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The role of the *g*-factor in the magnetism of rare-earth intermetallics

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Abstract

In previous papers, we have proposed reduced-level models in which we retain in the model Hamiltonian only the two lowest levels of the crystal field splitting scheme of a rare-earth ion in rare-earth intermetallics. This approach yields a simple analytical state equation, from which we may determine the value of the g-factor and molecular field parameter (λ). In this work we use a full model to study the spontaneous magnetization vs temperature of $PrAl_2$, using the values of g and λ obtained through a reduced model.

Keywords: Crystal field; Lande' factor; Rare earths; Magnetization; Molecular field parameter

1. Introduction

The study of the magnetism of rare-earth (RE) intermetallics is generally described by crystal field (CF) and magnetic interactions, through model Hamiltonians that include the complete set of levels of the ground multiplet of a RE ion in RE intermetallics (full models) [1,2].

In previous papers [3,4] we have proposed reduced-level models, in which we retained only the two lowest levels of the ground multiplet of the RE ion in a RE intermetallic. This model is a good theoretical framework for the study of magnetism of various RE intermetallics, since the relevant results of this study are obtained with the temperature range in which these results are not affected by the suppression of the highest levels. This model will be applied to the magnetism of $PrAl_2$, and in the case of this compound as well as for a group of other compounds that present a deep similarity with $PrAl_2$ ($PrMg_2$, $PrNi_2$, $PrRh_2$, $PrRu_2$, $PrIr_2$), the procedure mentioned above is correct, since the contribution of the highest excited levels of the splitting scheme for the values of the magnetic quantities may be neglected, as will be shown elsewhere. Generally, for heavy RE intermetallics, it is necessary to include the second or the third excited level, since the difference of energy between two neighbour levels in the CF splitting scheme of the ion is very small.

The reduced models provide analytical magnetic state equations from which we may perform an accurate study for each CF and magnetic parameter of interest related to a given RE intermetallic. However, the main contribution of the reduced model is the possibility of determining a specific value of the coupling-factor g for each RE intermetallic. In contrast with the reduced models, the full ones use the Lande factor g_J obtained from Hund's rules for an isolated ion of RE.

In this work we use a full model to study the spontaneous magnetization vs temperature related to $PrAl_2$ intermetallic, using the values of the g-factor and the molecular field constant λ obtained through a reduced model.

2. Theoretical outline

The Hamiltonian that describes a system of RE ions in a crystal interacting through an exchange interaction is given for both models by

$$H = H_{CF} + H_{mag} \tag{1}$$

where H_{CF} and H_{mag} are the crystal field and magnetic Hamiltonians respectively.

The nine-fold degenerate ground state of the Pr^{3+} ion in a Pr intermetallic in cubic symmetry is decomposed

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into Γ_1 (singlet), Γ_3 (doublet), Γ_4 (triplet) and Γ_5 (triplet) levels [5], with the ground state being Γ_3 [6].

In the full models the eigenfunction and the eigenvalues of energy are obtained from the operator equivalent method using the following Hamiltonian [1]:

$$H = g\mu_B(\vec{J}\cdot\vec{H}) + W[x(O_4/F_4) + (1-|x|)(O_6/F_6)] \tag{2}$$

The nomenclature corresponds to that used by Lea et al. [5], in which W is an energy parameter, x is the parameter that establishes the relative importance of the fourth- and the sixth-order contributions to the crystal field, the O_ms are Stevens' equivalent operators, and F_4 and F_6 are constants.

The total magnetic field, $\boldsymbol{H}$, includes the applied field H_0 and the molecular field H_m, given by:

$$H_m = -\lambda\mu_B^2 g^2 \langle\vec{J}\rangle \tag{3}$$

where λ is a molecular field constant. $\langle\vec{J}\rangle$ is an average value of the total angular momentum of a RE ion, and g is the gyromagnetic factor:

$$\langle\vec{J}\rangle = \frac{\sum\langle n|\vec{J}|n\rangle \exp(-E_n/KT)}{\sum \exp(-E_n/KT)} \tag{4}$$

The spontaneous magnetization, $\vec{M}$, for a single ion is expressed by

$$\vec{M} = gJ\mu_B\langle\vec{J}\rangle \tag{5}$$

For the reduced model the splitting is simplified, i.e. we keep only the two lowest levels of the ground multiplet of the RE ion in the RE intermetallic.

