The Fulde–Ferrell–Larkin–Ovchinnikov phase in the presence of pair hopping interaction

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Abstract

The recent experimental support for the presence of the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) phase in CeCoIn\(_5\) directed attention towards the mechanisms responsible for this type of superconductivity. We investigate the FFLO state in a model where on-site/inter-site pairing coexists with the repulsive pair hopping interaction. The latter interaction is interesting in that it leads to pairing with non-zero momentum of the Cooper pairs even in the absence of the external magnetic field (the so-called $\eta$ pairing). It turns out that, depending on the strength of the pair hopping interaction, the magnetic field can induce one of two types of the FFLO phase with different spatial modulations of the order parameter. It is argued that the properties of the FFLO phase may give information about the magnitude of the pair hopping interaction. We also show that $\eta$ pairing and d-wave superconductivity may coexist in the FFLO state. It holds true also for superconductors which, in the absence of magnetic field, are of pure d-wave type.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

An unconventional superconducting state with a non-zero total momentum of the Cooper pairs was predicted by Fulde and Ferrell [1] as well as by Larkin and Ovchinnikov [2] in the mid-1960s. Under particular conditions, this phase should occur at low temperatures and in strong magnetic fields. Due to the severe requirements for the formation of the FFLO state, this type of superconductivity has experimentally been observed only recently. In the FFLO state the superconducting order parameter (OP) oscillates in real space. This property, to some extent, resembles the unconventional superconductivity in strongly correlated systems [3], where the OP changes sign in the momentum space. The FFLO state has recently been analyzed in the context of heavy fermion systems [4–17], organic superconductors [18, 19], ultracold atoms [20, 21] and dense nuclear matter [22–24]. Although there is no direct experimental evidence for the spatial variation of the OP, suggestions for future experiments have been developed in [9, 25, 26].

The orbital (diamagnetic) pair breaking is a crucial mechanism that limits realization of the FFLO state. In the vast majority of superconducting materials it is a dominating pair breaking mechanism that destroys superconductivity for magnetic fields much weaker than the Clogston–Chandrasekhar limit ($H_{CC}$) [27, 28]. It holds true also for models appropriate to describe the short coherence length superconductors [29–31]. The significance of the diamagnetic pair breaking is usually described in terms of the Maki parameter $\alpha = \sqrt{2} H_{c2}^\text{orb}/H_{c2}^\text{CC}$, where $H_{c2}^\text{orb}$ is the upper critical field calculated without Zeeman splitting. There exist two general possibilities to reduce the destructive role of the orbital pair breaking. In the layered superconductors, formation of Landau orbits should be suppressed for magnetic fields applied parallel to the layers. This may explain possible observations of the FFLO state in some organic superconductors [18, 19]. The role of orbital pair breaking should also be limited in systems with narrow energy bands, like heavy fermion systems. The experimental evidence for the FFLO superconductivity in these systems seems to be the strongest [4–15].

In the context of recent investigations of the FFLO state, it is important to search for other mechanisms that stabilize superconductivity against the orbital pair breaking. Recently
it has been found that superconductivity originating from the repulsive pair hopping interaction is unique in that it is robust against this pair breaking mechanism [32]. The origin of this interaction may vary in different systems and therefore we do not specify a particular superconductor for which the following qualitative analysis can be directly applied. The repulsive pair hopping interaction can be derived from a general microscopic tight-binding Hamiltonian [33], but in this case the magnitude of the interaction is very small. However, since this interaction leads to superconductivity that is almost unaffected by the orbital effects, it may become more important close to the upper critical field, i.e. in the regime where the FFLO phase is expected to occur. We may also consider other sources of the pair hopping which give rise to much larger magnitudes of this interaction. For example, such a term may be included in the effective Hamiltonian describing Fermi gas in an optical lattice in the strong interaction regime [34, 35]. It also arises in a natural way in multifluidal models [36], though then the pairs hop between different orbitals on the same site. Nevertheless, we expect that some of our conclusions can still be valid.

