

Quantitative Temperature Dependence of Longitudinal Spin Seebeck Effect at High Temperatures

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We report temperature-dependent measurements of longitudinal spin Seebeck effects (LSSEs) in Pt/Y₃Fe₅O₁₂ (YIG)/Pt systems in a high temperature range from room temperature to above the Curie temperature of YIG. The experimental results show that the magnitude of the LSSE voltage in the Pt/YIG/Pt systems rapidly decreases with increasing the temperature and disappears above the Curie temperature. The critical exponent of the LSSE voltage in the Pt/YIG/Pt systems at the Curie temperature is estimated to be 3, which is much greater than that for the magnetization curve of YIG. This difference highlights the fact that the mechanism of the LSSE cannot be explained in terms of simple static magnetic properties in YIG.

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I. INTRODUCTION

The Seebeck effect converts a temperature difference into electric voltage in conductors [1]. Since the discovery of the Seebeck effect nearly 200 years ago, it has been studied intensively to realize simple and environmentally friendly energy-conversion technologies [2]. The Seebeck effect has been measured using various materials in a wide temperature range to investigate thermoelectric conversion performance and thermoelectric transport properties. Temperature-dependent measurements in a high temperature range are especially important in the investigation of the Seebeck effect, since thermoelectric devices are often used above room temperature [3,4].

In the field of spintronics [5–7], a spin counterpart of the Seebeck effect—the spin Seebeck effect (SSE)—was recently discovered. The SSE converts a temperature difference into spin voltage in ferromagnetic or ferrimagnetic materials. When a conductor is attached to a magnet under a temperature gradient, the thermally generated spin voltage in the magnet injects a spin current [8–10] into the conductor. Since the SSE occurs not only in metals [11–14] and semiconductors [15,16] but also in insulators [13,17–32], it enables the construction of “insulator-based

thermoelectric generators” [21] in combination with the inverse spin Hall effect (ISHE) [33–37], which was impossible if only the conventional Seebeck effect was used.

The observation of the SSE in insulators has been reported mainly in a longitudinal configuration [13,18–32]. The sample system for measuring the longitudinal SSE (LSSE) is a simple paramagnetic metal (PM)–ferrimagnetic insulator (FI) junction system. In many cases, Pt and Y₃Fe₅O₁₂ (YIG) are used as PM and FI, respectively. When a temperature gradient ∇T is applied to the PM-FI system perpendicular to the interface, the spin voltage is thermally generated and injects a spin current into the PM along the ∇T direction owing to thermal spin-pumping mechanism [38–46]. This thermally induced spin current is converted into an electric field \mathbf{E}_{ISHE} by the ISHE in the PM according to the relation

$$\mathbf{E}_{\text{ISHE}} \propto \mathbf{J}_s \times \boldsymbol{\sigma}, \quad (1)$$

where \mathbf{J}_s is the spatial direction of the thermally induced spin current and $\boldsymbol{\sigma}$ is the spin-polarization vector of electrons in the PM, which is parallel to the magnetization \mathbf{M} of FI [see Fig. 1(a)]. By measuring \mathbf{E}_{ISHE} in the PM, one can detect the LSSE electrically.

In the experimental research on the SSE, temperature-dependent measurements also have been used for investigating various thermospin transport properties, such as phonon-mediated effects [15,16,39,47], correlation between the SSE and magnon excitation [29,48], and effects of metal-insulator phase transition [13]. However, all the experiments

