Instability of Fulde-Ferrell-Larkin-Ovchinnikov states in atomic Fermi gases in three and two dimensions

Jibiao Wang,^{1,2,3} Yanming Che,^{1,2} Leifeng Zhang,^{1,2} and Qijin Chen^{1,2,4,*}

¹Department of Physics and Zhejiang Institute of Modern Physics, Zhejiang University, Hangzhou, Zhejiang 310027, China ²Synergetic Innovation Center of Quantum Information and Quantum Physics, Hefei, Anhui 230026, China

³Laboratory of Quantum Engineering and Quantum Metrology, School of Physics and Astronomy, Sun Yat-Sen University (Zhuhai Campus),

Zhuhai, Guangdong 519082, China

⁴James Franck Institute, University of Chicago, Chicago, Illinois 60637, USA

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The exotic Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) states have been actively searched for experimentally since the mean-field based FFLO theories were put forward half a century ago. Here, we investigate the stability of FFLO states in the presence of pairing fluctuations. We conclude that FFLO superfluids cannot exist in continuum in three and two dimensions, due to their intrinsic instability, associated with infinite *quantum* degeneracy of the pairs. These results address the absence of convincing experimental observations of FFLO phases in both condensed matter and in ultracold atomic Fermi gases with a population imbalance. We predict that the true ground state has a pair momentum distribution highly peaked on an entire constant energy surface.

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I. INTRODUCTION

The Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) states, which were first predicted by Fulde and Ferrell [1] (FF) and Larkin and Ovchinnikov [2] (LO) in an *s*-wave superconductor in the presence of a Zeeman field over fifty years ago, have attracted enormous attention in condensed matter physics [3–6], including heavy-fermion [7–9], organic [10–13], and high- T_c superconductors [14], nuclear matter [15] and color superconductivity [16], and, more recently, in ultracold Fermi gases [17]. Conventional BCS superfluidity originates from Bose-Einstein condensation (BEC) of Cooper pairs at zero momentum. In contrast, in these exotic states, Cooper pairs condense either at a finite momentum \mathbf{q} , with a one-plane-wave order parameter $\Delta(\mathbf{r}) = \Delta_0 e^{i\mathbf{q}\cdot\mathbf{r}}$ or at momenta $\pm \mathbf{q}$, with a standing-wave order parameter $\Delta(\mathbf{r}) = \Delta_0 \cos(\mathbf{q}\cdot\mathbf{r})$ for the FF and LO states, respectively.

These exotic superfluids have been actively searched for over the past half century. The strongest signatures of FFLO states come from heavy fermion [7,8,18–24], and organic superconductors [25–30]. References [3] and [31] give nice reviews of possible experimental observations as well as theoretical surveys of FFLO states.

For heavy fermions, Gloos *et al.* [18] and Huxley *et al.* [19] reported possible FFLO states in UPd₂Al₃ and CeRu₂, respectively. However, they were argued to be inconsistent with theory [32,33]. The majority of heavy fermion signatures come from CeCoIn₅. Radovan *et al.* [7] reported the first possible thermodynamic signature of FFLO states in CeCoIn₅ via heat capacity and magnetization measurements. However, they considered an FFLO wave vector perpendicular to the highly two-dimensional Fermi surface, which is rather different from

the case of a Fermi surface mismatch considered in the original FF and LO ideas. Bianchi et al. [22] reported specific heat measurements of CeCoIn₅ near the superconducting critical field H_{c2} , with magnetic fields in different directions, and at temperatures down to 50 mK, and interpreted an observed second specific heat anomaly as signatures of FFLO states, which seems to have been under doubt in later works [8]. Kumagai et al. [20] reported signatures that can be associated with FFLO states in CeCoIn₅ via NMR studies of ¹¹⁵In in a perpendicular field. Mitrovic et al. [21] observed a spin susceptibility enhancement in possible FFLO states in CeCoIn₅. Kenzelmann et al. [8] reported an observation of coexisting magnetic order and superconductivity in CeCoIn₅, and suggested a form of superconductivity (referred to as **Q** phase) that is associated with a nonvanishing momentum. However, they also noticed discrepancies between theory and their own experimental observations. Recently, Tokiwa et al. [23] performed high-precision studies of the isothermal field dependence of the entropy in CeCoIn5, derived from combined specific heat and magnetocaloric effect measurements, and did not observe an additional entropy contribution upon tuning at constant temperature by the magnetic field from the homogeneous superconducting into the presumed FFLO state. In addition, for $H \parallel [100]$, a reduction of entropy was found that quantitatively agrees with the expectation for spin-densitywave order without FFLO superconductivity. Very recently, Kim et al. [24] observed a reduction of thermal conductivity in the magnetic \mathbf{Q} phase in CeCoIn₅, and argued that an additional order (intertwined with possibly an FFLO state) is needed in order to account for the data [34].

In organic superconductors, Mayaffre *et al.* [25] reported enhancement of NMR relaxation rate in a possible FFLO phase in κ -(BEDT-TTF)₂Cu(NCS)₂, and argued that an Andreev bound state can be used as a hallmark of FFLO states. However, they failed to establish a hallmark of an Andreev bound state,

^{*}Corresponding author:qchen@uchicago.edu

such as a zero bias conductance peak. Koutroulakis et al. [26] reported an NMR study of the quasi-two-dimensional (2D) organic superconductor β'' -(ET)₂SF₅CH₂CF₂SO₃ and associated a possible second transition as a signature of FFLO states. Uji et al. [35] studied the interlayer resistance in high magnetic fields in λ -(BETS)₂FeCl₄, and reported results that are consistent with pinning interactions between the vortices penetrating the insulating layers and the order parameter of the FFLO state. Yonezawa et al. [28] observed anomalous in-plane anisotropy of the onset of superconductivity in (TMTSF)₂ClO₄, and related it to an occurrence of FFLO phases. Bergk et al. [29] reported magnetic torque evidence for the FFLO state in the layered organic superconductor κ -(BEDT-TTF)₂Cu(NCS)₂ from rf penetration depth measurements with a tunnel diode oscillator in a pulsed magnetic field. Coniglio et al. [30] reported signatures of FFLO states in λ -(BETS)₂GaCl₄ also from rf penetration depth measurements. Agosta et al. [13] analyzed these recent rf penetration depth measurements and found possible disagreement between rf penetration depth and specific heat measurements.

