MAGNETIC ORDER IN THE HEAVY FERMION SYSTEM $Ce(Cu_{1-x}Ni_x)_2Ge_2$

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The magnetic phase diagram of the heavy fermion (HF) systems $Ce(Cu_{1-x}Ni_x)_2Ge_2$ is discussed utilizing results of transport, thermodynamic and neutron-scattering measurements. While the Kondo temperature increases monotonically with x, a complex x-dependence is found for the Néel temperature, associated with a transition from local-moment to itinerant HF magnetism.

The tetragonal Ce-homologes of $ThCr_2Si_2$, while lacking direct 4f-wave function overlap, exhibit a distinct 4f-ligand hybridization. The latter, along with strong many-body renormalizations below the so called "lattice Kondo temperature" T^*, is effective in forming heavy-mass quasiparticles ("heavy fermions" (HF)). In the case of the compounds CeM_2X_2 (M : Cu, Ag, Au, Ru, Ni; X : Si, Ge), for large Ce-M distances r ($\geq$ 3.3Å), magnetic ordering between local moments occurs at T_m, whereas in the limit of small r ($\leq$ 3.2Å) a valence fluctuating state forms [1]. In the intermediate regime, the HF-compounds $CeNi_2Ge_2$, $CeRu_2Si_2$ and $CeCu_2Si_2$ are found, of which the latter is unique not only because it is a superconductor below 0.7K [2], but also it seems to exhibit an itinerant type of HF magnetism below approximately 0.8K (at zero magnetic field) and 7 Tesla (T→0) [3] .

In this communication, we report a brief summary of a systematic study on the quasibinary system $Ce(Cu_{1-x}Ni_x)_2Ge_2$, which reveals a transition from local-moment ordering to itinerant HF magnetism. These alloys are ideally suited for this purpose : The Kondo binding energy kT^* of $CeCu_2Ge_2$ ($T^*=8\pm2$K) is of the same order of magnitude as the magnetic interaction energy, kT_{RKKY} ($T_{RKKY}=7$K), at which short-range ordering sets in [4]. Long range magnetic order has been detected below $T_m = T_{N1} =$ 4.1K [4,5]. On the other hand, $CeNi_2Ge_2$ ($T^*=30$K) exhibits the signature of a Kondo lattice or HF compound with no long-range magnetic order [6]. For the samples prepared as described in [1], no parasitic phases have been detected by X-ray diffractometry or electron microprobe analysis.

Fig.1 shows the concentration dependence of the magnetic ordering temperature (in reduced units) for $Ce(Cu_{1-x}Ni_x)_2Ge_2$ as determined from thermal expansion, specific heat and magnetic susceptibility measurements [7,8]. Even small Ni concentrations cause a dramatic decrease of $T_{N1}(x)$, which extrapolates to zero near x=0.2. However, for larger x a second branch, $T_{N2}(x)$, develops with a maximum near x=0.5. This new magnetically ordered phase can be monitored up to approximately x=0.75. Bulk measurements show clear evidence for a second transition below T_{N1} and T_{N2}, respectively in the range $0.02 \leq x \leq 0.3$ (hatched region) as discussed in [8].

The magnetic structure and the size of the ordered moments have been determined by neutron powder-diffraction experiments for the compounds with concentrations x=0, 0.1, 0.28 and 0.5. Incommensurate modulated spin arrangements with magnetic Bragg reflections at $|Q| = |\tau_{hkl} \pm q_0|$, where (hkl) is a vector of the reciprocal chemical lattice and q_0 the propagation vector, have been found. They are described by the following values. For x=0: $q_0=(0.28, 0.28, 0.54)$ and the effective moment $\mu_S=0.74\ \mu_B$; for x=0.1 : $q_0=(0.28, 0.28, 0.41)$, $\mu_S=0.5\mu_B$; for x=0.28 : $q_0=(0.11, 0.11, 0.25)$ and finally for x=0.5 : $q_0=(0, 0, 0.14)$, $\mu_S=0.3\mu_B$. For this latter sample the magnetic Bragg reflections are much broader than the experimental resolution of the spectrometer which presumably reflects a disturbance of long-range magnetic order by single-site (Kondo) interactions.

