

Crystallographic spin torque conductivity tensor of epitaxial IrO₂ thin films for oxide spintronics

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Abstract

Unconventional spin-orbit torques arising from electric-field-generated spin currents in anisotropic materials have promising potential for spintronic applications, including for perpendicular magnetic switching in high-density memory applications. Here we determine all the independent elements of the spin torque conductivity tensor allowed by bulk crystal symmetries for the tetragonal conductor IrO₂, via measurements of conventional (in plane) anti-damping torques for IrO₂ thin films in the high-symmetry (001) and (100) orientations. We then test that rotational transformations of this same tensor can predict both the conventional and unconventional anti-damping torques for IrO₂ thin films in the lower-symmetry (101), (110), and (111) orientations, finding good agreement. The results confirm that spin-orbit torques from all these orientations are consistent with the bulk symmetries of IrO₂, and show how simple measurements of conventional torques from high-symmetry orientations of anisotropic thin films can provide an accurate prediction of the unconventional torques from lower-symmetry orientations.

Introduction

The linear-response spin current generated by an applied electric field within a material is described by a third-rank spin Hall conductivity (SHC) tensor σ_{ij}^s , where s is the index for the spin polarization direction, i for the spin flow direction, and j for the applied electric field direction. If the spin current is absorbed by an adjacent magnetic layer, it will apply a torque that can efficiently reorient the magnetization. The 27 elements within σ_{ij}^s are often highly constrained by symmetry, and in commonly-used high-symmetry materials most of the elements are zero because symmetries can require the spin polarization, spin flow, and electric field to be mutually orthogonal¹. We will term the torques generated by such a spin current as “conventional”. For some applications, however, unconventional spin torques are highly desired. In particular, out-of-plane anti-damping torques associated with tensor elements of the form σ_{zj}^z (where z is the direction normal to the device plane) are needed to drive efficient anti-damping switching of magnetic memory devices with perpendicular magnetic anisotropy. Such out-of-plane anti-damping torques have been realized using spin-source materials in which the symmetry constraints are relaxed by very low crystal symmetries or magnetic ordering^{2–13} or by interfacial effects^{14–16}. We have also recently proposed a simple alternative strategy for generating out-of-plane anti-damping torques -- to use a relatively high-symmetry but anisotropic material as the spin source (e.g., a tetragonal or orthorhombic structure) and to grow thin films with a growth axis tilted away from any high symmetry direction (e.g., tilted in a (101) or (111) orientation). In this case, the tilt of the crystal axes relative to the sample plane can break the necessary symmetries to allow a nonzero value for σ_{zj}^z and an associated unconventional torque. We demonstrated this qualitatively in ref. [13] for the tetragonal material IrO₂. Here we test this approach quantitatively. By measuring the electric-field generated spin torque for (001) and (100) thin films of IrO₂ we determine all of the independent elements of the spin torque conductivity tensor associated with the absorbed spin current, and then we test whether rotational transformations of this single tensor can provide consistent quantitative predictions for the torques in three lower-symmetry film orientations: (110), (101), and (111). We find excellent agreement between the measurements and the predictions of the rotated tensor for both conventional and unconventional torques in all 5 crystal orientations.

Results and Discussion

First, one note regarding notation. The electric-field-generated torque applied to the magnetic layer in a spin-source/magnet bilayer will differ from the spin current generated in the spin-source layer by an interfacial transmission coefficient which describes what fraction of the spin current is absorbed by the magnetic layer. Spin-torque experiments therefore do not measure the spin current directly. However, if the interfacial transmission coefficient is to a good approximation a constant ($T < 1$) that does not depend on the thin-film orientation or the spin orientation, the spin torque conductivity (STC) tensor is simply proportional to the bulk spin Hall conductivity tensor: $\tau_{ij}^s = T\sigma_{ij}^s$. Our analysis is not reliant on this assumption, however, and in fact one can view the purpose of the paper as being in part to test this assumption, i.e., to test the degree to which rotational transformations of a single STC tensor can give an accurate description of the measured torques for different crystal orientations.

