

Signature of Quantum Criticality in Photoemission Spectroscopy

M. Klein,¹ A. Nuber,¹ F. Reinert,^{1,2} J. Kroha,³ O. Stockert,⁴ and H. v. Löhneysen^{5,6}

¹Universität Würzburg, Experimentelle Physik II, Am Hubland, D-97074 Würzburg, Germany

²Forschungszentrum Karlsruhe, Gemeinschaftslabor für Nanoanalytik, D-76021 Karlsruhe, Germany

³Physikalisches Institut, Universität Bonn, Nussallee 12, D-53115 Bonn, Germany

⁴Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany

⁵Physikalisches Institut, Universität Karlsruhe (TH), D-76128 Karlsruhe, Germany

⁶Forschungszentrum Karlsruhe, Institut für Festkörperphysik, D-76021 Karlsruhe, Germany

(Received 13 October 2008; published 30 December 2008)

A quantum phase transition in a heavy-fermion compound may destroy the Fermi-liquid ground state. However, the conditions for this breakdown have remained obscure. We report the first direct investigation of heavy quasiparticle formation and breakdown in the canonical system $\text{CeCu}_{6-x}\text{Au}_x$ by ultraviolet photoemission spectroscopy at elevated temperatures without the complications of lattice coherence. Surprisingly, the single-ion Kondo energy scale T_K exhibits an abrupt step near the quantum critical Au concentration of $x_c = 0.1$. We show theoretically that this step is expected from a highly nonlinear renormalization of the local spin coupling at each Ce site, induced by spin fluctuations on neighboring sites. It provides a general high-temperature indicator for heavy-fermion quasiparticle breakdown at a quantum phase transition.

DOI: 10.1103/PhysRevLett.101.266404

PACS numbers: 71.27.+a, 71.10.-w, 71.28.+d, 79.60.-i

A quantum phase transition (QPT) is a second-order transition occurring at zero temperature, driven by a non-thermal control parameter such as composition or pressure. Here, two competing ground states are separated by a quantum critical point (QCP). Heavy-fermion (HF) systems, in particular, show striking deviations from Fermi-liquid behavior at a QCP between a magnetically ordered and a paramagnetic phase [1,2]. In the standard Hertz-Millis (HM) scenario [3,4], only the bosonic fluctuations of the order parameter become quantum critical, i.e., long-ranged in space and time, leading to anomalous behavior of physical quantities, but leaving the fermionic quasiparticles intact. However, in a HF system the order-parameter fluctuations always couple to the spin degree of freedom of the fermionic excitations, so that the latter may become critical as well and disintegrate. In this case, the formation of the Kondo spin singlet between the conduction electron and the local $4f$ magnetic moments is prevented, and hence the Kondo scale T_K vanishes at the QCP. This scenario has, therefore, been termed local quantum critical (LQC) [5,6]. $\text{CeCu}_{6-x}\text{Au}_x$ is one of the best characterized HF compounds [7–13] with a QPT between a fully Kondo-screened paramagnetic and an antiferromagnetically ordered phase at a critical Au concentration of $x_c = 0.1$. Based on inelastic neutron scattering experiments, it was suggested that the LQC scenario is realized in this compound [14], yet this issue has remained controversial.

In order to address the crucial question of whether Kondo screening persists or not at the QPT, we present in this work ultraviolet photoemission spectroscopy (UPS) measurements, which provide the most immediate access to the screening scale T_K by directly recording the Kondo resonance (KR) in the local Ce $4f$ spectrum [15,16]. By

performing the experiments at temperatures T well above the Néel temperature and above the lattice coherence temperature ($T > \{T_K, T_{\text{coh}}, T_N\}$), we are able to investigate the local spin screening effect *alone*, without the complications of Fermi-volume change due to lattice coherence, which influence the results of other experimental techniques.

The high-resolution UPS was performed with a Gamdata R4000 analyzer and a monochromatized vacuum ultraviolet lamp at $h\nu = 40.8$ eV (see Refs. [16,17] for more details). We cleaved the single crystalline samples *in situ* just before the measurement, already at the measurement temperature. The surface is known to be γ -Ce-like, meaning that the surface properties do not differ from the bulk properties [16].

