Non-Fermi-liquid behavior in an undoped single crystal of $CeRh_2Ga$

G. F. Chen,* I. Sakamoto, and S. Ohara
Department of Electrical & Computer Engineering, Graduate School of Engineering, Nagoya Institute of Technology, Nagoya 466-8555, Japan

T. Takami, H. Ikuta, and U. Mizutani
Department of Applied Physics, Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan

(Received 23 June 2003; published 28 January 2004)

The measurements of specific heat C, electrical resistivity ρ, magnetic susceptibility χ, magnetization M, Hall coefficient R_H, and thermoelectric power S were performed on a ternary compound of $CeRh_2Ga$ which crystallizes in the triple-hexagonal Na_3As-type structure with the space group $P6_3cm$. $CeRh_2Ga$ remains in a paramagnetic state down to 1.2 K and exhibits a non-Fermi-liquid behavior. The specific heat divided by temperature C/T shows a $T^{-1+\lambda}$ power-law temperature dependence with $\lambda=0.15$ (from 7 K to the lowest measured temperature 1.2 K). $\chi_\perp$ ($H\perp c$) exhibits the same power-law behavior up to 50 K with $\lambda=0.21$, while $\chi_\parallel$ ($H\parallel c$) shows the power-law behavior in the restricted temperature range up to 20 K with $\lambda=0.52$. At $T=1.8$ K and $H<1$ T, M varies linearly with H; above 1 T, M could be described by $M=\text{const}+H^\lambda$ with $\lambda=0.59$ and 0.20 for $H\parallel c$ and $H\perp c$, respectively. The power-law dependence behavior is consistent with the Griffiths phase scenarios. ρ increases linearly with decreasing temperature below 4 K, also signaling non-Fermi-liquid behavior. R_H shows a $1/T$ dependence below 20 K. S shows a $\ln T$ dependence between 4 and 20 K, followed by a negative minimum at ~ 3 K. The low-temperature unconventional behaviors in R_H and S for $CeRh_2Ga$ may be related to the non-Fermi-liquid anomalies observed in the specific heat and magnetic susceptibility.

DOI: 10.1103/PhysRevB.69.014420 PACS number(s): 75.20.Hr, 71.10.Hf, 71.27.+a

I. INTRODUCTION

The low-temperature anomalies for $U_{0.2}Y_{0.8}Pd_3$ with $C/T\propto \ln T$, $\rho\propto\rho_0+aT^{1.1}$ and $\chi\propto A\text{-}BT^{1/2}$ were first reported by Seaman *et al.* in 1991.[1] These behaviors deviate obviously from Landau-Fermi-liquid model, which predicts that C/T and χ are constant and ρ follows ρ_0+AT^2 at low temperatures.[2] Recently, a number of $4f$ and $5f$ electron systems have been reported to show such non-Fermi-liquid (NFL) behavior (see Ref. 3, and the references therein) and several theoretical models were proposed to explain the origin of NFL behavior. For example, in $Y_{1-x}U_xPd_3$,[1] the NFL behavior was suggested to arise from the two-channel Kondo effect due to the presence of the electrical quadrupolar interaction. In $CeIn_3$,[4] the NFL behavior occurs due to the coupling of conduction electrons to critical spin fluctuation in the proximity the quantum critical point (QCP).[5] A recent developed Griffiths phase model proposed the existence of a Griffiths phase (magnetic cluster) in the vicinity of a QCP.[6] This model predicts that the specific heat divided temperature and magnetic susceptibility in the paramagnetic phase follow a power-law temperature dependence $C/T\propto\chi\propto T^{-1+\lambda}(\lambda<1)$.

Doping, pressure, and magnetic field usually induce the NFL behavior in f-electron systems. However, there are only several undoped NFL systems in which NFL behavior occurs under normal conditions, such as $CeNi_2Ge_2$,[7] $YbRh_2Si_2$,[8] and the recently discovered heavy-fermion superconductors $CeTIn_5$ ($T=$Co, Rh, and Ir),[9] etc. In this paper, we report the preliminary measurement results on the flux-grown single crystals of $CeRh_2Ga$. Our results indicate that $CeRh_2Ga$ crystallizes in the hexagonal Na_3As-type structure,[10,11] and exhibits the NFL behavior at low temperatures, which could be well explained by Griffiths phase model.[6] Compared to $CeRh_2Ga$, the isostructural compound $CeIr_2Ga$ (Ref. 12) behaves as a mixed-valent system with an electronic specific heat coefficient $\gamma=77$ mJ/mol K^2.

