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HEAVY FERMION EFFECTS IN $Ce(Cu_{1-x}Ni_x)_2Ge_2$

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We report results of the specific heat, dc susceptibility, thermal expansion, thermopower and resistivity for a number of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ alloys. Upon increasing x, a non-linear increase of the heavy-fermion band width T^* and a strongly non-monotonic Néel temperature $T_N(x)$ are found ($T_N = 4.1$ K for $x = 0$ and $T_N < 2$ K for $x \geqq 0.75$).

Ce-based intermetallics with $ThCr_2Si_2$-structure have helped substantially to explore the properties of Kondo-lattice or heavy fermion systems. $CeCu_2Ge_2$ with Kondo temperature (or heavy fermion bandwidth) $T^* = 4$ K shows antiferromagnetic order between ("Kondo-reduced") local 4f-moments below $T_N = 4.1$ K [1]. $CeCu_2Si_2$ ($T^* = 10$ K) exhibits heavy fermion superconductivity below $T_c = 0.7$ K [2]. $CeRu_2Si_2$ ($T^* = 25$ K) remains Pauli paramagnetic at very low temperatures [3]. As was found recently [4], $CeNi_2Ge_2$ ($T^* = 29$ K) is phenomenologically closely related to $CeRu_2Si_2$. In order to monitor the transition between a local-moment system showing magnetic order and a Pauli paramagnetic system we have initiated a study of the quasi-ternary system $Ce(Cu_{1-x}Ni_x)_2Ge_2$ $CeCu_2Ge_2$ has been the first antiferromagnet, for which a structure in the Sommerfeld coefficient $\gamma(T)$ of the electronic specific heat was discovered well below the Néel temperature $T_N = 4.1$ K [1]. Such $\gamma(T)$ maxima are known to exist for $CeAl_3$, and normal state $CeCu_2Si_2$ [5] as well as for $CeNi_2Ge_2$ [4] and have been ascribed to coherence among the heavy fermions at low temperatures. Since an undisturbed Ce sublattice is commonly considered necessary for this feature to occur, our investigations offered the possibility to follow the effect of disorder only in the Cu/Ni sublattice on the coherence of the heavy-fermion band, as indicated by the $\gamma(T)$ peak.

The samples were prepared in an argon-arc furnace by mixing together appropriate amounts of the highly pure elements and remelting them several times. Microprobe and X-ray powder diffractometry could not resolve any strange phases (no annealing necessary). While the lattice parameter a kept almost constant ($a = 4.17$ Å for $x = 0$ and $a = 4.15$ Å for $x = 1$), the c-parameter shows an almost linear decrease from $c = 10.21$ Å ($x = 0$) to $c = 9.85$ Å ($x = 1$). The corresponding volume compression ($\bar{V} = a^2c$) amounts to 4.4%.

Our results of the specific heat (figs. 1a and b), dc-susceptibility (fig. 2a), thermal expansion (fig. 2b), thermopower (fig. 3a) and resistivity (fig. 3b) of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ can be summarized as follows:

1. Long-range magnetic order occurs up to, at least, $x = 0.65$. The thermal expansion data for the 75 at% Ni alloy suggest an ordering temperature to exist even below the temperature limit (2 K) of our susceptometer for this sample, too.
2. Both dc susceptibility [6] and specific heat data indicate that the transition is of antiferromagnetic type, cf. the magnetic field dependence at the

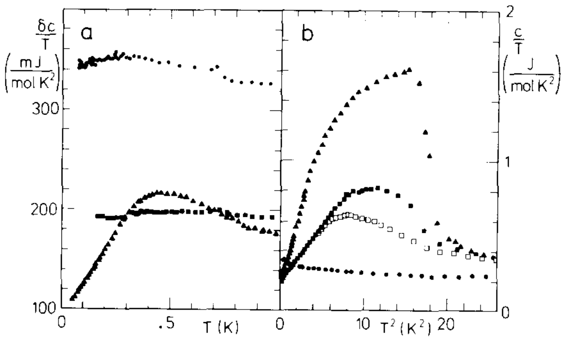

Fig. 1. Specific heat of $Ce(Cu_{1-x}Ni_x)_2Ge_2$. (a) $\delta C/T$ vs. T for $x = 0$(▲), 0.5(■), 1(●); $\delta C = C - C_{nuclear} - C_{magnon}$, cf. [1]; (b) C/T vs. T^2 for $x = 0$ at $B = 0$(▲); $x = 0.5$ at $B = 0$(■), 8 T(□) and $x = 1$ at $B = 0$(●). The T^3 dependence of $C(T)$ data points to antiferromagnetic magnons.

