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H/T scaling in disordered non-Fermi liquid materials Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ for $x=0.5$ and 0.6: Quantum Griffiths nature

Yoshikazu Tabata,* Toshifumi Taniguchi, and Yoshihito Miyako
Graduate School of Science, Osaka University, Toyonaka 560-0043, Osaka, Japan

O. Tegus and A. A. Menovsky
Van der Waals-Zeeman Institute, University of Amsterdam, The Netherlands

J. A. Mydosh
Kamerlingh Onnes Laboratory, Leiden University, The Netherlands and Max Planck Institute for Chemical Physics of Solid, D-01187 Dresden, Germany

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Low field ac-susceptibility experiments have been carried out to study the effect of “chemical” disorder and proximity to a magnetic quantum critical point (QCP) on the non-Fermi liquid (NFL) behavior in Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ for $x=0.5$ and 0.6 and CeCu$_{5.9}$Au$_{0.1}$. The susceptibility of strongly disordered NFL material Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ contains two components associated with different mechanisms; a disorder-driven component $\delta\chi$ and a mean-field (MF) quantum critical component $\chi_{\text{MF}}$. $\delta\chi$ exhibits $H/T$-scaling in the form of $T^{-\gamma}(H/T)$ with $\gamma$ depending on $x$. In contrast, the disorder-driven component has not been observed in weakly disordered NFL material CeCu$_{5.9}$Au$_{0.1}$. The results of the scaling analysis strongly suggest that $\delta\chi$ is due to the quantum Griffiths singularity.

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I. INTRODUCTION

Non-Fermi-liquid (NFL) phenomenon in strongly correlated electron systems is a subject of great current interest because it raises fundamental questions about the elementary excitations of these systems with respect to the Fermi-liquid (FL) nature in normal metals. Many heavy fermion materials show the NFL behavior as they approach a magnetic quantum critical point (QCP). These NFL materials have been characterized by different values of the exponents that describe $T$-dependencies of the specific heat, the magnetic susceptibility and the electrical resistivity from those in the FL state. The NFL behavior has been generally explained by the notion of the quantum phase transition at zero temperature or the effect of the “chemical” disorder.

In f-electron systems, two magnetic interactions originating from the hybridization of the localized electron state and the conduction electron state are competing with each other, the Kondo interaction and the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction. The Kondo interaction drives the conduction electrons to screen the localized spins and hence leads to the formation of the nonmagnetic FL state, whereas the RKKY interaction drives the localized spins to order magnetically and hence leads to magnetic ground states. The ratio of both interactions can be continuously tuned by substitution for a constituent element, applying hydrostatic pressure or external magnetic field, and the magnetic-nonmagnetic quantum phase transition is realized. Some theories have predicted anomalous thermodynamic or transport quantities as NFL critical behavior in the vicinity of the QCP, which exists in the pure f-electron systems, for instance CeNi$_2$Ge$_2$ or CePd$_2$Si$_2$ under pressure.

On the other, the origin of the NFL in alloy systems, for instance UCu$_{5-x}$Pd$_x$ (Ref. 6) or Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$, should be more complicated, because chemical disorder due to alloying affects thermodynamic or transport properties in such systems. Miranda et al. predicted the instability of FL caused by a wide distribution of local Kondo temperature $T_K^{\text{local}}$. When the weight of $T_K^{\text{local}}$ at 0 K is finite, a logarithmic divergence of $C(T)/T$ and $\chi(T)$ results. In their model, the inter-site interaction is completely neglected. Castro Neto et al. suggested the similarity between the disordered Kondo lattice system and the Ising spin glass system under transverse field, where the quantum Griffiths singularities is expected in the vicinity of the QCP at 0 K. Thermodynamic quantities diverge down to $T=0$ due to this singularity. The quantum Griffiths model may explain a wide class of the NFL phenomena in Kondo alloy systems.

