Letter

Enhancing Spin-Orbit Torque by Strong Interfacial Scattering From Ultrathin Insertion Layers

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Increasing dampinglike spin-orbit torque (SOT) is of fundamental importance for enabling new research into spintronics phenomena and also technologically urgent for advancing low-power spin-torque memory, logic, and oscillator devices. Here, we demonstrate that enhancing interfacial scattering by inserting ultrathin layers within spin Hall metals with intrinsic or side-jump mechanisms can significantly enhance the spin Hall ratio. The dampinglike SOT is enhanced by a factor of 2 via submonolayer Hf insertion, as evidenced by both harmonic response measurements and current-induced switching of in-plane magnetized magnetic memory devices with the record low critical switching current of approximately 73 μA (switching current density of approximately 3.6×10^6 A/cm^2). This work demonstrates a very effective strategy for maximizing dampinglike SOT for low-power spin-torque devices.

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I. INTRODUCTION

Spin-orbit torques (SOTs) generated by the spin Hall effect (SHE) can efficiently switch thin-film nanomagnet devices [1–4], excite magnetization oscillations [5], and drive skyrmion and chiral domain-wall displacement [6–8]. Increasing SOT efficiencies is of great importance for enabling new research into spintronics phenomena [1–9] and for advancing technological applications of SOTs [10–13]. Of particular interest in this effort is to develop heavy metals (HMs) that can simultaneously provide a large dampinglike SOT efficiency per current density (ξ_{DL}^j), easy growth, good chemical and thermal stability, and the capability to be readily integrated into complex experimental configurations and/or into manufacturing processes. A good representative of such HMs is Pt, which has giant spin Hall conductivity (σ_{SH}) arising from the Berry curvature of its band structure [14,15]. For the SHE, $\xi_{DL}^j \equiv (2e/\hbar)T_{int}\sigma_{SH}\rho_{xx}$, with e, $\hbar$, ρ_{xx}, and T_{int} being the elementary charge, the reduced Planck's constant, the HM resistivity, and the spin transparency of the HM-ferromagnet (FM) interface [9]. ξ_{DL}^j for Pt/FM systems is approximately 0.08 where $\rho_{xx} = 20\,\mu\Omega\,\mathrm{cm}$ [16]. Recently, impurity scattering has been demonstrated to increase ξ_{DL}^j via enhancing ρ_{xx} [17–20]. However, in all the previous work the increase of ξ_{DL}^j was limited (e.g., to $\xi_{DL}^j = 0.12 - 0.3$ for 4-nm Pt alloys) due to a fast decrease in σ_{SH} with doping level [19] and/or only a weak enhancement of ρ_{xx} [17,18]. Exploring new enhancement strategies that can better optimize the trade-offs between ρ_{xx} and $T_{int}\sigma_{SH}$ is of both fundamental interest and technological urgency (e.g., for low-power magnetic memories, logic, and oscillators).

In this work, we report that introducing strong interfacial electron scattering via the insertion of submonolayers of Hf into Pt can enhance ρ_{xx} of an approximate 4-nm Pt layer by a factor of 5, which beneficially results in 100% enhancement of ξ_{DL}^j (up to 0.37). The increase in ξ_{DL}^j by the ultrathin insertion layers is approximately twice as effective as a uniform alloying of Hf into Pt. This giant enhancement of ξ_{DL}^j by Hf insertion layers is reaffirmed by the deterministic switching of in-plane magnetic tunnel junctions (MTJs) at a low zero-temperature critical current of approximately 73 μA (current density of approximately 3.6×10^6 A/cm^2) as determined from ramp-rate measurements.

II. RESULTS AND DISCUSSIONS

A. Enhancing resistivity by interfacial scattering

The main idea of this work is schematically shown in Fig. 1. In a single metallic layer of Pt that is not too thin, e.g., 4 nm as typically used for spin-torque magnetic random-access memories (MRAMs) [10], the resistivity

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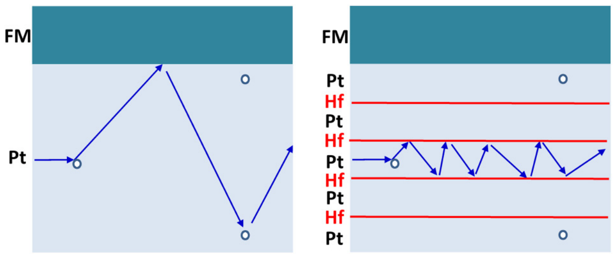

