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Magnetocrystalline anisotropy in a (110) (Tb_{0.27}Dy_{0.73})Fe₂ thin-film

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Abstract

Magnetic anisotropy measurements performed in a (110) (Tb_{0.27}Dy_{0.73})Fe₂ (Terfenol-D) film epitaxially grown on a sapphire substrate are presented. The magnetic torque curves have been determined by using a vectorial vibrating sample magnetometer, which allows us to measure the angular dependence of magnetization components parallel, M_{\parallel} , and perpendicular, M_{\perp} , to the applied field up to 2 T. The fourfold symmetry associated with the cubic structure within the (110) plane is clearly observed. The analysis of the experimental torque has been carried out considering magnetocrystalline anisotropy up to sixth order and magnetoelastic energy up to second order; so, the magnetocrystalline anisotropy constants in the (110) plane of the film, K_1 and K_2 , have been obtained. This allows us to determine the direction of the magnetization easy axis for (110) Terfenol-D thin-film: it is $[\bar{1}12]$ at RT, passes through $[\bar{3}34]$ at 140 K and then changes to $[\bar{1}20]$ at 40 K. It was completely impossible to explain the angular dependence of the experimental magnetic torque without including shear and tetragonal magnetoelastic stress parameters, b_2 and b_1 , respectively. This confirms the paramount role of the strain in the determination of the magnetic properties in this kind of Terfenol-D thin film.

The bulk alloy (Tb_{0.27}Dy_{0.73})Fe₂ (Terfenol-D) has been widely studied due to its remarkable technological applications at RT. Terfenol-D shows a low coercive field at RT and a high ratio between the cubic magnetoelastic (MEL) stress parameter, $b_2 \cong -0.2$ GPa, associated with the highly anisotropic cubic magnetostriction λ_{111} ($\lambda_{111} \gg \lambda_{100}$; λ_{100} is the non-cubic magnetostriction responsible for the volume and tetragonal strains: $\lambda_{111} = 1.64\%$ and

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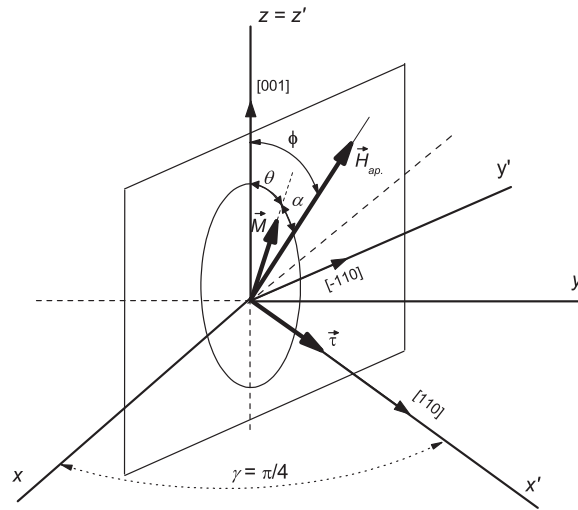


Figure 1. An outline representation of the main crystallographic directions with respect to the (110) plane of the Terfenol-D film.

$\lambda_{100} = 0.09\%$) and the lowest order magnetocrystalline anisotropy constant $K_1 \cong -3 \times 10^{-4}$ GPa [1]. On decreasing the temperature, Terfenol-D undergoes spin reorientation transitions: the magnetization easy axis (ea) is $\langle 111 \rangle$ at RT; between 250 and 175 K there exist unusual $\langle uvw \rangle$ magnetization eas, which suggested the possible influence of eighth order terms in the direction-cosines of the magnetization expansion of the magnetocrystalline energy [2]; down to 20 K the ea is $\langle 100 \rangle$; and below 20 K it is $\langle uv0 \rangle$ -type [2].

Over the last decade much effort has been made to study the magnetic and MEL properties of REFe₂ films (RE = rare earths), looking for possible modifications of their properties as compared with the bulk material and, hence, for some improvement of the magnetoelastic and magnetocrystalline properties [3]. Its potential uses as microdevices motivate such effort. The recent use of the molecular-beam epitaxy technique in the growth of Laves-phases thin-films has allowed the growth of epitaxial (110) REFe₂ thin films [4]. This opens the possibility of studying the influence of factors like stress, thickness, growing conditions, etc, in the magnetic behaviour of these systems. A spontaneous spin-reorientation transition of (110) Terfenol-D thin-film, which changes the magnetization ea from $\langle 331 \rangle$ above 220 K to $\langle 114 \rangle$ below 65 K has been observed [5].

