Crossover between "localized" and "itinerant" antiferromagnetic states in $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$ driven by chemical pressure

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Abstract

We have investigated the magnetization process of the pseudobinary Kondo-lattice system $Ce(Ru_{0.9}Rh_{0.1})_2$-$(Si_{1-y}Ge_y)_2$. The base material $Ce(Ru_{0.9}Rh_{0.1})_2Si_2$, which has a spin density wave(SDW) phase below $T_N = 5.0$ K, shows two-step metamagnetic behavior at H_c and H_m; the SDW-Fermi liquid (FL) transition occurs at H_c, and FL state becomes unstable and localized spin character recovers at H_m. By substituting Ge for Si, H_m reduces very rapidly and disappears for $y > 0.08$, where the magnetization curve is quite similar to those of localized Ising spin antiferromagnets(AF). This observation is interpreted as a manifestation of crossover of the character of the AF state from "itinerant" to "localized" induced by the negative chemical pressure.

Key words: heavy fermion; SDW; magnetization; chemical pressure

The magnetism of the heavy fermion(HF) systems is described by the competition between the Kondo effect and the RKKY interaction, both of which originate from the hybridization of the conduction electrons and the f-electrons (cf-hybridization). The amplitude of the cf-hybridization can be easily controlled and the magnetic instability can be caused experimentally, for instance by regular magnitude of hydrostatic pressure or by chemical pressure due to alloying. The magnetic instability in HF systems induces various attractive phenomenon, for example the exotic superconductivity[1], the non-Fermi liquid behavior[2].

In the antiferromagnetic HF compounds, there is consider to be two types of magnetic order; one is the order with the localized moment on each magnetic atom, which is formed by RKKY interaction, and the other is the itinerant magnetic order with polarized heavy quasi-particle band, which is the spin density wave(SDW) formed by the nesting of the Fermi surface. $CeRh_2Si_2$ [3] and $CePd_2Si_2$ [4] are the examples of the former case, and $Ce(Ru_{1-x}Rh_x)_2Si_2$ ($0.03 < x < 0.35$)[5] and $Ce_{1-x}La_xRu_2Si_2$ ($0.08 < x < 0.25$)[6] are the examples of the latter case. In the latter case, an evident energy-gap forming at Fermi surface was found in the resistivity [7], which is a typical feature of the SDW.

In this paper, we discuss on an effect of 'negative' chemical pressure to the SDW phase in $Ce(Ru_{0.9}Rh_{0.1})_2Si_2$ by substitution Ge for Si. The substitution Ge for Si is consider to be equal to be applying negative pressure in $CeRu_2(Si_{1-y}Ge_y)_2$ system [9]. It is the same even in $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$ [10]. The base material $Ce(Ru_{0.9}Rh_{0.1})_2Si_2$ has the SDW phase below $T_N = 5.0$ K with the tetragonal $TrCr_2Si_2$ type crystal structure[7]. Single crytalline samples of $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$ were grown with the Czochralski method in a tri- or tetra-arc furnace. All samples were annealed in vacuumed($\sim 5 \times 10^{-6}$ Torr) crystal tubes at 1000 °C for more than 120 hours. Magnetization measurements were

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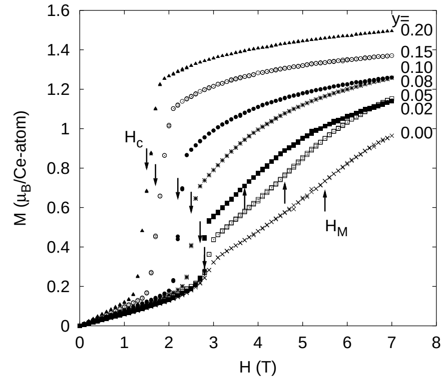

Fig. 1. The magnetization process of $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}$-$Ge_y)_2$ at 1.8 K are shown. The arrows represent the positions of H_m and HC for each Ge-concentration.