FIG. 1. Schematic depiction of interfacial scattering enhancement of the resistivity.

arises mainly from the electron scattering by impurities and thermal phonons inside the Pt layer and is hence relatively low, e.g., 20–50 $\mu\Omega$ cm at room temperature [16–21]. In contrast, if we "dice" the same Pt layer into several layers by inserting multiple ultrathin Hf layers during the deposition process, the new Pt/Hf interfaces should introduce strong additional interfacial scattering of electrons and hence greatly enhance the averaged ρ_{xx}. The Pt crystal structure between the interfaces can be disrupted less than would be the case for uniform alloying with Hf [19], thereby better preserving the large intrinsic σ_{SH} of Pt and better enhancing ξ_{DL}^{j}.

We sputter-deposit magnetic stacks of Ta(1.0 nm)/[Pt(d nm)/Hf(0.2 nm)]$_n$/Pt(d nm)/Co(t nm)/MgO(2.0 nm)/Ta(1.5 nm) with $d = 0.4$, 0.5, 0.6, 0.75, 1, 1.5, 2, and 4 nm. Here n (≤ 7) is chosen to be the integer that can make the total Pt thickness closest to 4 nm under the constraint that the total Hf thickness is no more than 1.4 nm (note that the spin diffusion length λ_s of the amorphous Hf is approximately 1 nm [22]). For the perpendicular magnetic anisotropy (PMA) samples, the Co thickness t is 0.83 nm for $d \geq 1$ and 0.63 nm for $d \leq 0.75$ nm; for in-plane magnetic anisotropy (IMA) samples, t is 1.3 nm for $d \geq 1$ and 0.93 nm for $d \leq 0.75$ nm. The samples are further patterned into 5×60 μm^2 Hall bars [see Fig. 2(a)] for resistivity and SOT measurements (see Supplemental Materials [23]).

As shown in Fig. 2(b), the average resistivity of the [Pt(d nm)/Hf(0.2 nm)]$_n$/Pt(d nm) multilayer is increased from 37 $\mu\Omega$ cm for $d = 4$ nm (pure Pt) to 191 $\mu\Omega$ cm for $d = 0.4$ **(**[Pt(0.4 nm)/Hf(0.2 nm)]$_7$/Pt(0.4 nm)**)**. Compared to that achieved by alloying or impurity doping (83 $\mu\Omega$ cm for $Au_{0.25}Pt_{0.75}$ and 110 $\mu\Omega$ cm for $Pt_{0.85}Hf_{0.15}$) [18–21], this is a remarkable resistivity enhancement despite the fact that the 0.2-nm Hf insertions are too thin to be distinguishable by either x-ray diffraction and/or reflectivity or scanning tunneling electron microscopy (STEM) and electron dispersive spectroscopy (EDS) measurements (see Figs. S1–S3 within the Supplementary Material [23]).

B. Magnifying spin torque by interfacial scattering

Figure 2(c) summarizes the values of ξ_{DL} determined from harmonic response measurements [24,25] on both

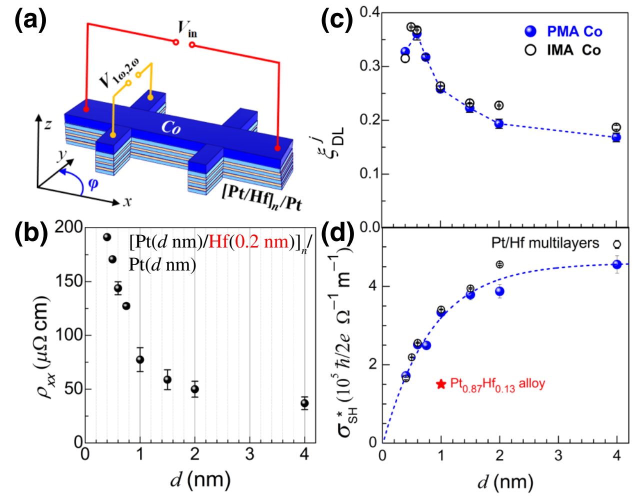

FIG. 2. (a) Geometry and coordinates for the SOT measurements; (b) measured resistivity of [Pt(d nm)/Hf(0.2 nm)]$_n$/Pt(d nm) multilayers as a function of the "slice" thickness d of the individual Pt layers; (c) the dampinglike spin-torque efficiency (ξ_{DL}^{j}); and (d) the apparent spin Hall conductivity (σ_{SH}^{*}) determined from harmonic-response measurements for both PMA (blue dots) and IMA (black circles) samples plotted as a function of d. The dashed lines are guides to the eye. The red star denotes the value of σ_{SH}^{*} for 4 nm of a spatially uniform $Pt_{0.87}Hf_{0.13}$ alloy [19].