II. EXPERIMENTAL

Single crystals of $CeRh_2Ga$ were grown from a Ga flux.[11] The resulting crystals have a hexagonal morphology with a length of ~ 5 mm and a diameter of ~ 2 mm. The crystals were characterized by a powder x-ray diffraction method using Cu-$K\alpha$ radiation at room temperature. The dc electrical resistivity and Hall coefficient measurements were performed by standard four- and five-probe methods, respectively. The specific heat was measured by the relaxation method. The magnetic susceptibility and magnetization measurements were performed using a quantum design superconducting quantum interference device (SQUID) magnetometer. The thermoelectric power (TEP) was measured using a four-wire dynamic method. All measurements were performed on the well-polished samples except the powder x-ray diffraction.

III. RESULTS AND DISCUSSION

The powder x-ray diffraction pattern of as-grown single crystals could be completely indexed on the basis of the triple-hexagonal Na_3As-type structure (the space group $P6_3cm$) in agreement with the presence of $CeIr_2Ga$.[11,12] We have not observed any additional lines arising from impurity phases. The lattice parameters of $CeRh_2Ga$ are $a=7.687$

 ©2004 The American Physical Society

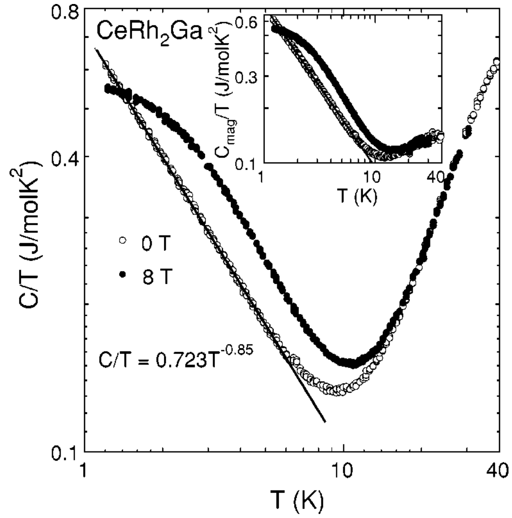

FIG. 1. Specific heat divided by temperature C/T vs temperature T plotted on log-log scales for single crystalline $CeRh_2Ga$ between 1.2 and 40 K in zero field and in 8 T ($H \| c$). The solid line indicates the power-law dependence of C/T for $CeRh_2Ga$ at low temperatures. Inset: Magnetic contribution to the specific heat in $CeRh_2Ga$.

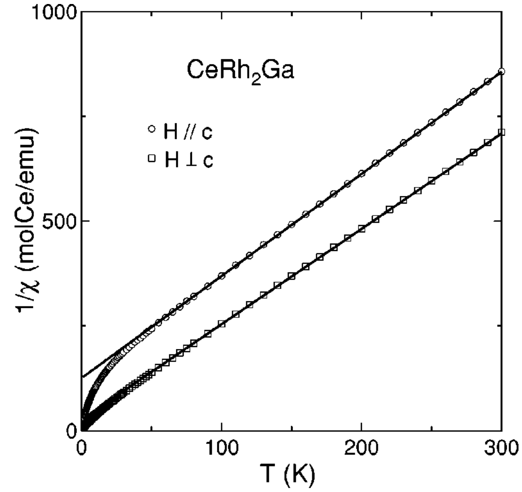

FIG. 2. Temperature dependence of inverse magnetic susceptibility for $CeRh_2Ga$ with magnetic field ($H = 0.1$ T) applied parallel or perpendicular to the c axis. The solid lines are the fits of χ^{-1} to the Curie-Weiss law with the parameters given in the text.

