Magnetization Process in Holmium: Easy Axis Spin Reorientation Induced by the Magnetostrictive Basal Plane Distortion

L. Benito,[1,*] M. Ciria,[1] C. de la Fuente,[1] J. I. Arnaudas,[1,†] R. C. C. Ward,[2] and M. R. Wells[2]

[1]*Departamento de Magnetismo de Sólidos-I.C.M.A., Departamento de Física de la Materia Condensada, and Instituto de Nanociencia de Aragón, Universidad de Zaragoza and C.S.I.C., 50071 Zaragoza, Spain*

[2]*Department of Physics, Clarendon Laboratory, Oxford OX1 3PU, United Kingdom*

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We report on the change of the easy axis direction in holmium, from the **a** to the **b** axis, under the application of a magnetic field in the basal plane. This spin reorientation is observed by measuring the magnetic torque in Ho_n/Lu_{15} superlattices (n and 15 are the number of atomic planes in the Ho and Lu blocks). We also observe that, at the field H_0 and temperature at which the reorientation occurs, both axes are easy directions. Based on the fact that the field H_0 depends on n in the same way as the field-induced magnetoelastic distortion does, we propose that this spin reorientation originates from the strong field-induced magnetoelastic deformation within the basal plane. The modulation of the α strains with sixfold symmetry originates a 12-fold term in the magnetic anisotropy energy.

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Field-induced magnetic order can be driven by decreasing the energy gap between quantum levels, as in quantum antiferromagnets [1], decreasing the Zeeman energy of one magnetic phase, as in the spin-flop transition [2], or modifying the magnetoelastic (ME) and exchange energies with respect to the zero field phase, which is the case of rare earth metals [3]. Thus, the field-induced spin reorientation can reveal the nature of the interactions that play a role in the magnetism of magnetic materials. For example, the Dzyaloshinskii-Moriya interaction has been addressed as the physical origin for the second order field-induced spin reorientation observed in weak antiferromagnetic oxides at temperatures below 4 K [4]. In rare earth metals, like terbium and holmium, the magnetic anisotropy energy density, e_a, is comprised of exchange, crystal-field, and ME energies, all of them with different strengths and temperature dependencies [5,6]. The strain dependence of the ME contribution to e_a opens up the possibility of studying the magnetic anisotropy in magnetic phases with different irreducible deformations and, therefore, different contributions to e_a: ferromagnetic phase with an orthorhombic distortion and helical phase with an undistorted hexagonal crystal lattice (clamping effect) [7]. Thus, a field-induced spin reorientation of the easy axis in the plane could be expected as long as the magnetic anisotropies in the ferromagnetic and helical phases had opposite sign.

In this Letter, we report on the observation of such a kind of spin reorientation in Ho_n/Lu_{15} superlattices (SL's), where the subscripts stand for atomic planes and $8 < n < 85$. Magnetic torque measurements, as a function of the applied magnetic field H_a, indicate that the absolute minimum of e_a depends on H_a in such a way that the **a** axis becomes easier than the **b** axis for H_a below a certain critical field H_0. Based on the dependencies of H_0 and the ME stress with n, we propose that the field-induced ME distortion, which is not a linear function of H_a, is the origin of the spin reorientation. Our experiments also indicate that, depending on H_a and T, not only **b** but also the **a** axis are simultaneously magnetic easy directions. This observation implies the existence of a 12-fold symmetry in e_a. We suggest that the second harmonic of the sixfold symmetry stems from the elastic response to the sixfold ME distortion that preserves the hexagonal symmetry.

In holmium, neutron diffraction and x-ray magnetic scattering experiments [8–11] show, at zero field, a basal plane helix below the Néel temperature $T_N = 132$ K. At $T_C = 18$ K a ferromagnetic component along the **c** axis is observed, forming a cone structure [8,11] that is suppressed in Ho/Lu [12] and Ho/Y [13] SL's. At low temperature, spin-slips, with the magnetic moments along the **b** axis, are observed [10]. The application of a magnetic field within the basal plane may change the helix phase into fan and helifan structures (depending on the temperature) before the Ho layers become ferromagnetic [10]. In Ho/Y SL's the ferromagnetic phase takes place at field values higher [14] than those observed for bulk Ho [9] at the same temperature; in Ho/Lu SL's the opposite occurs [15]. The latter SL's also show ferromagnetic order in the basal plane of the Ho blocks for $n < 20$, at temperatures below 40 K [12]. Neutron scattering experiments suggest that the ferromagnetic transition can only proceed after the helix becomes commensurate with the wave vector $(1/6)c^\star$ [16], where $c^\star$ is the reciprocal lattice vector.

