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

by

P.J. von RANKE[1], L. PALERMO[2] and X.A. da SILVA

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

[1]Instituto de Física - UERJ
Rua São Francisxo Xavier, 524
20550 - Rio de Janeiro, RJ - Brasil

[2]Instituto de Física - UFF
Rua São João Batista s/n - Centro
24020 - Niteroi, RJ - Brasil

ABSTRACT

We present a simple magnetic model for $PrA\ell_3$. 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 $PrA\ell_3$ given in the literature.

Key-words: Exchange; Crystal field; Magnetism; $PrA\ell_3$.

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 4f-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 $PrA\ell_3$ 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 4f-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 $PrA\ell_3$ is discussed and the parameters of the model are estimated using the experimental temperature dependence of the susceptibility of $PrA\ell_3$ [3].

2. Model Hamiltonian and magnetic quantities

In the molecular field approximation the model Hamiltonian is

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

where

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

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

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.

$$H^{i}_{exch} = - \mu_B h_i \sum_j g J^z_j \tag{3.a}$$

$$H^{e}_{exch} = - 2\mu_B h_e \sum_j s^z_j \tag{3.b}$$

where

$$\mu_B h_i = \mu_B h_0 + J_o \langle s^z \rangle \tag{4.a}$$

$$2\mu_B h_e = 2\mu_B h_0 + J_0 \langle J^z \rangle \tag{4.b}$$

In 3 and 4 μ_B is the Bohr magneton, h_o is an applied magnetic field, g 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.

H^{i}_{exch} and H^{e}_{exch} in expressions 3.a, 3.b come from the molecular field approximation of the actual exchange interaction

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

The parameter J_0 in 4 is

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

In this paper the H_{CF} will be constructed taking into account only the two lowest levels of Pr^{+3} in an hexagonal symmetry [2]. Fig. 1 shows the three first levels of the complete level scheme [3]. In the case of Pr^{+3} in $PrA\ell_3$, according to Mader et al. [3], these two levels are singlets separated by an energy gap Δ = 3,41 meV. The eigenfunctions of these levels are

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

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

Within the basis defined by 7

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

and

$$H^i_{exch} = -\ \mu_B h_i \begin{pmatrix} 0 & \alpha_0 \\ \alpha_0 & 0 \end{pmatrix} \tag{9}$$

where

$$\alpha_0 = \langle e_0 | gJ^z | e_1 \rangle \tag{10}$$

In what follows α_o 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 magnetizations is given by

$$\mu_B \langle gJ^z \rangle = - \frac{\sum_{j=0}^{1} \frac{\partial E_J}{\partial h_i} \exp(-\beta E_j)}{\sum_{j=0}^{1} \exp(-\beta E_j)} \tag{11}$$

where E_j are the engenvalues of (2.a), given in appendix A and $\beta = \frac{1}{k_B T}$.

The electronic magnetization is obtained from

$$\int_0^{\varepsilon_F} \frac{n(\varepsilon)d\varepsilon}{\exp\left[-\beta(\varepsilon-\mu_B h_e-\mu)+1\right]} \mp \int_0^{\varepsilon_F} \frac{n(\varepsilon)d\varepsilon}{\exp\left[-\beta(\varepsilon+\mu_B h_e-\mu)+1\right]} = \begin{cases} 2zN\langle s^z \rangle & (12.a) \\ 2N & (12.b) \end{cases}$$

In 12, $n(\varepsilon)$ 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 $n(\varepsilon)$.

$$n(\varepsilon) = \begin{cases} N/2\varepsilon_0 & \text{if } 0 \le \varepsilon \le \varepsilon_0 \\ 0 & \text{otherwise} \end{cases} \tag{13}$$

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 $n(\varepsilon)$ given by 13, equation 12 can be simplified (see appendix B). For the range of temperature and band width ε_0 of interest, we have

$$J_0\langle gJ^z\rangle + 2\mu_B h_0 = 4\varepsilon_F\langle s^z\rangle \tag{14}$$

where $\varepsilon_F = z\varepsilon_0$.

