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Thermal and Magnetic Properties in $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ Intermetallic Compounds

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The magnetic and thermal properties of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ compounds have been studied using specific heat, dc magnetization, and ac susceptibility measurements. All these compounds are isomorphic with the MgCu_2 Laves phase, and the lattice parameter decreases almost linearly with the increasing Er concentration x . The dc magnetic susceptibility follows the Curie-Weiss law, and the Weiss temperature continuously changes from $\Theta = -24$ K for $x = 0$ to $\Theta = 15$ K for $x = 1$, indicating a change from antiferro-magnetism to ferromagnetism. Θ changes from negative to positive at around $x = 0.2$ where a field-induced metamagnetic transition disappears. The magnetic ordering state continuously changes with the change in x from antiferromagnetic to ferromagnetic through a spin-glass-like behavior.

KEYWORDS: spin glass, heat capacity, susceptibility, CeAl_2 , ErAl_2

1. Introduction

The magnetic properties of intermetallic compounds containing rare-earth elements have been an object of great interest.¹⁻³⁾ The binary compounds of RAl_2 (R: rare earth) are the prototypes of rare-earth compounds with non magnetic metals, and show a wide range of electronic and magnetic properties by changing R. Many RAl_2 compounds melt congruently, so it is easy to grow them even in the form of single crystals. The crystal structure is relatively simple: isomorphic MgCu_2 type with the space group $Fd\bar{3}m$ (C15 Laves phase).⁴⁾ The interest

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in them is enhanced by the fact that compounds in which $4f$ electrons are localized are often model systems in which quantitative analysis can be performed, allowing for an accurate determination of the relevant interactions. Almost all RAl_2 compounds show ferromagnetic ordering with different Curie temperatures T_C .^{5,6)} It is well known that the exchange coupling between the localized $4f$ -electron shells of rare-earth ions is due to the conduction electrons and is called the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction.

On the other hand, CeAl_2 is a special case among the RAl_2 compounds. Since the f -level of CeAl_2 is energetically close to the conduction-electron band, both magnetic and hybridization interactions between f levels and the conduction-electron band are vital for the determination of the ground state. Considerable attention has been focused on CeAl_2 as a system that shows the competition between these magnetic and nonmagnetic interactions. An antiferromagnetic ordering due to a spin density wave occurs at $T_N = 3.8 \text{ K}$,⁷⁻⁹⁾ whereas Kondo-type conduction-electron screening of the localized moments occurs above T_N , which is suppressed by applying pressure since T_N is merged to the Kondo temperature T_K .⁸⁾ Below T_N , a pronounced jump in the magnetization has been observed at a magnetic field of approximately 5 T in a CeAl_2 single crystal. It indicates a metamagnetic phase transition from the antiferromagnetic ordering phase to the paramagnetic ordering phase.¹⁰⁾ At higher magnetic phases, the magnetization curve along the easy axis $[111]$ is the highest.

Therefore, the substitution of Ce by another rare-earth element R causes changes in the magnetic interaction. If we choose R such that RAl_2 shows a ferromagnetic transition, interesting magnetic properties are expected to be observed in the pseudobinary system $\text{Ce}_{1-x}\text{R}_x\text{Al}_2$ because of the competition between the ferromagnetic interaction of RAl_2 ($x = 1$) and the antiferromagnetic interaction of CeAl_2 ($x = 0$). In particular, ErAl_2 exhibits ferromagnetic ordering at approximately $T_C = 13 \text{ K}$,¹¹⁾ which is relatively close to the T_N of CeAl_2 compared with the T_C of the other RAl_2 compounds. Moreover, the easy axis of ErAl_2 is $[111]$,^{12,13)} which is the same as that of CeAl_2 in the antiferromagnetic ground state. These findings indicate that it is relatively easy to tune the ground state of the pseudobinary system $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ by changing the amounts of Er and Ce.

In this paper, we present the crystallography, magnetization, and specific heat of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ with $0 < x < 1$. We also describe the influence of Er substitution on the magnetic properties of the $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ system, which presents a complex magnetic phase diagram with antiferromagnetism, ferromagnetism, and spin-glass states.

2. Experimental Methods

Single crystals of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ were grown by the Czochralski pulling method from a melt of stoichiometric amounts of the constituent elements in a tetra-arc furnace. All ingots were confirmed to be single crystals from their Laue patterns. The crystal structure and lattice parameter were determined using X-ray powder diffraction analysis. The dc magnetization was measured in a magnetic field with the easy axis [111], using the Quantum-Design MPMS. The ac magnetic susceptibility was measured at a modulation field along [111] using the Quantum-Design PPMS. The specific heat was measured by thermal relaxation using the Quantum-Design PPMS.

