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LETTER TO THE EDITOR

Strong interfacial magnetoelastic stress in holmium–lutetium (0001) superlattices

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Abstract. The saturation magnetoelastic stress (MS), M^γ, breaking the basal plane cylindrical symmetry of the hexagonal structure, has been measured for the series of superlattices $(Ho_n/Lu_{15}) \times 50(0001)$ ($n = 8$ to 85 atomic planes, with separation $c/2$). The MS was directly measured using a low-temperature cantilever technique. The Ho block MSs in the superlattices are larger than in an Ho film of 10^4 Å thickness and in bulk Ho. An analysis accounting for the contributions to M^γ coming from: the volume, M^γ_{v0}, the *interface*, with magnetoelastic parameter M^γ_s, and the epitaxial strain shows that the interfacial magnetoelastic stress $2M^\gamma_s/n(c/2)$ is *strong*, up to about six times larger than M^γ_{v0} for $n = 8$, and of the opposite sign.

There exists considerable current interest about the magnetic properties of rare earth (RE) superlattices (SLs), where the magnetic blocks, with n_{RE} atomic planes (a.p.), are interleaved by blocks of a non-magnetic rare earth such as Lu or Y, with e.g. n_{Lu} a.p. [1–3]. It is well known that in bulk RE metals such as Dy and Ho the basal plane magnetoelastic energy is one of the agents driving the magnetic structure from helical to ferromagnetic [4]. In a previous letter [5] we presented the first magnetoelastic stress (MS) measurements in a unique $(Ho_6/Y_6) \times 100(0001)$ SL, using a purpose built cantilever capacitance device, working from 1.7 K at applied magnetic fields up to 12 T; this technique has been now used extensively in the present work. In the present letter we will focus on the cylindrical symmetry breaking MS measurements performed on a series of $(Ho_n/Lu_{15}) \times 50(0001)$ SLs, where n ranges from 8 to 85 atomic planes (a.p.), with separation $c/2$. Two thick Ho films of 5×10^3 Å and 10^4 Å thicknesses were also studied. The SLs were grown using a molecular beam epitaxy (MBE) technique, upon an Lu seed on Nb covered sapphire as substrate and capped by an Lu film. The epitaxial relationships were: $\{11\bar{2}0\}Al_2O_3 \parallel \{110\}Nb \parallel \{0001\}RE$. More details about the MBE technique, the characteristics of the samples and the method for their crystalline characterization are given in [3] and [5]. Good epitaxial growth was found in the similar $(Ho_{40}/Y_{15}) \times 50$ and $(Ho_{10}/Y_{10}) \times 50$ SLs, the misfit $(a(Y) - a(Ho))/a(Ho)$ for the hexagonal a lattice parameter at 45 K being 0.21% and 0.01% respectively. The average interdiffusion at the interfaces is estimated as ±2 a.p. over the 50 biblocks [3]. The crystalline coherence length is about 3.000 Å. [3].

The purpose of this research is mainly to investigate the MS originating in the Ho/Lu *interface*. A long time ago Néel [6] predicted that the symmetry breaking at the surface of a ferromagnetic transition metal crystal could be the source of an extra anisotropy. This model was extended by Chuang *et al* [7] and by de Lacheisserie and McGrath [8] considering an atom pair interaction, which for transition metals could be one of the sources of magnetoelastic (MEL) coupling. Very recently O'Handley and coworkers [9] have performed extensive magnetostriction measurements on Ni, NiFe thin films and Cu/Li/Cu sandwiches and analysed the anisotropy measurements of Lee *et al* [10] on Co/Cu superlattices putting forwards the existence of a surface or interfacial MS respectively, larger than the volume one by a factor as large as about −6 in Cu/Li/Cu sandwiches. For these systems the surface or interfacial MS comes from the symmetry reduction and chemistry variation at the surface or interface [9]. However in the present case of Ho/Lu SLs where the $\boldsymbol{c}$ axis is normal to the interface and the basal plane lattice misfit is $\Delta a/a_0 \approx 0.01$–$0.2\%$, a substantial breaking of hexagonal symmetry is hard to believe, and therefore the origin of the MS must be sought from another source.

