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Pseudogap and Superconducting Energy Gap in Single Crystals of URu_2Si_2 by Point Contact Spectroscopy

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Abstract We performed point contact spectroscopy measurements in the superconducting and normal state of a single crystal of the heavy fermion URu_2Si_2. The differential resistance as a function of bias voltage shows the features of the superconducting energy gap up to 1.37 K, above T_C we observe another feature that we identify as a pseudogap that persists up to 2 K. The superconducting gap does not fit a BCS behavior.

Keywords Superconductivity · Heavy fermions · Point contact spectroscopy

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Electronic correlations play an important role in the origin of the physical characteristics of superconducting materials. These effects can be readily observed, in different correlated materials; classical examples are superconductivity and magnetism. In the normal state of high temperature superconductors, it has been observed an extra feature; the so called pseudogap (PG), which is seen as a depletion on the electronic density of states at the Fermi level. This feature has been observed also in other superconducting materials. In heavy fermion superconductors also is presented the pseudogap, but the observation has not been so clear as in high temperature cuprates. The pseudogap feature has been observed using different experimental techniques, this characteristic is mainly shown in underdoped materials (see [1] and references therein). It has been found that the PG in general shows the onset temperature T^* well above T_C, and it is expected that this onset will be independent of the superconducting critical temperature [2]. Among the proposed models to explain the origin of

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the PG two important scenarios may be considered: The first one is based on the idea of uncorrelated electronic pairing that begins at T^*. This state changes to the superconducting state when the uncorrelated pairs get a global phase coherence at T_C. The second scenario, considers that the pseudogap appears as a consequence of magnetic instabilities, independent of the superconducting state, and it is believed that both phenomena are present below T_C (see [1, 3] and references therein). However, up to now there is no consensus about the origin of the pseudogap in the high T_C superconductors. For the case of heavy fermions superconductors, the strong electronic correlations arise at the coherence temperature, T_s, which is between 10 K and 100 K [4]. Below T_s two types of magnetic interactions between the d and f electronic populations compete. The two processes are the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, and the Kondo process. If the RKKY mechanism is dominant, then the system will be ordered magnetically, and at low temperatures this will coexist with the superconducting state. If this is not the case, the system evolves to a Kondo lattice [5]. The pseudogap feature has been observed in heavy fermion compounds by optical methods [6, 7], and the possibility that this feature could be a signature of heavy Fermions has been suggested [7].

URu_2Si_2 is a heavy fermion compound that shows superconductivity and magnetism. The magnetic ordering is explained by the formation of a spin density wave (SDW) [8–10], with a very small magnetic moment (0.02–0.04 μ_B) along the tetragonal **c** axis in the U sites. The SDW occurs at $T_N = 17.5$ K. The Fermi surface shows an imperfect nesting, which in turn produces an energy gap only in some small regions around the Fermi surface. At lower temperatures the system transits to the superconducting state, with a critical temperature of about $T_C \simeq 1.5$ K. The superconducting energy gap compete and opens on regions of the Fermi surface not used by the SDW. At temperatures below T_C the SDW and the superconductivity coexist. Moreover, the presence of a hidden order [11] suggests that in this compound may exist a pseudogap and a quantum critical point in its phase diagram, as in electron(hole)-doped high temperature superconductors [12].

Actually there are many different interpretations about the hidden order that emerges at a temperature of 17.5 K in URu_2Si_2. It has been proposed that may be related to different mechanisms, such as; unconventional spin density waves [8–10], orbital magnetism [13], magnetic helicity [14], octupolar ordering [15], etc. However until today still this hidden order remains as an open issue. What is now clear, is that in URu_2Si_2 the physical processes involving the hidden order and/or the magnetic behavior may be related to the coexistence of the two electronic subsystems that produce the complex behavior: One is related to localized $5f$ electrons, other to the itinerant $5f$ electrons. The localized $5f$ electrons sites are the responsible for the magnetic ordering, whereas the itinerant or delocalized $5f$ electrons determine the heavy fermion characteristics. The magnetic behavior in this compound, also was found that it is associated to two processes: One is the observed small magnetic moment of the order about 0.02 μ_B per U atom, that is now starting to be related with quality of the sample [16, 17], whereas the other, perhaps the most important, start to be related to the hidden order, this has a magnetic moment, with value about 0.3–0.4 μ_B/U when URu_2Si_2 is under pressure [18]. The antiferromagnetic ordering in this system also is presented by two different physics processes or effects: the anomalous nesting of the

