A magnetoresistance (MR) effect induced by the Rashba spin-orbit interaction was predicted, but not yet observed, in bilayers consisting of normal metal and ferromagnetic insulator. We present an experimental observation of this new type of spin-orbit MR (SOMR) effect in the Cu(Pt)/Y$_3$Fe$_5$O$_{12}$ (YIG) bilayer structure, where the Cu/YIG interface is decorated with nanosize Pt islands. This new MR is apparently not caused by the bulk spin-orbit interaction because of the negligible spin-orbit interaction in Cu and the discontinuity of the Pt islands. This SOMR disappears when the Pt islands are absent or located away from the Cu/YIG interface; therefore, we can unambiguously ascribe it to the Rashba spin-orbit interaction at the interface enhanced by the Pt decoration. The numerical Boltzmann simulations are consistent with the experimental SOMR results in the angular dependence of magnetic field and the Cu thickness dependence. Our finding demonstrates the realization of the spin manipulation by interface engineering.

**INTRODUCTION**

Relativistic spin-orbit interaction (SOI) plays a critical role in a variety of interesting phenomena, including the spin Hall effect (SHE) (1–3), topological insulators (4), and the formation of skyrmions (5, 6). In SHE, a pure spin current transverse to an electric current can be generated in conductors with strong SOI, such as Pt and Ta (7, 8). The inverse SHE (ISHE) is generally used to detect the spin current electrically by converting a pure spin current into a charge current (9, 10). It was recently discovered that the interplay of the SHE and ISHE in a nonmagnetic heavy metal (NM) with strong SOI in contact with a ferromagnetic insulator (FI) leads to an unconventional magnetoresistance (MR)—the spin Hall magnetoresistance (SMR), in which the resistance of the NM layer depends on the direction of the FI magnetization $M$ (11–13). SMR has been observed in several NM/FI systems and even in metallic bilayers (14–17). However, it has been argued that SMR may originate from the magnetic moment in the NM layer induced by the magnetic proximity effect (MPE) (18). These two mechanisms were proposed to be distinguished by the angular-dependent MR measurements (11, 13). Very recently, another type of MR, the Hanle MR (HMR), was demonstrated in a single metallic film with strong SOI owing to the combined actions of SHE and Hanle effect (19). HMR depends on the direction and the strength of the external magnetic field $H$, rather than that of $M$ in SMR. Within the framework of SMR, because of the negligible SOI in Cu (20), one would not expect any MR effect in a Cu/FI bilayer.

Recently, Grigoryan et al. (21) predicted a new type of MR effect in the NM/FI systems when a Rashba-type SOI is present at the interface between NM and FI. This new spin-orbit MR (SOMR) works even with light metals such as Cu or Al, with negligible bulk SOI, provided that the Rashba SOI is present at the NM/FI interface. However, because of the identical angular dependence on $M$ direction for SOMR and SMR, it is difficult to distinguish SOMR from SMR in systems like Pt/Y$_3$Fe$_5$O$_{12}$ (YIG), where both SOMR and SMR are present in principle. Here, we report the first observation of SOMR in a Cu/YIG bilayer, where the Rashba SOI at the Cu/YIG interface is enhanced by an ultrathin Pt layer (<1 nm). We also confirmed that SOMR almost disappears when Pt is placed inside or on the other side of the Cu layer, indicating that SMR from the ultrathin Pt layer cannot be the origin of the observed MR and that the Pt decoration of the Cu/YIG interface is crucial for SOMR. The observed SOMR has the same angular dependence as the SMR in Pt/YIG, in agreement with the SOMR prediction (21). The monotonous Cu thickness dependence of SOMR is different from the nonmonotonic dependence of SMR (13, 18). Both the angular and Cu thickness dependences of the observed MR are in good agreement with our Boltzmann simulations based on the SOMR mechanism. In addition, the MR shows two maxima as the Pt layer thickness increases, in sharp contrast to that of SMR (13, 22).

**RESULTS AND DISCUSSION**

**Sample morphology and structure**

The YIG films used in this study are 10 nm thick, unless otherwise stated, and grown by pulsed laser deposition (PLD) on Gd$_3$Ga$_5$O$_{12}$ (GGG) (111) substrates. The surface morphology of the YIG films was characterized by atomic force microscopy (AFM), as shown in Fig. 1A. The film is fairly smooth with a root mean square (RMS) roughness of 0.127 nm and a peak-to-valley fluctuation of 0.776 nm. The 0.4-nm-thick Pt layer, thinner than the peak-to-valley value of the YIG film, deposited on YIG by magnetron sputtering forms the nanosize islands with an RMS roughness of ~0.733 nm, as shown in Fig. 1B. This discontinuous Pt layer is nonconductive with a resistance over the upper limit of a multimeter. The surface roughness is reduced after the deposition of Cu onto Pt, as shown in fig. S1. Figure 1C presents the cross-sectional high-resolution transmission electron microscopy (HRTEM) image of the Au(3)/Cu(4)[Pt(0.4)]/YIG films, where the numbers are the thicknesses in nanometers. The YIG film is single-crystalline and smooth. The lattice constant of the YIG film is determined to be...
1.2234 nm, as compared to 1.2366 nm for the bulk YIG. A clear interface is observed between the metallic films and the YIG film. The metallic films are polycrystalline.

