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Citation for published version:

Clark, C, Huxley, A, Czerski, D & Record, P 2021, 'Anisotropy in thin film galfenol deposited on LiNbO₃ as measured by MOKE', *Journal of Magnetism and Magnetic Materials*, vol. 529, 167799, pp. 1-5.
<https://doi.org/10.1016/j.jmmm.2021.167799>

Digital Object Identifier (DOI):

[10.1016/j.jmmm.2021.167799](https://doi.org/10.1016/j.jmmm.2021.167799)

Link:

[Link to publication record in Edinburgh Research Explorer](#)

Document Version:

Peer reviewed version

Published In:

Journal of Magnetism and Magnetic Materials

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Anisotropy in Thin Film Galfenol Deposited on LiNbO₃ as Measured by MOKE

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Abstract

We report the magnetic anisotropy of films of the magnetostrictive alloy galfenol deposited on a LiNbO₃ substrate measured with the magneto-optic Kerr effect (MOKE). This anisotropy is present in 250nm thick films but is absent in 600nm thick films, most probably showing a loss in epitaxy with thickness. An anisotropy constant of $k_u=3.45\text{kJm}^{-3}$ is deduced. Magnetic stripe domains (width $30\mu\text{m}$) oriented along the easy axis, parallel to the substrate (1 $\bar{1}$ 00) axis, could be created and imaged.

Keywords: magnetic anisotropy, FeGa, thin film, MOKE

1. Introduction

Galfenol is an iron-gallium alloy with moderate magnetostriction ($\sim 400\mu\text{m/m}$) at small saturation fields ($\sim 10\text{mT}$) [1]. The ductility and machinability of galfenol mean it is promising for thin film applications in micro-electromechanical actuators/sensors [2], vibrational energy harvesters [3], and surface acoustic wave (SAW) devices [4]. To realise the potential of thin film galfenol, an enhancement of the magnetostriction by controlling the growth conditions and substrate choice is essential.

The magnetostrictive properties of galfenol are strongly dependent on the gallium concentration, with peaks in the single crystal saturation magnetostriction constant λ_{100} at 19 at.% and 27 at.% Ga. The thermal history also plays a key role in determining the magnetic properties of galfenol; the cooling rate and any annealing processes can control structural transitions [5, 6]. In galfenol, the disordered A2 (α -Fe) bcc phase may coexist with the ordered B2 fcc, L₁₂ fcc, DO₃ fcc superlattice, and DO₁₉ hcp phases [7, 5]. These phases are strongly linked to the magnetostrictive properties that manifest in bulk and thin film samples, with the DO₃ superlattice and L₁₂ fcc phases being detrimental

to magnetostriction and the presence of a single A2 phase being beneficial to magnetostriction [8, 9, 10].

One route to increase magnetostriction may be found by inducing a magnetic anisotropy. There will then be a maximum expansion when the external field is aligned along the hard axis as this causes the largest rotation of magnetic moments during magnetisation. Previously, a training field (50mT) has been used during growth to induce an in-plane uniaxial anisotropy in 660nm thick Fe₈₁Ga₁₉ [6]. Additionally, weak anisotropy has been induced during film growth by sputtering using low argon pressure ($\sim 3\mu\text{bar}$) [11], or alternatively by raising the substrate temperature to several hundred degrees Celsius during deposition [12, 13]. Doping Fe-Ga films with B, and N has also been found to induce a large stress which affects the magnetic and high frequency response of the films [14].

The choice of substrate has been shown to affect the structure of galfenol films. Commonly chosen Si (100) or SiO₂ substrates can control the surface texture of polycrystalline galfenol [15, 16]. Both GaAs (001) [17] and MgO (100) [18] have also been successfully used as substrates, with the latter resulting in an increased saturation magnetisation in comparison to Si (100) and SiO₂.

LiNbO₃ is an attractive substrate choice due to

Ar Pressure (μbar)	Time (mins)	Thickness (nm)	Fe %	Ga %
2	20	251	87.5	12.5
4	20	254	84.5	15.5
6	20	284	83.7	16.3
2	40	590	84.9	15.1
4	40	664	86.4	13.6
6	40	616	84.2	15.8

Table 1: Growth conditions and the relative compositions of the galfenol films using LiNbO_3 as a substrate. Films were RF sputtered using a power of 60W.

its piezoelectric properties. LiNbO_3 has an order of magnitude larger electromechanical coupling coefficient than SiO_2 [19], meaning it is far more efficient in transferring electric and kinetic energy. This is particularly relevant for increasing the efficiency for SAW applications. Enhancing magnetostriction by inducing an anisotropy through the choice of growth conditions and substrate can improve the efficiency and viability of thin film devices. In this article we report on the anisotropy of thin films of galfenol deposited on a LiNbO_3 substrate as measured by the magneto-optic Kerr effect (MOKE) - the rotation and ellipticity change of reflected polarised light. Additionally, magnetic domain images following AC demagnetisation along the hard axis reveal the presence of stripe domains of average width $\sim 30\mu\text{m}$ with magnetisation orientated along the easy axis.

