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Magnetic anisotropy terms in [110] MBE-grown REFe₂ films involving the strain term ϵ_{xy}

G J Bowden¹, P A J de Groot¹, B D Rainford¹, K Wang¹, K N Martin¹,
J P Zimmermann² and H Fangohr²

¹ School of Physics and Astronomy, University of Southampton, Southampton SO17 1BJ, UK

² Computational Engineering and Design Group, School of Engineering Sciences, University of Southampton, Southampton SO17 1BJ, UK

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Abstract

The magnetic anisotropy parameters in [110] MBE-grown films of REFe₂ (RE, rare earth) compounds are not the same as those in the bulk. This is due to the presence of a shear strain ϵ_{xy} , frozen-in during crystal growth. In this paper, magnetic anisotropy parameters for [110] MBE-grown REFe₂ films, that directly involve the shear strain ϵ_{xy} , are presented and discussed. In addition to the usual first-order Callen and Callen term \tilde{K}'_2 , there are nine second-order terms, six of which involve cross-terms between ϵ_{xy} and the cubic crystal field terms B_4 and B_6 . Two of the second-order cross-terms are identified as being important: $\tilde{K}''_{242}(T)$ and $\tilde{K}''_{264}(T)$. Of these, the rank-two term $\tilde{K}''_{242}(T)$ dominates over a large temperature range. It has the same angular dependence as the first-order term \tilde{K}'_2 , but with a more rapid temperature dependence. The correction at $T = 0$ K for TbFe₂, DyFe₂, HoFe₂, ErFe₂ and TmFe₂, amounts to $\sim +9.2\%$, -13.9% , -11.6% , $+14.3\%$, and 27.1% , respectively. Similar comments are made concerning the rank-four $\tilde{K}''_{264}(T)$ term.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Because of their numerous applications in magnetostriction, the REFe₂ (RE, rare earth) intermetallic compounds have a secure place in the annals of magnetism (Clark 1979, Engdahl 1999). Recently, interest in these compounds has been revived because of possible applications in the field of nano-magnetism. Specifically, epitaxial single crystal films of the REFe₂ compounds (~ 400 nm) have been grown by molecular beam epitaxy (MBE) on sapphire substrates (Odero *et al* 1996). These films exhibit a wide range of phenomena, from model magnetic exchange springs (Sawicki *et al* 2000a, 2000b, Dumesnil *et al* 2000) to giant magnetoresistance (Gordeev *et al* 2001).

Concomitantly, there has been renewed interest in the values of the magnetic anisotropy parameters, as input parameters for magnetic modelling of the epitaxial films (Mougin *et al* 2000, Bowden *et al* 2003). Long ago, estimates for the bulk REFe₂ compounds were given by Atzmony and Dariel (1976), who showed that in addition to the phenomenological K_1 and K_2 parameters for cubic symmetry, it is necessary to include a higher-order term K_3 . In addition, these authors found that some of the calculated K_1 and K_2 parameters changed sign as a function of temperature, a feature which could not be explained in the context of the Callen and Callen model (Callen and Callen 1965, 1966, Callen and Shtrikman 1965). This problem has recently been re-visited by Martin *et al* (2006), using an extended version of the Callen and Callen model. They were able to provide (i) an explanation for the change in sign of K_1 in HoFe₂ and K_2 in DyFe₂ and (ii) elucidate the origins of the K_3 , and higher-order terms. In particular, explicit expressions for K_1 – K_5 were given in terms of the RE-ion crystal field parameters B_4 and B_6 and their products B_4^2 , B_4B_6 and B_6^2 . The latter cross-terms dominate the higher-order K_3 – K_5 parameters, with K_3 and K_4 being of the same order of magnitude as K_1 and K_2 .

However, there are important difference between MBE-grown films and their free-standing equivalents. Mougin *et al* (2000) have shown that MBE-grown films exhibit a strain term, frozen-in during crystal growth. This term, which has the same form as the magneto-elastic Hamiltonian, is usually written:

$$H_{\text{ME}}(\varepsilon_{xy}) = b_2\varepsilon_{xy}\alpha_x\alpha_y. \quad (1)$$

Here b_2 is the temperature dependent magneto-elastic constant, ε_{xy} is the shear strain ($\sim -0.55\%$) and α_x and α_y are the direction cosines with respect to the [100] and [010] cubic axes, respectively. In the past, it has been assumed that both the strain term and the anisotropy parameters K_1 – K_2 are simply additive (Mougin *et al* 2000). However, given that cross-terms of the form B_4^2 , B_4B_6 and B_6^2 are important in the bulk, it is possible that cross-terms of the form $(b_2\varepsilon_{xy})^2$, $b_2\varepsilon_{xy}B_4$ and $b_2\varepsilon_{xy}B_6$ are important in their strained equivalents. Indeed, the latter may play a role in determining the direction of easy magnetization. These questions are addressed in this paper.

2. Theory: magneto-crystalline magnetic anisotropy

A more complete description of the basic theory used in this paper has already been given by Martin *et al* (2006). However, for convenience, an abbreviated account is set out below.