Combining 14 with 11, one obtains

$$\langle gJ^z\rangle = \frac{2\alpha_0^2\mu_B h_i}{\left[\Delta^2+4\alpha_0^2(\mu_B h_i)^2\right]^{1/2}} \tanh\left\{\frac{\left[\Delta^2+4\alpha_0^2(\mu_B h_i)^2\right]^{1/2}}{2k_B T}\right\} \tag{15}$$

From 15, 14 and 4.a, we also derive the ionic magnetic susceptibility

$$\chi = \frac{\alpha_0^2\mu_B^2(4\varepsilon_F - J_0)\tanh(\Delta/2k_B T)}{2\Delta\varepsilon_F - J_0^2\alpha_0^2\tanh(\Delta/2k_B T)} \tag{16}$$

The limit at T = OK of equation 15 is of interest

$$\langle gJ^2\rangle_0 = \frac{2\alpha_0^2\mu_B h_i}{\left[\Delta^2+4\alpha_0^2(\mu_B h_i)^2\right]^{1/2}} \tag{17}$$

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

$$J_0^c = \frac{(2\Delta\varepsilon_F)^{1/2}}{\alpha_0} \tag{18}$$

Equation 18 defines the boundary between the ferro and paramagnetic regions in the space of parameters $J_0/2\varepsilon_F$ versus $\Delta/2\varepsilon_F$.

Equation 18 is the starting point to study the magnetic behaviour in the para- and ferromagnetic regions. Figures 2 and 3 illustrate exchange enhancement and crystal field effects in the para and ferromagnetic phases respectively. Fig. 4 shows the inverse of χ versus k_BT/ε_F for different values of Δ/ε_F, J_0/ε_F and α_0.

5. Application to $PrA\ell_3$

The magnetic susceptibility of $PrAl_3$ was experimentally studied by Mader et al. [3]. Figure 5 shows the inverse susceptibility versus temperature, obtained using equation 16 together with the experimental points of Mader et al. [3]. The fitting is for $\varepsilon_F = 8.2$ 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 ε_F is taken from Jarlborg et al. [4], who computed the band structure of $LaA\ell_2$, $CeA\ell_2$ and $yA\ell_2$.

Finally it is interesting to note that in the space parameters $J_0/2\varepsilon_0$ versus $\Delta/2\varepsilon_0$, the point defined by the above values of J_0, Δ, ε_F and α_0 fall in the paramagnetic phase (see equation 18 and its interpretation).

APPENDIX A

Eingenvalues of ionic Hamiltonian (eq. 2.a)

In order to compute $\mu_B\langle gJ^z\rangle$, we need $-\frac{\partial E_j}{\partial h_1}$ (see equation (11). $E_j(j = 0,1)$ are calculed from

$$\begin{vmatrix} -E & -\alpha \\ -\alpha & \Delta-E \end{vmatrix} = 0 \tag{A.1}$$

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

$$E_1 = \frac{\Delta}{2} + \frac{\sqrt{\Delta^2+4\alpha^2}}{2} \tag{A.3}$$

where

$$\alpha = \mu_B h_1 \alpha_0$$

Appendix B

Electronic magnetic state equation for a rectangular energy density of states

Putting (1.3) into (12), we can solve for $<s^z>$, giving

$$2\beta\mu_B h_e = \mathrm{Ln}\,\frac{\sinh\beta\varepsilon_0(z-1/2) - \sinh\beta\varepsilon_0(1/2+2z<s^3>}{\sinh\beta\varepsilon_0(z-1/2) - \sinh\beta\varepsilon_0(1/2-2z<s^z>}\ . \qquad (B.1)$$

For low temperatures $\beta\varepsilon_0 >> 1$ and z around 0.5, (B.1) reduces to

$$2\mu_B h_e = 4\varepsilon_F<s^z> \qquad (B.2)$$

where

$$\varepsilon_F = z\varepsilon_0$$

Figure captions

Figure 1 The three first levels of Pr^{+3} in $PrA\ell_3$, extracted from the complete level structure from Mader et al. [3]. In this paper only the two first levels were used.

Figure 2 Magnetic response $<gJ^z>$ versus external magnetic field at T = OK. Curves (a) and (b) are for J_0/ε_F equal to 0.0 and 0.03162. For both curves $\Delta/\varepsilon_F = 0.0005$.

Figure 3 Ionic magnetization (in units of μ_B) versus J_0/ε_F at T = OK. Curves (a) and (b) are for Δ/ε_F equal to 0.0005 and 0.0004 respectively.

Figure 4 Reduced inverse magnetic susceptibility $\mu_B h_i/J_0<gJ^z>$ versus k_BT/ε_F. For curves (a) and (c) $\Delta/\varepsilon_F = 4.0 \times 10^{-5}$, $J_0/\varepsilon_F = 1.0 \times 10^{-2}$ and α_0 are respectively 2.0 and 1.0. For curves (b) and (c) $\alpha_0 = 1.0$, $J_0/\varepsilon_F = 0.5 \times 10^{-4}$ and Δ/ε_F are respectively 2×10^{-4}.and 4×10^{-4}. For curves (c) and (d) $\alpha_0 = 1.0$, $\Delta/\varepsilon_F = 4 \times 10^{-4}$ and J_0/ε_F are respectively 0.5×10^{-4} and 1.0×10^{-4}.

