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A SIMPLE LOCALIZED–ITINERANT MODEL FOR $PrAl_3$: CRYSTAL FIELD AND EXCHANGE EFFECTS

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We present a simple magnetic model for $PrAl_3$. The effects of crystal field are treated using a reduced set of levels and the corresponding wave functions are extracted from the actual crystal field levels of Pr^{+3} in a hexagonal symmetry. The exchange between $4f$- and conduction-electrons are dealt within a molecular field approximation. An analytical magnetic state equation is derived and the magnetic behaviour discussed. The parameters of the model are estimated from a fitting of the inverse susceptibility of $PrAl_3$ given in the literature.

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1. Introduction

The starting point to understand basic magnetic quantities of rare-earth intermetallics consists in considering on equal foot the splitting of the $4f$-levels of the rare-earth ions due to the crystal field and the exchange interaction between the 4*f*- and the conduction-electrons. Usually the crystal field Hamiltonian and level scheme are presented within the Lea–Leask–Wolf notation [1] and the effects of conduction electrons in an effective exchange interaction coupling the spins of the rare-earth ion [2].

In this paper we study the magnetic properties of $PrAl_3$ from a model in which the crystal field description is considerably simplified and the role of the conduction electrons, which produce an effective exchange magnetic field at the 4*f*-electrons of Pr^{+3}, is made explicit.

The structure of the paper is as follows. In Section 2 the model Hamiltonian and the magnetic quantities are presented, in Section 3 the magnetic state equation for the ionic and electronic magnetization are derived and an explicit expression for the ionic susceptibility is obtained in Section 4. Finally, in Section 5 an application of the results of the model to $PrAl_3$ is discussed and the parameters of the model are estimated using the experimental temperature dependence of the susceptibility of $PrAl_3$ [3].

2. Model Hamiltonian and magnetic quantities

In the molecular field approximation the model Hamiltonian is

$$H = H_{\text{ion}} + H_{\text{el}}, \tag{1}$$

where

$$H_{\text{ion}} = H_{\text{CF}} + H^{\text{i}}_{\text{exch}}, \tag{2a}$$

$$H_{\text{el}} = H_{\text{kin}} + H^{\text{e}}_{\text{exch}}. \tag{2b}$$

H_{CF} describes the crystal field effects and it is discussed elsewhere. H_{kin} is related to the dynamics of the conduction electrons; from it one can derive the electronic energy density of states which is of interest to the magnetism of the conduction band. In this paper we adopt a rectangular shape for the density of states.

$$H^{\text{i}}_{\text{exch}} = -\mu_{\text{B}} h_{\text{i}} \sum_j g J^z_j, \tag{3a}$$

$$H^{\text{e}}_{\text{exch}} = -2\mu_{\text{B}} h_{\text{e}} \sum_j s^z_j, \tag{3b}$$

where

$$\mu_{\text{B}} h_{\text{i}} = \mu_{\text{B}} h_0 + J_0 \langle s^z \rangle, \tag{4a}$$

$$2\mu_{\text{B}} h_{\text{e}} = 2\mu_{\text{B}} h_0 + J_0 \langle J^z \rangle. \tag{4b}$$

In Eqs. (3) and (4) μ_B is the Bohr magneton, h_0 is an applied magnetic field, g is the Landé factor and J^z and s^z are the z components of the total angular momentum of $4f$-electrons and the conduction-electron spin respectively.

Expressions $H^{\rm i}_{\rm exch}$ and $H^{\rm e}_{\rm exch}$ in Eqs. (3a) and (3b) come from the molecular field approximation of the actual exchange interaction:

$$H_{\rm exch} = -2J_{\rm exch}(g-1)\sum_i s_i^z J_i^z. \tag{5}$$

The parameter J_0 in Eq. (4) is

$$J_0 = \frac{J_{\rm exch}(g-1)}{g}. \tag{6}$$

In this paper the $H_{\rm CF}$ will be constructed taking into account only the two lowest levels of Pr^{+3} in a hexagonal symmetry [2] (Fig. 1 shows the three first levels of the complete level scheme [3]). In the case of Pr^{+3} in $\mathrm{PrAl_3}$, according to Mader et al. [3], these two levels are singlets separated by an energy gap $\Delta = 3.41$ meV. The eigenfunctions of these levels are:

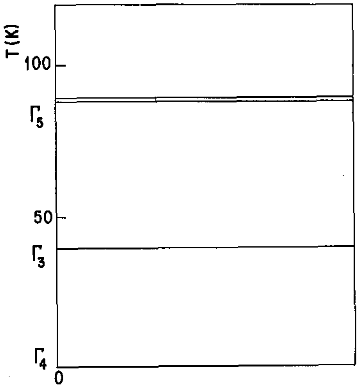


Fig. 1. The three first levels of Pr^{+3} in $\mathrm{PrAl_3}$, extracted from the complete level structure from [3]. In this paper only the two first levels were used.

$$|e_0\rangle = \frac{1}{\sqrt{2}}(|-3\rangle + |3\rangle). \tag{7a}$$

$$|e_1\rangle = \frac{1}{\sqrt{2}}(|-3\rangle - |3\rangle). \tag{7b}$$

Within the basis defined by (7)

$$H_{\rm CF} = \begin{pmatrix} 0 & 0 \\ 0 & \Delta \end{pmatrix} \tag{8}$$

and

$$H^{\mathrm{i}}_{\mathrm{exch}} = -\mu_{\mathrm{B}} h_{\mathrm{i}} \begin{pmatrix} 0 & \alpha_0 \\ \alpha_0 & 0 \end{pmatrix} \tag{9}$$

where

$$\alpha_0 = \langle e_0 | g J^z | e_1 \rangle . \tag{10}$$

In what follows α_0 is treated as a free parameter (g is an effective Landé factor).

Our main quantities of interest are the electronic and ionic magnetizations $2\langle s^z\rangle$ and $\langle gJ^z\rangle$ (in units of μ_{B}). In the next section magnetic state equations relating these quantities to the model parameters, the external magnetic field and temperature are derived.

3. Magnetic state equations

The ionic magnetization is given by

$$\mu_{\mathrm{B}}\langle gJ^z\rangle = -\frac{\sum_{j=0}^{1} \frac{\partial E_J}{\partial h_{\mathrm{i}}} \exp(-\beta E_j)}{\sum_{j=0}^{1} \exp(-\beta E_j)}, \tag{11}$$

where E_j are the eigenvalues of (2a), given in Appendix A and $\beta = 1/k_{\mathrm{B}}T$.

The electronic magnetization is obtained from

$$\int_0^{\mathcal{E}_{\mathrm{F}}} \frac{n(\mathcal{E})\mathrm{d}\mathcal{E}}{\exp\left[-\beta(\mathcal{E} - \mu_{\mathrm{B}} h_{\mathrm{e}} - \mu) + 1\right]} \mp \int_0^{\mathcal{E}_{\mathrm{F}}} \frac{n(\mathcal{E})\mathrm{d}\mathcal{E}}{\exp\left[-\beta(\mathcal{E} + \mu_{\mathrm{B}} h_{\mathrm{e}} - \mu) + 1\right]} = \begin{cases} 2zN\langle s^z\rangle & (12\mathrm{a}) \\ 2N\,. & (12\mathrm{b}) \end{cases}$$

In Eq. (12) $n(\mathcal{E})$ is the electronic density of states, μ — the chemical potential, N — the number of states in the band and z — the fraction of occupied states in the band. In what follows we take a rectangular shape for $n(\mathcal{E})$:

$$n(\mathcal{E}) = \begin{cases} N/2\mathcal{E}_0 & \text{if } 0 \le \mathcal{E} \le \mathcal{E}_0 \\ 0 & \text{otherwise.} \end{cases} \tag{13}$$

Equations (4), (11) and (12) define the magnetic state equations. In the next section we derive an explicit equation for the ionic magnetization and magnetic susceptibility.

4. Ionic magnetic state equation

For $n(\mathcal{E})$ given by (13), Eq. (12) can be simplified (see Appendix B). For the range of temperature and band width $\mathcal{E}_0$ of interest, we have:

$$J_0\langle gJ^z\rangle + 2\mu_{\mathrm{B}} h_0 = 4\mathcal{E}_{\mathrm{F}}\langle s^z\rangle , \tag{14}$$

where $\mathcal{E}_{\rm F} = z\mathcal{E}_0$.

