Spin-current-driven thermoelectric generation based on interfacial spin-orbit coupling

A. Yagmur,1,a) S. Karube,2,3 K. Uchida,1,4,b) K. Kondou,3 R. Iguchi,1 T. Kikkawa,1,5
Y. Otani,2,4 and E. Saitoh1,5,6,7
1Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
2Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
3Center for Emergent Matter Science, RIKEN, Wako, Saitama 351-0198, Japan
4PRESTO, Japan Science and Technology Agency, Saitama 332-0012, Japan
5WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
6Spin Quantum Rectification Project, ERATO, Japan Science and Technology Agency, Sendai 980-8577, Japan
7Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan

(Received 27 April 2016; accepted 2 June 2016; published online 14 June 2016)

The longitudinal spin Seebeck effect (SSE) in Bi2O3/Cu/yttrium-iron-garnet (YIG) devices has been investigated. When an out-of-plane temperature gradient is applied to the Bi2O3/Cu/YIG device, a spin current is generated across the Cu/YIG interface via the SSE and then converted into electric voltage due to the spin–orbit coupling at the Bi2O3/Cu interface. The sign of the SSE voltage in the Bi2O3/Cu/YIG devices is opposite to that induced by the conventional inverse spin Hall effect in Pt/YIG devices. The SSE voltage in the Bi2O3/Cu/YIG devices disappears in the absence of the Bi2O3 layer and its thermoelectric conversion efficiency is independent of the Cu thickness, indicating the important role of the Bi2O3/Cu interface. This result demonstrates that not only the bulk inverse spin Hall effect but also the spin–orbit coupling near the interface can be used for SSE-based thermoelectric generation. Published by AIP Publishing.

[http://dx.doi.org/10.1063/1.4953879]

Conversion between charge and spin currents is an essential phenomenon that plays an important role in spintronic devices.1 This phenomenon has been demonstrated mostly in metals2,3 and semiconductors4,5 possessing large spin–orbit coupling (SOC). In the spin Hall effect (SHE), a charge current flowing through a material generates a transverse spin current polarized perpendicular to the plane defined by the charge and spin currents.6 Its reciprocal effect, the inverse spin Hall effect (ISHE) has been widely used to convert a spin current into a charge current, enabling electric detection of spin-current phenomena.7–10 At surfaces, interfaces, or two-dimensional electron gases, another SOC called the Rashba-Edelstein effect (REE) appears.11 Recently, Rojas-Sánchez et al. demonstrated the spin-to-charge current conversion based on the reciprocal effect of the REE, the inverse Rashba-Edelstein effect (IREE),12 by using Bi(111)/Ag Rashba interfaces. In the IREE, when a spin current polarized along the y direction is injected vertically into the Rashba interface (in the x–y plane), the spin-split dispersion curve of the interface is shifted along the +kx or –kx axis depending on the spin polarization, resulting in the generation of a charge current along the x direction. The IREE has been observed in Bi(111)/Ag/Permalloy(Py) trilayer devices by using microwave spin pumping as a tool to inject a spin current.12 After this demonstration, several systematic researches were reported, which provide a crucial piece of information to understand the mechanism of the IREE.13–17 In Ref. 18, the spin-to-charge current conversion in Bi2O3/Cu/Py devices has been demonstrated by using the microwave spin-pumping method, where Bi2O3 is a nonmagnetic band insulator and its Bi atoms may generate large electric fields and SOC closed to the interface because of the heavy nucleus. This result indicates that the IREE can arise even at insulating oxide/metal interfaces; based on the spin-pumping measurements, the spin-to-charge current conversion coefficient for the Bi2O3/Cu interface on Py was estimated to be comparable to or larger than that for the Bi(111)/Ag interface.