The pseudobinary alloy system Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ is a good example for studying the two mechanisms for the NFL phenomena. This system exhibits a rich variety of magnetic ground state associated with strong electron correlation as shown in Fig. 1. The base material CeRu$_2$Si$_2$ is a FL heavy fermion compound with no evidence for magnetic ordering, whereas CeRh$_2$Si$_2$ is an antiferromagnetic compound with a Néel temperature $T_N=35$ K. With decreasing $x$ $T_N$ is suppressed, and vanishes for $x=0.6$. Another region of magnetic order in the phase diagram appears in the low Rh-concentration region (0.03 < $x$ < 0.35), where the ordering is a spin density wave (SDW). In the intermediate Rh-concentration region between both magnetic ordered phases, NFL behavior was observed, which is characterized by $C(T)/T=\log T$, $\chi(T)\sim 1-T^{1/3}$, $\rho(T)\sim T^{1.6}$; here the chemical disorder due to high degree of substitution of Ru by Rh coexists with the quantum critical fluctuation in the vicinity of magnetic instability points.

Recently the magnetoresistance of $x=0.5$ in the NFL region was measured, and was found to be well described by...
the dynamical mean-field theory (DMFT) of the spin glass (SG) QCP above 1 T.\textsuperscript{15} The resistivity can be scaled by \((T/\Delta)\) onto one universal curve. \(\Delta\) is a distance from the QCP and a function of temperature \(T\), magnetic field \(H\) and “chemical” distance from the critical concentration \(r\). On the other hand, several features in the lower field region could not be understood by the theory;\textsuperscript{14,15} (i) Divergent behaviors of the susceptibility and the specific heat divided by temperature were observed down to 40 mK. For the quantum spin glass, both quantities approach a finite value at 0 K. (ii) Rapid increase of the magnetoresistance and strong nonlinearity of the magnetization were observed below 1 T. These behaviors may be described by the disorder-driven mechanism, that is, the quantum Griffiths singularity.

This article reports measurements of ac-susceptibilities of the highly disordered NFL materials Ce(Ru\(_{1-x}\)Rh\(_x\))\(_2\)Si\(_2\) for \(x=0.5\) and 0.6 in dc-magnetic field. We investigate the scaling analysis of the susceptibilities and the nonuniversal behavior of the critical exponents for the variance of \(x\), which can be interpreted in terms of the quantum Griffiths effect. For comparison, we also report measurements of the ac-susceptibility of CeCu\(_{5.9}\)Au\(_{0.1}\), known to be a weakly disordered NFL material.

II. EXPERIMENT

Single crystalline samples for \(x=0.5\) and 0.6 were prepared by Czochralski pulling method using a tri-arc furnace. The size of the samples used in the present experiments was about 2\(\times\)2\(\times\)1 mm. A single crystalline sample of CeCu\(_{5.9}\)Au\(_{0.1}\) was grown by the zone melt method. The ac-susceptibility measurements were performed by a conventional mutual inductance method in the temperature range between 40 mK and 2.5 K. The amplitude and the frequency of the ac-field were 2.1 Oe and 130 Hz. We tested the dependence of the susceptibility for Ce(Ru\(_{1-x}\)Rh\(_x\))\(_2\)Si\(_2\) compounds and CeCu\(_{5.9}\)Au\(_{0.1}\) on ac-field amplitude between 0.5 and 5 Oe, and observed no significant ac-field dependence. Significant frequency dependence was also not observed below 500 Hz. Thus, we can conclude that the susceptibility, measured in the above conditions, is equivalent to that at \(H=0\) and \(\omega=0\). In order to track the magnetic field dependence of the susceptibility, we measured it under a dc-magnetic field applied by superconducting magnet.

Ce(Ru\(_{1-x}\)Rh\(_x\))\(_2\)Si\(_2\) has the tetragonal ThCr\(_2\)Si\(_2\)-type crystal structure (space group 14mm), and a strong uniaxial magnetic anisotropy along the \(c\)-axis.\textsuperscript{13} The susceptibility in the \(ab\)-plane for each Rh-concentration is much smaller than that along the \(c\)-axis \((\chi_{ab}/\chi_{c}\sim 1/10)\) and shows very weak temperature dependence above 1 K. CeCu\(_{5.9}\)Au\(_{0.1}\) has the orthorhombic CeCu\(_6\)-structure (space group \(Pmmn\)), and also shows strong uniaxial magnetic anisotropy along the \(c\)-axis.\textsuperscript{16} We neglect the small monoclinic distortion observed below 70 K.\textsuperscript{17} The temperature dependences of the susceptibility along magnetic hard axes in both compounds are very weak, and thus, we measured the susceptibilities only along the magnetic easy axis. The dc-field was also applied along the magnetic easy axis.

