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Decoupling between Field-instabilities of Antiferromagnetism and Pseudo-metamagnetism in Rh-doped CeRu₂Si₂ Kondo Lattice

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Doping Kondo lattice system CeRu₂Si₂ with Rh-8% (Ce(Ru_{0.92}Rh_{0.08})₂Si₂) leads to drastic consequences due to the mismatch of the lattice parameters between CeRu₂Si₂ and CeRh₂Si₂. A large variety of experiments clarifies the unusual properties of the ground state induced by the magnetic field from longitudinal antiferromagnetic (AF) mode at $H = 0$ to polarized paramagnetic phase in very high magnetic field. The separation between AF phase, paramagnetic phase and polarized paramagnetic phase varying with temperature, magnetic field and pressure is discussed on the basis of the experiments down to very low temperature. Similarities and differences between Rh and La substituted alloys are discussed with emphasis on the competition between transverse and longitudinal AF modes, and ferromagnetic fluctuations.

KEYWORDS: CeRu₂Si₂, Rh-doping, La-doping, antiferromagnetism, pseudo-metamagnetism, specific heat, resistivity, thermal expansion, magnetization, neutron diffraction, high pressure

1. Introduction

The effect of pressure on Ce heavy fermion systems close to the antiferromagnetic (AF) quantum phase transition is well known. Pressure drives the system from AF to paramagnetic (PM) ground state at a critical pressure P_c . If the transition is of second order with continuous suppression of the sublattice magnetization at P_c , P_c corresponds to a quantum critical point.¹⁾

In complex materials such as heavy fermion compounds, quite different pictures can be obtained for different materials as demonstrated in CeRu₂Si₂, CeCu₆ and YbRh₂Si₂. The first case is often referred as an example of global criticality where critical fluctuations are that of the magnetic order parameter, and two others of local criticality where quantum criticality is driven by local i. e. Q -independent magnetic fluctuations.^{1,2)} We focus here on the CeRu₂Si₂ series. In this tetragonal crystal, the Ce ions show a strong Ising character with an anisotropy of susceptibility between the easy-axis (c -axis) and the hard-axis with a factor of 15 at low temperatures. The effect of the volume change via a tuning parameter δ have been achieved first by pressure or by doping on Ce_{1-x}La_xRu₂Si₂.^{3,4)} The pure system CeRu₂Si₂ at $P = 0$ is already in a PM ground state; the effective critical pressure P_c is at a slightly negative pressure of a few kbar. Expanding the volume by La substitution on the Ce side pushes to reenter in the AF domain for $x > x_c \sim 0.075$; the AF-PM transition at $T = 0$ K corresponds to a critical volume V_c achieved at δ_c which is equivalent to P_c or x_c .^{5,6)}

Applying a magnetic field (H) will lead to cross the AF boundary $H_c(T)$ between AF and PM. For the Ising spin system of the CeRu₂Si₂ series the first order metamagnetic transition at $H_c(0)$ below P_c terminates at the critical endpoint H_c^* .^{1,4,7,8)} The magnetic field can lead to switch from dominant AF interactions at low field to a highly polarized paramagnetic phase (PPM) at high field via a strong interplay between AF and ferromagnetic (FM) coupling through H_c studied with La- and Ge-doped system.^{1,5,9)} Furthermore, due to the large uniform susceptibility χ_0 associated to the huge value of the Sommerfeld coefficient γ (directly linked to the strong local $4f$ character of the heavy fermion quasiparticle via Kondo fluctuation), the growth of the majority spin-up component at H_c is associated to a Fermi surface instability.¹⁰⁻¹³⁾ Roughly at $T = 0$ K, the magnetic polarization is given by the ratio of the induced magnetization ($\chi_0 H$) by the saturated magnetization. It reaches a critical value, $M_c \sim 0.6 \mu_B/\text{Ce}$ for $H_c \sim H_K$ Kondo field associated to the local spin fluctuations.

For $P > P_c$, the first order metamagnetic transition is replaced by crossover phenomena at H_m referred as a pseudo-metamagnetism. The effect can be quite sharp since close to P_c , Kondo fluctuation and AF intersite interaction have comparable strength. Approaching P_c , the crossover field $H_m \sim H_K$ is reduced and is comparable to H_c at P_c for $T = 0$ K.

The combination of pressure and magnetic field gives the opportunity to observe the interplay between $H_c(T, P)$ and $H_m(T, P)$ schematically shown in Figs. 1(a) and 1(b) assuming an unique AF instability at a wave vector \mathbf{k}_1 . For $P < P_c$ in the AF domain, the H_m crossover line joins the $H_c(T)$ one at finite temperature.

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For $P > P_c$ the H_m line marks the entrance in the PPM region. Measuring thermal expansion or specific heat for $P > P_c$ shows that two main clear crossover regimes delimited by a $\tilde{T}(H)$ line emerge:^{14,15)} a low field nearly AF (NAF) regime dominated by AF correlations and a high field one (PPM) above H_m governed by the strong local polarization of the electron. Just in the vicinity of H_c or H_m strong duality between AF at \mathbf{k}_1 and FM fluctuations at $k = 0$ fluctuations is observed.^{16–18)} The expected variation of $\gamma(H)$ for both cases are shown in Figs. 1(c) and 1(d). Sharp maxima of γ are expected at H_c and H_m on both sides of P_c in agreement with the theoretical framework on AF tricriticality. However, the first order nature of the metamagnetic transition can wipe out the sharp increase of γ on approaching H_c .

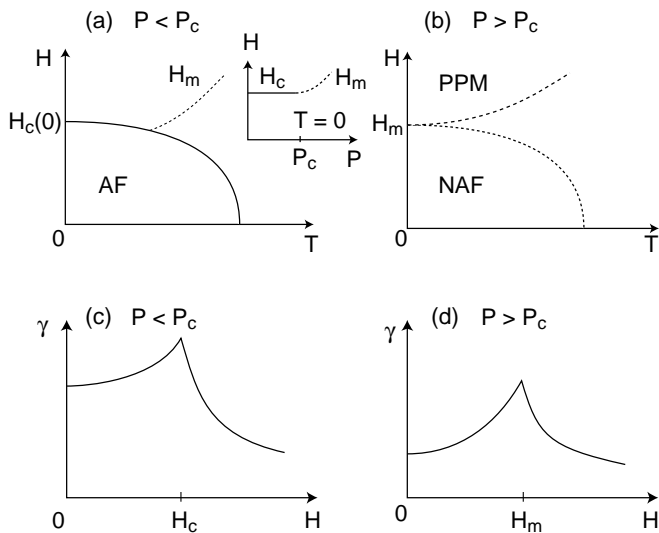


Fig. 1. Interplay between AF and pseudo-metamagnetism for (a) $P < P_c$ and (b) $P > P_c$. The inset of panel (a) shows the pressure variation of H_c and H_m at $T = 0$ K. Field variation of the Sommerfeld coefficient $\gamma(H)$ for (c) $P < P_c$ and (d) $P > P_c$. AF, PPM and NAF denote antiferromagnetism, polarized paramagnetism and nearly antiferromagnetism, respectively.