3. Results and Discussion

The crystal structures of the pseudobinary alloy system $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ are confirmed to be of the cubic C15 Laves phase. The lattice parameters a of CeAl_2 and ErAl_2 are 8.066 and 7.791 Å, respectively, which are comparable with those in a previous report.⁴⁾ For $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$, a decreases with increasing x , which is due to the lanthanoid contraction. Figure 1 shows the cell volume variation of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ as a function of the Er concentration x . A linear Vegard law is obtained without significant changes in volume.

Figure 2 shows the temperature dependence of the specific heat $C(T)$ of the samples with different x . For CeAl_2 , an anomaly occurs at $T_N = 3.8$ K owing the antiferromagnetic phase transition. Above T_N , the coefficient of the linear term in the specific heat is calculated to be $\gamma = 112$ mJ/mol·K², which is comparable to the previously reported value of $\gamma = 135$ mJ/mol·K².¹⁴⁾ The inset of Fig. 2 shows the detailed behavior at temperatures below 6 K. With increasing x , the sharp peak becomes broad, and the temperature at which the peak is observed decreases with increasing Er concentration. For $x = 0.4$, there is no peak in $C(T)$, indicating that the antiferromagnetic ordering is suppressed. On the other hand, a peak appears again on the $C(T)$ curve of $x = 0.6$ at approximately 6.6 K. As the amount of Er substitution increases from $x = 0.6$, the peak becomes sharper. The temperature of the peak increases and is determined to be 12.5 K for ErAl_2 ($x=1$), which is consistent with the ferromagnetic transition temperature $T_C = 13$ K.¹¹⁾

Figure 3 shows the magnetic field dependence of the magnetization $M(B)$ of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ at 2 K. For $x = 0$, $M(B)$ increases linearly with increasing magnetic field up to 5 T. The metamagnetic transition due to the collapse of antiferromagnetism is observed at $B_M \sim 4.6$ T. To clarify this anomaly, the first derivative of $M(B)$, dM/dB , was calculated at 2 K. As shown in the inset of Fig. 3, B_M corresponds to the magnetic field exhibiting the peak. With

Fig. 1. Concentration x dependence on the cell volume $V = a^3$ in $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$. The solid line is the least-squares fit of the function $V = 525.4 - 53.5 \times x$ ($\text{\AA}^3$).

increasing the amount of Er substitution, the anomaly becomes broad, and B_M is suppressed. This will be discussed later.

The magnetization of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ increases with increasing x , since the localized magnetic moment of Er^{3+} is larger than that of Ce^{3+} . For $x = 1$, the ferromagnetic behavior with hysteresis is observed at 2 K, which was not observed in previous studies. The magnetization at 7 T in ErAl_2 is $7.7 \mu_B/\text{Er}^{3+}$, which is in good agreement with previous reports.^{12,13)}

Figure 4 shows the temperature dependence of the inverse susceptibility $\chi^{-1}(T)$ of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$. In all compounds, $\chi^{-1}(T)$ follows the Curie-Weiss law above 50 K. For $x=0$, the effective magnetic moment μ_{eff} and the Weiss temperature Θ_W are observed to be $2.3\mu_B$ and -24 K, respectively. μ_{eff} is close to that of the Bohr magneton of Ce^{3+} , $\mu_{\text{Ce}^{3+}} = 2.54\mu_B$. As x increases, both μ_{eff} and Θ_W increase. For $x = 1$, μ_{eff} and Θ_W are observed to be $9.7\mu_B$ and 15 K, respectively. μ_{eff} is close to that of the Bohr magneton of Er^{3+} , $\mu_{\text{Er}^{3+}} = 9.59\mu_B$.

