Charge-Spin Interconversion in Epitaxial Pt Probed by Spin-Orbit Torques in a Magnetic Insulator

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L4	We measure spin-orbit torques (SOTs) in a unique model system of all-epitaxial ferrite/Pt bilayers
15	to gain insights into charge-spin interconversion in Pt. With negligible electronic conduction in the
16	insulating ferrite, the crystalline Pt film acts as the sole source of charge-to-spin conversion. A
17	small field-like SOT independent of Pt thickness suggests a weak Rashba-Edelstein effect at the
18	ferrite/Pt interface. By contrast, we observe a sizable damping-like SOT that depends on the Pt
19	thickness, from which we deduce the dominance of an extrinsic spin-Hall effect (skew scattering)

thickness, from which we deduce the dominance of an extrinsic spin-Hall effect (skew scattering) and Dyakonov-Perel spin relaxation the crystalline Pt film. Furthermore, our results point to a large internal spin-Hall ratio of ≈ 0.8 in epitaxial Pt. Our experimental work takes an essential step towards understanding the mechanisms of charge-spin interconversion and SOTs in Pt-based heterostructures, which are crucial for power-efficient spintronic devices.

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I. INTRODUCTION

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Spin-orbit torques (SOTs)^{1,2} have been recognized ⁵⁸ 25 as a viable means to manipulate magnetization in 59 26 thin-film heterostructures. A prototypical SOT-driven ⁶⁰ 27 medium consists of a ferro(ferri)magnetic metal (FM) 61 28 interfaced with a nonmagnetic heavy metal (HM) with 62 29 strong spin-orbit coupling (e.g., Pt). In a conventional 63 30 picture of SOTs in such a bilayer, an in-plane charge 64 31 current through the HM (or its surface) generates non-65 32 equilibrium spin accumulation via the spin-Hall effect 66 33 (or Rashba-Edelstein effect)¹⁻⁴. This charge-to-spin 67 34 conversion then results in SOTs^{1,2,5,6}, typically classified ⁶⁸ 35 into (1) a "damping-like" torque that either enhances 69 36 or counteracts damping in the magnetic layer and (2) a ⁷⁰ 37 "field-like" torgue that acts similarly to a torgue from an ⁷¹ 38 external magnetic field. 72 39

Although SOTs are often attributed to charge-to-spin 73 40 conversion effects in the HM, recent studies point to 74 41 other effects that impact SOTs in metallic FM/HM 75 42 bilayers⁷⁻²⁶. For example, current shunted through ⁷⁶ 43 the FM can generate additional SOTs through spin-77 44 dependent scattering within the FM or across the 78 45 FM/HM interface^{7-16,27}. Roughness at the interfaces 79</sup> 46 of FM/HM bilayers, which are typically disordered ⁸⁰ 47 (i.e., polycrystalline or amorphous), may also contribute 81 48 to SOTs^{17–19}. Even with atomically sharp FM/HM 82 49 interfaces, SOTs may be intrinsically impacted by spin-83 50 memory $loss^{20-24}$ and proximity-induced magnetism^{25,26} ₈₄ 51 due to orbital hybridization. 85 52

These possible complications in FM/HM bilayers make ⁸⁶ it difficult to elucidate the fundamental mechanisms of ⁸⁷ SOTs and, more generally, the underlying charge-to- ⁸⁸ spin conversion phenomena. These factors also impede reconciling the wide spread of reported spin transport parameters – particularly for the often-used HM of Pt, with its spin diffusion length in the range ~1-10 nm and its spin-Hall ratio ~ $0.01-1^{20,23,28-43}$.

Here, we demonstrate a clean ferrimagneticinsulator/heavy-metal (FI/HM) model system where SOTs originate solely in the HM layer, permitting analysis of charge-to-spin conversion a simpler Specifically, we investigate SOTs at mechanisms. room temperature in FI/HM bilayers where the FI is an epitaxial spinel ferrite film of MgAl_{0.5}Fe_{1.5}O₄ $(MAFO)^{44}$ and the HM is an epitaxial film of Pt, whose high crystallinity is enabled by its excellent lattice match to the spinel⁴⁵. The insulating nature of MAFO removes all complications from electronic spin transport in the magnetic layer $^{7-16,27}$, and the Pt layer with a sharp crystalline interface minimizes roughness-induced mechanisms^{17–19}. Spin-memory loss and proximityinduced magnetism are also expected to be significantly weaker in $\widetilde{\text{FI}/\text{HM}^{46-49}}$ compared to $\overline{\text{FM}/\text{HM}^{20-26}}$ due to weaker interfacial hybridization²².

We leverage the low damping of MAFO⁴⁴ to quantify both the damping-like and field-like SOTs in a straightforward manner through dc-biased spintorque ferromagnetic resonance $(ST-FMR)^{50-54}$. We observe a large damping-like SOT due to the spin-Hall effect in the bulk of Pt^{1,3}, along with an orderof-magnitude smaller field-like SOT attributed to the interfacial Rashba-Edelstein effect^{4,55}. Modeling the Pt thickness dependence of the damping-like SOT and spin-pumping damping indicates that the skew scattering^{1,3,37,56} and Dyakonov-Perel^{57,58} mechanisms

primarily govern charge-to-spin conversion and spin 89 relaxation, respectively, in epitaxial Pt. This 90 combination of mechanisms is distinct from the intrinsic 91 spin-Hall effect and Elliott-Yafet spin relaxation often 92 found in Pt-based systems^{38,39,41,42,59}. Our modeling 93 results point to a large internal spin-Hall ratio of 94 ≈ 0.8 in Pt, while a small spin-mixing conductance of 95 $\approx 1 \times 10^{14} \ \Omega^{-1} m^{-2}$ primarily limits the efficiency of the 96 damping-like SOT in the MAFO/Pt bilayer. Our work 97 demonstrates a unique material system and experimental 98 approach to uncover the mechanisms of charge-spin 99 interconversion in Pt. with minimal spurious influence 100 from the adjacent magnetic layer. 101

II. FILM GROWTH AND STRUCTURAL PROPERTIES

MAFO is a low-damping ferrimagnetic insulator with 104 a Curie temperature of ≈ 400 K, which can be grown 105 epitaxially on spinel MgAl₂O₄ (MAO) substrates⁴⁴. We 106 first deposit epitaxial MAFO films on (001)-oriented 107 single-crystal MAO by pulsed laser ablation. А 108 sintered ceramic target of stoichiometric MgAl_{0.5}Fe_{1.5}O₄ 109 is ablated in 10 mTorr of O_2 at a fluence of ≈ 2 110 J/cm^2 , repetition rate of 1 Hz, target-to-substrate 111 separation of ≈ 75 mm, and substrate temperature of 112 450 °C. No post-annealing at a higher temperature 113 is performed. All MAFO films are grown to be 13^{144} 114 nm thick, which is within the optimal thickness $\operatorname{range}^{^{145}}$ 115 that ensures coherently strained growth (i.e., without $^{\rm 146}$ 116 dislocations) and low Gilbert damping^{44,60}. Broadband¹⁴⁷ 117 ferromagnetic resonance (FMR) measurements $confirm^{148}$ 118 a Gilbert damping parameter of $\alpha\,\approx\,0.0017$ for these $^{^{149}}$ 119 MAFO films, similar to prior reports^{44,49,60}. Then, 3-19¹⁵⁰ 120 nm thick Pt layers are sputtered onto the MAFO films¹⁵¹ 121 in 3 mT orr of Ar at room temperature. To avoid ${\rm surface}^{^{152}}$ 122 damage, we used a low dc power of 15 W. 123

