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Highly efficient spin current generation by the spin Hall effect in Au_{1-x}Pt_x

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We report very efficient spin current generation by the spin Hall effect in the alloy $Au_{0.25}Pt_{0.75}$, which, as determined by two different direct spin-orbit torque measurements, exhibits a giant internal spin Hall ratio of ≥ 0.58 (anti-damping spin-orbit torque efficiency of ≈ 0.35 in bilayers with Co), a relatively low resistivity of $\approx 83 \ \mu\Omega$ cm, an exceptionally large spin Hall conductivity of $\ge 7.0 \times 10^5 \ \Omega^{-1} \ m^{-1}$, and a spin diffusion length of 1.7 nm. This work establishes $Au_{0.25}Pt_{0.75}$ as a milestone spin current generator that provides greater energy efficiency than that yet obtained with other heavy metals or with the topological insulators Bi_2Se_3 and $(Bi_3Se)_2Te_3$. Our findings should advance spin-orbit torque-based fundamental research and benefit the development of new fast, efficient spin-orbit torque-driven magnetic memories, skyrmion and chiral domain wall devices, and microwave and terahertz emitters.

I. INTRODUCTION

Current-induced spin-orbit torques (SOTs) in heavy metal/ferromagnet (HM/FM) systems have attracted considerable attention due to their potential for application in the efficient manipulation of magnetization at the nanoscale [1-6]. The pure transverse spin current density j_s arising from an applied longitudinal electrical density j_e via the spin Hall effect (SHE) in HMs can generate damping-like SOT sufficiently strong to excite magnetization dynamics at microwave or terahertz frequencies [3,4], create and move skyrmions [5], drive chiral domain wall displacement [6], or switch the magnetization of thin film nanomagnets [1,2]. Of particular technological promise are the HMs with a large spin Hall ratio (θ_{SH}), or more precisely with a large dampinglike SOT efficiency $\xi_{DL} \equiv T_{int}\theta_{SH} \equiv T_{int}(2e/\hbar)j_s/j_e$, which is exerted on an adjacent ferromagnetic layer, as these could allow for the development of very fast, deterministic and efficient magnetic random access memories (MRAMs). Here T_{int} (<1) is the spin transparency of the HM/FM interface. For example, the SHE in Pt (W) has been demonstrated recently to switch in-plane magnetic tunnel junctions with a critical switching current density of 4.0 (0.54) $\times 10^7$ A/cm² and a write error rate below 10^{-5} (10⁻⁶) with a 2 ns pulse duration [7,8]. Despite extensive efforts, the efficiency of present SOT operations are still limited by a relatively low $\xi_{\rm DL}$ and/or a very high resistivity (ρ_{xx}) of the HM. For instance, β -W has a large $|\xi_{DL}|$ of ~0.3 but is very resistive $(\rho_{xx} \approx 300 \ \mu\Omega \ \text{cm} \ \text{at} \ 4 \ \text{nm})[9]; \ |\xi_{\text{DL}}|$ generally reported for Pt and β -Ta is comparatively low (~ -0.12), and β -Ta is also very resistive ($\approx 180 \ \mu\Omega \ cm$)[1,10]. These factors lead to undesirably large energy dissipation.

It has been established that the SHE in Pt is generally dominated by the intrinsic band structure effect [11,12] with the spin Hall conductivity $\sigma_{SH} \equiv \sigma_{SH}^{intr} + \sigma_{SH}^{extr} \approx \sigma_{SH}^{intr}$. Here σ_{SH}^{intr} and σ_{SH}^{extr} are the intrinsic and extrinsic contributions. Hence ξ_{DL} (= T_{int} (2*e/h*) $\sigma_{SH}\rho_{xx}$) can be enhanced by raising ρ_{xx} provided that σ_{SH} is not degraded in the process. Some success has been achieved in this manner by alloying Pt with Hf or with Al, but the maximum ξ_{DL} is still below 0.16 even when ρ_{xx} is enhanced to 110 $\mu\Omega$ cm, presumably due to a decrease in σ_{SH} with increasing Hf or Al concentration [10]. Recently theoretical work has suggested that alloying Pt with Au would be particularly effective in increasing ξ_{DL} via raising ρ_{xx} , due to a slower reduction in σ_{SH} at high Pt concentration [13]. Here we report from direct SOT measurements that the Au_{1-x}Pt_x alloy is indeed an enhanced spin current generator, with a maximum ξ_{DL} of 0.35 while still maintaining a relatively low ρ_{xx} of $\approx 83 \ \mu\Omega$ cm and a relatively long spin diffusion length (λ_s) of 1.7 nm for the optimized composition x = 0.75. We also determined a lower bound for θ_{SH} of 0.58 and a lower bound for the σ_{SH} of 7.0×10⁵ Ω^{-1} m⁻¹ for Au_{0.25}Pt_{0.75}. While the variation of ξ_{DL} with Pt concentration x is qualitatively similar in form to the theoretical prediction both θ_{SH} and σ_{SH} are much greater, approximately twice, than both the theoretical prediction and the experimental results from the indirect spin pumping/inverse SHE measurements reported in Ref. [13].