± 0.002 Å, $c = 9.548 \pm 0.003$ Å, respectively, obtained by a least-squares fit to the experimental data. The crystalline structure can be described as a layered quasi-2D structure,[11] in which Ce-Ga layers (with hexagonal symmetry) are separated by Rh layers stacked along the c axis. This compound offers an interesting example to study NFL phenomenon in systems having lower than cubic symmetry.

Figure 1 shows the specific heat C of single crystalline $CeRh_2Ga$ measured in zero field and in 8 T plotted as $\ln(C/T)$ vs $\ln T$. In zero applied magnetic field, C/T shows a minimum at around 10 K and reaches to 612 mJ/mol K^2 at the lowest measured temperature $T = 1.2$ K. Below 7 K, C/T obeys well-defined power-law behavior $C/T \propto T^{-1+\lambda}$, with $\lambda = 0.15$. The small value of λ indicates that the strong enhancement of specific heat with decreasing temperature. By subtracting the specific heat of nonmagnetic YRh_2Ga from that of $CeRh_2Ga$, we obtained the magnetic part of the specific heat C_{mag} as plotted in the upper inset of Fig. 1. We found that the zero field data does not change the power-law behavior at low temperatures. Such power-law dependence is a typical characteristic of NFL systems. We have tried a logarithmic temperature dependence fit to C/T. However, the logarithmic dependence behavior is restricted in a narrow temperature range. It is well known that an external magnetic field will affects the specific heat significantly at low temperatures for NFL systems. We measured the specific heat in an applied magnetic field $H = 8$ T ($H \| c$) and plotted also in Fig. 1. Compared to the zero field data, C/T shows a tendency to saturate at lower temperatures. This behavior indicates that the NFL state is suppressed by an applied magnetic field.

The temperature dependence of inverse magnetic susceptibility of $CeRh_2Ga$ between 1.8 and 300 K is shown in Fig. 2 measured with a magnetic field $H = 0.1$ T applied parallel ($\chi_{\|}$) or perpendicular ($\chi_{\perp}$) to its c axis. The magnetic susceptibility shows a large anisotropy at low temperatures. Above 50 K, the susceptibility follows the Curie-Weiss law $\chi(T) = N\mu_{\mathrm{eff}}^2/3\kappa_B(T - \Theta)$, and yields Weiss temperature $\Theta = -50$ K (-12 K) and effective moment $\mu_{\mathrm{eff}} = 1.81\mu_B$ ($1.87\mu_B$) for $H \| c$ ($H \perp c$), respectively. The values of μ_{eff} are reduced from expected $2.54\mu_B$ for the free Ce^{3+} ion. These values are close to those of $CeIr_2Ga$, however, which is considered as a weakly mixed-valent compound.[12]

The magnetic susceptibility between 1.8 and 50 K is shown in Fig. 3(a) as log-log plots of χ vs T. These low temperature data show a power-law dependence $\chi \propto T^{-1+\lambda}$. $\chi_{\perp}$ could be described as $\chi = 107.0T^{-0.79}$ up to 50 K; for $H \| c$, this power-law behavior occurs only below 20 K with $\chi = 23.8T^{-0.48}$. Fits of χ to a $\ln T$ behavior are unsuccessful. In addition, we found the data also follows a modified Curie-Weiss law $1/\chi = 1/\chi_0 + AT^{\alpha}$, with $\alpha = 0.38$ (0.75) for $H \| c$ ($H \perp c$) in the temperature range 1.8–50 K for both directions, as shown in Fig. 3(b). Such a dependence behavior suggests a local deviation from Fermi-liquid behavior as well.[3]

The power-law temperature dependence of C/T and magnetic susceptibility χ is a characteristic of NFL systems expected on Griffiths phase model.[6] De Andrade *et al.*[13] found power-law fits for the specific heat and magnetic susceptibility of $Th_{1-x}U_xPd_2Al_3$, $Y_{1-x}U_xPd_3$, and $UCu_{5-x}M_x$ ($M = $ Pd, Pt) to be successful. However, for $CeRh_2Ga$, the values of λ obtained from magnetic susceptibility are much larger than the one obtained from specific heat. A further prediction of Griffiths phase model[14] gives that magnetization M behaves a linear field dependence $M \propto H$ for $H < H^*$ (H^*, a crossover field) and a power-law dependence $M \propto H^{\lambda}$ for $H > H^*$, where λ should have the same value obtained from magnetic susceptibility. To test this prediction, we plotted the magnetization of $CeRh_2Ga$ measured at 1.8 K in applied magnetic field up to 5 T in Fig. 4. For $H < 1$ T, M shows a linear field dependence for both directions. Above 1 T, a variation M is well described by H^{λ}. The best fitting

