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Nanometre-thick Si/Al Gradient Materials for Sustainable Spin Current Generation

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ABSTRACT

Green materials for electron spin manipulation are essential for common spintronic applications. Recent studies have documented the efficient generation of spin torque using spin-orbit interactions (SOIs); however, SOI use relies on the employment of rare metals such as platinum. Here, we demonstrate that a nanometre-thick gradient from silicon to aluminium, which consists of readily available elements from earth resources, can produce a spin torque over three times as large as that of platinum, despite the weak SOI of these compositions. The spin-torque efficiency can be improved by decreasing the thickness of the gradient, while a sharp interface was not found to increase the spin torque. Moreover, the electric conductivity of the gradient material can be up to twice as large as that of platinum, which provides a way to reduce Joule heating losses and signal delays in spintronic device circuits. Our findings represent a pathway for sustainable spin manipulation.
origin is still controversial\textsuperscript{29–32}. Because a conductivity gradient leads to a vortical electric current that acts on spins as an emergent magnetic field (Fig. 1b), the magnitude of the field should depend on the thickness of electron conductivity gradient in oxides. However, the thickness of the oxidation gradient is difficult to control atomically. This poses a problem not only for practical applications of spintronic devices but also for confirming the existence of emergent magnetic fields due to electric current vorticity.

In this study, we successfully fabricated a nanometre-thick artificial gradient from silicon (Si) to aluminium (Al), which constitutes the second- and third-most abundant components in the Earth’s crust (Fig. 1c), and we demonstrate its ability to generate a spin torque of greater magnitude than Pt. The magnitude of the spin torque increases when the compositional gradient steepens; however, an atomically sharp interface does not significantly increase the spin torque. Moreover, as reported in the study of a surface-oxidised copper (Cu) film\textsuperscript{28}, we observed a large non-reciprocity that is regarded as evidence for a SC generated via an emergent magnetic field due to electric current vorticity.

\section*{Fabrication and structural analysis of Si/Al gradient materials}

We fabricated Si(10)/Al(t\textsubscript{i}/2)/Si(t\textsubscript{i}/2)/Al(10)/Ni\textsubscript{85}Cu\textsubscript{5}(10)/SiO\textsubscript{2}(20) (unit: nm) multilayer strips on a thermally oxidised Si substrate by means of a conventional lift-off method using magnetron sputtering and photolithography. Here, \( t_i \) is the thickness of the interfacial Al/Si insertion, which varied from 0.5 to 2.0 nm at intervals of 0.5 nm. Al is a very conductive metal with an open 2p shell. Although Si is adjacent to Al in the periodic table, it is a semiconductor with much lower electrical conductivity than Al. Sputter deposition processes generally lead to atomic or metallographic disturbances at an interface due to the large kinetic energy of the sputtered particles. An insertion of a few nm of Al/Si therefore increases the mixed region at the interface between 10-nm-thick Si and Al layers. Indeed, microstructural analysis using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) revealed an increase in the thickness of the compositional gradient from Si to Al with increasing \( t_i \). Figure 2a,b displays cross-sectional HAADF-STEM images near the Si/Al interface with \( t_i = 2.0 \) and 1.0 nm, respectively. In comparison, a cross-section of a Si/Al interface without Al/Si insertion (i.e. \( t_i = 0 \) nm) is also shown in Fig. 2c, in which a sharp interface was clearly observed. The Al/Si insertion (with thickness \( t_i \)) obscures the Si/Al boundary. Moreover, aggregation of Al and/or Si was observed at the Si/Al interface for \( t_i = 2.0 \) nm. The formation of such aggregation features is attributed to non-solid solution atomic mixing between Si and Al. Energy dispersive spectroscopy (EDS) line profiles of each element (Si and Al) were obtained by averaging the signals in the dashed box area shown in Fig. 2d,e,f which explicitly confirm the formation of a Si/Al gradient with a transition thickness that systematically varies with \( t_i \). The transparent bold lines in Fig. 2g,h,i show the best fit to the following equation:

\[
\text{Composition} = \frac{C_1 + C_2}{2} \pm \frac{C_2 - C_1}{2} \tanh \left( \frac{z - z_{\text{int}}}{L} \right),
\]  

where \( C_1 \) and \( C_2 \) are the composition at each end of an EDS scan, \( z_{\text{int}} \) is the centre position of the interface, and \( L \) is the thickness of the compositional gradient from Si to Al. The fit of Eq. (1) indicates \( L \) values of 1.3 and 2.4 nm for \( t_i = 1.0 \) and 2.0 nm, respectively. As shown in Fig. 2, some compositional fluctuations occur along the interfaces, especially for the sample with \( t_i = 2.0 \) nm; thus, the value of \( L \) depends on both the size and position of the EDS signal-averaging window. The value of \( L \) determined in Fig. 2 should be regarded as an averaged thickness of the compositional gradient. It is known that Al and Si form a typical non-solid solution combination at equilibrium. Few intermetallic compound or intermediate phases of these elements exist, although certain non-equilibrium states can form during the sputter deposition process at room temperature. Figures 2j and 2k are nanobeam electron diffraction (NBED) patterns for 10-nm-thick Si and Al layers, respectively. A broad halo pattern from the amorphous structure of the Si layer is present, whereas diffraction spots corresponding to the polycrystalline nature of the Al layer appear in the high-resolution HAADF-STEM image in Fig. 2l.

\section*{Electric current-induced spin torque}

When an electric current is applied to the bilayer strip consisting of a nonmagnet (NM) and a ferromagnet (FM), part of the SC generated in the NM is transmitted towards the FM followed by a spin torque applied on the magnetisation of the FM. This torque is generally referred to as a damping-like (DL) torque, \( \tau_{\text{DL}} \), whereas a separate spin torque known as the field-like (FL) torque, \( \tau_{\text{FL}} \), arises from the SC reflected at the NM/FM interface. The effect of these torques on a magnetisation \( \mathbf{m} \) produced by a SC with a polarisation \( \mathbf{\sigma}_s \) is described by the following equation:

\[
\tau = \tau_{\text{DL}} + \tau_{\text{FL}} = \xi_{\text{DL}} \frac{\hbar}{2e} \frac{1}{\mu_0 M_s d_{\text{FM}}} \mathbf{m} \times (\mathbf{m} \times \mathbf{\sigma}_s) + \xi_{\text{FL}} \frac{\hbar}{2e} \frac{1}{\mu_0 M_s d_{\text{FM}}} \mathbf{m} \times \mathbf{\sigma}_s,
\]  

where \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) are the efficiencies of the DL and FL torques with respect to an electric current density \( j_c \), while \( e, \mu_0, \) and \( \hbar \) are the elementary charge, permeability of a vacuum, and the reduced Planck’s constant, respectively. \( M_s \) and \( d_{\text{FM}} \)
are the saturation magnetisation and FM thickness, respectively. Both these orthogonal torques play an important role in magnetisation switching, which has been widely investigated in applications for non-volatile magnetoresistive memory and magnetic logic devices.