While the existence of an FFLO phase is currently under active debate, we note that the experimental phase diagrams were inferred only from NMR, magnetic torque, and specific heat data, rather than from more definitive phase sensitive probes and Meissner effects. The phase diagram may be further complicated by possible pseudogap phenomena, which can persist above T_c , associated with these heavy-fermion or organic superconductors. Thus far, *there has been no solid experimental evidence for the FFLO superfluid states from condensed matter systems*.

With the easy tunability of various control parameters, especially population imbalance [36,37], ultracold Fermi gases have provided a much greater opportunity and led to a high expectation for finding the FFLO states. Despite many theoretical studies in this regard, both in a 3D homogeneous case [38–44] and in a trap [45–48], as well as in more complex systems, such as Fermi-Fermi mixtures [43,49,50] or optical lattices [51], the experimental search for these exotic states in atomic Fermi gases has not been successful [52,53].

In this paper, we investigate the stability of FFLO states with an *s*-wave pairing symmetry against ubiquitous and inevitable pairing fluctuations, and show that *FFLO states are intrinsically unstable at any finite temperature T due to pairing fluctuations*. We begin with simple arguments based on general physical grounds, and then demonstrate this instability with concrete calculations using several different pairing fluctuation theories, some of which [36,54] have been applied successfully to the BCS-BEC crossover physics. Furthermore, we predict that, in the mean-field FFLO regime in $D \ge 2$ (spatial) dimensions, the true ground state has a pair momentum distribution highly peaked on an entire (D - 1)-dimensional constant energy surface (CES), which can be readily tested experimentally.

We shall mainly work with the 3D continuum case, then generalize to 2D, and finally discuss possible complications in the presence of a lattice potential. We note that both the FF and LO states as well as higher-order crystalline states in the literature are essentially constructed at the mean-field level; their stability does not appear to have been *properly* tested against pairing fluctuations.

II. GENERAL PICTURE AND ARGUMENTS ON PHYSICAL GROUNDS

We first argue intuitively that, unlike the BCS case, there is no compelling constraint requiring condensation into a superfluid having a finite momentum. To see this, consider a single minority fermion in the presence of an isotropic majority Fermi sphere in a homogeneous 3D continuum, without a mass imbalance. For weak pairing, the ground state is a polaron moving in the Fermi sea. When the interaction becomes just strong enough, the minority fermion will pair with a majority fermion near the Fermi surface to form a (meta-)stable pair. To minimize the system's energy, the pair dispersion will reach a minimum at a finite momentum of $q \approx k_F^{\uparrow}$, where k_{E}^{\uparrow} is the majority Fermi wave vector [55]. Obviously, adding more minority fermions to form a two-component gas with a high population imbalance will not lead to abrupt changes, because the majority background is barely modified by a small percentage of minority fermions and the influence of the correlation among minority fermions is negligible. For weak interactions, the ground state will be minority polarons moving in the majority Fermi sea. An example of this case is the destruction of the mean-field FFLO solution by an overly strong magnetic field in a superconductor; the pairing strength is simply not strong enough to overcome the big Fermi surface mismatch, which is necessary to form a pair. For very strong interactions in the BEC regime, the two-body binding energy dominates the Fermi energy, so that a non-FFLO, but polarized (i.e., Sarma [56]) superfluid will emerge at low T [57].

For intermediate pairing strengths, where the majority Fermi surface still exists, pairs will first form in the normal state without phase coherence, with momenta evenly distributed in all possible directions. As the temperature drops from very high T into the Fermi degenerate regime such that the Fermi surface mismatch has a strong effect, the pair dispersion starts to develop a minimum at a finite momentum q. As T decreases further, the system will either phase separate into a 50-50 mixture forming a BCS superfluid plus a majority normal Fermi gas, or try to enter an FF or LO state. At issue is whether there is a compelling reason for these finite momentum pairs to Bose condense. For the latter case, since the pair energy reaches its minimum on a 2D surface in momentum space (which happens to be spherical surface \mathbb{S}^2 for a 3D homogeneous Fermi gas), one finds that no condensation is needed at any finite T, in order to satisfy the appropriate particle number constraints; pairing fluctuations destroy Bose condensation of the pairs.

More concretely, as a starting point, the mean-field FFLO phase can be regarded as a condensate of pairs at a finite momentum \mathbf{q} (or $\pm \mathbf{q}$). Importantly, near the minimum of the pair dispersion, the pairs have a finite density of states (DOS). This reflects an infinite degeneracy associated with all possible directions for momentum \mathbf{q} in the thermodynamic limit. As a result, these pairs cannot condense so that fluctuations destroy condensation into a single quantum state.