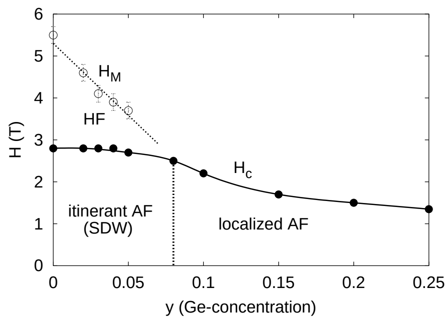

Fig. 2. The H-y phase diagram of $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}$-$Ge_y)_2$ at 1.8 K is shown. The solid line and the dashed line in the figure are guide to eyes.

performed by a commercial SQUID magnetometer (MPMS-7 manufactured by Quantum Design co., ltd.) at 1.8 K in the field range of $0 \leq H \leq 7$ T.

Figure 1 shows the magnetization processes of $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$ along the c-axis, which is magnetic easy axis, at 1.8 K. The magnetization processes are quite different qualitatively in low and high Ge-concentration region. In the low Ge-concentration region ($y < 0.08$), a two-step metamagnetic behavior at H_c and H_m is found; the SDW-Fermi Liquid(FL) transition occurs at H_c , and FL state becomes unstable and localized spin character recovers at H_m [8]. By substituting Ge for Si, H_m reduces very rapidly and disappears for $y > 0.08$. In the high ($y > 0.08$) Ge-concentration region, only one metamagnetic behavior at H_c is found, which is quite similar to the metamagnetic spin-flipping in the localized Ising spin antiferromagnets. In this region, the AF state seems to be constructed by well defined localized spins. The qualitative changes of magnetization process occurs continuously around $y = 0.08$.

The negative chemical pressure effect found in $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$ can be interpreted as follows. For $y = 0$, the Kondo temperature T_K is too high to be formed the magnetic order by RKKY interaction and the coherence of the heavy quasi-particle band is developed far above T_N . The SDW state is realized by the nesting of this heavy quasi-paricle band. T_K is decreasing by applying negative pressure, and the development of the coherence becomes unsatisfactory. Finally, the magnetic order of localized spins by RKKY interaction takes place before the development of the coherence of the quasi-particle band. The change of the character of the AF state seems to be crossover, not phase transition, with considering the continuous change of the magnetization process. We show the H-y phase diagram of $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$ at 1.8 K in Fig. 2.

On the basis of these experimental results we consider that we observed the crossover of the character of the AF state from "itinerant" to "localized" in $Ce(Ru_{0.9}Rh_{0.1})_2(Si_{1-y}Ge_y)_2$.

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References

[1] For example; R. A. Fisher, S. Kim, B. F. Woodfield, N. E. Phillips, L. Taillefer, K. Hasselbach, J. Flouquet, A. L. Giorgi, J. L. Smith, Phys. Rev. Lett. **62** (1989) 1411.

[2] For example; H. v. Löhneysen, J. Magn. Magn. Mater. **177-181** (1998) 12.

[3] S. Kawarazaki, M. Sato, Y. Miyako, N. Chigusa, K. Watanabe, N. Metoki, Y. Koike, M. Nishi, Phys. Rev. B **61** (2000) 4167.

[4] B. H. Grier, J. M. Lawrence, V. Murgai, R. D. Parks, Phys. Rev. B **29** (1984) 29.

[5] S. Kawarazaki, M. Sato, H. Kadowaki, Y. Yamamoto, Y. Miyako, J. Phys. Soc. Jpn. **66** (1997) 2473.

[6] P. Lejay et al., J. Phys. Cryst. Growth **130** (1993) 238

[7] Y. Miyako, T. Takeuchi, T. Taniguchi, S. Kawarazaki, K. Marumoto, R. Hamada, Y. Yamamoto, M. Ocio, P. Pari, J. Hammann, J. Phys. Soc. Jpn. **65** (1996) Suppl.B 12.

[8] C. Sekine, T. Sakakibara, H. Amitsuka, Y. Miyako, T. Goto, J. Phys. Soc. Jpn. **61** (1992) 4536

[9] H. Wilhelm, K. Alami-Yadri, B. Revaz, D. Jaccard, Phys. Rev. B **59** (1999) 3651

[10] K. Watanabe, Y. Tabata, T. Taniguchi, S. Kawarazaki, unpublished