the PMA and IMA multilayers as a function of d, with good agreement between the two types of measurements (see Figs. S4 and S5 within the Supplementary Material [23]). For both the PMA and IMA samples, ξ_{DL}^{j} increases quickly from 0.17 ± 0.01 at $d = 4$ nm (pure Pt) to a peak at $d = 0.6$ nm and then drops slightly as d increases further to 0.4 nm. The peak value of $\xi_{DL}^{j} = 0.37 \pm 0.01$ for $d = 0.6$ nm **(**i.e., [Pt(0.6 nm)/Hf(0.2 nm)]$_5$/Pt(0.6 nm) multilayers**)** is significantly higher than the values reported for $Pt_{0.85}Hf_{0.15}$ ($\xi_{DL}^{j} \approx 0.15$) [19], $Au_{0.25}Pt_{0.75}$ ($\xi_{DL}^{j} \approx 0.30$) [18], β-W ($\xi_{DL}^{j} \approx 0.2-0.3$) [13,26] and β-Ta ($\xi_{DL}^{j} \approx 0.12$) [3]. We attribute the increase of ξ_{DL} for Pt/Hf multilayers to the enhanced resistivity from interface scattering [see Fig. 2(b)]. Based on the comparisons in Fig. S8 and Table S2 within the Supplementary Material [23], the giant ξ_{DL}^{j} for Pt/Hf multilayers can provide very compelling current and energy efficiencies for spin-torque applications, for instance for SOT-MRAMs, with a current efficiency superior to any other known material for practical applications.

The interesting peak behavior of ξ_{DL}^{j} at $d \approx 0.6$ nm can be explained as being due to a competition between ρ_{xx} that increases quickly as a function of decreasing d [Fig. 2(b)] and the apparent spin Hall conductivity, $\sigma_{SH}^{*} \equiv T_{int}\sigma_{SH} = (\hbar/2e)\xi_{DL}^{j}/\rho_{xx}$, that decreases sharply as d decreases from 4 to 0.4 nm [Fig. 2(d)]. This decrease in σ_{SH}^{*} should be attributed partly to the enhanced attenuation of spin current in the Hf insertion layers. The amorphous Hf has a short λ_s of approximately 1 nm and does not contribute to the generation of the spin current due to its negligible SHE [22]. Therefore, in the multilayers with small d where the total Hf thickness reaches >1 nm, there should be a strong attenuation of the spin currents that diffuse to the FM interface from the bottom Pt layers to exert a SOT. In addition, the decrease of σ^*_{SH} with d could result in part from a strain-induced degradation of the Pt band structure (from a well-ordered fcc texture to a nearly armorphous structure, see Fig. S1). Nevertheless, in the Pt/Hf multilayers σ^*_{SH} is better preserved compared to that of uniformly doped Pt with Hf impurities. As shown in Fig. 2(d), σ^*_{SH} for the 4-nm $Pt_{0.87}Hf_{0.13}$ is 1.5×10^5 $(\hbar/2e)$ $\Omega^{-1}\,m^{-1}$ [19], which is a factor of 2 smaller than that of the Pt/Hf multilayers with similar Hf "concentration" (i.e., close to $[Pt(1\ nm)/Hf(0.2\ nm)]_3/Pt(1\ nm)$). This suggests that such HM multilayers with strong interfacial scattering can be generally advantagous over the corresponding impurity doping because in the latter σ_{SH} can be degraded more substantially by a stronger disturbance to the Pt band structure. We speculate that an enhancement of ξ^j_{DL} beyond the value of 0.37 that we obtain here should be possible if the increase of resistivity, the insertion layer attenuation of spin current, and the insertion-induced Pt strain can be better balanced, for instance, by using an insertion material that has a longer λ_s and an atomic radius closer to that of Pt (e.g., Ti) to minimize the disruption of the Pt crystal lattice and band structure.

