Giant spin Hall effect in perpendicularly spin-polarized FePt/Au devices

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Conversion of charge current into pure spin current and vice versa in non-magnetic semiconductors1–5 or metals6–8, which are called the direct and inverse spin Hall effects9–16 (SHEs), provide a new functionality of materials for future spin-electronic architectures17. Thus, the realization of a large SHE in a device with a simple and practical geometry is a crucial issue for its applications. Here, we present a multi-terminal device with a Au Hall cross and an FePt perpendicular spin injector to detect giant direct and inverse SHEs at room temperature. Perpendicularly magnetized FePt injects or detects perpendicularly polarized spin current without magnetic field, enabling the unambiguous identification of SHEs. The unprecedentedly large spin Hall resistance of up to 2.9 mΩ is attributed to the large spin Hall angle in Au through the skew scattering mechanism and the highly efficient spin injection due to the well-matched spin resistances of the chosen materials.

The origin of the spin Hall effect (SHE) is believed to be similar to that of the anomalous Hall effect (AHE) in ferromagnetic materials with spontaneous magnetization. According to theories of the AHE18, one of its origins is spin–orbit scattering of conduction electrons through skew and/or side-jump scattering by non-magnetic impurities, or through spin-disordered scattering19. A key to obtaining a large Hall angle ($\alpha_H$) is a large spin–orbit coupling parameter ($\eta_{so}$). The most distinguished point of the SHE is that it does not require ferromagnetic order. When an unpolarized charge current flows in a non-magnetic material with a large $\eta_{so}$, up- and down-spin electrons are scattered in opposite directions, thus generating a pure spin current in the transverse direction (without an accompanying charge current), which is called the direct SHE (DSHE). On the other hand, the flow of a spin current in the same material induces a transverse charge current, which is referred to as the inverse SHE (ISHE).

Theoretical studies based on the skew and/or side-jump scattering mechanisms20,11,15 and on the distinctive band structure in the presence of spin–orbit coupling21,22, and experimental studies on optically detected SHE in non-magnetic semiconductors such as GaAs (refs 1–4) and ZnSe (ref. 5) have stimulated much scientific interest and have raised the possibility of SHE-based devices. More recently, in non-magnetic metals such as Pt (refs 7,8) or Al (ref. 6), the SHE has been electrically detected by using the non-local spin-injection or spin-pumping technique. Pt is a well-known non-magnetic metal with a large $\eta_{so}$ and, indeed, a room-temperature SHE has been detected. However, its short spin diffusion length ($\lambda_s$) makes it very difficult to observe the SHE, so sophisticated device structures8 or measurement techniques are required. Al shows a relatively long $\lambda_s$, but a quite small $\eta_{so}$; therefore, the small ISHE is observed only at low temperature. In addition, a perpendicular magnetic field larger than the in-plane demagnetizing field is needed to magnetize a spin injector perpendicular to the film plane.

We fabricated a multi-terminal device consisting of a Au Hall cross and an FePt perpendicular spin injector as shown schematically in Fig. 1a. Au is a non-magnetic metal with a large $\eta_{so}$ and a simple electronic structure. The FePt spin injector with perpendicular magnetization generates perpendicularly (z directionally) polarized spin current flowing in the positive x direction without a perpendicular magnetic field. The spin current is reflected at the Au Hall cross, and charge accumulation occurs in the y direction, which is detected as the Hall voltage. The widths of the Hall cross and the spin injector are 150 nm and 200 nm, respectively, and the distance from the edge of the spin injector to that of the Hall cross (d) is varied in the range from 70 nm to 400 nm (Fig. 1b). The resistivities ($\rho$) of Au and FePt at 295 K are 2.7 $\mu$Ω cm and 36 $\mu$Ω cm, respectively.