We note that the infinite quantum degeneracy [58] and finite DOS on the CES occur at any dimensionality above 1D. In 1D, the "CES" becomes merely two points, and the degeneracy reduces to 2, so that the continuous *transverse* momentum integral on the "CES" becomes a discrete sum, and thus no divergence associated with a finite DOS will occur. Nevertheless, there is no long-range order in 1D in any case, due to the diverging DOS associated with the remaining *longitudinal* dimension (which is supposed to be perpendicular to the two-point "CES"). While one may consider such a situation as a stripe state, it is certainly not an FFLO superfluid, and thus is excluded from our consideration.

To be more concrete, one may approximate the pair dispersion in a balanced isotropic system by $\Omega_{\mathbf{p}} = p^2/(2M) - \mu_{\text{pair}}$ for small momentum p, with an effective pair mass M and a bosonic chemical potential μ_{pair} . In the case of high population imbalance, slightly above the FFLO superfluid transition T_c (if it exists), this dispersion can be approximated by

$$\Omega_{\mathbf{p}} \approx C(p^2 - q^2)^2 - \mu_{\text{pair}}, \qquad (1)$$

with a constant coefficient *C*, and *q* is the radius of the minimum pair energy surface. Starting from such a state, one wants to seek an FFLO superfluid solution such that μ_{pair} vanishes at a finite $T_c > 0$ while *q* remains nonzero. However, to satisfy the finite pair number constraint with the finite DOS at the lowest energy, it is necessary that $\mu_{\text{pair}} < 0$ at all T > 0 so that Bose condensation of the pairs (or spontaneous symmetry breaking) never occurs at any finite *T*.

Mathematically, this is similar to the derivation of the Mermin-Wagner theorem for the absence of long-range order in 2D, where the DOS of a quadratic dispersion is finite at zero momentum. The same conclusion can also be obtained equivalently following Hohenberg's argument [59] once the simple quadratic bosonic dispersion is replaced by one that has a minimum on a finite-size 2D surface.

III. INSTABILITY OF THE MEAN-FIELD FFLO STATE

A. Mean-field treatment of the one-plane-wave FFLO state

In this section, we examine what would happen if one presumed an FFLO state at low T with a wave vector \mathbf{q} pointing in a symmetry broken direction. We consider a two-component homogeneous Fermi gas with a contact potential [60] of strength U < 0 with a population imbalance η in isotropic 3D, and begin by presenting the mean-field solutions, and then show that the mean-field FFLO phase will eventually be destroyed by pairing fluctuations.

Consider the simplest symmetry breaking with only one wave vector \mathbf{q} , i.e., the (assumed) FF states. Momentum \mathbf{k} states pair with those having $\mathbf{q} - \mathbf{k}$ so that condensed Cooper pairs have a nonzero momentum \mathbf{q} (note that setting $\mathbf{q} = 0$ would give us the formalism for the Sarma superfluid state) with a free atom dispersion $\xi_{\mathbf{k},\sigma} = \mathbf{k}^2/2m_{\sigma} - \mu_{\sigma}$, where m_{σ} and μ_{σ} are the mass and chemical potential for (pseudo)spin $\sigma = \uparrow, \downarrow$, respectively. We set the volume V = 1, $\hbar = k_B = 1$, and use the four-vector notations $K \equiv (\mathbf{k}, i\omega_n)$, $P \equiv (\mathbf{p}, i\Omega_l)$, where ω_n (Ω_l) is the odd (even) Matsubara frequency.

The mean-field self-energy [36] takes the simple BCSlike form, $\Sigma_{\sigma}(K) = -\Delta^2 G_{0\bar{\sigma}}(Q - K)$, with $Q \equiv (0,\mathbf{q})$, and bare Green's function $G_{0\sigma}^{-1}(K) = i\omega_n - \xi_{\mathbf{k},\sigma}$. The full Green's



FIG. 1. Typical quasiparticle dispersion $E_{\mathbf{k},\sigma}$ in the FFLO phases, shown here along the $\mathbf{k} \parallel \mathbf{q}$ direction in the unitary case with population imbalance $\eta = 0.75$ and $T/T_F = 0.01$, with $q = 0.71k_F$ and $m_{\sigma} = m$. The lower branch, $E_{\mathbf{k},\downarrow}$, is gapless. The units for energy and momentum are E_F and k_F , respectively.

functions G(K) are given by

$$G_{\uparrow}(K) = \frac{u_{\mathbf{k}}^2}{i\omega_n - E_{\mathbf{k},\uparrow}} + \frac{v_{\mathbf{k}}^2}{i\omega_n + E_{\mathbf{k},\downarrow}},$$
 (2a)

$$G_{\downarrow}(K) = \frac{u_{\mathbf{q}-\mathbf{k}}^2}{i\omega_n - E_{\mathbf{q}-\mathbf{k},\downarrow}} + \frac{v_{\mathbf{q}-\mathbf{k}}^2}{i\omega_n + E_{\mathbf{q}-\mathbf{k},\uparrow}}, \qquad (2b)$$

where $u_{\mathbf{k}}^2 = (1 + \xi_{\mathbf{kq}}/E_{\mathbf{kq}})/2$, $v_{\mathbf{k}}^2 = (1 - \xi_{\mathbf{kq}}/E_{\mathbf{kq}})/2$, $E_{\mathbf{kq}} = \sqrt{\xi_{\mathbf{kq}}^2 + \Delta^2}$, $E_{\mathbf{k},\uparrow} = E_{\mathbf{kq}} + \zeta_{\mathbf{kq}}$, $E_{\mathbf{k},\downarrow} = E_{\mathbf{kq}} - \zeta_{\mathbf{kq}}$, $\xi_{\mathbf{kq}} = (\xi_{\mathbf{k},\uparrow} + \xi_{\mathbf{q}-\mathbf{k},\downarrow})/2$, and $\zeta_{\mathbf{kq}} = (\xi_{\mathbf{k},\uparrow} - \xi_{\mathbf{q}-\mathbf{k},\downarrow})/2$. Here, $E_{\mathbf{k},\sigma}$ may not be gapped. Shown in Fig. 1 are the quasiparticle dispersions along the $\mathbf{k} \parallel \mathbf{q}$ direction at unitarity with a population imbalance $\eta = 0.75$ (see below for definition) and $q/k_F = 0.71$ at low *T*. The lower branch, $E_{\mathbf{k},\downarrow}$, is gapless. It is the gapless region in the momentum space that accommodates the excessive majority fermions.

