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Journal

Applied Physics Letters, 127(3)

ISSN

0003-6951

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Publication Date

2025-07-21

DOI

10.1063/5.0278599

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Low damping (111) Oriented Lithium Aluminium Ferrite Thin Films for Spin Wave Applications

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(Dated: June 24, 2025)

Spin wave-based spintronics are an alternative to conventional electronics due to their potential for efficient energy consumption, improved processing speed and smaller device dimensions. Low damping magnetic insulators provide the medium for efficient propagation of spin waves for information transfer. We have synthesized by pulsed laser deposition epitaxial spinel structure ferrite thin films of $\text{Li}_{0.5}(\text{Al}_{1.0}\text{Fe}_{1.5})\text{O}_4$ (LAFO) on (111)-oriented MgAl_2O_4 that support isotropic magnon propagation in the film plane. Our ferromagnetic resonance measurements show low magnetic damping with a typical Gilbert damping parameter of $\alpha = 0.006$ and weak-spin orbit coupling with $g = 2.02$. These films have low effective magnetization, $\mu_0 M_{\text{eff}} = 20$ mT, similar to that of yttrium iron garnet (YIG), the gold standard of low loss magnetic insulators. Our findings show that LAFO is a good candidate as a spin wave medium since it can be grown at low temperatures in different crystal orientations.

Spin-based devices have been recognized as an alternative to conventional electronics but, to date, their promise of efficient energy consumption, information processing and smaller device dimensions has yet to be fully realized.[1–4] Current spintronics, or spin-based electronics, is based on spin polarized charge currents where the flow of charge still results in power dissipation similar to traditional microelectronics. Spin-wave based devices composed of magnetic insulators offer a more energy efficient route to spintronics. The success of spin-wave based devices relies on the ability to synthesize magnetic insulators (MIs) with low magnetic damping in which pure spin currents can propagate for long distances and can be interfaced with traditional microelectronics. Currently the gold standard in low damping MIs is the family of garnet ferrites, including yttrium iron garnet (YIG) [5–11]. YIG has a Gilbert damping parameter, α , that can be as low as 1×10^{-4} in thin films, indicating low magnetic loss, but requires high processing temperature (e.g., an annealing temperature of 900 °C [11–13]) and single crystal garnet structure substrates. Recently, a class of spinel structure ferrite thin films has been identified as a low damping medium for the propagation of spin waves. In particular, spinel ferrite $\text{Li}_{0.5}(\text{Al}_{1.0}\text{Fe}_{1.5})\text{O}_4$ (LAFO) thin films with a (001) orientation have been shown to exhibit low damping with $\alpha \sim 5 \times 10^{-4}$, low external saturation field requirements, and low growth temperature of 400 - 450 °C.[14–16] The low external field requirement in these (001) spinel ferrite thin films is due to the presence of a

giant in-plane magnetic anisotropy that is not present in (111) YIG films. To establish low damping LAFO with more isotropic spin wave propagation in the plane of the film, we explored the stabilization of (111)-oriented epitaxial LAFO films.

In this work, we have stabilized (111)-oriented LAFO thin films with excellent crystallinity and low damping for spin wave propagation. Atomic force microscopy shows atomically flat surfaces with an rms roughness of 0.29 nm. Static magnetic properties of these (111) LAFO films show saturation magnetization, $M_s = 145$ kA/m at 10 K which is similar to the bulk value and also show negligible in-plane magnetic anisotropy [17–19]. We deduce low damping of $\alpha = 0.006$ in (111) LAFO thin films from the frequency dependence of the ferromagnetic resonance linewidth. From the frequency dependence of the ferromagnetic resonance field, we deduce a spectroscopic g-factor close to 2.02. We confirm that Fe^{3+} , distributed among the tetrahedral and octahedral sites, to be the sole source of magnetism using x-ray magnetic circular dichroism. Together these properties make (111) LAFO films promising as a medium for spin wave-based electronics.

Epitaxial thin films of $\text{Li}_{0.5}\text{Al}_{1.0}\text{Fe}_{1.5}\text{O}_4$ were grown on (111) oriented single-crystalline MgAl_2O_4 (MAO) substrates using pulsed laser deposition. Before deposition, MAO substrates were ultrasonically cleaned in acetone then isopropanol for 10 minutes each. A polycrystalline pressed target of $\text{Li}_{0.6}\text{Al}_{1.0}\text{Fe}_{1.5}\text{O}_4$ (manufactured by Toshima Corporation) was sanded before deposition. The target with excess Li was used to compensate for Li loss during deposition due to the high volatility of Li.

