Origin of Strong Two-Magnon Scattering in Heavy-Metal/Ferromagnet/Oxide Heterostructures

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We experimentally investigate the origin of two-magnon scattering (TMS) in heavy-metal (HM)/ferromagnet (FM)/oxide heterostructures (FM = Co, Ni81Fe19, or Fe60Co20B20) by varying the materials located above and below the FM layer. We show that strong TMS in HM/FM/oxide systems arises primarily at the HM/FM interface and increases with the strength of the interfacial spin-orbit coupling and magnetic roughness at this interface. TMS at the FM/oxide interface is relatively weak, even in systems where spin-orbit coupling at this interface generates strong interfacial magnetic anisotropy. We also suggest that the spin-current-induced excitation of nonuniform short-wavelength magnons at the HM/FM interface may function as a mechanism of spin memory loss for the spin-orbit torque exerted on the uniform mode.

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I. INTRODUCTION

The magnetic damping (α) of magnetic thin-film systems is a key parameter in the determination of the relaxation time of magnetization dynamics [1], the propagation distance of spin waves [2], the speed of antidamping torque switching of a macrospin [3], the velocity of current-induced skyrmion motion [4], and the energy efficiency of spin-torque magnetic memories [5,6], oscillators [7], and logic [8]. For in-plane-magnetized heavy-metal (HM)/ferromagnet (FM)/oxide heterostructures, the variation of α with the FM thickness (tFM) has been widely used to determine the effective spin-mixing conductance (G↑↓eff) of HM/FM interfaces for the analysis of spin transport based on the widespread assumption that spin pumping is the dominant source of α [9–16]. Recently, we have demonstrated that this assumption fails badly for most sputter-deposited in-plane-magnetized HM/FM/oxide heterostructures in the nanometer-scale tFM regime of interest for spintronic devices [17], leading to far-reaching consequences, including considerable overestimation of the true G↑↓eff and large errors in any analysis that relies on the value of G↑↓eff. Instead, we found that α for these sputter-deposited multilayers is generally dominated by two-magnon scattering (TMS) [17], in which a uniform magnon of a precessional macrospin scatters into a degenerate nonuniform short-wavelength magnon induced by material imperfections [18–20].

II. BACKGROUND

In the work presented here, we investigate in detail the origin of TMS in HM/FM/oxide devices, as well as the potential influence of nonuniform magnons on the efficiency of spin-orbit torques (SOTs). By varying the underlayer and the overlayer for different FM metals (e.g., Co, Ni81Fe19, and Fe60Co20B20), we find that the TMS in our HM/FM/oxide devices arises primarily from the interfacial spin-orbit coupling (ISOC) and magnetic roughness of the HM/FM interface. In contrast, the TMS at the FM/oxide interface is much weaker even when strong ISOC at this interface generates a large interfacial perpendicular magnetic anisotropy (PMA). We also suggest that, when the ISOC is strong at the HM/FM interface, the spin-current-induced excitation of nonuniform short-wavelength magnons at this interface may influence SOT experiments as a mechanism of spin memory loss (SML).

Here g is the g-factor, µB the Bohr magnetron, h the Planck constant, and Ms the saturation magnetization of...
the FM layer. The second term of Eq. (1) is a combined contribution ($\alpha_{SP}$) from the spin-pumping spin current that is absorbed in the HM layer [9–14] and at the HM/FM interface due to SML [16,22,23], which for convenience we parameterize as an “effective SML conductance” $G_{SML}$. The third term of Eq. (1), $\alpha_{TMS} = \beta_{TMS} \delta_{FM}^{-2}$, is the TMS damping arising from the combination of ISOC and magnetic roughness (e.g., variations of the thickness and/or the ISOC strength) [17–20]. The coefficient $\beta_{TMS} = C_{TMS} (2K_{ISOC} / M_s)^2$ [17,18], where $C_{TMS}$ is a parameter related to the density and geometry of the scattering defects at the interfaces [19,20] and $K_{ISOC}$ is the interfacial magnetic energy density of the associated interface. In most HM/FM/oxide heterostructures with a $t_{FM}$ of only a few nanometers, $\alpha_{TMS} \gg \alpha_{SP}$ [17].

