Observation of the crossover between metallic and insulating regimes of the spin Hall effect

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1 **Observation of the crossover between metallic and insulating**
2 **regimes of the spin Hall effect**

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The physics of the anomalous and spin Hall effects is one of the most intriguing aspects of condensed matter physics. An important finding from a large collection of experimental and theoretical results is the universal scaling of the anomalous or spin Hall conductivity with the electric conductivity. This scaling has been successfully described by the intrinsic Berry curvature and extrinsic scattering mechanisms for metallic systems, revealing the topological nature of these effects. In contrast, the underlying physics in the opposite limit, the disordered insulating regime, is still unclear. In particular, it remains a major challenge to explore the spin Hall effect in the insulating regime. Here, we report the observation of the crossover between the metallic and insulating regimes of the spin Hall effect. The result demonstrates an important correspondence between the spin and anomalous Hall effects, which provides essential information for the fundamental understanding of spin transport.

INTRODUCTION

The family of Hall effects has played a central role in the development of condensed matter physics^{1–6}. An important member of this family is the spin Hall effect (SHE), which was theoretically predicted about half a century ago⁷. The SHE has attracted extensive attention for its fascinating topological, relativistic, and quantum mechanical nature, as well as spintronics applications^{3,8}. This effect enables electric generation and detection of spin currents in solid-state devices, providing an avenue for discovering a variety of spintronics phenomena³.

The SHE shares the same origin as the anomalous Hall effect (AHE). The SHE and AHE in metallic systems have intrinsic and extrinsic contributions². The intrinsic contribution originates from the Berry curvature associated with the Fermi surface and the band structure of the material, whereas the extrinsic mechanism originates from spin-dependent scattering on structural defects or impurities. For the AHE, a large collection of experimental results on a wide class of ferromagnets has revealed that there are three different regimes², characterized by the power-law relation, $\sigma_{xy} \propto \sigma_{xx}^\gamma$, between the anomalous Hall conductivity σ_{xy} and the longitudinal conductivity σ_{xx} ; (i) the superclean regime, where $\sigma_{xy} \propto \sigma_{xx}$ due to the dominant contribution from the skew-scattering, (ii) the moderately dirty regime, where the intrinsic mechanism is dominant, and σ_{xy} is roughly insensitive to σ_{xx} , and (iii) the

dirty regime with the scaling exponent γ generically larger than unity. The discovery of the crossover of the AHE has provided important information for the fundamental understanding of the physics of spin transport. Recently, the crossover between these regimes has also been confirmed for the SHE^{9–11}, demonstrating an important correspondence between the AHE and SHE in the metallic regime.

Although the existing theories have been successful in describing the AHE and SHE in metallic systems based on the intrinsic Berry curvature or the extrinsic scattering mechanisms, it remains a major challenge to understand the full-range phase diagram of these phenomena. The last important step is to explore the AHE and SHE in disordered insulating systems, where the carrier transport is dominated by hopping. In the insulating regime, the scaling relation is nontrivial because the mechanism of the AHE and SHE in hopping systems is clearly different from that in metallic systems; in the insulating regime, the AHE is attributed to a phase that a carrier gains when hopping around closed-loop paths in the presence of spin-orbit coupling (SOC) and background magnetization of the localized moments^{12,13}. Nevertheless, experimental observations have demonstrated that the scaling of the anomalous Hall conductivity with the electric conductivity prevails not only in the dirty metallic regime but also deep into the disordered insulating hopping regime². An analogous trend is expected for the SHE because of the similarity in the underlying physics. However, in contrast to the large collection of experimental results for the AHE, it remains a major challenge to explore the SHE in the disordered insulating regime.

In this work, we report the observation of the crossover between the metallic and insulating regimes of the SHE in Pt, a model system for the study of the SHE. To explore the SHE in the insulating regime, the electric conductivity of Pt films is varied by three orders of magnitude by incorporating oxygen. We find the scaling behavior of the spin Hall conductivity, which prevails not only in the dirty metallic regime but also in the disordered insulating regime, despite the distinct difference in the transport mechanism. The observed variation of the spin Hall conductivity over the wide range of electric conductivity is reminiscent of the scaling of the anomalous Hall conductivity, illustrating a direct correspondence between the SHE and AHE in both the metallic and insulating regimes.

