In-plane angular dependence of the upper critical field in $CeCoIn_5$

Franziska Weickert and Philipp Gegenwart
Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany

Hyekyung Won
Department of Physics,Hallym University, Chunchon 200-702, South Korea

David Parker* and Kazumi Maki
Department of Physics and Astronomy, University of Southern California, Los Angeles, California 90089-0484, USA
(Received 9 January 2006; revised manuscript received 3 August 2006; published 20 October 2006)

The tetragonal $CeCoIn_5$ is an unconventional d-wave heavy-fermion superconductor with T_c=2.3 K. Strong Pauli limiting causes a first-order superconducting transition for fields close to $H_{c2}(0)$ and evidence for a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state has been found at low temperatures. Here, we present a study of the in-plane angular dependence of the upper critical field. At T=0.1 K a fourfold oscillation of 1.2% amplitude with maxima along the [100] direction is observed. This points toward a $d_{x^2-y^2}$ order-parameter symmetry. The data are compared with theoretical calculations including the FFLO state and a tiny anisotropy of the g factor.

DOI: 10.1103/PhysRevB.74.134511 PACS number(s): 74.70.Tx, 74.20.Rp, 74.25.Fy, 71.27.+a

I. INTRODUCTION

The tetragonal heavy-fermion system $CeCoIn_5$ has attracted much interest because of its unusual normal and superconducting (SC) properties. An unconventional SC state below T_c=2.3 K is indicated by power-law behavior in specific heat and thermal conductivity.[1] Its d-wave nature has been deduced from its thermal conductivity studied in a field rotating in the tetragonal plane.[2] Further, highly anomalous behavior has been observed at magnetic fields close to the upper critical field $H_{c2}(0)$, which amounts to about 5 and 11.5 T along and perpendicular to the c axis, respectively. Strong Pauli limiting leads to a first-order superconducting transition at temperatures below 0.7 K for both field orientations. [2–5] Additionally, for fields aligned within the tetragonal plane, specific heat experiments have revealed an additional second-order phase transition in the vicinity of H_{c2} and for $T \leq 0.3$ K. [5,6] Since for this field orientation the upper critical field exceeds the Pauli limit and the system is in the clean limit with a quasiparticle mean free path much larger than the SC coherence length, the formation of a spatially modulated SC state, first predicted by Fulde and Ferrell as well as Larkin and Ovchinnikov[7] (FFLO) has been proposed. This has also been supported by ultrasound velocity [8] and NMR[9] measurements; in the latter a second resonance line in the FFLO state proves the emergence of normal quasiparticles.

The normal state of $CeCoIn_5$ is also very unusual. Specific heat[10] and electrical resistivity[11] experiments provide evidence for non-Fermi-liquid behavior which may hint at a magnetic quantum critical point (QCP) located very close to H_{c2}. Alternatively, from the observation of a giant Nernst effect[12] and the angular dependence of the normal state magnetoresistance[13] the formation of an unconventional d-wave density wave phase has been proposed in this region of the phase diagram.[14,15]

In this paper, we focus on the determination of the SC order parameter in $CeCoIn_5$. Although several experiments hint at a d-wave SC order parameter, its exact symmetry is still under debate. Clear fourfold oscillations have been observed in measurements of the thermal conductivity[2] and specific heat[16] when the field was applied in the plane perpendicular to the c axis. Whereas Izawa *et al.* deduce a $d_{x^2-y^2}$ order-parameter symmetry, Aoki *et al.* argue in favor of d_{xy} symmetry. Determination of the in-plane anisotropy of the upper critical field should resolve this issue as smaller values are expected along the gap node directions. However, there have been conflicting reports. Whereas Murphy *et al.* found H_{c2} minima along the [100] direction, [17] Bianchi *et al.* reported minima along the [110] direction.[5] Below we present a detailed study of the temperature and in-plane angular dependence of the upper critical field and compare our results with calculations including a FFLO SC state at low temperatures.

