

Magnetocrystalline and magnetoelastic basal plane anisotropies in Ho/Lu superlattices

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We report the temperature dependence of the effective magnetic anisotropy constant in the basal plane of the hcp structure ($K_6^{6,\text{eff}}$) of Ho/Lu superlattices. High-sensitivity vibration sample vector magnetometry was used to obtain the magnetic torque curves from which $K_6^{6,\text{eff}}$ was determined. It is shown, in the framework of the single-ion theory, the paramount role of the magnetoelastic contribution to $K_6^{6,\text{eff}}$ through both the magnetostrictive and epitaxial strains. The presence of epitaxial strain gives rise to a magnetoelastic contribution that reduces the strength of $K_6^{6,\text{eff}}$.

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Magnetic rare-earth superlattices (SL's) have attracted much interest¹⁻⁹ because they provide a unique manner of probing the nature of the mechanisms of interaction in localized magnetic moment systems. The main features of these artificial structures are the following: helical magnetic order is found to propagate even through nonmagnetic blocks and different magnetic phases are identified when a comparison with the bulk system is made. The induced strain, in the crystalline structure, by the mismatch between blocks of different elements is thought to be the origin of the new magnetic phases. The structural strain is coupled to the magnetization either by modifying the indirect exchange or by changing the energy balance between the exchange and magnetoelastic contributions. The earlier mechanism accounts for the suppression of the conical *c*-axis ferromagnetic transition in Er/Y (Ref. 3) and Ho/Y,⁴ and the latter one for the enhancement of the Curie temperature in Dy/Lu,⁵ when Dy lattice is compressed. Transition-metal-based thin films and SL's shown surface and strain contributions to the bulk magnetic anisotropy.^{10,11} Thus, it is also expected that the magnetic anisotropy constants would be altered in rare-earth SL's with respect to the bulk values due to the large magnetoelastic (ME) stress coefficients of rare earths. Moreover, because the thermal dependence of magnetic anisotropy and ME stresses in rare earths is well explained by using the single-ion theory,¹² the study of the temperature variation of the magnetic anisotropy arises as a powerful method to identify the physical origin of those new contributions to the magnetic anisotropy energy in thin films and SL's.

In this Brief Report, we report on torque curves measurements performed on RE SL's, which give account of the behavior of the magnetic anisotropy on the basal plane (BP) (0001) of the hcp structure, as a function of the temperature and rare-earth (RE) block thickness. The studied samples are $[\text{Ho}_n/\text{Lu}_{15}]_{50}$ ($n=14, 22, 30, 35, 40, 45, 85$), where n and 15 are the number of atomic layers of Ho and Lu, respectively, and 50 is the number of repetitions of the biblock. Both crystallize in an hexagonal unit cell with space group D_{3h} and the lattice parameters are $a_{\text{Ho}}=3.578 \text{ \AA}$, $c_{\text{Ho}}=5.618 \text{ \AA}$, $a_{\text{Lu}}=3.505 \text{ \AA}$, and $c_{\text{Lu}}=5.549 \text{ \AA}$. The mismatch in the basal-

plane, the epitaxial growth plane, is $e_o=(a_{\text{Ho}}-a_{\text{Lu}})/a_{\text{Ho}}=0.0204$. The growing procedure of the samples can be found elsewhere.⁴

The magnetic torque curves were performed in a ‘‘home-made’’ vector vibrating sample magnetometer,¹³ in which the sample is rotated with respect to the applied magnetic field and provides the longitudinal M_{\parallel} and transversal M_{\perp} components of the magnetization on the rotation plane, as a function of the experimental rotation angle ϕ . The magnetic field range is $\pm 20 \text{ kG}$ and temperature ranges between 4.2 and 300 K. The resolution is better than $2 \times 10^{-7} \text{ emu}$ and the sensitivity is $5 \times 10^{-6} \text{ emu}$. In Fig. 1, as an example, it is shown the longitudinal and transversal components of \mathbf{M} for the $[\text{Ho}_{14}/\text{Lu}_{15}]_{50}$ SL at three temperatures. In such a way, magnetic torque is determined straightly from the transversal component of the magnetization. If \mathbf{M} and \mathbf{B} are assumed to make angles ϕ and φ , respectively, with a coplanar direction (easy axis) in the crystal (sample) and α is the angle that