An advantage of the present multi-terminal device is that three kinds of Hall effect, that is, DSHE, ISHE and local Hall effect (LHE), are measured in the same device by simply changing the terminal configuration of the bias current ($I$) and the voltmeter. When $I$ flows from terminal A to B, $I^{A-B}$, and the voltage ($V$) is measured between terminals C and D, $V^{C-D}$, the terminal configuration corresponds to the ISHE geometry. The resistance of the ISHE ($R_{ISHE} = V^{C-D}/I^{A-B}$) versus magnetic field ($H$) applied in the z direction is shown in Fig. 1c. For comparison, the hysteresis loop of the resistance of the LHE ($R_{LHE} = V^{B-C}/I^{A-F}$) is also shown. The shape of the hysteresis loop of $R_{ISHE}$ obtained at 295 K is the same as that of $R_{LHE}$. The LHE mainly originates from the AHE in FePt, indicating that the change of $R_{ISHE}$ reflects the magnetization reversal of FePt. The voltage change of the ISHE ($\Delta V = V_t - V_d$) varies linearly with $I$, where $V_t$ and $V_d$ are the voltages when the magnetic moment of FePt points upward and downward, respectively. From the absolute value of $\Delta V$ versus $I$, the resistance change of the ISHE ($\Delta R_{ISHE}$) is found to be...
The temperature dependence of $\Delta R_{\text{ISHE}}$ differs from that for Pt, which shows an increase of spin Hall signals as the temperature decreases. Because of the non-degenerate band dispersion of Au, the large SHE originates from the skew or side-jump scattering mechanisms. Assuming that the interface condition is transparent (metallic contact), $\Delta R_{\text{ISHE}}$ is phenomenologically expressed as\cite{16}

$$\Delta R_{\text{ISHE}} = 2\alpha_{\text{Fe}} \frac{R_{\text{Au}}}{R_{\text{Fe}}} \exp(-d/\lambda_{\text{Au}}),$$

where $\lambda_{\text{Au}}$ is the thickness of the Au Hall cross and $\alpha_{\text{Fe}}$ is the ratio of the spin Hall conductivity to the electrical conductivity. $P$ is the effective current spin polarization and is described as\cite{16}

$$P = \left\{ \frac{P_{\text{d}}}{1 - P_{\text{d}}} \left( \frac{R_{\text{Au}}}{R_{\text{Fe}}} \right) \right\} \left\{ 1 + \frac{2}{1 - P_{\text{d}}} \left( \frac{R_{\text{Au}}}{R_{\text{Fe}}} \right) \right\},$$

where $P_{\text{d}}$ is the bulk spin polarization of FePt and $R_{\text{Au}}^{\text{FePt(Au)}}$ is the spin resistance of FePt (Au). $P$ is found to be $\sim 0.01$ assuming $P_{\text{d}} = 0.3$–0.4 (refs 21–23) and using $R_{\text{Au}}^{\text{FePt(Au)}} = 0.04$ (see the Methods section). From the results of $\Delta R_{\text{ISHE}}$ as a function of $d$ shown in Fig. 2b, $\lambda_{\text{Au}}$ at 295 K is estimated to be $86 \pm 10$ nm. Consequently, $\alpha_{\text{Fe}}$ is found to be $\sim 0.113$. As mentioned above, $\alpha_{\text{Fe}}$ is

\[ \alpha_{\text{Fe}} = \frac{P_{\text{d}}}{1 - P_{\text{d}}} \left( \frac{R_{\text{Au}}}{R_{\text{Fe}}} \right) \left( 1 + \frac{2}{1 - P_{\text{d}}} \left( \frac{R_{\text{Au}}}{R_{\text{Fe}}} \right) \right). \]

This temperature dependence of $\Delta R_{\text{ISHE}}$ differs from that for Pt, which shows an increase of spin Hall signals as the temperature decreases. Because of the non-degenerate band dispersion of Au, the large SHE originates from the skew or side-jump scattering mechanisms. Assuming that the interface condition is transparent (metallic contact), $\Delta R_{\text{ISHE}}$ is phenomenologically expressed as