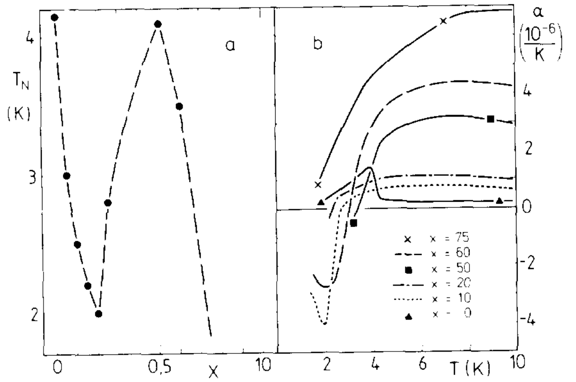

Fig. 2. (a) Néel temperature as derived from the peak in $\chi_{DC}(T)$ as a function of x for Ce $(Cu_{1-x}Ni_x)_2Ge_2$; (b) coefficient of thermal expansion $\alpha(T)$, for several $Ce(Cu_{1-x}Ni_x)_2Ge_2$ alloys.

transition temperature and the T^3 magnon dispersion relation of the specific heat for $x = 0.5$ (fig. 1b).

3. The particular type of antiferromagnetic order that occurs in the $CeCu_2Ge_2$ compound is efficiently changed already upon low Ni substitution. This is not only inferred from a rapid depression of T_N, but especially from the sign change in the phase transition anomaly in the thermal-expansion coefficient. According to thermodynamics this implies that the pressure derivative of the Néel temperature (as $p \to 0$) changes from a positive value (for $x = 0$) to a negative one.

4. T_N shows a pronounced maximum as a function of Ni concentration at $x = 0.5$.

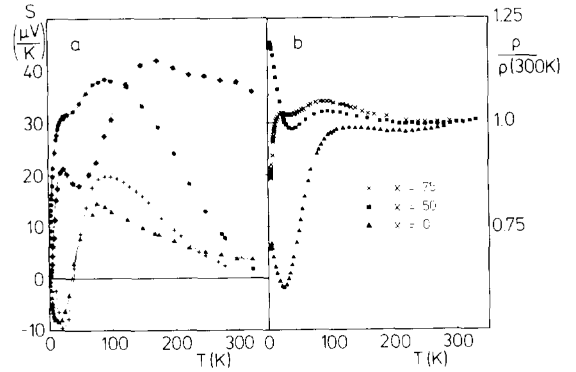

Fig. 3. (a) Thermopower as a function of temperature for $CeNi_2Ge_2$ (●) [4], $CeCu_2Ge_2$ (▲) [10], $CeRu_2Si_2$ (◆) and $CePd_2Si_2$ (+) [11] between $T = 2$ K and $T = 300$ K; (b) resistivity, $\rho(T)$, normalized to its room-temperature value for several $Ce(Cu_{1-x}Ni_x)_2Ge_2$ alloys.

5. From the position of a broad positive $\alpha(T)$ maximum, one concludes [7] that T^* does hardly change between $x = 0$ $x = 0.25$, but increases by almost a factor of three on going to $x = 0.75$. Not surprising, the magnetic entropy at $T = T_N$ decreases from $0.7R \ln 2$ [1] for $x = 0$ to $0.3R \ln 2$ for $x = 0.5$.

6. Though no disorder is introduced on the Ce-sublattice, typical "coherence properties" become weakened when the periodicity of the Cu/Ni sublattice is destroyed: The low-T resistivity peak is very pronounced (fig. 3b) and the $\gamma(T)$ peak almost suppressed (fig. 1a) for $x = 0.5$

7. While the thermopower of $CeCu_2Ge_2$ reproduce the typical behavior of most Ce-based Kondo-lattice systems, i.e., a positive CF-derived high-T peak and a negative giant peak near $T = T^*$, for $CeNi_2Ge_2$ also the low-T maximum has a positive sign which has been established before only for $CeCu_6$ and $CeRu_2Si_2$ [8].

To summarize, the transition from a local-moment antiferromagnetic state to a Pauli paramagnetic state has been monitored in the heavy-fermion system $Ce(Cu_{1-x}Ni_x)_2Ge_2$. Upon increasing Ni-concentration a non-linear increase of the single-ion Kondo temperature and a complex change in the magnetic characterisation including a non-monotonic concentration dependence of the Néel temperature are observed. We presume that these anomalies track the evolution of a multi-structured 3d-density of states at E_F. This makes photo-electron spectroscopy studies and band-structure calculations most desirable. The kind of local moment ordering established for $x < 0.75$ seems to be absent for the Ni-rich systems. $CeNi_2Ge_2$ shows similarities with both $CeRu_2Si_2$ (i.e. in the size of $\gamma(T)$ and in the temperature dependence of the thermopower $S(T)$) and $CeCu_2Si_2$ (i.e. in the pronounced $\gamma(T)$ structure near 0.3 K and in the T-dependence of the resistivity $\rho(T)$. Future investigations will have to show whether $CeNi_2Ge_2$ is indeed an enhanced Pauli paramagnet as currently assumed or belongs to the class of itinerant heavy-fermion antiferromagnets with extremely small ordered moments, e.g. UPt_3 [9].

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