Delta \neq 0$ and $q \neq 0$). Remarkably, in the presence of spin-orbit coupling (i.e., $\Omega_R \neq 0$) the FF superfluid is always more favorable in energy than the standard BCS pairing state at finite detunings [Fig. 2(b)]. It is easy to check that the superfluid density of the BCS pairing state in the axial direction becomes negative (i.e., $\delta \Omega / \Omega_R < 0$), signaling the instability towards a FF superfluid. Therefore, experimentally the Fermi cloud would always condense into a FF superfluid at finite Raman detunings. In Fig. 1, we report a low-temperature phase diagram that could be directly observed in current experiments. The FF superfluid occupies the major part of the phase diagram.

The greatly enlarged parameter space for a FF superfluid can be qualitatively understood from the change of the Fermi surface due to spin-orbit coupling, as discussed in the pioneering work by Barzykin and Gor’kov [30]. In our case, the single-particle energy spectrum takes the form $E_{k\pm} = \frac{\hbar^2 k^2}{2m} \pm \sqrt{(\Omega_R/2)^2 + (\lambda k_z + \delta/2)^2}$, where “±” stand for two helicity bands. At large Raman couplings ($\Omega_R \gg \lambda k_F, \delta$),

we find

$$E_{k\pm} \approx \frac{\hbar^2 (k_x^2 + k_y^2)}{2m} + \frac{\hbar^2}{2m} \left( k_x \pm \frac{q}{2} \right)^2 \pm \frac{\Omega_R}{2},$$

where $q = 2k_R \delta / \Omega_R$. Thus, the Fermi surfaces remain approximately circular but the centers of the two Fermi surfaces are shifted by $q/2$ along the $z$ axis in opposite directions. In this situation, we may gain condensate energy by shifting the two Fermi surfaces by an amount $\pm (q/2)e_z$, respectively (see Ref. [31] for a similar analysis for a 3D superconductor with Rashba spin-orbit coupling). Here $e_z$ is the unit vector in the $z$ axis. The fermionic pairing then occurs between single-particle states of $k + (q/2)e_z$ and $-k + (q/2)e_z$, leading to a FF order parameter that has a spatial distribution $\Delta(x) = \Delta e^{iqz}$. The direction of the FF momentum is uniquely fixed by the form of spin-orbit coupling (i.e., $\lambda k_z$) and its magnitude is roughly proportional to the detuning $\delta$.

At small Raman couplings $\Omega_R \sim 0$, however, the above argument becomes invalid. Indeed, at $\Omega_R = 0$ where the spin-orbit coupling is absent, the instability to inhomogeneous superfluidity is driven solely by the detuning. The FF superfluid occurs in a narrow window (not shown in Fig. 1): $\delta_{cs} \ll \delta \ll 1.204 E_F$, where $\delta_{cs} \approx 1.162 E_F$ is the Clogston limit for a unitary Fermi gas [15]. The direction of the FF momentum cannot be specified, indicating that a LO state with periodic stripe structure is more preferable [32]. In our calculations, we do not consider such a LO superfluid that exists in a small wedge near $\Omega_R \sim 0$ and $\delta \sim 1.2 E_F$. Due to this simplification, at low Raman coupling strengths the FF superfluid does not transform continuously into a normal gas. We note that, the competition between FF and LO phases in a Rashba spin-orbit-coupled 3D superconductor was recently studied by Agterberg and Kaur [31]. The LO stripe phase was found to cease to exist at large detunings.

The FF superfluid in Fig. 1 can have different topological structures in the quasiparticle excitation spectrum, as illustrated in Fig. 3. At small Raman couplings [Fig. 3(a)], the FF superfluid has a negligible center-of-mass momentum and therefore behaves very similar to a standard gapped superfluid.