The bulk crystal symmetries for the tetragonal structure of rutile IrO₂ dictate that the spin Hall conductivity tensor can be defined in terms of three independent elements. For the

corresponding STC tensor we will call these elements a , b , and c . If we define basis vectors in terms of the IrO_2 crystal axes as $X = [100]$, $Y = [010]$, and $Z = [001]$, the most general form allowed for the STC tensor is

$$\begin{array}{ccc} \tau^X & \tau^Y & \tau^Z \\ \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & b \\ 0 & -a & 0 \end{bmatrix} & \begin{bmatrix} 0 & 0 & -b \\ 0 & 0 & 0 \\ a & 0 & 0 \end{bmatrix} & \begin{bmatrix} 0 & c & 0 \\ -c & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \end{array}$$

Examples of spin currents corresponding to purely the a , b , and c processes are depicted in Fig. 1c. If a thin film of IrO_2 is grown in an orientation different than (001), one can perform a change of basis in order to define the STC tensor relative to basis vectors in the plane and perpendicular to the thin film. This is achieved through a rotational transformation of the form:

$$\tau_{ij}^S = \sum_{l,m,n} R_{il} R_{mj} R_{sn} \tau_{lm}^n \quad (1)$$

where R_{il} are the elements of the appropriate rotation matrix. For example, for a (100) oriented IrO_2 film, using the basis vectors $X = [010]$, $Y = [001]$, $Z = [100]$, the STC tensor takes the form

$$\begin{array}{ccc} \tau^X & \tau^Y & \tau^Z \\ \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & a \\ 0 & -b & 0 \end{bmatrix} & \begin{bmatrix} 0 & 0 & -c \\ 0 & 0 & 0 \\ c & 0 & 0 \end{bmatrix} & \begin{bmatrix} 0 & b & 0 \\ -a & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \end{array}$$

The transformed forms of the STC tensor appropriate for the (101), (110), and (111) thin-film orientations of IrO_2 are listed in the supplemental information.

To determine the full STC tensor in IrO_2 , the three elements a , b , and c must be experimentally measured. Spin-torque measurements in spin-source/magnet bilayer samples are only sensitive to spin currents flowing perpendicular to the sample plane, because only for this flow direction can the spin current be transmitted to the magnetic layer to exert a torque. This means that only the elements in the bottom rows (i.e., σ_{Zj}^S) of the STC tensor are accessible. The first element, a , can be measured using the (001)-orientated film, where an electric field is applied along the $[010]$ direction resulting in a $[\bar{1}00]$ -polarized spin current flowing in the out-of-plane $[001]$ direction as indicated in Fig. 1b. The second element, b , can be measured using the (100) orientation, with an electric field applied along the in-plane $[001]$ direction resulting in a $[0\bar{1}0]$ -polarized spin current flowing in the out-of-plane $[100]$ direction seen in Fig. 1e. The third and last term, c , can also be measured in the (100) orientation. An electric field applied along the $[010]$ direction results in a $[001]$ -polarized spin current flowing in the out-of-plane $[100]$ direction seen in Fig. 1g.

To measure the electric-field-generated torques experimentally, high crystalline thin films of IrO_2 were grown via RF magnetron sputtering on different orientations of TiO_2 substrates, then capped *in situ* with ferromagnetic permalloy (Py), and patterned into device structures using the same methods described in our previous report¹³. High resolution x-ray diffraction (HRXRD) demonstrated a single high crystalline phase for all five thin-film IrO_2 orientations studied in this paper: (001), (100), (110), (101), and (111) (see Fig. 2a-e), and scanning transmission electron

microscopy (STEM) also demonstrated sharp IrO₂/Py interfaces using STEM as shown in Fig. 2f-i. The spin-torque ferromagnetic resonance (ST-FMR) technique was used to characterize the SHC components by measuring ST-FMR resonance spectra as a function of sweeping the magnetic-field magnitude for a series of different directions of the magnetic-field within the plane of the sample. (See Methods for more details.)

The symmetric and antisymmetric ST-FMR amplitudes for the (001) and (100) orientations are shown in Fig. 1a,d, and f as a function of the angle of an in-plane applied magnetic field. The symmetric signals allow us to determine the three independent elements that define the anti-damping STC tensor of IrO₂ (see Methods). The first term a , determined in the (001) orientation, has a value of $520 \pm 19 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$. The second term b , determined in the (100) orientation with an electric field applied along the [001] direction, has a value of $238 \pm 5 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$. The third term c determined in the (100) orientation with electric field along the [010] direction, has a value of $493 \pm 15 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$. As required by symmetry for the (001) and (100) orientations with these electric-field directions (i.e., (001) with any in-plane E-field direction and (100) with E along [001] or [010]), we detect no unconventional torque components.

