Absence of the Thermal Hall Effect in Anomalous Nernst and Spin Seebeck Effects

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The anomalous Nernst effect (ANE) and the spin Seebeck effect (SSE) in spin caloritronics are two of the most important mechanisms to manipulate the spin-polarized current and pure spin current by thermal excitation. While the ANE in ferromagnetic metals and the SSE in magnetic insulators have been extensively studied, a recent theoretical work suggests that the signals from the thermal Hall effect (THE) have field dependences indistinguishable from, and may even overwhelm, those of the ANE and SSE. Therefore, it is vital to investigate the contribution of the THE in the ANE and SSE. In this work, we systematically study the THE in a ferromagnetic metal, Permalloy (Py), and magnetic insulator, an yttrium iron garnet (YIG), by using different Seebeck coefficients between electrodes and contact wires. Our results demonstrate that the contribution of the THE by the thermal couple effect in the Py and YIG is negligibly small if one includes the thickness dependence of the Seebeck coefficient. Thus, the spin-polarized current in the ANE and the pure spin current in the SSE remain indispensable for exploring spin caloritronics phenomena.

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The anomalous Nernst effect (ANE) and spin Seebeck effect (SSE), which give rise to spin separation of charge carriers in ferromagnetic metals (FMs) and pure spin current (J_S) in magnetic insulators (MIs), respectively, have attracted a great deal of attention [1,2]. The ANE was first realized soon after the anomalous Hall effect, whereas the SSE was a more recent advent, which was first reported in the transverse configuration with an in-plane temperature gradient ($\nabla_x T$) in a ferromagnetic thin film on a substrate in Fig. 1(a) [3] and subsequently in the longitudinal configuration with an out-of-plane temperature gradient ($\nabla_z T$) in Fig. 1(b) [4–6]. In the transverse spin Seebeck effect (TSSE), J_S driven by $\nabla_x T$ is generated from ferromagnetic materials [4–8] and may be detected in an adjacent normal metal by the inverse spin Hall effect (ISHE). The transverse ISHE electric field can be described by

$$E_{\mathrm{ISHE}} \propto J_S \times .\boldsymbol{\sigma}, \tag{1}$$

where $\boldsymbol{\sigma}$ is the spin direction. However, it has been shown that in the TSSE configuration for a FM on a substrate with an intended $\nabla_x T$, an unintentional $\nabla_z T$ also exists, and that unavoidably generates the ANE [9–14], which produces a transverse electric field of

$$E_{\mathrm{ANE}} \propto \nabla_z T \times \boldsymbol{m}, \tag{2}$$

where $\boldsymbol{m}$ is the magnetization vector. Therefore, in the case of FMs, the ISHE voltage, sharing the same field dependence in the TSSE, is indistinguishable from the ANE voltage. In the case of MIs, the ISHE voltage from $\nabla_x T$ in the TSSE is even more difficult to separate from that generated by an unintentionally $\nabla_z T$. In fact, Meier *et al.* recently have shown if a $\nabla_x T$ was indeed applied, there were no significant signals of the TSSE in MIs [13]. To date, the TSSE has not been experimentally established. The longitudinal spin Seebeck effect (LSSE) configuration with an unequivocal $\nabla_z T$ is the most important, and indeed the only established, geometry for SSE in ferromagnetic [4], paramagnetic [15], and antiferromagnetic materials [16,17]. In the normal metal-ferromagnetic conductor bilayer, the LSSE and ANE could coexist.

Recently, research suggests that the thermal Hall effect (THE), including the magnon Hall effect or the anomalous Righi-Leduc effect, may have been overlooked when one explores spin current phenomena in the SSE and ANE [18]. The anisotropic transverse temperature difference (ΔT_y) induced by the THE in ferromagnetic materials has the same angular dependence as those of the ANE and LSSE with a $\nabla_z T$ and those of the TSSE and planar Nernst effect

