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Y. Tokiwa, Philipp Gegenwart, D. Gnida, D. Kaczorowski

Angaben zur Veröffentlichung / Publication details:

Tokiwa, Y., Philipp Gegenwart, D. Gnida, and D. Kaczorowski. 2011. "Quantum criticality near the upper critical field of Ce2PdIn8." *Physical Review B* 84 (14): 140507(R).
https://doi.org/10.1103/physrevb.84.140507.

PHYSICAL REVIEW B **84**, 140507(R) (2011)

Quantum criticality near the upper critical field of Ce_2PdIn_8

Y. Tokiwa and P. Gegenwart
I. Physikalisches Institut, Georg-August-Universität Göttingen, D-37077 Göttingen, Germany

D. Gnida and D. Kaczorowski
Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 1410, PL-50-950 Wrocław, Poland

(Received 11 August 2011; revised manuscript received 6 October 2011; published 25 October 2011)

We report low-temperature specific-heat measurements in magnetic fields up to 12 T applied parallel and perpendicular to the tetragonal c axis of the heavy fermion superconductor Ce_2PdIn_8. In contrast to its quasi-two-dimensional (2D) relative $CeCoIn_5$, the system displays an almost isotropic upper critical field. While there is no indication for a high-field and low-temperature phase in Ce_2PdIn_8, the data suggest a smeared weak first-order superconducting transition close to $H_{c2} \approx 2$ T. The normal-state electronic specific-heat coefficient displays logarithmically divergent behavior, comparable to $CeCoIn_5$ and in agreement with 2D quantum criticality of spin-density-wave type.

DOI: 10.1103/PhysRevB.84.140507 PACS number(s): 74.70.Tx, 74.40.Kb, 74.20.Mn, 74.25.Bt

Unconventional superconductivity (SC) often occurs in systems with competing phases near a quantum critical point (QCP). Indicative for quantum criticality is, e.g., the observation of non-Fermi-liquid (NFL) behavior in the normal state at $T > T_c$ in zero field or down to the lowest accessible temperatures at fields above the upper critical magnetic field H_{c2}. Heavy fermion (HF) superconductors have sufficiently small H_{c2}, allowing for a detailed thermodynamic study of the normal-state properties down to very low temperatures to investigate NFL behavior. The layered tetragonal $CeCoIn_5$ with $T_c = 2$ K is a well-studied heavy fermion superconductor with a NFL normal state due to a field-induced QCP slightly below H_{c2}.[1–8] Its SC properties are also fascinating. The transition at $H_{c2}(T)$ becomes first order below 1 K due to strong Pauli limiting[9,10] and clear phase transition anomalies at the high-field and low-temperature (HFLT) corner in the SC phase diagram have been discovered,[11,12] which were thought to be related to a modulated SC Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state.[13,14] The origin of the HFLT SC state is under extensive debate, as subsequent NMR and neutron-diffraction experiments have revealed the existence of an antiferromagnetic (AF) ordering within the HFLT phase,[15–17] while a very recent NMR study suggests spatially distributed normal quasiparticle regions due to the formation of a FFLO state.[18]

The recently discovered HF superconductor ($T_c = 0.7$ K) Ce_2PdIn_8 (Refs. 19 and 20) belongs to the same class of Ce_nTIn_{3n+2} (T: transition metal, $n = 1$, 2, and ∞) systems,[21] containing a series of $CeIn_3$ and TIn_2 layers stacked along the c axis. While the $n = 1$ systems with $T =$ Co, $T =$ Ir (Ref. 22), and $T =$ Rh (Ref. 23) display anisotropic properties due to the layered, quasi-two-dimensional (2D) structure, cubic $CeIn_3$ ($n = \infty$) is a completely isotropic system. The bilayer ($n = 2$) systems are therefore expected to display intermediate behavior due to the more three-dimensional (3D) character of their crystal structure consisting of a stack of two $CeIn_3$ layers separated by one $PdIn_2$ layer. Ce_2PdIn_8 is an ambient-pressure HF superconductor of this class of materials. Its upper critical field H_{c2} is strongly Pauli limited with a large Maki parameter of 2.9.[24] Clean single crystals have become available, with a residual resistivity of $\sim$2.5 $\mu\Omega$ cm and a large mean free path of $l = 420$ Å,[24] exceeding the SC coherence length $\xi = 82$ Å. Thus, the system may fulfill the necessary conditions for the formation of the FFLO state, and it is interesting to investigate the high-field and low-temperature part of the SC phase by a thermodynamic probe. Furthermore, recent low-T electrical resistivity measurements at fields above the upper critical field have revealed strong similarities to $CeCoIn_5$, i.e., a quasilinear dependence at H_{c2} which turns into Fermi-liquid behavior only at larger fields,[24,25] suggesting a field-induced QCP near H_{c2}. Because of these common features, it is highly interesting to study the low-temperature specific heat of single-crystalline Ce_2PdIn_8 in magnetic fields applied both parallel and perpendicular to the tetragonal c axis.