To test whether this same tensor gives a quantitative description of both the conventional and unconventional torques for other thin-film orientations, we also performed ST-FMR measurements for (110), (101), and (111) IrO₂ thin films. For each thin-film orientation, the measurements were made using various directions (ψ) of in-plane electric field (and hence charge current), with ψ measured relative to the direction defined as the X axis for that orientation. For convenience in distinguishing the conventional and unconventional torques, we will plot the measured torque tensor components using a different set of coordinate axes (using lower-case x, y, and z), in which the x axis is along the applied electric field direction and z remains normal to the sample plane. The anti-damping torque components we measure for each orientation are then the conventional in-plane torque perpendicular to E (the tensor component τ_{zx}^y), the unconventional in-plane torque parallel to E (τ_{zx}^x), and the unconventional out-of-plane torque (τ_{zx}^z). The results are shown as the symbols within the Fig. 3 graphs. The solid lines in Fig. 3 are the predicted STC values from the rotated experimental tensor, using the values of a , b , and c as determined above with no adjustable fit parameters. We find that, by using the STC tensor elements from the experimental (001) and (100) ST-FMR results and assuming the torque is governed by the bulk symmetries of IrO₂, the tensor rotation gives a good description of both the conventional and unconventional torque components for the other orientations. (Supplemental Note 3 shows zoomed-in plots of the unconventional torques.) For the (100), (110), (101), and (111) orientations, an unconventional in-plane STC was observed and followed the expected $\sin(2\psi)$ angular dependence, with a magnitude within $30 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$ of the value predicted by the tensor rotation.. For the (101) and (111) orientations, unconventional out-of-plane anti-damping torque was also present, following an expected $\sin(\psi)$ dependence, with a magnitude within $10 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$ compared to the expectation from the tensor rotation. Both types of unconventional torques are significantly weaker than the conventional in-plane anti-damping torques, but this is fully consistent with the tensor rotation given the measured values of a , b , and c . For example, in the (111) orientation, the largest amplitude predicted by the tensor rotation for the out-of-plane anti-damping torque is predicted to be $(a - c)/2 = 14 \pm 24 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$ (see supplemental information), or less than 3% of the conventional torque for the (001) orientation.

For the conventional in-plane anti-damping torque, the tensor rotation correctly predicts the observed dependence on the electric-field angle, but the magnitudes in some cases show somewhat larger deviations than for the unconventional torques. For example, the predicted conventional in-plane torque for the (111) orientation is about 20% lower than the measurements. One possible explanation for this could be due to the surface quality of the (001) and (100) orientations compared to (111). Due to the low surface energy of the (111) orientation, the interface between IrO₂ and Py is much sharper compared to the (001) and (100) orientations which could change the spin transparency at the interface. Compared to previous measurements for IrO₂ (001) and (110) grown by reactive oxide molecular beam epitaxy, the torque magnitudes for our (110) samples are in close agreement, whereas the torques for (001) orientation in our films are about a factor of two smaller.¹⁷ This difference could be due to an Ir spacer layer included in the previous work or due to different growth techniques. The results in Fig. 3 correspond to anti-damping torque components. In addition, unconventional field-like torques were present in the (100), (110), (101), and (111) orientations which we attribute to anisotropic resistances within these orientations (see Fig. S9 in Supplemental Note 3)^{5,18}.

Despite the consistency of the experimentally-determined anti-damping spin-torque tensor for different crystal orientations, the experimental results are inconsistent with density-functional-theory (DFT) calculations of the spin Hall conductivity. The DFT predictions for the elements of the spin Hall conductivity tensor corresponding to the a , b , c parameters of the STC tensor are $a_{SHC} = 254 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$, $b_{SHC} = 162 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$, and $c_{SHC} = 18 \left(\frac{\hbar}{e} (\Omega \text{ cm})^{-1}\right)$. Despite the fact that the SHC magnitudes should be larger than the STC magnitudes on account of the interfacial spin transmission factor ($T < 1$), the spin Hall conductivities predict values that are too small, by more than a factor of 2 for the a parameter and by a factor of 27 for the c parameter. This leads to the measured conventional spin-orbit torques being significantly larger than expected from the DFT predictions. In contrast, the measured unconventional out-of-plane torque for low-symmetry crystal orientations is nevertheless much smaller than predicted by DFT. This is because the out-of-plane anti-damping torque is proportional to $a - c$, and this difference is much smaller for the torque parameters than the difference predicted by DFT, $a_{SHC} - c_{SHC}$. The poor agreement between measurements of spin-orbit torque and DFT predictions is true not only for IrO₂, but also for most materials including the prototypical spin source Pt^{19,20}. This indicates that essential physics is still missing from this comparison. Nevertheless, since our tensor-rotation analysis depends only on the bulk symmetries of IrO₂ with no assumptions about microscopic mechanisms, predictions based on the rotated-tensor analysis remain valid and accurate.