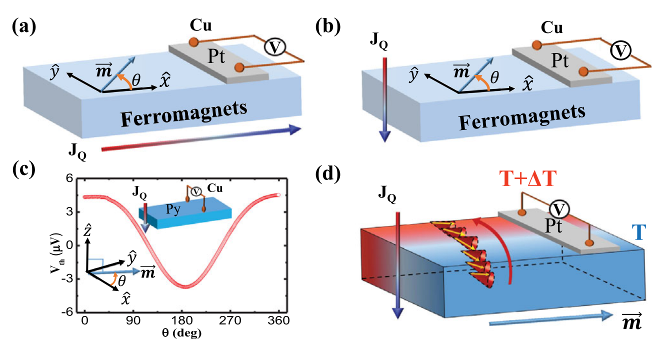

FIG. 1. (a) TSSE configuration with a $\nabla_x T$. (b) LSSE configuration with a $\nabla_z T$. (c) The angular dependence of the thermal voltage for Py (50 nm) with the setup of ANE measurement. (d) Schematic diagram of the THE measured by the LSSE configuration. The wires and the electrode serve as a thermocouple.

with a $\nabla_x T$. Surprisingly, this anisotropic field-dependent ΔT_y can be converted into a field-dependent thermal voltage in both the TSSE and LSSE configurations by the thermocouple effect. In fact, recent experiment results in Pt/YIG and Pt/Py with a $\nabla_x T$ indicate that the interpretation of the anisotropic field-dependent thermal voltage can be accounted solely by the THE *without* requiring spin current phenomena such as the ISHE, SSE, and ANE [19]. Furthermore, Tanabe *et al.* report that an appreciable ΔT_y induced by the magnon Hall effect has been observed at the sample edge in bulk yttrium iron garnet (YIG) [20]. These proposed effects, if true, can severely complicate the studies of spin-dependent thermal effects. Hence, it is vitally important to address the contribution, if any, of the THE in the SSE and ANE. In this work, we systematically investigate the contribution of the thermocouple effect in FM(Py) and MI(YIG) with a $\nabla_z T$. We found the contribution of the THE in the ANE of Py and in the SSE of YIG is *negligibly* small.

The samples are ultrasonically cleaned in acetone, isopropyl alcohol, and DI water before deposition. We use magnetron sputtering to fabricate thin films, whose magnetic properties and transport properties have been measured by a vibrating sample magnetometer and four-probe measurements, respectively [21]. The vertical temperature differences were measured by thermocouples, and the spin-dependent thermal voltages were measured by the nanovoltmeter. The uniform $\nabla_z T$ is generated by a heater and a copper block heat sink at near room temperature, of about 28 K/mm.

In Fig. 1(c), we show the voltage due to the ANE for a 50 nm Py, with an angular dependence of $\cos\theta$ as described in Eq. (2). However, in addition to electrons, quasiparticles including phonons and magnons can also carry heat current, which can cause the THE. The magnon Hall effect in a ferromagnetic insulator ($Lu_2V_3O_7$) due to the Dzyaloshinsky-Moriya (DM) interaction [22] and the phonon Hall effect in a paramagnetic insulator ($Tb_3Ga_5O_{12}$) due to the spin-phonon interaction [23] have both been reported. Therefore, when a heat current is applied perpendicular to a magnetic field, a ΔT_y might be induced by the deviated phonon and magnon flows. Recently, Wegrowe, Drouhin, and Lacour proposed that the ΔT_y induced by the THE is anisotropic in both insulating and conducting ferromagnets, based on the anisotropic properties of the heat transport [18]. Most importantly, they suggest that the ΔT_y can be converted into a spin-dependent thermal voltage in the TSSE and LSSE by the thermocouple effect due to the different Seebeck coefficient (ΔS) between the contacting wires and normal metals, which originally served as spin current detectors in the SSE, as shown in Figs. 1(a) and 1(b). For instance, under a $\nabla_x T$ or a $\nabla_z T$, the angular dependence of the transverse voltage (ΔV_y) with the in-plane magnetic field by the THE can be, respectively, described as

$$\Delta V_y \propto \Delta S J_{\mathrm{Qx}} \cos\theta \quad \text{and} \quad \Delta V_y \propto \Delta S J_{\mathrm{Qz}} \cos\theta, \qquad (3)$$

where J_{Qx} and J_{Qz} are the in-plane and perpendicular heat currents, respectively. Therefore, the experimental results in Ref. [19] suggest that the field-dependent thermal voltages with angular dependence in Eq. (3) for Py and YIG are due to the THE only, when a contact normal metal Pt is used to detect the thermal signal in Fig. 1(d). Using a similar argument, the $\cos\theta$ of Py in Fig. 1(c), measured by another material, such as Cu wires, with a different Seebeck coefficient could no longer be considered as an unambiguous proof of the ANE either. Since the validity of the SSE and ANE is at stake, it is therefore essential to investigate this proposed scenario.