Furthermore in this complex band structure case, the magnetic field can induce changes in the ordered AF wave vector as often different magnetic hot spots exist on the Fermi surface. Thus the selection of the AF wave vector can be modified by the magnetic field in the AF domain.

Inelastic neutron scattering experiments on the pure CeRu_2Si_2 system indicate that three AF hot spots exist at $\mathbf{k}_1 = (0.31, 0, 0)$, $\mathbf{k}_2 = (0.31, 0.31, 0)$ and $\mathbf{k}_3 = (0, 0, 0.35)$.¹⁹⁾ The two first \mathbf{k} vectors, \mathbf{k}_1 and \mathbf{k}_2 are transverse modes as the fluctuating moments are aligned along the c -axis of the tetragonal crystal, while \mathbf{k}_3 is a longitudinal one. Under magnetic field, a sharp pseudo-metamagnetic crossover occurs at H_m marked by a sharp effective mass enhancement right at H_m , a slowdown of the field induced FM fluctuation, and a spectacular Fermi surface change.^{1, 4, 10, 11, 17, 18)}

For the La substituted case, AF ordering at $H = 0$ occurs at a transverse wave vector \mathbf{k}_1 with respect to the sublattice magnetization aligned along c -axis.⁶⁾ At low

temperatures, the sharp pseudo-metamagnetic crossover at H_m is replaced by a first order metamagnetic transition at H_c . Increasing temperature, the field sweep below the Néel temperature T_N leads to detect both $H_c(T)$ and $H_m(T)$. At x_c for $T \rightarrow 0$ K, H_c is equal to H_m . The (H, T) phase diagram is more complex than that shown in Fig. 1 as at a critical field $H_a < H_c$, the AF ordered wave vector remains transverse but changes from incommensurate wave vector to another complex AF phase with a large hysteresis domain characterized on cooling by the appearance of the wave vector \mathbf{k}_2 and even a commensurate component $(1/3, 1/3, 0)$ at low temperature.^{20, 21)} For x larger than x_c , the schematic scheme of the (H, T) phase diagram measured by the elastic neutron diffraction is shown in Fig. 2(a).

In the previous studies with La substitution, the $H_c(T)$ and $H_m(T)$ phase diagram interfere, leading to an AF order to PPM phase boundary at H_c for $\delta < \delta_c$. There is no obvious basic arguments for the coincidence between H_c and H_m at δ_c . However, it is clear that the dominant competing wave vector are the AF transverse ones ($\mathbf{k}_1, \mathbf{k}_2, \dots$) and the uniform FM mode.

A striking point is that Rh substitution on the Ru site, namely $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ reveals a decoupling between H_c and H_m .^{22–24)} This drastic change may be driven by the fact that with Rh doping the ordered wave vector in the AF domain ($x > x_c = 0.05$) is now the longitudinal ($\mathbf{k}_3 = (0, 0, 0.35)$) wave vector instead of the transverse mode in the La doped alloys (Fig. 2(b)).^{25, 26)} Up to now there is no indication via neutron scattering experiments what will be the dominant AF spin fluctuation mode up to H_m .

Previous experiments on $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ were limited at temperatures above 1.5 K except for $x = 0.15$ which is quite higher than $x_c \sim 0.05$.^{27, 28)} Thus we performed new set of experiments on $x = 0.08$. To clarify the situation, we focus here on the thermodynamic and transport measurements at very low temperatures down to 100 mK. The first aim is to determine the field variation of the Sommerfeld coefficient γ through H_c and H_m and to compare the results with magnetization, specific heat and resistivity measurements. The second aim is to investigate precisely the crossover line $\tilde{T}(H)$ by high accurate thermal expansion measurements in the intermediate temperature range above 2 K. The third aim is to clarify the pressure evolution of H_m and the effective mass, comparing with those of CeRu_2Si_2 .

It will be shown that the substitution of Ru by Rh is a major perturbation due to the mismatch of the lattice parameter of CeRu_2Si_2 and CeRh_2Si_2 . In the extended field region $H_m - H_c = 3$ T assumed to be paramagnetic, the fancy point is the quasi-invariant value of γ close to the critical value γ_c observed at H_c^* or P_c at $H = 0$ in the La doped system. The validity of the hypothesis of PM ground state above H_c for longitudinal and transverse modes will be confirmed by new neutron scattering experiments.

2. Experimental

Single crystals of $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ were grown using the Czochralski method in a tetra-arc furnace. Start-

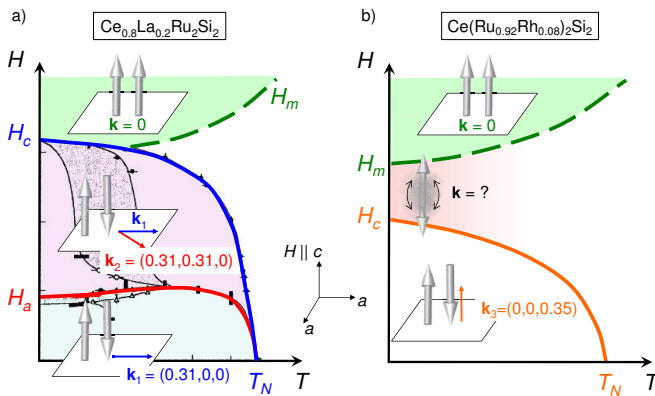


Fig. 2. (Color online) Schematic magnetic phase diagrams of $\text{Ce}_{0.8}\text{La}_{0.2}\text{Ru}_2\text{Si}_2$ and $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$.

ing materials of Ce (purity: 99.9%-3N), Ru(4N), Rh(4N) and Si(6N) with the ratio, 1 : 1.84 : 0.16 : 2 were melted under the high purity Ar gas for a polycrystalline ingot. The ingot was tuned over and was melted again. This process was repeated several times in order to obtain the homogeneous phase. The ingot was subsequently pulled with a pulling rate of 15 mm/hr. The obtained single crystal ingot was cut using a spark cutter, and was oriented by the X-ray Laue photograph. The first specific heat measurements were realized using the relaxation method at temperatures down to 0.42 K and at magnetic fields up to 9 T. The measurements were then pushed down to 0.21 K, using a homemade dilution refrigerator up to 14 T. Precise magnetization measurements were performed down to 80 mK and up to 8 T with a homemade SQUID magnetometer, which was successfully applied in our previous experiments CeRu_2Si_2 ¹⁵⁾ and CeCoIn_5 .²⁹⁾ Thanks to the Maxwell relation, $\partial\gamma/\partial H = \partial^2 M/\partial T^2$, the field dependence of γ can be determined by integrating $\partial^2 M/\partial T^2$ with field. Resistivity measurements were performed using four probe AC method at temperature down to 100 mK and at field up to 16 T. The pressure study was also realized by the resistivity measurements in a NiCrAl-CuBe pressure cell up to 3.8 kbar. The pressure was determined by the superconducting transition temperature of Pb. The $H_c(T)$ and $H_m(T)$ boundaries were confirmed by magnetostriction experiments using a strain gauge glued on the c -plane down to 2 K and up to 9 T. The search for $\tilde{T}(H)$ line was precisely investigated by high precision thermal expansion measurements using a capacitance dilatometer down to 2 K. For comparison, thermal expansion measurements were performed in $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$, as well. The single crystals obtained in the previous reports^{30,31)} were used. The precision of our measurements has been improved, compared with the previous works realized down to 1.2 K.^{32,33)} In order to confirm that H_c marks the AF-PM boundary, a neutron diffraction experiment was performed on the two-axis D23-CRG-CEA thermal neutron diffractometer equipped with a lifting detector at ILL in Grenoble. A copper monochromator provides an unpolarized neutron beam with a wavelength of $\lambda = 1.276 \text{ \AA}$. The single crys-

tal sample was put in the 12 T vertical field magnet with the c -axis along the field.