A possible explanation of this behavior is the polycrystallinity of our Py films. This may cause diffuse scattering at the interface which is largely independent of the IrO₂ crystal facet. It would be interesting to explore material systems where the ferromagnetic thin film is epitaxially grown on crystalline IrO₂ or other spin-current source material to measure spin torques. In this case, the symmetry of the whole bilayer structure would control the spin torque, and the effect of interface breaking symmetry of the bulk spin-current source material could be elucidated. We leave these studies for future research.

Conclusion

We have experimentally determined the full anti-damping spin torque conductivity tensor for IrO₂, and showed that this single tensor provides consistent and accurate results of the measured electric-field-driven torques for five different thin-film orientations, including both conventional

and unconventional torques. The good agreement between the experimental measurements of the anti-damping spin torques and the predictions from the tensor rotation in Fig. 3 confirm that the electric-field-induced torques generated by IrO₂ originate from bulk spin currents within the IrO₂, with perhaps a minor contribution to the conventional spin torque in the (111) orientation from an additional interfacial effect. We observe no indication of large differences in interfacial spin transmission for different crystal orientations that would invalidate the tensor analysis. A possible explanation of this behavior is the polycrystallinity of our Py films, which causes diffuse scattering at the interface that is largely independent of the IrO₂ crystal facet. In addition, the tensor analysis show that it is possible to fully characterize the spin-torque tensor of an anisotropic material using measurements of conventional spin-orbit torque for selected high-symmetry crystal orientations, and then to obtain accurate predictions of the unconventional torques for lower-symmetry crystal orientations by means of a simple tensor rotation.

Materials and Methods

Sample growth, fabrication, and characterization.

Epitaxial IrO₂ was grown on TiO₂ (001), (100), (110), (101) and (111) substrates by RF magnetron sputtering followed by *in situ* growth of ferromagnetic permalloy Ni₈₁Fe₁₉ (Py). The IrO₂ films were grown at 400°C at a pressure of 30 mTorr with 10% oxygen partial pressure. The target power was 20 W. After growth the sample was cooled in an O₂ atmosphere. Py was then grown in situ at room temperature, 4 mTorr of Ar, power of 35W, and a background pressure of 3E-7 Torr. The samples were then fabricated using photolithography and ion beam milling, followed by sputter deposition of 100 nm Pt/10nm Ti and lift off techniques for the electrodes.

ST-FMR measurements

During the ST-FMR measurements, a microwave current was applied at a fixed frequency (5-12 GHz) and fixed power (10-13 dBm) while sweeping an in-plane magnetic field through the Py resonance conditions from 0 to 0.15 T. The microwave current was modulated at a fixed frequency of 437 Hz and the mixing voltage across the device was measured using a lock-in amplifier. The mixing voltage was fitted vs applied field to extract the symmetric and antisymmetric Lorentzian components. For the angular-dependent ST-FMR, the applied field was rotated in-plane 360° and the symmetric and antisymmetric components were plotted as a function of angle. The out-of-plane (T_{\perp}) and the in-plane (T_{\parallel}) torques are proportional to the mixing voltage V_{mix} as the ferromagnetic layer goes through its resonance condition, which can be fitted as a sum of a symmetric and an antisymmetric Lorentzians:

$$V_{\text{mix},S} = -\frac{I_{rf}}{2} \left(\frac{dR}{d\varphi} \right) \frac{1}{\alpha(2\mu_0 H_{FMR} + \mu_0 M_{eff})} T_{\parallel}$$

$$V_{\text{mix},A} = -\frac{I_{rf}}{2} \left(\frac{dR}{d\varphi} \right) \frac{\sqrt{1+M_{eff}/H_{FMR}}}{\alpha(2\mu_0 H_{FMR} + \mu_0 M_{eff})} T_{\perp} .$$