In previous works, we have studied the magnetic and MEL properties of some REFe₂ thin-films (RE = Tb, Tb_{0.3}Dy_{0.7}) [6, 7]. Clear effects of epitaxial strain and interfaces on the magnetoelastic coupling were shown. One of the more relevant aspects we would like to mention about the magnetoelastic behaviour of Terfenol films is that the cubic magnetostriction slightly reduces its values with respect to the bulk, but the non-cubic one is notably increased.

In this work we present magnetic anisotropy measurements performed in a single (110) Terfenol-D thin-film by using a 2D-vector vibrating sample magnetometer [9], where the magnetic field, up to 2 T, is applied within the (110) plane of the film. The magnetic torque curves have been determined by using this magnetometer, which allows us to measure $\mu_0 M_{\parallel}$ and $\mu_0 M_{\perp}$ in the plane of the thin-film with respect to the angular dependence of the total applied field, $\mu_0 \mathbf{H}_{ap}$, and a reference crystallographic direction, [001]. Note that the torque exerted on the sample by the magnetic field is $\tau = |\mu_0 \mathbf{M} \times \mu_0 \mathbf{H}_{ap}| = \mu_0^2 H M_s \sin \alpha = \mu_0^2 H M_{\perp}$, where M_s is the saturation magnetization, $M_{\perp} = M_s \sin \alpha$, with α the angle between $\mu_0 \mathbf{M}$ and the applied magnetic field, $\mu_0 \mathbf{H}_{ap}$ (see figure 1).

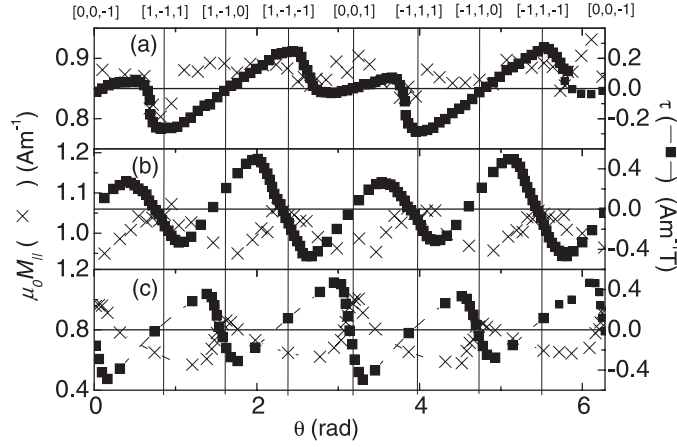


Figure 2. The θ -angular dependence of the M_{\parallel} and experimental torque at 2 T and at temperatures of 290 K (a), 140 K (b) and 40 K (c).

The (110) Terfenol-D thin film was grown by molecular-beam epitaxy, upon a (17 Å) thin Fe seed-layer deposited onto a (110) Nb layer covering a sapphire substrate, and capped with a (100 Å) thin Y film [4]. The (110) Terfenol-D layer had a thickness of 600 Å and was obtained by codeposition of the rare earth and iron constituents ($T_{\text{dep}} = 820$ K). X-ray scattering diffraction on Terfenol-D films showed a good epitaxial growth, a coherence length of about 200 Å, mosaic spread $\approx 1.5^\circ$, and roughness ≈ 25 Å [5]. Owing to the different thermal-expansion coefficients between the (110) Terfenol-D film and substrate, the cooling of the sample from the deposition temperature to RT produces a compressive shear deformation perpendicular to the (110) plane, $\epsilon_{[220]} \approx -0.64\%$, and a tensile and isotropic strain within the (110) plane, $\epsilon_{[004]} \approx +0.41\%$ [5].

In figure 2 we show the parallel magnetization and the magnetic torque at 2 T for 40 K, 140 K and RT as a function of the angle, θ , between \mathbf{M} and a reference crystallographic direction of the crystal, [001]. Note that the magnetization \mathbf{M} was experimentally measured as a function of the angle, ϕ , between the applied magnetic field \mathbf{H}_{ap} and the crystallographic direction [001]. The relationship between α , ϕ and θ is explained in figure 1, when \mathbf{M} and \mathbf{H}_{ap} are within the sample plane (110), where α can be calculated from the components of the magnetization M_{\parallel} and M_{\perp} as

$$\alpha = \arctan\left(\frac{M_{\perp}(\theta)}{M_{\parallel}(\theta)}\right). \quad (1)$$

Plotting the torque curves versus θ is the usual method to relate the experimental torque with the one deduced from the total free energy density of the sample [9].