To evaluate the strength of the spin torque produced by applying an electric current to the sample, we conducted spin-torque ferromagnetic resonance (ST-FMR) measurements\textsuperscript{15,16,18,33–35}. The theory of this technique is described in Supplementary note S1. In the ST-FMR experiment, an alternating current was applied to the strip using a microwave with amplitude of 20 dBm and a frequency of 20 GHz. The direct current voltage \( V_a \) magnitude due to the ST-FMR excitation was measured while sweeping the external field from 0 to 2.0 T. Figure 3a shows a typical ST-FMR spectrum for \( t_i = 0.5 \) nm, which can be reproduced by a combination of symmetric- and antisymmetric-Lorenzian functions, as shown in Fig. 3b. From the amplitude ratio between the symmetric- and antisymmetric-Lorenzian components, \( V_a/V_s \), we can evaluate the spin torque efficiency \( \xi_{\text{FMR}} \), which is commonly used as a rough estimate of \( \xi_{\text{DL}} \) when \( \xi_{\text{FL}} \) is negligible in ST-FMR. From Eq. (S1) in Supplementary note S1 and the value of \( V_a/V_s \) for the ST-FMR spectrum at a given resonant field \( B_r \), we obtain \( \xi_{\text{FMR}} = 0.029 \pm 0.004 \) for a sample with \( t_i = 0.5 \) nm, whose magnitude is about half the value for a Pt(10 nm)/Ni/Al(10 nm) bilayer film \( (\xi_{\text{FMR}} = 0.066 \pm 0.005) \). Figure 3c shows \( \xi_{\text{FMR}} \) as a function of \( t_i \). As shown in Fig. 3c, a lower \( t_i \) leads to a larger \( \xi_{\text{FMR}} \), although the \( \xi_{\text{FMR}} \) for \( t_i = 0 \) nm is similar to the value for a reference sample with an Al(10 nm)/Ni/Al(10 nm) structure fabricated on a thermally oxidised Si substrate. This result implies that most of the SC is produced in the 10-nm-thick Al region via SHE and that a sharp Si/Al interface does not result in additional SC generation. That is, the nanometre-thick gradient from Si to Al is the most important factor for SC generation in the sample.

In general, a large difference in electric conductance between NM and FM means that the contribution of \( \xi_{\text{FL}} \) cannot be ignored. As shown in Supplementary note S3, both \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) can be determined from the dependence of \( \xi_{\text{FMR}} \) on \( d_{\text{FM}} \). Furthermore, the ratio between \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) generally depends on the condition of a NM/FM interface, i.e. an Al/Ni/Al(10 nm) interface. In other words, the value of \( \xi_{\text{FL}}/\xi_{\text{DL}} \) is independently determined with respect to the thickness of the compositional gradient at the Si/Al interface. Figure 3d contains a plot of \( \xi_{\text{FMR}} \) as a function of \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \), that is expected from Eq.(S2) of Supplementary note S3. From the \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) for \( t_i = 1.0 \) nm in Supplementary note S3, \( \xi_{\text{FL}}/\xi_{\text{DL}} \) was determined to be 3 for the Al/Al(10 nm) interface. The dashed line in Fig. 3d indicates the condition \( \xi_{\text{FL}}/\xi_{\text{DL}} = 3 \). The solid circles on the dashed line in Fig. 3d indicate the experimental \( \xi_{\text{FMR}} \) values obtained in Fig. 3c. The \( x \)- and \( y \)-axes values for each circle in Fig. 3d therefore represent \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) of each \( t_i \), respectively. As shown in Fig. 3d, the magnitudes of both \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) clearly increase with decreasing \( t_i \), which is the thickness of the compositional gradient from Si to Al. Surprisingly, the \( \xi_{\text{DL}} \) value for \( t_i = 1.0 \) nm reaches a magnitude three times larger than that of Pt; further increase in \( \xi_{\text{DL}} \) would be expected for \( t_i = 0.5 \) nm, even though only weak SOI elements, i.e. Si and Al, were used for the NM. The ratio between \( \xi_{\text{DL}} \) and \( \xi_{\text{FL}} \) suggests that the reflected SC is three times larger than the transmitted SC. This is attributed to the fact that electric conductivity in the SC absorber, the Ni/Al(10 nm) layer, is an order of magnitude smaller than that of the SC injector, or the Al layer. Further increases in \( \xi_{\text{DL}} \) can therefore be realised using ferromagnetic materials with a higher electrical conductivity than Ni/Al(10 nm).

