A SIMPLE MODEL FOR LOCALIZED-ITINERANT MAGNETIC
SYSTEMS: CRYSTAL FIELD EFFECTS

by

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ABSTRACT

The magnetic behavior of a system consisting of localized electrons coupled to conduction electrons and submitted to an axial crystal field at T=0 K is investigated within the framework of the molecular field approximation. An analytical ionic magnetic state equation is deduced; it shows how the magnetization depends on the model parameters (exchange, crystal field, band occupation) and external magnetic field. A condition for the onset of spontaneous magnetic order is obtained and the ferro- and paramagnetic phases are studied. This study displays several features of real magnetic systems, including quenching or total suppression of the magnetic moments (depending on the relative value of the crystal field parameter) and exchange enhancement. The relevance of such model for the description of rare-earth intermetallic compounds is discussed.

Key-words: Crystal-field; Exchange interaction; Rare-earth intermetallics.

1. Introduction

In this paper we apply a simple model to a system consisting of conduction electrons interacting with localized electrons, these being under the action of a crystal field. We derive a magnetic state equation that contains the parameters that characterize the crystal field, the exchange interaction and the features of the conduction electron band. The interest of such study lies in the applicability to the description of magnetic properties of rare earth intermetallic compounds, particularly compounds containing light rare-earths, for which crystal-field effects are known to be more significant [1]. The same approach was used in an earlier study in the absence of crystal-field effects [2]. In this paper we concentrate on light rare earth intermetallics containing non-transition metals. In the compounds where transition elements are present, associated to strong anisotropies, as in the hard magnets, the exchange interactions between d electrons are important for the magnetic properties. The paper has the following structure: in Section 2, the model Hamiltonian and the quantities of interest are presented. In Section 3, within the molecular field approximation, magnetic state equations relating ionic and electronic magnetizations to the model parameters, applied magnetic field and temperature are derived. In Section 4, an ionic magnetic state equation at T = 0 K is obtained and it is shown that ferro- or paramagnetism occur according to the range of the model parameters. A detailed study is made for both phases. Finally, in Section 5, the main conclusions are summarized and the possible applications mentioned.

2. Model Hamiltonian and Magnetic Quantities

The model Hamiltonian is the following:

$$H = H_{cf} + H_{el} + H_{exch} + H_{mag} \qquad (1)$$

where H_{cf} is the crystal field Hamiltonian. In this work, to simplify matters, we assume an axially symmetric, one-parameter H_{cf}:

$$H_{cf} = D \, J_z^2 \qquad (2)$$

where D is the crystal field splitting. For $J = 1$, one has a singlet doublet level structure, for $D > 0$.

The second term H_{el} describes the dynamics of the conduction electrons

$$H_{el} = \sum_{k\sigma} \varepsilon_k \, C_{k\sigma}^+ \, C_{k\sigma} \qquad (3)$$

where ε_k is the electron energy spectrum, from which one derives the energy density of states. The latter is of interest for the description of band magnetism; in this work the density of states will be assumed to be rectangular in shape. $C_{k\sigma}^+$ ($C_{k\sigma}$) is the usual creation (annihilation) operator in the Bloch representation.

The third term in (1) is

$$H_{exch} = - 2 \, J_o \sum_i \vec{S}_i \cdot \vec{s}_i \qquad (4)$$

where J_o is the effective exchange integral that accounts for the interaction between the localized electrons (with total spin S) and the conduction electrons with spin s. The operator $\vec{s}_i$ is defined by its components

$$s_i^z = 1/2 \, (C_{i\uparrow}^+ C_{i\uparrow} - C_{i\downarrow}^+ C_{i\downarrow}) \qquad (5a)$$

$$s_i^+ = C_{i\uparrow}^+ C_{i\downarrow} \qquad (5b)$$

$$s_i^- = c_{i\downarrow}^+ c_{i\uparrow} \tag{5c}$$

where $C_{i\sigma} = (1/\sqrt{N}) \sum_k \exp(-i\vec{k}.\vec{R}_i) C_{k\sigma}$. The summation on $\vec{k}$ extends to the Brillouin zone, N is the number of electron states and $\vec{R}_i$ are the position vectors of the crystal lattice.

