Transport and magnetic properties of $CeNiGe_{2-x}Si_x$ single crystals

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Abstract

The electrical resistivity and the magnetic susceptibility have been measured for the series of intermetallic compounds $CeNiGe_{2-x}Si_x$. $CeNiGe_2$ is an antiferromagnetic compound with T_N =3.3 K. $CeNiSi_2$ is an intermediate-valence compound with cerium valence varying from 3.4 at room temperature to 3.75 at 1.8 K. With increasing x, the coupling constant J is enhanced and thus the Kondo temperature T_K increases.

Key words: $CeNiGe_{2-x}Si_x$; Kondo effect ; Valence fluctuation

Cerium based ternary intermetallic compounds of the type $CeTX_2$ (where T=transition metals and X=semimetallic elements) remain a subject of considerable interest because of their unusual ground state properties observed in many compounds of their family. For instance, heavy-Fermion behavior with γ=1.7 J/K^2·mol in $CePtSi_2$ [1], antiferromagnetic Kondo system with T_N=3.9 K in $CeNiGe_2$ [2] and non-magnetic intermediated valence material with a Kondo temperature $T_K\approx$500 K in $CeNiSi_2$ [2]. It is well known that their various ground states are governed by the competition between the RKKY and Kondo interactions. In this paper, we report the influence on the electrical resistivity and magnetic susceptibility as germanium is substituted by silicon in $CeNiGe_{2-x}Si_x$. The substitution is expected to change the coupling constant J between the $4f$ magnetic moments and the conduction electrons.

The single crystals of $CeNiGe_{2-x}Si_x$ have been grown by the Czochralski pulling method using a tetra-arc furnace in high purity (6N) argon atmosphere from the constituent elements (Ce 3N, Ni 4N, Ge 5N, and Si 6N). X-ray diffraction confirmed the single-phase nature of all the investigated samples and the crystal structure was to be the orthorhombic $CeNiSi_2$ structure type with the space group $Cmcm$. The lattice parameters decreased almost linearly with increasing x [3].

Fig. 1 shows the temperature dependence of inverse magnetic susceptibility ($1/\chi$) for H⊥b-axis. The peaks due to the long-ranged antiferromagnetic ordering are observed at T_N=3.3 K and 2.2 K in compositions with x=0 and x=0.2, respectively, which is not plotted in Fig. 1. Any anomaly due to the magnetic ordering, however, is not found down to 2 K in compositions with $x\geq$0.4. The $1/\chi$ curves of compositions with $x\leq$1.2 indicate that the Curie-Weiss law is followed between 150 K and 300 K and μ_{eff} values obtained by the least-squares fitting are in the range 2.4-2.7μ_B across the series compared to 2.54μ_B expected for Ce^{3+}. θ_P is also evaluated to be very large negatively and increases with x as shown in Fig. 3. Since this stems from the Kondo effect, T_K is thought to increase with x. The anomaly by the CEF effect is observed in low temperature regions.

A broad minimum around 100 K in $1/\chi$ curve of compositions with $x\geq$ 1.8 is a characteristic of valence fluctuating systems, as reported earlier [4]. The simple model, which describes the features for a fluctuating valence in a cerium material, was developed by Sales and Wohlleben [5]. The measured χ well obeys the model in the temperature regions from 50K to 300

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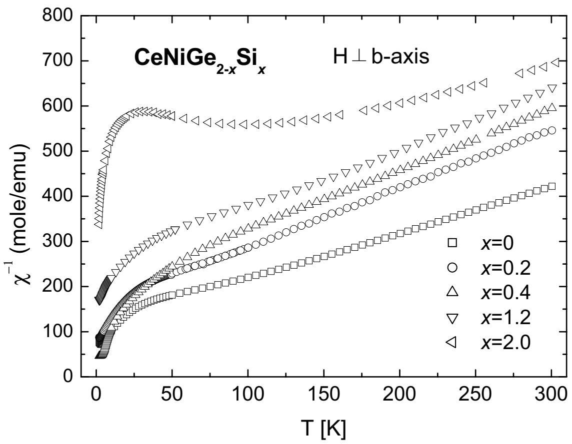

Fig. 1. Temperature dependence of inverse magnetic susceptibility ($1/\chi$) for CeNiGe$_{2-x}$Si$_x$.

