Evidence of a heavy fermion state in the disordered Ce-alloy system without translation symmetry

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We have measured the specific heat C_p and the resistivity ρ on sputtered amorphous $\operatorname{Ce}_x Y_{80-x} \operatorname{Mn}_{20}$ alloys. The low-temperature specific heat has been found to follow $C_p = \gamma T + \beta T^3$ for all the samples. The *T*-linear coefficient γ increases linearly with increasing Ce concentration and becomes very large ($\gamma > 100 \text{ mJ mol}^{-1} \text{ K}^{-2}$) in the Ce-rich region. The γ_{Ce} per Ce atom is about 200 mJ Ce-mol}^{-1} \text{ K}^{-2} and almost independent of the Ce concentration. The large γ_{Ce} has been interpreted in terms of the electronic specific heat owing to the 4*f*-electron mixed with conduction electrons. In the resistivity ρ , the $-\log T$ dependence has been observed at low temperature for $x \ge 51$, which suggests the formation of the dense Kondo state. The ρ vs *T* for $x \ge 62$ exhibits a maximum and decreases as $\rho \propto AT^2$ with a large coefficient *A* down to the low-temperature limit. These results suggest the formation of a heavy fermion state as the ground state for $x \ge 62$. We made the Kadowaki-Woods (KW) plot for $x \ge 62$ and found the A/γ^2 value to be much smaller than that of the conventional KW relation probably due to the *f*-orbital degeneracy.

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I. INTRODUCTION

Ce-based rare-earth and U-based actinoid intermetallic compounds show the Kondo lattice or the heavy fermion state, sometimes with secondary fascinating phases such as spin-density wave, non-Fermi liquid, and superconductivity.¹ The Kondo state is formed from mixing of the local f electron of the Ce or U atoms with conduction electrons as a local quasiparticle state. Then, the Kondo-lattice band takes place as the Bloch state at low temperature in case of the periodic coherent arrangement of the f-electron site, which is the Fermi-liquid heavy fermion ground state with a large effective mass of the f electron. Therefore, the experimental research for the heavy fermion has been done by using pure single crystals.

On the other hand, research on the heavy f electron of structure-disordered-type (amorphous) alloys has been reported by Sumiyama et al. for Ce-based amorphous (a-)Ce-Cu,²⁻⁴ a-Ce-Si,⁵ and a-Ce-Ru (Ref. 6) mainly in the Ce-poor region, where the heavy fermion nature appears in their crystalline counterpart. They showed the Curie-Weisstype paramagnetic behavior in the susceptibility down to the low-temperature limit without any definite magnetic transition. Their specific-heat results indicate the low-temperature specific heat $C_p = \gamma T + \beta T^3$ with the coefficient γ much larger ($\gamma > 100 \text{ mJ Ce-mol}^{-1} \text{ K}^{-2}$) than the case for usual amorphous magnetic alloys. The resistivity follows -log T dependence with a large residual resistivity, except for a-Ce-Ru 82at. %, which exhibits the superconductivity below 2.4 K.^{6,7} The authors suggested that the abovementioned characteristics show an evidence of a dense Kondo state due to the 4f electron of Ce at incoherent disordered sites. Furthermore, they claimed that the enhanced large γ value is due to effective-mass enhancement of itinerant 4f electrons at low temperature. It is possible that such itinerant electrons with a large γ arise from formation of a coherent-Kondo or heavy fermion state as the ground state even in structure-disordered alloys. In order to confirm this, it is necessary to find evidence of a Fermi-liquid band state, such as T^2 law in the low-temperature resistivity and *T*-linear dependent nuclear magnetic relaxation rate $1/T_1$ at low temperature. However, the T^2 dependence of the low-temperature resistivity in *a*-Ce-Cu and *a*-Ce-Si has not been observed.^{2–5} The authors pointed out that these results imply that the random atomic configuration suppresses the occurrence of not only the magnetic ordering but also the singlet state and masks the Fermi-liquid degeneracy of 4f electrons. Therefore, it is not clear whether the Fermi-liquid heavy fermion ground state exists or not in amorphous Ce alloys without translation symmetry.

