Magnetic order of CeNi$_x$Ga$_{4-x}$

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Abstract. We investigate the ternary Ce-Ni-Ga system with samples prepared by the flux-growth method in Ga-flux. The series CeNi$_x$Ga$_{4-x}$ crystallizes in the tetragonal BaAl$_4$ structure. The Ga-rich compounds of this series have previously been reported to exhibit ferromagnetism. Our systematic study shows that with increasing Ni content the lattice parameters shrink. The magnetic transition temperature, as identified from the sharp maximum in the magnetic susceptibility, increases slightly with increasing Ni content. On the other hand the absolute value of the magnetization at the transition drops by around one order of magnitude. We did not find any difference between field-cooled and zero-field cooled magnetization measurements nor a hysteresis in the magnetization versus field curves, suggesting that our samples order antiferromagnetically.

1. Introduction

During the past decades research on the magnetism of intermetallic rare-earth compounds has led to a rich variety of novel phases and new physics. A topic of special interest is that of magnetic quantum phase transitions. In $f$-electron systems the competition between magnetically ordered and paramagnetic groundstates arises from the competition between RKKY interaction and Kondo effect. At low temperatures quantum fluctuations may yield exotic magnetic ground states with new properties. In the class of cerium-based systems exhibiting a quantum critical point (QCP), most systems are antiferromagnetic while ferromagnetic systems are quite rare. In our quest for compounds with a ferromagnetic QCP a suitable candidate seemed to be the system CeNi$_x$Ga$_{4-x}$, which was reported previously to be ferromagnetic [1–3].

2. Sample Preparation and Experimental Details

Due to the low melting point of Ga the Ce-Ni-Ga systems are ideal candidates for crystal growth by the flux-growth method in Ga flux. With this method, the ingots of the crystal-to-grow are molten together with a flux that acts as solvent, lowering the melting points of the ingots. If the solution then is cooled at appropriate rates, single crystals form, which can be separated from the flux by chemical or mechanical means. While the flux-growth method is relatively simple the product is less predictable than with other crystal-growth methods, since there is only little control over what is incorporated in the crystal and what is remaining in the flux. For example, an ingot containing Ce, Ni and Ga in the ratio 1 : 1 : 10 yields Ce$_2$NiGa$_{12}$.

The samples prepared for this investigation were grown from ingots with the ratios 1 : 1.5 : 5, 1 : 2 : 5 and 1 : 2.5 : 5. Appropriate amounts of the starting materials according to the concentrations mentioned above were put into alumina crucibles and sealed in evacuated quartz...
tubes. They were subsequently heated to 1100° C for 2 hours and then cooled to 300° C with a cooling rate of 10° C per hour. The Ga flux was removed by centrifugation. The crystal structure was determined by powder x-ray diffraction. The Ni:Ga ratio was determined by atomic absorption spectroscopy (AAS) with diluted nitric acid as solvent. The magnetic measurements were conducted in a commercial vibrating sample magnetometer (VSM) in the temperature range between 2 and 300 K in magnetic fields up to 3 T.

3. Sample Characterization
The x-ray diffraction measurements showed the samples crystallizing in the tetragonal BaAl\textsubscript{4} structure, which is a variant of the well-known ThCr\textsubscript{2}Si\textsubscript{2} structure. We found a small amount of a non-magnetic Ga\textsubscript{3}Ni\textsubscript{2} impurity phase in all samples. It was reported in literature that the series CeNi\textsubscript{x}Ga\textsubscript{4−x} forms in a very limited concentration range around 0.4 ≤ x ≤ 1.15 only [1; 4]. According to AAS measurements our CeNi\textsubscript{x}Ga\textsubscript{4−x} samples have Ni concentrations between 0.63 and 1.55. In this assignment possible deviations due to Ga-Ni impurity phases are neglected.

Figure 1. Unit-cell volume (top), lattice parameter along the long axis c (middle) and lattice parameter along the short axis a (bottom) for CeNi\textsubscript{x}Ga\textsubscript{4−x}.

