NON-MAGNETIC GROUND STATE AND FIELD-INDUCED TRANSITION IN $PrInNi_4$*

Hiroyuki Suzuki, Naohito Tsujii, Osamu Suzuki
Hideaki Kitazawa, Hideki Abe, Motoharu Imai
and Giyuu Kido

National Institute for Materials Science, Tsukuba, Ibaraki, 305-0047, Japan

(Received July 10, 2002)

The magnetic properties of $PrInNi_4$ with the field-induced ferromagnetic transition have been investigated by the measurements of the magnetic susceptibility and the high-field magnetization up to 30 T. The experimental results have been described in terms of the single-site effect of the crystalline-electric-field and the inter-atomic exchange interaction with a ferromagnetic coupling.

PACS numbers: 75.30.Kz, 75.30.Cr

Systems including non-Kramers rare-earth ion under cubic symmetry are of special interest from the viewpoint of their characteristic crystalline-electric-field (CEF) states. For example, the Hund's rule ground state for $^3H_4$ of Pr^{3+} ion splits in a cubic CEF into one singlet, Γ_1, one doublet, Γ_3, and two triplet, Γ_4 and Γ_5. Here, it is noted that the non-Kramers doublet of Γ_3 is non-magnetic but has quadrupolar moments. When Γ_3 ground state with a relatively large CEF splitting is realized, it is suited for the investigation of a pure quadrupolar system. They are, however, rare cases, for example, $PrPb_3$ [1], $PrInAg_2$ [2] and PrPtBi [3]. Very recently, the polycrystalline sample of $PrInNi_4$ with the cubic $MgSnCu_4$-type structure has been prepared and its physical properties have been reported [4]. The magnetic susceptibility, $\chi(T)$, and the magnetization curve, $M(H)$, have revealed a non-magnetic CEF ground state and the field-induced ferromagnetic transition at low temperatures. In this paper, we discuss the experimental results of $\chi(T)$ and high-field $M(H)$ up to 30 T in terms of CEF and ferromagnetic interaction. Our analysis shows that the field-induced transition can be explained by a mean-field approximation model and that the CEF ground state is Γ_3.

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

As shown in Fig. 1, $1/\chi(T)$ increases almost linearly above 50K, *i.e.* it seems to follow the Curie–Weiss law, but with a small upward curvature, indicating a relatively large CEF splitting. Below about 20 K, $1/\chi(T)$ decreases rapidly. The electrical resistivity also shows a rapid decrease below 20 K [4]. These behavior suggest a characteristic energy scale of $T^* \sim 20$ K in this system. While the rapid decrease of $1/\chi(T)$ below T^*, *i.e.* an enhancement of χ, implies a development of magnetic interactions, a tendency of $\chi(T)$ to flatten around the lowest temperature suggests a non-magnetic ground state. These features are key-points to understand the origin of the field-induced ferromagnetic transition. This will be discussed later. The high-field $M(H)$ up to 30 T at $T = 2$ K, as shown in Fig. 2, is found to saturate above 20 T to a value of $\sim 2.4\ \mu_B$, which is smaller than that expected from Pr^{3+} (3.2 μ_B). This also indicates the relatively large CEF splitting. $M(H)$ shows a sharp jump at $H_m = 0.65$ T. In Ref. [4], it is reported that this H_m shifts to higher values with the increase of the temperature.