X-ray diffraction (XRD) measurements $indicate^{154}$ 124 epitaxy and high crystallinity of our MAFO/Pt samples.¹⁵⁵ 125 Figure 1(a) shows symmetrical scans for MAFO/Pt¹⁵⁶ 126 Strong Pt(111), Pt(222), and ¹⁵⁷ and MAFO samples. 127 MAFO(004) Bragg peaks indicate a high degree of out-128 of-plane epitaxy. The visible Laue oscillations around the 129 Pt(111) peak for the MAFO/Pt bilayers further indicate¹⁵⁸ 130 high structural quality of the Pt film. The degree of 131 crystallinity of the Pt layer is determined by performing¹⁵⁹ 132 a rocking curve measurement around the Pt(111) peak.¹⁶⁰ 133 The narrow rocking curve width of $\approx 0.4^{\circ}$ (Fig. 1(b)) 134 indicates a uniform out-of-plane orientation of Pt crystals₁₆₁ 135 with an only small mosaic spread. 136 162

The in-plane orientation of MAFO/Pt is investigated¹⁶³ by measuring asymmetrical (113) Bragg peaks for Pt,¹⁶⁴ MAFO, and MAO layers. The MAFO layer is fully¹⁶⁵ coherently strained to the MAO substrate as indicated¹⁶⁶ in the previous study⁴⁴. As can be seen from Fig. 1(c),¹⁶⁷ the MAFO layer and MAO substrate exhibit four-fold¹⁶⁸ symmetry that is expected from its cubic structures. The¹⁶⁹



FIG. 1. XRD analysis of samples. (a) XRD $2\theta/\omega$ scans of MAFO (13 nm)/Pt (5 nm), MAFO (13 nm)/Pt (3 nm), and MAFO (13 nm). (b) Rocking curve scan about the Pt (111) peak for the MAFO (13 nm)/Pt (5 nm) sample shown in (a), with FWHM $\approx 0.4^{\circ}$. (c) XRD ϕ scans on the (113) plane of the MAFO (13 nm)/Pt (5 nm) sample. Pink: MAFO. Green: MAO. (d) Lattice matching relationship between the Pt and MAFO (MAO) unit cells.

Pt(113) peak exhibits twelve maxima indicating a rather complex epitaxial relationship. Careful analysis of the Pt in-plane orientation on MAFO reveals a twinning pattern of the Pt domains, which is presented in Fig. 1(d). One can distinguish four Pt domains that match MAFO epitaxially and produce in total twelve Pt(113) peaks as shown in Fig. 1(c).

It should be noted that the epitaxial growth of Pt on MAFO is in contrast to polycrystalline or amorphous Pt on iron garnets^{33,61,62}. Further, X-ray reflectivity indicates a small roughness of <0.2 nm at the MAFO/Pt interface. Our structural characterization thus confirms that MAFO/Pt is a high-quality model system with a highly crystalline structure and sharp interface.

III. RESULTS AND DISCUSSION

A. DC-Biased Spin-Torque Ferromagnetic Resonance

The MAFO/Pt bilayers are lithographically patterned and ion-milled to 60 μ m × 10 μ m strips with the edges parallel to the in-plane (110) axes of MAFO. They are then contacted by Ti (5 nm)/Au (120 nm) ground-signalground electrodes to allow input of a microwave current for our ST-FMR measurements at room temperature, as illustrated in Fig. 2(a). We have verified that the magnetic properties of MAFO/Pt are unchanged by the lithographic patterning process (see Appendix A).



FIG. 2. ST-FMR measurement setup. (a) MAFO/Pt stack etched to a 60 μ m × 10 μ m strip. Magnetization, external field, rf field, and SOTs are shown as the arrows. The ground-signal-ground Au electrode connects MAFO/Pt to the external circuit. (b) FMR spectrum at 4 GHz. Red curve: symmetric Lorentzian contribution. Green curve: antisymmetric Lorentzian contribution. blue curve: total fit.

The microwave current in Pt induces SOTs and a 170 classical Oersted field torque on the magnetization in 171 the MAFO layer. ST-FMR spectra are obtained from 172 the rectified voltage due to magnetoresistance and spin-173 pumping signals 63,64 with field modulation 65 . Each 174 integrated ST-FMR spectrum (e.g., Fig. 2(c)) can be 175 fit with a superposition of symmetric and antisymmetric 176 Lorentzians to extract the half-width-at-half-maximum 177 linewidth ΔH and resonance field $H_{\rm res}$. 178

We use an additional dc bias current to directly extract 179 the damping-like and field-like $SOTs^{50-54}$ in MAFO/Pt. 180 This dc bias approach circumvents ambiguities of the 181 oft-used symmetric/antisymmetric Lorentzian ST-FMR²⁰⁴ 182 lineshape analysis (e.g., where the symmetric Lorentzian²⁰⁵ 183 can contain voltage signals from spin pumping and $^{\rm 206}$ 184 thermoelectric effects 63,64,66,67) and instead probes both²⁰⁷ 185 SOTs in a direct manner. In particular, the dc damping-²⁰⁸ 186 like SOT modifies the effective damping (\propto linewidth²⁰⁹ 187 ΔH) linearly with the dc bias current density $J_{\rm dc}$; the²¹⁰ 188 dc field-like torque shifts the resonance field $H_{\rm res}$ linearly 211 189 with $J_{\rm dc}$. Since all of the current flows in the Pt layer,²¹² 190 the classical Oersted field $H_{\rm Oe}$ is easily determined from $^{\rm 213}$ 191 $H_{\rm Oe}/J_{\rm dc} = t_{\rm Pt}/2$, where $t_{\rm Pt}$ is the Pt thickness, and²¹⁴ 192 subtracted from $dH_{\rm res}/dJ_{\rm dc}$ to extract the field-like SOT. $^{\rm 215}$ 193 Figure 3(a,b) shows the effect of $J_{\rm dc}$ on ΔH and²¹⁶ 194 $H_{\rm res}$. The linear dependence on current indicates²¹⁷ 195 that Joule heating contributions⁶⁸ are minimal in these²¹⁸ 196 measurements. By reversing the magnetization direction²¹⁹ 197 (external magnetic field direction), we observe a reversal²²⁰ 198 in the slope for ΔH (or $H_{\rm res}$) versus $J_{\rm dc}$ consistent with²²¹ 199 the symmetry of the $SOTs^{1,2}$. 200