The inset in Fig. 1(a) shows survey spectra for five Au concentrations x at $T = 15$ K. The energy range includes the spin-orbit (SO) feature at $E_B \approx 260$ meV and the tail of the Kondo resonance just below the Fermi energy E_F . A distinct crystal-field (CF) feature as observed, e.g., in CeCu_2Si_2 and CeNi_2Ge_2 [15,16], is expected near $E_B = 7$ meV but strongly broadened [12] and, hence, not discernible in $\text{CeCu}_{6-x}\text{Au}_x$. The SO peak does not show a significant x -dependent variation in position or intensity. This is expected, since it corresponds to an only virtually excited initial state of the UPS process [15,18]. In order to investigate the KR in more detail, we performed high-resolution measurements near E_F [Fig. 1(a)]. For all concentrations, one observes a significant rise in the intensity from $E_B \approx 20$ meV towards E_F , attributed to the tail of the KR which has its major spectral weight a few meV *above* the Fermi energy [15,16] suppressed by the Fermi-Dirac distribution function (FDD) in the UPS data. Normalizing

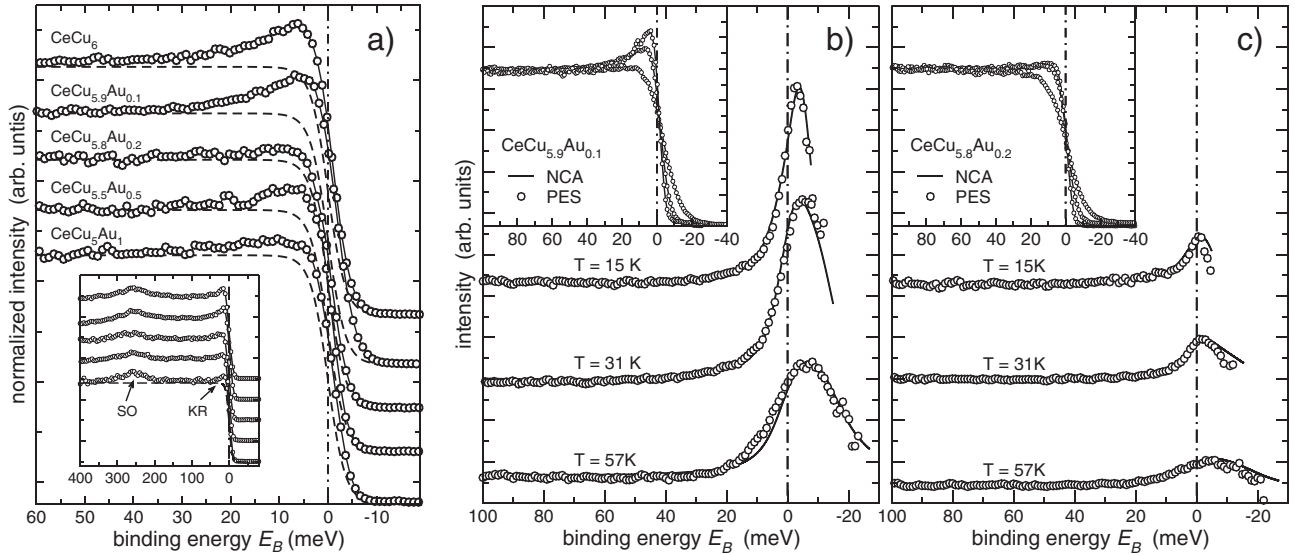


FIG. 1. (a) Near- E_F spectra of $\text{CeCu}_{6-x}\text{Au}_x$ for five different Au concentrations at $T = 15$ K ($h\nu = 40.8$ eV, $\Delta E = 4.9$ meV), normalized at $E_B \approx 100$ meV. The dashed lines describe the resolution-broadened FDD at $T = 15$ K. The inset shows a larger energy range including the SO partner at $E_B \approx 260$ meV ($\Delta E = 15$ meV). (b) and (c) show spectra for $x = 0.1$ and $x = 0.2$, respectively, divided by the FDD, at various T . The solid lines are best NCA fits (see Table I for the resulting model parameter values). The insets in (b) and (c) show the corresponding raw data.

to the FDD allows one to recover the thermally occupied spectrum up to $\approx 5k_B T$ above E_F [15,16] and reveals the KR. Figure 1 shows the FDD-renormalized spectra for $x = 0.1$ [Fig. 1(b)] and 0.2 [Fig. 1(c)]. In the low- T spectra ($T = 15$ K), its maximum is at about 3 meV above E_F for $x = 0.1$ and at 1 meV for $x = 0.2$. The striking feature seen in Figs. 1(b) and 1(c) is the significant drop of the spectral weight of the KR from $x = 0.1$ to 0.2 , i.e., in the vicinity of the critical concentration x_c . The differences in the shape and the T dependence between the $x = 0$ and 0.1 spectra as well as the differences between the $x = 0.2, 0.5$, and 1.0 spectra (not shown) are negligibly small compared to the jump close to x_c .

