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Nearly isotropic magnon transport in epitaxial lithium aluminum ferrite thin films

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ABSTRACT

Low-loss magnetic insulating thin films are promising for information transport via magnons, where isotropic in-plane magnon propagation is desirable. We report nonlocal measurements of electrically and thermally generated magnons in epitaxial (001) lithium aluminum ferrite $\text{Li}_{0.5}\text{Al}_{0.7}\text{Fe}_{1.8}\text{O}_4$ thin films with pronounced fourfold in-plane magnetic anisotropy. By measuring the inverse spin Hall signal as a function of the magnon diffusion distance, we deduce magnon diffusion lengths that are nearly identical along the [100] and [110] directions at 250 K. This isotropy is consistent with a nearly isotropic exchange stiffness. These results highlight spinel ferrites as viable platforms for isotropic magnon transport.

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Magnon transport in epitaxial thin films has been studied in a range of materials from conventional systems such as ferromagnets, antiferromagnets, and ferrimagnets to more exotic materials such as non-collinear antiferromagnets, synthetic antiferromagnets, and multiferroics.¹ Among these, ferrimagnets are especially promising because of their fast spin dynamics similar to antiferromagnets and easy control in switching magnetization similar to ferromagnets.² Moreover, ferrimagnetic insulators with low magnetic damping are excellent candidates for magnon propagation media as they give rise to minimal energy loss due to Joule heating compared to metals. The most well-studied ferrimagnetic insulators come from the family of garnet ferrites, including $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ³ and $\text{Tm}_3\text{Fe}_5\text{O}_{12}$.⁴ They exhibit Gilbert damping parameter as low as 2×10^{-4} in thin film form^{5–8} and even lower with post-deposition annealing in oxygen.⁹

More recently, spinel ferrite thin films have been found to be a promising alternative to garnet ferrite films as they can be grown at much lower deposition temperatures, have lower external field requirements,^{10–12} and have potential for integration with silicon.^{13,14} Examples include $\text{Mg}(\text{Al}, \text{Fe})_2\text{O}_4$ (MAFO) and $\text{Li}_{0.5}(\text{Al}, \text{Fe})_{2.5}\text{O}_4$

(LAFO) thin films,^{15–17} which have been made into heterostructure devices and demonstrated efficient spin transport.^{18–21} In magnesium aluminum ferrite $\text{MgAl}_{0.5}\text{Fe}_{1.5}\text{O}_4$ thin films, previous work found a small fourfold in-plane magnetic anisotropy on the order of 10 mT according to static magnetization measurements.¹⁵ However, nonlocal spin transport measurements show a spin diffusion length 30% greater for magnon propagation along the in-plane easier (110) axes compared to the harder (100) axes.²² It was proposed that the anisotropic magnon transport was not a result of the magnetic anisotropy; instead, it could be explained by an anisotropy in the exchange stiffness.²²

To understand if the anisotropy in spin transport exists more broadly within spinel ferrite thin films, we studied epitaxial lithium aluminum ferrite $\text{Li}_{0.5}\text{Al}_x\text{Fe}_{2.5-x}\text{O}_4$ (LAFO) for $x = 0.7$ thin films, which are spinel ferrites with larger in-plane magnetic anisotropy and even lower Gilbert damping than MAFO films. Epitaxial LAFO ($x = 0.7$) films on MgAl_2O_4 substrates are under compressive epitaxial strain. They exhibit a magnetically hard axis out-of-plane and a fourfold in-plane magnetic anisotropy as do the epitaxial MAFO thin

films, thus making them a suitable candidate to study spin transport for comparison with MAFO. Contrary to MAFO, we find a nearly isotropic spin diffusion length in the plane of the film. This phenomenon can be explained by nearly isotropic exchange energy in contrast to MAFO.²² This in-plane isotropic exchange is attributed to the tetragonally distorted crystal structure and weak spin-orbit coupling in the Fe^{3+} cations in LAFO. In any case, the nearly isotropic spin diffusion lengths found in LAFO are promising for magnon waveguide or interconnect applications.