### III. SAMPLE CONFIGURATIONS

As listed in detail in Table I, the magnetic stacks studied in the work presented here consist of six Co-based sample series (A1–A6), four Ni$_{81}$Fe$_{19}$-based sample series (B1–B4), and six Fe$_{60}$Co$_{20}$B$_{20}$-based sample series (C1–C6). In each series, different materials are deposited below and/or above the FM layer. All the stacks are deposited by dc/rf sputtering onto 4 inch oxidized silicon substrates. For most samples, the FM layer is prepared by oblique deposition to make a wedge, allowing devices with different FM thicknesses to be studied across a single wafer. Each FM wedge is 75 mm long. The thickness slopes are approximately $6.9 \times 10^{-5}$ Å/μm for the Co wedges, $4.6 \times 10^{-3}$ Å/μm for the Ni$_{81}$Fe$_{19}$ wedges, and $(5.9–9.0) \times 10^{-3}$ Å/μm for the Fe$_{60}$Co$_{20}$B$_{20}$ wedges. Some samples are also made with a constant ferromagnetic thickness by rotating the substrate during deposition, from which we verify that the oblique sputtering of the wedges does not affect the results (see below). A 1 nm Ta seed layer is deposited as part of the growth in all the sample series other than A1 and B1. All samples with a MgO layer were capped with 1.5 nm of Ta.

Sample series A1–A6, B1–B3, and C1 (Table I) are patterned into magnetic strips (10 $\times$ 20 µm$^2$) via photolithography and ion milling to measure $\alpha_0$, the effective demagnetization field ($4\pi M_{eff}$), and the SOT efficiency.

| Number | Magnetic heterostructure | $\beta_{TMS}$ (nm$^2$) | $2K_{ISOC}/M_s$ (T nm) | Technique |
|--------|--------------------------|------------------------|------------------------|-----------|
| A1     | Si/SiO$_2$/Co(1.8–6.6 nm, wedge)/Pt(4.5 nm) | 2.73 ± 0.42            | −3.48 ± 0.10           | ST-FMR    |
| A2     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 0.41 ± 0.04            | −2.69 ± 0.03           | ST-FMR    |
| A3     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Hf(0.3 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 0.32 ± 0.01            | −1.72 ± 0.08           | ST-FMR    |
| A4     | Si/SiO$_2$/Ta(1 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 0                      | −0.74 ± 0.01           | ST-FMR    |
| A5     | Si/SiO$_2$/Ta(1 nm)/Pt(5.3 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 1.38 ± 0.10            | −1.85 ± 0.05           | ST-FMR    |
| A6     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Co(1.8, 2, 3, 5 nm)/MgO(2 nm) | ...                    | ...                    | ST-FMR    |
| B1     | Si/SiO$_2$/Ni$_{81}$Fe$_{19}$(1.8–5 nm, wedge)/Pt(4.5 nm) | 0.41 ± 0.01            | −1.80 ± 0.07           | ST-FMR    |
| B2     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Ni$_{81}$Fe$_{19}$(1.8, 2, 3 nm)/MgO(2 nm) | ...                    | ...                    | ST-FMR    |
| B3     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Ni$_{81}$Fe$_{19}$(1.8–5 nm, wedge)/MgO(2 nm) | 0.30 ± 0.02            | −1.04 ± 0.05           | ST-FMR    |
| B4     | Si/SiO$_2$/Ta(1 nm)/MgO(2 nm)/Ni$_{81}$Fe$_{19}$(1.8–5 nm, wedge)/MgO(2 nm) | 0.04 ± 0.01            | −1.43 ± 0.07           | Flip-chip FMR |
| C1     | Si/SiO$_2$/Ta(1 nm)/Pt(4nm)/Fe$_{60}$Co$_{20}$B$_{20}$(1.6–5.7 nm, wedge)/MgO(2 nm) | 0.04 ± 0.01            | −1.35 ± 0.10           | ST-FMR    |
| C2     | Si/SiO$_2$/Ta(1 nm)/Ta(4 nm)/Fe$_{60}$Co$_{20}$B$_{20}$(3.0–10.7 nm, wedge)/MgO(2 nm) | 0.09 ± 0.01            | −2.70 ± 0.08           | Flip-chip FMR |
| C3     | Si/SiO$_2$/Ta(1 nm)/MgO(2 nm)/Fe$_{60}$Co$_{20}$B$_{20}$(1.6–5.7 nm, wedge)/MgO(2 nm) | 0.20 ± 0.01            | −3.26 ± 0.10           | Flip-chip FMR |
| C4     | Si/SiO$_2$/Ta(1 nm)/Au$_{25}$Pt$_{75}$(4 nm)/Fe$_{60}$Co$_{20}$B$_{20}$(1.6 nm)/MgO(2 nm) | ...                    | ...                    | Flip-chip FMR |
| C5     | Si/SiO$_2$/Ta(1 nm)/Au$_{25}$Pt$_{75}$(4 nm)/Pt(0.5 nm)/Hf(0.25 nm)/Fe$_{60}$Co$_{20}$B$_{20}$(1.6 nm)/MgO(2 nm) | ...                    | ...                    | Flip-chip FMR |
| C6     | Si/SiO$_2$/Ta(1 nm)/Au$_{25}$Pt$_{75}$(4 nm)/Pt(0.5 nm)/Hf(0.25 nm)/Fe$_{60}$Co$_{20}$B$_{20}$(1.6 nm)/Hf(0.1 nm)/MgO(2 nm) | ...                    | ...                    | Flip-chip FMR |