III. RESULTS AND ANALYSES

A. Susceptibility of Ce(Ru\(_{1-x}\)Rh\(_x\))\(_2\)Si\(_2\)

We show the results of the ac-susceptibility of Ce(Ru\(_{1-x}\)Rh\(_x\))\(_2\)Si\(_2\) for \(x=0.5\) and 0.6 with zero and finite dc-magnetic field in Fig. 2. An ac-susceptibility under dc-field represents a differential susceptibility \(\chi(T, H)=\partial M/\partial H\) at \(H=H_{dc}\). The susceptibility for each concentration at zero field diverges as \(T\to 0\) K. The divergence of \(\chi(T)\) is easily suppressed by applying an external magnetic field of about 50 Oe, and a broad maximum is found. The field dependence of \(\chi(T, H)\) found in Fig. 2 is too strong to be explained by the
existence of individually fluctuating spins. The suppression of $\chi(T, H)$ by such small fields indicates existence of huge magnetic clusters, which are estimated to be more than 10 $\mu_B$. This indicates the quantum Griffiths nature of these compounds. The fluctuation of the huge clusters at 0 K and zero field causes the divergence of the susceptibilities. The huge clusters can be frozen easily along the direction of $H$ in moderate magnetic field.

In the previous paper, we reported the nondivergent behavior of the susceptibility, $1-T^3/4$, above 1 kOe for $x=0.5$ and its analysis is based on the dynamical mean-field theory (DMFT). The experimental results quantitatively agree with the DMFT predictions. The divergent behavior observed in lower fields, as shown in Fig. 2, should be interpreted by the existence of another component due to the large fluctuating clusters, as argued above. Thus, we assume that the temperature dependencies of the susceptibilities for $x=0.5$ and 0.6 take the form of,

$$\chi(T, H) = \delta\chi(T, H) + \chi_{MF}(T).$$

The first term $\delta\chi(T, H)$ will be a disorder-driven component and expresses the divergent behavior at zero field. The $\delta\chi$ at zero field is written as $cT^{-\gamma}$ and exhibits strong field dependence. The second term $\chi_{MF}(T)$ is a nondivergent MF component and hardly shows any field dependence within the field region in the present works. The solid lines correspond to the best fits of the susceptibilities at zero field for $x=0.5$ and 0.6, with $\gamma=0.71$ and 0.60 and $c=0.013$ and 0.0038 emu/mol, respectively. $\chi_{MF}(T)$ for $x=0.5$, see in Ref. 15, is described by three parameters which are $T_0=20$ K, $H_0=13$ T, and $r=7\times10^{-3}$. $T_0$ and $H_0$ are characteristic temperature and field, respectively, and are proportional to the Kondo temperature $T_K$. $r$ is a chemical distance from the QCP. The $\chi_{MF}$ for $x=0.6$ is obtained from the analysis of the susceptibility at 1 kOe above 1.8 K, where $T_0$ and $H_0$ are fixed to the values of $x=0.5$. The value of $r$ for $x=0.6$ is $4.0\times10^{-3}$, which is smaller than that of $x=0.5$. It means that $x=0.6$ is closer to the QCP than $x=0.5$, which is in agreement with the phase diagram shown in Fig. 1.

$\gamma$, the exponent of the divergent term in the susceptibility, is smaller than 1, and depends on the value of $x$, which corresponds to a variance in the degree of chemical disorder or of a distance from the QCP. In order to understand this fact, it should be noticed that in the quantum Griffiths model the exponent $\lambda$, which describes the divergent behavior in several thermodynamic quantities, also depends on the distance from the QCP. In their notation, the exponent $\gamma$ is given by $1-\lambda$, and $\lambda=0.29$ and 0.40 for $x=0.5$ and 0.6 are obtained, respectively.

