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# Theory of a generalized Fulde–Ferrell–Larkin–Ovchinnikov state in heavy fermion and intermediate-valence superconductors

S. Takahashi <sup>a,\*</sup>, M. Tachiki <sup>a</sup>, R. Modler <sup>b</sup>, P. Gegenwart <sup>b</sup>, M. Lang <sup>b</sup>, F. Steglich <sup>b</sup>

<sup>a</sup> *Institute for Materials Research, Tohoku University 2-1-1 Katahira, Aoba-ku, Sendai 980, Japan*

<sup>b</sup> *Institut für Festkörperphysik, Technische Hochschule Darmstadt, 64289 Darmstadt, Hochschulstr. 8, Germany*

## Abstract

Anomalies in the magnetization and crystal dilatation have been observed in the mixed state of UPd<sub>2</sub>Al<sub>3</sub> and CeRu<sub>2</sub>. To explain the anomalies we study a superconducting system with large spin susceptibility. We predict a new superconducting state in which the superconducting order parameter has a periodic array of planar nodes perpendicular to the vortices. The nature of this generalized Fulde–Ferrell–Larkin–Ovchinnikov state is discussed in connection with the peak effect in the magnetization process of UPd<sub>2</sub>Al<sub>3</sub> and CeRu<sub>2</sub>.

## 1. Introduction

Recently, anomalies in the sample length and the magnetization process have been observed near  $H_{c2}$  in the mixed state of the heavy-fermion superconductor UPd<sub>2</sub>Al<sub>3</sub> [1–3] and the intermediate-valence superconductor CeRu<sub>2</sub> [4–8]. The magnetization process is anomalously reversible over a wide field range below a field  $H_i$  near  $H_{c2}$ , indicating weak vortex pinning. For fields  $H > H_i$ , however, an abrupt increase in the diamagnetic (paramagnetic) response for increasing (decreasing) fields appears as a consequence of trapped vortices inside the super-

conductors. This hysteretic peak (the so-called peak effect) indicates the onset of strong vortex pinning above  $H_i$ .

The UPd<sub>2</sub>Al<sub>3</sub> and CeRu<sub>2</sub> compounds belong to a class of clean type-II superconductors with large spin susceptibilities,  $\chi_{\text{spin}}$ , and the fields ( $H_i < H < H_{c2}$ ) at which the peak effect occurs are comparable to the paramagnetically limited field  $H_p = (4\pi\chi_{\text{spin}})^{-1/2}H_c$ ,  $H_c$  being the thermodynamic critical field [1–8]. Under these circumstances, one expects that the superconducting order parameter is modulated in order to induce the spin polarization as in the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state [9–11]. In the FFLO state, however, the magnetic field acts only on the spins of the conduction electrons. In this paper, we study the interplay between the vortices and the modulated order parameter by including orbital and paramagnetic effects, and show that the order parameter has a periodic array of planar nodes perpendicular to the vortices. We dis-

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\* Corresponding author. Fax: +81 22 215 2006;  
e-mail: takahashi@tacsun.imr.tohoku.ac.jp.

cuss the peak effect in the magnetization process on the basis of this new superconducting state.

## 2. Formulation

We start with the following BCS Hamiltonian of the clean limit which includes the orbital and paramagnetic terms:

$$H = \int d^3r \sum_{\sigma} \psi_{\sigma}^{\dagger}(r) \left[ -\frac{\hbar^2}{2m_e} \left( \nabla + \frac{2ie}{\hbar c} A \right)^2 - \epsilon_F + \sigma \mu_B B(r) \right] \psi_{\sigma}(r) - \int d^3r (\Delta(r) \psi_{+}^{\dagger}(r) \psi_{-}^{\dagger}(r) + \Delta^{*}(r) \psi_{-}(r) \psi_{+}(r)), \quad (1)$$

where  $\psi_{\sigma}(r)$  is the annihilation operator of a quasiparticle with spin variable  $\sigma$ ,  $A$  the vector potential,  $\epsilon_F$  the Fermi energy,  $\mu_B$  the Bohr magneton,  $B(r) = \nabla \times A$  the magnetic field, and  $\Delta(r)$  the superconducting order parameter.