Fermi surface, that may be related to spin density waves, with effects readily observed and correlated with the reduction of the electronic density of states, as seen in Hall measurements and in tunneling spectroscopy [11, 19, 20]. We call unconventional or anomalous spin density wave because the absence to identify an appropriated nesting vector. However in early tunneling experiments [11, 19, 20] we could detect an energy gap at the emergence of the hidden order that we correlated with a spin density wave. So, it is clear that the two magnetic effects observed in this compound are the weak antiferromagnetic order close to the hidden order, that workers [21] are related to an octupolar ordering, and/or to an unconventional spin density wave. Lastly it is clear that more work is necessary in order to better understand this complex heavy fermion system.

Among the most direct experimental techniques to study the superconducting state, particularly the energy gap and the pseudogap, electron tunneling and point contact spectroscopy (PC) are the most used tools. With the two mentioned techniques it is possible to study the electronic density of states, the phonon density of states, the temperature dependence of the energy gap, and also processes related to the scattering of qusiparticles injected into the superconducting state. In fact, it is also possible to observe features on the Fermi surface, that may be related to the superconducting state and to the normal state, as the mentioned pseudogap. Features related also to the nesting of the Fermi surface as SDW features clearly can be observed [22]. In the past PC has been used to study the energy gaps of URu_2Si_2 in both the antiferromagnetic and superconducting states [19, 20, 23–32].

In this work we report the study of the temperature dependency of the energy gap as determined by measurements using point contact spectroscopy in single crystals of URu_2Si_2. Our studies show that the superconducting energy gap does not follow the temperature dependence of the BCS model. In addition, measurements above T_C show that a feature persists. This behavior resembles the pseudogap feature observed in underdoped high T_C superconducting cuprates.

The point contact experiments were performed with single crystals growth by Czochralski method and annealed for one week at 850 °C, this technique is similar to that reported elsewhere [33]. The crystals have a platelet-like shape with the crystallographic **c** axis perpendicular to the plane of the platelet, typical dimension of the used crystals were approximately of $2 \times 3 \times 0.5$ mm^3.

The superconducting critical temperature was determined by resistance measurements as function of the temperature $R(T)$, using the ac four probe method. The applied current was 100 μA. The critical temperature of the crystal used for the PC measurements was determined to be $T_C(R = 0) = 1.37$ K. The transition temperature width was $\Delta T = 0.15$ K, and the ratio $R(300\ \text{K})/R(2\ \text{K}) = 38$ indicating a high degree of perfection of the crystal.

Point contacts were prepared at room temperature in the following manner: the single crystal was glued to a glass substrate using Oxford varnish. The point contacts were formed with the single crystal and a 5 μm tungsten wire plated with gold (Au(W)). The area of the contact junctions were estimated to be about 1 μm^2. In the reported experiments the point contacts were made over an edge of the crystal in such a manner that the electronic current is moving along the **a**–**b** crystallographic plane.

The differential resistance $\mathrm{d}V/\mathrm{d}I$ as function of the bias voltage V of the point contacts were measured between 0.325 K and 2.1 K, in a ^{3}He refrigerator using a

standard ac lock-in technique. The applied bias voltages were between ±2.5 mV, bias voltages in excess of ±2.5 mV produce heating in the contact region. The zero bias resistance of the different point contact junctions was between 0.3 Ω and 6 Ω. An important aspect when performing point contact studies, is the determination of the working regime. In order to determine this regime we estimated the radii a of the contacts using the Wexler's interpolation formula [34], in which we substituted the mean free electronic path $l \simeq 100$ Å [25, 26], the resistivity $\rho \sim 40$ μΩ-cm measured at 2 K [35] and the resistance of the point contacts measured at zero bias. The obtained values were between 320 Å and 3700 Å. These values indicate that the studied point contacts were in the diffuse regime where $a \simeq l$ [36]. (This is distinct to the ballistic regime where $a \ll l$ or the thermal regime where $a \gg l$.) An important experimental aspect considered in our study was the thermal stability of the contact junction. About this consideration, it is important to mention that our point contacts with resistances lower than 2 Ω were very stable respect to temperature changes. Contacts with higher resistances, are instable and changed its point contact resistance when cycled with temperature. In this work we report studies performed in more than 20 different contact junctions, all showed similar $\mathrm{d}V/\mathrm{d}I\,(V)$ curves.