1 RESULTS

2 Transport measurement.

3 We use spin-torque ferromagnetic resonance (ST-FMR)^{14–16} to study the SHE of PtO_x
4 films with various oxidation levels. Figure 1a shows a schematic illustration of the device.
5 The device structure is SiO₂(4 nm)/Ni₈₁Fe₁₉(*d*_F)/TiN(2 nm)/PtO_x(10 nm)/SiO₂-substrate,
6 where the numbers in parentheses represent the thickness (for details, see Methods). The
7 separation between the Ni₈₁Fe₁₉ and PtO_x layers by the insertion layer minimizes the in-
8 terfacial SOC effects, including the spin memory loss and interfacial spin-orbit torques^{17,18}.
9 To minimize possible oxidation effects on the insertion layer due to its proximity to the
10 PtO_x layer, we chose TiN, which is less susceptible to oxidation¹⁹, instead of other light
11 metals, such as Cu and Ti. We can neglect the spin relaxation in the insertion layer because
12 of the long spin diffusion length of around 40 nm in TiN due to the weak SOC²⁰. In the
13 Ni₈₁Fe₁₉/TiN/PtO_x device, to control the oxidation level of the PtO_x layer, argon and oxy-
14 gen gases were introduced into the chamber during the sputtering of the PtO_x film, and the
15 amount of oxygen gas in the reactive mixture, *Q*, was varied between 0 and 10%. As shown
16 in Fig. 1b, the electric resistivity ρ_N of the PtO_x film increases with *Q*, indicating that the
17 oxidation level of the PtO_x layer is controlled by tuning *Q*. The change in the oxidation
18 level induced by tuning *Q* is supported by x-ray photoelectron spectroscopy (XPS) spectra,
19 shown in Fig. 1c.

20 To characterize the carrier transport in the PtO_x films with different oxidation levels,
21 we measured temperature *T* dependence of the sheet resistance *R*_s for the PtO_x films.
22 Figure 2a shows the *T* dependence of *R*_s for the PtO_x film with *Q* = 1%. This result shows
23 that *R* decreases monotonically with decreasing *T*: $dR_s/dT > 0$, showing a typical metallic
24 behavior. By increasing the oxidation level, the *T* dependence of *R*_s is clearly changed. As
25 shown in Figs. 2b and 2c, *R*_s increases with decreasing *T* for the PtO_x film with *Q* = 5%
26 and *Q* = 10%: $dR_s/dT < 0$.

27 To clarify the transport mechanism in the PtO_x films around room temperature, we plot
28 $\log R_s$ as a function of $T^{-1/4}$ (see the inset to Figs. 2b and 2c). This result shows that, for
29 $200 \text{ K} \leq T \leq 300 \text{ K}$, the *T* dependence of *R*_s is well described by the Mott variable range
30 hopping (Mott-VRH) mechanism²¹:

$$R_s = R_{\text{Mott}} \exp \left[\left(\frac{T_{\text{Mott}}}{T} \right)^{1/4} \right], \quad (1)$$

where R_{Mott} and T_{Mott} are the resistance parameter and the Mott characteristic temperature, respectively. This result indicates that the PtO_x film with $Q = 5\%$ is near the crossover between the band and hopping transport regimes, and the transport in the PtO_x film with $Q = 10\%$ is dominated by the Mott-VRH.

Spin-torque ferromagnetic resonance.