The role of the pair hopping interaction in a multifluidal model is of particular interest because of its presence in the recently discovered iron pnictides [37]. Additionally, if the on-site repulsion exceeds the gap between the lowest and the next-lowest bands in the optical lattice, then the interband pair hopping seems to be important. Very recent quantum Monte Carlo calculations for TMTSF-salt [38] also suggest a significant role of the pair hopping processes in this system, which, on the other hand, probably exhibits the FFLO phase at high field [19, 39].

In the absence of magnetic field, the pair hopping interaction is responsible for the \( \eta \)-type pairing where the total momentum of the paired electrons is \( Q = (\pi, \pi) \) and the phase of the superconducting order parameter alters from one site to the neighboring one [40–44]. It has been shown that flux quantization and the Meissner effect appear in this state [45, 46]. Therefore, even in the absence of external magnetic field, the repulsive pair hopping interaction favors pairing with non-zero momentum of Cooper pairs. Although the pair hopping may not be the dominating pairing mechanism, its presence may affect the FFLO phase. In the following, the superconducting state with zero total momentum of the Cooper pair will be referred to as the BCS superconductivity. In that sense, both FFLO and \( \eta \) pairing will be referred to as the BCS superconductivity. As a consequence, the amplitude of the superconducting order parameter becomes a site-dependent quantity. Recent theoretical investigations of the FFLO phase in the attractive Hubbard model have been motivated mostly by the increasing interest in ultracold Fermi gases [47, 48]. These approaches may also be applicable to compounds other than the strongly correlated heavy fermion systems [49, 50]. Contrary to this, experimental results obtained for CeCoIn 5 indicate the anisotropic d-wave pairing. Therefore, in section 4 we study the case of inter-site attractive interaction that is responsible for the d-wave superconductivity. We show for the case of inter-site attraction that d-wave and \( \eta \)-pairing orders may coexist in the FFLO state. Such a coexistence is possible also for systems which, in the absence of a magnetic field, are in the pure d-wave state. For on-site and inter-site pairings, the potential of the pair hopping interaction \( J \) is assumed to be positive.

2. On-site pairing

We start our analysis with a model with on-site pairing interaction described by the following tight-binding Hamiltonian:

\[
H = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger c_{i\downarrow} c_{i\uparrow} + J \sum_{\langle i,j \rangle} c_{i\uparrow}^\dagger c_{j\downarrow}^\dagger c_{j\uparrow} c_{i\downarrow} + \sum_{i,\sigma} \epsilon(\sigma) c_{i\sigma}^\dagger c_{i\sigma},
\]

where \( t \) is the nearest-neighbor hopping integral, \( J \) is the pair hopping interaction, \( \mu \) is the chemical potential, \( s(\uparrow) = 1 \) and \( s(\downarrow) = -1 \). The Zeeman coupling is determined by \( h = g\mu_B \mathcal{H}/2 \), where \( g \) is the gyromagnetic ratio, \( \mu_B \) is the Bohr magneton and \( \mathcal{H} \) is the external magnetic field. The Hamiltonian (1) does not include the diamagnetic pair breaking. We refer to [51–60] for discussion of the influence of this mechanism on the FFLO state. Here, we focus on the role of the pair hopping interaction as well as on the properties of the \( \eta \) pairing that are robust against the orbital pair breaking. Nevertheless, the role of the orbital pair breaking will be briefly discussed. In this section we assume the simplest form of the effective on-site pairing interaction \( (U < 0) \) that is responsible for the s-wave superconductivity.

We apply the mean-field approximation and assume the order parameter in a general form:

\[
\Delta(R_j) \equiv \langle c_{j\uparrow}^\dagger c_{j\downarrow} \rangle = \sum_{m=1}^M \Delta_m \exp(iQ_m \cdot R_j).
\]

Then, the Hamiltonian in the momentum space takes the form

\[
\tilde{H}_\text{MF} = \sum_{k,\sigma} \tilde{\varepsilon}_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{m=1}^M U_{\text{eff}}(Q_m) \sum_k (\Delta_m^* c_{-k+Q_m}^\dagger c_{k\uparrow} + \text{h.c.}) - N \sum_m U_{\text{eff}}(Q_m) |\Delta_m|^2
\]

where

\[
\tilde{\varepsilon}_{k\sigma} = \varepsilon_k - \mu - s(\sigma)h.
\]
For arbitrary lattice geometry the dispersion relation is given by
\[ \varepsilon_k = -t/N \sum_j \exp(i \mathbf{R}_j \cdot k), \]
where the prime means summation over the nearest-neighbor sites. We have also introduced an effective pairing potential:
\[ U_{\text{eff}}(Q) = U - \frac{J s Q}{t}. \]  