To extract the Kondo screening scale T_K from the experimental results, one must bear in mind that the coherence temperature T_{coh} obtained from resistivity measurements [19] as well as the Kondo scale extracted from specific heat [20] or neutron scattering data [12,14] are well below our lowest experimental temperature. Moreover, quantum critical fluctuations, which in $\text{CeCu}_{6-x}\text{Au}_x$ extend up to about $T = 7$ K [14] and certainly become crucial at low T , are not expected to influence our experimental spectra. Thus, our data exhibit the onset of the *local* Kondo physics on the Ce atoms only. Therefore, the single-impurity Anderson model (SIAM) is employed to interpret the experimental data. To determine T_K we follow the procedure successfully applied to various Ce compounds in the past [15,16,21–24]: Using the noncrossing approximation (NCA) [25], we calculate the Ce $4f$ spectral function of the SIAM, including all CF and SO excitations. For each composition x , the NCA spectra are broadened by the experimental resolution and fitted to the experimental data, using a single parameter set for all experimental T . The

NCA spectra are then computationally extrapolated to $T \approx 0.1T_K$, where T_K is extracted from the Kondo-peak half width at half maximum (HWHM) of the NCA spectra. The resulting fit parameter values are shown in Table I. Figure 2 shows the x dependence of T_K for $\text{CeCu}_{6-x}\text{Au}_x$ obtained from our UPS data, compared to results of various other experiments [12,19,20]. We emphasize that, irrespective of a possible systematic ambiguity in the fit procedure, the surprisingly abrupt step of T_K near $x = x_c \approx 0.1$ is significant and already clearly visible in the raw data (Fig. 1).

What can be learned from this steplike behavior at elevated T about the nature of the QPT at $T = 0$ K? The method of extracting a Kondo scale from a single-ion picture described above mimics exactly a system of dense Kondo atoms that, however, would *not* undergo any lattice coherence or magnetic ordering at sufficiently low T . This scenario is different from the low- T behavior of the Kondo lattice and two-impurity Kondo problems, where Kondo

TABLE I. SIAM NCA fit parameters, CF splittings of the Ce $4f$ orbitals, $\Delta_{\text{CF}1}$, $\Delta_{\text{CF}2}$, $4f$ -conduction-band hybridization matrix element V , and the resulting Kondo temperatures T_K for different Au concentrations x . Fixed parameters: Conduction-band half-width $D_0 = 2.8$ eV (HWHM), single-particle $4f$ binding energy $\varepsilon_f = -1.05$ eV, and SO splitting $\Delta_{\text{SO}} = 250$ meV.

x	$\Delta_{\text{CF}1,2}$ (meV)	V (meV)	T_K (K)
0	7.2, 13.9	116	4.6
0.1	7.2, 13.9	116	4.6
0.2	8.7, 13.6	111	3.4
0.5	8.9, 13.6	109	3.3
1	9.2, 13.6	108	3.1

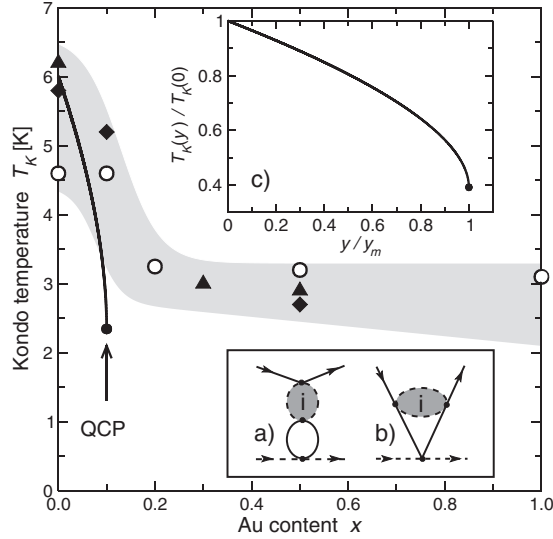


FIG. 2. Dependence of the Kondo temperature T_K on the Au content x , as determined by UPS (open circles), specific heat [20] (triangles), and neutron scattering [12,14] (diamonds). The shaded area is a guide to the eye. The insets (a) and (b) show diagrammatic representations of the RKKY corrections to the local Kondo vertex. The inset (c) and the solid line in the main panel show the universal curve $T_K(y)/T_K(0)$ vs y/y_m as given by Eq. (3).

spin screening would have to compete with the formation of interimpurity coherent states [26] or could be destroyed by critical fluctuations of a magnetic order parameter [27]. By contrast, our system is represented by an effective single-impurity Kondo model where the Kondo exchange coupling J is renormalized by the local spin fluctuations on the surrounding *identical* Kondo atoms through the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling. This problem can be treated in a controlled way by renormalization group (RG) enhanced perturbation theory which allows for definite predictions about the formation of HF quasiparticles at low T , as we will show in the following.