**B. Comparison study: Susceptibility of CeCu$_{5.9}$Au$_{0.1}$**

For comparison, we have measured the low field ac-susceptibility of the weakly disordered NFL material, CeCu$_{5.9}$Au$_{0.1}$ with the same experimental condition as those for Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$. The results are shown in Fig. 3(a). In contrast to the case of Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$, the susceptibility of CeCu$_{5.9}$Au$_{0.1}$ shows nondivergent behavior even at zero field as well as in the higher field measurements. This can be clearly seen in the log($-Td\chi/dT$) vs log $T$ plot, where the slope corresponds to an exponent of a leading term of the susceptibility. In Figs. 3(b) and 3(c), the plots for Ce(Ru$_{0.5}$Rh$_{0.5}$)$_2$Si$_2$ and CeCu$_{5.9}$Au$_{0.1}$ are shown, respectively. It is clearly found that the plot of Ce(Ru$_{0.5}$Rh$_{0.5}$)$_2$Si$_2$ shows a negative slope, corresponding to a negative exponent, and hence, the susceptibility diverges down to 0 K. On the other hand, a positive slope is found in the plot of CeCu$_{5.9}$Au$_{0.1}$, which indicates that the susceptibility in CeCu$_{5.9}$Au$_{0.1}$ is non-divergent.

The susceptibility in CeCu$_{5.9}$Au$_{0.1}$ at zero field can be well described by a functional dependence $\chi_0(1-aT^\alpha)$ with $\alpha=0.36$, which is represented by the solid line in Fig. 3(a). It is similar to the functional dependence $1-T^{aT}$ predicted by Hatatani et al. on the basis of the mode-mode coupling theory of the AF spin fluctuation with a magnetic propagation vector $Q=G$; $G$ being a reciprocal lattice vector. They pointed out that AF fluctuations with $Q=G$ also lead a $-log T$-dependence of $C/T$. A different approach to analyze the dynamical and the static susceptibilities of CeCu$_{5.9}$Au$_{0.1}$ resulted from the point of view of the strong coupling theory. Here the functional dependence of the static susceptibility at zero field is $C/(\theta^* + T^\alpha)$ with $\alpha' \neq 1$. The best fit of this function to the data at zero field is represented by the dashed line in Fig. 3(a), where $\alpha'=0.63$, which is smaller than 0.75 obtained from the data at 1 kOe. Both functions can describe the experimental data in the temperature region between 40 mK and 2.0 K.
C. Scaling analysis of $\delta \chi$

In Sec. III A, we have argued that the divergent behavior of the susceptibility in Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ could be due to the quantum Griffiths singularity. Here, we show the result of the scaling analysis of the susceptibility. As previously argued, we can conclude that the susceptibility in Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ has two components; the disorder-driven component $\delta \chi(T,H)$ and the MF component $\chi_{MF}(T)$. By considering the fact that $\delta \chi$ diverges down to 40 mK in zero field, we assume the following scaling form:

$$\delta \chi(T,H) = c T^{-\gamma} f\left(\frac{H}{T^\delta}\right).$$

(2)

Little freedom is left in the choice of parameters in order to make all of data collapse onto a single curve. In Fig. 5(a), the scaling plots of $\delta \chi$’s for $x=0.5$ and 0.6 are shown, where the scaled susceptibilities $\delta \chi(T,H)/c T^{-\gamma}$ are plotted vs the reduced field $h = H/T^\delta$ for $T < 1.4$ K. In the scaling plots, the values of $\gamma$ for $x=0.5$ and 0.6 are taken to be 0.71 and 0.60, respectively, which are the same values of the exponents in the divergent terms of the susceptibilities at zero field. The $c$-coefficients are also assigned the values obtained from the analysis at zero field. In order to see how the quality of the scaling plot varies with $\delta$, we have made a histogram of $\ln(f)$ with a stepsize of 0.1 on a $\ln(h) = \ln(H/T^\delta)$ scale between $\sim 3.5$ and $-10$ for each concentration and have calculated the mean square (log) deviation $\sigma_{ln(f)}$ from the mean step values as a function of $\delta$, as shown in Figs. 5(b) and 5(c). The values of $\sigma_{ln(f)}$ for $x=0.5$ and 0.6 are minimal for $\delta = 1.01 \pm 0.06$ and $0.96 \pm 0.08$, respectively. Thus, we can conclude that the values of $\delta$ for both concentrations are 1 within margin of errors. In Fig. 5(a), the values of $\delta$ are fixed to be 1. We found that all of the data for $x=0.5$ and 0.6 successfully collapse onto a single scaling curve.