Here I_{rf} is the RF current calibrated using Joule heating experiments, R is the resistance of the device, φ is the magnetization angle with respect to the applied current, α is the Gilbert damping

coefficient determined from the ST-FMR linewidth (giving values in the range $\alpha = 0.013 - 0.037$, $\mu_0 H_{FMR}$ is the resonance field, and $\mu_0 M_{eff}$ is the effective magnetization ranging from 0.6 - 0.85 T. The effective magnetization of Py is obtained using Kittel's equation $f = \frac{\gamma}{2\pi} \sqrt{(H_{FMR} + H_K)(H_{FMR} + H_K + M_{eff})}$ where γ is the gyromagnetic ratio and H_K is the in-plane anisotropy field. Unconventional torques are characterized based on the dependence of the measured torques on the angle φ of the magnetic field:

$$T_{\parallel} = T_{x,AD} \sin(\varphi) + T_{y,AD} \cos(\varphi) + T_{z,FL}$$

$$T_{\perp} = T_{x,FL} \sin(\varphi) + T_{y,FL} \cos(\varphi) + T_{z,AD}$$

$V_{mix,S}$ and $V_{mix,A}$ can then be expressed in the form of $\sin(2\varphi)(T_{x,AD} \sin(\varphi) + T_{y,AD} \cos(\varphi) + T_{z,FL})$ and $\sin(2\varphi)(T_{x,FL} \sin(\varphi) + T_{y,FL} \cos(\varphi) + T_{z,AD})$, respectively. Here the subscript x denotes a component associated with the spin polarization direction aligned with the applied electric field, and z the component associated with spin polarization normal to the sample plane. Additionally, here capital T represents the torque on the magnetic moment which is different than lowercase τ used to denote the spin torque conductivity in the main text. The spin torque conductivity τ_{jk}^i can then be determined using $\tau_{jk}^i = \frac{\theta_i}{\rho_{IrO_2}} \frac{\hbar}{2e}$, where ρ_{IrO_2} is the resistivity and θ_i is defined as:

$$\theta_i = T_{i,AD} \frac{2e\mu_0 M_s t_{FM}}{\gamma \hbar J}$$

where t_{FM} is the thickness of the ferromagnetic layer (Py or Ni) and J is the charge current density in the IrO₂. The resistivities were determined using Van der Pauw measurements and found to be 105 $\mu\Omega cm$ for (001), 226 $\mu\Omega cm$ for (100) along the [010] direction, 160 $\mu\Omega cm$ for (100) along the [001] direction, 100 $\mu\Omega cm$ for (101) along the [010] direction, 150 $\mu\Omega cm$ for (101) along the [10-1] direction, 95 $\mu\Omega cm$ for (111) along the [11-2] direction, and 105 $\mu\Omega cm$ for (111) along the [1-10] direction.

Theoretical Calculations

DFT calculations were performed using a Quantum-ESPRESSO code²¹. The plane-wave pseudopotential method with the fully relativistic ultrasoft pseudopotentials²² was employed in the calculations. The exchange and correlation effects were treated within the generalized gradient approximation (GGA)²³. The plane-wave cut-off energy of 40 Ry and a $16 \times 16 \times 16$ k-point mesh in the irreducible Brillouin zone were used in the calculations. Spin-orbit coupling was included in all the calculations.

The spin Hall effect is given by:

$$\sigma_{ij}^k = \frac{e^2}{\hbar} \int \frac{d^3 \vec{k}}{(2\pi)^3} \sum_n f_{n\vec{k}} \Omega_{n,ij}^k(\vec{k}),$$

$$\Omega_{n,ij}^k(\vec{k}) = -2Im \sum_{n \neq n'} \frac{\langle n\vec{k} | J_i^k | n'\vec{k} \rangle \langle n'\vec{k} | v_j | n\vec{k} \rangle}{(E_{n\vec{k}} - E_{n'\vec{k}})^2},$$

where $f_{n\vec{k}}$ is the Fermi-Dirac distribution for the n th band, $J_i^k = \frac{1}{2}\{v_i, s_k\}$ is the spin current operator with spin operator s_k , $v_j = \frac{1}{\hbar} \frac{\partial H}{\partial k_j}$ is the velocity operator, and $i, j, k = x, y, z$. $\Omega_{n,ij}^k(\vec{k})$ is referred to as the spin Berry curvature in analogy to the ordinary Berry curvature. In order to calculate the spin Hall conductivities, we construct the tight-binding Hamiltonians using PAOFLOW code^{24,25} based on the projection of the pseudo-atomic orbitals (PAO) from the non-self-consistent calculations with a $16 \times 16 \times 16$ k -point mesh. The spin Hall conductivities were calculated using the tight-binding Hamiltonians with a $48 \times 48 \times 48$ k -point mesh by the adaptive broadening method to get the converged values.