In Fig. 1 we show the differential resistance curves of the junction contact, $\mathrm{d}V/\mathrm{d}I$ *vs* V measured at different temperatures from 325 mK to 2.1 K. The main features in these curves is the dramatic depression around zero bias, this is the typical feature

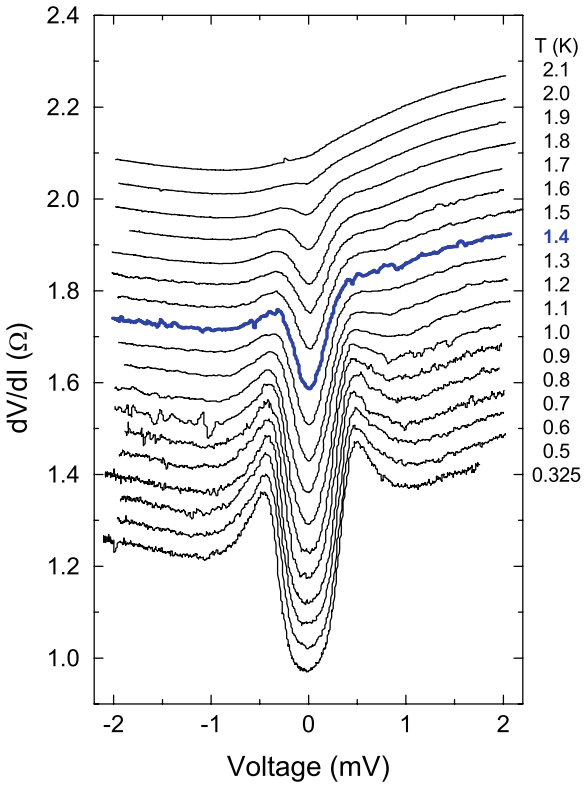

Fig. 1 (Color online) The differential resistance *vs* bias voltage at different temperatures for a URu_2Si_2-Au(W) contact junction. Note the clear presence of the superconducting energy gap, and the features above $T_C = 1.37$ K identified as the pseudogap. The thick line correspond to the transition temperature as determined by $R(T)$ measurements. For clarity, the curves have been vertically displaced

associated with the superconducting energy gap as observed in a superconductor-normal metal point contact. This feature is well defined at lower temperatures, but decreases and is smeared when the temperature is increased. However an extra feature in the differential resistance is observed; above T_C, the gap feature still can be seen. In Fig. 1 the curve measured at about 1.37 K is plotted with a thick line to separate the superconducting region of the normal state, according with the superconducting transition temperature as determined by $R(T)$ measurements. In addition, we observed clearly above the thick curve an extra feature, that we identify as a pseudogap. The curves show a marked asymmetry respect to zero bias; but this is a normal characteristic of heterojunctions [28, 29].

In order to determine the size of the superconducting gap, the Blonder et al. model (BTK) [22] is often used to fit the experimental data when the junctions are in the ballistic regime. The experimental data can not be well fitted to BTK model, our point contacts are in the diffusive regime. In this regime the magnitude of the spectra is reduced and broaden [36].

The width of the superconducting energy gap was determined from the separation of peaks observed in the $\mathrm{d}^2V/\mathrm{d}I^2$ characteristics as function of bias voltage. The superconducting energy gap value at 325 mK is $2\Delta = (580 \pm 7)$ μeV in agreement with values reported by other authors [23–26], but this is different to the BCS value, the ratio $2\Delta/k_BT_C$ is 4.9, indicative of a strong coupling limit. In Fig. 2 we show the symmetric part of the normalized differential resistance data respect to the value determined at 1.7 mVolts. The symmetric part is obtained adding the positive and negative parts of the $\mathrm{d}V/\mathrm{d}I\,(V)$ and dividing over 2. The size of the pseudogap feature at 1.8 K is (285 ± 25) μeV, note that this temperature is above T_C.