The cubic symmetry associated with the (110) plane of the sample is clearly observed in figure 2. The θ -angular comparison of the magnetic torque and the parallel magnetization help us to localize *in situ* easy and hard directions. When the value of τ is negligible and M_{\parallel} is maximum or minimum, the direction is either an easy one or a hard one. In this sense figure 2 shows that $[\bar{1}13]$, [001], $[\bar{1}11]$, and $[\bar{1}10]$, and their equivalents within the (110) plane, are the directions where the torque is negligible. Note how the magnetization \mathbf{e}_a at 2 T could be either the $[\bar{1}10]$ or [001] at RT (see figure 2), passes through $[\bar{1}11]$ at 140 K, and finally is [001] at 40 K. At temperatures close to RT it is difficult to establish the magnetization \mathbf{e}_a because the parallel magnetization hardly changes and is quite weak.

The magnetic torque can be easily calculated from the total phenomenological free energy density, which can be written as follows:

$$F = F_k + F_{\text{MEL}} + F_d + F_Z, \quad (2)$$

where the magnetocrystalline anisotropy energy density, F_k , expanded up to sixth order in the direction cosines, $\alpha_x, \alpha_y, \alpha_z$, of the magnetization, $M = M_s(\alpha_x, \alpha_y, \alpha_z)$, is

$$F_k = K_1(\alpha_x^2\alpha_y^2 + \alpha_x^2\alpha_z^2 + \alpha_y^2\alpha_z^2) + K_2(\alpha_x^2\alpha_y^2\alpha_z^2), \quad (3)$$

K_i being the magnetocrystalline anisotropy constants; F_{MEL} is the magnetoelastic energy density and is written up to second order in $\alpha_x, \alpha_y, \alpha_z$ and linearly with respect to the strain tensor components, ε_{ij} :

$$F_{\text{MEL}} = b_1(\alpha_x^2\varepsilon_{xx} + \alpha_y^2\varepsilon_{yy} + \alpha_z^2\varepsilon_{zz}) + b_2(\alpha_x\alpha_y\varepsilon_{xy} + \alpha_x\alpha_z\varepsilon_{xz} + \alpha_y\alpha_z\varepsilon_{yz}), \quad (4)$$

b_i being the magnetoelastic stress parameters and ε_{ij} the Cartesian strain tensor components; F_d is the magnetostatic energy density obtained for (110) epitaxial thin-films; $F_d = (1/4)(\mu_0 M_s)^2(\alpha_x + \alpha_y)^2$ with spontaneous magnetization partially oriented out of that growing plane, and, finally, F_Z is the Zeeman term,

$$F_Z = -\mu_0^2 H_{\text{ap}} M_s \cos(\phi - \theta). \quad (5)$$

Assuming that the Zeeman energy density is not large enough to orient the magnetic moments along the field direction but that it can force them within the (110) plane, the direction of magnetization can be written in the (x', y', z') reference system (see figure 1) as $\alpha_{x'} = 0$, $\alpha_{y'} = \sin\theta$ and $\alpha_{z'} = \cos\theta$, where θ changes from 0 to 2π . To obtain the corresponding direction cosines in the (x, y, z) reference system a z -rotation of value $\gamma = \pi/4$ must be done (see figure 1). Now, we can minimize the total free energy density against θ , $\partial F/\partial\theta = 0$, and then the magnetic torque is

$$\begin{aligned} \tau = \mu_0^2 H_{\text{ap}} M_s \sin(\phi - \theta) = & -\frac{1}{2} b_2 \varepsilon'_{yz} \cos(2\theta) \\ & + \left(\frac{K_1}{4} + \frac{K_2}{64} + \frac{b_1}{2} (\varepsilon'_{xx} + \varepsilon'_{yy} - 2\varepsilon'_{zz}) + \frac{b_2}{4} (-\varepsilon'_{xx} + \varepsilon'_{yy}) \right) \sin(2\theta) \\ & + \left(\frac{3K_1}{8} + \frac{K_2}{16} \right) \sin(4\theta) - \left(\frac{3K_2}{64} \right) \sin(6\theta). \end{aligned} \quad (6)$$

The magnetic torque $\mu_0^2 H_{\text{ap}} M_s \sin(\phi - \theta) \equiv \mu_0^2 H_{\text{ap}} M_s \sin(\alpha)$ is experimentally determined by measuring $\mu_0 M_{\perp}$ ($\tau = \mu_0^2 H_{\text{ap}} M_{\perp}$); on the other hand, ε'_{ij} are the deformations in the (x', y', z') reference system, where $\varepsilon'_{ii} = \varepsilon_{ii}$, $\varepsilon'_{ij} = 2\varepsilon'_{ij}$, while ε'_{ij} represents the strain tensor components. Note that the strain tensor $\{\varepsilon'\}$ is $\pi/4$ rotated around the [001] direction from the non-rotated Cartesian one, $\{\varepsilon\}$.