Note that it is often assumed in the literature that the LO states exhibit a gap in a standing wavelike pattern in real space, whereas the FF states are fully gapped with only an oscillating phase. Here we see that the order parameter may differ from the actual excitation gap completely.

The mean-field solution for the gap equation can be written as a Thouless criterion,

$$t^{-1}(0,\mathbf{q}) = U^{-1} + \chi(0,\mathbf{q}) = 0,$$
(3)

with $t(P) = U/[1 + U\chi(P)]$ representing the mean-field compatible *T* matrix, and

$$\chi(P) \equiv \sum_{K,\sigma} G_{0\sigma}(P-K)G_{\bar{\sigma}}(K)/2$$
(4)



FIG. 2. $T - \eta$ phase diagram of a homogeneous Fermi gas with equal mass at $1/k_F a = -0.5$, 0, and 0.1, corresponding to near-BCS, unitary, and near-BEC cases, respectively. Here, "PS" denotes phase separation. An FF phase (yellow shaded) exists in the low-T and relatively high- η regime, while they become unstable against phase separation in the dotted region. The gap vanishes along the black solid line, which separates the FF phase and normal Fermi gas phase.

being the pair susceptibility. (Here, spin $\bar{\sigma}$ is the opposite of spin σ .) This leads to the usual gap equation,

$$\frac{m_r}{2\pi a} = \sum_{\mathbf{k}} \left[\frac{1}{2\epsilon_{\mathbf{k}}} - \frac{1 - 2\bar{f}(E_{\mathbf{kq}})}{2E_{\mathbf{kq}}} \right],\tag{5}$$

where $\epsilon_{\mathbf{k}} = k^2/4m_r$ with reduced mass m_r . Here, $\bar{f}(x) = [f(x + \zeta_{\mathbf{kq}}) + f(x - \zeta_{\mathbf{kq}})]/2$, $f(x) = 1/(e^{x/T} + 1)$ is the Fermi distribution function and U has been replaced by the *s*-wave scattering length *a* via $U^{-1} = m_r/2\pi a - \sum_{\mathbf{k}} 1/2\epsilon_{\mathbf{k}}$.

From the number constraint $n_{\sigma} = \sum_{K} G_{\sigma}(K)$, we obtain the total number density $n = n_{\uparrow} + n_{\downarrow}$ and density difference $\delta n = n_{\uparrow} - n_{\downarrow} \equiv \eta n$,

$$n = \sum_{\mathbf{k}} \left[\left(1 - \frac{\xi_{\mathbf{k}\mathbf{q}}}{E_{\mathbf{k}\mathbf{q}}} \right) + 2\bar{f}(E_{\mathbf{k}\mathbf{q}}) \frac{\xi_{\mathbf{k}\mathbf{q}}}{E_{\mathbf{k}\mathbf{q}}} \right],\tag{6}$$

$$\eta n = \sum_{\mathbf{k}} [f(E_{\mathbf{k},\uparrow}) - f(E_{\mathbf{k},\downarrow})]. \tag{7}$$

The FFLO wave vector **q** can be determined via $\frac{\partial \chi(0,\mathbf{p})}{\partial \mathbf{p}}|_{\mathbf{p}=\mathbf{q}} = 0$, or equivalently via minimizing the thermodynamic potential with respect to **q** [43], as

$$\sum_{\mathbf{k}} \left[\frac{\mathbf{k}}{m_{\uparrow}} (n_{\mathbf{k}\mathbf{q}} + \delta n_{\mathbf{k}\mathbf{q}}) + \frac{\mathbf{q} - \mathbf{k}}{m_{\downarrow}} (n_{\mathbf{k}\mathbf{q}} - \delta n_{\mathbf{k}\mathbf{q}}) \right] = 0, \quad (8)$$

where $n_{\mathbf{kq}}$ and $\delta n_{\mathbf{kq}}$ represent the summands of Eqs. (6) and (7), respectively. Equations (5)–(8) form a closed set, and can be used to solve the mean-field solution of the one-plane-wave FFLO state, e.g., for $(\mu_{\uparrow}, \mu_{\downarrow}, T_c, \mathbf{q})$ with $\Delta = 0$, and for $(\mu_{\uparrow}, \mu_{\downarrow}, \Delta, \mathbf{q})$ at $T < T_c$.

B. Mean-field phase diagrams of the FFLO states

We present in Fig. 2 the calculated mean-field $T - \eta$ phase diagram for a homogeneous Fermi gas with equal mass for the near-BCS ($1/k_F a = -0.5$), unitary, and near-BEC ($1/k_F a = 0.1$) cases, respectively. A mean-field FF state in the low-*T* and relatively high- η regime exists for all three cases. For lower η , the FF states become unstable against phase separation (PS) at low *T* (dotted region), and these two phases are divided by the green line, as determined by the phase separation condition, which can be found in Refs. [38,43]. The red line denotes where **q** drops to zero. Along the black phase boundary, which

separates FF from the normal state, the gap vanishes, and q is finite and decreases continuously with T from $q = k_F^{\uparrow} - k_F^{\downarrow}$ at T = 0 till it intersects the q = 0 line. It is the small gap case near this boundary that was addressed by the LO paper [2]. Here we focus on the FF phase, not showing other phases at lower population imbalance and higher T. The FF state in the near-BEC case has a substantially smaller phase space, which quickly shrinks to zero towards the BEC regime.