Single crystals were grown by the self-flux method,[19] and we have confirmed by specific-heat and susceptibility measurements that they are not contaminated by $CeIn_3$. Very thin single-crystalline pieces were glued on the sample holder, and the basal plane orientation for the different pieces was not investigated. The specific heat at temperatures down to 70 mK and magnetic fields up to 12 T was measured in a dilution refrigerator with a SC magnet by employing the quasiadiabatic heat-pulse method.

Figure 1 shows the specific heat after subtraction of the nuclear contribution as $C_{el}(T)/T$, at magnetic fields up to H_{c2}. The nuclear contribution has been determined by fitting a $\alpha(H)T^{-3}$ contribution to C/T with $\alpha(H) \propto H^2$. At zero field, C_{el}/T increases upon cooling to $T_c = 0.68$ K, at which a typical mean-field-type anomaly is found. Within the SC state, it decreases linearly down to 0.2 K and saturates at a residual value of $C_{el}/T = 0.65$ J/mol K^2 (0.33 J/Ce mol K^2), which is much larger than the respective value of 0.04 J/mol K^2 found for $CeCoIn_5$,[26] indicating a larger quasiparticle density of states arising from pair breaking of defects. The specific heat at magnetic fields along different directions is rather similar and displays only a weak anisotropy in $H_{c2} = 2.0$ T and 2.2 T for $H \perp [001]$ and $H \parallel [001]$, respectively. At high fields, the SC transition changes its shape and becomes a rather symmetric peak different from the expectation for standard superconductors. This may indicate a smeared first-order transition. Note that in $CeCoIn_5$ a sharp and large peak of the

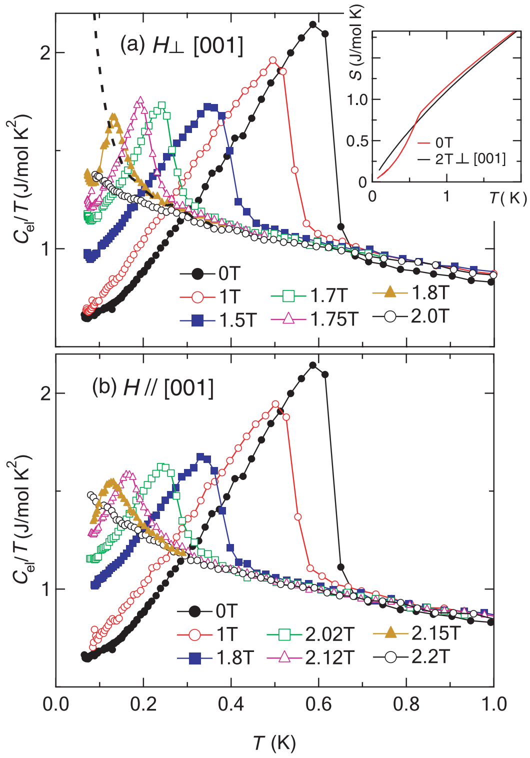

FIG. 1. (Color online) Electronic specific heat as $C_{el}(T)/T$ of Ce_2PdIn_8 at different magnetic fields perpendicular to (a) and along (b) the c axis, respectively. The dotted line displays the raw data at 2 T. The inset in (a) displays the entropy contributions, obtained from the integral of the electronic specific-heat coefficients at 0 and 2 T, extrapolated down to zero temperature by assuming a constant (for zero field) and logarithmic dependence for 2 T [cf. black solid line in Fig. 5(a)], respectively.