Figure 3a shows the ST-FMR spectra for the $\text{Ni}_{81}\text{Fe}_{19}(8\text{ nm})/\text{TiN}(2\text{ nm})/\text{PtO}_x(10\text{ nm})$ trilayers with $Q = 0, 6$, and 10% , measured at room temperature. For the measurement, a radio frequency (RF) charge current was applied along the longitudinal direction of the device, and an in-plane external magnetic field H was applied with an angle of $\theta = 45^\circ$ from the longitudinal direction (see also Fig. 1a). In the trilayer, the RF current generates damping-like (DL) and field-like (FL) spin-orbit torques, as well as an Oersted field, which drive magnetization precession in the $\text{Ni}_{81}\text{Fe}_{19}$ layer under the FMR condition. The magnetization precession induces an oscillation of the resistance due to the anisotropic magnetoresistance, resulting in the generation of a direct current (DC) voltage V_{DC} through the mixing of the RF charge current and oscillating resistance:^{14,15}

$$V_{\text{DC}} = V_{\text{sym}} \frac{W^2}{(\mu_0 H - \mu_0 H_{\text{res}})^2 + W^2} + V_{\text{antisym}} \frac{W(\mu_0 H - \mu_0 H_{\text{res}})}{(\mu_0 H - \mu_0 H_{\text{res}})^2 + W^2}, \quad (2)$$

where W is the linewidth and H_{res} is the FMR field. Here, V_{sym} and V_{antisym} are the magnitude of the symmetric and antisymmetric components, respectively; V_{sym} is proportional to the DL spin-orbit effective field H_{DL} , while V_{antisym} is proportional to the sum of the Oersted field H_{Oe} and FL spin-orbit effective field H_{FL} . As shown in Fig. 3a, the V_{DC} spectrum varies systematically by changing the RF frequency f . We have confirmed that the variation of the resonance field H_{res} is consistent with the Kittel formula: $(2\pi f/\gamma) = \sqrt{\mu_0 H_{\text{res}}(\mu_0 H_{\text{res}} + \mu_0 M_{\text{eff}})}$, where M_{eff} is the effective demagnetization field and γ is the gyromagnetic ratio. We also note that in-plane magnetic field angle θ dependence of the symmetric V_{sym} and antisymmetric V_{antisym} components, extracted by fitting the measured V_{DC} using Eq. (2), is consistent with the prediction of the ST-FMR, as shown in Fig. 3b.

The ST-FMR signals observed for the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films originate from the RF current flowing in the PtO_x layer. This is evidenced by the fact that the V_{DC} signal disappears in a $\text{Ni}_{81}\text{Fe}_{19}(8\text{ nm})/\text{TiN}(2\text{ nm})$ film, where the PtO_x layer is absent, as shown in Fig. 3c. This result shows that the anomalous spin-orbit torque in the $\text{Ni}_{81}\text{Fe}_{19}$ layer, as well as

the spin-orbit torques generated by the TiN layer is negligible in the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films. The interfacial spin-orbit torques originating at the TiN/PtO_x interface also play a minor role in the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ film because interfacial SOC effects are notable only when PtO_x is directly contacted with $\text{Ni}_{81}\text{Fe}_{19}$ ¹⁸. These results indicate that the sizable symmetric voltage V_{DC} observed for the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films originates from the DL torque generated by the SHE in the PtO_x layer.

The ST-FMR for the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films allows us to quantify the DL-torque efficiency due to the SHE in the PtO_x layer. For the trilayer, we define the effective FMR spin-torque efficiency as²²

$$\xi_{\text{FMR}}^{\text{eff}} = \frac{V_{\text{sym}}}{V_{\text{antisym}}} \frac{e\mu_0 M_s d_{\text{F}} d_{\text{N}}^{\text{eff}}}{\hbar} \sqrt{1 + \frac{\mu_0 M_{\text{eff}}}{\mu_0 H_{\text{res}}}}, \quad (3)$$