The leading perturbative corrections to J on a given Ce atom at lattice site $i = 0$ are depicted diagrammatically in insets (a) and (b) of Fig. 2. They obviously involve the full dynamical spin susceptibility $\chi_{4f}(T, \Omega)$ of a local Ce $4f$ spin on a neighboring site $i \neq 0$, where $\chi_{4f}(T, 0) = (g_L \mu_B)^2 N(0) D_0 / (4\sqrt{T_K^2 + T^2})$, with $N(0)$ the density of states at the Fermi level, $D_0 \approx E_F$ the band cutoff, and g_L and μ_B the Landé factor and the Bohr magneton, respectively. Summing over all lattice sites $i \neq 0$, one obtains for the direct and the exchange corrections [Figs. 2(a) and 2(b)], respectively, the spin-isotropic terms

$$\delta J^{(d)} = -y \frac{1}{4} J g_i^2 \frac{D_0}{\sqrt{T_K^2 + T^2}} \frac{1}{1 + (D/T_K)^2}, \quad (1)$$

$$\delta J^{(ex)} = -y \frac{1}{4} J g_i^2 \left(\frac{3}{4} + \frac{T}{\sqrt{T_K^2 + T^2}} \right). \quad (2)$$

Here J is the bare, local spin exchange coupling on site $i = 0$, and $g_i = N(0)J_i$ is the dimensionless, bare coupling on site $i \neq 0$. y is a dimensionless factor that describes the (experimentally not known) relation between the RKKY coupling strength and the Au content x . We assume a linear dependence across the QPT $y = \alpha(x + x_0)$, with adjustable parameters α and x_0 . As seen from Fig. 2(a), $\chi_{4f}(T, \Omega)$ imposes a soft cutoff on the energy exchange in $\delta J^{(d)}$, which appears as the last factor in Eq. (1) in terms of the running band cutoff $D \leq D_0$. In $\delta J^{(ex)}$ [Fig. 2(b)] there is no such cutoff; however, $\chi_{4f}(T, \Omega)$ restricts the conduction electron response to the Fermi energy and suppresses $\delta J^{(ex)}$ compared to $\delta J^{(d)}$ by an overall factor of $\sqrt{T_K^2 + T^2}/D_0$, as seen in Eq. (2). The one-loop RG equation for the local spin exchange coupling, including RKKY corrections Eqs. (1) and (2), reads $dJ/d \ln D = -2N(0) \times [J + \delta J^{(d)} + \delta J^{(ex)}]^2$. Note that in this RG equation the couplings g_i on sites $i \neq 0$ are not renormalized, since this is already included in the full susceptibility χ_{4f} . The essential feature is that in the low- T limit ($T \ll T_K$), for which T_K is to be extracted, the direct RKKY correction $\delta J^{(d)}$ [Eq. (1)] is inversely proportional to the renormalized Kondo scale $T_K(y)$ itself via $\chi_{4f}(0, 0)$. $T_K(y)$, including perturbative RKKY corrections, is defined as the scale where the solution of the RG equation diverges. This leads without further approximations to a highly nonlinear renormalization of T_K given by the self-consistency equation

$$\frac{T_K(y)}{T_K(0)} = \exp\left[-\left(\frac{1}{2g} + \ln 2\right) \frac{f(u)}{1 - f(u)}\right], \quad (3)$$