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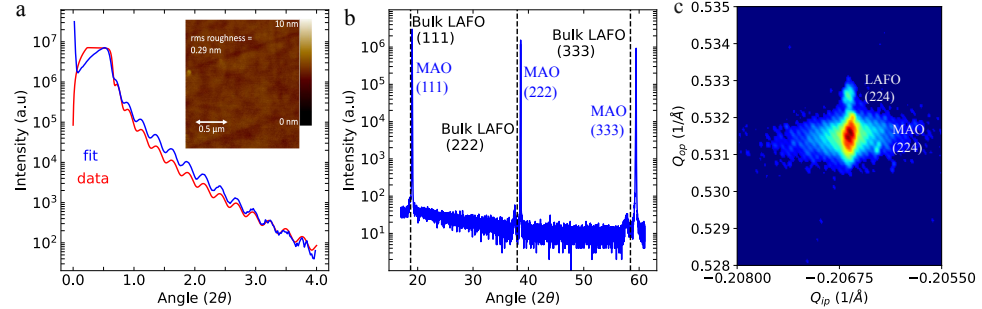


FIG. 1. Structural characterization of a 32 nm LAFO sample. (a) XRR scan with Kiessig fringes that indicates uniform film thickness and low surface roughness as confirmed by the AFM image. LAFO has atomically smooth surface with an rms roughness of 0.29 nm. (b) A symmetric x-ray diffraction scan showing film Bragg peaks to the left of the expected bulk LAFO peaks and (111) MAO peaks. (c) The reciprocal map scan around the (224) MAO peak. The MAO and LAFO peaks are perfectly aligned along the in plane q indicating coherent strain of the film to the substrate.

The target was ablated with a 248nm KrF laser at a fluence of 2.8 J/cm^2 and a repetition rate of 2 Hz. Films were grown at substrate temperature of 325°C and oxygen pressure of 10 mTorr, resulting in a deposition rate of 1 nm/min. Both the substrate temperature and oxygen pressure are lower compared to the optimum growth temperature and pressure of thin films of (001) LAFO, (001) $\text{Mg}(\text{Al,Fe})_2\text{O}_4$, and YIG [11–15, 20]. After deposition, the samples were cooled to room temperature in 150 Torr of oxygen.

Immediately after deposition, the sample's surface morphology was studied using atomic force microscopy (AFM) on a Digital Instruments Dimension 3100. X-ray diffraction (XRD), in the form of symmetric diffraction, reciprocal space map (RSM) and x-ray reflectivity (XRR), was performed on a Panalytical Empyrean diffractometer and Panalytical X'pert 2. Static magnetic characterization of the films were performed on a Quantum Design Evercool SQUID magnetometer. Dynamic magnetic properties were probed on a broadband ferromagnetic resonance (FMR) system in a co-planar waveguide geometry with a 23 dB RF power source at frequencies ranging from 3 GHz to 20 GHz, and field sweeps from 100 mT to 700 mT. X-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism measurements (XMCD) were performed at normal incidence in total electron yield (TEY) and total luminescence yield (TLY) modes at beamlines 4.0.2 and 6.3.1 of the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory (LBNL). Data was collected with a flying scan, where measurements were taken as the photon energy was continuously varied. Spectra were recorded with both positive and negative photon helicity and XMCD spectra were calculated from their difference. These measurements enabled us to probe Fe cation valence and distribution in samples within TEY probe depth of $\sim 5 \text{ nm}$.

To understand the structural properties of the films, we performed AFM, XRR and XRD measurements. The AFM measurement of a 32 nm LAFO film, shown in the inset of figure 1(a), indicates a smooth surface with a typical rms roughness of 0.29 nm which is roughly 1 % of the film thickness. The XRR scan in figure 1(a) indicates Kiessig fringes that can be fit with an interface roughness of 0.33 nm and a thickness of 32 nm. Figure 1(b) shows an XRD scan measured around the (111)-MAO substrate peaks. There is a broad shoulder to the left of the family of {111} substrate peaks corresponding to the family of {111} LAFO film peaks. We do not observe Laue oscillations surrounding (111) LAFO film peaks that appear in (001) LAFO films, thus suggesting that (111) films do not have as smooth a surface or interface with the substrate compared to (001) films. The shift of the film peaks from the expected bulk values indicates a 5.63% elongation of the out-of-plane lattice parameter. Figure 1(c) shows an RSM scan around the (224) film and substrate peaks. The alignment of the film and substrate peak at the same in-plane q indicates that the film is coherently strained to the substrate. Together these results indicate that the films are under coherent in-plane compressive strain by the substrate.

