

Enhanced unidirectional spin Hall magnetoresistance in a Pt/Co system with a Cu interlayerK. Hasegawa ^{1,2,*} T. Koyama ^{2,3} and D. Chiba ^{2,3,†}¹*Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo, Tokyo 113-8656, Japan*²*The Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan*³*Center for Spintronics Research Network (CSRN), Osaka University, 1-3, Machikaneyama-cho, Toyonaka, Osaka 560-8531, Japan*

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Spin-dependent scattering at the nonmagnet/ferromagnet interface plays a key role in determining the amplitude of unidirectional spin Hall magnetoresistance (USMR), similar to giant magnetoresistance (GMR). We report the enhancement of USMR by inserting a thin Cu interlayer into the Pt/Co interface, where the Cu/Co system is well known to exhibit a large GMR. A measurement of the spin-orbit torque shows that the spin current injection into the Co layer is not modulated by the Cu interlayer. In addition, USMR increases with the Cu thickness for the ultrathin regime as the Cu/Co interface is formed, reaching a peak before decreasing, owing to the shunting effect. Our results suggest an interfacial origin of the enhanced USMR and highlight the close similarities between USMR and GMR.

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Magnetoresistance is a central research theme in spintronics, and many phenomena have been found thus far, such as anisotropic magnetoresistance (AMR) [1], giant magnetoresistance (GMR) [2,3], and spin Hall magnetoresistance (SMR) [4]. The recently discovered unidirectional spin Hall magnetoresistance (USMR) in ferromagnet/nonmagnet heterostructures is completely different from these types of magnetoresistance in terms of its symmetry; USMR changes its sign when the current or magnetization direction is reversed [5–10]. Moreover, USMR increases with the current amplitude, unlike many other types of magnetoresistance. In addition to these unique features, USMR is also attractive as a simple readout method in magnetic random access memories because it can detect the magnetization direction of a single ferromagnetic layer without requiring any additional ferromagnetic pinned layer [9–11]. However, the magnitude of USMR is still too small for practical applications. Thus, an enhancement of its magnitude and a solid understanding of its physics is desirable.

As the origins of USMR, two mechanisms have been proposed [5,8,12,13]. One is electron-magnon scattering. Spin current absorption in the ferromagnetic layer causes the creation or annihilation of magnons depending on the relative orientation between spin polarization and magnetization. This modulates electron-magnon scattering, resulting in a resistance change in the ferromagnetic layer. The other is spin-dependent scattering, which is composed of bulk and interface contributions. The spin current injected into the ferromagnetic layer gives rise to USMR, owing to the spin-dependent conductivity in the ferromagnetic layer. In addition, the transmission and reflection probabilities of the spins at

the ferromagnet/nonmagnet interface depend on the spin orientation, leading to a unidirectional interface resistance, in analogy to GMR.

The magnon contribution is known to be significant when the applied magnetic field is small and is the main origin for a large USMR in topological insulator-based systems [7,8,10,11]. The effect of the bulk spin scattering on USMR has been clearly demonstrated by the thickness dependence of the USMR in the Pt/Co bilayer [12,14]. However, the USMR owing to interfacial spin scattering has not been sufficiently investigated [8,15].

In this Letter, we investigated the effect of the Cu interlayer on USMR in a Pt/Co system to focus on the role of interfacial scattering, where the Cu/Co system is a well-known GMR structure possessing a strong spin-dependent scattering potential at the interface [16–18]. We found that the USMR is enhanced by a factor of 1.5 with a thin insertion of Cu. Instead, the USMR is reduced when inserting Au, indicating the strong interfacial material dependence of USMR. From the measurement of the spin-orbit torque (SOT) and the interlayer thickness dependence of USMR, we conclude that the enhancement of USMR has an interfacial origin.