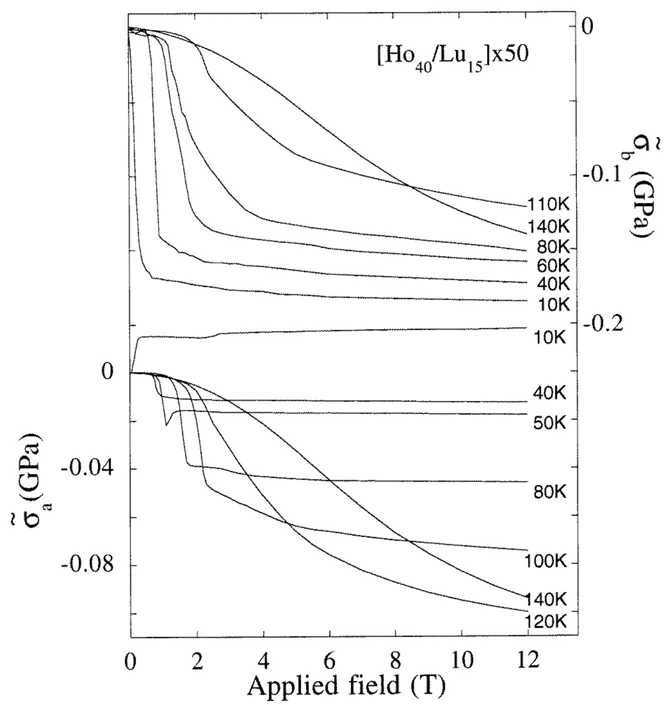

Figure 1. Magnetoelastic stress measured isotherms for the superlattice (SL) $(Ho_{40}/Lu_{15}) \times 50$. $\tilde{\sigma}_a$ and $\tilde{\sigma}_b$ respectively correspond to the SL clamping along the $\boldsymbol{a}$ and $\boldsymbol{b}$ axes of the hexagonal structure, with magnetic field applied along the $\boldsymbol{b}$ easy axis.

In figure 1 we present, as an example, MS isotherms for the sample clamped along the $\boldsymbol{a}$ and $\boldsymbol{b}$ axes of the hcp structure and with the applied magnetic field along the $\boldsymbol{b}$ easy axis, i.e. $\tilde{\sigma}_a$ and $\tilde{\sigma}_b$ respectively, for the SL $(Ho_{40}/Lu_{15}) \times 50$. As we can notice changes in slope are patent for certain applied magnetic fields. We ascribe the first to the helical–fan transition and the second to the fan–ferromagnetic transition. As we may observe saturation is practically accomplished at 12 T and 10 K. Such saturation indicates that we are measuring an MS of crystal electric field (CEF) origin, once the sample is ferromagnetic.

The concrete purpose of our work is to study the cylindrical symmetry breaking magnetoelastic stress in the basal plane, M^{γ}, coming from CEF interaction and therefore we will focus on the saturation MS. We will now mention how M^{γ} was obtained [5]. It can be shown that the experimental MS, $M^{\gamma}_{exp.}$, is obtained by

$$M^{\gamma}_{exp.} = \frac{h_{sa}^2}{3h_{SL}}\left(\frac{C_{xx}}{R_x} - \frac{C_{yy}}{R_y}\right) = 2(\tilde{\sigma}_a - \tilde{\sigma}_b) \tag{1}$$

where h_{sa} and h_{SL} are the sapphire substrate and SL thicknesses respectively, C_{xx} and C_{yy}, particular combinations of the sapphire elastic constants and R_x and R_y, the radii of curvature of the cantilever when respectively clamped along the $\boldsymbol{a}$ and $\boldsymbol{b}$ hexagonal axes. More details about the experimental method are given in [5]. Also in-plane magnetization measurements between 10 K and well above the Néel temperatures were performed at applied magnetic fields up to 12 T, with the field applied along the $\boldsymbol{b}$ easy axis.