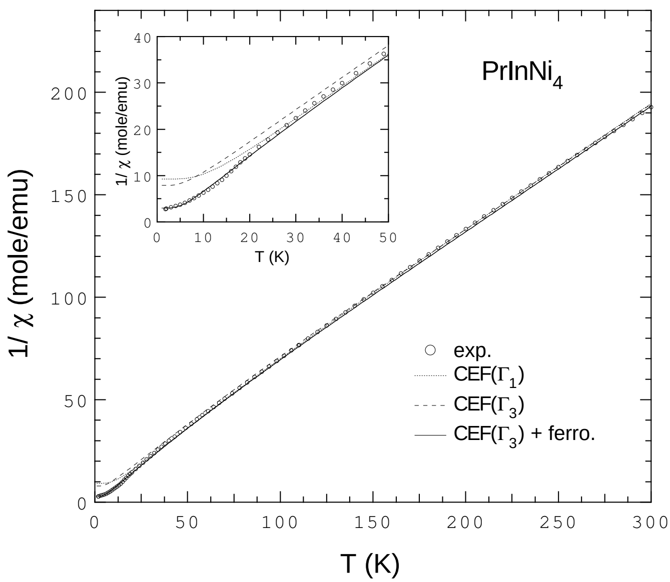

Fig. 1. The temperature dependence of $1/\chi$. The dots, dashed and solid lines represent the calculated results obtained by considering CEF with Γ_1 ground state (Γ_1), CEF (Γ_3) and CEF (Γ_3) + ferromagnetic interaction, respectively. For the CEF level schemes used in the calculation, see the text.

First, we discuss CEF level schemes. In possible CEF level schemes having a non-magnetic ground state, Γ_1 ground and Γ_5 1$^{\text{st}}$ excited state (Γ_1–Γ_5) and Γ_3–Γ_5 level schemes are excluded because $\chi(T)$ for these schemes with a relatively large CEF splitting decreases toward $T = 0$ K below the temperature of the 1st excited level, resulting in the appearance of a peak in $\chi(T)$, which is inconsistent with the experimental result. The obtained CEF level schemes that can fit $1/\chi(T)$ above 20 K and give the value of M ($\sim 2.4\,\mu_{\mathrm{B}}$/f.u.) around 30 T at 2 K are Γ_1(0 K), Γ_4(29 K), Γ_3(50 K), Γ_5(531 K) and Γ_3(0 K), Γ_4(18 K), Γ_1(43 K), Γ_5(481 K), corresponding to the CEF parameters, by Lea, Leask and Wolf [5], of $x \approx 0.88$, $W \approx -13$ and $x \approx 0.84$, $W \approx -12$, respectively. Note that the splitting between the ground and 1st excited states for Γ_3–Γ_4, Δ_1, is comparable with T^*. As can be seen from Fig. 1 and 2, the calculated results in terms of CEF can not explain the rapid decrease of $1/\chi(T)$ below 20 K and a rise in $M(H)$ at lower fields.

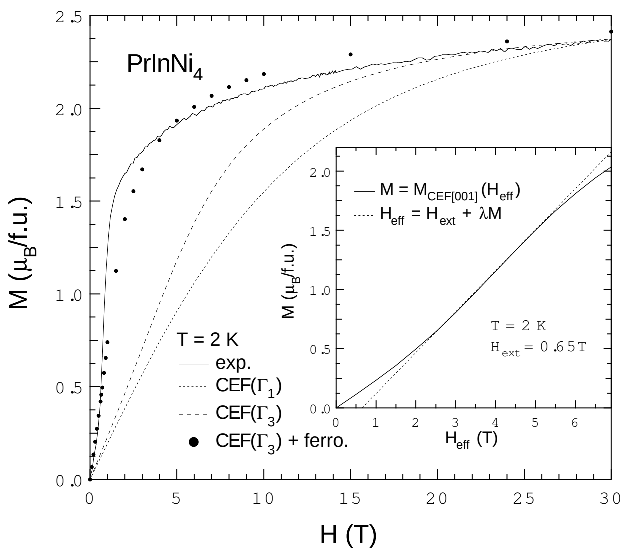

Fig. 2. High field magnetization at 2 K. The dots and dashed lines and solid circles represent the calculated results obtained by considering CEF with Γ_1 ground state (Γ_1), CEF (Γ_3) and CEF (Γ_3) + ferromagnetic interaction, respectively. For the CEF level schemes used in the calculation, see the text. The inset plots two equations of $M = M_{\mathrm{CEF}H||[001]}(H_{\mathrm{eff}})$ for Γ_3–Γ_4 at $T = 2$ K and $H_{\mathrm{eff}} = H_{\mathrm{ext}} + \lambda M$ at $\mathrm{H_{eff}} = 0.65$ T.