specific heat is found at low temperatures, which has proven to be due to a first-order transition.[9]

In Fig. 2, we compare the zero- and high-field SC transition anomalies of Ce_2PdIn_8 and $CeCoIn_5$ (Ref. 27) on a reduced temperature scale. At $H = 1.8$ T for the former and 11 T for the latter system, respectively, T_c is suppressed to 20% of its zero-field value. The peak at the SC to normal transition in

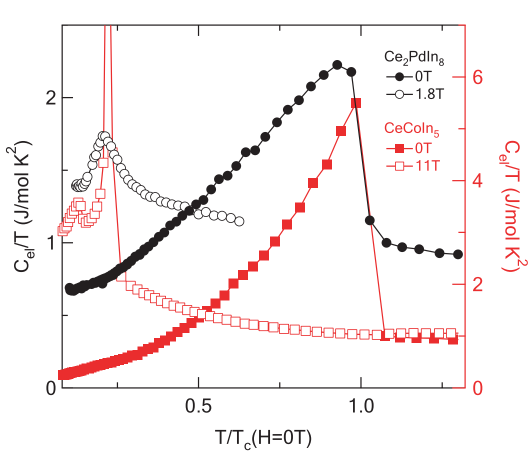

FIG. 2. (Color online) Comparison on $C_{el}(T)/T$ for Ce_2PdIn_8 (circles, $H \perp c$, left-hand axis) and $CeCoIn_5$ (squares, $H \parallel c$, right-hand axis) (Ref. 27), plotted against the reduced temperature.

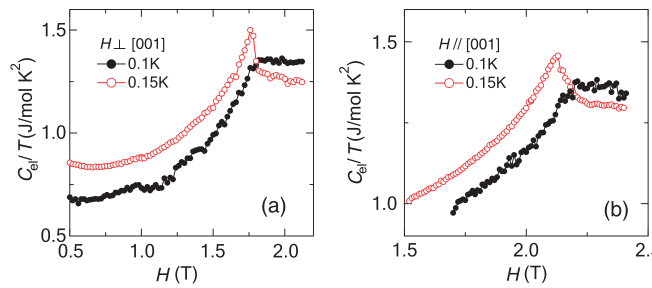

FIG. 3. (Color online) Electronic specific heat divided by temperature C_{el}/T for Ce_2PdIn_8 at 0.1 and 0.15 K as a function of magnetic field perpendicular to (a) and along (b) the tetragonal c axis, respectively.

the latter system is much sharper, indicating a much stronger first-order nature of SC transition in $CeCoIn_5$. The peak height for $CeCoIn_5$ has been reported to be very sensitive to impurity scattering. Only 0.22% of Cd doping suppresses significantly the sharp peak in specific heat and leads to a very similar C/T dependence as found for 1.8 T in Ce_2PdIn_8.[27] We may thus associate the reduced peak height for Ce_2PdIn_8 to the approximately ten-times-shorter electronic mean free path, compared to undoped $CeCoIn_5$ (4000 Å).[28] The HFLT phase in undoped $CeCoIn_5$ causes an additional jump in $C_{el}(T)/T$ of ~ 0.2 J/mol K^2 at 11 T, whereas in Ce_2PdIn_8 no anomaly could be detected within the measurement error of $\sim$0.02 J/mol K^2 at 0.1 K.

Measurements of the electronic specific-heat coefficient as a function of magnetic field, displayed in Fig. 3, also show no sign of an anomaly in addition to H_{c2}. According to the theoretical study by Ichioka and Machida,[29] the convex shape of residual C_{el}/T as a function of field, which is also observed in our experiments, is an indication of a strong Pauli-limiting effect.