II. EXPERIMENT

For the measurements of the in-plane angular dependence of the upper critical magnetic field of $CeCoIn_5$ a single axis manual drive Swedish rotator has been adapted to a dilution refrigerator with a 20 T superconducting magnet. We used a platelike single crystal of rectangular shape grown at Los Alamos. Careful Laue diffraction has been performed along the three main directions and compared with simulations. The resulting patterns prove that the orientation of the piece is such that the two longer and the shorter sides point along the crystallographic [100] and [001] directions, respectively [see sketch inside Fig. 1(a)]. The electrical resistivity of the plate has been measured using a low-frequency four-point ac method. Excitations currents of 0.1 mA have been applied along the [100] direction. The sample was mechanically and thermally anchored to a small silver plate mounted perpendicular to the rotation axis inside the plastic rotator. The angular position Θ in the tetragonal sample plane with respect to the applied magnetic field is determined with the

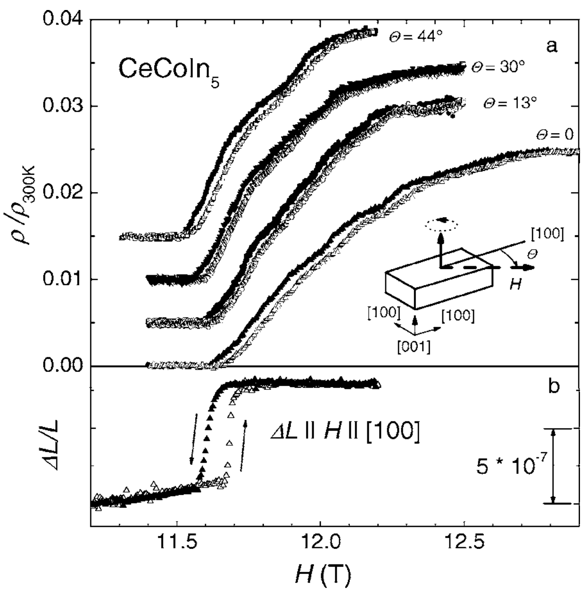

FIG. 1. Magnetic field dependence of the electrical resistivity of CeCoIn$_5$ at 0.1 K for various angles Θ between the applied field H and the [100] direction as indicated by the sketch (a). The open and closed symbols represent data obtained in increasing and decreasing H, respectively. Isothermal length change $\Delta L/L$ at 0.1 K along the [100] direction (b).

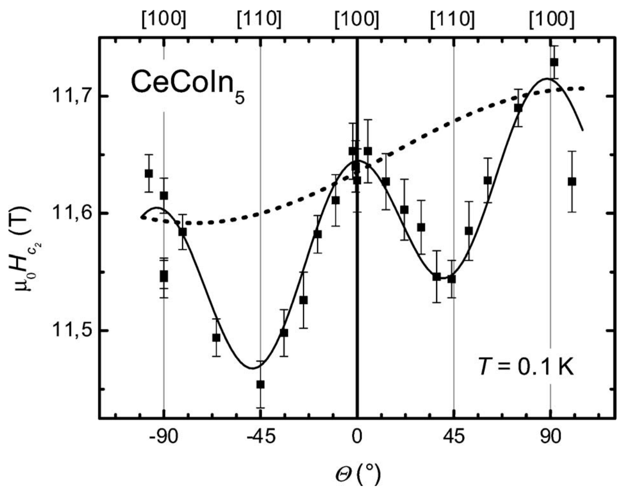

FIG. 2. Upper critical field H_{c2} derived from resistive transition vs angle Θ from [100] direction (see sketches in Fig. 1). The solid line is a fit of the data using the sum of two cosine functions with periodicity of 90° and 360°, respectively. The dotted line indicates the influence of the twofold oscillation due to the misalignment of the sample (see text).

help of a small coil (20 turns) mounted parallel to the sample surface along the [100] direction. An ac field modulation (0.3 mT applied with a superconducting coil mounted inside the main superconducting magnet) induces a voltage in this coil that is proportional to the cosine of Θ. This method allows us to resolve changes in Θ of less than 1°.