The x dependences of the metamagnetic transition field (B_M), effective Bohr magneton

Fig. 2. Temperature dependence of magnetic specific heat in $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$. The inset shows the extended figure at low temperature.

number (μ_{eff}), and paramagnetic Curie temperature (Θ_{W}) are shown in Figs. 5 (a), 5 (b), and 5 (c), respectively. The B_{M} of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ decreases linearly from $B_{\text{M}} = 4.6$ T for $x = 0$ as Er concentration increases. It appears that B_{M} disappears at $x \sim 0.1$. μ_{eff} is described by the function

$$\mu_{\text{eff}} = (\mu_{\text{Ce}^{3+}}^2 \times (1 - x) + \mu_{\text{Er}^{3+}}^2 \times x)^{\frac{1}{2}} (\mu_{\text{B}}/\text{atom}), \quad (1)$$

which is shown as the solid line in Fig. 5 (b). It indicates that the magnetic moment comes from the average of the magnetic moments of the rare earth elements in the $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ ion in the paramagnetic region. As shown in Fig. 5 (c), Θ_{W} also increases with x . The negative Θ_{W} for $x < 0.2$ is consistent with the existence of the antiferromagnetic ground state in CeAl_2 . We find that the sign of Θ_{W} changes from negative to positive at around $x = 0.2$, indicating that the antiferromagnetism of CeAl_2 changes to ferromagnetism by substituting Er for Ce.

To investigate the magnetic ordering state in detail, we performed ac susceptibility measurements for several frequencies. Figure 6 shows the temperature dependence on the real component $\chi'(T)$ of ac susceptibility for a frequency of 100 Hz. All $\chi'(T)$ curves of the $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ compounds show similar behaviors, and have a single maximum. Here, we de-

Fig. 3. Magnetic field dependence of magnetization at 2 K in $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$. Inset: the magnetic field dependence of first derivative of $M(B)$ at 2 K in CeAl_2 . The magnetic field was applied along the easy axis [111].

fine T_M as the temperature where the $\chi'(T)$ curve shows a maximum. For CeAl_2 ($x = 0$), a peak is clearly visible at $T_M \sim 3.8\text{ K}$, which corresponds to the antiferromagnetic transition temperature observed in the heat capacity in Fig 2. Although T_M decreases as Er concentration x increases from 0 to 0.2, it increases for $x > 0.2$ where antiferromagnetism ordering disappears.

It is found that a $\chi'(T)$ exhibits a significant frequency-dependent shift of T_M for several compounds. Figure 7 shows a typical example of the $\chi'(T)$ curve for $\text{Ce}_{0.9}\text{Er}_{0.1}\text{Al}_2$ below 4.0 K for several frequencies. All the $\chi'(T)$ curves exhibit pronounced maxima, and both amplitude and position depend on the frequency of the applied magnetic field, particularly in the low-frequency range. This result indicates the formation of a spin-glass state in $\text{Ce}_{0.9}\text{Er}_{0.1}\text{Al}_2$ with a spin freezing temperature of $T_M = 2.56\text{ K}$ (at a frequency of $f = 100\text{ Hz}$) determined from the peak position of $\chi'(T)$.

Fig. 4. Temperature dependence of the inverse magnetic susceptibility in $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$.

Here, we inspected the spin dynamics by applying a simple formula, which corresponds to the shift of the ac-susceptibility maxima per frequency decade,

$$\delta T_M = \frac{\Delta T_M}{T_M \Delta \log f}. \quad (2)$$

The obtained δT_M 's are 0.0047, 0.021, and 0.042 for $x = 0.08$, 0.1, and 0.2, respectively. These values are comparable to those reported for other metallic spin-glass systems, e.g., CuMn: 0.005,¹⁵⁾ AuFe: 0.010,¹⁵⁾ U_2RhSi_3 : 0.008,¹⁶⁾ URh_2Ge_2 ,¹⁷⁾ $\text{PrNi}_{1-x}\text{Cu}_x\text{Al}$: 0.006 - 0.012,¹⁸⁾ and Nd_2AgIn_3 : 0.015.¹⁹⁾ To investigate the nature of the spin-glass state in greater detail, the well-known Vögel-Fulcher law²⁰⁾ was applied to the data as follows:

$$f = f_0 \exp \frac{-E_a}{k_B(T_M - T_0)}, \quad (3)$$

where f is the applied frequency, f_0 is the characteristic frequency, E_a is the activation energy, which determines the energetic barrier for spins to align with the external magnetic field, T_0 is the Vögel-Fulcher temperature, which corresponds to the interspin or intercluster interaction, and k_B is the Boltzmann constant. We tested various f_0 values near the characteristic value of 10^{13} Hz, which is typical for a spin-glass system.²¹⁾ Although the activation energies change upon varying f_0 , they still lie in reasonable ranges when considering the freezing temperatures. Figure 8 shows the fit of freezing temperatures using Eq. (3) for the $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ series of $x = 0.08$, 0.1, and 0.2, where the f_0 parameter is fixed at 10^{13} Hz. The

Fig. 5. (a) Metamagnetic transition field B_M , (b) Bohr magneton μ_{eff} , and (c) Weiss temperature Θ_W vs Er concentration in $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$.

slope gives the activation energy E_a . The intercept with the y-axis corresponds to T_0 . The obtained parameters are summarized in Table I. In general, the activation energies E_a are one order higher than the values of T_0 for compounds that show the spin-glass-like behavior. In the case of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$, the data between $x=0.1$ and 0.4 provide evidence of the formation of a collective spin-glass-like state.