It should be noted that the green PS instability boundary corresponds to the temperature where the longitudinal superfluid density is rendered negative by collective modes in a very recent work [61]. The counterpart (and more complete) phase diagrams for Fermi-Fermi mixtures such as ⁶Li-⁴⁰K can be found in Ref. [43].

C. Instability of the mean-field solution within a G_0G approximation of the pairing fluctuations

Next, we consider pairing fluctuations in a fashion which is *consistent with this mean-field treatment* [54]. (This same approach was used to address a variety of ultracold Fermi gas experiments without [36,62,63] and with population [57,64] and/or mass imbalances [65,66].) We extract the *generic* pair dispersion $\Omega_{\mathbf{p}}$ via Taylor-expanding

$$t^{-1}(\Omega, \mathbf{p}) \approx a_0(\Omega - \Omega_{\mathbf{p}} + \mu_{\text{pair}}),$$
 (9)

after analytic continuation, where coefficients a_0 , B_{\parallel} , B_{\perp} , and μ_{pair} are self-consistently determined during the expansion, with $\mu_{\text{pair}} = 0$ at $T \leq T_c$, and $\mu_{\text{pair}} < 0$ above T_c [67]. Near $\mathbf{p} = \mathbf{q}$,

$$\Omega_{\mathbf{p}} = -[\chi(0, \mathbf{p}) - \chi(0, \mathbf{q})]/a_0 \approx B_{\parallel}(p_{\parallel} - q)^2 + B_{\perp} p_{\perp}^2 \,.$$
(10)

Here, the subscript $\parallel (\perp)$ denotes the direction parallel with (perpendicular to) the wave vector **q**. One can then deduce the number of noncondensed pairs, via

$$n_{\text{pair}} = \sum_{\mathbf{p}} b(\Omega_{\mathbf{p}} - \mu_{\text{pair}}), \qquad (11)$$

where $b(x) = 1/(e^{x/T} - 1)$ is the Bose distribution function.

We show in Fig. 3 a 3D plot of the pair dispersion Ω_p in the mean-field FFLO phase at unitarity, with $\eta = 0.75$



FIG. 3. Typical pair dispersion $\Omega_{\mathbf{p}}$ in the FFLO phases. Shown here is the unitary case with $\eta = 0.75$ and $T/T_F = 0.01$. The color coding is such that $\Omega_{\mathbf{p}}$ increases with the wavelength of the light. The units for energy and momentum are E_F and k_F , respectively.

and $T/T_F = 0.01$, which has a solution of $q = 0.71 k_F$ (and $\mu_{\text{pair}} = 0$). Here, $\mathbf{q} \parallel \hat{z}$, and θ measures the polar angle between pair momentum \mathbf{p} and the FFLO momentum \mathbf{q} . Evidently, the SO(3) rotational symmetry is broken. We find that, while in all radial directions, $\Omega_{\mathbf{p}}$ reaches a minimum near $p = |\mathbf{p}| \approx q$, this minimum itself reaches a maximum at $\theta = 0$. Therefore the $\mathbf{p} = \mathbf{q}$ point is *not* a global minimum of the pair energy, in contradiction with our assumption at the beginning. This conclusion is also manifested in an alternative presentation of this same pair dispersion as a function in the θ -p plane, as shown in Fig. 9 in Appendix.

The pair dispersions for the near-BCS and near-BEC cases are similar, which are also shown in Appendix (in Fig. 10). To see the pair dispersion with a better quantitative resolution, we plot in Fig. 4 the radial minimum, $\Omega_{p=q}$, as a function of θ (black solid curve), i.e., along the p = q (half-)circle (in Fig. 3). We also show the near-BCS case (red dashed line) at $1/k_F a = -1/2$ with $\eta = 0.4$. In *neither* case, the $\mathbf{p} = \mathbf{q}$ point is the global minimum of the pair dispersion, which contradicts



FIG. 4. Radial minimum of the pair energy, $\Omega_{p=q}$, as a function of θ (black solid line), i.e., along the bottom half circle in Fig. 3. Also shown is the near-BCS case (red dashed line) at $1/k_F a = -1/2$ with $\eta = 0.4$. Here, $T/T_F = 0.01$.

our presumption that the FFLO state is a spontaneously broken symmetry state. This means that the FFLO states found at the mean-field level are *not* stable once pairing fluctuations are taken into account.

D. Instability of the mean-field FFLO solution using alternative treatments of pairing fluctuations

While it is not entirely consistent to consider pairing fluctuations in other approximations, for completeness, we perform similar calculations using an alternative (i.e., GG) scheme with a substituted pair susceptibility

$$\chi_{GG}(P) = \sum_{K\sigma} G_{\sigma}(P - K)G_{\bar{\sigma}}(K)/2.$$
(12)

This has been known as the FLEX approximation [68], and has been used by various authors in the study of BCS-BEC crossover. This enables us to compare the results between these two schemes.

