Magnetic Order in the Heavy Fermion Compound $CeCu_6$ at mK Temperatures

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(Received 10 December 1999)

We have measured the ac magnetic susceptibility in various fields and thermal expansion of single crystals of the heavy fermion compound $CeCu_6$ at temperatures down to 250 μK. The susceptibility of $CeCu_6$ shows a peak at about 2 mK and has a large anisotropy. We also detected an anomaly of thermal expansion at the same temperature. The observed behaviors of the susceptibility and the thermal expansion in $CeCu_6$ indicate the occurrence of an antiferromagnetic order.

PACS numbers: 75.30.Mb, 71.27.+a, 75.30.Kz

Heavy fermion (HF) behavior is understood as a consequence of a competition between RKKY interaction and the Kondo effect. The complete screening of magnetic moments by the conduction electrons can lead to the paramagnetic Fermi-liquid (FL) state. $CeAl_3$ and $CeCu_6$ have been considered as the typical HF material of the largest effective mass. $CeAl_3$ exhibits magnetic order in single crystals [1] while polycrystals appear to be nonmagnetic [2]. The only HF compound whose magnetic ground state has not been determined is $CeCu_6$. The experimental results on $CeCu_6$ suggest the possibility of magnetic order. Short-range antiferromagnetic correlations were noted by neutron-scattering measurements in $CeCu_6$ [3,4]. During the last few years the measurements of ac susceptibility [5], Cu nuclear quadrupole resonance (NQR) [6], dc magnetization, and specific heat [7] on $CeCu_6$ were carried out down to submillikelvin temperatures. They suggest that the possible magnetic order occurs at about 2 to 3 mK.

In alloys of $CeCu_6$ where some Cu atoms are replaced by Au or Ag, the existence of the magnetic order has been clearly observed [8,9]. The Néel temperature T_N decreases with decreasing concentration x and the magnetic order is suppressed at the critical concentration $x_c = 0.1$ and 0.09 in $CeCu_{6-x}Au_x$ and $CeCu_{6-x}Ag_x$, respectively. At the critical point $x = x_c$ where the magnetic order is suppressed, non-Fermi-liquid (NFL) behaviors were observed [10,11]. NFL behavior is seen in the specific heat, where C/T diverges as $-\log T$, and in the susceptibility, where χ behaves like $1 - \sqrt{T}$ or $-\log T$ at low temperature [12]. This is in marked contrast to the FL behavior where C/T and χ are constant. NFL behavior has been referred to the proximity to quantum phase transition. Quantum phase transition is $T = 0$ phase transition between magnetic and nonmagnetic ground states. The magnetic order is suppressed to absolute zero by changing a parameter such as magnetic field, composition, or pressure which changes the amplitude of quantum fluctuations. The critical fluctuations in $CeCu_{6-x}Au_x$ were determined by inelastic neutron scattering [13,14]. In recent years, many quantum phase transitions have been found and they show a variety of interesting behavior in the neighborhood of the critical point.

The behaviors in a series of $CeCu_6$ suggest that $CeCu_6$ appears to be close to the quantum critical point (QCP). We have carried out the experiments of the ac susceptibility and thermal expansion on single-crystalline $CeCu_6$ along the three principal axes at ultralow temperatures in order to prove the magnetic order of $CeCu_6$ and investigate the behaviors near the QCP.

The single crystal sample of $CeCu_6$ was grown by the Czochralski pulling method and was cut into three pieces (about a $5 \times 5 \times 5$ mm cube) for the measurement in different crystal axes simultaneously. The cerium of 99.99% purity and the copper of 99.999% purity were provided as starting materials by Johnson-Matthey. The residual resistivity of the sample is of the order of 1 $\mu\Omega$ cm which is among the best values. $CeCu_6$ has an orthorhombic structure at room temperature, and changes into a monoclinic structure at about 200 K. Here the orthorhombic notation is used. The samples were cooled with a copper nuclear demagnetization refrigerator and a ^{3}He-^{4}He dilution refrigerator. The temperature of the samples was measured by a Pt NMR thermometer, a ^{3}He melting curve thermometer, and a Ge-resistance thermometer.