II. RESULTS AND DISCUSSIONS A. Samples and characterizations

The samples for this work include two series of multilayer stacks: (I) Ta 1.0/Au_{1-x}Pt_x 4.0 /Co 0.8-1.4/MgO 2.0/Ta 1.5 (numbers are thickness in nm) with x = 0, 0.25, 0.5, 0.65, 0.75, 0.85, and 1, respectively; (II) Ta $1.0/Au_{0.25}Pt_{0.75}$ 0, 2, 3, 4, 5, 6, and 8/Co 0.8-1.4/MgO 2.0/Ta 1.5 (Fig. 1(a)). All the stacks were sputtered at room temperature on SiO₂/Si wafers with argon pressure of 2 mTorr and a base pressure of below 1×10^{-8} Torr. The 1 nm Ta underlayer was introduced prior to co-sputtering of to improve the adhesion and uniformity of the $Au_{1,r}Pt_r$ layer but is expected to contribute negligible spin current into the Co layer as a consequence of the small thickness and high ρ_{xx} of Ta, the short λ_s of Au_{1-x}Pt_x, and the opposite signs of ξ_{DL} for Ta and Au_{1-x}Pt_x. The 1.5 nm Ta capping layer was fully oxidized upon exposure to atmosphere. X-ray diffraction studies show that, as is the case with the elemental metal Au and Pt, $Au_{1-x}Pt_x$ has a facecentered-cubic structure with the Au_{1-x}Pt_x (111) peak shifting from Pt to Au with increasing Au concentration (Fig. 1(b)), indicating the variation of lattice constant according to Vegard's Law (Fig. 1(c)). Vibrating sample magnetometer measurements as a function of Co thicknesses (t) show that there is no significant magnetic dead layer at the $Au_{1,r}Pt_r$ /Co and Co/MgO interfaces and that the Co layers have an almost x-independent saturation magnetization (M_s) of 1413 \pm 82 emu/cc [14], which is slightly smaller than the Co bulk value of ~1450 emu/cc and indicative of the absence of a

significant magnetic proximity effect [15].

As shown in Fig. 1(d), the stacks were patterned into 5×60 μ m² Hall bars by ultraviolet photolithography and argon ion milling. The Au_{1-x}Pt_x resistivity for each composition (x) and thickness (d) was determined by measuring the conductance enhancement of the corresponding stacks (d > 0 nm) with respect to the d = 0 nm stack. The Co resistivity was determined to be ~80 and 65 $\mu\Omega$ cm for 0.85 and 1.4 nm thicknesses, respectively, higher than bulk Co due to the strong interfacial scattering. For harmonic response measurements, a lock-in amplifier was used to source a sinusoidal voltage ($V_{in} = 4 \text{ V}$) onto the bar (length $L = 60 \text{ }\mu\text{m}$) orientated along the x axis and to detect the in-phase first and out-of-phase second harmonic Hall voltages, $V_{1\omega}$ and $V_{2\omega}$. The direct current switching of magnetization was measured with dc current sourced by a Yokogawa 7651 and with the differential Hall resistance detected by the lock-in amplifier $(V_{\rm in} = 0.1 \text{ V}).$

B. Composition dependence of Spin-orbit torques

Figure 1(e) shows three examples of the anomalous Hall voltage hysteresis loops of Au_{1-x}Pt_x 4 nm/Co 0.8 nm bilayers with x = 1, 0.75, and 0.25, where the fairly square loops and large H_c provide evidence of the strong perpendicular magnetic anisotropy (PMA) of these samples. All the samples with $x \ge 25\%$ exhibit good PMA, H_c larger than 500 Oe, and perpendicular anisotropy field H_k of 6-12 [14]. The strong PMA allows us to obtain high-quality out-of-plane harmonic response data. Pure Au/Co bilayers do not show PMA in the studied Co thickness range. We carried out inplane harmonic response measurement on Au_{1-x}Pt_x 4 /Co 1.4 bilayers with in-plane magnetic anisotropy (IMA) to reaffirm the SOT efficiencies for each x from 0 to 1.

