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1992 J. Phys.: Condens. Matter 4 4687

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Spin-wave model for systems with competing magnetic anisotropies and undergoing spin reorientation transitions

A del Moral

Laboratorio de Magnetismo, Departamento de Física de Materia Condensada and Instituto de Ciencia de Materiales de Aragón (ICMA), Universidad de Zaragoza and Consejo Superior de Investigaciones Científicas (CSIC), 50009 Zaragoza, Spain

Received 31 May 1991, in final form 23 December 1991

Abstract. An adapted spin-wave model has been developed to explain spin reorientation (SR) processes in rare-earth ferromagnetic (or ferrimagnetic) systems with competing axial-planar anisotropies. The model involves only *second* order in the effective spin S and single-ion crystal electric field (CEF) interactions, and several results of interest have been achieved, in particular the temperature dependence of the SR angle θ . The conditions required to have an SR transition have been established. Of importance is that the 0 K SR angle $\theta(0)$ becomes a consequence of the frozen-in uniform zero-point quantum spin fluctuations, and depends only on the ratio $\Delta = -(D_p/D_a)$ between the planar and axial second-order CEF strengths. A consequence is that the SR temperature T_{SR} and the 0 K second-order magnetic anisotropy free energy, $K_1(0) \sin^2 \theta(0)$, become proportional. The angle θ fluctuates critically for $T \leq T_{SR}$ and its exponent has been calculated, $\beta = 1/2$, as well as the dependence of T_{SR} with Δ and the exchange strength. At low temperatures the dependence of $\sin^2 \theta$ becomes of $T^{3/2}$ Bloch-like type. A comparison of these results with current experimental findings in hard magnetic intermetallics, such as $(RE'_x RE_{1-x})_2 Fe_{14}B$ and $RE'_x RE_{1-x} Co_5$, has been made. The effective anisotropy constant $K_1(0)$ and the magnon energy at the Brillouin zone boundary for some of the above series of intermetallics have been obtained from previous experiments, the $K_1(0)$ values being in good agreement with theory.

1. Introduction

In previous papers (Algarabel *et al* 1988, Ibarra *et al* 1988a, 1989a, b, del Moral *et al* 1989, 1990, Moze *et al* 1990a, b) it was shown that spin reorientation (SR) transitions are phase ones occurring between a high-temperature magnetically axial (A) structure and a low-temperature one, where the ferromagnetic (or ferrimagnetic) system becomes conical (C) or planar (P), the transition occurring at a critical temperature T_{SR} . The driving mechanism is believed to be the magnetocrystalline anisotropy energy, in systems where there exist two competing anisotropies, of A and P character respectively (Cullen and Callen 1985). In fact, such transitions occur most widely in rare-earth (RE) magnetically hard intermetallics, where we alloy two RE ions with crystal electric field (CEF) Stevens *second*-order coefficients α_j of opposite signs. Examples of such systems are the $(Er_x RE_{1-x})_2 Fe_{14}B$ (RE = Nd, Dy, Ho) and $RE'_x RE_{1-x} Co_5$ intermetallics, which present a crystallographic axial structure (tetragonal or hexagonal, respectively). It was shown

(del Moral *et al* 1989, 1990) that the SR angle θ , i.e. the angle formed by the average magnetization with the c axis, is the relevant order parameter.

In experiments (see all the references above) performed over a wide range of the mentioned pseudobinary and pseudoternary intermetallics we were able to determine the temperature (T) dependence of θ . As we shall see, the spin-wave (sw) approximation becomes a very satisfactory technique to deal with SR, allowing the calculation of $\theta(T)$ and predicting many other interesting features of the phenomenon amenable to comparison with experiment. Martynov and Saadrev (1981) earlier applied the sw theory to the region *above* T_{SR} , using the quantum-statistical-mechanics Green function method, and obtained a cumbersome implicit equation for the calculation of T_{SR} , which was difficult to compare with experiment. However, our approach, which brings in substantial modifications of the Holstein-Primakoff (HP) (1940) method, allows an explicit calculation of T_{SR} .

This paper is organized in the following way: In section 2 we justify and discuss our model hypothesis and adapt the HP method to deal with systems with competing anisotropies undergoing SR transitions. In section 3 we calculate $\theta(T)$ explicitly, including $T = 0$ K. The conditions needed for SR to occur are established. The critical scaling of θ near T_{SR} is made patent, explicitly calculating the exponent β and providing an expression for T_{SR} . The low-temperature variation of θ is obtained, finding a connection of $\theta(0)$ with T_{SR} . Finally, in section 4 we make a comparison of our findings with some of the experimental results available.

2. Spin-wave model for systems with competing magnetic anisotropies; justification and discussion

2.1. Physical justification and introductory remarks

Our model deals with substitutionally disordered alloys, i.e. RE intermetallics where we have two rare-earth ions with opposite-sign Stevens factors α_j , such as $(\text{RE}'_x\text{RE}_{1-x})_2\text{Fe}_{14}\text{B}$ and $(\text{RE}'_x\text{RE}_{1-x})\text{Co}_5$, where the RE and RE' ions occupy random positions. These kinds of compounds are really quite complicated magnetic systems, both from their large and complex unit cells and also from their magnetic structures (see e.g. Kirchmayr and Poldy 1982), with several non-equivalent RE-RE' and transition-metal (TM) sublattices (e.g. six for Fe and two for the RE-RE' in the case of the $(\text{RE}'_x\text{RE}_{1-x})_2\text{Fe}_{14}\text{B}$ compounds, the Fe atoms also carrying different atomic magnetic moments (Givord and Li 1985)). The application of the sw approximation to such systems by distinguishing the three (RE, RE' and TM) magnetic sublattices is a really formidable problem. To our knowledge the only attempt to deal with sw excitations in magnetically ordered *Ising* systems with multiple structurally ordered magnetic sublattices was done by Saenz (1962), but considering only axial anisotropies (no SR possible), through the simplifying assumption of introducing equivalent anisotropy fields, which is not a fully clear-cut microscopic description. Our problem, where the rare-earth sublattice is structurally disordered, is frankly much more complex, in fact formidable (Kaneyoshi 1984), and needs some reasonable simplifications if a fully *microscopic* model, such as the sw approximation, is to be used.

There have so far been two ways to tackle the SR problem. One is the well known *phenomenological* approach, where one necessarily needs to consider higher-order anisotropy constants to produce a continuous rotation of the magnetization. This

approximation does not distinguish among the different sublattice magnetizations and anisotropy constants, assuming a unique magnetocrystalline anisotropy free energy,

$$F_k = K_1(T) \sin^2 \theta + K_2(T) \sin^4 \theta.$$

For collinear ferromagnets the introduction of different $K_i(T)$, $i = 1, 2$, for each sublattice is clearly useless, and the use of only second-order constants, although competing and with different temperature dependences, i.e. $K_1^{\text{RE}}(T)$, $K_1^{\text{RE}'}(T)$ and $K_1^{\text{TM}}(T)$, clearly does not produce a temperature-dependent SR angle at all.