3. Results

3.1 $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$

Figure 3 shows the temperature dependence of C/T at different magnetic fields. AF transition occurs at $T_N = 4.2 \text{ K}$ at zero field. As for classical AF, T_N identified as a jump of C/T decreases with H . That allows to determine $H_c(0) \sim 2.8 \text{ T}$. Surprisingly between 3 T and 5.8 T, C/T is almost invariant against field in a regime assumed to be paramagnetic. At $H_m \sim 5.8 \text{ T}$, C/T abruptly drops as observed for CeRu_2Si_2 just above the pseudo-metamagnetic field $H_m \sim 7.8 \text{ T}$.

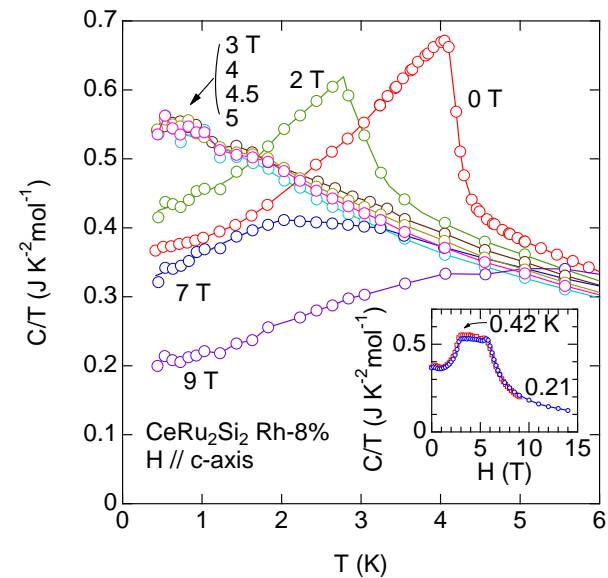


Fig. 3. (Color online) Temperature dependence of the specific heat in the form of C/T vs T at different fields in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. The inset shows the field dependence of the specific heat in the form of C/T vs H at 0.21 K and 0.42 K.

Figure 4 shows the field dependence of C/T at different temperatures with again the field decrease of the AF anomaly at $T_N(H)$ and the smearing out of the PPM domain on warming at $H_m(T)$.

Figure 5 represents the response of the strain gauge directly linked with the change of the length $\Delta L/L$ along the c -axis. Taking the field derivative $\lambda_c = d(\Delta L/L)/dH$, the positions of the lines $H_c(T)$ and $H_m(T)$ are well drawn (Fig. 6). The singular point is that the magnetostriction at H_m appears quite more broadened than the one observed for CeRu_2Si_2 at the PM-PPM boundary.^{1,3,4,14,15)} As discussed later, this smearing is caused by the doping which inhibits partly a full deformation of the lattice.

A new set of thermal expansion measurements α was realized to search for the crossover $\tilde{T}(H)$ line for $H \parallel c$, as shown in Fig. 7. As observed in the La-doped case,³⁴⁾ the same sign of thermal expansion is detected along c and a -axes with a ratio near 3. The AF $H_c(T)$ line is very well defined through the strong negative jump of α .

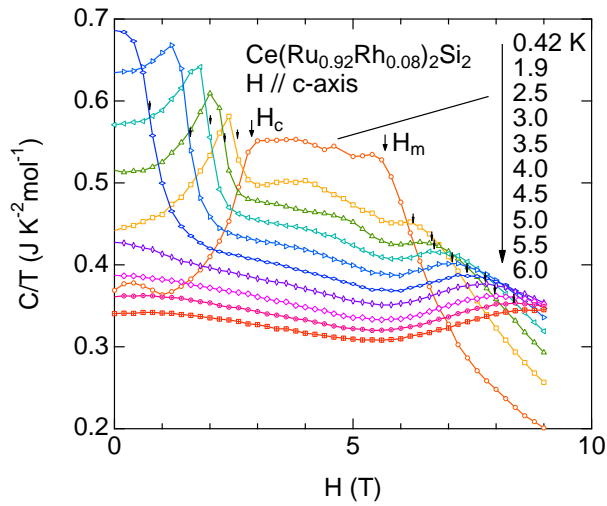


Fig. 4. (Color online) Field dependence of the specific heat in the form of C/T vs H at different temperatures in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. The small arrows indicate $H_c(T)$ and $H_m(T)$.

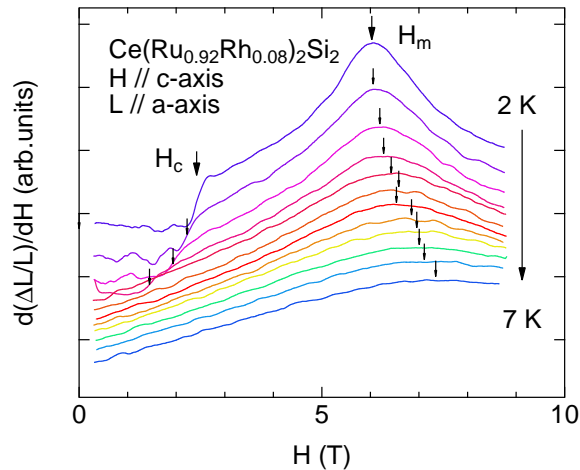


Fig. 5. (Color online) Field dependence of the magnetostriction in the form of $d(\Delta L/L)/dH$ vs H at different fields in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$.

The crossover line $\tilde{T}(H)$ can be drawn following the position of the extremum of thermal expansion at fixed field (Fig. 8). This indicates strongly that the region between H_m and H_c is paramagnetic. The (H, T) phase diagram can be reconstructed with a mixture of phase diagram shown in Fig. 1. A clear separation exists between H_c and H_m . The effect is obvious in the magnetostriction data $\lambda_V = (1/V)\partial V/\partial H$ shown in Fig. 9 even for $T \sim 2\text{K}$ equal only to $0.5T_N$.