In the free-standing REFe₂ inter-metallic compounds, the dominant anisotropy derives from the crystal field interaction at the RE ion, with the exceptions of Gd and Y. The Hamiltonian at the RE ion can be written:

$$H = H_{\text{EX}} + H_{\text{CF}} \quad (2)$$

where (i)

$$H_{\text{EX}} = 2(g_j - 1)\mu_B H_{\text{EX}}^{\text{Fe}} \mathbf{J} \cdot \mathbf{Z} = X \mathbf{J} \cdot \mathbf{Z} \quad (3)$$

and (ii)

$$H_{\text{CF}} = B_4[\mathbf{O}_{40} + 5\mathbf{O}_{44}^C] + B_6[\mathbf{O}_{60} - 215\mathbf{O}_{64}^C]. \quad (4)$$

(Cohen 1964, Bowden *et al* 1968, Atzmony and Dariel 1973, 1976). Here the exchange field at the RE site $H_{\text{EX}}^{\text{Fe}}$ is assumed to be isotropic, and to follow the temperature dependence of the Fe sub-lattice. At $T = 0$ K, $\mu_B H_{\text{EX}}^{\text{Fe}}/k = 100$ K (Atzmony and Dariel 1976).

For the purposes of this paper we choose not to use the operator equivalents \mathbf{O}_{40} , \mathbf{O}_{44}^C etc, (e.g. Abragam and Bleaney 1970, Hutchings 1964) but rather the set of tensor operators

\mathbf{T}_q^n given by Buckmaster *et al* (1972), Bowden and Hutchison (1986). The latter have clear advantages. First, it is possible to construct the entire tensor set starting from the basic building blocks $\mathbf{T}_0^1, \mathbf{T}_{-1}^1, \mathbf{T}_{+1}^1$. Second, the operators can be recast in terms of unit irreducible tensor operators $\hat{\mathbf{T}}_q^n$ where $\text{Tr}[\hat{\mathbf{T}}_q^n(\hat{\mathbf{T}}_{q'}^{n'})^*] = \delta_{nn'}\delta_{qq'}$. Third, formulae exist for recasting products of tensor operators $\hat{\mathbf{T}}_q^n\hat{\mathbf{T}}_{q'}^{n'}$ into single tensors $\hat{\mathbf{T}}_Q^N$ where $N \leq n + n'$ (Bowden and Hutchison 1986). Fourth, they obey well-known rotation laws (Edmonds 1957). Since tensor operator products arise in second-order perturbation theory (see below), they are the natural choice for the problem in hand. Within the Buckmaster formulation therefore, the crystal field takes the form:

$$H_{\text{CF}} = \tilde{B}_4 \left[\mathbf{T}_0^4 + \sqrt{\frac{5}{14}}(\mathbf{T}_4^4 + \mathbf{T}_{-4}^4) \right] + \tilde{B}_6 \left[\mathbf{T}_0^6 - \sqrt{\frac{7}{2}}(\mathbf{T}_4^6 + \mathbf{T}_{-4}^6) \right] \quad (5)$$

where

$$\tilde{B}_4 = 2\sqrt{70}B_4 \quad \text{and} \quad \tilde{B}_6 = 4\sqrt{231}B_6. \quad (6)$$

In the Callen and Callen model of magnetic anisotropy, and its extensions, the crystal field Hamiltonian H_{CF} is assumed to be small compared to the magnetic exchange H_{EX} . So the free energy of RE ion can be expanded in the form:

$$F = F_0 + F' + F'' + \dots \quad (7)$$

where $F_0 = F_{\text{EX}}$ is the free energy associated with the ‘dominant’ Fe–Fe magnetic exchange term, and $F' = \langle H_{\text{CF}} \rangle$ etc, where the expectation values are calculated using the Zeeman eigenvalues and functions of the magnetic exchange Hamiltonian H_{EX} . Here the use of a single (double) dash on any symbol signifies that it originates from first (second)-order perturbation theory, respectively.

In first order, for an arbitrary direction, we find:

$$F' = \langle H_{\text{CF}} \rangle = \sum_{n,m} \tilde{B}_m^n D_{0m}^n(\omega) \langle \mathbf{T}_0^n \rangle \quad (8)$$

where (i) the $D_{0n}^n(\omega)$ are the well-known rotation operators (Edmonds 1957) and (ii) ω is a shorthand notation for the Euler angles (α, β, γ) . For the bulk REFe₂ compounds therefore:

$$F' = \tilde{K}'_4(T) \left[Y_4^0(\theta, \phi) + \sqrt{\frac{5}{14}}(Y_4^4(\theta, \phi) + Y_4^{-4}(\theta, \phi)) \right] + \tilde{K}'_6(T) \left[Y_6^0(\theta, \phi) - \sqrt{\frac{7}{2}}(Y_6^4(\theta, \phi) + Y_6^{-4}(\theta, \phi)) \right] \quad (9)$$

where (i) we have set (β, γ) equal to (θ, ϕ) , respectively, and (ii) the anisotropy constants are given by:

$$\tilde{K}'_4(T) = \sqrt{\frac{4\pi}{9}} \tilde{B}_0^4 \langle \mathbf{T}_0^4 \rangle \quad (10)$$

$$\tilde{K}'_6(T) = \sqrt{\frac{4\pi}{13}} \tilde{B}_0^6 \langle \mathbf{T}_0^6 \rangle.$$

Equations (9) and (10) are the principal results of the Callen and Callen model of anisotropy (Callen and Callen 1965, 1966) for cubic compounds. In practice, the anisotropy parameters $\tilde{K}'_4(T), \tilde{K}'_6(T)$ decrease monotonically with increasing temperature, and do not change sign.

