PHYSICAL REVIEW B **69**, 180504(R) (2004)

Upper critical field and Fulde-Ferrell-Larkin-Ovchinnikov state in $CeCoIn_5$

Hyekyung Won,[1] Kazumi Maki,[2] Stephan Haas,[2] Niels Oeschler,[3] Franziska Weickert,[3] and Philipp Gegenwart[3]
[1]*Department of Physics, Hallym University, Chunchon 200-702, South Korea*
[2]*Department of Physics and Astronomy, University of Southern California, Los Angeles, California 90089-0484, USA*
[3]*Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany*
(Received 11 March 2004; published 6 May 2004)

Angle 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 $\vec{H}\|\vec{a}$ and $\vec{H}\|\vec{c}$. The data are 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 $\vec{H}\|\vec{a}$ is consistent with a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state at low temperatures, $T<0.7$ K, whereas for $\vec{H}\|\vec{c}$ 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.

DOI: 10.1103/PhysRevB.69.180504 PACS number(s): 74.70.Tx, 71.27.+a, 74.20.Rp, 74.25.Fy

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 nonuniform 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 κ, measuring the ratio of the magnetic penetration depth versus the superconducting coherence length, should be very large, i.e., $\kappa \gg 10$.

The recent synthesis of quasi-two-dimensional(quasi-2D) nodal superconductors, such as the high-T_c cuprates, the κ-$(ET)_2$ 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 λ-$(BETS)_2GaCl_4$, λ-$(BEDTS)_2FeCl_4$, and κ-(BEDT-TTF)$_2$Cu(NCS)$_2$.[7–11] In the first compound, a kink in the thermal conductivity points to a transition from a FFLO state to a vortex lattice. In the last 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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ by angle dependent magnetothermal conductivity measurements in a rotating magnetic field within the conducting crystal plane.[12,13] Moreover, it was observed that the upper critical field $H_{c2}(T)$ in the FFLO regimes decreases quasilinearly 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,[14] and it has a layered structure similar to the high-T_c cuprates. Angle dependent magnetothermal conductivity experiments indicate $d_{x^2-y^2}$-wave superconductivity in this material.[15] Furthermore, the temperature dependence of the upper critical field $H_{c2}(T)$ for both $\vec{H}\|\vec{a}$ and $\vec{H}\|\vec{c}$ was measured in single crystals of $CeCoIn_5$.[16] It was observed that $H_{c2}(T)$ for $\vec{H}\|\vec{a}$ exhibits a quasilinear 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 of $CeCoIn_5$ is determined from thermal expansion and magnetorestriction measurements with field orientations $\vec{H}\|\vec{a}$ and $\vec{H}\|\vec{c}$. 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 $\vec{H}\|\vec{c}$ 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 $\vec{H}\|\vec{a}$ 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 $T=0.7$ K. In particular, the quasilinear 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 further test and scrutinize this model, we also determine the corresponding quasiparticle density of states which should be accessible to scanning tunneling microscopy(STM) experiments.

II. $H_{c2}(T)$ FOR $\vec{H}\|\vec{c}$

Within the $d_{x^2-y^2}$-wave BCS model, the temperature dependence of the upper critical field along the crystallo-

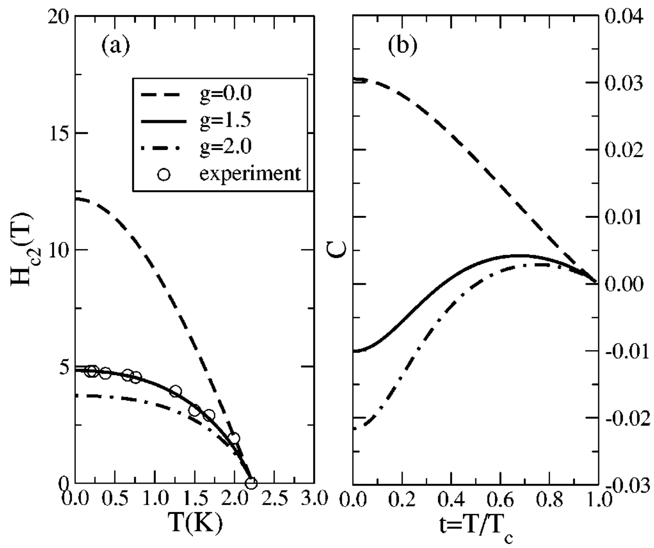

FIG. 1. Temperature dependence of (a) the upper critical field and (b) the admixture parameter C in a $d_{x^2-y^2}$-wave superconductor with g factors $g=0$, 1.5, and 2. The magnetic field is applied along the crystal c direction.

