Supplementary Materials for

Nonreciprocal surface acoustic wave propagation via magneto-rotation coupling

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Section S1. Theoretical modelling

We present a theoretical description of surface acoustic wave absorption by a ferromagnetic thin film. We model the experimental setup by an isotropic elastic material filling the half-space \( z < d/2 \) with its top layer of thickness \( d \) assumed to be ferromagnetic with magnetization \( M \). Although the elastic properties could be anisotropic and differ between the magnet and the substrate, this simplified model turns out to be sufficient for our purposes. Let \( u_{x,y,z} \) be the Cartesian components of the displacement vector field. The acoustic waves in isotropic media are characterized by Lamé constants \( \lambda, \mu \) through the elastic free energy

\[
F = \int d^3r \left\{ \frac{\lambda}{2} (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})^2 + \mu (\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + \varepsilon_{zz}^2 + 2\varepsilon_{xy}^2 + 2\varepsilon_{yz}^2 + 2\varepsilon_{zx}^2) \right\} \tag{S1}
\]

where \( \varepsilon_{ij} = (\partial_i u_j + \partial_j u_i)/2 \) are the components of the strain tensor. The dynamics of \( \mathbf{u} \) with the isotropic free energy is studied in any textbook on continuum mechanics. In particular, the longitudinal and transverse bulk acoustic waves travel at respective speeds of sound \( c_L = \sqrt{(\lambda + 2\mu)/\rho}, c_T = \sqrt{\mu/\rho} \) where \( \rho \) is the mass density, and the surface acoustic waves propagating in positive and negative \( x \) directions with respective wavenumbers \( \pm k, k > 0 \) along the surface \( z = d/2 \) are described by the solution

\[
\begin{align*}
(u_x^\pm & , u_z^\pm) = C \text{Re} \left[ \frac{1}{\mp i} \left\{ (1 - \xi_T^2/2) \left( 2e^{\kappa_L(z-d/2)} - (2 - \xi_T^2)e^{\kappa_T(z-d/2)} \right) \right\} e^{-i(\omega t \mp kx)} \right] \tag{S2}
\end{align*}
\]

where \( C \) is an arbitrary real constant, and the parameters \( \kappa_{L,T}, \xi_{L,T} \) are related to the bulk speeds of sound \( c_{L,T} \) and the surface speed of sound \( c_S \) by

\[
\kappa_{L,T} = \frac{k^2 - \omega^2}{c_{L,T}^2}, \xi_{L,T} = \frac{c_S}{c_{L,T}}. \tag{S3}
\]

Note that the value of \( c_S \) depends implicitly on \( c_{L,T} \) through the algebraic equation

\[
\xi_T^6 - 8\xi_T^4 + 8 \left( 3 - \frac{2c_T^2}{c_L^2} \right) \xi_T^2 - 16 \left( 1 - \frac{c_T^2}{c_L^2} \right) = 0. \tag{S4}
\]

The “spin”-momentum locking of surface acoustic waves manifests itself in the phase difference between \( u_x^\pm \) and \( u_z^\pm \) by \( \mp i = e^{i\pi/2} \), which changes the sign under \( k \rightarrow -k \).

We regard the surface acoustic wave solution (S2) with fixed \( \omega \) and \( k \) being given as an effective rf field and study how the magnet responds. Back reactions of magnetization dynamics onto acoustic waves are neglected. The purely magnetic part of the free energy is assumed to include an external magnetic field, cubic crystalline and interface-induced uniaxial magnetic anisotropies, and exchange, dipole-dipole and Dzyaloshinskii-Moriya interactions:

\[
W = \int d^3r \left[ -\mu_0 M_s H \cdot \mathbf{n} - \frac{A}{2} \mathbf{n} \cdot \nabla^2 \mathbf{n} + \frac{\mu_0 M_s^2}{8\pi} \int d^3r' (\mathbf{n} \cdot \nabla)(\mathbf{n}' \cdot \nabla') \frac{1}{|\mathbf{r} - \mathbf{r}'|} \right.
\]

\[
+ K_e (n_x^2 n_y^2 + n_y^2 n_z^2 + n_z^2 n_x^2) - K_L n_x^2 - K_H n_z^2 + D \mathbf{n} \cdot \left\{ \hat{x} \partial_y - \hat{y} \partial_x \times \mathbf{n} \right\}, \tag{S5}
\]
where $M_s$ is the saturation magnetization, $\mathbf{n} = \mathbf{M}/M_s$, $\mathbf{n}'$ is the value of $\mathbf{n}$ evaluated at $\mathbf{r}'$, $\nabla'$ is the spatial derivative with respect to $\mathbf{r}'$ and $\hat{x}', \hat{y}'$ are unit vectors in $x$ and $y$ directions. Note that $K_\perp$ arises from the interface inversion symmetry breaking while $K_\parallel$ is present due to the crystallographic $c$-axis alignment of the single crystalline LiNbO$_3$ substrate. Taking the external field to be spatially homogeneous and in-plane $\mathbf{H} = H(\cos \phi, \sin \phi, 0)$, the ground state magnetization is also in the plane $\mathbf{n} = (\cos \theta, \sin \theta, 0)$ and $\theta = \phi$ for sufficiently strong $\mathbf{H}$ if $K_c = K_\parallel = 0$. The spin waves excited by the surface acoustic waves $\mathbf{u}^\pm$ have wavevectors $\pm k\hat{x}$ respectively. In the thin film limit $kd \ll 1$, linear perturbation around the ground state yields the dispersion relation

