

# Spin current generation in highly conductive Ru/Cu epitaxial heterostructures

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Significant spin current generation from highly conductive materials is promising for spintronic applications, such as spin-orbit-torque magnetic random-access memories. In this work, we fabricated epitaxial Ru/Cu heterostructures with interface engineering where 1-nm-thick Ru and Cu layers were alternately deposited at the interface. The spin current generation in the heterostructures was evaluated by unidirectional spin Hall magnetoresistance and spin-torque ferromagnetic resonance. A sizable spin Hall efficiency ( $\sim 2\%$ ) was achieved in the Ru/Cu sample with a sharp interface which may result from the interface spin filtering effect. Increased spin Hall efficiency ( $\sim 4\%$ ) was observed in the interface-engineered samples which could be attributed to the intrinsic contribution from lattice distortion and local band structure near the interface. The effective spin Hall conductivity was estimated to be  $3\sim 5 \times 10^5 \frac{\hbar}{2e} \Omega^{-1} \text{m}^{-1}$ , which is comparable to that of Platinum.

**Key words**—Spin current, Interface spin-filtering, Intrinsic contribution, Ru/Cu heterostructures

## I. INTRODUCTION

THE generation of large spin current by spin-orbit effects, e.g., the spin Hall effect (SHE), in well-designed materials/heterostructures, is a key technology for the development of spintronic devices for energy-efficient electronic applications, such as magnetoresistive random access memories, spin-orbit oscillators, and spin-orbit magnetoresistance sensors. The spin angular momentum of the spin current can be transferred to an adjacent ferromagnetic (FM) layer, as spin-orbit torques (SOTs), to manipulate the magnetization of the FM layer. Also, the spin-dependent accumulation at the interface due to the spin current generation can contribute to a change in resistance of the spin-orbit heterostructures. For achieving practical applications of SOT-based devices, it is necessary to find or to design materials exhibiting a large efficiency of spin-charge conversion. Heavy  $5d$  transition metals and topological materials were reported to have large spin Hall angles due to their significant spin-orbit couplings; however, several complications, such as their high cost, phase instability, and high resistivity, hinder the integration of those materials to the state-of-the-art electronics.

Highly conductive copper is commonly used in electronic circuits, but its spin Hall angle is extremely small due to its weak spin-orbit coupling. However, a large spin current was generated in surface-oxidized copper films, which was interpreted by the interface effect [1] and the formation of spin vorticity resulting from a gradient conductivity in the films [2]. These results indicate nanostructure engineering and interfacial design in Cu-based heterostructures are effective approaches to realize high efficiencies of spin-current generation. In this work, we developed fully epitaxial Ru/Cu heterostructures and investigated the spin current generation by unidirectional spin Hall magnetoresistance (USMR) and spin-torque ferromagnetic resonance (ST-FMR), as illustrated in Fig.1.

## II. EXPERIMENTAL

All the thin films were deposited using magnetron sputtering with a base pressure of  $\sim 4 \times 10^{-7}$  Pa. The Ru/Cu-based heterostructures have the stacking structures of sample #1:  $\text{Al}_2\text{O}_3(0001)$  substrate//Ru (10)/Cu (10)/  $\text{Ni}_{81}\text{Fe}_{19}(\text{NiFe})$  (5)/cap, sample #2:  $\text{Al}_2\text{O}_3(0001)$  substrate//Ru (9)/[Cu (1)/Ru (1)]/Cu (9)/NiFe (5)/cap, sample #3:  $\text{Al}_2\text{O}_3(0001)$  substrate//Ru (8)/[Cu (1)/Ru (1)]<sub>2</sub>/Cu (8)/NiFe (5)/cap. The unit in parentheses is nanometer. Reflection high energy electron diffraction (RHEED) and atomic force microscopy (AFM) were used to examine the surface structure and morphology. High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) combining with nano-beam electron diffraction and energy dispersive X-ray spectroscopy (EDS) was used for microstructure analysis of sample cross sections. Hall bars and co-plane wave waveguide devices were patterned by UV lithography techniques. USMR signal was obtained by second harmonic measurements. ST-FMR was performed by a rf signal generator with amplitude modulation and a lock-in amplifier. The external magnetic field swept from 0 to 3 kOe at an in-plane angle  $\varphi = 0\sim 360^\circ$  during the measurements.

## III. RESULTS AND DISCUSSION

The characterization of surface and microstructure indicates that the Ru/Cu films show fully epitaxial growth and high crystallinity. For each layer of Ru, Cu, and NiFe in all the samples, sharp streaks with Kikuchi lines were observed in the RHEED patterns along the incident azimuth angles parallel to the  $\text{Al}_2\text{O}_3$   $[10\bar{1}0]$  and  $[11\bar{2}0]$  directions. The epitaxial growth orientation was confirmed as be Ru (0002)/Cu (111)/NiFe (111). Also, AFM images ( $1 \times 1 \mu\text{m}^2$  area) show very flat surfaces where the average roughness is  $\sim 0.2$  nm and the value of peak-to-valley is  $\sim 2.5$  nm. Moreover, atomic-resolution

HAADF-STEM observations reveal that the Ru shows a hcp structure with A-B-A-B lattice plane stacking while the Cu holds a fcc structure with A-B-C-A-B-C lattice planes. The sample #1 shows a sharp interface with perfect lattice matching between Ru (10 nm) and Cu (10 nm) which is also confirmed in the EDS elemental profiles. For sample #2, having a Ru (1 nm)/Cu (1 nm) intercalation, very clear atomic-level STEM images have also been collected with the bottom Ru and top Cu layers respectively showing hcp and fcc structures. Nevertheless, although relatively flat interfaces can be identified in the sample, there are more or less localized mixing or dislocations or atomic swapping at the interfaces.