Figure 1(a) shows the layer structure used in this study. For Cu-inserted systems, Ta(1.0 nm)/Pt(3.0 nm)/Cu(t_{Cu})/Co(2.5 nm)/MgO(2.8 nm)/Ta(0.7 nm) layers were deposited on a thermally oxidized Si substrate by rf sputtering in Ar gas. We also prepared a Au-inserted system as a reference. The Au(t_{Au}) layer was deposited through electron beam evaporation, and the other layers were deposited using rf sputtering. Hereafter, we refer to these systems as Pt/Cu(Au)/Co for $t_{\text{Cu(Au)}} \neq 0$ and Pt/Co for $t_{\text{Cu(Au)}} = 0$. All samples exhibited in-plane magnetic anisotropy.

For the measurement of USMR, the deposited films were processed into Hall-bar structures using photolithography and Ar-ion milling. The nominal width of the wire was 10 μm and the distance between the two Hall arms was 40 μm .

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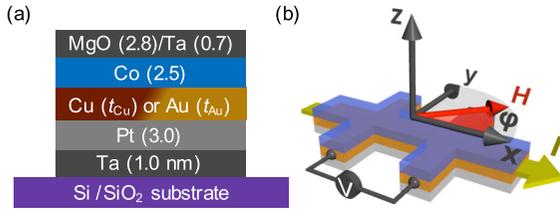


FIG. 1. (a) The layer structure of Pt/Cu(Au)/Co systems. (b) Schematic illustration of measurement geometry. φ is the azimuthal angle of the external magnetic field.

Figure 1(b) schematically depicts the experimental setup for the USMR measurement and coordinate system. An AC current with a frequency of 13.14 Hz was applied along the x direction. Then, the first (R_ω) and second ($R_{2\omega}$) harmonic longitudinal resistances were measured while rotating an external magnetic field of 4 T in the xy plane. Here, R_ω represents the conventional resistance, which does not depend on the current direction and amplitude, whereas $R_{2\omega}$ contains the USMR and thermoelectric signal. The applied external magnetic field is sufficiently large to saturate the magnetic moment and exclude the magnon contribution to the USMR. All measurements in this study were carried out at 300 K.

Figure 2(a) shows the angle dependence of the first harmonic resistance $\Delta R_\omega/R$ for Pt/Co and Pt/Cu(1.5 nm)/Co systems as a function of the in-plane magnetic field angle φ , where R and ΔR_ω are defined as $R_\omega(\varphi = 0)$ and $R_\omega - R$, respectively. Note that the value of R will be shown later [Fig. 4(b)]. In addition, $\Delta R_\omega/R$ shows the $-\sin^2 \varphi$ symmetry, which is a typical behavior of AMR and SMR [see the solid lines in Fig. 2(a) for fitting]. Moreover, $\Delta R_\omega/R$ for the Pt/Cu/Co system is smaller than that for the Pt/Co system, mainly because of the current shunting into the Cu interlayer [19–21].

Figure 2(b) shows the normalized pure USMR signal $R_{2\omega}^{\text{USMR}}/R$, where the thermoelectric signal owing to the thermal gradient along the z axis is subtracted (see Supplemental Material [22]). The angle dependence of $R_{2\omega}^{\text{USMR}}/R$ is well fitted by a sinusoidal function, showing that USMR is proportional to the y component of magnetization as expected [5]. The injected current density was 1×10^{11} A/m² for both samples, which was obtained by dividing the current intensity by the cross section of the Pt, Cu(Au), and Co layers. The current shunting into the top and bottom Ta layers is

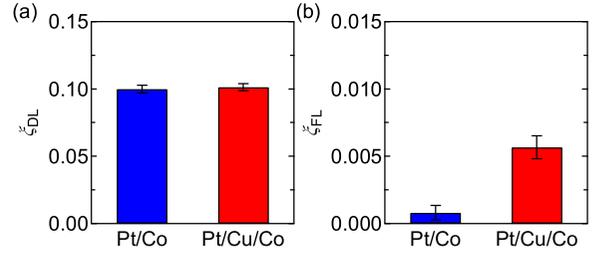


FIG. 3. (a) The DL and (b) FL SOT efficiencies for the same samples shown in Figs. 2(a)–2(c). Error bars are estimated from the fitting of second harmonic Hall resistance as a function of external magnetic field (see Supplemental Material [22]).