The spin Seebeck effect (SSE) is a thermoelectric conversion phenomenon that refers to the generation of a spin current as a result of a temperature gradient in magnetic materials. When a conductor is attached to a magnetic material, the spin current generated by the SSE is injected into the conductor.19–25 This spin current is then converted into an electric field via the SOC in the conductor. The SSE is attracting rapidly growing interest in spintronics owing to its potential application to thermoelectric conversion,24 and the investigation of the SSE using various mechanisms and materials is important for improving the thermoelectric conversion efficiency.21,22,25,26 However, in most of the SSE experiments to date, the spin-to-charge current conversion based on the bulk SOC, i.e., the ISHE, has been used,23 except for the experiments using topological insulators.27

In this letter, we report the observation of the SSE by means of the interfacial SOC using Bi2O3/Cu/yttrium-iron-garnet (YIG) trilayer systems. Here, YIG is one of the most widely used materials for spin-current studies since it has a small Gilbert damping constant, long spin-wave-propagation length, and high electrical resistivity.28 We select Cu as an interlayer since it has low bulk SOC and long spin-diffusion length,29 therefore, in the Bi2O3/Cu/YIG

a)ahmetyagmur@imr.tohoku.ac.jp
b)kuchida@imr.tohoku.ac.jp
systems, the spin-to-charge current conversion is expected to appear only at the Bi2O3/Cu interface, enabling the separation of the interfacial SOC from the conventional bulk ISHE. Importantly, since both YIG and Bi2O3 are good insulators, the output voltage can be generated only in the Cu layer or at the interfaces.

A schematic illustration of our Bi2O3/Cu/YIG trilayer sample is shown in Fig. 1(a). A single-crystalline YIG with the thickness of tYIG = 112 μm was grown on a single-crystalline Ge3Ga5O12 (GGG) (111) substrate with the thickness of 0.4 mm by a liquid-phase-epitaxy method. The lengths along the x and y directions of the YIG/GGG substrate are 7 mm and 5 mm, respectively. The Cu and Bi2O3 layers were fabricated on YIG by electron-beam evaporation in a high vacuum of about 10−6 Pa. The Bi2O3/Cu bilayer film was patterned into 5 strips with 5-mm length and 0.3-mm width by photolithography and lift-off methods to prepare 5 samples with different Cu thicknesses on the same YIG/GGG substrate. The thickness of the Cu layer is changed from tCu = 33 nm to 42 nm by using a linear shutter system, while the thickness of the Bi2O3 layer is fixed at 20 nm. To obtain good electrical contacts to the Cu layer, two Au(200 nm)/Ti(5 nm) electrodes were fabricated at the ends of the Bi2O3/Cu strips. In Fig. 1(b), we show an atomic force microscopy image and cross-sectional profile of the Bi2O3(20 nm)/Cu(33 nm)/YIG(112 μm) sample. The peak-to-valley height of the Bi2O3 layer was observed to be ~4 nm, which is larger than the surface roughness of Pt/YIG systems, where typical peak-to-valley height is ~1 nm. The Cu layer on YIG may also have surface roughness comparable to that of the Bi2O3 layer; probably due to the surface roughness, the electrical resistivity of the Cu films on YIG (6 μΩ cm) is greater than that on metals (4.5 μΩ cm).18 Therefore, we fabricated the Cu films with the thickness of tCu > 30 nm, much larger than their roughness.

To detect the interfacial spin-to-charge current conversion induced by the SSE in the Bi2O3/Cu/YIG samples, we performed thermoelectric voltage measurements in a longitudinal configuration [see Fig. 1(a)].30 Here, when a temperature gradient ∇T is applied to the Bi2O3/Cu/YIG sample perpendicular to the interfaces, a spin current is thermally generated in the Cu layer along the ∇T direction. This spin current reaches the Bi2O3/Cu interface owing to the long spin-diffusion length of Cu. If the Bi2O3/Cu interface exhibits the SOC, the spin current is converted into a charge current. When the magnetization of YIG is along the y direction, the charge current is expected to be generated along the x direction, which is of the same symmetry as the SSE-induced ISHE [see Fig. 1(a)]. This interfacial spin-to-charge current conversion can be detected by measuring electric voltage (SSE voltage) in the Cu layer.

The experimental setup used in this study is similar to that used in the conventional SSE studies.30 To apply ∇T along the z direction, the Bi2O3/Cu/YIG samples were sandwiched between two AlN heat baths of which the temperatures were stabilized to 300 K + ΔT and 300 K. A Peltier thermoelectric module was used to generate the temperature difference ΔT, which was measured with two thermocouples. By applying ∇T and an external magnetic field H (with the magnitude H) to the samples, we measured the voltage V between the Au/Ti electrodes connected to the Cu layers of the Bi2O3/Cu/YIG samples. When |H| > 100 Oe, the magnetization of the YIG layer is aligned along the H direction [see Fig. 2(a)].

