Possible Fulde-Ferrell-Larkin-Ovchinnikov State in CeCoIn$_5$

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Angular dependent magnetothermal conductivity experiments on CeCoIn$_5$ indicate that this compound is a $d_{x^2-y^2}$-wave superconductor. In this study, the low-temperature behavior of the upper critical field is measured in a single crystal of CeCoIn$_5$ along the directions $\vec{H} \parallel \vec{a}$ and $\vec{H} \parallel \vec{c}$. The data is compared with model calculations of the upper critical field in a $d_{x^2-y^2}$-wave superconductor. It is found that the observed $H_{c2}(T)$ along $\vec{H} \parallel \vec{a}$ is consistent with a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state at low temperatures, $T < 0.7$K, whereas for $\vec{H} \parallel \vec{c}$ the FFLO state appears to be absent in CeCoIn$_5$. Furthermore, it is predicted that the quasiparticle density of states in the FFLO state exhibits a complex peak structure which should be observable by scanning tunneling microscopy.

I. Introduction

Recent measurements on CeCoIn$_5$ have led to a renewed discussion of a possible high-field Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state in unconventional superconductors. [1,2] In this state, the coupling of the magnetic field to the quasiparticle spins dominates over the orbital coupling, leading to pairing between exchange-split Fermi surfaces, and hence to a spatially non-uniform superconducting order parameter. For conventional superconductors, its realization appears to be practically impossible because of two reasons. First, the sample quality has to be in the superclean limit, i.e. the quasiparticle mean path needs to be much larger than the coherence length. Second, the Ginzburg-Landau parameter $\kappa$, measuring the ratio of the magnetic penetration depth vs. the superconducting coherence length, should be very large, i.e. $\kappa \gg 10$.

The recent synthesis of quasi-two-dimensional nodal superconductors, such as the high-$T_c$ cuprates, the $\kappa$-(ET)$_2$ salts, and CeCoIn$_5$ has changed this situation dramatically. It appears that the above two conditions can be met in high-quality single crystal samples of these compounds. [3–6] These systems are quasi-two-dimensional, leading to a large Ginzburg-Landau parameter in a planar magnetic field. Furthermore, unlike in the conventional s-wave superconductors, the stability region of the FFLO state is much more extended in $d_{x^2-y^2}$-wave superconductors compared to conventional ones. [3,4]

Indications for possible FFLO states in organic superconductors were already reported in $\lambda$-(BEDTS)$_2$GaCl$_4$ and $\kappa$-(BEDT-TFF)$_2$Cu(NCS)$_2$. [7,8] In the first compound, a kink in the thermal conductivity points to a transition from a FFLO state to a vortex lattice. In the second material a similar feature in the magnetization was identified. Moreover, recent evidence for $d_{x^2-y^2}$-wave order parameter symmetry was found in $\kappa$-(BEDT-TFF)$_2$Cu(NCS)$_2$ by angular dependent magnetothermal conductivity measurements in a rotating magnetic field within the conducting crystal plane. [9,10] Moreover, it was observed that the upper critical field $H_{c2}(T)$ in the FFLO regimes decreases quasi-linearly with temperature, in contrast to the rather weak temperature dependence of $H_{c2}$ in the absence of a FFLO state as $T \to 0$.

More recently, a new heavy fermion compound, CeCoIn$_5$, was discovered. This material superconducts below a critical temperature $T_c = 2.3$K, [11] and it has a layered structure similar to the high-$T_c$ cuprates. Angular dependent magnetothermal conductivity experiments indicate $d_{x^2-y^2}$-wave superconductivity in this material. [12] Furthermore, the temperature dependence of the upper critical field $H_{c2}(T)$ for both $\vec{H} \parallel \vec{a}$ and $\vec{H} \parallel \vec{c}$ was measured in single crystals of CeCoIn$_5$. [13] It was observed that $H_{c2}(T)$ for $\vec{H} \parallel \vec{a}$ exhibits a quasi-linear temperature dependence in the proposed FFLO regime, $T < 0.7$K.

