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Engineering antiferromagnetic magnon bands through interlayer spin pumping

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Spin pumping, a central phenomenon in spintronics used to source pure spin currents, is best understood in collinear magnetic multilayers. To date, there is not yet a unified Landau Lifshitz Gilbert (LLG) theory that captures the fieldlike and dampinglike torques in a generic non-collinear magnetic multilayer. Here, we theoretically expand the LLG phenomenology to incorporate both dynamic fieldlike and dampinglike torques arising from spin pumping within noncollinear magnetic materials. We find that often overlooked dynamic fieldlike torques are capable of unveiling inversion asymmetries present in magnetic multilayers. Consequently, spin pumping can be used to lift the spectral degeneracy between various magnon modes in noncollinear antiferromagnets. We experimentally confirm this new magnon-magnon interaction in a synthetic antiferromagnetic tetralayer, which has highly non-collinear magnetization configurations when under the influence of an external field. Thus, we demonstrate how spin-pumping can facilitate a new magnon-magnon interaction, significantly expanding how magnonic interactions can be engineered into antiferromagnets and magnetic metamaterials.

Introduction. In magnetically ordered materials, the precession of magnetization can lead to “spin pumping”, i.e., the transfer of angular momentum from the magnetically ordered material into adjacent non-magnetic materials in the form of a pure spin current[1, 2]. The confirmation of spin pumping in magnetic heterostructures[3–5] marked a paradigm shift in the field of spintronics, triggering a transition from the traditional charge-based electronics towards a spin-oriented approach and has emerged as a cornerstone for developing a new generation of spintronic devices[6, 7]. In multilayer systems, spin pumping can be used to generate torques when a spin current generated by one magnetic layer is absorbed by an adjacent layer[8, 9]. Early theoretical works have identified two types of spin pumping torques in the LLG equations of motion governing coherently-driven collinear magnetizations dynamics, i.e., a dampinglike and a fieldlike torque associated with, respectively, the real and imaginary part of the spin-mixing conductance [1, 2, 8, 10]. Dampinglike torques stem from the transfer of longitudinal spin angular momentum and they have been under intense investigation as they uniquely provide a mechanism to transfer spin angular momentum in magnetic heterostructures[11, 12]. Fieldlike torques, instead, arise from the transfer of transverse spin angular momentum, and, while they do not affect the magnitude of the magnetization, they can cause a rotation of the magnetization direction around the effective field. Despite reports of significant field like torques at some interfaces [13–15], the fieldlike torque is commonly neglected as it was suggested that the imaginary part of the spin-mixing conductance must be exceedingly smaller than its real part at metallic interfaces. Here, using synthetic antiferromagnets (SAF) as a model system, we will show that spin pumping fieldlike torques can instead play a dominant role in magnon band engineering.

SAF structures are comprised of ferromagnetic layers separated by normal metal spacer layers that promote an antiferromagnetic, exchange-like coupling via the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction[16, 17]. In such materials, magnetization dynamics can pump spin currents between magnetic layers through the spacer material. While earlier works have addressed spin injection in diverse non-collinear structures [18–20], there is not yet an unified LLG theory that captures the fieldlike and dampinglike torques in a generic non-collinear magnetic multilayer. A strong motivation for filling this gap in our understanding of spin pumping is being driven by the attention that SAFs are generating as a hybrid magnonic material[21–23]. **Specifically, in materials with modest interlayer exchange interactions, such as SAFs and van der Waals magnets[24–29], optical and acoustic magnons can be accessed at GHz frequencies[30–32]. By accessing both optical and acoustic magnons at GHz frequencies, these recent studies have demonstrated how layered antiferromagnets have a unique potential for having “tunable” magnon-magnon interactions.**