**Origin of spin torque accompanied by Si/Al gradient materials**

Below we discuss the origin of spin torque in the Si/Al/Ni/Al(10 nm) trilayer film. Similarly to a torque generated via SHE and/or REE with an electric current along the \( x \)-axis, the variations of \( V_a \) and \( V_s \) with respect to the direction of the external magnetic field can be explained by assuming that the electron spin is polarised along the \( y \)-axis (see Supplementary note S4)\textsuperscript{36–38}. In the Si/Al/Ni/Al(10 nm) trilayer film, most of the electric current flows not within the Si or Ni/Al(10 nm) layer, but in the Al layer because the electric conductivity of Al is much larger. However, the spin torque produced by the electric current flow in the Al layer via SHE can be ignored because the spin Hall angle for bulk Al is only 0.028,\textsuperscript{39} which is 10 times smaller than the value of \( \xi_{\text{DL}} \) in our sample with \( t_i = 1.0 \) nm. Inversion symmetry breaking at the Al/Ni/Al(10 nm) interface can also produce a spin torque. However, this effect should be independent of \( t_i \) because the atomic and/or structural asymmetry condition at the Al/Ni/Al(10 nm) interface is determined by the sequential sputtering processes of Al and Ni/Al(10 nm), which are commonly used in all samples. Consequently, the dependence of \( \xi_{\text{DL}} \) on \( t_i \) is not attributed to a spin torque caused by either bulk or interface SOI. An inversion symmetry breaking leading to an orbital angular momentum (OAM) flow is also enhanced at the Si/Al interface by decreasing the transition width and must be strongest at \( t_i = 0 \) nm, while the smallest \( \xi_{\text{DL}} \) was observed at \( t_i = 0 \) nm, as shown in Fig. 3d. Therefore, generation of OAM flow is not the primary cause of the increase in \( \xi_{\text{DL}} \) for the compositional gradient from Si to Al. Similarly, interface-generated SCs caused by either spin-orbit filtering or spin-orbit precession\textsuperscript{40} can also be excluded.

An additional possible explanation for the increase in \( \xi_{\text{DL}} \) is SC generation from electric current vorticity via spin vorticity coupling (SVC)\textsuperscript{22,23,41,42}. A non-uniform distribution of electric current arises in the nanometre-thick Si/Al gradient from the large difference in electrical conductivity between Si and Al. Similarly to the inversion symmetry breaking, the degree of electric current non-uniformity increases with decreasing compositional gradient thickness. Conversely, specular electron scattering, which occurs at sharp interfaces, would create a uniform electric current distribution near the interface. Consequently, the electric current vorticity will be maximised when compositional gradient thickness is comparable to the effective vorticity length尺度.
mean free path of the electron. The optimisation of $\xi_{DL}$ at a certain compositional gradient thickness seems to support the SVC mechanism. Indeed, as shown in Supplementary note S5, the ratio of $\xi_{DL}$ increases with respect to the compositional gradient thickness by a similar order of magnitude to the value obtained by evaluating a simple model based on SVC theory. Fluctuations in the compositional gradient that disrupt the electric current vorticity are likely to be small because the strength of $\xi_{DL}$ clearly scales with the averaged compositional gradient thickness, as shown in Fig. 3d.

Highly non-reciprocal SC generation in Si/Al gradient materials

A highly non-reciprocal conversion between electric current and SC can be considered additional evidence that SVC contributes to SC generation. In the case of typical SOI-based phenomena, both intrinsic and extrinsic spin-dependent scattering sources exist in the material regardless of the applied electric current. On the contrary, the vorticity gradient needed for SC generation via SVC appears only when a net drift velocity exists. Namely, the SC is not accompanied by a net drift velocity and, consequently, cannot be converted into electric current.

