Tunable magnon-magnon coupling in synthetic antiferromagnets

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In this work, we study magnon-magnon coupling in synthetic antiferromagnets (SyAFs) using microwave spectroscopy at room temperature. Two distinct spin-wave modes are clearly observed and are hybridised at degeneracy points. We provide a phenomenological model that captures the coupling phenomena and experimentally demonstrate that the coupling strength is controlled by the out-of-plane tilt angle as well as the interlayer exchange field. We numerically show that a spin-current mediated damping in SyAFs plays a role in influencing the coupling strength.

Generating new spin-wave states can be an enabling role for developing future spintronic/magnonic devices. While individual spin-wave modes can be tailored by changing material parameters of host magnets, a novel approach of creating new spin-wave states is to couple two modes coherently by tuning them into resonance, where physical parameters of the coupled modes can also be modified. Although the coupling phenomena could be phenomenologically explained by a classical coupled-oscillator picture in general, microscopic descriptions of this type of hybridisation are rich, offering novel functionalities of state control and energy/information transfer. For example, strong coupling of light-matter interaction is envisaged to offer fast and protected quantum information processing. Within this expanding research domain, strong coupling between microwave photons and collective spins in magnetically-ordered systems has been extensively studied in recent years.

Magnon-magnon coupling has an advantage over the light-matter interaction, in terms of coupling strength. The coupling strength of light-matter interactions is sometimes significantly reduced by a lack of spatial mode overlapping of the two, and so scientists have made considerable efforts to achieve large coupling strength by designing optimum geometries for efficient mode-volume overlapping. On the other hand, magnon-magnon interaction does not suffer from this since two modes normally reside within the same host media, providing mode overlapping of 100% or close to. While magnon-magnon coupling has been studied in single magnets and magnetic bi-layers, magnon-magnon interaction in highly tunable material systems could offer unexplored parameter spaces on which to tailor the coupling phenomena. Here, we focus on synthetic antiferromagnets (SyAFs) as a host of magnon-magnon coupling and report clear hybridisation of two distinct SyAF modes arising from interlayer exchange coupling between two magnetic layers. We provide a full phenomenological model for the mode coupling, magnetic relaxation and coupling strength as a function of different material parameters for SyAF modes. Aided by these derived relationships, we demonstrate that the interlayer exchange field strength, which can be controlled by sample growth, allows the engineering of the coupling strength. We further numerically show that the spin-current mediated damping plays a role in influencing the coupling strength. Our demonstration and full details of the magnon-magnon coupling phenomena in SyAFs will act as a springboard for further research along this avenue.

Low-energy spin-wave modes in synthetic antiferromagnets in their canted regime are acoustic and optical modes where two coupled moments precess in-phase (acoustic) and out-of-phase (optical) as shown in Fig. 1(a). The acoustic (optical) mode is excited by perpendicular (parallel) configuration between microwave and applied magnetic fields. There are a number of reports in which these two modes in different SyAFs have been studied in great detail. For example, mutual spin pumping within the coupled modes has been proposed and experimentally demonstrated.

Since the resonant frequency of two modes shows different magnetic field dependence (as discussed more later), we can find the degeneracy point of the two modes by tuning experimental conditions. When the two moments are canted within the plane, the motion of the optical and acoustic modes can be decoupled, meaning that the two modes are not allowed to hybridise. This restriction can be lifted when we tilt the moments towards the out-of-plane direction and we will be able to hybridise them (see Fig. 1(b) for schematic understanding). The strength of hybridisation is defined by which represents a rate of energy transfer between the two modes. When this rate is fast, compared to mode dissipation rates of individual modes, we expect well-defined coupled modes before the excited states are relaxed. Control of the coupling strength in-situ and ex-situ will be potentially useful to a scheme of reconfigurable energy and information transfer using coherent coupling.

The SyAF stacked films used in this study were prepared by magnetron co-sputtering at a base pressure of...
Two moments \( m_1 \) and \( m_2 \) are coupled antiferromagnetically and canted at equilibrium. Under microwave irradiation, they precess in-phase (acoustic mode) and out-of-phase (optical mode) at different angular frequencies \( \omega_{ac} \) and \( \omega_{op} \), respectively. We define \( \theta_0 \) as in the figure, where the \( z \) axis is the film growth direction. (b) Schematics of the magnon-magnon coupling phenomena with the optical and acoustic modes. When the exchange field \( B_{ex} \) is small or two moments are within the film plane, the coupling strength \( g \) is zero, so the two modes do not couple. We can validate the coupling strength by tuning \( B_{ex} \) and \( \theta_0 \) and achieve strong magnon-magnon hybridisation, as shown on the right panel. (c) Microwave absorption spectrum for \( \theta_0 = 90^\circ \), measured at 13.4 GHz. Two magnetic field directions \( B_\parallel \) and \( B_\perp \) are defined as per the inset. Microwave transmission spectrum as a function of frequency and applied field for two configurations of applied magnetic fields \( B_\parallel \) and \( B_\perp \) for \( \theta_0 = 90^\circ \). Best fit curves using Eq. (1) and (2) represent as dashed curves in blue and in red, respectively.