Experimental Results. Using DC magnetron sputtering, we prepared both bilayer and tetralayer SAF films comprised of alternating permalloy/ruthenium (Py/Ru) layers that are grown between two platinum (Pt) layers that are 6 nm thick. The Pt layers serve two purposes; preventing surface oxidation and also, removing a residual ferromagnetic moment which can occur if the first permalloy layer is grown directly onto the substrate. As illustrated in Fig. 1 (a) and (b), bilayers and tetralayers have two and four Py layers respectively. Our samples exhibit antiferromagnetic interlayer coupling between 5 nm Py layers when the Ru layers are fixed at 1 nm. The antiferromagnetic interaction is verified with magnetometry as seen in Fig. 1 (a) and (b). Antiferromagnetic sam-

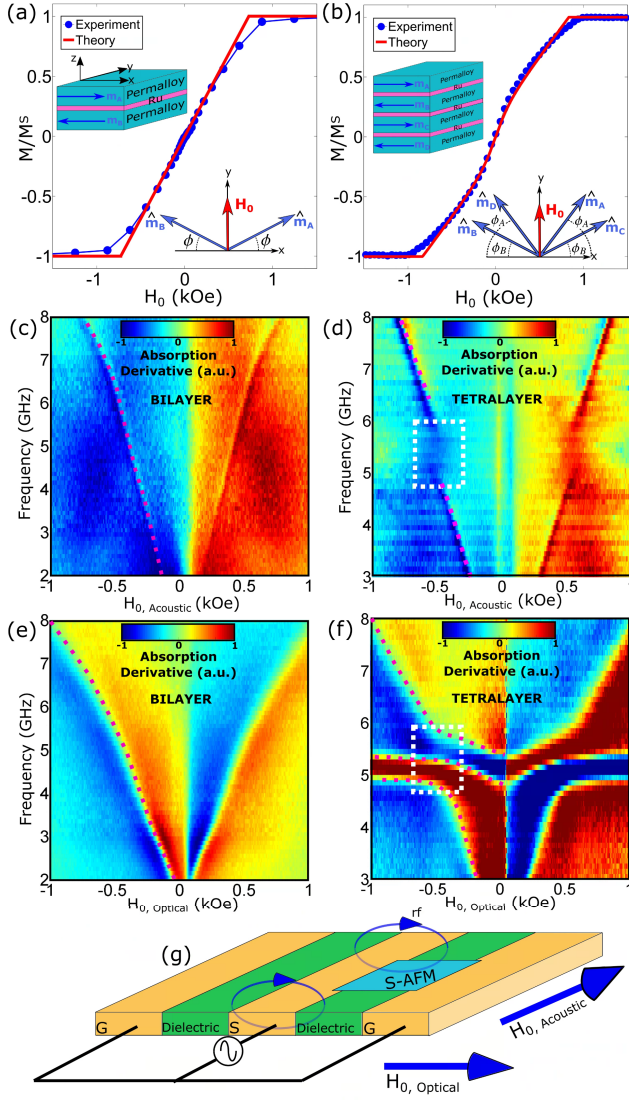


FIG. 1. (a), (b), the normalized magnetization of bilayers and tetralayers are plotted as a function of field. Insets illustrate both the structure, as well as the static equilibrium orientations of each sample. (c), (d), the acoustic magnon spectra for the bilayer and tetralayer are shown. (e), (f), the optical magnon spectra for the bilayer and tetralayer are shown. Pink dashed lines are drawn as a guide to indicate the various magnon branches. In (d) and (f), a dashed box is drawn around a region of interest where magnon branches tend to mutually avoid one another. (g) illustrates how samples are placed on the CPW, and how the external field, H_0 , should be oriented relative to the rf-field so that acoustic and optical magnons are excited.

ples have no magnetic hysteresis, and the magnetization increases with field linearly for the bilayer, and non-linearly for the tetralayer. By fitting the normalized magnetization of our samples versus magnetic field (mathematical details are found in Supplemental material, Section 2.1[33]) we are able to extract an interlayer antiferromagnetic exchange field of $\mu_0 H_E = 28.1$ mT and $\mu_0 H_E = 24.4$ mT for the bilayer and tetralayer respectively. Importantly, our magnetometry data is explained

