

Spin Density Wave Accompanied by Anisotropic Energy Gap Opening

Yoshihito MIYAKO, Tetsuya TAKEUCHI¹, Toshifumi TANIGUCHI,
Shuzo KAWARAZAKI, Kazuhiro MARUMOTO, Ryuji HAMADA,
Yoshiyuki YAMAMOTO, Miguel OCIO², Patrick PARI² and Jacques HAMMANN²

*Department of Earth and Space Science, Faculty of Science, Osaka University,
Toyonaka 560, Osaka*

¹*Low Temperature Center, Osaka University, Toyonaka 560, Osaka*

²*Service de Physique de l'Etat Condense, C.E.N. Saclay,
Orme des Merisiers, 91191 Gif-sur Yvette Cedex, France*

Experimental results suggesting spin density wave of itinerant heavy electrons are shown in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and possible nesting of hole bands is discussed. In $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$, the 4f localized magnetic moment around and above $0.2 \mu_B$ orders dominantly by means of the magnetic exchange interactions, although it is suggested from the resistivity measurement that a nesting effect exists. The details of the magnetic ordering in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ ($0.05 < x < 0.3$) are discussed.

KEYWORDS: SDW, Nesting, Heavy Fermion, Kondo Lattice, Antiferromagnetic Order

§1. Introduction

Magnetic phase transitions in heavy fermions exhibit a large variety and are an interesting research subject. The low temperature phase is classified into magnetic and non-magnetic states according to the size of the characteristic energies: i) the binding energy of the Kondo screening, ii) the RKKY interaction energy and iii) crystalline field energy.

In addition to above characteristic energies, quadrupolar coupling¹⁾ and the effect of band structure (Nesting effect)²⁾ contribute to the phase transition in some compounds. In the Haldane state, a non-magnetic state is formed by the nearest neighbour coupling between the magnetic atoms with integer spin. However, the role of the RKKY interactions for the formation of non-magnetic ground state in the coherent Kondo state is not known so well.

Flouquet and his collaborators studied the magnetic instability in Ce heavy fermion compounds by applying pressure and magnetic field³⁾. Coqblin *et al.*⁴⁾ studied theoretically the effects of magnetic short range orders on the Kondo state near the magnetic instability.

CeRu_2Si_2 is one of the most interesting heavy fermion compounds, showing Pauli paramagnetic down to 40 mK as derived in neutron scattering experiment⁵⁾. Recently, a tiny magnetic moment $1/10^3 \mu_B$ was found in μSR experiments⁶⁾. The characteristic feature of CeRu_2Si_2 compound is a strong magnetic anisotropy. The susceptibility shows a Curie-Weiss behavior at high temperatures along the *c*-axis but it is almost independent of temperature along the *a*-axis. The magnetization is also strongly anisotropic. This magnetic anisotropy is reflected in the magnetic ordering. $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ ($0.07 < x < 0.3$) and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ ($0.1 < x < 0.3$) show an antiferromagnetic phase transition. The antiferromag-

netic state in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ is a spin density wave (SDW) formed by nesting of hole bands in heavy fermion state. Anisotropic gap opening in the Fermi surface occurs perpendicular to the *c*-axis in the reciprocal lattice space and the resistivity exhibits anisotropic anomaly associated with the SDW transition⁷⁾. In $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$, on the contrary, the anomaly in the resistivity occurs along the *a*-axis at T_N but there is no anomaly along the *c*-axis. In this system, T_K varies largely with *x* and becomes closer to the temperature T_N . As a result, from Kondo screened non-magnetic state, localized magnetic moments begin to appear more rapidly than in the case of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ at Ce sites with La impurities.

Here, we show the magnetic properties of two $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ compounds from the view point of the ordering mechanism.

