Spin Fluctuation in Heavy Fermion CeRu$_2$Si$_2$

Hiroaki Kadowaki, Masugu Sato, and Shuzo Kawarazaki

1Department of Physics, Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-0397, Japan
2MSD, JASRI, 1-1-1 Kouto Mikazuki-cho Sayo-gun, Hyogo 679-5198, Japan
3Department of Earth and Space Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

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Spin fluctuations of the archetypal heavy-fermion compound CeRu$_2$Si$_2$ have been investigated by neutron scattering in an entire irreducible Brillouin zone. The dynamical susceptibility is remarkably well described by the self-consistent renormalization (SCR) theory of the spin fluctuation in a phenomenological way, proving the effectiveness of the theory. The present analysis using the SCR phenomenology has allowed us to determine fourteen exchange constants, which show the long-range nature of the Ruderman-Kittel-Kasuya-Yosida interaction.

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Effects of the strong correlation of $d$- and $f$-electron systems are exhibited in dual aspects of localized and itinerant characters [1, 2, 3]. In heavy-fermion systems, observations with energies larger than a small scale, Kondo temperature $T_K$, show local-moment behavior, such as the Curie-Weiss susceptibility, and antiferromagnetic correlations [3]. While at lower energy scales $f$ and conduction electrons form composite quasiparticles with a large mass renormalization $m^*/m \propto C/T$ by a factor of up to a few thousands. This large effective mass has been ascribed to the local Kondo effect and to nearness to a quantum critical point (QCP) at $T = 0$, which separates the heavy Fermi-liquid (FL) state from an antiferromagnetic phase of local moments interacting with Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interactions. Theoretical treatment of the both localized and itinerant characters has been a difficult and central issue for heavy fermions [3]. In fact, a focus of recent experimental [3] and theoretical [4] studies on systems close to QCP, or non-Fermi-liquid behavior, is directed toward unveiling the nature of the fixed point, i.e., whether it is a spin-density-wave type [7, 8, 9] or a locally critical quantum phase transition [6, 10, 11].

Attempts to dealing with spin fluctuations in heavy fermions including the both localized and itinerant characters in a wide region close to QCP have been carried out by the self-consistent renormalization (SCR) theory of the spin fluctuation [5]. This extends the SCR theory, well established for weak ferromagnets and antiferromagnets of $d$-electron systems [1, 2], to $f$-electron heavy-fermions using the same form of the dynamical susceptibility $\chi(q, E)$ in the phenomenological way [2, 5]. Experimental results of bulk properties, such as temperature dependence of $C/T$, have been successfully analyzed using the SCR theory for many heavy fermions especially for paramagnetic states [8, 12]. However, an experimental test [13, 14] of the SCR theory using the archetypal heavy-fermion CeRu$_2$Si$_2$ [15, 16] by means of neutron scattering and bulk quantities showed that measured $\chi(q, E)$ is only semi-quantitatively consistent with $C/T$, posing a question about the phenomenology. Extending this work, the present study is aimed at performing a more rigorous test of the SCR theory on CeRu$_2$Si$_2$ by comprehensive measurements of neutron scattering. We have shown that the SCR theory describes $\chi(q, E)$ remarkably well, and that it also provides a rewarding method to determine a number of exchange constants.

CeRu$_2$Si$_2$, which crystallizes in a body-centered tetragonal structure [the ThCr$_2$Si$_2$ structure, see Fig. 1(a)], is an archetypal heavy-fermion compound with enhanced $C/T \simeq 350$ mJ/K$^2$ mol [15, 16]. It shows Kondo behavior with $T_K \simeq 24$ K and remains in a paramagnetic state down to the lowest temperature investigated. The local moments with strongly uniaxial anisotropy develop antiferromagnetic correlations with the energy scale of $k_B T_K$ [17, 18]. For $T \ll T_K$ CeRu$_2$Si$_2$ exhibits FL behavior, for example $\rho \propto T^2$ [17], with renormalized heavy quasiparticles, proved by the de Haas-van Alphen (dHvA) effect [19, 20]. An intriguing property of this compound is the metamagnetic behavior under a magnetic field $H_M = 7.7$ T, showing a sharp crossover of magnetic states [19, 20, 21, 22].