III. IN-PLANE ANISOTROPY OF H_{c2}

Figure 1(a) shows the low-temperature (0.1 K) electrical resistivity of CeCoIn$_5$ upon sweeping the magnetic field between 11 and 13 T. We find a clear hysteresis indicative of a first-order phase transition, compatible with previous experiments.[4] At this large magnetic field, the resistive transition is broadened significantly. We have determined H_{c2} from the field at which the resistivity reaches zero, as these values for $\mathbf{H}\|[100]$ agree well with the ones obtained from magnetostriction experiments[18] performed on the same sample [see Fig. 1(b)]. Clearly, H_{c2} along [100] exceeds the critical field along [110] in agreement with Ref. 5 but opposite to the results reported by Murphy *et al.*[17] Since for an unconventional superconducting order parameter, $H_{c2}(\Theta)$ maxima occur when the magnetic field in the real space is parallel to the antinodal directions,[19] these data are compatible with a $d_{x^2-y^2}$ order parameter but inconsistent with the d_{xy} symmetry proposed in Ref. 16.

Upon rotating the magnetic field in the tetragonal plane perpendicular to [001], a clear fourfold oscillation of $H_{c2}(\Theta)$ is observed, where Θ denotes the angle from the [100] direction (see Fig. 2). The amplitude of this oscillation amounts to 1.2%. Superposed on the fourfold oscillation is an oscillation with periodicity of 2π which arises from a slight misalignment of the sample with respect to the rotation axis and center of magnetic field. For the following analysis, we use $\tilde{H}_{c2}(\phi_0)$, representing the data after subtraction of this latter contribution (ϕ_0 is the angle with respect to the [100] direction in radians).

IV. MODEL CALCULATIONS

Here we shall analyze $\tilde{H}_{c2}(T,\phi_0)$ within the weak-coupling BCS theory for d-wave superconductors as employed in Ref. 18. Although an enhanced specific heat jump $\Delta C/\gamma T_c \approx 4.5$ has been observed in CeCoIn$_5$,[20] this cannot be taken as an indication for strong-coupling superconductivity. In this material, the normal state specific heat coefficient $C(T)/T$, measured at $H=H_{c2}$, is not constant, as expected for a Landau Fermi liquid, but strongly increases with decreasing temperatures, reaching about 1 J/K^2 at 0.1 K. Taking this value as a lower bound for the normal state γ value, also justified by the entropy balance between the normal and SC states,[20] yields a reduced jump height of only 1.3. This justifies the weak-coupling approach used in the following. First, the formula for H_{c2} for s-wave superconductors in Ref. 21 has to be generalized for unconventional superconductors.[22] Second, the layered structure has to be incorporated following Ref.19. Furthermore, the expression for H_{c2} for d-wave superconductors including the FFLO state[23] has to be extended to incorporate the orbital effect following Gruenberg and Gunther.[24] Then, for an arbitrary angle ϕ_0 between the field direction and the [100] direction, $\tilde{H}_{c2}(\phi_0,T)$ is determined from

$$-\ln t = \int_0^\infty \frac{du}{\sinh u}\{1-\langle[1+\cos(4\phi_0)\cos(4\phi)] \times e^{-\rho u^2|s|^2}\cos[h(1-p\cos\phi)u](1+2C\rho u^2 s^2)\rangle\}, \tag{1}$$

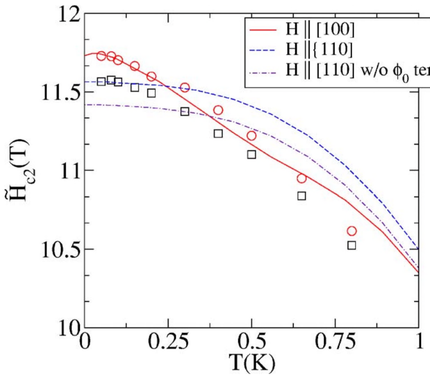

FIG. 3. (Color online) Comparison between experimental $\tilde{H}_{c2}(T)$ data of $CeCoIn_5$ for $\phi_0=0$ ($H\|[100]$, red open circles) and $\phi_0=\pi/4$ ($H\|$ [110], black open squares) with theoretical calculations indicated by lines. Dash-dotted and dashed lines represent calculation for $g=0.624$ and $0.6195+0.0045\cos(4\phi_0)$, respectively.