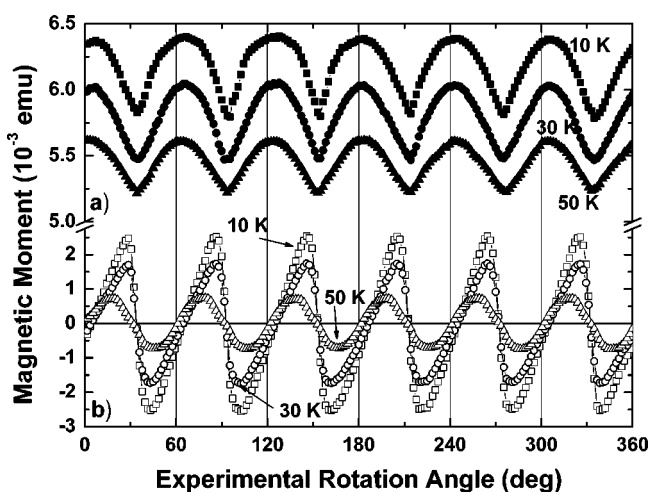


FIG. 1. (a) Longitudinal and (b) transversal (proportional to the magnetic torque) components of magnetization with respect to the applied magnetic field (20 kOe) on the rotation plane (0001) for a $[\text{Ho}_{14}/\text{Lu}_{15}]_{50}$ superlattice.

makes \mathbf{M} with \mathbf{B} , then, at equilibrium, the torque balance per unit volume of the crystal (neglecting demagnetization energy) is given by $\partial E_{\text{total}}/\partial\phi=0=-BM \sin \alpha + \partial E_A/\partial\phi$, where E_{total} is the sum of the Zeeman energy term plus an effective anisotropy energy density E_A . Then, the magnetic anisotropy torque is

$$L(\phi) = -\frac{\partial E_A}{\partial\phi} = -BM \sin \alpha = -M_{\perp}(\alpha)B. \quad (1)$$

Experimental measurements provide $M_{\perp}(\phi)$ and, to express the exerted torque by the sample as a function of angle ϕ , we make use of the relationship $\phi = \alpha - \varphi$. In our case, the angle α is also experimentally determined from $M_{\parallel}(\varphi)$ and $M_{\perp}(\varphi)$ as follows: $\alpha(\varphi) = \arctan [M_{\perp}(\varphi)/M_{\parallel}(\varphi)]$. Therefore, no assumption about the value of the magnetization at saturation is necessary, unlike in conventional torque meters.¹⁴

For a crystal of hexagonal symmetry, the free energy density is written in terms of the spherical coordinates (θ, ϕ) , which specify the magnetization direction relative to the crystallographic axes, as $F(\theta, \phi) = F_0(\theta, \phi) + K_2^{\text{eff}} P_2(\cos \theta) + K_4^{\text{eff}} P_4(\cos \theta) + K_6^{\text{eff}} P_6(\cos \theta) + K_6^{\text{eff}} \sin^6 \theta \cos 6\phi$, where F_0 includes the magnetostatic and exchange energy densities. The effective macroscopic anisotropy constants K_2^{eff} , K_4^{eff} , K_6^{eff} , and K_6^{eff} include magnetocrystalline (MC) and ME contributions.¹² $P_n(\cos \theta)$ are the Legendre polynomials of n order, θ and ϕ are, respectively, the angles between the magnetization and the \mathbf{c} axis, and between the projection of the magnetization in the BP and the \mathbf{a} axis. In our experimental situation the magnetic field, applied in the BP, is large enough to reach the ferromagnetic phase for all the studied SL's.¹⁵ Thus, $\theta = \pi/2$ and then the torque exerted by the sample can be written as $L(\phi) = -\partial F/\partial\phi = 6K_6^{\text{eff}} \sin(6\phi)$. Now, from the experimental torque values, we can obtain the values of K_6^{eff} at different temperatures and fields following the standard torque analysis methods.^{16,17} It was determined the magnetic torque for three different fields (15, 17.5, and 20 kG), between 10 and 90 K. At some temperatures, we obtained the magnetic torque at four fields to be sure of the quality of the $B^{-1}=0$ extrapolation.¹⁶ It can be observed that, below 30 K, the magnetic anisotropy is so large that, even at 20 kG, $M_{\perp}(\varphi)$ is sheared (see Fig. 1). The magnetization curves show sixfold symmetry, which is expected for hexagonal rare earths when torque is measured in the BP of the hcp structure.