neglected because both Ta layers are expected to be oxidized and become poor conductors. The amplitude of $R_{2\omega}^{\text{USMR}}/R$ for the Pt/Co system is comparable to previously reported values [14,23]. Contrary to the reduced AMR and SMR, the USMR for Pt/Cu(1.5 nm)/Co is approximately 1.5 times larger than that for the Pt/Co system. We note that USMR is reduced when the interlayer is Au (1.5 nm) instead of Cu, as shown later.

We also carried out harmonic resistance measurements at different current amplitudes. Figure 2(c) shows the current density dependence of the USMR, where $\Delta R_{2\omega}^{\text{USMR}}/R = |R_{2\omega}^{\text{USMR}}(90^\circ) - R_{2\omega}^{\text{USMR}}(270^\circ)|/R$. The USMR for both Pt/Co and Pt/Cu(1.5 nm)/Co systems scales linearly with the current density. This linear dependence is consistent with the spin-dependent scattering mechanism because the spin accumulation is proportional to the injected current.

To clarify the role of interfacial spin-dependent scattering, the effect of the Cu interlayer on the spin injection should be excluded. For this purpose, we measured the SOT for Pt/Co and Pt/Cu/Co systems using the harmonic Hall method [24]. Figures 3(a) and 3(b) show the dampinglike (DL) and fieldlike (FL) SOT efficiencies for Pt/Co and Pt/Cu(1.5 nm)/Co systems, respectively. Here, the DL(FL) SOT efficiency is expressed as follows:

$$\xi_{\text{DL(FL)}} = \frac{2e}{\hbar} \mu_0 M_s t_{\text{FM}} \frac{H_{\text{DL(FL)}}}{j}, \quad (1)$$

where M_s , t_{FM} , and $H_{\text{DL(FL)}}$ are the saturation magnetization, thickness of the ferromagnetic layer, and DL(FL) effective field, respectively. ξ_{DL} is known as the effective spin Hall angle and has a strong correlation with the

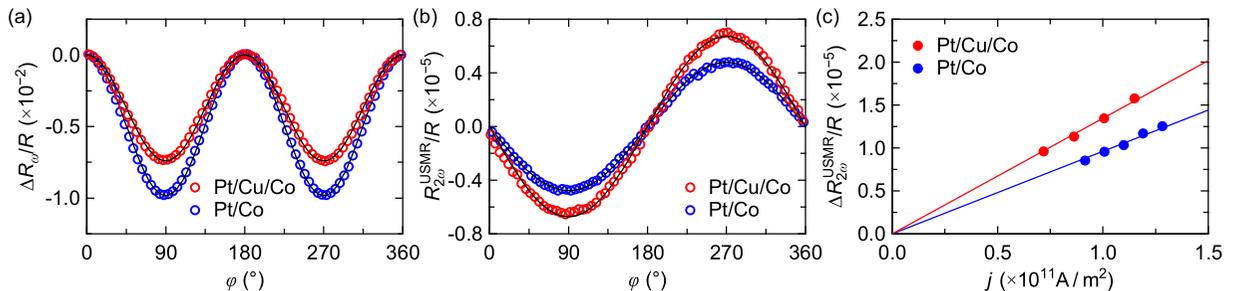


FIG. 2. (a) Angle dependence of normalized first harmonic resistance. (b) Angle dependence of normalized second harmonic resistance at current density of 1×10^{11} A/m², where the thermoelectric contribution is subtracted. (c) Normalized USMR as a function of current density. The solid lines in (a)–(c) show the fits to the data.