In second order (Bowden 1977, Martin *et al* 2006), we obtain:

$$F'' = -\frac{1}{2}\beta \left\{ \sum_{n,m} \sum_{n',m'} \tilde{B}_m^n \tilde{B}_{m'}^{n'} \sum_{N,M} (2N+1) \begin{pmatrix} n & n' & N \\ m & m' & M \end{pmatrix} D_{0M}^N(\omega)^* \right. \\ \left. \times \sum_q \begin{pmatrix} n & n' & N \\ q & -q & 0 \end{pmatrix} \langle \mathbf{T}_q^n; \mathbf{T}_{-q}^{n'} \rangle \right\} \quad (11)$$

where (i) $\langle \mathbf{T}_q^n : \mathbf{T}_{q'}^{n'} \rangle$ is a shorthand notation for:

$$\begin{aligned} \langle \mathbf{T}_q^n : \mathbf{T}_{q'}^{n'} \rangle &= \left\langle \mathbf{T}_q^n \int \mathbf{T}_{q'}^{n'} \right\rangle - \langle \mathbf{T}_q^n \rangle \langle \mathbf{T}_{q'}^{n'} \rangle \\ &= \text{Tr} \left[\mathbf{T}_q^n \int_0^1 ds \rho^{1-s} \mathbf{T}_{q'}^{n'} \rho^s \right] - \langle \mathbf{T}_q^n \rangle \langle \mathbf{T}_{q'}^{n'} \rangle. \end{aligned} \quad (12)$$

(ii) $\beta = 1/kT$ and (iii) ρ is the density matrix, calculated using the magnetic exchange Hamiltonian alone. In practice, it is advantageous to recast equation (11) in the form:

$$F'' = -\frac{1}{2}\beta \left\{ \sum_{n,m} \sum_{n',m'} \tilde{B}_m^n \tilde{B}_{m'}^{n'} \sum_{N,M} (2N+1) \begin{pmatrix} n & n' & N \\ m & m' & M \end{pmatrix} D_{0M}^N(\omega)^* \alpha_{nn'N}(T) \right\} \quad (13)$$

where the temperature dependent $\alpha_{n,n',N}(T)$ coefficients are given by:

$$\alpha_{nn'N}(T) = \sum_q \begin{pmatrix} n & n' & N \\ q & -q & 0 \end{pmatrix} \langle \mathbf{T}_q^n : \mathbf{T}_{-q}^{n'} \rangle. \quad (14)$$

Note that the behaviour of the second-order terms as a function of temperature is governed by two terms; the $\alpha_{nn'N}(T)$ coefficients together with the global $\beta = 1/kT$ term appearing outside all of the terms in equation (13). In practice, the $\beta\alpha_{nn'N}(T)$ coefficients converge to zero as $T \rightarrow \infty$, and to finite values as $T \rightarrow 0$ K. More details concerning the properties of $\alpha_{nn'N}(T)$ etc, can be found in appendix A of Martin *et al* (2006).

From equation (13) it is clear that second-order perturbation theory will lead to terms which are proportional to $(\tilde{B}_4)^2$, $(\tilde{B}_6)^2$ and cross-terms $\tilde{B}_4\tilde{B}_6$. In particular, the $(\tilde{B}_4)^2$, $\tilde{B}_4\tilde{B}_6$ and $(\tilde{B}_6)^2$ terms give rise to spherical harmonics with rank ≤ 8 , ≤ 10 and ≤ 12 , respectively. To illustrate the terminology used in this paper, consider the second-order term involving $(\tilde{B}_4)^2$. We obtain:

$$F''(4,4) = \tilde{K}_{440}''(T)Y_0^0(\theta, \phi) + \tilde{K}_{444}''(T)Y_4^C(\theta, \phi) + \tilde{K}_{446}''(T)Y_6^C(\theta, \phi) + \tilde{K}_{448}''(T)Y_8^C(\theta, \phi) \quad (15)$$

where (i) the combinations of spherical harmonics $Y_N^C(\theta, \phi)$ with cubic symmetry are listed in table 3 of Martin *et al* (2006) and (ii) the anisotropy coefficients $\tilde{K}_{nn'N}''(T)$ are given by