As said above, K_6^{eff} contains MC and ME contributions due to the large ME coupling in the rare earths (which play a relevant role in their magnetic properties). As is well explained in Ref. 12, the hexagonal symmetry is broken in the BP by orthorhombic strains (γ strains), which for rare earths can be large and are the origin of the main ME contribution to K_6^{eff} . For the bulk material, the magnetic behavior is well explained in terms of the single-ion theory;¹² thus, it can be shown that the minimization of the free energy density with respect to the γ strains leads to the following expression for the temperature dependence of the volume anisotropy constant:

$$K_{6,V}^6(T) = K_{6,V}^{6,MC} \hat{I}_{13/2}[\hat{m}] + K_{6,V}^{6,ME} \hat{I}_{5/2}[\hat{m}] \hat{I}_{9/2}[\hat{m}], \quad (2)$$

where $\hat{I}_{l+1/2}$ are the reduced hyperbolic Bessel functions $\hat{m} = \mathcal{L}^{-1}[m(T)]$, \mathcal{L}^{-1} being the inverse Langevin function and m the reduced magnetization of the Ho blocks [$m = M(T)/M(0)$].¹⁸⁻²⁰ $K_{6,V}^{6,MC}$ is the MC anisotropy constant and $K_{6,V}^{6,ME} = -B^{\gamma^2} B^{\gamma^4} (c_{11} - c_{12})^{-1}/2$, is the ME anisotropy constant, B^{γ^2} and B^{γ^4} are the macroscopic ME parameters associated with the orthorhombic distortion in the BP, and c_{11} , c_{12} are Cartesian elastic constants. We have determined the BP magnetic anisotropy constant for a 500 nm thick film of Ho.²¹ Using Eq. (2) to analyze its temperature variation, we get $K_{6,V}^{6,MC} = 4.8 \times 10^6 \text{ erg/cm}^3$ and $K_{6,V}^{6,ME} = 9.4 \times 10^6 \text{ erg/cm}^3$. The purpose is to obtain reference values for $K_{6,V}^{6,MC}$ and $K_{6,V}^{6,ME}$, that is, not affected by the influence of strain and interfaces on the magnetism of the holmium blocks in the SL. We have also attempted to perform a fit of the temperature dependence of $K_{6,V}^6$ for bulk Ho, but now deduced from magnetization measurements along the \mathbf{a} and \mathbf{b} axes of Ref. 22, the low resolution of this method to obtain $K_{6,V}^6$ results in an unsatisfactory fit.

Another ME contribution to $K_{6,V}^6$ comes from the fact that the strain state in thin films and superlattices depends strongly on the thickness of the magnetic species. In the $[\text{Ho}_n/\text{Lu}_{15}]_{50}$ series the lattice symmetry is preserved, as the holmium lattice is compressed in the basal plane and enlarged along the c axis. That deformation results in volume ϵ_{a1} and tetragonal ϵ_{a2} strains that quantify the difference between the lattice parameters in the SL's with respect to those in the holmium bulk lattice. Therefore, in an hexagonal crystal, the terms in the K_6^6 coming from the ME energy with a sixfold dependence are $B_{a1}^{66} \epsilon_{a1} + B_{a2}^{66} \epsilon_{a2} (= K_{6,\text{strain}}^{6,MC})$,¹² where B_{a1}^{66} and B_{a2}^{66} are ME stress coefficients. The thermal dependence for these ME contributions is the $\hat{I}_{13/2}[\hat{m}]$ law.¹² Thus, in systems where the lattice parameters can be varied, the expression for $K_{6,V}^6$ is

$$K_{6,V}^6(T) = (K_{6,V}^{6,MC} + K_{6,\text{strain}}^{6,MC}) \hat{I}_{13/2}[\hat{m}] + K_{6,V}^{6,ME} \hat{I}_{5/2}[\hat{m}] \hat{I}_{9/2}[\hat{m}]. \quad (3)$$