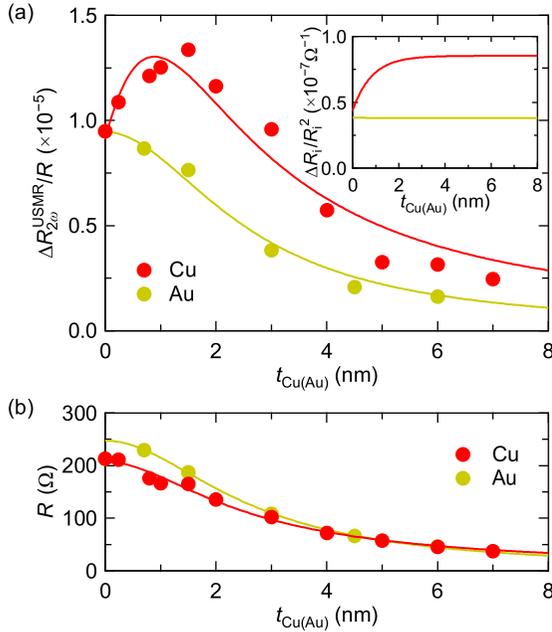


FIG. 4. (a) Normalized USMR and (b) longitudinal resistance as a function of Cu(Au) thickness together with fitting curves. The inset in (a) is the Cu(Au) thickness dependence of $\Delta R_i/R_i^2$ determined from the fitting and Eq. (3).

amplitude of current-induced spin accumulation at the ferromagnet/nonmagnet interface. Therefore, the result in which ξ_{DL} for Pt/Co and Pt/Cu(1.5 nm)/Co are the same within the error bars suggests that the modulation of spin accumulation cannot explain the enhanced USMR. This is reasonable because the spin current from the Pt layer shows little dissipation in the thin Cu layer owing to its long spin diffusion length.

Unlike the DL torque, the FL torque is clearly larger in the Pt/Cu(1.5 nm)/Co system, indicating the modulation of the imaginary part of the spin mixing conductance or the Rashba–Edelstein effect at the interface [25]. It has been reported that the Rashba–Edelstein effect can cause unidirectional magnetoresistance [9,26]. However, the FL torque is an order of magnitude smaller than the DL torque. Moreover, no apparent relationship between the FL torque and USMR was reported for a series of nonmagnet/Co systems [23]. Thus, we conclude that the change in FL torque is not directly related to the enhanced USMR in our system.

Considering the same amplitude of the spin injection, the plausible origin of the enhanced USMR is the increase in spin-dependent scattering at the nonmagnet/Co interface. To further investigate this interfacial effect, we measured the USMR by varying the interlayer thickness. Figure 4(a) shows the normalized USMR as a function of $t_{Cu(Au)}$. The USMR for the Pt/Cu/Co system initially increases with t_{Cu} , followed by a decrease for $t_{Cu} > 1.5$ nm. This result strongly demonstrates that forming a Cu/Co interface is responsible for the enhancement of USMR. The decay for the thick Cu regime is ascribed to the current shunting into the Cu layer, which does not contribute to the USMR. In contrast to Cu insertion, USMR monotonically decreases with t_{Au} in the Pt/Au/Co system. This means that the presence of an intervening nonmagnetic layer does not necessarily enhance the USMR. Thus, the ap-

propriate material combination is crucial for improving the USMR through interfacial engineering.

We will now discuss the $t_{Cu(Au)}$ dependence of the USMR quantitatively. We first assume that the resistance of the film consists of two independent resistors in parallel. One is the combined resistance of the Co layer and the nonmagnet/Co interface, which exhibits USMR. We describe this combined resistance and its resistance change as R_i and ΔR_i , respectively. The other is the combined resistance of the other metallic layers, which does not depend on the current. In this parallel circuit, the normalized USMR is written as follows [5,27]:

$$\frac{\Delta R_{2\omega}^{USMR}}{R} = R \frac{\Delta R_i}{R_i^2}. \quad (2)$$

The normal resistance R decreases with $t_{Cu(Au)}$, as shown in Fig. 4(b), leading to reduced USMR for thick Cu(Au) owing to the shunting effect. Note that the small increase in R by an ultrathin Au insertion can be attributed to diffusive scattering at the interface [28]. The solid lines in Fig. 4(b) represent the fitting result, where the resistivity of the Cu(Au) layer is assumed to be inversely proportional to $t_{Cu(Au)}$ (see Supplemental Material [22]). The fitting curves reproduced the experimental data well, and we obtained a bulk Cu(Au) resistivity of $4.7(1.5) \mu\Omega$ cm, which is in good agreement with the reported values [29,30].

