Magnetic order and properties in heavy fermion \((\text{Ce}_{1-x}\text{Gd}_x)\text{Ni}\) single crystal

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Abstract. Electronic states of Ni and Ce in the heavy fermion \((\text{Ce}_{1-x}\text{Gd}_x)\text{Ni}\) system were studied employing magnetic properties. The samples \((x=0.15\) and \(0.20\) showed clear magnetic order, however, the magnetizations in \(x=0.15\) and \(0.20\) unexpectedly decreased rapidly with the increase of temperature. The Curie temperatures were clearly determined by the Arrott-plot and the values were 11 K and 17 K, respectively for \(x=0.15\) and \(0.20\). The inverse susceptibilities were not linear and showed the characteristic curves for ferri-magnetism. This behaviour suggests that the Ni and/or Ce may be magnetic elements in \(x=0.15\) and \(0.20\). Based on the above-mentioned results, molecular-field analysis was carried out within the localized picture and it was found that the Gd-Gd exchange interaction was strongly suppressed and this suppression reflected the rapid decrease of magnetization.

1. Introduction

CeNi is composed of rare-earth (RE) and transition-metal (TM) and belongs to essentially the RE-TM compound system. However, CeNi shows no clear magnetic order and the unexpected and peculiar behavior of magnetism in comparison with those of other RE-TM systems and it is known to be an enhanced Pauli-paramagnetism [1]. Since Ni has been known to be non-magnetic in RENi (i.e. 1:1 content ratio) [2] and it follows that the Ce owes to its magnetism in CeNi and retains no clear magnetic moment, which is quite different from the magnetism in RE. Further, the effective mass of the electrons of CeNi is much larger than those of normal metals and CeNi has been classified rather as a heavy fermion material.

On the other hand, some recent studies have made it clear that the Ni in GdNi is magnetic and does retain the magnetic moment [3, 4, 5]. Considering that the structure of CeNi has the same as that of GdNi, the fact that the Ni and Gd in CeNi becomes non-magnetic and CeNi becomes non-magnetic is self-evident and this is worth reconsidering. According to this scenario, we have made a preliminary study in the \((\text{Ce}_{1-x}\text{Gd}_x)\text{Ni}\) system and it was found that the sample with \(x=0.03\) seems to retain a magnetic order at 2 K [6].

In this study, the content of Gd was further increased to 0.15 and 0.20 and the variations of magnetic properties were investigated in detail. Furthermore, a molecular field analysis was carried out and the analyzed results were compared with the experimental ones.
2. Experimental and analytical procedure

Single crystals of the $x=0.15$ and 0.2 in (Ce$_{1-x}$Gd$_x$)Ni were grown by the Bridgman method. The structural analysis was carried out by using Laue photographs. The samples were cut as long as possible (axial ratio $>1/3$) in order to reduce the effect of the shape anisotropy.

Magnetization measurements were performed from 2 K to 280 K in magnetic fields between 50 and 10,000 Oe employing the commercial Quantum Design SQUID MPMS system. The saturation magnetization was determined by a $1/H$ plot.

The temperature dependence of magnetization $M(T)$ was calculated and simulated within the frame of two-sublattice model [7, 8, 9]. This model describes both ferro-magnetism (for only one kind of magnetic element) and ferri-magnetism (for two kinds of magnetic elements). When three kinds of elements are magnetic, the calculation of $M(T)$ is beyond the power of this model. The parameters in the calculation were three exchange interaction energies $J_{ij}$s ($=J_{RE-RE}$, $J_{RE-TM}$ and $J_{TM-TM}$) including $J_{ij}=0$.

3. Results and Discussion

3.1. Ce$_{0.85}$Gd$_{0.15}$Ni

The temperature dependence of magnetization $M(T)$ is shown in figure 1 and the inset therein is a $M-H$ plot at 2 K. In $M(T)$, the applied magnetic fields are 10,000, 8,000, 6,000, 4,000 and 2,000 Oe. From figure 1 including the inset, the value of magnetization is great enough and this sample is considered to retain some magnetic ordering, however, the magnetization is extraordinarily sensitive to the applied field. Accordingly, the Curie temperature $T_c$ and the saturation magnetization $M_s$ seem to be not easy to determine uniquely. First, the $1/H$-plot was employed in order to determine the saturation magnetization $M_s$ and the value of $M_s$ was estimated to be 26.25 emu/g. If the Gd is the only magnetic element in this sample, the saturation magnetization is expected to become 29.17 emu/g under the assumption that Gd has $7 \mu_B$/atom. That is, this estimated value of $M_s$ suggests that some other elements except Gd should be magnetic and couple anti-parallel with the Gd moment.