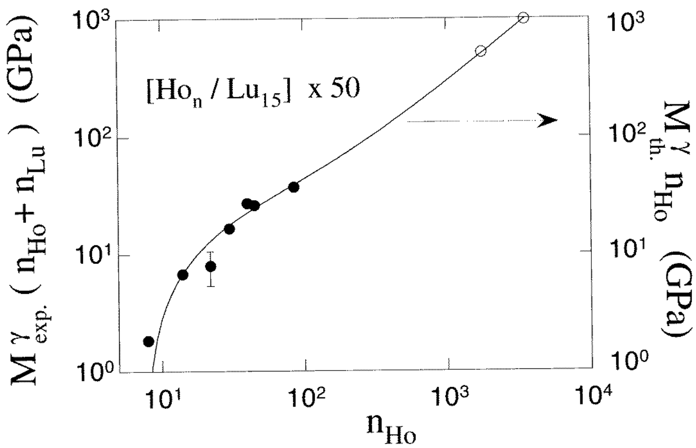

Figure 2. The variation of the basal plane cylindrical symmetry breaking magnetoelastic stress, $M^{\gamma}_{exp.}$, at 10 K and at an applied magnetic field of 12 T, multiplied by $(n_{Ho}+n_{Lu})$ (●), against n_{Ho} (where n_{Ho} and n_{Lu} respectively are the number of atomic planes in the Ho and Lu blocks), for the $(Ho_n/Lu_{15}) \times 50$ superlattices (SLs). $M^{\gamma}_{exp.}$ multiplied by the same factor is plotted for 5×10^3 Å and 10^4 Å thick Ho films (○). The line is the fit by the theoretical model (4), with the right-hand side multiplied by n_{Ho}, using the MEL parameter values given in the text.

Dealing with REs the source of the MEL coupling must be of single-ion origin, coming from the coupling of the RE spin to the strain. In order to interpret our results we assume two sources of MS: one coming from the volume, i.e. M^{γ}_{v}, and the other from the interface, i.e. $\sigma_s = 2M^{\gamma}_{s}/t_{Ho}$, this latter arising in the way explained below and where t_{Ho} is the Ho thickness in the biblock. We also assume that because of the basal plane isotropic strain, $\epsilon = \epsilon_{xx} = \epsilon_{yy}$ [5], produced in the Ho blocks by the misfit, the M^{γ}_{v} parameter is modified in the way [5]

$$M^{\gamma}_{v} = M^{\gamma}_{v0} + D^{\gamma}_{v}\epsilon \tag{2}$$

where $D^{\gamma}_{v} = (\partial M^{\gamma}_{v}/\partial\epsilon)_{\epsilon=0}$. It is easy to show that for free x–y interfaces with no stress component in the z direction, $\epsilon = -(C_{33}/2C_{13})\epsilon_{zz}$, where C_{13} and C_{33} are Ho elastic stiffness constants. Here $\epsilon_{zz} = (c - c^b)/c^b$, where c^b and c are the $\boldsymbol{c}$ axis parameters for the bulk (b) and the superlattice respectively. Now in order to calculate ϵ we write the misfit elastic energy for the Ho/Lu biblock, of respective thicknesses t_{Ho} and t_{Lu}, in terms of the Cartesian strains and elastic constants [11] and minimize it, so obtaining

$$\epsilon = e' \frac{t_{Lu}}{\alpha t_{Ho} + t_{Lu}}. \tag{3}$$

e' is the differential strain between the Ho and Lu blocks in the superlattice, $e' = \epsilon_{Ho} - \epsilon_{Lu} \cong (a^{SL}_{Ho} - a^{SL}_{Lu})/a^{SL}_{Lu} - (a^{b}_{Ho} - a^{b}_{Lu})/a^{b}_{Lu}$, which becomes the lattice constant misfit between bulk Lu and Ho, i.e. $e = (a^{b}_{Lu} - a^{b}_{Ho})/a^{b}_{Lu}$, for *perfect* superlattice epitaxy, i.e. $a^{SL}_{Ho} - a^{SL}_{Lu} = 0$. Also, $\alpha = C^{Ho}_{eff}/C^{Lu}_{eff}$ with $C_{eff} = C_{11} + C_{12} - 2(C^2_{13}/C_{33})$, where C_{ij} are elastic constants. From the Ho and Lu elastic constants [12], we obtain $\alpha = 0.94$. The resulting expression for the overall MEL stress is

