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PHYSICAL REVIEW B **67**, 180407(R) (2003)

ac susceptibility and static magnetization measurements of $CeRu_2Si_2$ at small magnetic fields and ultralow temperatures

D. Takahashi,[1,*] S. Abe,[1] H. Mizuno,[1] D. A. Tayurskii,[1,2] K. Matsumoto,[1] H. Suzuki,[1] and Y. Ōnuki[3]
[1]*Department of Physics, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan*
[2]*Physics Department, Kazan State University, Kremlevskaya str., 18, Kazan, 420008, Russia*
[3]*Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan*
(Received 11 December 2002; published 30 May 2003)

The magnetic properties of $CeRu_2Si_2$ at microkelvin temperatures (down to 170 μK) and ultrasmall magnetic fields ($0.02\sim6.21$ mT) are investigated experimentally. The simultaneously measured ac susceptibility and static magnetization show neither evidence of the magnetic ordering, superconductivity down to the lowest temperatures nor conventional Landau Fermi-Liquid behavior. The results imply the magnetic transition temperature in undoped $CeRu_2Si_2$ is very close to absolute 0 K. The possibility for proximity of $CeRu_2Si_2$ to the quantum critical point without any doping is discussed.

DOI: 10.1103/PhysRevB.67.180407 PACS number(s): 75.30.Cr, 71.10.Hf, 71.27.+a

The unusual properties of heavy fermion (HF) systems are determined by the competition between intersite spin couplings, Ruderman-Kittel-Kasuya-Yosida interaction, and intrasite Kondo interaction.[1] In a system dominated by the Kondo effect, the Pauli paramagnetic (PP) state with massive quasiparticles is achieved through screening of the f electron's magnetic moments by conduction electrons below the characteristic temperature T_K. The physical properties of the HF compounds below T_K are well understood within the framework of the Landau Fermi-liquid (LFL) theory.

Recently, however, non-Fermi-liquid (NFL) behavior was observed in a large class of HF compounds near the quantum critical point (QCP).[2,3] NFL systems exhibit anomalous temperature dependence of the physical quantities in contrast to the LFL theory, such as specific heat $\Delta C/T \propto -\ln T$, resistivity $\Delta\rho \propto T^{\epsilon}(1 \leqslant \epsilon < 2)$, and magnetic susceptibility $\Delta\chi \propto$ either $1-\sqrt{T}$ or $-\ln T$. In general, the quantum (zero-temperature) phase transition is driven by a control parameter other than temperature, for example, composition, pressure, or magnetic field, and is accompanied by a qualitative change in the correlations in the ground state. The second order quantum phase transitions and QCPs in HF systems can be classified into two types. (i) The long-wavelength fluctuations of the order parameter are the only critical degrees of freedom and the quantum criticality is developed as spin-density wave instability,[4,5] here the zero-temperature spin fluctuations are given by the Gaussian fluctuations of the order parameter. (ii) Local critical modes coexist with long-wavelength fluctuations of the order parameter and there is non-Gaussian distribution of the fluctuations.[6] These are the so-called locally critical phase transitions where the quantum criticality of $CeCu_{(6-x)}Au_x$ (Ref. 7) and $YbRh_2Si_2$ (Ref. 8) are regarded as type-(ii) QCP.[6]