The SL's studied in this work have been grown by using a molecular beam epitaxy system. The growing procedure for which the description can be found elsewhere [13], is designed to obtain holmium and lutetium layers with the basal plane parallel to the substrate plane. The measurement of the magnetic anisotropy was performed by means of a vectorial magnetometer by applying the magnetic field within the basal plane. The magnetometer measures the in-plane components of the magnetization, **M**; longitudinal

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$M_{\parallel}$, and transversal $M_{\perp}$, to H_a, as a function of the azimuthal angle ϕ, formed by **M** and the **a** axis. The sample is rotated 360 degrees about the **c** axis, although in the figures, for clarity, only one third of the rotation is shown. The applied field ranges from 10 to 20 kOe and the temperature is varied between 10 and 100 K. The anisotropy constants are obtained from the Fourier analysis of the magnetic torque: $\Gamma_m = H_a M_{\perp} = -\partial e_a/\partial\phi$.

In a previous paper [17] we have determined, in the Ho_n/Lu_{15} series, the magnetic anisotropy constant associated with the in-plane anisotropy K_6^6. The dependence of K_6^6 with n is explained including a term due to the ME distortion that breaks the hexagonal symmetry by γ strains, K_γ, and crystal-field and ME terms invariable under the α representation of the hexagonal symmetry, K_α [17,18]. The latter ME terms preserve the hexagonal symmetry since they are associated with the isotropic expansion $\epsilon_{\alpha 1}$, and with the tetragonal distortion that changes the c/a ratio, $\epsilon_{\alpha 2}$. The K_γ term has a positive value, which means that the **b** axis is the easy direction, independently of n, but K_α could be either positive or negative depending strongly on the epitaxial strain induced on the holmium layers. Nevertheless, K_6^6 at 0 K is positive for all the samples studied [17]. This fact is presented in Fig. 1, which shows $M_{\parallel}(\phi)$ and $M_{\perp}(\phi)$ for two SL's with $n = 22$ and 85, at $T = 30$ K for $H_a = 20$ kOe; at this field **M** is saturated and the SL's are in a ferromagnetic state. We observe that the curves have a sixfold periodicity: $M_{\parallel}$ has a maximum every 60 degrees and $M_{\perp}$ crosses through zero at the angles for which $M_{\parallel}$ is either maximum or minimum. In this representation a negative slope in the $M_{\perp}(\phi)$ curve means that e_a has a minimum along the corresponding crystallographic direction, that for the curves in Fig. 1 is the **b** axis.

At temperatures higher than 30 K we observe that the $M_{\perp}(\phi)$ curves exhibit a shoulder between two easy directions, as it is illustrated in Fig. 2(a) for the Ho_{30}/Lu_{15} SL, for $H_a = 20$ kOe and $T = 50$ K, at the direction where the **a** axis is located. Modeling this feature requires the introduction of a 12-fold contribution in $e_a(\phi)$. Eventually, for lower fields [see Fig. 2(b), $H_a = 15$ kOe] or higher temperatures [see Fig. 2(c) for $H_a = 20$ kOe and Fig. 2(d) for $H_a = 15$ kOe, at $T = 60$ K], the $M_{\perp}(\phi)$ curve around the **a** axis becomes characteristic of an easy direction since the slope of $M_{\perp}$ is negative, as happens at the **b** axis. Thus, the observation of not six but 12 easy directions for **M** in the plane suggests the existence of a 12-fold symmetry. To determine the easier axis, besides analyzing the torque curve, $M_{\parallel}(\phi)$ can be evaluated. Thus, from Fig. 3 that shows $M_{\parallel}(\phi)$ and $M_{\perp}(\phi)$, at 50 K, for the Ho_{85}/Lu_{15} SL, for $H_a = 12.5$, 15, 17.5, and 20 kOe, a shift in the maxima of $M_{\parallel}(\phi)$ from the **a** axis at 12.5 kOe [see Fig. 3(a)] to the **b** axis at the remaining H_a values, Figs. 3(b)–3(d), prove the existence of a field-induced spin reorientation.