$$\omega_k = |\gamma| \mu_0 \sqrt{[H \cos(\theta - \phi) + H_v(\theta)] [H \cos(\theta - \phi) + H_z(\theta)] + \gamma \mu_0 D_{\text{DMI}}(\theta)}, \quad (S6)$$

where $\gamma < 0$ is the gyromagnetic ratio and

$$H_v(\theta) = \frac{Ak^2}{\mu_0 M_s} + M_s \left(1 - \frac{1-e^{-kd}}{kd}\right) \sin^2 \theta + \frac{2K_\perp \cos 2\theta - K_c (1 - 3 \cos^2 2\theta)}{\mu_0 M_s}, \quad (S7)$$

$$H_z(\theta) = \frac{Ak^2}{\mu_0 M_s} + M_s \frac{1-e^{-kd}}{kd} - \frac{2K_\perp - 2K_c \cos^2 \theta - K_c (1 + \cos 2\theta)}{\mu_0 M_s}, \quad (S8)$$

$$H_{\text{DMI}}(\theta) = \pm \frac{2Dk \sin \theta}{\mu_0 M_s}. \quad (S9)$$

Note that we throughout use the convention to take frequencies to be positive. In the main text, we chose not to discuss $K_\parallel$ and $K_c$, which we shall see are small, and denoted the total perpendicular anisotropy by $K_u = K_\perp - \mu_0 M_s^2/2$. For a given driving frequency $\omega$, the resonance field is determined by

$$H_{\text{res}} \cos(\theta - \phi) = -\left(\frac{H_v + H_z}{2} + \sqrt{(H_v - H_z)^2 + 4(H_{\text{DMI}} - \omega/\gamma \mu_0)^2}\right). \quad (S10)$$

and expanding the square root to linear order in $H_{\text{DMI}}$ (and setting $\theta = \phi$) yields Eq. (2) in the main text.

There are a variety of ways in which $\mathbf{M}$ interacts with acoustic waves. For cubic crystals, one usually includes the conventional magnetoelastic coupling in the free energy

$$L_1 = \int d^3r \left\{b_1(n_x^2 \varepsilon_{xx} + n_y^2 \varepsilon_{yy} + n_z^2 \varepsilon_{zz}) + 2b_2(n_x n_y \varepsilon_{xy} + n_y n_z \varepsilon_{zx} + n_z n_x \varepsilon_{xx})\right\}. \quad (S11)$$

It turns out to be insufficient, however, for explaining the large non-reciprocal response seen in our experiment. For this purpose, we consider free energy terms that describe interactions between $\mathbf{M}$ and the rotation of elastic deformations $\omega_{ij} = (\partial_j u_i - \partial_i u_j)/2$. There are many possible mechanisms for magneto-rotation coupling, and here we discuss those that are directly related to the purely magnetic free energy $W$. As first pointed out by Maekawa and Tachiki(12), magnetic anisotropy fields induce magneto-rotation couplings through reorientations of crystalline axes, which for uniaxial and cubic anisotropies read

$$L_2 = 2K_\perp \int d^3r \left\{\omega_{zx} n_x + \omega_{zy} n_y \right\}n_x + 2K_c \int d^3r \left(\omega_{xy} n_y + \omega_{xz} n_x \right)n_x$$

$$+ 2K_c \int d^3r \left[n_x n_u (n_z^2 - n_y^2) \omega_{xy} + n_y n_z (n_x^2 - n_z^2) \omega_{yx} + n_x n_z (n_x^2 - n_y^2) \omega_{yz}\right] \quad (S12)$$

Similarly, the dipolar shape anisotropy results in a magneto-rotation coupling via change of the surface normal directions induced by SAWs. Based on the model of coupling between magnons and surface deformations given in (21), one derives the interaction energy.
\[ I_3 = \frac{\mu_0 M_s^2}{8\pi} \int d^3r \int d^3r' \left[ \sum \left( \frac{\delta \left( z - \frac{d}{2} \right)}{2} - \frac{\delta \left( z + \frac{d}{2} \right)}{2} \right) u_z(r) 
+ \left\{ \frac{\delta \left( z' - \frac{d}{2} \right)}{2} - \frac{\delta \left( z' + \frac{d}{2} \right)}{2} \right\} u_z(r') \right] (n \cdot \nabla)(n' \cdot \nabla) \frac{1}{|r - r'|}. \]  

(\text{S13})