where $d_{\text{N}}^{\text{eff}} = d_{\text{N}} + (\rho_{\text{N}}/\rho_{\text{I}})d_{\text{I}}$ is the effective thickness of the nonmagnetic layer. In Eq. (3), $d_{\text{N}} = 10$ nm is the thickness of the PtO_x layer; $d_{\text{I}} = 2$ nm and $\rho_{\text{I}} = 462 \mu\Omega\text{cm}$ are the thickness and resistivity of the TiN layer, respectively. Here, M_s is the saturation magnetization, which is independent of Q as shown in Fig. 3d. The negligible change in M_s with Q shows that the $\text{Ni}_{81}\text{Fe}_{19}$ layer is not affected by the change of the oxidation level of the PtO_x layer, supporting that the $\text{Ni}_{81}\text{Fe}_{19}$ and PtO_x layers are well separated by the TiN insertion layer. We have also confirmed that $\xi_{\text{FMR}}^{\text{eff}}$, extracted by fitting the V_{DC} spectra, is independent of the frequency f of the applied RF current, as shown in Fig. 3e. The negligible change in $\xi_{\text{FMR}}^{\text{eff}}$ with f shows that the observed voltage is dominated by the ST-FMR, and possible spin pumping and thermoelectric contributions are negligible in the ST-FMR spectra²³. From the effective FMR spin-torque efficiency $\xi_{\text{FMR}}^{\text{eff}}$, the DL(FL) torque efficiencies per unit applied electric field E , $\xi_{\text{DL(FL)}}^E = (2e/\hbar)\mu_0 M_s d_{\text{F}} H_{\text{DL(FL)}}/E$, can be determined using^{22,24}

$$\frac{1}{\xi_{\text{FMR}}^{\text{eff}}} = \frac{1}{\xi_{\text{DL}}^E} \left(\frac{1}{\rho_{\text{N}}} + \frac{\hbar}{e} \frac{\xi_{\text{FL}}^E}{\mu_0 M_s d_{\text{F}} d_{\text{N}}^{\text{eff}}} \right). \quad (4)$$

To determine ξ_{DL}^E using Eq. (4), we measured the ST-FMR for the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films with different $\text{Ni}_{81}\text{Fe}_{19}$ layer thicknesses d_{F} , as shown in Figs. 4a and 4b. By fitting the measured spectra using Eq. (2), we obtain $1/d_{\text{F}}$ dependence of $1/\xi_{\text{FMR}}^{\text{eff}}$ for the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films with different oxidation levels, as shown in Figs. 4c-f. Figures 4c-f show that $1/\xi_{\text{FMR}}^{\text{eff}}$ changes linearly with $1/d_{\text{F}}$, enabling us to determine the DL torque efficiency ξ_{DL}^E using Eq. (4).

1 DISCUSSION

2 Using the ST-FMR result, we investigate the variation of the spin Hall conductivity σ_{SH}
 3 in the PtO_x films, where the longitudinal electric conductivity σ_{N} is varied by three orders
 4 of magnitude. Since the SHE in the PtO_x layer dominates the observed DL torque, the spin
 5 Hall conductivity σ_{SH} of the PtO_x layer can be determined from the extracted values of ξ_{DL}^E
 6 using

$$\sigma_{\text{SH}} = \frac{1}{T_{\text{int}}} \left(\frac{\hbar}{2e} \right) \xi_{\text{DL}}^E. \quad (5)$$

7 For the metallic PtO_x , the spin transparency T_{int} in the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ film is described
 8 by the framework based on the spin diffusion model²⁵:

$$T_{\text{int}} = \frac{1 - \text{sech}(\delta_{\text{N}})}{G_{\text{I}} \sinh(\delta_{\text{I}}) + G_{\text{N}} \cosh(\delta_{\text{I}}) \tanh(\delta_{\text{N}})} \frac{G_{\text{F}} \tanh \delta_{\text{F}}}{1 + (G_{\text{F}}/G'_{\text{ext}}) \tanh \delta_{\text{F}}}, \quad (6)$$