Thus, from the measurements of $K_{6,V}^6$ as a function of the temperature it can be possible to separate the contribution due to the orthorhombic distortion in the basal plane $K_{6,V}^{6,ME}$, while the measurements of $K_{6,V}^6$ as a function of the holmium thickness allow us to distinguish the crystal field $K_{6,V}^{6,MC}$ from the epitaxial strain contribution $K_{6,\text{strain}}^{6,MC}$.

For the SLs, to take into account the contribution of the atomic layers that belong to the interfaces, which behave in a different manner with respect to the ones belonging to the inside of the Ho blocks, we have included in K_6^{eff} an interface term, $2K_{6,S}^6(T)/t_{\text{Ho}}$,²³ where $t_{\text{Ho}} (=nc_{\text{Ho}}/2)$ is the thickness of the Ho blocks. $K_{6,S}^6(T)$ follows a thermal dependence with a power of the reduced magnetization $m^{\alpha}(T)$, where, at low temperature ($m \approx 1$) $\alpha = 36$ and at high temperature ($m \ll 1$) $\alpha = 2$.²⁴ So that, the thermal dependence of K_6^{eff} reads

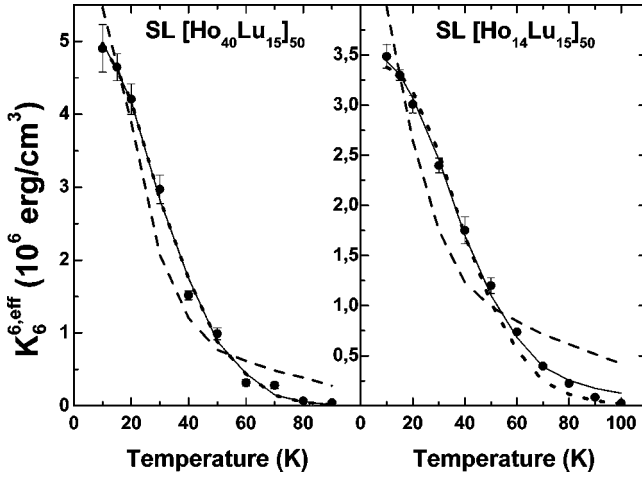


FIG. 2. Temperature dependence of the effective anisotropy constant K_6^{eff} for two $[\text{Ho}_n/\text{Lu}_{15}]_{50}$ superlattices. The lines represent three fittings, including different contributions to K_6^{eff} : (—) magnetocrystalline, magnetoelastic, and interfacial; (···) magnetocrystalline and magnetoelastic, and (---) magnetocrystalline and interfacial terms. The latter fails for all the samples. Just for the SL having thinner Ho blocks, $n=14$, there is a small difference between the first and second fittings at high temperatures ($T > 60$ K), for the rest of the samples both practically coincide.

$$K_6^{\text{eff}}(T) = K_{6,V}^6(T) + \frac{2K_{6,S}^6}{t_{\text{Ho}}} m^{\alpha}(T). \quad (4)$$

The analysis of the temperature variation of $K_6^{\text{eff}}(T)$ in the Ho/Lu SLs has been done by using Eq. (4). In Fig. 2, it is shown, as an example, the fittings for two SL's. The main conclusions obtained from our fits are (see values of $K_{6,\text{strain}}^{6,\text{MC}}$ and $K_{6,V}^{6,\text{ME}}$ in Table I): (a) $K_{6,\text{strain}}^{6,\text{MC}}$ is negative while the bulk value $K_{6,V}^{6,\text{ME}}$ is positive, (b) $K_{6,V}^{6,\text{ME}}$ does not change too much along the series, (c) the inclusion of an interfacial term does not substantially affect the quality of the fitting, except for the $n=14$ SL, for which taking $K_{6,S}^6(T) = -0.03 \text{ erg/cm}^2$ improves the fit at high temperatures (see Fig. 2), and (d) $K_{6,V}^{6,\text{ME}}$ is even larger than $|K_{6,\text{strain}}^{6,\text{MC}}|$.