$$\begin{aligned} \tilde{K}_{440}''(T) &= -\frac{1}{2}\beta \left[+\frac{4\sqrt{4\pi}}{7}(\tilde{B}_4)^2\alpha_{440}(T) \right] \\ \tilde{K}_{444}''(T) &= -\frac{1}{2}\beta \left[+\sqrt{\frac{4\pi}{9}}\frac{6\sqrt{14}}{\sqrt{11.13}}(\tilde{B}_4)^2\alpha_{444}(T) \right] \\ \tilde{K}_{446}''(T) &= -\frac{1}{2}\beta \left[-\sqrt{\frac{4\pi}{13}}\frac{4}{7}\sqrt{\frac{65}{11}}(\tilde{B}_4)^2\alpha_{446}(T) \right] \\ \tilde{K}_{448}''(T) &= -\frac{1}{2}\beta \left[+\sqrt{\frac{4\pi}{17}}\frac{3}{7}\sqrt{\frac{5.11.17}{26}}(\tilde{B}_4)^2\alpha_{448}(T) \right]. \end{aligned} \quad (16)$$

Note that (i) the fourth-order crystal field terms ($n = n' = 4$) can give rise to a magnetic anisotropy term with rank $N = 8$ and (ii) the suffices $nn'N$ for the $\tilde{K}_{nn'N}''(T)$ and $\alpha_{nn'N}(T)$ coefficients are the same.

Proceeding in this fashion, explanations have been provided for (i) the change in sign of K_1 in HoFe₂ and K_2 in DyFe₂, (ii) the origin of the K_3 and (iii) the need for even higher-order terms (Martin *et al* 2006). In particular, explicit expressions are given for phenomenological magnetic

anisotropy constants K_1 – K_5 , in terms of the RE-ion crystal field parameters B_4 and B_6 and their products B_4^2 , B_4B_6 and B_6^2 . The latter dominate the higher-order K_3 – K_5 parameters, with K_3 and K_4 being of the same order of magnitude as K_1 and K_2 .

But, as noted earlier, there are important differences between the bulk and MBE-grown REFe₂ thin films. In MBE-grown films, there is a strain term (ε_{xy}), in addition to the usual cubic crystal Hamiltonian H_{CF} . So cross-terms of the form $(b_2\varepsilon_{xy})^2$, $b_2\varepsilon_{xy}B_4$, and $b_2\varepsilon_{xy}B_6$ will occur.

3. The magneto-elastic Hamiltonian

First, we establish a general operator form of the magneto-elastic Hamiltonian, which is usually written in the form:

$$H_{ME} = b_2\varepsilon_{xx}\alpha_x^2 + b_2\varepsilon_{yy}\alpha_y^2 + b_2\varepsilon_{zz}\alpha_z^2 + b_2\varepsilon_{xy}\alpha_x\alpha_y + b_2\varepsilon_{xz}\alpha_x\alpha_z + b_2\varepsilon_{yz}\alpha_y\alpha_z. \quad (17)$$

Using standard methods (Hutchings 1964, Abragam and Bleaney 1970) we obtain:

$$H_{ME} = B_{xx}\mathbf{J}_x^2 + B_{yy}\mathbf{J}_y^2 + B_{zz}\mathbf{J}_z^2 + B_{xy}\frac{1}{2}(\mathbf{J}_x\mathbf{J}_y + \mathbf{J}_y\mathbf{J}_x) + B_{xz}\frac{1}{2}(\mathbf{J}_x\mathbf{J}_z + \mathbf{J}_z\mathbf{J}_x) \\ + B_{yz}\frac{1}{2}(\mathbf{J}_y\mathbf{J}_z + \mathbf{J}_z\mathbf{J}_y) \quad (18)$$

where:

$$B_{ij} = \left[\frac{b_2\varepsilon_{ij}}{J(J+1)} \right]. \quad (19)$$

However, in place of the spin operators \mathbf{J}_α , we choose to use the tensor operators \mathbf{T}_q^k . Thus equation (18) is transformed to:

$$H_{ME} = B_{xx} \left\{ \frac{1}{2} [\mathbf{T}_2^2 + \mathbf{T}_{-2}^2] - \frac{1}{2} \left[\sqrt{\frac{2}{3}} \mathbf{T}_0^2 - \frac{2}{3} J(J+1) \right] \right\} \\ + B_{yy} \left\{ -\frac{1}{2} [\mathbf{T}_2^2 + \mathbf{T}_{-2}^2] + \frac{1}{2} \left[\sqrt{\frac{2}{3}} \mathbf{T}_0^2 - \frac{2}{3} J(J+1) \right] \right\} \\ + B_{zz} \left\{ \left[\sqrt{\frac{2}{3}} \mathbf{T}_0^2 - \frac{1}{3} J(J+1) \right] \right\} \\ - B_{xy} \frac{i}{2} [\mathbf{T}_2^2 - \mathbf{T}_{-2}^2] - B_{xz} [\mathbf{T}_1^2 - \mathbf{T}_{-1}^2] + B_{yz} i [\mathbf{T}_1^2 + \mathbf{T}_{-1}^2]. \quad (20)$$

In the [110] MBE films in question only the ε_{xy} term is important. So:

$$H_{ME}(\varepsilon_{xy}) = -B_{xy} \frac{i}{2} [\mathbf{T}_2^2 - \mathbf{T}_{-2}^2]. \quad (21)$$

We are now in a position to calculate the magnetic anisotropy parameters which involve the strain term.