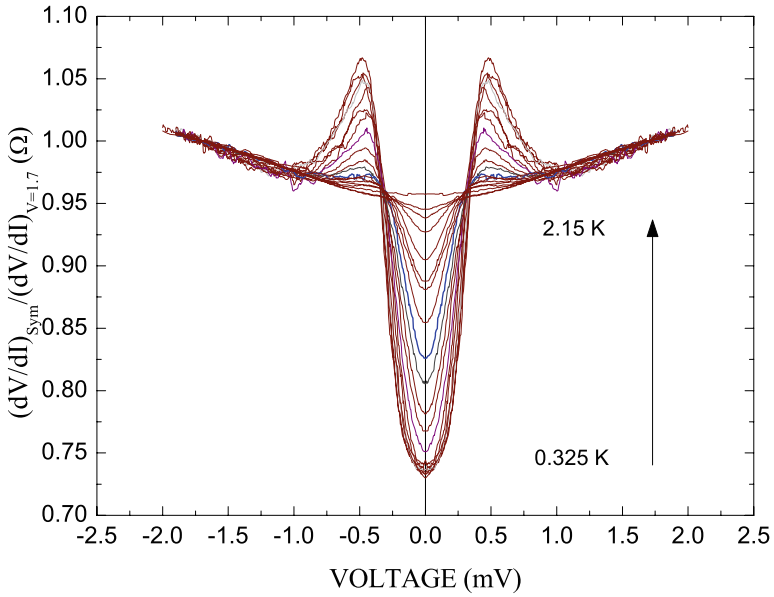

Fig. 2 (Color online) The symmetric contribution of the differential resistance as a function of temperature, normalized with $\mathrm{d}V/\mathrm{d}I$ ($V = 1.7$ mV)

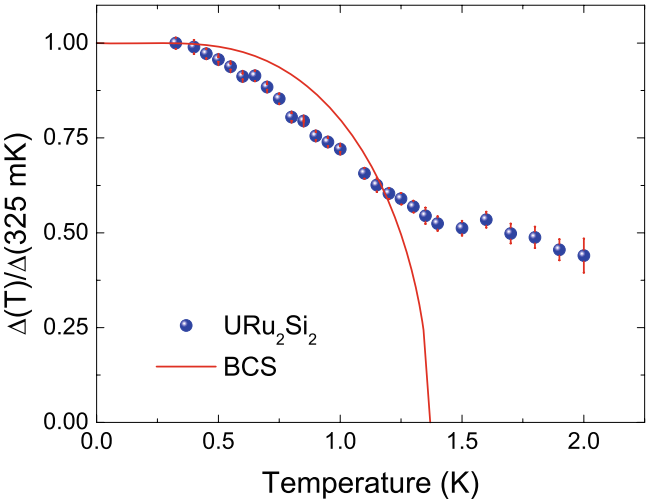

Fig. 3 (Color online) Temperature dependence of superconducting energy gap normalized to the gap at $T = 325$ mK. Above T_C the pseudogap feature is present. The continuous line is the energy gap BCS prediction

In order to see the temperature behavior of the superconducting gap, we plotted in Fig. 3 the energy gap normalized to the value of 2Δ obtained from the curve measured at 325 mK, as a function of T. The continuous line represents the energy gap temperature dependence predicted by the BCS model. As clearly seen the superconducting energy gap does not follow the behavior predicted by BCS, this decreases in different form. The decreasing manner of the gap looks quite linear with increasing temperature. Above T_C another feature persists, clearly notable. This structure decreases with the temperature and closes as the temperature rises, at about 2 K the feature is faint but still exists. Furthermore, the width of this feature is almost temperature independent, this resembles the pseudogap observed by tunneling spectroscopy in high temperature bismuth based superconductors [37, 38]. Taking into account the fact that the high T_C superconductors and the heavy Fermions present strong electronic correlations, we may consider that this feature may be characterized as a pseudogap.

An important aspect related with these point contact experiments is the effects caused by the tip of the contact into the superconducting material. The effect of local pressure clearly could modified the superconducting and normal behavior, as was observed by other workers. Rodrigo et al. [32] have reported point contact measurements in the antiferromagnetic state of URu_2Si_2, and they found that the Neel temperature, T_N, obtained from the PC experiments is 22 K instead of 17.5 K. They explain that the reason of this change was the pressure exerted by the tip on the specimen. They estimated a point contact radius of 5 nm and that the applied pressure about 1 GPa. Similarly Steglich et al. [39] have studied $CeCu_2Ge_2$ using point contacts formed by a tungsten tip, pressed onto the $CeCu_2Ge_2$. This compound is not a superconductor under normal pressure conditions, but it changes to the superconducting state with $T_C = 0.68$ K at pressures higher than 70 kbar. The experiments performed by Steglich and co-workers showed superconductivity in the $CeCu_2Ge_2$,

and they concluded that the local pressure produced by the tip on the sample was higher than 70 kbar.