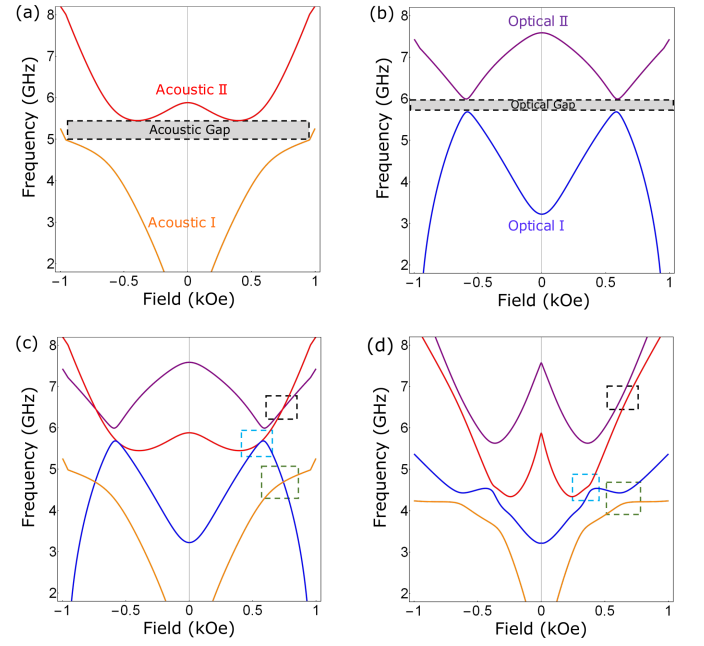


FIG. 2. (a), (b), and (c) represent the acoustic, optical, and combined magnon spectrum in the absence of spin pumping for the tetralayer. Due to the dynamic exchange field, the two acoustic magnons (a), and the two optical magnons (b), “self-hybridize” with one another. This creates the acoustic and optical gap that is indicated in (a) and (b), respectively. In (c), we observe that no interactions exist between optical and acoustic magnons, as seen in the field-frequency relationship regions highlighted by the dashed boxes. In (d) we calculate the magnon spectrum of the tetralayer for $\alpha_{f,AB} = \alpha_{f,BC} = 0.1$, $\alpha_{f,CD} = 0.15$, $\alpha_{d,AB} = \alpha_{d,BC} = 0.01$, $\alpha_{d,CD} = 0.015$ and $\alpha = 0.01$. Due to the asymmetry between the uppermost and the lowermost interface, i.e., $\alpha_{f,AB} \neq \alpha_{f,CD}$, the three energy-level crossings between acoustic and optical magnon branches are lifted, and new branches emerge as the result of the interface-driven magnon-magnon hybridization.

by a model which assumes that the interlayer exchange field between every magnetic layer is the same, i.e., there is no evidence of any inversion asymmetry found in the interlayer exchange interactions.

To experimentally characterize the magnon energy spectra of our samples we employ a broadband magnetic resonance technique where samples are flipped on top of a co-planar waveguide, see Fig. 1. Our most compelling results arise in an experimental geometry where samples are placed on the dielectric region between the signal and ground line of the CPW. Here, the CPW generates an in-plane rf-field that is perpendicular to the signal line, as well as an out-of-plane rf-field [see Fig. 1 (g)]. In Fig. 1 (c) and (d), we show the experimentally excited magnon energy spectrum in the “acoustic configuration”, where the external field is applied along the length of the signal line. In this configuration, all dynamic components of the rf-field are perpendicular to the external field, and we only excite acoustic magnons. For the bilayer, an unbroken linear magnon branch is observed, but for the tetralayer an avoided energy level crossing near 5.5 GHz breaks the main

linear branch into two branches.