Neutron-scattering measurements were performed on the triple-axis spectrometers HER and GPTAS at JAERI. Typical energy resolutions using final energies of $E_f = 3.1$ and 13.7 meV were 0.1 and 1.0 meV (full width at half maximum), respectively, at elastic positions. Four single crystals with a total weight of 19 g were grown by the Czochralski method. They were aligned together and mounted in a He flow cryostat. All the data shown are converted to the dynamical susceptibility and corrected for the magnetic form-factor and the orientation factor. It is scaled to absolute units by comparison with the intensity of a standard vanadium sample.

The dynamical susceptibility $\chi(q, E)$ is assumed, in the SCR theory [5], to be described by

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

where 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 $J_{r,r'}$, and $J(q) = \sum_{r \neq 0} J_{r,0} \exp(iq \cdot r)$. In the standard treatment \cite{3}, Eq. (1) is expanded around an antiferromagnetic wave-vector $Q$, which is appropriate and has been used for weak itinerant antiferromagnets of $d$-electron systems \cite{1,2}. In stead of this expansion, we directly apply Eq. (1) for the present analysis. The necessity of the nonexpansion form was suggested for heavy fermions because of much weaker magnetic correlations peaked at $T = 5 \text{ K}$ is shown on the surface of the irreducible Brillouin zone. Antiferromagnetic spin correlations show three peaks at wave vectors $k_1 = (0.3, 0, 0)$, $k_2 = (0.3, 0.3, 0)$, and $k_3 = (0, 0, 0.35)$. (c) Calculated intensity map using the fit to the SCR theory.

FIG. 1: (a) Constant-$Q$ scans at several wave vectors reduced to the irreducible Brillouin zone, which is illustrated on the right side together with the body-centered tetragonal structure. Curves are calculated by the fit to the SCR theory, i.e., Eqs. (2). (b) An intensity map of constant-$E$ scans taken with $E = 1 \text{ meV}$ at $T = 1.5 \text{ K}$ is shown on the surface of the irreducible Brillouin zone. Antiferromagnetic spin correlations show three peaks at wave vectors $k_1 = (0.3, 0, 0)$, $k_2 = (0.3, 0.3, 0)$, and $k_3 = (0, 0, 0.35)$. (c) Calculated intensity map using the fit to the SCR theory.

TABLE I: Exchange constants $J_{r,0}$ between magnetic moments at $r = xa + yb + zc$ and 0. They are defined by Eq. (1) and determined by the fit of constant-$Q$ and -$E$ scans shown in Fig. 1 to the SCR theory [cf. Eqs. (2)]. A positive constant $J_{r,0}$ represents a ferromagnetic coupling with an exchange energy $-J_{r,0}\sigma_r\sigma_0$ between Ising variables ($\sigma_r = \pm 1$).

| $x$ | $y$ | $z$ | $J_{r,0}$ (K) | $x$ | $y$ | $z$ | $J_{r,0}$ (K) |
|-----|-----|-----|-------------|-----|-----|-----|-------------|
| 0   | 0   | 2   | -0.90       | 3   | 0   | 0   | 0.22        |
| 1   | 0   | 0   | 0.73        | 0   | 0   | 3   | 0.21        |
| 0   | 0   | 1   | 0.66        | 2   | 1   | 1   | -0.12       |
| 0   | 1   | 0   | 0.56        | 1/2 | 1/2 | 1/2 | 1.18        |
| 2   | 0   | 1   | -0.54       | 3/2 | 1/2 | 1/2 | -0.58       |
| 2   | 0   | 0   | 0.47        | 3/2 | 1/2 | 3/2 | -0.39       |
| 1   | 0   | 1   | -0.42       | 3/2 | 3/2 | 3/2 | -0.24       |
higher temperatures: to compare observations with the SCR theory. As discussed in Ref. 8, the most important temperature dependence of $\chi(q, E)$ originates from that of one parameter $\chi_L(T)$, and the temperature dependence of the other parameters can be neglected in a low temperature range of $k_B T \ll \Gamma_L$. Under this assumption the $T$ dependence of $\chi_L(T)$ can be evaluated by solving

$$\sum_q \int_0^{E_c} \frac{dE}{\pi} \left[1 + \frac{2}{\exp(\beta E) - 1}\right] \text{Im}[\chi(q, E)] = \mu^2$$

where $E_c$ and $\mu$ are the cutoff energy and effective moment, respectively. This is a sum rule simply implying that the total magnetic scattering crosssection integrated in $q$ and $E$ spaces is a constant determined by the magnetic moment of the crystal-field ground-doublet.