The other, and more powerful, approach is the *mean-field crystal-field* (MF-CEF) approximation (Ibarra *et al* 1989a). This description lies between an atomistic one, when describing the CEF upon the RE by a Hamiltonian H_{CEF} , and a very simplified mean-field one to deal with the exchange interactions. However, in order to produce a continuous SR process, H_{CEF} must again include higher-order CEF operators, i.e.

$$H_{\text{CEF}} = B_2^0 O_2^0 + B_2^2 O_2^2 + B_4^0 O_4^0$$

and although the model distinguishes between the two RE species, it treats the weaker TM anisotropy within the phenomenological model. The limitation of this approach, although very valuable, is that it is a computational one, fitting the temperature variation of θ by an adequate choice of the parameters involved, but hardly revealing the detailed physical processes involved in the complexity of the SR phenomenon.

Therefore, the ultimate reason to choose the SW approximation is that, as we shall see, it predicts a continuous temperature variation of θ , but, in contrast with the phenomenological or MF-CEF models, without invoking *higher-order* anisotropy or CEF terms. This assumption is quite important because, for RE, RE' ions with competing α_J factors, the main mechanism for SR to occur must be more the result of this competition and less the result of higher-order interactions. But, there is also the important aspect of a deeper understanding of the physics involved in the SR phenomenon, in terms of the many predictions liable to experimental verification, which were briefly mentioned in section 1.

Considering the mentioned complexities of our RE'_uRE_{1-u}TM_v intermetallics, in order to use the SW approximation we have made the simplification of assuming an average magnetic ion, with effective spin S , defined through the experimentally determined magnetic moment per unit cell, $\mu = g\mu_B S$, the resultant of the coupling among the RE', RE and TM moments, i.e.

$$\mu = n[\pm u\mu_{\text{RE}'} \pm (1-u)\mu_{\text{RE}} + v\mu_{\text{TM}}]$$

with + for light and - for heavy rare-earth ions respectively and n the number of molecules per unit cell; g is the corresponding effective Landé factor. However, such a severe simplification can be well justified to a reasonable extent. The RE'-RE sublattice is substitutionally random and it is justified to consider an average magnetic moment between $\mu_{\text{RE}'}$ and μ_{RE} . Moreover, in our kinds of intermetallics, the strongest exchange interaction is between the TM atoms, which is really quite strong according to the large Curie temperatures observed (~ 600 K) (Wallace 1973, Buschow 1980, Coey *et al* 1985). Therefore, the exchange between the overall spins S is, within our model, the resulting one between the TM spin contributions to S . Now, the formation of the overall magnetic moment is believed to be due to the exchange polarization of the RE and RE' spins by the TM ones (Campbell 1972), giving rise to a magnetic moment quite rigid under rotation (Ibarra *et al* 1991). On the other hand the TM anisotropy energy is, in general, less relevant compared with the competing RE-RE' ones (Ibarra *et al* 1989a) and can, to some

extent, be neglected within our model. Finally, the source of the anisotropy can be described in the following way. The RE and RE' ions are respectively submitted to axial and planar CEF, and in this way the overall effective spin S is submitted to the competing CEF but through the intermediary action of the RE and RE' ionic contributions to S .

Summarizing, we propose a scenario, for our pseudoternary or pseudobinary intermetallics, of a lattice of unit cells with effective spins S , coupled to one another through the strongest TM exchange and submitted to competing second-order CEF, coming from the RE(axial)-RE'(planar) sublattice. In this sense, although the magnetic moment structural fluctuation is neglected, the most important one in dealing with the SR phenomenon, namely the competing character of the RE anisotropies, is truly taken into account.

2.2. Basic Hamiltonian and rotation to a frame having the magnetization direction as quantization axis

Therefore, we will assume a crystal-field Hamiltonian, referred to the crystal axes frame, for a lattice of N spins, given by

$$H_{\text{CEF}} \equiv H_a + H_p = -\sum_l [D_a \bar{O}_2^0(S_l) + D_p \bar{O}_2^2(S_l)] \quad (1)$$

where D_α ($\alpha = a, p$) are the respective CEF strength parameters and l the lattice points. The definitions of the CEF operators are currently confusing; we will adhere to the ones of Buckmaster (1962) and accordingly take extended Stevens operators \bar{O}_n^m and \hat{O}_n^m (Rudowicz 1985). The axes frame has z parallel to the crystal c axis (tetragonal, orthorhombic or hexagonal), $x \parallel a$ and $y \perp a$, within the basal plane. The Hamiltonian H_p in (1), although strictly representing an anisotropy within the basal plane, within our model also becomes an anisotropy capable of tilting S away from the c axis, in competition with the axial CEF represented by H_a . Finally, we will express the exchange interaction through the Heisenberg Hamiltonian

$$H_{\text{ex}} = -\frac{1}{2} \sum_{l,m} J(l-m) S_l \cdot S_m \quad (2)$$

where $J(l-m)$ is the effective exchange interaction between the spins.

The SW approximation naturally assumes small spin deviations from the quantization axis (QA). For a system undergoing SR, the natural QA is the average magnetization (M) direction. Therefore, we shall rotate our axes frame by Eulerian angles ψ and θ , such that the new axis $z' \parallel M$, θ and ψ becoming the polar angles of M . The Hamiltonian $H = H_{\text{ex}} + H_a + H_p$ will be rotated accordingly, although H_{ex} indeed remains invariant. According to Rudowicz (1985) and Buckmaster *et al* (1972),

$$[\bar{O}_2^0]_{\text{cf}} = \frac{1}{2} \sin^2 \theta \bar{O}_2^2 - 3 \sin^2 \theta \bar{O}_2^1 + \frac{1}{2} (3 \cos^2 \theta - 1) \bar{O}_2^0 \quad (3a)$$

$$[\bar{O}_2^2]_{\text{cf}} = \frac{1}{2} \cos(2\psi) (\cos^2 \theta + 1) \bar{O}_2^2 + \cos(2\psi) \sin(2\theta) \bar{O}_2^1 + \frac{1}{2} \cos(2\psi) \sin^2 \theta \bar{O}_2^0 \\ - 2 \sin(2\psi) \sin \theta \hat{O}_2^1 - \sin(2\psi) \cos \theta \hat{O}_2^2 \quad (3b)$$

where the label cf refers to the crystal frame axes and the primed one to the rotated frame.