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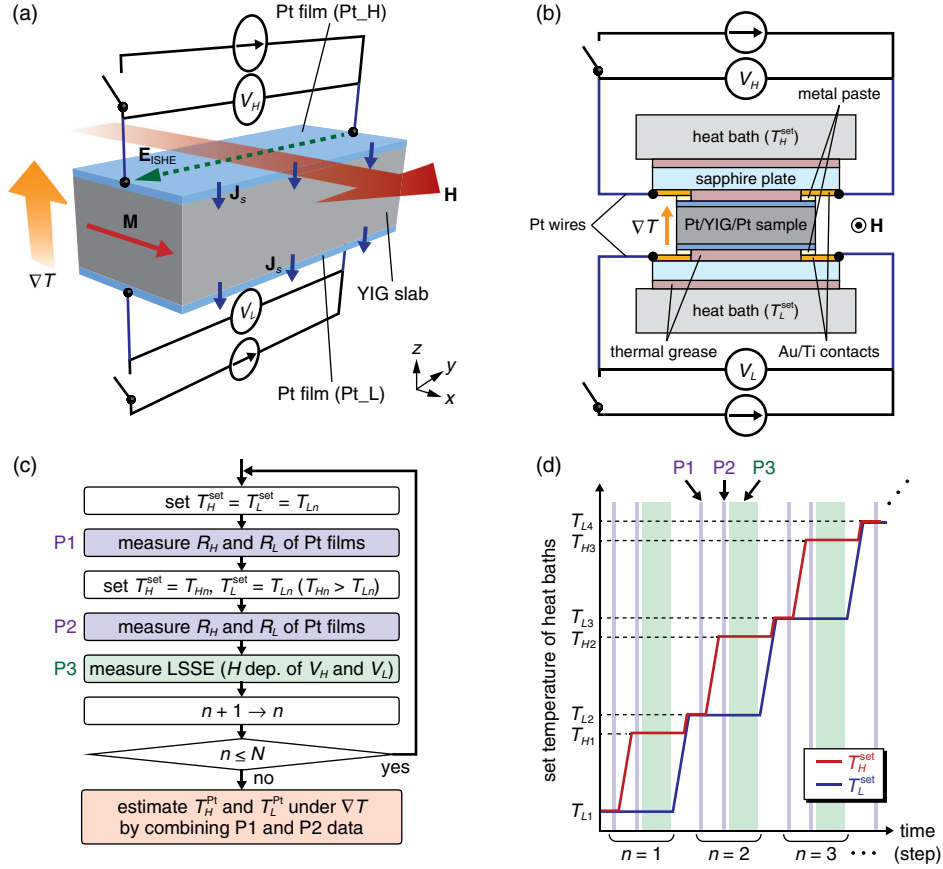


FIG. 1. (a) A schematic illustration of the Pt/YIG/Pt sample. ∇T , \mathbf{H} , \mathbf{M} , \mathbf{E}_{ISHE} , and \mathbf{J}_s denote the temperature gradient, magnetic field (with the magnitude H), magnetization vector, electric field induced by the ISHE, and spatial direction of the thermally generated spin current, respectively. The electric voltage V_H (V_L) and resistance R_H (R_L) between the ends of Pt_H (Pt_L) are measured using a nanovolt, micro-ohm meter (Agilent 34420A) in the “dc-voltage” and “2-wire-resistance” modes, respectively. The dc-voltage (2-wire-resistance) mode corresponds to the switch-off (switch-on) state in this schematic illustration. (b) Experimental configuration for measuring the LSSE used in the present study. (c) Flow chart of the measurement processes. (d) Schematic graph of the set temperatures, T_{Hn} and T_{Ln} , of the heat baths as a function of time or the measurement step number n . Process P1 is performed under the isothermal condition, while processes P2 and P3 are under the temperature gradient. Before starting processes P1 and P2, we wait for ~ 30 min at each n to stabilize the T_H^{set} and T_L^{set} values.

on the SSE to date have been performed around and below room temperature. In this article, we report quantitative temperature-dependent measurements of the LSSE in Pt/YIG systems in the high temperature range from room temperature to above the Curie temperature of YIG.

II. EXPERIMENTAL PROCEDURE

The sample system used in this study consists of a single-crystalline YIG slab covered with Pt films. One difference from conventional samples is that the Pt films are put on both the top and bottom surfaces of the YIG slab [Fig. 1(a)], while only the top surface of YIG is covered with a Pt film in conventional samples [18,23,27,32]. The lengths of the YIG slab along the x , y , and z directions are 3, 7, and 1 mm, respectively. Pt films 10 nm thick are sputtered on the whole of the $3 \times 7 \text{ mm}^2$ (111) surfaces of the YIG. The top and bottom Pt films are electrically insulated from each

other because YIG is a very good insulator. Since YIG has a large charge gap of 2.7 eV [49], thermal excitation of charge carriers in YIG is vanishingly small even at the high temperatures.