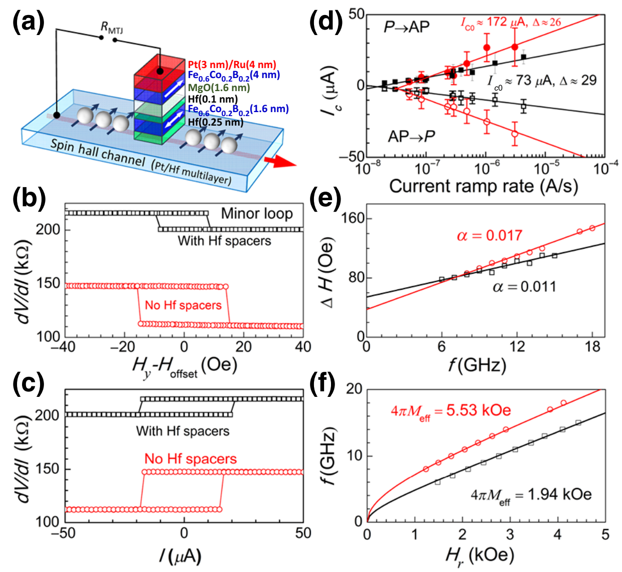

FIG. 3. (a) Schematic of the three-terminal MRAM device; (b) minor loop for switching by an in-plane applied magnetic field; (c) direct current switching loop; (d) critical current for $P \rightarrow AP$ (solid) and $AP \rightarrow P$ (open) switching as a function of current ramp rate; (e) FMR linewidth ΔH as a function of the resonance frequency f; (f) FMR resonance field H_r for the 1.6-nm $Fe_{0.6}Co_{0.2}B_{0.2}$ magnetic free layers for device A (red) and device B (black). The solid lines in (d), (e), and (f) denote the best fits of data to Eq. (1), $\Delta H = \Delta H_0 + (2\pi/\gamma)\alpha f$, and $f = (\gamma/2\pi)\sqrt{H_r(H_r + 4\pi M_{eff})}$, respectively. ΔH_0 and γ are the inhomogeneous broadening of the FMR linewidth and the gyromagnetic ratio, respectively.

C. Spin-torque switching of magnetization

Now we show that our optimal Pt/Hf multilayer with strong interfacial scattering, $[Pt(0.6\ nm)/Hf(0.2\ nm)]_5/Pt(0.6\ nm)$, is a particularly compelling spin Hall material for SOT research and technological applications. As the first example, we show the switching of a PMA Co layer ($j_e = 1.7 \times 10^7$ A/cm^2, coercivity $H_c = 0.43$ kOe) enabled by the giant ξ^j_{DL} due to the SHE of the $[Pt(0.6\ nm)/Hf(0.2\ nm)]_5/Pt(0.6\ nm)$ multilayer (Fig. S4). As an independent check of the effectiveness of the enhancement of ξ^j_{DL} by Pt/Hf interfaces, we demsonstrate antidamping switching of in-plane magnetized SOT-MRAM devices with $Fe_{0.6}Co_{0.2}B_{0.2}$-MgO MTJs. We fabricate two types of MRAM devices, devices A and B. Each MRAM device consists of a 300-nm-wide spin Hall channel of $[Pt(0.6\ nm)/Hf(0.2\ nm)]_n/Pt(0.6\ nm)$ ($n = 5$ for device A and 6 for device B), an elliptical MTJ pillar of $Fe_{0.6}Co_{0.2}B_{0.2}$(1.6 nm)/MgO(1.6 nm)/$Fe_{0.6}Co_{0.2}B_{0.2}$(4 nm) (190×45 nm^2 for device A or 190×74 nm^2 for device B), and protective capping layers of Pt(3 nm)/Ru(4 nm) (see the schematic in Fig. 3(a) and the cross-section STEM and EDS imaging results in Fig. S3). All devices are annealed at 240 °C. For device B, 0.25-nm and 0.1-nm Hf spacers are inserted at the bottom and top of the 1.6-nm $Fe_{0.6}Co_{0.2}B_{0.2}$ free layer, respectively, to suppress the magnetic damping constant (α) [27] and reduce the effective magnetization ($4\pi M_{eff}$), thereby reducing the critical current for antidamping switching [11]. The long axis of the elliptical MTJ pillars is along the y direction, transverse to the spin Hall channel and the write-current flow (x direction). In Figs. 3(b)–3(f), we compare the magnetization switching behaviors, α, and $4\pi M_{eff}$ of two representative MRAM devices without (device A, red) and with (device B, black) the two Hf spacers. Figure 3(b) shows the sharp switching minor loops of the MTJs under an in-plane magnetic field along the long axis of the MTJ pillar (H_y). The minor loops are artificially centered after subtraction of the dipole fields ($H_{offset} \approx 150$ Oe for device A and 180 Oe for device B) of the 4-nm $Fe_{0.6}Co_{0.2}B_{0.2}$ reference layers. H_c of the free layer is 36 Oe for device A and 9 Oe for device B. The apparent tunnel magnetoresistance ratio (approximately 40% for devices A and approximately 7% for device B) is not very high, which is attributed to a large background resistance caused during the device fabrication process (i.e., the oxidization of the Ti adhesion layer between the MTJ pillars and the top Pt contact as indicated in Fig. S3).