*Sample series A1–A6, B1–B4, and C1–C6 are heterostructures based on Co, Ni$_{81}$Fe$_{19}$, and Fe$_{60}$Co$_{20}$B$_{20}$, respectively. For the sample series grown with a wedge-shaped ferromagnetic layer, measurements as a function of ferromagnet thickness allow measurements of the coefficient of two-magnon scattering ($\beta_{TMS}$) and the ratio of the interface magnetic anisotropy to the saturation magnetization ($2K_{ISOC}/M_s$).*
using spin-torque ferromagnetic resonance (ST-FMR) [24,25], and also into Hall bars (5 × 65 µm²) for determination of the SOT efficiency using the “in-plane” harmonic Hall-response technique [26–28]. α and 4πMeff for sample series B4 and C2–C6 (Table I) are obtained from unpatterned pieces using flip-chip FMR with the magnetic pieces face-side down on a coplanar waveguide.

IV. SOURCE OF TWO-MAGNON SCATTERING IN HM/FM/OXIDE STRUCTURES

Using ST-FMR [24,25] and flip-chip FMR in the radio-frequency regime with a frequency (f) from 7 to 18 GHz, we measure α and 4πMeff for each sample from the best fits of the FMR linewidth (ΔH, half width at half maximum) and resonance field (Hr) to the relations [29]

\[ ΔH = ΔH₀ + 2παf/γ, \]

\[ f = (γ/2π)\sqrt{H_r(H_r + 4πMeff)}, \]

where ΔH₀ is the inhomogeneous broadening of the FMR linewidth and γ is the gyromagnetic ratio. In Fig. 1(a), we plot the values of α for SiO₂/Co/Pt (series A1), Pt/Co/MgO (series A2), Pt/Hf/Co/MgO (series A3), and Ta/Co/MgO (series A4) as a function of tCo⁻¹. We find that α for Ta/Co/MgO (series A4) remains small and almost constant as a function of tCo⁻¹, corresponding to αint = 0.0126 ± 0.0001 for the Co layer, with negligible amounts of both TMS and spin pumping. From this, we can exclude both the Ta/Co and the Co/MgO interfaces as strong sources of TMS. This result is consistent with previous measurements of weak damping enhancement at Ta/FM interfaces [14,15] and the weak ISOC of Ta/Co (see below).

In contrast, the values of α for sample series A1–A3 vary proportionally to tCo⁻², indicating that TMS is the dominant mechanism of magnetic damping in these heterostructures. From the best fits of the data in Fig. 1(a) to

\[ α = α_{\text{int}} + β_{\text{TMS}}t_{\text{FM}}^{-2}, \]

we obtain the values listed in Table I for βTMS, which parameterizes the strength of the TMS. βTMS is substantial in all three sample series and is almost one order of magnitude larger for SiO₂/Co/Pt (A1) than for Pt/Co/MgO (A2) and Pt/Hf/Co/MgO (A3). As expected [17,18], the strength of the TMS is correlated with the strength of the ISOC and the magnetic roughness. We can determine the total interfacial magnetic-anisotropy density (Ks, the
sum arising from both the interfaces of the FM) and the saturation magnetization $M_s$ using fits of $4\pi M_{\text{eff}}$ vs $t_{\text{Co}}^{-1}$ [Fig. 1(b)] to the relation [29]

$$4\pi M_{\text{eff}} \approx 4\pi M_s + 2K_s/M_s t_{\text{Co}}.$$  

(5)