Figure 5 Inverse of susceptibility $1/\chi$ (in mol/emu units) for $PrA\ell_3$ versus temperature (in K units). The curve was computed from parameters presented in the text, and the experimental points were taken from Mader el al. [3].

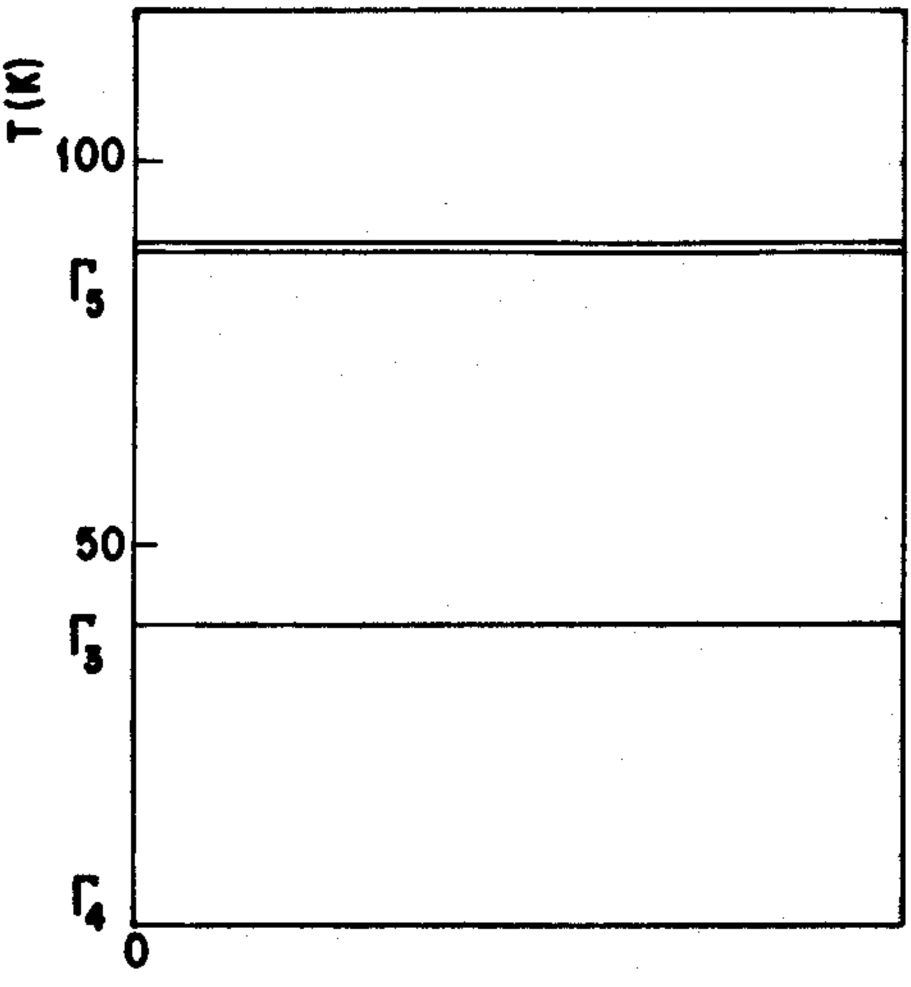

Fig. 1

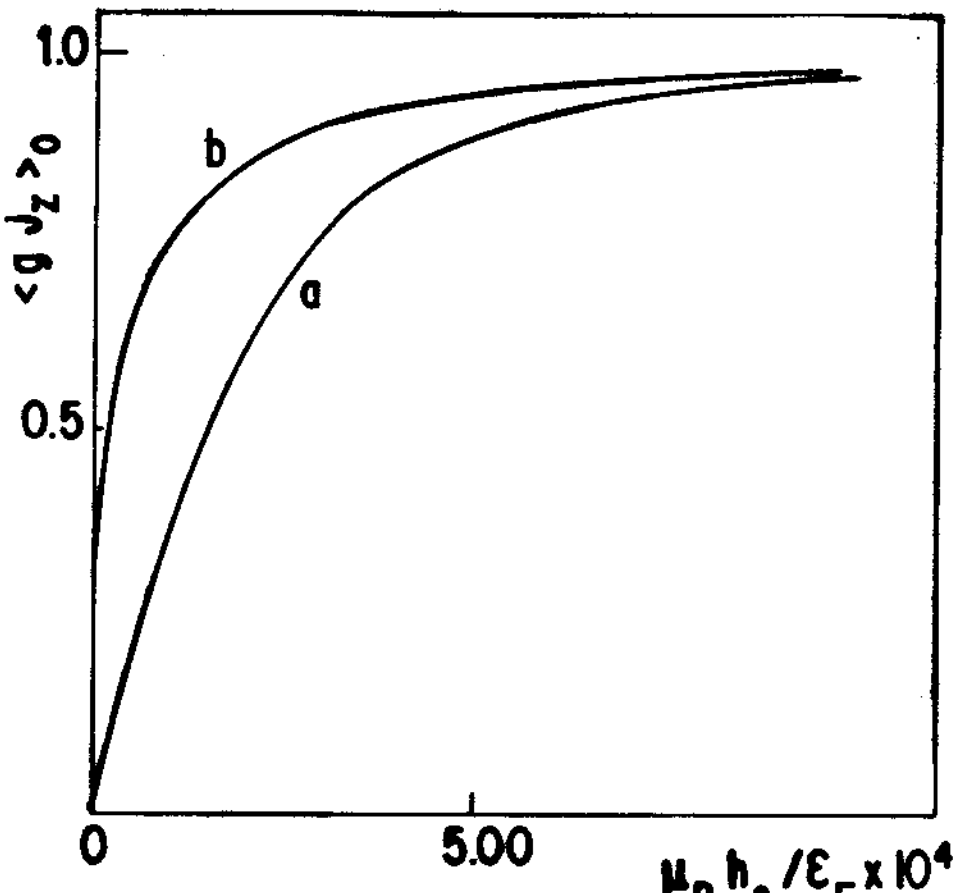

Fig. 2

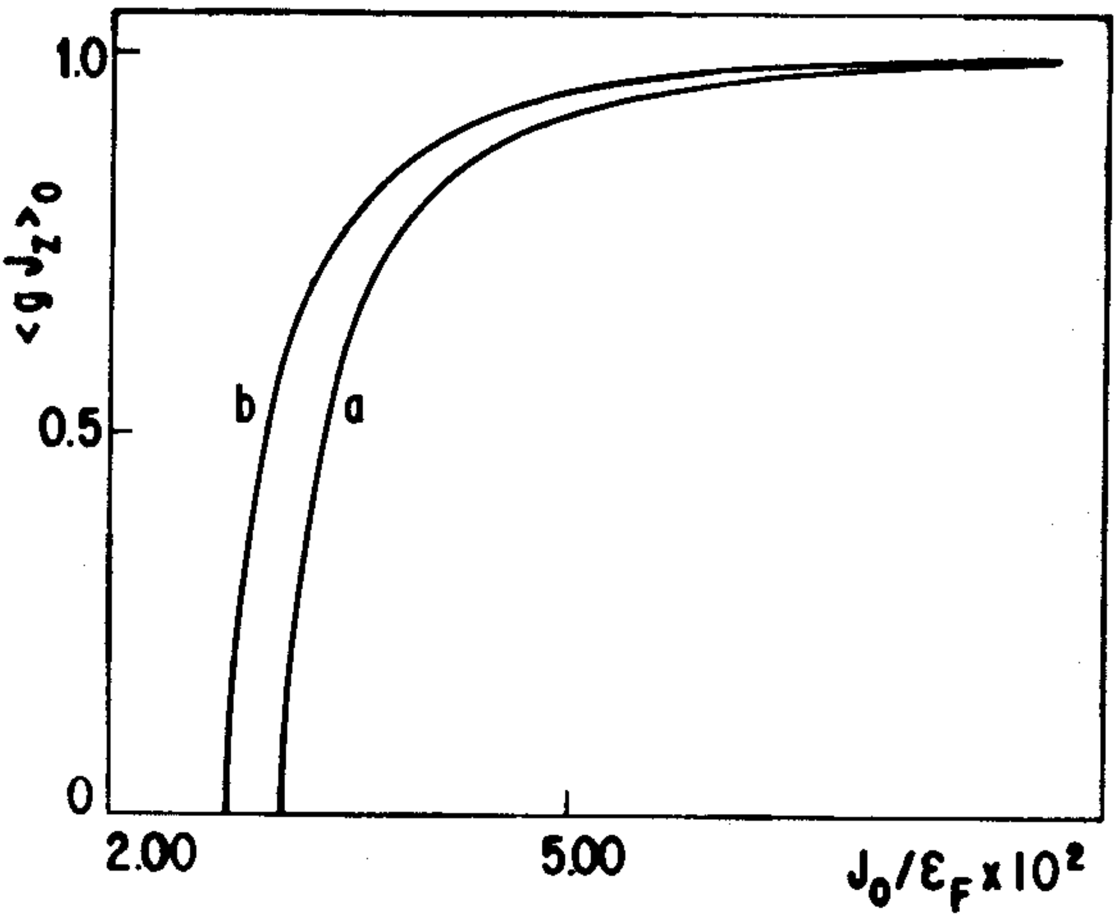
1.0
0.5
0
2.00
5.00
b
a
$\langle g J_z \rangle_0$
$J_0/\varepsilon_F \times 10^2$