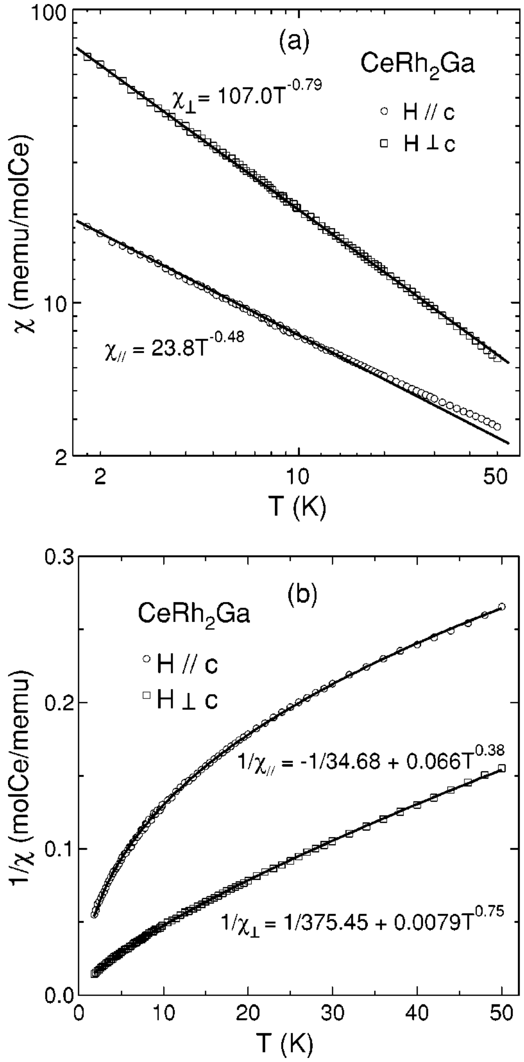

FIG. 3. (a) Log-log plots of magnetic susceptibility χ vs temperature T for $CeRh_2Ga$ with magnetic field applied parallel or perpendicular to the c axis between 1.8–50 K. The solid lines are the best fits of power law $\chi = aT^{-1+\lambda}$. (b) Inverse magnetic susceptibility $1/\chi$ vs temperature T for $CeRh_2Ga$, showing that $1/\chi$ can be fit to $1/\chi_0 + AT^{\alpha}$, in the temperature range 1.8–50 K for both directions.

gives $\lambda = 0.59$ for $H \| c$ or $\lambda = 0.20$ for $H \perp c$, respectively. The values of λ are comparable to those obtained from magnetic susceptibility data.

The electrical resistivity ρ measured with the current parallel to the c axis ($J \| c$) is illustrated in Fig. 5. The resistivity increases with decreasing temperature, similarly to the behaviors observed in UCu_4Pd and $U_{0.2}Y_{0.8}Pd_3$.[1,15] The upper inset of Fig. 5 shows the low-temperature data. The linear dependence of $\rho_{\| c}$ can be described as $\rho = \rho_0 - AT$ with $\rho_0 = 375\ \mu\Omega$ cm and $A = 8.5\ \mu\Omega$ cm/K, which are comparable to those of UCu_4Pd.[15] We also plotted the $\rho_{mag} + \rho_0$ vs $\ln T$ for $CeRh_2Ga$ in the lower inset of Fig. 5. Here $\rho_{mag} + \rho_0$ is taken to be the total resistivity minus that of YRh_2Ga. With decreasing temperature, the $\rho_{mag} + \rho_0$ shows a $-\ln T$ dependence down to 40 K, indicative of the single-impurity Kondo scattering. The ab-plane resistivity $\rho_{\perp c}$ exhibits a similar temperature dependence behavior as that of $\rho_{\| c}$, but the absolute values of $\rho_{\perp c}(T)$ could not be determined accurately owing to the small size in the present work.