Here, \( B_{ex}, B_\parallel, B_\perp \) and \( \gamma \) are the exchange field, the demagnetisation magnetization, the resonance field and the gyromagnetic ratio, respectively. We found that our best fits produce \( B_{ex}, B_\parallel \) and \( \gamma/2\pi \) to be 0.14 T, 1.5 T and 29 GHz/T respectively. Fits using Eqs. (1) and (2) can reproduce our experimental results very well, strongly supporting that we can experimentally observe and study the coupled SyAF modes. Since the frequency of the two modes show different magnetic field dependences, it is possible to study mode coupling of the two by tuning the mode frequencies. In Fig. 1(d), we observe a clear crossing of the two modes at \( B_0 \approx 0.2 \) T. This "crossing" means that the two modes are not able to hybridise due to mode symmetry. We can break this symmetry by tilting the moment towards the out-of-plane direction. We therefore repeated similar experiments for \( \theta_0 \neq 90^\circ \) as shown in Fig. 2(a-c). The two modes start to show an avoided crossing as \( \theta_0 \) is decreased, indicating mode

\[
\omega_{ac} = \gamma B_0 \sqrt{1 + \frac{B_{ex}}{2B_{ex}}},
\]

\[
\omega_{op} = \gamma \sqrt{2B_{ex}B_\parallel \left(1 - \frac{B_0}{2B_{ex}}\right)^2}.
\]
FIG. 2. (a-c) Microwave transmission as a function of frequency and applied field for different \( \theta_B \). The avoided crossing starts to appear and the frequency gap increases as \( \theta_B \) is decreased. (d)-(f) Simulation results for the same experimental condition as (a)-(c), respectively. (g) The coupling strength \( g/2\pi \) as a function of \( \theta_B \). We plot results from two samples with the Ru thickness of 0.5 nm and 0.6 nm. The 0.5 nm sample shows sizable \( g/2\pi \) compared to much smaller \( g/2\pi \) for 0.6 nm. The red curves are produced by Eq. (3) in the main text.

This correctly captures our experimental observation as \( g/2\pi \) grows with decreasing \( \theta_B \). The red curve in Fig. 2(g) is calculated by this equation and there is quantitative agreement between experiments and theory, despite marginal deviation at small \( \theta_B \). To further attest the validity of this equation for our experiments, we performed similar measurements on a SyAF sample having the Ru thickness of 0.6 nm since Eq. (3) suggests that the coupling strength can be tuned by \( B_{ex} \). For this sample, we found that \( B_{ex} \) is decreased to 30 mT due to a weaker interlayer coupling and accordingly, as expected, we observed a significant decrease of \( g/2\pi \) as summarised in Fig. 2(g). These results show the tunability of the mode coupling strength in SyAFs by both thin-film growth engineering (ex-situ) as well as out-of-plane tilt angle (in-situ).

Next we focus on the relaxation of the SyAF modes. Figure 3(a-b) represent plots of the half width at half maximum (HWHM) linewidth (\( \Delta B \)) extracted for individual sweeps for both modes. \( \Delta B \) of the acoustic mode increases with increasing magnetic field, with a characteristic anomaly around the field where the two modes hybridise. \( \Delta B \) of the optical mode however shows a different magnetic field dependence as it decreases with increasing magnetic field. This is primarily due to the relationship of the magnetic-field-domain linewidth and

hybridisation which can be quantitatively discussed by using the coupling strength \( g/2\pi \), the half of the minimum frequency gap. We plot the \( \theta_B \) dependence of \( g/2\pi \) in Fig. 2(g) where \( g/2\pi \) grows with the out-of-plane component, with the highest value excessing 1 GHz.