9 where $\delta_i = d_i/\lambda_i$, $G_i = \sigma_i/\lambda_i$, and $G'_{\text{ext}} = G_{\text{I}} \left[\frac{G_{\text{I}} \coth(\delta_{\text{N}}) + G_{\text{N}} \coth(\delta_{\text{I}})}{G_{\text{I}} \coth(\delta_{\text{N}}) \coth(\delta_{\text{I}}) + G_{\text{N}}} \right]$. Here, λ_i is the spin
 10 diffusion length and σ_i is the longitudinal electric conductivity of the i (=F, I, and N) layer,
 11 where F, I, and N correspond to $\text{Ni}_{81}\text{Fe}_{19}$, TiN, and PtO_x , respectively. Using^{26,27} $G_{\text{F}} = 1.1 \times$
 12 $10^{15} \Omega^{-1}\text{m}^{-2}$ and $G_{\text{N}} = 1.6 \times 10^{15} \Omega^{-1}\text{m}^{-2}$ with measured values of σ_i , we obtain $T_{\text{int}} = 0.35$
 13 for $Q = 0\%$. By increasing Q , the transparency T_{int} increases due to suppression of the
 14 spin conductance G_{N} because G_{N} is proportional to the carrier density and independent
 15 of the carrier scattering time¹¹. By taking into account the change of the carrier density,
 16 determined from Hall measurements, and assuming a dominant Elliott-Yafet spin relaxation
 17 mechanism, we obtain $T_{\text{int}} = 0.44$ for $Q = 1\%$. In Fig. 5, we plot the spin Hall conductivity,
 18 determined from $\sigma_{\text{SH}} = (1/T_{\text{int}})(\hbar/2e)\xi_{\text{DL}}^E$. Figure 5 shows that σ_{SH} is almost independent
 19 of σ_{N} when σ_{N} is in the range of around $2 \times 10^4 \Omega^{-1}\text{cm}^{-1}$ to $8 \times 10^4 \Omega^{-1}\text{cm}^{-1}$. This result
 20 is consistent with the prediction of the SHE in the moderately dirty regime, where the
 21 dominant mechanism of the SHE is the intrinsic mechanism and the spin Hall conductivity
 22 is insensitive to the electric conductivity².

23 The situation is clearly changed by further decreasing the electric conductivity of the PtO_x
 24 layer. For the heavily oxidized PtO_x devices, a full description of the spin transparency
 25 requires a theoretical framework describing hopping spin transport. However, developing
 26 such a framework remains a major challenge for theoretical study. In this work, we adopt the
 27 spin-diffusion model, which has been justified in organic semiconductors where the transport
 28 is dominated by carrier hopping, as well as metals and inorganic semiconductors²⁸. Under
 29 this assumption, $T_{\text{int}} = 0.44$, for $Q = 1\%$, is the lower bound of the transparency for

1 $Q \geq 5\%$ because the decrease of the carrier density decreases G_N , which increases T_{int} . In
 2 Fig. 5, we plot the spin Hall conductivity σ_{SH} obtained under the assumption of $T_{\text{int}} = 0.44$
 3 for $Q \geq 5\%$. This result demonstrates that σ_{SH} decreases rapidly with decreasing σ_N for
 4 $Q \geq 5\%$, showing $\sigma_{\text{SH}} \propto \sigma_N^\gamma$ with $\gamma = 0.8$. Since T_{int} is expected to increase with decreasing
 5 Q under the above assumption, $\gamma = 0.8$ is the lower bound; γ can be larger than unity when
 6 $T_{\text{int}}(Q = 10\%)/T_{\text{int}}(Q = 5\%) > 1.9$.

7 Notable is that the scaling behavior of the spin Hall conductivity prevails not only in the
 8 dirty metallic regime but also in the disordered insulating regime. Figure 5 shows that the
 9 observed variation of the spin Hall conductivity of the PtO_x film in the insulating hopping
 10 regime ($\sigma_N < 10^3 \Omega^{-1}\text{cm}^{-1}$) is similar to that for Pt and Pt-based alloys in the dirty metallic
 11 regime (see the data obtained by the present and previous studies in the range of around
 12 $2 \times 10^3 \Omega^{-1}\text{cm}^{-1}$ to $2 \times 10^4 \Omega^{-1}\text{cm}^{-1}$ in Fig. 5). This result is reminiscent of the variation of
 13 the anomalous Hall conductivity in the dirty metallic and disordered insulating regimes. In
 14 the dirty metallic regime, the anomalous Hall conductivity is suppressed by the disorder due
 15 to the influence of finite-lifetime disorder broadening on the intrinsic contribution²⁹, which
 16 explains the scaling of the anomalous Hall conductivity in this regime. This mechanism is
 17 also responsible for the scaling of the spin Hall conductivity in the dirty metallic regime¹¹.
 18 For the AHE, experimental studies have uncovered that the same scaling holds even in the
 19 disordered insulating regime, despite the distinct difference in the transport mechanism².
 20 In the insulating hopping regime, the AHE arises from interference between direct ($i \rightarrow j$)
 21 and indirect ($i \rightarrow k \rightarrow j$) hoppings in a triad, where i and j are pairs of hopping sites, and
 22 k is the intermediate hopping site¹³. A similar mechanism can also give rise to the SHE in
 23 the insulating regime; the SHE in this regime arises when the hopping via an intermediate
 24 site is considered in addition to the hopping between pairs of sites³⁰. The observed scaling
 25 of the spin Hall conductivity over the wide range of electric conductivity demonstrates an
 26 important correspondence between the SHE and the AHE in both the metallic and insulating
 27 regimes.