Epitaxial LAFO ($x = 0.7$) films were grown on MgAl_2O_4 (MAO) (001) substrates using pulsed laser deposition with a 248 nm KrF laser operating at a fluence of 2.8 J/cm^2 . These epitaxial films were grown at substrate temperatures of $400\text{--}450^\circ\text{C}$ in O_2 atmospheres of 2.0 Pa, similar to previous studies on LAFO.^{16,23} X-ray diffraction measurements, performed on a Panalytical Empyrean x-ray diffractometer, indicate that the LAFO films grow epitaxially on MAO substrates under 2.2% biaxial compressive epitaxial strain (see [supplementary material](#) Sec. I). Films show excellent crystallinity with a typical rocking curve linewidth of $\Delta\omega = 0.0183^\circ$ [see [supplementary material](#) Figs. S1(a)–S1(b)]. X-ray reflectivity (XRR) measurements show film thicknesses on the order of 10–15 nm for films grown for this study [[supplementary material](#) Fig. S1(c)]. Moreover, films are extremely smooth with a root mean square roughness of $\sim 0.2 \text{ nm}$ as deduced from atomic force microscopy [[supplementary material](#) Fig. S1(d)].

We studied the magnetic anisotropy of our films via static magnetization measurements on a superconducting quantum interference device (SQUID) magnetometer. Epitaxial (001) LAFO films grown under compressive strain have a magnetically easier $\langle 110 \rangle$ family of axes and a magnetically harder $\langle 100 \rangle$ family of axes, with the out-of-plane $[001]$ axis being the hardest.²³ Field dependence of the magnetization in our LAFO thin films shows square hysteresis loops with coercive fields on the order of 0.5–1.0 mT along the $[110]$ in-plane axes at 250 K (Fig. 1). Along the magnetically harder $[100]$ axis, the LAFO films exhibit fast domain switching, followed by slow domain rotation until they saturate²⁴ (Fig. 1). By fitting to the portion of the magnetization M vs magnetic field $\mu_0 H$ curve that corresponds to domain rotation (see [supplementary material](#) Sec. II), we can extract the saturation magnetization $M_s = 165.3 \pm 0.2 \text{ kA/m}$ and the saturation field $H_s = 62 \pm 3 \text{ mT}$. In the case of (001) oriented films of

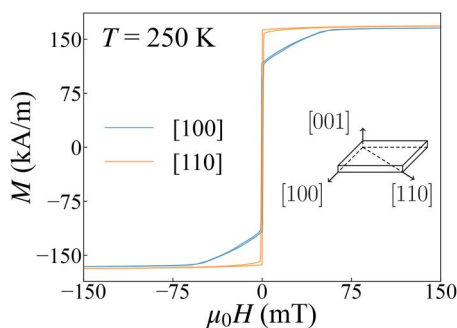


FIG. 1. Field dependence of the magnetization for a (001) LAFO film on (001) MAO along the in-plane $[100]$ and $[110]$ directions at 250 K. (inset) Schematic diagram of the LAFO film showing the $[001]$ out-of-plane axis and the $[100]$ and $[110]$ in-plane axes.

nominally cubic materials under biaxial epitaxial strain, the strain anisotropy does not affect the overall in-plane magnetic anisotropy. In addition, our sample is a $5 \times 5 \text{ mm}^2$, so shape anisotropy is negligible for in-plane measurements. What remains is the magnetocrystalline anisotropy, the energy of which can be computed as $K_1 = \frac{-H_s M_s}{2} = (-5.1 \pm 0.3) \times 10^3 \text{ J/m}^3$.²⁴

Broadband ferromagnetic resonance (FMR) measurements demonstrate magnetic anisotropy values consistent with SQUID magnetometry measurements. FMR was measured in a coplanar waveguide geometry at frequencies between 5 and 20 GHz at 250 K. In the presence of a small modulating field of 0.2 mT and 700 Hz, we measured the RF power reflected from the end of the waveguide with a lock-in amplifier. The derivative of the reflected RF power with respect to the modulating field was measured as a function of the static external magnetic field, as illustrated by the FMR spectrum in Fig. 2(a). We extract the resonance field H_{FMR} at 5 GHz by fitting the FMR spectrum to the generalized Lorentzian derivative.

By measuring the frequency dependence of the resonance field H_{FMR} , we can extract the Landé g -factor, effective magnetization M_{eff} , and fourfold in-plane anisotropy field $H_{4\parallel}$ via the in-plane Kittel equation^{25,26} [see Fig. 2(b)]:

$$f = \frac{g\mu_B\mu_0}{h} [H_{\text{FMR}} + H_{4\parallel} \cos(4\phi)]^{1/2} \times \left[H_{\text{FMR}} + M_{\text{eff}} + \frac{1}{4}H_{4\parallel}(3 + \cos(4\phi)) \right]^{1/2}, \quad (1)$$

