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Some clues in the investigation of the FFLO phase in superconductors

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Abstract

The Fulde–Ferrell–Larkin–Ovichinnikov (FFLO) phase is investigated in a two-dimensional superconductor described by a negative-U Hubbard model in the presence of a magnetic field. The parameter space defined by interparticle attraction and band filling is investigated and a search is performed for the FFLO phase therein, so as to provide clues to experiments designed to confirm the existence of a nonuniform spatial nature of the superconducting state. Our results convincingly demonstrate periodic modulation of the local pairing gap in real space. Heavy fermions, considered as a probable candidate that hosts the FFLO phase, are found in a metallurgically clean state and shows extreme type-II behaviour. In our calculations both these conditions are satisfied for a certain magnetic field range and the range expands for large interacting strengths and particle densities. The cleanliness condition is met as the coherence length becomes very small (compared to the mean free path) and the extreme type-II behaviour shows up via a large Ginzburg–Landau parameter.

1. Introduction

The study of superconductivity in the presence of a magnetic field commenced nearly half a century ago with the works of Clogston and Chandrasekhar [1, 2]. The subject was intermittently revived to discuss the bounds on the upper critical field and its effect on the phase boundary where paramagnetic effect governs the physics. More of the numerous implications of the presence of an external magnetic field are elucidated by Fulde and Ferrel [3] and by Larkin and Ovichinnikov [4], where a possibility of finite momentum pairing between the different participation species of electrons is explored. The first experimental realization of a finite momentum pairing was obtained in a heavy fermion compound (UPd_2Al_3) via thermal expansion of magnetostriction measurements [5]. Soon after many other heavy fermion compounds also reported the FFLO phase [5–7].

The factor aiding the heavy fermion compounds to be candidates for realizing Cooper pairing with a nonzero momentum can possibly be attributed to the extreme type-II behaviour, a high effective electron mass, m^*, with a large Ginzburg–Landau and Maki parameters and their availability in metallurgically clean state. All these qualities put together imply a very large upper critical field and thus underscore the ascendancy of paramagnetism over the orbital effect.

An alternative route to achieve the supremacy of paramagnetic effect is to use a layered structure in a strong magnetic field applied parallel to the layers, thereby undermining the orbital pair breaking effect further and augmenting the parameter space where the FFLO phase can exist [8]. The organic superconductors strongly fit into these requirements and hence are considered as ideal candidates for the FFLO phase [9–11]. Apart from these compounds, signatures of FFLO phases are also observed in other materials such as neutron stars [12] and ultracold atomic gases [13]. An attempt to elaborate on these topics will lead us to digress from the main focus of the paper and there are many good reviews on the subject. Only a few are listed here [14, 15].

To make the introductory discussion self-contained, we briefly mention a few experimental results, particularly in heavy fermion compounds which provide support to the FFLO phase present there. In $CeCoIn_5$, the heat capacity data as a function of magnetic field, h, with h applied along the ab-plane, shows two phase transitions, a second order one within the superconducting (SC) state at low field values and a higher field first order transition at H_{c_2} [16], the intervening regime acquiring a nonuniform nature. However with the external magnetic field acquiring an angle with the ab-plane, the orbital effect starts playing a role. The large field transition goes away rendering an absence of the nonuniform or the FFLO state. The above conclusions were promptly contested by Movshovich *et al* [17, 18].

Other experiments such as anisotropic magnetothermal measurements [19, 20], ultrasound velocity measurements [21] etc provide only indirect and cursory evidence supporting the presence of an FFLO state.

More recently, ^{115}In NMR studies on $CeCoIn_5$ with the applied field parallel to ab plane revealed a dramatic asymmetry in the NMR spectrum for a field greater than the upper critical field when compared with a field less than that [22]. Further, an unusual temperature dependence of the Knight shift of ^{115}In is noted for the latter case. These facts are correlated with direct evidence of the FFLO phase and a simulation of the NMR spectrum with a spatially modulated gap function, seeming to satisfactorily explain the experimental findings [23]. Even these results were challenged in the light of other NMR studies [24].