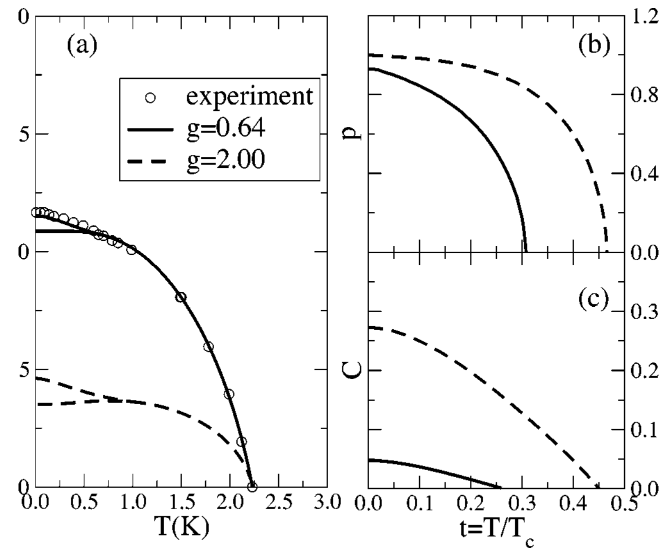

FIG. 2. Temperature dependence of (a) the upper critical field, (b) the $\vec{v}\cdot\vec{q}/(2H)=p\cos\phi$ term, and (c) the admixture parameter C in a $d_{x^2-y^2}$-wave superconductor with g factors $g=0.64$ (solid lines) and 2 (dashed lines). Here $t\equiv T/T_c$ is the reduced temperature. In (a)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

graphic c direction can be obtained from two coupled integral equations,[17]

$$-\ln t=\int_0^\infty \frac{du}{\sinh u}[1-\exp(-\rho u^2)\cos(hu)(1+2\rho^2u^4C)], \tag{1}$$

$$-C\ln t=\int_0^\infty \frac{du}{\sinh u}\left\{C-\exp(-\rho u^2)\cos(hu)\left[\frac{\rho^2u^4}{12}+C\left(1-8\rho u^2+12\rho^2u^4-\frac{16\rho^3u^6}{3}+\frac{2\rho^4u^8}{3}\right)\right]\right\}, \tag{2}$$

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

$$|\Psi\rangle=[1+C(a^\dagger)^4]|0\rangle, \tag{3}$$

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.[17,18]

In Fig. 1(a), experimental data of $H_{c2}(T)$ along the c axis in $CeCoIn_5$ are 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.[19–21] For temperatures above $T_0\approx0.7$ K, 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 $\partial\Delta l/\partial B$, as shown in Fig. 3 of Ref. 20. Below T_0 sharp jumps in Δl are observed upon crossing $H_{c2}(T)$, indicative of a first-order phase transition (cf. Fig. 3 in Ref. 21). The error bars of the so-derived $H_{c2}(T)$ are smaller than the size of the symbols in Figs. 1(a) and 2(a). The best fit with a numerical solution of Eqs. (1) and (2) is obtained with $v=3.2738\times10^8$ 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 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 $g\geqslant1.2$, 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 $\vec{H}\parallel\vec{c}$. A similar instability to a square vortex lattice was previously predicted for the high-T_c cuprates in the vicinity of H_{c2}.[17] Both SANS (Ref. 22) and STM (Ref. 23) on the vortex state of yttrium barium copper oxide (YBCO) single crystals indeed indicate significant deviations from a hexagonal towards a square vortex lattice in this compound.[24] Furthermore, the above theory was recently extended to lower magnetic fields.[18,25] The predicted square vortex lattice was observed by SANS in a single crystal of LSCO at $H=2$ T.[26]

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

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.[27] Again, the equations for the upper critical field can be derived from weak-coupling $d_{x^2-y^2}$-wave BCS theory. The differences of these results from the corresponding conventional *s*-wave superconductors are (i) the assumption of a quasi-2D Fermi cylindrical Fermi surface, and (ii) the admixture of higher Landau levels, as was first proposed by Luk'yanchuk and Mineev.[28] Here we have extended this formalism to include (i) the $d_{x^2-y^2}$-wave symmetry of the superconducting order parameter, (ii) Pauli paramagnetism, (iii) FFLO pairing, and (iv) the orbital effect via the ansatz of Gruenberg and Gunther.[29] The inclusion of the FFLO state leads to a new set of coupled integral equations,