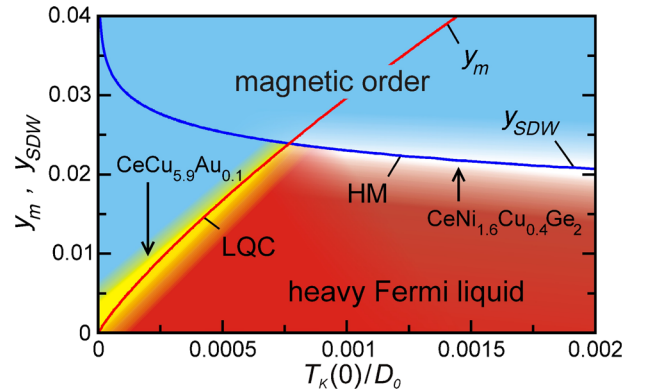


FIG. 3 (color online). Schematic $T = 0$ phase diagram of a HF system with a magnetic QPT driven by RKKY coupling or a SDW instability in the plane of single-ion Kondo scale $T_K(0)/D_0$ and RKKY coupling strength y , as drawn from the high- T analysis. Red line (y_m): Maximum RKKY coupling strength beyond which complete Kondo screening ceases to exist [Eq. (4)]. At the red line, a step occurs in $T_K(y)$. Blue line (y_{SDW}): RKKY coupling strength at which the heavy Fermi liquid becomes unstable towards a SDW, assuming a spin-spin coupling $J_{SDW} = J$. The cases $y_m < y_{SDW}$ and $y_m > y_{SDW}$ distinguish whether the QPT is LQC-like (yellow) or HM-like (white), respectively. The arrows indicate estimates [16] for $T_K(0)/D_0$ for $\text{CeCu}_{6-x}\text{Au}_x$ and for $\text{CeNi}_{2-x}\text{Cu}_x\text{Ge}_2$.

with $g = N(0)J$, $f(u) = u - u^2/2$, and $u = yg^2D_0/[4T_K(y)]$. The single-ion Kondo scale without RKKY coupling is $T_K(0) = D_0 \exp[-1/2g]$. We have verified that this perturbative RG treatment of the RKKY corrections is controlled in the sense that the effective perturbation parameter $f(u) \leq 0.1$ for all self-consistent solutions, i.e., the exponent in Eq. (3), remains small. The solution of Eq. (3) is universal in the dimensionless variables $T_K(y)/T_K(0)$ and y/y_m and turns out to exist only for y smaller than a maximum value y_m . For $y/y_m \rightarrow 1$ it behaves as $T_K(y)/T_K(0) = r + b\sqrt{1 - y/y_m}$, with numerically determined constants $r \equiv T_K(y_m)/T_K(0) \approx 0.391$ and $b \approx 0.517$ [cf. Fig. 2(c)]. The maximum RKKY parameter y_m beyond which a solution of Eq. (3) ceases to exist can be deduced in terms of $\tau_K = T_K(0)/D_0$ as

$$y_m = 8r\tau_K(\ln\tau_K)^2 \left[2 - \ln\frac{\tau_K}{2} - \sqrt{\left(2 - \ln\frac{\tau_K}{2}\right)^2 - 4} \right] \quad (4)$$

(Fig. 3, red line). For $y > y_m$, the Kondo spin exchange coupling including RKKY corrections does not diverge under the RG. Hence, as the essential result of this analysis, in a dense system of Kondo ions, complete Kondo screening ceases to exist above a critical RKKY coupling strength $y > y_m$, even if magnetic ordering does not occur. For comparison, the well-known Doniach criterion [28] pertains to the breakdown of Kondo screening due to magnetic ordering and reads $T_K(0) \approx y_m N(0)J^2$. It is similar, albeit not equivalent, in numerical values to Eq. (4) and does not allow for a determination of the composition-dependent screening scale $T_K(y)$. By identifying y_m with the QCP, as done in Fig. 2, the x dependence of $T_K[y(x)]$ near this critical end point explains the abrupt step of T_K observed experimentally in the high- T spectral data. The small KR spectral weight seen for $x > x_c$ should be interpreted merely as the logarithmic onset of Kondo screening which does not fully develop even for $T \rightarrow 0$. Consequently, the small, residual local moments must order in dimensions $d > 2$ at sufficiently low T .

Hence, the theory predicts two possible scenarios, depending on the size of $T_K(0)/D_0$: (i) Magnetic ordering at $T = 0$ occurs for an RKKY parameter $y = y_{SDW} < y_m$, i.e., without breakdown of Kondo screening. The ordering may arise in this case from a $T = 0$ spin-density-wave (SDW) instability of the underlying heavy Fermi liquid. This corresponds to the HM scenario, depicted in Fig. 3 as the white region. In this case, a steplike behavior of T_K as extracted from the high- T UPS spectra is still predicted from Eq. (3) at $y = y_m$, i.e., inside the region where magnetic ordering occurs at $T < T_N$. (ii) Magnetic ordering does not occur for $y < y_m$. In this case, Eq. (3) indicates a breakdown of Kondo screening at the magnetic QCP, and quantum critical fluctuations (not considered in our theory) will suppress the low- T spin screening scale to zero at this point. This is the LQC scenario, shown in Fig. 3 as the yellow region.