Figure 2(b) shows V in the Bi2O3/Cu(tCu = 33 nm)/YIG sample as a function of H at ΔT = 6 K for θ = 90° and θ = 0°, where θ denotes an angle between H and the x direction in the x-y plane [see Fig. 1(a)]. We found that a clear V signal appears for θ = 90°, while it disappears for θ = 0°. The measured V-H curve for θ = 90° clearly reflects the magnetization curve of the YIG layer [compare Figs. 2(a) and 2(b)]. The magnitude of the V signal is proportional to ΔT and its sign for finite values of ΔT is reversed in response to the magnetization reversal of the YIG layer [see Figs. 2(c) and 2(d)]. These behaviors are in good agreement with the features of the SSE in the longitudinal configuration.20 Importantly, the V signal observed here is opposite in sign to the SSE voltage in the conventional Pt/YIG system [see the inset of Fig. 2(b)], which is consistent with the spin-pumping experiments using the Bi2O3/Cu/Pt devices.18 Therefore, the SSE voltage in the Bi2O3/Cu/YIG sample cannot be explained by the bulk ISHE in Cu since the sign of the spin Hall angle of Cu is shown to be the same as that of Pt.31

To clarify the origin of the SSE voltage in the Bi2O3/Cu/YIG sample, we also fabricated two control samples and performed the same measurements using these samples. The first sample is a Cu/YIG system without the Bi2O3 layer. We
found that the SSE voltage disappears in the absence of the Bi$_2$O$_3$ layer [see Fig. 3(b)], confirming that the contribution of the ISHE in the Cu layer is negligibly small due to its low bulk SOC. This result also implies that possible contribution of the interfacial SOC at the Cu/YIG interface is irrelevant to the observed $V$ signal. The other control sample is an Al$_2$O$_3$/Cu/YIG system in which the Bi$_2$O$_3$ layer replaced with an Al$_2$O$_3$ film with the thickness of 20 nm [see Fig. 3(c)]. The SSE voltage was found to disappear also in the Al$_2$O$_3$/Cu/YIG sample, indicating the important role of the heavy-element-based oxide in the voltage generation. These observations allow us to conclude that the SSE voltage in the Bi$_2$O$_3$/Cu/YIG sample is attributed to the interfacial SOC at the Bi$_2$O$_3$/Cu interface.

We also investigated the SSE voltage in the Bi$_2$O$_3$/Cu/YIG samples by changing the thickness of the Cu layer $t_{Cu}$. We found that the magnitude of the $V$ signal in the Bi$_2$O$_3$/Cu/YIG samples gradually and monotonically decreases with increasing $t_{Cu}$ [see Fig. 4(a)]. In these samples, the resistivity of the Cu layer $\rho_{Cu}$ slightly decreases with increasing $t_{Cu}$, and the $t_{Cu}$ dependence of the sheet resistance of the Cu layer $R_s$ ($=\rho_{Cu}/t_{Cu}$) is similar to that of the $V$ signal [see Fig. 4(b)]. By combining the $V$ and $R_s$ data, we estimated the $t_{Cu}$ dependence of the charge current $I_C$ ($=V/R_s$) generated in the Bi$_2$O$_3$/Cu/YIG samples. Significantly, as shown in Fig. 4(c), the $I_C$ values were found to be independent of the thickness of the Cu layer. Since the resistivity variation in our Bi$_2$O$_3$/Cu/YIG samples is very small, the $t_{Cu}$ dependence of $I_C$ observed here is consistent with the characteristic of the IREE induced by the spin pumping reported in Ref. 18, on the basis that the thickness of the Cu layers is