In order to couple our CPW to optical magnon modes, we rotate the external field so that it is parallel to the in-plane component of the rf-field; this configuration is illustrated in (g). We call this the “optical configuration” because it is known to excite optical magnons[24–26]. However, acoustic magnons are still excited due to the out-of-plane component of the rf-field. The magnon spectrum in this configuration for the bilayer and tetralayer is shown in Fig. 1 (e) and (f). For the bilayer, we more strongly observe the low energy acoustic branch and do not couple to the optical magnon. For the tetralayer we clearly observe *three* magnon branches. The lowest energy linear magnon branch is observed to dramatically flatten out as the external field increases. A second,

relatively flat, new magnon mode with a frequency of 4.9 GHz at zero field is observed. Finally, a third magnon branch with a zero field frequency near 5.5 GHz is excited. In addition to the surprising overall “flatness”, i.e., the independence of the magnon energy gaps on the external magnetic field for the two lower energy magnon branches, all three branches are experimentally observed to avoid one another.

Theoretical Model. To explain these experimental measurements, we develop a general phenomenology for spin pumping in noncollinear magnetic layered structures incorporating both fieldlike and dampinglike spin pumping torques, consistent with Onsager reciprocity and symmetry principles. **A detailed derivation is provided in Supplemental material, Section 1[33].** We describe the magnetization dynamics of the tetralayer using four modified and coupled LLG equations:

$$\begin{aligned} \frac{d\mathbf{m}_{A(D)}}{dt} = & -\mu_0\gamma\mathbf{m}_{A(D)} \times [H_0\hat{y} - H_E\mathbf{m}_{B(C)} - M_s(\mathbf{m}_{A(D)} \cdot \hat{z})\hat{z}] \\ & + \mathbf{m}_{A(D)} \times \left[\frac{d\mathbf{m}_{B(C)}}{dt} \times (\alpha_{f,AB(CD)}\mathbf{m}_{A(C)} + \beta_{f,AB(CD)}\mathbf{m}_{B(D)}) \right] + (\alpha_{A(D)} + \alpha_{d,AB(CD)})\mathbf{m}_{A(D)} \times \frac{d\mathbf{m}_{A(D)}}{dt} \\ & - \alpha_{d,AB(CD)}\mathbf{m}_{A(D)} \times \left(\mathbf{m}_{B(C)} \times \frac{d\mathbf{m}_{B(C)}}{dt} \right) \times \mathbf{m}_{A(D)}, \end{aligned} \quad (1)$$

$$\begin{aligned} \frac{d\mathbf{m}_{B(C)}}{dt} = & -\mu_0\gamma\mathbf{m}_{B(C)} \times [H_0\hat{y} - H_E(\mathbf{m}_{A(B)} + \mathbf{m}_{C(D)}) - M_s(\mathbf{m}_{B(C)} \cdot \hat{z})\hat{z}] + \mathbf{m}_{B(C)} \times \left[\frac{d\mathbf{m}_{A(B)}}{dt} \times (\alpha_{f,AB(BC)}\mathbf{m}_{A(B)} + \beta_{f,AB(BC)}\mathbf{m}_{B(C)}) \right] \\ & + \mathbf{m}_{B(C)} \times \left[\frac{d\mathbf{m}_{C(D)}}{dt} \times (\alpha_{f,BC(CD)}\mathbf{m}_{B(C)} + \beta_{f,BC(CD)}\mathbf{m}_{C(D)}) \right] + (\alpha_{B(C)} + \alpha_{d,AB(CD)} + \alpha_{d,BC})\mathbf{m}_{B(C)} \times \frac{d\mathbf{m}_{B(C)}}{dt} \\ & - \alpha_{d,AB(BC)}\mathbf{m}_{B(C)} \times \left(\mathbf{m}_{A(B)} \times \frac{d\mathbf{m}_{A(B)}}{dt} \right) \times \mathbf{m}_{B(C)} - \alpha_{d,BC(CD)}\mathbf{m}_{B(C)} \times \left(\mathbf{m}_{C(D)} \times \frac{d\mathbf{m}_{C(D)}}{dt} \right) \times \mathbf{m}_{B(C)}, \end{aligned} \quad (2)$$

