All-oxide spin Seebeck effects

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We report the observation of longitudinal spin Seebeck effects (LSSEs) in an all-oxide bilayer system comprising an IrO2 film and an Y3Fe5O12 film. Spin currents, which are generated by a temperature gradient across the IrO2/Y3Fe5O12 interface, were detected as a voltage via the inverse spin Hall effect in the conductive IrO2 layer. This voltage is proportional to the magnitude of the temperature gradient; its magnetic field dependence is consistent with the characteristics of LSSEs. This demonstration may lead to the realization of low-cost, stable, transparent spin-current-driven thermoelectric devices. © 2015 The Japan Society of Applied Physics

The spin Seebeck effect (SSE) generates a spin voltage in a magnetic material as a result of a temperature gradient.3–21) Because the thermally generated spin voltage induces a spin current across the interface between the magnetic material and an adjacent conductive material, it can be detected as a voltage via the inverse spin Hall effect (ISHE) in the conductive layer. Therefore, a magnetic/conductive bilayer system is commonly used for studying the SSE.9–21) The SSE has attracted increasing attention because of possible applications in thermoelectric conversion and spintronic devices.18,22) Investigation of the SSE in various materials is important for further improvement of thermoelectric and thermo-spin conversion efficiency. However, to date, all experimental studies on the SSE have been performed using simple metals as conductive layers; although, a wide variety of materials have been investigated for the magnetic layer.9,10,21)

As alternate conductive materials for SSE devices, conductive oxides are good candidates because of their low cost, good chemical stability, and ease of fabrication. In addition, many conductive oxide films are transparent. Therefore, conductive oxides enable the construction of transparent thermoelectric and thermo-spin devices, which are more suitable for applications requiring transparency, such as applications to smart windows.

In this work, we report the observation of the SSE in an all-oxide bilayer system comprising a conductive IrO2 layer and a ferrimagnetic insulator Y3Fe5O12 (YIG). YIG is one of the most widely used materials for spin-current studies, because it has a small Gilbert damping constant, long spin-wave-propagation length, and high electrical resistivity.11–19,23–25) We select IrO2 for detecting the SSE because a relatively high spin-Hall angle has been reported in this conductive oxide.26–28) Importantly, IrO2 is an n-type transparent semiconductor with a work function that is very close to that of metals such as Ag,29) making it easier to form an Ohmic contact between IrO2 and metallic electrodes.28) This property is useful for detecting the ISHE in IrO2 if the output voltage is on the order of microvolts or less. In contrast, the work function of another typical conductive oxide, indium tin oxide (ITO), is sensitive to the interface and preparation conditions,30) which makes it difficult both to create Ohmic contacts between ITO and metallic electrodes and to observe the SSE using ITO films.

To investigate the SSE in an all-oxide system, a longitudinal configuration is employed in this work.12,13) In Fig. 1(a), we present a schematic illustration of the experimental configuration and the sample structure of the IrO2/YIG bilayer film used for measuring the longitudinal SSE (LSSE). The single-crystalline YIG film was grown on a 0.5-mm-thick (111) Gd3Ga5O12 (GGG) substrate using a liquid phase epitaxy method. The thickness of the YIG film is roughly 4.5 µm. A 30-mm-thick IrO2 film was then deposited on the YIG film using an RF sputtering method at room temperature. As shown in Fig. 1(b), the light transmittance of the 30-mm-thick IrO2 film is much higher than that of a conventional 10-mm-thick Pt film in the visible light range.

The LSSE measurements were performed using an experimental setup similar to that described in Ref. 13. The IrO2/YIG sample, with a size of 2 × 6 mm2, was sandwiched between two AlN heat baths with temperatures that were stabilized at 300 K + ΔT and 300 K, respectively. (b) Comparison of light transmittance spectra of a 30-mm-thick IrO2 film and a 10-mm-thick Pt film. The films were formed on glass substrates; the contribution from the light transmittance of the substrates is subtracted.