![Fig. 2](image-url) (Color online) (a) Landscape of the thermodynamic potential $[\Omega(\Delta, q) - \Omega(0, 0)](N E_F)$ at $\Omega_R = 2E_F$ and $\delta = 0.68E_F$. The chemical potential is fixed to $\mu = -0.471E_F$. The competing ground states include (i) a normal Fermi gas with $\Delta = 0$; (ii) a fully paired BCS superfluid with $\Delta \neq 0$ and $q = 0$; and (iii) a finite momentum paired FF superfluid with $\Delta \neq 0$ and $q \neq 0$. (b) The free energy of different competing states as a function of the detuning at $\Omega_R = 2E_F$. The inset shows the detuning dependence of the order parameter and momentum of the FF superfluid state.
BCS superfluid, while at large Raman coupling [Fig. 3(b)], the sizable FF momentum features a gapless spectrum. The nodal points with zero-excitation energy are located close to the $k_z = 0$ axis, along which the dispersion relations take the form $E(k_\perp, k_z) \approx \pm \sqrt{\Delta_0^2 + (\delta E)^2 + \Omega_{1R}^2/2} \pm \sqrt{\Delta_0^2 + (h^2 k_0^2/(2m) - \mu + h^2 q^2/8m)^2}$. Apparently, these nodal points would appear when the effective Zeeman field $\sqrt{\Delta_0^2 + \Omega_{1R}^2/2}$ is sufficiently large. In Fig. 1, the gapped and gapless FF superfluids are separated by a dot-dashed line. The topological transition from gapped to gapless phases is continuous and could be revealed by thermodynamic measurements through the atomic compressibility, spin susceptibility, and momentum distribution.

We now consider how to experimentally detect a FF superfluid through momentum-resolved rf spectroscopy [20], which is a cold-atom analog of the widely used angle-resolved photoemission spectroscopy in solid-state systems. In such a measurement, one uses a rf field with frequency $\omega$ to break a Cooper pair and transfer the spin-down atom to an unoccupied third hyperfine state (3). The momentum distribution of the transferred atoms is then measured by absorption imaging after a time-of-flight expansion. The rf Hamiltonian can be written as $\mathcal{H}_{\text{rf}} \propto \int dx [e^{-ikz\hat{\Sigma}_3(x)} \Psi(x) + \text{H.c.}]$, where $\Psi_3(x)$ is the field operator which creates an atom in (3). The momentum transfer $k_{RF} \hbar e_z$ in $\mathcal{H}_{\text{rf}}$ arises from the gauge transformation. For a weak rf drive, the number of transferred atoms can be calculated using linear response theory

$$\Gamma(k, \omega) = A_{+1} \left[ k + \left( k_R - \frac{q}{2} \right) e_z \hat{\Sigma}_k - \omega \right] f(\xi_k - \omega).$$

Here $A_{+1}(k, \omega) = \sum_{q} | u_{k0(\pm)} |^2 \delta(\omega - E_{k0})$ is the single-particle spectral function of spin-down atoms, $\xi_k = \hbar^2 k^2/(2m) - \mu$, and $f(x) = 1/(e^{x/k_B T} + 1)$ is the Fermi-Dirac distribution function. For a FF superfluid, as Cooper pairs now carry a finite center-of-mass momentum $q = q e_z$, the transferred atoms acquire an overall momentum $q/2$. As a result, there is a $q/2$ shift in the transferred strength Eq. (5), which in principle could be experimentally measured.

In Fig. 4, we report the momentum-resolved rf spectroscopy along the $z$ direction $\Gamma(k, \omega) \equiv \sum_{k_z} \Gamma(k, \omega)$ at $\Omega_{RF} = 2E_F$ on a logarithmic scale. Quite generally, there are two contributions to the spectroscopy, corresponding to two different final states: After a Cooper pair is broken by a rf-photon, the remaining spin-up atoms can be in different helicity bands [33]. These two contributions are well separated in the frequency domain, with peak positions indicated by the symbols “+” and “−”, respectively. Interestingly, at finite detuning with a sizable FF momentum $q$, the peak positions of the two contributions are shifted roughly in opposite directions by an amount $q/2$. This provides clear evidence for observing the FF superfluid.

In summary, we have shown that the condensate state of a spin–orbit-coupled Fermi gas is the long-sought inhomogeneous Fulde-Ferrell superfluid. Our prediction can be readily examined in current experiments, where a three-dimensional spin-orbit-coupled atomic Fermi gas is created using a two-photon Raman process [18,19]. The finite center-of-mass momentum carried by the inhomogeneous superfluid can be unambiguously measured by momentum-resolved radio-frequency spectroscopy. Our work complements relevant studies of solid-state systems, in which inhomogeneous superfluidity was predicted to be enhanced by Rashba spin-orbit interaction [30], but was difficult to confirm experimentally. The controllable setting of a spin-orbit-coupled atomic Fermi gas opens a new direction to explore the fascinating inhomogeneous superfluidity.

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INHOMOGENEOUS FULDE-FERRELL SUPERFLUIDITY IN . . .

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