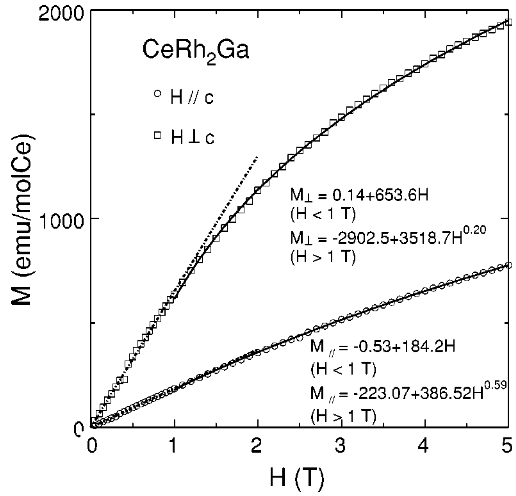

FIG. 4. The field dependence of magnetization of $CeRh_2Ga$ with magnetic field applied parallel or perpendicular to the c axis at 1.8 K. The dashed lines indicate the linear dependence for low field. The solid curves are the fits using the expression $M = \text{const} + H^{\lambda}$.

Figure 6 shows the Hall coefficient R_H vs T for $CeRh_2Ga$ between 3 and 300 K measured with $J \| c$ and $H \perp c$ ($H = 1.3$ T). R_H is positive over the measured temperature range. Above 100 K, it is nearly independent on temperature. With decreasing temperature, R_H decreases and exhibits a minimum at around 30 K and followed by a steep increase toward lower temperatures. In the impurity Kondo systems, the Hall coefficient can be divided into two terms: the normal Hall coefficient R_0 and the anomalous Hall coefficient R_a arising from skew scattering of conduction electrons.[16]

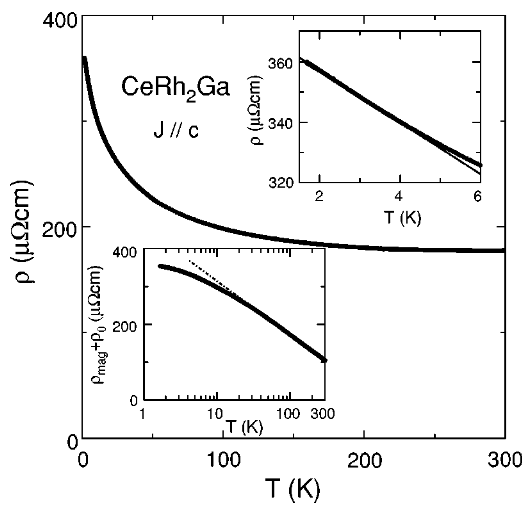

FIG. 5. Temperature dependence of electrical resistivity of $CeRh_2Ga$ with the current along the c axis between 1.6–300 K. Upper inset: The electrical resistivity on an expanded scale for the lowest temperatures. The solid line is a fit of $\rho = \rho_0 - AT$ with the parameters given in the text. Lower inset: Magnetic resistivity on a semilog plot. The dashed line is a guide for the eye.

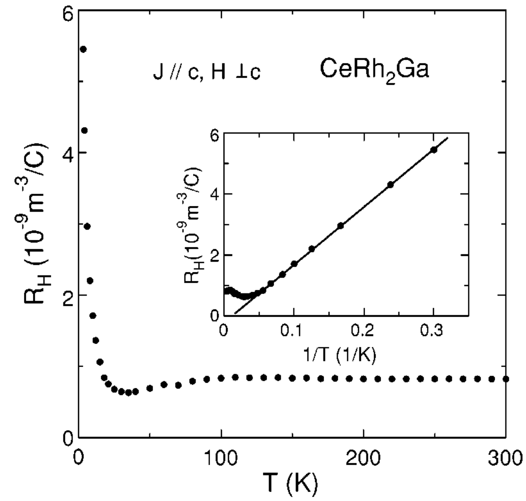

FIG. 6. Temperature dependence of Hall coefficient R_H for $CeRh_2Ga$ with current along the c axis and magnetic field applied perpendicular to the c axis. Inset: R_H plotted vs $1/T$. The solid line is a guide for the eye.