4. Magnetic anisotropy terms involving ε_{xy}

The first-order change to the free energy due to the magneto-elastic term is given by:

$$F' = \langle H_{ME}(\varepsilon_{xy}) \rangle = \sum_{n,m} \tilde{B}_m^n D_{0m}^n(\omega) \langle \mathbf{T}_0^n \rangle = \tilde{K}'_2 i [Y_2^{-2}(\theta, \phi) - Y_2^2(\theta, \phi)] \quad (22)$$

where:

$$\tilde{K}'_2 = \sqrt{\frac{\pi}{5}} B_{xy} \langle \mathbf{T}_0^2 \rangle_{EX}. \quad (23)$$

Note that the combination of spherical harmonics appearing in equation (22) is real:

$$i[Y_2^{-2}(\theta, \phi) - Y_2^2(\theta, \phi)] = \frac{1}{2}\sqrt{\frac{15}{2\pi}} \sin^2 \theta \sin 2\phi. \quad (24)$$

In practice, \tilde{K}'_2 decreases monotonically with increasing temperature, in accord with expectations based on the Callen and Callen model. However, as we shall see, this is not necessarily the case for some of the second-order terms.

In second-order perturbation theory, *beating* occurs between $H_{\text{ME}}(\varepsilon_{xy})$ and the fourth- and sixth-order cubic crystal field terms H_{CF} of equation (5). This gives rise to additional anisotropy terms, which do not possess cubic symmetry. There are three contributions, discussed separately below.

First, there are terms proportional to B_{xy}^2 . These generate contributions to the free energy with rank $N = 0, 2$ and 4:

$$F''(2, 2) = \tilde{K}''_{220}(T)Y_0^0(\theta, \phi) + \tilde{K}''_{222}(T)Y_2^0(\theta, \phi) + \tilde{K}''_{224}(T) \\ \times \left[Y_4^0(\theta, \phi) - \sqrt{\frac{35}{2}}(Y_4^4(\theta, \phi) + Y_4^{-4}(\theta, \phi)) \right] \quad (25)$$

where:

$$\begin{aligned} \tilde{K}''_{220}(T) &= -\frac{1}{2}\beta \left[+\sqrt{\frac{\pi}{5}}(B_{xy})^2\alpha_{220}(T) \right] \\ \tilde{K}''_{222}(T) &= -\frac{1}{2}\beta \left[+\sqrt{\frac{2\pi}{7}}(B_{xy})^2\alpha_{222}(T) \right] \\ \tilde{K}''_{224}(T) &= -\frac{1}{2}\beta \left[+\sqrt{\frac{\pi}{70}}(B_{xy})^2\alpha_{224}(T) \right]. \end{aligned} \quad (26)$$

Here the numbering on the $\tilde{K}''_{nn'N}(T)$ and $\alpha_{nn'N}(T)$ coefficients refers to the rank of the strain term ($n = n' = 2$) which couples vectorially to give a final rank $N = 0, 2, 4$. Note that none of the terms appearing in equation (25) has the same form as the spherical harmonics appearing in first order (see equation (24)).

Secondly, there are cross-terms involving $B_{xy}\tilde{B}_4$. These generate terms with rank $N = 2, 4$ and 6:

$$F''(2, 4) = \tilde{K}''_{242}(T)i[Y_2^{-2}(\theta, \phi) - Y_2^2(\theta, \phi)] + \tilde{K}''_{244}(T)i[Y_4^{-2}(\theta, \phi) - Y_4^2(\theta, \phi)] \\ + \tilde{K}''_{246}(T)i \left\{ [Y_6^{-2}(\theta, \phi) - Y_6^2(\theta, \phi)] + \frac{3\sqrt{55}}{13}i[Y_6^{-6}(\theta, \phi) - Y_6^6(\theta, \phi)] \right\} \quad (27)$$

where:

$$\begin{aligned} \tilde{K}''_{242}(T) &= -\frac{1}{2}\beta \left[-\frac{4}{3}\sqrt{\frac{2\pi}{7}}B_{xy}\tilde{B}_4\alpha_{242}(T) \right] \\ \tilde{K}''_{244}(T) &= -\frac{1}{2}\beta \left[+4\sqrt{\frac{3\pi}{77}}B_{xy}\tilde{B}_4\alpha_{244}(T) \right] \\ \tilde{K}''_{246}(T) &= -\frac{1}{2}\beta \left[+\frac{26}{33}\sqrt{\frac{11\pi}{14}}B_{xy}\tilde{B}_4\alpha_{246}(T) \right]. \end{aligned} \quad (28)$$

Note that (i) in writing equation (27) care has been taken to include both the cross-terms $B_{xy}\tilde{B}_4$ and \tilde{B}_4B_{xy} and (ii) the $\tilde{K}''_{242}(T)$ term has the same functional form as the first-order term of equation (22).