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strongly correlated with $\eta_{\text{Au}}$. The dimensionless spin–orbit coupling parameter ($\eta_{\text{Au}}$) is given by \[ \eta_{\text{Au}} = \frac{3\sqrt{3\pi}}{4} \frac{1}{\rho_{\text{Au}}} \frac{R_{\text{S}}}{k_f^2}, \]

where $R_{\text{S}}$ is the resistance quantum (25.8 k$\Omega$) and $k_f$ is the Fermi momentum of Au ($1.2 \times 10^{10}$ cm$^{-1}$). In the case of the present Au, $\eta_{\text{Au}}$ is calculated to be 0.3. Surprisingly, $\alpha_H$ for Au ($\sim 0.113$) is significantly larger than the value of 0.0037 for Pt (ref. 8). In the case when the side-jump scattering is dominant, $\alpha_H = \eta_{\text{Au}}/k_f I$, where $I$ is the electron mean free path, is calculated to be $\sim 1.5 \times 10^{-3}$, which is much smaller than the experimental value. On the other hand, $\alpha_H$ of the skew scattering is given by $\alpha_H = (2\pi/3)\eta_{\text{Au}}(V_{\text{imp}} \cdot N(0))$, where $V_{\text{imp}}$ and $N(0)$ are the impurity potential and the density of states. Using $\alpha_H = 0.113$ and $\eta_{\text{Au}} = 0.3$, $\langle V_{\text{imp}} \cdot N(0) \rangle$ is estimated to be 0.18. This small impurity potential is presumably reasonable because the low $\rho_{\text{Au}}$ indicates the high purity of the present Au. Therefore, we conclude that the main mechanism causing the large $\alpha_H$ in Au is the skew scattering. $\alpha_H$ weakly depends on temperature (Fig. 2a, inset), which supports that the skew scattering contribution is dominant because $\alpha_H$ decreases with $T$ in contrast with $\alpha_{\text{S}}$ not depending on $T$. We also emphasize that the well-matched $R_s$ leads to highly efficient spin injection. Au and FePt show a better match for $R_s$ compared with a conventional materials combination (see the Methods section), which is an important parameter for the large SHE as well as the large $\alpha_H$ in Au.

The terminal configuration of $I^{\text{C-D}}$ and $V^{\text{A-B}}$ is used for the detection of the DSHE (Fig. 3a, inset). In this configuration, the FePt spin injector serves as a spin detector for measuring the spin-dependent electrochemical potentials in Au induced by the DSHE (Fig. 3c), which is the same manner as the detection in a conventional non-local spin valve24,25. Figure 3a shows the resistance of DSHE ($R_{\text{DSHE}} = V^{\text{A-B}}/I^{\text{C-D}}$) while applying the current $I = -0.6$ mA, compared with $R_{\text{LHE}}$ as a function of $H$. As in the case of the ISHE, the observed $R_{\text{DSHE}}$ follows the magnetization reversal of FePt. $\Delta R_{\text{DSHE}}(2.2 \mu\Omega)$ is larger than $\Delta R_{\text{ISHE}}(1.5 \mu\Omega)$. For $I = \pm 0.6$ mA, however, $\Delta R_{\text{DSHE}}$ is quite small ($\sim 0.3$ m$\Omega$). To clarify the reason for the enhancement and reduction of $\Delta R_{\text{DSHE}}$ for $-I$ and $+I$, respectively, $V^{\text{A-B}}$ with $I^{\text{C-D}}$ was also measured. Even in this terminal configuration, hysteresis loops of $V$ were observed in both current directions. However, the polarity of $\Delta V$ did not depend on the current direction. In addition, $\Delta V$ increased as a parabolic function of $I$. We consider that $V^{\text{A-B}}$ with $I^{\text{C-D}}$ is the transverse voltage ($y$ direction), which is known as the anomalous Nernst–Ettingshausen effect26,27. As this thermomagnetic effect shows identical voltage polarity regardless of the current direction, we eliminate the extra effect by averaging $\Delta V_{\text{DSHE}}$, that is, $\Delta V_{\text{DSHE}} = (\Delta V_{\text{DSHE}} + \Delta V_{\text{DSHE}})/2$. The average $\Delta V_{\text{DSHE}}$ is found to be $\sim 1$ m$\Omega$, which is almost equal to $\Delta R_{\text{DSHE}}$. This thermomagnetic effect does not overlap on the ISHE because the Au Hall cross does not have spontaneous magnetization, which was confirmed using the terminal configuration of $V^{\text{C-D}}$ and $I^{\text{A-B}}$ (see the Supplementary Information).