To attach electrodes to both the Pt films symmetrically and to generate a uniform temperature gradient, we make the configuration shown in Fig. 1(b). Here, the Pt/YIG/Pt sample is sandwiched between two 0.5-mm-thick sapphire plates of which the surface is covered with two separated Au/Ti contacts. The distance between the two Au/Ti contacts is ~ 6 mm. To extract voltage signals in the Pt films, both the ends of the Pt films are connected to the Au/Ti contacts via sintering metal paste, which can be used up to 900 °C, and thin Pt wires with diameter of 0.1 mm are attached to the end of the contacts [see Fig. 1(b)]. The sapphire plates are thermally connected to heat baths of which the temperatures are controlled with an accuracy of < 0.6 K by using proportional-integral-derivative

temperature controllers. We attach thermal paste to both the surfaces of the sapphire plates, except for the regions of the Au/Ti contacts, to improve the thermal contact [see Fig. 1(b)]. During the measurements of the LSSE, the temperature of the upper heat bath T_H^{set} is set to be higher than that of the lower one T_L^{set} . According to the direction of the temperature gradient, hereafter, the top and bottom Pt films of the Pt/YIG/Pt sample are referred to as “Pt_H” and “Pt_L,” respectively. In this setup, we can measure the electric voltage V_H (V_L) and resistance R_H (R_L) between the ends of Pt_H (Pt_L) without changing electrodes, wiring, and measurement equipment [see also the caption of Fig. 1]. An external magnetic field \mathbf{H} (with the magnitude H) was applied along the x direction.

To investigate the temperature dependence of the LSSE quantitatively, it is important to estimate the temperature difference between the top and bottom of the YIG sample. However, in conventional experiments, the temperatures of the heat baths, not the sample itself, are usually monitored [20]. Therefore, the measured temperature difference includes the contributions from the interfacial thermal resistance between the sample and heat baths and from small temperature gradients in the sample holders. To avoid this problem, in this study, we use the Pt films not only as spin-current detectors but also as temperature sensors [28,31]; we can know the temperatures of the Pt films from the temperature dependence of the resistance of the films, enabling the estimation of the temperature difference between the top and bottom of the sample during the LSSE measurements.

Figure 1(c) shows the flow chart of the measurement processes. The measurement comprises the three processes P1–P3, and the processes are repeated N times. Hereafter, T_{Hn} and T_{Ln} denote the set temperatures for each measurement step number n ($= 1, 2, \dots, N$) (see Table I, where the values of T_{Hn} and T_{Ln} for each n are shown). Process

TABLE I. Set temperatures of the heat baths for each measurement step number n ($\leq N$). Here, the T_{Hn} and T_{Ln} values are increased by 10 K with every n increase, while $T_{Hn} - T_{Ln}$ is fixed at 8 K. The single dots represent the abbreviation of the n , T_{Hn} , and T_{Ln} values.

Step number n	T_{Ln} (K)	T_{Hn} (K)
1	290	298
2	300	308
3	310	318
4	320	328
5	330	338
.	.	.
.	.	.
.	.	.
31	590	598
32	600	608
33	610	618
34 ($= N$)	620	628

P1 is the measurement of the resistance of Pt_H and Pt_L, R_H and R_L , in the isothermal condition, where the temperatures of the heat baths are set to the same temperature: $T_H^{\text{set}} = T_L^{\text{set}} = T_{Ln}$. In this isothermal condition, the temperature of the Pt/YIG/Pt sample is uniform and very close to the set temperature of the heat baths irrespective of the presence of the interfacial thermal resistance. Next, we apply a temperature gradient to the Pt/YIG/Pt sample by increasing the temperature of the upper heat bath, where $T_H^{\text{set}} = T_{Hn}$ and $T_L^{\text{set}} = T_{Ln}$, with $T_{Hn} > T_{Ln}$ (see Table I). After waiting until the temperatures are stabilized, we measure the resistance of the Pt films under the temperature gradient: this is process P2. Processes P1 and P2 are performed without applying \mathbf{H} . Immediately after process P2, we proceed to process P3; the LSSE, i.e., the H dependence of V_H and V_L , is measured with keeping the magnitude of $T_H^{\text{set}} - T_L^{\text{set}}$ constant. After finishing the LSSE measurements, we go on to the next step and increase n by 1, where the values of T_{Hn} and T_{Ln} are increased as shown in Table I. These measurement processes are summarized in Fig. 1(d).