2. Experimental

All samples were grown by RF sputtering following a similar method to [2] but with a LiNbO_3 substrate. In particular, galfenol from a $\text{Fe}_{81}\text{Ga}_{19}$ target was deposited at room temperature on 1mm thick 128° X-cut LiNbO_3 (Roditi International) with approximate dimensions $10\text{mm} \times 10\text{mm}$. Table 1 lists the thickness and relative composition of the six galfenol films that were measured. They are broadly split into two categories depending on the time allowed for deposition: ‘thin’ films of thickness $\sim 250\text{nm}$ and ‘thick’ films $\sim 600\text{nm}$. Detailed discussion here is focused on two film thicknesses which are each representative of the two thickness regimes.

Longitudinal MOKE measurements were made with a dual photoelastic modulator setup [20] that

Figure 1: Field dependence of the longitudinal Kerr rotation (a) and ellipticity angle (b) for light reflected from 254nm thick $\text{Fe}_{84.5}\text{Ga}_{15.5}$ at different film orientations (ϕ) relative to the y axis of the LiNbO_3 substrate, as indicated by the inset.

is capable of synchronously measuring the Kerr rotation θ_K and ellipticity angle ϵ_K of reflected light. All MOKE signals were measured with incident s-polarised light ($\lambda=635\text{nm}$) focused to a $4\mu\text{m}$ spot with an incidence angle of 25° . The sample temperature was held at 295K during all measurements and the magnetic field was aligned with the film surface and the plane of incidence.

3. Results & Discussion

The longitudinal Kerr rotation and ellipticity change of light reflected from 254nm thick $\text{Fe}_{84.5}\text{Ga}_{15.5}$ were measured for various orientations of the film. The angle ϕ , as illustrated by the inset

of Figure 1(b), defines the angle between the plane of incidence and the 128° X-cut LiNbO_3 substrate y axis ($1\bar{1}00$). Field sweeps were carried out every $\sim 5^\circ$ from $\phi = -50^\circ$ to $\phi = 115^\circ$ and a subset of these are plotted in Figure 1. Each curve is the average of seven individual field sweeps, and it should be noted that the general shape of the magnetic hysteresis was independent of the position that light was focused on the sample surface (away from edges).

At $\phi = 0^\circ$, when the incidence plane, field direction and y axis of the LiNbO_3 substrate are aligned, both $\Delta\theta_K$ and $\Delta\epsilon_K$ show sharp square-like hysteresis that is characteristic of a magnetic easy axis. As ϕ is increased from zero, the field dependence of both $\Delta\theta_K$ and $\Delta\epsilon_K$ smooths out and the coercivity reduces to a minimum value at $\phi = 90^\circ$, representing a magnetic hard axis. A notable distinction between the $\Delta\theta_K$ and $\Delta\epsilon_K$ field dependence is the saturation signals as ϕ increases from 0° to 90° : $\Delta\theta_K^{sat.}$ decreases (by $\sim 28\%$) from 0.39mrad to 0.28mrad whereas $\Delta\epsilon_K^{sat.}$ increases (by $\sim 50\%$) from 0.08mrad to 0.17mrad . The observed anisotropy may be due to a magnetostriction in the galfenol film arising from the differing lattice parameters between galfenol and LiNbO_3 . The lattice parameter of cubic A2 (α -Fe) FeGa (which is assumed due to the low gallium concentration in these films) is roughly 0.287nm at 300K , and LiNbO_3 is hexagonal with $a=b=0.515\text{nm}$, and $c=1.386\text{nm}$. The magnetic easy axis in thin film FeGa would be along $\langle 001 \rangle$, the same as bulk FeGa. The lattice mismatch ($(d_{\text{LiNbO}_3} - d_{\text{FeGa}})/d_{\text{LiNbO}_3}$) for commensurate ratios would be -11.5% along $\phi = 0^\circ$ and, taking the $\langle 110 \rangle$ direction to be the hard axis aligned with the (0001) projection of the 128° X-cut LiNbO_3 , the mismatch would be -7.7% along $\phi = 90^\circ$. The large difference in the saturation values of θ_K and ϵ_K would then be a result of a change in the allowed inter- and intra-band transitions that would be a consequence of a magnetostrictive-dependent change in structure.

