Enhanced spin–orbit torques by oxygen incorporation in tungsten films

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The origin of spin–orbit torques, which are generated by the conversion of charge-to-spin currents in non-magnetic materials, is of considerable debate. One of the most interesting materials is tungsten, for which large spin–orbit torques have been found in thin films that are stabilized in the A15 (β-phase) structure. Here we report large spin Hall angles of up to approximately –0.5 by incorporating oxygen into tungsten. While the incorporation of oxygen into the tungsten films leads to significant changes in their microstructure and electrical resistivity, the large spin Hall angles measured are found to be remarkably insensitive to the oxygen-doping level (12–44%). The invariance of the spin Hall angle for higher oxygen concentrations with the bulk properties of the films suggests that the spin–orbit torques in this system may originate dominantly from the interface rather than from the interior of the films.
here has been considerable recent interest in the controlled manipulation of magnetic moments using spin–orbit torques for the purposes of building novel spintronic devices\textsuperscript{1–4}. Spin–orbit torques are derived from pure spin currents that are generated from charge currents flowing through non-magnetic metals via spin–orbit coupling\textsuperscript{5–8}. The conversion efficiency of the charge-to-spin current can be described by the spin Hall angle (SHA). Although small effects were first observed in semiconductors a decade ago (\(\sim 2 \times 10^{-4}\) in GaAs\textsuperscript{9}), recently much larger SHAs have been surprisingly observed in simple metals, making them potentially useful for spintronic applications. Indeed, reliable methods to measure the spin currents involve their diffusion into adjacent magnetic layers on which they can exert significant spin torques\textsuperscript{4,10,11}. These spin–orbit torques have been shown to be sufficiently large to induce the motion of magnetic domain walls\textsuperscript{12–16}, excite precessional magnetization dynamics\textsuperscript{10,17} or switch a uniform magnetic layer\textsuperscript{3,4,18}.

Although the exact origins responsible for these spin–orbit torques in conventional metals are not understood in detail, existing theories ascribe them to combinations of intrinsic mechanisms and extrinsic spin-dependent scattering from within the bulk of the materials studied\textsuperscript{19}. Most typically, extrinsic mechanisms will likely dominate because of innate bulk disorder in the thin films that have been studied to date. Largely neglected has been the role of interface scattering (which is known to dominate the origin of giant magnetoresistance\textsuperscript{20}), although electronic discontinuities at interfaces between two distinct materials are known to give rise to Rashba effects that can result in charge-to-spin conversion\textsuperscript{21–23}.

One of the most efficient materials exhibiting spin–orbit torques is the highly resistive \(\beta\)-phase of tungsten, where SHAs of up to approximately \(-0.35\) have been reported\textsuperscript{24–26}. In contrast, the \(\alpha\)-phase of tungsten exhibits a much smaller SHA. Consequently, one would expect that the SHA should scale strongly with significant changes in the microstructure of the materials under consideration. A detailed understanding of the role of materials’ microstructure on the spin–orbit torques is thus crucial for enhancing the efficiency of these effects, as well as giving insight towards the underlying mechanisms responsible for the spin–orbit torques.

Here we demonstrate that by doping oxygen into tungsten thin films, large spin–orbit torques are attained that are rather insensitive to considerable changes in the resistivity and film microstructure. The spin–orbit torques that we observe, when quantified as a SHA, are the largest to be reported thus far for a conventional metal-based system (approximately \(-0.5\)), and could be of considerable technological interest for spintronic applications. Furthermore, our findings suggest that these very-efficient spin–orbit torques are largely interfacial in origin.

