Collapse-Like Decrease of RKKY Interaction and Kondo Effect in Heavy Fermion Compounds (Ce_{1-x}Gd_x)Ni (0.03≤X ≤0.20)

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Abstract. Small amounts of Ce were substituted by Gd and the (Ce-Gd)Ni heavy fermion compounds were investigated in detail from viewpoint of magnetism in Gd-poor region between 3 at% and 20 at%. Magnetizations showed peculiar linear relationship as a function temperature and were sensitive to the applied magnetic field in common. These characteristic natures were analyzed by employing molecular field analysis by changing three exchange interaction energies, \( J_{\text{Gd-Gd}} \), \( J_{\text{Gd-Ni}} \) and \( J_{\text{Ni-Ni}} \). The unique solution for the linear relationship in the temperature dependence of magnetization \( M(T) \) was found to be attributed to the collapse-like decrease of \( J_{\text{Gd-Gd}} \) and the decrease of \( J_{\text{Gd-Gd}} \) can explain the sensitivity to applied magnetic field. However this peculiar decrease of \( J_{\text{Gd-Gd}} \) did not effect on the inverse susceptibilities above the Curie temperatures and the \( \chi^{-1}(T) \) behaved normally as ferri-magnet.

1. Introduction

It is well-known that CeNi is one of the most famous heavy fermion compounds in strongly correlated electron systems and the investigation for compound systems has been a classic theme. From a magnetic point of view, CeNi shows no clear magnetic ordering and is classified as an enhanced Pauli-paramagnet [1]. On the other hand, CeNi belongs to basically to rare-earth (RE)-transition metal (TM) systems and vast amount of investigations have been piled up for RE-TM systems [2, 3]. From the magnetism in RE-TM systems, CeNi seems to be an exceptional case under the assumption that the Ce is trivalent in periodic table because the Ce\(^{3+}\) should be magnetic according to Hund rule. Considering that the Ni is well-known to decrease its magnetic moment as the increase of RE content and loses it at 33.3 at% of RE and in CeNi, where RE is 50 at%, Ni should be nonmagnetic and therefore the Ce is the only magnetic element. It follows that the Ce takes probably Ce\(^{4+}\) state and 4f electron is considered to be excited in higher levels and negligible in 4f level.

On the other hand, we have made it clear that the Ni does not lose its magnetic moment even in GdNi [4, 5, 6], which has the same structure as that of CeNi, and it becomes not self-evident that the Ni is nonmagnetic in CeNi. Therefore, it is worth reconsidering the electronic states of Ni including Ce and we have investigated the (Ce-Gd)Ni system from magnetic properties [7, 8].

In this study, we analyzed the temperature dependence of magnetization at low contents of Gd between 3 at% and 20 at% employing molecular field analysis and derived out the unique solution for showing the unusual linear relationship in \( M(T) \) and the sensitivity to applied magnetic field.
2. Experimental and analytical procedure

Single crystals of \(x=0.15\) and 0.20 were grown by the Bridgman method and the \(x=0.03\) was grown by the Cz pulling method in RF furnace. The structural analysis was carried out by using Laue photographs. The samples were cut as long as possible in direction of b-axis in order to reduce the demagnetization factor.

Magnetization measurements were performed from 2 K to 280 K under the magnetic field between 50 and 10,000 Oe employing the commercial Quantum Design SQUID MPMS system. The values of saturation magnetization were determined by a \(1/H\) plot. The Curie temperatures were obtained by a reciprocal susceptibility.

The temperature dependence of magnetization \(M(T)\) was calculated by molecular field analysis with two-sublattice model [9, 10, 11]. This model describes well the \(M(T)\) of ferri-magnet where two kinds of magnetic elements couple antiparallel. When three kinds of elements are magnetic, this model cannot calculate the magnetization and in this paper, the Ce is assumed to be nonmagnetic, whose assumption seems to be realistic in Ce-rich region. The parameters in calculation were three exchange interaction energies \(J_{ij}\)s (\(J_{Gd-Gd}\), \(J_{Gd-Ni}\) and \(J_{Ni-Ni}\)).

3. Results and Discussion

3.1. Temperature dependence of magnetization; \(M(T)\)

The temperature dependence of magnetizations for three kinds of contents are shown in figure 1, where (a) is for Gd content \(x=0.03\) (3 at %), (b) is for \(x=0.15\) and (c) is for \(x=0.20\). In the \(M(T)\)s for three contents, the applied magnetic fields are 10,000, 8,000, 6,000, 4,000 and 2,000 Oe. The typical behavior in \(M(T)\) is observed for \(x=0.15\) and the magnetization decreases linearly as the increase of temperature. This linear behavior in \(M(T)\) is not popular and is characteristic of the low contents of Gd (=0.03-0.20). These characteristic linear relationships in \(M(T)\) are analyzed by molecular field analysis in next section 3.2. Another characteristic feature in these contents (0.03-0.2) is that the magnetizations are considerably sensitive to the applied magnetic field. However, the Curie temperatures \(T_C\) can be determined by a reciprocal susceptibility as shown in figure 3 and the values are estimated to be about 2 K (\(x=0.03\)), 11-12 K (\(x=0.15\)) and 18-19 K (\(x=0.20\)), respectively.