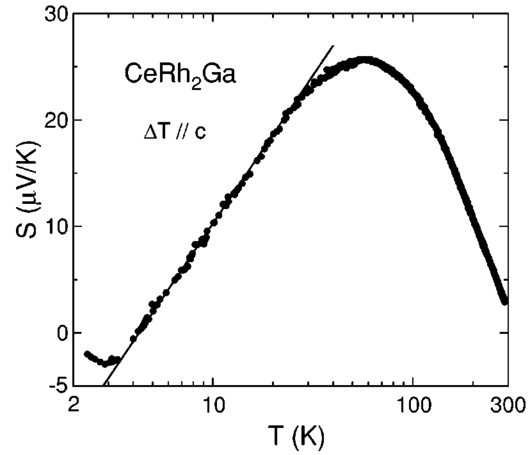

FIG. 7. Semilog plots of thermoelectric power S vs temperature T for $CeRh_2Ga$ with thermal gradient parallel to the c axis. The solid line is a guide for the eye.

However, for $CeRh_2Ga$, the temperature-independent behavior of R_H ($T>100$ K) indicates the skew scattering could be neglected. For low temperatures, we plotted the R_H vs $1/T$ and found a well $1/T$ dependence between 3 (the lowest measured temperature) and 20 K, as shown in the inset of Fig. 6. Note that the $1/T$ dependence of R_H was also observed in the NFL systems of heavy-fermion superconductor $CeCoIn_5$ (Ref. 17) and was attributed to the presence of antiferromagnetic spin fluctuations near the QCP in 2D systems. This Curie-Weiss-like behavior of R_H bears striking resemblance to the high-T_c cuprates in the normal state.[18]

The temperature dependence of thermoelectric power S (with the thermal gradient parallel to c axis) on semilog plots between 2 and 300 K is presented in Fig. 7. S increases with decreasing temperature and exhibits a broad positive maximum S_{max} ($\sim 26\ \mu$V/K) roughly at 60 K, which may be ascribed to the effect of the crystalline electric field (CEF) splitting.[19] At 4 K, S changes sign and then shows a negative minimum S_{min} ($\sim -3\ \mu$V/K) at about 3 K. This minimum is similar to those observed in many heavy fermion systems,[20,21] which were predicted theoretically by Fischer[22] and Kim *et al.*[23] Note that another prominent feature in the thermoelectric power of $CeRh_2Ga$ is the logarithmic temperature dependence between 4 and 20 K, as shown in Fig. 7 by the solid line. We suggested that the $\ln T$ dependence is associated with the unconventional behaviors observed in χ, C/T and R_H in the same temperature region. However, there are few available data to be compared on thermoelectric power in the typical NFL systems till now. For $U_{0.2}Y_{0.8}Pd_3$,[24] it also shows a broad maximum in S at high temperatures but no anomaly was observed at lower temperatures. Sato *et al.*[25] have observed a sharp minimum in S near 8 K for $Y_{1-x}U_xRu_2Si_2$. Thus, more experimental results on TEP of NFL systems are required to identify the open question about the behavior of TEP of NFL system.

In summary, we have grown a ternary compound of $CeRh_2Ga$ and measured the physical properties. $CeRh_2Ga$ exhibits NFL behavior at low temperatures. From the power-law dependence of $C/T \propto \chi \propto T^{-1+\lambda}$ and $M \propto H^{\lambda}$, we speculated that the NFL behavior observed in $CeRh_2Ga$ may be attributed to the existence of a Griffiths phase in the vicinity of a QCP, although the values of λ obtained from magnetic susceptibility are not consistent with the one obtained from specific heat. The observed low-temperature unconventional behaviors in thermoelectric power and Hall coefficient may be related to NFL anomalies. To further clarify the origin of the NFL behaviors, more detailed studies of specific heat, magnetic susceptibility, and electrical resistivity at lower temperatures are currently in progress.

