Onset of magnetic ordering in heavy fermion CeNi and (Ce$_{0.97}$Gd$_{0.03}$)Ni

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Abstract. Electronic states of Ce and Ni in heavy fermion CeNi were studied from both a macroscopic magnetization measurement and a microscopic way of magnetic Compton profile MCP. The behaviour of magnetization as a function of temperature resembles to that of $\alpha$-Ce in the range between 4 K and 150 K and no clear magnetic ordering was observed. The maximum value of magnetization corresponded to 0.01 $\mu_B$ of the constituent. The MCP measurement at 20 K coincided with that of magnetization. The substitution of only 3 at% of Gd for Ce was found to cause a kind of magnetic ordering and the tentative Curie temperature was assumed to be about 2 K. The magnetic structure of the substituted material (Ce$_{0.97}$Gd$_{0.03}$)Ni was possible to be a ferri-magnet from the inverse susceptibility measurement.

1. Introduction

Rare Earth (RE)-transition metal (TM) compounds have been investigated intensively and vast amounts of data have been piled up since both elements are magnetic however their mechanism of the magnetism are utterly contrast [1, 2]. Through these studies, some important principles have been proposed in order to understand the magnetic properties in RE-TM systems. One of these principles is a charge transfer model and this explains well the experimental results that the magnetic moments of TM decrease as the increase of RE content and lose at Laves phase RENi$_2$. However, very recently, this principle is not true of Gd-Ni compound system and the Ni is found to retain its magnetic moment even at GdNi of 50 at% of Gd [3, 4, 5].

CeNi should have been classified essentially to RE-TM compound since the Ce belongs to RE and the Ni is one of the transition metals. The CeNi, however, has been rather famous for a heavy fermion compound and is known to show an enhanced Pauli paramagnetism. That is, the CeNi does not order magnetically and both constituents of the Ce and Ni are not magnetic. Taking into account of the above-mentioned that the GdNi is magnetic and the Ni in GdNi does order ferro-magnetically, the Pauli-paramagnetism in CeNi seems to have a room to be reconsidered.

In this study, at first, the CeNi of single crystal was investigated from macroscopic point of view (magnetization measurement) and next, a microscopic method of magnetic Compton profile MCP
measurement [6] was carried out for the first time. Based upon these results, the substitution effect of Gd for Ce in CeNi, that is (Ce-Gd)Ni, was investigated magnetically and compared with those of CeNi in detail. Here only 3 at% substitution of Gd for Ce was discussed where the structure of the sample remains unchanged.

2. Experimental procedure
Single crystals of CeNi and (Ce0.97Gd0.03)Ni were grown by CZ pulling method in RF furnace. They were cut as long as possible in the direction of b-axis employing by the electrical discharge cutter in order to avoid the mechanical stress as much as possible.

The magnetization measurements were carried out from 2 K to 300K under the magnetic field between 50 and 20,000 Oe employing the commercial Quantum Design SQUID MPMS system.

The magnetic Compton profile was measured at BL-08W in Spring-8. The temperature was kept at 20 K under the magnetic field of 10,000 Oe. Circularly polarized synchrotron X-rays were monochromatized and focused on the sample by a single channel-cut bent Si crystal. The energy of the incident X-rays were tuned to 175 keV. The energy spectra of Compton scattered X-rays were measured by a solid-state detector (SSD). Conventionally, two energy spectra I+ and I- were measured repeatedly, where the I+ and I- denote that the direction between the sample magnetization and the polarized X-rays is parallel and anti-parallel, respectively. By taking the difference I+−I−, we can obtain only the spin-dependent Compton scattered component [6, 7].

3. Results and Discussion

3.1. CeNi
The magnetization as a function of temperature is shown in figure 1. The results shown in the figure correspond to those for magnetic fields of 5,000, 10,000 and 20,000 Oe, respectively. As the increase of temperature, each magnetization tends to decrease slightly until about 20 K and then increases and takes a maximum value around 150 K. With further increase of temperature, each one decreases again gradually. The profiles between 2 K and 150 K resemble to that of α-Ce [8], however the values are found to be much larger than those of α-Ce. The maximum value of the magnetization at 150 K correspond to about 0.01 μB for Ce or Ni atom. Such behavior as a function of temperature is difficult to be explained by the existing models at present and remains to be a hard nut to crack [9].

Figure 1. Magnetizations as a function of temperature for single crystal of CeNi. Applied magnetic fields were selected as H=20,000, 10,000 and 5,000 Oe, respectively.

Figure 2. Magnetization as a function of H/T for single CeNi crystal. The plots were carried out for three temperatures.

Figure 2 shows the H/T-plot for the CeNi single crystal. The paramagnetism is well known to obey a unique line for H/T-plot and is described by Brillouin function [10]. From the figure 2, the H/T-plot
for this sample is found not to obey to a unique line and to be sensitive to the temperature. It is considered to be characteristic of this sample.

The magnetic Compton profile MCP for the single crystal CeNi was measured at 20 K where the magnetization takes minimum from figure 1, and the result is shown in figure 3. The MCP reflects the wave-functions of magnetic electrons (that is, electrons which contribute to magnetism) and the MCP of magnetic electrons tends to populate highly in the region of \( P_z < 2 \) (a.u.) and then to decrease gradually to become nearly zero in \( P_z > 5 \) (a.u.) [6, 7, 11]. From this figure, it can be seen that the MCP in CeNi is effectively small and negligible and this sample does not order magnetically. This result coincides with the result shown in figure 1.

![Figure 3: Magnetic Compton profile MCP for a single CeNi crystal at 20 K. Applied magnetic field was selected as 10,000 Oe.](image)

![Figure 4: Magnetization as a function of temperature applying magnetic fields of 10,000 Oe for single crystal (Ce0.97Gd0.03)Ni.](image)

3.2. \((Ce_{0.97}Gd_{0.03})Ni\)

The temperature dependence of magnetization \( M(T) \) for \((Ce_{0.97}Gd_{0.03})Ni\) single crystal is shown in figure 4. From the figure, only 3 at% substitution of Gd for Ce does cause a drastic change in \( M(T) \) and the sample seems to retain a kind of magnetic ordering to some extent. The value of magnetization at 2 K in 10,000 Oe corresponds to over \( 6 \mu_B \), if the magnetic moment is responsible only for Gd atoms. The Arrott-plot was tentatively carried out for the same sample since the electronic state of the sample may take a color of heavy fermion and cannot be described perfectly in terms of magnetic localized model. The result is shown in figure 5 and the figure shows that the tentatively determined Curie temperature \( T_c \) for this sample is considered to be about 2 K.

![Figure 5: Tentative Arrott-plot for single crystal (Ce0.97Gd0.03)Ni.](image)

![Figure 6: Inverse susceptibility as a function of temperature for single crystal (Ce0.97Gd0.03)Ni.](image)
the electronic state of the sample may take a color of heavy fermion and cannot be described perfectly in terms of magnetic localized model. The result is shown in figure 5 and the figure shows that the tentatively determined Curie temperature $T_c$ for this sample is considered to be about 2 K.

Figure 5 is an inverse susceptibility as a function of temperature $\chi^{-1}(T)$. The form of the $\chi^{-1}(T)$ is characteristic of ferri-magnetism [12] and the sample substituted by only 3 at% Gd for Ce can be a ferri-magnet. It follows that at least two elements may order magnetically.

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