The rest of section 2.2 is straightforward. We will first perform the transformations, for large spin S (Wallace 1973, Coey *et al* 1985), $S_i^- = S - a_i^\dagger a_i$, $S_i^+ = \sqrt{(2S)} a_i$, $S_i^- = \sqrt{(2S)} a_i^\dagger$, where a_i^\dagger and a_i respectively are creation and annihilation spin deviation

operators. It is simple to obtain from equations (1)–(3), keeping terms only up to second order in a_l^\dagger, a_l , a rotated Hamiltonian $H' = H'_{\text{ex}} + H'_a + H'_p$, with coefficients that are in general complex functions of θ and ψ . Transforming H' to the magnon representation (Holstein and Primakoff 1940, Keffer 1966), in terms of a_k^\dagger and a_k operators of wave-vector k , we obtain the Hamiltonian

$$H' = H_0 + \sum_k (A_k a_k^\dagger a_k + \frac{1}{2} B_k a_k a_{-k} + \frac{1}{2} B_k^\dagger a_k^\dagger a_{-k}^\dagger) + (\rho_0 a_0 + \rho_0^\dagger a_0^\dagger) \quad (4)$$

where

$$H_0 = -ND_a(S/2)(S/2 - 1)(3 \cos^2 \theta - 1) - ND_p(S/2)(S/2 - 1) \cos(2\psi) \sin^2 \theta \quad (5)$$

and where the functions A_k and B_k are given by

$$A_k = \varepsilon_k + \frac{3}{2} D_a S (3 \cos^2 \theta - 1) + \frac{3}{2} D_p S \cos(2\psi) \sin^2 \theta \quad (6a)$$

$$B_k = -(\sqrt{6}/4) [\frac{3}{2} D_a S \sin^2 \theta + \frac{1}{2} D_p S \cos(2\psi) (\cos^2 \theta + 1) + i D_p S \sin(2\psi) \cos \theta] \quad (6b)$$

with an unperturbed magnon energy, for Bravais lattices of effective spins S (Kittel 1967),

$$\varepsilon_k = \frac{1}{2} JSz \left(1 - \frac{1}{z} \sum_{\delta} e^{ik \cdot \delta} \right). \quad (7)$$

The summation in (7) is restricted to the z nearest neighbours at distances $l - m \equiv \delta$ of the probe ion, the exchange being assumed uniform, J . There also appear in H' the important $k = 0$ terms a_0 and a_0^\dagger , with coefficient

$$\rho_0 = \sqrt{N} [\frac{3}{2} \sqrt{2} S^{3/2} D_a \sin(2\theta) - \frac{1}{2} \sqrt{2} S^{3/2} D_p \cos(2\psi) \sin(2\theta) - i \sqrt{2} S^{3/2} D_p \sin(2\psi) \sin(2\theta)]. \quad (8)$$

2.3. Hamiltonian diagonalization and consequences

The second term of Hamiltonian (4) has the well known HP form and can be diagonalized using the usual HP diagonalization transformations. However, the linear term in a_0^\dagger, a_0 is new and has to be diagonalized using a novel transformation (Cullen 1987), in its present form,

$$a_k = \alpha_k + c_k \delta_{k,0} \quad \text{and} \quad \text{cc} \quad (9)$$

where the Fourier transform c_0 of $c_k \delta_{k,0}$ represents frozen-in *uniform* spin deviations. The diagonalization of Hamiltonian (4) by (9) yields

$$c_0 = -(\rho_0^\dagger A_0 - \rho_0 B_0^\dagger) / (A_0^2 - |B_0|^2) \quad \text{and} \quad \text{cc}. \quad (10)$$

Now, the new diagonalized Hamiltonian H'' contains a $k \neq 0$ term identical to (4), but with a_k^\dagger, a_k substituted by the $\alpha_k^\dagger, \alpha_k$ operators, plus a new $k = 0$ term of the form

$$H''_0 = H_0 + A_0 |c_0|^2 + \frac{1}{2} B_0 c_0^2 + \frac{1}{2} B_0^\dagger (c_0^\dagger)^2 + \rho_0 c_0 + \rho_0^\dagger c_0^\dagger \quad (11)$$

which is just a c -number. Hamiltonian H'' again has the standard HP form (Keffer 1966) and can be diagonalized using the two-stage HP diagonalization transformations

(Holstein and Primakoff 1940, Keffer 1966). The fully diagonalized Hamiltonian becomes, in the final form,

$$H'' = H_0'' + \sum_k [(\tilde{\beta}_k^+ \tilde{\beta}_k + \frac{1}{2})\tilde{\epsilon}_k - \frac{1}{2}A_k] \quad (12)$$

with the *perturbed*, by the CEF, magnon energy

$$\tilde{\epsilon}_k = (A_k^2 - |B_k|^2)^{1/2} \quad (13)$$

which indeed includes the $k = 0$ CEF magnon energy gap, immediately obtained from equations (6a, b) and (13).

Before continuing we will assume that SR takes place within the (c, a) plane, i.e. $\psi = 0$ in equations (5), (6a, b) and (8), which is a simplifying real assumption in many intermetallic systems (see references of section 1). In such a situation $H_0'' = H_0 + c_0\rho_0$. Also $\psi = 0$ is the condition for minimum anisotropy energy, for $D_p > 0$, within the basal plane if we treat Hamiltonian H'' within the classical limit, where simply $H'' = H_0$.

It is now worthwhile to notice that Hamiltonian (12) contains three kinds of spin fluctuations: zero-point quantum fluctuations, represented by the term

$$H_1 = \sum_k \frac{1}{2}(\tilde{\epsilon}_k - A_k) \equiv \frac{1}{2} \sum_k [(A_k^2 - |B_k|^2)^{1/2} - A_k] \quad (14)$$

thermally excited magnons, represented by

$$\sum_k \tilde{\beta}_k^+ \tilde{\beta}_k \tilde{\epsilon}_k$$

and, most importantly, a term $c_0\rho_0$ and CC dealing with the uniform ($k = 0$) spin canting from the c axis, which constitutes our main pursuit.

3. Spin reorientation angle

The spontaneous SR angle θ is a static equilibrium quantity, obtained by minimization of the system free energy $F = -(1/\beta) \ln Z$, with $\beta \equiv 1/k_B T$, Z being the partition function, obtained from (14) as $Z = \text{Tr} \exp(-\beta H'')$; thus

$$F = F_0 + F_k(T) = F_0 - \frac{1}{\beta} \ln \prod_k \frac{1}{1 - \exp(-\beta \tilde{\epsilon}_k)} \quad (15)$$

with $F_0 = H_0'' + H_1$. Again, in (15) there are three contributions to the free energy; the static ground-state energy $H_0'' = H_0 + c_0\rho_0 \equiv H_0 - \rho_0^2/(A_0 + B_0)$; the zero-point quantum fluctuation energy H_1 ; and the thermally excited magnon free energy $F_k(T)$. There are two temperature regions of interest, in order to calculate θ , as now discussed.

