Journal of Physics: Conference Series

La substitution effect to the heavy-fermion state in structure-disordered Ce-Ru alloys

To cite this article: Y Amakai *et al* 2012 *J. Phys.: Conf. Ser.* **391** 012002

Journal of Physics: Conference Series **391** (2012) 012002 doi:10.1088/1742-6596/391/1/012002

La substitution effect to the heavy-fermion state in structure-disordered Ce-Ru alloys

Y. Amakai[1], D. Yoshii[1], S. Murayama[1], H. Takano[1], N. Momono[1], Y. Obi[2] and K. Takanashi[2]

[1]Graduate School of Engineering, Muroran Institute of Technology, Muroran, Hokkaido, 050-8585, Japan

[2]Institute for Materials Research, Tohoku University, Sendai, 980-8577, Japan

E-mail: a-rain@mmm.muroran-it.ac.jp

Abstract. We have measured the magnetic susceptibility χ, specific heat C_p and resisitivity ρ on sputtered structure-disordered $Ce_{80-y}La_yRu_{20}$ alloys. The χ for $y \leq 70$ follows the Curie-Weiss law in the measurement temperature region. The paramagnetic effective magnetic moment p_{eff} is about 1.8 μ_B/Ce-atom for $y = 0$ and decreases with increasing the La-concentration (0.6 μ_B/Ce-atom for $y = 70$). The C_p follows linear relation of temperature above 7 K for all the samples. The specific heat coefficient γ shows about 200 mJ/molK^2 for $y = 0$ and decreases rapidly with increasing La-concentration. We found the -log T dependence of the ρ in the La-substituted samples. Magnitude and temperature range of the -log T contribution to the whole resistivity increase with increasing the La concentration. On the other hand, we have observed superconducting behaviors of the resistivity $\rho = 0$ and a large diamagnetization at low temperature ($T < 3$ K) for $y = 80$.

1. Introduction

Amorphous alloy system CeRu shows a heavy-fermion like behavior in the Ce-rich side and a superconductivity in the Ru-rich side, which was reported by Homma *et al*.[1] Recently, we have reported the resistivity[2], the specific heat[3] and susceptibility[4] for the structure-disordered (a-)Ce_xRu_{100-x} alloys in the wide Ce-concentration range x. In the Ce-rich side, the electronic specific heat coefficient γ shows a very large value ($\gamma \sim 220$ mJ/Ce-molK^{-2}, at $x = 80$). The resistivity of a-$Ce_{80}Ru_{20}$ follows a T^2 law with a large coefficient A in the low temperature region. However, the γ decreases rapidly with decreasing the Ce-concentration. In the Ru-rich region, the γ becomes less than 10 mJ/molK^{-2} and the resistivity becomes zero (T_c ~3.6 K, at $x = 15$). These facts suggest that a heavy fermion state and a superconducting state appeared, respectively, in the Ce-rich and Ru-rich sides of the structure-disordered system even with no translation symmetry. The susceptibility for $x \geq 39$ shows a Curie-Weiss paramagnetic behavior. The effective magnetic moment p_{eff} is about 1.8 μ_B/Ce-atom at $x = 80$ and decreases rapidly with decreasing the Ce-concentration ($p_{eff} \sim 0.6$ μ_B/Ce-atom, at $x = 39$). The susceptibility for $x \leq 23$ is almost independent of temperature except in the superconducting region. The valence of Ce estimated from recent measurement of x-ray absorption near edge structure is +3.1 in $x = 80$ and +3.45 in $x = 9$.[5] Therefore, it is considered that the 4f-electron of Ce is trivalent configuration in the Ce-rich region of the heavy-fermion state and mixed valence state in the Ru-rich

Journal of Physics: Conference Series **391** (2012) 012002 doi:10.1088/1742-6596/391/1/012002