In this work a two-level model will be applied to $PrAl_2$, in cubic symmetry, within the temperature range 0–33 K, in which the ground and the first excited levels are respectively the Γ_3 (doublet) and the Γ_4 (triplet), whose eigenvectors are [5]

$$\Gamma_3 = |e_0\rangle = 0.5400|4\rangle - 0.6450|0\rangle + 0.5400|-4\rangle \tag{6a}$$

$$|e_1\rangle = 0.7071|2\rangle + 0.7071|-2\rangle \tag{6b}$$

$$\Gamma_4 = |e_2\rangle = 0.3536|3\rangle + 0.9351|-1\rangle \tag{6c}$$

$$|e_3\rangle = 0.3536|-3\rangle + 0.9351|1\rangle \tag{6d}$$

$$|e_4\rangle = 0.7071|4\rangle + 0.7071|-4\rangle \tag{6e}$$

Following Frauenheim *et al.* [1], the difference energy between the four levels of the CF splitting scheme of the Pr^{+3} ion in $PrAl_2$ are $\Gamma_3(0)$; Γ_4(2.36 meV); Γ_1(5.67 meV) and Γ_5(13.7 meV).

As our reduced-level model retains only levels Γ_3 and Γ_4, the CF Hamiltonian is described by:

$$H_{CF} = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \Delta & 0 & 0 \\ 0 & 0 & 0 & \Delta & 0 \\ 0 & 0 & 0 & 0 & \Delta \end{bmatrix} \tag{7}$$

where Δ is the difference of the crystalline energy between levels Γ_3 and Γ_4.

As the direction of easy magnetization is $\langle 001\rangle$ [1], we may describe the Hamiltonian in the molecular field approximation by

$$H_{mag} = \begin{bmatrix} 0 & 0 & 0 & 0 & -\alpha \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \gamma & 0 & 0 \\ 0 & 0 & 0 & -\gamma & 0 \\ -\alpha & 0 & 0 & 0 & 0 \end{bmatrix} \tag{8}$$

where $\alpha = \alpha_0(g\mu_B h^z)$ and $\gamma = \gamma_0(g\mu_B h^z)$, with

$$g\mu_B h^z = \lambda\mu_B^2 g^2\langle J^z\rangle + g\mu_B h_0 \tag{9}$$

Here μ_B is the Bohr magneton, h^z is the magnetization vector in the z direction, h_0 is the external magnetic field, J^z is the total angular momentum in the $\langle 001\rangle$ direction, $\alpha_0 = \langle e_0|J^z|e_4\rangle = 3.05$, $\gamma_0 = \langle e_2|J^z|e_2\rangle = -\langle e_3|J^z|e_3\rangle = 0.50$. Putting matrices (7) and (8) into the Hamiltonian (1), we obtain the following eigenvalues of energy:

$$E_0 = \frac{\Delta}{2} - \frac{(\Delta^2+4\alpha^2)^{1/2}}{2} \tag{10a}$$

$$E_1 = 0 \tag{10b}$$

$$E_2 = \Delta + \gamma \tag{10c}$$

$$E_3 = \Delta - \gamma \tag{10d}$$

and

$$E_4 = \frac{\Delta}{2} + \frac{(\Delta^2+4\alpha^2)^{1/2}}{2} \tag{10e}$$

The average magnetization is given by

$$M = \sum_n f_n\left(\frac{-\partial E_n}{\delta h^z}\right) \tag{11}$$

where f_n is the Boltzman factor.

Putting the Eqs. (10) into Eq. (11) we obtain

$$M = \left(\frac{S}{PQ}\right)\sinh\left(\frac{\beta Q}{2}\right) \tag{12}$$

where

$$S = 2\alpha_0^2\mu_B^2 g^2 h^z$$

$$P = \cosh\left(\frac{\beta Q}{2}\right) + \exp\left(\frac{-\beta\Delta}{2}\right)\cosh(\beta\beta_0\mu_B h^z) + \exp\left(\frac{\beta\Delta}{2}\right)$$

$$Q=[\Delta^2+4\alpha_0^2(g\mu_B h^z)]^{1/2}$$

and

$$R=g\mu_B\exp\left(\frac{-\beta\Delta}{2}\right)\sinh(\beta\beta_0 g\mu_B h^z)$$

In the case of spontaneous magnetization we take $h_0=0$ in Eq. (9).

Eq. (12) at $T=0$ K reduces to

$$\frac{M}{\mu_B}=\frac{g\alpha_0(\eta^2-1)}{\eta} \tag{13}$$

where

$$\eta=\frac{2\alpha_0^2\lambda\mu_B^2g^2}{\Delta} \tag{14}$$

The parameter η measures the ratio of the exchange interaction to the CF splitting.

In the limit $\langle J^z\rangle\rightarrow 0$ $(T\rightarrow T_c)$ we obtain from Eq. (12) with $h_0=0$ the following equation for Curie temperature (T_c):

$$\eta=\frac{4\cosh[\Delta/(2KT_c)]+\exp[-\Delta/(2KT_c)]}{2\sinh[\Delta/(2KT_c)]} \tag{15}$$

3. Application to $PrAl_2$

The specific values of g and λ were obtained through Eqs. (13), (14) and (15), in which we used the experimental values of $\Delta=2.36$ meV [1]; $T_c=33$ K and $(M/\mu_B)=2.85$ [2]; plus $\alpha_0=3.05$.

