APPLIED PHYSICS

Spin-orbit torque manipulated by fine-tuning of oxygen-induced orbital hybridization

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Current-induced spin-orbit torques provide an effective way to manipulate magnetization in spintronic devices, promising for fast switching applications in nonvolatile memory and logic units. Recent studies have revealed that the spin-orbit torque is strongly altered by the oxidation of heterostructures with broken inversion symmetry. Although this finding opens a new field of metal-oxide spin-orbitronics, the role of the oxidation in the spin-orbit physics is still unclear. Here, we demonstrate a marked enhancement of the spin-orbit torque induced by a fine-tuning of oxygen-induced modification of orbital hybridization. This is evidenced by a concomitant enhancement of the interface spin-orbit torque, interface spin loss, and interface perpendicular magnetic anisotropy within a narrow range of the oxidation level of metallic heterostructures. This result reveals the crucial role of the atomic-scale effects in the generation of the spin-orbit torques, opening the door to atomic-level engineering of the spin-orbit physics.

INTRODUCTION
Oxide electronic materials promise a wide range of applications, such as transparent electronics, optoelectronics, and power harvesting (1). Among this class of materials, Cu oxide is one of the first semiconducting materials to be investigated as early as the 1920s (2). Because of its low cost, good carrier mobility, and direct energy gap, Cu oxide has been used in the fabrication of a variety of electronic devices, including solar cells, resistive switching memories, and transistors (3).

Recently, Cu oxide has become attractive in spin-orbitronics, which aims to discover novel phenomena and functionalities arising from spin-orbit coupling (SOC) (4). The fundamental building block of spin-orbitronics is the generation of spin-orbit torques (SOTs), which enable electric manipulation of magnetization (5–11). Although Cu has been believed to generate negligible SOTs because of the weak SOC, a recent study has shown that Cu becomes an efficient SOT generator through oxidation (12). The oxidation-induced enhancement of the SOTs has been observed in various systems recently (13–16). These findings illustrate the essential role of the oxidation effect in exploring the physics and technology of spin-orbitronics. However, a fundamental understanding of the oxidation effect on the spin-orbit physics is still unclear. The great flexibility of the oxidation level of Cu makes it a model system to study the physics behind the generation of the SOTs manipulated by oxidation.

In this work, we demonstrate that the SOT in spin-orbitronic heterostructures is markedly enhanced through a fine-tuning of oxygen-induced orbital hybridization. By measuring the SOTs for Ni81Fe19/CuOx bilayers with various oxidation levels, we found that the magnitude of the damping-like (DL) SOT is substantially enhanced, whereas the sign of the field-like (FL) SOT is reversed, only within a narrow range of the oxidation level of CuOx. We show that this anomaly appears concomitantly with the enhancement of the effective spin-mixing conductance and interface perpendicular magnetic anisotropy (PMA). These results demonstrate the essential role of the atomic-scale and chemical bonding effects in the interface spin-orbit physics, providing a crucial piece of information for a fundamental understanding of the SOT generation.

RESULTS
Spinorque ferromagnetic resonance
We used the spin-orbit ferromagnetic resonance (ST-FMR) to quantify the SOTs generated by CuOx films with various oxidation levels. A schematic illustration of the device structure is shown in Fig. 1A. We deposited a Ni81Fe19 (10 nm)/CuOx (10 nm) bilayer, covered by a SiO2 (4 nm) capping layer, on a SiO2 substrate by radio frequency (RF) magnetron sputtering, where the numbers in parentheses represent the thickness. For the sputtering of the CuOx film, we introduced the mixture of argon and oxygen gas into the chamber. The amount of the oxygen gas flow Q in the reactive mixture varied between 0 and 1.5% (see Materials and Methods). As shown in Fig. 1B, the electrical resistivity p of the CuOx film increases markedly with Q, showing that the oxidation level of the CuOx film is controlled by the oxygen gas flow. The bilayer films were patterned into rectangular strips with a width of 20 μm and length of 150 μm using the photolithography and lift-off techniques. For the ST-FMR measurements, an RF charge current was applied along the longitudinal direction of the device, and an in-plane external magnetic field H was applied with an angle of 45° from the longitudinal direction of the device. In the bilayer, the RF current generates the DL- and FL-SOTs, as well as an Oersted field, driving magnetization precession in the adjacent ferromagnetic Ni81Fe19 layer (17). The magnetization precession induces an oscillation of the resistance in the ferromagnetic layer by the anisotropic magnetoresistance, resulting in the generation of a direct current (DC) voltage Vmix through the mixing of the RF charge current and oscillating resistance (18, 19): $V_{mix} = V_{sym}W^2/\left(\left(\mu_0H - \mu_0H_{FMR}\right)^2 + W^2\right) + V_{ant}W\left(\mu_0H - \mu_0H_{FMR}\right)/\left(\left(\mu_0H - \mu_0H_{FMR}\right)^2 + W^2\right)$, where $V_{sym}$ and $V_{ant}$ are the magnitude of the symmetric component, the magnitude of the antisymmetric component, and the spectral width, respectively. $H_{FMR}$ is the ferromagnetic resonance field. Here, in the $V_{mix}$ signal, the symmetric part is proportional to the DL spin-orbit effective field $H_{DL}$, and the antisymmetric part is due to the sum of the Oersted field $H_{Oe}$ and FL spin-orbit effective field $H_{FL}$ (17). In Fig. 1C, we show the $V_{mix}$
signal for the Ni81Fe19/CuO bilayers with Q = 0.5, 1.0, and 1.25%, measured by using a bias tee at room temperature. This result shows that the sign of Vmax is reversed by reversing the external magnetic field direction, as expected for the voltage generation induced by the ST-FMR.