Finally, the last term in (1) contains the interaction of the localized electrons and conduction electrons with an applied magnetic field h_o:

$$H_{mag} = -g\, \mu_B \vec{h}_o . \sum_i \vec{J}_i - 2\, \mu_B \vec{h}_o . \sum_i \vec{s}_i \tag{6}$$

where g is the Landé factor of the localized spin, $\vec{J}$ is the total localized angular momentum and μ_B is the Bohr magneton. We will make use of the de Gennes relation $S = (g-1)\, J$.

The quantities of interest are the statistical averages $\langle \vec{J} \rangle$ and $\langle \vec{s} \rangle$, which are related to the ionic and electronic magnetizations. These quantities are functions of the temperature and applied magnetic field.

3. Magnetic State Equations

In what follows, we make use of the molecular field approximation:

$$H_{exch} \cong -J_o(g-1)\langle \vec{J} \rangle . \sum_i \vec{s}_i - J_o(g-1)\langle \vec{s} \rangle . \sum_i \vec{J}_i \tag{7}$$

By means of (7), we get from (1) two effective Hamiltonians

$$\hat{H}_{el} = H_{el} - 2\, \mu_B\, \vec{h}_e . \sum_i \vec{s}_i \tag{8a}$$

$$\hat{H}_{ion} = H_{cf} - g\, \mu_B \vec{h}_i . \sum_i \vec{J}_i \tag{8b}$$

where

$$2 \mu_B \hat{h}_\bullet = 2 \mu_B \hat{h}_o + J_o(g-1)\langle \hat{J} \rangle \qquad (9a)$$

$$g \mu_B \hat{h}_\iota = g\mu_B \hat{h}_o + J_o(g-1)\langle \hat{s} \rangle \qquad (9b)$$

Assuming that the easy direction of magnetization occurs along the x axis, the eigenvalues of $\hat{H}_{ion}$ are, for $J = 1$ [3]

$$E_o = [D - (D^2 + 4 \alpha^2)^{1/2}]/2 \qquad (10a)$$

$$E_\iota = D \qquad (10b)$$

$$E_2 = [D + (D^2 + 4 \alpha^2)^{1/2}]/2 \qquad (10c)$$

where

$$\alpha = g\mu_B h_\iota \qquad (11)$$

The magnetic moments of these levels are given by $m_j = -dE_j/dh_\iota$ $(j = 0, 1, 2)$

$$m_o = 2g\mu_B \alpha/(D^2 + 4 \alpha^2)^{1/2} \qquad (12a)$$

$$m_\iota = 0 \qquad (12b)$$

$$m_2 = - 2g\mu_B \alpha/(D^2 + 4 \alpha^2)^{1/2} \qquad (12c)$$

The ionic magnetization $g\mu_B\langle J_x \rangle$ is then obtained as

$$g\mu_B\langle J_x \rangle = \frac{\Sigma_\iota m_\iota \exp(-\beta E_\iota)}{\Sigma_\iota \exp(-\beta E_\iota)} \qquad (13)$$

The electronic magnetization $2\mu_B\langle s_x \rangle$ is derived from

$$f(\mu+\mu_B h_\bullet) - f(\mu-\mu_B h_\bullet) = 2\gamma N \langle s_x \rangle \qquad (14a)$$

$$f(\mu+\mu_B h_\bullet) + f(\mu-\mu_B h_\bullet) = \gamma N \qquad (14b)$$

where

$$f(x) = \int_{-\varepsilon_o}^{\varepsilon_o} \frac{n(\varepsilon)d\varepsilon}{\exp[\beta(\varepsilon-x)] + 1} \qquad (15)$$

In (13), (14) and (15), $\beta = (k_B T)^{-1}$; $n(\varepsilon)$ is the energy density of states, μ is the chemical potential and γN is the number of electrons in the conduction band. Equations 13 and 14, coupled via (9) give rise to the magnetic state equations; they allow us to analyze the magnetic behavior of the localized-itinerant electrons system described by the present model.

4. Ionic Magnetic State Equation at 0 K

Using a rectangular density of states $n(\varepsilon)$, of width $2\varepsilon_o$:

$$n(\varepsilon) = \begin{cases} N/4\varepsilon_o & \text{if } -\varepsilon_o < \varepsilon < \varepsilon_o \\ \\ 0 & \text{otherwise} \end{cases} \qquad (16)$$

equations (14) can be solved analytically for any temperature and the result is given in Appendix A.