Concerning the pressure effects in URu_2Si_2 it has been observed that this modify the superconducting and the SDW transition temperatures [40–43]. Measurements performed on single crystals have shown that T_C is increased when pressure is applied in the **c** direction of the crystal, whereas if the pressure is applied parallel to the **a** axis T_C decreases with a ratio of 35 mK/kbar. However T_N changes inversely to the T_C behavior [43]. High pressure studies reported by Amitsuka et al. [18] on URu_2Si_2 have shown the changes in the magnetic characteristics under pressure. They performed neutron elastic scattering experiments, at 1.4 K, that show that the magnetic moment (μ_0) in the (100) direction shows a notable increment under pressure. The magnetic moment changes almost linearly with pressure at a rate of about 0.25 μ_B/GPa between 0 and 1.3 GPa, reaching a value of 0.17 μ_B. At 1.5 GPa the magnetic moment increases to 0.40 μ_B. This value is 10 times higher than the value of 0.04 μ_B obtained under normal conditions. Even with this modification in the magnetic moment, T_N changes from 17.5 K to about 22 K. These authors suggested that there is a phase transition at $P_C = 1.5$ GPa that can be understood on the base of magnetic instabilities. In our PC experiments, the wire used as the electrode of the point contact applies a pressure on the sample parallel to the **a**–**b** plane, in agreement to the high pressure measurements the expected behavior is that T_C should decrease, however the PC curves show additional structure above T_C. According to this behavior the additional structure is not related to the superconducting energy gap, but to a pseudogap feature. The pressure exerted by the wire in our point contacts modify the electronic density of states around the Fermi level, showing a pseudogap in the normal state that resemble the pseudogap in the high T_C materials. According to the above discussion, in relation to our point contact experiments, it is clear that the superconducting and magnetic properties of URu_2Si_2 are very sensitive to the application of external pressure.

In summary, we have performed point contact spectroscopy measurements in the heavy fermion superconductor URu_2Si_2. Our results show the energy gap and the existence of a pseudogap feature. The width of the pseudogap is almost temperature independent and disappears at about 2 K. The superconducting energy gap has a linear dependence with temperature and does not follow BCS behavior.

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References

1. T. Timusk, B. Statt, Rep. Prog. Phys. **62**, 61 (1999)
2. D. Pines, Physica C **282–287**, 273 (1997)
3. V.M. Loktev, R.M. Quick, S.G. Sharapov, Phys. Rep. **349**, 1 (2001)
4. L. Degiorgi, Rev. Mod. Phys. **71**, 687 (1999)
5. G.J. Nieuwenhuys, in *Hand Book of Magnetic Materials*, vol. 9, ed. by K.H.J. Buschow. Elsevier (1995)
6. S. Donovan, A. Schwartz, G. Grüner, Phys. Rev. Lett. **79**, 1401 (1997)
7. M. Dressel, B. Gorshunov, N. Kasper, B. Nebendahl, M. Huth, H. Adrian, J. Phys.: Condens. Matter **12**, L633 (2000)

