Quantum Critical Point of Itinerant Antiferromagnet in the Heavy Fermion Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$

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Abstract. A focus of recent experimental and theoretical studies on heavy fermion systems close to antiferromagnetic (AFM) quantum critical points (QCP) is directed toward revealing the nature of the fixed point, i.e., whether it is an itinerant antiferromagnet [spin density wave (SDW)] type or a locally-critical fixed point. The relevance of the local QCP was proposed to explain the $E/T$ scaling with an anomalous exponent observed for the AFM QCP of CeCu$_{5.9}$Au$_{0.1}$. In this work, we have investigated an AFM QCP of another archetypal heavy fermion system Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ with $x = 0$ and 0.03 ($-x_c$) using single-crystalline neutron scattering. Accurate measurements of the dynamical susceptibility $\text{Im} \chi(T, E)$ at the AFM wave vector $Q = 0.35c^*$ have shown that $\text{Im} \chi(T, E)$ is well described by a Lorentzian and its energy width $\Gamma(Q)$, i.e., the inverse correlation time depends on temperature as $\Gamma(Q) = c_1 + c_2 f^{3/2} T_0^4$, where $c_1$ and $c_2$ are $x$ dependent constants, in low temperature ranges. This critical exponent $3/2$ proves that the QCP is controlled by the SDW QCP in three space dimensions studied by the renormalization group and self-consistent renormalization theories.

INTRODUCTION

Quantum critical points (QCP) separating ferromagnetic or antiferromagnetic states from paramagnetic Fermi liquid states in strongly correlated electron systems have been investigated for decades. Successful descriptions of quantum critical behavior were provided by the self-consistent renormalization (SCR) theory of spin fluctuations [1, 2] for $d$-electron systems based on the Hubbard model. The mean-field type approximations in this theory were justified by the renormalization group studies [3, 4] using Hertz’s effective action above upper critical dimensions. For the ferromagnetic QCP, theoretical predictions were supported by experimental studies of $d$-electron systems [1]. In contrast there is little experimental understanding of the antiferromagnetic QCP [2].

A recent intriguing issue of QCP under controversial debate is directed toward revealing relevant fixed points for antiferromagnetic QCPs in heavy-fermion systems [5]. For energy scales much lower than the Kondo temperature $T_K$, $f$ and conduction electrons form composite quasiparticles with a large mass renormalization in paramagnetic heavy fermions. By tuning a certain parameter, e.g., pressure or concentration, an antiferromagnetic long range order emerges from the Fermi liquid state. In a weak coupling picture, it has been hypothesized that the same QCP as the $d$-electron itinerant antiferromagnet, referred to as spin density wave (SDW) type QCP, is relevant to the heavy fermion QCP [4, 5].

However despite a number of experimental studies of heavy-fermion systems showing non-Fermi liquid behavior, none of them definitively supports this QCP [7, 8, 9, 10]. This stems partly from experimental difficulty in measuring weakly divergent quantities around a QCP, especially for bulk properties, which has been also the case for $d$-electron itinerant antiferromagnets [2]. On the contrary, several recent experiments suggest the possibility of a novel strong coupling picture of the QCP [5, 11, 12]. Among these studies, single-crystalline neutron scattering investigations of the heavy fermion CeCu$_{5.9}$Au$_{0.1}$ provided interesting insight [11]. On the basis of the observed $E/T$ scaling with an anomalous exponent [11], and effective two space dimensions [13], a scenario of a locally critical QCP was proposed [5, 14].