Thus it is fairly evident that the experimental signatures of the FFLO phase in real systems can still be questioned. This provides a motivation for us to look for the spatially modulated profile of the order parameter, a hallmark of the FFLO phase. Thus, the stringency of the number of conditions to be met simultaneously, that impedes the scope of observing the FFLO phase in experiments, is explored [25].

Starting with a weak coupling BCS superconductor in a magnetic field, we solve the mean field Bogoliubov–de Gennes (BdG) equations for an attractive Hubbard model in two dimensions. The mean field order parameter thus obtained in real space shows periodic modulation, a signature of a finite momentum Cooper pairing and hence a FFLO phase. The dependence of this modulated phase on the (attractive) Hubbard interaction, $|U|$, and band filling, μ (or particle densities, n), is investigated in detail to comment on the possible difficulties in accessing this phase experimentally. Some of the characteristic properties of a superconductor are calculated, such as the coherence length (related to the wavefunction of modulation in the order parameter [16, 21]) and the penetration depth (related to the superfluid density or stiffness [33]) etc to provide strong evidence in favour of observing the FFLO phase. The relevance of our results to real materials, such as heavy fermion compounds are discussed.

We organize the paper as follows. The model and the BdG formalism are briefly discussed in section 2. The details are skipped here since they have been discussed earlier in the literature [26–28]. Section 3 contains a convincing demonstration of the periodic profiles of the order parameter and local magnetization for certain choice of parameters. It is worth mentioning here that the previous reports have been restricted to a very narrow parameter range [29, 30] and thus a detailed study of the effect of electronic interaction strength and density on the FFLO phase was missing. The results further include an estimation of characteristic lengths, such as the coherence length and the penetration depth, which yields a large Ginzburg–Landau parameter. This information nicely correlates with the requirements for the existence of the FFLO phase in physical systems, such as heavy fermion compounds etc.

2. Model and formalism

We consider a two-dimensional Hubbard model with $|U|$ as the magnitude of the onsite attractive interaction,

$$\mathcal{H} = -t\sum_{\langle ij\rangle,\sigma}(c^\dagger_{i\sigma}c_{j\sigma} + \text{h.c.}) - |U|\sum_i (n_{i\uparrow} - \tfrac{1}{2})(n_{i\downarrow} - \tfrac{1}{2}) + \sum_{i,\sigma}(\sigma h - \mu)n_{i\sigma}. \quad (1)$$

$c^\dagger_{i\sigma}$ $(c_{i\sigma})$ is the creation (destruction) operator for an electron with spin σ, which can assume values ± 1 at a site $\mathbf{r}_i$, h is the magnetic field which couples with the spin, σ, of electrons via Zeeman coupling, $n_{i\sigma} = c^\dagger_{i\sigma}c_{i\sigma}$ and μ denotes the chemical potential. Here t is the transfer integral. Other parameters such as, U, h and μ are expressed in units of t. t is typically of the order of $1ev$.

Hartree–Fock decomposition of the interaction term in equation (1) yields,

$$\mathcal{H}_{\text{eff}} = \sum_{ij,\sigma}\mathcal{H}_{ij\sigma}(c^\dagger_{i\sigma}c_{j\sigma} + \text{H.c.}) + \sum_i[\Delta_i c^\dagger_{i\uparrow}c^\dagger_{i\downarrow} - \Delta^*_i c_{i\uparrow}c_{i\downarrow}]. \quad (2)$$

Here $\mathcal{H}_{ij\sigma} = -t\delta_{i\pm 1j} - (\mu + U\delta n_{i\bar{\sigma}} - \sigma h)\delta_{ij}$, where $\delta n_{i\bar{\sigma}} = n_{i\bar{\sigma}} - 1/2$ with $\bar{\sigma} = -\sigma$. The local pairing amplitude, $\Delta_i = -|U|\langle c_{i\downarrow}c_{i\uparrow}\rangle$ is the order parameter.

The following transformations are used to diagonalize equation (2),

$$c_{i\sigma} = \sum_n[\gamma_{n\sigma}u_n(\mathbf{r}_i) - \sigma\gamma^\dagger_{n\bar{\sigma}}v^*_n(\mathbf{r}_i)], \quad (3)$$

where $\gamma_{n\sigma}$ and $\gamma^\dagger_{n\sigma}$ are the quasiparticle operators, $u_n(\mathbf{r}_i)$ and $v_n(\mathbf{r}_i)$ are the BdG eigenvectors.