§2. Magnetic Properties of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$

2.1 T_K and T_N vs. *x* Magnetic Phase Diagram of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$

The magnetic phase diagram of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ is shown in Fig. 1. The SDW, with the magnetic wave vector propagating along the *c*-axis, occurs in the range of about $x = 0.05$ to 0.25 in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ as reported in the preceding paper⁸⁾ and the ordering temperature, T_N , is maximum at $x = 0.15$. The Kondo temperature, T_K , and the volume of the unit cell are almost constant in the concentration range where the SDW appears. Nearly constant T_K supports the theoretical model given by Coqblin *et al.*⁴⁾ who claim that antiferromagnetic correlations stabilize the Kondo state near magnetic instability and enhance the density of states. Here, T_K is estimated from the peak temperature of the Schottky anomaly in the specific heat.

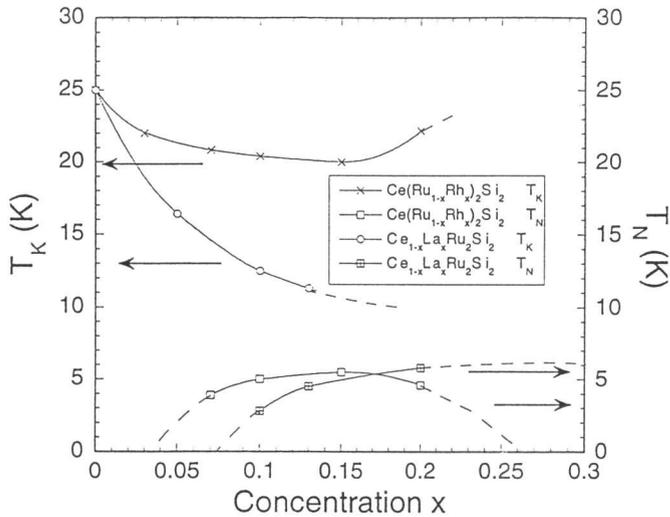


Fig. 1. Magnetic phase diagram of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$.

In pure CeRu_2Si_2 , the magnetic susceptibility has a maximum at about 10 K and decreases slowly below the peak temperature⁹). The C/T of the specific heat is also constant below about 5 K¹⁰). These results indicate Pauli paramagnetic properties. Basically, these Pauli paramagnetic properties with antiferromagnetic correlations are sustained in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ ²), although a SDW ordering sets in below about 5 K. On the other hand, the T_K for $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ varies largely closer to the value of T_N . The antiferromagnetic order changes from sinusoidal to squared modulation with the same magnetic wave vector $\mathbf{q}(0.309,0,0)$ in the similar x range as $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$. For $0.4 < x < 0.9$, the magnetic order is reported, but the details of the properties are not known¹¹).

2.2 Magnetic Ordering of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$

Fig. 2 shows the resistivity around the antiferromagnetic transition temperature for $\text{Ce}(\text{Ru}_{0.9}\text{Rh}_{0.1})_2\text{Si}_2$ and $\text{Ce}_{0.87}\text{La}_{0.13}\text{Ru}_2\text{Si}_2$, respectively. A hump at T_N is observed in the c -axis for $\text{Ce}(\text{Ru}_{0.9}\text{Rh}_{0.1})_2\text{Si}_2$ while there is no anomaly in the a -axis. However, in $\text{Ce}_{0.87}\text{La}_{0.13}\text{Ru}_2\text{Si}_2$, a small increase of the resistivity occurs below T_N along the a -axis and the resistivity along the c -axis decreases smoothly. This different behavior is associated with the magnetic ordered state. Neutron scattering experiment on $\text{Ce}(\text{Ru}_{0.85}\text{Rh}_{0.15})_2\text{Si}_2$ revealed that the antiferromagnetic ordered moments align along the c -axis with the magnetic wave vector $\mathbf{q}(0,0,0.42)$ and are modulated sinusoidally with an amplitude of $0.65 \mu_B$ ²). The sinusoidal modulation of the ordered moment is inferred to be the magnetic ground state from the fact that it remains down to 0.4 K, *i.e.* 1/10 of T_N (5.5 K). The magnetic wave vector \mathbf{q} changes as a function of x as shown in Fig. 3. These results predict the spin density wave (SDW) of itinerant heavy electrons in Kondo screened state. We suggested²) that the dominant driving force of the SDW ordering is the