The calibration method of the sample temperatures is as follows. From process P1, we obtain the temperature (T_{Ln}) dependence of R_H and R_L under the isothermal condition. By comparing the resistance under the temperature gradient, obtained from process P2, with the isothermal $R_{H,L} - T_{Ln}$ curves, we can calibrate the temperature of the Pt films, T_H^{Pt} and T_L^{Pt} , under the temperature gradient, allowing us to estimate the average temperature $T_{\text{av}} [= (T_H^{\text{Pt}} + T_L^{\text{Pt}})/2]$ and the temperature difference ΔT ($= T_H^{\text{Pt}} - T_L^{\text{Pt}}$) in the YIG slab during the LSSE measurements. The T_{av} and ΔT values are free from the contributions from the interfacial thermal resistance between the sample and heat baths and from the temperature gradients in the sapphire plates, enabling quantitative evaluation of the LSSE at various temperatures. Although the experimental protocol proposed here cannot estimate the contributions of the interfacial thermal resistance at the Pt/YIG interfaces and temperature gradients in the Pt films, they are negligible compared with the temperature difference applied to the YIG slab [26,32].

III. RESULTS AND DISCUSSION

Figures 2(a) and 2(b), respectively, show the H dependence of V_H and V_L in the Pt/YIG/Pt sample for each step number n , measured when $T_{Hn} - T_{Ln} = 8$ K. Around room temperature, we observe clear voltage signals in both the Pt films; the signs of V_H and V_L are reversed in response to the reversal of the magnetization direction of the YIG slab. Since the contribution of anomalous Nernst effects induced by proximity ferromagnetism in Pt [50] is negligibly small in Pt/YIG systems, the voltage signals observed here are purely due to the LSSE [23,27,32]. The sign of the LSSE voltage in Pt_H is observed to be the same as that in Pt_L, a situation consistent with the scenario of

