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(H, T) PHASE DIAGRAM OF A UNIAXIAL DIPOLAR FERROMAGNET: LiHoF4

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Abstract. – We report on the first determination of the (H, T) magnetic phase diagram of a uniaxial dipolar ferromagnet: LiHoF₄ below T_c =1.53 K. It is deduced from the evolution of the magnetic domain pattern (branched stripes structure, bubbles), visualized by Faraday rotation down to 1.3 K.

The domain structure and magnetic phase diagram of thin ferromagnetic films have been extensively studied in the two last decades but only few data was reported on highly anisotropic thick magnets. In this paper we present the (H,T) phase diagram and the evolution of the field dependent magnetic domain structure in thick slabs of an uniaxial dipolar ferromagnet, LiHoF₄. Results are discussed from the theories [1-4] valid for an Ising magnet confined in a slab of thickness D with magnetization normal to the sheet, which rules out closure domains but accounts for domain branching [3-7].

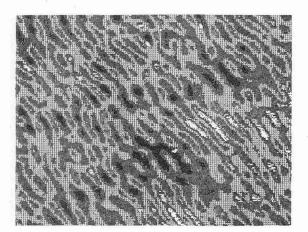
As shown by Kittel [8], thin samples ($D < D_c \sim 0.2~\mu m$ for LiHoF₄ [4]) in zero field must spontaneously order in a stripe domain pattern. If an upward magnetic field is applied the "up" domains grow at the expense of the "down" ones by domain wall movements. Since stripe domains cannot shrink to zero, they suddenly transform into cylindrical domains or "bubbles" above a critical field $H_{\rm B}^+$. These bubbles are stable over a narrow field range above $H_{\rm B}^+$ and collapse suddenly at a field H_c^+ associated with a further transition towards the uniformly magnetized state. In thicker samples ($D > D_c$) it is more favourable to form surface or branched domains of reverse magnetization inside the stripes [3-7].

LiHoF₄ is optically transparent and, below $T_c = 1.53$ K, the Ising-like Ho³⁺ spins orient themselves along the tetragonal crystallographic c-axis. The formation of domains in LiHoF₄ has been already detected by Faraday rotation [9], but no detailed analysis of the domain structure has been reported so far. The studied samples show very good optical quality and are shaped as disks of 6 mm in diameter $(0.2 \le D \le 2.7 \text{ mm})$, cut perpendicular to the c-axis.

A sensitive Faraday rotation experimental set-up was built to visualize the magnetic domain structure at low temperature. The image was digitized on 4 bits from a COHU-CCD camera and sometimes averaged on an ATARI microcomputer. The quality of the magnetic image may be significantly improved by subtracting the associated non magnetic pattern. We generally visualize the domain structure at the $\lambda=0.5~\mu{\rm m}$ wavelength at the central part of the sample.

As expected from calculations [3, 4], a branched stripe pattern appears below T_c in all samples. The ramified pattern observed after cooling the sample through T_c in zero field is shown in figure 1. For this thickness (D=0.67 mm) the theory predicts [3, 4] about four generations of branching. The pattern is not much more complex for thicker samples because the number of branching varies as Ln(D) at low temperature [3]. As previously pointed out [1], the stripe lattice is broken by dislocations and fluctuations which induce a smecticlike structure with only an orientational order. The orientation of stripes corresponds to a twofold crystal symmetry axis.

From the development of the in-field domain structure we have determined the (H,T) phase diagram of LiHoF₄ (Fig. 2). It is qualitatively comparable with that calculated for magnetic anisotropic films [2] or for dipolar ferromagnetic thin plates [1]. Along the H_c (T) line the spontaneous magnetization M_s (T) may be de-



50 µm

Fig. 1. – Zero field cooled magnetic domain pattern at T=1.3 K for the D=0.67 mm thick sample. The image is digitized and restored with an arbitrary grey scale. (Light level intensities are 0 to 7, 8, 9, 10, 11 to 15 for the 5 successive grey levels on the image, going from black to white).

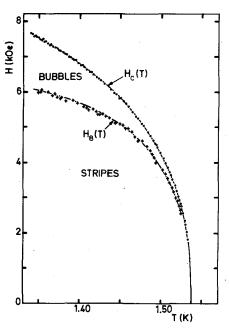


Fig. 2. -(H,T) phase diagram of LiHoF₄, determined in decreasing field, for the D=0.2 mm thick sample.

duced from the relation $M_s(T) = H_c(T) / N$, where N is the demagnetizing factor. We checked this relation for all samples we studied using the appropriate mean value of N for cylinders.

Let us consider now in more detail the magnetic domain morphology of the sample with D = 0.67 mm. The mean periodicity of its domain pattern does not differ much from that measured at H = 0 (Fig. 1) under the application of a moderate magnetic field up to 3 KOe along the c-axis, in agreement with the predicted evolution of the branched structure in a field [3]. Only the number of ramifications n_s at the surface is decreased but since the $n_s = 3$ or $n_s = 4$ generations of branching are expected to be narrow [4] the magnetic image of the central part of the sample cannot be affected much. Only the transition between the $n_s = 1$ to $n_s = 0$ branched striped structure is expected to be easily evidenced in our type of crystal, but unfortunately metastability prevents its observation during an increase of the magnetic field. The use of a special procedure bringing into play an inversion of the applied field is able to reveal this transition: the periodicity of reversed domains at the surface is reduced by a factor three in agreement with simple theoretical arguments [3, 6]. We wish to point out the slow dynamic aspect of this transition.

As discussed above a stripe to bubble transition takes place at $H_{\rm B}^+$ and the magnetization continuously reverses when increasing the field up to the collapse field value $H_{\rm c}^+$. Their number is preserved during this process while they nucleate progressively for a field decrease starting from the uniform magnetized state at

 $H_{\rm c}^-$. In this case their magnetization is close but opposite to the value at saturation. A quasi amorphous bubble structure is observed, reasonably due to a high density of dislocations as a source of nucleation. Below the field value $H_{\rm B}^-$, as already discussed for thin films [2], they distort elliptically and run back into stripes when lowering progressively the applied field. Coming back to H=0 the domain pattern looks different from that obtained previously for a H=0 cooled sample (Fig. 1). Strong irreversibilities and hysteresis are then evidenced for branched configurations, as opposed to what happens in thin samples where identical stripe patterns are observed, using the two procedures.

An interesting and simple aspect of the magnetic structure is realized just in the vicinity of the $H_c^-(T)$ line. For thick enough samples ($D \ge 0.67 \mu m$) we followed and studied the formation of bubbles in decreasing field starting from the saturated magnetic phase. The nucleation process for bubbles is initiated at the surface by spikes with opposite magnetization. This process is favourable since the local demagnetizing field is higher at the surface. Cylindrical domains form afterwards from these nuclei to shield the interior of the sample from the demagnetizing field. The bubble diameter increases on decreasing the field below H_c^- accordingly to calculations [2]. We have shown that the mean collapse diameter varies like d(mm) =9.5 $D^{2/3}$ (µm), a reasonable power law variation similar to that reported for a branched stripe structure [5]. More results and their analysis will be given in a forthcoming paper.

Acknowledgments

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