![FIG. 2. (a) The $M$-$H$ curve, normalized by the saturation magnetization $M_S$, of the YIG crystal used in this study, which was measured with a vibrating sample magnetometer. (b) $H$ dependence of $V$ in the Bi$_2$O$_3$(20 nm)/Cu($t_{Cu} = 33$ nm)/YIG(112 l m) sample at $\Delta T = 6$ K, measured when the $H$ direction was along the $y$ ($\theta = 90^\circ$) and $x$ ($\theta = 0^\circ$) directions. $\nabla T$ was applied along the $+z$ direction. The inset to (b) shows the $H$ dependence of $V$ in the Pt(5 nm)/YIG(112 l m) sample at $\Delta T = 6$ K, measured when the $H$ direction was along the $y$ ($\theta = 90^\circ$) direction. (c) $\Delta T$ dependence of $V$ in the Bi$_2$O$_3$/Cu/YIG sample at $H = +200$ Oe and $\theta = 90^\circ$. (d) $H$ dependence of $V$ in the Bi$_2$O$_3$/Cu/YIG sample for various values of $\Delta T$ at $\theta = 90^\circ$.](#)

![FIG. 3. $H$ dependence of $V$ in the (a) Bi$_2$O$_3$/Cu/YIG, (b) Cu/YIG, and (c) Al$_2$O$_3$/Cu/YIG samples at $\Delta T = 6$ K and $\theta = 90^\circ$.](#)

![FIG. 4. $t_{Cu}$ dependence of (a) $|V|/\Delta T$ in the Bi$_2$O$_3$/Cu/YIG samples at $H = +200$ Oe, (b) the resistivity of the Cu layer $\rho_{Cu}$, and (c) $|I_C|/\Delta T$ ($=V/R_s\Delta T$) at $H = +200$ Oe. The inset to (b) shows the $t_{Cu}$ dependence of the sheet resistance of the Cu layer $R_s$ ($=\rho_{Cu}/t_{Cu}$).](#)
much smaller than the spin-diffusion length and the spin current injected from YIG reaches the Bi$_2$O$_3$/Cu interfaces without decaying in the Cu layers. This result supports our interpretation that the SSE voltage in the Bi$_2$O$_3$/Cu/YIG samples is due to the SOC near the Bi$_2$O$_3$/Cu interface.

Finally, we mention the magnitude of the SSE voltage in the Bi$_2$O$_3$/Cu/YIG samples. As shown in Fig. 2(b), the SSE voltage in the Bi$_2$O$_3$/Cu/YIG without decaying in the Cu layers. This result supports our interpretation that the SSE voltage in the Bi$_2$O$_3$/Cu/YIG and Al$_2$O$_3$/Cu/YIG devices is attributed to the SOC near the Bi$_2$O$_3$/Cu interface, not to the conventional bulk ISHE or other artifacts, by comparing the magnitude of the SSE voltage induced by the interfacial SOC and by investigating the Cu-thickness dependence. Although the magnitude of the SSE voltage induced by the interfacial SOC in the Bi$_2$O$_3$/Cu/YIG devices is much smaller than that induced by the ISHE in conventional Pt/YIG devices, our thermoelectric generation in Bi$_2$O$_3$/Cu/YIG devices. We confirm that the SSE voltage in the Bi$_2$O$_3$/Cu/YIG devices is attributed to the SOC near the Bi$_2$O$_3$/Cu interface, not to the conventional bulk ISHE or other artifacts, by comparing the voltage with that in Cu/YIG and Al$_2$O$_3$/Cu/YIG devices and by investigating the Cu-thickness dependence. Although the magnitude of the SSE voltage induced by the interfacial SOC in the Bi$_2$O$_3$/Cu/YIG devices is much smaller than that induced by the ISHE in conventional Pt/YIG devices, our results will expand the choice of materials, including noble-metal-free materials, for SSE-based thermoelectric generation and provide new strategies for improving its efficiency because the interfacial SOC depends on oxide/metal combinations. Furthermore, the SSE device based on the interfacial SOC can be potentially tuned by applying an external electric field to the interface, which cannot be realized in conventional bulk metals; its demonstration is one of the future challenges.

The authors thank T. Seki and K. Takanashi for their assistance in magnetometry measurements. This work was supported by PRESTO “Phase Interfaces for Highly Efficient Energy Utilization” from JST, Japan, Grant-in-Aid for Scientific Research (A) (No. 15H02012), Grant-in-Aid for Scientific Research on Innovative Area, “Nano Spin Conversion Science” (No. 26103002, 26103005) from MEXT, Japan, NEC Corporation, the Noguchi Institute, and E-IMR, Tohoku University. S.K. is supported by JSPS through Program for Leading Graduate Schools (MERIT). T.K. is supported by JSPS through a research fellowship for young scientists (No. 15J08026).