Applying the above transformations in equation (2), we get the BdG equations in a matrix form as

$$\begin{pmatrix} \mathcal{H}_{ij\sigma} & \hat{\Delta}_i \\ \hat{\Delta}^*_i & -\mathcal{H}^*_{ij\bar{\sigma}} \end{pmatrix}\begin{pmatrix} u_n(\mathbf{r}_i) \\ v_n(\mathbf{r}_i) \end{pmatrix} = E_{n\uparrow}\begin{pmatrix} u_n(\mathbf{r}_i) \\ v_n(\mathbf{r}_i) \end{pmatrix}, \quad (4)$$

where $E_{n\uparrow}$ are the eigenvalues. We start with initial guesses for the pairing amplitude, Δ_i and the density of up and down-spin electrons, $\langle n_{i\uparrow}\rangle$ and $\langle n_{i\downarrow}\rangle$ respectively. Subsequently, the eigenvalues, $E_{n\uparrow}$ and the eigenvectors $(u_n(\mathbf{r}_i), v_n(\mathbf{r}_i))$ are determined numerically from equation (4). The local pairing amplitudes at sites $\mathbf{r}_i$ and the density of up and down-spin electrons in terms of $u_n(\mathbf{r}_i)$ and $v_n(\mathbf{r}_i)$ are calculated from,

$$\Delta(\mathbf{r}_i) = -|U|\sum_n[u_n(\mathbf{r}_i)v^*_n(\mathbf{r}_i)f(E_{n\uparrow}) - u_n(\mathbf{r}_i)v^*_n(\mathbf{r}_i)f(-E_{n\downarrow})] \quad (5)$$

and

$$\langle n_{i\sigma}\rangle = \sum_n[|u_n(\mathbf{r}_i)|^2 f(E_{n\sigma}) + |v_n(\mathbf{r}_i)|^2 f(-E_{n\bar{\sigma}})], \quad (6)$$

where $f(E_{n\sigma})$ is the Fermi distribution function. In this paper, we present results only at zero temperature, where $f(E_{n\sigma})$ is unity. The entire process is iterated with new guesses for the above quantities until self-consistency is achieved for all of them simultaneously.

As discussed later, a number of self-consistent solutions may exist for the pairing amplitude, $\Delta(\mathbf{r}_i)$, corresponding to one set of parameters. The winner among these will be decided by computing the free energies, $\mathcal{F}$, computed with respect to the free energy in vacuum (at zero temperature) and is given by [29],

$$\mathcal{F} = \sum_{n\sigma} E_{n\sigma}\left[f(E_{n\sigma}) - \sum_i |v_n(\mathbf{r}_i)|^2\right] + |U|\sum_i \langle n_{i\uparrow}\rangle\langle n_{i\downarrow}\rangle + \frac{1}{|U|}\sum_i \Delta_i^2 - \frac{|U|N}{4}. \quad (7)$$

For example, consider one particular parameter set, $|U| = 2.5$, $\mu = -0.5$ and $h = 0.35$, all in units of t (same as elsewhere in the paper). The free energies are computed using equation (7) corresponding to BdG solutions that yield uniform, one period and two period modulations for the local pairing gap, Δ_i and are obtained as -1.2539, -1.2543 and -1.2520 respectively (again in units of t) for a two-dimensional lattice of size 32×16 (see discussion below). Since one period modulation for Δ_i yields the lowest energy, it is considered as the energetically favourable solution. We have carried out similar studies for all choices of U and μ corresponding to various values of h used in the paper to pin down the stable solution.

Next we comment on the choice of parameters. We investigate the behaviour of the pairing amplitude for different values of μ corresponding to a few representative values of onsite interaction strengths, $|U| = 1$, 2.5 and 4. The rationale behind the choice of U is to get an insight into the stability of the FFLO phase at weak to moderate interparticle attraction strengths (mean field studies prohibit large U) for various densities. All our calculations are carried on a two-dimensional lattice of size 32×16. The reason behind choosing a rectangular lattice instead of a square one, is as follows. The FFLO order parameter undergoes a *one-dimensional modulation* with a period that is commensurate with the lattice size [29, 30]. Thus, the choice of a rectangular lattice allows us to increase the lattice size along one direction such as to accommodate more periods of the pairing gap.