To test the induced thermal voltage by the thermocouple effect when a device is composed of a thin metal layer connected with two electrode wires, we investigate a Pt-glass single-layer sample. Two Cu wires are connected to the sample to measure the developed thermal electromotive force (emf) with respect to an intentional ΔT_y [Fig. 2(a)]. In Fig. 2(b), we indeed observe a significant thermal voltage in response to the ΔT_y when one end of the electrodes is heated. However, when Pt instead of Cu wires is used, the thermal emf becomes even larger and opposite in sign [in Fig. 2(c)]. These results highlight the fact that the Seebeck coefficients for the thin film and that for the bulk are *different*. According to the Mott formula $S = (\pi^2 K_B^2 T/3e\sigma)(\partial\sigma/\partial E)E_F$, the Seebeck coefficient depends on the derivative of the electric conductivity σ at the Fermi level (E_F) [24,25]. In thin films, it is well known that when the thickness is comparable to or less than the carrier mean free path, σ significantly reduces due to surface scattering. There is a corresponding effect in the S, which, according to the effective mean-free path theory, can be described as $S(t) = S_g(1\text{-}b/t)$, where S_g is the Seebeck coefficient of the infinitely thick film, b is the coefficient related to the surface

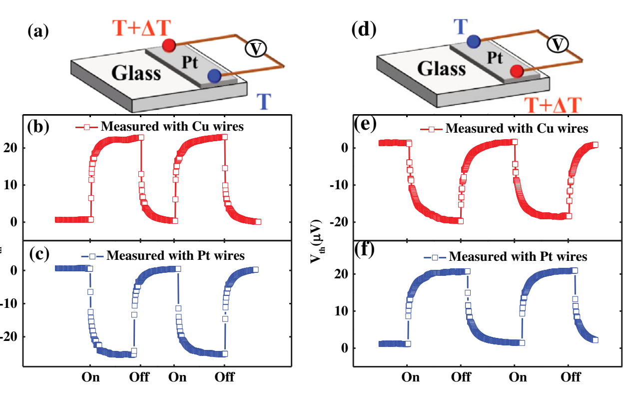

FIG. 2. (a) Schematic diagram of the thermocouple effect composed of a Pt (50 nm) thin film and two wires with a heat source on a top electrode. The thermal emf measured by two Cu wires in (b) and Pt wires in (c). (d) Schematic diagram of the thermocouple effect with a heat source on a bottom electrode. The thermal emf measured by two Cu wires in (e) and Pt wires in (f).

and grain-boundary scattering, and t is the thickness [26,27]. Here, the S of the 50 nm Pt of about $-1.4\ \mu\mathrm{V/K}$ is smaller than $S_{\mathrm{Pt}} = -5.3\ \mu\mathrm{V/K}$ for the Pt wire, which are measured by using two stable distinct temperatures of 0° and 100 °C. Together with Cu wire of $S_{\mathrm{Cu}} = 1.83\ \mu\mathrm{V/K}$, the ΔS between the Pt thin film and Cu wires is about $\Delta S_{\text{Cu-Pt(50 nm)}} = 3.23\ \mu\mathrm{V/K}$, whereas the difference between the Pt thin film and Pt wires is completely different at $\Delta S_{\text{Pt-Pt(50 nm)}} = -3.9\ \mu\mathrm{V/K}$. If we heat the other contact, the sign of thermal emf between Figs. 2(b) and 2(c) and Figs. 2(e) and 2(f) are reversed due to the inversion of the in-plane temperature gradient. Therefore, a device simply composed of a thin metal layer with two electrode wires can serve as a thermocouple, even if all materials are the *same*. This pitfall due to thin film could lead to completely erroneous results in temperature measurements.

Because the ANE and THE have the same angular field dependence, their contributions cannot be separated from the spin-dependent thermal signals. However, the ANE signal associated with spin-polarized current is proportional to the magnetization, whereas the THE signal that resulted from the thermocouple effect is proportional to the ΔS. If we use different contacting leads to measure the signal, the signal from the THE can be dramatically influenced, but the voltage from the spin current will remain the same.