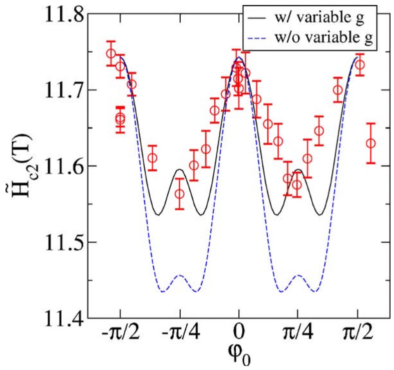

FIG. 4. (Color online) Comparison between $\tilde{H}_{c2}(\phi_0)$ data of $CeCoIn_5$ at $T=0.1$ K (red open circles) and theoretical calculations for $g=0.621$ (dashed line) and $0.61675+0.00425\cos(4\phi_0)$ (solid line).

$$-C\ln t=\int_0^\infty \frac{du}{\sinh u}\{C-\langle[1+\cos(4\phi_0)\cos(4\phi)]\times e^{-\rho u^2|s|^2}\cos[h(1-p\cos\phi)u]\times[\rho u^2 s^2+C(1-4\rho u^2|s|^2+2\rho^2u^4|s|^4)]\rangle\} \quad (2)$$

$$0=\int_0^\infty \frac{du}{\sinh u}\{\langle[1+\cos(4\phi_0)\cos(4\phi)]e^{-\rho u^2|s|^2}\times\sin[h(1-p\cos\phi)u](1+2\rho u^2 s^2)u\cos(\phi)\rangle\}, \quad (3)$$

where $t=T/T_c$, $h=(g\mu_B H)/(2\pi T)$, and $s=\sin\chi+i\sin\phi$. ϕ is the angle made by the quasiparticle velocity within the plane perpendicular to [001] with H, C is the weight of the $N=2$ Landau wave function, $\rho=vv_c eH/2(2\pi T)^2$, $p=v|q|/2h$, where $2\pi/q$ is the period of the spatial oscillation of $\Delta(\mathbf{r})$, $\chi=ck_z$, and $\langle\cdots\rangle$ means the average over ϕ and χ. The optimization of H_{c2} in terms of p gives Eq. (3). As previously we make use of the Gruenberg-Guenther ansatz[24] to construct the solution

$$|\Psi\rangle=\cos(\mathbf{q}\cdot\mathbf{r})[1+C(a^\dagger)^2]|0\rangle \quad (4)$$

where $\mathbf{q}\|\mathbf{H}$, $|0\rangle$ is the Abrikosov solution,[25] and $a^\dagger$ is the raising operator. In other words the vortex state wave function $\Delta(\mathbf{r})$ in d-wave superconductors is constructed from the $N=0$ and $N=2$ Landau wave functions.[19,22]

The two fitting parameters used in the theoretical description of the upper critical field are the gyromagnetic ratio g and the Fermi velocity v (incorporated into ρ). The values of $\tilde{H}_{c2}(\phi_0,T)$, the admixture parameter C, and the FFLO coefficient p are determined by solving for the unique solution of Eqs. (1)–(3), subject to the above fitting parameters. Figure 3 displays $\tilde{H}_{c2}(\phi_0,T)$ for $\phi_0=0$ (i.e., $\mathbf{H}\|$ [100]) and $\phi_0=\pi/4$ ($\mathbf{H}\|$ [110]) together with the experimental data, which show a peculiar linear increase with decreasing temperature for both field orientations. Since the overall fitting is excellent as in Ref. 18, we concentrate here on the region $T\leq 1$ K. The solid curve is the fit to the data for $\mathbf{H}\|$ [100], which gives $g=0.624$. However, this ϕ_0-independent g gives a rather poor fitting of $\tilde{H}_{c2}(T)$ for $\mathbf{H}\|$ [110] and $\tilde{H}_{c2}(\phi_0)$ at $T=0.1$ K. Therefore we have chosen a ϕ_0-dependent g factor: $g(\phi_0)=0.6195+0.0045\cos(4\phi_0)$, corresponding to a tiny anisotropy of the g factor. This allows us to fit $\tilde{H}_{c2}(\pi/4,T)$ for $T<0.2$ K fairly well, although the theoretical curve is rather flat at low temperatures. Of course the calculated $\tilde{H}_{c2}(\phi_0,T)$ for $\phi_0=0$ and $\phi_0=\pi/4$ are somewhat reminiscent of the Pauli limit discussed in Ref. 23.

In Fig. 4 we focus on the angular dependence of the upper critical field at $T=0.1$ K, i.e., well inside the FFLO state. The experimental data show a $\cos(4\phi_0)$ modulation. On the other hand the theoretical result suggests the presence of an eightfold term in the FFLO state which has not been observed experimentally.