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The LSSE measurements were performed using an experimental setup similar to that described in Ref. 13. The IrO2/YIG sample, with a size of 2 × 6 mm2, was sandwiched between two AlN heat baths with temperatures that were stabilized to 300 K + ΔT and 300 K, where the temperature of the heat bath connected to the top of the IrO2 layer is higher than that connected to the bottom of the GGG substrate. The temperature difference ΔT was generated using a Peltier thermoelectric module and detected by using thermocouples.

The temperature gradient across the IrO2/YIG interface induces a spin current in the IrO2 layer along the direction...
normal to the interface if the magnetic moments in YIG and conduction-carriers’ spins in IrO$_2$ are coupled via the spin-mixing conductance. This spin current is converted into an electric field $E_{\text{ISHHE}}$ by the ISHE in the IrO$_2$ layer along a direction determined by the following relation:

$$ E_{\text{ISHHE}} = g_t^f l \frac{\theta_{\text{ISHHE}} \rho}{d} j_s \times \frac{\mathbf{M}}{|\mathbf{M}|}, $$

(1)

where $j_s$, $\theta_{\text{ISHHE}}$, $\rho$, $\lambda$, and $d$ denote the spatial direction of the thermally generated spin current, spin-Hall angle, resistivity, spin-diffusion length, and thickness of the IrO$_2$ film, respectively. $g_t^f$ is the real part of the spin-mixing conductance at the IrO$_2$/YIG interface and $\mathbf{M}$ is the magnetization vector of the YIG film. In Eq. (1), we neglect the diffusion term of the spin current in the IrO$_2$ film because $d (= 30 \text{ nm})$ of our sample is much greater than $\lambda$ of IrO$_2$ ($= 3.8 \text{ nm}^{29}$). The LSSE-induced $E_{\text{ISHHE}}$ value in the IrO$_2$ layer can be detected as a voltage signal $V_{\text{ISHHE}} = E_{\text{ISHHE}} l$ with $E_{\text{ISHHE}}$ and $l$ being the magnitude of $E_{\text{ISHHE}}$ and the effective sample length, respectively. To measure $V_{\text{ISHHE}}$ induced by the LSSE in the IrO$_2$/YIG sample, two silver-paste electrodes were attached to the ends of the IrO$_2$ layer at an interval of $l = 4 \text{ mm}$; the electric voltage difference $V$ between the two electrodes was measured while sweeping an external magnetic field $H$ at various values of $\Delta T$. Here, silver paste is used to attach Au lead wires to the ends of the IrO$_2$ film, which creates good Ohmic contact (note that the work function of silver is close to that of IrO$_2$).

Figure 2(b) shows the $H$ dependence of $V$ for various values of $\Delta T$ for the IrO$_2$/YIG sample, which is measured when the magnetic field was applied along the $y$-direction. As $\Delta T$ increases, a clear voltage signal was found to appear in the IrO$_2$ layer in response to the magnetization reversal of the YIG layer, whereas no signal was observed at $\Delta T = 0 \text{ K}$. The voltage signal is proportional to $\Delta T$; its linear fitting line shows that the thermopower in the IrO$_2$/YIG sample is $V/\Delta T = 0.021 \mu \text{V K}^{-1}$ [see Fig. 2(c)]. These results indicate that the voltage signal can be attributed to the ISHE in the IrO$_2$ layer, which is generated by the LSSE in the YIG layer.

To further confirm the origin of the thermoelectric voltage in the IrO$_2$/YIG sample, we measured $V$ while changing the angle of the external magnetic field. Figure 3 shows the $H$ dependence of $V$ for the IrO$_2$/YIG sample at $\Delta T = 9 \text{ K}$ for various values of the out-of-plane field angle $\theta_{\text{H}}$, where $\theta_{\text{H}}$ is defined as in the upper left panel of Fig. 3. When $\theta_{\text{H}} \neq 0^\circ$, finite-valued $V$ signals appear; their magnitude and sign systematically change with $\theta_{\text{H}}$. By contrast, no $V$ signal was observed when the magnetic field is perpendicular to the film’s surface: $\theta_{\text{H}} = 0^\circ$. The $\theta_{\text{H}}$ dependence of $V$ in the IrO$_2$/YIG sample is well represented by Eq. (1), which includes with static demagnetizing fields in the YIG film (see Fig. 4), which is consistent with the symmetry of the ISHE voltage induced by the LSSE.