The solid line in Fig. 5(a) corresponds to a scaling function

$$f(h) = \frac{1}{(1 + h^\epsilon)^2}$$

(3)

with $\epsilon=1.5$. Although Eq. (3) lacks theoretical foundation, nevertheless, it describes the experimental data very well. The scaling function $f(h)$ approaches 1 for $h \rightarrow 0$, which represents that the $\delta \chi$ asymptotes to the divergent behavior $c T^{-\gamma}$ as $H \rightarrow 0$. $f(h)$ also asymptotes to $h^{-1.5}$ as $h \rightarrow \infty$, which rep-
resolves \( \delta \chi(T, H) \to cH^{-1.5}T^{1.5-\gamma} \) as \( T \to 0 \). The values of \( \gamma \) are smaller than 1.5, thus, \( \delta \chi(T, H) \)’s in finite field vanish at 0 K.

Note that the values of \( \delta \) for \( x=0.5 \) and 0.6 are fixed to be 1. In the quantum Griffiths model\(^{22}\) the magnetization has a scaling form

\[
M(H, T) = \frac{H}{T^{3-\delta}8} \left( \frac{H}{T} \right) \tag{4}
\]

which leads the scaling form of the susceptibility as in Eq. (2) with \( \delta=1 \), where \( f(h) = g(h) + hg'(h) \). Thus, the fact of \( \delta=1 \) offers strong evidence that \( \delta \chi(T, H) \) originates from the quantum Griffiths singularity.

IV. DISCUSSION

In the last section, we argued that the susceptibility of the disordered NFL material \( \text{Ce(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) contains two components, the MF quantum critical components \( \chi_{\text{MF}} \) and the disorder-driven components \( \delta \chi \), which could originate from the quantum Griffiths singularity. \( \delta \chi \) has strong field dependence, and is distinguished from \( \chi_{\text{MF}} \) by applying a very small magnetic field.

It is not a trivial question why the susceptibility of \( \text{Ce(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) can be separated into two different components. Ohashi and Suga predicted the possibility of a “non-trivial” distribution function of \( T_{K_{\text{local}}} \) in a two-dimensional disordered electron system based upon the quantum Monte Carlo method.\(^{24}\) They pointed out that the distribution function \( P(T_{K_{\text{local}}}) \) has a broad maximum in a higher \( T_{K_{\text{local}}} \) region, and separately, has a finite weight at \( T_{K_{\text{local}}}=0 \). In a three-dimensional system, a similar distribution function was obtained.\(^{25}\) Their calculation was done in a finite-size system. At the thermodynamic limit, the system may have a continuous distribution with large density at \( T_{K_{\text{local}}}=0 \), as shown in Fig. 6.