Figure 5 depicts the temperature and angular dependence of the admixture coefficient C and the FFLO coefficient p. In

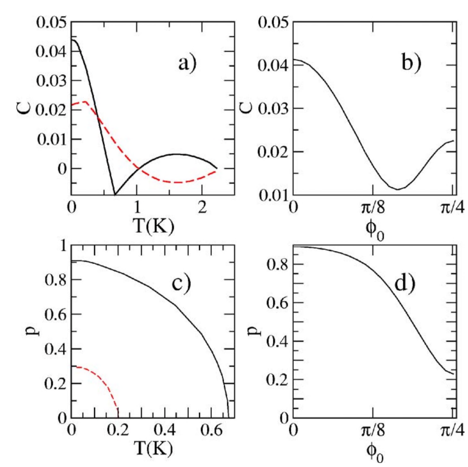

FIG. 5. (Color online) Temperature dependence of the coefficient C for $\mathbf{H}\|$ [100] (solid) and $\mathbf{H}\|$ [110] (dashed) (a). Angular dependence of C at $T=0.1$ K (b). Temperature dependence of p $(=\mathbf{v}\cdot\mathbf{q}/2h)$ for $\mathbf{H}\|$ [100] (solid) and $\mathbf{H}\|$ [110] (dashed) (c). Angular dependence of p at $T=0.1$ K (d).

particular, a sharp cusp in C at $T=0.67$ K for $\mathbf{H}\parallel[100]$, and a smaller cusp at 0.2 K for $\mathbf{H}\parallel[110]$ are evident. These cusps are directly associated with the abrupt onset of the FFLO state at these temperatures [see Fig. 5(c)]. C and p also show significant angular variation at 0.1 K, which lies within the FFLO state for all angles. Fourfold as well as eightfold and higher harmonic angular dependence is present for both these parameters. The eightfold and higher angular dependencies arise from the mixing of the FFLO term $\cos[h(1-p\cos\phi)u]$ in Eqs. (1) and (2) with the other, fourfold-dependent terms in these equations. Outside the FFLO state $\widetilde{H}_{c2}(\phi_0,T)$ has only a fourfold dependence.[19]

The comparison with the experimental data indicates that within our simple model we obtain an excellent fit for $\mathbf{H}\parallel[100]$. On the other hand, the fitting of $\widetilde{H}_{c2}(T)$ for $\mathbf{H}\parallel[110]$ is not so perfect. This suggests that there is still a missing element in our understanding of the FFLO state for $\mathbf{H}\parallel[110]$. Possibly, the proposed unconventional d-wave density wave[14,15] coexisting with d-wave superconductivity may have some additional effect on the angular dependence of the upper critical field.

V. CONCLUSIONS

Our study of the in-plane angular dependence of the upper critical field in $CeCoIn_5$ at very low temperatures has revealed a fourfold oscillation of the upper critical field with maxima for fields applied parallel to the [100] direction. This is compatible with a $d_{x^2-y^2}$ order-parameter symmetry in this system but excludes the d_{xy} symmetry proposed in Ref. 16. Furthermore, the temperature dependence of the upper critical field has been determined at low temperatures for $\mathbf{H}\parallel[100]$ and $\mathbf{H}\parallel[110]$. At low temperatures a peculiar linear increase with decreasing temperature is found for both field orientations. Model calculations have been performed within the weak-coupling BCS theory for d-wave superconductivity including the FFLO state. Using the Gruenberg-Gunther ansatz and assuming a possible small in-plane anisotropy of the g factor gives an excellent description of the $\mathbf{H}\parallel[100]$ data, whereas the fitting of $\widetilde{H}_{c2}(T)$ for $\mathbf{H}\parallel[110]$ is not perfect. In particular, the theory predicts an additional eightfold oscillating term which is not observed experimentally. We speculate that the origin of this discrepancy is related to the unusual normal state properties of $CeCoIn_5$ with strong magnetic fluctuations, caused by incipient antiferromagnetism and a hidden QCP.[10] Such behaviors which have not been modeled here, may tend to wash out small-angle effects such as the eightfold term.

