ISSN 0029 - 3865

CBPF-NF-014/85

MAGNETIC ORDER OF SINGLET-SINGLET IONS COUPLED TO CONDUCTION ELECTRONS: BAND STRUCTURE EFFECTS

by

L. Palermo* and X.A. da Silva

Centro Brasileiro de Pesquisas Físicas - CBPF/CNPq
Rua Dr. Xavier Sigaud, 150
22290 - Rio de Janeiro, RJ - Brasil

*Instituto de Física da UFF
Outeiro de São João Batista, s/nº - Centro
24210 - Niteroi, RJ - Brasil

Abstract

The onset of magnetic order of a system consisting of conduction electrons interacting with localized f-electrons having only two singlet levels is analysed. The effects of band structure, crystal field and exchange parameters are deduced from a parametric study of the model. We suggest that the combined role of these effects may be of importance in explaining drastic differences in the magnetic properties of light rare-earth intermetallics.

Key-words: Singlet-singlet ions; Crystal field; Conduction electrons; Magnetic order; Hight rare-earth intermetallics.

1 Introduction

Previously (Palermo and Silva 1980) we have studied various magnetic quantities associated to a system consisting of light rare-earth ions, coexisting with conduction electrons. Under the action of a crystal field, the f-ground state level is splitted, for simplicity, into two singlet levels, but with off-diagonal of the total angular momentum non zero; the conduction electrons interacting by exchange with the local f-electrons. Despite the simplifications in the description of the band structure and the local symmetry of the crystal field, we have applied (Palermo and Silva 1981) the results of a parametric study of the model to the available experimental data of $PrAl_2$ and also, with the inclusion of intra-band electron interaction, to $PrFe_2$ (Palermo and Silva 1984). A similar study, in the absence of crystal field effects, was also performed in connection with the heavy rare-earth intermetallics (Iannarella et al 1982).

In this paper, in connection with the above mentioned model, we want to explore band effects. These are simulated by adopting several energy density of states shapes, which can be filled with different number of electrons per band. At $T = 0K$, we calculate the ionic and electronic magnetizations versus the exchange parameter. Our parametric study shows that the combined role of exchange interaction, crystal field and the band structure of the conduction electrons is responsible for the existence or not of magnetic order. The ionic magnetization saturates quickly as a function of the exchange

parameter, except in the case of higher values of crystal field parameter. We suggest that differences in the magnetic properties of $PrAl_2$ ($T_c = 34K$) and $PrNi_2$ ($T_c = 0.27K$) may be due to differences in crystal field, exchange and most probably in band structure details.

2 Ionic and Electronic Magnetizations at T = 0K

The model Hamiltonian is

$$H = H_{e\ell} + H_{ion} + H_{exch} \tag{1}$$

where $H_{e\ell}$ is the conduction electron Hamiltonian, H_{ion} is the pseudo spin Hamiltonian for the singlet-singlet ions, given in (Palermo and Silva 1980), H_{exch} is the local ion-conduction electron exchange.

From (1) one can compute, in the molecular field approximation, the ionic and electronic magnetization. At T = 0K the electronic magnetization $\langle S^z \rangle$ is given by

$$\int_{-\infty}^{\mu+\mu_B h_{e\ell}} n(\varepsilon)d\varepsilon - \int_{-\infty}^{\mu-\mu_B h_{e\ell}} n(\varepsilon)d\varepsilon = 2zN\langle S^z \rangle \tag{2-a}$$

$$\int_{-\infty}^{\mu+\mu_B h_{e\ell}} n(\varepsilon)d\varepsilon + \int_{-\infty}^{\mu-\mu_B h_{e\ell}} n(\varepsilon)d\varepsilon = zN \tag{2-b}$$

where μ is the chemical potential, $n(\varepsilon)$ is the density of

states of the conduction electrons, z is the number of electrons per band and N is the number of atoms in the crystal. In the molecular field approximation,

$$\mu_B h_{e\ell} = \frac{J}{2} < s^z > \tag{3}$$

the ionic magnetization, at T = 0K, is

$$2< s^z > = \frac{4g\alpha\mu_B h_{ion}}{\left[(4g\alpha\mu_B h_{ion})^2 + \Delta^2\right]^{1/2}} \tag{4}$$

where Δ is the energy difference between the two singlet levels: In the molecular field approximation

$$2g\alpha\mu_B h_{ion} = \frac{J}{2} < s^z > \tag{5}$$

In order to determine $< s^z >$ and $< s^z >$ from equations 2 and 4, one has to give the density of states $n(\varepsilon)$. We will work with different model shapes of density of states:

2.1 Rectangular band

$$n(\varepsilon) = \begin{cases} \dfrac{N}{2\varepsilon_o} & \text{if} \quad -\dfrac{\varepsilon_o}{2} \leq \varepsilon \leq \dfrac{\varepsilon_o}{2} \\ 0 & \text{Otherwise} \end{cases} \tag{6}$$

For this case, eqs. 2 and 4, give

$$\left(\frac{J}{\varepsilon_o}\right)^2 = 4z\left[\left(\frac{J}{\varepsilon_o}\right)^2 \xi^2 + 4\left(\frac{\Delta}{\varepsilon_o}\right)^2\right]^{1/2} \tag{7}$$

where $\xi = 2< s^z >$

Curve d of Figures 1 and 2 give the plot of ξ versus J/ε_o for $\Delta/\varepsilon_o = 1 \times 10^{-3}$ and $z = \frac{1}{2}$. The corresponding $2\langle S^z \rangle$ versus J/ε_o is given, using eq. 4, in the curve d of Figure 3.

2.2 Triangular band

Now,

$$n(\varepsilon) = \begin{cases} \frac{2N}{\varepsilon_o}(\varepsilon_o + \varepsilon) & \text{if} \quad -\frac{\varepsilon_o}{2} \leq \varepsilon \leq 0 \\ \\ \frac{2N}{\varepsilon_o}(\varepsilon_o - \varepsilon) & \text{if} \quad 0 \leq \varepsilon \leq \frac{\varepsilon_o}{2} \end{cases} \tag{8}$$

Equations 2 and 4, give

$$\begin{cases} \frac{z\xi}{2} = \frac{\mu_B h_{e\ell}}{\varepsilon_o} - \left(\frac{\mu}{\varepsilon_o}\right)^2 - \left(\frac{\mu_B h_{e\ell}}{\varepsilon_o}\right)^2 & \text{(8-a)} \\ \\ \frac{\mu}{\varepsilon_o} = \frac{2z-1}{4} \cdot \frac{1}{1 - \frac{2\mu_B h_{e\ell}}{\varepsilon_o}} & \text{(8-b)} \end{cases}$$

using equations 8, 3, 4 and 5, one obtains ξ and $2\langle S^z \rangle$ versus J/ε_o for a given Δ/ε_o and z. Curve a, in Figures 1,2 and 3 gives the plot of electronic and ionic magnetizations versus J/ε_o, for $\Delta/\varepsilon_o = 1 \times 10^{-3}$ and $z = \frac{1}{2}$.

2.3 Parabolic band

In this case

$$n(\varepsilon) = \frac{3N}{4\varepsilon_0^{2/3}}\,\varepsilon^{1/2} \tag{9}$$

Equations 2 and 4, now give

$$\left(\frac{2z+\xi}{2}\right)^{2/3} - \left(\frac{2z-\xi}{2}\right)^{2/3} = \frac{2\mu_B h_{e\ell}}{\varepsilon_0} \tag{10}$$

using eq. 3, 4 and 5, one obtains

$$\left(\frac{2z+\xi}{2}\right)^{1/2} = \left(\frac{2z-\xi}{2}\right)^{3/2} - \frac{\left(\frac{J}{\varepsilon_0}\right)^2 \xi}{2\left[\left(\frac{J}{\varepsilon_0}\right)^2 \xi^2 + 4\left(\frac{\Delta}{\varepsilon_0}\right)^2\right]^{1/2}} \tag{11}$$

curves c and f in Fig. 1, and curve c in Fig. 2 give, for $z = \frac{1}{2}$ and $\Delta/\varepsilon_0 = 1 \times 10^{-3}$ and $\Delta/\varepsilon_0 = 0.2$ respectively the plot of ξ versus J/ε_0. In figure 3, curves c and f, gives $2\langle S^z \rangle$ versus J/ε_0.

2.4 Linear band

Now

$$n(\varepsilon) = \frac{N}{\varepsilon_0}\,\varepsilon \tag{12}$$

From equations 2-a and 2-b, one obtains

$$z^{1/2}\left[(1+\xi)^{1/2} - (1-\xi)^{1/2}\right] = \frac{2\mu_B h_{e\ell}}{\varepsilon_0} \tag{13}$$

Using equations 3,4 and 5 one has

$$\bar{z}^{1/2}\left[(1+\xi)^{1/2}-(1-\xi)^{1/2}\right]=\left(\frac{J}{\varepsilon_0}\right)^2\cdot\frac{\xi}{2\left[\left(\frac{J}{\varepsilon_0}\right)^2\xi^2+4\left(\frac{\Delta}{\varepsilon_0}\right)^2\right]^{1/2}} \qquad (14)$$

curves b and e in figures 1 and 2, show for $z=\frac{1}{4}$ and $z=\frac{1}{2}$ respectively the plot of ξ versus J/ε_0, for $\Delta/\varepsilon_0 = 1\times10^{-3}$. Curves b and e in figure 3 show the corresponding plots for $2\langle S^z\rangle$ versus J/ε_0.

