Magneto-crystalline Anisotropy and non-Fermi-liquid Behavior in CeNi$_{1-x}$Co$_x$Ge$_2$

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Abstract

We present results of magnetization, AC susceptibility and heat capacity measurements on polycrystalline CeNi$_{1-x}$Co$_x$Ge$_2$ samples ($x = 0$, 0.025, 0.05, 1) which were prepared by arc melting and on crystal CeNiGe$_2$ grown by optical floating zone method in four mirror furnace. The parent compound CeNiGe$_2$ is an antiferromagnetic Kondo system that orders magnetically at $T_N = 3.8$ K and undergoes a spin structure rearrangement at $T_J = 3.2$ K while CeCoGe$_2$ is a nonmagnetic heavy-fermion Kondo compound with $j = 5/2$ ground state and large Kondo temperature $T_K > 200$ K. Our measurements showed that the phase transition from the paramagnetic to the antiferromagnetic state was suppressed to lower temperatures with an increasing concentration of dopant.

Keywords: CeNi$_{1-x}$Co$_x$Ge$_2$ compound, arc melting, AC susceptibility, magnetization, heat capacity

1 Introduction

Based on the available magnetization measurements it is known that the compound CeNiGe$_2$ has a transition from antiferromagnetic to the paramagnetic state at about 3.8 K and the CeCoGe$_2$ compound is a paramagnet in the whole temperature range. The main goal of our study is the preparation of CeNi$_{1-x}$Co$_x$Ge$_2$ single crystals using floating zone method. In most cases, the floating zone method is suitable for synthesis of high quality single crystals forming substitutional solid solution. In the first step we concentrated on preparation of CeNi$_{1-x}$Co$_x$Ge$_2$ polycrystalline materials, which were synthetized by arc melting in sufficiently wide range of concentrations. We assumed that the substitution of Ni atoms with Co leads to suppression of magnetic ordering of in this system and that will allows us to study the physical properties in the vicinity of quantum critical point (Jung, 2005, Im,
2006, Pikul 2004, Mun 2004). We performed an attempt to grow single crystal of the parent compound CeNiGe$_2$ by the optical floating zone method and Czochralski technique, respectively.

## 2 Experimental Details

Polycrystalline samples of CeNi$_{1-x}$Co$_x$Ge$_2$ ($x = 0, 0.025, 0.05, 1$) were prepared by arc melting in a mono-arc furnace under an argon atmosphere. Since the cerium has a high affinity for oxygen and cerium oxides can affect the magnetic properties of these compounds, which can lead to misinterpretation of results, it is necessary to remove the oxides from samples. Our scanning electron microscopy investigation (SEM) including energy dispersive analysis (EDX) revealed that in heavy polluted samples CeO$_2$ forms long strips in the whole volume of the sample. We tried to remove these oxides from the sample by multiple re-melting of cerium by arc melting in Ar atmosphere. After any melting procedure oxides were deposited on the surface of the sample and we removed them by subsequent grinding. We repeated this procedure at least 5 times to achieve the lowest oxide content in cerium. In next step we added remaining constituent elements with high-purity products (99.99%) in order to obtain the desired stoichiometry. Prepared samples were characterized by SEM using a secondary electron (SE) and backscattered electron (BSE). For the qualitative and quantitative determination of the phases we carried out EDX analysis. Because the EDX analysis and X-ray diffraction data probe small volume of the material related to the surface of material, we performed magnetization measurements, which allowed us to obtain satisfactory good information from the whole volume of samples. The parent compound of CeNiGe$_2$ was grown by Czochralski method in tetra-arc furnace. A wolfram wire was used as a seed and Ti as a getter, the growth procedure was performed in Ar atmosphere. The grown crystal was characterized by X-ray diffraction method using 3 axes goniometer in transmission mode on edges of the crystal. From obtained diffraction pattern we were not able to construct diffraction matrix and that is why we claim that the crystal was of quite poor quality. The crystal growth by floating zone method was very difficult because the melting zone was always created under an impurity – oxide envelope which disturbed the crystal growth process rapidly and we grew always only very short ingot approximately 1 cm long. The lamp power was chosen 60% of total power. The floating zone passes the feed material with rate of mirrors of 8mm/h and consumption of feed was regulated by additional movement of feed rod with rate of 6mm/h. Melting zone contained usually a lot of solid particles, which melted uniformly. Process of crystal growth swept about three hours, pulled crystal had length of about 20 mm. The EDX analysis showed the ingot was single phase of CeNiGe$_2$ with high homogeneity. X-ray powder diffraction confirmed that the crystal was single phase. The crystal was inspected by X-ray Laue’s method. Laue pattern indicates that the sample has a monocristalline nature, but the crystal contains lot of defects like twins and small-angle boundaries.