The lattice parameters and unit-cell volumes determined from x-ray powder patterns are presented in fig. 1. Overall we observe a clear tendency of shrinking lattice parameters with increasing Ni concentration. There appears to be a maximum in c for Ni concentrations around x ≈ 0.8 as well as a minimum in a for Ni concentrations around x ≈ 1.4. These features were
4. Magnetic properties

The magnetic dc susceptibility $\chi$ vs. temperature in an external field of 0.1 T is shown in the left-hand panel of fig. 2. No difference between field-cooled (fc) and zero-field-cooled (zfc) measurements is observed. In the samples with Ni concentrations between $0.63 \leq x \leq 1.01$ a pronounced maximum indicating an antiferromagnetic phase transition is visible. The Néel temperature as shown in the inset of the left-hand panel of fig. 2, increases with increasing Ni content from $T_N \approx 3.6 \pm 0.1$ K for $x \approx 0.65$ to 4.7 K for $x \approx 1.0$. The scatter of $T_N$ for $x = 0.63$ and 0.66 might be due to uncertainties in the determination of the Ni concentration. At the same time the absolute value of the magnetization right at the transition drops by roughly one order of magnitude. The alloys with $x = 1.4$ and 1.55 do not show any indication of a phase transition in the temperature range investigated here, suggesting that they might order antiferromagnetically at lower temperatures. This assumption is corroborated by the downward curvature visible at below 20 K in $1/\chi$ as shown in the right-hand panel of fig. 2. The absolute value of $\chi$ at high temperatures (above 100 K) has no clear dependence on the Ni content. Such a scattering was also observed by Grin et al. [1] and Sampathkumaran et al. [2].

Figure 2. Left: Magnetic dc susceptibility $\chi$ vs temperature of CeNi$_x$Ga$_{4-x}$. The symbols present fc measurements, while the lines depict the zfc measurements. Inset: Néel temperatures $T_N$ vs. Ni concentration $x$ for the magnetically ordered compounds. Right: Inverse magnetic dc susceptibility $1/\chi$ vs temperature of CeNi$_x$Ga$_{4-x}$.

Inspection of $1/\chi$ over the entire temperature range $2 - 150$ K (fig. 2, right) shows that the effective paramagnetic moment is approximately $1.6 \mu_B$, which is about two thirds of the full moment of free Ce$^{3+}$. However, for the sample with $x = 0.63$ a clear upward curvature is observed, which might indicate a transition to a higher effective moment.

The magnetization as a function of the external magnetic field at $T = 2.5$ K is plotted in the left-hand panel of fig. 3. We could not find any signs of a hysteresis in the magnetization versus field curves, confirming our assignment of antiferromagnetism rather than ferrromagnetism in our samples. The magnetization of the sample with $x = 0.63$ shows two distinct kinks at 0.5 T and 1 T. The samples with $x = 0.66$ and 1.01 show distinct features at approximately $\pm 1$ T. These anomalies are indicative of metamagnetic transitions, similar as observed in the series RNi$_2$Ge$_2$ [5]. In the samples with $x = 1.4$ and 1.55 such features are absent, as illustrated in the right-hand panel of fig. 3. This is of course expected because these latter samples are in the
paramagnetic state.

5. Conclusion
Our CeNi$_x$Ga$_{4-x}$ samples exhibit properties clearly different from previously reported results [1–4; 6], which might be attributed to the different methods of sample preparation, since it was already noted before that the series CeNi$_x$Ga$_{4-x}$ is very sensitive to preparative conditions [7]. While in previous studies a Curie-Weiss behavior with the full effective paramagnetic moment of Ce and indications of ferromagnetism was found, our samples show a reduced effective magnetic moment at high temperatures followed by antiferromagnetism towards low temperatures. In agreement with previous studies increasing the Ni content and thus decreasing the lattice parameters suppresses the magnetic order. This is analogous to CeCu$_{6-x}$Au$_x$, which orders antiferromagnetically for sufficiently large Au concentrations, but goes into the non-magnetic heavy-fermion state of CeCu$_6$, when the Au content is reduced to $x < 0.1$ (Ref. [8]). Therefore the system CeNi$_x$Ga$_{1-x}$ can be viewed as an additional system in the Doniach phase diagram with the Ni concentration as control parameter.

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