The ac susceptibility of $CeCu_6$ was measured by a mutual inductance method. The experimental setup is similar to the arrangement used by Herrmannsdörfer *et al.* [15]. The samples were silver epoxied to the copper cold finger that was bolted to the copper nuclear demagnetization stage. Each sample has its own secondary coil (8-mm diam, 4.5-mm length, 10 000 turns). The primary coil and static field coil were placed inside a Nb shield, and these were thermally anchored to the mixing chamber of a dilution refrigerator. The whole assembly was surrounded by a mu metal shield to suppress external stray fields. The measurements of both components of the complex susceptibility $\chi = \chi' - i\chi''$ were performed by a commercial mutual inductance bridge [16] at a frequency of 16 Hz in a static field $0 \leq B \leq 11$ mT. The ac excitation field was less than 1 μT and parallel to the external static field. The susceptibility signal was calibrated by the Meissner effect in an indium sample.

The thermal expansion of $CeCu_6$ was measured by the capacitance method. The samples were glued to the sample cell made of high purity copper. The copper capacitor plate of 12 mm in diameter was attached to the sample with Stycast 2850FT epoxy of 0.1 mm thick. The fixed plate was mounted in a holder which was attached to the body of the cell. The capacitance was measured using a bridge with a ratio transformer and a reference capacitor. The change in length Δl of the sample was derived from the measured capacitance. We neglected the change in length of copper in the holder, since the thermal expansion coefficient α of the $CeCu_6$ is larger than that of Cu by 2 orders of magnitude below 10 K [17]. The thermal expansion coefficient α was obtained by differentiating the value of $\Delta l/l$.

The real part of the susceptibilities χ' of $CeCu_6$ along the three principal axes and the imaginary part of the susceptibility χ'' along the a axis in the fields up to 11 mT are shown in Fig. 1. The susceptibility was measured during a slow cooling and warming of 4 μK/h and the data show no appreciable hysteresis. The susceptibility displayed in the figures is the net susceptibility which is obtained by subtracting the background susceptibility. The values of χ' are normalized to the absolute value at 0.6 K taken from the previous study [18]. The susceptibility of $CeCu_6$ is strongly anisotropic and shows a peak at about 2 mK following a plateau due to the Pauli susceptibility.

We find that the susceptibility above its peak temperature obeys the Curie-Weiss law. The results of this analysis are shown as solid lines in Fig. 1 and the temperature dependence of the inverse susceptibility along the a axis is shown in Fig. 2. The Weiss temperature $\theta = -1.49$ and -1.05 mK and the Curie constant $C = 1.57 \times 10^{-4}$ and 1.11×10^{-5} K are obtained by the fitting between 3 and 80 mK along the a and the b axes, respectively. Along the c axis, the susceptibility obeys the Curie-Weiss law in the narrower temperature region, between 3 and 50 mK. The obtained values are $\theta = -1.98$ mK and $C = 9.76 \times 10^{-6}$ K along the c axis. The negative Weiss temperature indicates antiferromagnetic behavior of $CeCu_6$. The Curie constant of the a axis corresponds to $7.9 \times 10^{-2} \mu_B$/Ce.