We first discuss the SOTs in 4 nm $Au_{1-x}Pt_x$ samples with different x. Figure 2(a) shows an example of $V_{1\omega}$ and $V_{2\omega}$ to an applied ac current for perpendicularly magnetized $Au_{1-x}Pt_x$ 4/Co 0.8 bilayers ($x \ge 0.25$) as a function of in-plane bias fields H_x and H_y , from which the damping-like and field-like effective spin-torque fields are determined by $H_{\rm DL(FL)}$ = - $2\frac{\partial V_{2\omega}}{\partial H_{x(y)}}/\frac{\partial^2 V_{1\omega}}{\partial^2 H_{x(y)}}$. As summarized in Fig. 2(c), both H_{DL} and $H_{\rm FL}$ are tuned significantly by x. Figure 2(d) shows $\xi_{\rm DL}$ and field-like SOT efficiency ξ_{FL} calculated by $\xi_{DL(FL)}$ = $2e\mu_0 M_s t H_{DL(FL)}/\hbar j_e$, with e, μ_0, t, \hbar , and $j_e = V_{in}/L\rho_{xx}$ being the elementary charge, the permeability of vacuum, the ferromagnetic layer thickness, the reduced Planck constant, and the charge current density, respectively. Most notably, $\xi_{\rm DL}$ climbs up quickly from 0.03 at x = 0 (pure Au) to the peak value of 0.30 at x = 0.75 and then gradually drops to 0.18 at x = 1 (pure Pt). We consistently observed the same x dependence with in-plane harmonic measurements [14] of ξ_{DL} on Au_{1-x}Pt_x 4/Co 1.4 IMA samples (Fig. 2(d)). We note here that ξ_{DL} of 0.18 for the 4 nm pure Pt sample is larger than 0.12 as previously reported for Pt/Co bilayers [16], but in that case ρ_{xx} was 28 $\mu\Omega$ cm due to specific details of the thin film growth process. The giant ξ_{DL} of 0.30 for the 4 nm Au_{0.25}Pt_{0.75} is comparable to the high value reported for β -W [9] and 3 times higher than that of β -Ta [1]. One of the important factors that determines the non-monotonic x dependence is the variation in ρ_{xx} of the alloys. As can be clearly seen in Fig. 2(e), ρ_{xx} of 4 nm Au_{1-x}Pt_x thin film varies significantly and peaks at x = 0.75 due to enhanced electron scattering at that alloy concentration. We note that the maximum ρ_{xx} is 83 μ Ω cm for Au_{0.25}Pt_{0.75}, approximately one fourth that of β -W [9], which represents a considerable improvement of energy efficiency for SOT applications. ξ_{FL} is negative and small in magnitude, relative to ξ_{DL} , and scales with ξ_{DL} , consistent with the SHE in Au_{1-x}Pt_x alloys being the dominant source of both spin torques.

In Fig. 2(f) we plot the apparent spin Hall conductivity $\sigma_{SH}^* \equiv T_{int}\sigma_{SH} = (2e/\hbar)\mu_0 M_s t H_{DL}\rho_{xx}/j_e$ as a function of *x* using the experimental results shown in Figs. 2(c) and 2(e). Assuming that T_{int} is approximately independent of *x* in the high Pt concentration regime, this plot indicates that σ_{SH} remains constant or even initially increases slightly for decreasing *x* in the Pt rich regime, $0.6 < x \le 1.0$. Moreover, as also shown in Fig. 2(f), the measured σ_{SH}^* for the 4 nm samples has a functional *x* dependence similar to a recent calculation of σ_{SH}^{intr} for bulk Au_{1-x}Pt_x[13], which supports the conclusion that in this system the intrinsic spin Hall contribution arising from the topology of the electronic band structure is dominant.