3. Results

It is evident from our earlier discussions that magnetic field induces a nontrivial Cooper pairing and hence an unconventional superconductivity when all associated conditions are simultaneously satisfied. In an attempt to get a deeper understanding of this novel superconducting state, we compute few important length scales, *namely* the coherence length, ξ and the penetration depth, λ that characterize a condensate, in the subsequent discussion. Since the modulation of the order parameter is suggestive of a FFLO phase, we first compute Δ_i for the parameter values discussed earlier. The self-consistent Δ_i thus obtained show interesting variations as the magnetic field is increased, that is, starting with a uniform order at

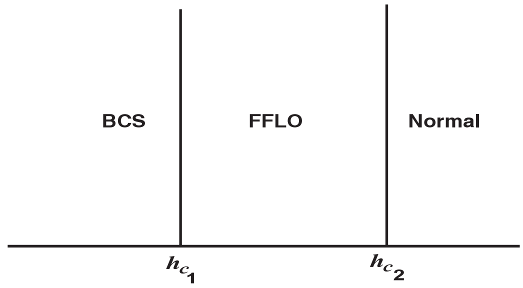

Figure 1. A schematic representation of the FFLO phase is shown as a function of the magnetic field. This phase, obtained via minimization of the corresponding free energy, is intermediate to a BCS superconductor (homogeneous Δ_i) and a normal phase ($\Delta_i = 0$). The boundaries of the FFLO phase are marked by h_{c_1} and h_{c_2}, the lower and upper critical magnetic fields respectively. The diagram is valid over a large regime of U–μ parameter space, except at low U and densities where the intermediate space is vanishingly small.

small h values, the order parameter shows periodic modulation at intermediate fields before vanishing at large fields. The modulating part between lower and upper threshold magnetic field values, *namely* h_{c_1} and h_{c_2} respectively, represents the FFLO phase and is central to our discussion. The scenario is schematically shown in figure 1.

Before we proceed with the discussion on the effect of interparticle attraction, we note that the FFLO phase is not so sensitive to the carrier density. We are able to observe the existence of the FFLO phase at almost all densities, except for very low ones where superconductivity itself becomes very weak. To arrive at the above conclusion, we have scanned a large $U - \mu$ parameter space. Thus we have fixed μ at $\mu = -0.5$ such that the density is fixed at a value around quarter filling.

The interaction effects are invoked via a comparison between $|U| = 2.5$ and 4, which yields a broader FFLO phase for the larger $|U|$. For $|U| = 4$, the region intervening two critical fields (h_{c_1} and h_{c_2}) is wider than that for $|U| = 2.5$, thereby establishing the fact that the FFLO state is stable for strong interaction strengths. To quote some values for extending support to the above argument, h_{c_1} and h_{c_2} are obtained as 0.35 and 0.55 respectively for $|U| = 2.5$, whereas the same for $|U| = 4$ are 0.9 and 1.88. The periodic modulation of the pairing amplitude presented in figure 2, suggests that Δ_i has a larger amplitude for stronger U. Also note that with increasing h, the amplitude decreases and also more periods are accommodated. The latter can be understood as follows. The rise in the number of broken Cooper pairs ($\Delta_i = 0$) results in increase in the number of nodes in the spatial profile of the order parameter. At still lower values of U, e.g. $|U| = 1$, a direct transition from the superconducting to the normal phase is obtained, mainly because of weak superconducting correlations.