**Spin-torque generation efficiency**

To quantitatively evaluate the influence of the oxidation level of the CuO layer on the DL and FL spin-torque efficiencies, we measured the ST-FMR for the Ni81Fe19(dF)/CuOx(10 nm) bilayers with various Ni81Fe19 layer thicknesses dF as shown in Fig. 2A. For the bilayers, the FMR spin-torque generation efficiency, ξFMR = (Vsym/Vana)(eq0MsdF/ds/h)\sqrt{1 + Meff/HFMR}, is related to the DL and FL spin-torque efficiencies, ξDL and ξFL, as (17)

$$\frac{1}{\xi_{\text{FL(FM)}}} = \frac{1}{\xi_{\text{DL}}} \left(1 + \frac{\xi_{\text{FL}}}{\xi_{\text{DL}}} \frac{dF}{dF} \right)$$

where ξFL(FM) = (2e/h)μoMsdF/HDL/FL(eF/\hbar)² (see the Supplementary Materials). Here, jF is the charge current density in the CuO layer and dO is the thickness of the CuO layer. M, and Meff are the saturation magnetization and effective demagnetization field of the Ni81Fe19 layer, respectively. Using Eq. 1, ξDL(FL) can be determined by measuring dF dependence of ξFMR under the assumption that ξDL(FL) does not have a strong dependence on dF in the range examined. In Fig. 2B, we show 1/ξFMR as a function of 1/dF for different Q.

Figure 2B demonstrates that only the bilayer with Q = 1% shows a different trend in the SOT generation; 1/ξFMR increases linearly with 1/dF for Q = 1%, whereas 1/ξFMR decreases linearly with 1/dF for Q = 0.5, 0.75, and 1.25%. The opposite sign of the slope in the linear relation between 1/ξFMR and 1/dF indicates that the direction of the FL-SOT generated in the bilayer with Q = 1% is opposite to that in the bilayer with Q = 0.5, 0.75, and 1.25% (see Eq. 1). We also note that the intercept of the linear fitting of 1/ξFMR for the bilayer with Q = 1% is smaller than that for the bilayer with Q = 0.5, 0.75, and 1.25%, indicating a larger DL-SOT generation efficiency when Q = 1%. To study the SOT generation around Q = 1%, we further measured the SOTs by slightly changing Q around 1%. In the following, we discuss the ST-FMR results with respect to the electrical resistivity ρ of the CuO layer because ρ changes sensitively and monotonically with Q as shown in Fig. 1B, enabling the characterization of the oxidation level of the CuOx films.

