Non-Fermi-liquid behavior in $CeNiGe_{2-x}Si_x$ single crystals

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Abstract

The specific heat in low temperature regions was measured for several pseudoternary compounds of the $CeNiGe_{2-x}Si_x$ system. Substitution of silicon for germanium enhances the coupling constant J and the Kondo interaction. The antiferromagnetism in $CeNiGe_2$ is ultimately suppressed around x=1 by the enhanced Kondo effect. A non-Fermi liquid behavior has been observed in x=1 composition.

Key words: $CeNiGe_{2-x}Si_x$; specific heat ; Kondo effect ; non-Fermi liquid

The competition between the RKKY interaction and the Kondo interaction in Ce-based compounds was successfully described by Doniach[1], taking into account that both depend on the same coupling constant J between the $4f$-local moments and the conduction electrons. The former tends to drive a system into a magnetically ordered state, while the latter tends to drive it into a nonmagnetic state. A compound having a proper magnitude of J shows non-Fermi liquid (NFL) such as $C/T \propto \ln T$, $\rho(T) \propto T$, and $\chi(T) \propto (1\text{-}T^{1/2})$ through a quantum critical point (QCP) [2,3].

The aim of this paper is to report the result of an investigation on the effective suppression of the long-range magnetic order by the increase of the Kondo interaction by a progressive substitution of Si for Ge in $CeNiGe_{2-x}Si_x$.

It is expected that Si-substitution generates the normal chemical pressure and enhances the coupling constant J. For $CeNiGe_2$ Pecharsky *et al*. reported that an antiferromagneticall ordered state below T_N=3.9 K coexists with a heavy-Fermion state (γ=98 mJ/K^2·mol) and the anomaly below T_N due to the modulation of the antiferromagnetism appears at 3.2 K in magnetic susceptibility and specific heat [4]. On the other hand, $CeNiSi_2$ is an intermediated valence material with T_K ≈ 500 K [4].

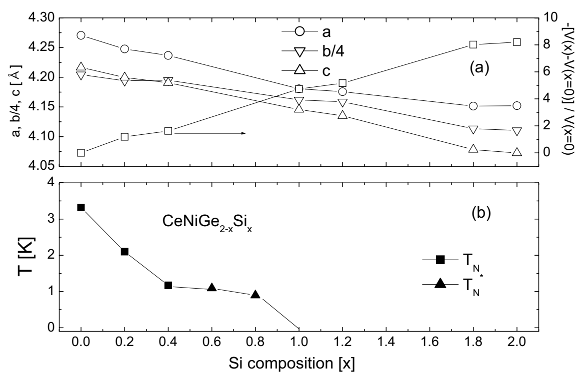

Fig. 1. Lattice parameters a, b, c, volume change of unit cell -[V(x) - V(x=0)]/V(x=0) (a) and Néel temperature(T_N) or hump temperature($T^*{}_N$) (b) as a function of Si concentration x in $CeNiGe_{2-x}Si_x$.

The single crystals of $CeNiGe_{2-x}Si_x$ have been prepared by the Czochralski pulling method using a tetra-arc furnace in an argon atmosphere. The specific heat was measured by a quasi-adiabatic heat-pulse method using a ^{3}He refrigerator from 0.5 K to 30 K.

The samples were examined by X-ray powder diffrac-

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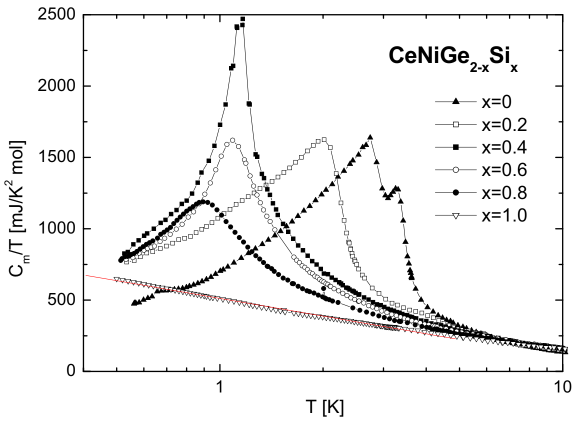

Fig. 2. C_m/T vs. log$T$ curves for $CeNiGe_{2-x}Si_x$.