The values obtained for $K_s$ and $M_s$ for sample series A1–A4 are summarized in Table II. If we assume that the small $K_s$ for Ta/Co/MgO (A4) is due mostly to the Co/MgO interface (i.e., $K_{s,\text{Co/MgO}} \approx 0.34$ erg/cm$^2$) and that $K_s$ is zero for the SiO$_2$ interface, we can estimate $K_{s,\text{ISOC}}$ for the individual HM/FM interfaces to be $2.31 \pm 0.05$ erg/cm$^2$ for Co/Pt (A1), $1.44 \pm 0.01$ erg/cm$^2$ for Pt/Co (A2), and $0.86 \pm 0.03$ erg/cm$^2$ for Pt/Hf/Co (A3) (see also Table II). In Fig. 1(c), we plot $\beta_{\text{TMS}}$ for these four series of Co samples as a function of $(2K_{s,\text{ISOC}}/M_s)^2$. For the three sample series (A2–A4) deposited with a Ta seed layer, we find an accurate linear scaling. This is consistent with the expectation for TMS [i.e., $\beta_{\text{TMS}} = C_{\text{TMS}}(2K_{s,\text{ISOC}}/M_s)^2$] and indicates similar magnetic roughnesses of these HM/Co interfaces ($C_{\text{TMS}} \approx 0.08$ T$^2$). In contrast, $\beta_{\text{TMS}}$ for SiO$_2$/Co/Pt (A1) is fourfold higher than the value extrapolated from the linear fit in Fig. 1(c), suggesting a considerable increase in magnetic roughness. Cross-sectional transmission-electron-microscopy (TEM) images of the samples are shown in Figs. 1(d)–1(f). In SiO$_2$/Co/Pt [A1, Fig. 1(d)], the Co layer, whose nominal “thickness” is 2.3 nm, has a granular texture and is thus magnetically very rough. This granularity arises because Co has a much higher surface energy than SiO$_2$, while the Pt grows coherently on the Co grains. In contrast, for Pt/Co/MgO [A2, Fig. 1(e)] the Co layer is atomically smooth at both interfaces and coherently follows the Pt lattice. When a 0.3 nm Hf layer is inserted at the Pt/Co interface in sample series A3 [Fig. 1(f)], the Co layer still grows in a relatively smooth manner, while its coherent growth is substantially interrupted by the Hf insertion, as indicated by the distortion of the lattice planes in the Co layer near the interface with the Pt/Hf. In both Pt/Co/MgO (A2) and Pt/Hf/Co/MgO (A3), the Co/MgO interface is atomically sharp [Figs. 1(e) and 1(f)], consistent with a negligible TMS at the Co/MgO interface. The relatively strong TMS at the atomically smooth Pt/Co interface in Pt/Co/MgO (A2) and Pt/Hf/Co/MgO (A3) suggests that the observed TMS in those samples is due mainly to the variation in the ISOC induced by the polycrystalline texture (e.g., the different orientations and dimensions of crystalline grains) rather than a thickness-induced roughness. The much stronger TMS in SiO$_2$/Co/Pt (A1) may also have a contribution from the much larger roughness in those samples.

Our results for samples with Ni$_{81}$Fe$_{19}$ magnetic layers (sample series B1–B4, shown in Fig. 2) are similar to those for the Co-based samples. The TMS at the Ni$_{81}$Fe$_{19}$/MgO interfaces is minimal; see the results for MgO/Ni$_{81}$Fe$_{19}$/MgO (B4) in Fig. 2(a). The presence of a Pt/Ni$_{81}$Fe$_{19}$ interface enhances the TMS for Pt/Ni$_{81}$Fe$_{19}$/MgO (B2, B3), and the TMS is largest in the sample series without a smoothing Ta seed layer, i.e., SiO$_2$/Ni$_{81}$Fe$_{19}$/Pt (B1). The value of $\beta_{\text{TMS}}$ is small for MgO/Ni$_{81}$Fe$_{19}$/MgO (B4) despite the fact that $(2K_{s,\text{ISOC}}/M_s)^2$ is 1.5 times larger for this sample than for Pt/Ni$_{81}$Fe$_{19}$/MgO (B2 and B3) [see Table I and Fig. 2(b)]. Within the usual model of TMS [17, 18], this suggests that the Ni$_{81}$Fe$_{19}$/MgO interfaces are magnetically smooth, with small values of $C_{\text{TMS}}$.

As we show in Fig. 3(a), the TMS in the HM/Fc$_{60}$Co$_{20}$B$_{20}$/MgO sample series is weaker than that in Co and Ni$_{81}$Fe$_{19}$ samples, but nevertheless it is clearly measurable. The small values of $\beta_{\text{TMS}}$ for the Fe$_{60}$Co$_{20}$B$_{20}$ samples (e.g., 0.04 nm$^{-2}$ for Pt/Fe$_{60}$Co$_{20}$B$_{20}$/MgO and 0.09 nm$^{-2}$ for Ta/Fe$_{60}$Co$_{20}$B$_{20}$/MgO) are consistent with a relatively weak ISOC [Fig. 3(b)]. We continue to find that $\alpha$ in the HM/Fe$_{60}$Co$_{20}$B$_{20}$/MgO systems is strongly dependent on the details of the HM/Fe$_{60}$Co$_{20}$B$_{20}$ interface but insensitive to even a strong ISOC at the Fe$_{60}$Co$_{20}$B$_{20}$/MgO interface. For instance, as we show in Figs. 4(a)