Recently, we have reported several low-temperature properties for a-Ce-Mn alloys.⁸⁻¹⁰ Temperature dependence of the susceptibility χ (Ref. 8) in *a*-Ce-Mn alloys on the Cepoor side shows a typical Curie-Weiss behavior above 25 K. With decreasing temperature below 25 K, it shows a maximum, and a separation between field-cooled and zero-fieldcooled susceptibilities below around the maximum temperature. These behaviors indicate occurrence of a spin-glass phase in the Ce-poor side. This is probably due to the magnetic interaction between the Mn magnetic moments because the spin-glass transition temperature T_g increases with increasing Mn concentration. However, the spin-glass transition disappears and the Curie-Weiss-type paramagnetic behavior remains down in the low-temperature limit when the Ce concentration increases more than 60 at. %. The lowtemperature specific heat of a-Ce-Mn alloys corrected for the phonon contribution depends on a T-linear law with a very large coefficient $\gamma > 100$ mJ mol⁻¹ K⁻² on the Ce-rich paramagnetic side.⁹ The resistivity ρ of *a*-Ce-Mn is large and has small temperature dependence in the whole temperature region as in the case for usual amorphous alloys.¹⁰ On the Ce-rich side the low-temperature resistivity follows a T^2 law with a large coefficient A. From these results, it has been suggested that an itinerant heavy fermionlike 4f state occurs at low temperature as a Fermi-liquid ground state in the structure-disordered system after formation of a dense Kondo state.

In this work, in order to confirm the heavy fermionlike behavior on the Ce-rich side of a-Ce-Mn, we have investigated the Ce-concentration dependence of the lowtemperature specific heat and resistivity on amorphous $(a-)Ce_{x}Y_{80-x}Mn_{20}$ alloys (x=10, 30, 50, 60, 70, and 80), where the dense Kondo state of the 4f electron should be affected by the substitution of nonmagnetic Y for Ce. We found a large electronic specific-heat coefficient γ_{Ce} per Ce atom, $-\log T$ dependence of the resistivity, and a large coefficient A for the T^2 dependence of the resistivity in the lowtemperature limit on *a*-Ce_xY_{80-x}Mn₂₀ alloys in the Ce-rich region. From the magnitude of γ , ratio $\Delta \rho / \rho$ of the contribution of the $-\log T$ dependence to the resistivity and A, respectively, we concluded that the observed lowtemperature behaviors for the present disordered Ce alloys in the Ce-rich region are due to the formation of a heavy fermion band state as the ground state after a local Kondo effect.

II. EXPERIMENTS

Bulk ingots of $Ce_x Y_{80-x} Mn_{20}$ (x=10, 30, 50, 60, 70, and 80) were made by arc melting from nominal amount of Ce 99.9%, Y 99.9%, and Mn 99.9%, in an argon-arc furnace. Amorphous $Ce_{x}Y_{80-x}Mn_{20}$ was prepared by a dc high-rate sputtering method from the arc-melted ingots onto the watercooled Cu substrate. After removal of the copper substrate by mechanical polishing, the chemical compositions of these alloys were determined by induction-coupled plasma analysis as Ce₁₀Y₇₀Mn₂₀, Ce₂₈Y₅₂Mn₂₀, Ce₅₁Y₃₀Mn₁₉, Ce₆₂Y₁₉Mn₁₉, Ce₇₆Y₉Mn₁₅, and Ce₈₁Mn₁₉, respectively. From the x-ray diffraction measurement on each obtained sample, we confirmed a typical amorphous halo pattern without definite crystalline peaks. The specific heat was measured by using a commercial light-chopped ac calorimeter from 5 to 300 K. A pure Cu plate (5N) was used as a reference material in order to estimate the absolute value of the specific heat. The electrical resistivity was measured by using the four-terminal ac method from 2 to 280 K.

III. RESULTS

Figure 1 shows the specific heat C_p over T vs T^2 plots for the present a-Ce_xY_{80-x}Mn₂₀ alloys, where we present the observed Ce concentration as x, and specify the Y concentration as 80-x and the Mn concentration as 20, for convenience. As shown in this figure, C_p follows a function of $\gamma T + \beta T^3$ above 10 K for all the samples as usually observed in amorphous magnetic alloys, where the first term is an electronic or magnetic contribution and the second term should be a phonon contribution. Figure 2 shows the Ceconcentration dependence of the coefficient γ in the unit of 1 mol of Ce+Y+Mn atoms and γ_{Ce} in 1 mol of Ce for a-Ce_xY_{80-x}Mn₂₀ alloys. The value of γ for x=10 is 20 mJ mol⁻¹ K⁻², which is a little larger than the γ for usual amorphous magnetic alloys. The γ value increases almost



FIG. 1. C_p/T vs T^2 plots at low temperature for a-Ce_xY_{80-x}Mn₂₀ alloys. The dotted lines show $C_p/T = \gamma + \beta T^2$ relation.