To gain further insight into this competition of single-site and inter-site interactions, the magnetic relaxation rate via quasielastic neutron scattering has been investigated in the temperature range $1.5K \leq T \leq 200K$. The lattice Kondo temperature as a function of Ni concentration, as read off the residual quasielastic line widths at low temperatures ($T \geq T_N$), is also plotted in Fig.1 and compared to T^* as determined from thermal expansion and resistivity

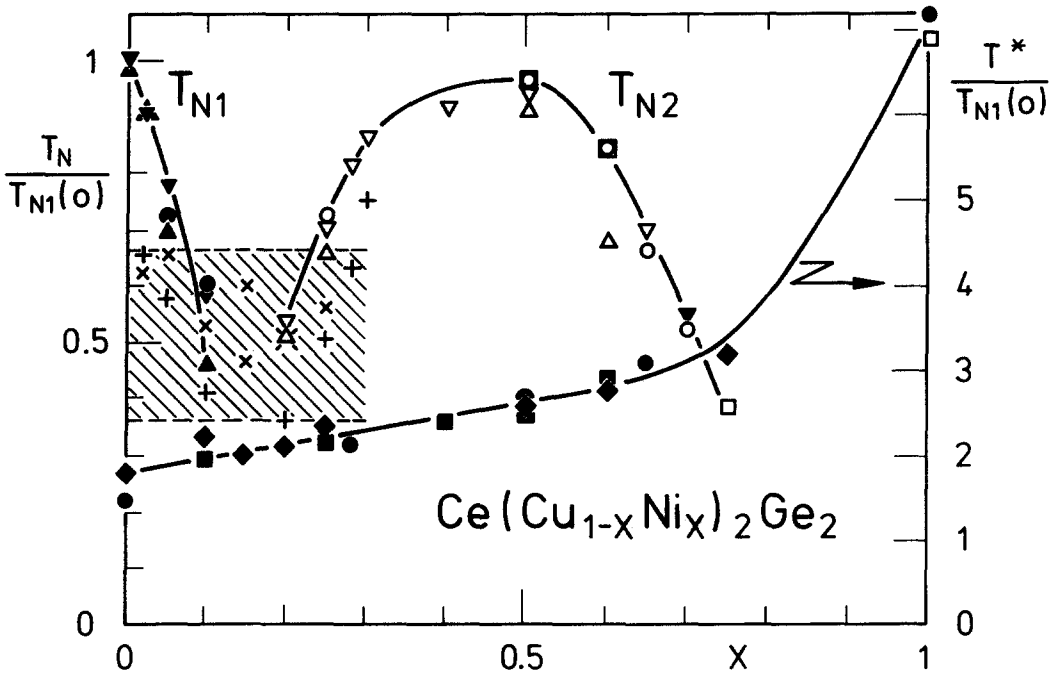

Fig.1. LEFT SCALE : x, T_N phase diagram for the quasi-binary compound $Ce(Cu_{1-x}Ni_x)_2Ge_2$ as determined from specific heat (▼, ▽, +), thermal expansion (▲, △, x) and dc susceptibility (●,O) measurements [9]. RIGHT SCALE : T^* as determined from the residual quasielastic neutron line width (●), thermal expansion (▪) and resistivity (◆) peaks. Positions of the latter are scaled by factors 1.5 and 1.9, respectively (T^* and T_N are normalized to T_{N1} for x=0).

maxima. With increasing x, T^* increases continuously and, for x=0.65, the Kondo binding energy is found to be larger than the energy of the inter-site interactions by almost a factor of four.

The most exiting result of the present study is the occurence of an extremely short propagation vector for the x=0.5 sample which characterizes a static spin wave extending over almost ten lattice constants. Such short ordering wave vectors have been predicted by Grewe and Welslau [9], if HF band magnetism develops in a Kondo lattice. Recent calculations of Welslau and Grewe within this model [10] revealed the development of T_N and T^* as a function of the local exchange coupling constant $g=N_F J$ (N_F : conduction band density of states at the Fermi level; J<0 : exchange integral) in the full range between $T_N \gg T^*$ and $T_N \ll T^*$. These calculations are corroborated by our results near x=0 where $T_N \approx T^*$ and for $0.5 < x \leq 0.75$ where $T_N \ll T^*$.

The intermediate composition range, in which

(i) two subsequent transitions are observed as a function of T for each sample ($0.02 \leq x \leq 0.3$) and

(ii) $T_{N2}(x)$ shows a monotonic increase ($0.2 \leq x \leq 0.5$), challenges further investigation.

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