**Results**

**Spin torque ferromagnetic resonance of W(O) | CoFeB structures.**

In this study we were deposited at room temperature using d.c. magnetron sputtering onto oxidized silicon substrates consisting of the layer-structure Si substrate | SiO\(_x\) (25) | W(O) (6) | Co\(_{40}\)Fe\(_{40}\)B\(_{20}\) (6) | TaN (2) (with film thicknesses in nm in parentheses). Hereafter, we shall refer to oxygen-doped tungsten as W(O) and Co\(_{40}\)Fe\(_{40}\)B\(_{20}\) as CoFeB. The amount of oxygen gas flow in the reactive mixture during the sputtering, \(Q\), was varied between 0 and 3%. A 20-Å-thick highly resistive TaN layer (5.08 \(\Omega\)cm for 50 nm thickness) was used for capping the magnetic CoFeB film (109 \(\mu\)Ωcm for 6 nm thickness; Fig. 1a,b). The TaN layer was \(\sim 10^4\) times more resistive than the most resistive W(O) film considered in this study, ensuring that current primarily flows in the W(O) and CoFeB layers. The atomic oxygen concentration \(n\) was determined in films specially deposited on graphite substrates using Rutherford backscattering spectrometry (RBS). (d) Oxygen concentration versus gas flow \(Q\) for W(O) films of different thicknesses. (e) Microscope image and schematic of the electrical circuit used for STFMR measurements.

We use the spin transfer torque (STT) ferromagnetic resonance technique\textsuperscript{10} to determine an effective SHA, \(\Theta_{SH}^{eff}\), which we use to assess the value of the damping-like spin–orbit torque. Figure 1e shows a microscope image of a representative microstrip device (10 \(\mu\)m × 80 \(\mu\)m) with a 45° tilt (for more information on the fabrication, see Methods) and a schematic illustration of the experimental set-up used for this measurement. A radiofrequency (RF) current is passed through the device, which generates spin–orbit torques as well as an Oersted field, in the presence of an externally applied magnetic field \(H_{ext}\). These torques cause a sustained precession of the magnetization of the magnetic layer, which is measured through the mixing d.c. voltage \(V_{mix}\) that is generated from the oscillating anisotropic magnetoresistance and spin Hall magnetoresistance\textsuperscript{25} signals and the applied RF current. The RF source has been set to an output power of 5 dBm and then amplified to 22 dBm. We extract the magnitude of \(\Theta_{SH}^{eff}\) by performing two different types of analyses based on fitting \(V_{mix}\) to...
a Lorentzian function consisting of symmetric ($F_S$) and asymmetric ($F_A$) components:

$$V_{\text{mix}} = V_0(SF_S(H_{\text{ext}}) + AF_A(H_{\text{ext}})),$$

where

$$V_0 = -\frac{1}{4}\frac{R}{\Delta} \gamma \mu_0 I_{\text{RF,tot}} \cos \phi,$$

$$F_S(H_{\text{ext}}) = \frac{\Delta^2}{\Delta^2 + (H_{\text{ext}} - H_0)^2},$$

and

$$F_A(H_{\text{ext}}) = \frac{F_S(H_{\text{ext}})(H_{\text{ext}} - H_0)}{\Delta}.$$

Here $\Delta$ is the linewidth and $H_0$ is the resonance field. The prefactors, $S$ and $A$, are the contributions of the symmetric and asymmetric parts, respectively, and $V_0$ is a constant prefactor. In equation (2), $R$ represents the resistance of the device, which, because of a combination of anisotropic magnetoresistance and spin Hall magnetoresistance, depends on the angle $\phi$ between the current and the magnetization (which for our measurements is either at $45^\circ$ or $-135^\circ$, depending on the applied field direction), $\gamma$ is the gyromagnetic ratio, $\mu_0$ is the magnetic permeability of free space, $I_{\text{RF,tot}}$ is the RF current through the device and $f$ is the frequency of the applied RF current. Figure 2a shows representative data for $V_{\text{mix}}$ ($n=12.1\%$, $Q=0.3\%$) for an RF current applied at 9 GHz, along with the symmetric and asymmetric components of the data extracted from the fitting.