A subtle point needs mentioning in the preceding discussion. The fully self-consistent solutions for the BdG equations demand simultaneous self-consistencies of Δ_i, $\langle n_{i\uparrow}\rangle$ and $\langle n_{i\downarrow}\rangle$ and thus require more computational time in the vicinities of h_{c_1} and h_{c_2} owing to the existence of different

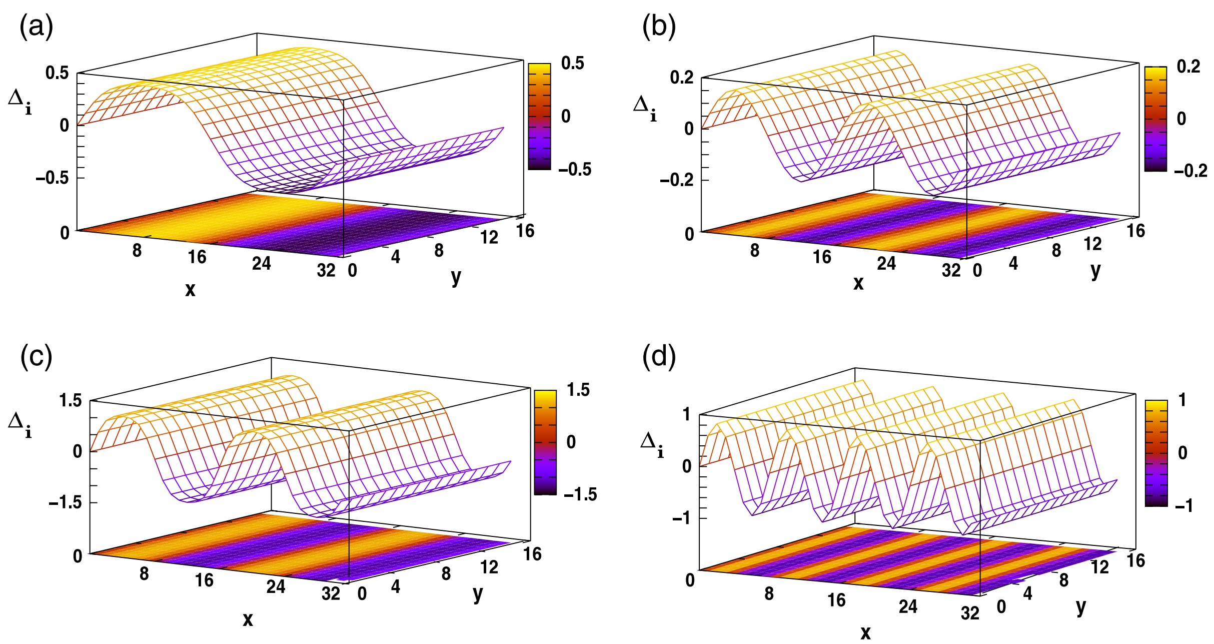

Figure 2. Local pairing amplitude modulation is shown for $|U| = 2.5$ ((a) and (b)) and $|U| = 4$ ((c) and (d)) for magnetic field values between h_{c_1} and h_{c_2}. The four figures correspond to $h = 0.35$ (a), 0.5 (b), 0.9 (c) and 1.2 (d). The band filling is chosen to be $\mu = -0.5$ and the system size is $L_x \times L_y = 32 \times 16$ and the same are considered for other figures. All the parameters are in units of t (true for all other figures) and our calculations are done at $T = 0$.

(This figure is in colour only in the electronic version)

competing solutions. The difficulty can only be partially taken care of by clever choices of the initial guesses for the above quantities.

We now focus on the coherence length, ξ, which is the separation between the Bloch walls of broken Cooper pairs ($\Delta_i = 0$) [16]. More concretely, ξ is of the order of the wavelength of the order parameter and can be computed from the modulation seen in Δ_i presented in figure 2. It is seen that ξ undergoes appreciable reduction from a large value (practically infinite corresponding to homogeneous Δ_i in the BCS phase) to a few lattice spacings at the onset of the FFLO phase. This result is elucidated in figure 3, which also shows ξ reducing further within the FFLO phase. We note that the reduction in the magnitude of ξ as h increases is more pronounced for $|U| = 4$ (than for $|U| = 2.5$) which accommodates more periods of the order parameter and hence is characterized by even shorter ξ. A short coherence length makes it easy for the condition $l \gg \xi$ (l being the electron mean free path) to be met, a requirement laid out for realizing the FFLO phase.