From this calculation, we obtained the values of 0.95 for the g-factor and 148.5 kG/μ for λ. With these values we can calculate the spontaneous magnetization vs temperature of $PrAl_2$ using a full model.

Fig. 1 shows two theoretical curves determined from full models and the experimental points [1] for spontaneous magnetization vs temperature of $PrAl_2$. The dashed curve was obtained by Frauenheim *et al.* [1] using $\lambda=145.0$ kG/μ_B and $g_J=0.8$ for the Pr^{3+} ion. The solid curve was obtained by the authors, taking $\lambda=148.5$ kG/μ_B and $g=0.95$, estimated by the reduced model.

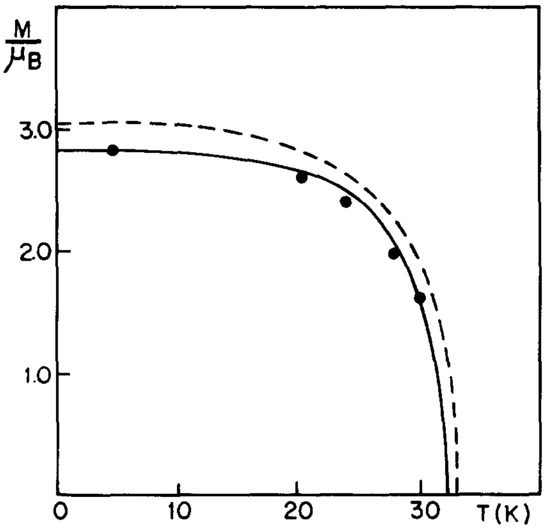

Fig. 1. Temperature dependence of spontaneous magnetization vs. temperature for $PrAl_2$. The experimental points (full circles) were determined by Purwins et al. [2]. The dashed curve was constructed by Frauenheim et al. [1], with $g_J=0.8$ and $\lambda=145.0$ kG/μ_B, and the solid curve was constructed by the authors with $g=0.95$ and $\lambda=148.5$ kG/μ_B.

4. Conclusions

The correctness of the g-factor obtained from a two-level model depends on how this parameter is affected by the levels of the RE ion neglected in the model. As the g-factor is determined from the results of spontaneous magnetization versus temperature, we must verify the dependence of this magnetic quantity in relation to the second excited level Γ_1 not included in the model. The energy of this level in the CF splitting scheme of the Pr^{+9} ion in $PrAl_2$ measured from the ground level is $\Delta'=5.67$ meV [1]. The level Γ_1 is non-magnetic and from the level Γ_1 there is only one transition allowed to Γ_4 represented by the non-diagonal matrix element $\delta_0=\langle e_4|J^z|e_1\rangle$, where the expressions for the eigenvectors are found in [7]. Including in this model the level Γ_1, we have two 6×6 matrices in the model Hamiltonian (1), and the energy eigenvalues determined numerically at 30 K are: $E_0=0$; $E_1=4.47$ meV; $E_2=6.04$ meV; $E_9=7.53$ meV; $E_4=7.63$ meV; $E_5=13.8$ meV. These values for energy were obtained taking $\beta_0=0.5$, $\alpha_0=3.05$, $\delta_0=2.56$, $\Delta=2.36$ meV, $\Delta'=5.67$ meV, and $J_0=0.77$ meV, and the experimental point of spontaneous magnetization versus temperature $M/\mu_B=1.6$ at 30 K determined by Purwins et al. [2] for $PrAl_2$. Putting the energy eigenvalues mentioned above into the Boltzmann factors, we achieve the contribution of the energy state E_5 associated with Γ_1 for the spontaneous magnetization at 30 K. The resultant value is about 4%. This value shows clearly that in this case the second excited level Γ_1 may be neglected.

Generally the g-factor used in the literature for determining the values of the magnetic quantities related to RE intermetallics is calculated from Hund's rules for a single ion of RE, and does not take into account the effects of the crystalline environment in which the ion is located: as for example the quenching effect due to the crystal field on the orbital contribution to the moment. On the contrary, as the g-factor used in the reduced models is determined from experimental data it includes the effects due to the crystalline environment and so it may be calculated particularly for each RE intermetallic. From a reduced model, it is also possible to determine analytically a λ value for a RE intermetallic, as we have for $PrAl_2$ in this work. Finally, using the values of g-factor and λ obtained through our reduced model in a full model, we can obtain a fit with the experimental points better than that achieved by Frauenheim et al. [1] using $g_J = 0.8$ and $\lambda = 145.0$ kG/μ_B, as is shown in Fig. 1.

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