C. Spin diffusion length and spin Hall conductivity

The spin diffusion length is a key parameter for the theoretical understanding of a spin Hall material, for determining $\theta_{\rm SH}$ via inverse SHE experiments and for optimizing SOT effectiveness. We determined λ_s for $Au_{0.25}Pt_{0.75}$ by out-of-plane harmonic measurements of H_{DL} and ξ_{DL} for a series of Au_{0.25}Pt_{0.75} d/Co 0.8 bilayers as a function of thickness d. The results plotted in Figs. 3(a) and 3(b) show that $H_{\rm DL}$ and $\xi_{\rm DL}$ increase as d increases, as expected from the "bulk" SHE, with ξ_{DL} saturating at ≈ 0.35 by d = 8 nm. In Fig. 3(c) we plot the resistivity of the Au_{0.25}Pt_{0.75} layers as a function of d showing that ρ_{xx} reaches its bulk value of 83 $\mu\Omega$ cm by $d \approx 3$ nm, consistent with a high resistivity material having a short bulk mean-free-path. In Fig. 3(d) we plot σ_{SH}^* of the Au_{0.25}Pt_{0.75} alloy. If we assume that the SOT arises entirely from the SHE of $Au_{0.25}Pt_{0.75}$, as indicated by the *d* dependence of ξ_{DL} and that the interfacial spin mixing conductance $G^{\uparrow\downarrow} \approx \text{Re}G^{\uparrow\downarrow} \gg \text{Im}G^{\uparrow\downarrow}$, we can obtain and $\sigma_{\rm SH}$ following $\sigma_{\rm SH}^* = \sigma_{\rm SH} (1-\operatorname{sech}(d/\lambda_s))$ $\lambda_{\rm s}$ $(1+\tanh(d/\lambda_s)/2\lambda_s\rho_{xx}\text{Re}G^{1})^{-1}$ [17]. This assumes that T_{int} is set by the spin backflow of an ideal Au_{0.25}Pt_{0.75}/Co interface with no interfacial spin-flip scattering (spin memory loss). We use $\rho_{xx} = 83 \ \mu\Omega \ \text{cm}$ and, as an approximation, $G^{12} \approx 5.9 \times 10^{14} \ \Omega$ 1^{1} m⁻² as calculated for the Pt/Co interface [17] to fit the data as shown by the solid line in Fig. 3(d). This fit ignores the slight variation in ρ_{xx} for small d and any change in λ_s with that variation. From this fit we determine $\lambda_s \approx 1.7 \pm 0.1$ nm, $\sigma_{\rm SH} \approx (7.0 \pm 0.1) \times 10^5 \ \Omega^{-1} \ {\rm m}^{-1}$, and an internal spin Hall ratio $\theta_{\rm SH}$ = 0.58 ± 0.01. This value for $\lambda_{\rm s}$ together with the measured ρ_{xx} corresponds to a Au_{0.25}Pt_{0.75} spin conductance $1/\lambda_{\rm s}\rho_{xx} \approx 0.71 \times 10^{15} \,\Omega^{-1} {\rm m}^{-2}, \sim 40\%$ less than $1.25 \times 10^{15} \,\Omega^{-1}$ ¹m⁻² obtained for pure Pt from a similar thickness dependent study [12]. Given the large value for $\sigma_{\rm SH}$ obtained from this thickness-dependent measurement which as it assumes an ideal $Au_{1-x}Pt_x/Co$ interface is perhaps just a lower bound, the proximity of the measured σ_{SH}^* value for $d \approx 8$ nm to the theorectically calculated value [13] also shown in Fig. 2(d) is fortutious. Instead our measurements are indicating that internal σ_{SH} for Au_{0.25}Pt_{0.75} is much larger than yet calculated, in accord with the results previously found for pure Pt [12]. Finally we note that the value for λ_s here is considerably larger than ~0.2 nm (less than one atomic layer) used in the extraction of θ_{SH} in Au_{1-x}Pt_x/Ni₈₁Fe₁₉ bilayers from spin pumping and inverse SHE measurements [13]. This latter technique also does require a quite accurate determination of G^{tl} , which can be challenging to achieve, particularly if there is significant spin memory loss at the interface [16,18].