We first investigate the contribution of the THE and ANE in a 50 nm Py by using two different wires, Cu and Pt. The signals are simultaneously measured by the two wires from the same Py sample under the same experimental conditions, including the same $\nabla_z T$ and the same distance between the two contacting leads [in Fig. 3(a)]. The thermal voltage of Py is clearly dominated by the ANE. A subtraction between these two signals is drawn as a black line, where the signal of the ANE is eliminated, because the spin signal caused by the ANE is independent of the materials of the contacting wires. However, the THE signal still has a contribution, because it originates from the junction consisting of two dissimilar materials with different S. By using Pt and Cu wires with opposite sign in S, the sensitivity to detect the THE signal can be further enhanced. The generation of a thermal emf by the Seebeck effect is linear with the ΔT_y between the two junctions. The ΔT_y caused by the THE can be evaluated by $|\Delta T_y| = |\Delta V/\Delta S|$, where ΔV is the difference of the saturation voltage between the signals measured by Cu and Pt wires and ΔS is between Cu and Pt, whose S is 1.83 and $-5.3\ \mu\mathrm{V/K}$, respectively. In this experiment, the corresponding ΔT_y is calculated to be around 70.1 mK if the two different voltages are from the temperature difference.

However, according to Ohm's law, the electric field induced by the spin-orbit coupling in ANE is also proportional to electrical resistance. Since it is unlikely that we can have two identical contacts, the variation of contact resistance should be taken into account. The contact

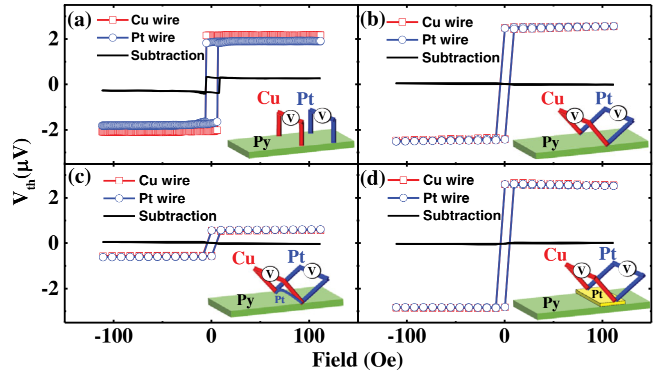

FIG. 3. A series of geometries for the field-dependent thermal voltage measurement with a uniform vertical temperature gradient in the Py (50 nm) sample, which are (a) parallel connecting Cu and Pt wires in different positions, (b) connecting two wires in the same position, (c) connecting two wires in the same position with an additional short Pt wire, and (d) connecting two wires in the same position of the Pt(5 nm)/Py(50 nm) bilayer sample.

resistances for the Cu and Pt wires are 7.6 and 6.8 Ω, respectively. We normalize the thermal voltage by dividing the contact resistance and obtain a similar ratio of 0.56 for both $\Delta V_{\mathrm{Cu}}/R_{\mathrm{Cu}}$ and $\Delta V_{\mathrm{Pt}}/R_{\mathrm{Pt}}$. After eliminating the contribution of the shunting effect, the ΔT_y by the THE is largely reduced to 14.0 mK. This indicates that the subtracted signal in Fig. 3(a) has a large part caused by the different contact resistance instead of the THE. To avoid the parasitic shunting effect, we connect two wires in one contact to make the contact resistance identical in Fig. 3(b). Interestingly, the signal difference between Cu and Pt contact wires becomes essentially nonmeasurable. With the same contact, ΔV measured by Cu and Pt is 35 nV, and the corresponding ΔT_y is estimated about 4.9 mK, which has a similar value when we take the shunting effect into account for the results in Fig. 3(a). The contribution of the THE to the ANE is only around 1.8% for Py measured by typical Cu wires.

Since the contribution of the THE in Py is very small, next we design one measurement to reduce the magnitude of the ANE but to enhance the ratio between the THE and ANE. An additional Pt wire was added between the two contacts to compose an extra Cu-Pt thermocouple to detect the ΔT_y in Fig. 3(c) and decrease the ANE signal by the shunting effect. With the resistance ratio of the Py film to the short Pt wire, $R_{\mathrm{Py}}/R_{\mathrm{pt}} = 4.8$, the ANE in Fig. 3(c) is 5 times smaller than the signal in Fig. 3(b). However, we obtain a similarly large THE signal and $\Delta T_y \cong 5.8$ mK with $\Delta V = 41$ nV. We also investigate the device with a Pt thin film on the Py layer in Fig. 3(d), where the configuration is widely adopted to study the TSSE and LSSE. By replacing the shorting Pt wire with a strip-patterned Pt (5 nm), the thermal contact can also be improved. Moreover, since the S is thickness dependent, the thermocouple effect is much larger for the thinner Pt. The S of the

