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The magnetism of $PrPd_2Ga_3$

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Abstract

The mean-field approach and crystal-field theory were employed in this work to study the magnetism of $PrPd_2Ga_3$. We have derived several expressions for the magnetization induced by an external magnetic field and have determined the condition for occurrence of spontaneous magnetic ordering. Our theoretical analyses explain well why the system is paramagnetic in absence of a magnetic field and suggest that the system is always magnetically ordered if an external field is applied in the *ab*-plane, but remains paramagnetic for the magnetic field applied along *c*-direction.

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1. Introduction

The compounds $PrPd_2Al_3$ and $PrPd_2Ga_3$ crystallize in hexagonal $PrNi_2Al_3$-type of crystal structure, which consists of Pr–Ni layers, alternating along the *c*-direction with isolated Al layers. Both compounds were reported to remain paramagnetic down to very low temperatures [1,2]. The concluded paramagnetic ground state was attributed to specific crystal-field schemes. For example, the lowest crystal-field (CF) level is a *non-magnetic* singlet $|0\rangle$, and the first excited CF level is a doublet $|\pm 1\rangle$. However, since the matrix elements of $J_\pm$ between the two CF levels are non-zero, the systems would in certain cases order magnetically in the *ab*-plane according to the established theory [3,4]. On the other hand, one cannot downright conclude that the magnetic ordering would occur only when the energy of the RKKY (Ruderman–Kittel–Kasuya–Yosida) coupling [5–7] is comparable to the energy separation between the two CF levels. Additionally, it is still not clear how such special systems, if they are intrinsically anisotropic, will respond to an external magnetic field when it is applied in the *ab*-plane or the *c*-direction.

To answer these questions and to understand the magnetic behavior of these materials quantitatively, we took $PrPd_2Ga_3$ as an example in this work to study the effects of the CF interaction, of the RKKY coupling between the neighboring magnetic ions and of application of an external magnetic field on the magnetic state. In the following, we will apply the mean-field approach and perturbation theory.

2. Induced and spontaneous magnetic ordering within mean-field theory

As indicated above, the lowest CF level of $PrPd_2Ga_3$ is a non-magnetic singlet $\Gamma_1 = |0\rangle$. The first excited CF level is a doublet $\Gamma_5 = |\pm 1\rangle$, which is 5.2 meV above Γ_1. The other levels are well separated from the first excited level. Thus at very low temperatures, only the two lowest levels are thermally populated, so that we only consider these CF states in following formulation, and we denote them as $|g\rangle = |0\rangle$ and $|e_{1,2}\rangle = |\pm 1\rangle$, respectively. We expect from this CF-level scheme that, if the crystal orders magnetically, it orders in the *ab*-plane. When an external magnetic field is applied along *x*-axis, situated in the *ab*-plane, the

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Hamiltonian becomes in the mean-field approximation

$$\mathscr{H} = \mathscr{H}_{\rm CF} - \tfrac{1}{2}[(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}g_J B)J_+ + (\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J)J_-], \quad (1)$$

where $\mathscr{H}_{\rm CF}$ denotes the CF interaction, and $\mathscr{I}_{ff}$ is the RKKY exchange integral.

In vector space with the three states as the base, the Hamiltonian matrix is given by

$$\mathscr{H} = \begin{pmatrix} \varepsilon_1^{(0)} & -\sqrt{5}(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B) & -\sqrt{5}(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}B) \\ -\sqrt{5}(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}g_J B) & \varepsilon_2^{(0)} & 0 \\ -\sqrt{5}(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B) & 0 & \varepsilon_2^{(0)} \end{pmatrix}, \quad (2)$$

where $\varepsilon_1^{(0)}$ and $\varepsilon_2^{(0)}$ are the eigenenergies of the pure CF interaction. For convenience, we set $\Delta = \varepsilon_2^{(0)} - \varepsilon_1^{(0)}$. By diagonalizing this matrix, we have the eigenenergies:

$$\begin{aligned} \varepsilon_1 &= \Delta, \\ \varepsilon_2 &= \tfrac{1}{2}\left[\Delta + \sqrt{\Delta^2 + 40(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}g_J B)}\right], \\ \varepsilon_2 &= \tfrac{1}{2}\left[\Delta - \sqrt{\Delta^2 + 40(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}g_J B)}\right], \end{aligned} \quad (3)$$

and the corresponding eigenstates

$$\begin{aligned} |\varphi\rangle = &\frac{1}{\sqrt{(\Delta-\varepsilon)^2 + 5(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)^2 + 5(\mathscr{I}_{ff}\ J_-\rangle - \mu_{\rm B}g_J B)^2}} \\ &\times[(\Delta-\varepsilon)|g\rangle + \sqrt{5}(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}g_J B)|e_1\rangle \\ &+ \sqrt{5}(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)|e_2\rangle] \end{aligned} \quad (4)$$

in which the expressions for ε have been substituted. We notice from the above equations that new states are formed due to both the RKKY coupling and the external magnetic field. In particular, the original excited states are mixed into the new lowest one, which consequently becomes magnetic; one new level (e_1) remains at Δ, but the other two (e_2 and e_3) are shifted, producing a new level below the lowest CF state if magnetic ordering really occurs.