We will discuss the peaks in the ac susceptibility in $CeCu_6$ shown in Fig. 1. It is seen that a quite distinct behavior takes place in each of the three directions. In a zero magnetic field χ'_a along the a axis passes through a very large peak at 2.3 mK and below 0.5 mK decreases to about 1/10 of the peak value. The peak of χ''_a is observed at 1.5 mK which gives the steepest slope in χ'_a. This fact confirms that χ is measured correctly. The maximum values seen in χ'_b and χ'_c are about 20 times smaller than that in χ'_a. With a decreasing temperature below 0.5 mK χ'_b decreases to about 2/3 of the peak value; χ'_c is constant down to the lowest temperature. The behavior in χ'_c is likely to be an antiferromagnet in which χ perpendicular to the

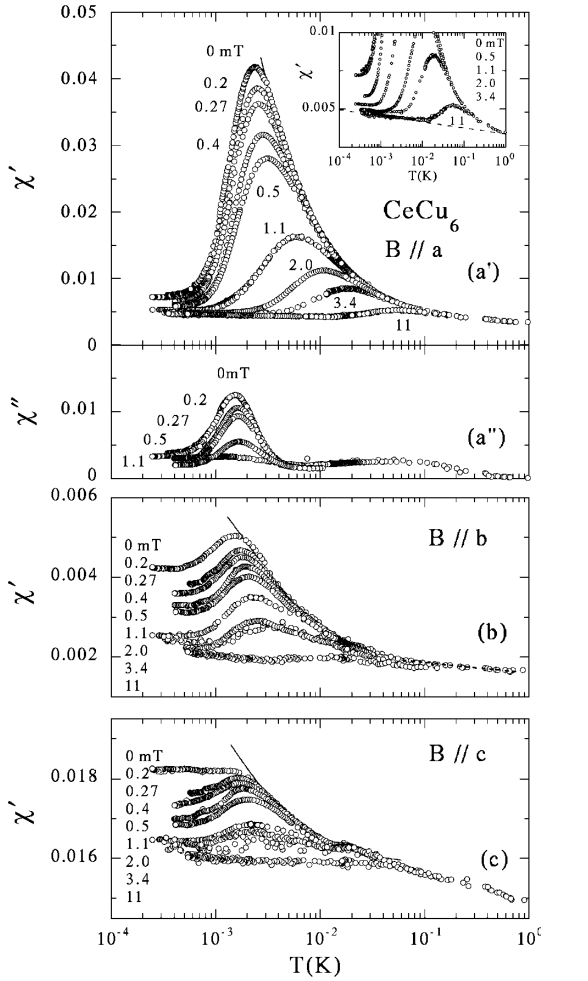

FIG. 1. (a′),(b),(c) The real part of the susceptibility of $CeCu_6$ along the a, the b, and the c axes, respectively. (a″) The imaginary part of the susceptibility of $CeCu_6$ along the a axis. The volume susceptibility is given in dimensionless SI units. The solid lines are fit to the Curie-Weiss law. The numbers in the figure show the magnetic field. The dashed lines in the inset of (a′) and in (b) show $\chi \propto -\log T$.

sublattice magnetization stays constant below T_N. The very large peak seen in χ_a suggests that the direction of preferred spin alignment is along the a axis. With an increasing field the temperature of the peak in χ''_a does not change and the peak in χ''_a disappears in the field larger than 0.5 mT. The peaks of χ'' along the b and the c axes were barely detectable. The temperature giving maximum χ', T_M, follows the linear dependence in the fields that are larger than 0.3 mT. The linear increase of T_M with B means a simple paramagnet. Such fields produced little effect on χ'_b and χ'_c but changed χ'_a dramatically. In the smaller field T_M deviates from the linear dependence and is nearly constant along three axes, which shows the occurrence of the magnetic order. These results indicate that the antiferromagnetic order occurs in small fields, and the magnetic order is suppressed by the field $B \geq 0.3$ mT. The ordering temperature $T_N = 2.0$ mK is determined by

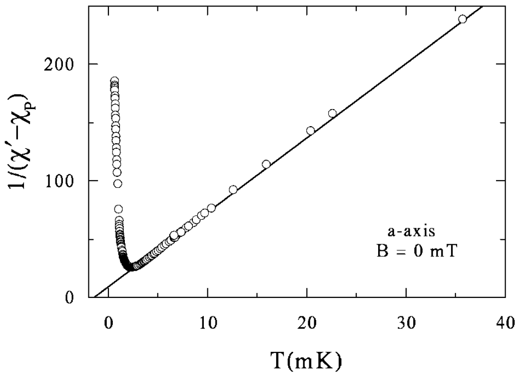

FIG. 2. The inverse susceptibility of $CeCu_6$ along the a axis in zero magnetic field. The solid line shows the Curie-Weiss law. χ_P is the Pauli susceptibility, estimated from the fitting by $\chi' = \chi_P + C/(T - \theta)$.

a intersection of the Curie-Weiss law and the extrapolation of susceptibility from the lowest temperature.