Field-dependent magnetization and transport measurements

Here, all the measurements were performed at room temperature. The YIG film is almost isotropic in the film plane, with a coercivity of about 0.4 Oe, as shown in Fig. 2A. Because of the large paramagnetic background of the GGG substrate, the magnetization of a thin YIG/GGG film in the out-of-plane geometry is difficult to measure. We measured the 400-nm-thick YIG/GGG(111) film instead. As shown in Fig. 2B, the magnetization is saturated at ~1800 Oe. The saturation magnetization $M_s$ of our YIG film is determined to be 164.5 emu/cm$^3$, as measured by ferromagnetic resonance (FMR) (see the Supplementary Materials). In comparison, the bulk YIG is 140 emu/cm$^3$.

Figure 2 (C and D) presents the resistivity $\rho$ as a function of $H$ for the Cu(2)[Pt(0.4)]/YIG(10) sample. In experiments, $H$ was applied (i) along the direction of the current $I$ (x axis), (ii) in the sample plane and perpendicular to the current direction (y axis), and (iii) perpendicular to the sample plane (z axis). The MR effects are present in all measurements. For $H$ along x and y directions, $\rho$ shows two peaks around the coercive fields of YIG. For $H$ along z direction, $\rho$ shows a minimum at $H = 0$ and remains almost a constant value above the saturation field. These features indicate that the MR effects are intimately correlated with $M$, meaning that the observed MR effects are not HMR.

Angular-dependent MR measurements

To further study the anisotropy of the MR effects in Cu[Pt]/YIG, we performed the angular-dependent MR measurements. Figure 3A shows $\Delta\rho/\rho$ of the Cu(3)[Pt(0.4)]/YIG(10) sample with rotation of $H$ in the xy ($\alpha$ scan), yz ($\beta$ scan), and xz ($\gamma$ scan) planes, where $\alpha$, $\beta$, and $\gamma$ are the angles between $H$ and x, z, and y directions, respectively, as defined in the inset of Fig. 3A. The applied magnetic field strength ($H = 1.5$ T) is large enough to align $M$ with $H$. The MR effect is anisotropic. The MR ratio, defined as $\Delta\rho/\rho = [\rho(\text{angle}) - \rho(\text{angle} = 90^\circ)]/\rho(\text{angle} = 90^\circ)$, in $\alpha$ and $\beta$ scans is about 0.012%, which is comparable to the SMR ratio in Pt/YIG (see fig. S3) (11, 13, 23).

Next, we investigated the origin of the observed MR effect. Considering that Pt on YIG may suffer from the MPE-induced ferromagnetic moment and the corresponding anisotropic MR (AMR) (24), we replaced Pt by a 0.4-nm-thick Au layer, which is well known to have a negligible MPE (25). The MR effect of 0.002% still appears as shown in Fig. 3B, comparable to the SMR ratio in Au/YIG (see fig. S4), ruling out MPE as the origin of the observed MR. Furthermore, the MR ratios of the Cu(3)[Pt(0.4)]/YIG(10) sample in $\alpha$ and $\beta$ scans are comparable and almost one order of magnitude larger than the MR ratio in the $\gamma$ scan. This is different from AMR of a ferromagnetic metal, where the MR ratio in $\alpha$ and $\gamma$ scans is much larger than that in the $\beta$ scan (11, 14, 24). Therefore, the MPE-induced AMR can be ruled out.

The behaviors of the MR angular dependence follow the SMR scenario well (11, 13–15, 17). However, with several control experiments, we can unambiguously exclude SMR as the explanation for our observations.