In this work, we have studied the antiferromagnetic QCP of another heavy-fermion system Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ ($x = 0$, 0.03) using single-crystalline neutron scattering [15]. Stoichiometric CeRu$_2$Si$_2$ is an archetypal paramagnetic heavy-fermion with enhanced
$C/T \simeq 350 \text{ mJ/K}^2 \text{ mol}$ and $T_K \simeq 24 \text{ K}$. Extensive neutron scattering studies of CeRu$_2$Si$_2$ have shown that spin fluctuations possessing three-dimensional ($d = 3$) character are excellently described by the SCR theory for heavy fermions. A small amount of Rh doping, $x > x_c \simeq 0.04$, induces an antiferromagnetic phase (see the inset of Fig. 3) of the sinusoidally modulated structure with the wave vector $k_3 = 0.35c^t$ [19]. Samples nearly tuned to the lowest concentration QCP ($x \sim x_c$) show non-divergent $C/T$ ($T \to 0$) and $\Delta \rho \propto T^{3/2}$ [15], which are consistent with the SDW QCP in $d = 3$. Thus one can expect that Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ ($x \lesssim x_c$) is suited to investigate the SDW QCP without disorder effects.

**EXPERIMENTS AND ANALYSES**

Neutron-scattering measurements were performed on the triple-axis spectrometer HER at JAERI. It was operated using final energies of $E_F = 3.1$ and 2.4 meV providing energy resolutions of 0.1 and 0.05 meV (full width at half maximum), respectively, at elastic positions. Single crystals with a total weight of 19 g (x = 0) and 17 g (x = 0.03) were grown by the Czochralski method. Two sets of multi-crystal samples aligned together were mounted in a He flow cryostat so as to measure a $(h0l)$ scattering plane. All the data shown are converted to the dynamical susceptibility $\text{Im} \chi(q,E)$. It is scaled to absolute units by comparison with the intensity of a standard vanadium sample. We note that a new point of the present work is unprecedented experimental accuracy in determining the critical exponent using large samples and long counting time. This has enabled us to determine the singularity of the QCP and to make qualitative conclusions of the universality class, which was very difficult in the pioneering work using the related compound Ce$_{1-x}$La$_x$Ru$_2$Si$_2$ [6,9].

**Previous Study of CeRu$_2$Si$_2$**

Let us first make brief comments on our previous neutron scattering study of CeRu$_2$Si$_2$ [17]. We showed that spin fluctuations of CeRu$_2$Si$_2$ are reasonably well described by the SCR theory for heavy fermions [6]. This result has the following two implications in connection with the SDW QCP. First, the spin fluctuations at low temperature $T = 1.5 \text{ K}$ can be parametrized by the SCR form [6]

$$\chi(q,E)^{-1} = \chi_L(E)^{-1} - J(q).$$

This equation means that the local dynamical susceptibility $\chi_L(E) = \chi_L/(1 - iE/\Gamma_L)$, expressing the local quantum fluctuation by the Kondo effect, is modulated by the intersite exchange interactions $J_{r,r} = \sum_{r \neq 0} J_{r,0} \exp(iq \cdot r)$. A number of constant-$Q$ and -$E$ scan spectra can be reproduced by Eq. (1) with the adjustable parameters of $\chi_L$, $\Gamma_L$, and 14 exchange parameters [17]. In Fig. 1 we show observed and calculated intensity maps of constant-$E$ scans with $E = 1 \text{ meV}$ at $T = 1.5 \text{ K}$. One can see that the calculated intensity reproduces the complicated antiferromagnetic spin fluctuations with the three peaks and weaker structures around the Z and N points.

Second, the temperature dependence of the spin fluctuations of CeRu$_2$Si$_2$ [17] can be approximately described by the SCR scenario [6], in which the $T$ dependence of Eq. (1) is brought about by the single $T$ dependent parameter $\chi_L(T)$. The temperature dependence of $\chi_L(T)$ is determined by the self-consistent relation [6]. The existence of this single $T$ dependent parameter indicates that the underlying mechanism is controlled by a neighboring SDW QCP. In fact, the numerically calculated $\chi_L(T)$, showing $\chi_L(T) \propto \text{const} - T^{3/2}$ [17] in a low temperature range, agrees with the computation [Eq. (2)] by the renormalization group theory of the SDW QCP [21]. Along this line, the purpose of this work is to check whether the $T^{3/2}$ dependence of $\chi_L(T)$ really occurs in CeRu$_2$Si$_2$ and in the nearly tuned sample Ce(Ru$_{0.97}$Rh$_{0.03}$)$_2$Si$_2$.