Similar to the G_0G scheme of the *T*-matrix approximation, one can also extract the pair dispersion from the expansion of the counterpart inverse *T* matrix in the *GG* and G_0G_0 schemes, as in Eq. (9), leading to $\Omega_{\mathbf{p}} = -[1/U + \chi_{GG}(0, \mathbf{p})]/a_0$. With no doubt, the coefficient a_0 will be quantitatively different. To make different plots comparable in numerical values, here we use the a_0 from the G_0G scheme to plot the pair dispersion. The result at unitarity is shown in Fig. 5, where the values of the chemical potentials μ_{σ} , the gap Δ , and the vector \mathbf{q} were the same as in Fig. 3 for the G_0G case. Evidently, the pair dispersion for the *GG* case is similar to that of the G_0G case, confirming that the $\mathbf{p} = \mathbf{q}$ point is only a saddle point, rather than a global minimum of $\Omega_{\mathbf{p}}$.

Shown in Fig. 6 are representative pair dispersions Ω_p as a function of p along the radial directions at three representative angles, $\theta = 0$ (black), $\pi/2$ (red), and π (blue), for both G_0G (solid) and GG (dashed lines) schemes. Both were calculated using the same unitary mean-field solution as in Fig. 3. Note that for the GG scheme, the pair dispersion along $\theta = 0$ does *not* touch zero at its minimum, due to its inconsistency with



FIG. 5. Typical pair dispersion Ω_p in the mean-field FFLO phases for the *GG* scheme of pairing fluctuation theories. Shown here is the unitary case with $\eta = 0.75$ and $T/T_F = 0.01$. The conventions on color coding and units are the same as in Fig. 3.



FIG. 6. Pair dispersion Ω_p in the mean-field FFLO phase at unitarity with $\eta = 0.75$ as a function of p along different polar angles $\theta = 0$ (black), $\pi/2$ (red), and π (blue), for both the G_0G (solid) and GG (dashed lines) schemes of T-matrix theories. For neither scheme the minimum energy along the **q** direction is a global minimum. Here the specific parameters are labeled.

the mean-field BCS gap equation. Nevertheless, common to both schemes is that the minimum pair energy along $\theta = 0$ is higher than along other directions, indicating that $\mathbf{p} \parallel \mathbf{q}$ is not a stable symmetry breaking direction.

Similar instability of the FFLO states is also expected from the G_0G_0 scheme of *T*-matrix approximation, with the pair susceptibility $\chi_0(P) = \sum_K G_0(P - K)G_0(K)$, as in the Nozieres–Schmitt-Rink theory [69]. The corresponding pair dispersion reaches a minimum at a finite *p*, as shown in Fig. 7.

Note that there is an obvious difference between the G_0G_0 case and the other two; the pair dispersion has no angle dependence. This can be easily understood since the pair susceptibility $\chi_0(P)$ is isotropic, independent of the gap Δ and the wave vector **q**, due to the lack of feedback effect. This is a defect of the G_0G_0 approximation. Nevertheless, this angle *independence* does suggest that the pair energy reaches a



FIG. 7. Typical pair dispersion Ω_p in the mean-field FFLO phases for G_0G_0 approximation of the pairing fluctuation theories. Shown here is the unitary case with $\eta = 0.75$ and $T/T_F = 0.01$. The conventions on color coding and units are the same as in Fig. 3.

minimum on a finite momentum sphere so that no spontaneous symmetry breaking or Bose condensation would take place for the pairs. Indeed, using such a theory, Ohashi also find that the FF state is unstable in 3D homogeneous Fermi gases for a similar reason [70]. Like the *GG* scheme, the pair dispersion for the G_0G_0 scheme does not vanish at $\mathbf{p} = \mathbf{q}$, because it is incompatible with the BCS mean-field gap equation, either.

E. Extending to the 2D case and higher order crystalline states

Extending the 3D results to the 2D case, we note that the pair dispersion reaches its minimum on a 1D ring, leading to an infinite degeneracy, which shall destroy FFLO type of superfluidity. Furthermore, there is no true long-range order (LRO) in 2D, even for a zero-momentum condensate.

While our calculations were done with the FF states, we argue that the infinite degeneracy induced finite DOS effects hold for the LO and higher-order FFLO states as well. We shall also point out that the pair momentum **p** takes value in the entire space \mathbb{R}^3 , unlike the vector field in a nonlinear sigma model, whose value in 3D is restricted to the sphere \mathbb{S}^2 . For the latter, the transverse and longitudinal components cannot vary independently, allowing spontaneous symmetry breaking to occur. Indeed, except for translational invariance, continuous spontaneous symmetry breaking is often associated with a compact symmetry group, whose variables, e.g., the angle parameters of an SU(2) spin, are not quantum numbers, unlike the pair momentum **p** in the present case.

In extending to possible higher crystalline FFLO states, we notice that one may understand the instability of the FFLO states from a different perspective. At the mean-field level, it is known that the LO states have *slightly* lower energy than the corresponding FF states at low T in 2D and 3D; the latter doubles the number of pair momenta for condensation. Meanfield calculations in both 2D [71–74] and 3D [75–77] show that condensation at two pairs of q's forming a square in the momentum space shall further lower the energy. This suggests that condensation at 3, 4, 6, and 8 pairs of momenta and so on should have a progressively lower energy (while T_c becomes progressively lower as well). Indeed, this has been shown to be true in 2D [73]. In 3D, it has been shown to hold for up to 3 pairs [75]. It is promising that with a higher number of pairs that preserve proper symmetries, the system may have a lower energy at lower T. Eventually, it leads to the conclusion that the lowest energy solution would be "condensation" on the entire 2D constant-energy surface in 3D or a constant-energy ring in 2D, on which the pair dispersion reaches its minimum. This is no longer a condensed, FFLO state.