D. Energy efficiency for spin-torque applications

Finally, we point out that $Au_{0.25}Pt_{0.75}$ is a particularly notable spin Hall material for SOT research and technological applications. As an example, we first show in Fig. 4(a) the deterministic switching of a 0.8 nm thick perpendicularly magnetized Co layer with a large PMA ($H_k \approx$ 6620 Oe) and a high coercivity ($H_c \approx 800$ Oe) enabled by the giant ξ_{DL} generated by the SHE of Au_{0.25}Pt_{0.75}. The switching current of ~3 mA corresponds to $j_e = 1.2 \times 10^7 \text{ A/cm}^2$ in the Au_{0.25}Pt_{0.75} layer. A small magnetic field of \pm 50 Oe was applied along the current direction to overcome the Dzyaloshinshii-Moriya interaction at the Au_{0.25}Pt_{0.75}/Co interface. For technological applications, e.g. MRAMs, very efficient SOT switching of a FeCoB thin layer is of great interest. Figure 4(b) compares the write energy efficiency for SOT-MRAM applications based on various strong spin Hall channel materials whose ξ_{DL} , ρ_{xx} , and σ_{SH}^* are listed in Table I. Here we calculate a MRAM device with a $400 \times 200 \times 4 \text{ nm}^3$ spin Hall channel and a $110 \times 30 \times 1.8$ nm³ FeCoB free layer (resistivity $\rho_{FeCoB} = 130 \ \mu\Omega$ cm) by taking into account the current shunting into the free layer and by using the parallel resistor model (see the inset of Fig. 4(b))[14]. The write power consumption for Au_{0.25}Pt_{0.75} is 5, 8, 10 and 30 times smaller than that for Pt, $Pt_{0.85}Hf_{0.15}$, β -W, and β -Ta, respectively. The SOT-MRAMs based on topological insulators Bi₂Se₃ and (Bi₂Se)₂Te₃ which are reported to have ξ_{DL} of 0.16-3.5 [19,20] also have much higher power consumption than Au_{0.25}Pt_{0.75} due to the considerable current shunting effect that is a consequence of their giant resistivity (e.g. 4020 $\mu\Omega$ cm for (Bi,Se)₂Te₃). We also note that $Au_{0.25}Pt_{0.75}$ is compatible with sputtering techniques and the use of Si substrates which are preferable for integration technology, whereas Bi₂Se₃ and (Bi,Se)₂Te₃ require costly molecular beam epitaxy growth and GaAs or sapphire substrates [19,20]. The high ρ_{xx} of β -W, β -Ta, Bi₂Se₃, or (Bi,Se)₂Te₃ is also problematic for applications that require energy efficiency, e.g. the prospective implementation of SOT devices in cryogenic computing systems [11]. For example, use of a spin Hall material with a large resistivity will result in a high write impedance that is difficult for superconducting circuits in a cryogenic computing system to accommodate. Therefore, the combination of the giant ξ_{DL} the relatively low resistivity, and the compatibility with sputtering and silicon integration technology makes Au_{0.25}Pt_{0.75} a milestone spin Hall material for SOT research and technological applications.

III. CONCLUSIONS

We have found Au_{0.25}Pt_{0.75} to be a strong spin Hall material with, a giant $\theta_{\rm SH}$ of > 0.58 ($\xi_{\rm DL}$ of 0.35 in bilayers with Co), a relatively low resistivity of ~83 $\mu\Omega$ cm, a large σ_{SH} of > $7.0 \times 10^5 \ \Omega^{-1} \ \mathrm{m}^{-1}$, and λ_{s} of ~1.7 nm (assuming $G^{\mathrm{s}} \approx 5.9 \times 10^{14}$ Ω^{-1} m⁻²). This giant $\theta_{\rm SH}$ and $\xi_{\rm DL}$ arising from the intrinsic contribution of the SHE in Au_{1-x}Pt_x alloys and the low resistivity make Au_{0.25}Pt_{0.75} more energy-efficient than all the other conventional HMs and the topological insulators Bi₂Se₃ and (Bi,Se)₂Te₃ whose spin Hall properties have yet been reported. Our direct demonstration of the SOT efficiency of this milestone spin Hall material Au_{0.25}Pt_{0.75} which simultaneously combines a giant charge-spin conversion efficiency, low resistivity, and chemical stability, with excellent processing compatibility for device integration, paves the way for further fundamental research that will benefit from the efficient generation of spin currents and of spin-orbit torques, and for the development of new highly efficient SOT-driven magnetic memories, skyrmion and chiral domain wall devices, and microwave and terahertz emitters.