We describe the magnon-magnon coupling phenomena in SyAFs by a 2×2 matrix eigenvalue problem derived from the coupled LLG equations with mutual spin pumping terms (see Supplemental Material for detailed derivation):

\[
\begin{bmatrix}
\omega^2 - \omega_{op}^2 + i(\nu_{o1} + \nu_{o2})\omega & (i\omega - \nu_{o1}gB_2)\eta m_{z0} \\
(-i\omega + \nu_{a2}gB_2)\eta m_{z0} & \omega^2 - \omega_{ac}^2 + i(\nu_{a1} + \nu_{a2})\omega
\end{bmatrix}
\]

Here, \( \eta \) = \( 2B_{ex}/B_2 \), \( m_{z0} \) = \( B_0\cos\theta_B/(B_2 + 2B_{ex}) \), \( \nu_{o1} \) = \( (\alpha_0 + \alpha_{sp})(1 - m_{z0}^2) - \alpha_{sp}(1 - m_{z0}^2 - (B_0^2\sin^2\theta_B/4B_{ex}^2))(m_{z0}^2/m^2) \), \( \nu_{o2} \) = \( \alpha_0(1 - B_0^2\sin^2\theta_B/4B_{ex}^2) \), \( \nu_{a1} \) = \( \alpha_0(1 - m_{z0}^2 - B_0^2\sin^2\theta_B/4B_{ex}^2) \) and \( \nu_{a2} \) = \( \alpha_0(\eta + 1)(1 - m_{z0}^2) \), respectively, with \( \alpha_0 \) and \( \alpha_{sp} \) being the standard Gilbert damping constant and one arising from mutual spin pumping between the two magnetic layers. The real part of the eigenvalues gives the resonance frequencies and the imaginary part represents the loss rates of the two modes. We numerically solved the eigenvalue problem with parameters described above and found that the coupled equations can model our experimental observation well for each experimental set, such as Figs. 2(d)-(f) reproducing corresponding experimental results. We simplified the 2×2 matrix by neglecting the damping terms to calculate the eigenvalues and found an analytical expression for the coupling strength as (see derivation in Supplemental Material):

\[
g = \frac{\gamma B_{ex}B_2}{2B_2 + 4B_{ex}}\cos\theta_B.
\]
frequency-domain linewidth as given by:

$$\Delta B_{\text{op(ac)}} = \left| \frac{d\omega_{\text{op(ac)}}}{dB} \right|^{-1} \frac{1}{\tau_{\text{op(ac)}}}.$$  \hspace{1cm} (4)

When the resonance field is low, $|d\omega_{\text{op}}/dB|$ becomes small, which can extrinsically enhance the observed $\Delta B$ in our experiments. In order to extract material-specific parameters such as $\alpha_0$ from our data, we solved the eigenvalue problem and compared the imaginary part with experimental results. We found that the linewidth calculated from the imaginary part models excellently for experimental results. We found that the linewidth calculated by our numerical simulations from the eigenvalue problem as shown in Figs. 3(a-b). This linewidth averaging is similar to ones discussed in spin-photon coupling systems. We went on to quantify the loss rates for both modes by using Eq. 4. First of all, we estimated the loss rate of individual modes at the avoided crossing point (open circles in Fig. 3 (d)), by extrapolating from the values outside the coupling regime. Both show a very weak angular dependence, which can be understood that the damping (Fig.3 (c)) has no angular dependence with a subtle change of the mode-crossing frequency when $\theta_B$ is decreased. By contrast, loss rates for the hybridised modes (solid circles in Fig. 3 (d)), estimated by our eigenvalue problem, exhibit clear attraction as the coupling strength is increased by changing $\theta_B$. After $\theta_B = 60^\circ$, the loss rates of the two modes coalesce into a single number which is exactly the average of the two rates $1/\tau_{\text{mix}}=(1/2)(1/\tau_{\text{ac}}+1/\tau_{\text{op}})$ where $1/\tau_{\text{mix}}$ is the loss rate of the hybridised states. Furthermore, through the course of our simulation study, we found that $\alpha_{sp}$ can have an effect on $\alpha$, suggesting that the magnon-magnon coupling is partially mediated by spin currents. We observe that for large $\alpha_{sp}$ the coupling between the two modes can be completely suppressed (see Supplemental Material). We highlight that this damping-mediated coupling control cannot be achieved by simply changing $\alpha_0$ in our system, something specific for the magnetic relaxation via spin pumping to the coupling and the energy exchange. Although it is not possible to control $\alpha_{sp}$ in our experiments, it could act as an extra parameter to define the magnon-magnon coupling strength in SyAFs. Finally, we highlight that the highest $g/2\pi$ achieved (1.0 GHz) outnumbers the loss rates of the individual modes, indicating that this magnon-magnon coupling starts to
enter the strong coupling regime in our experiments. Although our experiments are just at the onset of the strong coupling regime, here we briefly discuss potential improvements and control of the coupling strength against the individual loss rates. Equation (3) can be simplified as $g/2\pi \propto B_{ex}/B_s$, suggesting that a sample with a higher $B_{ex}$ as well as a smaller $B_s$ shows a large coupling strength. Achieving similar coupling with low-damping materials could be another plausible path.