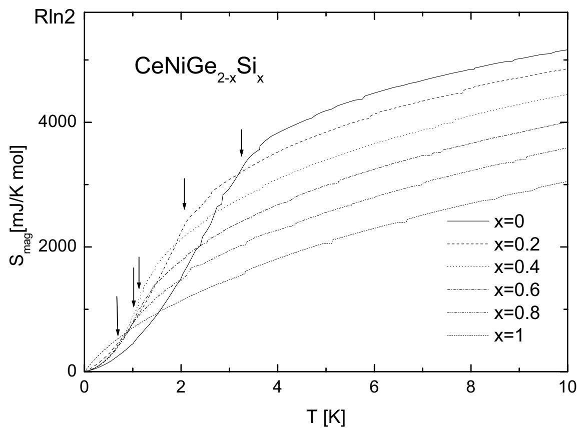

Fig. 3. Temperature dependence of the magnetic entropy (S_{mag}) in $CeNiGe_{2-x}Si_x$. The arrows indicate the T_N and $T^*{}_N$.

tion and metallography. X-ray diffraction confirmed the single-phase nature of all the investigated samples and the crystal structure was confirmed to be the orthorhombic $CeNiSi_2$ structure type with the space group *Cmcm*. The composition dependence of the lattive parameters a, b, c and unit-cell volume V is plotted in Fig. 1(a). The parameters decrease almost linearly with increasing x as expected.

Fig. 2 shows the magnetic contribution to the specific heat divided by temperature, C_m/T. The two peaks due to magnetic transitions in x=0 composition are observed at T_N=3.3 and 2.7 K as reported in Ref.4. The former is due to the antiferromagnetic ordering, since the peak in the magnetic susceptibility is observed at 3.3 K. The latter seems to arise from the modulation of the antiferromagnetism as mentioned above and disappears in compositions with $x>0.2$. T_N shifts to lower temperature with increasing x in compositions with $x\leq 0.4$. This decrease might come from the enhanced Kondo effect according to the coupling constant J increasing from the shrink of unit-cell volume. In compositions with x=0.6 and x=0.8, however, the hump observed below 1 K is different from the peak arising due to the long-range antiferromagnetic ordering in compositions with $x\leq 0.4$, since the hump is small and is not very sharp. As shown in Fig. 3, the magnetic entropy of compositions with x=0 and x=0.2 at T_N is ∼50 % of Rln2 expected for complete removal of the twofold degeneracy of a CEF ground doublet. The reduced entropy value might be due to the substantial Kondo-derived reduction of the Ce moments. On the other hand, the entropy in compositions with x=0.6 and x=0.8 is as small as ∼15% of Rln2. This value suggests that the hump is not due to the long-range magnetic ordering but short-range magnetic one. The short-range order seems to stem from the site disorder generated by the Si-substitution, which may have a significant influence on the low-temperature behavior through a modification of local Kondo interaction. It is reported that such behaviors occur in $CeCo(Ge_{1-x}Si_x)_3$ [5].

The composition dependence of T_N and temperature of the hump are plotted in Fig. 1(b). On the other hand, $CeNiGe_{2-x}Si_x$ does not possess any peak due to the magnetic transition down to 0.5 K but exhibits the -ln$T$ dependence of C_m/T in a wider temperature range from 0.5 K to 5 K, which is said be one of the characteristic features of non-Fermi liquid behaviors. The entropy of x=1 approaches Rln2 at 31 K. This is in contrast with the result of the quadrupolar Kondo model including a purported missing entropy of 0.5Rln2 at low temperatures [6]. The non-Fermi liquid behavior in this system may be thought to arise from the depression of the short-range magnetic order mentioned above.

In conclusion, $CeNiGe_{2-x}Si_x$ is a heavy Fermion system with a competition between the RKKY interaction and the Kondo interaction and exhibits the non-Fermi liquid behavior that comes from the magnetic disorder.

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).

References

[1] S. Doniach, Physica B **91** (1977) 231.

[2] B. Andraka and G. R. Stewadt, Phys. Rev. B **47** (1993) 3208.

[3] M. C. Aronson *et al.*, Phys. Rev. Lett. **75** (1995) 725.

[4] V. K. Pecharsky *et al.*, Phys. Rev. B **43** (1991) 10906

[5] D. Eom *et al.*, J. Phys. Soc. Jpn **67** (1998) 2495

[6] C. L. Seaman, Phys. Rev. Lett. **67** (1991) 2882