The first type of analysis, which we call here the line-shape analysis ($\theta_{1W}^\text{LS}$), associates the symmetric part of the Lorentzian function to the antidamping-like STT and the asymmetric part to the Oersted field in the system. Meanwhile, the line-width analysis is based on measuring changes in the linewidth ($\theta_{1W}^\text{LS}$),

![Figure 2](image-url)

**Figure 2 | Line-shape analysis.** (a) $V_{\text{mix}}$ along with the fitted (green), symmetric ($F_S$, red) and asymmetric ($F_A$, blue) Lorentzian functions used for the fitting for $n=12.1\%$ ($Q=0.3\%$). (b) Frequency as a function of the resonant field for $n=12.1\%$ ($Q=0.3\%$) used in the Kittel formula fitting. (c) The magnetization $M$ determined by VSM and effective demagnetization field $M_{\text{eff}}$ from the Kittel formula fitting versus oxygen concentration $n$. (d) $V_{\text{mix}}$ normalized to either its minimum or maximum value for different oxygen concentrations at 9 GHz. The coloured dotted line denotes the corresponding maximum/minimum value of $V_{\text{mix}}$. The dotted line (dashed grey line) indicates the zero level. (e) SHA calculated from the line-shape analysis $\theta_{1W}^\text{LS}$ as a function of the oxygen concentration $n$. 

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that is, damping, as a function of a superimposed d.c. current. We note that the SHAs we report here are effective values because we do not take into account the transparency\textsuperscript{29}, whose role is minor in the materials under consideration here because of their relatively high resistivities.

**Line-shape analysis.** The determination of the SHA based on the line-shape analysis is described by the following formula\textsuperscript{10}:

\[ \theta_{LS}^{eff} = \frac{I_{S,eff}}{I_{C,eff}} = \frac{S 2e\mu_0 M_{S} H_0}{A} \beta_{oc} \sqrt{1 + M_{eff} / H_0} \]  

(5)

\[ \beta_{oc} = \frac{d}{2}, \]  

(6)

where \( e \) represents the electron charge and \( h \) represents Planck’s constant above. We deduce the thicknesses of the magnetic (t) and non-magnetic (d) layers by determining the film deposition rates from measurements of the thicknesses of nominally 50-nm-thick calibration films with a profilometer. The saturation magnetization \( M_s \) was measured by vibrating sample magnetometry (VSM) and the effective demagnetization field \( M_{eff} \) was calculated by fitting the frequency versus the resonance field, according to the Kittel formula (Fig. 2b; see Supplementary Table 1 for a summary of the parameters above for the films under study):

\[ f = \frac{\gamma H_0}{2\pi} \sqrt{H_0 / (H_0 + M_{eff})} \]  

(7)

Figure 2c illustrates a weak decrease in \( M_0 \) with increasing oxygen concentration.

Figure 2d shows the fitting of normalized \( V_{\text{mix}} \) at 9 GHz for different amounts of oxygen incorporated into the W(O) film. The data are normalized either to the minimum or maximum value of \( V_{\text{mix}} \). The corresponding maximum/minimum of \( V_{\text{mix}} \) is indicated by the coloured dotted line, as a guide to the eye. Using the data analysis given above, large SHAs imply a large symmetric component relative to the asymmetric component. Consequently, the closer the dotted line is to zero (grey dashed line in figure), the higher the SHA is. The highest value we reach with this analysis is \( \theta_{LS}^{eff} = -0.67 \). Moreover, the variation of the SHA as a function of the oxygen concentration is rather significant (Fig. 2e).

We further note that this commonly used analysis technique requires careful attention from possible artefact voltages. In particular, the line-shape analysis assumes that the symmetric component arises completely from the antidamping STT. However, a contribution to the symmetric component arises completely from the antidamping STT.