linearly with increasing Ce concentration and becomes $\gamma = 172 \text{ mJ mol}^{-1} \text{ K}^{-2}$ for Ce₈₁Mn₁₉. The γ_{Ce} per Ce atom is about 200 mJ Ce-mol⁻¹ K⁻² and almost independent of Ce concentration as shown in Fig. 2. Therefore, the large γ observed in the Ce-rich region of the present alloys should be due to the Ce contribution. Temperature dependence of the susceptibility χ for a-Ce_xY_{80-x}Mn₂₀ on the Ce-poor region of $x \leq 51$ shows a spin-glass transition at low temperature as has been reported in a-Ce-Mn.^{8,11} The χ on the Ce-rich side of $x \geq 62$ shows a Curie-Weiss-type paramagnetic behavior in the measurement temperature region. Therefore, the large γ in the Ce-rich side would probably arise from the electronic contribution of 4f electrons of Ce mixed with conduction electrons, namely, a dense Kondo state.

Figure 3 shows the resistivity ρ as a function of log T for *a*-Ce_xY_{80-x}Mn₂₀ alloys. The ρ exhibits small temperature



FIG. 2. The specific-heat coefficient γ in 1 mol of Ce+Y+Mn atoms and γ_{Ce} in 1 mol of Ce vs Ce concentration x for a-Ce_xY_{80-x}Mn₂₀ alloys.



FIG. 3. Temperature dependence of the resistivity ρ as a function of log *T* for *a*-Ce_xY_{80-x}Mn₂₀ alloys. The solid lines show a logarithmic guideline for the eyes.

change within about 15% in the whole temperature region. Such small temperature dependence is one of the characteristics for amorphous alloys. The ρ on the Ce-poor side of $x \le 28$ shows a large residual resistivity $\rho_0 > 150 \ \mu\Omega \ {\rm cm}$ and a monotonic increase with decreasing temperature as is observed in typical amorphous alloys. In the Ce-rich side of $x \ge 51$, the $\rho_0 \sim <150 \ \mu\Omega$ cm and the ρ decreases with decreasing temperature on the higher temperature side above 120 K. These behaviors are in agreement with the Mooij's empirical law.¹² For $x \ge 51$, $-\log T$ dependence of ρ is found in the lower temperature side below 30 K for each sample. In the range of $x \ge 62$, ρ -T curve makes a maximum at T_{max} after obeying the $-\log T$ dependence and then decreases with further decreasing temperature. In the low-temperature limit the ρ for $x \ge 62$ obeys the relation $\rho - \rho_0 = AT^2$ as shown in Fig. 4 with the coefficient A that is much larger compared with usual metals with exchange-interacting conduction electrons. Since we have found the large γ value of the specific heat in the Ce-rich paramagnetic region due to the 4f electron mixed with conduction electrons, the $-\log T$ dependence in the resistivity strongly suggests formation of the dense Kondo state. The large AT^2 term of ρ in the low-temperature limit after the $-\log T$ dependent region is often observed in typical heavy fermion systems.¹³ The present behavior of the resistivity in the Ce-rich side of the a-Ce_xY_{80-x}Mn₂₀ alloys suggests formation of a coherent-Kondo or a heavy fermion state as the ground state.



FIG. 4. The resistivity $\rho - \rho_0$ vs T^2 plots at low temperature for the Ce-rich side of a-Ce_xY_{80-x}Mn₂₀ alloys.

IV. DISCUSSION

The T^2 dependence in the low-temperature resistivity has been sometimes observed in amorphous alloys as shown in amorphous Ni-P (Ref. 14) and Pd-Ni-P,¹⁵ which has been explained by well-known Ziman picture based on the Debye-Waller amorphous structure factor. In these cases, the coefficient A of the T^2 dependence is about $10^{-4} \ \mu\Omega$ cm K⁻². The $-\log T$ dependence of the resistivity also has been observed in several amorphous alloys. In case of nonmagnetic amorphous alloys,¹⁶ the ratio $\Delta \rho / \rho$ of the $-\log T$ contribution to the whole resistivity is estimated as small as the order of 10^{-5} . This has been explained by a local lattice-vibration model based on the disordered structure. The $\Delta \rho / \rho$ for the magnetic amorphous alloys¹⁷ have been observed as $10^{-3}-10^{-4}$, which is larger than the case of nonmagnetic amorphous alloys. In this case, the magnetic disorder factor is supposed to be concerned. On the other hand, Hasegawa et al. have found a large $-\log T$ dependence of $\Delta \rho / \rho \approx 10^{-2}$ in *a*-Pd-Si alloy with small amount of Cr or Mn.¹⁸ Since $\Delta \rho / \rho$ is $10^{-1}-10^{-2}$ in usual crystalline Kondo alloys, they pointed out that the $-\log T$ term in *a*-Pd-Si is due to the Kondo effect in the amorphous alloy.