The susceptibility of the a axis is 10 times larger than the value expected from impurity concentration of the starting materials. Therefore the peaks of susceptibility are not due to the effect of magnetic impurities. In addition, we have measured the thermal expansion which is not appreciably affected by the impurities in order to rule out the effect of the magnetic impurities. The data were taken in several runs which allowed us to correct drift and jumps of the signal in time. The thermal expansion coefficient α, obtained by differentiating the value of $\Delta l/l$ of the a axis, is shown in Fig. 3. The thermal expansion coefficient shows an abrupt change in the slope (anomaly) at 2.0 mK which is in agreement with the temperature of peaks in the susceptibility. The anomaly in the thermal expansion shows the magnetic order of $CeCu_6$. This kind of anomaly is known to show the magnetic order as in the case of the magnetic order in UPt_3 observed around 18 mK [19].

The susceptibility measured in our experiment is similar to the previous reports. Jin *et al.* found peaks at 3 and 2 mK along the a and the b axes, respectively. And the polycrystalline sample has a broad peak at 2.5 mK

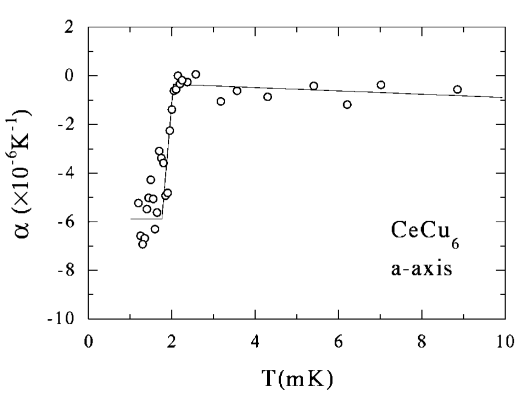

FIG. 3. The thermal expansion coefficient of $CeCu_6$ along the a axis. The line guides the eye.

[5]. Similar features have been shown by Schuberth *et al.* around 3 mK [7]. In the NQR measurement Pollack *et al.* observed deviation from the Korringa law in the spin-lattice relaxation times below 5 mK [6]. The fact that a similar structure was found in different samples from different sources indicates that the magnetic order in $CeCu_6$ is robust. In the previous reports, the ordering temperatures were not discussed. The temperature where the abrupt change was observed in the thermal expansion coefficient, $T = 2.0$ mK, can be regarded as the ordering temperature, because there is a simple relation between the expansion coefficient and the specific heat. The ordering temperature in the thermal expansion coefficient is consistent with $T_N = 2.0$ mK determined by the susceptibility measurements.

We will now consider the ordering temperature by applying the self-consistent renormalization (SCR) theory. The SCR model can be applied to any itinerant systems where spin fluctuations are localized around the collective q vector and dominate low energy excitations. Recently the SCR theory has been applied to heavy fermion systems [20]. The Néel temperature T_N is described by

$$T_N = 0.1376 m_Q^{4/3} T_A^{2/3} T_0^{1/3},$$

where m_Q is the spontaneous magnetic moment in μ_B at $T = 0$, T_A and T_0 are characteristic temperatures in the q space and ω space. An upper limit of an ordered moment $0.01\mu_B$ was inferred from μSR measurements performed down to 40 mK [21]. If we use the derived values of $T_A = 5.5$ K and $T_0 = 3$ K [22] obtained from specific heat and magnetic susceptibility measurements, we can obtain $T_N = 1.3$ mK. The estimation by means of the SCR theory gives good agreement to the experimental Néel temperature. The agreement with the SCR theory suggests the antiferromagnetic order in $CeCu_6$ is explained well by the spin fluctuation effect. The small magnetic moment and the low Néel temperature suggest that the weak antiferromagnetic order occurs in $CeCu_6$.