The three galfenol films grown with a deposition time of 20 mins, according to Table 1, showed similar evolution of the field dependence of $\Delta\theta_K$ and $\Delta\epsilon_K$ with increasing ϕ . No dependence on the argon growth pressure was observable in the MOKE measurements, as has previously been noted [11], though this is likely due to the anisotropy resulting from the LiNbO_3 -galfenol interface being much greater than a residual stress due to the argon pressure. Additionally, no out of plane MOKE signal was measurable, indicating that the magnetisation

Figure 2: Field dependence of the longitudinal Kerr rotation (a) and ellipticity angle (b) for light reflected from 664nm thick $\text{Fe}_{86.4}\text{Ga}_{13.6}$ at different film orientations (ϕ) relative to the y axis of the LiNbO_3 substrate. The average $\Delta\theta_K$ and $\Delta\epsilon_K$ from 254nm thick $\text{Fe}_{84.5}\text{Ga}_{15.5}$ from Figure 1 is also plotted.

is confined within the film plane.

Figure 2 shows longitudinal MOKE field sweeps on a 664nm thick $\text{Fe}_{86.4}\text{Ga}_{13.6}$ film for various ϕ . As this galfenol film is rotated from $\phi = 0^\circ$ to 90° , the change in shape of the hysteresis loops is far less marked than that of the thinner film in Figure 1. For comparison, the weighted field dependence of $\Delta\theta_K$ and $\Delta\epsilon_K$ averaged over ϕ for the thinner 254nm $\text{Fe}_{84.5}\text{Ga}_{15.5}$ film is also plotted in Figure 2. The saturation $\Delta\theta_K$ is identical for both galfenol thicknesses however, the average coercivity of the thinner film is larger than the thicker film (4.3mT compared to 1.2mT). Both $\Delta\theta_K^{sat.}$ and $\Delta\epsilon_K^{sat.}$ increase with increasing ϕ for the thicker film in con-

trast to the inverse correlation for the thinner film. There is also a large difference in the magnitude of $\Delta\epsilon_K^{sat.}$ between the two film thicknesses, with the saturation signal varying between 0.22mrad and 0.33mrad in the 664nm thick film compared to the average saturation of 0.10mrad for the 254nm thick film. The penetration depth of visible light is typically 10's of nm in similar metals, far less than the film thicknesses here. This cannot, therefore, explain the reduction of $\Delta\epsilon_K^{sat.}$ in the thinner film. The growth conditions, as detailed in Table 1, were identical for both films. Hence, the presence of magnetic anisotropy only in the thinner galfenol films suggest that a stress induced at the LiNbO₃-FeGa interface decays with increasing galfenol thickness.

Figure 3 is a plot of the remanence to saturation ratio (M_r/M_s) for both θ_K and ϵ_K from the 254nm thick Fe_{84.5}Ga_{15.5} film, as obtained from loops in Figure 1. The MOKE signal is proportional to the component of magnetisation along the plane of incidence. The remanence measured by θ_K follows a $|\cos(\phi)|$ dependence. Hence, the magnetisation is confined to point along the easy axis at zero field i.e. for all ϕ , $M_r = M_s \cos(\phi)$. This suggests that the anisotropy in this galfenol film is strong enough to prevent domains forming along a direction other than the easy axis. As the magnetic field is reduced the moments rotate smoothly towards the closest easy axis.

The Stoner-Wohlfarth model describes magnetic reversal of a single domain ferromagnet with uniaxial anisotropy. The free energy density may be expressed in terms of the angle between the easy axis and applied field (ϕ) as well as the angle between the easy axis and the saturation magnetisation (ψ), as

$$E = k_u \sin^2(\phi) - M_s H \cos(\phi - \psi). \quad (1)$$

k_u is an anisotropy constant, and the second term is the Zeeman term. The anisotropy constant k_u is related to the anisotropy field [21]

$$H_k = \frac{2k_u}{M_s}. \quad (2)$$

H_k is the field value at which saturation of θ_K is achieved for field along the hard axis ($\phi=90^\circ$). For the 254nm Fe_{84.5}Ga_{15.5} film $\mu_0 H_k=15\text{mT}$. The saturation magnetisation M_s can be obtained from the hard axis hysteresis loop measured in a SQUID magnetometer. For this film, $\mu_0 M_s=580\text{mT}$ at 295K, which gives $k_u=3.45\text{kJm}^{-3}$. This value is slightly higher than would typically be expected

Figure 3: Ratio of the remanence to saturation values of θ_K and ϵ_K for the 254nm thick Fe_{84.5}Ga_{15.5} film. Blue points are the orange points rotated by 180°.