F. Nature of the normal and ground state

Finally, we investigate the nature of this unusual normal state. The pairing correlation function for the 3D continuum case is given by

$$C(r) \propto \int \frac{\mathrm{e}^{i\mathbf{p}\cdot\mathbf{r}}\mathrm{d}^{3}p}{\xi^{2}(p-q)^{2}+\tau}$$
$$\approx \frac{1}{4\pi r\xi^{2}}\sqrt{\frac{4\xi^{2}q^{2}+\tau}{\tau}}\mathrm{e}^{-r\sqrt{\tau}/\xi}\sin(qr)\,,\qquad(13)$$



FIG. 8. Momentum distribution of the pairs in the (p_x, p_y) plane in the Bose metal state, for the unitary case with the pair dispersion minimized near $p/k_F = 0.71$.

where $\xi^2 = a_0 B_{\parallel}$ is the screening length (squared), and $\tau = -a_0 \mu_{\text{pair}} > 0$, with $\mu_{\text{pair}} \propto T$ near zero *T*. (Note here that we consider an isotropic pair dispersion.) Apart from the oscillating behavior, the correlation length is given by $\xi/\sqrt{\tau} \propto \xi/\sqrt{T}$. When $T \rightarrow 0$, the exponential decay will disappear, leaving an r^{-1} power-law decay at large distances at T = 0 so that the pairs approach an algebraic Bose liquid. This oscillating behavior due to a finite *q* is very unusual, manifesting the tendency to form a wavelike pairing order. Without superfluidity, such a Bose liquid is a Bose metal in the ground state, where μ_{pair} approaches 0 at zero *T*. In addition, the major part of the system is composed of the excessive majority fermions, which add to the metallic character of the system. We shall call this phase "anomalous metal."

The Bose metal state of the pairs can be probed experimentally using a time-of-flight imaging technique, with a quasiuniform trapping potential [78]. The momentum distribution of the pairs, given by the Bose distribution function, $n_B(\mathbf{p}) \propto b(\Omega_{\mathbf{p}} - \mu_{\text{pair}})$ is highly peaked on the constant energy surface (or ring in 2D). The peak height is controlled by $T/|\mu_{\text{pair}}|$, which has only a weak *T* dependence at low *T*. To visualize this, we suppress the \hat{z} direction, and plot $n_B(\mathbf{p})$ as a function of (p_x, p_y) in Fig. 8. This is in sharp contrast with the Bose distribution of conventional Bose gas, which has a single peak at zero momentum.

IV. DISCUSSIONS

One may argue that including impurities and/or higherorder interpair interactions may change the conclusion. However, while they may have a quantitative influence on the mean-field phase boundaries, we remark that the effects of impurities and higher-order interactions may either shift the minimum of the pair dispersion down to zero momentum or leave it at a finite momentum $p \neq 0$. For the former, a Sarma type of breached pair superfluid will result. For the latter, the situation remains. Either way, a stable FFLO superfluid state will *not* appear.

It should be noted that we have restricted ourselves to *s*-wave pairing only. Nonconventional pairing symmetries such

as p wave are often related to anisotropy associated with spin-orbit coupling, and/or lack of inversion symmetry, etc. This often introduces a preferential direction, and thus is not considered here. For *d*-wave pairing, which is mainly relevant for high T_c superconductors, there have been no strong indications for the existence of an FFLO state in the cuprate phase diagram [79]. We note that the pairing symmetry is an internal degree of freedom within a pair, while the **q** vector reflects the center-of-mass motion of the pair. These two are largely decoupled. Of course, the pairing symmetry has also a large effect on the pair dispersion [80]. It may or may not induce an anisotropy in the pair dispersion, and this will be investigated in a future work.

The instability of FFLO states has been investigated by a few other groups both at finite T [72,81–84] and zero T [5]. Baym and Friman [81] found that at finite T pion condensates with order parameter varying in one dimension (i.e., FFLO states) are prohibited, but are stable at all temperatures when varying in two and three dimensions (higher order crystalline FFLO states).

Shimahara [72,82] has found that phase fluctuations destroy LRO for the FFLO states in isotropic 3D and 2D, consistent with our results. However, he also found a quasi-LRO (QLRO) in isotropic 3D. We note that this is most likely a consequence of their presumed mean-field approximation, which necessarily leads to a zero pair chemical potential μ_{pair} . In contrast, we have shown here that, without presuming a mean-field solution at the beginning, one will obtain a normal state instead with a nonzero μ_{pair} for all T > 0. The nonzero μ_{pair} leads to an oscillatory exponential decay in the pairing correlation function, Eq. (13), indicating that neither LRO or QLRO exists in 3D. [Indeed, setting $\tau = 0$ in Eq. (13) would have led to a power law decay and hence a QLRO.] Note that we do not invoke topological excitations such as vortex-antivortex pair fluctuations.

Samokhin *et al.* [5] found a divergent spin susceptibility at T = 0 in isotropic 3D and 2D due to quantum fluctuations, not inconsistent with our findings here. However, their analysis involves diagrammatic pair propagators without the self-energy feedback effect, as well as an approximation assuming a small gap.

Radzihovsky and Vishwanath [83,85] found that the LO phase is unstable, which is consistent with our findings here. Starting with the unstable LO state, they further argued that fermion pairs may pair again to form a nematic charge-4 SF₄ superfluid phase. However, the fact that the interactions between fermion pairs are usually repulsive may render it unlikely to form such an SF₄ phase. Lee *et al.* [86] found that the FF phase is unstable in isotropic 3D in the context of quark matter, but reported a QLRO, for a reason similar to that of Shimahara [72,82]. By adding collective mode contributions to the mean-field solution, the latest work of Boyack *et al.* [61] has reported that the superfluid density of a mean-field FF state vanishes in the direction transverse to the wave vector **q**, consistent with our findings here.