Another quantity, the local magnetization, m_i ($=n_{i\uparrow} - n_{i\downarrow}$), also suggestive of a nonuniform phase, is considered. m_i exhibits modulation, reminiscent of the behaviour of the pairing gap. It is interesting to note that the period of modulation of m_i is half of that of the order parameter, as lattice sites with nonzero order parameter corresponds to weak magnetization, while large m_i is obtained at the nodal lines, which have broken pairs [30], thereby causing a phase difference between the two. As the magnetization data do not convey anything new to aid our analysis, for brevity the corresponding plots are not presented here.

A useful quantity that is indicative of a FFLO phase in real materials is the Ginzburg–Landau parameter, κ. A large κ denotes extreme type-II behaviour, a requisite for the nonuniform phase. Since computation of κ needs another characteristic length i.e. the penetration depth, λ, we proceed to calculate λ. The experimental scenario [31] has suggested a large penetration of magnetic flux through the nodal lines (where the order parameter is zero) in the FFLO phase.

λ can be evaluated from the superfluid stiffness D_s ($\propto \frac{1}{\lambda^2}$ [32]), which denotes the phase rigidity of the condensate and is given by the long wavelength ($\mathbf{q} \to 0$) and static ($\omega = 0$) limits of the Kubo linear response formula [33],

$$\frac{D_s}{\pi} = \langle -K_x \rangle - \Lambda_{xx}(q_x = 0, q_y \to 0, \mathrm{i}\omega = 0), \tag{8}$$

where the first term $\langle -K_x \rangle$, defined as

$$\langle -K_x \rangle = \left\langle \sum_\sigma -t[(c^\dagger_{i+\hat{x}} c_i + c^\dagger_i c_{i+\hat{x}})] \right\rangle \tag{9}$$

is the average kinetic energy along the x-direction and represents the diamagnetic response to an external magnetic field. The second term, a transverse current–current correlation at different times, is the paramagnetic response and is given by

$$\Lambda_{xx}(\mathbf{q}, \mathrm{i}\omega_n) = \sum_{\mathbf{r}_i} \int_0^\beta \mathrm{d}\tau \langle j_x(\mathbf{r}_i, \tau) j_x(0, 0)\rangle \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\mathbf{r}} \mathrm{e}^{-\mathrm{i}\omega_n \tau}. \tag{10}$$

Equations (9) and (10) are written in terms of quasiparticle operators, γ (see equation (3)), whose expectations are evaluated between the BdG states (u_n and v_n) to compute D_s from equation (8).

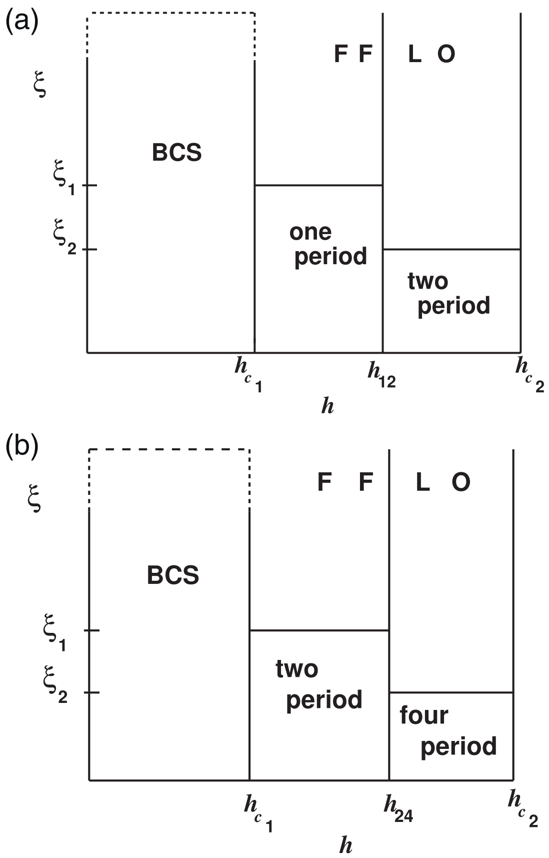

Figure 3. The coherence length, ξ, (in units of t) measured from the modulation profile of Δ_i (figure 2) is schematically shown for $|U| = 2.5$ (a) and $|U| = 4$ (b) as a function of the magnetic field. In (a), $h_{c_1} = 0.35$ marks the onset of a one period modulation (FFLO) with $\xi_1 = 32$. At $h_{12} = 0.4$, a transition from a one period to a two period modulation ($\xi_2 = 16$) is obtained, which persists until $h_{c_2} = 0.55$. In (b), the FFLO regime commences with a two period modulation at $h_{c_1} = 0.9$ with $\xi_1 = 16$ (the one period solution does not exist), then crosses over to a four period solution (with $\xi_2 = 8$) at $h_{24} = 1.2$ and continues until $h_{c_2} = 1.88$.