MacLaughlin et al. analyzed the magnetic susceptibility and the \( \mu \text{SR} \) spectra data for \( x=0.5 \) in a high (\( > 2 \) K) temperature region within the framework of the quantum Griffiths model and the Kondo-disorder model.\(^{21}\) The distribution functions \( P(\Delta) \)’s they obtained on a basis of both models have no weight at \( \Delta=0 \); \( \Delta \) is the tunneling energy associated with a magnetic cluster in the quantum Griffiths model, whereas \( \Delta \) is \( T_{K_{\text{local}}} \) in the Kondo-disorder model. Their results indicate the nonsingular properties of the compound at \( T=0 \). However, singular properties were found at lower temperatures, as presented in this article. Taking account of our experimental results, \( \text{Ce(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) is considered to have a distribution of \( T_{K_{\text{local}}} \) with a “tail” in a low \( T_{K_{\text{local}}} \)-region, as shown in Fig. 6. \( P(T_{K_{\text{local}}}) \) obtained in Ref. 19 corresponds to a broad distribution in a high \( T_{K_{\text{local}}} \)-region. Here the tail of \( P(T_{K_{\text{local}}}) \) was left out at lower temperature. It is noted that the mean-value of \( T_{K_{\text{local}}} \) they obtained (\( \sim 30 \) K) is compatible with the value of \( T_{\text{dil}}(=20 \) K), which represent \( T_{K_{\text{local}}} \) in the model.\(^{19,20}\) obtained from the MF analysis as given in Ref. 15. Consequently, we can speculate that Ce-spins in \( \text{Ce(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) for \( x=0.5 \) and 0.6 are distinguished into two groups, namely, Ce-spins whose \( T_{K_{\text{local}}} \) belonging to the broad distribution in a high \( T_{K_{\text{local}}} \)-region and Ce-spins whose \( T_{K_{\text{local}}} \) belonging to the tail of \( P(T_{K_{\text{local}}}) \) in a low \( T_{K_{\text{local}}} \)-region, represented by a shaded portion in Fig. 6. The Ce-spins in the former group couple with the spins of the conduction electrons through the Kondo interaction of order of 20 K and couple with each other through the RKKY interaction, and give the \( \lambda_{\text{MF}} \) in the susceptibility. On the other hand, the Ce-spins in the latter group form magnetic clusters because their \( T_{K_{\text{local}}} \) is almost 0 K, and give the \( \delta \chi \).

The field dependence of \( \delta \chi(T, H) \) can be described by the scaling form derived in Sec. III C. The results of the scaling analysis strongly suggests that \( \delta \chi(T, H) \) is due to the quantum Griffiths singularity, namely, (i) the exponent \( \gamma \) depends on the value of \( x \), and (ii) the exponent \( \delta \) is 1.0. And also, we found the universal scaling function of \( f(h) \), which is not affected by the value of \( x \) within the margin of error. Castro Neto and Jones predicted the scaling form of the magnetization in the quantum Griffiths regime\(^{22}\) as Eq. (4), which leads to the \( H/T \)-scaling of the susceptibility. According to the arguments in Ref. 22, the scaling function of the magnetization \( g(h) \), and also \( f(h) \), depend on the value of \( \lambda \) in contradiction to the results of our scaling analysis. They derived that \( g(h) \) and \( f(h) \) asymptote to a constant value for \( h \to 0 \) and to \( h^{-1+\lambda} \) for \( h \to \infty \). We found in Fig. 5(a) that the experimental \( f(h) \) extrapolates to 1 for \( h \to 0 \) and to \( h^{-1.5} \) for \( h \to \infty \), which is \( \gamma \)-independent, that is, \( \lambda \)-independent. Hence, the asymptotic behaviors of the experimental and the theoretical \( f(h) \) for \( h \to \infty \) are inconsistent with each other. In a higher \( h \) region, i.e., in a lower temperature region, \( f(h) \)’s for \( x=0.5 \) and 0.6 could deviate from each other and may show \( \lambda \)-dependent behavior.

V. CONCLUSION

We have measured the low field ac-susceptibilities of the strongly disordered and the weakly disordered NFL materials, \( \text{Ce(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) and \( \text{CeCu}_2\text{Si}_2 \), in dc-magnetic fields. The susceptibilities of \( \text{Ce(Ru}_{1-x}\text{Rh}_x)\text{Si}_2 \) contain two components; the disorder-driven component \( \delta \chi \) and the MF quantum critical component \( \chi_{\text{MF}} \). The disorder component has not been observed in the weakly disordered materials.
CeCu$_2$Au$_{0.1}$. In Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ $\delta \chi$ diverges down to 0 K with the $x$-dependent exponent $\gamma$ at zero field, and is suppressed by small magnetic field. $\delta \chi$ also exhibits the $H/T$-scaling. These results can be well interpreted in terms of the quantum Griffiths model, however the universal scaling curve $f(h)$ obtained from the scaling analysis of the present experimental data disagrees with the $\lambda$-dependent scaling curve predicted by the model.

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9 Electronic address: yang@ltfridge.ess.sci.osaka-u.ac.jp


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