Table 1. The RE ion parameters after (Martin *et al* 2006).

RE	B_4 (K/ion)	B_6 (K/ion)	X (K/ion)
Tb	$+6.45 \times 10^{-3}$	$+8.73 \times 10^{-6}$	150
Dy	-3.02×10^{-3}	-7.22×10^{-6}	100
Ho	-1.15×10^{-3}	$+8.03 \times 10^{-6}$	75
Er	$+1.86 \times 10^{-3}$	-1.13×10^{-6}	60
Tm	$+6.33 \times 10^{-3}$	$+2.72 \times 10^{-6}$	50

Table 2. Estimates of the magneto-elastic term b_2 and B_{xy} at 0 K.

	b_2 (J m ⁻³)	b_2 (K/ion)	B_{xy} (K/ion)
Tb	-6.41×10^8	-2.29×10^3	0.300
Dy	-6.11×10^8	-2.17×10^3	0.187
Ho	-2.33×10^8	-0.84×10^3	0.071
Er	$+2.19 \times 10^8$	$+0.76 \times 10^3$	-0.066
Tm	$+5.39 \times 10^8$	$+1.92 \times 10^3$	-0.242

Third, there are cross-terms involving $B_{xy}\tilde{B}_6$, this time with rank $N = 4, 6$ and 8 :

$$\begin{aligned}
F''(2, 6) = & \tilde{K}_{264}''(T) i [Y_4^{-2}(\theta, \phi) - Y_4^2(\theta, \phi)] \\
& + \tilde{K}_{266}''(T) \left\{ i [Y_6^{-2}(\theta, \phi) - Y_6^2(\theta, \phi)] - \frac{1}{5} \sqrt{\frac{11}{5}} i [Y_6^{-6}(\theta, \phi) - Y_6^6(\theta, \phi)] \right\} \\
& + \tilde{K}_{268}''(T) \left\{ i [Y_8^{-2}(\theta, \phi) - Y_8^2(\theta, \phi)] - \frac{1}{3} \sqrt{\frac{1001}{15}} i [Y_8^{-6}(\theta, \phi) - Y_8^6(\theta, \phi)] \right\}
\end{aligned} \tag{29}$$

where:

$$\begin{aligned}
\tilde{K}_{264}''(T) &= -\frac{1}{2} \beta \left[+16 \sqrt{\frac{3\pi}{143}} B_{xy} \tilde{B}_6 \alpha_{264}(T) \right] \\
\tilde{K}_{266}''(T) &= -\frac{1}{2} \beta \left[+10 \sqrt{\frac{\pi}{11}} B_{xy} \tilde{B}_6 \alpha_{266}(T) \right] \\
\tilde{K}_{268}''(T) &= -\frac{1}{2} \beta \left[+3 \sqrt{\frac{3\pi}{26}} B_{xy} \tilde{B}_6 \alpha_{268}(T) \right].
\end{aligned} \tag{30}$$

Note the appearance of the rank 8 spherical harmonics $Y_8^{\pm 2}(\theta, \phi)$ and $Y_8^{\pm 6}(\theta, \phi)$. These possess the same rank as the \tilde{K}_8 terms of Martin *et al* (2006), but not the same functional form.

To make further progress, estimates are required for B_{xy} , B_4 and B_6 , for the five heavy REs in question.

5. Calculations

Values of the crystal field parameters B_4 and B_6 and the RE–Fe magnetic exchange field parameter X have been given by Martin *et al* (2006), and references contained therein. For convenience, these are reproduced in table 1.

Estimates for the magneto-elastic term b_2 for MBE-grown TbFe₂, DyFe₂ and ErFe₂ have been given by Mougin *et al* (2000). However, in table 2 we list the parameters used in this work. These were obtained using table 15.2 of Clark (1979), together with a value of the elastic

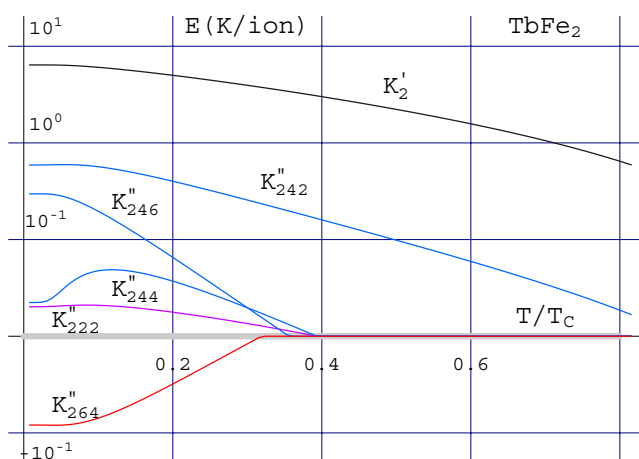


Figure 1. Anisotropy parameters for TbFe_2 which involve the shear term $b_2\varepsilon_{xy}$. The curves are labelled individually, but for colour on-line the black curve is first order, while the curves in purple, blue and red originate from B_{xy}^2 , $B_{xy}\tilde{B}_4$ and $B_{xy}\tilde{B}_6$, respectively. Curves not shown have been suppressed either for clarity or because they are small. Following Atzmony and Dariel (1976), anisotropy values between -10^{-2} and $+10^{+2}$ (K/ion) have been set equal to zero (thick line).