From the linear slope of linewidth ΔH versus J_{dc}^{223} (Fig. 3(a)), the damping-like SOT efficiency θ_{DL} is²²⁴ readily quantified with^{50,52}

$$\left|\theta_{\rm DL}\right| = \frac{2|e|}{\hbar} \frac{(H_{\rm res} + M_{\rm eff}/2)\mu_0 M_s t_{\rm M}}{|\sin\phi|} \left|\frac{d\alpha_{\rm eff}}{dJ_{\rm dc}}\right|, \quad (1)$$



FIG. 3. Measurement of SOT efficiencies. (a) Dependence of linewidth ΔH on dc bias current density $J_{\rm dc}$ for MAFO (13 nm)/Pt (5 nm). Linewidths and linear fits under positive (blue boxes and line) and negative (red dots and line) magnetic fields are shown. (b) Resonance field change $\Delta H_{\rm res}$ as a function of $J_{\rm dc}$ for the MAFO (13 nm)/Pt (5 nm). Resonance fields and linear fits under positive (purple dots and line) and negative (green dots and line) magnetic fields are shown. The Oersted field contributions are shown as purple (positive) and green (negative) dashed lines. (c,d) Pt thickness dependence of (c) $\theta_{\rm DL}$ and (d) $\theta_{\rm FL}$ for MAFO/Pt. Note the different vertical scales for $\theta_{\rm DL}$ and $\theta_{\rm FL}$. The error bars in (c) and (d) are derived from the linear fits of linewidth and resonance field change vs. $J_{\rm dc}$.

where $\alpha_{\rm eff} = |\gamma|\Delta H/(2\pi f), |\gamma|/(2\pi) = 29 \text{ GHz/T}$ is the gyromagnetic ratio of MAFO⁴⁴, f is the microwave frequency (e.g., f = 4 GHz in Figs. 2 and 3), $t_{\rm M} =$ 13 nm is the MAFO thickness, and $\phi = 45^{\circ}$ or 225° is the in-plane magnetization orientation with respect to the current axis (x-axis in Fig. 2(a)). In applying Eq. 1, we account for the sample-to-sample variation in the saturation magnetization $M_{\rm s} = 90 - 95$ kA/m (determined by SQUID magnetometry) and the effective magnetization $\mu_0 M_{\rm eff} = 1.2 - 1.5$ T (determined by fitting the frequency dependence of resonance field⁴⁴). The large effective magnetization of epitaxial MAFO arises due to significant magnetoelastic easy-plane anisotropy⁴⁴.

The $t_{\rm Pt}$ dependence of $\theta_{\rm DL}$ is summarized in Fig. 3(c). The increase in $\theta_{\rm DL}$ with $t_{\rm Pt}$ up to ≈ 5 nm (Fig. 3(c)) suggests that the spin-Hall effect in the Pt bulk is the dominant source of the damping-like SOT^{6,38}. The decrease in $\theta_{\rm DL}$ at higher $t_{\rm Pt}$ might seem surprising, but a similar trend has been observed in prior experiments³⁸.

We also quantify the field-like SOT efficiency $\theta_{\rm FL}$ from the linear shift of $H_{\rm res}$ with $J_{\rm dc}$ (Fig. 3(b)) and subtracting the Oersted field contribution^{19,52}

$$\theta_{\rm FL}| = \frac{2|e|}{\hbar} \frac{\mu_0 M_s t_{\rm M}}{|\sin\phi|} \left(\left| \frac{dH_{\rm res}}{dJ_{\rm dc}} \right| - \frac{t_{\rm Pt}}{2} |\sin\phi| \right), \qquad (2)$$

where the term proportional to $t_{\rm Pt}$ accounts for the₂₈₁ 226 Oersted field. As shown in Fig. 3(d), the constant value₂₈₂ 227 of $\theta_{\rm FL}$ with Pt thickness implies that the field-like SOT₂₈₃ 228 arises from the MAFO/Pt interface, e.g., via the Rashba-284 229 Edelestein effect^{4,55,69}. However, this field-like SOT is₂₈₅ 230 weak, i.e., similar in magnitude to the Oersted field286 231 (Fig. 3(b)). Indeed, we find that $\theta_{\rm FL} \sim 0.01$ is about₂₈₇ 232 an order of magnitude smaller than $\theta_{\rm DL}$. 233

Based on the dominance of the strongly $t_{\rm Pt}$ -dependent²⁸⁹ 234 damping-like SOT over the $t_{\rm Pt}$ -independent field-like 235 SOT, we conclude that charge-spin interconversion²⁹⁰ 236 processes in the bulk of Pt dominate over those at the²⁹¹ 237 MAFO/Pt interface. It has been proposed that a field-²⁹² 238 like SOT could arise from the bulk of Pt in the presence²⁹³ 239 of an imaginary part of the spin-mixing conductance,²⁹⁴ 240 $\operatorname{Im}[G_{\uparrow\downarrow}]^{70}$. A substantial $\operatorname{Im}[G_{\uparrow\downarrow}]$ would manifest in $a_{_{295}}$ 241 shift in the gyromagnetic ratio (or g-factor) in MAFO₂₉₆ 242 with the addition of a Pt overlayer⁷¹. Since such a shift 243 is not observed, we rule out this scenario of a field-like297 244 SOT of "bulk" origin. In other words, the damping-like₂₉₈ 245 torque is the predominant type of SOT that arises from 246 the bulk of Pt. Therefore, in the following sections, we²⁹⁶ 247 use the damping-like SOT as a measure of charge-to-spin³⁰⁰ 248 conversion in Pt. 301 249

B. Modeling the Pt-Thickness Dependence of the 304 Spin-Pumping Damping and Damping-Like Spin-Orbit Torque 305

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We employ a model similar to the one used by Berger³⁰⁷ 253 et al.²³ to assess charge-spin interconversion mechanisms³⁰⁸ 254 This model estimates key parameters that³⁰⁹ in Pt. 255 govern charge-spin interconversion by fitting the $t_{\rm Pt}^{310}$ 256 dependence of two experimentally measured quantities:³¹¹ 257 the Gilbert damping parameter α and the damping-like₃₁₂ 258 SOT conductivity σ_{DL} . 259 313 We have measured the damping parameter α by₃₁₄ 260 coplanar-waveguide-based FMR and ST-FMR, which 261 yield consistent results for unpatterned and patterned₃₁₅ 262

