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# Magnetic-field-induced decrease of spin Peltier effect in Pt/Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> system at room temperature

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## Abstract

We report the observation of magnetic-field-induced decrease of the spin Peltier effect (SPE) in a junction of a paramagnetic metal Pt and a ferrimagnetic insulator Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> (YIG) at room temperature. For driving the SPE, spin currents are generated via the spin Hall effect from applied charge currents in the Pt layer, and injected into the adjacent thick YIG film. The resultant temperature modulation is detected by a commonly-used thermocouple attached to the Pt/YIG junction. The output of the thermocouple shows sign reversal when the magnetization is reversed and linearly increases with the applied current, demonstrating the detection of the SPE signal. We found that the SPE signal decreases with the magnetic field. The observed decreasing rate was found to be comparable to that of the spin Seebeck effect (SSE), suggesting the dominant and similar contribution of the low-energy magnons in the SPE as in the SSE.

## I. INTRODUCTION

Thermoelectric conversion is one of the promising technologies for smart energy utilization<sup>1</sup>. Owing to the progress of spintronics in this decade, the spin-based thermoelectric conversion is now added to the scope of the thermoelectric technology<sup>2-7</sup>. In particular, the thermoelectric generation mediated by flow of spins, or spin current, has attracted much attention because of the advantageous scalability, simple fabrication processes, and flexible design of the figure of merit<sup>4,8-12</sup>. This is realized by combining the spin Seebeck effect (SSE)<sup>13</sup> and spin-to-charge conversion effects<sup>14-16</sup>, where a spin current is generated by an applied thermal gradient and is converted into electricity owing to spin-orbit coupling.

The SSE has a reciprocal effect called the spin Peltier effect (SPE), discovered by Flipse *et al.* in 2014 in a Pt/yttrium iron garnet ( $\text{Y}_3\text{Fe}_5\text{O}_{12}$ : YIG) junction<sup>5,17</sup>. In the SPE, a spin current across a normal conductor (N)/ferromagnet (F) junction induces a heat current, which can change the temperature distribution around the junction system.

To reveal the mechanism of the SPE, systematic experiments have been conducted<sup>17-19</sup>. Since the SPE is driven by magnetic fluctuations (magnons) in the F layer, detailed study on the magnetic-field and temperature dependence is indispensable for clarifying the microscopic relation between the SPE and magnon excitation and the reciprocity between the SPE and SSE<sup>20-30</sup>. A high magnetic field is expected to affect the magnitude of the SPE signal via the modulation of spectral properties of magnons. In fact, the SSE thermopower in a Pt/YIG system was shown to be suppressed by high magnetic fields not only at low temperatures but also at room temperature; the decreasing rate is much greater than the conventional theoretical expectation based on the equal contribution over the magnon spectrum. It highlights the dominant contribution of sub-thermal magnons, which possess lower energy and longer propagation length than thermal magnons<sup>22-24,29,31-34</sup>. Thus, the experimental examination of the field dependence of the SPE is an important task for understanding the SPE. Although the SPE has recently been measured in various systems by using the lock-in thermography (LIT)<sup>17</sup>, it is difficult to be used at high fields and/or low temperatures. For investigating the high-magnetic-field response of the SPE, an alternative method is required.

In this paper, we investigate the magnetic field dependence of the SPE up to 9 T at 300 K by using a commonly-used thermocouple (TC) wire. As revealed by the LIT experiments<sup>17,19</sup>,

the temperature modulation induced by the SPE is localized in the vicinity of N/F interfaces. This is the reason why the magnitude of the SPE signals is very small in the first experiment by Flipse *et al.*<sup>5</sup>, where a thermopile sensor is put on the bare YIG surface, not on the Pt/YIG junction. Here, we show that the SPE can be detected with better sensitivity simply by attaching a common TC wire on a N/F junction. This simple SPE detection method enables systematic measurements of the magnetic field dependence of the SPE, since it is easily integrated to conventional measurement systems. In the following, we describe the details of the electric detection of the SPE signal using a TC, the results of the magnetic field dependence of the SPE signal in a high-magnetic-field range, and its comparison to that of the SSE thermopower.