For a general form of the order parameter (equation (2)) diagonalization of the mean-field Hamiltonian usually cannot be reduced to an eigenproblem of a finite Hermitian matrix. The first one was originally proposed by Fulde and Ferrel (FF), whereas the second was by Larkin and Ovchinnikov (LO). In the former one, it is assumed that \( M = 1 \), so the absolute value of \( \Delta \mathbf{R}_j \) is constant, but the phase changes from site to site. In the latter case \( M = 2, \Delta_1 = \Delta_2 \) and \( Q_1 = -Q_2 \). Then, one gets \( \Delta \mathbf{R}_j = 2 \Delta_0 \cos(Q \cdot \mathbf{R}_j) \), where we use \( \Delta_0 \equiv \Delta_1 \) and \( Q \equiv Q_1 \). However, one should keep in mind that at low temperature and high magnetic field FFLO phases with \( M > 2 \) may be thermodynamically more stable [61, 62].

For the FF phase, the mean-field Hamiltonian can be diagonalized by means of the Bogoliubov transformation.

Straightforward calculations lead to the following form of the grand canonical potential \( \Omega = -kT \ln \text{Tr} \exp(-\beta H) \):
\[ \Omega = -kT \sum_{\alpha=\pm} \sum_k \ln[1 + \exp(-\beta E_{k,\alpha})] + \sum_k \varepsilon_{-k+Q} - NU_{\text{eff}}(Q)|\Delta_0|^2, \]
where
\[ E_{k,\pm} = \frac{1}{2} \left[ \varepsilon_{k\uparrow} - \varepsilon_{-k\downarrow} \right] \pm \sqrt{ \left( \varepsilon_{k\uparrow} + \varepsilon_{-k\downarrow} \right)^2 + 4U_{\text{eff}}(Q)^2|\Delta_0|^2}. \]

In the case of LO superconductivity, the Hamiltonian cannot be diagonalized analytically. However, the pairing term links the one-particle states with momenta lying along a single line in the Brillouin zone. Therefore, one can solve the resulting eigenproblem for relatively large clusters. Namely, for an \( L \times L \) cluster, one has to diagonalize an \( 2L \times 2L \) Hermitian matrix.

2.1. Numerical results

We start our discussion with the simplest case \( M = 1 \) (FF state), which allows one to estimate the boundaries of the non-BCS superconducting phases. However, the presence of the LO superconductivity will be discussed as well. The thermodynamically stable phase has been determined through minimization of the grand canonical potential with respect to the superconducting order parameter \( |\Delta_0| \) and \( Q \). The calculations have been carried out for a square lattice with \( \mu = 0 \) as well as for a triangular lattice with \( \mu = 2t \). These chemical potentials correspond to maxima in the density of states and, therefore, to the highest superconducting transition temperatures. A comparison of results obtained for both cases allows one to check the role of the lattice geometry.