$CeRu_2Si_2$ with a $ThCr_2Si_2$-type crystal structure is well known to be a typical HF compound with an electronic specific-heat coefficient $\gamma = 350$ mJ/K^{2}mol below $T_K = 20$ K.[9,10] This compound exhibits the pseudometamagnetic transition into the ferromagnetically ordered state induced by the magnetic field at $H_M = 7.8$ T below 10 K.[11–15] The neutron-scattering measurements note short-range antiferromagnetic (AFM) correlations in $CeRu_2Si_2$ even below T_K. These time-fluctuating correlations are described by different incommensurate wave vectors.[16,17] A μSR experiment shows an ultrasmall static moment of the order of $10^{-3}\mu_B$.[18] It is most remarkable that the alloying compound systems $Ce_{(1-x)}La_xRu_2Si_2$ and $Ce(Ru_{(1-x)}Rh_x)_2Si_2$ show an incommensurate spin density wave (SDW) ground state in a concentration range $x > 0.08$ (Ref. 19) and $0.03 < x < 0.4$ (Ref. 20). This long-range ordering has the form of a sine-wave modulated structure with the short-range correlation of $CeRu_2Si_2$, as described above. At the critical concentrations of $x_c = 0.075$ for La and $x_c = 0.03$ for Rh doping, the SDW transition vanishes but T_K remains a finite temperature. Therefore, these small critical concentrations of La and Rh suggest that $CeRu_2Si_2$ might be located in the vicinity of the type-(i) QCP, and AFM spin fluctuations are expected to play a key role in this magnetic ground state. The C, χ, and ρ measurements show the conventional LFL ground state for $CeRu_2Si_2$ below T_K down to 20 mK.[11] All magnetization and susceptibility measurements have been performed at the magnetic field above 1 T and at low temperatures. In this paper, we report the results of ac susceptibility and static magnetization measurements at microkelvin temperatures (down to 170 μK) and ultrasmall magnetic fields ($0.02 \sim 6.21$ mT). The obtained magnetic field and temperature dependence of the susceptibility and magnetization provide evidence of NFL behavior and allow us to think about the proximity of $CeRu_2Si_2$ to QCP.

The single crystal of $CeRu_2Si_2$ was prepared by a Czochralski pulling method with starting materials Ce (99.99%), Si ($>$99.999%), and Ru (99.99%) and purified by a solid state transport method. The sample size was $11\times4.2\times1.5$ mm^3. The sample was cooled with a copper nuclear demagnetization refrigerator and a ^{3}He-^{4}He dilution refrigerator. It was sandwiched between two silver plates which were parts of the thermal link to the copper nuclear stage. The temperature was measured by a Pt NMR thermometer, a

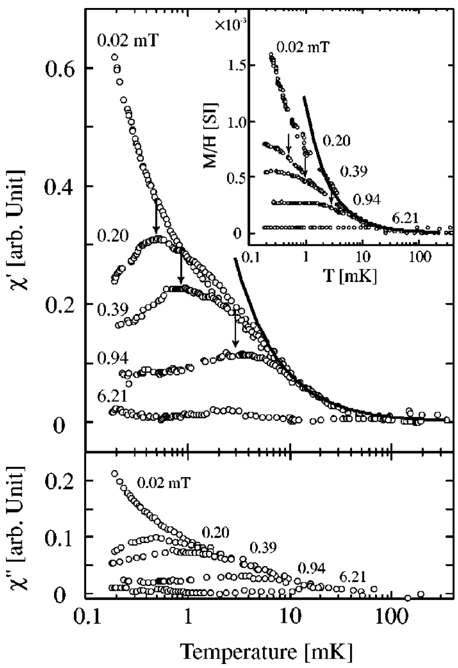

FIG. 1. Temperature dependence (on a logarithmic scale) of the ac susceptibility ($\partial M/\partial H$) at different applied fields as indicated in the figure. The inset shows the static susceptibility (M/H) in the same temperature range. The arrows and solid line indicate the peak temperature observed by ac susceptibility and the Curie law at each figure, respectively.

^{3}He melting curve thermometer,[21] and a RuO_2 resistance thermometer. All these thermometers and the thermal link were attached to the same flange of the nuclear stage. The temperature difference between sample and heat bath was estimated to be less than the order of 0.1% at all temperatures.