ACKNOWLEDGMENTS

The authors would like to thank Dr. Biwa and Mr. Kamatani for their assistance in the specific heat measurements.

*Electronic address: r04elc07@edsys.center.nitech.ac.jp

[1] C. L. Seaman, M. B. Maple, B. W. Lee, S. Ghamaty, M. S. Torikachvili, J. S. Kang, L. Z. Liu, J. W. Allen, and D. L. Cox, Phys. Rev. Lett. **67**, 2882 (1991).
[2] L. D. Landau, Sov. Phys. JETP **3**, 920 (1957).
[3] G. R. Stewart, Rev. Mod. Phys. **73**, 797 (2001).
[4] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. W. Haselwimmer, and G. G. Lonzarich, Nature (London) **394**, 39 (1998).
[5] A. J. Millis, Phys. Rev. B **48**, 7183 (1991).
[6] A. H. Castro Neto, G. Castilla, and B. A. Jones, Phys. Rev. Lett. **81**, 3531 (1998).
[7] S. R. Julian, C. Pfleiderer, F. M. Grosche, N. D. Mathur, G. J. McMullan, A. J. Diver, I. R. Walker, and G. G. Lonzarich, J. Phys.: Condens. Matter **8**, 9675 (1996).
[8] O. Trovarelli, C. Geibel, S. Mederle, C. Langhammer, F. M. Grosche, P. Gegenwart, M. Lang, S. Sparn, and F. Steglich, Phys. Rev. Lett. **85**, 626 (2000).
[9] J. S. Kim, J. Alwood, G. R. Stewart, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B **64**, 134524 (2001).

[10] P. Hafner, K. J. Range, J. Alloys Compd. **216**, 7 (1994); K. J. Range, R. Ehrl, and P. Hafner, *ibid.* **240**, 19 (1996).

[11] C. Petrovic, M. F. Hundley, R. Movshovich, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and Z. Fisk, J. Alloys Compd. **325**, 1 (2001).

[12] C. Petrovic, M. F. Hundley, R. Movshovich, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, Z. Fisk, A. Garcia, E. Granado, I. Torriani, and C. Rettori, J. Magn. Magn. Mater. **225**, 317 (2001).

[13] M. C. de Andrade, R. Chau, R. P. Dickey, N. R. Dilley, E. J. Freeman, D. A. Gajewski, M. P. Maple, R. Movshovich, A. H. Castro Neto, G. Castilla, and B. A. Jones, Phys. Rev. Lett. **81**, 5620 (1998).

[14] A. H. Castro Neto and B. A. Jones, Phys. Rev. B **62**, 14 975 (2000).

[15] B. Andraka and G. R. Stewart, Phys. Rev. B **47**, 3208 (1993).

[16] A. Fert and P. M. Levy, Phys. Rev. B **36**, 1907 (1987).

[17] Y. Nakajima, K. Izawa, Y. Matsuda, S. Uji, T. Terashima, H. Shishido, R. Settai, Y. Onuki, and H. Kontani, cond-mat/0305203 (unpublished).

[18] H. Kontani, K. Kanki, and K. Ueda, Phys. Rev. B **59**, 14 723 (1999).

[19] A. K. Bhattacharjee and B. Coqblin, Phys. Rev. B **13**, 3441 (1976).

[20] N. B. Brandt and V. V. Moshchalkov, Adv. Phys. **33**, 373 (1984).

[21] F. Steglich, C. D. Bredl, W. Lieke, U. Rachschwalker, and G. Sparn, Physica B **126**, 82 (1984).

[22] K. H. Fischer, Z. Phys. B: Condens. Matter **76**, 315 (1989).

[23] T. S. Kim and D. L. Cox, Phys. Rev. Lett. **75**, 1622 (1995).

[24] J. McCarten, S. E. Brown, C. L. Seaman, and M. P. Maple, Phys. Rev. B **49**, 6400 (1994).

[25] H. Sato, J. Urakawa, Y. Aoki, T. Namiki, H. Sugawara, Y. Onuki, H. Amitsuka, and T. Sakakibara, Physica B **259–261**, 390 (1999).