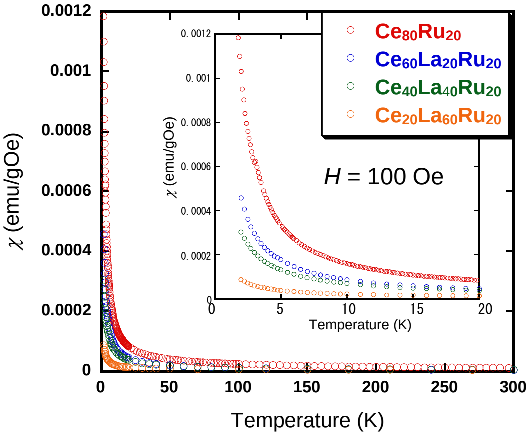

Figure 1. Temperature dependence of susceptibility χ for *a*-$Ce_{80\text{-}y}La_yRu_{20}$ alloys. The inset is lower temperature susceptibility.

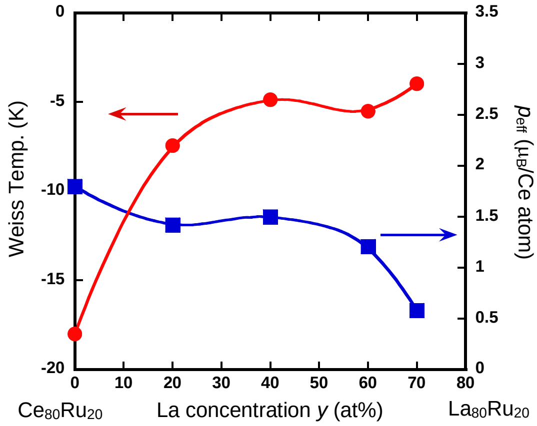

Figure 2. La-concentration dependence of the paramagnetic effective moment p_{eff} and Weiss temperature θ estimated from the Curie-Weiss law.

region of superconducting state. In this study, in order to clarify the effect of 4*f*-electron in *a*-Ce-Ru alloys system, we have measured the susceptibility χ, specific heat C_p and resistivity ρ for the *a*-$Ce_{80\text{-}y}La_yRu_{20}$ alloys (y = 0 - 80) substituted by La.

2. Experimental

Bulk ingots of $Ce_{80\text{-}y}La_yRu_{20}$ (y = 0, 20, 40, 60, 70 and 80) were made by arc melting from nominal amount of Ce 99.9 %, La 99.9 % (NIPPON YTTRIUM CO., LTD) and Ru 99.95 % (RARE METALLIC CO., LTD), in argon arc furnace. Amorphous $Ce_{80\text{-}y}La_yRu_{20}$ alloys were prepared by a dc sputtering method from the arc-melted ingots onto water-cooled Cu substrate. We confirmed a typical amorphous halo pattern from the x-ray diffraction measurement. The magnetic susceptibility was measured by using a commercial SQUID magnetometer (Quantum Design MPMS) from 2 to 300 K. The specific heat measurement was measured by using a relaxation method (Quantum Design PPMS) from 1.9 to 40 K. The electrical resistivity was measured by using a typical four-terminal method (Quantum Design PPMS) from 1.9 to 300 K.

3. Results and Discussion

Figure 1 shows temperature dependence of the susceptibility χ for *a*-$Ce_{80\text{-}y}La_yRu_{20}$ alloys. The χ for $y \leq 60$ increases with decreasing temperature, which follows the Curie-Weiss (CW) law in the measurement temperature region. The value of χ decreases with increasing La-concentration. Figure 2 shows the La-concentration dependence of the paramagnetic effective magnetic moment p_{eff} and Weiss temperature θ estimated from the CW law $N_{Ce}(p_{\mathrm{eff}})^2/3k_B(T\text{-}\theta)$, where N_{Ce} is the number of Ce-atom per gram. The value of p_{eff} is about 1.8 μ_B/Ce-atom for y = 0, and decreases with increasing the La-concentration. However, the p_{eff} for $20 \leq y \leq 60$ exhibits almost constant value ($p_{\mathrm{eff}} \approx 1.5$ μ_B/Ce-atom). With increasing La-concentration further, the p_{eff} decreases again and becomes about 0.6 μ_B/Ce-atom for y = 70. The value of θ shows about -18 K for y = 0 and the absolute value decreases with increasing the La-concentration. The θ for y >20 is almost constant value ($\theta \approx 5$ K). Figure 3 shows low temperature specific heat C_p over T *vs.* T^2 plots for *a*-$Ce_{80\text{-}y}La_yRu_{20}$ alloys. As shown in this figure, the C_p/T follows almost a T^2-linear relation above 7 K for all the samples. However, the C_p/T for all the samples increases at low temperature with decreasing temperature. We have estimated the γ-value extrapolated from a linear part above 7 K of C_p/T versus T^2 to T = 0. The inset of fig. 3 shows the La-concentration dependence of the specific coefficient γ and value of C_p/T at 2 K for