In Fig. 5, we show a comparison of the LSSE signals between the IrO$_2$/YIG sample and the conventional Pt/YIG sample. The Pt/YIG sample was prepared by sputtering a 10-nm-thick Pt film on a YIG/GGG wafer, where the YIG films for the IrO$_2$/YIG and Pt/YIG samples were grown at the same time. Both samples had the same size (2 $\times$ 6 mm$^2$) and the LSSE measurements were carried out under the same conditions at $\Delta T = 9 \text{ K}$. The magnitude of the LSSE signal in the IrO$_2$/YIG sample was found to be 68 times smaller than that in the Pt/YIG sample. This small LSSE signal in the IrO$_2$/YIG sample is attributed not only to the larger thickness of the IrO$_2$ layer but also to the small spin-mixing

![Fig. 2](image-url)  
(a) A schematic illustration of the IrO$_2$/YIG sample. (b) $H$ dependence of the voltage $V$ in the IrO$_2$/YIG sample for various values of $\Delta T$. (c) $\Delta T$ dependence of $V$ at $H = 450 \text{ Oe}$ in the IrO$_2$/YIG sample, measured when $VT$ was applied along the $+z$-direction.

![Fig. 3](image-url)  
$H$ dependence of $V$ in the IrO$_2$/YIG sample at $\Delta T = 9 \text{ K}$ for various values of the out-of-plane field angle $\theta_{\text{H}}$.

![Fig. 4](image-url)  
$\theta_{\text{H}}$ dependence of $V$ in the IrO$_2$/YIG sample at $H = 450 \text{ Oe}$. The solid line was obtained by fitting the experimental data using Eq. (1) combined with static demagnetizing fields in the YIG film.
The spin-mixing conductance at the IrO2/YIG interface; using Eq. (1) with $\theta_{\text{SHE}} \cdot \lambda = 0.152 \, \text{nm}^2$ ($0.188 \, \text{nm}^2$) and $\rho = 2 \times 10^{-4} \, \Omega \, \text{cm}$ ($4.5 \times 10^{-5} \, \Omega \, \text{cm}$) for the IrO2 (Pt) film and $g_{1}^{\text{ir}} = 1.3 \times 10^{18} \, \text{m}^{-2}$ at the Pt/YIG interface, the spin mixing conductance at the IrO2/YIG interface is estimated to be $g_{1}^{\text{ir}} = 1.2 \times 10^{16} \, \text{m}^{-2}$. Therefore, improvement of the spin-mixing conductance at the conductive-oxide/magnetic-insulator interface is useful for realizing efficient all-oxide SSE devices, which may be achieved, for example, by improving the crystalline structure of the interface by annealing treatment, by modulating carrier density in the conductive oxide layer, or by inserting magnetic interlayers between the conductive-oxide and magnetic-insulator layers.

In summary, we measured the longitudinal spin-Seebeck effect (LSSE) in the all-oxide IrO2/Y3Fe5O12 (YIG) bilayer film. The temperature-difference, magnetic-field, and field-angle dependences of the thermoelectric voltage in the IrO2/YIG sample are consistent with the characteristics of the inverse spin-Hall effect in the IrO2 layer induced by the LSSE in the YIG layer. The LSSE voltage in the IrO2/YIG sample was observed to be much smaller than that in a conventional Pt/YIG sample, which is attributed to the small spin-mixing conductance at the IrO2/YIG interface if the spin-Hall angle and spin-diffusion length of our IrO2 film are assumed to be comparable to those reported in previous studies. Although an all-oxide system is a promising candidate for realizing low-cost, stable, transparent LSSE thermospin devices, significant improvement of the spin-mixing conductance at conductive-oxide/magnetic-insulator interfaces is necessary.

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Fig. 5. $H$ dependence of $V$ in the IrO2/YIG (a) and Pt/YIG (b) samples at $\Delta T = 9 \, \text{K}$. The thickness of the IrO2 (Pt) layer of the IrO2/YIG (Pt/YIG) sample is 30 nm (10 nm).