At $T = 0$ K, the ionic magnetization is given by (12a). Combining expressions (12a), (9a), (9b) and (A2), one obtains:

$$2\frac{\mu_B h_o}{2\varepsilon_o} \left[\frac{(g-1)J_o}{2\varepsilon_o} \frac{1}{2\gamma} + g \right] = $$

$$\left\{ \frac{D}{2\varepsilon_o} \frac{1}{(1-\langle J_x \rangle^2)^{1/2}} - \left[\frac{(g-1)J_o)}{2\varepsilon_o} \right]^2 \frac{1}{2\gamma} \right\} \langle J_x \rangle \qquad (17)$$

Equation 17 is the ionic magnetic state equation at $T = 0$ K. It allows a ferro- or paramagnetic solution, according to the range

of the model parameters.

With $h_o = 0$ one can obtain a ferromagnetic solution in which case $\langle J_x \rangle \neq 0$; this last condition implies:

$$\left\{ \frac{D}{2\varepsilon_o} \frac{1}{(1-\langle J_x \rangle^2)^{1/2}} - \left[\frac{(g-1)J_o)}{2\varepsilon_o} \right]^2 \frac{1}{2\gamma} \right\} = 0 \qquad (18)$$

The condition for the onset of magnetic order, derived from (18) is given by

$$D/2\varepsilon_o = \frac{1}{2\gamma} \left[\frac{(g-1)J_o}{2\varepsilon_o} \right]^2 \qquad (19)$$

Relation (19) is the starting point to study the magnetic behavior of the model. This equation may be interpreted as a phase diagram in the parameter space $(D/2\varepsilon_o, (g-1)J_o/2\varepsilon_o)$. A generalization of Eq. 19 for an arbitrary density of states is presented in Appendix B. Above the curve (19) we have ferromagnetism, below it paramagnetism. From (18) we may compute $\langle J_x \rangle$ versus $(g-1)J_o/2\varepsilon_o$ for given values of $D/2\varepsilon_o$. Figure 1 illustrates the role of exchange and crystal field parameters in determining the spontaneous ionic saturation magnetization. In the paramagnetic region of the phase diagram we may compute the induced magnetization $\langle J_x \rangle$ as a function of the external magnetic field. Figure 2 shows the results of such computation for three different values of the exchange parameter.

5. Conclusions

The extreme simplicity of the model and the approximations employed (e.g. rectangular band shape, axial crystal field, etc) allow us to obtain an analytical magnetic state equation at $T = 0$ K (Eq. 17) which depends explicitly on the exchange, crystal field and band parameters that determine the magnetic properties of the system. A condition for magnetic order derived from Eq. 19 also involves these parameters.

In deriving the magnetic state equations from the model

presented here we had in mind the description of the magnetic properties of rare-earth intermetallic compounds. Of course the simplicity of the model and the approximations used do not allow a detailed numerical comparison with the data of real systems. However, some qualitative remarks can be made: 1)$PrAl_2$ and $PrNi_2$ are, respectively, ferro- and paramagnetic [4,5]; according to Eq. (B2), this difference may be due to differences in the band structure associated to Al and Ni; 2) $PrNi_2$ and $NdNi_2$ are both paramagnetic [5]; magnetization versus applied magnetic field at low temperature (Figs. 1 and 8, of reference 5) show that exchange enhancement effects are more pronounced in $NdNi_2$, suggesting that crystal field quenching is more effective in the case of $PrNi_2$. One should note that although Ni is a 3d transition element, in the above mentioned compounds it has no magnetic moment [6], and behaves as a normal metal.

Another system of interest are the amorphous rare earth alloys. In these systems the quadratic contribution to the crystal field (involving the O_2^0 and O_2^2 Stevens operators) exceeds by one order of magnitude the effect of higher order terms [7]. In this way, simplified crystal field Hamiltonians may be appropriate in discussing models for amorphous systems.

The main conclusions that can be drawn from this work are:

1. there is a quenching of the ionic magnetic moment at $T = 0$ K that depends on the crystal field parameter D. For higher values of D the magnetic order is suppressed.

2. in the paramagnetic region, an external magnetic field induces an ionic magnetization that shows exchange enhancement effects.

3. for reasonable values of the band parameters the contribution of the band magnetization to the total magnetization is very small. However, the conduction electrons play a significant role in triggering the magnetic order of the ionic moments.