Figure 4 shows the phase diagram of Ce_2PdIn_8 for two perpendicular magnetic field directions, revealing an almost isotropic field effect on superconductivity. The strongly convex shape of $H_{c2}(T)$ indicates a pronounced effect of Pauli limiting, dominating over orbital limiting. In $CeCoIn_5$, H_{c2} is also strongly Pauli limited and furthermore displays a pronounced anisotropy, which could be related to the anisotropic normal-state spin susceptibility. For Ce_2PdIn_8, by contrast,

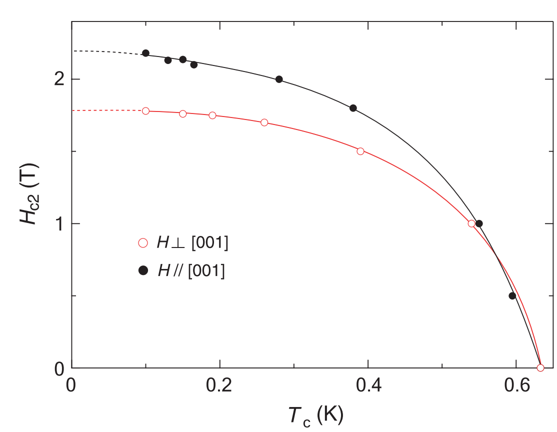

FIG. 4. (Color online) SC phase diagram of Ce_2PdIn_8 for two perpendicular field orientations. The solid lines are guides to the eyes.

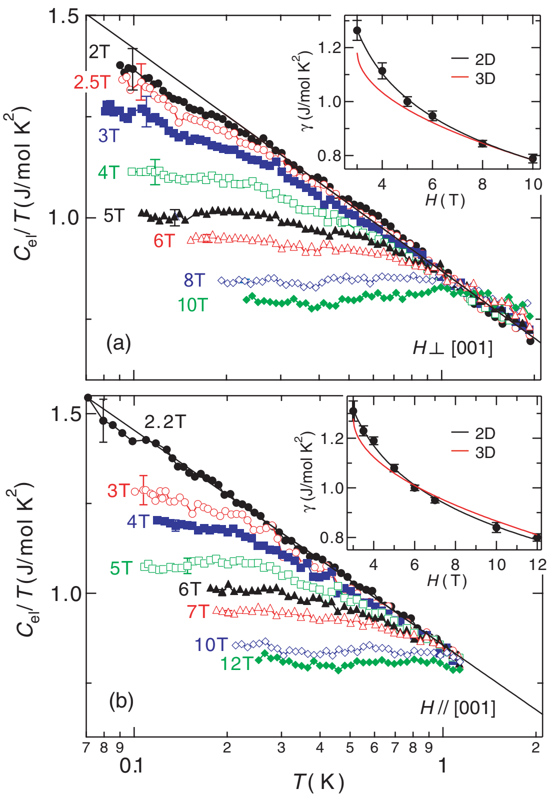

FIG. 5. (Color online) Electronic specific heat divided by temperature $C_{el}(T)/T$ (on a logarithmic scale) of Ce_2PdIn_8 at magnetic fields above H_{c2}, perpendicular to (a) and along (b) the tetragonal c axis, respectively. The solid lines represent a $\ln(1/T)$ dependence. The insets display the field dependence of the Sommerfeld coefficient (see text). The colored solid lines represent the fitted curves using theoretical functions for 2D and 3D AF QCPs (Ref. 31).

the upper critical field is almost isotropic, which is consistent with the much weaker anisotropy of the reported normal-state susceptibility.[30]