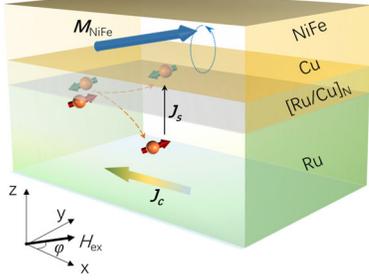


Fig. 1. Illustration of epitaxial Ru/Cu sample stack and spin current generation. FMR signal can be induced by the transfer of spin angular momentum from an oscillating spin current to the moment of NiFe layer.

In order to examine the spin current generation in the Ru/Cu samples, we carried out USMR and ST-FMR measurements. The USMR originates from the spin accumulation at the spin source/FM interface due to the spin current generation. The longitudinal resistance changes depending on the configuration of the spin polarization vector of spin current and the magnetization of FM. Obvious USMR signals were observed in all samples. Interestingly, an increase in USMR was achieved in the interface-engineered samples (#2 and #3). A linear relationship between charge current density and USMR ratio is also shown, which suggests that the origin of USMR is related to spin-dependent accumulation. Furthermore, by applying a rf current in the Ru/Cu layer, an oscillating transverse spin current was generated. FMR signals were induced by the transfer of spin angular momentum of the spin current to the FM layer. The FMR signal contains a symmetric Lorentzian line shape ( $V_S$ ) and an antisymmetric Lorentzian line shape ( $V_A$ ). The  $V_S$  is related to the damping-like torque from the spin current while the  $V_A$  results from the Oersted field and the field-like spin torque. All the samples show clear ST-FMR spectra. Figure 2 is a representative ST-FMR spectrum measured at  $\varphi = 45^\circ$  with frequency  $f = 8$  GHz and power = 11 dBm for sample #3. Based on our ST-FMR results, we quantitatively estimated the efficiency of spin current generation in the three samples. A sizeable spin Hall efficiency of  $\sim 2\%$  was achieved in the sample #1. The absolute value is much larger than  $+0.6\%$  and  $\sim 0$  in pure Ru [3] and pure Cu [2], respectively. Also, the sign is opposite to that in pure Ru. The results suggest that the interface may play an important contribution to the generation of spin current in the epitaxial Ru/Cu bilayers, other than the bulk contribution. Since the weak spin-orbit coupling of both Ru and Cu, the spin-filtering effect [4] at the interface, related to spin-dependent

transmission and reflection electrons from the spin-orbit scattering and relative magnitude of charge current in the two layers [5], may be responsible for the spin current generation. For the interface-engineered samples (#2 and #3), it is calculated that the spin Hall efficiency increases to  $\sim 4\%$ , which may be due to the intrinsic contribution of SHE from the lattice distortion and the local band structure change near the interfaces [6]. The effective spin Hall conductivity is estimated to be  $3\sim 5 \times 10^5 \frac{\hbar}{2e} \Omega^{-1} \text{m}^{-1}$ , which is as large as that of platinum.

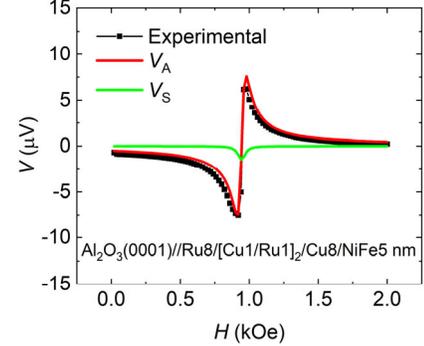


Fig. 2. Representative ST-FMR spectrum ( $f = 8$  GHz,  $P = 11$  dBm, and  $\varphi = 45^\circ$  and) in the sample of Ru8/[Cu1/Ru1]<sub>2</sub>/Cu8/NiFe5 nm.  $V_A$  represents the fitting of antisymmetric Lorentzian line shape of the ST-FMR signal from  $H_{\text{Oersted}}$  and  $H_{\text{FL}}$  while  $V_S$  is the fitting of symmetric Lorentzian line shape induced by the oscillating transverse spin current.

#### IV. CONCLUSION

In summary, we deposited fully epitaxial Ru/Cu heterostructures with interface engineering by alternating nanometer Ru/Cu bilayers at the interface and investigated the spin current generation in the heterostructures by USMR and ST-FMR in combination with a ferromagnetic NiFe layer. Sizable spin Hall efficiencies of around  $\sim 2\% \sim 4\%$  and large spin Hall conductivities of  $3\sim 5 \times 10^5 \frac{\hbar}{2e} \Omega^{-1} \text{m}^{-1}$  are achieved. The large spin Hall conductivities may be useful for SOT-based spintronic device applications.

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