The equations for the Gor'kov Green functions derived from Eq. (1) are transformed into the quasi-classical equations by integrating the Green functions over the quasiparticle energy as [12,13]

$$\left[ \omega_s + \frac{v_F}{2} \hat{k}_F \cdot \left( \nabla + \frac{2ei}{\hbar c} A(r) \right) \right] \times f(r, \hat{k}_F, \omega_s) = \Delta(r) g(r, \hat{k}_F, \omega_s), \quad (2)$$

$$\left[ \omega_s - \frac{v_F}{2} \hat{k}_F \cdot \left( \nabla - \frac{2ei}{\hbar c} A(r) \right) \right] \times f^{+}(r, \hat{k}_F, \omega_s) = \Delta^{*}(r) g(r, \hat{k}_F, \omega_s), \quad (3)$$

$$g(r, \hat{k}_F, \omega_s) = \left[ 1 - f(r, \hat{k}_F, \omega_s) f^{+}(r, \hat{k}_F, \omega_s) \right]^{1/2}, \quad (\text{Re } g > 0) \quad (4)$$

where  $\omega_s = \omega_l + i\mu_B B$  with  $\omega_l = (2l+1)\pi k_B T$ ,  $\hat{k}_F$  is the unit vector in the direction of the Fermi momentum  $k_F$ , and  $v_F$  is the Fermi velocity. Eqs.

(2)–(4) are completed by the self-consistency equations for the order parameter  $\Delta(r)$ ,

$$\Delta(r) \ln \frac{T}{T_c} + 2\pi T \sum_{l=0}^{\infty} \left[ \frac{\Delta(r)}{\omega_l} - \frac{1}{2} \int \frac{d^2 \hat{k}_F}{4\pi} \times \left( f(r, \hat{k}_F, \omega_s) + f^2(r, \hat{k}_F, \omega_s^{*}) \right) \right] = 0, \quad (5)$$

and the magnetic field

$$\nabla \times B(r) = \frac{8\pi e}{\hbar c} 2\pi TN(0) \times \sum_{l=0}^{\infty} \int \frac{d^2 \hat{k}_F}{4\pi} v_F \hat{k}_F \text{Im } g(r, \hat{k}_F, \omega_s). \quad (6)$$

The difference in the Gibbs free energies between the superconducting and normal states is given by

$$G = \int d^3r \left\{ \frac{(B-H)^2}{8\pi} + \Delta^2 N(0) \ln \frac{T}{T_c} + 2\pi TN(0) \times \sum_{l=0}^{\infty} \left[ \frac{\Delta^2}{\omega_l} - \text{Re} \int \frac{d^2 \hat{k}_F}{4\pi} \frac{\Delta f^{+} + \Delta^{*} f}{1+g} \right] \right\}, \quad (7)$$

where  $H$  is an applied magnetic field in the  $z$ -direction.

Near  $H_{c2}$ , if we linearize the Eilenberger equations (2) and (3) with respect to  $\Delta(r)$ , we find a solution of the form

$$\Delta(r) = \Psi_v(x, y) \sin Qz, \quad (8)$$

where  $\Psi_v(x, y)$  is the order parameter of the Abrikosov vortex lattice and  $Q$  is a wave number [14]. Below  $H_{c2}$  we assume the order parameter to be of the form

$$\Delta(r) = \Psi_v(x, y) \eta(z), \quad (9)$$

as a generalization of Eq. (8), and obtain  $\eta(z)$  by solving the Eilenberger equations (2)–(6) self-consistently by numerical computation. A more detailed calculation will be published elsewhere.