In summary, we experimentally show the magnon-magnon coupling in SyAF CoFeB/Ru/CoFeB multilayers. Clear magnon-magnon hybridisation has been observed when the optical and acoustic modes are tuned into resonance. The magnon-magnon coupling strength has been controlled by bringing the moments into the out-of-plane direction, which breaks the orthogonality of the two modes. In addition, the interlayer exchange coupling is found to tune the coupling strength. The loss rate of two modes exhibits an averaging effect upon hybridisation. Our eigenvalue problem approach serves to provide the analytical expression of the coupling strength as well as numerical explanations/predictions of the experimental data. We envisage that in the present study will be transferable to other weakly-coupled antiferromagnetic systems since the phenomenological descriptions of their spin-wave modes should be identical to our model developed.

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I. SAMPLE CHARACTERIZATION BY VIBRATING SAMPLE MAGNETOMETER

In this study, we use the following free energy expression which includes of linear and quadratic exchange coupling contributions\(^\text{31-34}\) to describe static magnetisation direction in a synthetic antiferromagnet (SyAF):

\[
F = \sum_{j=1}^{2} \left[ M_s B \cdot m_j + \frac{1}{2} M_s B_s (m_j \cdot z)^2 \right] + \frac{2 J_{\text{ex1}}}{d} m_1 \cdot m_2 + \frac{2 J_{\text{ex2}}}{d} (m_1 \cdot m_2)^2. \tag{S1}
\]

Here, \(M_s\), \(B\), \(m_1(2)\), \(B_s\), \(J_{\text{ex1(2)}}\) are the saturation magnetisation, external magnetic field vector, the unit vector of individual moments in a SyAF, demagnetisation field, the linear and quadratic antiferromagnetic interlayer exchange coupling constants, respectively; \(d\) is the thickness of two ferromagnetic layers which are identical in the present case. Figure S1 shows magnetometry characterization of two samples used in the present study. The red lines in the figure are calculated by using \(M(B) = M_s \cos \phi(B)\)\(^\text{2,3}\) where \(\phi(B)\) is the angle between the applied magnetic field direction and equilibrium direction of individual moments which is obtained for all field values by minimizing Eq. S1 reiteratively until we achieve good matching to experimental data. The red curves in Fig. S1 were generated by the linear and quadratic exchange fields of 140 (30) ± 1.2 (0.6) mT and 7 (2) ± 0.1 (0.03) mT for the 0.5 (0.6) nm Ru thickness sample, together with \(M_s = 1400 (1300)\) kA/m for the 0.5 (0.6) nm Ru sample. The effective magnetic field acting on both moments can be given by differentiating the exchange coupling terms \(F_{\text{ex}}\) in Eq. S1 with respect to \(m_1(2)\):

\[
B_{\text{ex,1(2)}} = \frac{1}{2M_s} \frac{\partial F_{\text{ex}}}{\partial m_{1(2)}} = -\frac{J_{\text{ex1}}}{d} m_{2(1)} - \frac{2 J_{\text{ex2}}}{d} (m_1 \cdot m_2) m_{2(1)}, \tag{S2}
\]

\(m_1 \cdot m_2\) is a scalar value defined by the relative angle between \(m_1\) and \(m_2\). Note that we incorporate this second-order exchange coupling term within \(B_{\text{ex}}\) for our analysis in our study.

![Graph](image-url)

FIG. S1. (a-b) Magnetization curve of the CoFeB (3 nm)/Ru (t nm)/CoFeB (3 nm) measured by vibrating sample magnetometer for (a) \(t = 0.5\) and (b) \(t = 0.6\). The black (red) curve is the experimental (calculation) results.
II. ADDITIONAL MAGNETISATION-DYNAMICS RESULTS IN THIS STUDY

This section provides supplementary results used in our study to support our claims in the main text. In Fig. S2, we show 2D plots of frequency vs magnetic field for different $\theta_B$ from the sample with the Ru thickness of 0.5 nm. This supplements Fig. 2 in the main text and further supports our observation of crossing/avoided-crossing feature.