The anisotropy energy density, relative to the basal plane, is written as: $e_a(\phi) = K_6^6 \cos 6\phi + K_{12}^{12} \cos 12\phi$. Since $M_{\perp} = \Gamma_m(\phi)/H_a$, the fit of $(1/H_a)(-\partial e_a(\phi)/\partial\phi)$ to $M_{\perp}(\phi)$ [see the continuous line in Figs. 3 and 4] provides the values of K_6^6 and K_{12}^{12} for each H_a and T. The data indicate that K_6^6 changes from negative to positive values with increasing H_a. Figure 4 shows, as a function of n, for $T = 50$ K, the values of H_a for which $K_6^6 = 0$, namely H_0 (solid circles), and the critical fields [19] that separate magnetic phases (open circles). For $n < 30$ there is only one critical field marking the transition from the helix to the ferromagnetic state, if $n > 30$ two points are identified, which are interpreted as the boundaries for an intermediate fan phase. We note that in bulk Ho the fan phase appears at about 40 K [9]. Also in Fig. 4, the lines correspond to the field values for which the ME γ stress is about 15%, 85%, and 90% of the saturation value [20]. We observe that H_0 is located in the area of the ferromagnetic phase and follows the trend of the ME stress, suggesting a correlation between H_0 and the ME stress. Based on this observation, we propose that the spin reorientation stems from the variation of the ME anisotropy energy due to the strong γ-lattice distortion induced by the applied field, which favors the **b** axis as an absolute minimum in e_a.

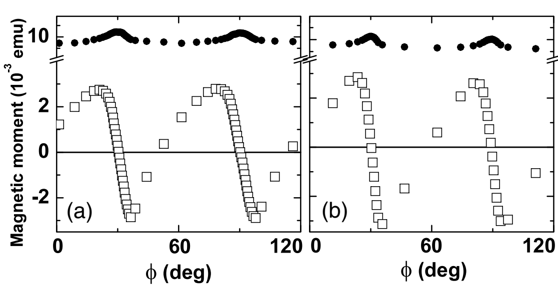

FIG. 1. Longitudinal, $M_{\parallel}$ (solid circles), and transversal, (open squares) $M_{\perp}$, (proportional to the magnetic torque) components of **M** with respect to the applied magnetic field on the plane at $T = 30$ K and for $H_a = 20$ kOe for (a) Ho_{22}/Lu_{15} and (b) Ho_{85}/Lu_{15} (ϕ is the angle formed by **M** and the **a** axis).

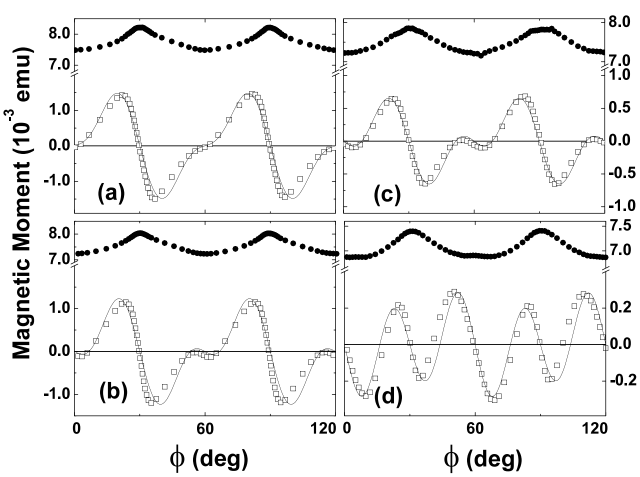

FIG. 2. $M_{\parallel}$ (solid circles) and $M_{\perp}$ (open squares), for a Ho_{30}/Lu_{15} superlattice at $T = 50$ K and H_a (a) 20 kOe, (b) 15 kOe and $T = 60$ K (c) 20 kOe, (d) 15 kOe.

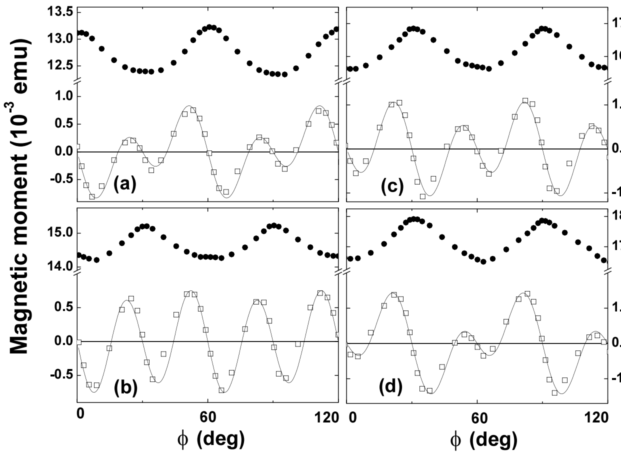

FIG. 3. $M_\parallel$ (solid circles) and $M_\perp$ (open squares), for a Ho_{85}/Lu_{15} superlattice at $T = 50$ K and H_a (a) 12.5 kOe, (b) 15 kOe, (c) 17.5 kOe, and (d) 20 kOe.