The ac susceptibility and static magnetization of $CeRu_2Si_2$ were measured simultaneously in a static field $0.02 \leqslant B \leqslant 6.21$ mT by an ac impedance bridge using a SQUID magnetometer. The applied static field declined a few tens of degrees from the crystalline c axis. All of the ac susceptibility measurements were performed at a frequency of 16 Hz with an excitation field below 0.75 μT parallel to the static field. The primary coil, secondary coil, and static field coil were placed inside a Nb superconducting magnetic shield, which was surrounded by a μ metal magnetic shield to suppress any external stray field. The static magnetization was calibrated against the absolute value measured by another magnetometer in the temperature range from 4 to 2 K.

The ac susceptibility was measured during cooling and warming, and the results showed no appreciable hysteresis. The static magnetization was measured in the warming procedure. Figure 1 shows the temperature dependence of the inphase (χ') and quadrature (χ'') components of the ac susceptibility ($\partial M/\partial H$) at different magnetic fields below 400 mK. The inset of Fig. 1 shows the temperature dependence of the static susceptibility (M/H) derived from the static magnetization. We calibrated all data against the temperature independent PP susceptibility which observed above $\sim$50 mK. Below $\sim$50 mK, we observed an excess

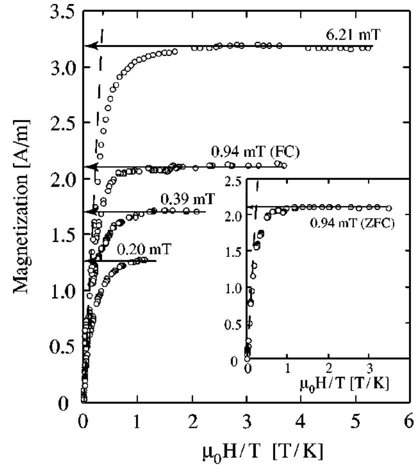

FIG. 2. H/T (H is the applied field) dependence of the static magnetization of $CeRu_2Si_2$ above 0.20 mT at ultralow temperatures. The inset shows the H/T dependence of magnetization at 0.94 mT obtained in the zero field cooling experiment. Solid lines (saturated magnetization) and dashed lines (Curie-law) are guides for the eye.

susceptibility obeying the Curie law.

The ac susceptibility shows a peak at the magnetic field between 0.20 and 0.94 mT. The peak temperatures T_P shift to higher values and the ac susceptibility is suppressed with increase in the applied magnetic field. In particular, the ac susceptibility at 6.21 mT is suppressed almost to the level of the PP susceptibility. The ac and static susceptibilities deviate from the Curie law as they approach T_P. The Curie constant C can be written in the form $C = N_A \mu_0 \mu_p^2 / 3k_B V_{mol}$. The effective magnetic moment μ_p turns out to be 0.020 $\pm 0.003\mu_B$/unit cell from the static susceptibility and is independent of applied magnetic fields. The value of μ_p is in agreement with the ultrasmall static moment observed in the μSR experiment.[18] The static susceptibility, however, becomes flat with no peaks in fields higher than 0.20 mT.

Figure 2 shows the H/T dependence of the static magnetization below 400 mK. The dashed line corresponds to the Curie law with $\mu_p = 0.02\mu_B$. In the fields above 0.20 mT, the magnetization clearly shows the saturation. The saturated magnetic moment μ_s can be evaluated in each field using the following relation: $M_s = N_A \mu_s / V_{mol}$. The calculated μ_s are $1.20\times10^{-5}, 1.60\times10^{-5}, 1.95\times10^{-5}$, and $2.98\times10^{-5}\mu_B$/unit cell, and the ratio of μ_p to μ_s is derived as $1.80\times10^3, 1.15\times10^3, 0.98\times10^3$, and 0.73×10^3 at 0.20, 0.39, 0.94, and 6.21 mT, respectively. Figure 2 suggests that the magnetization cannot be attributed to an impurity effect. At very small concentrations, the magnetic impurities contribution to the total magnetization should behave according to a Brillouin function. With increasing concentration, a locally ordered state like spin-glass can be formed. The impurity effect on the static magnetization in $CeCu_6$ at low temperatures is one example for the first case.[22] It indicates that the ratio of μ_p, as deduced by Curie law, to μ_s has to be of the order of 1. However, this does not agree with our results, $\mu_p/\mu_s \sim 10^3$. In the second case, many compounds with a spin glass transition show quite different magnetization be-