Combining (11) with (A2) and (A3), one obtains

$$\langle gJ^z \rangle = \frac{2\alpha_0^2 \mu_{\rm B} h_{\rm i}}{[\Delta^2 + 4\alpha_0^2(\mu_{\rm B} h_{\rm i})^2]^{1/2}} \tanh \left\{ \frac{[\Delta^2 + 4\alpha_0^2(\mu_{\rm B} h_{\rm i})^2]^{1/2}}{2k_{\rm B}T} \right\}. \tag{15}$$

From (15), (14) and (4a) we also derive the reduced inverse ionic magnetic susceptibility:

$$\frac{\mu_{\rm B} h_0}{\langle gJ^z \rangle \mathcal{E}_{\rm F}} = \frac{2\frac{\Delta}{\mathcal{E}_{\rm F}} - \left(\frac{J_0}{\mathcal{E}_{\rm F}}\right)^2 \alpha_0^2 \tanh[\Delta/(2k_{\rm B}T)]}{\alpha_0^2 \left(4 + 2\frac{J_0}{\mathcal{E}_{\rm F}}\right) \tanh[\Delta/(2k_{\rm B}T)]}. \tag{16}$$

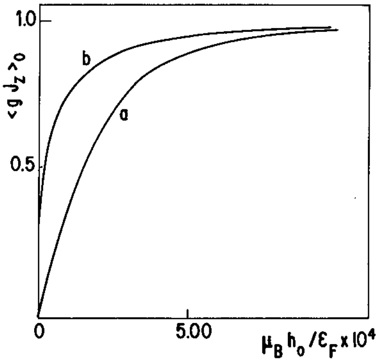
The limit at $T = 0$ K of Eq. (15) is:

$$\langle gJ^z \rangle_0 = \frac{2\alpha_0^2 \mu_{\rm B} h_{\rm i}}{[\Delta^2 + 4\alpha_0^2(\mu_{\rm B} h_{\rm i})^2]^{1/2}}. \tag{17}$$

From (17) one obtains, using (4a) and (14), in the limit $\langle gJ^z \rangle_0 = 0$, the onset condition for spontaneous magnetic order:

$$J_0^c = \frac{(2\Delta\mathcal{E}_{\rm F})^{1/2}}{\alpha_0}. \tag{18}$$

Equation (18) defines the boundary between the ferro- and paramagnetic regions in the space of parameters $J_0/2\mathcal{E}_{\rm F}$ versus $\Delta/2\mathcal{E}_{\rm F}$.



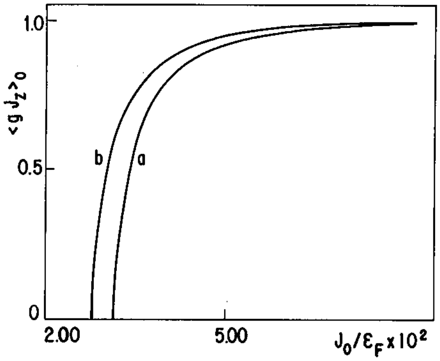


Fig. 2. Magnetic response $\langle gJ^z \rangle$ versus external magnetic field at $T = 0$ K. Curves (a) and (b) are for $J_0/\mathcal{E}_{\rm F}$ equal to 0.0 and 0.03162 respectively. For both curves $\Delta/\mathcal{E}_{\rm F} =$ 0.0005.

Fig. 3. Ionic magnetization (in units of $\mu_{\rm B}$) versus $J_0/\mathcal{E}_{\rm F}$ at $T = 0$ K. Curves (a) and (b) are for $\Delta/\mathcal{E}_{\rm F}$ equal to 0.0005 and 0.0004 respectively.

Equation (18) is the starting point to study the magnetic behaviour in the para- and ferromagnetic regions. Figures 2 and 3 illustrate exchange enhancement

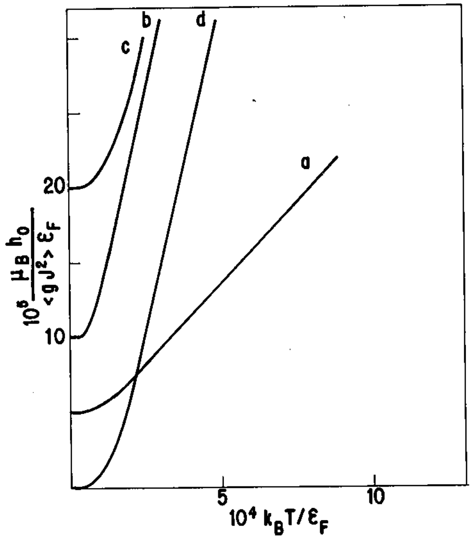


Fig. 4. Reduced inverse magnetic susceptibility $\mu_B h_0/\mathcal{E}_F \langle gJ^z \rangle$ versus $k_B T/\mathcal{E}_F$. For curves (a) and (c) $\Delta/\mathcal{E}_F = 4.0 \times 10^{-4}$, $J_0/\mathcal{E}_F = 0.5 \times 10^{-4}$ and α_0 is respectively 2.0 and 1.0. For curves (b) and (c) $\alpha_0 = 1.0$, $J_0/\mathcal{E}_F = 0.5 \times 10^{-4}$ and $\Delta/\mathcal{E}_F$ is respectively 2×10^{-4} and 4×10^{-4}. For curves (c) and (d) $\alpha_0 = 1.0$, $\Delta/\mathcal{E}_F = 4 \times 10^{-4}$ and $J_0/\mathcal{E}_F$ is respectively 0.5×10^{-4} and 2.8×10^{-2}.