FIG. S2. (a-d) Extra data plots of Microwave transmission as a function of frequency and applied field, for the sample with the Ru thickness of 0.5 nm for different $\theta_B$. Large coupling gap can be seen at low angles. Figures (e-h) plot simulation results for the same experimental conditions as Fig. (a-d).

FIG. S3. (a-d) Extra data plots of linewidth of the two modes as a function of magnetic field for different $\theta_B$ for the sample with the Ru thickness of 0.5 nm. Solid lines represent simulation results from the theoretical model we used in this study.
controlled by the out-of-plane angle $\theta_B$ in the main text. Furthermore, Fig. S3 represents the magnetic-field-domain linewidth ($\Delta B$) as a function of frequency measured for different $\theta_B$. Theory curves plotted were produced by the imaginary part of eigenvalues discussed in the main text and Section 3 in this document. Damping parameters and loss rates plotted in Fig. 3(d) in the main text have been extracted from the parameters in the eigenvalue problem.

We repeated similar measurements for the sample with the Ru thickness of 0.6 nm. The same analysis procedure and plots have been carried out for experimental data and shown in Figs. S4 and S5. In Fig. S4, we notice that there exist magnetic-field independent background signals around 5 GHz which we consider as transmission losses unrelated to magnetisation dynamics. Nevertheless, we here highlight that the gap opening is much weaker than those measured for the sample with the Ru thickness of 0.5 nm. This is attributed to the size of exchange coupling, which has been independently quantified by VSM as explained above.

![Graph showing microwave transmission as a function of frequency and applied field for different $\theta_B$.](image)

**FIG. S4.** (a-d) Microwave transmission as a function of frequency and applied field, for the sample with the Ru thickness of 0.6 nm for different $\theta_B$. Small gap opening corresponds to the weak exchange coupling of the sample. Figures (e-h) plot simulation results for the same experimental conditions as Fig. (a-d).

![Graph showing linewidth as a function of magnetic field for different $\theta_B$.](image)

**FIG. S5.** (a-b) Linewidth of the two modes as a function of magnetic field for $\theta_B$ of (a) 90° and (b) 25° for the sample with the Ru thickness of 0.6 nm. We show our simulation results as solid lines. (c) Extracted values of damping parameters. (d) Calculated loss rates of each mode at the crossing point as well as those of the hybridised modes.
III. THE EIGENVALUE PROBLEM AND ANALYTICAL EXPRESSIONS

In this section, the two-coupled Landau-Lifshitz-Gilbert (LLG) equations at the macrospin limit are employed to model magnetic dynamics of optical and acoustic modes in SyAFs. We recognize that similar approaches have been taken by others previously\cite{S3,S5} but not specifically for magnon-magnon coupling phenomena in SyAFs as we detail below. We consider a canted regime of two individual moments \( \mathbf{m}_1 \) and \( \mathbf{m}_2 \) which are coupled antiferromagnetically by the exchange interaction with the strength of \( B_{\text{ex}} \). These two moments reside in thin-film magnets subjected to a demagnetisation field \( B_d \) and we apply an external magnetic field \( \mathbf{B} \) within the \( x-z \) plane with angle \( \theta_B \) from the \( z \) axis which is the sample growth direction in our case. Following convention, we first define Kittel and Neel vectors as \( \mathbf{B} = \langle m_1 + m_2 \rangle / 2 \) and \( \mathbf{n} = \langle m_1 - m_2 \rangle / 2 \), respectively. Dynamics of these two coupled moments are given by\cite{S6},

\[
\frac{dm}{dt} = -\Omega_L m \times \mathbf{u} + \Omega_B \left[ (m \cdot z) m \times z + (n \cdot z) n \times z \right] + \tau_m, \tag{S3}
\]

\[
\frac{dn}{dt} = -\Omega_L n \times \mathbf{u} + \Omega_B \left[ (m \cdot z) n \times z + (n \cdot z) m \times z \right] + 2\Omega_{\text{ex}} n \times m + \tau_n. \tag{S4}
\]