A few papers have addressed or briefly mentioned the effect of anisotropy or lattice. Shimahara [72,82] claimed that there exists a stabilized LRO in the presence of anisotropy in 2D. Samokhin *et al.* [5] stated that a lattice will remove the divergence in 3D. Ohashi also noted that crystal lattice

may stabilize the FFLO phase. While these claims may be right, however, only generic forms of possible solutions were constructed [72], without self-consistent determination of the gaps, the \mathbf{q} -vectors, etc., on other parameters such as interaction strength and population imbalance. Further fully self-consistent, quantitative determination of the FFLO phase diagram in the presence of anisotropy and/or lattice effect is needed to substantiate these claims.

There have also been theoretical studies of possible FFLO (or stripe) states in Fermi gases with spin-orbit coupling (SOC) [87,88]. We point out that SOC forces a preferential direction, and/or leads to topologically distinct Fermi surfaces, making the system drastically different from the conventional FFLO physics. There are also studies of FFLO phases in 1D Fermi gases, which, however, do not possess long-range order at all.

Finally, we note that, despite many works in the literature on the LO and higher-order crystalline FFLO states, it is problematic to go beyond the FF case without a small gap expansion even at the mean-field level. A Ginzburg-Landau type of free energy with a combination of different \mathbf{q} order parameters are often assumed [2,3,6,34,72,83,84], but the evaluation of the coefficients of the various terms of the free energy either is left undetermined as free parameters [72] or has to be done using a small gap expansion, as in the original LO treatment [2], so that the noninteracting fermionic Green's function can be used. Such an approach is appropriate only near the FFLO/normal phase boundaries in Fig. 2. It will fail at least quantitatively for a large gap with a strong pairing interaction, such as in a unitary Fermi gas. Indeed, Samokhin et al. [5] have shown that the magnitude of quantum fluctuation corrections is determined by $(\Delta/E_F)^2$, which will no longer be small for a unitary Fermi gas. Thus one needs to go beyond the Ginzburg-Landau type of approach (which is appropriate only for small order parameters anyway). In other words, most Ginzburg-Landau-based treatments of the FFLO phases in the literature are inadequate for unitary Fermi gases.

A mean-field ansatz beyond the small gap expansion would be to write down the self-energy in the form

$$\Sigma(K) = -\sum_{i} \frac{\Delta_{\mathbf{q}_{i}}^{2}}{i\omega_{n} + \xi_{\mathbf{q}_{i}-\mathbf{k}}}.$$
(14)

When there are more than one \mathbf{q}_i , the simple BCS-like form of the full Green's function G(K), such as in Eq. (2), necessarily breaks down. Even with the LO states it becomes very complicated, as shown in Ref. [64].

Diagrammatically, the combination of different superconducting vertices associated with each $\Delta_{\mathbf{q}_i}$ order parameter (and their hermitian conjugates) necessarily generates a hierarchy of self-energy processes associated with an arbitrary combination of these \mathbf{q}_i 's. There will also be finite momentum pairing fluctuations associated with each one of them. This will also spoil the BCS-like form of the Green's function so that there is no simple many-body theoretical approach for addressing LO and higher-order FFLO states. A fully numerical approach seems to be necessary.

V. CONCLUSIONS

In summary, we have studied the effects of pairing fluctuations on the mean-field FFLO phases, and found that FFLO phases are intrinsically unstable against pairing fluctuations in continuum in 3D and 2D, and thus do not exist experimentally. This conclusion holds on general physical grounds, independent of specific pairing fluctuation theories, and is applicable for both quantum gases and isotropic condensed matter systems. We predict that the momentum distribution of the pairs are highly peaked on an entire CES, which can be easily tested experimentally.

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APPENDIX: PAIR DISPERSION IN THE MEAN-FIELD FFLO PHASES FROM NEAR-BCS THROUGH NEAR-BEC REGIMES

In this section, we will present more results of the pair dispersion at high population imbalances in the *mean-field* FFLO phase from near-BCS through near-BEC regimes. Starting with the unitary case, Fig. 9 shows the pair dispersion Ω_p in the FFLO phase with a population imbalance $\eta = 0.75$ and equal masses at temperature $T/T_F = 0.01$. This is just an alternative 3D plot of Fig. 3 in the main text, treating the angle θ between pair momentum **p** and the FFLO wave vector **q** as a Cartesian coordinate. This makes it easier to see that the minimum value of Ω_p (as a function of *p*) decreases as θ varies from 0 to π , revealing that the point **p** = **q** is indeed merely a saddle point



FIG. 9. Alternative 3D plot of the pair dispersion Ω_p in the FFLO phases at unitarity with population imbalance $\eta = 0.75$ and temperature $T/T_F = 0.01$. The conventions on color coding and units are the same as in Fig. 2 of the main text.



FIG. 10. Pair dispersion Ω_p in the FFLO phases for the near-BCS and near-BEC cases, with $(1/k_Fa, \eta, T/T_F) = (-1/2, 0.45, 0.01)$ and (0.1, 0.75, 0.01) for the left and right panels, respectively. The conventions on color coding and units are the same as in Fig. 1 of the main text.

of Ω_p . Now, we show the counterpart plot of the near-BCS and near-BEC cases in Fig. 10, as the left and right panels,

respectively. Despite the different radii of the bottom (half) circle, both confirm that the $\mathbf{p} = \mathbf{q}$ point is a saddle point of $\Omega_{\mathbf{p}}$.

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