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## Disappearance of hyperscaling at low temperatures in non-Fermi-liquid CeCu<sub>5.2</sub>Ag<sub>0.8</sub>

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Following the recently published results (down to 0.3 K) on magnetic-field induced non-Fermi-liquid behavior in polycrystalline  $\text{CeCu}_{6-x}\text{Ag}_x$  (0.09<x<1.2) we present here specific heat *C* measurements down to T=70 mK on a  $\text{CeCu}_{5.2}\text{Ag}_{0.8}$  single crystal. In the single crystal we find that in a critical magnetic field  $B_c$  only along the magnetic easy (c) axis, where the long-range antiferromagnetic order ( $T_N=0.7$  K in B=0 T) is just suppressed to T=0, a quantum critical point is reached with non-Fermi-liquid properties down to our lowest temperature of measurement. The lowest temperature C/T data,  $B\parallel c$ , for  $B>B_c$  are proportional to 1  $-aT^{0.5}$ , a temperature dependence proposed in the Gaussian ( $\Leftrightarrow$  weakly interacting spin fluctuations) selfconsistent renormalized spin-fluctuation theory. In contrast, the data at higher temperatures—including scaling with  $B/T^{\beta}$  of both *C* and magnetization as a function of *B*—require a non-Gaussian strongly interacting spin-fluctuation model. In order to test this apparent crossover in behavior we have examined the scaling of C(B) for T<0.2 K and find that the high-temperature scaling with  $B/T^{\beta}$  (''hyperscaling'') does not function at the lowest temperatures. [S0163-1829(98)51648-7]

### **INTRODUCTION**

Alloying of the nonmagnetic heavy fermion compound CeCu<sub>6</sub> with Ag leads to a linear increase of the unit cell volume of the orthorhombic Pnma structure and therefore changes the hybridization between the conduction electrons and the Ce-4f moments in the system. For Ag concentrations higher than x = 0.09, the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange coupling overcomes the single-site Kondo screening and long-range antiferromagnetic order appears. The ordering temperature  $T_N$  increases linearly with further substitutions up to x=1 with  $T_N=0.8$ .<sup>1,2</sup> At a critical concentration  $x_c = 0.09$ , just where  $T_N = 0$  in CeCu<sub>6-x</sub>Ag<sub>x</sub>, typical non-Fermi-liquid (nFl) properties (for a review of NFl experiment and theory, see Ref. 1 and the references therein) are found,<sup>1</sup> similar to Au doping.<sup>3</sup> C/T is proportional<sup>1</sup> to  $-\log T$  and the data are nearly identical to those at the critical concentration  $x_c = 0.1$  in CeCu<sub>6-x</sub>Au<sub>x</sub>.<sup>3</sup>

Reference 1 reported that magnetic field is an effective tool to reach the quantum critical point (QCP) with nFl properties down to 0.3 K in a wide range of the magnetic part (x>0.09) of the phase diagram of CeCu<sub>6-x</sub>Ag<sub>x</sub>. Changing of the magnetic field results in a change of the relevant coupling of the spin fluctuations and does not change the atomic disorder. The first measurements<sup>1</sup> were on polycrystalline samples which, because of the magnetic anisotropy of the orthorhombic system, leads to a deviation from a pure temperature dependence in the specific heat at T < 0.3 K, because there is a mix of several directions present in the samples. Therefore we present here measurements of specific heat, as well as discussing recently published resistivity results, on a single crystal of CeCu<sub>5.2</sub>Ag<sub>0.8</sub> with magnetic field applied along the c and the b axes. Furthermore we compare our results at lowest temperatures with the theoretical predictions of the self-consistent renormalized (SCR) theory,<sup>4</sup> which describes well the results for  $Ce_{1-x}La_xRu_2Si_2$  (Ref. 5) and CeCu<sub>2</sub>Si<sub>2</sub>.<sup>6</sup> The SCR model predicts that at lowest temperatures  $\rho = \rho_0 + cT^{1.5}$  and that  $C/T \propto 1 - aT^{0.5}$ , while C/T  $\propto -\log T$  for a limited (~60% of a decade) region at higher temperature. The SCR theory has limitations (e.g., 3-dimensional character vs the quasi-2-dimensional character observed<sup>7</sup> in the comparable CeCu<sub>6-x</sub>Au<sub>x</sub>, as well as not including the magnetic field—central to the present results—in the effective Hamiltonian) making the utility of the theory for comparison to the present work only qualitative.