where ϕ is the angle between the in-plane magnetic field and the $[100]$ axis. Note that $M_{\text{eff}} = M_s + H_{2\perp}$, where M_s is the saturation magnetization and $H_{2\perp}$ is the uniaxial out-of-plane anisotropy field. Fitting along the $[110]$ axis yields $g = 1.98 \pm 0.02$, $\mu_0 M_{\text{eff}} = 1.70 \pm 0.03 \text{ T}$, and $\mu_0 H_{4\parallel} = -51.1 \pm 0.2 \text{ mT}$. Since it is harder to fully saturate the sample along the $[100]$ axis (see [supplementary material](#) Sec. III), the best-fit parameters along the $[100]$ axis have larger uncertainties. We do not expect M_s or $H_{2\perp}$ to vary in the (001) plane, so we can restrict $\mu_0 M_{\text{eff}}$ to 1.65–1.75 T for fitting along the $[100]$ axis. This yields $g = 1.99 \pm 0.04$, $\mu_0 M_{\text{eff}} = 1.65 \pm 0.07 \text{ T}$, and $\mu_0 H_{4\parallel} = -53.2 \pm 0.2 \text{ mT}$. The magnitude of fourfold in-plane anisotropy field $|\mu_0 H_{4\parallel}|$ of $\sim 50 \text{ mT}$ is in quantitative agreement with the saturation field $\mu_0 H_s = 62 \pm 3 \text{ mT}$ from the static magnetometry measurements.

Nonlocal magnon transport measurements, performed on LAFO films along the magnetically easier and harder in-plane directions, indicate nearly isotropic magnon transport. We used a nonlocal measurement geometry to probe magnon transport similar to

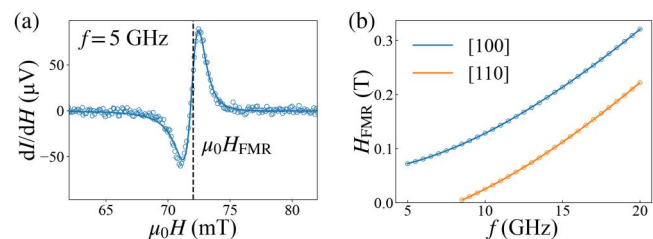


FIG. 2. (a) Field dependence of the FMR signal at 5 GHz and 250 K. (b) Frequency dependence of H_{FMR} at 250 K can be fit to the Kittel equation to deduce the Landé g -factor, effective magnetization M_{eff} , and in-plane anisotropy field $H_{4\parallel}$ along the $[100]$ and $[110]$ axes.

previous studies on garnet ferrites.^{27–31} As shown in Fig. S4, we deposited pairs of parallel Pt electrodes on the surface of LAFO films along the [100] and [110] directions and applied an AC at $\omega/2\pi = 9.8$ Hz to one of the electrodes, creating a vertical spin current via the spin Hall effect (SHE). This leads to spin accumulation at the Pt/LAFO interface, thereby applying an anti-damping torque and exciting magnons in the LAFO film. These magnons propagate toward the second Pt electrode a distance d away and are then detected electrically at this detector Pt electrode via the inverse spin Hall effect (ISHE). Additionally, the injection current generates a thermal gradient near the first electrode, producing magnons via the spin Seebeck effect (SSE). All magnons that arrive at the second electrode produce an electrical voltage via the ISHE. Since the SHE is proportional to the time-varying injection current $I(t) = I_0 \cos(\omega t)$, we detected electrically generated magnons from the first harmonic voltage $V_{1\omega}$. The SSE is proportional to the square of the injection current $I^2(t) = I_0^2 \cos^2(\omega t)$, so thermally injected magnons were detected from the second harmonic voltage $V_{2\omega}$. The nonlocal resistance is defined as $R_{\text{SHE}} = V_{1\omega}/I_0$ and $R_{\text{SSE}} = V_{2\omega}/I_0^2$, both of which are independent of the injection current I_0 .