At $T=0$, only the new lowest state is involved in magnetic ordering. Therefore, we obtain the following expectation values of J_+ and J_-:

$$\begin{aligned} \langle J_+\rangle &= \frac{10(\Delta-\varepsilon_3)(\mathscr{I}_{ff}\langle J_+\rangle + \mathscr{I}_{ff}\langle J_-\rangle - 2\mu_{\rm B}g_J B)}{(\Delta-\varepsilon_3)^2 + 5(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)^2 + 5(\mathscr{I}_{ff}\langle J_-\rangle - \mu_{\rm B}g_J B)^2} \\ &= \langle J_-\rangle. \end{aligned} \quad (5)$$

Inserting ε_3 into above expression gives,

$$\langle J_+\rangle = \frac{40[\Delta + \sqrt{\Delta^2 + 40(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)^2}](\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)}{[\Delta + \sqrt{\Delta^2 + 40(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)^2}]^2 + 40(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)^2}, \quad (6)$$

an equation for evaluating $\langle J_+\rangle$ with arbitrary values of the external magnetic field and $\mathscr{I}_{ff}$.

For isostructural crystal $NdPd_2Ga_3$, the Heisenberg-like coupling constant has been estimated to be 1.14 K by fitting its transition temperature [8]. Applying the de Gennes rule, we obtain $\mathscr{I}_{ff} = 0.61$ K for $PrPd_2Ga_3$. By taking $\mathscr{I}_{ff} = 0.61$ K and $\Delta = 5.2$ meV, but varying the external field along the x-axis, we have calculated the expectation values of $\langle J_+\rangle$ as shown in Fig. 1. Surprisingly, the system always exhibits magnetic ordering in the presence of the external magnetic field no matter how weak the field is and irrespective of the energy separation between the CF levels.

For the initial magnetization, both B and $\langle J_+\rangle$ must be considerably small so that

$$\frac{40(\mathscr{I}_{ff}\langle J_+\rangle - \mu_{\rm B}g_J B)^2}{\Delta^2} \ll 1. \quad (7)$$

Neglecting the higher-order terms in above expression, we obtain

$$\alpha\langle J_+\rangle^3 + \beta\langle J_+\rangle^2 + \gamma\langle J_+\rangle + \delta = 0, \quad (8)$$

with the coefficients

$$\begin{aligned} \alpha &= 45\mathscr{I}_{ff}^2, \\ \beta &= -90\mu_{\rm B}g_J B\mathscr{I}_{ff}, \\ \gamma &= 2\Delta^2 - 40\mathscr{I}_{ff}\Delta + 45(\mu_{\rm B}g_J B)^2, \\ \delta &= 40\mu_{\rm B}g_J B\Delta. \end{aligned} \quad (9)$$

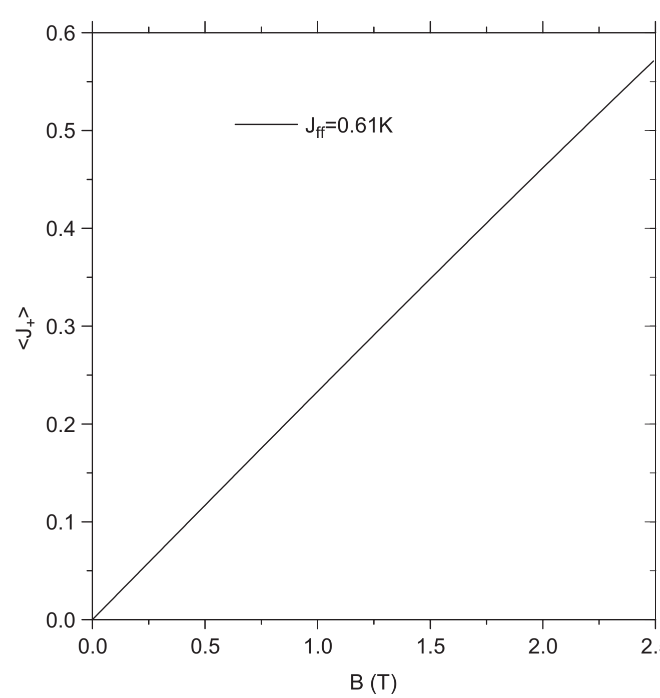

Fig. 1. Calculated $\langle J_+\rangle$ in the external field at $T=0$, where $\mathscr{I}_{ff} = 0.61$ K.