**Line-width analysis.** The Gilbert damping \( \alpha \) can be deduced from the dependence of \( \Delta \) on frequency that is given by

\[ \Delta = \Delta_0 + \frac{2\pi \alpha}{\gamma} \]  

(8)

where \( \Delta_0 \) represents the inhomogeneous broadening. We observe that the Gilbert damping remains relatively constant with respect to the oxygen concentration, with an average value of \( \alpha = 0.0085 \) (Fig. 3). We have also compared the change in the Gilbert damping with the presence of the W(O) layer to deduce the effective spin-mixing conductance as a function of \( n \), and find it to be relatively constant (see Supplementary Note 2 and Supplementary Fig. 3).

We further measured the dependence of \( \Delta \) on an applied d.c. current through the device (Fig. 3b):

\[ \Delta = \frac{2\pi \alpha}{\gamma} \left( \alpha + \frac{\sin \phi}{(H_{\text{ext}} + 0.5 M_{\text{eff}} / \mu_0 M_s t)^2} \right) J_s \]  

(9)

To determine \( \theta_{\text{eff}}^{\text{LW}} \) originating from the antidamping-like STT\textsuperscript{31}, one has to use the slopes \( \delta \Delta / \delta J_{\text{LS}} \) from Fig. 3b:

\[ \theta_{\text{eff}}^{\text{LW}} = \frac{\delta \Delta / \delta J_{\text{LS}}^{\text{LW}}}{R_{\text{FM}} + R_{\text{W}(O)} A_C} \]  

(10)

\( R_{\text{FM}} \) and \( R_{\text{W}(O)} \) are the resistances of the ferromagnetic layer and W(O) layer, respectively, and \( A_C \) is the cross-sectional area of the device. From this line-width analysis we find that \( \theta_{\text{eff}}^{\text{LW}} \) increases abruptly on the introduction of oxygen, but varies slightly with the addition of more oxygen into the tungsten film, as shown in Fig. 3c. Although the dependence of \( \theta_{\text{eff}}^{\text{LW}} \) and \( \theta_{\text{LS}}^{\text{eff}} \) on oxygen concentration is qualitatively similar, the variation in \( \theta_{\text{LS}}^{\text{eff}} \) with higher oxygen concentrations is much more pronounced when using the line-shape analysis (Fig. 2e). The SHA values reached here are at most \( \theta_{\text{LS}}^{\text{eff}} = -0.49 \) where \( n = 12.1\%\). The SHA in the pure tungsten film, by comparison, is \( \theta_{\text{LS}}^{\text{eff}} = -0.14 \), which is consistent with a previous report for a 6-nm-thick film where both the \( \alpha\)- and \( \beta\)-phase tungsten films were found to co-exist\textsuperscript{24}. The difference between the line-shape and line-width analyses stems mostly from the contributions of the spin-pumping and field-like torques. To quantitatively reconcile the values based on the two analysis techniques, an accurate determination of the RF current through the device is required (see Supplementary Note 1 and Supplementary Figs 1 and 2). Figure 3d shows the comparison between the line-shape and line-width analyses after accounting for the spin-pumping and field-like torques. Here we shall focus on the line-width analysis when drawing conclusions regarding the SHA.

The role of thickness of the W(O) on the SHA was examined for a gas flow \( Q = 1.2\% \) (Fig. 3e). However, one important point to note is that the amount of oxygen that is actually incorporated in the film varies significantly with the thickness of the grown film, as mentioned above (Fig. 1d). Thus, the SHA of these films can be plotted as a function of oxygen content, irrespective of their thickness, and compared with data from above where the thickness remains constant and the oxygen content is intentionally varied (Fig. 3f). On the basis of this analysis, we find that the SHA does not change significantly despite large changes in thickness. Furthermore, based on a volume origin of the SHE, the SHA as a function of thickness varies as

\[ \frac{J_s(d)}{J_s(\infty)} = 1 - \frac{d}{\lambda_s} \]  

(11)