We performed nonlocal measurements as a function of the in-plane orientation of the LAFO magnetization $\mathbf{M}_s$, which is completely saturated by an external magnetic field of 300 mT, well above the in-plane saturation field of 62 mT (see Fig. 1). This allows us to eliminate domain boundaries and study nonlocal transport in the uniformly saturated state. The angle between magnetization $\mathbf{M}_s$ and the direction of magnon propagation is denoted by γ . Figure 3 shows the γ

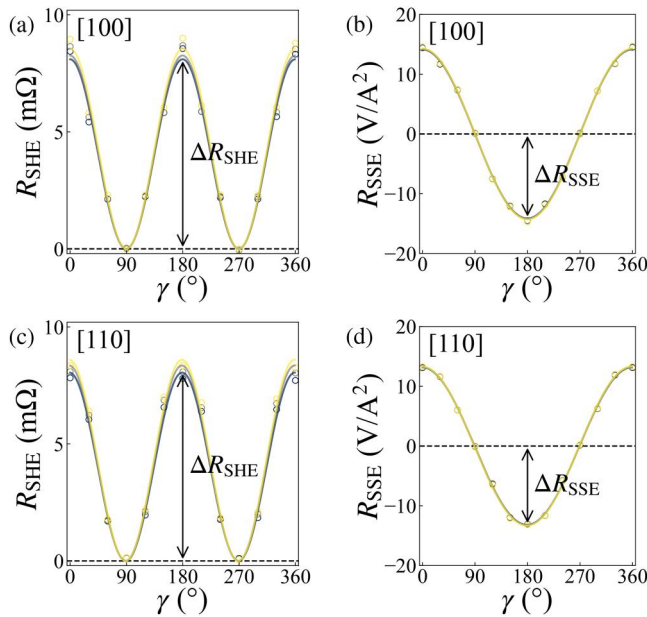


FIG. 3. Nonlocal spin transport measurements along the [100] directions show (a) SHE nonlocal resistance R_{SHE} and (b) SSE nonlocal resistance R_{SSE} as a function of the angle γ between the magnetization and the magnon propagation direction. Similar SHE and SSE measurements are shown for magnon propagation along the [110] direction in (c) and (d). These measurements were taken on a device with Pt electrodes a distance $d = 0.5 \mu\text{m}$ apart at 250 K and 300 mT with 3 different injection currents I_0 : 0.4 mA (yellow), 0.5 mA (gray), and 0.6 mA (blue).

dependence of R_{SHE} and R_{SSE} . R_{SHE} has a $\cos^2\gamma$ dependence since the SHE and the ISHE each contribute a factor of $\cos\gamma$. R_{SSE} only has a $\cos\gamma$ dependence from the ISHE because the SSE is independent of the direction of magnetization. By fitting to such γ dependence, we extracted the amplitudes ΔR_{SHE} and ΔR_{SSE} . We performed this measurement for magnons propagating along the [100] and [110] directions. For current values ranging from 0.4 to 0.6 mA, there is no significant difference in ΔR .

By measuring the nonlocal resistance on an array of parallel Pt electrodes with different separation distances d , we have deduced the spin diffusion length in LAFO films. Since the thickness of the LAFO film around 12 nm is much smaller than the distance d between electrodes, and the distance d is much smaller than the length of each electrode at $40 \mu\text{m}$, we can assume that magnons propagate in one dimension. The nonlocal resistance ΔR as a function of the separation distance d is therefore described by the one-dimensional magnon diffusion equation:²⁷

$$\Delta R \propto \frac{e^{-d/\lambda}}{1 - e^{-2d/\lambda}}, \quad (2)$$

where λ is the spin diffusion length. This equation encompasses both the diffusion-limited regime for small d and the relaxation-limited regime for d up to 3–5 times λ ,^{27,30} consistent with the range of d in our devices.

We measured the d dependence of ΔR_{SHE} and ΔR_{SSE} as shown in Fig. 4. By fitting the d dependence of ΔR to Eq. (2), we can estimate λ along the [100] and [110] directions. Table I lists the spin diffusion lengths λ for SHE and SSE generated magnons along the [100] and

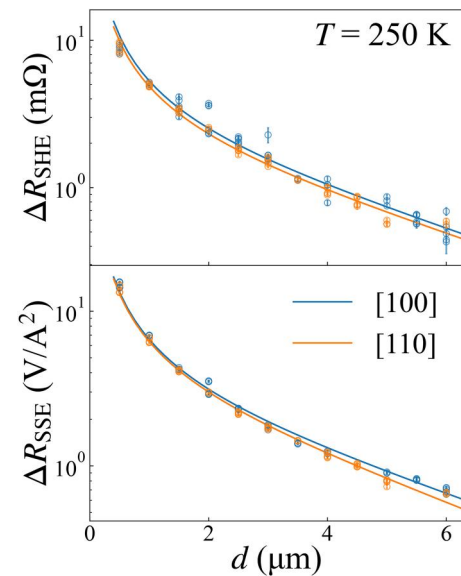


FIG. 4. Amplitudes of the SHE nonlocal resistance ΔR_{SHE} (upper panel) and amplitudes of the SSE nonlocal resistance ΔR_{SSE} (lower panel) as a function of the magnon propagation distance d along the [100] and [110] directions at 250 K. Measurements were taken with an injection charge current of $I_0 = 0.4$ mA, 0.5, and 0.6 mA on each device and one or two devices at the same distance d . A single best-fit curve is plotted for each orientation and for each type of nonlocal resistance.