In this paper, the upper critical field $H_{c2}(T)$ in a single crystal sample of CeCoIn$_5$ is determined from thermal expansion and magnetoresistance measurements with field orientations $\vec{H} \parallel \vec{a}$ and $\vec{H} \parallel \vec{c}$. A $d_{x^2-y^2}$-wave model calculation is used to explain the temperature dependence of $H_{c2}(T)$ along both directions, considering the orbital effect and the Pauli term. In particular, the possibility of a FFLO state is addressed. For $\vec{H} \parallel \vec{c}$ it is found that...
the temperature dependence of $H_{c2}(T)$ can be fitted consistently to the experimental data without invoking a FFLO state. On the other hand, for $\tilde{H} \parallel \vec{c}$ we observe that the inclusion of a $\vec{v} \cdot \vec{q}$ term arising from a FFLO state is crucial for a consistent description of the observed $H_{c2}(T)$ below $T = 0.7K$. In particular, the quasi-linear $T$-dependence of $H_{c2}(T)$ at low temperatures can be understood within this framework, which provides compelling evidence for a FFLO state in CeCoIn$_5$. In order to help to further test and scrutinize this model, we also determine the corresponding quasiparticle density of states which should be accessible to scanning tunneling microscopy experiments.

II. $H_{c2}(T)$ for $\tilde{H} \parallel \vec{c}$

The temperature dependence of the upper critical field along the crystal $c$-direction can be obtained from two coupled integral equations [14]

\begin{align}
-\ln t &= \int_0^\infty \frac{du}{\sinhu} \left[ \bar{1} - \exp(-\rho u^2) \exp(\hbar u)(1 + 2\rho^2 u^4 C) \right], \\
-C \ln t &= \int_0^\infty \frac{du}{\sinhu} \left\{ C - \exp(-\rho u^2) \cos(hu) \left[ \frac{\rho u^2}{12} + C \left( 1 - 8\rho u^2 + 12\rho^2 u^4 - \frac{\rho^4 u^6}{3} \right) \right] \right\},
\end{align}

where $t = T/T_c$, $\rho \equiv (v^2eH)/(8\pi^2 T^2)$, and $h \equiv (g\mu_B H)/(2\pi T)$. Here it is assumed that the wave function of the Abrikosov state for a $d_{x^2-y^2}$-wave superconductor can be written as

$$|\Psi\rangle = \left( 1 + C \left( a^\dagger \right)^{3} \right) |0\rangle,$$

where the “vacuum” $|0\rangle$ is the Abrikosov state of an $s$-wave superconductor, and $a^\dagger$ is the raising operator of the Landau level. In other words, $|0\rangle$ is a combination of the $N = 0$ Landau states. For $d_{x^2-y^2}$-wave superconductors an admixture of higher Landau states that are allowed by symmetry needs to be included in order to account for structural changes in the vortex lattice.

![FIG. 1. Temperature dependence of (a) the upper critical field and (b) the admixture parameter $C$ in a $d_{x^2-y^2}$-wave superconductor with $g$-factors $g=0$, 1.5, and 2. The magnetic field is applied along the crystal $c$-direction.](image)

In Fig. 1(a), experimental data of $H_{c2}(T)$ along the $c$-axis in CeCoIn$_5$ is compared with the numerical solution of Eqs. 1 and 2, describing a $d_{x^2-y^2}$-wave vortex lattice. The temperature dependence of the upper critical field $H_{c2}(T)$ is extracted from low-temperature thermal expansion, $\Delta l(T, B = \text{const})$, and magnetostriction $\Delta l(B, T = \text{const})$ measurements utilizing a high-resolution capacitive dilatometer at temperatures down to 15 mK and in magnetic fields up to 18 T. [15] For temperatures above $T_0 \approx 0.7K$, the superconducting-to-normal phase transition is of second order and the $H_{c2}(T)$ is determined from the midpoint of idealized jumps in $\partial\Delta l/\partial T$ and $\partial\Delta l/\partial B$. Below $T_0$ sharp jumps in $\Delta l$ are observed upon crossing $H_{c2}(T)$, indicative of a first order phase transition [24]. The best fit with a numerical solution of Eqs. 1 and 2 is obtained with $v = 3.2738 \times 10^8$ cm/sec, and $g=1.5$. For comparison, solutions with the same Fermi velocity $v$, but $g= 0$ and 2 are also shown. The fit to the experiment appears to be very good in

\[ T_K \]
the low-temperature regime $T < 0.7 K$. Therefore, there are presently no obvious indications for a FFLO state in this material by measurements of $H_{c2}(T)$ along the crystal $c$-direction.