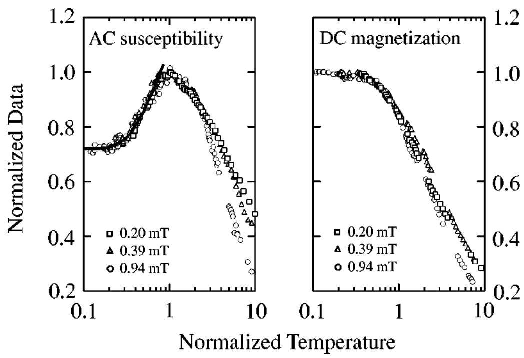

FIG. 3. Scaling behavior of the ac susceptibility and static magnetization in the fields between 0.20 and 0.94 mT. The vertical axes represent the normalized susceptibility and the normalized magnetization. The horizontal axis is the normalized temperature. Details of normalization and the solid line are explained in the text.

havior between zero field cooling (ZFC) and field cooling (FC) measurements through the transition temperature. The inset in Fig. 2 shows the magnetization at 0.94 mT measured by ZFC. The results of ZFC and FC do not indicate different behavior below T_P in this field. Consequently, the possibility of spin glass transition is also strongly denied.

Below T_P, there is unique scaling behavior of the ac susceptibility and static magnetization from 0.20 to 0.94 mT, as shown in Fig. 3. We normalized the susceptibility by its peak height and the magnetization by the saturated value at each field. And the temperature is normalized by T_P. The scaling behavior provides evidence of the proximity of $CeRu_2Si_2$ at small magnetic fields and ultralow temperatures to some phase transition.[23] The nature of such transition is determined by interplaying between ferromagnetic (FM) and AFM fluctuations observed by neutron scattering experiments.[14–17] And this type of interplaying was evidently observed around H_M.[14] In order to shed light on the nature of this transition we analyzed this scaling behavior from the viewpoint of up-to-date theories for ordering in HF compounds.

According to the mean-field theory, the temperature dependence of the susceptibility with the SDW ground state below Néel temperature obeys the following expression: $\chi(T)=\chi_0+B\exp(-a/T)$, where χ_0 and a are the $\chi(T\rightarrow 0)$ and an energy gap, respectively. There, the ratio of the gap energy to T_N should be above 1.76.[24] In our case, the normalized temperature T^* dependence of the normalized ac susceptibility $\chi^*(T^*)$ is obeyed above the expression with $a/T^*\approx 1.0$ at the peak temperature, as shown in Fig. 3. This ratio is in contrast to the SDW state case and there is no indication for the AFM transition at T_P. The scaling behavior and the exponent type temperature dependence of χ^* below T_P, however, suggest that $CeRu_2Si_2$ is in some magnetic field arranged state between 0.20 and 0.94 mT. We speculate that the physical background of this scaling behavior is the quantum critical fluctuation effect of $CeRu_2Si_2$ which is in proximity to the QCP discussed below.