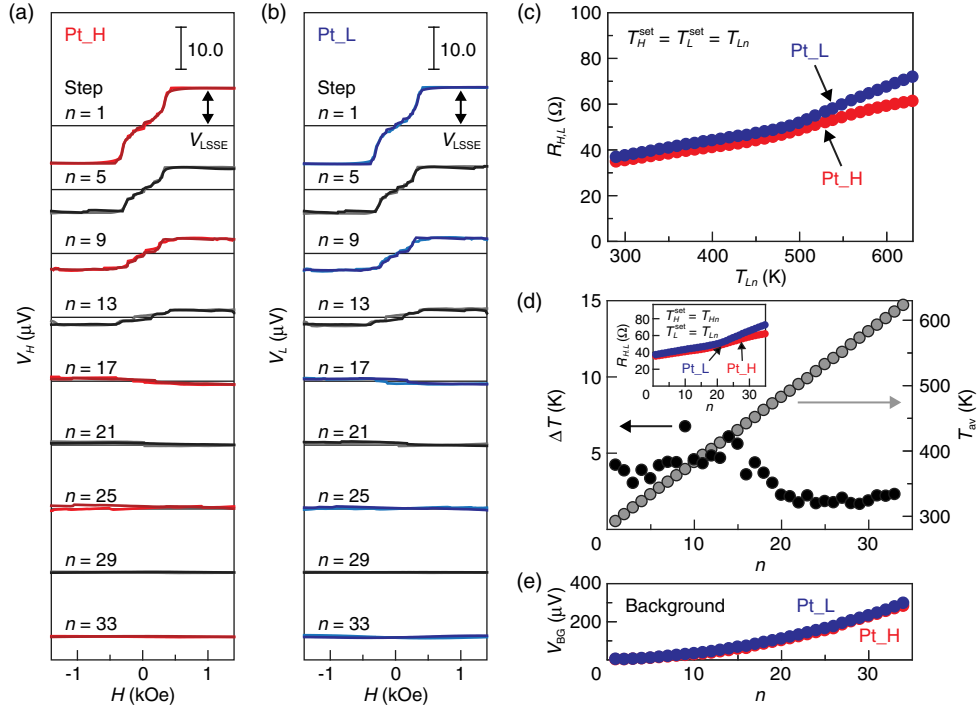


FIG. 2. (a) H dependence of V_H for Pt_H in the Pt/YIG/Pt sample for various values of n . (b) H dependence of V_L for Pt_L. The LSSE voltage V_{LSSE} for Pt_H (Pt_L) is defined as V_H (V_L) at $H = 1$ kOe. (c) T_{Ln} dependence of R_H (R_L) for Pt_H (Pt_L) in the isothermal condition: $T_H^{\text{set}} = T_L^{\text{set}} = T_{Ln}$. (d) The average temperature T_{av} and temperature difference ΔT of the Pt/YIG/Pt sample during the LSSE measurements as a function of n . The inset of (d) shows R_H (R_L) for Pt_H (Pt_L) as a function of n under the temperature gradient: $T_H^{\text{set}} = T_{Hn}$ and $T_L^{\text{set}} = T_{Ln}$ with $T_{Hn} > T_{Ln}$. (e) The background voltage V_{BG} for Pt_H and Pt_L as a function of n under the temperature gradient.

the SSE [38,40,44,51]. Here, we note that, although the large background voltage V_{BG} appears due to unavoidable thermopower differences in the wires with increasing n [Fig. 2(e)], the noise level in the V_H and V_L signals does not increase and the drift of V_{BG} is small [Figs. 2(a) and 2(b)]. Therefore, we can extract the LSSE voltage simply by measuring the H dependence of V_H and V_L . We also find that the magnitude of the LSSE voltage in the Pt/YIG/Pt sample monotonically decreases with increasing the temperature.

To quantitatively discuss the temperature dependence of the LSSE voltage in the Pt/YIG/Pt sample, we estimate T_{av} and ΔT at each step number n by using the method explained in Sec. II. Figure 2(c) shows the T_{Ln} dependence of R_H and R_L for the Pt films measured under the isothermal condition. By combining the isothermal $R_{H,L}-T_{Ln}$ curves with the R_H and R_L data under the temperature gradient [the inset of Fig. 2(d)], we obtain the T_{av} and ΔT values at each n [Fig. 2(d)]. Importantly, the calibrated values of ΔT are dependent on n and always smaller than the temperature difference applied to the heat baths due to the interfacial thermal resistance and temperature gradients in the sapphire plates (note that $T_{Hn} - T_{Ln} = 8$ K for all the measurements, as shown in Table I).

Figure 3(c) shows the LSSE voltage normalized by the calibrated temperature difference applied to the Pt/YIG/Pt sample, $V_{LSSE}/\Delta T$, as a function of T_{av} . Here, V_{LSSE} denotes V_H (V_L) for Pt_H (Pt_L) at $H = 1$ kOe. We confirm again that the magnitude of $V_{LSSE}/\Delta T$ monotonically decreases with increasing the temperature and disappears above the Curie temperature T_c of YIG, where T_c of our YIG slab is experimentally estimated to be 553 K (see Appendix A). This behavior is observed not only in one sample but also in our different samples as exemplified in Fig. 3(c). Interestingly, the temperature dependence of $V_{LSSE}/\Delta T$ is significantly different from the magnetization ($4\pi M$) curve of the YIG slab [compare Figs. 3(a) and 3(c)]; the magnitude of $V_{LSSE}/\Delta T$ rapidly decreases with a concave-up shape, while the magnetization curve of YIG exhibits a standard concave-down shape. We also check that the strong temperature dependence of the LSSE voltage in the Pt/YIG/Pt sample cannot be explained by the weak temperature dependence of the thermal conductivity of YIG (see Appendix B). A similar difference in the temperature-dependent data between the ISHE voltage and magnetization is also observed in Pt/GaMnAs systems in the measurement of the transverse SSEs [15,16].