Figure 1. Magnetization as a function of temperature for $x=0.15$. The inset is a $M-H$ curve.

Figure 2. Inverse susceptibility as a function of temperature for $x=0.15$. The inset is an Arrott-plot.

Figure 2 shows the inverse susceptibilities $\chi^{-1}(T)$ together with the Arrott-plot in the inset. The $\chi^{-1}(T)$ clearly deviates from the linear relationship and resembles that for ferri-magnetism [10]. That is, it is suggested that the sample with $x=0.15$ is a ferri-magnet and some other elements (Ce and/or Ni) are magnetic and couple(s) anti-parallel with that of Gd. This result does coincide with the result obtained by the $1/H$ plot and the magnetic structure of this sample is very likely ferri-magnetic.
inset in the figure shows that the Arrott-plot is linear and the Curie temperature $T_c$ can be clearly
determined. The linearity of the Arrott-plot means that the electronic state of this sample is of
localized nature and the value of $T_c$ is between 11 K and 12 K. After knowing that the Curie
temperature $T_c$ is about 11 K and seeing the figure 1, it is found that the value of magnetization at 11K
(=$T_c$) in magnetic field of 10,000 Oe is much larger than half of the $M_s$ and even in 4,000 Oe the value
is about half. This high value of magnetization at $T_c$ is not regular and this is considered to reflect that
this sample is considerably sensitive to the magnetic field.

3.2. Ce$_{0.80}$Gd$_{0.20}$Ni

Figure 3 shows the temperature dependence of magnetization $M(T)$ and the $M$-$H$ curves at 2 K in the
inset. Qualitatively, the $M(T)$ and $M$-$H$ curve are nearly the same as those in figure 1 and the inset,
respectively. By using the $1/H$ plot, the saturation magnetization $M_s$ was also estimated to be 37.00
emu/g and this value is a little smaller than the value of 38.68 emu/g, which corresponds with that of
Gd atoms under the assumption of $7 \mu_B$/atom for Gd. This value also suggests that Ni and/or Ce atoms
become(s) magnetic and couple anti-parallel with Gd atoms. In this content, the magnetizations are
also considerably sensitive to magnetic field, however, less suppressed in comparison with that in
$x=0.15$.

![Figure 3. Temperature dependence of magnetization for $x=0.20$. The inset is a $M$-$H$ curve at 2 K.](image1)

![Figure 4. Temperature dependence of inverse susceptibility for $x=0.20$. The inset is an Arrott-plot.](image2)

The reciprocal susceptibility $\chi^{-1}(T)$ is shown in figure 4 together with the Arrott-plot in the inset.
From these two figures, the $T_c$ of this sample is found to be about 17 K. In addition, the Arrott-plot
follows fairly well the linear relationship and can be described by the Brillouin function near the Curie
temperature. It follows that the electronic state of the $x=0.20$ can be also described well by the
localized model.

3.3. Molecular field analysis

As mentioned above in 3.1 and 3.2, it is found that the electronic states regarding the magnetism in
$x=0.15$ and 0.20 can be described in terms of a localized model. In this section, a molecular field
analysis was employed and was tried to reproduce qualitatively the characteristic decrease of $M(T)$
within the localized two-sublattice model. Here, the model employed is that the Gd and Ni atoms are
magnetic and couples anti-parallel to each other. The Ce was assumed to be non-magnetic in this case.
Figure 5 is the calculated result and three parameters $J_{ij}$’s ($J_{\text{Gd-Gd}}$, $J_{\text{Gd-Ni}}$ and $J_{\text{Ni-Ni}}$) used in calculation
are shown in the figure.
The calculated result seems to qualitatively reproduce the rapid decrease of $M(T)$ observed in $x=0.15$ and $x=0.20$. It is found that the $M(T)$ decreases more rapidly as the decrease of $J_{\text{Gd-Gd}}$. Normally observed values of exchange interaction energy $J_{\text{Gd-Gd}}$ are between 1 K and 5 K [9] and in GdNi$_2$, for example, the $J_{\text{Gd-Gd}}$ is 1.75 K [5]. It may be surprising that the value $J_{\text{Gd-Gd}}$ used for the calculation is much smaller and suppressed in comparison with the normal values. It follows that the suppression of $J_{\text{Gd-Gd}}$ may be essential in the drastic decrease of magnetization in $x=0.15$ and 0.20.

![Figure 5. Simulated magnetization as a function of temperature $M(T)$. Only Gd-Gd exchange interaction $J_{\text{Gd-Gd}}$ was changed, with other two parameters $J_{\text{Gd-Ni}}$ and $J_{\text{Ni-Ni}}$ being kept unchanged.](image)

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