3.1. Zero-point SR angle

At 0 K, the spontaneous SR angle $\theta(0)$ is obtained by minimization against θ of the zero-point free energy f_0 (per spin),

$$f_0 = F_0/N = -(S/2)(S/2 - 1)[D_p \sin^2 \theta + D_a(3 \cos^2 \theta - 1)] + H_1/N + c_0\rho_0. \quad (16)$$

It is now essential to notice that, in the pure classical limit, the magnetocrystalline anisotropy free energy becomes

$$f_0^c = -(S/2)(S/2 - 1)[D_p \sin^2 \theta + D_a(3 \cos^2 \theta - 1)] \quad (17)$$

and therefore $\partial f_0^c / \partial \theta = 0$ yields the solution $\sin \theta \cos \theta = 0$, which has only two equilibrium states, $\theta = 0$ (for $D_p/D_a < 3$) and $\theta = \pi/2$ (for $D_p/D_a > 3$), with no intermediate

minimum for the SR angle. Even considering structural fluctuations, as discussed in section 2.1, by introducing different spins in the classical anisotropy free energy (17), i.e. S_a and S_p for the RE and RE' sublattices respectively, the result would be the same. Again, this means that classically second-order anisotropy terms alone are unable to produce a continuous SR angle θ . Therefore, the spin fluctuation terms $c_0\rho_0 + H_1/N$ in equation (16) are, in principle, responsible for the formation of a 0 K finite SR angle. Calling, for convenience,

$$\begin{aligned}\psi_1(\theta) &= D_a(3 \cos^2 \theta - 1) + D_p \sin^2 \theta \\ \psi_2(\theta) &= \psi_1(\theta) - 2(D_a + D_p)\end{aligned}\quad (18)$$

we will now evaluate the term $f_1 = H_1/N$, given by equation (14). We will assume that at the temperatures considered, well below the Curie point T_c (T_{SR} are at least of the order of $T_c/2$ for the intermetallics mentioned in section 1 (Ibarra *et al* 1989a, Marquina 1990, Algarabel *et al* 1988, Coey *et al* 1985, Wallace 1973)), only low-energy magnons will be excited. Then a series expansion of (7) for a simple cubic (sc) lattice of effective spins S (which is the simple way in which we have modelled the complex tetragonal or hexagonal lattices of our intermetallics) gives, as a first-order approximation, for the unperturbed magnon energy, $\varepsilon_k = Ak^2$, with $A \equiv \frac{1}{2}JSa^2$ being the sw stiffness constant (a is an average lattice constant). Assuming that the anisotropy term $B_k \equiv B_0$ (see (6b)) is small compared with A_k (i.e. in the large exchange limit, which is the situation for the present systems), we can write

$$f_1 \equiv \frac{H_1}{N} \cong -\frac{1}{4} \sum_k \frac{|B_k|^2}{A_k}$$

which can be evaluated in the continuous- k integral approximation, i.e.

$$f_1 = -\left(\frac{3v_a S^2}{16^2 \pi^2}\right) \frac{\psi_2(\theta)^2}{A} \int_0^{k_c} \frac{k^2}{\varepsilon_k + \frac{3}{2}S\psi_1(\theta)} dk \quad (19)$$

where $k_c = (\pm\pi/a, 0, 0)$ and its symmetric reciprocal lattice points are the limits of the average cubic Brillouin zone (BZ) boundary; v_a is the lattice volume per spin. The exact result for f_1 is quite cumbersome to handle, i.e.

$$\begin{aligned}f_1 &= -(3v_a S^2/16^2 \pi^2)(\psi_2^2/A)\{k_c \\ &\quad - (3S/2)^{1/2}(\psi_1/A)^{1/2} \tan^{-1}[k_c/(3S/2)^{1/2}(\psi_1/A)^{1/2}]\}.\end{aligned}\quad (20)$$

However, it can be shown (del Moral *et al* 1992) that $\xi = ((2/3S)(A/\psi_1))^{1/2}$ is the magnetic correlation length characterizing small-angle neutron scattering (SANS), which is rather large, of the order of several hundred ångströms, as determined in the $(Er_xNd_{1-x})_2Fe_{14}B$ compounds (del Moral *et al* 1991, 1992). Therefore, $k_c\xi$ is large and f_1 can be simplified (see the last term in equation (21) below). Therefore, from equations (16), (17) and (20), the zero-point free energy becomes

$$\begin{aligned}f_0 &= -(S/2)(S/2 - 1)\psi_1 + 2D_a S^2/(3D_a - D_p)\{\dot{\psi}_1^2/[3\psi_1 + (\sqrt{6}/2)\psi_2]\} \\ &\quad - (3v_a S^2/16^2 \pi^2)(\psi_2^2/A)(k_c - \pi/2\xi)\end{aligned}\quad (21)$$

with $\dot{\psi}_1 \equiv d\psi_1/d\theta$. Minimization of $f_0 = f_0^0 + c_0\rho_0 + f_1$ against θ shows that $\partial f_1/\partial\theta$, the torque on the effective magnetic moment μ , coming from the $k \neq 0$ spin quantum

fluctuations, becomes completely negligible compared with the other two torques coming from the classical and uniform $k = 0$ spin fluctuations. For large S , the resulting equation from the minimization is

$$\dot{\psi}_1 \left[\frac{2}{3 + \Delta} \frac{1}{(3 + \sqrt{6}/2)\psi_1 + \sqrt{6}(\Delta - 1)} \left(2\ddot{\psi}_1 - \frac{(3 + \sqrt{6}/2)\dot{\psi}_1^2}{(3 + \sqrt{6}/2)\psi_1 + \sqrt{6}(\Delta - 1)} \right) \right] - \frac{\psi_1}{4} = 0 \quad (22)$$

where we have introduced the important parameter within our model, $\Delta \equiv -(D_p/D_a)$. Notice that $\psi_1(\theta) = -f_0^c/[(S/2)(S/2 - 1)]$. A first solution of (22) is $\dot{\psi}_1 = 0$ (or $\partial f_0^c/\partial \theta = 0$), which is the classical one already discussed. But, most importantly, there is a new solution for $\theta(0)$, cast in the form

$$A^* \sin^4 \theta - B^* \sin^2 \theta + C^* = 0 \quad (23)$$

where the coefficients are functions of the parameter Δ alone, which means that $\theta(0)$ is controlled only by the ratio D_p/D_a , independently of the individual strengths of the CEF and spin S . Physically, these coefficients respond to the effect of the zero-point quantum spin fluctuations in the formation of $\theta(0)$, and explicitly are

$$\begin{aligned} A^*(\Delta) &\equiv 32(3 + \sqrt{6}/2)(3 + \Delta) + (3 + \sqrt{6}/2)^2(3 + \Delta)^2 \equiv 32c_2 + c_2' \\ B^*(\Delta) &\equiv 32(12 + 2\sqrt{6}\Delta) + 12(3 + \sqrt{6}/2)(3 + \Delta)(\Delta + \sqrt{6}/6) \equiv 32c_1 + c_1' \\ C^*(\Delta) &\equiv 32(6 + \sqrt{6}\Delta) + 4(3 + \sqrt{6}/2)(\sqrt{6}\Delta + 3 - \sqrt{6}/2) + 6(\Delta - 1)^2 \equiv 32c_3 + c_3'. \end{aligned} \quad (24)$$

The splitting into c_i and c_i' terms has no physical meaning, only being done for convenience when evaluating later the evolution of θ at finite temperatures. The new non-classical solution for the zero-point SR angle then becomes

$$\sin^2 \theta(0) = (B^*/2A^*)[1 \pm (1 - 4A^*C^*/B^{*2})^{1/2}]. \quad (25)$$

The important conclusion reached is that a stable minimum with $0 < \theta(0) < \pi/2$, given by equation (25), is possible besides the two classical solutions $\theta = 0$ and $\theta = \pi/2$, and, most importantly, assuming only *second-order* competing CEF interactions. Essentially responsible is the $k = 0$ frozen-in spin fluctuation, represented by c_0 (see equations (9) and (10)). Quantum fluctuations with $k \neq 0$ seem to have little effect on the canting, although probably reducing the marginal value of $\sin^2 \theta(0)$ obtained for $\Delta = 0$, inasmuch as $f_1 < 0$ (see (23)) further reduces the free energy.