**QCP of Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$**

In the scenario of the SDW QCP in $d = 3$, which was well established by the renormalization group theory [6,4,21], the wave-vector dependent susceptibility for the tuned sample ($x = x_c$) diverges as $\chi(k_3) \propto T^{-3/2}$. 

![FIGURE 1. Observed (a) and calculated (b) intensity maps of constant-$E$ scans taken with $E = 1 \text{ meV}$ at $T = 1.5 \text{ K}$ for the sample with $x = 0$ [17]. They are shown on the surface of the irreducible Brillouin zone.](image-url)
or the characteristic energy of spin fluctuation, i.e., the inverse correlation time, depends on temperature as \( \Gamma(k_3) \propto \chi(k_3)^{-1} \propto T^{3/2} \). By taking the detuning effect \((x < x_0)\) into account, the leading two terms of \( \Gamma(k_3) \) computed by the renormalization group theory \(^2\) are given by

\[
\Gamma(k_3) = c_1 + c_2 T^{3/2},
\]

where \(c_1\) and \(c_2\) are \(T\) dependent constants. This equation is an approximation in the temperature range \(T_{FL} \ll T \ll T_K\), where \(T_{FL}\) is a crossover temperature below which the system shows the Fermi liquid behavior \(^2\).

The imaginary part of the dynamical susceptibility at \(Q = k_3 + q\) with small \(|q|\) and \(|E|\) is approximated \(^2\) by

\[
\text{Im}\chi(k_3 + q, E) = \frac{\chi(k_3) \Gamma(k_3) E}{E^2 + \Gamma(k_3 + q)^2},
\]

\[\Gamma(k_3 + q) = D_i \left[ \kappa_3^2 + q_3^2 + F(q_0^2 + q_3^2) \right] \] (3b)

[a quadratic expansion of Eq. \(^1\)] where \(D_i\) and \(F\) are \(T\) independent parameters, and \(\kappa_3\) is the inverse correlation length along the \(c\)-axis. In Eq. \(^3\) the product \(\chi(k_3) \Gamma(k_3)\) is theoretically \(T\) independent. The two parameters \(D_i\) and \(F\) were determined by using constant-\(Q\) and -\(E\) scans for both samples with \(x = 0\) \(^1\) and \(0.03\) at \(T = 1.5\) K. These data were fit to Eqs. \(^3\) convolved with the resolution functions. In Fig. \(^2\) we show constant-\(E\) scans through the antiferromagnetic wave vector \(Q = (101) - k_3\), and the fit curves for the sample with \(x = 0.03\). The good quality of fitting indicates that Eqs. \(^3\) well describe the experimental data at \(T = 1.5\) K. We obtained \(x\) independent values of the parameters \(D_i = 98 \pm 4\) (meV Å\(^2\)) and \(F = 0.12 \pm 0.01\).

The temperature dependence of \(\Gamma(k_3) = D_i \kappa_3^2\) for both samples has been determined by performing constant-\(Q\) scans taken at \(Q = (101) - k_3\). These scan data were fit to Eqs. \(^3\) convolved with the resolution functions, where there are two adjustable parameters \(\Gamma(k_3)\) and \(\chi(k_3)\). Several fit results of the constant-\(Q\) scans for \(x = 0.03\) are shown in Fig. \(^3\) where one can see that the quality of fitting is excellent. We also checked the \(T\) independence of the parameters \(D_i\) and \(F\) by comparing the constant-\(E\) scans in Fig. \(^2\) at \(T = 4\) and \(8\) K with those calculated using the \(T\) dependent \(\Gamma(k_3)\) and \(\chi(k_3)\) determined by the constant-\(Q\) scans. The calculated curves in Fig. \(^2\) agree reasonably well with the observations. Thus we conclude that the theoretical approximation of Eqs. \(^3\) has been experimentally confirmed, and that the fit parameter \(\Gamma(k_3)\) has been determined very precisely.