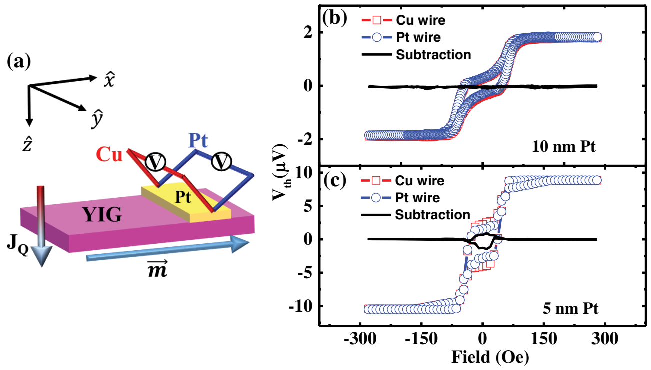

FIG. 4. (a) Schematic diagram of the field-dependent thermal voltage measured with a uniform vertical temperature gradient in the Pt/YIG sample. Field dependence of the thermal voltage on (b) Pt (10 nm) /YIG and (c) Pt(5 nm)/YIG measured by connecting Cu and Pt wires in the same position.

5-nm Pt is measured at about 1.34 μV/K. Here, we found that even for the Pt/Py sample the ΔT_y caused by the THE is also about 5.6 mK with $\Delta V = 40$ nV. The results in our four special measurement configurations clearly reveal that the thermal voltage induced in Py is dominated by the ANE. We have thus conclusively demonstrated that the field-dependent thermal signal in Py with a uniform $\nabla_z T$ is due to spin-orbit coupling, that deflects the spin-polarized current by the ANE.

Next, we address the THE in magnetic insulators, where only the magnons and phonons contribute to heat conduction. The THE contribution in the TSSE configuration was proposed by Wegrowe, Drouhin, and Lacour [18], but that was excluded from the ISHE by Meier *et al.* [13]. Since the thermal voltage induced by the THE also exists in insulators, it will be very important to investigate the contribution of the THE in the magnetic insulator YIG. In addition, the Pt/YIG bilayer structure is widely used to study the thermal spin current injection with the ISHE by the LSSE. In order to detect the electromotive force by the LSSE and THE, a 10 nm Pt is deposited on a polycrystalline YIG slab. Similar to the above measurement setups, Cu and Pt wires are used as the contacting leads to measure the spin-dependent thermal voltage with two identical contacts in Fig. 4(a). Although YIG is an insulator, the thermocouple composed of the Pt-film electrode and contacting wires can also convert the ΔT_y from the THE into the thermal voltage on YIG. However, the signal in Fig. 4(b) measured by Pt wires is almost the same as that measured by Cu wires, indicating that the ISHE signal by the LSSE dominates in the Pt/YIG. The maximum ΔT_y is estimated at 13.3 mK. To test the thickness dependence of the THE, we reduce 10 nm Pt to 5 nm in Fig. 4(c). It is known that the ISHE of the LSSE is a function of thickness associated with the spin diffusion length of normal metal, although the proponents of the THE [19] suggest that the thickness-dependent thermal voltage can also be solely explained by the decrease of the THE due to the thermal shunting effect. However, in Fig. 4(c), the signal induced by the thermocouple effect for the thinner Pt remains negligibly small. The ISHE voltage of 5 nm Pt is 5 times larger than that of 10 nm Pt due to the short spin diffusion length in Pt. The ΔT_y induced by the THE is 15.0 mK for 5 nm Pt with $\Delta V = 0.09$ μV. The similar ΔT_y but the dramatically