It is now quite important to determine under which conditions 0 K spin canting can occur. In section 2.3 we assumed that the SR was in plane (c, a) , i.e. for $\psi = 0$. For the ground state this situation corresponds, within the classical approximation, to $D_p > 0$. Now, it is easily shown that $\partial^2 f_0/\partial \theta^2 > 0$ (i.e. minimum for f_0) is fulfilled, and SR at 0 K is possible for the situations summarized in table 1. The Δ axis was explored in the wide range $-20 \leq \Delta \leq 20$, and the calculated variation, from equation (25), of $\sin^2 \theta(0)$ in the intervals where a solution exists is shown in figures 1(a)-(c). Here $|\Delta| = 3$ is the threshold value, within the classical approximation, for the only allowed Δ -driven first-order SR transition from $\theta = 0$ to $\theta = \pi/2$ (see figure 1(a)). Notice the existence of such a first-order transition also for the non-classical solution (figures 1(b) and (c)). Besides, there appears a gap between $\Delta = -2.45$ and $+2.87$, where 0 K uniform spin canting is forbidden.

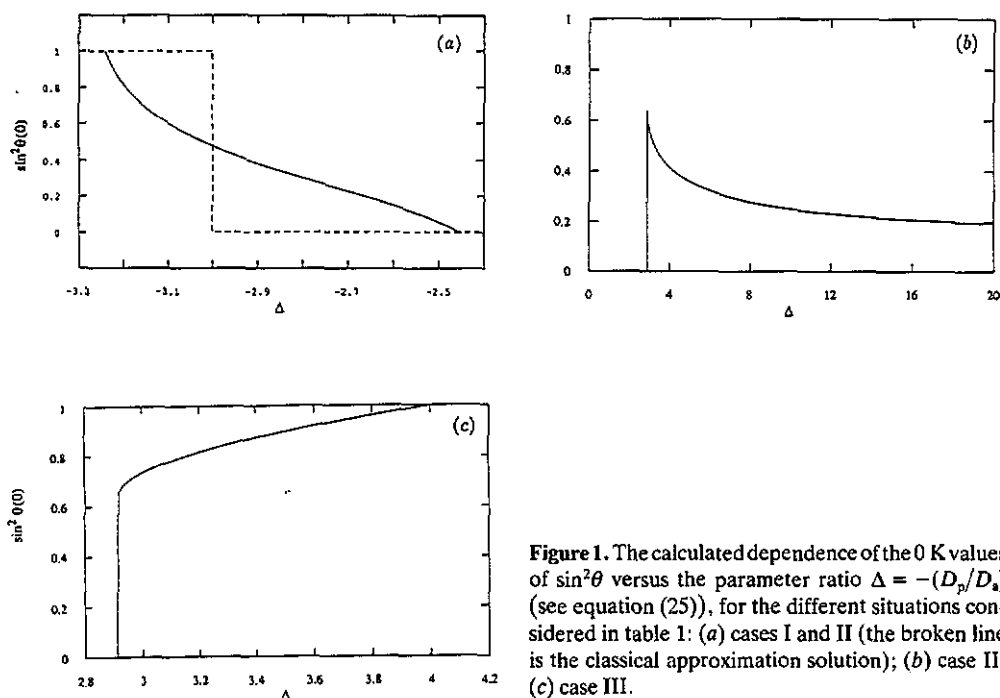


Figure 1. The calculated dependence of the 0 K values of $\sin^2\theta$ versus the parameter ratio $\Delta = -(D_p/D_a)$ (see equation (25)), for the different situations considered in table 1: (a) cases I and II (the broken line is the classical approximation solution); (b) case II; (c) case III.

Table 1. Situations where a spin reorientation process at 0 K happens within the (c, a) plane (i.e. for basal plane crystal field (CEF) parameter $D_p > 0$), in terms of the sign of D_a (axial CEF parameter) and of the ratio $\Delta = -(D_p/D_a)$, with indication of the sign of the square root, $\pm[1 - (4A^*C^*)/B^{*2}]^{1/2}$ in equation (25), giving $\sin^2\theta(0)$.

Situation	D_a	Δ	Sign of the square root	Possibility of SR
I	+	$\Delta < -3$	-	Yes
II	-	$\Delta > -3$	-	Yes
III	+	$\Delta > -3$	+	Yes
IV	-	$\Delta < -3$	+	No

3.2. Variation of the SR angle with temperature, SR temperature and critical behaviour

At finite temperatures the minimization of the free energy F given by (15) immediately gives rise to

$$\frac{\partial F_0}{\partial \theta} + S\psi_1 \sum_k \frac{1}{\exp(\beta\tilde{\epsilon}_k) - 1} \frac{\sqrt{6}B_k - 3A_k}{2\tilde{\epsilon}_k} = 0 \quad (26)$$

which again possesses the classical solution $\psi_1 = 0$. We will now consider two extreme relevant temperature limits for the resolution of equation (26):

3.2.1. *High temperatures ($T \ll T_{SR}$) and critical scaling.* For high enough temperatures, i.e. $\beta \bar{\epsilon}_k \ll 1$, and in the large exchange limit, i.e. when $B_k \ll A_k$, equation (26) becomes

$$\frac{\partial F_k(T)}{\partial \theta} \approx 3 k_B T S \sum_k \frac{1}{A_k} \quad (27)$$

where the summation can again be evaluated in the continuous- k integral approximation (see equation (19)). Then for $k_c \xi \gg 1$, from (27), an equation identical to the 0 K one (22) results, removing the factor ψ_1 and adding to the left-hand side a term of the form

$$\left(\frac{3}{S}\right) \left(\frac{k_B T}{A}\right) \left(\frac{v_a}{2\pi^2}\right) k_c.$$

Therefore the resulting equation is the same as (23), but for substituting the 0 K coefficients $c'_i(0)$ in (24) by $c'_i(T) = c'_i(0)(1 - \alpha T)$, with

$$\alpha = (6/\pi^2 S)(v_a k_c^3) (k_B/Ak_c^2) = (6\pi/S)(Ak_c^2/k_B)^{-1} \quad (28)$$

where $v_a = a^3$ for an sc lattice. Physically, α^{-1} roughly represents the temperature required to excite the highest-energy, $\epsilon(k_c) = Ak_c^2$, magnons. Notice also that $\alpha T \approx 6\pi(k_B T/Ak_c^2)$ represents the importance of the available lattice thermal energy compared with $\epsilon(k_c)$, and in this way α alone determines the temperature dependence of the SR angle. In fact, in the same way as equation (23) was derived from (22), it is now possible to deduce from the $T \neq 0$ version of (23) the temperature dependence of θ , in the high-temperature limit (i.e. for $\beta \bar{\epsilon}(k_c) \ll 1$), i.e.

$$\sin^2 \theta(T) = \frac{B^* - \alpha c'_1 T}{2(A^* - \alpha c'_2 T)} \left[1 - \left(1 - \frac{4(A^* - \alpha c'_2 T)(C^* - \alpha c'_3 T)}{(B^* - \alpha c'_1 T)^2} \right)^{1/2} \right] \quad (29)$$

a result that constitutes another goal of our model.