Figure 10 shows the magnetization curve extrapolated to 0 K from the temperature dependence down to 75 mK at different fields. A weak hysteresis is detected only at H_c , as it was reported on $\text{Ce}(\text{Ru}_{0.85}\text{Rh}_{0.15})_2\text{Si}_2$.²⁴⁾ Taking the H derivative $\chi(H) = \partial M/\partial H$ of the magnetization (inset of Fig. 10), one can observe that the transition at H_c and H_m are very broad by comparison to

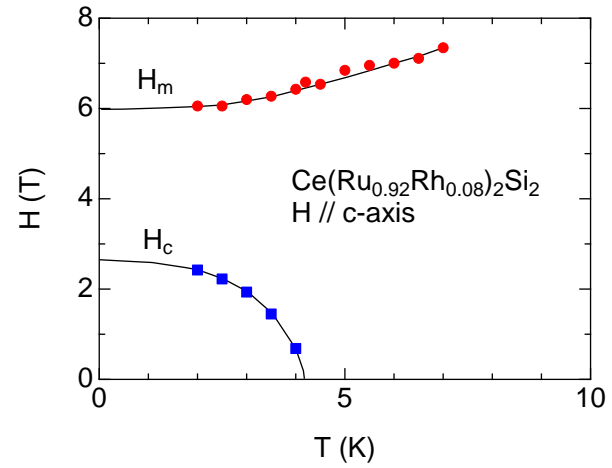


Fig. 6. (Color online) (H, T) phase diagram obtained by the magnetostriction measurements in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$.

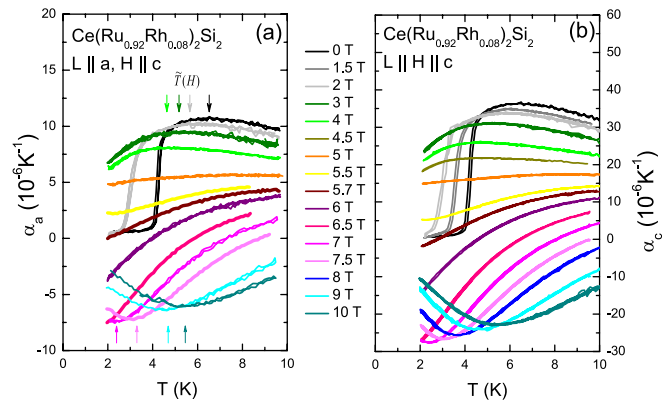


Fig. 7. (Color online) Thermal expansion of $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ with high accuracy at the intermediate temperature regime ($T > 2\text{K}$) at different fields for $L \parallel a$ -axis and $L \parallel c$ -axis.

the first order metamagnetic transition detected for example on $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ ($T_N = 5\text{K}$) at H_c and to the pseudo-metamagnetic transition of CeRu_2Si_2 where $\chi(H_m)$ reaches $1.6 \mu_B/\text{T}$ at 100 mK.^{1,30)} This reduction points out that the magnetostriction is strongly affected as the susceptibility at constant pressure (χ_P) is linked to the susceptibility at constant volume χ_V by the relation:

$$\chi_P = \chi_V + \lambda_V^2 \frac{V_0}{\kappa}, \quad (1)$$

where V_0 is the molar volume and κ is the compressibility. A quite similar attenuation is observed when CeRu_2Si_2 is doped with La and Y in $\chi_P(H_m)$ and $\lambda_V = (1/V)\partial V/\partial H$.³⁾ It is interesting to notice that in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ the switch from PM to PPM phase occurs at a critical value $M_c \sim 0.7 \mu_B$, just slightly higher than the one measured in CeRu_2Si_2 .

The absence of hysteresis between H_c and H_m , the broad susceptibility anomaly at H_m , and the observation of crossover $\tilde{T}(H)$ line give strong supports that the H_m - H_c window corresponds to a paramagnetic ground state. The paradox is that in this paramagnetic domain a quasi-

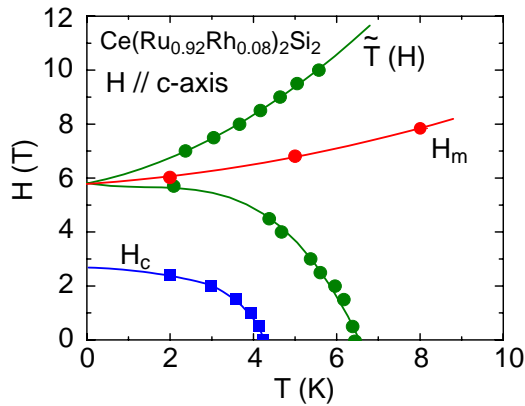


Fig. 8. (Color online) (H, T) phase diagram of $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ determined by the precise thermal expansion measurements.

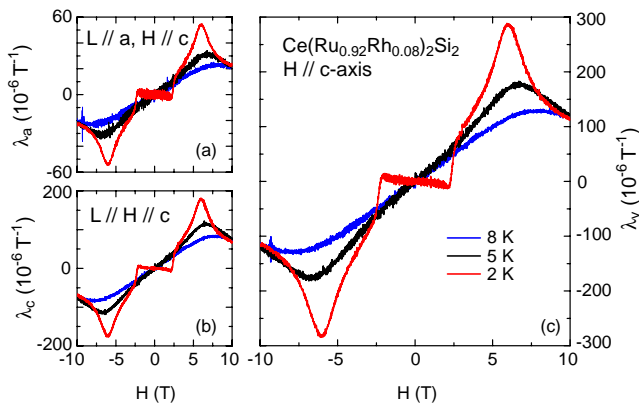


Fig. 9. (Color online) Field dependence of the magnetostriction $\lambda_V = (1/V)(\partial V/\partial H)$ on $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$.

constant field value of γ is observed as derived with great accuracy from the application of the Maxwell relation to the temperature dependence of the magnetization at constant field, namely $\partial\gamma/\partial H = \partial^2 M/\partial T^2$ and also by direct specific heat measurements (Fig. 11).

In order to establish that AF disappears at H_c , we have performed new elastic scattering experiments on a single crystal extracted from the same batch. The aim was to verify the possible interplay between the longitudinal and transverse mode. Figure 12 shows the representative Q scan performed around $Q = (1, 1, 0) + \mathbf{k}_3$ (Fig. 12(a)), $Q = (1, 1, 0) - \mathbf{k}_1$ (Fig. 12(b)) and $Q = (1, 1, 0) - \mathbf{k}_2$ (Fig. 12(c)) at different field at 2.3 K. These data indicate that the magnetic ordering with the wave vector \mathbf{k}_3 disappears at H_c , while no signal is induced at \mathbf{k}_1 and \mathbf{k}_2 for higher magnetic fields above H_c and H_m . Figure 12(d) shows the magnetic field dependence of the integrated intensity measured for \mathbf{k}_3 . The value of H_c at 2.3 K is approximately 2.3 T in good agreement with the phase diagram drawn in Fig. 8. Short range correlations are observed up to 2.8 T. Figure 12(e) gives the field dependence of the l component of \mathbf{k}_3 which is found to be 0.36 at low field.

Transport measurements were realized as shown in

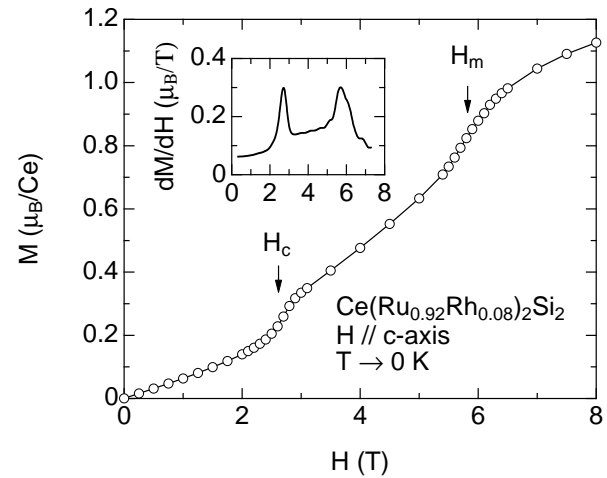


Fig. 10. Magnetization curve extrapolated to 0 K obtained by the temperature dependence at different fields in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. The inset shows the field derivative of the magnetization curve.