A reduction of the total magnetic moment in the (110) plane is observed below 40 K. This indicates firstly the existence of an out-of-plane ea, and secondly that the Zeeman energy, at 2 T, is unable to rotate the magnetic moments towards the (110) plane. Thus, equation (6) does not hold for these temperatures. However, for $T \geq 40$ K, the applied magnetic field is strong enough to confine the total magnetization in the (110) plane (the $y'z'$ -plane in figure 1), and the experimental torque is adequately described by equation (6). Now, using it, fixing the magnetoelastic coupling coefficients, b_1 and b_2 , to the values previously obtained (see table 1), and taking as initial values of the deformations ε'_{xx} and $\varepsilon'_{yy}, \varepsilon'_{zz}$ those obtained from XRD at zero field [5], we obtain the set of parameters (see table 1) giving the best fit to the experimental torque curves at 2 T. Figure 3 shows the fit for the torque measured at 140 K and 2 T.

Now, the calculation of the free energy density by using the parameters listed in table 1 allow us to elucidate what is the minimum of the total density free energy when the torque

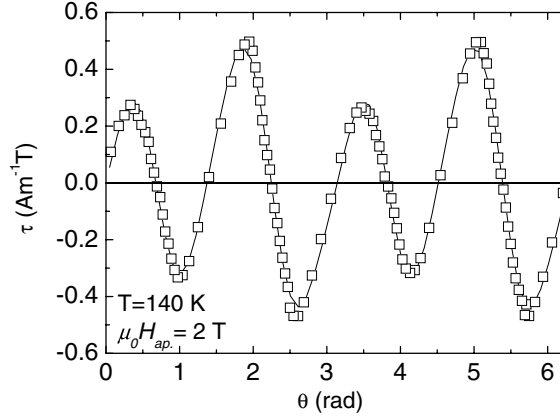


Figure 3. The torque curve measured at 140 K and 2 T and a continuous line, which represents the best fit to equation (6) by using the parameters displayed in table 1.

Table 1. Parameters used in the fit to the torque curves at 2 T by using equation (6) and the anisotropy constants obtained from extrapolation to saturation. The anisotropy constants K_i and the magnetoelastic parameters b_i are given in MPa. The strains are given in per cent.

T (K)	K_1	K_2	b_1^a	b_2^a	ϵ'_{xx}	ϵ'_{yy}	ϵ'_{zz}	ϵ'_{yz}
40	1.45	-1.45	-180	-180	-0.27	1.02	0.54	0.012
140	-0.975	-0.356	-135	-145	-0.415	0.8	0.715	0.166
290	-0.525	1.543	-80	-85	-0.643	0.404	0.502	-0.019

^a See [7].

and parallel magnetization measurements are not conclusive. This is the situation for the RT measurements. The result shows that $[\bar{1}10]$ is the magnetization e_a at 2 T.

We should mention that attempts to fit the torque curves without including the MEL contributions were completely unsuccessful. This points to the essential role of the MEL coupling in the magnetic anisotropy of the present system, which was also previously highlighted by Mössbauer studies at zero field in REFe₂ thin-films [5]. In fact, the values of $\epsilon_{[220]} = \epsilon'_{xx}$ and $\epsilon_{[004]} = \epsilon'_{yy}$ appearing in table 1 are different from the initial zero-field values, except for the signs. We also see that the deformations within the (110) plane ($y'z'$ -plane in figure 1) are different, $\epsilon'_{zz} \neq \epsilon'_{yy}$, unlike the XRD results at zero-field, which indicate that they are isotropic, $\epsilon'_{zz} = \epsilon'_{yy}$. The shear deformations within the (110) plane, ϵ'_{yz} , cannot be compared with the XRD results; however, they are small, except at 140 K. The reason for the differences found between our values for the strains, obtained under a magnetic field, and the XRD zero-field values can be ascribed to the magnetostriction, which is large due to the strong magnetoelastic coupling in this system [7]. For instance, at 2 T the magnetization e_a at 140 K is $[111]$, and the large magnetostriction must be the origin of the large *shear* strain, ϵ'_{yz} , obtained at this temperature; as to the *non-shear* strains, at 40 K, where the magnetization e_a is $[001]$ and the magnetostriction is larger, we obtain a high value for the ϵ'_{zz} strain; whereas at RT the magnetization e_a is $[\bar{1}10]$, because at this temperature the magnetostriction is small [7], and the strain ϵ'_{yy} is not very different from the zero-field value.