The variation in $R_{\text{DSHE}}$ under a tilted magnetic field with respect to the $z$ axis is shown in Fig. 3b, where the magnetic field is rotated at an angle of $\phi$ in the $z$–$y$ plane. No change in $R_{\text{DSHE}}$ is observed for $\phi = \pi/2$, and the polarity reversal of $R_{\text{DSHE}}$ occurs between $\phi = 0$ and $\pi$. For the LHE, DSHE and ISHE, the switching field ($H_{\text{sw}}$) gradually increases with $\phi$. The variations in $H_{\text{sw}}$ normalized by $H_{\text{sw}}$ at $\phi = 0$ follow the $1/\cos\phi$ law (Fig. 3d), supporting that all of the Hall effects are signals from FePt showing magnetization reversal through domain wall propagation. All of the hysteresis loops of the ISHE (Fig. 1) and DSHE (Fig. 3) saturate with the magnetization of FePt, and the ordinary Hall effect induced by the external magnetic field is negligible in the Au Hall cross. In addition, the polarity of $R_{\text{DSHE}}$ for $I^{\text{A-B}}$ is the same as that for $I^{\text{C-D}}$, which indicates that there is no normal Hall effect owing to the stray field and the non-zero charge current in Au.

A giant $\Delta R_{\text{DSHE}}$ of 2.9 m$\Omega$ is achieved for $d = 70$ nm (Fig. 2b, inset), the magnitude of which is more than 10 times that of previous signals in other non-magnetic metals\textsuperscript{8,9}. The SHE signal can be further enhanced by higher current spin polarization using the tunnel spin injection\textsuperscript{26,25,27}. We believe that the large SHE in the present multi-terminal device paves the way for SHE-based spin-electronic devices.

**METHODS**

**THIN-FILM PREPARATION**

A continuous film with a stacked structure of FePt(10)/Au(10)/Pt(2) (in nanometres) was prepared on a MgO (001) single-crystal substrate using an ultrahigh-vacuum magnetron sputtering system (ULIVAC) with high-purity targets of Fe (99.99%), Pt (99.99%) and Au (99.9%). The base pressure was below $1 \times 10^{-9}$ torr and high-purity argon gas (>99.9999%) was introduced during sputtering. A typical deposition rate was 0.01 nm s$^{-1}$. First, the FePt layer was epitaxially grown on the MgO substrate heated to 573 K, and subsequently annealed at 773 K to promote the formation of the L1$_{0}$ ordered structure. The epitaxial growth of the FePt layer with the (001) preferential crystallographic
Figure 3 DSHE. a, Resistance of DSHE geometry $R_{\text{DSHE}}$ versus $H$ for the device with $d = 150$ nm superimposed on $R_{\text{LHE}}$ (red lines). The measurement is carried out at room temperature while applying $I = -0.6$ mA (top panel) and $+0.6$ mA (bottom panel). Inset: Schematic diagram of the terminal configuration for the DSHE. b, Angular dependence of $R_{\text{DSHE}}$ and $R_{\text{LHE}}$. The tilt angle $\phi$ with respect to the $z$ direction is rotated in the $x$-$y$ plane. c, Spatial map of electrochemical potentials for electrons with up- (blue) and down- (red) spin moments in Au and FePt for the DSHE with negative $I$. When FePt shows an upward local magnetic moment at a positive $H$, the electric potential in FePt is lower than that in Au. d, Normalized switching field $H_{\text{sw}}$ as a function of $\phi$ for $R_{\text{DSHE}}$, $R_{\text{GMR}}$, and $R_{\text{LHE}}$. $H_{\text{sw}}$ is normalized by $H_{\text{sw}}$ at $\phi = 0$. The dashed curve denotes the $1/\cos \phi$ law representing magnetization reversal through domain wall motion.