Fig. 3

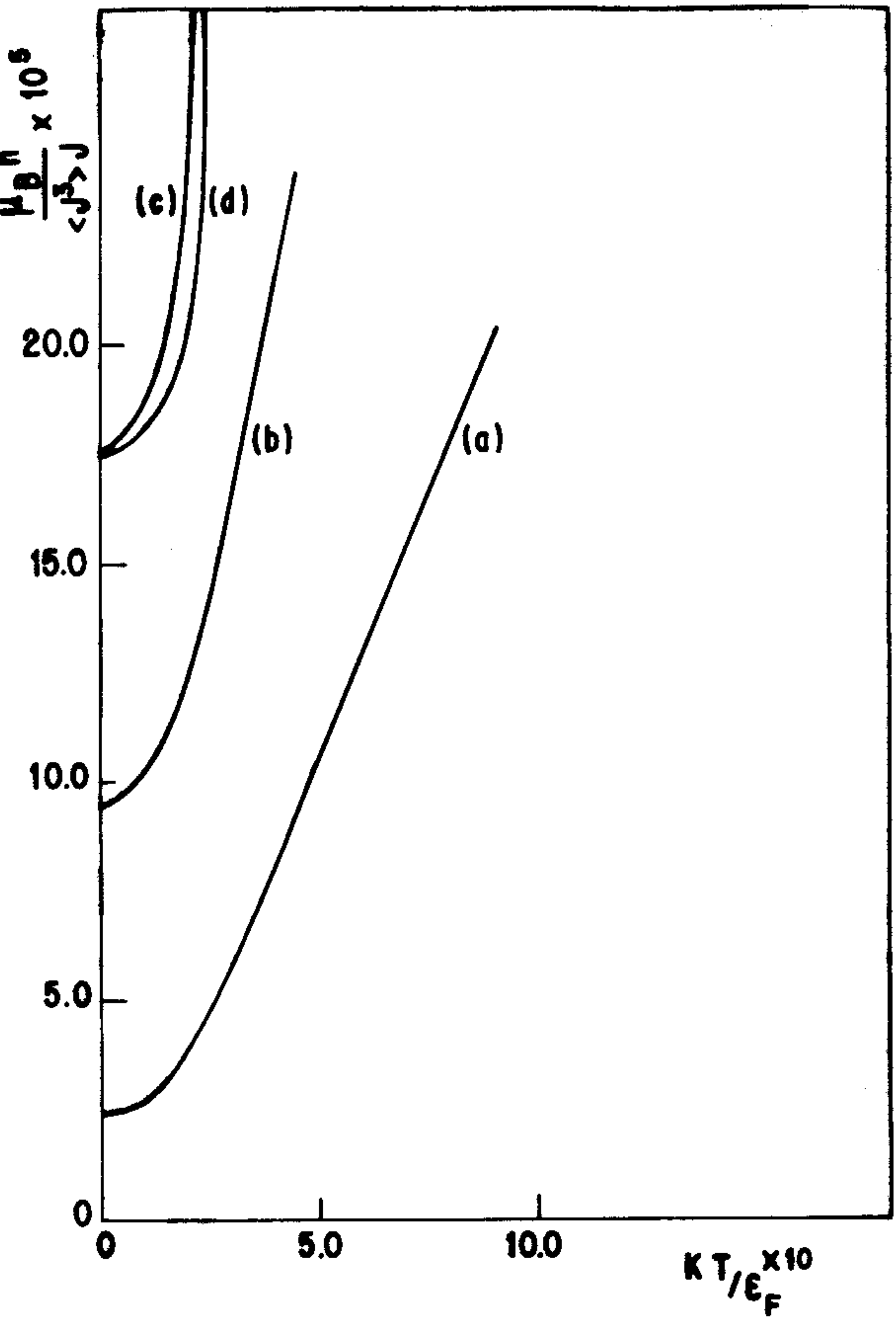
$\frac{\mu_B h}{\langle J^3 \rangle J} \times 10^5$
20.0
15.0
10.0
5.0
0
(c)
(d)
(b)
(a)
0
5.0
10.0
$KT/\epsilon_F \times 10$