$$M^{\gamma}_{th.} = M^{\gamma}_{v0} + \frac{2M^{\gamma}_{s}}{t_{Ho}} + D^{\gamma}_{v} e' \frac{t_{Lu}}{\alpha t_{Ho} + t_{Lu}} \tag{4}$$

where the third term is the misfit stress, σ_{ε}. In figure 2 we have plotted the variation of the experimental quantity $M^{\gamma}_{exp.}(n_{Ho} + n_{Lu})$, at 10 K and 12 T, against n_{Ho}, where n_{Ho} and n_{Lu} are the numbers of a.p. along the $\boldsymbol{c}$ axis of the Ho/Lu biblock. In the same figure we show the fit of $M^{\gamma}_{exp.}(n_{Ho} + n_{Lu})$ by $M^{\gamma}_{th.} n_{Ho}$ where $M^{\gamma}_{th.}$ is given by (4), taking for $e' = e = -0.020$ [13], i.e. assuming perfect epitaxy. The factors multiplying $M^{\gamma}_{exp.}$ and $M^{\gamma}_{th.}$ are the result of multiplying $M^{\gamma}_{th.}$ by $n_{Ho}/(n_{Ho} + n_{Lu})$ to normalize to the unit volume of SL, in order to compare with the experiment. The parameters ensuing from the fitting are: $M^{\gamma}_{v0} = +0.275$ GPa, $M^{\gamma}_{s}/(c/2) = -7.0$ GPa and $D^{\gamma}_{v} = -116$ GPa, where c is the $\boldsymbol{c}$ axis Ho lattice parameter in the SL. The interface magnetoelastic stress σ_s is very *strong* compared with the volume MS, up to 6.4 times larger for $n_{Ho} = 8$ and of the opposite sign. Also the misfit stress, σ_{ε}, is strong, up to $5.5M^{\gamma}_{v0}$ for $n_{Ho} = 8$, because the misfit e is very large. This contribution should be negligible in the bulk; therefore measuring the MEL stress in the SLs is a way to evidence such a non-linear effect on the MEL energy. Finally notice that the value of M^{γ}_{v0} is the same as for bulk Ho, within the experimental error [14]. An attempt to fit $M^{\gamma}_{exp.}(n_{Ho} + n_{Lu})$ adding to (4) a term reflecting the possible dependence of M^{γ}_{s} on the epitaxial strain, i.e. $2D^{\gamma}_{s} e' t_{Lu}/t_{Ho}(\alpha t_{Ho} + t_{Lu})$, was unsuccessful, apparently showing the insensitivity of the interface MS to the epitaxial strain.

Before discussing any interpretation of the large interface MS we should address the origin of the MEL coupling in the present SLs. Dealing with RE the origin should be sought in the interaction of the Ho^{3+} ion with the distorted *crystal electric field*. It is well known that at any temperature M^{γ}_{v0} and D^{γ}_{v} should vary as $\hat{I}_{5/2}(L^{-1}(m))$, where m is the reduced saturation magnetization $m \equiv M(T)/M(0)$, $\hat{I}_{5/2}$, the reduced Bessel function and L^{-1}, the inverse Langevin function [15]. On the other hand, at the interface the low-temperature m^3 power dependence for the MS should be changed to m^4, if the spin dimension were reduced to two (2D) [16]. Therefore M^{γ}_{s} should vary as m^4 and as m^2 at low and high temperatures respectively [15, 16]. In seeking this origin, in figure 3, as some examples, we plot the thermal variation, at 12 T, of $M^{\gamma}_{exp.}(n_{Ho} + n_{Lu})/n_{Ho}$, now normalizing to the Ho volume in the biblock, for the superlattices Ho_n/Lu_{15} with $n = 14$, 30, 40, 45. In the same graph we plot the expected MS stress thermal variation

$$M^{\gamma}_{th.}(m) = (M^{\gamma}_{v0} + D^{\gamma}_{v}\epsilon)\hat{I}_{5/2}(L^{-1}(m)) + \frac{2M^{\gamma}_{s}}{t_{Ho}} m^{\alpha} \tag{5}$$

using for M^{γ}_{v0}, D^{γ}_{v} and M^{γ}_{s} the *same* above-obtained values (slightly modified, less than 6%) and taking $\alpha = 4$ below a certain temperature and $\alpha = 2$ above it (see the temperatures in the caption of figure 3). The fits obtained are reasonably satisfactory. This confirms the relevance of the *interfacial* stress contribution to the magnetoelastic stress.