### Table II: Saturation magnetization ($M_s$), total interfacial magnetic-anisotropy density ($K_s$), and interfacial magnetic-anisotropy density of individual HM/FM interfaces ($K_{s,\text{ISOC}}$) for the sample series A1–A4.

| Number | Magnetic heterostructure | $M_s$ (emu/cm$^3$) | $K_s$ (erg/cm$^2$) | $K_{s,\text{ISOC}}$ (erg/cm$^2$) |
|--------|--------------------------|-------------------|-------------------|-----------------------------|
| A1     | Si/SiO$_2$/Co(1.8–6.6 nm, wedge)/Pt(4.5 nm) | 1417 ± 30 | 2.31 ± 0.05 | 2.31 ± 0.05 |
| A2     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 1314 ± 8 | 1.78 ± 0.01 | 1.44 ± 0.01 |
| A3     | Si/SiO$_2$/Ta(1 nm)/Pt(4.5 nm)/Hf(0.3 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 1352 ± 12 | 1.21 ± 0.03 | 0.86 ± 0.03 |
| A4     | Si/SiO$_2$/Ta(1 nm)/Co(1.8–6.6 nm, wedge)/MgO(2 nm) | 1134 ± 16 | 0.34 ± 0.04 | ≈0 |

*aThe $K_{s,\text{ISOC}}$ values for the Co/Pt (A1), Pt/Co (A2), Pt/Hf/Co (A3), and Ta/Co (A4) interfaces are estimated by assuming that the small $K_s$ for Ta/Co/MgO (A4) is due mostly to the Co/MgO interface (i.e., $K_{s,\text{Co/MgO}} \approx 0.34$ erg/cm$^2$) and that $K_s$ is zero for the SiO$_2$ interface.
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FIG. 2. Results for Ni81Fe19-based heterostructures. (a) $\alpha$ vs $t_{\text{NiFe}}^{-1}$ and (b) $4\pi M_{\text{eff}}$ vs $t_{\text{NiFe}}^{-1}$ for SiO2/Ni81Fe19 (tNiFe)/Pt(4.5 nm) (series B1), Pt(4.5 nm)/Ni81Fe19(tNiFe)/MgO (B2), Pt(4.5 nm)/Ni81Fe19(1.8, 2, 3 nm)/MgO(2 nm) (B3), MgO(2 nm)/Ni81Fe19 wedge/MgO (B4). The lines in (a) represent the best fits of the data to Eq. (4); the straight lines in (b) represent the best linear fits of the data to Eq. (5).

and 4(b), $\alpha$ of Au25Pt75(4 nm)/Fe60Co20B20(1.6 nm)/MgO (C4) is reduced markedly when a spacer bilayer of Pt(0.5 nm)/Co(0.25 nm) is inserted at the Au25Pt75/Fe60Co20B20 interface (sample C5), likely due to a reduction in the ISOC [as indicated by an increased value of $4\pi M_{\text{eff}}$, Fig. 4(b)]. However, $\alpha$ remains similar when $4\pi M_{\text{eff}}$ is reduced by up to 50% due to the insertion of a 0.1-nm Hf spacer at the Fe60Co20B20/MgO interface (sample C6). The same is true after the samples are annealed at 450°C [Figs. 4(c) and 4(d)]. This is consistent with previous measurements of a substantial reduction of $\alpha$ in (Pt(5 nm)/Co40Fe40B20)/MgO by insertion of an ultrathin Hf spacer between the HM and FM layers [30,31].

An earlier pump-probe magneto-optical-Kerr-effect experiment [14] also indicated that the increase in the in-plane damping with $t_{\text{Fe}}^{-1}$ is approximately a factor of 2 faster for Ta(5 nm)/Co40Fe40B20/Ta(5 nm) (annealed at 250°C) than for Ta(5 nm)/Fe40Co40B20/MgO (annealed at 250°C). These observations consistently reveal that the TMS of HM/Fe-Co-B/MgO arises primarily from the HM/Fe-Co-B interface, while the Fe-Co-B/MgO interface is magnetically smoother and contributes minimally to $\alpha$ despite the fact that it is the primary source of the total ISOC and the interfacial PMA. This is a technologically interesting observation because it indicates that HM/Fe-Co-B/oxide devices can be tuned to have a low $\alpha$ and low $4\pi M_{\text{eff}}$ (high PMA) at the same time by separately reducing the ISOC at the HM/Fe-Co-B interface and enhancing the ISOC at the Fe-Co-B/oxide interface.