where Eq. (1) describes the dynamics of the magnetization on the top (bottom) surface layers $\mathbf{m}_{A(D)}$, while Eq. (2) represents the upper (lower) interior layers $\mathbf{m}_{B(C)}$. H_0 and H_E are the external and interlayer exchange fields, respectively. The torque term proportional to the saturation magnetization, M_s , parameterizes the shape-dependent demagnetizing field of each thin film layer. The parameter α_i accounts for the local Gilbert damping of the i th layer, which we assume to be the same within each layer, i.e., $\alpha_i \equiv \alpha$, with $i = A, B, C, D$. Terms $\propto \alpha_{d,ij}$ describe the dampinglike torques associated with spin pumping at the interface between the i th and j th layers and reduce to the well-known results of Ref. [8] for a collinear magnetization configuration, i.e., $\mathbf{m}_i \parallel \mathbf{m}_j$. The terms $\alpha_{f,ij}$ and $\beta_{f,ij}$

parameterize the strength of the fieldlike torques at the interface between the i th and j th layers [34], **which have been neglected by previous works generalizing LLG phenomenology to noncollinear magnetic heterostructures[35, 36].** In what follows, we treat each individual Py|Ru|Py bilayer as symmetric, which allows us to set $\alpha_{f,ij} = \beta_{f,ij}$ for a given (i, j) pair of layers. It is important to note that the dynamical fieldlike torques contribute to the effective field $\mathbf{H}_{i,\text{eff}}$ acting on the magnetic order parameter $\mathbf{m}_i$, and thus influence the equilibrium orientation around which the latter coherently precesses. By solving self-consistently Eqs. (1) and (2), the effective field $\mathbf{H}_{i,f}$ acting on $\mathbf{m}_i$ due to spin pumping can be written, to first order, as:

$$\mathbf{H}_{A(D),f} = \mathbf{m}_{B(C)} \times [H_0\hat{y} - H_E(\mathbf{m}_{A(B)} + \mathbf{m}_{C(D)}) - M_s(\mathbf{m}_{B(C)} \cdot \hat{z})\hat{z}] \times \alpha_{f,AB(CD)}(\mathbf{m}_{A(C)} + \mathbf{m}_{B(D)}) \quad (3)$$

$$\begin{aligned} \mathbf{H}_{B(C),f} = & \mathbf{m}_{A(D)} \times [H_0\hat{y} - H_E\mathbf{m}_{B(C)} - M_s(\mathbf{m}_{A(D)} \cdot \hat{z})\hat{z}] \times \alpha_{f,AB(CD)}(\mathbf{m}_{A(C)} + \mathbf{m}_{B(D)}), \\ & + \mathbf{m}_{C(B)} \times [H_0\hat{y} - H_E(\mathbf{m}_{B(C)} + \mathbf{m}_{D(A)}) - M_s(\mathbf{m}_{C(B)} \cdot \hat{z})\hat{z}] \times \alpha_{f,BC}(\mathbf{m}_B + \mathbf{m}_C). \end{aligned} \quad (4)$$