Figure 5 shows the dimensionless surface $e(\phi) = K\cos 6\phi - \cos 12\phi$, for K ranging from -8 to 8; $e(\phi)$ can be related to $e_a(\phi)$ by changing $K \rightarrow K_6^6/|K_{12}^{12}|$ and $e(\phi) \rightarrow e_a(\phi)/|K_{12}^{12}|$. Thus, assuming that e_a had the angular dependence drawn in Fig. 5, the $\Gamma_m(\phi)$ curves measured for the Ho/Lu SL's could be obtained in terms of a single parameter. For $K > 4$, the minima and maxima of e are at the **b** and **a** directions, respectively. This case corresponds to the curves presented in Fig. 1. For $K \gtrsim 4$ the maxima of $e(\phi)$ at the **a** directions are quite flat, while below $K = 4$ (see Fig. 5) the maxima split into two peaks whose angular separation increases as K decreases, appearing minima at the **a** directions. To this region in the $e(\phi)$ surface corresponds the measurements showed in Figs. 2(a)–2(c), 3(c), and 3(d). Because of the presence of a 12-fold symmetry, at $K = 0$, $e(\phi)$ shows a clear $\cos 12\phi$ shape, as it is reflected in the torque curves of Figs. 2(d) and 3(b). For $K < 0$, the absolute minima of the $e(\phi)$ is at the **a** axis; this is the situation shown in Fig. 3(a). For $K < -4$, the **b** direction becomes the maxima of $e(\phi)$. The main physical reason for changing K can be associated with the contribution to K_6^6 due to the γ-ME stress of the Ho/Lu SL's (K_γ). Thus, starting at the field-induced ferromagnetic state at 0 K, if H_a decreases or T increases, the ME γ distortion decreases and also K_γ. Therefore, K_α, which favors the **a** axis as minimum in e_a, may overcome K_γ because of the much stronger field dependence of the γ strain compared with the α strain. Thus, this competition between ME contributions to e_a yields the field-induced spin reorientation observed in Fig. 3.

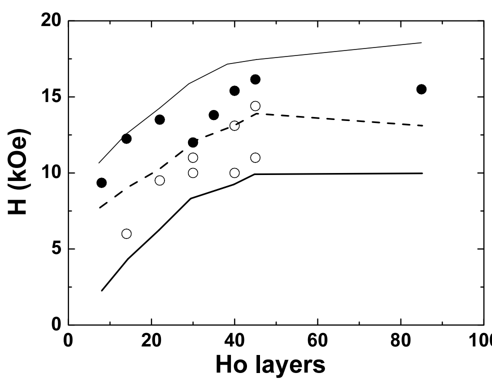

FIG. 4. H_0 (solid circles) and critical fields for magnetic phase transitions (open circles) for Ho_n/Lu_{15} SL's as function of the holmium thick spacer n. The lines represent the field for which the ME γ stress is about 15% (thick continuous line), 85% (dashed line) and 90% (thin continuous line) of the saturation value. All the quantities are evaluated at $T = 50$ K.

In order to explain the presence of the 12-fold symmetry, we consider the existence of a sixfold ME stress and the accompanying elastic reaction. This intrinsic contribution has its maximum strength at the **a** and **b** axes that are the directions at which the ME stress produces the maximum striction, either with tensile or compressive character. This contribution is usually described for cubic materials [21], but can be straightforwardly extended to the hexagonal symmetry. We take as starting point the elastic and ME energy densities belonging to the α representation, Γ^α. By symmetry considerations these strains may contain a $\cos 6\phi$ term. Thus, if **M** is within the basal plane, the sum of the ME and elastic energy densities belonging to Γ^α is $e_\alpha = -(B_{\alpha 1}^{66}\epsilon_{\alpha 1} + B_{\alpha 2}^{66}\epsilon_{\alpha 2})\cos 6\phi + (1/2)c_{\alpha 1}\epsilon_{\alpha 1}^2 + c_{\alpha 3}\epsilon_{\alpha 1}\epsilon_{\alpha 2} + (1/2)c_{\alpha 2}\epsilon_{\alpha 2}^2 + \mathcal{B}$ [5]. The B's and the c's are, respectively, phenomenological irreducible ME and elastic coefficients, and $\mathcal{B}$ includes ME terms without a ϕ dependence. Minimization of e_α provides the equilibrium strains (magnetostriction), $\bar{\epsilon}_\alpha$, which are