We do find a significant TMS ($f_{\text{TMS}} \approx 0.2$ nm$^{-2}$) in MgO/Fe60Co20B20/MgO [C3, Fig. 3(a)], suggesting a large magnetic roughness and an enhanced ISOC at the MgO/Fe60Co20B20 interface. This is similar to the roughness obtained from SiO2/Co [A1, Fig. 1(a)], SiO2/Ni81Fe19 [B1, Fig. 2(a)], and previous measurements on SiO2/Ni50Fe50 [19]. The increased magnetic roughness is likely to be because the surface energy of the metallic FMs is higher than that of MgO and SiO2.

V. INSENSITIVITY TO OBLIQUE GROWTH OF THE FM LAYER

It is well established that the strength of TMS can vary when the magnetic precession axis is oriented along different directions with respect to an anisotropic surface defect [18–20]. Oblique deposition of thin-film wedges has the potential to induce anisotropic tilting of crystalline grains
As-grown and annealed at 450°C. (a) Dependence of the ferromagnetic-resonance linewidth ($\Delta H$) on frequency ($f$) and (b) $f$ vs FMR resonance field ($H_r$) for Au$_{25}$Pt$_{75}$, Au$_{25}$Pt$_{75}$/Pt(0.5 nm)/Hf(0.25 nm)/Fe$_{60}$Co$_{20}$B$_{20}$/MgO (sample C4), Au$_{25}$Pt$_{75}$/Pt(0.5 nm)/Hf(0.25 nm)/Fe$_{60}$Co$_{20}$/B$_{20}$/MgO (C5), and Au$_{25}$Pt$_{75}$/Pt(0.5 nm)/Hf(0.25 nm)/Fe$_{60}$Co$_{20}$/B$_{20}$/Hf(0.1 nm)/MgO (C6).

(c) and (d) The same quantities after annealing the samples at 450°C for 1 h. All of the data are taken with flip-chip FMR on unpatterned sample pieces. The lines represent the best fits of the data to Eq. (2) in (a) and (c) and to Eq. (3) in (b) and (d).

as well as variations in film thickness [32–35]. Here we affirm that our analysis of TMS is not affected by the oblique deposition that we use to make wedge-shaped samples.

We first pattern identical ST-FMR microstrips (20 $\times$ 30 $\mu$m$^2$) with different orientations [$\varphi = 0°$, $±30°$, $±45°$, $±60°$, and 90° relative to the wedge gradient; see Fig. 5(a)] for each fixed value of $t_{FM}$ obtained from the Pt(5.3 nm)/Co(1.8–6.6 nm)/MgO samples (sample series A5). In all cases, ST-FMR is performed with an applied field oriented at a fixed angle of 45° from the microstrip axis. As shown in Figs. 5(b) and 5(c), the ST-FMR
measurements of both $\alpha$ and $4\pi M_{\text{eff}}$ are independent of $\varphi$ within our experimental sensitivity, indicating the absence of any anisotropy due to the orientation of the magnetic precession axis relative to the wedge direction. In Fig. 5(c), we show that the $\alpha$ values for the wedge-shaped sample (A2) agree with those for sample A6, where the Co layers are grown uniformly with substrate rotation during growth. We also find no indication of any sensitivity to oblique deposition in $\alpha$ or $4\pi M_{\text{eff}}$ for the Ni$_{81}$Fe$_{19}$ layer in the Pt/Ni$_{81}$Fe$_{19}$ bilayers (samples B1 and B2). From these observations, we can safely conclude that the oblique deposition of the FM wedge is not an important source of the observed TMS in our HM/FM/oxide systems.

VI. INFLUENCE OF SHORT-WAVELENGTH MAGNONS ON SPIN TORQUE

It is known that magnons can efficiently interconvert with spin currents [36–40] and generate an inverse spin Hall voltage [36,37]. It is therefore an important question as to how nonuniform short-wavelength magnons at a HM/FM interface affect the SOT efficiency in a heterostructure. As schematically shown in Fig. 6(a), nonuniform short-wavelength magnons might affect a SOT measurement via four possible processes: (i) they can be excited directly by the spin current from the HM layer and relax into the lattice; (ii) they can be excited by relaxation of the uniform magnon mode and then subsequently relax into the lattice; (iii) they can be excited by the spin current from the HM layer and then relax by transferring spin angular momentum to the uniform mode; and (iv) they can be excited by the rf Oersted field and then relax by transferring angular momentum to the uniform mode. In the first process, the nonuniform magnons would behave as a source of SML that reduces the efficiency of the SOT; in the second process, the short-wavelength magnons would provide an additional channel for damping in the second process. Processes (iii) and (iv) would enhance the SOT efficiency for the uniform mode if the interconversion of the spin current is more efficient for the short-wave magnons than for the uniform mode (e.g., as indicated for yttrium iron garnet/HM structures [36,37]).