For finite spin pumping strength, i.e., $\alpha_{f,ij} \neq 0$, we find the

(dynamical) equilibrium angles by minimizing the free energy

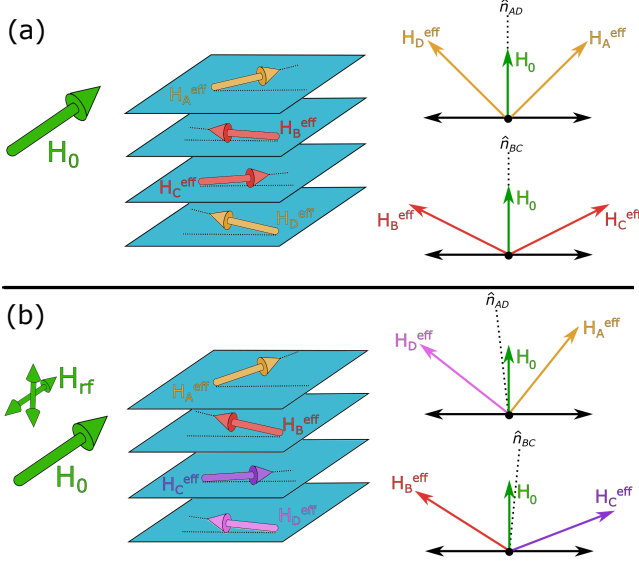


FIG. 3. (a) illustrates the conventional magnetostatic picture where, if an external field is applied along the y -direction, the effective magnetic field directions that the surface layers experience (H_A^{eff} & H_D^{eff}) are mirror symmetric about the y -axis. Similarly, the effective fields experience by the interior layers (H_B^{eff} & H_C^{eff}) are also mutually mirror symmetric about the external field direction. In (b), we consider a more realistic situation where there is some asymmetry between the metallic interface separating A & B , compared to the interface separating C & D . Here, when both an external field and driving rf-field are present, *interlayer spin pumping can break the symmetry of the system*. This broken symmetry is illustrated in the rightmost panel of (b) where the mirror symmetry axes of the surface layers and interior layers ($\hat{n}_{AD}$ & $\hat{n}_{BC}$) rotate in opposite directions away from the external field direction.

while accounting for Eqs. (3) and (4), i.e., $\mathbf{H}_{i,\text{eff}} \rightarrow \mathbf{H}_{i,\text{eff}} + \mathbf{H}_{i,f}$. To obtain the magnon energy spectrum we linearize the coupled LLG equations, seeking solutions of the form: $\mathbf{m}_i = (e^{i\omega t} \delta m_i^x, e^{i\omega t} \delta m_i^y, 1) = (0, 0, 1) + \delta \mathbf{m}_i(t)$ in the local coordinates. Here δm_i^x and δm_i^y are the small dynamic amplitudes that describe the elliptical precession of the magnetization around the direction of (dynamical) equilibrium.

Discussion. To parse the different magnon-magnon interactions, we first discuss the magnon frequency-field dependence without spin pumping. In this limit, Fig. 2 (a) and (b) show that there are a pair of, respectively, acoustic and optical magnon branches. In the absence of spin pumping, these “pure” optical and acoustic magnons can be differentiated in the theory (see Supplemental Section 2.2[33]), and both the acoustic pairs and optical pairs tend to “self-hybridize” with one another due to the RKKY interaction $\propto H_E$. This self-hybridization generates two avoided energy level crossings, one for each pair of modes. In earlier work, we used micro-magnetic simulations to show that near the avoided energy level crossing, the low-energy acoustic magnon branch resides on the interior layers, and the low-energy optical magnon branch primarily resides on the surface layers. In contrast, the high-energy acoustic magnon branch resides on the surface

layers, and the high-energy optical magnon branch resides on the interior layers[37]. Thus, even if only considering the RKKY interaction, pairs of magnons sharing the same character (acoustic or optical) are found to interact with one another. In Fig. 2 (c), we plot the combined magnon spectrum, and we observe three level crossings, i.e., between acoustic I and optical I, optical I and acoustic II, acoustic II and optical II, and no additional interactions between any acoustic-optical magnon pair.

By comparing the experimental results from Fig. 1 (d) with Fig. 2 (a), we see agreement between experiment and theory in that the acoustic gap is centered near 5.5 GHz, with comparable gap sizes between theory and experiment. Thus, when experimentally driving the tetralayer in the “acoustic configuration”, the RKKY interaction is responsible for the avoided energy level crossing we observe in Fig. 1 (d). In contrast, when comparing the theoretical spectrum shown in Fig. 2 (c) with the experimentally measured spectrum in the “optical configuration” [Fig. 1 (f)], it is clear that the model sans spin pumping is unable to reproduce (1) the flattening of the lowest magnon branch and (2) the mutual avoidance among all three magnon branches. In order to explain these experimental observations we use the extended model, where we find that acoustic and optical modes interact via the dynamic fieldlike and dampinglike torques exerted on one layer by the neighboring layers via spin pumping. The dampinglike torque amounts to a non-Hermitian contribution to the effective Hamiltonian derived upon linearization of Eqs. (1) and (2) [38]. The latter manifests as a change in the magnon linewidth and as magnon level attraction [8, 39–41], which can lead to energy coalescence and, thus, exceptional points [38, 42]. The fieldlike torques amount instead to additional fields acting on the magnetization dynamics, i.e., see Eqs. (3) and (4), leading to level repulsion between acoustic and optical modes.