3 Final comments and possible applications

In this section we summarize the results obtained, putting emphasis on the role of the model parameters in determinning its magnetic behaviour.

Figures 1,2 and 3 show, within the range of the employed parameters (which are physically meaningful), that the significant contribution to the total magnetization comes essentially from the ionic counterpart; the conduction electrons, however, playing a fundamental role in the magnetic ordering process.

The onset of magnetic order is a function of the shape of the electronic density of states, number of electrons per band and the ratio between the exchange and crystal field parameters:

1 - for a given density of states and a fixed number of electrons per band, the onset of magnetic order is more difficult (one needs a bigger J/ε_0) the greater the crystal field parameter. This is illustrated in Fig. 1 by curves c and f;

2 - for a given shape of density of states and same crystal field parameter, the onset of magnetic order is easier for smaller number of electrons per band. This is shown in Figure 2 by curves b and e;

3 - for a given crystal field parameter and same number of electrons per band, the onset of magnetic order is easier for the triangular density of states, and increases in the following order of shapes of density of states: parabolic, retangular and linear. This can be seen in the curves a, c, d and e of Figure 2;

4 - once one reaches the magnetic order, given in terms of a minimum exchange parameter, the ionic magnetization quickly saturates for a small increase of J/ε_0. This is shown in Figure 3.

The magnetic behaviour deduced from the proposed model suggests that in evaluating the magnetic properties of light rare-earth intermetallics, crystal field effects should be complemented by band structure details. It is interesting to point out the drastic differences in the magnetic properties of $PrAl_2$ (T_c = 34K) and $PrNi_2$ (T_c = 0.27K), printed in Table II of reference 5. Differences in the electronic structure associated to Al and Ni give rise to different shapes of density of states and effective number of electrons per band; according to our study the onset of magnetic order is very sensitive to such changes. In the case of $PrCO_2$ and $PrFe_2$, also listed in reference 5, both ferromagnetic, there should also be a significant contribution of the band magnetism.

Figure Captions

Fig. 1 - Electronic magnetization versus exchange parameter for different crystal field parameter and energy density of states: triangular a, linear b and e, parabolic c and f, rectangular d. For curves a, c, d, e and f, $z = \frac{1}{2}$; for curve b, $z = \frac{1}{4}$. For curves a, b, c, d and e $\Delta = 1{,}0 \times 10^{-3}$; for curve f, $\Delta = 0.2$.

Fig. 2 - Same as Fig. 1, but for much smaller values of exchange parameter.

Fig. 3 - Ionic magnetization at T = 0K versus exchange parameter. The shapes of density of state and crystal field parameters correspond to the curves given in Fig. 1.