Although this contains both strain and rotation, we shall see shortly that for in-plane magnetization and surface acoustic waves, the strain can be neglected. The Dzyaloshinkii-Moriya interactions are also affected by the crystalline orientation and consequently generate couplings between magnet and lattice deformations;

\[ I_4 = D \int d^3r n \cdot \left\{ \sum_{a=x,z} (\varepsilon_{xa} - \omega_{xa}) \hat{f}_a \partial_y - \sum_{a=x,z} (\varepsilon_{ya} - \omega_{ya}) \hat{f}_a \partial_x 
+ \hat{x} \sum_{a=x,z} (\varepsilon_{ya} - \omega_{ya}) \partial_a - \hat{y} \sum_{a=x,z} (\varepsilon_{xa} - \omega_{xa}) \partial_a \right\} \times n \right\}, \]  

(\text{S14})

where \( \hat{f}_x = \hat{x}, \hat{f}_y = \hat{y}, \hat{f}_z = \hat{z} \). Again, the strain couplings are discarded later. Finally, when the microscopic magnetic moments may be considered fixed on individual atomic sites and adiabatically following the motion of the lattice, there will be an analogue of Coriolis force called spin-rotation coupling (22) given by

\[ I_5 = \frac{hS}{V} \int d^3r \left( n_x \partial_t \omega_{xy} + n_y \partial_t \omega_{xz} + n_z \partial_t \omega_{xy} \right), \]  

(\text{S15})

where \( S/V \) is the effective length of spin per unit cell.

Each of the interaction terms introduces an effective rf field \( h_a \) acting on the magnetization \( M \) where \( h_a = -\delta l_a / (\mu_0 M_s n), a = 1, \cdots, 5. \) Evaluating these fields for the ground state configuration of \( M \) yields

\[ h_1 = \frac{b_1 (\varepsilon_{xx} - \varepsilon_{yy}) \sin 2\theta - 2b_2 \varepsilon_{xxy} \cos 2\theta}{\mu_0 M_s} \hat{v} - \frac{2b_2 (\varepsilon_{xx} \cos \theta + \varepsilon_{yy} \sin \theta)}{\mu_0 M_s} \hat{z}, \]  

(\text{S16})

\[ h_2 = 2K_{\parallel} \cos 2\theta - 2K_c \cos 4\theta \]  

(\text{S17})

\[ h_3 = M_s \frac{\hat{v} \cdot \nabla + \hat{z} \partial_z}{4\pi} \]  

(\text{S18})

\[ h_4 = \frac{D}{\mu_0 M_s} \left[ \partial_y (\varepsilon_{xz} + \omega_{xz}) - \partial_x (\varepsilon_{yz} - \omega_{yz}) \right] \hat{v} 
+ \partial_x \left[ (\varepsilon_{xx} + \omega_{xx}) \cos \theta + (\varepsilon_{yy} - \omega_{yz}) \sin \theta \right] \hat{z}, \]  

(\text{S19})

\[ h_5 = -\frac{hS}{\mu_0 M_s V} \partial_t \left[ (\omega_{xx} \cos \theta - \omega_{yz} \sin \theta) \hat{v} + \omega_{xy} \hat{z} \right], \]  

(\text{S20})
where $\hat{v} = -\hat{x}\sin\theta + \hat{y}\cos\theta \perp \hat{n}_0$ and the in-plane components have been projected onto this axis. To obtain Eq. (1) of the main text, we set $K_t = K_c = 0$ and discarded $h_4$ and $h_5$, which is justified in the next section. The main contribution of $h_3$ is to replace $K_\perp$ in $h_2$ by $K_u$. For the surface acoustic waves $u^\pm$ propagating in the positive and negative $x$ directions, their non-vanishing components are given as follows:

$$
\varepsilon_{xx}^\pm = \pm iC \left(1 - \frac{\xi_T^2 \xi_L}{2}\right) \left\{ e^{\kappa_L (z - \frac{d}{2})} - \left(1 - \frac{\xi_T^2 \xi_L}{2}\right) e^{\kappa_T (z - \frac{d}{2})} \right\}, \quad (S21)
$$

$$
\varepsilon_{zz}^\pm = \mp iC \left(1 - \frac{\xi_T^2 \xi_L}{2}\right) \left\{ (1 - \xi_T^2) e^{\kappa_L (z - \frac{d}{2})} - \left(1 - \frac{\xi_T^2 \xi_L}{2}\right) e^{\kappa_T (z - \frac{d}{2})} \right\}, \quad (S22)
$$

$$
\varepsilon_{zz}^\pm = C \left(1 - \frac{\xi_T^2 \xi_L}{2}\right) \sqrt{1 - \xi_T^2} \left\{ e^{\kappa_L (z - \frac{d}{2})} - e^{\kappa_T (z - \frac{d}{2})} \right\}, \quad (S23)
$$

$$
\omega_{xx}^\pm = -C \frac{\xi_T^2 \xi_L}{2} \sqrt{1 - \xi_T^2} e^{\kappa_T (z - \frac{d}{2})}. \quad (S24)
$$

Note that we now omit the operation of taking real parts. Due to the free surface boundary conditions used to derive the solution, $\varepsilon_{xz}$ vanishes at the surface and is smaller by a factor of $k\ell$ in the magnetic region than $\varepsilon_{xx,zz}$ and $\omega_{xx}$. Thus in the thin film limit, one can ignore $\varepsilon_{xz}$ in $h_1, h_3$ and $h_4$.