We studied static field dependent magnetization of the films at 300 K for magnetic fields applied out-of-plane along the (111) direction as well as in-plane along the directions (112) and (110) and at 10 K for magnetic field applied in the plane direction (112). Figure 2(b) shows the hysteresis loop at 10 K with $M_s = 145 \text{ kA/m}$ which is similar to the bulk value [17–19]. Figure 2(a) shows a hysteresis loop with negligible coercivity for a magnetic field applied along the in-plane (112) direction and out-of-plane (111) direction at 300 K, well below the bulk Curie temperature of 750 K[17–19]. The in-plane and out-of-plane scans are indistinguishable, indicating negligible magnetic anisotropy. The inset in figure 2(a) shows

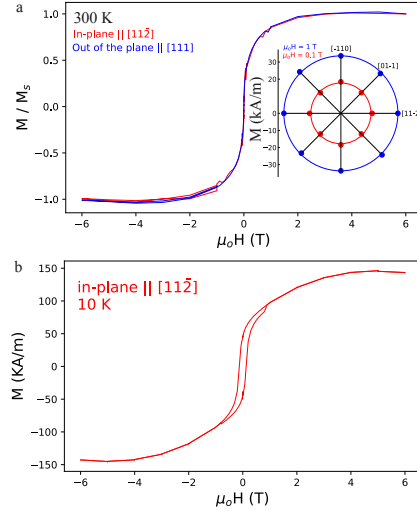


FIG. 2. SQUID measurements of a 32 nm sample of LAFO. (a) Magnetization versus field ($\mu_0 H$) along the in-plane and out-of-plane directions with $M_s = 40$ kA/m. Insert shows the magnetization values at different in-plane orientations for fixed fields, $\mu_0 H = 0.1$ T, and 1 T at 300 K. (b) Hysteresis loop measurement at 10 K where $\mu_0 H \parallel [112]$ with $M_s = 145$ kA/m.

that the magnetization values at 0.1 T and 1 T (corresponding to 3.3 and 33% of the saturation field), are also isotropic in the plane, indicating the absence of magnetic anisotropy. These results indicate that (111) LAFO films are excellent candidates for supporting isotropic spin wave propagation.

To understand the origin of magnetism in LAFO, we performed XAS and XMCD measurements. XAS is performed at the L_3 and L_2 edges of Fe with circularly polarized light (figure 3(a)). We take the difference of the signal of right and left circularly polarized light to obtain the XMCD signal (figure 3(b)). By comparing our data with standard Fe based samples, we determine that Fe^{3+} cations with octahedral and tetrahedral coordination contribute to the magnetism in (111) LAFO. The peak at 709.2 eV in the XMCD spectrum in figure 3(b) arises from Fe^{3+} tetrahedrally (Fe_T^{3+}) coordinated with respect to the oxygen atoms and the second peak at 710.0 eV arises from Fe^{3+} octahedrally (Fe_O^{3+}) coordinated with respect to the oxygen atoms [21, 23–25]. It can also be seen from figure 3(b) that the Fe_T^{3+} and Fe_O^{3+} have opposite signs of the XMCD signal, indicating that Fe_T^{3+} and Fe_O^{3+} moments are aligned anti-parallel to one another.[14, 15] This data was compared to the reference materials shown in figure 3(c), GaFeO_3 with 100% Fe_O^{3+} [22] and $\gamma\text{-Fe}_2\text{O}_3$ with 37.5% Fe_T^{3+} and 62.5% Fe_O^{3+} [21]. Through fit-

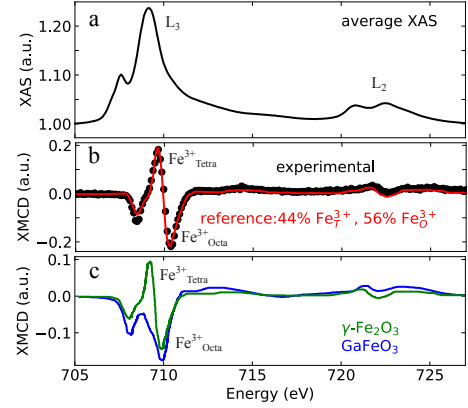


FIG. 3. X-ray absorption spectroscopy and x-ray magnetic circular dichroism spectra of 13 nm LAFO at the Fe $L_{2,3}$ edges. (a) An average of the right and left circularly polarized light x-ray absorption spectra. (b) In black, XMCD spectra of LAFO and in red, fit to the reference XMCD. (c) Reference XMCD spectra of $\gamma\text{-Fe}_2\text{O}_3$ (62.5% Fe_O^{3+} , 37.5% Fe_T^{3+}) [21] and GaFeO_3 (100% Fe_O^{3+}) [22] used to fit the spectra in (b).

ting the data to a weighted sum of the reference data, we determine the relative ratios of 56% Fe_O^{3+} and 44% Fe_T^{3+} . From this ratio, the calculated $M_s = 126$ kA/m is compared to the $M_s = 145$ kA/m value from SQUID measurements at 10 K shown in figure 2(b).