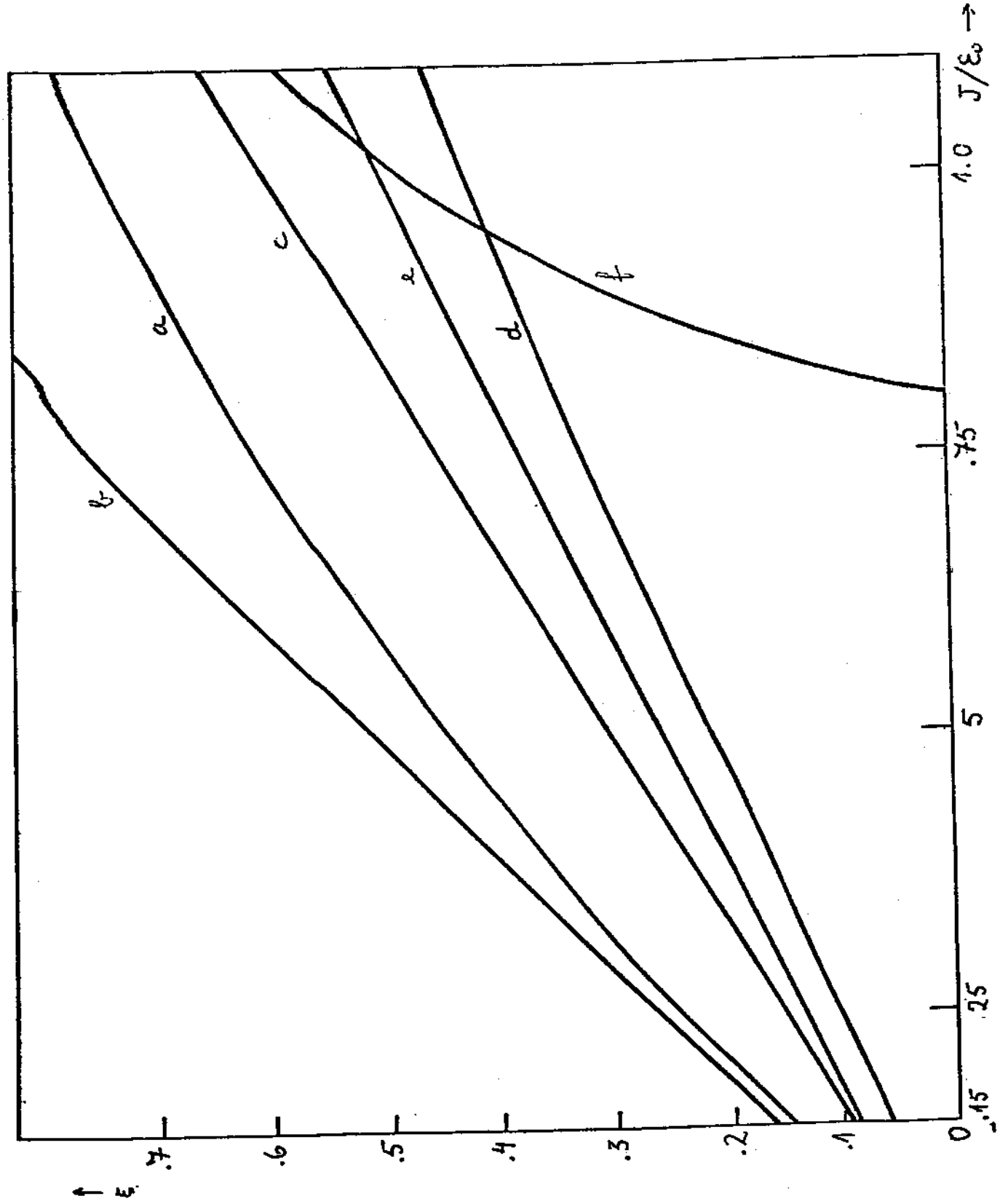

FIG. 1

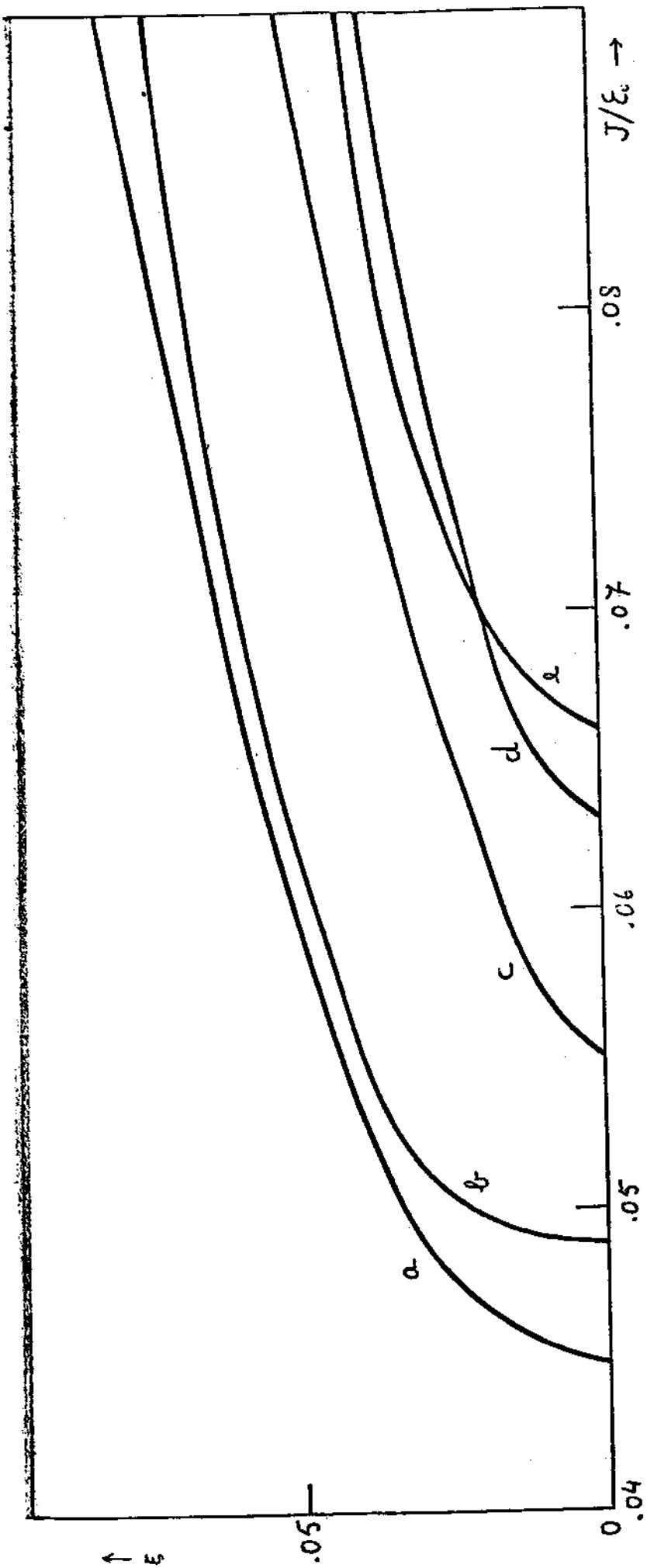

FIG. 2

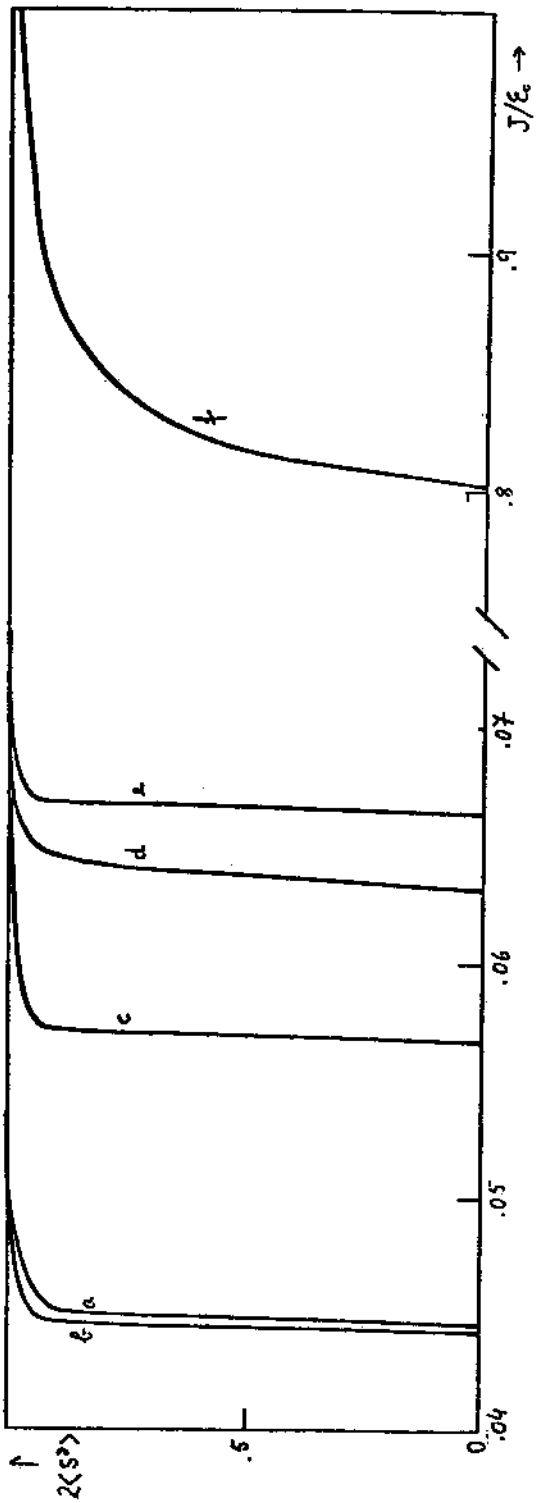

FIG. 3

References

L. Palermo and X.A. da Silva, Phys. Stat. Sol. (b), 102, 661. (1980)

L. Palermo and X.A. da Silva, Phys. Stat. Sol. (b), 103, 419 (1981)

L. Palermo and X.A. da Silva, Journal of Mag. and Mag. Materials, 43, 308 (1984).

L. Iannarella; A.P. Guimarães and X.A. da Silva, Phys. Stat. Sol. (b) 114 (1982) 255

F.J.A.M. Greidanus; L.J. de Jongl; W.J. Huiskamp; A. Furrer and K.H.J. Buschow, p. 13, in Crystalline Electric Field Effects in f-Electron Magnetism, Edited by R.P. Guertin, W. Suski and Z. Zolnierek-Plenum Press, N.Y., 1982.