The linearized Landau-Lifshitz-Gilbert equation with Gilbert damping $\alpha$, after Fourier transforms in time and space, reads

$$
\begin{align*}
\begin{pmatrix}
\frac{d}{dt}v \\
\frac{d}{dt}z
\end{pmatrix}
&= \frac{1}{(H + H_v)(H + H_z) - \alpha^2 H_0^2 + i\alpha H_\omega (2H + H_v + H_z) - (H_\omega - H_{DMI})^2} \\
&\times \begin{pmatrix}
H + H_\omega + i\alpha H_\omega & -i(H_\omega - H_{DMI}) \\
i(H_\omega - H_{DMI}) & H + H_v + i\alpha H_\omega
\end{pmatrix}
\begin{pmatrix}
h_v \\
h_z
\end{pmatrix}, \quad (S25)
\end{align*}
$$

where we set $\theta = \phi$ for simplicity (i.e. ignoring the effect of cubic anisotropy on the ground state), introduced $H_\omega = \omega/\gamma\mu_0$, and the components of the total effective field $h^\pm$ from SAW with wavenumber $\pm k$ are given by

$$
\begin{align*}
h_v^\pm &= iC \cos\phi \frac{\xi_T^2 \xi_L}{2} \left\{ -b_1 (2 - \xi_T^2) \sin\phi + \frac{h_\omega S}{V} \sqrt{1 - \xi_T^2} \right\}, \quad (S26)
\end{align*}
$$

$$
\begin{align*}
h_z^\pm &= \frac{C \cos\phi}{\mu_0 M_s} \left\{ \left(K_\perp - \frac{\mu_0 M_s^2}{2} - K_\parallel - K_c \cos^2\phi \right) \xi_T^2 \sqrt{1 - \xi_T^2} - Dk\xi_T^2 \left(1 - \frac{\xi_T^2}{2}\right) \right\}. \quad (S27)
\end{align*}
$$

The constant $C$ is irrelevant as it multiplies the overall magnitude of the excited spin waves. The energy absorbed from SAW into spin waves per unit time is expected to be proportional to the power $P_{\pm k}$ dissipated by the spin waves, which is equal to the amount of work done by $h^\pm$

$$
\begin{align*}
P &= \mu_0 M_s \left\{ \text{Re} \left[ \frac{\partial}{\partial t} \right] \cdot \text{Re}[h^\pm] \right\}, \quad (S28)
\end{align*}
$$

where the angled bracket denotes averaging over one period of SAW. Substituting Eqs., (S25), (S26) and (S27), one derives the formula for SAW absorption.
\[ P_{\pm k} = \frac{C^2 \alpha \omega^2 \xi_T^4 \cos^2 \phi / 2 |\gamma| \mu_0 M_s}{\{(H_\omega - H_{DME})^2 - (H + H_0)(H + H_z) + \alpha^2 H_0^2\}^2 + \alpha^2 H_0^2(2H + H_v + H_z)^2 \times \{[(H + H_z)^2 + (H_\omega - H_{DME})^2 + \alpha^2 H_0^2](\pm \rho_{ME} \sin \phi + \rho_{SR})^2\}^2 + 2(H_\omega - H_{DME})(2H + H_v + H_z)(\pm \rho_{ME} \sin \phi + \rho_{SR})\rho_{MR} + \{(H + H_v)^2 + (H_\omega - H_{DME})^2 + \alpha^2 \Omega^2\}\rho_{MR}^2, \quad (S29) \]

The parameters \( \rho_{ME,SR,MR} \) measure, in unit of energy density, the contributions from magneto-elastic, spin-rotation and magneto-rotation couplings respectively, defined by

\[ \rho_{ME} = b_1 \left(1 - \frac{\xi_T^2}{2}\right), \rho_{SR} = \frac{\hbar \omega}{2V} \sqrt{1 - \frac{\xi_T^2}{2}}, \quad (S30) \]
\[ \rho_{MR} = \left(K_\perp - \frac{\mu_0 M_s^2}{2} - K_\parallel - K_c \cos^2 \phi\right) \sqrt{1 - \frac{\xi_T^2}{2}} - \frac{Dk_E \xi_T^2}{2\xi_T^2} \left(1 - \frac{\xi_T^2}{2}\right), \quad (S31) \]

It can be clearly observed that nonreciprocity, i.e. dependence of \( P_{\pm k} \) on the sign \( \pm \), comes from cross terms between \( \rho_{ME} \) and \( \rho_{SR} \) or \( \rho_{ME} \) and \( \rho_{MR} \). Any \( \pm \) dependence is accompanied by a factor \( \sin \phi \), which is dictated by the time-reversal symmetry. Note that in Eq. (S29), the dependence of the absorption on the strength of external magnetic field has been all explicitly spelt out so that it may be directly used to fit the SAW transmission data.