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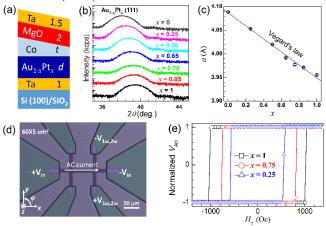


FIG. 1. (a) Schematic depiction of the magnetic stacks with *t* and *d* being the thicknesses of Co and Au_{1-x}Pt_x layers (the thicknesses are in nm); (b) X-ray diffraction patterns for different Au_{1-x}Pt_x composition *x*; (c) Optical microscopy image of a Hall bar device showing the geometry and the measurement coordinate; (d) Normalized anomalous Hall voltage for Au_{1-x}Pt_x 4/Co 0.8 bilayers (x = 1, 0.75, and 0.5, respectively).

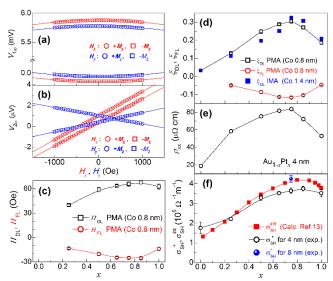


FIG. 2. (a) The first $(V_{1\omega})$ and (b) second $(V_{2\omega})$ harmonic voltages plotted as a function of in-plane fields H_x (red) and H_y (blue), respectively (for x = 0.75); Composition dependence of (c) effective spin-torque fields H_{DL} and H_{FL} , (d) spin-torque efficiencies ξ_{DL} and ξ_{FL} , (e) Au_{1-x}Pt_x resistivity ρ_{xx} , and (f) *calculated* intrinsic spin Hall conductivity σ_{SH}^{intr} for bulk Au_{1-x}Pt_x (red squares) and experimentally measured effective spin Hall conductivity $\sigma_{SH}^* \equiv T_{int}\sigma_{SH}$, where $T_{int} \cong 0.6$ if there is no spin memory loss at the Au_{1-x}Pt_x/Co interfaces, for 4 nm (black circle) and

Au_{0.25}Pt_{0.75} 0.35 80 4.4 This work

8 nm Au_{0.25}Pt_{0.75} (blue solid dot), respectively. For clarity, the H_y -dependent $V_{1\omega}$ data (the blue points) in (a) are artificially shifted by ±0.001 mV for $\mp M_z$, respectively.

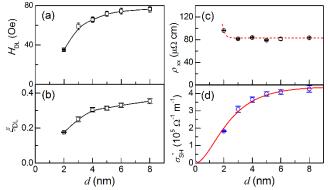


FIG. 3. Au_{0.25}Pt_{0.75} thickness dependence of (a) H_{DL} , (b) ξ_{DL} ,(c) ρ_{xx} , and (d) σ_{SH}^* , respectively.

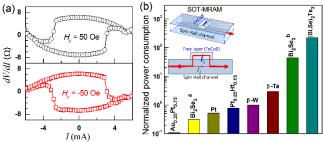


FIG. 4. (a) Deterministic direct current switching of a 0.8 nm perpendicualrly magnetized Co layer with coercivity of 800 Oe and perpendicular anisotropy field of 6620 Oe via the damping-like SOT genearated by the giant SHE of a 4 nm $Au_{0.25}Pt_{0.75}$ layer. Magnetic fields of \pm 500e were applied along the current direction, respectively. (b) Normalized power consumption as calculated for a typical SOT-MRAM device with different spin Hall channel materials listed in Table 1. Insets: schematics of a SOT-MRAM device and its side-view highlighting the increase of the write current due to the current shunting into the magnetic free layer.

TABLE I. Comparison of ξ_{DL} , ρ_{xx} , and σ_{SH}^* of various spin Hall materials.

	ζdl	$ ho_{xx}$ ($\mu\Omega$ cm)	$\sigma_{\rm SH}^{*}$ (10 ⁵ Ω^{-1} m ⁻¹)	Ref.
β-Ta	0.12	190	0.63	[1]
β -W	0.3	300	1.0	[9]
Pt	0.12	50	2.4	[12]
$Pt_{0.85}Hf_{0.15}$	0.16	110	1.5	[10]
Bi ₂ Se ₃ ^a	3.5	1755	2.0	[18]
Bi ₂ Se ₃ ^b	0.16	1060	0.15	[19]
(Bi,Se) ₂ Te ₃	0.4	4020	0.1	[19]