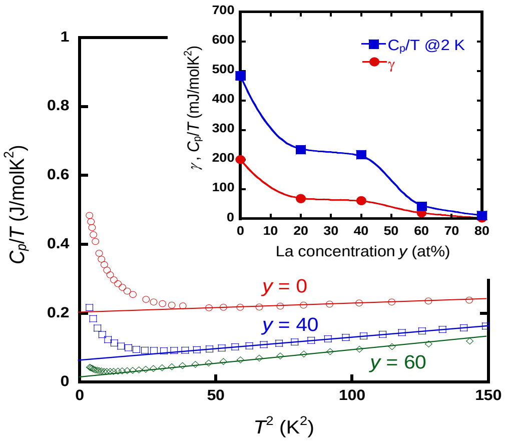

Figure 3. Low temperature specific heat C_p over T *vs.* T^2 plots for a-$Ce_{80-y}La_yRu_{20}$ alloys. The inset is La-concentration dependence of the specific heat coefficient γ and value of C_p/T at 2 K.

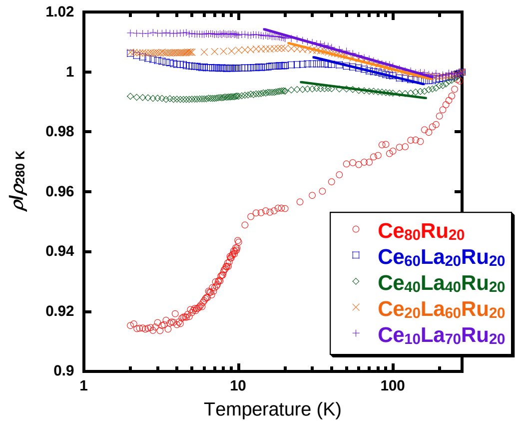

Figure 4. Temperature dependence of the resistivity for a-$Ce_{80-y}La_yRu_{20}$ alloys. The solid lines show a logarithmic guideline for the eyes.

a-$Ce_{80-y}La_yRu_{20}$ alloys. The γ-value shows about 200 mJ/molK^2 for $y = 0$ and decreases rapidly with increasing La-concentration. The γ for $y = 20$ and 40 is almost constant value (~ 50 mJ/molK^2). With increasing the La-concentration further, the γ decreases again and reach about 10 mJ/molK^2 at $y >$ 60. The C_p/T at 2 K exhibits the La-concentration dependence similar to the γ. However, the value of C_p/T at 2 K for $y \leq 40$ is a much larger than γ-value. On the other hand, the C_p/T at 2 K and γ for $y \geq 60$ are almost the same value. Figure 4 shows temperature dependence of the resistivity ρ/ρ_{280K} normalized at 280 K for $y \leq 70$. The ρ for each sample exhibits small temperature change in the whole temperature region and decreases with decreasing the T above 150 K. The ρ for $y = 0$ decreases with decreasing the temperature and a plateau in 10 K $\leq T \leq$ 20 K probably due to the Kondo effect. We found the –log T dependence of ρ in the higher temperature region by the La substitution for $y \geq 20$. The –log T contribution to the whole resistivity increases with increasing the La-concentration. This behavior suggests that the Kondo-scattering effect is enhanced by the La substitution. The ρ at $T <$ 7 K for $y = 0$ obeys the T^2 dependence with a large coefficient A. Since we have found the large γ value of the specific heat in $y = 0$, the T^2 dependence of the ρ in the low temperature region suggests formation of the heavy-fermion state. However, the T^2 dependence of the ρ was not observed by the La substitution. Therefore, the coherence necessary for the formation of the heavy-fermion state disappeared by the La substitution.