Several constant-$E$ and -$Q$ scans were carried out at higher temperatures. In Fig. 2 we show constant-$E$ scans along the $\Sigma$ and $\Lambda$ lines (see Fig. 1). Curves predicted by the SCR theory, i.e., Eqs. (2) with $\chi_L(T)$, show agreement with the observation within $30\%$ in a temperature range $T < 40$ K. The constant-$Q$ scans were fitted to the single Lorentzian [Eq. (2a)] with $\chi(q)$ and $\Gamma(q)$ determined at each $q$ and $T$. Examples of this fitting are shown in Fig. 3(c) for $q = k_3$. The resulting $\chi(q)$ and $\Gamma(q)$ measured at $q = k_1$, $k_3$, and the $Z$ point are plotted as a function of temperature in Figs. 3(a) and (b), where the uniform susceptibility is also shown. The predicted curves in these figures are in agreement with the observations within $30\%$ up to $T = 40$ K. From these comparisons shown in Figs. 2 and 3 although there are systematic discrepancies to a certain extent, we conclude that the overall agreement of the temperature dependence of the observed $\text{Im}[\chi(q, E)]$ with the SCR predictions is fairly good in view of the simple assumption.

The specific heat is dominated by spin fluctuations at low temperatures, and has been used as an important quantity in applying the SCR theory 8,12. Hence it is interesting to compare the observed specific heat $C/T$ with the SCR theory 27. The calculated $C/T$ is compared with the observations in Fig. 3(d), which shows agreement within $50\%$. Considering that the theory employs the same method as that for d-electron systems, which is not strictly justified for $f$ electrons, the agreement is fairly good. However, the SCR theory on $C/T$ has to be improved for more quantitative purposes, which should agree with a FL theory of renormalized quasiparticles 14 at $T = 0$.

We have shown that the SCR theory 8 provides a useful description of the spin fluctuations, i.e., $\chi(q, E)$, for the archetypal heavy fermion CeRu$_2$Si$_2$. It seems that this phenomenology will be applicable to a certain class of heavy-fermions. The success for CeRu$_2$Si$_2$ is partly based on the fact that Eq. (1) is a good approximation at low temperatures. In fact, almost the same form of equation was derived by a $1/d$-expansion theory 28 on the periodic Anderson model. At high temperatures the SCR theory proposed the simple assumption that only one parameter $\chi_L(T)$ depends on temperature, and used...
the exact sum rule of Eq. (3). In the limit $T \to \infty$, this recipe gives the correct Curie-Weiss susceptibility $\chi(q) = \mu^2/k_B T - J(q)$. This natural extension may account for why we obtain the acceptable agreement up to 40 K (see Figs. 2 and 3) beyond the original expectation, $T \ll T_L/k_B \simeq 51$ K [23].

The three antiferromagnetic wave-vectors $k_1$, $k_2$, and $k_3$ are determined by $J(q) = \max$ [cf. Eq. (1)]. In the SCR theory $J(q)$ is ascribed mainly to the RKKY mechanism and is weakly temperature dependent. However one also might attribute the wave-vectors to nestings of the quasiparticle bands. According to Ref. [28], the nesting mechanism can be incorporated in Eq. (1) by replacing $J(s(q) + J_Q(q, E))$, where the first term represents exchange interactions involving intermediate states of high-energy excitations (including the RKKY interaction), and the second term involving low-energy excitations of the quasiparticles. This second term possesses the energy scale $k_B T_L$, and consequently would show larger $T$ dependence [28]. We speculate that $J_Q(q, E)$ is smaller than $J(s(q)$ for CeRu$_2$Si$_2$ (in zero magnetic field), because nesting wave-vectors close to $k_1$, $k_2$, and $k_3$ cannot be easily seen in the Fermi surfaces of the band structure [14], and because $T$ dependence of $J(q)$ could be neglected in our analysis. On the other hand, the quasiparticle mechanism supposedly brings about ferromagnetic exchange interactions under magnetic fields close to the metamagnetic crossover [22]. Therefore, to resolve these problems on the quasiparticle contribution, improved SCR theories based on microscopic models [24] are awaited. It will be also interesting to apply them to those in which the quasiparticles play essential roles in spin fluctuations, e.g. UPt$_3$ [31] and CeNi$_2$Ge$_2$ [31].