## II. EXPERIMENTAL

The spin current for driving the SPE is generated via the spin Hall effect (SHE) from a charge current applied to N<sup>15,16</sup>. The SHE-induced spin current then forms spin accumulation at the N/F interface, whose spin vector representation is given by

$$\boldsymbol{\mu}_s \propto \theta_{\text{SHE}} \mathbf{j}_c \times \mathbf{n}, \quad (1)$$

where  $\theta_{\text{SHE}}$  is the spin Hall angle of N,  $\mathbf{j}_c$  the charge-current-density vector, and  $\mathbf{n}$  the unit vector normal to the interface directing from F to N.  $\boldsymbol{\mu}_s$  at the interface exerts spin-transfer torque to magnons in F via the interfacial exchange coupling at finite temperatures, when  $\boldsymbol{\mu}_s$  is parallel or anti-parallel to the equilibrium magnetization ( $\mathbf{m}$ )<sup>35,36</sup>. The torque increases or decreases the number of the magnons depending on the polarization of the torque ( $\boldsymbol{\mu}_s \parallel \mathbf{m}$  or  $\boldsymbol{\mu}_s \parallel -\mathbf{m}$ ), and eventually changes the system temperature by energy transfer, concomitant with the spin-current injection<sup>5,17,30</sup>. The energy transfer induces observable temperature modulation in isolated systems, which satisfies the following relation

$$\Delta T_{\text{SPE}} \propto \boldsymbol{\mu}_s \cdot \mathbf{m} \propto (\mathbf{j}_c \times \mathbf{n}) \cdot \mathbf{m}. \quad (2)$$

A schematic of the sample system and measurement geometry is shown in Fig. 1(a). The sample system is a Pt strip on a single-crystal YIG. The YIG layer is 112- $\mu\text{m}$ -thick and grown by a liquid phase epitaxy method on a 500- $\mu\text{m}$ -thick  $\text{Gd}_3\text{Ga}_5\text{O}_{12}$  substrate with the lateral dimension  $10 \times 10 \text{ mm}^2$ , where small amount of Bi is substituted for the Y-site of

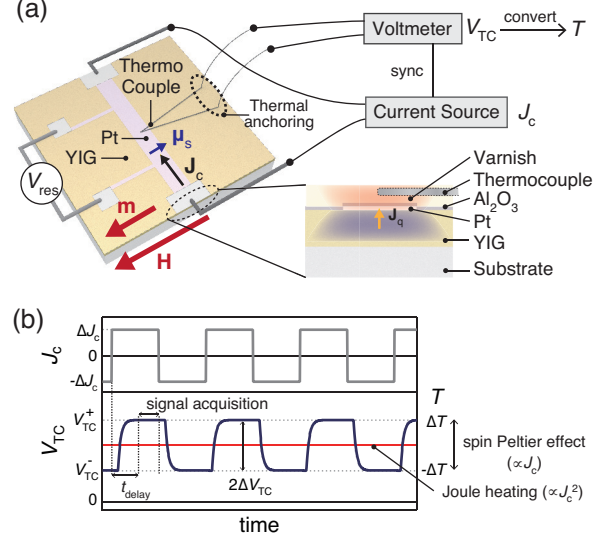


FIG. 1. (a) Schematic of the Pt/YIG sample and measurement system with a thermocouple wire.  $\mathbf{J}_c$ ,  $\boldsymbol{\mu}_s$ ,  $\mathbf{m}$ ,  $\mathbf{H}$ , and  $\mathbf{J}_q$  denote the applied current, the spin accumulation, the unit vector of the equilibrium magnetization, the magnetic field, and the heat current concomitant with the spin-current injection. In an isolated system,  $\mathbf{J}_q$  induces a temperature gradient by accumulating heat, which can be detected by the thermocouple. The resistance of the Pt strip is obtained by measuring  $V_{\text{res}}$ . The measurements were carried out by using the physical property measurement system, Quantum Design. (b) Expected responses due to the SPE and Joule heating when  $J_c$  is periodically changed from  $\Delta J_c$  to  $-\Delta J_c$  with the zero offset current  $J_c^0 = 0$ . Corresponding voltage signals  $V_{\text{TC}}^+$  and  $V_{\text{TC}}^-$  are obtained after the time delay  $t_{\text{delay}}$ . The SPE signal is extracted from the difference  $\Delta V_{\text{TC}} = (V_{\text{TC}}^+ - V_{\text{TC}}^-) / 2$ .