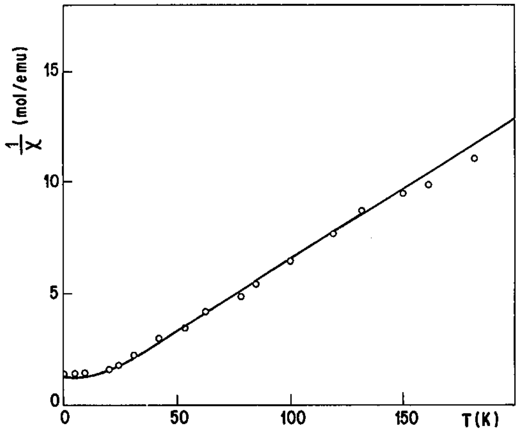


Fig. 5. Inverse of susceptibility $1/\chi$ for $PrAl_3$ versus temperature. The curve was computed from parameters presented in the text, and the experimental points were taken from Ref. [3].

and crystal field effects in the para- and ferromagnetic phases respectively. Figure 4 shows the reduced inverse ionic susceptibility given by Eq. (16) versus $k_BT/\mathcal{E}_F$ for different values of $\Delta/\mathcal{E}_F$, $J_0/\mathcal{E}_F$ and α_0.

5. Application to PrAl$_3$

The magnetic susceptibility of PrAl$_3$ was experimentally studied by Mader et al. [3]. Figure 5 shows the inverse susceptibility versus temperature, obtained using Eq. (16) together with the experimental points of Mader et al. [3]. The fitting is for $\mathcal{E}_F = 8.2$ eV, $\Delta = 3.41$ meV, $J_0 = 6.82$ meV and $\alpha_0 = 1.95$. The value of Δ is that of Mader et al. [3] and the $\mathcal{E}_F$ is taken from Jarlborg et al. [4], who computed the band structure of LaAl$_2$, CeAl$_2$ and YAl$_2$.

Finally, it is interesting to note that in the space parameters $J_0/2\mathcal{E}_0$ versus $\Delta/2\mathcal{E}_0$, the point defined by the above values of J_0, Δ, $\mathcal{E}_F$ and α_0 falls in the paramagnetic phase (see Eq. (18) and its interpretation).

Appendix A. Eigenvalues of ionic Hamiltonian (Eq. (2.a))

In order to compute $\mu_B\langle gJ^z\rangle$, we need $-\partial E_j/\partial h_i$ (see Eq. (11)). The values $E_j (j = 0, 1)$ are calculated from

$$\begin{vmatrix} -E & -\alpha \\ -\alpha & \Delta - E \end{vmatrix} = 0, \tag{A1}$$

$$E_0 = \frac{\Delta}{2} - \frac{\sqrt{\Delta^2 + 4\alpha^2}}{2}, \tag{A2}$$

$$E_1 = \frac{\Delta}{2} + \frac{\sqrt{\Delta^2 + 4\alpha^2}}{2}, \tag{A3}$$

where

$$\alpha = \mu_B h_i \alpha_0.$$

Appendix B. Electronic magnetic state equation for a rectangular energy density of states

Putting (13) into (12) we can solve for $\langle s^z\rangle$, giving

$$2\beta\mu_B h_e = \ln\frac{\sinh\beta\mathcal{E}_0(z - 1/2) - \sinh\beta\mathcal{E}_0(1/2 + 2z\langle s^z\rangle)}{\sinh\beta\mathcal{E}_0(z - 1/2) - \sinh\beta\mathcal{E}_0(1/2 - 2z\langle s^z\rangle)}. \tag{B1}$$

For low temperatures $\beta\mathcal{E}_0 \gg 1$ and z around 0.5 and Eq. (B1) reduces to

$$2\mu_B h_e = 4\mathcal{E}_F \langle s^z \rangle, \tag{B2}$$

where

$$\mathcal{E}_F = z\mathcal{E}_0.$$

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