Here, \( \Omega_L = \gamma B_0 \), \( \Omega_B = \gamma B_s \), and \( \Omega_{\text{ex}} = \gamma B_{\text{ex}} \) where \( \gamma \) and \( B_0 \) are the gyromagnetic ratio and the resonance field, respectively; \( \mathbf{u} = \sin \theta_B \mathbf{x} + \cos \theta_B \mathbf{z} \) represents the applied field direction in the \( x-z \) plane, given that \( \mathbf{x} \) and \( \mathbf{z} \) are unit vectors for the corresponding axes. The last terms in Eqs. (S4) and (S5) account for damping torques which are expressed as:

\[
\tau_m = \alpha_0 \left( m \times \frac{dm}{dt} + n \times \frac{dn}{dt} \right), \tag{S5}
\]

\[
\tau_n = (\alpha_0 + \alpha_{sp}) \left( m \times \frac{dm}{dt} + n \times \frac{dn}{dt} \right) - \alpha_{sp} \left[ m \cdot \left( n \times \frac{dn}{dt} \right) \right] n + n \cdot \left( m \times \frac{dn}{dt} \right) n \tag{S6}
\]

where \( \alpha_0 \) and \( \alpha_{sp} \) are the standard Gilbert damping constant and one arising from mutual spin pumping between the two magnetic layers. With approximation of small angle precession, we can separate the equilibrium \( \langle m_0 \rangle \) and time-dependent \( \langle \delta m(t) \rangle \) and \( \langle \delta n(t) \rangle \) terms as\( \langle m(t) \rangle = m_0 + \delta m(t) \) and \( \langle n(t) \rangle = n_0 + \delta n(t) \) and here we define each vector component in a standard manner, e.g. \( m_0 = (m_{0x}, m_{0y}, m_{0z}) \). Substituting \( \langle m(t) \rangle \) and \( \langle n(t) \rangle \) into Eqs. (S4)-(S7) and keeping the first order of the time-dependent and damping terms, we obtain the following six coupled equations:

\[
\frac{1}{\Omega_L} \frac{dm_0}{dt} = n_0 \eta \delta n_z - \alpha_0 \eta \left( m_{0z}^2 + n_{0y}^2 \right) \delta m_x - \eta m_{0x} \delta m_y + \alpha_0 (\eta + 1) m_{0x} m_{0z} \delta m_z, \tag{S7}
\]

\[
\frac{1}{\Omega_L} \frac{d\delta m_x}{dt} = \alpha_0 m_{0x} m_{0y} \delta n_z + \eta m_{0x} \delta m_x - \alpha_0 \eta m_{0x} ^2 \delta m_y - (\eta + 1) m_{0x} \delta m_z, \tag{S8}
\]

\[
\frac{1}{\Omega_L} \frac{d\delta m_y}{dt} = \alpha_0 \eta m_{0x} m_{0z} \delta m_x + \eta m_{0z} \delta m_y - \alpha_0 (\eta + 1) \left( m_{0x}^2 + n_{0y}^2 \right) \delta m_z, \tag{S9}
\]

\[
\frac{1}{\Omega_L} \frac{d\delta m_z}{dt} = \alpha_0 \alpha_{sp} m_{0x} m_{0z} \delta n_x + \alpha_0 \eta m_{0y} m_{0z} \delta m_y + (\eta + 1) m_{0y} \delta m_z + \alpha_{sp} \frac{m_{0x} m_{0x} m_{0z} n_{0y}^2}{m_0^2} \delta n_z, \tag{S10}
\]

\[
\frac{1}{\Omega_B} \frac{d\delta n_x}{dt} = -m_{0x} \delta n_z + \alpha_0 \eta m_{0x} m_{0y} \delta m_x + \alpha_0 (\eta + 1) m_{0x} m_{0y} \delta m_z, \tag{S11}
\]

\[
\frac{1}{\Omega_B} \frac{d\delta n_y}{dt} = -\left( \alpha_0 \alpha_{sp} \right) \left( m_{0x}^2 + n_{0y}^2 \right) \delta n_z - \eta m_{0y} \delta m_x + \alpha_0 \eta m_{0x} m_{0y} \delta m_y + \alpha_{sp} \frac{m_{0x}^2 n_{0y}^2}{m_0^2} \delta n_z, \tag{S12}
\]

where the equilibrium conditions and a new parameter \( \eta \) are introduced: \( \langle m_0 \rangle = (m_{0x}, m_{0y}, m_{0z}) \), \( \langle n_0 \rangle = (0, n_{0y}, 0) \) and \( \eta = 2\Omega_{yz} / \Omega_L \). Further simplification has been made by using additional parameter definitions as well as time derivative treatment:

\[
\nu_{01} = (\alpha_0 + \alpha_{sp}) \left( m_{0x}^2 + n_{0y}^2 \right) - \alpha_{sp} \frac{m_{0x}^2 n_{0y}^2}{m_0^2}, \tag{S13}
\]

\[
\nu_{02} = \alpha_0 \eta \left( m_{0z}^2 + n_{0y}^2 \right), \tag{S14}
\]
\[ \nu_{a1} = \alpha_0 \eta m_0^2, \quad (S15) \]

\[ \nu_{a2} = \alpha_0 (\eta + 1) (m_0^2 + n_0^2), \quad (S16) \]

\[ \frac{1}{\nu_{a1}} \frac{d}{d \nu_{a1}} \rightarrow -i \nu_{a1}^{-1} \equiv -i \nu. \quad (S17) \]

These produce the following equations of motion for \( \delta m_x, \delta m_y, \delta m_z \) and \( \delta n_z \):

\[ -i \Omega \delta m_x = n_0 y \delta n_z - \nu_{a2} \delta m_x - \eta m_0 \delta m_y + \alpha_0 (\eta + 1) m_0 \delta m_z, \quad (S18) \]

\[ -i \Omega \delta m_y = \alpha_0 m_0 n_0 y \delta n_z + \eta m_0 \delta m_x - \nu_{a1} \delta m_y - (\eta + 1) m_0 \delta m_z, \quad (S19) \]

\[ -i \Omega \delta m_z = \alpha_0 m_0 n_0 \delta m_x + \eta m_0 \delta m_y - \nu_{a2} \delta m_z, \quad (S20) \]

\[ -i \Omega \delta n_z = -\nu_{a1} \delta n_z - \eta m_0 \delta m_x + \alpha_0 m_0 n_0 \delta m_y. \quad (S21) \]

We write these as a matrix form as follows:

\[ \Omega \begin{pmatrix} \delta n_z \\ \delta m_x \\ \delta m_y \\ \delta m_z \end{pmatrix} = i \begin{pmatrix} -\nu_{o1} & -\eta n_0 y & \alpha_0 m_0 n_0 & 0 \\ n_0 y & -\nu_{o2} & -\eta m_0 & \alpha_0 (\eta + 1) m_0 m_0 & \end{pmatrix} \begin{pmatrix} \delta n_z \\ \delta m_x \\ \delta m_y \\ \delta m_z \end{pmatrix} \quad (S22) \]

We can obtain the eigen mode frequency \( \omega_{op/ac} \) and relaxation time \( \tau_{op/ac} \) by solving the complex eigenvalue problem, which can be expressed as,

\[ \Omega_{op(ac)} = \frac{1}{i \nu} (\omega_{op(ac)} - i \tau_{op(ac)}^{-1}). \quad (S23) \]

If we neglected damping terms in the off-diagonal components, we obtained the following relations:

\[ \delta n_z \sim -\frac{\eta n_0 y}{-\nu_{o1} + \nu_{o1}} \delta m_x \quad (S24) \]

\[ \delta m_z \sim \frac{\eta m_0}{-\nu_{o2} + \nu_{o2}} \delta m_y. \quad (S25) \]

Using these relations, we reduced Eq. (S23) into an eigenvalue problem with a 2-by-2 matrix form given by:

\[ \begin{pmatrix} \Omega^2 - \Omega_{op}^2 + i(\nu_{o1} + \nu_{o2})\Omega & - (\nu_{o1} + \nu_{o2}) \eta m_0 \\ -i(\nu_{o1} + \nu_{o2}) \eta m_0 & \Omega^2 - \Omega_{ac}^2 + i(\nu_{o1} + \nu_{o2})\Omega \end{pmatrix} \begin{pmatrix} \delta m_x \\ \delta m_y \end{pmatrix} = 0 \quad (S26) \]

During this process, we disregarded higher-order terms in the diagonal elements, such as \( \nu_{o1} \nu_{o2} \). This is the matrix we show in the main text after converting \( \Omega \) into \( \omega \) using Eq. S25. Here, \( \Omega_{op} \) and \( \Omega_{ac} \) are the eigen frequencies for optical mode and acoustic modes given by:

\[ \Omega_{op} = \frac{n_0 y \sqrt{\eta}}{2} - i \frac{1}{2} (\nu_{o1} + \nu_{o2}) \quad (S27) \]

\[ \Omega_{ac} = \sqrt{\eta^2 + \eta m_0} - i \frac{1}{2} (\nu_{a1} + \nu_{a2}) \quad (S28) \]