Next, we turn to the signatures of NFL behavior in the normal state. As shown in Fig. 5, for fields slightly above H_{c2}, a logarithmic divergence of the specific-heat coefficient is found over more than one decade in T. The entropy release in zero field at $T_c = 0.68$ K due to the formation of the SC state is almost fully balanced at $H = 2$ T [cf. inset of Fig. 1(a)], indicating that the same degrees of freedom are responsible both for the NFL behavior and the SC state. At $H > H_{c2}$ for both magnetic field directions, Fermi-liquid behavior is gradually recovered, as seen from the evolution toward a temperature-independent specific-heat coefficient C_{el}/T. In the insets of Fig. 5, we plot the magnetic field dependence of the Sommerfeld coefficient $\gamma(H)$ obtained from the saturated C_{el}/T values. Upon reducing the field from large values down to H_{c2}, divergent behavior is found in $\gamma(H)$. This indicates the existence of a field-induced QCP in close vicinity to the upper critical field of superconductivity, similar to $CeCoIn_5$.[2] In order to obtain information on the position of the QCP, as well as on the nature of the underlying quantum critical fluctuations, we have carefully compared the data with the predictions of the Hertz-Millis theory for an AF QCP in both 2D and 3D. The predicted field dependences are $\gamma_0 + \gamma_h \ln[1/(H - H_c)]$ (2D) and $\gamma_0 - \gamma_h\sqrt{H - H_c}$ (3D), respectively.[31] We have performed least-squares fitting of our data using γ_0, γ_h, and H_c as free parameters. The field dependence expected for 3D critical fluctuations appears to be too weak to explain the experimental γ values at sufficiently large fields. A much better description could be obtained over one decade in $h = H - H_c$, using the expectation for 2D critical behavior. The obtained quantum critical fields for the 2D description are $H_c = 2.0 \pm 0.2$ T and 1.7 ± 0.3 T for $H \perp$ and $\parallel$ to [001], respectively. These critical fields are very close to H_{c2}. Note that recent electrical resistivity measurements have also suggested a field-induced QCP very close to H_{c2} in this system.[24] Furthermore, at $H \approx H_{c2}$ both the linear temperature dependence of the electrical resistivity,[24] as well as the logarithmic temperature dependence of the specific-heat coefficient, are in agreement with the theoretical prediction for a 2D AF QCP.

In summary, we have studied the effect of magnetic fields on the SC and normal-state behavior in Ce_2PdIn_8 by measuring the specific heat at low temperatures and high magnetic fields. Even though the upper critical field $H_{c2}(T)$ is strongly Pauli limited, a phase transition into a HFLT phase as seen in the relative $CeCoIn_5$ has not been detected. It has been recently reported that an extremely small amount of impurities destroys (or smears out) this phase transition in $CeCoIn_5$. For 0.05% Cd-doped or 0.08% Sn-doped $CeCoIn_5$ it could not be detected by specific-heat measurements anymore.[27,32] It has also been shown theoretically that a very small amount of impurities smears out the FFLO phase.[33] Although Ce_2PdIn_8 is a clean limit superconductor, the electronic mean free path of the investigated single crystal is approximately ten times smaller compared to $CeCoIn_5$. This seems to be reflected in a much larger residual specific-heat coefficient C_{el}/T at zero field and reduced peak height of the SC transition in high fields. From a comparison of the respective specific-heat data, we find the investigated Ce_2PdIn_8 single crystals comparable to 0.22% Cd-doped $CeCoIn_5$.[27] Thus, a possible HFLT transition in Ce_2PdIn_8 already could be destroyed or smeared out by impurity scattering. Furthermore, we note that the upper critical field of the latter compound is almost isotropic, whereas it displays a pronounced anisotropy in $CeCoIn_5$. The observed NFL normal-state behavior in Ce_2PdIn_8 appears to be rather similar to that found in $CeCoIn_5$. We have observed a logarithmic divergence of C_{el}/T as a function of temperature at fields near H_{c2} and a logarithmic field dependence of the Sommerfeld coefficient, both of which are compatible with 2D AF fluctuations due to a QCP near H_{c2}, in close similarity to $CeCoIn_5$.

This work has been supported by the German Science Foundation through FOR 960 (quantum phase transitions).

[1] C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, and Z. Fisk, J. Phys. Condens. Matter **13**, L337 (2001).

[2] A. Bianchi, R. Movshovich, I. Vekhter, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **91**, 257001 (2003).

[3]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).

[4]J. Paglione, M. A. Tanatar, D. G. Hawthorn, F. Ronning, R. W. Hill, M. Sutherland, L. Taillefer, and C. Petrovic, Phys. Rev. Lett. **97**, 106606 (2006).

[5]S. Zaum, K. Grube, R. Schäfer, E. D. Bauer, J. D. Thompson, and H. v. Löhneysen, Phys. Rev. Lett. **106**, 087003 (2011).

[6]J. G. Donath, F. Steglich, E. D. Bauer, J. L. Sarrao, and P. Gegenwart, Phys. Rev. Lett. **100**, 136401 (2008).