First, we focus on the influence of the pair hopping interaction on the properties of the FFLO phase. Figure 1 shows how the ground state of the system depends on \( J \) and \( h \). In figures 2 (square lattice) and 3 (triangular lattice) we present the values of \( Q \) that minimize \( \Omega \) for some particular values of \( J \) and \( h \). In the absence of a magnetic field there are two stable superconducting phases for both the lattice geometries. For small \( J \) there exists an isotropic BCS phase, which will be referred to as s-wave superconductivity (see figures 2(a) and 3(a)). The \( \eta \)-pairing phase occurs for larger \( J \). In the case of a bipartite square lattice, \( \eta \) pairing corresponds to the total momentum of Cooper pairs \( Q = (\pi, \pi) \), where the phase of the superconducting order parameter changes from one lattice site to the neighboring one (see figure 2(c)). Stability of this phase obviously follows from the fact that the pair hopping interaction involves sites which belong to different sublattices. Then, the oscillating \( \Delta_1 \) minimizes the energy of the system for the physically relevant repulsive interaction. In this context, the problem of \( \eta \) pairing on a non-bipartite triangular lattice is interesting even in the absence of a magnetic field [63]. We have found that, also for this geometry, the repulsive pair hopping interaction may lead to a thermodynamically stable phase with \( Q \neq 0 \). The ground
state energy is minimal when \( Q \) represents one of the corners of the hexagonal first Brillouin zone (see figure 3(c)). It is easy to check that, for such a value of \( Q \), the phase of \( \Delta_i \) takes on three different values, namely \( \Delta_i = \Delta_0 \), \( \Delta_i = \Delta_0 \exp(i \frac{2\pi}{3}) \) or \( \Delta_i = \Delta_0 \exp(-i \frac{2\pi}{3}) \), depending on \( i \). In the presence of a sufficiently strong magnetic field there exist four different superconducting phases. Apart from the above discussed s-wave and \( \eta \)-pairing states there are two other phases, which will be referred to as FFLO1 and FFLO2. Investigation of the total momenta of Cooper pairs (see panels ‘b’ and ‘d’ in figures 2 and 3) allows one to link FFLO1 and FFLO2 to s-wave and \( \eta \) pairing, respectively. Namely, \( Q \) obtained for the FFLO1 is relatively close to the origin of the Brillouin zone, whereas in the FFLO2 phase \( Q \) remains on the edges of the zone. One can see that the FFLO2 phase occurs for lower magnetic fields than the FFLO1. FFLO1 evolves from the s-wave superconductivity when a sufficiently strong magnetic field is applied. It holds true for both the lattice geometries. On a square lattice, FFLO2 evolves from the \( \eta \)-pairing state under the same conditions. However, on a triangular lattice this phase is stable only for moderate values of the pair hopping interactions as well as for moderate magnetic fields. It is surprising that increasing the magnetic field may cause two phase transitions: the first one from s-wave to FFLO2 is discontinuous and the second from FFLO2 to \( \eta \) pairing is a continuous transition (see figure 4). We have also found that the transitions from the s-wave phase to the FFLO1 state are discontinuous, whereas the transitions from the FFLO1 and FFLO2 phases to the normal state are continuous. Finally, for a square lattice the transition from the \( \eta \)-pairing state to the FFLO2 phase is discontinuous as well. The field dependence of the superconducting order parameter in various phases can be inferred from figure 4.

In the absence of pair hopping interactions, it is well known that the LO phase has a lower ground state energy.
Figure 4. $|U_{\text{eff}}(Q)\Delta_0|$ as a function of magnetic field. The parameters as well as the lattice geometry are explicitly shown in the figure.

Figure 5. $|\Delta_0|$ as a function of magnetic field and temperature for a square lattice with $J=0.35t$. than the FF one [2]. In our case, this result directly applies to the FFLO1 phase. We have found that, in the case of the FFLO2 state with $M = 2$, it has an energy lower than that with $M = 1$. Therefore, also in this case $|\Delta_i|$ is spatially inhomogeneous. This result has been obtained on the basis of numerical diagonalization of $200 \times 200$ clusters with periodic boundary conditions. Consequently, there exists a simple criterion to distinguish between FFLO1 and FFLO2 phases. In both cases $\Delta_i \sim \cos Q \cdot R_i$ but $|Q| \ll 1$ for FFLO1, whereas $|Q| > \pi$ for FFLO2. Therefore, the periods of spatial modulations of the order parameters relevant to FFLO1 and FFLO2 are very different. In the latter case it is of the order of the lattice constant. It is instructive to examine in more detail the spatial modulation of $\Delta_i$ in the FFLO2 phase on a square lattice. As the total momentum of Cooper pairs $Q$ is close to $\Pi = (\pi, \pi)$, one can introduce $Q' = \Pi - Q$ and note that $|Q'| \ll 1$. Then $\Delta_i \sim \cos[(\Pi - Q') \cdot R_i] = \cos(\Pi \cdot R_i)\cos(Q' \cdot R_i)$. The spatial profile of $\Delta_i$ is determined by two oscillating functions. Due to the first one the phase of the superconducting order parameter alters from one site to the neighboring one. It means that the FFLO2 phase retains the basic properties of the $\eta$-pairing superconductivity. The second factor is responsible for a slow variation of the magnitude of the superconducting order parameter $|\Delta_i|$, which is a hallmark of the LO-type of superconductivity. On the basis of our analysis one cannot exclude that the FFLO2 phase with $M > 2$ is more stable. Therefore, the actual boundaries of the FFLO2 state may cover a slightly wider range of magnetic fields than presented in figure 1.