To characterize the dynamic magnetic properties, we performed broadband FMR measurements with the field applied in-plane at 300 K. Figure 4(a) depicts a typical ferromagnetic resonance peak at 13 GHz probed along the (110) direction. We perform a Lorentzian fit to the FMR peak to extract the half-width-half-max, $\mu_0 \Delta H_{\text{hwhm}} = 11.0$ mT and FMR field, $\mu_0 H_{\text{fmr}} = 451$ mT. The typical linewidth for these (111) LAFO thin films is approximately an order of magnitude higher than (001) LAFO films [14]. This broader FMR linewidth is consistent with the broader and lower intensity Bragg peaks in the XRD (Figure 1(b)). These broader and lower intensity peaks suggest increased defects and inhomogeneities. Similar linewidth broadening was observed in previous work on spinel structure MAFO and was correlated with increased disorder [20].

Frequency dependence of the FMR linewidth indicates low magnetic damping in our (111) LAFO films. The slope of the frequency dependence of the half-width-half-maximum linewidth (ΔH_{hwhm}) provides a measure of α of the samples determined by the following:

$$\Delta H_{\text{hwhm}} = \Delta H_0 + \alpha \frac{h}{g \mu_B \mu_0} f \quad (1)$$

where ΔH_0 is a constant magnetic inhomogeneous broad-

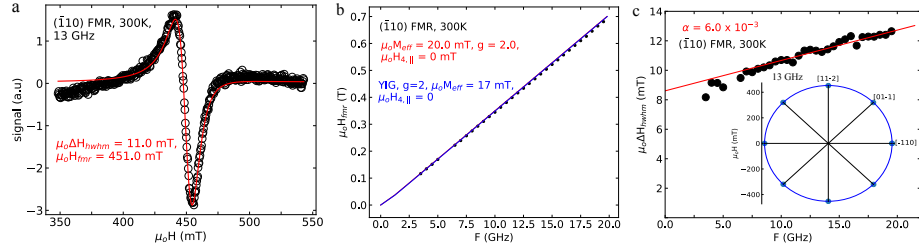


FIG. 4. FMR measurements. (a) An example of an FMR scan at 13 GHz and a fit to the derivative of a Lorentzian equation to obtain $\mu_0 \Delta H_{hwhm} = 11$ mT and $\mu_0 H_{fmr} = 451$ mT. (b) $\mu_0 H_{fmr}$ as a function of frequency and in red, a fit to the Kittel equation shown in equation 2. In blue is a graph of the equation 2 for a (111) YIG film with the constants given as $g = 2$, $\mu_0 M_{eff} = 17$ mT and $\mu_0 H_{4||} = 0$ [26]. (c) Half-width at half max ($\mu_0 \Delta H_{hwhm}$ versus frequency (F) and a fit to equation 1 in red. From the fit, we extract a low damping parameter, α , of 0.006. The inset is plot of $\mu_0 H_{fmr}$ in different in-plane directions at a fixed frequency of 13 GHz and in blue, is a fit to the a circle with a radius of 452.7 mT

ening term, α is the Gilbert damping parameter, g is the g-factor, μ_B is the Bohr magneton, μ_0 is the permeability of free space and h is the Planck constant. We do not see evidence of two-magnon scattering since the frequency dependence of the linewidth is linear [27–29]. Figure 4(c) shows the data for a 32 nm (111) LAFO along with a fit to equation 1 to deduce a Gilbert damping parameter of $\alpha = 6 \times 10^{-3}$ with an inhomogeneous broadening term $\Delta H_0 = 8.61$ mT. This damping parameter value is a factor of 6 higher than the reported values of LAFO grown on 001-oriented MAO [14] and may be a reflection of the more disordered nature of (111) growth versus (001) growth. The inhomogeneous broadening value is approximately an order of magnitude higher than the value obtained for (001) LAFO. We attribute this to the increased disorder in (111) LAFO compared to (001) LAFO [14].