ACKNOWLEDGMENTS

We thank J.L. Sarrao for providing a high-quality $CeCoIn_5$ single crystal and N. Caroca-Canales and R. Cardoso for helpful advice concerning the determination of the sample orientation. We have benefited from useful discussions with A. Bianchi, S. Haas, M. Nicklas, N. Oeschler and P. Thalmeier. K.M., D.P., and H.W. acknowledge the hospitality and support of Max-Planck Institute for Physics of Complex Systems at Dresden, where a part of this work was done. H.W. acknowledges support from the Korean Research Foundation (KRF) through Grant No. R05-2004-000-10814.

*Present address: Max-Planck Institute for Physics of Complex Systems, D-01187 Dresden

[1] R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **86**, 5152 (2001).
[2] K. Izawa, H. Yamaguchi, Yuji Matsuda, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. Lett. **87**, 057002 (2001).
[3] T. Tayama, A. Harita, T. Sakakibara, Y. Haga, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. B **65**, 180504(R) (2002).
[4] A. Bianchi, R. Movshovich, N. Oeschler, P. Gegenwart, F. Steglich, J. D. Thompson, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **89**, 137002 (2002).
[5] A. Bianchi, R. Movshovich, C. Capan, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **91**, 187004 (2003).
[6] H. A. Radovan, N. A. Fortune, T. P. Murphy, S. T. Hannahs, E. C. Palm, S. W. Tozer, and D. Hall, Nature (London) **425**, 51 (2003).
[7] P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964); A. I. Larkin and Y. N. Ovchinnikov, Sov. Phys. JETP **20**, 762 (1965).
[8] T. Watanabe, Y. Kasahara, K. Izawa, T. Sakakibara, Y. Matsuda, C. J. van der Beek, T. Hanaguri, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. B **70**, 020506(R) (2004).
[9] K. Kakuyanagi, M. Saitoh, K. Kumagai, S. Takashima, M. Nohara, H. Takagi, and Y. Matsuda, Phys. Rev. Lett. **94**, 047602 (2005).
[10] A. Bianchi, R. Movshovich, I. Vekhter, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **91**, 257001 (2003).
[11] J. Paglione, M. A. Tanatar, D. G. Hawthorn, E. Boaknin, R. W. Hill, F. Ronning, M. Sutherland, L. Taillefer, C. Petrovic, and P. C. Canfield, Phys. Rev. Lett. **91**, 246405 (2003).
[12] R. Bel, K. Behnia, Y. Nakajima, K. Izawa, Y. Matsuda, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. Lett. **92**, 217002 (2004).
[13] T. Hu, H. Xiao, T. A. Sayles, M. B. Maple, Kazumi Maki, B. Dóra, and C. C. Almasan, Phys. Rev. B **73**, 134509 (2006).
[14] B. Dóra, K. Maki, A. Virosztek, and A. Ványolos, Phys. Rev. B **71**, 172502 (2005).
[15] B. Dóra, K. Maki, and A. Virosztek, Europhys. Lett. **72**, 624 (2005).
[16] H. Aoki, T. Sakakibara, H. Shishido, R. Settai, Y. Onuki, P. Miranovic, and K. Machida, J. Phys.: Condens. Matter **16**, L13 (2004).
[17] T. P. Murphy, D. Hall, E. C. Palm, S. W. Tozer, C. Petrovic, Z. Fisk, R. G. Goodrich, P. G. Pagliuso, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B **65**, 100514(R) (2002).
[18] H. Won, K. Maki, S. Haas, N. Oeschler, F. Weickert, and P. Gegenwart, Phys. Rev. B **69**, 180504(R) (2004). Note the presence of several misprints in this paper.

[19] H. Won and K. Maki, Europhys. Lett. **34**, 453 (1996).
[20] C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, Z. Fisk, and P. Monthoux, J. Phys.: Condens. Matter **13**, L337 (2001).
[21] L.P. Gor'kov, Sov. Phys. JETP **10**, 59 (1960).
[22] I. A. Luk'yanchuk and V. P. Mineev, Sov. Phys. JETP **66**, 1168 (1987).
[23] K. Maki and H. Won, Czech. J. Phys. **46**, 1035 (1996).
[24] L. W. Gruenberg and L. Gunther, Phys. Rev. Lett. **16**, 996 (1966).
[25] A. A. Abrikosov, Sov. Phys. JETP **5**, 1174 (1957).