MAFO/Pt (see Appendix A). As can be seen in316 263 Fig. 4(b,c), MAFO/Pt bilayers exhibit higher α than₃₁₇ 264 the bare MAFO films with $t_{\rm Pt} = 0$. In Sec. III C, we 265 attribute this damping enhancement to spin pumping⁷¹,³¹⁸ 266 i.e., due to the loss of spin angular momentum pumped³¹⁹ 267 from the resonantly excited MAFO layer to the adjacent³²⁰ 268 spin sink layer of Pt. In Sec. III D, we further consider $_{321}$ 269 an additional contribution to the enhancement of α due₃₂₂ 270 to spin-memory loss or two-magnon scattering. 271

To parameterize the strength of the damping-like 272 SOT, we employ the "SOT conductivity," $\sigma_{\rm DL} =_{324}$ 273 $\theta_{\rm DL}/\rho_{\rm Pt}$. Normalizing $\theta_{\rm DL}$ by the Pt resistivity $\rho_{\rm Pt}$ makes³²⁵ 274 explicit the relationship between the SOT and $\mathrm{electronic}_{326}$ 275 transport. We also remark that $\sigma_{\rm DL}$ is equivalent to the $_{^{327}}$ 276 SOT efficiency per unit electric field ξ_{DL}^E in Refs. 38, 42.328 277 The $t_{\rm Pt}$ dependence of $\rho_{\rm Pt}$ (fit curve in Fig. 4(a)) is₃₂₉ 278 interpolated by using the empirical model outlined in₃₃₀ 279 Appendix D. 331 280

In contrast to Ref. 23 that studies FM/Pt bilayers where electronic spin transport in the FM can generally yield additional effects that impact SOTs, our MAFO/Pt system restricts the source of SOTs to Pt. We are therefore able to examine the spin-Hall effect of Pt without any complications from an electrically conductive FM.

To model our experimental results, we consider two types of spin-Hall effect^{1,3}:

- the *intrinsic* mechanism, where the internal spin-Hall ratio $\theta_{\rm SH}$ i.e., the charge-to-spin conversion efficiency within the Pt layer itself is proportional to $\rho_{\rm Pt}$, with a constant internal spin-Hall conductivity $\sigma_{\rm SH} = \theta_{\rm SH}/\rho_{\rm Pt}$;
- the skew scattering mechanism, where $\theta_{\rm SH}$ is independent of $\rho_{\rm Pt}$.

We also consider two mechanisms of spin relaxation that govern the spin diffusion length λ_s in Pt^{35,57,58}:

- Elliott-Yafet (EY) spin relaxation, where spins depolarize *during* scattering such that $\lambda_{\rm s}$ scales inversely with $\rho_{\rm Pt}$, i.e., $\lambda_{\rm s} = \lambda_{\rm s}^{\rm bulk} \rho_{\rm Pt}^{\rm bulk} / \rho_{\rm Pt}$;
- Dyakonov-Perel (DP) spin relaxation, where spins depolarize *between* scattering events such that λ_s is independent of $\rho_{\rm Pt}$ (as outlined by Boone *et al.*³⁵).

Thus, we model four combinations of the abovelisted charge-to-spin conversion and spin relaxation mechanisms, as shown in Fig. 4(b-e).

Similar to Ref. 23, we self-consistently fit α vs. $t_{\rm Pt}$ (Fig. 4(b,c)) and $\sigma_{\rm DL}$ vs. $t_{\rm Pt}$ (Fig. 4(d,e)) by using standard spin diffusion models^{6,35,71}, as elaborated in Appendix **E**, with four free parameters:

- spin diffusion length λ_s in the case of DP spin relaxation, or its bulk-limit value λ_s^{bulk} in the case of EY spin relaxation;
- internal spin-Hall ratio $\theta_{\rm SH}$ of Pt in the case of skew scattering, or its bulk-limit value $\theta_{\rm SH}^{\rm bulk} = \sigma_{\rm SH} \rho_{\rm Pt}^{\rm bulk}$ in the case of intrinsic spin-Hall effect;
- real part of the spin-mixing conductance $G_{\uparrow\downarrow}$ at the MAFO/Pt interface, neglecting the imaginary part as justified in Sec. III A;
- effective damping enhancement α_{SML} due to interfacial spin-memory loss or two-magnon scattering, as discussed in detail in Sec. III D.

A key assumption here is that the spin-pumping damping and damping-like SOT share the same values of λ_s , $G_{\uparrow\downarrow}$, and $\alpha_{\rm SML}$. This is justified by the enforcement of Onsager reciprocity on the charge-spin interconversion processes of spin pumping and SOT^{23,72}. We also assume a negligible interfacial contribution to the spin-Hall effect in Pt⁷³, which would yield a finite value of $\sigma_{\rm DL}$ when $t_{\rm Pt}$ is extrapolated to zero. Indeed, as shown in Fig. 4, the $_{332}$ $t_{\rm Pt}$ dependence of $\sigma_{\rm DL}$ is adequately modeled without $_{333}$ incorporating the interfacial spin-Hall effect.

For simplicity, we first proceed by setting $\alpha_{\rm SML} = 0$ in Sec. III C. This is a reasonable assumption considering that interfacial spin-memory loss is likely much weaker in MAFO/Pt than in all-metallic FM/Pt systems²⁰⁻²⁶. Nevertheless, we also discuss the consequence of $\alpha_{\rm SML} >$ 0 in Sec. III D.

C. Mechanisms and Parameters for Charge-Spin Interconversion in Pt: Zero Spin-Memory Loss

Our modeling results under the assumption of zero 342 spin-memory loss are summarized in Fig. 4 and Table I. 343 We find that the combination of skew scattering and 344 DP spin relaxation (solid green curves in Fig. 4(c,e)) 345 best reproduces the $t_{\rm Pt}$ dependence of both α and $\sigma_{\rm DL}$. 346 Although this observation does not necessarily rule out 347 the coexistence of other mechanisms^{23,43,57,58}, it suggests 348 the dominance of the skew scattering + DP combination 349 in the epitaxial Pt film. Skew scattering in highly 350 crystalline Pt is consistent with what is expected for 351 "superclean" Pt, in contrast to the intrinsic spin-Hall 352 effect that is dominant in "moderately dirty" Pt³⁷. 353

The dominance of DP spin relaxation – i.e., 354 spin depolarization (dephasing) from precession about 355 effective spin-orbit fields - is perhaps surprising, since 356 it is usually thought to be inactive in centrosymmetric 357 metals (e.g., Pt). Indeed, in the context of spin transport 358 in Pt, it is typical to assume EY spin relaxation where 359 spins depolarize when their carriers (e.g., electrons) 360 are scattered^{38,39,41,42,59}. However, a recent quantum 361 transport study indicates the dominance of DP spin 362 relaxation in crystalline Pt⁵⁷, which is in line with our 363 conclusion here. Possible origins of the DP mechanism 364 include symmetry breaking between the substrate and 365 the surface of the crystalline Pt film^{74} and strong $\mathrm{spin}^{^{387}}$ 366 mixing caused by the distinct band structure (large spin³⁸⁸ 367 Berry curvature) of Pt⁵⁸. DP spin relaxation may also³⁸⁹ 368 be more pronounced when proximity-induced magnetism³⁹⁰ 369 in Pt is negligible⁵⁸, as is likely the case for Pt interfaced³⁹¹ 370 with the insulating MAFO⁷⁵. We also note that DP³⁹² 371 spin relaxation has been previously used to model the $^{\scriptscriptstyle 393}$ 372 angular dependence of spin-Hall magnetoresistance^{76,77394} 373 in MAFO/Pt⁴⁹. The combination of skew scattering and³⁹⁵ 374 DP spin relaxation, though not reported in prior SOT³⁹⁶ 375 experiments, is reasonable for MAFO/Pt. 376