In Fig. 1(b) we show the numerical solution for the admixture parameter $C$. Interestingly, for $g \geq 1.2$, $C$ changes its sign as the temperature is decreased. Consequently, for $g = 1.5$ one finds that the conventional hexagonal vortex lattice, which is stable at high temperatures, may change into a square vortex for $T/T_c < 0.3$. This transition should be observable by small angle neutron scattering (SANS) with $\vec{H} \parallel \hat{c}$. A similar instability to a square vortex lattice was previously predicted for the high-$T_c$ cuprates in the vicinity of $H_{c2}$. [14] Both, SANS [16] and STM [17] on the vortex state of YBCO single crystals indeed indicate significant deviations from a hexagonal towards a square vortex lattice in this compound. [18] Furthermore, the above theory was recently extended to lower magnetic fields. [19] The predicted square vortex lattice was observed by SANS in a single crystal of LSCO at $H = 2$ Tesla. [20]

III. $H_{c2}(T)$ for $\vec{H} \parallel \hat{a}$

In order to match the experimental data for $H_{c2}(T)$ along the crystal $a$-axis, we explore the effect of a $\vec{v} \cdot \vec{q}$ term arising from the formation of a FFLO state. [21] This leads to a new set of coupled integral equations,

$$-\ln t = \int_0^\infty \frac{du}{\sinhu} \left[ 1 - \langle \exp(-\rho u^2|\vec{a}|^2) \text{cos} [h(1 - p \text{cos} \phi)u] (1 + \text{cos}(4 \phi)) (1 - 2\rho u^2 s^2 C) \rangle \right]$$

$$-C \ln t = \int_0^\infty \frac{du}{\sinhu} \left[ C - \langle \exp(-\rho u^2|\vec{a}|^2) \text{cos} [h(1 - p \text{cos} \phi)u] (1 + \text{cos}(4 \phi)) (\rho u^2 s^2 + C (1 - 4\rho u^2|\vec{a}|^2 + 2\rho^2 u^4|\vec{a}|^4)) \rangle \right],$$

where $s \equiv \sin \chi + i \sin \phi$, $\rho \equiv (v_{vc} \epsilon H)/(8\pi^2 T^2)$, $p \text{cos} \phi \equiv (\vec{v} \cdot \vec{q})/(2h)$, $\chi \equiv c k_2$, and (...) is the angular average over $\phi$ and $\chi$. Here $\sqrt{v_{vc}} = 1.63 \times 10^8 \text{cm/s}$ is used, and following Gruenberg and Gunther [22], we chose $\vec{q} \parallel \hat{a}$. In this configuration, the vortex state is represented by

$$|\Psi\rangle = \left(1 + C \left(a^+\right)^2\right)|0\rangle,$$

where the vacuum $|0\rangle$ is again the Abrikosov state of a simple $s$-wave superconductor [22], but mixing now occurs with the $N=2$ Landau level.

![Figure 2](image-url)

**FIG. 2.** Temperature dependence of (a) the upper critical field, (b) the $\vec{v} \cdot \vec{q}/(2\vec{H}) = p \text{cos} \phi$ term, and (c) the admixture parameter $C$ in a $d_{x^2-y^2}$-wave superconductor with $g$-factors $g = 0.64$ (solid lines) and 2 (dashed lines). In (a) the lower curves represent $p = 0$, i.e. absence of FFLO, whereas the upper curves have $p = 0.9$. The magnetic field is applied along the crystal $a$-direction. The experimental data (circles) is best described by $g = 0.64$ and $p = 0.9$.