The behavior of physical quantities near continuous phase transitions can be described by critical exponents

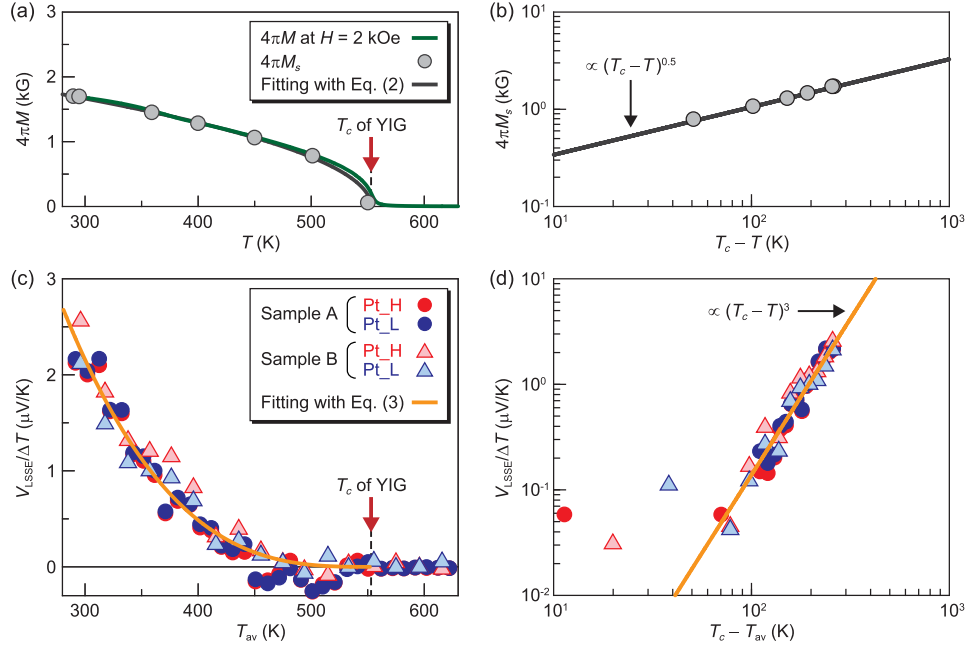


FIG. 3. (a) T dependence of the bulk magnetization $4\pi M$ of the YIG slab. The green curve shows the $4\pi M$ data at $H = 2$ kOe, measured with a vibrating sample magnetometer. The gray circles show the saturation magnetization $4\pi M_s$ of the YIG slab. The gray curve is obtained by fitting the $4\pi M_s$ data with Eq. (2). The values of $4\pi M_s$ and Curie temperature $T_c (= 553$ K) of the YIG slab are estimated by using the Arrott plot method (see Appendix A). (b) Double logarithmic plot of the $T_c - T$ dependence of $4\pi M_s$ of the YIG slab. (c) T_{av} dependence of $V_{LSSE}/\Delta T$ for two different Pt/YIG/Pt samples A (circles) and B (triangles). The experimental results shown in Fig. 2 are measured using the Pt/YIG/Pt sample A. The orange curve is obtained by fitting the $V_{LSSE}/\Delta T$ data with Eq. (3). (d) Double logarithmic plot of the $T_c - T_{av}$ dependence of $V_{LSSE}/\Delta T$ for the Pt/YIG/Pt samples A and B.

in general. Here, we compare the critical exponents for the observed temperature dependences of the LSSE voltage in the Pt/YIG/Pt sample and the magnetization of YIG. First, we check that the magnetization curve of YIG is well reproduced by a standard mean-field model [52]:

$$4\pi M_s = A(T_c - T)^{0.5}, \quad (2)$$

where the critical exponent is fixed at 0.5 and A is an adjustable parameter [see Figs. 3(a) and 3(b)]. The critical exponent γ for the LSSE is estimated by fitting the experimental data in Fig. 3(c) with the following equation:

$$\frac{V_{LSSE}}{\Delta T} = S(T_c - T)^\gamma, \quad (3)$$

where both S and γ are adjustable parameters and T_{av} is regarded as T for the LSSE data. We find that the observed temperature dependence of $V_{LSSE}/\Delta T$ for the Pt/YIG/Pt sample is well fitted by Eq. (3) with $\gamma = 3$, which is much greater than the critical exponent for the magnetization curve [see also the double logarithmic plot in Fig. 3(d)]. This big difference in the critical exponents between the LSSE and magnetization emphasizes the fact

that the LSSE is not attributed solely to static magnetic properties in YIG.