It is worth pointing out that no simple MC term or MC plus surface terms account for the observed temperature variation of K_6^{eff} in the studied samples (see Fig. 2) and, as well, that the ME term is mandatory to explain the experimental data. We have also attempted to plot $K_6^{\text{eff}}/t_{\text{Ho}}$ vs t_{Ho} at each temperature (as is usually done) to obtain the interface contribution. From this analysis, it was not possible to draw any reliable conclusion about the interface term in these samples due to the existence of two large competing contributions MC and ME to the magnetic anisotropy. Probably,

TABLE I. Values of the MC and ME anisotropy constants, in $(\text{erg/cm}^3) \times 10^6$, for the $[\text{Ho}_n/\text{Lu}_{15}]_{50}$ superlattices.

n	14	22	30	35	40	45	85
$K_{6,\text{strain}}^{6,\text{MC}}$	-6.6	-6.2	-3.9	-3.7	-3.3	-3.0	-0.4
$K_{6,V}^{6,\text{ME}}$	10.0	9.4	10.2	9.5	8.5	9.6	6.6

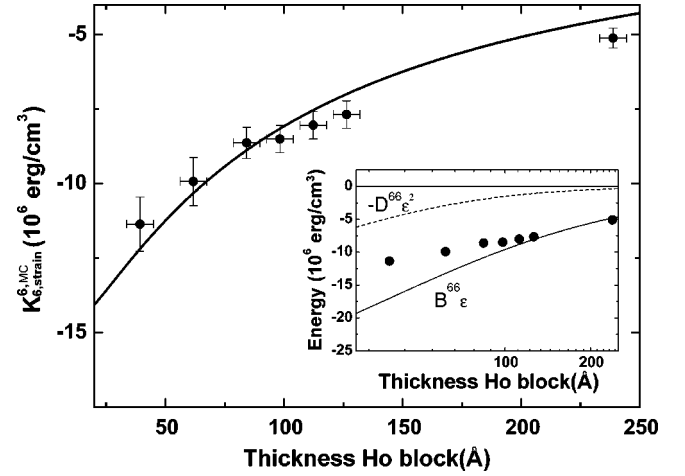


FIG. 3. Variation of $K_{6,\text{strain}}^{6,\text{MC}}$ vs Ho thickness. $K_{6,V}^{6,\text{MC}}$ is the value obtained from the Ho film. The continuous line is a fitting of the experimental data making use of an elastic model for the in-plane strain of Ho blocks (see text). The inset shows the $B^{66}\epsilon$ (continuous line) and $D^{66}\epsilon^2$ (dashed line) contributions to $K_{6,\text{strain}}^{6,\text{MC}}$.

heavy rare earths SL's are not the most suitable systems to study the influence of the surfaces/interfaces on the magnetic anisotropy. Therefore, we considered using the obtained values in the thermal fittings without a surface term.

Now, we examine the dependence of $K_{6,V}^{6,\text{ME}}$ and $K_{6,\text{strain}}^{6,\text{MC}}$, obtained from the thermal dependence analysis, with the holmium block thickness. It is remarkable that $K_{6,\text{strain}}^{6,\text{MC}}$ varies as the thickness of the Ho blocks is reduced from 85 to 14 atomic planes while $K_{6,V}^{6,\text{ME}}$ remains almost constant. To explain this fact, we should point out that $K_{6,V}^{6,\text{ME}}$ is related to the orthorhombic strain induced in the ferromagnetic phase, while $K_{6,\text{strain}}^{6,\text{MC}}$ is related to the $\epsilon_{\alpha 1}$ and $\epsilon_{\alpha 2}$ modes that do not break the hexagonal symmetry. Thus, the experimental data suggests the presence of a strain effect that does not break the hexagonal symmetry and therefore changes $K_{6,\text{strain}}^{6,\text{MC}}$ but not $K_{6,V}^{6,\text{ME}}$. This conclusion is supported by previous measurements of the lattice parameters in Ho/Lu SL's (Ref. 7) that describes the effect of the Lu blocks on the Ho ones as an isotropic compression within the BP and an expansion along the c axis.