Singularities of QCPs in itinerant magnets have been thought to be described by the SCR theory [1] or equivalently by the spin-density-wave type fixed point [3]. However a recent study of criticality of a heavy-fermion system CeCu$_{6-x}$Au$_x$ tuned to a QCP [10] caused controversy on a possibility of a locally-critical fixed point [8, 11]. A key observation, supporting this fixed point, is the dependence of high-energy excitations (including the RKKY interaction), and the second term involving low-energy excitations of the quasiparticles. This second term possesses the energy scale $k_B T_L$, and consequently would show larger $T$ dependence [28]. We speculate that $J_Q(q, E)$ is smaller than $J(s(q)$ for CeRu$_2$Si$_2$ (in zero magnetic field), because nesting wave-vectors close to $k_1$, $k_2$, and $k_3$ cannot be easily seen in the Fermi surfaces of the band structure [14], and because $T$ dependence of $J(q)$ could be neglected in our analysis. On the other hand, the quasiparticle mechanism supposedly brings about ferromagnetic exchange interactions under magnetic fields close to the metamagnetic crossover [22]. Therefore, to resolve these problems on the quasiparticle contribution, improved SCR theories based on microscopic models [24] are awaited. It will be also interesting to apply them to those in which the quasiparticles play essential roles in spin fluctuations, e.g. UPt$_3$ [31] and CeNi$_2$Ge$_2$ [31].

A key observation, supporting this fixed point, is the $T/E$ scaling form $\text{Im}[\chi(Q, E)] = T^{-\alpha}g(E/T)$ at the antiferromagnetic wave vector $Q$ [10]. In contrast, the SCR theory predicts the $T^{3/2}$ scaling form $\text{Im}[\chi(Q, E)] = T^{-3/2}g(E/T^{3/2})$, which one can see using $\chi(Q)^{-1} \propto T^{3/2}$ at the QCP [2] and Eqs. (2). For the present SCR theory of CeRu$_2$Si$_2$, located slightly off the QCP, this characteristic $T^{3/2}$ dependence appears approximately as $\Gamma(Q = k_i) \simeq A + BT^{3/2}$ in a low temperature range $0 < T < 10$ K. Hence we speculate that it may be interesting to accurately measure $T$ dependence of $\Gamma(Q)$ to determine whether it shows the SCR $T^{3/2}$-dependence or is closer to $T^{1}$-dependence, which would suggest the $E/T$ scaling.

In conclusion, we have demonstrated that the SCR theory remarkably well describes the spin fluctuations of the archetypal heavy-fermion CeRu$_2$Si$_2$. The analysis using magnetic excitation data covering the entire irreducible Brillouin zone has enabled us to determine the fourteen exchange constants.

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[25] Since Eq. (2a) does not account for an experimental fact that $\Gamma(q) \sim 0$ close to $q \sim 0$, we modify it to ad hoc $\Gamma(q)^{-\alpha} = \chi(q)[c_1 + c_2/(h a^2) + (b k^2) + c_3/(c k^2)]^{1/2}$. This is an interpolation between Eq. (2a) for large $q$ and $\Gamma(q) \propto q$ for small $q$, which is neglected in Ref. 8 for antiferromagnetic cases. Hence the actual fit parameters are $\chi_L$, $c_i$ $(i = 1, 2, 3)$, and $J_{r,0}$. The values of $c_i$ obtained by the fit at $T = 1.5$ K are $c_1 = 3.4 \pm 0.2$ [meV emu/(mol Ce)]$^{-1}$, $c_2 = 1.5 \pm 0.1$ [meV A emu/(mol Ce)]$^{-1}$, and $c_3 = 2.8 \pm 0.4$. The energy scale $\Gamma_L$ is approximated as $\Gamma_L \approx 1/(c_1 \chi_L) = 4.4$ meV. When $J_{r,0}$ in Table II is used in Eq. (2a), a conversion factor 0.58 [K emu/(mol Ce)]$^{-1}$ is multiplied.
[26] The single Lorentzian [cf. Eq. (2a)] is the simplest approximation for $\text{Im}[\chi(Q, E)]$ in a low energy range. We think the quality of fitting using the single Lorentzian shown in Fig. I(a) is good enough for the present purpose. It should be noted that the double Lorentzian form for $\text{Im}[\chi(q, E)]$ used in Ref. 15 has been somewhat overemphasized. In fact, the single Lorentzian provides good quality of fitting [Refs. 14, 21; see Fig. 5(c)].
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