To evaluate the conversion efficiency from a SC to electric current, as shown in Supplementary note S6, we measured the inverse SHE due to the SC \(^{43,44}\), which was produced by applying an alternating magnetic field \(^{45-48}\). Figure 4a shows the conversion efficiency $\theta_{j_k\rightarrow j_s}$ of the SC to an electric current as a function of $\xi_{DL}$, which is proportional to the conversion efficiency $\theta_{j_k\rightarrow j_s}$ from the charge current to the SC. In general, an SC source with a two-dimensional geometry, such as an interfacial SOI, suppresses $\theta_{j_k\rightarrow j_s}$ rather than $\theta_{j_s\rightarrow j_k}$, although the value of $\theta_{j_s\rightarrow j_k}/\theta_{j_k\rightarrow j_s}$ cannot exceed $10^{49,50}$. If the SC source does produce such a geometrical effect in the Si/Al gradient material, the non-reciprocity is expected to be intermediate between that of a bulk and an interfacial SOI because of its fuzzy interface. However, as shown in Fig. 4a, the sample with $t_i = 0.5$ nm exhibits a $\xi_{DL}/\theta_{j_k\rightarrow j_s}$ value much larger than 10. On the other hand, $\xi_{DL}/\theta_{j_s\rightarrow j_k}$ for $t_i = 0$ nm is unity, at which the highest two-dimensional geometry is expected. The fact that strong non-reciprocity is observed, not for $t_i = 0$ nm but for $t_i = 0.5$ nm, therefore suggests that the SC is generated by neither interfacial SOI nor inversion symmetry breaking at the Si/Al interface, but by angular momentum conversion from macroscopic vorticity in the electric current at the compositional gradient.

Figure of merit for spin torque switching capability of Si/Al gradient materials

To suppress the power-supply voltage for spin torque switching, the product of $\xi_{DL}$ and the electrical conductivity $\sigma_e$ of the NM must increase. Moreover, the magnitude of $\sigma_e$ itself should increase to reduce resistive-capacitive delays, which hinder the high-speed operation of integrated circuits. The relative abundance of elements $P_n$ is also an important parameter for sustainable development of SC-generating materials. To satisfy these demands, a figure of merit (FOM) for SC-generating materials can be determined as the product of these values as follows:

$$FOM = (\xi_{DL}\sigma_e) \times \sigma_e \times P_n = [\text{Spin torque efficiency}] \times [\text{Circuit performance}] \times [\text{Element sustainability}]$$

From the statistical data for $P_n$ in Fig. 1a, $P_{Si/Al} = P_{Si} \times P_{Al} = 2.24 \times 10^{-2}$, which is seven orders of magnitude larger than the value for Pt ($P_n$ of $5 \times 10^{-9}$). Si/Al gradient materials therefore offer a highly superior solution for sustainable materials development. Figure 4b displays a double logarithmic graph with respect to $\sigma_e$ and $\xi_{DL}\sigma_e$ for the Si/Al gradient samples. The data for a Pt/NiCu bilayer film are also plotted as a closed square for comparison. $\sigma_e$ is shown as a function of $t_i$ in Supplementary note S8. The dashed lines in Fig. 4b represent contours of $\xi_{DL}\sigma_e^2$. In addition to $\xi_{DL}\sigma_e$, we can also increase $\sigma_e$ in our Si/Al gradient materials by decreasing $t_i$ from 2.0 to 0.5 nm. This is surprising because strong SOI materials such as Pt, W, and Ta, typically exhibit low $\sigma_e$. More specifically, $\xi_{DL}\sigma_e^2$ can be larger than the typical value for strong-SOI materials even at the same value of $\xi_{DL}$. Indeed, we achieved a $\xi_{EMR}\sigma_e^2$ value of 14.3 (MS/m)\(^2\) for $t_i = 0.5$ nm, which is seven times larger than that in Pt. Moreover, the FOM for this material was $3.2 \times 10^{11}$ (S/m)\(^2\), which is seven orders of magnitude greater than that of Pt ($1.0 \times 10^4$ (S/m)\(^2\)). The overwhelmingly higher FOM for our fabricated films demonstrates the high potential for Si/Al gradient materials to be widely used in low-energy consumption and sustainable spin devices.
Fig. 1 | Green and rare-metal-free composition gradient material for spin current generation.