Although it is clear that without the knowledge of $\epsilon_{\alpha 1}$ and $\epsilon_{\alpha 2}$ as a function of the Ho block it is difficult to analyze the thickness dependence of $K_{6,\text{strain}}^{6,\text{MC}}$, we have made an analysis by using a simple elastic model for the strain:¹⁵ the equilibrium basal-plane strain can be obtained from the minimization of the elastic energy density of a single bilayer resulting in the following expression for the in-plane strain of the Ho block: $\epsilon = e_0 t_{\text{Lu}} / (c t_{\text{Lu}} + t_{\text{Ho}})$, where e_0 is the mismatch between lattice parameters of Ho and Lu, c is a combination of elastic constants with a value close to 1, and t_{Ho} and t_{Lu} are the thicknesses of Ho and Lu blocks, respectively. If the out-of-plane strain is related to the in-plane strain by elastic constants, $K_{6,\text{strain}}^{6,\text{MC}}$ results in a contribution proportional to the in-plane strain: $B^{66}\epsilon$, where B^{66} is an effective coefficient that includes elastic constants and the $B_{\alpha 1}^{66}$ and $B_{\alpha 2}^{66}$ coefficients.

Figure 3 shows the dependence with t_{Ho} of $K_{6,\text{strain}}^{6,\text{MC}}$ that

has been evaluated taking for $K_{6,V}^{6,MC}$ the value obtained for the 500 nm thick holmium film. The expression $B^{66}\epsilon = B^{66}e_0 t_{Lu}/(ct_{Lu} + t_{Ho})$ fails to adjust the experimental data. In order to obtain a good fit, we have to include a second-order term $D^{66}\epsilon^2$ in $K_{6,strain}^{6,MC}$. Thus, the continuous line in Fig. 3 is obtained with $B^{66} = 0.16 \times 10^{10}$ erg/cm³ and $D^{66} = 4 \times 10^{10}$ erg/cm³. Notice that $B^{66}\epsilon$ is about four times $D^{66}\epsilon^2$ for $n_{Ho} = 14$, (see Fig. 3, inset). The physical origin for D^{66} would be the strong tetragonal distortion that the hcp lattice undergoes due to the epitaxial strain as the Ho thickness decreases. We have to note that the use of a model for $\epsilon(t_{Ho})$ may introduce an uncertainty in the analysis of $K_{6,strain}^{6,MC}$, since the actual dependence of $\epsilon(t_{Ho})$ is unknown. Thus, if $\epsilon(t_{Ho})$ would need a term such as $(ct_{Lu} + t_{Ho})^{-2} \propto \epsilon^2$ to fit the thickness dependence, then a term proportional to $B^{66}\epsilon^2$ will appear in the analysis of $K_{6,strain}^{6,MC}$. On the other hand, even without the exact knowledge of the strain state in the holmium block, second-order effects have been widely reported in the literature not only for rare earths but also for 3d metals such as nickel and cobalt¹¹ and can be expected in the magnetic anisotropy of rare earth SL's. However, whatever the origin for D^{66} is, we think that the reduction in the absolute value of $K_{6,strain}^{6,MC}$, see Table I, is explained by compression on the BP of the Ho blocks that increases as t_{Ho} decreases. As a result it

is observed that the epitaxial strain induces a competition between the MC and ME anisotropy energies, where MC anisotropy tends to align the magnetic moments along the **a** axis whereas ME anisotropy does it along the **b** axis.

Summarizing, we have obtained, by measuring the angular dependence of the magnetization vector, the magnetic anisotropy constant in the basal plane $K_6^{6,eff}$ for the superlattices $[Ho_n/Lu_{15}]_{50}$. From the temperature variation of the $K_6^{6,eff}$, we have separated its MC and ME contributions, showing the paramount role of the ME contribution to the magnetic anisotropy. The epitaxial strain not only decreases the absolute value of the magnetocrystalline anisotropy contribution to the total magnetic anisotropy in the basal plane, but also it induces a competition between magnetocrystalline and ME anisotropy energies. We have also obtained the ME stress coefficients B^{66} and D^{66} , which give an account of the tetragonal deformation in the basal plane in hexagonal symmetry.

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