Appendix A Electronic Magnetic State Equation for a Rectangular Density of States

Using (16) in (14), after some algebraic manipulations, one obtains

$$2\beta\mu_B h_e = \ln \left[\left(\frac{\exp(2a\gamma) - \exp(-4a\gamma x)}{\exp(2a\gamma) - \exp(4a\gamma x)} \right) \left(\frac{\exp[-2a(1-\gamma)] - \exp(4a\gamma x)}{\exp[-2a(1-\gamma)] - \exp(-4a\gamma x)} \right) \right]$$

(A1)

where $x = \langle s_x \rangle$ and $a = \beta\varepsilon_0$.

In the limit $T = 0$ K, (A1) becomes

$$2\mu_B h_e = 8\varepsilon_0 \gamma x$$

(A2)

Appendix B Condition of Spontaneous Magnetic Order for an Arbitrary Density of States

Starting from Equations (14), in the limit $T = 0$ K and $\langle J_x \rangle = 0$, we obtain

$$n(\mu) = \lim_{h_e \to 0} \frac{2\gamma N \langle s_x \rangle}{2\mu_B h_e}$$

(B1)

Using relations (9) and (12a) one finally obtains

$$D/2\varepsilon_0 = \frac{2\varepsilon_0 n(\mu)}{\gamma N} \left[\frac{(g-1)J_0}{2\varepsilon_0} \right]^2$$
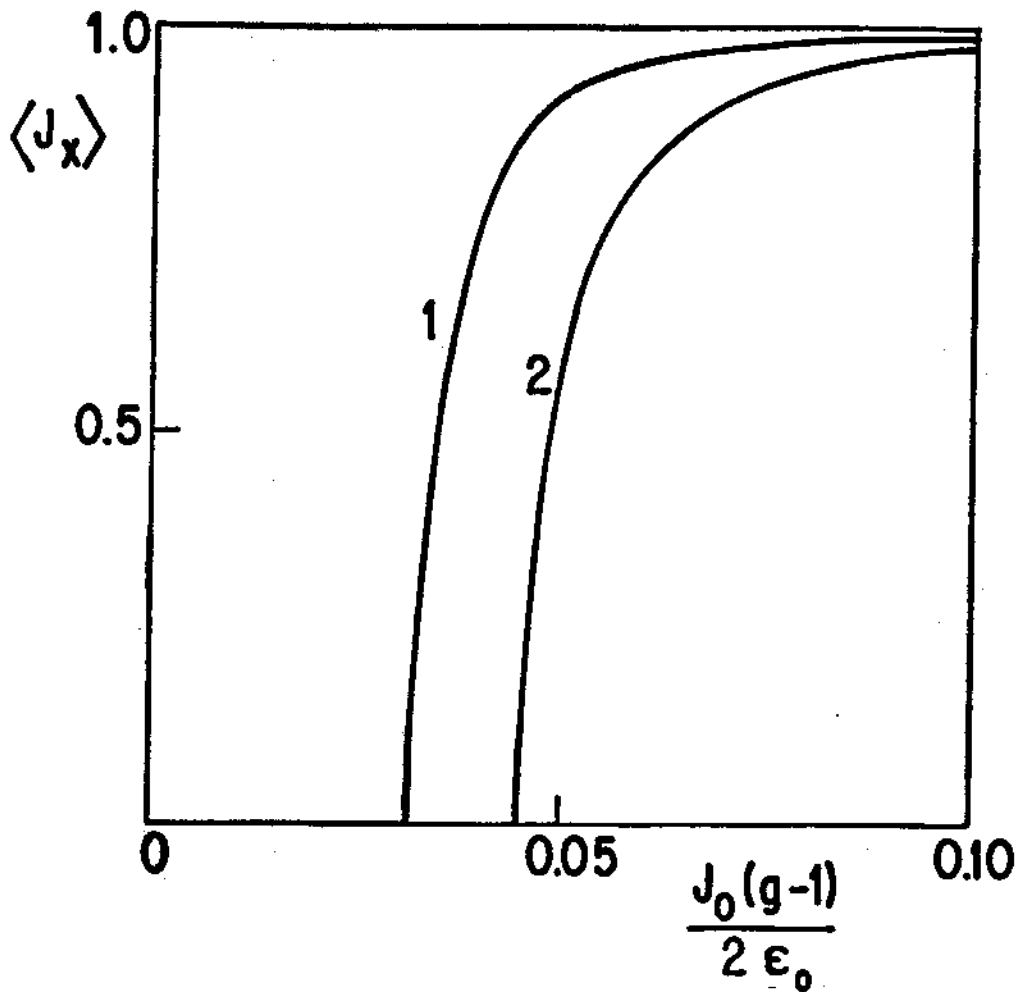
(B2)