Figure 3(c) shows the characteristic switching behavior of devices A and B as the write current in the spin Hall channel is ramped quasistatically (an in-plane field equal to H_{offset} is applied along the pillar long axis to compensate the dipole field from the reference layer). The MTJs show abrupt switching at write currents of 16 μA for device A and 20 μA for device B. Since thermal fluctuations assist the reversal of a nanoscale MTJ device during slow current ramps, we carry out ramp-rate measurements [Fig. 3(d)]. Within the macrospin model, the switching current I_c should scale with the ramp rate ($\dot{I}$) following [28]

$$I_c = I_{c0}\left(1 + \frac{1}{\Delta}\ln\frac{\tau_0\Delta|\dot{I}|}{|I_{c0}|}\right). \tag{1}$$

Here I_{c0} is the critical switching current in the absence of thermal fluctuations, Δ is the stability factor that represents the normalized magnetic energy barrier for reversal between the P and AP states, and τ_0 is the thermal attempt time, which we assume to be 1 ns. By fitting to Eq. (1), we obtain $|I_{c0}| \approx 172 \pm 18$ μA and $\Delta \approx 26$ for device A and $|I_{c0}| \approx 73 \pm 15$ μA and $\Delta \approx 29$ for device B after averaging the critical currents for P $\rightarrow$ AP and AP $\rightarrow$ P switching. The small critical switching currents are consistently reproduced by other devices. Considering a parallel resistor approximation, the current shunted into the $Fe_{0.6}Co_{0.2}B_{0.2}$ free layer and Hf spacers ($\rho_{\mathrm{Pt/Hf}} \approx 144\,\mu\Omega$ cm, $\rho_{Fe_{0.6}Co_{0.2}B_{0.2}} \approx \rho_{\mathrm{Hf}} \approx 130\,\mu\Omega$ cm) can be estimated to be approximately equal to $0.2I_{c0}$ for both devices (see Fig. S6 and Table S1 within the Supplementary Material [23]). The critical switching density in the Pt spin Hall channel is therefore $j_{c0} \approx (1.0 \pm 0.1) \times 10^7$ A/cm^2 for device A (no Hf spacers) and $j_{c0} \approx (3.6 \pm 0.7) \times 10^6$ A/cm^2 for device B (with Hf spacers). Both the total critical switching and the low switching current density obtained from device B are much lower than those previously reported for in-plane [2,10,11,20,26] or perpendicular [12] spin-torque MTJs (see Table I).

According to the macrospin model, j_{c0} for antidamping torque switching of an in-plane magnetized MTJ is given by $j_{c0} = (2e/\hbar)\mu_0 M_s t\alpha(H_c + 4\pi M_{\mathrm{eff}}/2)/\xi_{\mathrm{DL}}^j$ [29]. With α of 0.017 (0.011), $4\pi M_{\mathrm{eff}}$ of 5.54 (1.94) kOe, and M_s of 1240 emu/cm^2 for the magnetic free layer of device A (B) as calibrated from ferromagnetic resonance (FMR) [see Figs. 3(e) and 3(f)] and vibrating sample magnetometry measurements on unpatterned thin-film stacks, we estimate ξ_{DL}^j to be approximately 0.29 for device A and 0.17 for device B. The slight reduction of ξ_{DL}^j for device B compared to device A is mainly attributed to the spin-current attenuation due to the insertion of the 0.25-nm Hf layer in device B between the Pt/Hf multilayer and the $Fe_{0.6}Co_{0.2}B_{0.2}$ layer (spin diffusion length of Hf is approximately 1 nm [22]). Despite this reduction, this Hf spacer layer is still beneficial in that the suppression of α and the reduction of $4\pi M_{\mathrm{eff}}$ for the free-layer interface more than compensates for the decrease in ξ_{DL}^j. The value of $\xi_{\mathrm{DL}}^j \sim 0.29$ for device A is significantly higher than those previously obtained in similar studies for MRAM devices based on β-W ($\xi_{\mathrm{DL}}^j = -0.15$) [11], $Pt_{0.85}Hf_{0.15}$ ($\xi_{\mathrm{DL}}^j = 0.098$) [20], and Pt ($\xi_{\mathrm{DL}}^j = 0.12$) [27]. We do note that $\xi_{\mathrm{DL}}^j = 0.29$ from the MRAM ramp-rate experiment is approximately 20% less that the value determined from harmonic response measurements [see Fig. 2(c)]. This difference may be partly attributed to an increased magnetic damping of nanoscale devices compared to thin-film stacks due to, e.g., the ion-beam damage and the side-wall oxidation of the nanopillar during the device fabrication process. Tapering of the free layer, which is formed during the ion milling process due to the resist shielding effect (see more details in Fig. S3), can significantly increase the effective volume of the free layer of the MRAM device and lead to additional current shunting into the free layer. This current shunting into the tapering area is not taken into account in our calculation. For the same reasons, ξ_{DL}^j of spin Hall materials is generally found to be underestimated in the ramp-rate results of other nanoscale MRAM devices compared to those in direct SOT measurements on microscale Hall bars [10,11,20] (e.g., for W, ξ_{DL}^j is approximately 0.15 from MRAM ramp-rate measurements and approximately 0.20 from bilayer spin-torque measurements [11]).