We now discuss the parameters quantified with our model, as summarized in the "skew scatt.+DP" row in Table I. The value of $G_{\uparrow\downarrow} \approx 1 \times 10^{14} \ \Omega^{-1} \mathrm{m}^{-2}$ is comparable to those previously reported for FI/Pt interfaces^{33,49,78,79}, and $\lambda_{\rm s} \approx 3$ nm is in the intermediate regime of $\lambda_{\rm s} \sim 1-10$ nm in prior reports on Pt^{20,23,28-43}.

We find a large internal spin-Hall ratio of $\theta_{\rm SH} \approx$ 0.8. While a few studies have alluded to $\theta_{\rm SH}$ on the order of unity in transition metals^{23,33,42,80,81}, our experimental study is the first to derive such a large

FIG. 4. Pt thickness dependence of: (a) resistivity $\rho_{\rm Pt}$, with the solid curve showing the fit obtained with the model described in Appendix D; (b,c) Gilbert damping parameter α , with the black horizontal dashed line indicating the average damping parameter of uncapped MAFO; (d.e) damping-like SOT conductivity $\sigma_{\rm DL}$. Modeling results based on Elliott-Yafet (EY) spin relaxation are shown in (b,d), whereas those based on Dyakonov-Perel (DP) spin relaxation are shown in (c,e). The dotted curves are based on the intrinsic spin-Hall effect, and the solid curves are based on skew scattering. The modeling results in (b-e) are obtained by assuming zero spinmemory loss and two-magnon scattering (i.e., $\alpha_{\rm SML} = 0$). In (b-e), the error bars are comparable to or smaller than the symbol size and are derived from the linear fits of FMR linewidth vs. frequency (b,c) and dc bias current density (d,e).

value in Pt without uncertainties from a conductive $FM^{23,42,80,81}$ or microwave calibration^{23,33,80,81}. Our finding of θ_{SH} approaching unity is also distinct from previously reported spin-Hall ratios < 0.1 in all-epitaxial $FM/Pt^{59,82-85}$. This discrepancy may be partially explained by the conductive FM reducing the apparent charge-to-spin conversion efficiency, or by the indirect nature of the measurements in these reports. With direct SOT measurements on the model-system MAFO/Pt bilayers, our study points to the possibility of a strong

model	$\alpha_{\rm SML}$	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s}~({\rm nm})$	θ_{SH}
intrinsic + EY	0	2.5×10^{14}	21	0.21
skew scatt. $+ EY$	0	1.1×10^{14}	4.7	1.2
intrinsic + DP	0	1.8×10^{14}	5.7	0.25
skew scatt. $+$ DP	0	$1.3 imes10^{14}$	3.3	0.83

TABLE I. Parameters for the modeled curves in Fig. 4. For charge-to-spin conversion = intrinsic (for spin relaxation = EY), $\theta_{\rm SH}$ ($\lambda_{\rm s}$) is the value at $\rho_{\rm Pt} = \rho_{\rm Pt}^{\rm bulk} = 1.1 \times 10^{-7} \ \Omega {\rm m}$.



397 spin-Hall effect in highly crystalline Pt in the skew-448

³⁹⁸ scattering regime, where the charge-to-spin conversion⁴⁴⁹

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³⁹⁹ efficiency could be greater than the limit set by the

 $_{400}$ intrinsic spin-Hall effect 1,3,37,42 .

D. Mechanisms and Parameters for Charge-Spin Interconversion in Pt: Finite Spin-Memory Loss

A natural question at this point is how finite spin-459 403 memory loss at the MAFO/Pt interface impacts the₄₆₀ 404 parameters quantified in our modeling. Moreover, while₄₆₁ 405 bare MAFO exhibits negligible two-magnon scattering⁴⁴,₄₆₂ 406 an overlayer (Pt in this case) on top of MAFO may₄₆₃ 407 give rise to two-magnon scattering at the interface⁸⁶.464 408 Both spin-memory loss and two-magnon scattering would₄₆₅ 409 have the same consequence in that they enhance the $_{466}$ 410 apparent damping parameter, α , independent of $t_{\rm Pt}^{23,87}$. 411 We therefore model spin-memory loss and two-magnon₄₆₈ 412 scattering with a phenomenological parameter, $\alpha_{\rm SML}$. 413 460

Figure 5 and Table II summarize our modeling results⁴⁷⁰ 414 incorporating finite spin-memory loss or two-magnon⁴⁷¹ 415 scattering (i.e., $\alpha_{SML} > 0$). Finite α_{SML} does not⁴⁷² 416 improve the fit quality in $\sigma_{\rm DL}$ vs. $t_{\rm Pt}$ of the EY models⁴⁷³ 417 (Fig. 5(a,b)). By contrast, the fit quality is improved for⁴⁷⁴ 418 the DP models with increasing $\alpha_{\rm SML}$, particularly in α^{475} 419 vs. $t_{\rm Pt}$ (Fig. 5(c,d)). We therefore focus on the results⁴⁷⁶ 420 for the DP models. 477 421

As shown in Table II, increasing $\alpha_{\rm SML}$ significantly₄₇₉ 422 decreases $G_{\uparrow\downarrow}$, consistent with the reduced share of $\text{spin}_{_{480}}$ 423 pumping in the damping enhancement. To compensate $_{_{481}}$ 424 for the smaller $G_{\uparrow\downarrow}$, the internal spin-Hall ratio $\theta_{\rm SH_{482}}$ 425 must increase to reproduce the $t_{\rm Pt}$ dependence of $\sigma_{\rm DL_{483}}$ 426 (Ref. 87). In the "skew scattering + \overline{DP} " model, shown 427 to be most plausible in Sec. III C, θ_{SH} increases to values 428 exceeding unity with finite α_{SML} . At a sufficiently large $^{*\circ}_{496}$ 429 $\alpha_{\rm SML}$ of $\gtrsim 0.002$, the "intrinsic + DP" model appears to 430 becomes plausible (see Fig. 5(c)), but this scenario also₄₈₈ 431 vields $\theta_{\rm SH} > 1$. 432