**a.** Schematic illustration of the Stern-Gerlach experiment. Quantisation of spin angular momentum $\sigma_s$ was observed when a beam of silver atoms was split in two as it passed through a gradient of a static magnetic field $B$. **b.** Schematic image of a fabricated Si/Al gradient material. The Cartesian coordinate system used throughout this article is also shown in (b). A nanometre-thick compositional gradient from Si to Al exists along the $z$-axis with much conductivity in the Al layer than the Si layer. When an electric current $j_x$ is applied along the $x$-axis, an electric current vorticity $\omega = \nabla \times j_x$ appears, whose vector points along the $y$-axis. The electron spin $\sigma_s$ in the gradient material is polarised along the $y$-axis by an emergent magnetic field due to the vortical electric current. As a consequence, the gradient of $\omega$ leads to a density gradient in the non-equivalent electron spin, which produces a spin current along the $z$-axis. **c.** A plot of relative elemental abundance in the Earth’s crust. Si and Al represent the most abundant semiconductor and metal materials, respectively, while typical large spin current source materials such as Ta, W, and Pt are very rare.
Fig. 2 | Microstructural and compositional characterisation of gradients across the Si/Al interface. a-c.
HAADF-STEM images of samples with (a) $t_i = 2.0$, (b) 1.0, and (c) 0.0 nm. d-f. Enlarged views of the sample near the area analysed by EDS. EDS profiles were generated from within the orange dashed boxes. g-i. Averaged profiles of the atomic concentration of Si and Al for each sample along the arrows in each box. j,k, Nanobeam diffraction patterns for Si and Al layers, respectively. l, High-resolution HAADF-STEM image of the Al layer. Dashed lines indicate the grain boundaries.
**Fig. 3 | ST-FMR experiment for evaluating electric-current-induced spin torque.**

**a.** ST-FMR spectrum measured for a Si/Al/Ni$_{95}$Cu$_{5}$ trilayer film with $t_i = 0.5$ nm at a microwave frequency of 20 GHz. The dashed curve represents the best-fit result for a combination of symmetric- and antisymmetric-Lorentzian functions.  

**b.** Symmetric (purple) and antisymmetric (blue) Lorentzian components included in (a). 

**c.** Spin torque efficiency $\xi_{\text{FMR}}$ as a function of $t_i$. The vertical bars indicate the standard deviation calculated from the least-squares deviation of the fitting parameters used in the calculation of $\xi_{\text{FMR}}$. The closed circles indicate the $\xi_{\text{FMR}}$ values of samples with $t_i$ ranging from 0 to 2.0 nm. The horizontal dashed line shows the $\xi_{\text{FMR}}$ for a reference sample with an Al(10 nm)/Ni$_{95}$Cu$_{5}$(10 nm) structure. 

**d.** Colour plot of $\xi_{\text{FMR}}$ as a function of $\xi_{\text{DL}}$ and $\xi_{\text{FL}}$. The dashed line indicates the condition $\xi_{\text{FL}} = 3\xi_{\text{DL}}$, which was determined by measuring the Ni$_{95}$Cu$_{5}$ layer thickness dependence of $\xi_{\text{FMR}}$ for a sample with $t_i = 1.0$ nm (see Supplementary note S3). The closed circles are each labelled with the $t_i$ value.
Fig. 4 | Figure of merit for Si/Al gradient material. a. Conversion efficiency from SC to charge current $\theta_{j_s \rightarrow j_c}$ as a function of $\xi_{DL}$. The circles show data for samples with various $t_i$, whereas the square represents values for a Pt(10 nm)/Ni$_{95}$Cu$_{5}$(10 nm) bilayer film. The vertical bars indicate the standard deviation calculated from the least-squares deviation of the fitting parameters used to calculate $\theta_{j_s \rightarrow j_c}$. b. Double logarithmic plot of electric conductivity $\sigma_e$ and $\xi_{DL}\sigma_e$ for samples with $t_i$ ranging from 0 to 2.0 nm. The data for a Pt(10 nm)/Ni$_{95}$Cu$_{5}$(10 nm) bilayer film is also plotted for comparison(square). Dashed lines are contours of $\xi_{DL}\sigma_{e}^{2}$. 
Methods