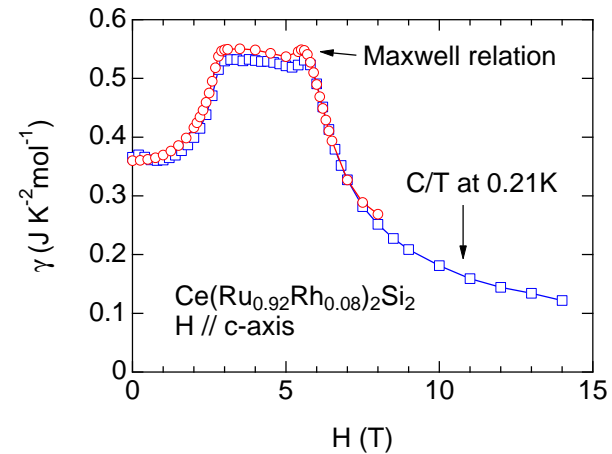


Fig. 11. (Color online) Field dependence of the Sommerfeld coefficient γ obtained by the Maxwell relation from the magnetization measurements assuming $\gamma(0) = 0.36 \text{ J K}^{-2} \text{ mol}^{-1}$. The result of direct specific heat measurements at 0.21 K is plotted as well for comparison.

Figs. 13 and 14. The (H, T) phase diagram is well reproduced with a characteristic drop of the residual resistivity at $H_c(0)$ followed by a wide maximum at $H_m(T)$. In agreement with the validity of the Kadowaki-Woods relation, the A coefficient of the T^2 resistivity law roughly follows the field dependence of γ according to the relation $A \propto \gamma^2$. In the case of doping, additional deviation from the Kadowaki-Woods relation can occur due to the T dependence of the impurity scattering and also due to the additional modification in the carrier number.^{35,36)}

Finally, preliminary pressure studies were realized to test the pressure response of H_c and H_m up to 3.8 kbar, as shown in Fig. 15. With increasing pressure, the plateau of the resistivity A coefficient between H_m and H_c decreases in value. H_c disappears rapidly at $P \sim 1.5$ kbar, while H_m monotonously increases with pressure. In this

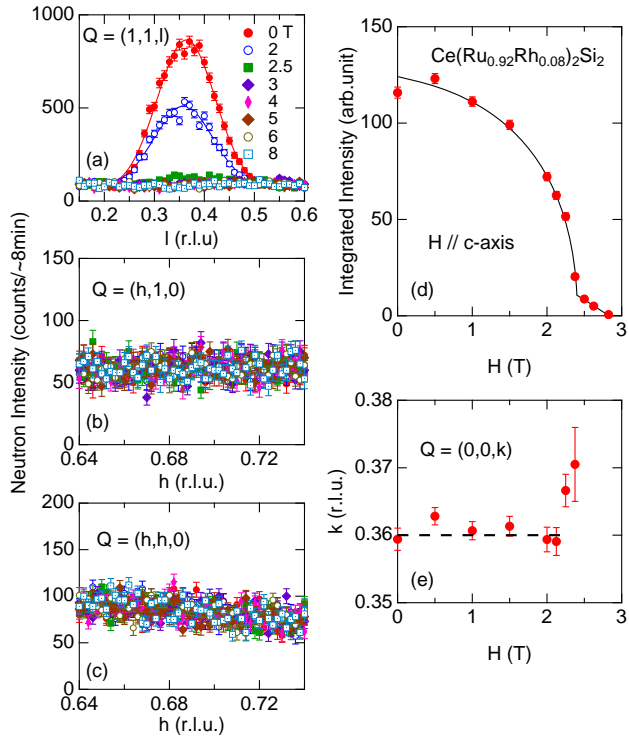


Fig. 12. (Color online) (a)–(c) Neutron diffraction profiles of $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ at 2.3 K at different fields for $H \parallel c$ -axis along $(1, 1, l)$, $(h, 1, 0)$ and $(h, h, 0)$ corresponding to \mathbf{k}_3 , \mathbf{k}_1 and \mathbf{k}_2 , respectively. (d) Field dependence of the integrated intensity of $(0, 0, k)$ at 2.3 K. (e) Corresponding field dependence of k .

preliminary experiments, it is not possible to conclude that H_c terminates at P_c as a critical endpoint, as it is in the La-doped case at x_c .¹⁾ A fine tuning through P_c is necessary. Close to the critical pressure ($P \sim 1.5$ kbar) where the longitudinal ordering collapses, the critical value of γ_c may be quite lower than the value of $\gamma \sim 650 \text{ mJ K}^{-2} \text{ mol}^{-1}$ in $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ reported for the AF instability of the transversal mode,^{30,31,37} as $A^{\text{Rh}}(P_c) \sim 1.4A^{\text{Rh}}(P=0)$ at $H=0$ leading to $\gamma_c \sim 450 \text{ mJ K}^{-2} \text{ mol}^{-1}$. Above P_c , the field variation of A is quite identical to that reported for CeRu_2Si_2 at $P=0.22$ GPa ($H_m \sim 11$ T).³¹⁾ The weak maxima of A at H_m reflects the duality between AF and FM correlations. Above H_m , good scaling of $A(H)/A(H_m)$ is observed under pressure. As seen in Fig. 16, assuming $A \sim (m^*)^2$ at H_m , the pressure dependence of electronic Grüneisen parameter $\Omega(H_m) = -\frac{\partial \ln m^*}{\partial \ln V}$ is quite similar between the pure system CeRu_2Si_2 and the Rh-doped system $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. A more accurate value of Ω determined by thermodynamic measurements is known to be $\Omega(H_m) \sim 200$ at ambient pressure in CeRu_2Si_2 . It is interesting to note that $\Omega(H_c)$ in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ strongly decreases with pressure by comparison to $\Omega(H_m)$ both in CeRu_2Si_2 and in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$.