We also could attribute the origin of the anisotropy of the deformations within the (110) plane to the magnetostriction, and specifically to its anisotropy. Unfortunately, in our earlier magnetoelastic stress measurements in the present film, we did not perform any experiment

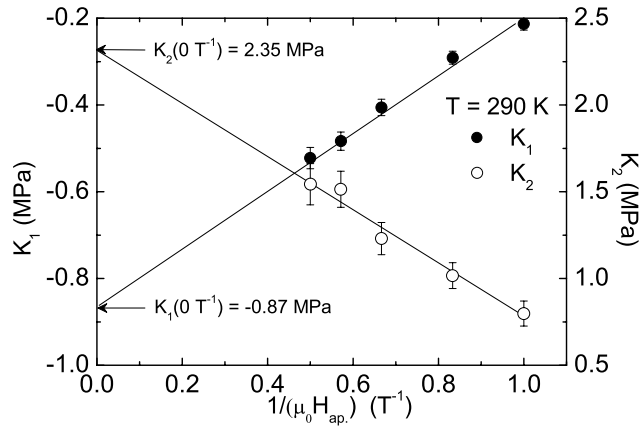


Figure 4. The inverse field dependence of the anisotropy parameters K_1 , and K_2 obtained at 290 K at fields of 1, 1.2, 1.5, 1.7 and 2 T. The continuous lines represent guide lines, which help us to obtain an extrapolation at the infinite field limit to get the right magnetic anisotropy constants.

Table 2. The first columns on the left show the magnetocrystalline anisotropy constants obtained from extrapolation at infinite field. The abbreviation ea represents the easy axis.

T (K)	K_1 (MPa)	K_2 (MPa)	ea	$\mu_0 M_s$ (T) ^a
40	2.54	-3.19	$[\bar{1}20]$ $[1\bar{2}0]$ $[\bar{1}\bar{2}0]$ $[120]$	1.326
140	-1.74	-1.35	$[\bar{3}34]$ $[3\bar{3}4]$ $[\bar{3}\bar{3}4]$ $[3\bar{3}\bar{4}]$	1.196
290	-0.87	2.35	$[\bar{1}12]$ $[1\bar{1}2]$ $[\bar{1}\bar{1}\bar{2}]$ $[1\bar{1}\bar{2}]$	0.945

^a See [7].

applying the magnetic field along directions other than $[\bar{1}10]$ and, therefore, we have no independent evidence of such anisotropy. However, the anisotropy of magnetostriction has been experimentally observed within the hexagonal basal plane in metallic rare earths [10], and it could be also present in our system.

Figure 4 shows the anisotropy constants obtained at different magnetic fields at RT. At 40 and 140 K we also get field-dependent anisotropy constants. The analysis of the torque curves has been done after correcting them for the angle α , as we explained above (see equation (1)). In principle, this correction should give field-independent values of the anisotropy constants, although it is usual to find some dependence on the field, ascribed to some spurious effect, as having a non-totally homogeneous magnetization in the sample, or to uncertainty in the α -correction [9]. In this situation, it is a customary procedure to extrapolate the anisotropy constants to infinite field linearly, especially when the anisotropy fields at different temperatures are of the same order or smaller than the applied fields. In our case, magnetization hysteresis loops indicate that the anisotropy fields are below 1.5 T [7]. Therefore, we have proceeded with such an extrapolation to get the *true* anisotropy constants (see figure 4, for the RT case). In table 2 we summarize the extrapolated values of K_1 and K_2 for the different measuring temperatures. By using these values, the magnetoelastic stress parameters b_1 and b_2 given in table 1, and the zero-field strains [5], we can calculate the ea of magnetization by searching for the minima of the free energy density at zero applied field. In figure 5 we display a contour plot of isoenergy lines in a grey-scale plot (the grey scale shows darker colouring for lower energy) at RT, $\hat{\theta}$ and $\hat{\phi}$ being the spherical coordinates of the magnetization ea with respect to the $OXYZ$ reference system. The four maxima and four minima found determine the angular positions of