**Section S2. Details of the attenuation fitting**

Although the expression (S29) can, in principle, be directly compared with the measured attenuation of SAWs, fitting of the data in practice is not entirely straightforward due to the relatively large number of fitting parameters. It should also be mentioned that the modeling is oversimplified in some aspects so that some discrepancies between the data and the theory are inevitable due to the neglected factors including crystalline anisotropy of LiNbO\(_3\), roughness of the interfaces, and the difference in elastic properties between the different materials among other things. To extract the salient features of the relevant physics under discussion in this article, we split the fitting procedure into several steps, which is guided by the underlying physics. Throughout, fitting functions are denoted by block capitals and the corresponding script letters are used for the experimental data to be fitted.

**Step 1: Lorentzian fitting.**

Let \( P_{\pm k}(H, \phi) \) be the attenuation data as a function of the external field \( H \) for a fixed angle \( \phi \) and wavenumber \( \pm k \). We fit each data set \( P_{\pm k}(H, \phi) \) by a function \( P_{\pm k}(\phi) \) with three parameters \( \mathcal{A}_{\pm k}(\phi), \mathcal{H}_{\pm k}^{res}(\phi), \Delta \mathcal{H}_{\pm k}(\phi) \):

\[ P_{\pm k}(\phi) = \frac{1}{2\pi} \frac{\mathcal{A}_{\pm k}(\phi) \Delta \mathcal{H}_{\pm k}(\phi)}{(H - \mathcal{H}_{\pm k}^{res}(\phi))^2 + \Delta \mathcal{H}_{\pm k}(\phi)^2}/4 \quad (S32) \]

The best fit values for the parameters are now fed into the next steps as the data points.

**Step 2: Resonance field fitting.**

We determine five parameters \( A, K_\perp, K_\parallel, K_c, D \) by fitting the data \( \mathcal{H}_{\pm k}^{res}(\phi) \) as a function of \( \phi \) by the function

\[ \mathcal{H}_{\pm k}^{res}(\phi) = \frac{-H_\nu(\phi) - H_z(\phi) + \sqrt{[H_\nu(\phi) - H_z(\phi)]^2 + 4\{\omega/\gamma \mu_0 - H_{DME}(\phi)\}^2}}{2} \quad (S33) \]

where \( H_{\nu,z,DME}(\phi) \) are given by Eqs. (S7) - (S9). We use the value \(|\gamma| = 29.4 \) quoted from Ref. and measure \( M_s \) independently by MPMS. The best fit values for the parameters
are given in TABLE. S1 and plotted in Fig. S1. The DMI constant value presented in the main text was obtained at this step.

### Table S1. Summary of resonance field fitting.

| Parameter  | Value                   |
|------------|-------------------------|
| $A$ ($J/m$) | $9.138 \times 10^{-11}$ |
| $K_\perp$ ($J/m^3$) | $6.134 \times 10^5$ |
| $K_\parallel$ ($J/m^3$) | $7.212 \times 10^3$ |
| $K_c$ ($J/m^3$) | $-9.023 \times 10^3$ |
| $D$ ($J/m^2$) | $8.9 \times 10^{-5}$ |

Note that the value of $A$ is presumably overestimated due to the unquantifiable spatial variation across the film thickness costing significant exchange energy.

### Step 3: Linewidth fitting.

In our simple phenomenology, the linewidth is independent of the angle and given by $\Delta H = \alpha |\omega/2\gamma\mu_0|$. We determine the value of Gilbert damping constant $\alpha$ by equating $\Delta H$ to the $\phi$- and $\pm k$-average of $H_{\pm k}(\phi)$. The estimated value is $\alpha = 0.059873$, and the fitting results are plotted in Fig. S2.

### Step 4: Amplitude fitting.

Finally, we carry out the fitting of the amplitude data $A_{\pm k}(\phi)$ by the function
The functions and parameters appearing in the above equation have all been obtained in the previous steps except for those to be fitted, i.e. $\tilde{C}$, $r_1$ and $r_2$. The overall constant $\tilde{C}$ does not contain any meaningful information. The other two $r_{1,2}$ are the central objects of interest in this study, which respectively measure the ratio of the magneto-rotation and spin-rotation coupling energies $\rho_{MR}$, $\rho_{SR}$ to the magnetoelastic coupling energy $\rho_{ME}$: $r_1 = \rho_{MR}/\rho_{ME}$, $r_2 = \rho_{SR}/\rho_{ME}$ (c.f. Eqs. (S30) and (S31)). The nonreciprocity arises from the terms that are linear in $r_{1,2}$. It turns out that these two parameters are highly degenerate: both of them can fit the data equally well on their own and when being fitted at the same time, the error bars tend to be much greater than when only one of them is fitted. The results of the fitting are given in TABLE. S2 and plotted in Fig. S3. In the end, we convert $A_{\pm k}(\phi)$ into $P_{\pm k}(\phi)$ and plot $P_{\pm k}(\phi)$ and rectifier ratio $[P_{+ k}(\phi) - P_{+ k}(\phi)]/[P_{+ k}(\phi) + P_{+ k}(\phi)]$ in Fig. S4.