In both of the above DP scenarios, substantial spin-490 433 memory loss or two-magnon scattering apparently leads⁴⁹¹ 434 to an unphysically large value of internal spin-Hall ratio⁴⁹² 435 in Pt exceeding unity. It is then reasonable to conclude⁴⁹³ 436 that spin-memory loss and two-magnon scattering is494 437 negligibly small in epitaxial MAFO/Pt. This is in₄₉₅ 438 stark contrast to the large spin-memory loss deduced for₄₉₆ 439 all-metallic FM/Pt bilayers²³. The small spin-memory⁴⁹⁷ 440 loss in MAFO/Pt also suggests fundamentally different498 441 spin-transport mechanisms between FM/Pt and FI/Pt499 442 systems, which could be exploited for more efficient SOT₅₀₀ 443 devices in the future. Our finding motivates further⁵⁰¹ 444 studies to test whether the negligible spin-memory loss⁵⁰² 445 is due to the crystalline growth or due to the absence of 503 446 proximity-induced magnetism. 504 447

E. Implications of the Large Internal Spin-Hall Ratio in Pt

From our analysis in Sec. IIIC, we have arrived at a large internal spin-Hall ratio of $\theta_{\rm SH} \approx 0.8$ in epitaxial Pt. Yet, the observed spin-torque efficiency of $\theta_{\rm DL} \lesssim 0.15$ implies an interfacial spin transparency ratio $\theta_{\rm DL}/\theta_{\rm SH}$ of \lesssim 0.2. In other words, at most only 20% of the spin accumulation generated by the spin-Hall effect in Pt is transferred to the magnetic MAFO layer as the damping-like SOT. The origin of this inefficient spin transfer, according to the spin diffusion model employed here, is the small spin-mixing conductance of $G_{\uparrow\downarrow} \sim 1 \times 10^{14} \ \Omega^{-1} \mathrm{m}^{-1}$, which is several times lower than $G_{\uparrow\downarrow}$ computationally predicted for FM/Pt interfaces^{88–90}. The small $G_{\uparrow\downarrow}$ results in a substantial spin backflow^{87,91} that prevents efficient transmission of spin angular momentum across the MAFO/Pt interface. We emphasize that spin-memory loss is likely negligible at the MAFO/Pt interface (see Sec. III D) and hence not responsible for the inefficient spin transfer.

There may be an opportunity to enhance the spin transparency – and hence the SOT efficiency – by engineering the interface. One possible approach is to use an ultrathin insertion layer of NiO, which has been reported to increase the spin transparency ratio to essentially unity in FM/Pt systems⁹¹. However, it remains to be explored whether the ultrathin NiO insertion layer can increase the spin transparency without causing substantial interfacial spin scattering⁸⁶ in FI/Pt bilayers. An increased spin transparency (via enhanced $G_{\uparrow\downarrow}$) also leads to higher spin-pumping damping^{71,92}, which may not be desirable for applications driven by precessional switching or auto-oscillations.

Another striking implication of the large internal spin-Hall ratio is a large maximum spin-Hall conductivity $\sigma_{\rm SH} = \theta_{\rm SH}/\rho_{\rm Pt}^{\rm bulk}$ of $\approx 8 \times 10^6 \ \Omega^{-1} {\rm m}^{-1}$, which is at least an order of magnitude greater than $\sigma_{\rm SH} \sim 10^4 - 10^5 \ \Omega^{-1} {\rm m}^{-1}$ typically predicted from band structure calculations^{93–97}. It should be noted, however, that these calculations are for the *intrinsic* spin-Hall effect, whereas our experimental data are best captured by the *extrinsic* spin-Hall effect of skew scattering. We thus speculate that this difference in mechanism could account for the discrepancy in $\sigma_{\rm SH}$ derived from our experimental work and from prior calculations.

Finally, we comment on remaining open fundamental questions. Comparing MAFO/epitaxial-Pt and MAFO/*polycrystalline*-Pt could reveal the critical role of crystallinity in charge-spin interconversion, spin relaxation, and the internal spin-Hall ratio in Pt. This comparison study is precluded here due to the difficulty in growing polycrystalline Pt on MAFO; Pt has a strong tendency to be epitaxial on MAFO due to the excellent lattice match, even when Pt is sputter-deposited with the substrate at room temperature. Moreover, while the epitaxial Pt film on MAFO is single-crystalline in the sense that its out-of-plane crytallographic orientation



FIG. 5. Pt thickness dependence of the Gilbert damping parameter α and the damping-like SOT conductivity σ_{DL} , taking into account different strengths of spin-memory loss or two-magnon scattering (parameterized by α_{SML}), for the four combinations of charge-to-spin conversion and spin relaxation mechanisms: (a) intrinsic spin-Hall effect + Elliott-Yafet (EY), (b) skew scattering + EY, (c) intrinsic spin-Hall effect + Dyakonov-Perel (DP), and (d) skew scattering + DP. The error bars are comparable to or smaller than the symbol size; they are derived from the linear fits of FMR linewidth vs. frequency (for α) and ST-FMR linewidth vs. dc bias current density (for σ_{DL}).

	intrinsic + EY			skew scatt. $+ EY$			intrinsic + DP			skew scatt. $+$ DP		
$\alpha_{\rm SML}$	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s}~({\rm nm})$	$\theta_{\rm SH}$	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s}~({\rm nm})$	$\theta_{\rm SH}$	$G_{\uparrow\downarrow} (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s}~({\rm nm})$	$\theta_{\rm SH}$	$G_{\uparrow\downarrow} (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s}~({\rm nm})$	$\theta_{\rm SH}$
0	2.5×10^{14}	21	0.21	1.1×10^{14}	4.7	1.2	1.8×10^{14}	5.7	0.25	$1.3 imes10^{14}$	3.3	0.83
0.001	1.5×10^{14}	23	0.40	$0.7 imes 10^{14}$	4.7	2.7	1.0×10^{14}	6.2	0.53	0.9×10^{14}	3.6	1.5
0.002	$0.6 imes 10^{14}$	26	1.3	0.4×10^{14}	5.0	7.5	0.6×10^{14}	7.1	1.2	0.5×10^{14}	3.8	4.1

TABLE II. Parameters for the modeled curves in Fig. 5. For charge-to-spin conversion = intrinsic (for spin relaxation = EY), $\theta_{\rm SH}$ ($\lambda_{\rm s}$) is the value at $\rho_{\rm Pt} = \rho_{\rm Pt}^{\rm bulk} = 1.1 \times 10^{-7} \ \Omega {\rm m}$.