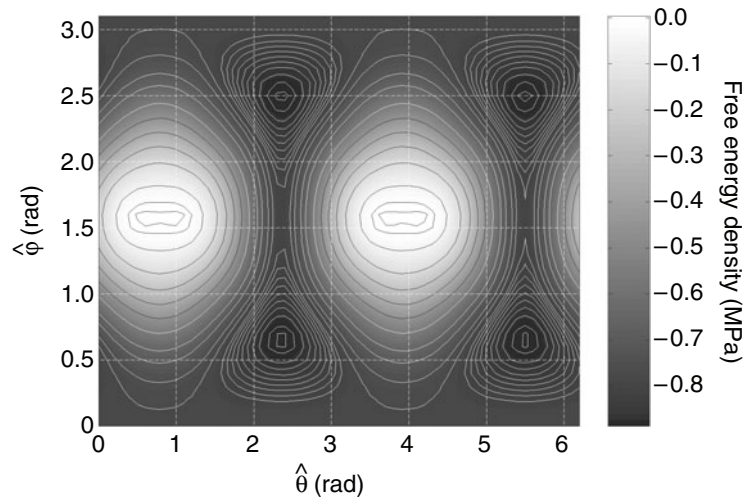


Figure 5. Contour plot of isoenergy lines at RT by using the b_1 and b_2 magnetoelastic stress parameters [7], the zero-field strains measured by x-rays [5], and the anisotropy constants from table 2. The grey scale shows darker colouring for lower energy. This scale was downshifted 0.45 MPa for plotting purposes.

the hard and easy axes, respectively. The magnetization e_a are summarized in table 2 for the three different measuring temperatures. Our results show that, at 40 K, the magnetization is 19° out of the plane of the film but, at higher temperatures, 140 K and RT, it lies on the (110) plane. Mössbauer experiments [5] shown that the magnetic moments at low temperatures, $T < 65$ K, are along the $[\bar{1}41]$ -direction, which is 32° out of the (110) plane; on increasing the temperature, two equally probable domains are formed: at 150 K the domains of magnetizations are $[\bar{1}42]$ and $[\bar{2}41]$ directions, which form angles smaller than 32° with the $[110]$ direction, and at RT the magnetization e_a is along $[\bar{1}33]$ and $[\bar{3}31]$. There is some discrepancy between the directions obtained by us and those deduced from Mössbauer data but, qualitatively, both coincide in the fact that the magnetization rotates towards the plane of the film on increasing the temperature. The magnetoelastic and magnetostatic contributions to the density free energy favour in-plane magnetization. In contrast, the magnetocrystalline anisotropy constants given in table 2 will favour an e_a out of the (110) plane. The different temperature dependence of all the above contributions gives rise to the spin reorientation observed at different temperatures (see table 2). Note that the magnetization e_a obtained at 40 K is out of the (110) plane. This fact can be related to the predominance of the magnetocrystalline term at low temperature over the other density energy terms.

A large magnetocrystalline anisotropy energy contribution has also been argued to explain the Mössbauer results in the Terfenol-D film [5], but in this case this contribution has been ascribed merely to a large value of the eighth order terms in F_K , which we have not considered in our expression of the magnetocrystalline energy given in equation (3). This kind of high-order term is introduced phenomenologically in F_K , and it was useful to explain most of the spin-orientation diagrams of the REFe₂ compounds, but was unable to precisely describe the Mössbauer results for bulk Terfenol-D [2]. However, the inclusion of the lower order magnetoelastic energy associated with the large b_1 and b_2 stresses (determined after the publication of [5]), is enough to account for the changes in the easy directions, without the use of an eighth-order term in F_K that, on the other hand, is not connected with any microscopic parameter, unlike K_1 , K_2 , b_1 and b_2 .

In summary, we have deduced the magnetic anisotropy constants K_1 and K_2 in a (110) Terfenol-D film from magnetic torque measurements. Notice that the easy axis directions obtained under a magnetic field applied within the plane of the film (our experimental situation) differ strongly from those we have obtained at zero field by using the deduced anisotropy constants. It is worth mentioning that, by including a Zeeman term in the free energy applying the magnetic field within the plane of the sample, we recover the magnetization easy directions obtained in the torque experiments. Moreover, we have shown that the magnetoelastic stress plays an essential role in setting up the magnetization easy direction of the sample; the competition between the magnetoelastic and magnetocrystalline anisotropy energies, which have different temperature variations, give rise to the experimentally observed changes in the magnetization easy direction.

Acknowledgments

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