## 3. Nature of the generalized FFLO state

Using the relaxation method, we solve the Eilenberger equations (2) and (3) and the gap equation (5)

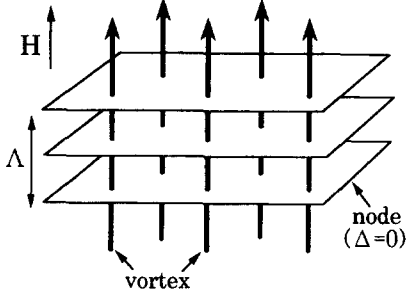


Fig. 1. Schematic diagram of the GFFLO state. The planar nodes of  $\Delta(r)$  are periodically arrayed with  $\Lambda/2$ .

by assuming  $\eta(z) = \eta(z + \Lambda)$ ,  $\Lambda$  being the wavelength of the oscillation. The solutions are used to calculate the Gibbs free energy, Eq. (7).  $\Lambda$  is determined by minimizing the Gibbs free energy with respect to  $\Lambda$ .

The paramagnetic effect relative to the orbital effect is characterized by the parameter  $\alpha = (H_{c2}^{(\text{orb})}/H_p)/\sqrt{2}e^{2+\gamma}$ , where  $\gamma = 0.57721$  is the Euler and  $H_{c2}^{(\text{orb})}$  is the orbital critical field. The numerical calculation shows that for  $\alpha > 0.07$  the order parameter  $\Delta(r)$  oscillates along the  $z$ -axis. As the applied magnetic field decreases from  $H_{c2}$ ,  $\Delta(r)$  changes its shape from a sinusoidal to a rectangular one [11]. Thus,  $\Delta(r)$  has a regular array of planar nodes perpendicular to the vortices, as shown in Fig. 1. We call the superconducting state having the structure in Fig. 1 the *generalized* FFLO state (referred to as the GFFLO state). The GFFLO state resembles the vortex state of layered oxide superconductors in magnetic fields perpendicular to the  $\text{CuO}_2$ -planes.

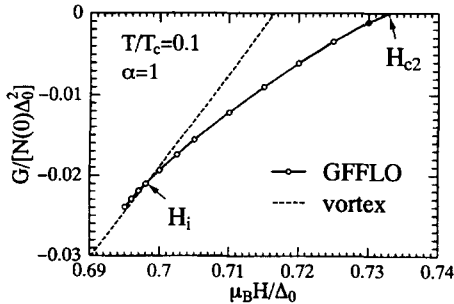


Fig. 2. Gibbs free energy of the GFFLO state measured from that of the normal state. The solid and dashed curves show the free energies of the GFFLO state and the Abrikosov vortex state, respectively.

The Gibbs free energy of the GFFLO state is shown for  $\alpha = 1$  in Fig. 2. When the applied magnetic field increases, the superconductor undergoes a first-order phase transition at  $H_i$  from the vortex state to the GFFLO state, and at  $H_{c2}$  from the GFFLO state to the normal state. The wavelength  $\Lambda$  increases monotonically from  $\Lambda \sim 14\xi_0$  to  $\Lambda \sim 40\xi_0$  as the temperature decreases from  $H_{c2}$  to  $H_i$ .

In the GFFLO state the spin susceptibility of the quasiparticles is recovered around the planar nodes and the vortex cores, causing a spin polarization there. The spin density of quasiparticles  $m = \langle \psi_+^\dagger \psi_+ \rangle - \langle \psi_-^\dagger \psi_- \rangle$  is given by

$$m = m_N - 4\pi N(0)T \sum_{l=0}^{\infty} \int \frac{d^2 \hat{k}_F}{4\pi} \text{Im} \left[ g(r, \hat{k}_F, \omega_l + i\mu_B B) \right], \quad (10)$$

where  $m_N$  is the spin density in the normal state. The spin polarizations appearing around the nodes and the vortex cores are shown in Figs. 3(a) and (b). The spin density is well reproduced by the formula  $m \approx m_N(1 - [\Delta(r)/\Delta_0]^2)$  if the calculated values of  $\Delta(r)$  are used. The spin polarization is expected to be observable by neutron diffraction experiments in these compounds.