To examine the possible effects of nonuniform short-wavelength magnons on the SOT exerted on the uniform mode, we determine the dampinglike SOT efficiency for the Co/Pt (A1), Pt/Co (A2), and Pt/Hf/Co (A3) samples using both ST-FMR measurements [24] and harmonic response measurements [26,27]. For the ST-FMR determination, if we employ the standard macrospin analysis and assume a negligible spin-pumping effect, we can define an effective FMR spin-torque efficiency $\xi_{\text{FMR}}$ [24] from the ratio of the symmetric ($S$) and antisymmetric ($A$) components of the FMR signal.
components of the magnetoresistance response of the ST-FMR resonance. $S$ is proportional to $H_{DL}$, and $A$ is due to the sum of the Oersted field and the fieldlike SOT effective field. The dampinglike and fieldlike SOT efficiencies per unit applied electric field, $\xi_{DL}^E$ and $\xi_{DL}^H$, respectively, can then be obtained from the linear dependence of $\xi_{FMR}^{-1}$ on $t_{FM}^{-1}$ when $\xi_{DL}^E$, $\xi_{DL}^H$, the HM resistivity ($\rho_{HM}$), and $M_s$ are approximately constant over the $t_{FM}$ regime studied [24]:

$$\frac{1}{\xi_{FMR}} = \frac{1}{\xi_{DL}^H \rho_{HM}} \left( 1 + \frac{h}{e} \mu_0 M_s t_{FM} \right).$$

Here, $\rho_{xx}$ for the 4.5 nm Pt layer is 61 $\mu$Ω cm for the Co/Pt samples, 35 $\mu$Ω cm for the Pt/Co samples, and 40 $\mu$Ω cm for the Pt/Hf/Co samples. As plotted in Fig. 6(b), $\xi_{DL}^E$ is estimated from the ST-FMR measurement to be $0.98 \pm 0.03 \times 10^5 \Omega^{-1} \text{m}^{-1}$ for the Co/Pt samples (A1), $1.62 \pm 0.04 \times 10^5 \Omega^{-1} \text{m}^{-1}$ for the Pt/Co samples (A2), and $2.64 \pm 0.29 \times 10^5 \Omega^{-1} \text{m}^{-1}$ for the Pt/Hf(0.3 nm)/Co samples (A3).

For the harmonic response measurements on Co(2.3 nm)/Pt(4.5 nm) (A1), Pt(4.5 nm)/Co(2.5 nm) (A2), and Pt(4.5 nm)/Hf(0.3 nm)/Co(2.3 nm) (A3), the second-harmonic Hall-voltage response ($V_{a2}$) is measured as a function of the in-plane orientation of the magnetization ($\varphi$) under different fixed magnitudes of an in-plane magnetic bias field ($H_{an}$) of 1–3.5 kOe, with a low-frequency sinusoidal electric field (61.5 kV/m, 1.327 kHz) applied on Hall bars. As described in more detail in Refs. [27,28], the cos $\varphi$-dependent component ($V_a$) of $V_{a2}$ follows

$$V_a = -H_{DL} V_{AH}/2(H_{in} + H_{k}) + V_{ANE},$$

where $H_{DL}$ is the dampinglike SOT effective field, $V_{AH}$ the anomalous Hall voltage, $H_{k}$ the anisotropic field, and $V_{ANE}$ the anomalous Nernst voltage. Using the values of $H_{DL}$ given by the slope of the linear fits of the $V_a$ data to Eq. (7) [Fig. 6(d)], we determine $\xi_{DL}^E$ for the samples following $\xi_{DL}^E = (2e/h) \mu_0 H_{DL} M_{s} C_{xx} / E$. As plotted in Fig. 6(d), $\xi_{DL}^E$ obtained from the harmonic response measurement increases from $(3.17 \pm 0.11) \times 10^5 \Omega^{-1} \text{m}^{-1}$ for Co(2.3 nm)/Pt(4.5 nm) (A1) to $(4.58 \pm 0.05) \times 10^5 \Omega^{-1} \text{m}^{-1}$ for Pt(4.5 nm)/Co(2.5 nm) (A2) and to $(5.69 \pm 0.87) \times 10^5 \Omega^{-1} \text{m}^{-1}$ for Pt(4.5 nm)/Hf(0.3 nm)/Co(2.3 nm) (A3).