The magnetization measurements were carried out by SQUID magnetometer (MPMS) in magnetic field with induction up to 0.1 T and in the temperature range from 2.0 K to 300 K. Heat capacity measurements were performed on PPMS.

## 3 Experimental results

The temperature dependences of susceptibility follow the Curie-Weiss law and the negative Curie paramagnetic temperature (Figure 2) indicates an antiferromagnetic exchange interactions. The effective magnetic moment $\mu_{\text{eff}}$ has approximately the same value (2.55$\mu_B$) for all samples and the paramagnetic Curie temperature $\theta$ changes from -5.59 K to -80 K. In each case is value of $\mu_{\text{eff}}$ is close to theoretical one calculated for free Ce$^{3+}$ ion, thus indicating the presence of very well localized
cerium magnetic moments. The obtained value of $\mu_{\text{eff}}$ is very similar to the already published results on CeNiGe$_3$ (Pikul 2012) or results on CeCoGe$_2$ showing 2.54 $\mu_B$ and $\theta = -145$ K (Rotundu CR 2006); the higher value of $\theta$ in this case can be attributed to magneto-crystalline anisotropy because the measurements were performed on an oriented single crystal. Magneto-crystalline anisotropy has been observed on CeNiGe$_2$ (Pikul 2004) and depending on the crystallographic axis the effective moment ranges from 2.53 $\mu_B$ to 2.64 $\mu_B$ and the paramagnetic Curie temperature from -161 K to 23 K. We can conclude that low value of $\theta$ for CeCoGe$_2$ is not indicating the strongest antiferromagnetic interaction in this sample but reflecting magneto-crystalline anisotropy which is due to preferred orientation of crystals in the sample accidentally correlated with direction of magnetic field.

AC magnetic susceptibility measurements (Figure 1 and 2b) revealed that compound CeNiGe$_2$ has the magnetic transition temperature at $T_N = 3.8$ K. This temperature decreases with increasing concentrations of cobalt to 3.5 K for CeNi$_{0.975}$Co$_{0.025}$Ge$_2$, to 2.6 K for CeNi$_{0.95}$Co$_{0.05}$Ge$_2$ and finally completely disappears in the case of CeCoGe$_2$ as it is indicated from a maximum in $\chi'$ but a small bump can be seen at about 7 K, which can indicate the presence of CeO$_2$ impurities. Such a maximum is visible in $\chi''$ at about 7 K and the maxima at lower temperature we associate with magnetic phase transition. The heat capacity measurement in zero magnetic fields clearly shows doublet peak at about 2.3 K and 3.8 K (Figure 3). The presence of cerium oxide in the volume of material can be indicating by a small bump at about 6 K.

**Figure 1:** Temperature dependence of the inverse magnetic susceptibility for CeNi$_{1-x}$Co$_x$Ge$_2$.

**Figure 2:** Temperature dependence of the AC magnetic susceptibility for CeNi$_{1-x}$Co$_x$Ge$_2$: (a) real part, (b) imaginary part.
4 Summary

We have prepared and characterized polycrystalline samples of CeNi$_{1-x}$Co$_x$Ge$_2$ system by arc melting and we made an attempt to grow single crystal of CeNiGe$_2$ sample by optical floating zone technique and Czochralski method. All our prepared samples are mostly single phases but from bulk magnetization measurements we have got results indicating presence of CeO$_2$ in samples. The single crystals are not of very good quality. Our results signal that the zone melting is better for the crystal growth of CeNiGe$_2$ but we have to use cerium of higher quality with smaller content of oxides. Our magnetic measurements confirmed suppression of the antiferromagnetic ordering with Co–doping, but we did not find the right concentration of Co, which leads to total suppression of antiferromagnetic ordering in the system.

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References

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Figure 3: Temperature dependence of heat capacity for CeNiGe$_2$ crystal.