Our data thus imply that either the spin diffusion length \( \lambda_s \) would have to be significantly smaller compared with 4.4 nm or alternatively the spin–orbit torque has an interfacial origin.
that increasing the oxygen concentration in the films leads to remaining nanocrystalline at a critical and show similar trends to the multilayer films (see Supplementary Note 3 and Supplementary Fig. 4). The transition indicates that the W(O) material becomes oxygen concentration-independent grain size for higher oxygen concentrations. The X-ray diffraction measurements (Fig. 4a,b) reveal that the tungsten film grown without any oxygen exhibits a predominantly $\alpha - W$ phase with a (100) orientation (strong peak at $2\theta = 40.3^\circ$) with some contributions from a $\beta - W$ structure with (200) and (211) crystal orientations ($2\theta = 35.4^\circ$ and $43.7^\circ$, respectively). The sample with the highest SHA, corresponding to $n = 12.1\%$, exhibits a peak at $2\theta = 39.8^\circ$ that is distinct from the $\alpha - W$ (100) peak and corresponds to the (210) peak for $\beta - W$. Thus, the X-ray data show that the amount of $\beta - W$ is increased significantly for $n = 12.1\%$ compared with $n = 0\%$ and there is no evidence for any remaining $\alpha - W$. The X-ray diffraction measurements also show that increasing the oxygen concentration in the films leads to eventually significantly broader and weaker X-ray diffraction peaks, indicating an increasing nanocrystallization of the W(O) films. The grain size can be calculated using the Scherrer equation

$$L_{\text{Grain}} = \frac{K\lambda}{\beta\cos\theta},$$

with $K = 1$ (shape factor), wavelength $\lambda = 0.154$ nm, $\beta$ being the half intensity width and $\theta$ being diffraction angle. The grain size versus oxygen concentration is shown in Fig. 4c. Interestingly, we observe a sudden decrease in the grain size at $n = 25.5\%$ to an oxygen concentration-independent grain size for higher $n$. The transition indicates that the W(O) material becomes nanocrystalline at a critical $n$. The X-ray diffraction measurements of the actual films used in the STT ferromagnetic resonance technique study as well as much thicker films, nominally $\sim 50$ nm thick, are shown in the Supplementary Section, and show similar trends to the multilayer films (see Supplementary Note 3 and Supplementary Fig. 4).
The observed effects (see Supplementary Note 4 and Supplementary temperature study to investigate transport mechanisms underlying 11.2 (extremely sensitive to the oxygen incorporation during the studies, which have shown that the formation of $\beta$- W is in the range of 100–300 μm2 cm, which corroborates our X-ray diffraction measurements that indicate that the 6-nm-thick pure tungsten film contains β– W. We have also performed a resistance versus temperature study to investigate transport mechanisms underlying the observed effects (see Supplementary Note 4 and Supplementary Fig. 5)26,36. With an increasing oxygen content, 50-nm-thick films become less metallic, until when $n$ is greater than ~25.5%, the film resistance increases with a decreasing temperature. For the thinner films, the resistance increases with decreasing temperature for all films. For $n < 31.7\%$, indications of superconductivity are observed below ~2K that give strong evidence of the A15 phase that is known to exhibit a superconducting transition near this temperature.

Discussion

Our materials’ characterization shows that the incorporation of oxygen stabilizes the β– W. This is consistent with previous studies, which have shown that the formation of β– W is extremely sensitive to the oxygen incorporation during the growth process37–41, and an oxygen concentration of only 10% is sufficient to stabilize β– W. In our experiments, at $n = 12.1\%$, we obtain a SHA of $\theta^{eff}_{LV} = -0.49$, which is higher than any previous measurements in conventional metal-based systems.