The large anisotropy can be described by the nesting of the Fermi surface. We can obtain the energy gap $\Delta = 2.58$ mK by fitting the susceptibility data along the a axis below 1 mK using the expression $\chi = \chi_0 + A\exp(-\Delta/T)$. Here, χ_0 is interpreted as the residual Pauli paramagnetic contribution coming from the ungapped Fermi surface at 0 K. The second term suggests the longitudinal susceptibility arising from the gapped part of the Fermi surface described by mean-field theory [23]. If we use the Néel temperature $T_N = 2.0$ mK, the ratio $\Delta/T_N = 1.3$ is calculated, which is close to the minimum value of 1.764 for the two-band model of itinerant antiferromagnetism [23]. A spin-density-wave transition is observed in $Ce(Ru_{1-x}Rh_x)_2Si_2$ due to a partial gap opening in the Fermi surface following the nesting of the Fermi surface [24]. The anisotropic susceptibility in $CeCu_6$ is described with an itinerant antiferromagnetic picture based on the Fermi-surface nesting along the a

axis, in a heavy fermion conduction band. Incidentally the preferred spin alignment along the a axis is in contrast with the fact that the c axis is the easy direction above 1 K, which is due to the crystal field.

Finally, let us discuss the quantum critical phenomena in $CeCu_6$. We observed that the susceptibility leveled off with the increasing fields at 1 and 3.5 mK. Therefore the susceptibility for a 11 mT field at lower temperatures is regarded as the background susceptibility without the influence of the peak. The susceptibility of $\chi \propto -\log T$ along the a axis was observed at 11 mT between 1 and 10 mK as shown by the dashed line in the inset of Fig. 1(a′). This behavior can be regarded as NFL effects. Similar behavior was observed for the b axis between 0.07 and 0.7 K as shown in Fig. 1(b). FL behavior appeared for the b and the c axes with the application of magnetic fields which suppress the magnetic order. These results suggest that $CeCu_6$ stays near the quantum critical point and Fermi-liquid behavior appears with decreasing the amplitude of quantum fluctuations by the magnetic fields.

In conclusion, we obtained clear evidence that $CeCu_6$ orders antiferromagnetically at 2 mK from the measurements of the ac susceptibility in the various fields and the thermal expansion at ultralow temperatures. The magnetic order in $CeCu_6$ occurs by the spin fluctuation.

This work was supported by a Grant-in-Aid for the International Scientific Research Program of the Ministry of Education, Science, Sports, and Culture (MESSC) and one of us (Y. Ōnuki) was financially supported by COE Research (10CE 2004) of MESSC. We acknowledge helpful discussions with T. Matsuura, Y. Kuroda, A. Kobayashi, Y. Miura, T. Matsushita, A. Sawada, M. W. Meisel, N. S. Sullivan, and E. D. Adams and experimental assistance from S. Inoue, W. Itoh, and T. Kurokawa.

[1] G. Lapertot, R. Calemczuk, C. Marcenat, J. Y. Henry, J. X. Boucherle, J. Flouquet, J. Hammann, R. Cibin, J. Cors, D. Jaccard, and J. Sierro, Physica (Amsterdam) **186B–188B**, 454 (1993).

[2] O. Avenel, J. S. Xia, B. Andraka, C. S. Jee, M-F. Xu, Y. J. Qian, T. Lang, P. L. Moyland, W. Ni, P. J. C. Signore, E. D. Adams, G. G. Ihas, M. W. Meisel, G. R. Stewart, N. S. Sullivan, and Y. Takano, Phys. Rev. B **45**, 5695 (1992).