Equations (1) and (2) in the main text can be obtained by the real part of these two equations by using the equilibrium conditions: \( (m_0, n_0, m_0, n_0) = (B_0 \sin \theta_B/2B_{ex}, 0, B_0 \cos \theta_B/2(B_0 + 2B_{ex}) \) and \( (n_0, m_0, n_0, m_0) = (0, \sqrt{1 - m_0^2} - m_0^2, 0, 0) \) with \( \theta_B = \pi/2 \). We note here that our numerical solutions of (S23) and (S27) are almost identical and therefore we decided to show the simpler 2x2 matrix in the main text. In order to provide the coupling constant \( g \) in the main text, we further take out the damping terms and solved the following eigen problem,

\[ \begin{pmatrix} \Omega^2 - \Omega_{op}^2 & i\Omega m_0 \\ -i\Omega m_0 & \Omega^2 - \Omega_{ac}^2 \end{pmatrix} \begin{pmatrix} \delta m_x \\ \delta m_y \end{pmatrix} = 0 \quad (S29) \]
By defining the crossing (dimensionless) frequency as $\Omega_{op} = \Omega_{ac} = \Omega_0$, we can find the energy gap ($\Delta \Omega_{\text{gap}}$) using Eq. (S32).

$$\Delta \Omega_{\text{gap}} = 2(\Omega - \Omega_0) = \pm \frac{2\eta_m \Omega_0}{\Omega + \Omega_0} \approx \pm \eta_m \Omega_0.$$  

Therefore,

$$g = \frac{1}{2} \Delta \Omega_{\text{gap}} \gamma B_s = \frac{\gamma B_{ex} B_0}{2B_s + 4B_{ex}} \cos\theta_B$$  

(S32)

Note that this is only valid when $\Delta \Omega_{\text{gap}} \ll \Omega_0$.

**IV. IMPACT OF THE MUTUAL SPIN PUMPING DAMPING ON THE COUPLING**

Our theory model allows to explore parameter regimes beyond experimental conditions. In an effort to understand the coupling of the two magnetic resonance modes in a SyAF, we investigated qualitatively the dependence of the damping parameters on the coupling strength. Especially, we focused on the effect of the damping arising from the mutual spin pumping between the two ferromagnets. This is because of our derived matrix form in Eq. S26 in which $\alpha_{sp}$ exists in the off-diagonal term, strongly suggesting that this parameter can contribute to the coupling of optical and acoustic modes. Figure S6 shows the simulated loss rate and resonance frequency of the acoustic and optic modes as a function of the magnetic field $B$ for $\theta_B = 27^\circ$. The plots show the results of the full eigenvalue problem defined in Eq. S22 for several values of $\alpha_{sp}$. For zero $\alpha_{sp}$, the loss rate of the acoustic and optic mode cross at a lower magnetic field than the point of minimal frequency separation between the resonance frequencies of the two modes. With increasing $\alpha_{sp}$, the crossing of the loss rates shifts higher in magnetic field until it appears at the same field as the minimal frequency separation of the resonance frequencies. For the highest $\alpha_{sp}$ the loss rate no longer cross and are separated. The dispersion of the resonance frequencies remains mostly unchanged for $\alpha_{sp} = 0$ and $\alpha_{sp} = 0.0255$. We observe that for higher $\alpha_{sp}$, the coupling strength reduces and eventually goes to zero for the highest $\alpha_{sp}$ we

![FIG. S6. Simulated loss rate (left panel) and resonance frequency (right panel) of the acoustic (red) and optic (blue) modes as a function of applied magnetic field at an angle of $\theta_B = 27^\circ$ for different multiples of the mutual spin pumping damping $\alpha_{sp} = 0.0255$. These are produced by solving Eq. S22 with other parameters of $B_0 = 1.583 \, \text{T}$, $B_{ex,1} = 0.14 \, \text{T}$, $B_{ex,2} = 0.0065 \, \text{T}$, $\gamma/2\pi = 29 \, \text{GHz/T}$ and $\alpha_0 = 0.0155$.](image-url)
plot. From the change of the coupling strength with $\alpha_{sp}$, we conclude that the coupling is partially mediated by spin currents. This change in the coupling behaviour can only be achieved by changing $\alpha_{sp}$. In contrast, a change of the Gilbert damping $\alpha_0$ has no influence on the coupling strength and only affects the loss rate of the modes. Our theory model suggests that the coupling between the acoustic and optic modes of a synthetic antiferromagnet is not fully described by a classical coupled harmonic oscillator model.

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