#### **EXPERIMENTAL DETAILS**

The CeCu<sub>5.2</sub>Ag<sub>0.8</sub> single crystal was grown by the Czochralski method in a tri-arc furnace out of a stoichiometric melt with a small Ag excess to prevent a Ag deficiency during the growth process. An x-ray analysis of the beginning and the end of the crystal shows a single phase orthorhombic *Pnma* structure equal to CeCu<sub>6</sub> and to the polycrystalline samples.<sup>1</sup> By comparing with the well-known dependencies of the lattice constants *a*, *b*, *c* and of  $T_N$  with Ag doping,<sup>1</sup> the composition of the single crystal is found to be exactly CeCu<sub>5.2</sub>Ag<sub>0.8</sub>, with *a*=8.325 Å, *b*=5.093 Å, and *c* = 10.377 Å. Along the growth direction no Ag-concentration gradient was detectable. For the measurements of the specific heat a 1-mg sample was prepared from the crystal and mounted on a sapphire disk; the orientation was confirmed via measurements of the anisotropic dc susceptibility.

Magnetic susceptibility measurements down to 1.8 K and up to 7 T were made using a Quantum Design magnetometer. The specific heat and resistivity in magnetic fields up to 13 T were measured in a dilution refrigerator down to T = 50 mK using established methods.<sup>2,8</sup>

### **RESULTS AND DISCUSSION**

As expected from the existing data<sup>1</sup> on  $\text{CeCu}_{6-x}\text{Ag}_x$  for x=0.8, the antiferromagnetic phase transition occurs at  $T_N = 0.7$  K (see Fig. 1) with a sharp peak in the specific heat C. In accordance with Ref. 9, almost the same anisotropy of the

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FIG. 1. Specific heat *C* vs log *T* of a CeCu<sub>5.2</sub>Ag<sub>0.8</sub> single crystal for small magnetic fields applied parallel to the easy *c* direction, shown in (a), and to the *b* axis as shown in (b). The inset gives the magnetic-field dependence of the *afm* ordering temperature  $T_N$  for B||c and B||b, determined from the peak in *C*.

magnetic susceptibility, smaller than observed<sup>10</sup> for  $CeCu_{6-x}Au_x$ , is found (data not shown) with the *c* axis being the easy magnetic axis with the highest  $\chi(2 \text{ K})$ . In Fig. 1 the specific heat C(T) is shown for small magnetic fields parallel to the *c* axis in Fig. 1(a) and the the *b* axis in Fig. 1(b). In contrast to Ref. 9 the peak in C(T) is shifted continuously to lower temperatures with increasing field and remains sharp. No second phase transition is observed in our single crystal. Extrapolating the magnetic-field dependence for the easy direction the afm transition vanishes in  $B_c || c \sim 2.3 \text{ T}$  (see Fig. 1 inset). Measurements of the specific heat with fields parallel to the *b* direction yields a critical magnetic field  $B_c || b$  of  $\sim 5.7 \text{ T}$ .

In Fig. 2 C/T is plotted for B||c in fields  $B \ge B_c||c|$ =2.3 T, in addition to the zero field data. Non-Fermi-liquid behavior is observed at the critical magnetic field  $B_c$ =2.3 T, where  $T_N$  is just suppressed to T=0, with C/T $\propto -\log T$  (straight line in Fig. 2) in an intermediate tempera-



FIG. 2. Specific heat *C* of a CeCu<sub>5.2</sub>Ag<sub>0.8</sub> single crystal plotted as *C*/*T* vs log *T* between *T*=0.07 and 3 K for magnetic fields *B* = 0, 2.3 (=*B<sub>c</sub>*), 2.5, 2.75, 3, and 5 T, applied parallel to the easy *c* axis. For all measurements the hyperfine contribution caused by the Cu content in the sample is subtracted (Ref. 13). In *B<sub>c</sub>*=2.3 T nFl behavior is found between 0.07 and 2 K. The straight line is a guide to the eye for  $\Delta C/T \approx -\log T$  behavior for the *B*=*B<sub>c</sub>* data, while the curved line through the 2.3 T data below 0.2 K is a fit *C*/*T* =  $\gamma_0 - aT^{0.5}$  ( $\gamma_0$ =4.89 J/mole K<sup>2</sup>, *a*=3.67 J/mole K<sup>2.5</sup>).