Up to this point we have analyzed a two-dimensional system, where the influence of the orbital pair breaking can be neglected provided the applied magnetic field is parallel to the plane. However, if the magnetic field has a non-zero component perpendicular to the plane, as well as in the case of three-dimensional systems, the destructive role of the orbital effects has to be taken into account. This problem has been analyzed, for example, in [29–32, 51–60]. On the one hand, it is the dominating pair breaking mechanism for $s$-wave and FFLO1 superconductivity [29–31, 51–60]. On the other hand, it is very ineffective in destroying $\eta$-type
superconductivity [32]. Therefore, we assume that FFLO2 will also be robust against orbital pair breaking. This property may lead to significant modifications of the phase diagram presented in figure 1. Namely, the dashed lines show the boundaries of the \( \eta \) and FFLO2 types of superconductivity obtained under the assumption that the s-wave and FFLO1 phase are destroyed by orbital effects. Note that even a weak pair hopping interaction should lead to the onset of \( \eta \) and FFLO2 phases for fields sufficiently strong to destroy the conventional superconductivity. However, this conjectural result should be confirmed by calculations for the FFLO2 phase with the diamagnetic pair breaking explicitly taken into account.

Finally, we discuss the standard \((kT, h)\) phase diagram for a square lattice with \( J = 0.35t \). It is the value of the pair hopping interaction for which the ground state in the absence of a magnetic field is the \( \eta \)-type superconductivity (see figure 1). Figure 5 shows the results. It is interesting that, despite the unconventional character of the \( \eta \) pairing, the phase diagram is very similar to the analogous phase diagram for BCS–FFLO superconductors. Namely, the FFLO2 phase occurs only in the presence of strong magnetic field and at low temperatures. The phase transition from the \( \eta \) to FFLO2 phase is discontinuous, whereas the transition from FFLO2 to the normal state is continuous.

3. Inter-site pairing

In this section we extend our previous study by allowing for the anisotropic d-wave superconductivity. In this case, we add the nearest-neighbor attraction term to the Hamiltonian (1):

\[
H \rightarrow H + V \sum_{\langle i,j\rangle,\sigma} c_{i\sigma}^\dagger c_{j\sigma,\sigma} - c_{j,\sigma,\sigma}^\dagger c_{i\sigma} - h.c.,
\]

where we assume \( V < 0 \). For simplicity, we restrict ourselves to FF superconductivity on a square lattice. Then, the mean-field Hamiltonian in the momentum space is

\[
H_{MF} = \sum_{k,\sigma} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k} \left[ (U_{eff}(Q)\Delta_0^d + Vd(k)\Delta_0^d) c_{-k+Q,\downarrow} c_{k\uparrow} \right]
\]

where \( U_{eff}(Q)\Delta_0^d \) vanishes above the dotted line.

Figure 7. \(|U_{eff}(Q)\Delta_0^d|\) (upper panel) and \(|V\Delta_0|\) (lower panel) for various \( J \) and \( h \) at \( T = 0 \). \( V = -2.5t \) has been assumed. Dashed lines show the boundaries of the FFLO1 and FFLO2 phases. In the upper panel \(|U_{eff}(Q)\Delta_0^d|\) vanishes above the dotted line.
The presence of two order parameters $\Delta_0$ and $\Delta_d$ makes the problem numerically much more complicated than for the on-site pairing only. Though the coexistence of the $s$-wave and $d$-wave pairings in the FFLO phase may significantly enhance the upper critical field [64], for the sake of simplicity and to avoid too many model parameters we restrict our further analysis to the case $U = 0$.