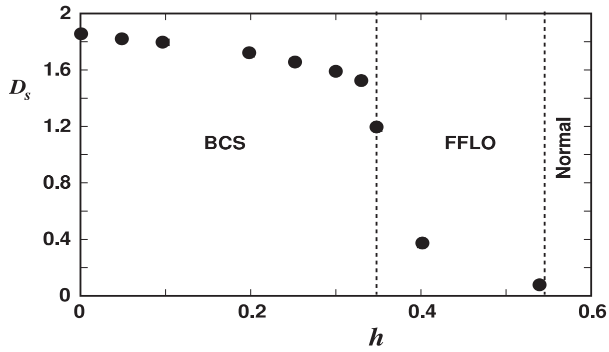

Figure 4. The superfluid stiffness, D_s, (in units of t) is shown for $|U| = 2.5$ and $\mu = -0.5$ as a function of magnetic field, h. It shows negligible variation in the BCS regime, while the FFLO phase is characterized by a large drop in D_s. The phase boundaries are shown by dotted lines.

In figure 4, we present D_s, which shows a considerable decrease within the FFLO phase (between h_{c_1} and h_{c_2} and marked by dotted lines in figure 4) as the phase rigidity is progressively destroyed with increasing magnetic field.

The lowering of D_s as a function of h indicates a rise in the penetration depth (in view of the inverse-square relation between D_s and λ). This, along with the lowering of the coherence length yields large κ values, a feature expected to demonstrate the existence of a FFLO phase.

4. Conclusions

We summarize the important results obtained here. The presence of a FFLO phase is investigated in the context of a two-dimensional superconductor in the presence of a magnetic field. The existence of a phase characterized by a modulated local pairing amplitude (FFLO) for a number of parameter values, i.e., electronic interaction, $|U|$ and band filling, μ (or particle density) is convincingly demonstrated. Weak to moderate values of U, such as $|U| = 1$, 2.5 and 4 are considered and, except for $|U| = 1$, we obtained a FFLO phase for the other two representative values with the larger one among them showing brisk modulation (more periods) in the spatial profile of the pairing amplitudes as the magnetic field is increased. Thus larger values of the interaction parameter facilitate a realization of the FFLO phase. All of these features are present for a large range of band filling, excepting the ones for which the particle density becomes very small.

The implications of these results to real materials are elucidated in the following manner. The sharp drop of the coherence length, ξ, at h_{c_1}, marked by the onset of a modulated local pairing amplitude underscores the cleanliness of the sample where the condition $l \gg \xi$, a requirement for the FFLO phase can easily be met. Moreover, ξ further reduces between h_{c_1} and h_{c_2}, owing to a more dramatic change in the period of modulation, making room for the above condition to be satisfied with a greater ease. Also the Ginzburg–Landau parameter, κ, increases as the magnetic field is enhanced, due to a simultaneous decrease in ξ and an increase in the penetration depth, λ. A large κ is suggestive of an extreme type-II behaviour and thus a condition that needs to be satisfied to realize a FFLO phase.

The conditions mentioned above seem to be satisfied to a large extent in heavy fermion compounds and organic superconductors, and thus these are considered as suitable candidates to achieve a modulated phase, a theoretical prediction that was made nearly five decades ago. In a simple model for a two-dimensional superconductor, we have demonstrated how some of these conditions are met in the presence of a magnetic field rendering support to the candidature of heavy fermion and other systems where the FFLO phase may be realized experimentally. Possibly in ultracold atomic superfluids, some of these requirements are met easily and hence demonstrate signatures of the FFLO phase in experiments more convincingly than their fermionic counterparts [13].

Acknowledgments

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