The temperature dependence of \(\Gamma(k_3)\) is shown in Fig. \(^4\) by plotting data as a function of \(T^{3/2}\). At low temperatures the observed data clearly agree with the linear behavior of Eq. \(^4\). In fact, by least squares fitting we obtained: \(\Gamma(k_3) = (0.67 \pm 0.01) + (0.0095 \pm 0.0021)T^{1.53 \pm 0.08}\) (in units of meV) in the range \(1.5 < T < 16\) K for the sample with \(x = 0\), and \(\Gamma(k_3) = (0.129 \pm 0.007) + (0.020 \pm 0.003)T^{1.49 \pm 0.07}\) in the range \(1.5 < T < 8\) K for \(x = 0.03\). Therefore we conclude that the observed critical exponent of \(3/2 \pm 0.1\) is in agreement with the theoretical value of \(3/2\) and, consequently,
that the temperature dependence of spin fluctuation is controlled by the SDW QCP in $d = 3$. The same exponent for both $x = 0$ and 0.03 samples ensures that randomness due to Rh doping does not affect the criticality.

**DISCUSSION AND CONCLUSION**

The constant $c_1$ in Eq. (2) is proportional to the theoretical tuning parameter and, hence, $c_1 \propto x_c - x$ is normally assumed [4]. This assumption is consistent with the observed values of $c_1$ and the critical concentration $x_c = 0.04 \pm 0.005$. On the other hand, the constant $c_2$ is theoretically assumed to be weakly dependent on $x$ [4]. However, we observed appreciable $x$ dependence $c_2(x = 0.03)/c_2(x = 0) \sim 2$, which may suggest certain unknown perturbations. Despite this problem, we think that the critical exponent of 3/2, which is determined solely by basic characteristics of the system (the space dimension $d = 3$ and the dynamical exponent $z = 2$), is more important and decisive to conclude the nature of the QCP.

An advantage of the present neutron scattering study is that Eq. (2) holds in a wider temperature range compared to those of indirect measurements of bulk properties. For example, the leading terms of the specific heat $C/T = \gamma_0 - \alpha T^{1/2}$ were shown to have too narrow $T$ ranges to be clearly observed [6,4]. In Fig. 3 the dashed line reproduces the SCR computation of $\Gamma(k_3)$ for CeRu$_2$Si$_2$ [17]. Apart from discrepancy of the coefficient $c_2$, one can see that the $T^{3/2}$ dependence of Eq. (2) is a good approximation for the SCR curve in the $T$ range $2.5 < T < 13$ K ($4 < T^{3/2} < 47$).

In connection with the neutron scattering study of CeCu$_{5.9}$Au$_{0.1}$ [14], it was theoretically predicted [14] that the locally critical QCP is relevant for the two-dimensional spin fluctuation. This theory also predicted that the SDW QCP is relevant for the three-dimensional spin fluctuation, being in accord with the present results. Finally we note that, to our knowledge, the present work is the first clear experimental verification of the SDW QCP among single-crystalline neutron scattering studies on the QCPs of heavy fermions and d-electron systems. Assuming that criticalities of QCPs are classified into a limited number of universality classes, we expect that the SDW QCP remains to be observed in other systems.

In summary, we have demonstrated that the quantum critical behavior of the heavy fermion Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ is controlled by the SDW type QCP in three space dimensions. The inverse correlation time, i.e., energy width $\Gamma(k_3)$ of the dynamical susceptibility, shows a $T^{3/2}$ dependence predicted by the renormalization group and SCR theories.

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