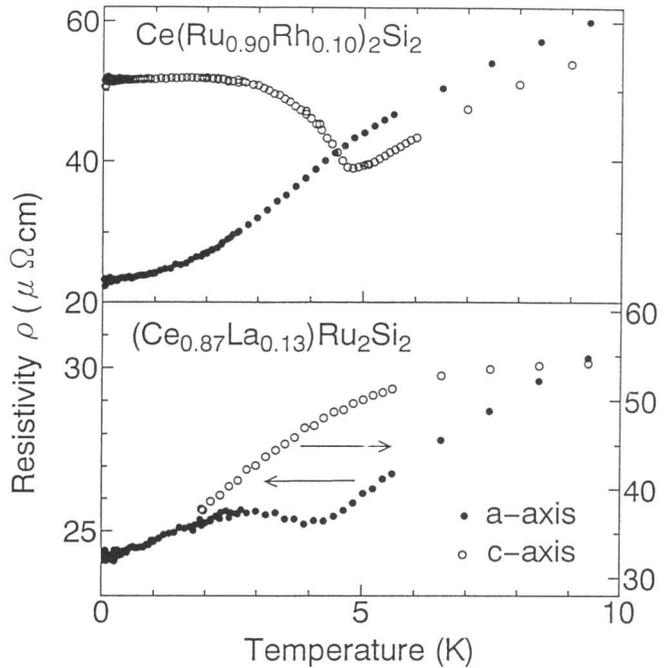


Fig. 2. Anomalous behavior of the resistivity around the antiferromagnetic phase transition temperature in $\text{Ce}(\text{Ru}_{0.9}\text{Rh}_{0.1})_2\text{Si}_2$ and $\text{Ce}_{0.87}\text{La}_{0.13}\text{Ru}_2\text{Si}_2$, respectively.

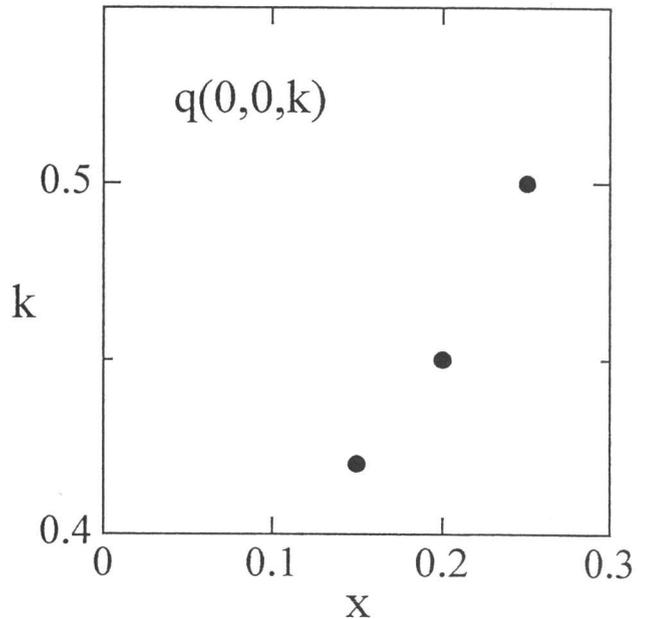


Fig. 3. The magnetic wave vector $\mathbf{q}(0,0,k)$ vs. x in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$. The \mathbf{q} for $x = 0.25$ was measured by Haen *et al.*¹³).

nesting of the hole bands. The Fermi surface of the quasi-particle band calculated by Zwicky¹²) is shown in Fig. 4 (a) and (b). The experiments of neutron scattering and resistivity predict the anisotropic gap opening in the Fermi surface of the quasi-particle bands. However, it is difficult to assign the Fermi surface for nesting with the magnetic wave vector $\mathbf{q}(0,0,0.42)$ for $\text{Ce}(\text{Ru}_{0.85}\text{Rh}_{0.15})_2\text{Si}_2$ on the basis of the band calculation.