![Fig. 1 Temperature dependence of magnetization for \(x=0.03\) (a), \(x=0.15\) (b) and \(x=0.20\) (c), respectively. Applied magnetic fields are 10,000, 8,000, 6,000, 4,000 and 2,000 Oe.](image)

3.2. Molecular field analysis

In this section, the characteristic linear relationship behavior of \(M(T)\) in low content of Gd (\(x=0.15, 0.20\)) is analyzed within the frame of 2 sublattice model (Gd and Ni=magnetic, Ce=nonmagnetic) and the unique solution for showing this linear relationship is investigated in detail.

As parameters in calculation, three exchange interaction energies \(J_{ij}\)s (\(J_{Gd-Gd}\)=\(J_{11}\), \(J_{Gd-Ni}\)=\(J_{12}\) and \(J_{Ni-Ni}\)=\(J_{22}\)) are necessary to be determined and at first, the values for amorphous Gd_{50}Ni_{50} alloy were employed. In order to find out the conditions for showing the linear behavior in \(M(T)\), each \(J_{ij}\) was varied independently.
Figure 2(a)-(c) show the calculated results of $M(T)$ for the Gd content $x=0.15$ (15 at %). In (a), (b) and (c), $J_{11}$, $J_{12}$ and $J_{22}$ were varied, respectively. From figure 2(a), as the decrease of the exchange interaction energy $J_{11}$ ($=J_{\text{Gd-Gd}}$), the $M(T)$ becomes gradually linear and in case of $J_{11}=0.1$ K, the $M(T)$ takes a linear relationship and this is found to be one possible condition for a linear behavior in $M(T)$. That is, in this case, $J_{\text{Gd-Gd}}$ ($=\text{RKKY interaction energy}$) is found to be considerably suppressed. In the figure 2(b), the $J_{12}$ ($=J_{\text{Gd-Ni}}$) is varied from -5 K to -0.1 K (- means antiparallel coupling between Gd and Ni) and this $J_{12}$ is found to effect little on the behavior of $M(T)$. Accordingly, the $J_{12}$ is found to be not an essential parameter for the linear behavior. Furthermore in figure 2(c), the $J_{22}$ ($=J_{\text{Ni-Ni}}$) was varied from 100 K to 40 K. From the figure, the $J_{22}$ is also found to change little the form of $M(T)$ and this parameter is also fond to be not an essential candidate for the linear variation. After all, the unique solution for showing the linear relationship in $M(T)$ is found to be attributed to the collapse-like decrease of $J_{\text{Gd-Gd}}$, that is, RKKY interaction. Further, this collapse-like decrease of RKKY interaction can explain the sensitivity of magnetizations to applied magnetic field, as shown in figure 1, since the decrease of the $J_{11}$ causes the decrease of molecular field and consequently the magnetization becomes sensitive to applied magnetic field. That is, the reason why the magnetizations are sensitive to magnetic field can be ascribed to the collapse-like decrease of $J_{\text{Gd-Gd}}$ ($=\text{RKKY}$) interaction.

![Graph](image1)

![Graph](image2)

![Graph](image3)

Fig. 2 Calculated magnetization behaviors as a function temperature. (a); $J_{11}$ is a parameter, (b); $J_{12}$ is variable, (c); $J_{22}$ is a parameter.

3.3. Reciprocal susceptibility
Considering that the linear relationship of $M(T)$ stems from the unusual collapse-like decrease of $J_{11} (=J_{Gd-Gd})$, it is interesting to check whether this unusual decrease of $J_{11}$ causes some kind of irregular behaviors in reciprocal susceptibilities $\chi'(T)$ or not.

Figure 3 (a), (b) show the reciprocal susceptibilities for $x=0.15$ and 0.20 and the solid lines are guides for eyes. At a glance, both reciprocal susceptibilities resemble to those of ferrimagnet and there seem to be no conspicuous irregular behaviors in $\chi'(T)$ and they are rather normal for ferrimagnet [12]. Consequently, in the $\chi'(T)$ for both $x=0.15$ and 0.20, no irregular behaviors are found. This result is rather reasonable since the exchange interactions including some kind of unusual $J_{11} (=J_{Gd-Gd})$ are almost negligible over Curie temperature $T_c$.

![Fig. 3](image-url)

**Fig. 3** Temperature dependence of reciprocal susceptibility for $x=0.15$ (a) and $x=0.20$ (b).

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