In the GFFLO state we expect a bound state around the nodes and the vortex centers, since the order parameter changes its sign there. The density

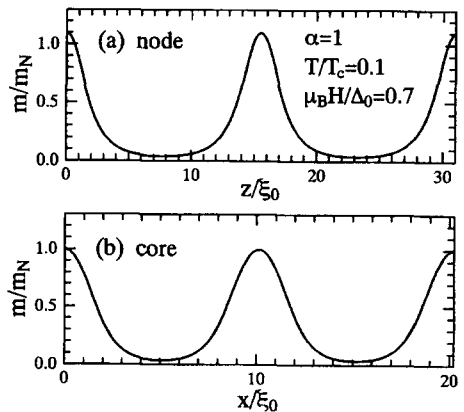


Fig. 3. Spin polarization induced around the nodes (a) and the vortex cores (b). The spin density  $m$  is normalized to that in the normal state  $m_N$ .

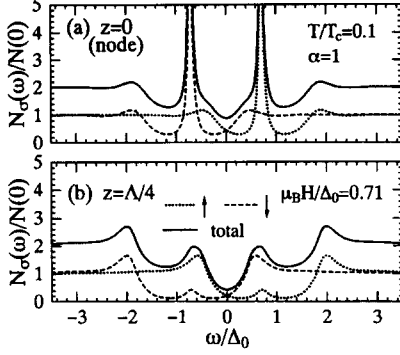


Fig. 4. Local densities of states at the node ( $z = 0$ ) and away from the node ( $z = \Lambda/4$ ). The dotted and dashed curves indicate those for up and down spins, respectively, and the solid curve indicates the sum of them.

of states for the quasiparticles  $N_\sigma(\omega)$  with spin  $\sigma$  is calculated from the expression

$$N_\sigma(\omega) = N(0) \int \frac{d^2 \hat{k}_F}{4\pi} \times \text{Re} \left[ g(r, \hat{k}_F, \omega - \sigma \mu_B H) \right]. \quad (11)$$

Fig. 4 shows the local densities of states at different positions. In Fig. 4(a) we see the sharp peaks associated with the bound state split by the Zeeman energy  $2\mu_B H$ , which may be observed in the  $I$ - $V$  characteristics as taken by scanning tunneling spectroscopy.

#### 4. Discussions

We briefly discuss the peak effect in the magnetization process mentioned in the Introduction on the basis of the generalized FFLO state. The reversible magnetization process in a wide range of the vortex state below  $H_i$  is explained as follows. The superconducting order parameter in the vortex state vanishes inside the vortex cores, causing a loss of the superconducting condensation energy. In the superconductors having large  $\chi_{\text{spin}}$ , the spin polarization, recovered inside the vortex core, causes a decrease of the magnetic energy. If the vortex core is approximated by a cylindrical core of radius  $\xi_0$ , the core energy is given by

$$E_{\text{core}} = \pi \xi_0^2 \left[ \frac{H_c^2}{8\pi} - \frac{1}{2} \chi_{\text{spin}} B^2(0) \right], \quad (12)$$

where  $B(0)$  is the magnetic field at the vortex core. At high magnetic fields  $B(0) \approx H$ , so that  $E_{\text{core}} \approx \pi \xi_0^2 (H_c^2/8\pi) [1 - (H/H_p)^2]$ . Thus, for fields near  $H_p$ , the core energy almost vanishes owing to the cancellation between the loss of the condensation energy and the gain of the spin-polarization energy. In this case, the vortex pinning force acting on the straight (rigid) vortices becomes very weak.

On the other hand, in the GFFLO state above  $H_i$ , the vortices are cut by the planar nodes of the order parameter into segments of length  $\Lambda$ . As a consequence, the vortices become flexible in a qualitatively similar way to the pancake vortices in high- $T_c$  cuprate superconductors. If the vortices are subject to a weak disorder potential as in the presence of point defects, we expect the individual vortex segments to accommodate themselves more easily to the pinning potential, thereby becoming efficiently pinned by the collective action of the pinning centers [15]. This pinning mechanism significantly enhances the pinning capability of the GFFLO state and may explain the peak effect in  $\text{UPd}_2\text{Al}_3$  and  $\text{CeRu}_2$ .

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