We also confirm the low magnetic anisotropy of these films, obtained via static magnetic measurements, by fitting the frequency dependence of the FMR field to the Kittel equation 2,

$$f = \frac{g\mu_B\mu_0}{h} \sqrt{(H_{fmr} + H_{4||})(H_{fmr} + M_{eff} + H_{4||})} \quad (2)$$

where $H_{4||}$ is the in-plane fourfold magnetocrystalline anisotropy and M_{eff} is the effective magnetization which is given by $M_{eff} = M_s + H_{2\perp}$, includes the saturation magnetization, M_s , and out of plane uniaxial anisotropy field, $H_{2\perp}$. [20, 29, 30]. From the fit shown in figure 4(b), we determine the value of the effective magnetization, $\mu_0 M_{eff} = 20 \pm 2$ mT, to be smaller than the saturation magnetization, $\mu_0 M_s = 40$ mT, obtained from the static magnetometry at 300 K. From the difference, we can deduce a negligible uniaxial anisotropy field of $\mu_0 H_{2\perp} = -20$ mT which is an order of magnitude lower than that observed in other low damping spinel ferrites such as (001) LAFO and (001) MAFO [14, 15]. The negligible values of both $H_{4||}$ and $H_{2\perp}$ agree with the SQUID data that shows no distinction between in-plane or out-

of-plane. Furthermore, we performed in-plane FMR measurements in different directions at a fixed frequency of 13 GHz; the inset in figure 4(c) shows that the $\mu_0 H_{fmr}$ is isotropic in the plane with an average value of 452.7 mT. From the fit to the Kittel equation, we also extract $g = 2.02 \pm 0.001$ which reveals negligible orbital angular momentum consistent with single ion anisotropy of Fe^{3+} cations.

In comparison to known magnetic insulators with low magnetic damping, we believe (111) LAFO has several advantages that make it an attractive option for device integration and applications where isotropic spin wave propagation is essential. Firstly, (111) LAFO films have undetectable in-plane magnetic anisotropy compared to (001) spinel ferrite films, such as (001) LAFO and (001) MAFO, hexaferrite films (e.g., such as $\text{BaFe}_{12}\text{O}_{19}$) and perovskite metal $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films [14, 15, 20, 31–35]. Secondly, the optimal growth temperature of (111) LAFO films is lower than growth temperatures of YIG, (001) LAFO and (001) MAFO, thus keeping the thermal budget low [26, 36–38]. Finally spinels, such as (111) LAFO, can be interfaced with oxide perovskites which, in turn, have been demonstrated to grow on silicon, thus providing a path to integration with silicon microelectronics [39, 40].

In summary, we have demonstrated the growth of (111) LAFO with low magnetic damping and in-plane magnetic anisotropy which are ideal for supporting isotropic spin wave propagation. They exhibit effective magnetization, which is similar to saturation magnetization, indicating negligible out-of-plane magnetic anisotropy. These films can be grown on MAO at temperatures as low as 325 °C and an oxygen pressure of 10 mTorr with atomically smooth surfaces. Our findings show the robustness of LAFO to grow in different crystal orientations at low growth temperatures which will be essential in integrating LAFO in spin wave based devices.

ACKNOWLEDGMENTS

This work is supported by the U.S. Department of Energy, Director, Office of Science, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Contract No. DESC0008505. X.Y.Z. was supported by the Air Force Office of Scientific Research under Grant No. FA 9550-20-1-0293. D.O. was supported by the National Science Foundation on Award DMR-2037652. Part of this work was performed at nano@stanford, supported by the National Science Foundation under award ECCS-2026822. This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under contract no. DE-AC02-05CH11231. Low-temperature magnetic properties measurements performed at Stanford University were supported by the Air Force Scientific Research Award FA9550-20-1-0252.

AUTHORS DECLARATION

Conflict of interest

The authors have no conflict of interest.

Authors contribution

Lerato Takana; Conceptualization (equal); Data curation (lead); Formal analysis (lead); Investigation

(lead); Writing – original draft (lead). **Sanyum Channa**; Data curation (supporting); Investigation (supporting); Writing – review & editing (supporting). **Xin Yu Zheng**; Data curation (supporting); Investigation (supporting **Daisy O'Mahoney**; Data curation (supporting). **Sauviz Alaei**; Data curation (supporting); Investigation (supporting). **Yuntian Li**; Data curation (supporting). **Arturas Vailionis**; Data curation (supporting). **Padraic Shafer**; Resources (supporting); Supervision (supporting). **Alpha T. N'Diaye**; Resources (supporting); Supervision (supporting). **Christoph Klewe**; Resources (supporting); Supervision (supporting). **Ian Fisher**; Funding acquisition (lead). **Yuri Suzuki**; Conceptualization (equal); Funding acquisition (lead); Supervision (equal); Writing – review & editing (equal)

DATA AVAILABILITY

The data supporting the findings of this study is available from the corresponding authors upon reasonable request.

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PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0278599

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