With the addition of further oxygen-doping, we observe the formation of a nanocrystalline structure. Despite the large changes we have observed in the bulk microstructure (as evidenced by a grain size change by more than a factor of 2), and a resistivity change by a factor of 2, we see that the SHA still remains very large. Our observation of the independence of the SHA despite significant changes in the bulk properties, namely the oxygen concentration, the resistivity, the microstructure as well as thickness, suggests that the mechanism responsible for the observed highly efficient spin–orbit torque may originate at the W(O)/ferromagnet interface. Our observations are consistent with theoretical proposals of antidamping-like torques from interfacial spin–orbit coupling such as the Rashba effect42–46. Such interfacial spin–orbit torques have also been reported in the Pt | oxidized CoFeB system47. One important difference in our experiment is that we do not intentionally oxidize the CoFeB. Furthermore, W is a heavy metal with large spin–orbit coupling, which gives rise to a large Rashba effect in combination with a 3d ferromagnet, such as CoFeB (ref. 48). For the Gd(0001) surface, it has been shown that on oxidation the Rashba parameter was enhanced49. Such a phenomenon may also play a role in our experiments. It has been postulated that spin memory loss (SML) at the interface could reduce the magnitude of the spin current50. It is difficult to estimate the magnitude of any SML but from the weak dependence of G_eff on oxygen content (Supplementary Fig. 3), it therefore follows that the SML would also be weakly dependent on the oxygen content, and, therefore, cannot account for the variation of effective SHA with oxygen content that we observe. In any case, any contribution from SML would simply mean that the values of the SHA that we report would be even larger. We have also examined the role of the inhomogeneous broadening on the SHA, and observe that it is uncorrelated to any changes we observe in the SHA (see Supplementary Note 5 and Supplementary Fig. 6).

We thus postulate that the first SHA peak at $n = 12.1\%$ is caused by β-phase stabilization and the large SHA observed at the higher oxygen concentrations to arise from interfacial spin–orbit torques. It is still possible, however, that for $n = 12.1\%$ there is already a considerably large interfacial spin–orbit torque that explains why we obtain larger values for β– W compared with previous studies. Furthermore, we note that one alternative explanation for the large SHA at high oxygen content may arise from amorphization of the W(O) material (Fig. 4c) and the SHA could be enhanced because of extrinsic effects in analogy with the large anomalous Hall effects found in amorphous magnetic materials51. However, in this case, one would expect the SHA to increase with higher oxygen concentration and scale with the resistivity. We instead observe a nearly constant SHA towards very high oxygen concentrations and thus believe that it is more likely that interfacial spin–orbit torques account for our observations.

Our results illustrate an intriguing path towards enhancing the magnitude of spin–orbit torques, and also serve to bridge the link between oxide electronics with spintronics.

Methods

Sample growth and preparation. The films were sputtered in the presence of Argon gas on undoped oxidized silicon with (100) orientation. Two-inch-diameter targets were used with the target to substrate distance kept fixed at 12.5 cm. The base pressure before deposition was less than $10^{-8}$ torr and the pressure during deposition was 3 mTorr. The W(O) layers were formed by introducing oxygen gas into the Ar-sputtering gas atmosphere during the film deposition. The gas flow of oxygen Q was varied between 0 and 3%. The highly resistive TaN-capping layer was fabricated by introducing 50% Nitrogen into the Argon gas flow.
Device-patterning was performed with optical lithography and ion milling, followed by a 14-nm-thick AlOx refill. Subsequently, 5 nm ruthenium and 65 nm gold were deposited and patterned for electrical contacts to the device.

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Author contributions

K.-U.D., T.P. and S.S.P.P. conceived the experiments. K.-U.D., T.P. and W.Z. performed the experiments and analysed the data. B.F.H. fabricated the samples. S.-H.Y. grew the films. A.K. performed the Rutherford backscattering spectrometry studies. K.-U.D., T.P. and S.S.P.P. wrote the manuscript. S.S.P.P. supervised the project. All authors discussed the results and implications.

Additional information

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Competing financial interests

The authors declare no competing financial interests.

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