28 In summary, we have demonstrated the crossover between the metallic and insulating
 29 regimes of the SHE by tuning the oxidation level of PtO_x . We found that the spin Hall
 30 conductivity in the lightly oxidized PtO_x is almost independent of the electric conductivity,
 31 which is consistent with the prediction of the intrinsic SHE in the moderately dirty metallic
 32 regime. By further increasing the oxidation level, the PtO_x film enters the dirty regime,

1 where the spin Hall conductivity decreases with decreasing the electric conductivity. We
2 found that the spin Hall conductivity varies systematically despite the drastic change of the
3 transport mechanism; the scaling of the spin Hall conductivity with the electric conductiv-
4 ity prevails not only in the dirty metallic regime but also in the insulating hopping regime.
5 This result is reminiscent of the scaling of the anomalous Hall conductivity. Previous stud-
6 ies have shown that the oxygen incorporation in Pt allows to explore SOC phenomena at
7 ferromagnetic-metal/PtO_x interfaces^{17,18,31–33}. Our result shows that the oxidation-level tun-
8 ing of PtO_x also offers a promising way to explore the phenomena arising from the bulk SOC
9 in spintronic devices. We believe that the observed crossover of the SHE provides essential
10 information for a deeper fundamental understanding of spin-orbit physics and stimulates
11 in-depth theoretical studies of the physics of spin transport in disordered systems.

1 Methods

2 Device fabrication.

3 The $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$ films were deposited on SiO_2 substrates by radio frequency (RF)
4 magnetron sputtering. We first deposited the PtO_x layer on the SiO_2 substrate in a mixed
5 argon and oxygen atmosphere. The amount of the oxygen gas in the reactive mixture Q
6 was varied between 0 and 10%. After the PtO_x deposition, the chamber was evacuated to
7 2×10^{-5} Pa. Then, the TiN layer was fabricated by introducing 10% nitrogen into the argon
8 gas flow. On the top of the TiN layer, the $\text{Ni}_{81}\text{Fe}_{19}$ film and a SiO_2 capping layer were
9 sputtered in a pure argon atmosphere. The resistivity of the TiN layer was determined from
10 measured resistance of $\text{Ni}_{81}\text{Fe}_{19}(10 \text{ nm})/\text{TiN}(2 \text{ nm})$ and $\text{Ni}_{81}\text{Fe}_{19}(10 \text{ nm})$ films by assuming
11 a two layer parallel circuit model. For the ST-FMR measurement, the $\text{Ni}_{81}\text{Fe}_{19}/\text{TiN}/\text{PtO}_x$
12 films were patterned into rectangular strips with a width of 10 μm and length of 100 μm
13 using the photolithography and lift-off techniques.

14 Data availability

15 The data that support the findings of this study are available from the corresponding author
16 upon reasonable request.

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7 **Additional information**

8 The authors declare no competing financial interests.

9 **Author contributions**

10 H.M. fabricated devices. H.M., A.M., S.H. collected and analyzed the data. K.A. designed
11 the experiments. K.A. and H.M. developed the explanation and wrote the manuscript. All
12 authors discussed results and reviewed the manuscript.