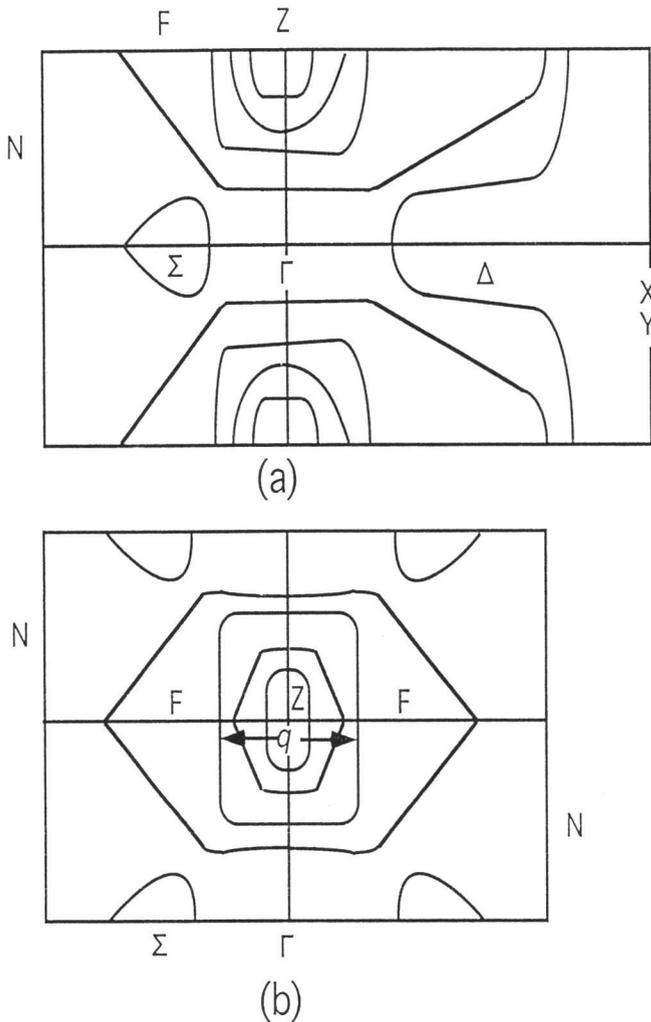


Fig. 4. Intersections of the Fermi surfaces (a) and (b) in the Brillouin zone for CeRu_2Si_2 (After G.Zwicknagl¹²).

On the other hand, in $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ the magnetic short range ordering with magnetic wave vector $q(0.309,0,0)$ as in CeRu_2Si_2 , is stabilized for $0.07 < x < 0.3^4$). In $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$, T_K decreases with x more rapidly than in $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$. The magnetic moment of $\sim 1 \mu_B$ is estimated for $x = 0.13$ from neutron scattering experiments⁴). The magnetic modulation with magnetic wave vector $q(0.309,0,0)$ is sinusoidal for $x = 0.13$, but it shows squaring for $x = 0.20$ with the same magnetic wave vector. From this neutron experiment, it seems that the magnetic order changes from a SDW of itinerant electrons for $x = 0.13$ to an antiferromagnetic state of localized 4f-electrons for $x = 0.2$ and the magnetic wave vector, $q(0.309,0,0)$, of the incommensurate SDW is possibly determined by the nesting as shown in Fig. 4(b). However, μSR experiment in ordered state for $x = 0.13$ suggested that the magnetic order is due to the 4f-localized moment at Ce sites¹⁴). In a SDW state of itinerant electrons, muon spin feels an internal field via hyperfine coupling. Amato estimated the hyperfine field, $A_C = 0.86 \text{ kG}/\mu_B$, in CeRu_2Si_2 ⁵). In our μSR experiment, we could not observe the effect due to the hyperfine field in $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ ($x = 0.05, 0.13$ and 0.25)¹⁴).