TABLE I. Spin diffusion length λ for SHE and SSE generated magnons propagating along the [100] and [110] axes at 250 K.

Orientation	λ_{SHE} (μm)	λ_{SSE} (μm)
[100]	3.21 ± 0.21	3.25 ± 0.10
[110]	3.24 ± 0.15	2.93 ± 0.07

[110] directions. At 250 K, λ is independent of the magnon propagation direction ([100] vs [110]) and the generation mechanism (SHE vs SSE). Our results show that the in-plane spin diffusion length is similar along the [100] and [110] directions.

Our nonlocal measurements provide compelling evidence that magnon transport in (001) LAFO films exhibit isotropic behavior with spin diffusion lengths on the order of $3 \mu\text{m}$. This result can be understood by considering the spin diffusion length λ classically as:³² $\lambda = \sqrt{D\tau_{\text{nc}}}$, where $D = \frac{1}{3}\langle v_g^2 \tau_c \rangle$ is the diffusion constant, v_g is the magnon group velocity, and τ_{nc} and τ_c are the magnon non-conserving and magnon conserving relaxation times, respectively. If τ_{nc} and τ_c have any orientation dependence, then we would observe deviations from the expected behavior of $R_{\text{SHE}} \sim \cos^2(\gamma)$ and $R_{\text{SSE}} \sim \cos(\gamma)$. The lack of such orientation dependence means that both types of magnon relaxation time are isotropic, similar to MAFO.²²

Next, we consider the crystallographic orientation dependence of the magnon group velocity v_g in LAFO. The crystallographic orientation dependence of v_g of magnons is predominantly determined by the anisotropy of the exchange stiffness, which has a significant effect on magnons with larger wavenumber k .²² To estimate the anisotropy in the exchange stiffness, we consider the exchange interaction between neighboring Fe^{3+} ions mediated by O^{2-} anions. Since our LAFO films stabilize with a cubic crystal structure with slight tetragonal distortion, we expect little, if any, in-plane anisotropy in the exchange interaction, i.e., $J_{xx} = J_{yy} = J_{zz}$.^{33–36} The anisotropy of the exchange stiffness can therefore arise from either the spinel crystal structure or an anisotropic exchange interaction J_{xy} . However, the complex network of exchange paths in the LAFO spinel structure provides strong angular averaging of the exchange kernel, leading to nearly isotropic exchange stiffness. Similar angular averaging arising from complex exchange paths also underlies the nearly isotropic magnon dispersion observed in YIG.^{37,38} The anisotropy in exchange interaction can also arise when spin-orbit coupling (SOC) is incorporated into the virtual hopping processes underlying exchange. We consider the anisotropic exchange interaction that is symmetric in the spin indices, e.g., $J_{xy} \neq 0$. Its magnitude can be estimated as $\sim (\Lambda_{\text{SOC}}/\Delta)^2$, where Λ_{SOC} is the strength of the spin-orbit coupling in Fe^{3+} and Δ is the exchange splitting of the Fe^{3+} ions.^{34,39} In our LAFO films, the SOC strength is on the order of $\Lambda_{\text{SOC}} \sim 70 \text{ meV}$ for an Fe^{3+} ion,⁴⁰ and the energy scale of the exchange splitting Δ is on the order of 1 eV for 3d transition metals.⁴¹ Using these values, we estimate the symmetric exchange anisotropy to be less than 1% of the isotropic exchange interaction. In addition, SOC causes the Landé g -factor to deviate from the free-electron value resulting in $(\Lambda_{\text{SOC}}/\Delta) \sim (g-2)$.³⁴ Experimentally, we find $|g-2| \leq 0.1$ [see Fig. 2(b) and supplementary material Fig. S2(a)], which is also consistent with the aforementioned estimate. The anisotropy of the exchange stiffness can be further verified by magnon dispersion experiments, such as Brillouin light scattering^{42,43} and resonant

inelastic x-ray scattering,⁴⁴ performed along different crystallographic directions. Complementary *ab initio* density functional theory modeling of the magnon dispersion could also shed light on the anisotropic exchange stiffness.⁴³