Acknowledgements

Fig. 1 Ionic magnetization (in units of $g\mu_B$) versus $J_o(g-1)/2\varepsilon_o$. We have taken $\gamma = 0.5$, $g = 2$ and $D/2\varepsilon_o$ is equal to 10^{-3} and 2×10^{-3} for curves 1 and 2, respectively. Note that the crystal field quenching is more pronounced for larger values of $D/2\varepsilon_o$ (curve 2).
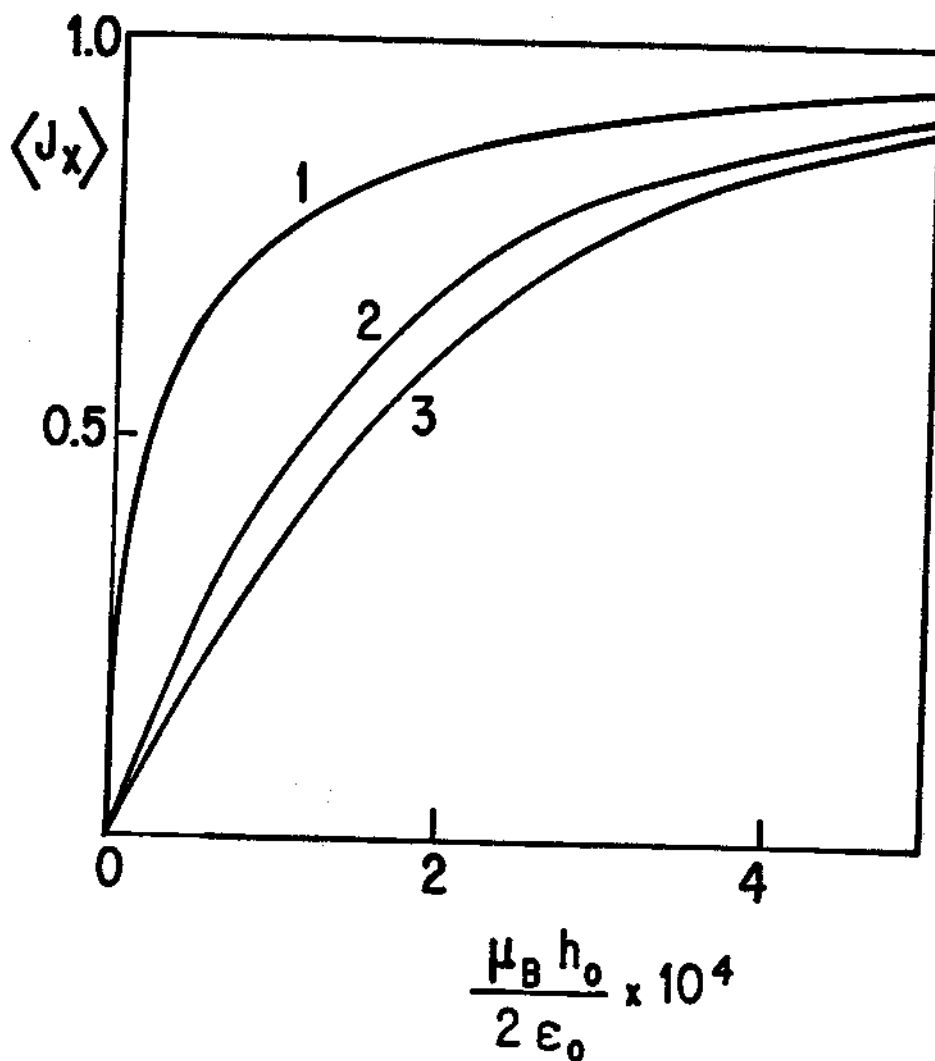
Fig. 2 Ionic magnetization (in units of $g\mu_B$) versus applied magnetic field. Again $g = 2$, $\gamma = 0.5$ and $D/2\varepsilon_0 = 1.0 \times 10^{-8}$. Curves 1,2 and 3 are for $J_0(g-1)/2\varepsilon_0$ equal to 0.0316, 0.0158 and zero, respectively. Curves 1 and 2 show exchange enhancement.

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