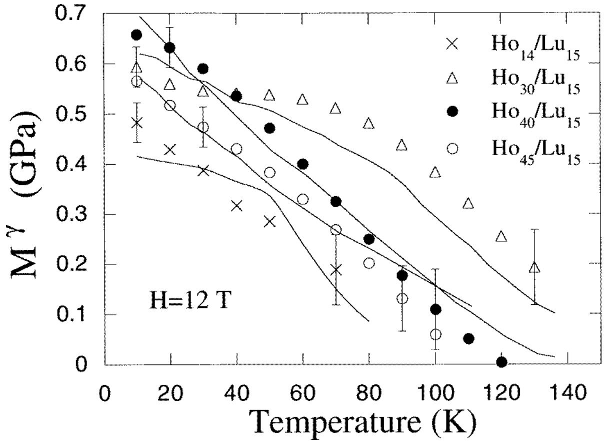

Figure 3. The variation with temperature of the magnetoelastic stress (MS) $M^{\gamma}_{exp.}(12\ \mathrm{T})$, multiplied by $(n_{Ho}+n_{Lu})/n_{Ho}$, for the $(\mathrm{Ho}_n/\mathrm{Lu}_{15})\times 50$ superlattices, with $n_{Ho}=14$ (×), 30 (△), 40 (●), 45 (○). The lines are the scalings with the reduced magnetization, $m(T)$, according to (5), with the *same* set of MEL parameters given in the text. $\alpha=4$ for temperatures smaller than 50, 80, 55 and 40 K (for $n_{Ho}=14$, 30, 40 and 45, respectively) and $\alpha=2$ for temperatures respectively higher than the above values. This particular $m^{\alpha}(T)$ dependence strongly indicates the existence of the interface MS.

In order to obtain further insight into the origin of the large interfacial MS we have performed theoretical calculations of M^{γ}_{v0} and M^{γ}_{s} [17]. Our model assumes that the origin of both MEL stresses stems from a variation of the Ho^{3+} crystal electric field energy because of the lattice deformation by magnetostriction. The calculation distinguishes between the Ho^{3+} ion at the Ho/Lu interface and within the Ho block. To start, the simple point charge model fails, giving for $M^{\gamma}_{s}/(c/2)$ a value -1.4% smaller than M^{γ}_{v0} and of the same sign. We have then considered a Gaussian charge distribution for the Ho^{3+} ligands and screened the deformed CEF by the conduction band electrons [18]. For bare ions and also for the free electron approximation for screening the model also fails. However screening the deformed CEF under the Hartree–Fock or Linhard approximation [18] for the reciprocal space dielectric constant explains very satisfactorily the found values for M^{γ}_{v0} and $M^{\gamma}_{s}/(c/2)$. To some extent the success of the mentioned theoretical explanation gives further support to the analysis of the MS results by (4), and to the finding of values for M^{γ}_{v0} and $M^{\gamma}_{s}/(c/2)$, differing by one order of magnitude and being of opposite signs.

Summarizing, we have shown that for $(\mathrm{Ho}_n/\mathrm{Lu}_{15})\times 50$ superlattices, where n ranges between 8 and 85 (0001) atomic planes a *strong interfacial* contribution σ_s (up to about six times larger than the volume one for $n_{Ho}=8$) to the h.c.p. basal plane cylindrical symmetry breaking magnetoelastic stress appears, being also of the opposite sign to the volume contribution. Also the large epitaxial strain increases the unstrained volume magnetoelastic stress M^{γ}_{v0} by a contribution, σ_{ε}, up to five times larger than M^{γ}_{v0} for $n_{Ho}=8$. The thermal variation of the overall magnetoelastic stress, M^{γ}, provides further credit to the CEF origin for M^{γ}_{v0} and M^{γ}_{s}.

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