For temperatures close to T_{SR} , a number of series expansions within (29) give, for small θ , the result

$$\theta \approx A_- t^{1/2} \quad (30)$$

where t is the reduced temperature, $t = (T_{SR} - T)/T_{SR}$, and the amplitude $A_- = (C^*/B^*)^{1/2}$. Besides, the SR temperature T_{SR} becomes

$$T_{SR} \approx (1/\alpha) A^* C^* / (C^* c'_2 - A^* c'_3). \quad (31)$$

Some comments about these two important results are worthwhile. Equation (30) shows that θ suffers critical fluctuations near T_{SR} , as it should do, and scales with a critical exponent $\beta = 1/2$. Therefore the sw approximation gives quantum-mechanical *microscopic support* to the phenomenological MF-Ginzburg-Landau theory for SR transitions (del Moral 1987). It is quite revealing to notice the influence of the exchange strength J , through α , on T_{SR} , and therefore the prediction of a proportionality between T_{SR} and the Curie temperature T_C , a fact actually observed in the $(Er_x Dy_{1-x})_2 Fe_{14} B$ intermetallics (Ibarra *et al* 1989a). The variation of the scaled SR temperature, αT_{SR} , with Δ , for the same intervals considered in figures 1(a)-(c), are respectively shown in figures 2(a)-(c).

3.2.2. *Low-temperature $T^{3/2}$ Bloch law.* At temperatures near to 0 K, we have to compute fully the second term in (26). Now, in the large exchange limit,

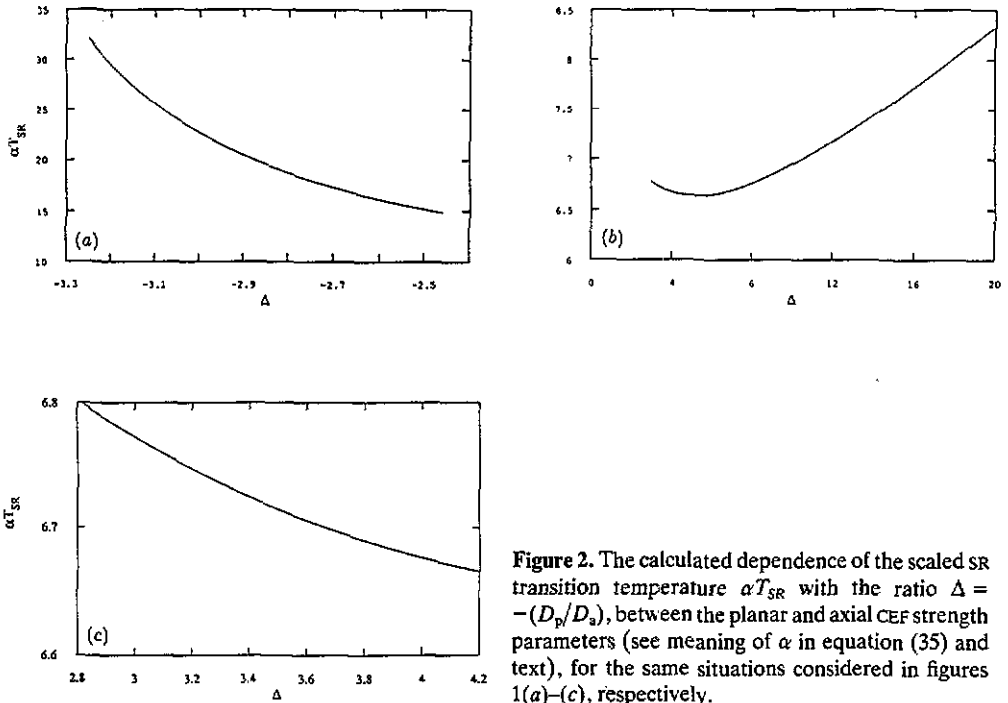


Figure 2. The calculated dependence of the scaled SR transition temperature αT_{SR} with the ratio $\Delta = -(D_p/D_a)$, between the planar and axial CEF strength parameters (see meaning of α in equation (35) and text), for the same situations considered in figures 1(a)–(c), respectively.

$(\sqrt{6} B_k - 3A_k)/2A_k = -3/2$ and $\bar{\epsilon}_k \approx A_k = Ak^2$ (i.e. the CEF magnon gap can be neglected) and therefore we obtain from (26)

$$\frac{\partial f_0}{\partial \theta} + \frac{3S\psi_1}{2} \frac{v_a}{8\pi^3} \int_0^{k_c} \frac{d^3k}{\exp(\beta Ak^2) - 1} = 0. \tag{32}$$

At low enough temperatures $Ak_c^2 \gg k_B T$ and therefore the upper limit of the integral can be taken as ∞ ; then, from (32) we again obtain an equation identical to (22) but with the additional temperature-dependent term $(3\kappa/S)(v_a/8\pi^2)(k_B T/A)^{3/2}$ on the left-hand side, with $\kappa = \Gamma(3/2)\zeta(3/2; 1)$. Then, the equation giving $\sin^2\theta(T)$ is again identical to (29) but with $c'_i(0)$ now substituted by

$$c'_i(T) = c'_i(0)(1 - \alpha' T^{3/2})$$

and where

$$\alpha' = (3\kappa/2\pi^{1/2}S)(k_B/Ak_c^2)^{3/2}. \tag{32a}$$

A transformation entirely similar to the one performed with (29), taking into account (31), gives

$$\sin^2\theta(T) = (C^*/B^*)[1 - (\alpha'/\alpha T_{SR})T^{3/2}] \tag{33}$$

which indeed constitutes the equivalent to the magnetization $T^{3/2}$ Bloch law for the order parameter $\theta(T)$, a result that shows the relevance of spin-wave excitations at low temperatures to explain properly the θ angle reduction. In other words, the classical phenomenological approach based on the free energy $K_1(T)\sin^2\theta + K_2(T)\sin^4\theta$ is unable to conclude with such a Bloch-like law, as happens with the mean-field theory with

respect to the magnetization reduction. From (33) it is clear that only for temperatures $T \leq (\alpha T_{\text{SR}}/\alpha')^{3/2}$ is the Bloch law fulfilled.