[7]F. Ronning, C. Capan, E. D. Bauer, J. D. Thompson, J. L. Sarrao, and R. Movshovich, Phys. Rev. B **73**, 064519 (2006).

[8]S. Singh, C. Capan, M. Nicklas, M. Rams, A. Gladun, H. Lee, J. F. DiTusa, Z. Fisk, F. Steglich, and S. Wirth, Phys. Rev. Lett. **98**, 057001 (2007).

[9]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).

[10]T. Tayama, A. Harita, T. Sakakibara, Y. Haga, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. B **65**, 180504 (2002).

[11]A. Bianchi, R. Movshovich, C. Capan, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **91**, 187004 (2003).

[12]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).

[13]P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964).

[14]A. I. Larkin and Y. N. Ovchinnikov, J. Exp. Theor. Phys. **47**, 1136 (1964) [Sov. Phys. JETP **20**, 762 (1965)].

[15]M. Kenzelmann, T. Strässle, C. Niedermayer, M. Sigrist, B. Padmanabhan, M. Zolliker, A. D. Bianchi, R. Movshovich, E. D. Bauer, J. L. Sarrao *et al.*, Science **321**, 1652 (2008).

[16]B.-L. Young, R. R. Urbano, N. J. Curro, J. D. Thompson, J. L. Sarrao, A. B. Vorontsov, and M. J. Graf, Phys. Rev. Lett. **98**, 036402 (2007).

[17]G. Koutroulakis, M. D. Stewart, V. F. Mitrović, M. Horvatić, C. Berthier, G. Lapertot, and J. Flouquet, Phys. Rev. Lett. **104**, 087001 (2010).

[18]K. Kumagai, H. Shishido, T. Shibauchi, and Y. Matsuda, Phys. Rev. Lett. **106**, 137004 (2011).

[19]D. Kaczorowski, A. P. Pikul, D. Gnida, and V. H. Tran, Phys. Rev. Lett. **103**, 027003 (2009).

[20]D. Kaczorowski, D. Gnida, A. Pikul, and V. Tran, Solid State Commun. **150**, 411 (2010).

[21]J. Thompson, R. Movshovich, Z. Fisk, F. Bouquet, N. Curro, R. Fisher, P. Hammel, H. Hegger, M. Hundley, M. Jaime *et al.*, J. Magn. Magn. Mater. **226**, 5 (2001).

[22]C. Petrovic, R. Movshovich, M. Jaime, P. G. Pagliuso, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Europhys. Lett. **53**, 354 (2001).

[23]H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).

[24]J. K. Dong, H. Zhang, X. Qiu, B. Y. Pan, Y. F. Dai, T. Y. Guan, S. Y. Zhou, D. Gnida, D. Kaczorowski, and S. Y. Li, Phys. Rev. X **1**, 011010 (2011).

[25]V. H. Tran, D. Kaczorowski, R. T. Khan, and E. Bauer, Phys. Rev. B **83**, 064504 (2011).

[26]R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **86**, 5152 (2001).

[27]Y. Tokiwa, R. Movshovich, F. Ronning, E. D. Bauer, P. Papin, A. D. Bianchi, J. F. Rauscher, S. M. Kauzlarich, and Z. Fisk, Phys. Rev. Lett. **101**, 037001 (2008).

[28]G. Seyfarth, J. P. Brison, G. Knebel, D. Aoki, G. Lapertot, and J. Flouquet, Phys. Rev. Lett. **101**, 046401 (2008).

[29]M. Ichioka and K. Machida, Phys. Rev. B **76**, 064502 (2007).

[30]K. Uhlířova, J. Prokleška, V. Sechovský, and S. Daniš, Intermetallics **18**, 2025 (2010).

[31]L. Zhu, M. Garst, A. Rosch, and Q. Si, Phys. Rev. Lett. **91**, 066404 (2003).

[32]Y. Tokiwa, R. Movshovich, F. Ronning, E. D. Bauer, A. D. Bianchi, Z. Fisk, and J. D. Thompson, Phys. Rev. B **82**, 220502 (2010).

[33]R. Ikeda, Phys. Rev. B **81**, 060510 (2010).