The $\xi_{DL}^E$ values measured using either ST-FMR or the harmonic response decrease with $K_{s}$ in approximately a linear manner, which, together with our previous observations on HM/Co bilayers annealed under different conditions [41], indicates a linear decrease in the spin transparency of the interface ($T_{int}$). This is because $\xi_{DL}^E = T_{int} \sigma_{SH}$ for a HM/FM bilayer and the spin Hall conductivity of the HM ($\sigma_{SH}$) is constant when $\rho_{xx}$ is constant [27,28].

$T_{int}$ for a SOT process should be given by

$$T_{int} = G_{HM/FM}^{↑\downarrow} / (G_{HM/FM}^{↑\downarrow} + G_{SML} + G_{HM}/2),$$

where $G_{HM/FM}^{↑\downarrow}$ is the bare spin-mixing conductance of the interface and $G_{HM} = 1/\rho_{xx} \lambda_s$ is the spin conductance of the HM, and where $\lambda_s$ is the spin diffusion length of the HM. $G_{HM}$ should be constant within the Elliot-Yafet spin-relaxation mechanism [42,43], while $G_{HM/FM}^{↑\downarrow}$ and $G_{SML}$ can be modulated by changes at the interface [44,45]. The monotonic decrease in $T_{int}$ with $K_{s}$ should indicate an increase in $G_{SML}$ with $K_{s}$. This might be suggestive of the possibility that nonuniform magnons are excited directly from the HM layer by the spin current and relax into the lattice [the aforementioned process (ii)]. The decrease in $T_{int}$ with $K_{s}$ is less likely to suggest a decrease in $G_{HM/FM}^{↑\downarrow}$ here, because previous studies have indicated that magnetic roughness (e.g., induced by diffusion) may increase $T_{int}$ via moderately enhancing $G_{HM/FM}^{↑\downarrow}$ [44,45]. Finally, while processes (iii) and (iv), which should increase $\xi_{DL}^E$, are still possible, they seem to be a weaker effect than the SML process, as indicated by the decrease in $T_{int}$ with increasing $K_{s}$.

It is also an interesting observation that the values of $\xi_{DL}^E$ obtained from the standard ST-FMR analysis are more than a factor of 2 smaller than those obtained from the harmonic response measurements [Fig. 6(d)], with this ratio getting larger as the ISOC becomes greater. However, this difference cannot be fully attributed to the excitation of short-wavelength magnons, because the difference still seems to exist at zero ISOC, as indicated by the extrapolation of the data to zero $K_{s}$ [straight lines in Fig. 6(d)]. We note that the ST-FMR measurements here are accompanied by significant spin pumping in the thick-Co regime, as indicated by the deviation of $\xi_{FMR}^{-1}$ from the linear $t_{FM}^{-1}$ dependence [Fig. 6(c)], while spin pumping seems negligible in the thin-Co regime, where we take data to determine $\xi_{DL}^E$ according to Eq. (6). Future efforts to unveil the cause of the different values of $\xi_{DL}^E$ are warranted, but that is beyond the scope of the present work. Here, it worth mentioning that the “in-plane” harmonic Hall-response measurements, if performed carefully [26,27], yield results for $\xi_{DL}^E$ that are consistent with those obtained from “out-of-plane” harmonic Hall-response measurements [46,47] and antidamping SOT switching of in-plane-magnetized three-terminal magnetic tunnel junctions [48,49].

VII. CONCLUSION

We show that the strong extrinsic damping in HM/FM/oxide systems arises predominantly from TMS due to the coexistence of ISOC and magnetic roughness at the HM/FM interface, while FM/oxide interfaces and the oblique growth of the FM layer are largely irrelevant to this damping. These results indicate that the energy effi-
ciency of SOT-driven magnetic memories, oscillators, and logic devices, where a HM/FM/oxide structure is the core ingredient, can be substantially improved by separately reducing the ISOC at the HM/FM interface and enhancing the ISOC at the FM/oxide interface through interface engineering. We also suggest that short-wavelength magnons may be excited in the FM layer by a spin current from the HM layers and subsequently relax into the lattice, and thus function as a source of SML in a SOT process. These results indicate that the ISOC at and magnetic roughness of the HM/FM interfaces should be minimized in spin-torque memories and logic, where high spin-torque efficiency and low damping are required to reduce the power consumption.

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