Here, we qualitatively discuss the origin of the temperature dependence of the LSSE voltage in the Pt/YIG/Pt sample. According to the thermal spin-pumping mechanism [38,44] and phenomenological calculation of the ISHE combined with short-circuit effects [53], the magnitude of the LSSE voltage is determined mainly by the following factors: the spin-mixing conductance [54–57] at the Pt/YIG interfaces, spin-diffusion length and spin-Hall angle of Pt, and the difference between an effective magnon temperature in YIG and an effective electron temperature in Pt. Since the LSSE voltage is proportional to the spin-mixing conductance [38], it can contribute directly to the observed temperature dependence of the LSSE voltage. Recently, Ohnuma *et al.* formulated the relation between the spin-mixing conductance and interface s - d interaction at paramagnet-ferromagnet interfaces and predicted that the spin-mixing conductance is proportional to $(4\pi M_s)^2$ of the ferromagnet [58]. By combining this prediction with Eq. (2), the spin-mixing conductance is proportional to $T_c - T$, of which the critical exponent ($= 1$) is greater than the exponent for the magnetization curve. Although the temperature dependence of the spin-mixing conductance can explain the facts that the LSSE voltage monotonically decreases with increasing the temperature and disappears at T_c , it is still much weaker than the observed $(T_c - T)^3$

dependence of the LSSE voltage. Furthermore, if the spin-diffusion length of Pt decreases with increasing the temperature [59], it can also contribute to reducing the LSSE voltage at high temperatures, while the spin-Hall angle of Pt was shown to exhibit weak temperature dependence [60] (note that the magnitude of the ISHE voltage monotonically decreases with decreasing the spin-diffusion length when the spin-Hall angle is constant [53]). The effective magnon-electron temperature difference could also be an important factor, but there is no clear framework to determine its temperature dependence at the present stage. Therefore, more elaborate investigations are necessary for complete understanding of the temperature dependence of the LSSE voltage.

IV. CONCLUSION

In this study, we report the longitudinal spin Seebeck effects in $\text{Y}_3\text{Fe}_5\text{O}_{12}$ slabs sandwiched by two Pt films in the high temperature range from room temperature to above the Curie temperature T_c of YIG. To investigate the temperature dependence of the LSSE quantitatively, we use the Pt films not only as spin-current detectors but also as temperature sensors. The measurement processes we use enable accurate estimation of the average temperature and temperature difference of the sample, being free from thermal artifacts. We found that the magnitude of the LSSE in the Pt/YIG/Pt sample rapidly decreases with increasing the temperature and disappears above T_c of YIG; the observed LSSE voltage exhibits the $(T_c - T)^3$ dependence of which the critical exponent ($= 3$) is much greater than that of the magnetization of YIG ($= 0.5$). Although more detailed experimental and theoretical investigations are required to clarify the microscopic origin of this discrepancy, we anticipate that the quantitative temperature-dependent LSSE data at high temperatures will be helpful for obtaining full understanding of the mechanism of the LSSE.

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APPENDIX A: ESTIMATION OF CURIE TEMPERATURE OF YIG

The Curie temperature T_c of the YIG slab used in the present study is estimated from vibrating sample magnetometry and Arrott-plot analysis [52,61]. The inset of Fig. 4(a) shows the H dependence of the magnetization $4\pi M$ of the YIG slab for various values of T . From this result, we obtain the Arrott plots, i.e., $H/4\pi M$ dependence of $(4\pi M)^2$, of the YIG slab [see Fig. 4(a)]. The saturation magnetization $4\pi M_s$ of the YIG at each temperature is extracted by extrapolating the $(4\pi M)^2$ data in the high-magnetic-field range to zero field [see red dotted lines in Fig. 4(a)]. As shown in Fig. 4(b), the T dependence of $(4\pi M_s)^2$ of the YIG slab is well fitted by a linear function; the horizontal intercept of the linear fit line corresponds to T_c . The fitting result shows that the Curie temperature of our YIG slab is $T_c = 553$ K, which is consistent with literature values [62,63].