In the present *a*-Ce_xY_{80-x}Mn₂₀ alloys for $x \ge 62$, the coefficient *A* of the T^2 dependence is found to be $10^{-2} \ \mu\Omega$ cm K⁻². This value is very large in contrast to the case of above-mentioned amorphous alloys and almost the same order as in the case of intermediate heavy fermion such as CeRu₂ (Ref. 19) and CePd₃ (Ref. 20). Also, the ratio $\Delta \rho / \rho$ of the $-\log T$ contribution to the whole resistivity in a-Ce_xY_{80-x}Mn₂₀ for $x \ge 51$ is about 10⁻², which is larger than the case of nonmagnetic amorphous case and almost agrees with the case of *a*-Pd-Si with Cr or Mn. It should be noted that in case of the low Ce concentration for a-Ce_xY_{80-x}Mn₂₀ single-site Kondo-scattering effect at the Ce site is weak and that the $-\log T$ contribution would be smeared out by the spin-glass freezing. Thus, we can conclude that the present large T^2 dependence after the large $-\log T$ dependence in the Ce-rich region originates from magnetic scattering under formation of a coherence and itinerancy for the mixed 4f state after the dense Kondo effect at each Ce site. We believe that the observed large γ in specific heat, the T^2 law, and $-\log T$ dependence in resistivity are in total the first clear evidence of a heavy fermion state as the ground state in the non-Bloch system without translation symmetry. Very recently, a study of ⁵⁵Mn NMR in *a*-Ce₆₂Y₁₉Mn₁₉ by Niki *et al.*²¹ shows that the spin-lattice relaxation rate $1/T_1$ has almost T-linear temperature dependence at low temperature, namely, the Korringa relation, suggesting existence of the density of states of heavy fermion band on the Fermi surface and a Fermi-liquid state of 4f electrons as the ground state.

We consider that the resistivity maximum below the $-\log T$ -dependent temperature region is due to initial evolution of the coherence between the Kondo 4*f* electrons in the disordered system. Then, we can define the temperature T_{max} at the resistivity maximum as an initial measure of the formation of a heavy fermion state for the disordered system, namely, a Fermi-liquid state in the non-Bloch Kondo system. Figure 5 shows the Ce-concentration dependence of the T_{max} and coefficient *A* of the T^2 dependence in ρ for the



FIG. 5. T_{max} and A vs Ce concentration for the Ce-rich side of a-Ce_xY_{80-x}Mn₂₀ alloys. The inset is A vs $(1/T_{\text{max}})^2$ plots.

Ce-rich side. The $T_{\rm max}$ increases with increasing Ce concentration and the *A* decreases linearly with the Ce concentration. We found a linear relation between $(1/T_{\rm max})^2$ and *A* as shown in the inset of Fig. 5. This fact states that the resistivity ρ is scaled as $\rho - \rho_0 = \alpha (T/T_{\rm max})^2$ with a composition-independent coefficient α in the scaled temperature region of $T/T_{\rm max} < 0.4 - 0.6$ in the present case. Therefore, the $T_{\rm max}$ would be one of the important indices for determining the *A* value in the present disordered heavy fermion system.