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

$$-C\ln t=\int_0^\infty \frac{du}{\sinh u}\{C-\langle \exp(-\rho u^2|s|^2)\cos[h(1-p\cos\phi)u][1+\cos(4\phi)][\rho u^2 s^2+C(1-4\rho u^2|s|^2+2\rho^2u^4|s|^4)]\rangle\}, \tag{5}$$

where $s\equiv\sin\chi+i\sin\phi$, $\rho\equiv(vv_ceH)/(8\pi^2T^2)$, $p\cos\phi \equiv(\vec{v}\cdot\vec{q})/(2h)$, $\chi\equiv ck_z$, and $\langle\cdots\rangle$ is the angular average over ϕ and χ. Here $\sqrt{vv_c}=1.63\times10^8$ cm/s is used, and following Gruenberg and Gunther,[29] we chose $\vec{q}\parallel\vec{H}\parallel\vec{a}$. In this configuration, the vortex state is represented by

$$|\Psi\rangle=[1+C(a^\dagger)^2]|0\rangle, \tag{6}$$

where the vacuum $|0\rangle$ is again the Abrikosov state of a simple *s*-wave superconductor,[29] but mixing now occurs with the $N=2$ Landau level.

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 $p(t=0)=0.9$, i.e., the zero-temperature limit of the FFLO coefficient $p(t)$ is treated as a fit parameter. 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) the temperature dependence of $p(t)$ 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.[30]

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 $T\lesssim0.8$ K.[15,16,21,31] 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.[32–34]

Very recently, a second-order phase transition has been observed inside the vortex state for temperatures $T\leqslant0.3$ K in specific-heat[31,35] and ultrasound velocity measurements.[36] This has been suggested to indicate the transition from the vortex state into the FFLO state. At this transition, a possible change in the magnetostriction $\Delta L(B)/L$ does not exceed our noise level of 10^{-8}, and therefore is at least 50 times smaller than the change observed at H_{c2}.[19]

Moreover, the reported specific-heat jump at the zero-field superconducting transition, $\Delta C/\gamma T_c=4.5$, has to be considered with care, because the normal state specific-heat coefficient C/T (measured at $H=H_{c2}$) is not constant at 2.2 K, but strongly increases with decreasing temperature. According to Ref. 14, it reaches about 1 J/K^2 mol at 0.1 K. Taking this value as a lower bound for the normal state γ value (as justified also by the analysis of the entropy) results in a reduced jump height of only 1.3. This calls into question simple estimates of whether $CeCoIn_5$ is a strong coupling superconductor (see also Ref. 14).

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 $\vec{H}\parallel\vec{a}$ this observable is well approximated by[5]

$$\frac{N(E)}{N_0}-1=\frac{\Delta^2}{4\sqrt{\pi}}\sum_{\pm}\left\langle\int_{-\infty}^{\infty}du\frac{\exp(-u^2)\cos^2(2\phi)}{[E\pm\tilde{H}(1-p\cos\phi)-\epsilon|s|u]^2}\right\rangle, \tag{7}$$

where $\tilde{H}\equiv(\mu_B gH)/2$, $|s|=\sqrt{\sin^2(\phi)+\sin^2(\chi)}$, and $\epsilon\equiv\sqrt{vv_ceH}$. Again, a finite p indicates the presence of a FFLO state. This quasiparticle density of states as a function of energy is plotted in Fig. 3. For the parameters, we have chosen $p=0.9$ and $\epsilon/H=0.2$, appropriate for $CeCoIn_5$ in the low-temperature regime $T\lesssim0.1$ K. The magnetic field is fixed at a value close to $H_{c2}\approx12$ T. In the absence of the FFLO state [Fig. 3(a)], there are two sharp resonances close to $E=\pm H$, corresponding to the two poles in Eq. (7). In the presence of the FFLO state, more structure appears in the spectral response, as shown in Fig. 3(b), with resonances at $E=\pm H(1\pm p)$. These additional features arise due to the contribution of the $p\cos\phi$ term in the denominator of Eq.

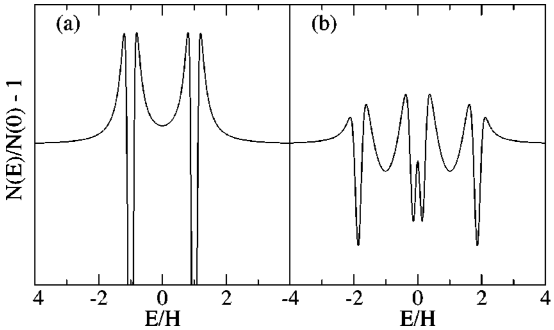

FIG. 3. Quasiparticle density of states of a $d_{x^2-y^2}$-wave superconductors in a magnetic field (a) in the absence of the FFLO state ($p=0$), and (b) in the FFLO state ($p=0.9$).

(7), and are therefore most pronounced for values of p close to unity, as it appears to be the case for $CeCoIn_5$ at low temperatures and high fields close to H_{c2}. Precision measurements of this quasiparticle density of states in an applied magnetic field can thus provide a clear signal for the presence of FFLO states and the symmetry of the underlying superconducting order parameter. It should therefore be of great interest to conduct a scanning tunneling microscope study of the quasiparticle density of states in $CeCoIn_5$ at $T < 0.7$ K in order to further scrutinize the proposed FFLO state.

