Resistance and Thermal Expansion Anomaly near the Martensitic Transformation in GdCu under Pressure

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Abstract

The thermal expansion and electrical resistivity of GdCu have been measured under pressure. A large anomaly in the temperature dependence of thermal expansion is observed at 239 K (= M_s) at ambient pressure. M_s is found to decrease rapidly by applying pressure at a rate of $dMs/dP \sim$ -160 K/GPa.

Key words: martensitic transition; GdCu; pressure

1. Introduction

In the equiatomic RM compounds (R= rare earth, M= non magnetic transition metal), the changes in the electron density could be responsible for the existence of structual transformations. For example, GdCu shows a structual phase transformation from the cubic CsCl-type structure to a FeB-type one at low temperature[1,2]. This transition has been identified as a diffusionless martensitic transformation being characterised by a large thermal hyteresis at the pronounced volume and electrical resistivity anomalies. In the present work, we report the temperature dependence of thermal expansion of GdCu under pressure in detail and discuss the pressure-temperature phase diagram of the phase transition.

2. Experimental

Several pieces of pollycristalline GdCu have been prepared by melting constituent elements in an arc furnace. The thermal expansion was measured by using strain gage method, in which Mo piece in purity 5N was used as a standard material. Hydrostatic pressure up to 1.2 GPa was generated by using piston-cylinder device and 1:1 mixture of Fruorinert FC 70 and FC 77 as a pressure transmitting medium. The details of high pressure apparatus has been reported previously[3,4].

3. Results and Discussions

Fig. 1 shows the temperature dependence of the linear thermal expansion ($\Delta L/L$) under pressure. Here the sample was cooled from room temperature to 77 K and then heated to room temperature. On cooling, there is a discontinuous change in $\Delta L/L(T)$ at M_s= 239 K at ambient pressure. By further cooling below M_s, this transformation is found to occur gradually in wide temperature range, where the linear thermal expansion coefficient becomes negative, and stops near 110 K (=M_f). It is suggested that the structural transformation is accompanied by considerable structual disorder in the lattice through the buildup of internal pressure, which comes from CsCl→ FeB transformation. Similar to the anomalies in $\Delta L/L(T)$, M_s and M_f are also observed in the temperature dependent

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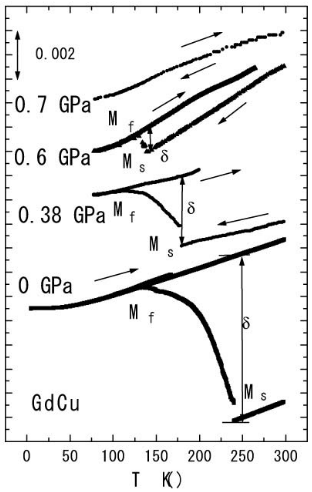

Fig. 1. The temperature dependence of the linear thermal expansion ($\Delta L/L$) of GdCu under pressure.

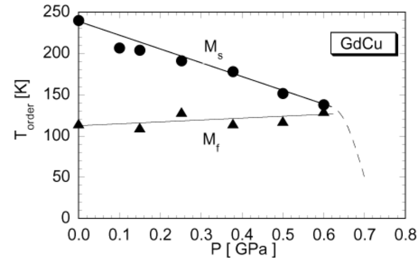

Fig. 2. The pressure dependence of M_s and M_f

electrical resistivity[5].

The length change of $\delta = 7 \times 10^{-3}$ is observed upon the transformation at M_s. This anomaly is not observed in the heating process because of the large hysteresis. To recover the initial state (CsCl structure), it is necessary to anneal the sample above 600 K[1].

As pressure increases, δ decreases and disappears around 0.7 GPa, indicating that the phase transition disappears. Fig. 2 shows the pressure dependence of M_s and M_f. M_s rapidly decreases by applying pressure at a rate of $dM_s/dP \sim$ -160 K/GPa, while M_f increases slightly at a rate of $dM_f/dP \sim$ 20 K/GPa. Above 0.7 GPa, where M_s seems to be coincident with M_f, M_s is not observed in the temperature region above 77 K.

In the case of $GdCu_{1-x}Ga_x$, M_s of $x=$ 0 (=250 K) decreases with an addition of Ga and disappears near $x=$ 0.03[1]. It is attributed to the valency difference between Cu and Ga, i.e., Ga provides two additional conduction electrons and a strong influence is expected on the band structure. However, the volume of the unit cell increases as increasing Ga concentration in $GdCu_{1-x}Ga_x$, which is in conflict with the case where GdCu is pressurised, i.e., the unit cell volume of GdCu is decreased.

Finally, we discuss briefly the thermodynamic property on the slope of the phase boundary at M_s. By using Clausius-Clapeyron relation,

$$\partial M_s/\partial P = \Delta S/\Delta V$$

the discontinuity of the entropy ΔS is estimated to be 3.4 J/mol K, where ΔV is obtained from the the discontinuity of the thermal expansion, $\delta = 7 \times 10^{-3}$ at ambient pressure, lattice constant a_0= 3.502 Åof CsCl structure at ambient pressure, and Avogadro number $N_0 = 6.02 \times 10^{23}$.

$$\Delta V = ((1-\delta)^3 - 1)a_0^3 N_0 (< 0)$$

The latent heat is estimated to be $\Delta Q = M_s \Delta S$= 810 J/mol, which is comparable to $\Delta Q \sim$ 1800 J/mol by the result of the scanning calorimeter[1].

4. Summary

From the measurements of the thermal expansion of GdCu at high pressure, the pressure-temperature phase diagram for the martensitic phase transformation was obtained. The result is compared with that of $GdCu_{1-x}Ga_x$.

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