The relaxation rate of muon spins is independent of temperature below 100 K in $\text{Ce}_{0.95}\text{La}_{0.05}\text{Ru}_2\text{Si}_2$, when the spins of the incident muons beam are polarized along the c -axis of the crystal, while they slowly decay for spins polarized along the a -axis. This behavior is the reflection of the strong magnetic anisotropy observed in the susceptibility. The magnetic moments order along the c -axis. We calculated the dipolar field at the possible 11 muon sites in a $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ unit cell, taking into account the random distribution of La^{14}). We assumed sinusoidal (for $x = 0.13$) and squared (for $x = 0.25$) modulations with the magnetic wave vector, $q(0.309,0,0)$, for the localized magnetic moment at Ce sites, respectively. The x, y, z -components of the dipolar field, which are parallel to the a, b and c -axis of the crystal, are different in magnitude and distribution. The relaxation of muon spins is explained by the dipolar field from the 4f-localized magnetic moment at Ce sites in the mixed compounds $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ ($x = 0.13$ and $x = 0.25$).

The anisotropic anomaly in the resistivity is related to the anisotropic gap opening in the Fermi surface. In $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$, the magnetic moment drastically increases with x , but the magnetic wave vector, $q(0.309,0,0)$, does not change with x . Therefore, it is considered that magnetic exchange interactions bring into play for the magnetic order when the localized magnetic moment at Ce sites exceeds the critical value, about $0.2 \mu_B$.

2.3 Specific heat, Susceptibility and Magnetization of $\text{Ce}(\text{Ru}_{0.85}\text{Rh}_{0.15})_2\text{Si}_2$ and $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$

The log-log plot of the temperature dependence of the magnetic specific heat, C_{mag} , of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ for $x = 0.1$ and 0.15 is shown in Fig. 5. The specific heat of samples of about 10 mg were measured by a thermal relaxation method. Below the peak of the specific heat at temperature about 4 K, the C_{mag} obeys approximately the power law

$$C_{mag} = AT^n \quad (2.1)$$

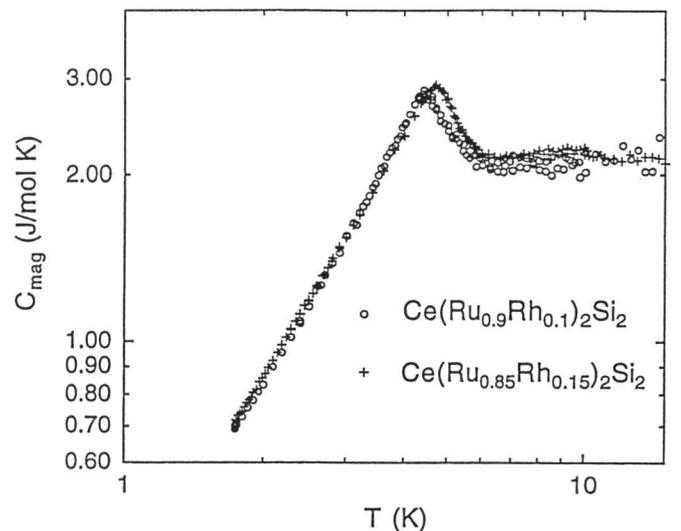


Fig. 5. Log-Log plot of C_{mag} vs. T for $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ ($x = 0.1$ and 0.15).

with $n = 1.5 \sim 2$, rather than having an exponential form. This may be correlated with an anisotropic gap opening at the Fermi surface due to the ordering of the SDW or may be reminiscent of the so-called non Fermi liquid properties often found on the paramagnetic side of the magnetic non-magnetic transition. If we fit the experimental data with the formula, $C/T = \gamma + AT$, we find that $C = 200(\text{mJ}/\text{mole}\cdot\text{K}^2)T + 100(\text{mJ}/\text{mole}\cdot\text{K}^3)T^2$ in the range between the peak temperature of C/T (4.3 K) and 2.3 K. Below 2.3K, A becomes smaller in this fitting. To study the thermal excitations at very low temperatures, more detailed measurements are necessary.

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