from single crystal galfenol. Bulk or single crystal galfenol can be annealed whilst under stress or in a magnetic field to enhance the anisotropy constant. Under stress annealing at elevated temperatures (219MPa at 635°C), k_u for Fe_{81.6}Ga_{18.4} can increase from a compressive -0.45kJm^{-3} to an expansive $+13.6\text{kJm}^{-3}$ [22]. Field annealing polycrystalline Fe_{81.6}Ga_{18.4} (1.2T at 550°C) can increase k_u from 1.6kJm^{-3} to 7.2kJm^{-3} [23]. Thus the anisotropy observed in the 254nm Fe_{84.5}Ga_{15.5} film here may possibly be enhanced further. Indeed, if stress caused by the FeGa-LiNbO₃ interface does decay with increasing film thickness, then reducing the thickness could increase k_u , resulting in a larger maximum magnetostriction.

Magnetic domains may be imaged in the MOKE signals by scanning the measurement position across the film surface. For galfenol, the Kerr rotation response is larger than the ellipticity angle change between opposing magnetisation directions. Figure 4(a) shows a map of $\Delta\theta_K$ for the plane of incidence parallel to the easy axis over the Fe_{84.5}Ga_{15.5} surface under a constant 150mT field along the hard axis. This is sensitive to moments along the easy axis and highlights the variation in Kerr rotation across the galfenol film when the magnetisation is fully saturated along the hard axis. Figure 4(b) & 4(c) show the change in Kerr rotation across the same area of the Fe_{84.5}Ga_{15.5} surface following two separate AC demagnetisation sweeps ending with zero field. In both figures, stripe do-

Figure 4: The Kerr rotation measured along the vertical (easy) axis across the same area on 254nm thick $\text{Fe}_{84.5}\text{Ga}_{15.5}$ with (a) constant +150mT along the horizontal (hard) axis, and (b) & (c) repeated scans after separate AC demagnetisation along the horizontal (hard) axis.

mains can be clearly distinguished when compared to the saturated scan in Figure 4(a). The magnitude of $\Delta\theta_K \approx \pm 0.4 \text{ mrad}$ is identical to the saturation signal in Figure 1. In these domain images, positive/negative changes in Kerr rotation correspond to domains with net magnetisation pointing along the easy axis in opposite directions. The average domain width is roughly $30 \mu\text{m}$, and they extend $> 150 \mu\text{m}$ along the easy axis.

The placement of domains across Figure 4(b) & 4(c) is broadly similar on the right half of the scan (above $400 \mu\text{m}$). Differences are evident in the left half, however, with domains having a positive Kerr rotation being preferred in Figure 4(c). These similarities between the two scans indicate that in this galfenol film, domains are not generally pinned however, some may be confined by local surface composition variations or defects.

The strong anisotropy that has been observed in 254nm thick galfenol films here is most likely caused by a stress induced by the lattice mismatch between the film and the 128° X-cut LiNbO_3 substrate. Rather than a magnetostriction brought about by the interface, an alternative explanation for the anisotropy could be a microstructure with elongated grains aligned along the LiNbO_3 y axis. Such a columnar growth has previously been ob-

served in galfenol films up to 100nm thick [6, 16, 18], with grain diameters up to 120nm. If galfenol sputtered on 128° X-cut LiNbO_3 grows with similar sized grains then the magnetic domain size should correspond to this. However, the comparatively large magnetic domain width of $30 \mu\text{m}$ measured here suggests that the grain shape is not the source of anisotropy. A microstructural study of galfenol films deposited on substrates that induce magnetostriction in galfenol, such as LiNbO_3 , could better identify the source of the anisotropy. As has previously been reported, the exact phase composition is likely an important controlling factor of the magnetostriction in these galfenol films [5, 8, 9, 10].

4. Conclusion

Magnetic anisotropy from the field dependence of θ_K and ϵ_K has been measured in $\sim 250 \text{ nm}$ thick galfenol films aligning with the crystal directions of the 128° X-cut LiNbO_3 substrate. This anisotropy was not observed in $\sim 600 \text{ nm}$ thick films grown under identical conditions, suggesting that a strain caused by the FeGa-LiNbO_3 interface relaxes within the galfenol. At zero field, the magnetisation in $\sim 250 \text{ nm}$ galfenol films points along the y axis of the LiNbO_3 substrate, forming stripe domains of

300 30 μ m width. Additionally, the anisotropy constant 360
is calculated as $k_u=3.45\text{kJm}^{-3}$.

We wish to gratefully acknowledge funding from
the EPSRC (grant number EP/R013004/1). The
authors declare no competing interests. 365

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