In order to explain the field-induced transition and the above mentioned disagreement between experiment and calculation, we introduce ferromagnetic interaction into the analyses. Here, we focus on the calculated CEF $M(H)$ ($H||[001]$) for Γ_3–Γ_4, $M_{\mathrm{CEF}[001]}(H)$, which is the only curve with a downward curvature, as shown in the inset of Fig. 2, in all $M_{\mathrm{CEF}}(H)$ curves for the three symmetry axes for Γ_1–Γ_4 and Γ_3–Γ_4. In the mean field approximation of the ferromagnetic interaction, the effective magnetic field can be described as $H_{\rm eff} = H_{\rm ext} + \lambda M$, where $H_{\rm ext}$ is the external field and λ is the ferromagnetic exchange parameter. As can be seen from the inset of Fig. 2, the two equations described in the inset intersect at two or three points with given $H_{\rm eff}$ and λ, resulting in that the magnetization moves to a higher point to gain the energy. Using the condition that these two equations meet in two points at $H_{\rm ext}$ $(= H_m) = 0.65$ T and $T = 2$ K, the exchange energy is estimated as $J^* = \lambda/(g\mu_{\rm B})^2 \sim 1.25$ K. The temperature dependence of H_m can be explained qualitatively by this model. The calculated magnetization of the powder, averaged over three symmetry axes, as plotted in Fig. 2, can reproduce roughly experimental data.

Next we perform the calculation of $\chi(T)$ for the Γ_3–Γ_4 case. Here, taking into account of the development of the magnetic interaction observed in $\chi(T)$ below T^* and $T^* \sim \Delta_1$ for Γ_3–Γ_4, the ferromagnetic interaction is introduced only into the van-Vleck term of Γ_3–Γ_4 in the calculation of $\chi(T)$. The calculated results with J^*, as plotted in Fig. 1, can explain well the rapid decrease of $1/\chi(T)$ below 20 K. In a preliminary specific heat measurement, a phase transition at $T_{\rm c} = 0.8$ K and a Schottky-type peak with a maximum of about 5 J/mole K at 7 K were observed. The calculated results using the mean field approximation with J^* predict a spontaneous magnetization around $T = 1$ K, which is comparable with $T_{\rm c}$. The latter Schottky-type peak can be also reproduced by the calculated result in terms of doublet-triplet (Γ_3–Γ_4) thermal excitation with the splitting of 18 K. These agreements strongly support our analyses in terms of the CEF and the ferromagnetic interactions.

In $\rm PrInNi_4$, a balance between the CEF splitting with the non-magnetic Γ_3 ground state and the ferromagnetic interaction introduces the field-induced ferromagnetic transition. For more quantitative analysis, the measurement on the single crystal is necessary. The preparation of the single crystal and the measurements of the specific heat and transport properties are now in progress and their results will be published elsewhere.

REFERENCES

[1] W. Gross, K. Knorr, A.P. Murani, K.H.J. Buschow *Z. Phys.* **B37**, 123 (1980).

[2] R.M. Galera, J. Pierre, E. Siaud, A.P. Murani *J. Less-Common Met.* **97**, 151 (1984).

[3] M. Kasaya, H. Suzuki, D. Tazawa, M. Shirakawa, A. Sawada, T. Osakabe *Physica B* **281-282**, 579 (2000).

[4] N. Tsujii, H. Kitazawa, H. Suzuki, M. Imai, G. Kido *J. Phys. Soc. Jpn.* **71**, 1852 (2002).

[5] K.R. Lea, M.J.M. Leask, W.P. Wolf *J. Phys. Chem. Solids* **23**, 1381 (1962).