3.2 Comparison with $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$

Next we compare the results of $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ with those of $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$, focusing on the 10 %

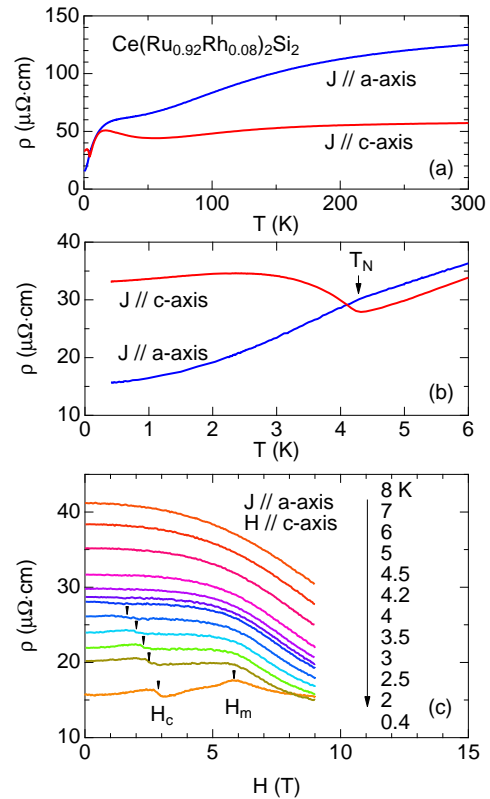


Fig. 13. (Color online) (a) Temperature dependences of the resistivities for the current along a and c -axes and (b) the corresponding low temperature resistivities. (c) Magnetoresistance at different temperatures for $H \parallel c$ -axis and $J \parallel a$ -axis

doped crystal, as it presents similar impurity scattering, as known from the value of ρ_0 .^{5,30)} Its Néel temperature T_N is around 2.5 K but the specific heat anomaly at T_N is much broader than the one measured for the previous Rh doped case, as it is closer to AF–PM instabilities. Focus is given here on recent magnetization data published in Ref. 31. As seen in Fig. 17 for the transverse AF ordered mode, a clear metamagnetic transition occurs at H_c without the separation between H_c and H_m at low temperatures. However, on warming, a marked difference between H_c and H_m was already observed since $H_c(T)$ decreases strongly. The transition between the two transverse modes at $H_a \sim 1$ T is marked by a rather broad maximum of $\partial M / \partial H$. Let us notice the large difference in shape and amplitude in $\partial M / \partial H$ detected at H_c between $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ and $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$. The inset of Fig. 17 shows the field variation of $\gamma(H)$ as derived from the Maxwell relation. For both cases, $\gamma(H)$ shows a drastic decrease of γ on entering in the PPM state.

Precise thermal expansion measurements were also realized on $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ down to 2 K (Fig. 18). Due to the weakness of $T_N \sim 2.5$ K and also the rather broad specific heat anomaly associated to the onset of the ordering, the AF boundary is difficult to define. However as for $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$, clear extrema of α in temperature allow to determine the $\tilde{T}(H)$ crossover (Fig. 19). The corresponding phase diagram is shown in Fig. 20. Quite

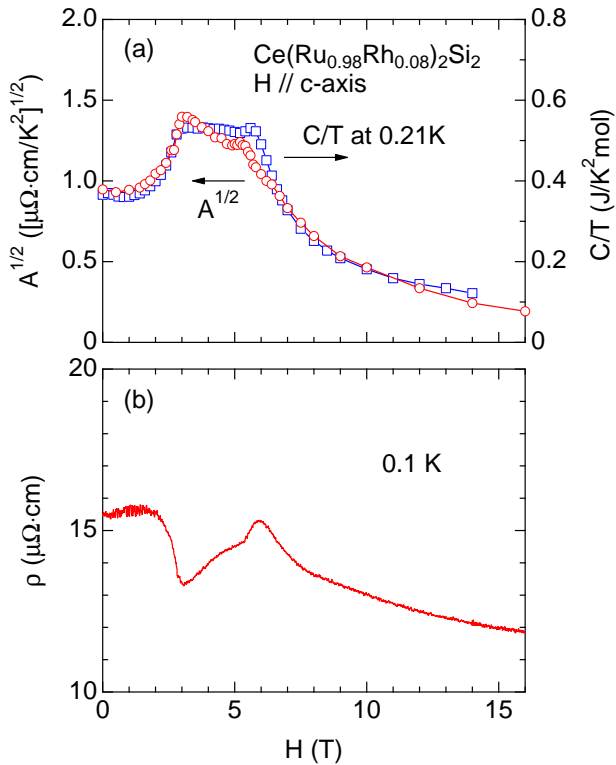


Fig. 14. (Color online) (a) Field dependence of the resistivity A coefficient and the specific heat at 0.21 K in the form of \sqrt{A} vs H and C/T vs T in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. (b) Field dependence of the resistivity at 0.1 K.

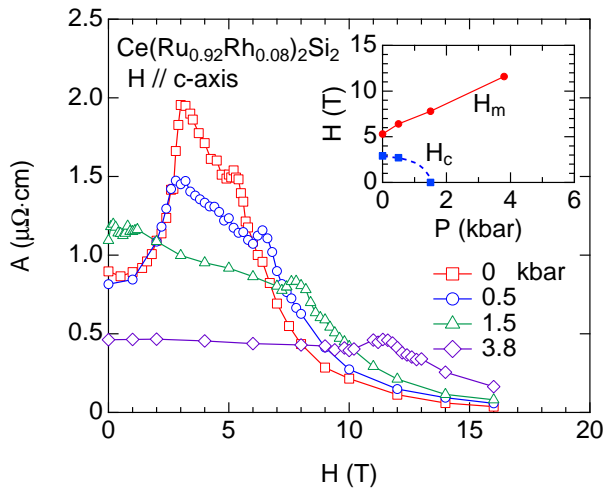


Fig. 15. (Color online) Field dependence of the A coefficient of resistivity at different pressures for $H \parallel c$ -axis in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. The inset shows the pressure dependence of H_m and H_c .

similar behaviors emerge with the sign that AF correlations will be initially stronger for $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ in good agreement with the ranking of their T_N .

4. Discussion

In $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$, the image is that H_c marks the transition from AF to PM phases. Increasing the

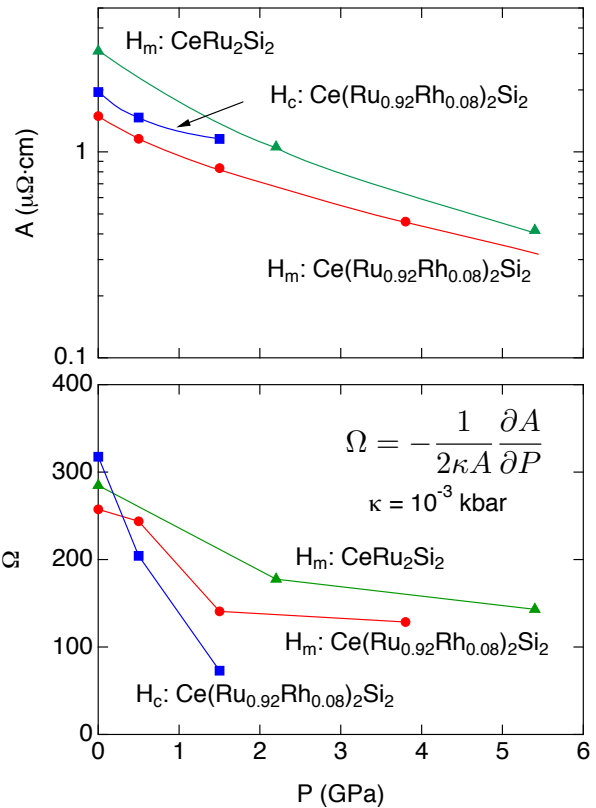


Fig. 16. (Color online) (a) Pressure dependence of the A coefficient at H_m and H_c in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ and CeRu_2Si_2 . (b) Pressure dependence of the effective Grüneisen parameter Ω for \sqrt{A} at H_m and H_c in CeRu_2Si_2 and $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. Ω is defined as $\Omega \equiv -\frac{\partial \ln m^*}{\partial \ln V} \sim -\frac{1}{2\kappa A} \frac{\partial A}{\partial P}$, where κ and m^* are compressibility and effective mass, respectively