orientation was confirmed by X-ray diffraction. After the deposition of FePt, the substrate was cooled to ambient temperature, and the Au and Pt layers were deposited. The magnetization curve of the continuous film, which was measured using a superconducting quantum interference device magnetometer and the polar magneto-optical Kerr effect, showed strong perpendicular magnetic anisotropy. An important point in the present thin-film preparation is that the interface between the FePt and Au was formed in an ultrahigh-vacuum condition before the microfabrication process, which enabled us to keep the interface clean as it was not exposed to air during microfabrication.

DEVICE FABRICATION AND ELECTRICAL MEASUREMENT

Instead of a conventional lift-off process, an ion etching process was used for device fabrication. A negative-type electron beam resist (Tokyo Ohka Kogyo) was spin-coated on the continuous film, and was patterned into the shape of the FePt spin injector using electron beam lithography (ELS-7500, ELIONIX). After patterning of the electron beam resist, the continuous film was completely etched by argon ions through the resist mask. Subsequently, the patterned sample was returned to the ultrahigh-vacuum sputtering chamber to deposit a 10-nm-thick Au layer on the sample surface. Finally, the Au layer was patterned into the shape of the Hall cross by electron beam lithography and ion etching. The ion etching was monitored by secondary-ion mass spectroscopy.

The device resistance was simply measured by a nanovoltmeter (34401A, Agilent) while applying a d.c. current. The external magnetic field was applied perpendicularly to the sample plane. Measurements were carried out at 295 K, 77 K and 4.2 K.

CALCULATION OF SPIN RESISTANCE

The spin resistance $R_S$ is given by $R_S = (\rho \lambda)/A$, where $\rho$, $\lambda$ and $A$ are the resistivity, the spin diffusion length and the effective cross-sectional area, respectively. $R_{\text{LHE}}$ is found to be 1.62 $\Omega$ with $A_{\text{Cu}} = 1.5 \times 10^{-12}$ m$^2$ (thickness $\times$ width). The case of $R_{\text{DSHE}}$, on the other hand, the current distribution should be taken into account because of the short $\lambda_{\text{FePt}} = 5$ nm, which is estimated from the current-perpendicular-to-plane giant magnetoresistance in the FePt layers. Using $A_{\text{FePt}} = 3 \times 10^{-14}$ m$^2$ (contact area between FePt and Au), $R_{\text{DSHE}}$ is found to be 0.06 $\Omega$. Therefore, $R_{\text{DSHE}}/R_{\text{LHE}}$ is 0.04.

Note that the ratio of $\rho_{\text{Cu}}/\lambda_{\text{FePt}}$ to $\rho_{\text{FePt}}/\lambda_{\text{Cu}}$ is quite high (0.7) compared with that of a conventional materials combination, for example, $\rho_{\text{Pd}}/\rho_{\text{Cu}}$ or $\rho_{\text{Cu}}/\rho_{\text{Fe}}$ is around 0.04 for permalloy (Py) and Cu, where the values reported in ref. 8 are used to calculate Py/Cu and Au is taken into account because it depends on the device geometry and size. These calculations indicate that the combination of FePt and Au is well suited for highly efficient spin injection.

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References