First, the observed MR amplitude cannot be explained by SMR. In our samples, the 0.4-nm-thick ultrathin Pt layer is nonconductive, and the conductivity of bulk Pt is about one order of magnitude smaller than that of bulk Cu, meaning that the current mainly passes through the Cu layer. We prepared a 3-nm-thick single-layer Cu on YIG without interface decoration and performed the MR angular-dependent measurement in the $\alpha$ scan. MR is not observed, as shown in Fig. 3B, evidencing that the Pt-decorated interface is indispensable. A conductive 0.4-nm-thick Pt layer is not available experimentally. Considering that a small fraction of current may flow in the Pt islands, there is a possibility of the occurrence of SMR from the Pt islands. According to the reported SMR results in Pt/YIG bilayers, the SMR ratio in Pt/YIG decreases rapidly with decreasing Pt thickness when the Pt thickness is less than about 3 nm (13, 18). The SMR ratio of Pt(0.4)/YIG is extrapolated to be well below 0.01% from the previously reported $\Delta\rho/\rho$ versus Pt thickness data (13, 18). Considering the pronounced shunting current of the highly
To identify the physical origin of the observed unusual MR, we carried out the Cu thickness induced SMR. Thus, we conclude that the observed MR effect is not SMR. Because both samples are fabricated under the same conditions as the Cu inside the Cu layer: [Pt(0.4)]Cu(3)/YIG and Cu(1)[Pt(0.4)]Cu(3)/YIG. These results rule out the Pt-Cu alloying MR vanishes in the [Pt(0.4)]Cu(3)/YIG and Cu(1)[Pt(0.4)]Cu(3)/YIG samples, the intermixing of Pt and Cu should be similar. The reason for this is that the Pt decoration enhances the Rashba SOI at the Cu/YIG interface. The monotonous NM thickness dependence decreases with increasing Cu, highlighting the importance of the Pt-Cu/YIG interface.
Finally, to further differentiate SOMR from SMR, we carried out the Pt thickness-dependent MR measurements. The interface scattering has minor contribution to the reduction of the interface roughness and the rapid decrease of the MR ratio extracted from Fig. 4A. This result means that the rough surface alone cannot cause the SOMR.

CONCLUSIONS
In conclusion, we report the first observation of the SOMR effect predicted recently [21] at room temperature in Cu/YIG films with the Pt decoration at the interface. We show that this MR effect is caused by the enhanced Rashba SOI at the Pt-decorated interface. The angular dependence of SOMR is similar to that of SMR, but all other features are different, such as the increasing MR with decreasing Cu thickness.

The amplitude of the SOMR ratio is comparable to that of the SMR ratio in Pt/YIG, highlighting the importance of the NM/FI interfaces. Our finding demonstrates the possibility of realizing spin manipulation by interface decoration.

MATERIALS AND METHODS
The single-crystalline YIG films were epitaxially grown on GGG (111) substrates by the PLD technique using a KrF excimer laser with a wavelength of 248 nm. The PLD system was operated at a laser repetition rate of 4 Hz and an energy density of 10 J/cm². The distance between the substrate and the target is 50 mm. Before film deposition, the chamber was evacuated to a base pressure of 1 × 10⁻⁷ torr. The YIG films were deposited at ~730°C in an oxygen pressure of 0.05 torr. The growth of the YIG films was monitored by in situ reflection high-energy electron diffraction. The structure was further examined by x-ray diffraction and HRTEM. The magnetic properties of all YIG films were characterized using a vibration sample magnetometer. Then, we used magnetron sputtering to deposit polycrystalline metallic films onto the YIG films via dc sputtering at room temperature with a shadow mask to define 0.3-mm-wide and 3-mm-long Hall bars. The deposition rate was calibrated by x-ray reflectivity. After the metallic film deposition, the samples were immediately mounted and transferred into a vacuum chamber for the transport measurements to minimize the metal oxidation. The resistance was measured by a Keithley 2002 multimeter in a four-probe mode. For the angular-dependent MR measurements with a magnetic field larger than 5000 Oe, the resistance was monitored as the magnetic field was rotated. The angular-dependent MR measurements with a magnetic field larger than 5000 Oe were performed in a physical property measurement system equipped with a rotatory sample holder.

SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/1/eaao3318/DC1

section S1. AFM images of Cu(3)/Pt(0.4)/YIG(10)/GGG(111).
section S2. Magnetic properties of the YIG films.
section S3. SMR in Pt/YIG.
section S4. SMR and AFM image of Au/YIG.
section S5. First-principles calculations.
section S6. Boltzmann simulations.
fig. S1. AFM images of Cu(3)/Pt(0.4)/YIG(10)/GGG(111).
fig. S2. FMR of the YIG films.
fig. S3. SMR in Pt/YIG.
fig. S4. AFM image of Au/YIG.
fig. S5. The band structures of Cu, Au/Cu/Au, and Pt/Cu/Pt.
fig. S6. The spin textures of outer and inner bands.
fig. S7. The Rashba splitting in Cu/Pt/Cu.
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Observation of spin-orbit magnetoresistance in metallic thin films on magnetic insulators
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