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Author Contributions:

M.P., D.R., and C.B.E. conceived the project. M.P. carried out the thin film growth, device fabrication, and spintronic measurements. D.P. and X.H. carried out additional device fabrication and supporting spintronic measurements. G.N. carried out the STEM experiments. G.G. performed theoretical calculations. C.B.E., M.S.R., D.C.R., S.Y.C., and E.Y.T. supervised the study. All authors discussed the results and commented on the manuscript. C.B.E. directed the research.

Competing Interest Statement:

The authors declare no conflict of interest.

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Figures and Tables

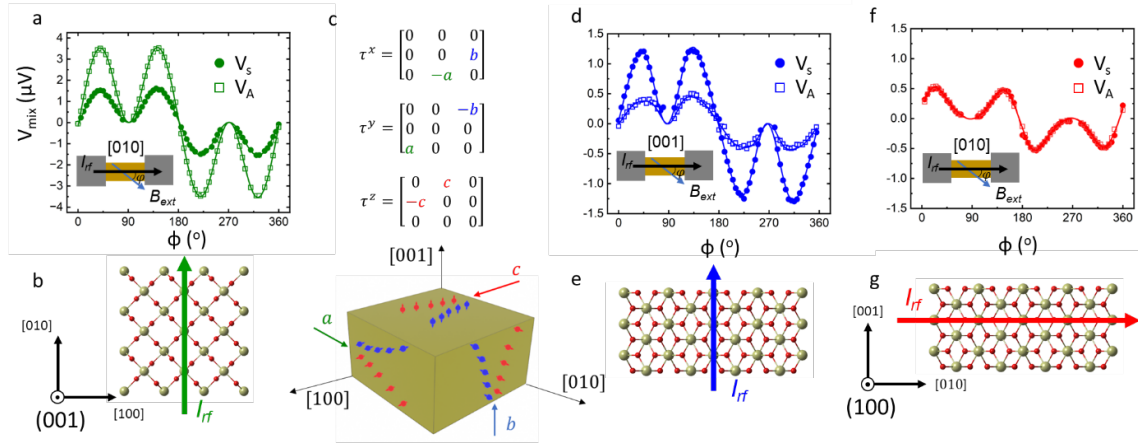


Figure 1. a, Symmetric and antisymmetric ST-FMR amplitudes as a function of magnetic-field angle for the (001) orientation with charge current along the [010] direction as depicted in b. c, experimental determination of the 3 non-zero STC elements using the (001) and (100) orientations. d, ST-FMR amplitudes as a function of magnetic-field angle for the (100) orientation with charge current along the [001] direction, as depicted in e. f, ST-FMR amplitudes as a function of magnetic-field angle for the (100) orientation with charge current along the [010] direction, as depicted in g.