**Table S2. Summary of amplitude fitting.**

| Fitting            | $C$      | $C_{err}$ | $r_1$    | $r_{1err}$ | $r_2$   | $r_{2err}$ |
|--------------------|----------|-----------|----------|------------|---------|------------|
| Fitting with $r_2 = 0$ | 192.340259 | 1.8981    | 0.187254 | 0.013432   | N/A     | N/A        |
| Fitting with $r_1 = 0$ | 192.229955 | 1.88400   | N/A      | N/A        | -0.08537 | 0.006042   |
| 3-parameter fitting | 1.717393  | -5.190078 | 0.976959 | -2.441063  | 0.442856 | 1.717393   |
Fig. S3. Fitting for $A_{+k}(\phi)$, $A_{-k}(\phi)$ and $[A_{+k}(\phi) - A_{-k}(\phi)]/[A_{+k}(\phi) + A_{-k}(\phi)]$ under conditions $r_2 = 0$, $r_1 = 0$ and $r_1, r_2 \neq 0$, respectively.

Fig. S4. Fitting for $P_{+k}(\phi)$, $P_{+k}(\phi)$ and $[P_{+k}(\phi) - P_{+k}(\phi)]/[P_{+k}(\phi) + P_{+k}(\phi)]$ under the condition $r_2 = 0$.

Even though the nonreciprocity data alone is insufficient to decide which of the magneto-rotation and spin-rotation couplings is the dominant mechanism, we can argue in favor of the former by considering how plausible the best fit values of $r_1, r_2$ are. First of all, we note that the value of $\rho_{MR}$ is completely known from the resonance field fitting and estimated to be $\rho_{MR} \sim -10^6$ J/m$^3$. In order to estimate $\rho_{SR}$, one would need to know the effective spin density $S/V$ for CoFeB thin films. Although it cannot be precisely determined due to the uncertainties in the microscopic magnetic structure, one could safely assume $S/V < 10^{30}$ m$^3$ since $S \sim O(1)$ and the unit cell size cannot be smaller than 1 Å. Thus for $\omega = 2\pi \times 6.1$ GHz, one obtains $\rho_{SR} < 6\pi \times 10^5$ J/m$^3$. Therefore, $|r_2|$ would be at best comparable to $|r_1|$ even in the most optimistic scenario. While we do not know the value of $b_1$ for our sample, typical values for transition metals are of order $10^7$ J/m$^3$ (23) so that the best fit value $r_1 \approx 0.2$ is very reasonable while achieving $r_2 \approx -0.1$ would require a significantly lower magnetostriction for CoFeB than Co or Fe alone. These estimates also suggest that the shear strain mechanism should be far less effective than the magneto-rotation coupling for our thin films as $b_2$ should be of the same order as $b_1$ and $kd \sim 1/500$. Therefore, we conclude that the observed giant nonreciprocity is mainly due to the
magneto-rotation coupling induced by the uniaxial anisotropy field \( K_u = K_\perp - \mu_0 M_s^2 / 2 \). Although we are unable to exclude a contribution from the spin-rotation coupling, it would be at most of similar order of magnitude to the contribution from the magneto-rotation coupling.

Section S3. Influence of the substrate anisotropy on the angular dependence of the SAW attenuation

The theoretical model used for fitting the data above assumes an isotropic elastic medium. However, LiNbO₃ is trigonal and its elastic properties are anisotropic. Here we consider possible corrections to the SAW attenuation signal arising from the substrate anisotropy. The power dissipated by spin waves is given by

\[
P = -\frac{\alpha M_s \omega^2}{2\gamma} \left[ \frac{1}{((H + H_v)(H + H_z) - (H_\omega - H_{DMI})^2)^2 + \alpha^2 (2H + H_v + H_z)^2 H_\omega^2} \right]^{-1}
\]

\[
= -\frac{\alpha M_s \omega^2}{2\gamma} \left[ \frac{1}{((H + H_v)(H + H_z) - (H_\omega - H_{DMI})^2)^2 + \alpha^2 (2H + H_v + H_z)^2 H_\omega^2} \right]^{-1}
\]

\[
\times \left[ ((H + H_z) h_v + i(H_\omega - H_{DMI}) h_z)^2 \right.
\]

\[
+ \left. \left( (H + H_v) h_z - i(H_\omega - H_{DMI}) h_v \right)^2 \right] \right], \quad (S35)
\]

This is essentially Eq. (S29), written in terms of \( h_{vz} \) instead of \( \rho_{ME,MR,SR} \) via Eqs. (S26) and (S27). Assuming near resonance \( H \sim H^{res} \), we separate it into the Lorentzian and the residual amplitude

\[
P \approx \frac{\alpha |H_\omega| A / \pi}{(H - H^{res})^2 + \alpha^2 H_\omega^2}, \quad (S36)
\]

\[
A = \frac{\pi}{2} \frac{\mu_0 M_s |\omega|}{(2 H^{res} + H_v + H_z)^2}
\]

\[
\times \left[ ((H + H_z) h_v + i(H_\omega - H_{DMI}) h_z)^2 \right.
\]

\[
+ \left. \left( (H + H_v) h_z - i(H_\omega - H_{DMI}) h_v \right)^2 \right], \quad (S37)
\]