Figure 1. Device structure. **a** Schematic illustration of the SiO₂/Ni₈₁Fe₁₉/TiN/PtO_x/SiO₂-substrate device used for the ST-FMR measurement. **b** Oxygen-gas flow ratio Q dependence of the electric resistivity ρ_N , measured by the standard four-probe method, of the PtO_x film. **c** XPS spectra for the PtO_x films with $Q = 1, 5$, and 10% . The gray curve is the experimental data, and the red curve is the fitting result. The binding energies of the Pt 4f_{7/2} peak for Pt, PtO, and PtO₂ are around 71.3, 72.3, 74.0 eV, respectively³⁴.

Figure 2. Transport mechanism. Temperature T dependence of the sheet resistance R_s for the PtO_x films with (a) $Q = 1\%$, (b) $Q = 5\%$, and (c) $Q = 10\%$. We also compare the T dependence of R_s for the films with different Q in the inset to (a). The insets in (b) and (c) show $T^{-1/4}$ dependence of $\log R_s$, where $200 \text{ K} \leq T \leq 300 \text{ K}$. The solid circles are the experimental data and the solid lines are the linear fitting results.

Figure 3. Spin-torque ferromagnetic resonance. **a** Magnetic field H dependence of the DC voltage V_{DC} for the $Ni_{81}Fe_{19}/TiN/PtO_x$ films with $Q = 0, 6$, and 10% at the frequencies from 5.0 to 8.0 GHz. The $Ni_{81}Fe_{19}$ -layer thickness is $d_F = 8$ nm. **b** In-plane magnetic field angle θ dependence of the V_{sym} (red) and $V_{antisym}$ (blue) components of the ST-FMR signal for the $Ni_{81}Fe_{19}/TiN/PtO_x$ film with $Q = 10\%$. Here, θ is the angle from the longitudinal direction of the device. The circles are the experimental data. The solid curves are the fitting result using a function proportional to $\sin 2\theta \cos \theta$. **c** H dependence of V_{DC} for the $Ni_{81}Fe_{19}/TiN$ film, where the PtO_x layer is absent. **d** Q dependence of $M_s/M_s(Q = 0)$, where $M_s(Q = 0)$ is the saturation magnetization of the $Ni_{81}Fe_{19}/TiN/PtO_x$ film with $Q = 0\%$. **e** Frequency f dependence of ξ_{FMR}^{eff} for the $Ni_{81}Fe_{19}/TiN/PtO_x$ film with $Q = 10\%$ and $d_F = 12$ nm.

Figure 4. Spin-torque ferromagnetic resonance for devices with different thickness. ST-FMR spectra for the $Ni_{81}Fe_{19}/TiN/PtO_x$ films with (a) $Q = 0\%$ and (b) $Q = 6\%$ at 6.5 GHz, where d_F is the thickness of the $Ni_{81}Fe_{19}$ layer. H_{res} is the FMR field. The solid circles are the experimental data and the solid curves are the fitting results using Eq. (2). $1/d_F$ dependence of $1/\xi_{FMR}^{eff}$ for the devices with (c) $Q = 0\%$, (d) $Q = 6\%$, (e) $Q = 7\%$, and (f) $Q = 10\%$. The open circles are the experimental data. The solid lines are the linear fitting results.

Figure 5. Scaling of spin Hall conductivity. The spin Hall conductivity σ_{SH} of Pt and Pt-based alloys plotted as a function of the longitudinal electric conductivity σ_{N} . The solid circles in red are the experimental result of this work. Other data are taken from published papers: Pt^{9,10,23,35}, PtN³⁶, Pd_xPt_{1-x}³⁷, Pt_x(MgO)_{1-x}¹¹, and Au_xPt_{1-x}^{38,39}. The open circles and open squares represent the data obtained from SHE and inverse SHE measurements, respectively. For results where σ_{SH} is not explicitly shown, we estimated σ_{SH} from the spin Hall angle. The grey line is a guide for the eyes. The different background colors represent different regimes of the SHE: the superclean regime (blue), moderately dirty metallic regime (light blue), dirty metallic regime (orange), and disordered insulating regime (pink).