Generally, the grand canonical potential should be minimized with respect to five variables: two components of the wavevector $Q$, magnitudes of two order parameters and the relative phase $\phi$ between $\Delta_0$ and $\Delta_d$. If the orders do not coexist, i.e. either $\Delta_0$ or $\Delta_d$ vanishes, $\Omega$ is independent of $\phi$. However, in order to analyze whether coexistence is possible, we have taken $\phi \in \{0, \pi, \pm \pi/2\}$ and minimized $\Omega$ with respect to the remaining variational parameters. The resulting phase diagram is presented in figure 6. Of course, the exact boundaries could be a bit different from those presented in this figure if one allows for an arbitrary value $\phi$.

A sufficiently strong magnetic field drives the system into the FF state. Depending on the values of $Q$, one can distinguish between two phases marked in figure 6 as FFLO1 and FFLO2. In the former case $\Delta_d \neq 0$, $Q = (0, Q_y)$ and $Q_y \ll \pi$. Then, $U_{\text{eff}}(Q)$ is positive and $\Delta_0 = 0$. However, in the FFLO2 state $Q = (\pi, Q_y)$, $Q_y \ll \pi$. We have found that in the FFLO2 phase both the order parameters may simultaneously be non-zero. This strongly contrasts with the results obtained in the absence of a magnetic field when the system is either in the pure $d$-wave superconducting state or in the pure $\eta$-pairing state. Note that for moderate (presumably realistic) values of $J$ and in the absence of magnetic field the ground state is of purely $d$-wave type.

In order to study the coexisting orders in more detail, we have calculated $|U_{\text{eff}}(Q)\Delta_0|$ (see the upper panel in figure 7) and $|V\Delta_d|$ (see the lower panel in figure 7) in the FFLO2 phase. In figure 8 we present these data for $J = 0.3t$ together with the field dependence of the wavevector $Q$. Although the dominating contribution to the superconducting gap comes from the $d$-wave pairing, $\Delta_0$ is non-negligible in the FFLO2 phase. For sufficiently strong inter-site pairing the boundaries of the FFLO2 phase are almost independent of $J$, which can be inferred from the lower panel in figure 6. This result can be explained in the following way: increase of the magnetic field shifts the wavevector $Q$ and, in this way, modifies $U_{\text{eff}}(Q)$. This potential may eventually vanish causing $U_{\text{eff}}(Q)\Delta_0 = 0$ (see panel ‘a’ in figure 8). Then, the upper critical field is determined only by the inter-site pairing. This effect may also be responsible for a non-monotonic field dependence of $\Delta_d$. Neither this non-monotonicity nor $J$-independent upper critical field occur for the weaker inter-site attraction, shown in the upper panel of figure 6. Here, both the order parameters are non-zero within the entire FFLO2 phase.

4. Concluding remarks

Our aim was to investigate the role of the pair hopping interaction for FFLO superconductivity. Probably this
interaction is not a dominating pairing mechanism. However, as we have argued in section 1, in some superconducting systems it may become important in the high field regime whether the FFLO phase is expected. Motivated by the pairing symmetry in these systems we have separately studied two models where the pair hopping interaction coexists with on-site and inter-site attractions. In the former case, the pair hopping interaction lowers the magnetic field corresponding to the onset of the FFLO state. In the presence of the inter-site pairing, sufficiently strong magnetic field allows for a coexistence of d-wave and $\eta$-pairing states even though such a coexistence does not occur in the absence of a magnetic field. It is instructive to compare this result with the recent experimental and theoretical data concerning the coexistence of superconductivity and spin-density waves in CeCoIn$_5$ [65–67]. One may formulate a general conjecture that field-induced breaking of the translational invariance of the superconducting phase gives way to other competing orders.

For a sufficiently strong pair hopping interaction one may expect an $\eta$-pairing state that is robust against the diamagnetic pair breaking. According to the best of our knowledge this phase has not been identified in any known superconducting system. However, upon application of an external magnetic field such a system should exhibit the FFLO state. In contradistinction to the BCS type of pairing, this phase should occur independently of the bandwidth and the orientation of the magnetic field. Investigating the spatial modulation of the superconducting order parameter, one can distinguish whether the FFLO phase originates from BCS or $\eta$ pairing. In the first case the period of modulation is much larger than the lattice constant. In the latter case it is of the order of the lattice constant and the phase of the order parameter retains its oscillating character typical for $\eta$-pairing superconductivity.

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