8. M.B. Maple, J.W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, M.S. Torikachvili, M.W. McElfresh, J.D. Thompson, Phys. Rev. Lett. **56**, 185 (1986)
9. H. Ikeda, Y. Ohashi, Phys. Rev. Lett. **81**, 3723 (1998)
10. V.P. Mineev, M.E. Zhitomirsky, Phys. Rev. B **72**, 014432 (2005)
11. Y.S. Oh, K.H. Kim, P.A. Sharma, N. Harrison, H. Amitsuka, J.A. Mydosh, Phys. Rev. Lett. **98**, 016401 (2007)
12. L. Alff, Y. Krockenberger, B. Welter, M. Schonecke, R. Gross, D. Manske, M. Naito, Nature **422**, 698 (2003)
13. P. Chandra, P. Coleman, J.A. Mydosh, V. Tripathi, Nature (London) **417**, 831 (2002)
14. C.M. Varma, L. Zhu, Phys. Rev. Lett. **96**, 036405 (2006)
15. K. Hanzawa, J. Phys.: Condens. Matter **19**, 072202 (2007)
16. J. Levallois, K. Behnia, J. Flouquet, P. Lejay, C. Proust, arxiv: 0809.4715vl
17. A. Villaume, F. Bourdarot, E. Hassinger, S. Raymmond, V. Tauford, D. Aoki, J. Flouquet, Phys. Rev. B **78**, 012504 (2008)
18. H. Amitsuka, M. Sato, N. Metoki, M. Yokoyama, K. Kuwahara, T. Sakakibara, H. Morimoto, S. Kawarazaki, Y. Miyako, J.A. Mydosh, Phys. Rev. Lett. **83**, 5114 (1999)
19. R. Escudero, F. Morales, P. Lejay, Phys. Rev. B **49**, 15271 (1994)
20. F. Morales, R. Escudero, P. Lejay, in *New Trends in Magnetism, Magnetic Materials, and Their Applications*, ed. by J.L. Morán-López, J.M. Sanchez (Plenum Press, New York, 1994), p. 337
21. K. Hanzawa, N. Watanabe, J. Phys.: Condens. Matter **17**, L419 (2005)
22. G.E. Blonder, M. Tinkham, T.M. Klapwijk, Phys. Rev. B **25**, 4515 (1982)
23. Y.G. Naidyuk, A. Nowack, I.K. Yanson, P.N. Chubov, Sov. J. Low Temp. Phys. **17**, 614 (1991)
24. A. Nowack, Y.G. Naidyuk, P.N. Chubov, I.K. Yanson, A. Menovsky, Z. Phys. B Condens. Matter **88**, 295 (1992)
25. K. Hasselbach, J.R. Kirtley, P. Lejay, Phys. Rev. B **46**, 5826 (1992)
26. K. Hasselbach, J.R. Kirtley, P. Lejay, Physica B **186–188**, 201 (1993)
27. A. Nowack, Y.G. Naidyuk, E. Ulbrich, A. Freimuth, W. Schlabitz, I.K. Yanson, A.A. Menovsky, Z. Phys. B **97**, 77 (1995)
28. Y.G. Naidyuk, O.E. Kvitnitskaya, I.K. Yanson, A. Nowack, A.A. Menovsky, Low Temp. Phys. **21**, 236 (1995)
29. Y.G. Naidyuk, O.E. Kvitnitskaya, I.K. Yanson, A. Nowack, A.A. Menovsky, Europhys. Lett. **33**, 557 (1996)
30. St. Thieme, P. Steiner, L. Degiorgi, P. Wachter, Y. Dalichaouch, M.B. Maple, Europhys. Lett. **32**, 367 (1995)
31. Y.G. Naidyuk, H.v. Löhneysen, G. Goll, I.K. Yanson, A.A. Menovsky, Europhys. Lett. **33**, 557 (1996)
32. J.G. Rodrigo, F. Guinea, S. Vieira, F.G. Aliev, Phys. Rev. B **55**, 14318 (1997)
33. K. Hasselbach, P. Lejay, J. Flouquet, Phys. Lett. A **156**, 313 (1991)
34. G. Wexler, Proc. Phys. Soc. **89**, 927 (1966)
35. U. Rauchschwalbe, Physica **147B**, 1 (1987)
36. A.M. Duif, A.G.M. Jansen, P. Wider, J. Phys.: Condens. Matter **1**, 3157 (1989)
37. M. Suzuki, T. Watanabe, A. Matsuda, Phys. Rev. Lett. **82**, 5361 (1999)
38. M. Suzuki, T. Watanabe, Phys. Rev. Lett. **85**, 4787 (2000)
39. F. Steglich, C. Geibel, K. Gloos, G. Olesch, C. Schank, C. Wassilev, A. Loidl, A. Krimmel, G.R. Stewardt, J. Low Temp. Phys. **95**, 3 (1994)
40. T. Kagayama, G. Oomi, K. Iki, N. Mōri, Y. Ōnuki, T. Komatsubara, J. Alloys Compd. **213/214**, 387 (1994)
41. K. Iki, G. Omi, Y. Uwatoko, H. Takahashi, N. Mōri, Y. Ōnuki, T. Komatsubara, J. Alloys Compd. **181**, 71 (1992)
42. M.W. Mc Elfresh, J.D. Thompson, J.O. Willis, M.B. Maple, T. Kohara, M.S. Torikachvili, Phys. Rev. B **35**, 43 (1987)
43. K. Bakker, A. de Visser, E. Brück, A.A. Menovsky, J.J.M. Franse, J. Magn. Magn. Mater. **108**, 63 (1992)