Fig. 6. Temperature dependence of ac susceptibility χ' at 100 Hz for $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$.

Table I. Values of parameters obtained from fitting of freezing temperatures using the Vögel-Fulcher law for $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ compounds and several spin-glass systems.

	E_a/k_B (K)	T_0 (K)
$\text{Ce}_{0.96}\text{Er}_{0.04}\text{Al}_2$	0	3.18
$\text{Ce}_{0.92}\text{Er}_{0.08}\text{Al}_2$	3.33	2.60
$\text{Ce}_{0.9}\text{Er}_{0.1}\text{Al}_2$	14.3	2.00
$\text{Ce}_{0.8}\text{Er}_{0.2}\text{Al}_2$	28.8	1.36
$\text{Ce}_{0.6}\text{Er}_{0.4}\text{Al}_2$	13.1	3.42
$\text{Ce}_{0.4}\text{Er}_{0.6}\text{Al}_2$	5.42	6.32
$\text{PdMn}_{8\%}$ ¹⁵⁾	39	3.6
$\text{AuFe}_{10\%}$ ¹⁵⁾	81	29.1
$\text{La}_{1-x}\text{Gd}_x\text{Al}_2$ ¹⁵⁾	4.6	0
$\text{PrNi}_{0.5}\text{Cu}_{0.5}\text{Al}$ ¹⁸⁾	28.8	3.0
Nd_2AgIn_3 ¹⁹⁾	111.4	9.7

Finally, considering the present data, the magnetic phase diagram of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ is compiled in Figure 9. In general, the magnetic susceptibility of Kondo compounds increases with decreasing temperature, reaches a maximum, and then decreases to a modestly large value at $T = 0$.²²⁾ This general temperature dependence is described well by a Bethe Ansatz solu-

Fig. 7. Temperature dependence of ac susceptibility $\chi'(T)$ of $\text{Ce}_{0.9}\text{Er}_{0.1}\text{Al}_2$ for several frequencies.

Fig. 8. Plots of T_M vs $100/\ln(f_0/f)$ for the $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ series of $x = 0.08, 0.1$, and 0.2 . The solid line is the least-squares fit using Eq. (3). f_0 was fixed at 10^{13} Hz. The obtained parameters are summarized in Table I.

Fig. 9. Schematic phase diagram of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$ systems.

tion of the Coqblin-Schrieffer model of Kondo impurities.^{23,24)} This theory predicts a single-impurity Kondo temperature T_K of approximately $3T_M$ for Ce compounds. However, in the case of CeAl_2 ($x = 0$), T_M can be influenced not only by the Kondo effect but also by antiferromagnetic interactions, since T_K is close to $T_N = 3.8$ K.⁸⁾ T_M decreases as Er concentration increases from $x = 0$. Since the coefficient dT_M/dx approaches dT_N/dx , which is estimated from the sharp peak of the temperature dependence of the specific heat. This means that the x dependence on T_M corresponds to that on T_N , at least up to x of approximately 0.08.

The $T_M - x$ curve shows a minimum at around $x = 0.2$. As discussed before, a spin-glass-like-behavior appears between $x = 0.1$ and 0.4 , where the values of E_a are one order higher than that of T_0 . It seems that T_M corresponds to the freezing temperature from the paramagnetic state to the spin-glass state. For $x > 0.6$, on the other hand, the spin-glass-like behavior disappears when E_a is smaller than T_0 . In this region, T_M increases with increasing x and is close to the Curie temperature T_C obtained from the results of the specific heat measurement. This result indicates that the ferromagnetic state is stable and that T_M corresponds to T_C for $x > 0.6$.

4. Summary

In this work, we performed specific heat, dc magnetization, and ac susceptibility measurements on single crystals of $\text{Ce}_{1-x}\text{Er}_x\text{Al}_2$. The magnetic behavior upon changing from an anti-ferromagnetic (CeAl_2) ground state to a ferromagnetic (ErAl_2) ground state was determined. The magnetic ordering temperature changed continuously as a function of Er concentration x . Spin glass like behavior was observed at around $x = 0.1 - 0.4$.

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