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TRANSPORT AND CRYSTAL PROPERTIES OF $\alpha$- AND $\beta$- Ce$_3$Al

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Abstract. - The electrical resistivity, the thermopower, the X-ray powder diffraction and the magnetic susceptibility of $\alpha$- and $\beta$- Ce$_3$Al were studied. $\alpha$- Ce$_3$Al was found to undergo a crystal phase transition of a first order, at about 100 K, keeping the same crystal structure but with a change of its unit cell volume of an amount of 1.8 %.

Among intermetallic compounds of Ce and Al, CeAl$_2$ and CeAl$_3$ have been most extensively studied in relation to their magnetic properties, especially the Kondo and the heavy Fermion properties [1]. However, only a few works were reported, very curiously, on the magnetic properties of CeAl and Ce$_3$Al [2-4]. The last compound was reported to have two phases, a hexagonal $\alpha$ - Ce$_3$Al phase with a Ni$_3$Sn type crystal and a cubic $\beta$ - Ce$_3$Al phase with a Cu$_3$Au type crystal [5].

On $\alpha$ - Ce$_3$Al, an antiferromagnetic order with $T_N$ equal to 2.6 K and a phase transition of an unknown origin at about 100 K were reported from the measurements of magnetic susceptibility, $\chi$, and electric susceptibility, $\rho$ [3]. The pressure dependence of $\rho$ was reported in [4]. We report, in this study, the thermoelectric power, $S$, and the X-ray powder diffraction in addition to $\chi$ and $\rho$.

We prepared $\alpha$ - Ce$_3$Al by a rapid cooling from the melt, while $\beta$ - Ce$_3$Al by annealing the former at 600 C for three days respectively. However, the description of the two phases in the reference [5] is the other way around and puzzling. Most of the $\alpha$ - Ce$_3$Al samples were annealed subsequently for homogenization at 200 C for eight days. These samples were found by X-ray powder diffraction to be a single phase and to have the reported crystal structures. Powder sample was obtained by filing the ingots and repeatedly annealed to relieve the strain at 150 C for four days.

In figure 1, $\rho$ of a sample of $\alpha$ - Ce$_3$Al with the heat treatment is plotted as the function of temperature, $T$. Although the structures of $\rho$ appearing on this curve are similar to those of the samples without the heat treatment, the former is much more prominent than the latter.

Low temperature part of $\rho$ shown in an inset of figure 1 are characterized by a minimum at 25 K, a Kondo type increase for decrease of $T$ below 25 K and a sharp drop due to an antiferromagnetic order at $T_N$ found to be equal to 2.8 K. Besides, at which we about 100 K, a sudden change of $\rho$ appears. These behaviours of $\rho$ were the same to those reported [3]. In addition, we found this change of $\rho$ at about 100 K to show a temperature hysteresis of about 10 K as seen in figure 1.

Fig. 1. - The electric resistivity, $\rho$, of $\alpha$- and $\beta$ - Ce$_3$Al plotted as the function of temperature, $T$. The insets shows $\rho$ of $\alpha$ - Ce$_3$Al in the low $T$ region.

From this fact, the transition is understood to be of the first kind.

In figure 2, $S$ of $\alpha$ - Ce$_3$Al is plotted as the function of $T$. Low temperature part of $S$ shown in an inset of figure 2 manifested a Kondo type increase for decrease of $T$ below 20 K and a sudden drop at $T_N$. A sudden change of $S$ is seen to appear again at about 100 K, which showed also a temperature hysteresis. Thus, the behaviours of $\rho$ and $S$ are in a very good correspondence.

Fig. 2. - The thermoelectric power, $S$, of $\alpha$- and $\beta$ - Ce$_3$Al plotted as the function of temperature, $T$. The insets show $S$ the both samples in the low $T$ region.
In figure 3, the relative expansion of lattice constants, $\Delta a/a$ and $\Delta c/c$, determined by the X-ray powder diffraction is plotted as the function of $T$. The hexagonal crystal structure was found to be identical above and below the transition. However, lattice constants were found to jump when the temperature of sample goes across the transition, the values of the jumps of $\Delta a/a$ and $\Delta c/c$ being 0.4 % and 1.0 %, respectively. Thus, the volume of crystal above the transition is larger than that below by an amount of 1.8 %.

![Graph of fig.3](image3)

Fig. 3. – The relative expansion of lattice constants, $\Delta a/a$ and $\Delta c/c$ for $\alpha$–Ce$_3$Al plotted as the function of temperature, $T$.

However, the transition does not proceed all at once. Instead, there was a temperature range of the coexistence of the two phases of about 40 degrees, where double peaks of X-ray diffraction appeared with a gradual interchange of their intensities.

In the phase transition of the first order, an energy barrier between the two phases above the transition and below is understood to cause the temperature hysteresis. A distribution of this energy barrier may be anticipated especially in a powder sample due to the surface effect, and it may give rise the temperature range of coexistence of the two phases.

The magnetic susceptibility, $\chi$, and its inverse, $1/\chi$, are shown in figure 4. The magnetic moment of the sample above the transition and below is very near to the value of Ce (4f). In addition, we remind of a large volume change of an amount of 1.8 %. Hence, neither the valence change nor the quadrupolar ordering of Ce (4f) electrons seems to be the origin of the transition at 100 K. A deviation from the Curie-Weiss law reported in [3] around the transition at 100 K is weakly seen also in figure 4.

![Graph of fig.4](image4)

Fig. 4. – The magnetic susceptibility, $\chi$, and its inverse, $1/\chi$, of $\alpha$– and $\beta$–Ce$_3$Al is plotted as the function of temperature, $T$. The inset shows $\chi$ of $\alpha$–Ce$_3$Al for the low $T$ region.

As for the other compound, $\beta$–Ce$_3$Al, its $\rho$, $S$ and $\chi$ are also shown in figures 1, 2 and 4 respectively. $\rho$ decreases monotonically with the decrease of $T$. It shows a sudden decrease at 9 K, and $S$ in figure 2 also shows a bent at the same temperature, which is probably an antiferromagnetic order, $T_N$, being equal to 9 K. The value of $S$ above 100 K is seen to be small and constant, similarly to $S$ of Ce compounds having well localised (4f) electron, CeAl$_2$ for example [6]. Magnetic moment from $\chi$ shown in figure 4 is near to that of Ce (4f). Hence, $\beta$–Ce$_3$Al is concluded to be one of Ce compounds with a (4f) electron having a well localized state. The thermal expansion, $\Delta a/a$, although not shown, was found to be normal.