is exclusively (111), it is yet unclear how the twin528
domains (discussed in Sec. II) influence charge-spin529
interconversion in Pt. Determining the impact of 530
microstructure on spin-Hall and related effects in Pt
remains a subject of future work.
Furthermore, we acknowledge the possibility that 532
the model appleued in our present study (arthined in 533

the model employed in our present study (outlined in⁵³³ 511 Sec. III B and Appendix E) is incomplete. For instance,⁵³⁴ 512 we have assumed that the damping-like SOT and spin-535 513 pumping damping are reciprocal phenomena with shared⁵³⁶ 514 $G_{\uparrow\downarrow}$ and $\lambda_{\rm s}$. This commonly made assumption²³ – with⁵³⁷ 515 prior studies suggesting that such reciprocity holds^{46,52538} 516 - is necessary for constraining the fitting of the limited 517 number of experimental data points. Further studies are⁵³⁹ 518 required for confirming whether the damping-like $\mathrm{SOT}^{^{540}}$ 519 and spin-pumping damping can be captured by the same $^{\rm 541}$ 520 542 values of $G_{\uparrow\downarrow}$ and λ_s . 521 543

IV. SUMMARY

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We have measured SOTs in a low-damping, epitaxial⁵⁴⁷ insulating spinel ferrite (MgAl_{0.5}Fe_{1.5}O₄, MAFO)⁵⁴⁸ interfaced with epitaxial Pt. This model-system bilayer⁵⁴⁹ enables a unique opportunity to examine charge-spin⁵⁵⁰ interconversion mechanisms in highly crystalline Pt,⁵⁵¹ while eliminating complications from electronic transport in (or hybridization with) a magnetic metal. Our key findings are as follow.

- 1. Charge-to-spin conversion in Pt appears to be primarily a bulk effect, rather than an interfacial effect. A sizable damping-like SOT, which depends strongly on the Pt thickness, arises from the spin-Hall effect within Pt. An order-of-magnitude smaller field-like SOT, independent of the Pt thickness, is attributed to the Rashba-Edelstein effect at the MAFO/Pt interface.
- 2. In crystalline Pt, the extrinsic spin-Hall effect of skew scattering and the Dyakonov-Perel spin relaxation mechanism likely dominate. This is in contrast to the combination of the intrinsic spin-Hall effect and Elliott-Yafet spin relaxation typically reported for Pt-based systems.
- 3. The internal spin-Hall ratio deduced for crystalline Pt is large, i.e., $\theta_{\rm SH} \approx 0.8$. While a similar magnitude has been suggested before from experiments on all-metallic FM/Pt bilayers, greater confidence may be placed in our result owing to the cleanliness of the MAFO/Pt system, the direct nature of the SOT measurement method, and

- the self-consistent modeling of the SOT and spin-595 552 pumping damping. 553
- 4. Spin-memory loss appears to be minimal in 554 the epitaxial MAFO/Pt system. Modeled⁵⁹⁷ 555 scenarios with substantial spin-memory loss yield598 556 unphysically large internal spin-Hall ratios that⁵⁹⁹ 557 exceed unity. 600 558
- 5. The factor limiting the damping-like SOT efficiency₆₀₂ 559 in the MAFO/Pt bilayer, despite the apparently₆₀₃ 560 large $\theta_{\rm SH}$, is the small spin-mixing conductance 561 $G_{\uparrow\downarrow}$. Enhancing $G_{\uparrow\downarrow}$ while keeping spin-memory $_{605}^{\circ\circ\circ}$ 562 loss minimal could increase the SOT efficiency. 563

of⁶⁰⁷ Overall, our work demonstrates the utility 564 ${\rm heterostructures}^{608}$ epitaxial insulating-ferrite-based 565 for understanding spin-transport phenomena in the $^{\rm 609}$ 566 widely-used spin-Hall metal of Pt, as well as for⁶¹⁰ 567 611 engineering materials for efficient spintronic devices. 568 612

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building the ST-FMR system. 594

Appendix A: Effect of Sample Processing on the Magnetic Properties of MAFO

We have used both broadband FMR (i.e., with unpatterned films placed on a coplanar waveguide, see Ref. 44 for details) and ST-FMR (i.e., with microwave current injected through patterned $10-\mu$ m-wide strips) to measure the frequency dependence of FMR linewidth and resonance field. Thus, it is important to confirm the consistency of measurements between the two techniques.

Figure 6(a) plots the linewidth vs. frequency data for a bare MAFO film (13 nm) that we started with, the MAFO (13 nm)/Pt (5 nm) film after Pt deposition, and the ST-FMR pattern with MAFO (13 nm)/Pt (5 nm) after the microfabrication processes. The damping constants of the MAFO/Pt unpatterned film and patterned strip are essentially identical, confirming the consistency of the broadband FMR and ST-FMR measurements.

We also show in Fig. 6(b) that the frequency dependence of resonance field is unaltered before and after microfabrication. The fit using the Kittel equation⁴⁴ indicates negligible ($\ll 5\%$) difference in the effective magnetization (dominated by mangnetoelastic easy-plane anisotropy) and gyromagnetic ratio for the unpatterned film and patterned strip. The results in Fig. 6 therefore confirm that the microfabrication processes have little to no effect on the essential magnetic properties of MAFO/Pt.



Frequency dependence of (a) linewidth and (b) FIG. 6. resonance field a bare MAFO film (13 nm), unpatterned MAFO (13 nm)/Pt (5 nm) film, and patterned MAFO (13 nm)/Pt (5 nm) ST-FMR strip.

Appendix B: Microwave Power Dependence of the 623 Spin-Torque Ferromagnetic Resonance Signal 624

Figure 7 shows the dependence of the ST-FMR signal 625 amplitude on the microwave power. The ST-FMR 626 voltage increases linearly with the microwave power, 627 indicating that all the measurements are done in the 628 linear regime in this present study. 629



(a) Exemplary ST-FMR spectra at $\operatorname{different}^{649}$ FIG. 7. microwave powers. (b) ST-FMR amplitude vs. microwave 650 651 power.

Appendix C: Frequency Dependence of the **Spin-Orbit Torque Efficiencies**

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FIG. 8. Dependence of SOTs in MAFO (13 nm)/ Pt (5 nm). (a) Damping-like torque efficiency θ_{DL} . (b) Field-like torque⁶⁵⁷ efficiency $\theta_{\rm FL}$.

We have carried out a frequency dependence study of 632 damping-like and field-like SOT efficiencies. The dc-633 biased ST-FMR method is used to extract each data 634 point. Figure 8 shows that both the damping-like and 635 field-like SOT efficiencies are nearly constant across the 636 frequency range of 3-8 GHz. This verifies that the $\mathrm{SOT}^{\scriptscriptstyle 658}$ 637 efficiencies are independent of the microwave frequency. $^{\rm 659}$ 638