TABLE I. Comparison of SOT-MRAM devices. Both the critical switching current (I_c) and the critical switching current density (j_{c0}) for our Pt/Hf multilayer device are the lowest among all spin Hall materials demonstrated in room-temperature SOT-MRAM devices. Here $[\mathrm{Pt/Hf}]_n$ represents the multilayers of $[\mathrm{Pt(0.6\ nm)/Hf(0.2\ nm)}]_6$/Pt(0.6 nm).

Materials	SOT device	I_{c0} (mA)	j_{c0} (MA/cm^2)	Refs.
$[\mathrm{Pt/Hf}]_n$	In-plane MTJ	0.073	3.6	This work
W	In-plane MTJ	0.15	5.4	[11]
W	In-plane MTJ	0.95	18	[26]
Pt	In-plane MTJ	0.67	40	[10]
Ta	In-plane MTJ	2.0	32	[3]
$Pt_{0.85}Hf_{0.15}$	In-plane MTJ	0.56	14	[20]
Ta	PMA MTJ	>20	>50	[12]

We point out that the record-low critical switching current (current density) of the SOT-MRAMs based on Pt/Hf multilayers is a technologically interesting achievement. The three-terminal SOT-MRAM is an advantageous current- and energy-efficient cache-memory candidate because the separation of the read and write channels in the three-terminal geometry offers additional advantages over the conventional two-terminal spin-transfer-torque geometry, e.g., unlimited endurance, faster write (subnanosecond [11]), faster readout without read disturbance, lower write energy, and an allowance for a thick MgO barrier for enhanced tunnel magnetoresistance ratio.

III. CONCLUSION

In conclusion, we demonstrate, from direct spin-torque measurements and also spin-torque switching experiments of magnetic layers with both perpendicular and in-plane magnetic anisotropy, that introducing additional interface electron scattering within Pt by inserting submonolayer layers of Hf can significantly increase ξ_{DL}^{j}. For example, we show an increase of ξ_{DL}^{j} from 0.17 ± 0.01 for a simple 4-nm-thick single Pt layer to 0.37 ± 0.01 for a [Pt(0.6 nm)/Hf(0.2 nm)]$_7$/Pt(0.6 nm) multilayer despite the attenuation of spin current from Pt by the Hf insertion layers. Taking advantage of this interface-scattering-enhanced spin Hall ratio in the Pt/Hf multilayers, we demonstrate deterministic switching of IMA MRAM devices with a zero-temperature critical switching current of approximately 73 μA and a critical switching current density of approximately 3.6×10^6 A/cm^2, both of which are much lower than the previously reported results. Our optimized multilayer, [Pt(0.6 nm)/Hf(0.2 nm)]$_5$/Pt(0.6 nm) (with $\xi_{\mathrm{DL}}^{j} = 0.37$, $\rho_{xx} = 144\,\mu\Omega\,\mathrm{cm}$), represents a highly efficient generator of spin-orbit torque that is also compatible with integration technology (e.g., allowing easy growth with standard sputtering techniques on Si substrates) for development of low-power magnetic memories, oscillators, and logic. Our findings also provide an alternative strategy with the potential to magnify SOTs generated by other heavy metals, e.g., the low-resistivity Pd-Pt [17] or Au-Pt [18].

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