Furthermore, let us note that for the purpose of In Fig. 2(a) $H_{c2}(T)$ is shown for $g = 0.64$ and 2 along with the measurements of the upper critical field along the $a$-axis of CeCoIn$_5$. We find that without the FFLO state ($p = 0$) one obtains a good fit to the experiment down to $T = 0.7 K$ with $g = 0.64$. However, for $T < 0.7 K$ the measured upper critical field is approximately linear in temperature. This feature can be reproduced by including a $\vec{v} \cdot \vec{q}$ term due to
the FFLO state with $p = 0.9$. For comparison, we also show results for $g = 2$ which yield a zero-temperature critical field that is less than half the value detected in the experiment. In Fig. 2(b) $p(t)$ is shown for $g = 0.64$ and $g = 2$. From this plot it is clear that the FFLO region expands as $g$ is increased. In Fig. 2(c) the coefficient $C(t)$ is shown. Here we observe that $C$ exhibits a significant temperature dependence for $g = 2$, whereas for $g = 0.64$ the admixture is almost negligible. [23]

Furthermore, let us note that for the purpose of the present discussion it was assumed that the transition at $H = H_{c2}$ is of second order. However, a number of experiments on CeCoIn$_5$ indicate a possible first order transition, and onset of magnetic order at $T < 0.8K$. [12,13,24,25] At the moment the nature of this magnetic order is unknown. In case it is a spin density wave, the condensation energy is expected to be relatively small, and consequently its effect on $H_{c2}(T)$ should be small as well. [28]

IV. Quasiparticle Density of States

Let us conclude this discussion of a possible FFLO state in CeCoIn$_5$ by calculating the shape of the associated quasiparticle density of states. In the vicinity of $H = H_{c2}$ and for $\vec{\alpha} || \vec{\alpha}$ this observable is well approximated by [6] 

$$
\frac{N(E)}{N_0} - 1 = \frac{\Delta^2}{4\sqrt{\pi}} \sum_{\pm} \left< \int_{-\infty}^{\infty} du \frac{\exp(-u^2) \cos(2\phi)}{E \pm \tilde{H}(1 - p \cos \phi) - \epsilon |s| u} \right>,
$$

where $\tilde{H} \equiv (\mu_B g H)/2$, $|s| = \sqrt{\sin^2(\phi) + \sin^2(\chi)}$, and $\epsilon \equiv \sqrt{\nu_\epsilon e H}$. Again, a finite $p$ indicates the presence of a FFLO state. This quasiparticle density of states is plotted in Fig. 3. For the parameters, we have chosen $p = 0.9$ and $\epsilon / \hbar = 0.2$, appropriate for CeCoIn$_5$ in the temperature regime $T < 0.1K$. In the absence of the FFLO state (Fig. 3(a)), there are two sharp resonances close to $E = \pm H$. In the presence of the FFLO state, more structure appears in the spectral response, as shown in Fig. 3 (b), with resonances at $E = \pm H(1 \pm p)$. Therefore, precision measurements of the quasiparticle density of states can provide a clear signal for the presence of FFLO states and the symmetry of the underlying superconducting order parameter. It should thus be of interest to conduct a scanning tunneling microscope study of the quasiparticle density of states in CeCoIn$_5$ at $T < 0.7K$ in order to further scrutinize the proposed FFLO state.

FIG. 3. Quasiparticle density of states of a $d_{x^2-y^2}$-wave superconductors in a magnetic field, (a) in the absence of the FFLO state ($p = 0$), and (b) in the FFLO state ($p = 0.9$).

V. Conclusions

In summary, the model calculation in this study incorporates consistently (i) the $d_{x^2-y^2}$-wave symmetry of the superconducting order parameter, (ii) the orbital effect, and (iii) a $\vec{v} \cdot \vec{q}$-term due to the formation of a FFLO state. The model appears to describe well the observed temperature dependence of the upper critical field in CeCoIn$_5$. Furthermore, it indicates a significant renormalization of the $g$-factor in this compound, as well as the presence of
a FFLO state at low temperatures if the applied field has an in-plane component. In this phase, the quasiparticle density of state is predicted to have a more complex structure. In order to further scrutinize the proposed model, it will be interesting to determine further relevant properties, such as the specific heat and the thermal conductivity.

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