Transport and magnetic properties of CeCo$_x$Ni$_{1-x}$Ge$_2$ system

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Abstract

Electrical resistivity and the magnetic susceptibility have been measured for the series of intermetallic compounds CeCo$_x$Ni$_{1-x}$Ge$_2$ ($x=0.1, 0.2, 0.5, 0.7, 0.8, 0.9$ and $1$). CeNiGe$_2$ is an antiferromagnetic compound with $T_N=3.3$ K. CeCoGe$_2$ is an intermediate-valence compound with cerium valence varying from 3.2 at room temperature to 3.8 at 1.8 K. The valence fluctuations are quenched at $x=0.5$, while the heavy Fermion behavior appears in compositions with $x<0.5$.

**Key words:** CeCo$_x$Ni$_{1-x}$Ge$_2$ ; Kondo effect ; Valence fluctuation

The cerium based intermetallic ternary compounds remain a subject of considerable interest because of their unusual ground state properties such as heavy Fermion, valence fluctuation and non-Fermi liquid behaviors. The low temperature behavior of these compounds can be fairly well accounted for by the competition between the RKKY-type interaction among 4$f$ local moments and the Kondo-type interaction between local moments and conduction electrons. The ground state properties are normally dominated by the relative magnitude of either the RKKY or Kondo energy.

The aim of this paper is to report the results of an investigation on the effective suppression of the long-range magnetic order by the Kondo effect and the concomitant development of the intermediate valence state by a progressive substitution of Co for Ni in CeCo$_x$Ni$_{1-x}$Ge$_2$. It is expected that the Co substitution generates the normal chemical pressure, enhances the density of states (DOS) at the Fermi level and thus continuously evolve from the antiferromagnetic state of the Kondo-lattice compound CeNiGe$_2$ ($T_N=3.3$ K) [1] into the valence fluctuating state.

All the samples investigated in this present work have been prepared by direct melting of the high purity elements in a tetra-arc furnace under a purified argon atmosphere followed by an annealing treatment at 950 °C for five weeks.

The powder X-ray diffraction studies show that the samples with $x>0.5$ have 2% to 3% other phases. The
diffraction patterns show that all samples have the orthorhombic CeNiSi$_2$-type structure with the group space Cmcm. As shown in Fig. 1, the change of unit-cell volume by Co substitution is very small as about 0.8%.

As shown in Fig. 2, the $\chi$ vs. $T$ curve for $x>0.7$ composition shows a broad peak around 50 K such as that of CeSn$_3$ which is a typical valence fluctuation material [2]. Below 20 K the $\chi$ increases with decreasing temperature, which may be attributed to a small amount of paramagnetic impurities not detected by the X-ray diffraction. A simple model, which describes the features for a fluctuating valence in a cerium material, was developed by Sales and Wohlleben [3].

The measured $\chi$ vs. $T$ behaviour obeys the model in the temperature regions from 50 K to 300 K. As a result, the cerium valence effectively increases in magnitude from mostly +3 at room temperature (a mean value of +3.2) to mostly +4 at 50 K (a mean value of +3.65) for $x=1$. On the other hand, the $\chi$ vs. $T$ curves for $x \leq 0.5$ compositions indicate that the Curie-Weiss law is followed between 150 K and 300 K and values of $\mu_{eff}$ obtained by least squares fitting vary in the range of 2.4$\mu_B$ to 2.7$\mu_B$ across the series compared to 2.54$\mu_B$ expected for Ce$^{3+}$. The antiferromagnetic ordering observed in CeNiGe$_2$ is depressed below 2 K by the Co substitution. This seems to be due to the enhanced Kondo effect, arising from the enhanced DOS at Fermi level, since the lattice parameters have hardly changed with Co substitution.

Fig. 3, shows the temperature dependence of the resistivity ($\rho$) of CeCo$_{x}$Ni$_{1-x}$Ge$_2$. The magnetic contribution to $\rho$ in $x \leq 0.2$ composition, which is not plotted in Fig. 3, exhibits the -ln$T$ behavior in two temperature regions. This is due to the Kondo scattering considering the CEF effect. The temperature of $\rho_{min}$ in lower temperature region gradually increases with $x$.

This indicates that the Kondo temperature ($T_K$) is enhanced with increasing $x$. On the other hand, in $x \leq 0.5$ composition $\rho$ increases as -ln$T$ with decreasing temperature and then decreases as $T^2$ in low temperature region. The temperature of the crossover between the two shifts to high temperature side with increasing $x$. These behaviors occur in the intermediate valence materials with $T_K$>CEF [4] and consistent with behavior of the magnetic susceptibility mentioned above.

The overall behavior of the present system is qualitatively understood as a competition between the Kondo effect and the RKKY interaction within the framework of Doniach’s phase diagram [5]; the DOS at Fermi level is strongly enhanced by the Co substitution for Ni. Consequently the antiferromagnetism in CeNiGe$_2$ is depressed by the substitution of Co and is changed into an intermediate valence state beyond $x=0.5$. The present system, hence, provides us with a convenient model case to further investigate how a Kondo-lattice compound evolves from a Kondo regime into a valence-fluctuation regime.

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References