[3] G. Aeppli, H. Yoshizawa, Y. Endoh, E. Bucher, J. Hufnagl, Y. Ōnuki, and T. Komatsubara, Phys. Rev. Lett. **57**, 122 (1986).

[4] J. Rossat-Mignod, L. P. Regnault, J. L. Jacoud, C. Vettier, P. Lejay, J. Flouquet, E. Walker, D. Jaccard, and A. Amato, J. Magn. Magn. Mater. **76 & 77**, 376 (1988).

[5] C. Jin, D. M. Lee, L. Pollack, E. N. Smith, J. T. Markert, M. B. Maple, and D. G. Hinks, Physica (Amsterdam) **194B–196B**, 207 (1994).

[6] L. Pollack, M. J. R. Hoch, C. Jin, E. N. Smith, J. M. Parpia, D. L. Hawthorne, D. A. Geller, D. M. Lee, and R. C. Richardson, Phys. Rev. B **52**, R15 707 (1995).

[7] E. A. Schuberth, J. Schupp, R. Freese, and K. Andres, Phys. Rev. B **51**, 12 892 (1995).

[8] H. G. Schlager, A. Schröder, M. Welsch, and H. v. Löhneysen, J. Low Temp. Phys. **90**, 181 (1993).

[9] A. K. Gangopadhyay, J. S. Schilling, H. D. Yang, and R. N. Shelton, Phys. Rev. B **36**, 4086 (1987).

[10] H. v. Löhneysen, T. Pietrus, G. Portisch, H. G. Schlager, A. Schröder, M. Sieck, and T. Trappmann, Phys. Rev. Lett. **72**, 3262 (1994).

[11] K. Heuser, E.-W. Scheidt, T. Schreiner, and G. R. Stewart, Phys. Rev. B **58**, R15 959 (1998).

[12] See, for instance, *Proceedings of the Conference on Non-Fermi Liquid Behavior in Metals* [J. Phys. Condens. Matter **8**, 9675 (1996)].

[13] A. Schröder, G. Aeppli, E. Bucher, R. Ramazashvili, and P. Coleman, Phys. Rev. Lett. **80**, 5623 (1998).

[14] O. Stockert, H. v. Löhneysen, A. Rosch, N. Pyka, and M. Loewenhaupt, Phys. Rev. Lett. **80**, 5627 (1998).

[15] T. Herrmannsdörfer, H. Uniewski, and F. Pobell, J. Low Temp. Phys. **97**, 189 (1994).

[16] LR 700 bridge, Linear Research Inc., 5231 Cushman Place, Suite 21, San Diego, CA 92119-3910.

[17] G. Oomi, Y. Ōnuki, and T. Komatsubara, J. Phys. Soc. Jpn. **59**, 803 (1990).

[18] Y. Ōnuki, K. Shibutani, T. Hirai, T. Komatsubara, A. Sumiyama, Y. Oda, H. Nagano, H. Sato, and K. Yonemitsu, J. Phys. Soc. Jpn. **54**, 2804 (1985).

[19] A. Sawada, T. Kubo, Y. Fujii, T. Komatsubara, Y. Ōnuki, N. Kimura, E. Yamamoto, and Y. Haga, Czech. J. Phys. **46**, 803 (1996).

[20] T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. **64**, 960 (1995).

[21] A. Amato, R. Feyerherm, F. N. Gygax, D. Jaccard, A. Schenck, J. Sierro, E. Walker, and U. Zimmermann, Physica (Amsterdam) **186B–188B**, 273 (1993).

[22] S. Kambe, J. Flouquet, and T. E. Hargreaves, J. Low Temp. Phys. **108**, 383 (1997).

[23] P. A. Fedders and P. C. Martin, Phys. Rev. **143**, 245 (1966).

[24] S. Murayama, C. Sekine, A. Yokoyanagi, K. Hoshi, and Y. Ōnuki, Phys. Rev. B **56**, 11 092 (1997).