$$\bar{\epsilon}_{\alpha 1} = \frac{B_{\alpha 1}^{66}c_{\alpha 2} - B_{\alpha 2}^{66}c_{\alpha 3}}{c_{\alpha 1}c_{\alpha 2} - c_{\alpha 3}^2}\cos 6\phi + \bar{\mathcal{B}}_{\alpha 1} \qquad (1)$$

where $\bar{\mathcal{B}}_{\alpha 1}$ does not depend on ϕ [$\bar{\epsilon}_{\alpha 2}$ is obtained by exchanging $\alpha 1 \leftrightarrow \alpha 2$ in the subscripts of Eq. (1)]. Now, inserting the equilibrium strains in e_α, an expression like

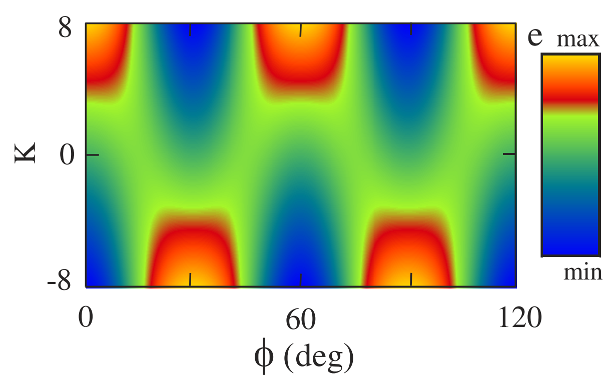

FIG. 5 (color online). Dimensionless density energy surface, $e(\phi) = K\cos 6\phi - \cos 12\phi$. This surface can be related to $e_a(\phi)$ if $K = K_6^6/|K_{12}^{12}|$ and $e(\phi) = e_a(\phi)/|K_{12}^{12}|$.

$K^6_{6,\mathrm{ME}}\cos 6\phi + K^{12}_{12}\cos 12\phi$ is obtained, with

$$K^{12}_{12} = \frac{2B^{66}_{\alpha 1}B^{66}_{\alpha 2}c_{\alpha 3} - (B^{66}_{\alpha 1})^2 c_{\alpha 2} - (B^{66}_{\alpha 2})^2 c_{\alpha 1}}{4(c_{\alpha 1}c_{\alpha 2} - c^2_{\alpha 3})}. \quad (2)$$

Sixfold exchange anisotropy has been reported in other rare earth like terbium [6] and cannot be excluded in the analysis of the spin reorientation observed in the Ho/Lu SL's. For this reason the K and B coefficients reflect the hexagonal symmetry in a phenomenological way, since including the exchange anisotropy is a complex issue. For instance, in terbium, the strength of this contribution depends not only on the temperature but also on the **q** vector of the spin wave excitations [6].

The complex phase diagram of holmium [9] has been described assuming that the **a** axis is a hard direction. Nevertheless, we have shown that in the helix and fan phases, which correspond to a field interval in which the spin reorientation is observed, the magnetic state has both the **a** and **b** directions as easy axes. We think that this observation provides a new framework to explain the Ho phase diagram for magnetic field applied in the basal plane since, as deduced from our experiments, the field variation of the magnetic anisotropy energy has to be taken into account to determine the stable structure that satisfies the exchange and anisotropy energies in bulk Ho and SL's.

In conclusion, a spin reorientation, from the **a** to **b** axis, is identified as a function of the strength of the applied magnetic field in the plane. We explain this magnetic phase transition as a result of the competition between the ME and magnetocrystalline contributions to the hexagonal magnetic anisotropy. An unexpected symmetry, that corresponds to the second harmonic of the sixfold plane symmetry, is also observed. We propose that its origin is the modulation of the ME strain, belonging to Γ^{α}, together with the hexagonal symmetry and the strong ME coupling. The main result is that, contrary to conventional wisdom, the magnetic easy axes directions can simultaneously be along the **a** and **b** axes due to the ME strain.

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*Present address: Cavendish Laboratory, Department of Physics, University of Cambridge
†Corresponding author: arnaudas@unizar.es

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