The a-Ce_xY_{80-x}Mn₂₀ alloys in the Ce-rich region are thus found to indicate characteristic behaviors of the heavy fermion in the low-temperature limit. In order to complete the heavy fermion physics for the present disordered system, we have attempted to make the Kadowaki-Woods (KW) plot²² from the specific-heat coefficient γ and coefficient A of T^2 law in resistivity of the present alloys as shown in Fig. 6. The solid line is the KW relation of $A/\gamma_{Ce}^2 = 1.0 \times 10^{-5} \ \mu\Omega$ cm Ce-mol² K² mJ⁻², which is satisfied for usual heavy fermion systems as shown for typical compounds in this figure. As shown in this figure, the present A/γ_{Ce}^2 values of $0.42 \times 10^{-6} \ \mu\Omega$ cm Ce-mol² K² mJ⁻² for a-Ce₈₁Mn₁₉, $1.1 \times 10^{-6} \ \mu\Omega$ cm Ce-mol² K² mJ⁻² for a-Ce₇₆Y₉Mn₁₅, and $1.8 \times 10^{-6} \ \mu\Omega$ cm Ce-mol² K² mJ⁻² for a-Ce₆₂Y₁₉Mn₁₉ located under the KW relation. They seem to approach the KW relation accordingly with substituting Y for Ce. The deviation from the KW relation has been observed in the Yb-based heavy fermion compounds so far.²³ In order to explain the deviation, Kontani and Tsujii et al. have recently proposed the generalized KW relation $A/\gamma^2 = 1.0 \times 10^{-5}/[N(N-1)/2]$, where the degree of the orbital degeneracy N of f electrons was taken into account.^{24,25} By using the generalized KW relation with the degeneracy N=2-8, the experimental data of Yb-based heavy fermion compounds have been well explained. This generalized formula converges the conventional KW relation $A/\gamma^2 = 1.0 \times 10^{-5} \ \mu\Omega \ \text{cm} \ \text{Ce-mol}^2 \ \text{K}^2 \ \text{mJ}^{-2}$ when N=2, which means no orbital degeneracy for typical heavy f electrons. In case of the full degeneracy in Ce-based heavy fermion compounds with the total angular momentum J=5/2, N is equals to six and the A/γ^2 is calculated as



FIG. 6. Log *A* vs log γ plots (the Kadowaki-Woods plot) for the Ce-rich side of a-Ce_xY_{80-x}Mn₂₀ alloys. The open squares present usual heavy fermion compounds. The filled circles present a-Ce_xY_{80-x}Mn₂₀ alloys. The solid line is the Kadowaki-Woods relation. The dotted line represents the generalized Kadowaki-Woods relation where the orbital degeneracy *N* is six.

 $0.67 \times 10^{-6} \ \mu\Omega$ cm Ce-mol² K² mJ⁻² shown by dotted line in Fig. 6. Our experimental result approximately agrees with this value. In the present structure-disordered system, the local atomic environment effect might be homogenized because of the random arrangement of the surrounding atoms and the resulting total environment would become highly symmetric. In such a case, the effective crystalline field is supposed to be small and the 4*f* orbital of the Ce ion can be fully degenerated as N=6 (Ref. 25). Since the $T_{\rm max}$ of the low-temperature resistivity decreases with increasing Y concentration as shown in Fig. 5, substitution of Y in *a*-Ce-Mn tends the prevention of the c-*f* mixing or the formation of the heavy fermion state. Then, the heavy *f* electron would be localized with the resolution of the *f*-orbital degeneracy due to the crystalline-field splitting. When, the orbital degeneracy is resolved, the c-*f* mixing and the Kondo temperature might further decrease as Yamada *et al.* have pointed out.²⁶ Thus, the initial full degeneracy of N=6 for *a*-Ce-Mn is expected to decrease with the Y concentration and the A/γ^2 would approach the line of conventional KW relation (N=2) with increasing Y concentration.

V. CONCLUSION

In conclusion, we have investigated the low-temperature specific heat and the resistivity for the a-Ce_xY_{80-x}Mn₂₀ alloys with the substitution of the nonmagnetic Y. The specific heat C_p has been found to follow a function of $\gamma T + \beta T^3$ at low temperature in each Ce concentration, where the electronic specific-heat coefficient γ increases linearly with increasing Ce concentration and becomes very large in the Cerich paramagnetic region. The γ_{Ce} per Ce atom is about 200 mJ Ce-mol⁻¹ K^{-2} and almost independent of the Ce concentration. In the resistivity ρ in the Ce-rich region of $x \ge 51$, a large -log T dependence has been observed, which suggests formation of a dense Kondo state. The ρ -T curve for $x \ge 62$ exhibits a maximum at T_{max} and then decreases with further decreasing temperature according to T^2 dependence with a large coefficient A. These experimental results are clear evidence of the formation of the heavy fermion state as the ground state even in the structure-disordered system. The resistivity is scaled as $\rho - \rho_0 = \alpha (T/T_{\text{max}})^2$ with a composition-independent coefficient α in the scaled temperature region of $T/T_{\text{max}} < 0.4-0.6$. The KW plot from the coefficients A and γ for $x \ge 62$ shows the A/γ^2 value that is much smaller than that of the normal KW relation probably because of the *f*-orbital degeneracy. The A/γ^2 tends to approach the KW relation due to the resolution of the degeneracy by substituting Y for Ce.

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