Fig. 4

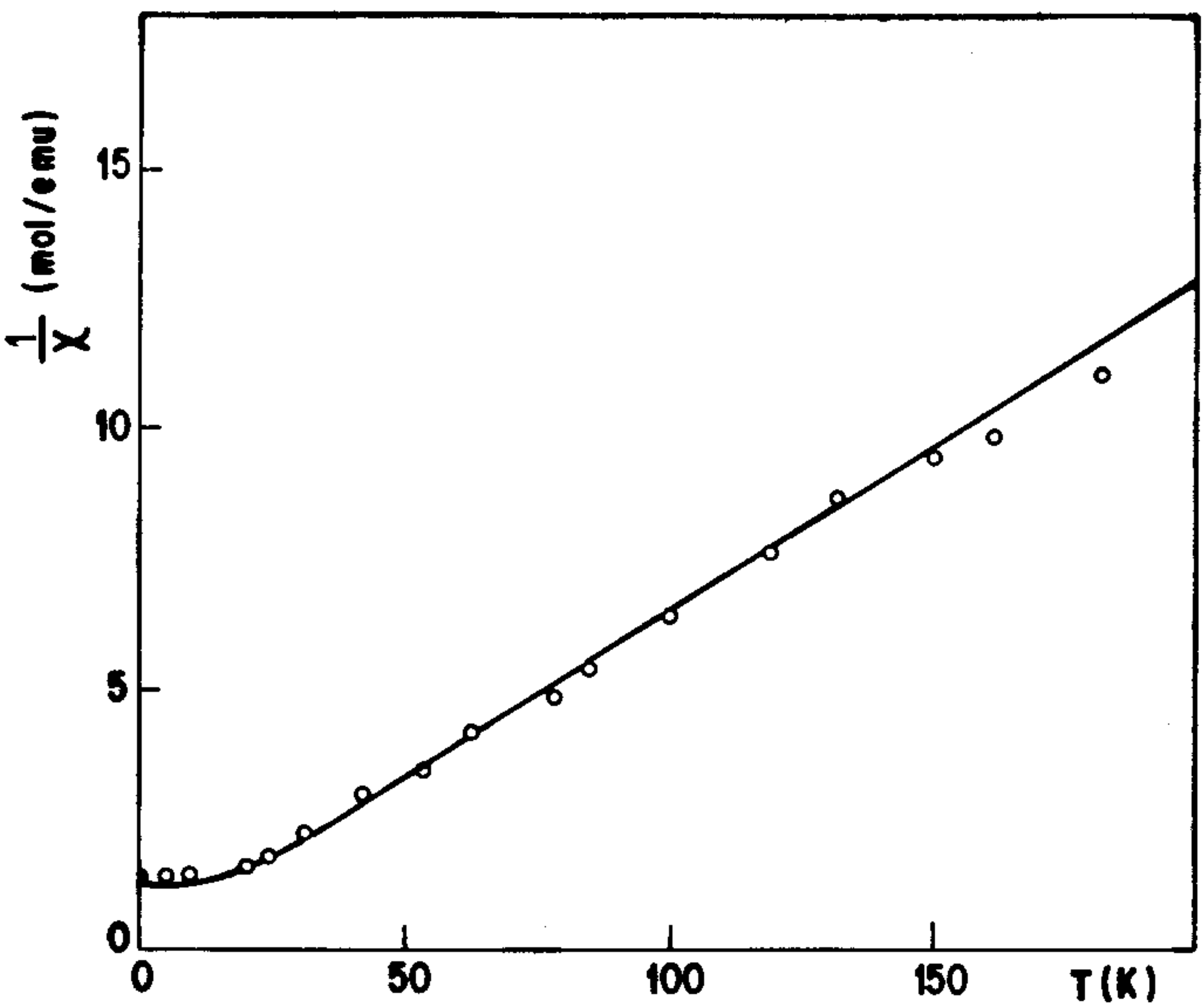

Fig. 5

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