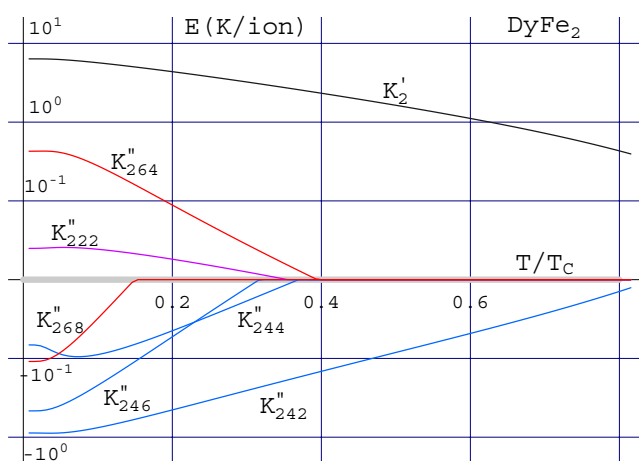


Figure 2. Anisotropy parameters for DyFe_2 which involve the shear term $b_2\varepsilon_{xy}$. Comments as per figure 1.

constant $C_{44} = 4.86 \times 10^{10} \text{ J m}^{-3}$ derived from ultrasonic experiments on bulk $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ (Rinaldi *et al* 1977).

The calculated strain-related anisotropy parameters for $\text{DyFe}_2\text{-TmFe}_2$ can be seen in figures 1–5, respectively. In preparing these logarithmic plots we have followed the system adopted by Atzmony and Dariel (1976) in that values between -10^{-2} and $+10^{+2}$ (K/ion) are considered to be zero (the thick horizontal line in figures 1–5). Note that the sign of \tilde{K}'_2 in Er and Tm is opposite to that in Tb, Dy and Ho, in accord with the sign of the magneto-elastic term B_{xy} of table 2.

The estimates shown in figures 1–5 can now be used, in conjunction with the cubic multipolar constants \tilde{K}_N ($N = 4\text{--}12$) of Martin *et al* (2006), as a starting point for the magnetic

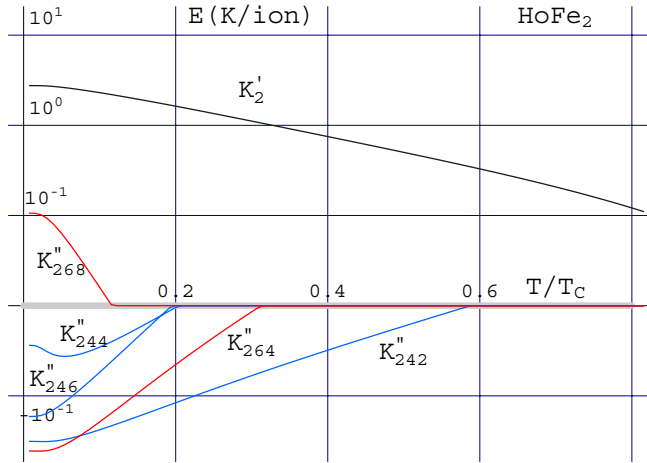


Figure 3. Anisotropy parameters for HoFe₂ which involve the shear term $b_2\varepsilon_{xy}$. Comments as per figure 1.

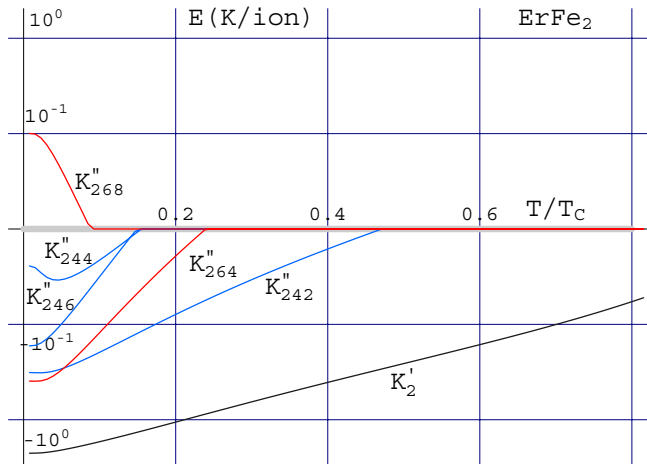


Figure 4. Anisotropy parameters for ErFe₂ which involve the shear term $b_2\varepsilon_{xy}$. Comments as per figure 1.