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.

ACKNOWLEDGMENTS

We thank J.L. Sarrao for providing a high-quality $CeCoIn_5$ single crystal. Furthermore, we are grateful to Y. Matsuda, A. Ardavan, T. Ishiguro, R. Movshovich, M.-S. Nam, T. Roscilde, and J.L. Sarrao for useful discussions on the FFLO state. S.H. acknowledges financial support by the Petroleum Research Foundation and the National Science Foundation, Grant No. DMR-0089882.

[1] P. Fulde and R.A. Ferrell, Phys. Rev. **135**, A550 (1964).
[2] A.I. Larkin and Y.N. Ovchinnikov, Sov. Phys. JETP **20**, 762 (1965).
[3] K. Maki and H. Won, Czech. J. Phys. **46**, 1035 (1996).
[4] K. Yang and S.L. Sondhi, Phys. Rev. B **57**, 8566 (1998).
[5] K. Maki and H. Won, Physica B **322**, 315 (2002).
[6] H. Shimahara, J. Phys. Soc. Jpn. **67**, 736 (1998).
[7] M.A. Tanatar *et al.*, Phys. Rev. B **66**, 134503 (2002).
[8] L. Balicas *et al.*, Phys. Rev. Lett. **87**, 067002 (2002).
[9] H. Shimahara, J. Phys. Soc. Jpn. **71**, 1644 (2002); H. Shimahara, Physica B **329-333**, 1442 (2003).
[10] M. Houzet *et al.*, Phys. Rev. Lett. **88**, 227001 (2002).
[11] J. Singleton *et al.*, J. Phys.: Condens. Matter **12**, L641 (2000).
[12] K. Izawa *et al.*, Phys. Rev. Lett. **88**, 027002 (2002).
[13] H. Won and K. Maki, Physica B **312-313**, 44 (2002).
[14] C. Petrovic *et al.*, J. Phys.: Condens. Matter **13**, L337 (2001).
[15] K. Izawa *et al.*, Phys. Rev. Lett. **87**, 057002 (2001).
[16] T. Tayama *et al.*, J. Phys. Chem. Solids **63**, 1155 (2002).
[17] H. Won and K. Maki, Phys. Rev. B **53**, 5927 (1996).
[18] A.J. Berlinsky *et al.*, Phys. Rev. Lett. **75**, 2200 (1995).
[19] N. Oeschler, Ph.D. thesis (2003).
[20] N. Oeschler *et al.*, Phys. Rev. Lett. **91**, 076402 (2003).
[21] A. Bianchi *et al.*, Phys. Rev. Lett. **89**, 137002 (2002).
[22] B. Keimer *et al.*, Phys. Rev. Lett. **73**, 3459 (1994).
[23] I. Maggio-Aprile *et al.*, Phys. Rev. Lett. **75**, 2754 (1995).
[24] While the observed apex angle of the vortex lattice is not 90° but rather 73°–77°, this deviation can be attributed to the orthorhombic structural distortion in YBCO. Moreover, the spatial anisotropy of the coherence length $\xi_a/\xi_b=1.5$ is likely due to the presence of Cu-O chains in this compound. Precise STM measurements have revealed an elliptical shape of the vortices, consistent with this ratio of ξ_a/ξ_b, and (Ref. 23) the observed deviation of the apex angle from 90° is a likely consequence of this asymmetry.
[25] J. Shiraishi, M. Kohmoto, and K. Maki, Phys. Rev. B **59**, 4497 (1999).
[26] R. Gilardi *et al.*, Phys. Rev. Lett. **88**, 217003 (2002).
[27] H. Won and K. Maki, Europhys. Lett. **34**, 453 (1996).
[28] I.A. Luk'yanchuk and V.P. Mineev, Sov. Phys. JETP **66**, 1168 (1987).
[29] L.W. Gruenberg and L. Gunther, Phys. Rev. Lett. **16**, 996 (1966).
[30] Furthermore, for larger g the admixture coefficient with the $N=2$ is greatly enhanced.
[31] A. Bianchi *et al.*, Phys. Rev. Lett. **91**, 187004 (2003).
[32] K. Yamaji, J. Phys. Soc. Jpn. **52**, 1361 (1983).
[33] T. Ishiguro and K. Yamaji, *Organic Superconductors* (Springer Berlin, 1990).
[34] B. Dora and A. Virosztek, Eur. Phys. J. B **22**, 167 (2001).
[35] H.A. Radovan *et al.*, Nature (London) **425**, 51 (2003).
[36] T. Watanabe *et al.*, cond-mat/0312062 (unpublished).