In contrast to R , the thickness dependence of $\Delta R_i/R_i^2$ in Eq. (2) cannot be determined experimentally. Thus, we introduce the second assumption that the $t_{Cu(Au)}$ dependence of $\Delta R_i/R_i^2$ can be expressed as follows:

$$\frac{\Delta R_i}{R_i^2} = \Delta G \exp\left(-\frac{t_{Cu(Au)}}{t_0}\right) + \Delta G_0, \quad (3)$$

where ΔG , ΔG_0 , and t_0 are the variation of $\Delta R_i/R_i^2$ by inserting the Cu(Au) layer, $\Delta R_i/R_i^2$ for infinitely thick Cu(Au), and the characteristic decay length, respectively. This assumption means that $\Delta R_i/R_i^2$ becomes a constant when a continuous interface is formed. Note that this exponential approximation can well describe the spacer thickness dependence of GMR [31].

By combining Eqs. (2) and (3), we can fit the $t_{Cu(Au)}$ dependence of the USMR, and the experimental results are reproduced well, as shown in Fig. 4(a). The inset in Fig. 4(a) shows the $t_{Cu(Au)}$ dependence of $\Delta R_i/R_i^2$ calculated from Eq. (3). The Au interlayer does not modulate $\Delta R_i/R_i^2$, and thus USMR in the Pt/Au/Co system scales simply with R . This means that the t_{Au} dependence of USMR is completely described by the current shunting effect. By contrast, the Cu interlayer doubles $\Delta R_i/R_i^2$ with $t_0 = 0.8 \pm 0.4$ nm, indicating that USMR in the Pt/Cu/Co system is determined by increasing $\Delta R_i/R_i^2$ and decreasing R . For thin Cu interlayer, the modulation of $\Delta R_i/R_i^2$ is dominant, resulting in enhancement of USMR. For the thick Cu region, however, $\Delta R_i/R_i^2$ is saturated and a decrease in R dominates the t_{Cu} dependence of USMR. The acquired t_0 value is comparable to the characteristic decay length for the t_{Cu} dependence of the effective out-of-plane anisotropy field (see Supplemental Material [22]). This consistency substantiates our argument that replacing the Pt/Co interface with Cu/Co is essential

for the enhancement of USMR because the anisotropy field reflects the modulation of the perpendicular magnetic anisotropy at the nonmagnet/Co interface.

The enhancement of $\Delta R_i/R_i^2$ in the Pt/Cu/Co system should originate from the Cu/Co interface because the resistance and magnetoresistance in the Co layer cannot be modulated by a Cu insertion. Moreover, the fitting result in Fig. 4(b), which agrees well with the experimental data at $t_{\text{Cu}} = 0$ nm, also indicates that the nonmagnet/Co interface resistance does not change. Therefore, the enhancement of $\Delta R_i/R_i^2$ is attributed to a large ΔR_i at the Cu/Co interface. Because the Cu/Co structure is known to exhibit a large GMR, our result highlights the striking similarity between USMR and GMR.

In summary, we achieved an enhancement of USMR in a Pt/Co system by inserting a Cu interlayer. In contrast to Cu, Au insertion decreases the USMR, meaning that a modulation

of the USMR strongly depends on the spacer material. The spin current injection is not modulated by a Cu insertion, and the enhancement of the USMR is limited to the thin Cu regime. Therefore, the strong spin-dependent scattering at the Cu/Co interface is likely to play a crucial role in enhancing the USMR.

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