The drastic change of the SOTs observed in the Ni81Fe19/CuOx bilayer appears only within a narrow range of the oxidation level. In Fig. 3A, we show the DL and FL spin-torque efficiencies, ξDL and ξFL, determined from the 1/dF dependence of 1/ξFMR, as a function of ρ. Figure 3A shows that ξDL is substantially enhanced only around ρ = 9 × 10⁻⁵ Ω cm, which is concomitant with the sign reversal of ξFL. ξDL ~ 0.05 at ρ ~ 9 × 10⁻⁵ Ω cm is comparable to ξDL for a Ni81Fe19/Pt bilayer, demonstrating efficient SOT generation using CuOx. Here, the drastic change of the SOTs is irrelevant to phase transformation in the CuOx layer. The principal phases of Cu oxide are cuprous oxide (Cu2O) and cupric oxide (CuO). However, from x-ray diffraction and x-ray photoelectron measurements, we found that all the CuOx films with Q = 0.50, 0.75, 1.00, and 1.25% are a mixture of Cu and Cu2O; the CuO phase is absent (see the Supplementary Materials).
**DISCUSSION**

The marked change of the SOTs is induced by the interface SOC, which is maximized only within the narrow range of the oxidation level. In the moderately oxidized CuO (Q ≤ 1.25%), both bulk and interface SOC can generate the SOTs because the applied change current can flow both in the CuO, bulk and in Ni81Fe19/CuO, interface layers. The bulk contribution can be eliminated by further increasing the oxidation level of the CuO layer because the spin Hall conductance decreases with the oxidation, where l_{SH} and l_{SS} are the charge current flowing in the CuO layer and the total charge current flowing in the Ni81Fe19/CuO bilayer, respectively. In the bilayer with ρ ∼ 10⁻⁴ Ω cm (Q = 1.5%), the resistance of the CuO layer is three orders of magnitude larger than that of the Ni81Fe19 layer, and thus the SOTs can only be generated by the interface SOC (see the Supplementary Materials). We note that the sign of the SOTs in the bilayer with ρ ∼ 9 × 10⁻⁵ Ω cm, ξ_{DL} > 0 and ξ_{FL} > 0, is the same as that in the bilayer with ρ ∼ 10⁻¹ Ω cm (Q = 1.5%), where the interface SOC is responsible for the SOTs (see the Supplementary Materials). This indicates that the interface SOC is responsible for the observed anomaly of the SOTs at ρ ∼ 9 × 10⁻⁵ Ω cm. This is also supported by the fact that the anomalous SOT disappears by inserting an ultrathin Cu layer between the Ni81Fe19 and CuO with Q = 1% (see the Supplementary Materials). In contrast to the highly oxidized Ni81Fe19/CuO bilayer with ρ ∼ 10⁻¹ Ω cm, the dominant mechanism of the SOT generation in the bilayer within the resistivity range studied in Fig. 3 is determined by the competition between the bulk and interface SOT generation efficiencies, rather than the spatial distribution of the applied charge current. The reason for this is that the change of l_{SH} l_{SS} is not substantial, l_{SH} l_{SS} ∼ 50% for ρ ∼ 9 × 10⁻⁵ Ω cm and l_{SH} l_{SS} ∼ 30% for ρ ∼ 23 × 10⁻⁵ Ω cm, and both bulk and interface SOC can generate sizable SOTs in all the films studied in Fig. 3. This indicates that the SOTs can be dominated by the interface SOC only when the interface SOT generation efficiency is substantially higher than that of the bulk SOT generation efficiency in the bilayers studied in Fig. 3. Thus, the sign reversal of the FL-SOT only at ρ ∼ 9 × 10⁻⁵ Ω cm demonstrates that the interface SOT efficiency is markedly enhanced by the oxidation only at ρ ∼ 9 × 10⁻⁵ Ω cm, and the dominant mechanism of the SOT generation is changed only around this oxidation level.

The marked enhancement of the interface SOC, within the narrow range of the oxidation level, is further evidenced by angular momentum dissipation from the Ni81Fe19 layer. Figure 3B shows the ρ dependence of the effective spin-mixing conductance g_{eff}^1, which characterizes the angular momentum dissipation (20). Here, g_{eff}^1 was obtained from d_{l} dependence of the magnetic damping constant α (20)