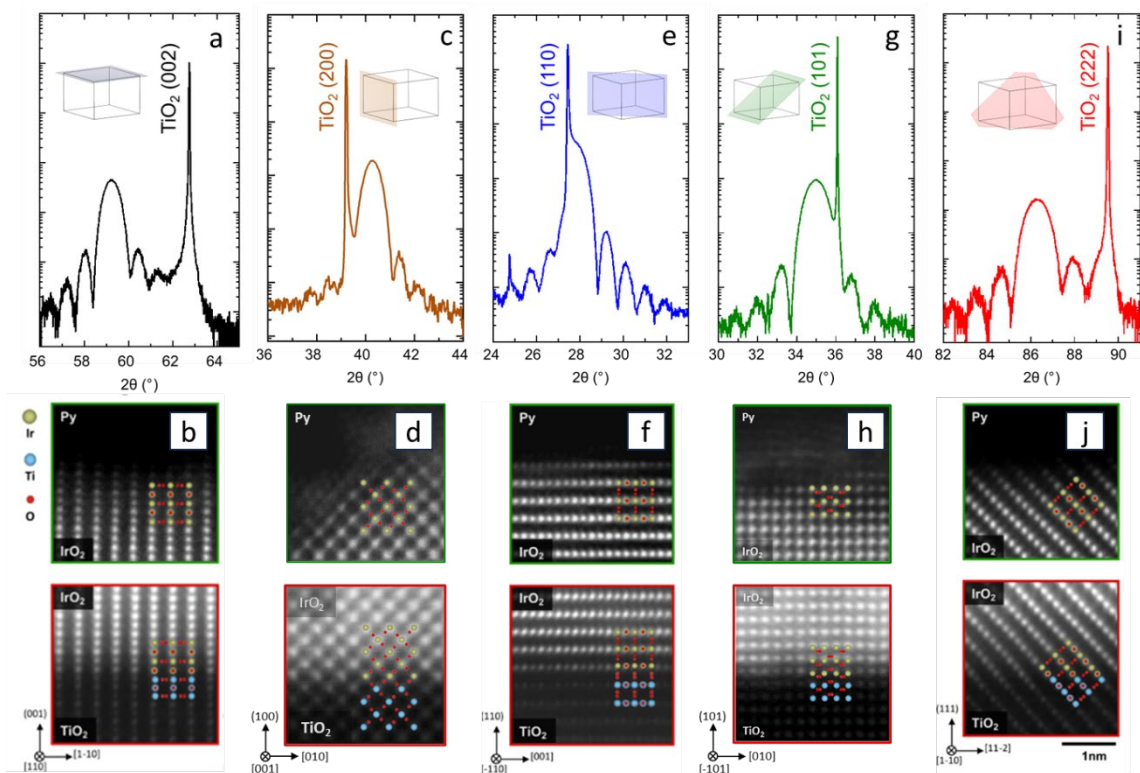


Figure 2. a, HR-XRD of the out of plane (002) peak and b, STEM of the interface between IrO₂ and Py and TiO₂ and IrO₂ with [1-10] zone axis. c, HR-XRD of the out of plane (200) peak and d, STEM of the interface between IrO₂ and Py and TiO₂ and IrO₂ with [010] zone axis. e, HR-XRD of the out of plane (110) peak and f, STEM of the interface between IrO₂ and Py and TiO₂ and IrO₂ with [001] zone axis. g, HR-XRD of the out of plane (101) peak and h, STEM of the interface between IrO₂ and Py and TiO₂ and IrO₂ with [010] zone axis. And i, HR-XRD of the out of plane (222) peak and j, STEM of the interface between IrO₂ and Py and TiO₂ and IrO₂ with [11-2] zone axis.

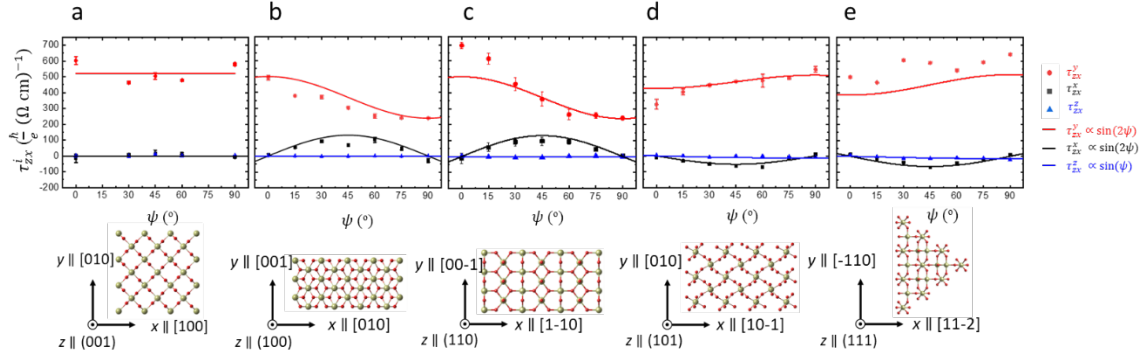


Figure 3: a-e, (001), (100), (110), (101), and (111) conventional and unconventional spin torque conductivities as a function of the angle ψ of the in-plane electric field with respect to the x axis shown for each projection. Solid lines represent predictions based on rotating the experimental STC tensor, using the tensor elements determined from the ST-FMR results in the (001) and (100) orientations and the bulk symmetries of IrO_2 .