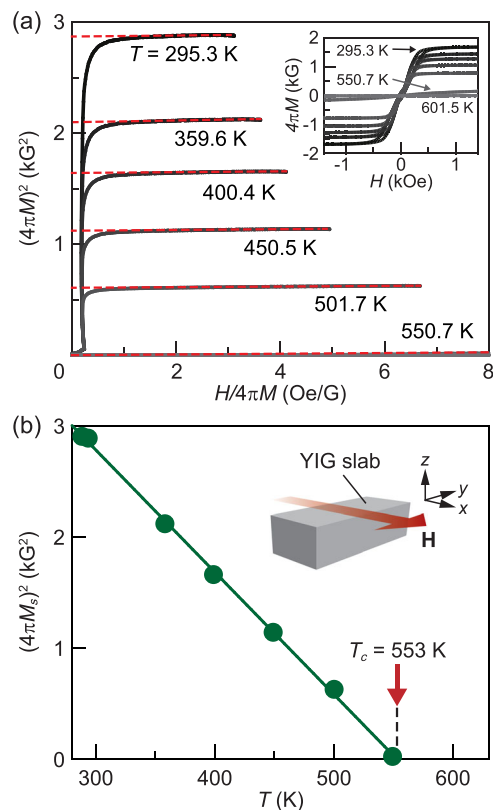


FIG. 4. (a) Arrott plots of the YIG slab for various values of T . The inset of (a) shows the H dependence of $4\pi M$ of the YIG slab for various values of T , measured with a vibrating sample magnetometer. The lengths along the x , y , and z directions of the YIG slab used for the magnetometry measurements are 3, 7, and 1 mm, respectively. \mathbf{H} is applied along the x direction. (b) T dependence of $(4\pi M_s)^2$ of the YIG slab.

APPENDIX B: TEMPERATURE DEPENDENCE OF THERMAL CONDUCTIVITY OF YIG

Figure 5(a) shows the thermal conductivity κ of the YIG slab used in the present study as a function of T . The κ values are obtained by the combination of thermal diffusivity measured by a laser-flash method and specific heat C measured by a differential scanning calorimetry. Here, we measure the thermal diffusivity along the [111] direction of the single-crystalline YIG slab, which is parallel to the ∇T direction in the LSSE setup. As shown in Fig. 5(b), the measured C values are consistent with the Dulong-Petit (DP) law [1]; the difference of the C values from the DP specific heat of YIG, $C_{DP} = 0.676$ J/gK, is less than 10% of C_{DP} for $T > 350$ K. The observed T dependence of κ is well fitted by $\kappa \propto T^{-1}$, indicating that the thermal conductivity of the YIG is dominated by phonons in this temperature range [see also the inset of Fig. 5(a)]. The κ value at 300 K is consistent with literature values [64–66]. We also confirm that the T dependence of κ is much weaker than that of the LSSE voltage in the Pt/YIG/Pt sample [compare Figs. 3(c) and 5(a)] and shows no anomaly around T_c .

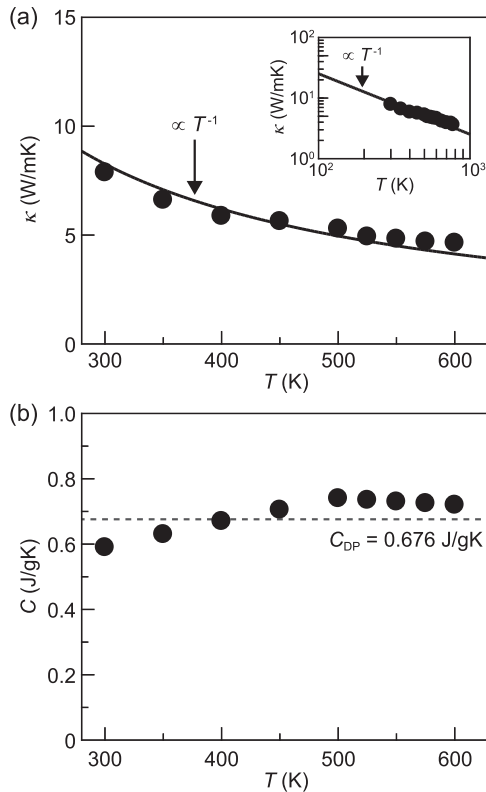


FIG. 5. (a) T dependence of the thermal conductivity κ of the YIG slab. The black circles show the measured thermal conductivity and the black curve shows the fitting of the experimental data with $\kappa = KT^{-1}$, where K is an adjustable parameter. The inset shows the double logarithmic plot of the T dependence of κ . (b) T dependence of the specific heat C of the YIG slab. The gray dotted line shows the DP specific heat of YIG.

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