Appendix D: Model for the Pt Thickness 639 **Dependence of Resistivity** 640

We model the Pt thickness dependence of resistivity 666 641 $\rho_{\rm Pt}$ by using an approach similar to that reported by 667 642 Berger et al.²³. This model takes into account the668 643

conductivity σ as a function of position along the film thickness axis z, expressed as the sum of bulk and interfacial contributions,

$$\sigma(z) = \frac{1 - \exp\left(-\frac{z}{L}\right)}{\rho_{\rm Pt}^{\rm bulk}} + \frac{\exp\left(-\frac{z}{L}\right)}{\rho_{\rm int}},\qquad({\rm D1})$$

where $\rho_{\rm Pt}^{\rm bulk} = 1.1 \times 10^{-7} \ \Omega^{-1} {\rm m}^{-1}$ is the bulk resistivity of Pt, $\rho_{\rm int}$ is the interfacial resistivity, and L is an empirical characteristic length scale capturing the decay of the interfacial contribution to resistivity. The resistivity of the Pt film with thickness $t_{\rm Pt}$ is then given by,

$$\rho_{\rm Pt}(t_{\rm Pt}) = \left(\frac{1}{t_{\rm Pt}} \int_0^{t_{\rm Pt}} \sigma(z) dz\right)^{-1} = \frac{\rho_{\rm Pt}^{\rm bulk}}{1 + \frac{L}{t_{\rm Pt}} \left(\frac{\rho_{\rm Pt}^{\rm bulk}}{\rho_{\rm int}} - 1\right) \left(1 - \exp\left(-\frac{t_{\rm Pt}}{L}\right)\right)}.$$
 (D2)

The fit curve for the experimentally measured $t_{\rm Pt}$ dependence of $\rho_{\rm Pt}$ (Fig. 4(a)) is obtained with Eq. D2 with $\rho_{\rm int} = 1.3 \times 10^{-6} \ \Omega^{-1} {\rm m}^{-1}$ and L = 10. nm.

Appendix E: Equations for the Pt Thickness **Dependence of** α and $\sigma_{\rm DL}$

We fit the $t_{\rm Pt}$ dependence of α with³⁵

$$\alpha(t_{\rm Pt}) = \alpha_0 + \alpha_{\rm SML} + \alpha_{\rm SP}$$

= $\alpha_0 + \alpha_{\rm SML} + \frac{g\mu_{\rm B}\hbar}{2e^2M_{\rm s}t_{\rm M}} \left[\frac{1}{G_{\uparrow\downarrow}} + 2\rho_{\rm Pt}\lambda_{\rm s} \coth\left(\frac{t_{\rm Pt}}{\lambda_{\rm s}}\right)\right]^{-1},$
(E1)

0.0017 is the mean value for the where α_0 = five bare MAFO films $(t_{\rm Pt} = 0)$ prior to Pt deposition for the MAFO/Pt bilayers, $\alpha_{\rm SP}$ is the spin-pumping contribution to Gilbert damping, $\alpha_{\rm SML}$ is the phenomenological parameter capturing the $t_{\rm Pt}$ independent enhancement of damping (from interfacial spin-memory loss or two-magnon scattering), g = 2.05 is the spectroscopic g-factor⁴⁴, $M_{\rm s} = 93$ kA/m is the mean value of the saturation magnetization of MAFO used in this study, and $t_{\rm M} = 13$ nm is the thickness of MAFO.

The $t_{\rm Pt}$ dependence of $\sigma_{\rm DL}$ is fit with⁶

$$\sigma_{\rm DL}(t_{\rm Pt}) = \frac{\theta_{\rm DL}}{\rho_{\rm Pt}} = \frac{\theta_{\rm SH}}{\rho_{\rm Pt}} \left\{ \frac{(1 - e^{-t_{\rm Pt}/\lambda_{\rm s}})^2}{(1 + e^{-2t_{\rm Pt}/\lambda_{\rm s}})} \frac{G'}{G' + \tanh^2\left(\frac{t_{\rm Pt}}{\lambda_{\rm s}}\right)} \right\} \frac{\alpha_{\rm SP}}{\alpha_{\rm SML} + \alpha_{\rm SP}},\tag{E2}$$

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where $G' = G_{\uparrow\downarrow} 2\rho_{\rm Pt} \lambda_{\rm s} \tanh(t_{\rm Pt}/\lambda_{\rm s})$. We also remark₆₇₀ that $\rho_{\rm Pt}$ is dependent on $t_{\rm Pt}$ as given by Eq. D2.



FIG. 9. Exemplary fit results for the Pt thickness dependence of the Gilbert damping parameter α and the damping-like SOT conductivity σ_{DL} with several values of $G_{\uparrow\downarrow}$. The solid curves (parameterized by the values in bold font in Table III) are the curves shown in Fig. 4. The error bars are comparable to or smaller than the symbol size; they are derived from the linear fits of FMR linewidth vs. frequency (for α) and ST-FMR linewidth vs. dc bias current density (for σ_{DL}).

	intrinsic $+ EY$			skew scatt. $+$ EY			intrinsic $+$ DP			skew scatt. $+$ DP		
	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s} \ ({\rm nm})$	θ_{SH}	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s} \ ({\rm nm})$	θ_{SH}	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s} \ ({\rm nm})$	θ_{SH}	$G_{\uparrow\downarrow} \ (\Omega^{-1} \mathrm{m}^{-2})$	$\lambda_{\rm s} \ ({\rm nm})$	$\theta_{\rm SH}$
(a)	1.5×10^{14}	16	0.24	1.0×10^{14}	0.1	48	1.2×10^{14}	5.7	0.32	1.1×10^{14}	3.2	0.96
(b)	2.0×10^{14}	19	0.22	1.1×10^{14}	4.7	1.2	1.6×10^{14}	5.6	0.27	1.2×10^{14}	3.3	0.89
(c)	2.5×10^{14}	21	0.21	1.2×10^{14}	5.7	0.96	$1.8{ imes}10^{14}$	5.7	0.25	$1.3{ imes}10^{14}$	3.3	0.83
(d)	3.0×10^{14}	22	0.20	1.3×10^{14}	6.7	0.80	2.0×10^{14}	5.8	0.23	1.4×10^{14}	3.4	0.77
(e)	5.0×10^{14}	24	0.19	1.4×10^{14}	6.1	0.68	2.5×10^{14}	6.1	0.21	1.5×10^{14}	3.5	0.72

TABLE III. Parameters for the modeled curves in Fig. 9. For charge-to-spin conversion = intrinsic (for spin relaxation = EY), $\theta_{\rm SH}$ ($\lambda_{\rm s}$) is the value at $\rho_{\rm Pt} = \rho_{\rm Pt}^{\rm bulk} = 1.1 \times 10^{-7} \ \Omega m$.

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⁶⁷¹ By using Equations E1 and E2, along with the⁶⁷² interpolated ρ_{Pt} (Eq. D2), we find the values of $G_{\uparrow\downarrow,675}$ ⁶⁷³ λ_{s} , and θ_{SH} that adequately capture the experimentally⁶⁷⁶ measured $\alpha(t_{\rm Pt})$ and $\sigma_{\rm DL}(t_{\rm Pt})$. In particular, $\alpha(t_{\rm Pt})$ and $\sigma_{\rm DL}(t_{\rm Pt})$ are fit simultaneously⁹⁸ with a series of fixed values for $G_{\uparrow\downarrow}$ (e.g., Figs. 9 and Table III).

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