modelling of epitaxial thin films. In general, the largest second-order terms are $\tilde{K}_{242}''(T)$ and $\tilde{K}_{264}''(T)$, which are of similar magnitude. At high temperatures, for say $T/T_C > 0.2$, $\tilde{K}_{242}''(T)$ dominates. As noted earlier, this term has the same angular dependence as the first-order term \tilde{K}_2' , but its temperature dependence is more rapid. At lower temperatures, the $\tilde{K}_{264}''(T)$ term becomes important. Thus we write:

$$\begin{aligned}
 F_A = F' + F'' &\approx \left(\tilde{K}_2' + \tilde{K}_{242}''(T) \right) i \left[Y_2^{-2}(\theta, \phi) - Y_2^2(\theta, \phi) \right] \\
 &+ \tilde{K}_{264}''(T) i \left[Y_4^{-2}(\theta, \phi) - Y_4^2(\theta, \phi) \right] \\
 &= \tilde{K}_{\text{eff}}''(T) \frac{1}{2} \sqrt{\frac{15}{2\pi}} \sin^2 \theta \sin 2\phi + \tilde{K}_{264}''(T) \frac{3}{4} \sqrt{\frac{5}{2\pi}} (7 \cos^2 \theta - 1) \sin^2 \theta \sin 2\phi.
 \end{aligned} \tag{31}$$

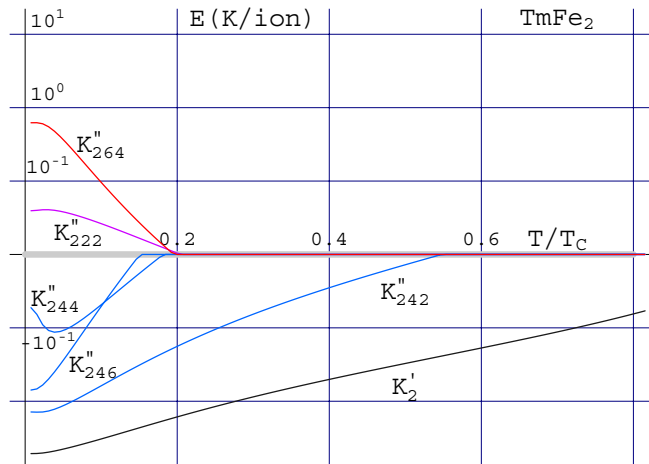


Figure 5. Anisotropy parameters for TmFe₂ which involve the shear term $b_2\varepsilon_{xy}$. Comments as per figure 1.

The correction at $T = 0$ K to \tilde{K}'_2 arising from $\tilde{K}''_{242}(T)$ amounts to +9.2%, -13.9%, -11.6%, +14.3% and +27.1%, for TbFe₂, DyFe₂, HoFe₂, ErFe₂ and TmFe₂, respectively. The percentage value of the $\tilde{K}''_{264}(T)$ term relative to \tilde{K}'_2 is -1.3%, +6.7%, -14.8%, +17.5% and -12%, respectively. Clearly the first-order term is dominant but the second-order terms can give rise to changes of up to $\sim 25\%$.

6. Conclusions and discussion

In summary, of the nine second-order terms which involve the strain ε_{xy} , only the $\tilde{K}''_{242}(T)$ and $\tilde{K}''_{264}(T)$ terms are important. The former arises from the cross-term $B_{xy}\tilde{B}_4$ and possesses the same angular form of the principal first-order contribution \tilde{K}'_2 . The second term, proportional to $B_{xy}\tilde{B}_6$, has a relatively fast temperature dependence and is only important at low temperatures.

However, some words of caution are in order. One, the strain term ε_{xy} is itself temperature dependent (see figure 2 of Mougin *et al* 2000), decreasing by $\sim 25\%$ on reaching room temperature. This will give a more rapid temperature dependence than that shown in figures 1–5. However, this effect can be readily taken into account, since all the data shown in figures 1–5 are linearly proportional to ε_{xy} . Two, it has been assumed that the elastic constant C_{44} is independent of both RE and temperature. Clearly more measurements will be required to shed light on these two features. Three, any distortions of the lattice should give rise to changes in the fourth- and sixth-order coefficients \tilde{B}_4 and \tilde{B}_6 . In this regard, we note that the point charge model has been used to show that these two coefficients hardly change for shear distortions (Bowden *et al* 2004, section 4). Thus we conclude, tentatively, that all the principal terms, linearly proportional to ε_{xy} , have been included. However, a more sophisticated calculation of the coefficients \tilde{B}_4 and \tilde{B}_6 , based say on a density-functional approach (e.g. Richter *et al* 1992), may well be in order.

Finally, given that the cross-terms involving strain do not amount to more than 25% or so, we believe that it is reasonable to assume, as a first approximation, that the dominant anisotropy terms in MBE-grown REFe₂ thin films are the cubic anisotropy terms $\tilde{K}_4 - \tilde{K}_{10}$ (figures 1–5 of Martin *et al* 2006) and the first-order strain terms \tilde{K}'_2 (figures 1–5 of this paper).

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