FIG. 3. Specific heat *C* of a CeCu<sub>5.2</sub>Ag<sub>0.8</sub> single crystal plotted as *C*/*T* vs log *T* between T=0.07 and 3 K for B=0, 4, 5, 5.7 $(=B_c||b)$ , and 13 T, applied parallel to the *b* axis. nFl behavior (strongly divergent *C*/*T*) is never observed, but rather *C*/*T*  $\rightarrow$  const as  $T\rightarrow 0$ . The straight line through the 5.7 T data (open circles) shows the  $C/T \propto -\log T$  behavior between 0.3 and 2 K.

ture region between 0.2 and 1.2 K. For T < 0.2 K, the C/T data deviate from the log *T* increase, with  $C/T = \gamma_0 - aT^{0.5}$ . Increasing the magnetic field to 2.5 T shows already a cross-over behavior in the specific heat with a distinct deviation from  $C/T \propto -\log T$  for T < 0.3 K. In B = 3 T, C/T starts to be constant at lowest temperatures, reaching the Fermi-liquid regime. These results, in the region (T > 0.3 K) where they overlap agree with our previous<sup>1</sup> polycrystalline data.

One would expect that by applying the critical magnetic field  $B_c = 5.7$  T along the *b* direction of the single crystal, the same behavior will be observable. This is in fact true for the data above T=0.3 K as shown in Fig. 3. However, at lower temperatures C/T for B||b increases only up to C/T (T=0.1 K)=3 J/mole K<sup>2</sup>, whereby for B||c, a value of C/T (T=0.1 K)=3.7 J/mole K<sup>2</sup> is reached. The differences between the two field directions are already visible in Fig. 1 for smaller magnetic fields, i.e., for B||c the value for C at lowest temperatures in Fig. 1 increases continuously with the field while for B||b, the value of C in the field never breaks through the zero field curve below  $T_N(B)$  and  $C(T \rightarrow 0)$  decreases continuously with the magnetic field. These results show that the anisotropy of the spin-fluctuation spectrum is not fixed to the anisotropy of the ordering moments.

Consider now the resistivity measurements<sup>11</sup> (not shown) with the magnetic field also applied in the *c* direction of the crystal. In accordance with the specific heat results nFl behavior is also observed for the resistivity in the critical field  $B_c || c = 2.3$  T.  $\rho$  clearly shows a linear temperature dependence between 0.17 and 0.3 K, while at lowest temperatures  $\rho = \rho_0 + cT^{1.4\pm0.1}$ . This is in agreement with the prediction of the SCR theory, used above to describe the specific heat data, that  $\rho = \rho_0 + cT^{1.5}$  at lowest temperatures. A further increase of the field *B* yields the usual Fermi-liquid behavior with  $\rho = \rho_0 + AT^2$ .

Inspired by the question of whether the crossover from an intermediate temperature range behavior  $(C/T \propto -\log T \text{ and } \rho \propto \rho_0 + BT)$  above ~0.2 K to a low temperature, noninteracting fluctuation SCR theory behavior  $(C/T \propto 1 - aT^{0.5} \text{ and } \rho = \rho_0 + cT^{\sim 1.5})$  implies a change in the nature of the fluctuations responsible for the nFl behavior in CeCu<sub>5.2</sub>Ag<sub>0.8</sub>,

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FIG. 4. (a) Scaling of the change of the specific heat divided by temperature of CeCu<sub>5.2</sub>Ag<sub>0.8</sub> with applied field  $B \parallel c$  vs  $B - B_c$  (= $\Delta B$ ) divided by  $T^{1.6}$  in the intermediate temperature range. The points for all fields,  $B > B_c$ , scale onto one universal curve. (b) The same as for (a), but for the lowest temperature data. Note that the data do not even tend toward falling onto one curve.

we consider now the scaling behavior of  $C(B)/T - C(B_c)/T$  for both temperature regimes. In Fig. 4(a), for T > 0.2 K, the scaling data demonstrate definite scaling with  $B/T^{\beta}$  with  $\beta = 1.6 \pm 0.1$ . This is consistent with the scaling data for  $C(B)/T - C(B_c)/T$  for T > 0.3 K and for M for T > 1.8 K observed<sup>1</sup> for polycrystalline CeCu<sub>6-x</sub>Ag<sub>x</sub>. It should be stressed that this type of scaling, known as hyperscaling, may be taken as proof that there exist above  $B_c$  strongly interacting fluctuations that require a non-Gaussian theoretical approach. Thus, the qualitative agreement of the SCR theory prediction (that  $C/T^{\alpha} - \log T$  in an intermediate temperature range) with our data above 0.2 K is negated by the fact that the data obey hyperscaling in this temperature regime—the data taken as a whole are not compatible with the SCR theory in this temperature range.