magnetic field above H_c may lead to escape from AF criticality and thus will correspond to a strong field-decrease of γ as proposed in Fig. 1. However, as observed in $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ where H_a marks a switch from one AF phase to another AF phase only by changing its wave vector, the γ -value is basically field-invariant between H_c and H_m in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$. Furthermore its value is quite close to the maximum value of $\gamma_c \sim 650 \text{ mJ K}^{-2} \text{ mol}^{-1}$ reached at P_c or at H_c^* in the $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ family. This suggests that above H_c in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ the low energy AF spin dynamics may be dominated by the fluctuations of the transverse mode. However, when the polarization reaches a critical value at H_m corresponding to a critical value of magnetization $M_c \sim 0.7 \mu_B$, a strong field decrease of γ is observed quite similar to that detected on the PPM side of CeRu_2Si_2 for $M = M_c = 0.6 \mu_B$. Figure 21 shows the comparison for the relative field variation of $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ and CeRu_2Si_2 ^{1,4,7} in a γ/γ_m vs H/H_m representation. For CeRu_2Si_2 a sharp maximum occurs at H_m as the magnetic field close to H_m wipes out the AF correlations, while the growth of uniform magnetization is associated with an increase of the FM correlations which slows down just at the vicinity of H_m .¹⁾ Increasing further the magnetic field above H_m reinforces the local character and thus leads to recover the proper-

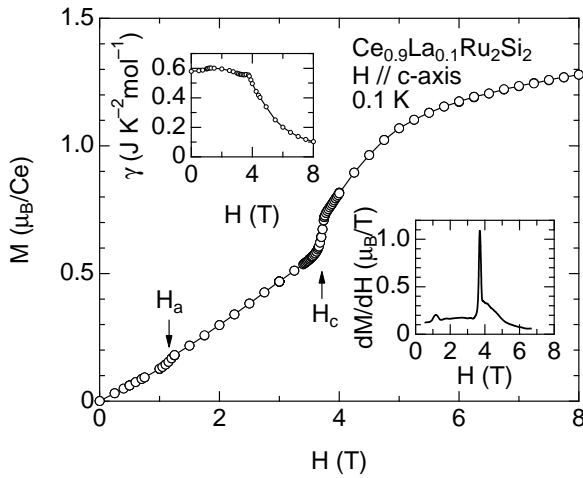


Fig. 17. Magnetization curve for $H \parallel c$ -axis at 0.1 K in $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$. The inset at right-bottom is the field-derivative of the magnetization curve. The inset at left-top is the field dependence of the γ -value obtained from the temperature dependence of the magnetization using the Maxwell relation.

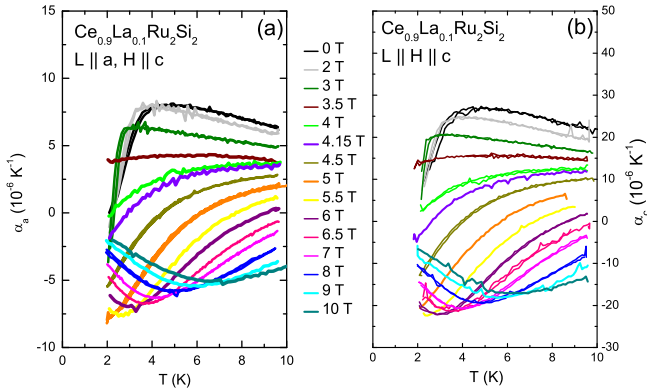


Fig. 18. (Color online) Thermal expansion of $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ with high accuracy at the intermediate temperature regime ($T > 2$ K) at different fields for $L \parallel a$ -axis and $L \parallel c$ -axis.

ties of polarized Kondo centers.

Previous results on $\text{Ce}(\text{Ru}_{0.85}\text{Rh}_{0.15})_2\text{Si}_2$ ($T_N = 5.5$ K, $k_3 = 0.42$, $H_c = 3.5$ T, $H_m = 5$ T) give an extrapolation of $\gamma(H)$ with only a rounded maxima near H_m ($\gamma(H_m) = 550 \text{ mJ K}^{-2} \text{ mol}^{-1}$, while the zero field value reaches $\gamma(0) \sim 300 \text{ mJ K}^{-2} \text{ mol}^{-1}$; those two values are quite close to the ones found in the present experiment for $x = 0.08$. Approaching x_c leads to an increase of the H_c - H_m window (by a factor 2) and thus to an excellent decoupling between AF and pseudo-metamagnetic instabilities. As mentioned later, low doping allows also to minimize the effect of lattice mismatch.

The novelty in the Rh doped case is the mismatch in the lattice parameters of CeRu_2Si_2 and CeRh_2Si_2 , which is tabulated in Table I together with the comparison with LaRu_2Si_2 . In La-doped case, both a and c lattice parameters expand. The driving force is the volume and the local perturbation is only moderated. In Rh-doped case, the value of a decreases, while the value of c increases. This contradictory behavior boosts the local perturbation now

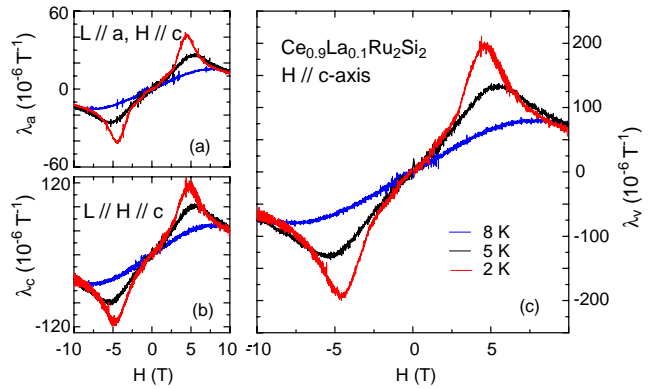


Fig. 19. (Color online) Field dependence of the magnetostriction $\lambda_V = (1/V)(\partial V/\partial H)$ on $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$.