For $T = 0$ K, from (31) and (33), and in the large exchange limit, we obtain the relation

$$\sin^2\theta(0) = (-\alpha c'_3/B^*)T_{\text{SR}}. \quad (34)$$

This proportionality was to be expected, because it represents the overall overcoming of the 0 K anisotropy energy by the thermal critical fluctuations at T_{SR} . But, the usefulness of equation (34) is that it provides the effective 0 K anisotropy constant (per ion and in kelvin)

$$K_1(0) = [-\alpha c'_3(\Delta)/B^*(\Delta)]^{-1} \quad (35)$$

which depends on Δ , but also increases with the unperturbed magnon energy, Ak_c^2 . In particular, $\theta(0)$ is expected to decrease with the Curie temperature, T_c (see section 4), as is actually observed in the $(\text{Er}_x\text{Dy}_{1-x})_2\text{Fe}_{14}\text{B}$ series (Ibarra *et al* 1989a).

4. Comparison with available experimental results on SR transitions in hard RE intermetallics; discussion and general conclusion

Although we should stress strongly that the present work is basically intended as a theoretical one, which aims to show how SR processes can be explained as due to spin-wave fluctuations, being the result of competing axial-planar *second-order* CEF alone, a comparison of the main findings achieved with available experimental results is indeed highly desirable, and in this sense some qualitative verifications of the model have already been pointed out in section 3.2. Unfortunately the number of experimental results on SR processes in rare-earth intermetallics is scarce and incomplete (see references at the beginning of the paper and references therein), which makes such a comparison necessarily limited, and therefore more experimental work is clearly needed in the future in order to check the present model further.

The first physically relevant result obtained is the prediction of a $T^{3/2}$ Bloch law for the variation of $\sin^2\theta$ at low temperatures (equation (33)), which provides the ratio

$$(\alpha/\alpha')^2 = (4\pi^3/k)(Ak_c^2/k_B)$$

i.e. precisely the magnon energy $\varepsilon(k_c)$, at the average cubic BZ boundary. Therefore, in figures 3(a) and (b) we have plotted $\sin^2\theta(T)$ against $T^{3/2}$ for some intermetallics where we possess enough and precise $\theta(T)$ data points at low temperatures, a difficult measurement in itself (del Moral *et al* 1988, Ibarra *et al* 1988a, Joven *et al* 1990). The plots are clearly linear at low enough temperatures, indicating how the SR process is really controlled by sw excitations. The values of $\varepsilon(k_c)$ so obtained are collected in table 2. Inelastic neutron scattering values determined for $\varepsilon(k_c)$ would be highly valuable for comparison, but such information is, to our knowledge, lacking.

Another direct test of the present model is through the relation (34) between T_{SR} and $\sin^2\theta(0)$. In figure 4, we have plotted the measured T_{SR} vs $\sin^2\theta(0)$ values, for the different series of pseudobinary and pseudoternary hard intermetallics considered here; the plots are clearly linear as predicted. Notice that equation (34) agrees with experiment even for $\theta(0) = \pi/2$, when the crossing from axial to planar structures is known to be through two transitions, one from axial to conical (at T_{SR}) and the other from conical to planar (at T'_{SR}) (see e.g. Ibarra *et al* 1989a, Moze *et al* 1990a). However, in this case the

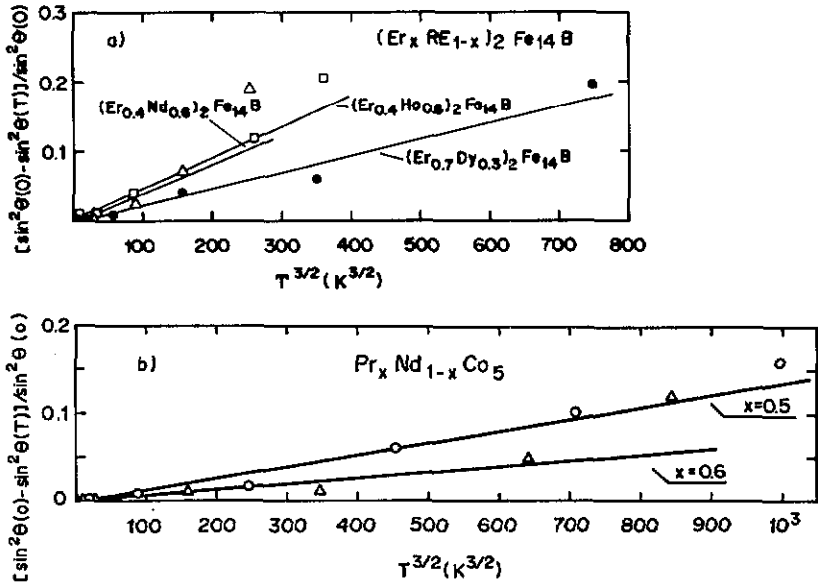


Figure 3. Plots of $[\sin^2\theta(0) - \sin^2\theta(T)]/\sin^2\theta(0)$ against $T^{3/2}$ (Bloch-like law) for: (a) $(Er_xRE_{1-x})_2Fe_{14}B$ (RE = Nd, Ho, Dy) series; (b) $Pr_xNd_{1-x}Co_5$ series. From the slopes of the straight lines, the magnon energies $\varepsilon(k_c)$ (see values in table 2), at the average cubic Brillouin zone boundary points $(\pi/a, 0, 0)$ and equivalent, are determined (see equation (33) and section 4 for details).

Table 2. The magnon energies $\varepsilon(k_c)$, at the average cubic Brillouin zone boundary $(\pi/a, 0, 0)$ and equivalent points, for some pseudobinary and pseudoternary hard intermetallic compounds, as obtained from the slopes of the straight lines of figures 3(a) and (b), and the use of equation (33). Also quoted are the second-order effective anisotropy constants at 0 K, $K_1(0)$, for each series, as obtained from the slopes of the straight lines of figure 4 (experiment), and the calculated values from equation (35) (theory) (see text for details).