What is found for T < 0.2 K for scaling of C(B), Fig. 4(b), offers a sharp contrast to the higher temperature scaling in Fig. 4(a). The data do not scale onto one curve, neither for  $\beta = 1.6$  nor for the "best fit" (Fig. 5) using  $\beta = 1.1$ . Thus, our data and their scaling offer evidence that, as the QCP is approached from higher temperature, although the interme-

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- <sup>1</sup>K. Heuser, E.-W. Scheidt, T. Schreiner, and G. R. Stewart, Phys. Rev. B **57**, 4198 (1998).
- <sup>2</sup>G. Fraunberger, B. Andraka, J. S. Kim, U. Ahlheim, and G. R. Stewart, Phys. Rev. B 40, 4735 (1989).
- <sup>3</sup>H. von Löhneysen, J. Phys.: Condens. Matter 8, 9689 (1996).
- <sup>4</sup>T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. **64**, 960 (1995).
- <sup>5</sup>S. Kambe, S. Raymond, L.-P. Regnault, J. Flouquet, P. Lejay, and P. Haen, J. Phys. Soc. Jpn. **65**, 3294 (1996).
- <sup>6</sup>F. Steglich, B. Buschinger, P. Gegenwart, M. Lohmann, R. Helfrich, C. Langhammer, P. Hellmann, L. Donnewert, S. Thomas, A. Link, C. Geibel, M. Lang, G. Sparn, and W. Assmus, J. Phys.: Condens. Matter 8, 9909 (1996).
- <sup>7</sup> A. Schröder, G. Aeppli, E. Bucher, R. Ramazashvili, and P. Coleman, Phys. Rev. Lett. **80**, 5623 (1998).



FIG. 5.  $[C(B)/T - c(B_c)/T]$  vs  $(B - B_c)/T^{\beta}$ , at lowest temperatures for CeCu<sub>5.2</sub>Ag<sub>0.8</sub>,  $B \parallel c$ , for  $\beta = 1.1$ , which brings the data closest to lying on one common curve. Even with this "best" fit, the data clearly do not scale in this lowest temperature range.

diate and low temperature regimes both demonstrate nFl behavior, the behaviors are strongly differentiated. The strong, non-Gaussian fluctuation behavior at higher temperatures is replaced as  $T \rightarrow 0$  by weak, Gaussian fluctuations. This may be theoretically pictured<sup>12</sup> as a depopulation of the fluctuation phase space as temperature ( $\Leftrightarrow$  thermal energy) is lowered, causing a weakening of the fluctuation interactions. Work is underway to determine the temperature at which this crossover between hyperscaling and Gaussian behavior occurs as a function of silver concentration in CeCu<sub>6-x</sub>Ag<sub>x</sub>.<sup>13</sup>

As a final remark, our observation of the failure at lowest temperature of hyperscaling is additional evidence of the existence of hyperscaling in the higher temperature data. Our data are clearly precise enough to allow the correct determination of the scaling behavior.

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- <sup>8</sup>W. W. Kim, J. S. Kim, B. Andraka, and G. R. Stewart, Phys. Rev. B **47**, 12 403 (1993).
- <sup>9</sup>O. Stockert, A. Neubert, and H. von Löhneysen, Physica B 230-232, 250 (1997).
- <sup>10</sup>H. G. Schlager, A. Schröder, M. Welsch, and H. von Löhneysen, J. Low Temp. Phys. **90**, 181 (1993).
- <sup>11</sup>A short description of the resistivity data, without the analysis given here, will be available in E.-W. Scheidt, T. Schreiner, K. Heuser, and G. R. Stewart, in Proceedings of the SCES, Paris, 1998 [Physica B (to be published)].
- <sup>12</sup>A. M. Tsvelik (private communication).
- <sup>13</sup>It should be mentioned that the internal field is enhanced by a factor of 1.17, determined by specific heat measurements in high magnetic fields ( $B \ge 5$  T), where Fermi-liquid behavior occurs at lowest temperatures.