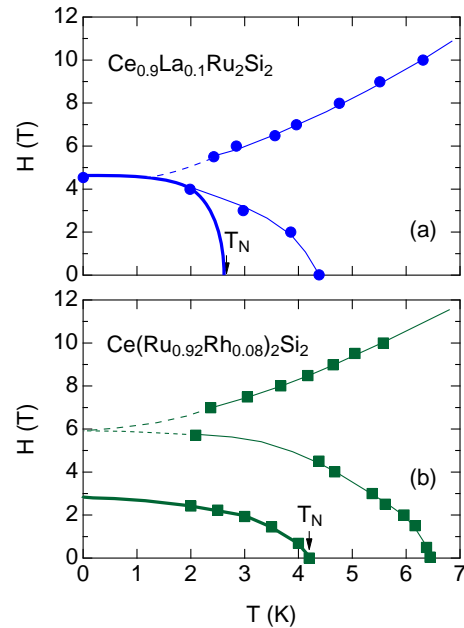


Fig. 20. (Color online) (H, T) phase diagram of (a) $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ and (b) $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ obtained by the precise thermal expansion measurements. Lines are guides to the eyes. Thin lines correspond to the crossover from results of the temperature dependence of thermal expansion. Thick lines correspond to the AF boundary. The data of AF boundary for $\text{Ce}_{0.9}\text{La}_{0.1}\text{Ru}_2\text{Si}_2$ are extrapolated from the previous low temperature measurements.³⁰⁾

not reduced to sole volume effects. Furthermore Fermi level may change by the substitution of Ru by Rh as well as the local fluctuations due to an increase of the carrier number of d electrons. The volume change between CeRu_2Si_2 and CeRh_2Si_2 is only 4×10^{-3} corresponding to a compression by 4 kbar. Despite this volume reduction, T_N in CeRh_2Si_2 reaches 36 K and H_c is 26 T. Furthermore AF occurs for a commensurate wave vector $(1/2, 1/2, 0)$ quite different from $(0, 0, 0.35)$ of Rh-doped CeRu_2Si_2 . Estimation of the Kondo temperature $T_K \sim 50$ K for CeRh_2Si_2 instead of $T_K \sim 25$ K for CeRu_2Si_2 . In the popular image of the Doniach collapse of AF for a Kondo lattice, the Ruderman-Kittel-Kasuya-Yosida (RKKY) in-

teractions in CeRh_2Si_2 must be quite larger than that of CeRu_2Si_2 . Thus a source of competing interactions is created via the Rh substitution, plus additional change in the carrier number caused by the Rh doping. At least an important consequence is that at $H = 0$ the magnetic ordered mode became the longitudinal one. It was recently observed that slight changes of the Fermi surface occur on doping with La or Ge substitution.^{36,38} It seems indirectly here that Rh doping has more drastic effect which promotes the \mathbf{k}_3 instabilities.

Evidences of strong nesting properties with partial gap opening for the Rh-doped system were given in the resistivity measurements, when the current J is applied along the c -axis as shown in Fig. 13(a)(b). For $\text{Ce}(\text{Ru}_{0.85}\text{Rh}_{0.15})_2\text{Si}_2$, similar data can be found in ref. 39. No jump of resistivity for $J \parallel c$ -axis was observed in the La-doped case, while with Ge-doping, the nesting is detected for $J \parallel a$ -axis.⁴⁰ We have verified that the nesting persists also in the present system $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$, as shown in Fig. 13(b).

Table I. Lattice parameters and volumes of CeRu_2Si_2 , LaRu_2Si_2 and CeRh_2Si_2 .

	CeRu_2Si_2	LaRu_2Si_2	CeRh_2Si_2
a	4.192 Å	4.215 Å	4.09 Å
c	9.78 Å	9.93 Å	10.18 Å
c/a	2.32	2.20	2.48
V	171 Å ³	176.4 Å ³	170.3 Å ³

A difficult enigma is what would be the phase diagram of a pure lattice of CeRu_2Si_2 (without induced disorder), if the ground state at $H = 0$ would be an AF longitudinal mode; would the transition from AF to PM at H_c be replaced by a switch between two AF structures as observed for the La substitution? As pointed out in Fig. 2, the AF regime^{20,21} is rather complex at $T_N(H = 0)$. A new question is if, due to the Rh substitution, above H_c , the system will switch from AF phase dominated by a longitudinal mode to a PM nearly AF phase dominated by a transverse mode between H_c and H_m before becoming governed by the crossover to the PPM phase. A strong indication is given by the pressure resistivity measurements suggesting quite different critical values of γ_c for the longitudinal and transversal instability.

Obviously, an open question is the interplay between the different modes on the spin dynamics. It will require a new generation of inelastic experiments, in which it is expected that the transverse mode may govern the spin dynamics above H_c . The main difficulty is to grow a large homogenous Rh-doped single crystal, as the physical properties are very sensitive to the development of concentration gradient. Our choice will be to remain in the AF domain with $x \sim 0.08$ as we can verify the homogeneity from the size and shape of the specific heat anomaly at T_N for $H = 0$. Another microscopic probe will be to succeed in detecting the Fermi surfaces. Here application of the magnetic field along the basal plane gives some hope to track the Fermi surface change, despite the fact that the electronic mean free path could

be damped by doping.

Some of the figures with H_m-H_c window are reminiscent of the effects observed in the cases such as $\text{Sr}_3\text{Ru}_2\text{O}_7$ ⁴¹ or recently UCoAl .⁴² In $\text{Sr}_3\text{Ru}_2\text{O}_7$ it was proposed that a nematic phase appears due to the strength of spin-orbit coupling, and that weak disorder preempts this establishment at the profit of a smearing into a metamagnetic quantum critical point. In UCoAl , a plateau of $\gamma(H)$ was recently detected above the quantum critical endpoint. At least, strong evidences are given here that the large H plateau of constant γ is the result of the competition between AF instability and field-driven FM instability.

5. Conclusion

Doping CeRu_2Si_2 with Rh instead of La or Ge leads to a drastic change of (H, T) phase diagram of the AF phases. The achievement of very low temperature allowed to characterize the field variation of γ inside the AF domain below $H_c = 2.8$ T inside the “unstable” PM phase between H_c and $H_m = 5.8$ T and in the PPM phase. The pressure study suggested that two critical values of γ are associated respectively to longitudinal (\mathbf{k}_3) and transversal AF instability. A key ingredient is the lattice mismatch between CeRu_2Si_2 and CeRh_2Si_2 . Above H_c , the occurrence of a PM ground state with Rh-doping is in excellent agreement with the lattice contraction by comparison to the pure system CeRu_2Si_2 .

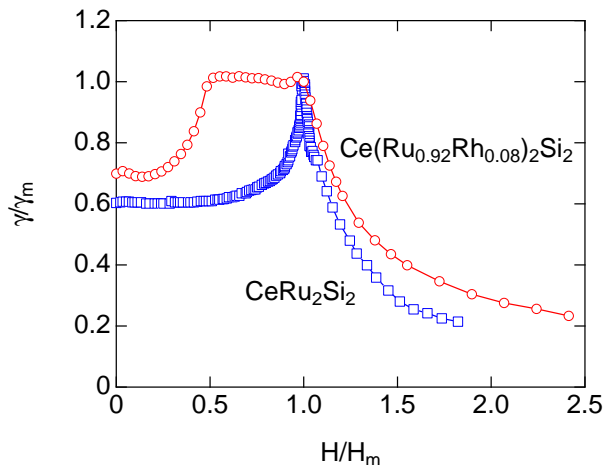


Fig. 21. (Color online) Field dependence of the Sommerfeld coefficient γ in $\text{Ce}(\text{Ru}_{0.92}\text{Rh}_{0.08})_2\text{Si}_2$ and CeRu_2Si_2 . The field and γ are scaled with H_m and the γ -value at H_m , respectively. The data of CeRu_2Si_2 are cited from ref. 7

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