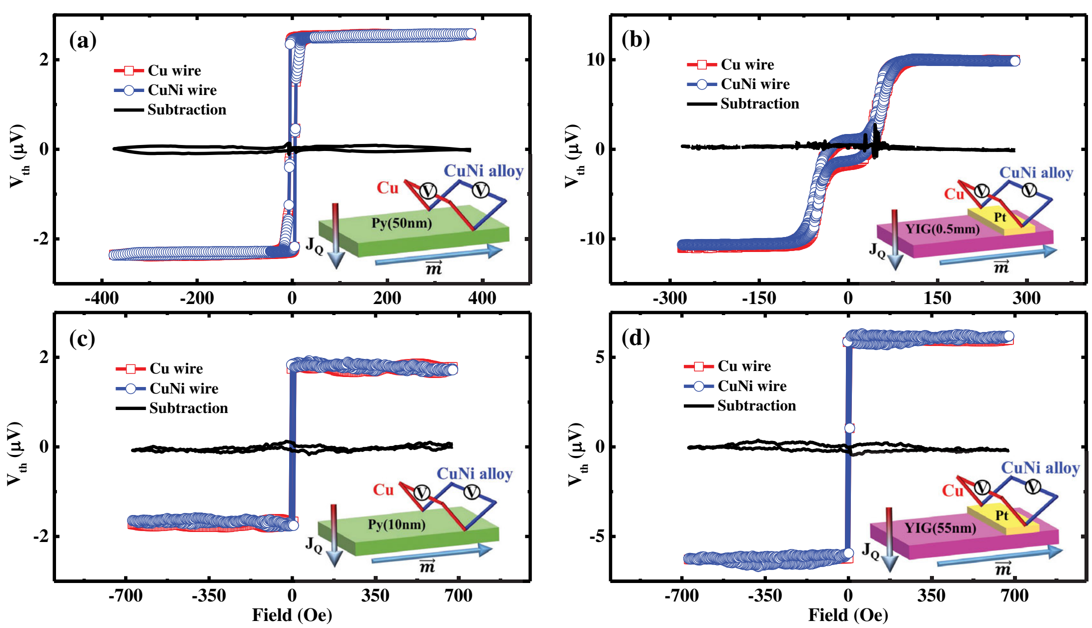

FIG. 5. Schematic diagram of using a T-type thermocouple (i.e., Cu wire and $Cu_{55}Ni_{45}$ wire) to measure the field-dependent thermal voltage with a uniform vertical temperature gradient for (a) Py (50 nm), (b) Pt(5 nm)/YIG (0.5 mm), (c) Py (10 nm), and (d) Pt(5 nm)/YIG(55 nm).

different ISHE voltage signals between different Pt thicknesses reveal that the thermal shunting effect is not dominating.

According to Eq. (3), the THE is uniquely proportional to ΔS. In Fig. 5, instead of typical electric wires, we use a $Cu_{55}Ni_{45}$ (constantan) wire of large negative Seebeck coefficient $S_{CuNi} = -39\ \mu V$ and a Cu wire of $S_{Cu} = 1.83\ \mu V$ to form a type-T thermocouple with a very high temperature sensitivity of $40.83\ \mu V/K$. Although we use atypical electrical wires to connect Py and YIG, the magnitude of the thermal voltage is consistent with the results in Figs. 3 and 4, measured by typical Cu wires. While ΔS increases more than 5 times with a type-T thermocouple, the thermal emf induced by the THE remains as a small value. Here, the ΔT_y in Py is derived about 0.5 mK from the $\Delta V = 20$ nV between the $Cu_{55}Ni_{45}$ and Cu wires in Fig. 5(a). The smaller ΔT_y than the results shown in Fig. 3 is due to a higher signal to noise ratio by the type-T thermocouple. In the case of YIG, ΔT_y is about 11.5 mK with $\Delta V = 470$ nV in Fig. 5(b). It is also worth mentioning that we observed similar results by reducing the thickness of Py and on the epitaxial YIG thin film in Fig. 5. Future work investigating the THE in materials with a strong DM interaction and inversion symmetry breaking would be of great interest.

In summary, we demonstrate that the thermocouple effect is inevitable in the ANE and SSE configuration. We design several special measurement geometries and use different Seebeck coefficients between the electrodes and wires to investigate the contribution of the THE in ferromagnetic metal Py and magnetic insulator YIG. In our measurement, the upper limit of a transverse thermal emf due to the thermocouple effect is negligibly small, which is less than 0.19% of ANE in Py and 0.03% of SSE in YIG. Future work investigating the THE by LSSE methods in materials with a strong DM interaction would be of great interest. However, if the thickness dependence of the Seebeck coefficients is overlooked, one can easily draw a different and erroneous conclusion. The spin-polarized current in the ANE and the pure spin current in the SSE remain as indispensable elements to explore these two spin caloritronics phenomena.

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