$$\alpha = \alpha_0 + \frac{\gamma h}{4\pi M_r} g_{eff}^1,$$

where α0 is the intrinsic magnetic damping of the Ni81Fe19 film and γ is the gyromagnetic ratio. α for the Ni81Fe19/CuO bilayers was determined by fitting the microwave frequency f dependence of the ST-FMR spectral width W, using W = (2πα/γ) + W_{ext} where W_{ext} is the inhomogeneous linewidth broadening of the extrinsic contribution (see the Supplementary Materials). Figure 3B shows that g_{eff}^1 is also enhanced around the oxidation level where ξ_{DL} is maximized and the sign of ξ_{FL} is reversed. In the Ni81Fe19/CuO bilayer, g_{eff}^1 can be associated with the angular momentum absorbed in the bulk and interface: g_{eff}^1 = g_{bulk}^1 + g_{int}^1, where the interface contribution, g_{int}^1, is known as the spin memory loss due to the interface SOC (21). The bulk contribution is expressed as g_{bulk}^1 = g_{eff}^1 [1 + (2c^2/h)(\lambda_N/\sigma_N)N g_{eff}^1 c (d_N/\lambda_N)^{-1}], which is the bare spin-mixing conductance, and \lambda_N and σ_N are the spin diffusion length and conductivity of the CuO layer, respectively (20). g_{eff}^1 can be changed with the oxidation through the change of g_{eff}^1, because the observed oxidation of g_{eff}^1 can be expressed in terms of the enhancement of g_{eff}^1 or the interface SOC, consistent with the marked change of the interface SOTs.

Our experimental finding is that the SOT is markedly changed through the enhancement of the SOC at the Ni81Fe19/CuO interface only within the narrow range of the oxidation level; the interface SOC and SO are insensitive to the oxidation level of the CuO layer. The maximization of the interface SOT through the fine-tuning of the oxidation has not been reported previously. Here, the physics behind the interface SOC, or the Bychkov-Rashba SOC, is the orbital hybridization due to the broken inversion symmetry and atomic SOC. Ab initio calculations show that the strength of the interface SOC is dominated by the asymmetry of the interface-state wave function, or the strength of the hybridization, near the position of the nucleus of the interface atoms in conjunction with the atomic SOC (22–29). Because the orbital hybridization plays a key role in the Rashba physics, several experimental and theoretical studies have proposed that the strength of the Rashba splitting can be tuned by adatom adsorption (28–31).

The critical enhancement of the SOC at the Ni81Fe19/CuO interface can be attributed to oxygen-induced deformations of the interface-state wave functions. Since oxygen is an electronegative atom, the charge transfer toward oxygen atoms should be substantial; although O-2p states are weakly spin-orbit–coupled, they can dramatically modify the hybridization of the Ni, Fe, and Cu orbitals near the interface, resulting in the change of the wave-function symmetry near the nucleus (32). This oxygen-induced wave-function asymmetry can result in the enhancement of the Rashba effect (28). In the Ni81Fe19/CuO bilayer, the strength of the atomic SOC of Ni, Fe, and Cu is comparable (33), and thus the wave-function deformation of these atoms near the Ni81Fe19/CuO interface can contribute to the observed interface SOC (25). In contrast, since the atomic SOC of O is quite small, the primary
MATERIALS AND METHODS

The Ni81Fe19 layer was deposited on thermally oxidized Si substrates by RF magnetron sputtering at room temperature. The base pressure in the chamber before the deposition was lower than 1 × 10⁻⁶ Pa, and the deposition pressure was 0.2 Pa. The Ni81Fe19 and SiO₂ films were deposited by applying argon gas with a flow rate of 10 standard cubic centimeters per minute (sccm). For the CuO₃ deposition, a 99.99% purity Cu target was used; argon gas flow rate was fixed with 20 sccm, and the oxygen gas was applied with the reactive mixture ratio Q. The film thicknesses were controlled by the deposition time with a precalibrated deposition rate for each oxidation level. For the fabrication of the devices used for the ST-FMR experiment, the substrates were patterned into a 20 μm × 150 μm rectangular shape by standard photolithography before deposition, and lift-off technique was used to take off the remaining part of the films after deposition. For the Ni₈₁Fe₁₉ layer thickness dependence of the ST-FMR, a linear shutter was used to fabricate the Ni₈₁Fe₁₉ layer with various thicknesses. For the x-ray diffraction and x-ray photoelectron measurements of the CuO₃ films, the 50-nm-thick films were deposited on glass substrates with different oxygen reactive mixture rates Q by RF magnetron sputtering at room temperature.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/11/eaax4278/DC1

Section S1. Spin-torque efficiency
Section S2. X-ray diffraction and x-ray photoelectron measurements
Section S3. Interface SOTs
Section S4. Spin-torque ferromagnetic resonance for the Ni₈₁Fe₁₉/Cu/CuO trilayer
Section S5. Magnetic damping constant
Section S6. Interface oxidation level characterized by x-ray reflectivity
Fig. S1. Characterization of CuO₃ films.
Fig. S2. Interface SOTs.
Fig. S3. ST-FMR for trilayer.
Fig. S4. Magnetic damping constant.
Fig. S5. X-ray reflectivity.
Fig. S6. Auger electron spectroscopy.

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