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

Yoshikazu Tabata $^{a,1}$, Kenji Watanabe $^{a,2}$, Toshifumi Taniguchi $^{a}$ Chikahide Kanadani $^{a}$ Shuzo Kawarazaki $^{a}$

$^{a}$Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

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$_{0.1}$)$_2$Si$_2$, 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$_2$Si$_2$ [3] and CePd$_2$Si$_2$ [4] are the examples of the former case, and Ce(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ (0.03 < $x$ < 0.35)[5] and Ce$_{1-x}$La$_x$Ru$_2$Si$_2$ (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$_{0.1}$)$_2$Si$_2$ 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$_{0.1}$)$_2$Si$_2$ has the SDW phase below $T_N = 5.0$ K with the tetragonal TrCr$_2$Si$_2$ type crystal structure[7]. Single crystalline 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(∼5 × 10$^{-6}$ Torr) crystal tubes at 1000 °C for more than 120 hours. Magnetization measurements were

1 E-mail: yang@ltfridge.ess.sci.osaka-u.ac.jp
2 Present address: Technical Research and Development Institute, Japan Defence Agency, 2-2-1, Nakameguro, Meguro-ku, Tokyo 153-8630, Japan
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 $T_K$. The SDW state is realized by the nesting of this heavy quasi-particle 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$_y$)$_2$.

Acknowledgements

This work was supported by a Grant-In-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture, Japan.

References