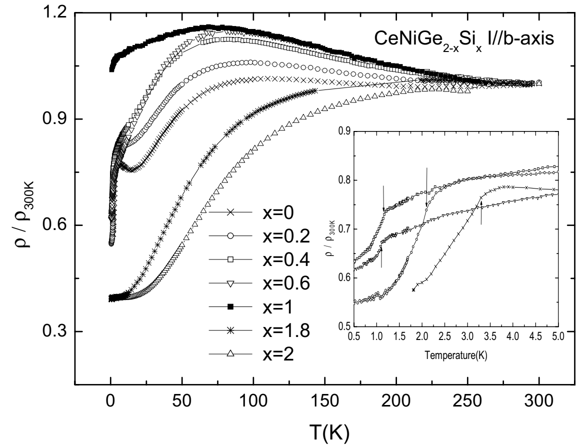

Fig. 2. Temperature dependence of electrical resistivity (ρ) of CeNiGe$_{2-x}$Si$_x$. The arrows in the inset indicate the point of inflection.

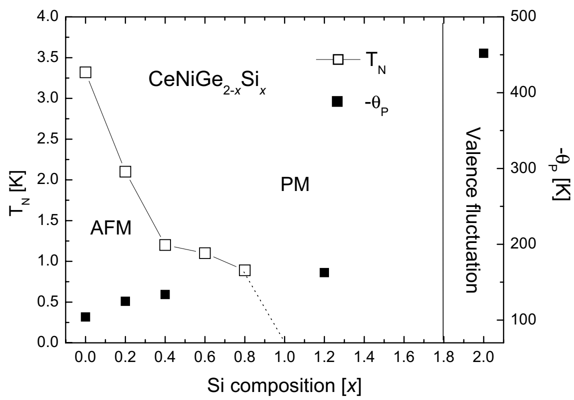

Fig. 3. Phase diagram and θ_P plot of CeNiGe$_{2-x}$Si$_x$.

K. As a result, the cerium valence effectively increases in magnitude from mostly 3 at room temperature (a mean value of +3.4) to mostly 4 at 1.8 K (a mean value of +3.75).

Fig. 2 shows the temperature dependence of the electrical resisitivity (ρ) for I$\|$ b-axis. The magnetic part of ρ in x=0 composition, which is not plotted in Fig. 2, exhibits the -ln$T$ behaviors in two temperature regions. The minimum of ρ around 20 K gradually disappears with increasing x, and the -ln$T$ behavior only appears in high temperature regions in x=1 composition. These behaviors indicate that T_K is enhanced by the Si substitution, according to the enhancement of the coupling constant J between the $4f$ magnetic moments and the conduction electrons which can be attributed to the lattice contraction, and is nearly equal to the magnitude of CEF in x=1 composition. The ρ for composition with $x \leq 1.8$ shows the behavior of valence fluctuation similar to $CeSn_3$[4]. As shown in the inset of Fig. 2, a point of inflection in $CeNiGe_2$ is observed at about 3.3 K. This is due to the antiferromagnetic ordering, since the ordering is observed at the same temperature in the magnetic susceptibility curve. The decrease in ρ occurs from a bit higher temperature than T_N. This seems to arise from the formation of coherent Kondo ground state. As shown in Fig. 3, the T_N shifts to low temperature region with increasing x but is not observed down to 0.5 K in compositions with $x \geq 1$. The quantum critical point is expected to be found near x =1 composition.

In conclusion, 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 [6]; the coupling constant J is strongly enhanced by the Si substitution for Ge. Consequently the antiferromagnetism in $CeNiGe_2$ is depressed by the progressive substitution of Si and is changed into an intermediate valence state beyond x=1.8.

This work was supported by Center for Strongly Correlated Materials Research (**CSCMR**) at Seoul National University and by the Korea Research Foundation Grant (KRF-2001-DPO139).

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