System	Composition x (at. %)	$\varepsilon(k_c)$ (meV)	$K_1(0)$ (K/unit cell)	
			Exp.	Theor.
$Pr_xNd_{1-x}Co_5$	0.5	55	96	—
	0.6	267		
$(Er_xDy_{1-x})_2Fe_{14}B$	0.7	31	232	253
$(Er_xNd_{1-x})_2Fe_{14}B$	0.4	9	250	239
$(Er_xHo_{1-x})_2Fe_{14}B$	0.4	16	276	242

agreement is only for the lowest-concentration (x) compounds suffering the double transition. The values of the effective anisotropy constant $K_1(0)$, obtained from the slopes of the straight lines in figure 4, are quoted in table 2, for each series of compounds. A good test of the model is through the comparison of the experimental $K_1(0)$ values with the theoretical ones provided by equation (35). Clearly the value of the effective

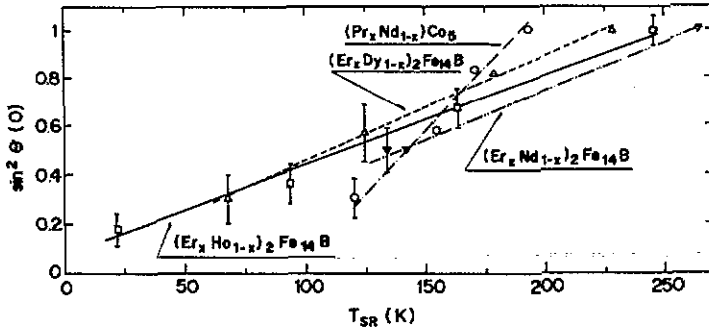


Figure 4. Plots of the measured $\sin^2\theta(0)$, at 0 K, versus the measured SR transition temperatures T_{SR} , for different series of RE pseudobinary and pseudoternary hard intermetallics. The slopes of the straight lines provide the values of the 0 K anisotropy constants quoted in table 2 under 'experiment'. The error bars correspond to the maximum precision achieved in the cone angle measurements.

spin S is needed; a determination of S from magnetization data, our only available results (Coe *et al* 1985, Wallace 1973), is not possible because S and the Landé factor g are combined in the magnetic moment per unit cell, $\mu = g\mu_B S$. However, we can make use of the MF theory, where the exchange constant is given by $J = 3k_B T_C / 8zS(S+1)$, and take for the sw stiffness constant, $A = \frac{1}{2}JSa^2$. Then for large S , $S(S+1) \approx S^2$, and for an sc lattice, we obtain from equation (28) that $\alpha = 61.1/T_C$ (T_C values can be found for the $(Er_x RE_{1-x})_2 Fe_{14} B$ and $Pr_x Nd_{1-x} Co_5$ compounds in Coe *et al* (1985) and Wallace (1973) respectively).

The determination of Δ , needed to evaluate the function $c_3^2(\Delta)/B^*(\Delta)$, which appears in (35), was made from the experimental values of αT_{SR} (see values of T_{SR} in references mentioned at the beginning of section 1) and the use of graphs in figures 2(a)–(c). This method of determination of Δ was preferred to the alternative one based on the $\theta(0)$ experimental values and the use of the graphs in figures 1(a)–(c), owing to the large uncertainty in the $\theta(0)$ values, of $\approx \pm 5^\circ$ (del Moral *et al* 1989). The calculated values of $K_1(0)$ are shown in table 2 for the $(Er_x RE_{1-x})_2 Fe_{14} B$ series of compounds, the agreement with experiment being quite good, if one considers all the simplifications made in the model. In the case of the pseudobinaries $(Pr_x Nd_{1-x}) Co_5$, there was no such agreement. The reason could be found in the fact that the Pr^{3+} and Nd^{3+} ions have α_j coefficients of the same sign and then the competing anisotropies are between the RE and Co sublattices, ferromagnetically coupled (Algarabel *et al* 1988, Ibarra *et al* 1991, Wallace 1973). Whether the interaction of the Co atoms with the CEF can be treated within the single-ion scheme is doubtful (Ibarra *et al* 1989a, 1991), and could make our model only applicable in a broad sense, considering that there are other features where it behaves correctly.

Now, a comparison with directly measured values of $K_1(0)$ is unfortunately not possible, owing to the lack of measurements at 4.2 K (the experimental data go down to 77 K only) and besides K_1 appears in linear combinations with K_2 and K_3 , casting the measured equivalent axial and planar anisotropy fields (Algarabel *et al* 1989b, Ibarra *et al* 1989a, Marusi *et al* 1990).

A last comparison made with experiment is regarding the predicted critical scaling of θ near T_{SR} (equation (30)). Such scaling is well fulfilled by the present series of

Table 3. Critical exponent β for the order parameter SR angle θ for some series of pseudobinary and pseudoternary magnetically hard rare-earth intermetallics.

System	Composition x (at.%)	β	References
$\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$	0.80	0.60 ± 0.10	Moze <i>et al</i> (1990a) ^a
	0.60	0.50 ± 0.10	
	0.50	0.70 ± 0.10	
	0.40	0.50 ± 0.10	
$(\text{Er}_x\text{Dy}_{1-x})_2\text{Fe}_{14}\text{B}$	0.70	0.60 ± 0.20	Ibarra <i>et al</i> (1989a)
	0.80	0.50 ± 0.20	del Moral <i>et al</i> (1989)
	0.90	0.80 ± 0.20	Ibarra and del Moral (1990)
$(\text{Er}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$	0.40	0.80 ± 0.20	Ibarra <i>et al</i> (1988a) del Moral <i>et al</i> (1989)
$(\text{Er}_x\text{Ho}_{1-x})_2\text{Fe}_{14}\text{B}$	0.40	0.70 ± 0.15	Marquina <i>et al</i> (1989)

^a The thermal dependences of the SR angles, from where the β exponents were obtained, are quoted in this reference and the ones below.

intermetallics, actually revealing the existence of critical fluctuations of the θ order parameter. In table 3, we have collected the values of the determined critical exponent β . The measured values are, in general, higher than the predicted one of 0.5, but much higher than the more reliable, in principle, renormalization group (RG) calculation value, $\beta = 0.367$. The reason why our sw (or the MF) approximation β value is closer to the experiment than the RG one is not yet well understood, and probably needs further theoretical work. Suffice it to say that the determination of θ , made through the measurement of the magnetization perpendicular to the applied field (Joven *et al* 1990), makes it quite difficult very near to T_{SR} , where the θ values are indeed quite small. This could introduce large systematic errors in the β values. On the other hand, although the sw approximation is a low-temperature one, we are far from T_{C} and this circumstance could explain the success in the prediction of a critical scaling for θ at $T \leq T_{\text{SR}}$.

As a general conclusion, our present model only claims to open some avenues to deal with the mechanisms underlying the complex and not yet well understood spin reorientation phenomenon in ferromagnetic (or ferrimagnetic) systems formed by two RE ionic species submitted to second-order competing crystal fields. Besides, more experimental work is clearly needed to test fully our model predictions and main hypothesis made of assuming an average spin per unit cell in $\text{RE}'_u\text{RE}_{1-u}\text{TM}_v$ intermetallics; and, most importantly, whether the quantum spin fluctuations are fully responsible for the 0 K frozen-in magnetization canting.

Acknowledgments

I acknowledge very useful discussions with Dr M R Ibarra, especially about section 4. The financial support of the Spanish CICYT under grants MAT88/552 and MAT 88/689 of the DGICYT under grant PB90-1014 and of the Project BREU-68-C of the EEC are greatly acknowledged. I am grateful to J Cullen (NSWC, Washington) for many

early discussions about spin-wave theory. The help of the undergraduate student L Morellón with the numerical computations is also appreciated.

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