In order to reproduce the flattening of the low energy magnon branch as well as the mutual avoidance amongst all three experimentally observed magnon branches, we set $\alpha_{f,AB} = \alpha_{f,BC} = 0.1$ and $\alpha_{f,CD} = 0.15$. We set the damping-like terms as $\alpha_{d,AB} = \alpha_{d,BC} = 0.01$, and $\alpha_{d,CD} = 0.015$, noting that a smaller magnitude in the damping like torques is needed to best reproduce experiment. The local Gilbert damping is set to $\alpha = 0.01$ for our calculations. In Fig. 2 (d) the resulting magnon branches are plotted. As indicated by dashed boxes, the original level crossings have turned into avoided level crossings, thereby creating mutual avoidance between the three lowest magnon branches. Furthermore, the two lowest energy magnon branches flatten out as the field increases, which. Thus, the measured experimental spectrum in the optical configuration shown in Fig. 1 (f) is most consistent with calculations incorporating interlayer fieldlike torques arising from interlayer spin pumping.

The torque parameters best describing our experimental results imply that an asymmetry exists between the top and bottom interfaces, i.e., $\alpha_{f,AB} \neq \alpha_{f,CD}$. If spin pumping creates fieldlike torques, this asymmetry lifts the degeneracies of the magnon spectrum and opens energy gaps between op-

tical and acoustic magnons; an extended discussion can be found in Supplemental material, Section 1[33]. The consequence of this asymmetry, visualized in Fig. 3, is that the symmetry axes describing the effective fields the surface and interior layers experience rotates away from the external field direction in opposite directions, i.e., $\mathbf{H}_{A,\text{eff}} \neq C_{2y}\mathbf{H}_{D,\text{eff}}$ and $\mathbf{H}_{B,\text{eff}} \neq C_{2y}\mathbf{H}_{C,\text{eff}}$. Crucially, the rotation of the surface and the interior layer symmetry axes away from one another only transpires if dynamic fieldlike torques are present. When the system is in a static equilibrium condition, i.e., during magnetometry measurements, we observe no experimental evidence that suggests the symmetry axes are misaligned. Thus, the energy-level avoidance between the acoustic and optical magnon branches is generated by dynamic fieldlike torques, but requires structural asymmetry within the tetralayer that does not affect the interlayer exchange interaction.

Evidence for inversion asymmetry in the structure of the sample itself is found by a series of atomic force microscopy measurements shown in the Supplemental material, Section 5[33]. Atomic force microscopy characterization of the surface roughness before and after metal deposition shows that the overall surface roughness decreases from 203 pm to 157 pm (averaged over three samples). The reduction in the surface roughness as the film thickness increases suggests that the interface between the ruthenium and permalloy at the bottom of the tetralayer is not equivalent to the interface at the top of the tetralayer due to the substrate.

Conclusions. In this work, the oft-neglected dynamic fieldlike torques generated by spin pumping were able to hybridize acoustic and optical magnons in S-AFM tetralayers. Looking ahead, the flexibility of the S-AFM material platform should allow for alternative multilayer structures where spin pumping can be either suppressed or enhanced, which leads to a new manner in which magnons can be manipulated and tuned in synthetic magnets. The phenomenology we have developed is not limited to just tetralayers, and can be applied to any other noncollinear magnetic multilayer structure. By using this phenomenology to describe other multilayers with specifically defined interfaces, we suggest that synthetic antiferromagnets are a playground not just for their flexibility in allowing experimentalists to control magnon-magnon interactions, but also for non-Hermitian phenomena. Future experiments that are able to harness dynamic antidamping torques associated with spin pumping may provide the key in unlocking the experimental ability to engineer exceptional points directly into the energy spectrum of synthetic antiferromagnetic materials.

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