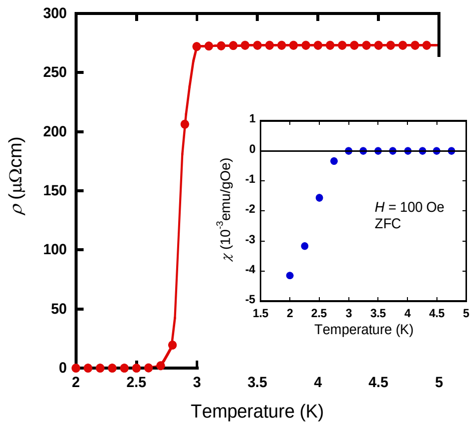

Figure 5. Temperature dependence of the low temperature resistivity ρ and susceptibility χ for $y = 80$.

Figure 5 shows the low temperature resistivity ρ and susceptibility χ for $y = 80$. The ρ at $T >$ 3K is almost independent of temperature. We have observed a rapid decrease of ρ in $T <$ 3 K and found zero resistivity in $T \sim$ 2.6 K. The χ for $y = 80$ shows a small value almost independent of temperature like the Pauli paramagnetic behavior except in the low temperature region. As shown in the inset of Fig. 5, a large diamagnetization was observed at $T < 3$ K. The zero resistivity with a rapid decrease of the ρ and a large diamagnetization of the χ suggest that the superconducting transition have occurred at $T = 2.6$ K.

In the a-Ce_xRu_{100-x} alloys case, according as the γ and p_{eff} rapidly decrease with decreasing the Ce-concentration, the superconductivity appeared in the Ru-rich region. The p_{eff} of the present a-$Ce_{80-y}La_yRu_{20}$ alloys does not show rapid decrease. Although the p_{eff} decreases down to 0.6 μ_B/Ce-atom in $y = 70$, the superconductivity does not occur up to $y = 80$. The γ for the present alloys decreases with decreasing the Ce-concentration, but does not become small very much in comparison to the case of the a-Ce-Ru alloy in the same Ce-concentration. The $\gamma = 61$ and 19 mJ/molK^2 for $y = 40$ and 60, respectively, whereas, $\gamma = 9.6$ and 3.9 mJ/molK^2 for the same Ce-concentration in a-Ce-Ru alloys with the superconductivity at $T_c = 1.1$ and 3.0 K, respectively.[3] The –log T dependence of ρ is observed in the higher temperature region and becomes larger depending on the La substitution. Therefore, the La substitution to the a-Ce-Ru alloy would make an increase of the Kondo temperature and make the local Kondo scattering effect clearer in the Ce diluted alloys.

Acknowledgements
This work was performed under the inter-university cooperative research program of the Institute for Materials Research, Tohoku University, and supported by a grant for Kidorui (Rare Earth) Program (2011) through Muroran Institute of Technology.

References

[1] Homma Y, Sumiyama K, Yamaguchi H and Suzuki K 1997 *J. Phys. Soc. Jpn.* **66** 1457
[2] Obi Y, Murayama S, Amakai Y, Okada Y and Asano K 2006 *Physica B* **378-380** 857
[3] Amakai Y, Murayama S, Takano H, Mizutani M, Asano K, Obi Y and Takanashi K 2007 *J. Magn. & Magn. Mater.* **310** 416
[4] Amakai Y, Murayama S, Obi Y, Takano H, Momono N and Takanashi K 2009 *J. Phys.:Conf. Ser.* **150** 042004
[5] Li Y, Onishi N, Nakai I, Amakai Y and Murayama S 2009 *J. Phys. Soc. Jpn.* **78** 094717