Similar to LAFO, MAFO also has a cubic crystal structure and weak SOC from Fe^{3+} ions, indicating a nearly isotropic exchange interaction. In contrast to the isotropic magnon transport in the plane in LAFO films, a 30% anisotropy in the spin diffusion length has been reported in MAFO, which was attributed to an anisotropy in the exchange stiffness.²² Note that MAFO has a smaller in-plane magnetocrystalline anisotropy, with an in-plane anisotropy field of 10 mT at room temperature,¹⁵ compared to LAFO of around 40 mT [see supplementary material Fig. S2(a)]. The SOC in MAFO is therefore on the same order as LAFO, if not smaller. Therefore, SOC cannot explain the anisotropic exchange stiffness in MAFO.

Nevertheless, MAFO has a smaller inhomogeneous broadening than LAFO, so the averaging of the exchange kernel may be less effective. The inhomogeneous broadening is usually quantified by the zero-frequency FMR linewidth ΔH_0 , which is consistently less than 0.2 mT in MAFO¹⁵ but higher in LAFO about 0.4–1.2 mT [see supplementary material Fig. S2(b)]. The increased inhomogeneous broadening in LAFO can be attributed to the volatile nature of the Li^+ ions, which may have a non-uniform composition throughout the sample and lead to magnetic inhomogeneities. The presence of fewer inhomogeneities in MAFO might provide less averaging of the exchange kernel, leading to anisotropic magnon dispersion despite nearly isotropic exchange interaction J . However, the anisotropy of the magnon dispersion in MAFO still requires experimental verification.

We further highlight another distinction in the crystallographic dependence of magnon transport between LAFO and MAFO. Apart from an anisotropic spin diffusion length, MAFO also has an anisotropy in the injected magnon densities, for both SHE and SSE generated magnons. Notably, the nonlocal signal at small spacings d between the two electrodes is consistently larger in the harder [100] direction compared to the easier $[1\bar{1}0]$ direction in MAFO, indicating a higher injected magnon density along the harder [100] direction.²² This is in contrast to LAFO, where we find no systematic difference in the nonlocal signal along the [100] and [110] directions at all spacings, as shown in Fig. 4. Indeed, at small d , the magnon densities generated by the two injection methods tend to converge, indicating that magnons are equally injected for electrodes oriented along the [100] and [110] directions. This crucial difference between MAFO and LAFO leads us to suspect that the magnon injection mechanisms are dependent on the crystallographic direction in MAFO, which may underlie the divergent behavior at small d and might also lead to an anisotropic spin diffusion length.

In summary, we have demonstrated that magnon transport in epitaxial LAFO thin films with low magnetic loss is nearly isotropic with respect to the film plane, contrary to epitaxial MAFO thin films. Spin diffusion lengths of both types of magnons measured along the in-plane [100] axis are similar to those measured along [110] axis by nonlocal magnon transport technique, despite the in-plane magnetocrystalline anisotropy measured by both static SQUID magnetometry and dynamic broadband FMR. The nearly isotropic magnon transport in low damping LAFO thin films, while expected from a nearly isotropic exchange energy, is promising for incorporation into magnon waveguides and interconnects.

See the [supplementary material](#) for additional structural and magnetic characterizations of LAFO as well as the nonlocal measurement setup.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Yiming Li: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Katya Mikhailova:** Conceptualization (equal); Data curation (equal); Investigation (equal); Methodology (equal); Writing – review & editing (equal). **Lerato Takana:** Conceptualization (equal); Data curation (equal); Investigation (equal); Methodology (equal); Writing – review & editing (equal). **Daisy O'Mahoney:** Data curation (supporting); Formal analysis (supporting); Investigation (supporting); Writing – review & editing (supporting). **Sauviz P. Alaei:** Data curation (supporting); Investigation (supporting). **Guanxiong Qu:** Formal analysis (equal); Writing – review & editing (equal). **Dominic Petruzz:** Data curation (supporting); Writing – review & editing (supporting). **Samuel Crossley:** Data curation (supporting). **Harold Y. Hwang:** Resources (supporting); Supervision (supporting). **Ian R. Fisher:** Resources (supporting); Supervision (supporting). **Clare C. Yu:** Resources (supporting); Supervision (supporting); Writing – review & editing (supporting). **Yuri Suzuki:** Conceptualization (equal); Funding acquisition (lead); Supervision (lead); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are openly available in the Stanford Digital Repository at <https://doi.org/10.25740/dn330pg4198>, Ref. 45.

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