We are doing this splitting because this model evidently fails to fit the observed anisotropic linewidth data (which is down to the back reaction of spin waves onto SAWs) so that the amplitude part should be isolated in comparing the theory with the data. If the magnetic resonance is isotropic, i.e. \( H_z = H_v \) one also has \( (H + H_v)^2 = (H + H_z)^2 = (H_\omega - H_{DMI})^2 \) at the resonance. Although this approximation is not very good in the present setup where the dipolar shape anisotropy is clearly visible, for simplicity we take it here. Conventionally choosing \( H_\omega > 0 \), one obtains at the resonance

\[
A = \frac{\pi}{4} \mu_0 M_s |\omega||h_v + i h_z|^2, \quad (S38)
\]

With the cubic magneto-elastic coupling and out-of-plane uniaxial anisotropy, the effective magnetic field generated by acoustic waves is given by
\[ h_v = \frac{1}{\mu_0 M_s^2} \left\{ b_1 (\varepsilon_{xx} - \varepsilon_{yy}) \sin 2\phi - 2b_2 \varepsilon_{xy} \cos 2\phi \right\}, \quad (S39) \]
\[ h_z = -\frac{2}{\mu_0 M_s} \left\{ (b_2 \varepsilon_{xx} + K_\perp \omega_{xx}) \cos \phi + (b_2 \varepsilon_{yy} + K_\perp \omega_{yy}) \sin \phi \right\}, \quad (S40) \]

Suppose that the surface acoustic wave propagates in the \( x \) direction, but still has a nonzero \( y \) component of the deformation. In our original analysis, we did not include this component since it is absent for SAWs in isotropic media. The boundary conditions force \( \varepsilon_{zx} = \varepsilon_{zy} \) at the boundary, and the effective magnetic field reduces to
\[ h_v = \frac{1}{\mu_0 M_s} \left\{ b_1 \varepsilon_{xx} \sin 2\phi - 2b_2 \varepsilon_{xy} \cos 2\phi \right\}, \quad (S41) \]
\[ h_z = -\frac{2K_\perp}{\mu_0 M_s} \left( \omega_{xx} \cos \phi + \omega_{yy} \sin \phi \right), \quad (S42) \]

We cannot derive analytical expressions for the strain and vorticity tensor components in general anisotropic media, but here the purpose is to capture the qualitative trend. First of all, let us assume \( \varepsilon_{xx}, \omega_{xx} \) are given by those of the SAWs in isotropic media, meaning they are of a similar order of magnitude and have a phase difference of \( \pm \pi/2 \) for \( \pm k \) respectively. Next, \( \varepsilon_{xy} = \partial_x u_y/2, \omega_{zy} = \partial_z u_y/2 \) arise from the anisotropy correction so that they are expected to be smaller than \( \varepsilon_{xx}, \omega_{xx} \). For surface localized waves, one expects \( \partial_x \sim ik, \partial_z \sim \kappa > 0 \), where \( k, \kappa \) are real so that it is reasonable to assume the relative phase between \( \varepsilon_{xy} \) and \( \omega_{xx} \) is also \( \pm \pi/2 \). Hence we introduce the following parameterisation:
\[ b_1 \varepsilon_{xx} = ia, \quad 2K_\perp \omega_{xx} = b, \quad 2b_2 \varepsilon_{xy} = ice^{i\delta}, \quad 2K_\perp \omega_{yy} = de^{i\delta}, \quad (S43) \]
where \( a, b, c, d, \delta \) can be taken to be real. The experimental data already suggested \( |b/a| \sim 0.35 \) and \( c, d \) represent the anisotropy correction so that \( |c|, |d| \ll |a| \). \( a \) and \( b \) are even and odd with respect to \( \pm k \) respectively, while the behavior under \( +k \to -k \) is not known for \( c, d \). However, given \( \varepsilon_{xy} \sim ik u_y/2, \omega_{yy} \sim \kappa u_y/2 \), it is expected that one is odd and the other is even. One obtains
\[ A = \frac{\pi |\omega|}{4 \mu_0 M_s} |a \sin 2\phi - b \cos \phi - (c \cos 2\phi + d \sin \phi) e^{i\delta}|^2, \quad (S44) \]
where the cross term between \( a \sin 2\phi \) and \( b \cos \phi \) gives the main nonreciprocal term in the amplitude, while the anisotropy corrections may have angular dependencies that do not appear from the isotropic part, i.e. a term proportional to \( \sin 4\phi \). These terms can explain at least parts of the features in Fig. 3 that are not accounted for by our fitting curve.