In conclusion, the theoretical analysis predicts generally that an abrupt step of the Kondo screening scale extracted from high- T spectral data should occur in any HF compound with competing Kondo and RKKY interactions, as long as the single-ion Kondo screening scale is larger than the magnetic ordering temperature. Whether this distinct feature is located at the quantum critical control parameter value x_c or inside the magnetically ordered region constitutes a general high- T criterion to distinguish the LQC and HM scenarios. The sharp step of T_K occurring in our UPS data very close to x_c (see Fig. 2) strongly supports that $\text{CeCu}_{6-x}\text{Au}_x$ falls into this class, as was previously inferred indirectly from inelastic neutron scattering data [14].

We thank F. Assaad, S. Kirchner, and M. Vojta for fruitful discussions. This work was supported by DFG through Re 1469/4-3/4 (M. K., A. N., and F. R.), SFB 608 (J. K.), and FOR 960 (H. v. L.).

-
- [1] H. v. Löhneysen, A. Rosch, M. Vojta, and P. Wölfle, *Rev. Mod. Phys.* **79**, 1015 (2007).
 - [2] P. Gegenwart, Q. Si, and F. Steglich, *Nature Phys.* **4**, 186 (2008).
 - [3] J. A. Hertz, *Phys. Rev. B* **14**, 1165 (1976).
 - [4] A. J. Millis, *Phys. Rev. B* **48**, 7183 (1993).
 - [5] Q. Si, S. Rabello, and J. L. Smith, *Nature (London)* **413**, 804 (2001).
 - [6] P. Coleman, C. Pépin, Q. Si, and R. Ramazashvili, *J. Phys. Condens. Matter* **13**, R723 (2001).
 - [7] H. v. Löhneysen *et al.*, *Phys. Rev. Lett.* **72**, 3262 (1994).
 - [8] H. v. Löhneysen, *J. Phys. Condens. Matter* **8**, 9689 (1996).
 - [9] H. v. Löhneysen *et al.*, *J. Magn. Magn. Mater.* **177–181**, 12 (1998).
 - [10] O. Stockert *et al.*, *Phys. Rev. Lett.* **80**, 5627 (1998).
 - [11] H. v. Löhneysen *et al.*, *Eur. Phys. J. B* **5**, 447 (1998).
 - [12] B. Stroka *et al.*, *Z. Phys. B* **90**, 155 (1993).
 - [13] K. Grube *et al.*, *Phys. Rev. B* **60**, 11 947 (1999).
 - [14] A. Schröder *et al.*, *Nature (London)* **407**, 351 (2000).
 - [15] F. Reinert *et al.*, *Phys. Rev. Lett.* **87**, 106401 (2001).
 - [16] D. Ehm *et al.*, *Phys. Rev. B* **76**, 045117 (2007).
 - [17] M. Klein *et al.*, *Phys. Rev. Lett.* **101**, 046406 (2008).
 - [18] J. Kroha *et al.*, *Physica (Amsterdam)* **18E**, 69 (2003).
 - [19] H. v. Löhneysen *et al.*, *J. Alloys Compd.* **408–412**, 9 (2006).
 - [20] H. G. Schlager, A. Schröder, M. Welsch, and H. v. Löhneysen, *J. Low Temp. Phys.* **90**, 181 (1993).
 - [21] F. Patthey *et al.*, *Phys. Rev. B* **42**, 8864 (1990).
 - [22] M. Garnier *et al.*, *Phys. Rev. Lett.* **78**, 4127 (1997).
 - [23] J. Allen *et al.*, *J. Appl. Phys.* **87**, 6088 (2000).
 - [24] D. Ehm *et al.*, *Acta Phys. Pol. B* **34**, 951 (2003).
 - [25] T. Costi, J. Kroha, and P. Wölfle, *Phys. Rev. B* **53**, 1850 (1996); N. E. Bickers, D. L. Cox, and J. W. Wilkins, *ibid.* **36**, 2036 (1987).
 - [26] B. A. Jones and C. M. Varma, *Phys. Rev. Lett.* **58**, 843 (1987).
 - [27] H. Maebashi, K. Miyake, and C. M. Varma, *Phys. Rev. Lett.* **95**, 207207 (2005).
 - [28] S. Doniach, *Physica (Amsterdam)* **91B+C**, 231 (1977).