Further, we compare $CeRu_2Si_2$ with $CeCu_6$ which is also a typical HF compound and very similar to $CeRu_2Si_2$ in its $4f$ electron behavior below T_K. Recently, it has been shown that magnetic ground state of $CeCu_6$ is the SDW state and T_N is in fair agreement with the estimated one by the self-consistent renormalization (SCR) spin fluctuation theory.[25] The SCR theory for the HF system predicts the value of T_N as an equation: $T_N=0.1376p_Q^{4/3}T_A^{2/3}T_0^{1/3}$, where p_Q is the staggered spontaneous moment in μ_B at $T=0$ K, T_A, and T_0 are the characteristic temperatures in the $\boldsymbol{q}$ and ω space, respectively.[5] If we use the derived values of $T_A=16$ K, $T_0=14.1$ K (Ref. 26), and $p_Q=7\times10^{-3}\ \mu_B$ (Ref. 18) for $CeRu_2Si_2$, the predicted T_N is estimated as $T_N\sim 2.8$ mK. However, the magnetic properties of $CeRu_2Si_2$ do not show any indication of the magnetic ordering in the smallest applied magnetic field. The estimation of T_N for SDW state in framework of SCR theory takes into account only AFM characteristic wave vectors while our data indicate an existence of both FM and AFM fluctuations. And these two types of fluctuations can lead to some disordered state in the smallest field. This means that the magnetic transition temperature in $CeRu_2Si_2$ is possibly close to $T=0$ K and the spin system of $CeRu_2Si_2$ under our conditions is in the vicinity of the QCP.

In the case of type-(i) QCP, we cannot explain the temperature dependence of the magnetic susceptibility in $CeRu_2Si_2$ only from AFM fluctuations which have been observed in neutron experiments.[16,17] Based on the SCR theory, the uniform susceptibility of an itinerant weak AFM compound should not indicate the Curie-Weiss behavior without taking account of the FM fluctuations.[29] The large value of the μ_p/μ_s also suggests the weak FM properties in this compound.

We consider two known examples of type-(ii) QCP for the $4f$ electron system. The well-known type-(ii) QCP doped compound is $CeCu_{5.9}Au_{0.1}$, which exhibits the critical scaling behavior for the differential susceptibility $(\partial M/\partial H)$ in the form $(\partial M/\partial H)^{-1}=\chi_0^{-1}+CT^{\alpha}g(H/T)$. Here α is the critical exponent and the universal scaling function $g(H/T)$ is given by Schröder *et al.*[7] This scaling function, however, does not lead to the peak for $\partial M/\partial H$; for this reason, it does not explain our results. On the other hand, our results are very similar to the case of undoped $YbRh_2Si_2$, which is classified as a type-(ii) QCP compound. A plateau in the Knight shift below 1 K is observed in $YbRh_2Si_2$, while the ac susceptibility shows a peak at the marginal temperature with the magnetic field dependence.[8,27,28] The Curie-Weiss behavior of magnetic susceptibility $\chi(T)$ for $YbRh_2Si_2$, however, hints to large fluctuating localized Yb^{3+} moments, while that for our system indicates very tiny fluctuating moments with itinerant nature.

We speculate that the magnetic properties of $CeRu_2Si_2$ at small magnetic fields and ultralow temperatures are determined by competition between FM and AFM fluctuations. The narrow range of the applied magnetic fields above 0.20 mT can modulate the FM fluctuations and show a magnetic field arranged state below T_P. The FM fluctuations, however, decrease with increasing magnetic field because the magnetic ground state is recovered nearly to the LFL state at the field above 6.21 mT. In the field of 0.02 mT, the FM and AFM fluctuations compete strongly and show a nonanalytical temperature dependence. This possible scenario of the QCP in our compound shows that $CeRu_2Si_2$ at small magnetic fields and ultralow temperatures should be considered as one of the candidates for investigations of quantum phase transitions at ambient pressure and without any doping. Similar to the type-(ii) QCP compound $YbRh_2Si_2$, the external magnetic field is the control parameter for that transition. The NMR measurements for $CeRu_2Si_2$ under the conditions described above would be very useful in identifying of the nature of the itinerant $4f$ electron system and that QCP.

This work was partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan. D.T. is grateful to the JSPS. The authors are also grateful to H. Tsujii and Y. Tabata for valuable discussions and to K. Mukai, T. Tsunekawa, and K. Nunomura for technical support.

*Present address: Low Temperature Physics Laboratory, Discovery Research Institute, RIKEN (The Institute of Physical and Chemical Research), Hirosawa 2-1, Wako, Saitama 351-0198, Japan.

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