**Section S4. Details of 100% nonreciprocity**

In the angular dependence spectrum near \( \phi =180^\circ \), there is an abrupt change of nonreciprocity (Fig. 3.). It is also the region where the nonreciprocity reaches its maxima. According to the theory, the maxima are indeed 100% as we shall demonstrate now. Defining nonreciprocity by \( (A_+ - A_-)/(A_+ + A_-) \) where \( A_\pm \) corresponds to \( A \) in Eq. (S44) evaluated for \( \pm k \), i.e. \( \pm b \) respectively, it is expected that 100% nonreciprocity may be achieved at angles where either \( A_+ \) or \( A_- \) is equal to zero. By considering the isotropic case with \( c = d = 0 \), this angle can be determined by the condition
\[2a \sin \phi + b = 0 . \quad (S45)\]

Since \(|b/2a| < 0\) in our sample, this always has a solution near \(\phi = 0, \pi\) and if \(b > 0\) one gets \(A_+ = 0\) at a \(\phi < 0\) for instance. And obviously \(A_+ = 0\) implies \((A_+ - A_-)/(A_+ + A_-) = -1\), i.e. 100% non-reciprocity.

Fig. S5. The nonreciprocity ratio of the absorption amplitude \(A\) (Eq. (S45)) when the SAW is assumed isotropic, i.e. \(c = d = 0\). We set \(a = 1, b = \pm 0.35\). The non-reciprocity reaches 100% at an angle very close to \(\phi = 0\).

In the experiment, we rotated the magnetic field angle from \(\phi = 172^\circ\) to \(\phi = 188^\circ\), tracking the variation in the nonreciprocity (Fig. S6). From the spectra, we confirmed the rapid change of nonreciprocity amplitude and sign. Also, interestingly, when \(\phi = 184^\circ\), we observed a total flat line for SAW(-k), i.e. a vanishing \(A_-\), while maintaining SAW(+k) with a robust peak, namely 100% nonreciprocity ratio in accordance with the theory.
Fig. S6. (A–I) Absorption spectra at $\phi = 172^\circ, 174^\circ, 176^\circ, 178^\circ, 180^\circ, 182^\circ, 184^\circ, 186^\circ, 188^\circ$, respectively.

Section S5. Characterization of Dzyaloshinskii-Moriya interaction via Brillouin light scattering spectroscopy

Dzyaloshinskii-Moriya interaction (DMI) is the antisymmetric exchange coupling, which favours the canting alignment of the neighboring magnetic spins $S_i$ and $S_j$. In recent years, due to its intriguing application in stabilizing magnetic skyrmions and chiral domain walls, DMI has attracted intensive research. In the magnetic heterostructure, DMI appears as a consequence of the broken structural inversion symmetry in the magnet. Among the experimental methods for investigating DMI, Brillouin light scattering (BLS) spectroscopy has been most widely used due to its high sensitivity.

In the presence of the DMI, because of the different canting arrangement, spin waves with wavenumbers $\pm k$ give opposite contributions to the total energy, which results in an asymmetric spin wave dispersion relation. And this asymmetry in $\pm k$ leads to Eq. (S45) (10, 14, 24, 25) for estimating DMI constant $D$:

$$\Delta \omega = \frac{\omega(-k) - \omega(k)}{2\pi} = \frac{2D|\gamma|}{\pi M_s}$$

(S46)

$$k = \text{sgn}(M_x) \frac{4\pi \sin \Theta}{\lambda_{\text{laser}}}$$

(S47)

where we take gyromagnetic ratio $|\gamma| = 29.4$ GHz/T (26), saturation magnetization $\mu_0 M_s = 1.5$ T, wavelength of the laser $\lambda_{\text{laser}} = 473$ nm and $\text{sgn}(M_x)$ is the polarity of the $x$ component of static magnetization, $\Theta$ the angle between incident light and sample plane.
Fig. S7. **Characterization of Dzyaloshinskii-Moriya interaction via Brillouin light scattering spectroscopy.** (A) Schematics of Brillouin light scattering geometry, with scattering plane (in blue), and Cartesian coordinates. (B) Brillouin spectra of the Ta/CoFeB(1.6nm)/MgO film measured at incident angle $\Theta = 65^\circ$. Red and blue dots represent spectra measured under applied field $\mu_0 H = 50$ mT along respective $+x$ and $-x$ directions. Solid lines represent Lorentzian fitting of spectra. $k_M$ is the magnitude of wavenumber $k$. Stokes and anti-Stokes peaks were normalized to a peak amplitude of 1, respectively.

Fig. S8. Frequency difference $\Delta \omega$ of $\pm k$ spin waves as a function of wavevector $k$. Purple circles and solid line denote measured data and fitting by Eq. (S46)

In order to estimate $D$, we performed BLS measurement on the Ta/CoFeB(1.6nm)/MgO thin film in Damon-Eshbach geometry (as depicted in Fig. S7A). Figure Fig. S7B shows measurement of BLS spectra at $\Theta = 65^\circ$ while applying magnetic field $\mu_0 H$ of $\pm 50$mT. Owing to in-plane momentum conservation of the light scattering process, spin waves travelling with the wavenumber $\pm k$ appear as anti-Stokes and Stokes peaks, respectively. The difference of spectra center frequency $\Delta \omega$ in $\pm k$ are plotted in Fig. S8. By fitting with Eq. (S46), we obtain DMI constant $D = 0.063 \pm 0.0023$ mJ/m$^2$, which is in a good agreement with the estimation from acoustic ferromagnetic resonance $D_{a-FMR} = 0.089 \pm 0.011$ mJ/m$^2$. 
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