Sample Preparation
The films were fabricated on thermally oxidised silicon substrates by magnetron sputtering at room temperature. The chamber base pressure prior to deposition was less than $5.0 \times 10^{-4}$ Pa. The deposition pressure was 0.22 Pa with an argon (Ar) flow rate of 4.0 sccm. The Al layer creation used radio-frequency (RF) deposition at 13.56 MHz with a power density of 1.4 W/m² and a deposition rate of 0.043 nm/s from a 99.9%-pure Al target. The Si layer was deposited by RF-sputtering with a power density of 3.5 W/m² and a deposition rate of 0.062 nm/s from a non-doped Si target. The Ni₉₅Cu₅ layer was deposited by direct current (DC) sputtering with a power density of 1.4 W/m² and a deposition rate of 0.2 nm/s from a 99.9%-pure Ni₉₅Cu₅ alloy target. The SiO₂ capping layer was deposited by RF-sputtering with a power density of 3.5 W/m² and a deposition rate of 0.044 nm/s from a 99.99%-pure SiO₂ target. The thin films were patterned into strips 10-µm-wide and 100-µm-long by photolithography and lift-off processes. An electrically shorted coplanar waveguide made from 70-nm-thick Au was connected to both ends of the strip to conduct the ST-FMR measurements. All measurements were conducted at room temperature.

Electrical Measurements
For ST-FMR measurements, we applied 20-dBm continuous sinusoidal signals with a frequency of 20 GHz in the longitudinal direction ($x$-axis) of the film by a signal generator. An in-plane external magnetic field was applied with an amplitude ranging from 0 to 2.0 T at a fixed angle of $\pi/4$ with respect to the $x$-axis. We then measured the rectified DC voltage $V_{dc}$ by a nanovoltmeter from a DC port of bias tee. The resulting spectra were fit by the expression $V_{dc} = V_{s}f_s(B) + V_{a}f_a(B)$, where $f_s(B)$ and $f_a(B)$ are the symmetric- and antisymmetric-Lorentzian functions, respectively. The inverse SHE measurement used a general spin-pumping experiment setup. We applied a continuous sinusoidal signal with amplitude of 20 dBm and a frequency of 5 GHz into the coplanar waveguide fabricated on the sample, which was patterned into a Hall-bar shape. The sample and coplanar waveguide were insulated by the insertion of 120-nm-thick SiO₂ film. The external magnetic fields with amplitudes ranging from 0 to 2.0 T were applied in the $x$-$y$ plane at an angle ranging between 0 and $2\pi$ from the $x$-axis. The DC inverse spin Hall voltage was measured using a nanovoltmeter.

Electron microscopy characterisation
We prepared cross-sectional thin specimens for HAADF-STEM characterisation using a focused ion beam with a Ga⁺ ion source on a FEI Helios G4 UX instrument. Before the milling process, a ~5 nm-thick Au layer was deposited on the film surface to protect and enhance the conductivity of sample during milling. The lift-out lamellae were thinned to ~100 nm at 30 kV with a current decreasing from 0.75 nA to 90 pA, followed by final polishing at 2 kV and 17 pA. HAADF-STEM and NBED images were obtained using a Cs-corrected FEI Titan G2 80–200 equipped with a Super-X EDS. The HAADF-STEM images were collected using a convergence semi-angle of 18 mrad and an inner-collection semi-angle of 55 mrad. EDS mapping was performed using a Bruker Esprit analysis system with automatic drift correction during the collection process, which augments the reliability of the compositional analysis. Integrated line profiles were conducted across the heterostructure in EDS maps to enhance the signal-to-noise ratio. The Cliff–Lorimer analysis method was applied to quantify the EDS line-scan results.

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**Author contributions**

T.H. and Y.N. planned the study and wrote the manuscript. T.H. performed film deposition and fabricated the devices. C.H. and T.O. performed the microscopy experiments with the help of K.H. T.H. carried out the measurements and analysed the data with the help of K.Y. and Y.N. T.H., J.F., M.M., and Y.N. performed the numerical calculation based on SVC theory. All the authors discussed the results and commented on the manuscript.
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