the YIG to compensate the lattice mismatching to the substrate. The Pt strip, connected to four electrodes, is 5-nm-thick, 0.5-mm-wide, fabricated by a sputtering method, and patterned with a metal mask. Then, the whole surface of the sample except the electrodes is covered by a highly-resistive  $\text{Al}_2\text{O}_3$  film with a thickness of  $\sim 100$  nm by means of an atomic layer deposition method. We attached a TC wire to the top of the Pt/YIG junction, where the wire is electrically insulated from but thermally connected to the Pt layer owing to the  $\text{Al}_2\text{O}_3$  layer. We used a type-E TC with a diameter of 0.013 mm (Omega engineering CHCO-0005), and fixed its junction part on the middle of the Pt strip using varnish. The rest of the TC wires were fixed on the sample surface for thermal-anchoring and avoiding thermal leakage from the top of the Pt/YIG junction [see the cross sectional view in Fig.

1(a)]. The expected thickness of the varnish between the TC and the sample surface is in the order of  $10\text{ }\mu\text{m}$ <sup>37</sup>. Note that the thickness of the varnish layer does not affect the magnitude of the signal while it affects the temporal response of the TC<sup>38</sup>. The ends of the Pt strip are connected to a current source and the other two electrodes are used for measuring resistance based on the four-terminal method. The magnetic field  $\mathbf{H}$  is applied in the film plane and perpendicular to the strip, thus is along  $\boldsymbol{\mu}_s$ , satisfying the symmetry of the SPE [Eq. (2)]. The TC is connected to electrodes on a heat bath, which acts as a thermal anchor, and further connected to a voltmeter via conductive wires. The measurements were carried out at 300 K and  $\sim 10^{-3}$  Pa.

For the electric detection of the SPE, we measured the amplitude difference ( $\Delta V_{\text{TC}}$ ) of the TC voltage  $V_{\text{TC}}$  in response to a change ( $\Delta J_c$ ) in the current  $J_c$  (so-called Delta-mode of the nanovoltmeter Keithley 2182A). After the current is set to  $J_c = J_c^0 \pm \Delta J_c$ , time delay ( $t_{\text{delay}}$ ) is inserted before measuring the corresponding voltages  $V_{\text{TC}}^{\pm}$ . Then,  $\Delta V_{\text{TC}}$  is obtained as  $\Delta V_{\text{TC}} = (V_{\text{TC}}^+ - V_{\text{TC}}^-)/2$ . The time delay  $t_{\text{delay}}$  is necessary because the temperature modulation occurred at the Pt/YIG junction takes certain time to reach and stabilize the TC. The appropriate value of  $t_{\text{delay}}$  can be determined from the delay-dependence of the SPE signal, which will be shown in Sec. III. For the SPE measurements, we set no offset current ( $J_c^0 = 0$ ) so that  $\Delta V_{\text{TC}}$  is free from Joule heating ( $\propto J_c^2$ ), and the SPE ( $\propto J_c$ ) is expected to dominate the  $\Delta V_{\text{TC}}$  signal [see Fig. 1(a)]. By using the measured values of  $\Delta V_{\text{TC}}$ , the temperature modulation ( $\Delta T$ ) is estimated via the relation  $\Delta T = S_{\text{TC}}\Delta V_{\text{TC}}$ , where  $S_{\text{TC}}$  is the Seebeck coefficient of the TC. For the low-field measurements ( $\mu_0 H < 0.1$  T), the reference value of  $S_{\text{TC}} = 61\text{ }\mu\text{V/K}$  at 300 K is used, while, for the high-field measurements, the field dependence of  $S_{\text{TC}}$ , determined by the method shown in Appendix A, is used.

### III. RESULTS AND DISCUSSION

First, we demonstrated the electric detection of the SPE at low fields. Figure 2(a) shows  $\Delta V_{\text{TC}}$  as a function of the field magnitude  $H$  at  $\Delta J_c = 10$  mA and  $t_{\text{delay}} = 50$  ms.  $\Delta V_{\text{TC}}$  clearly changes its sign when the field direction is reversed and the appearance of the hysteresis demonstrates that it reflects the magnetization curve of the YIG, showing the symmetry expected from Eq. (2)<sup>5,17</sup>. The small offset of  $\Delta V_{\text{TC}}$  may be attributed to the temperature modulation by the Peltier effect appearing around the current electrodes, Joule heating due

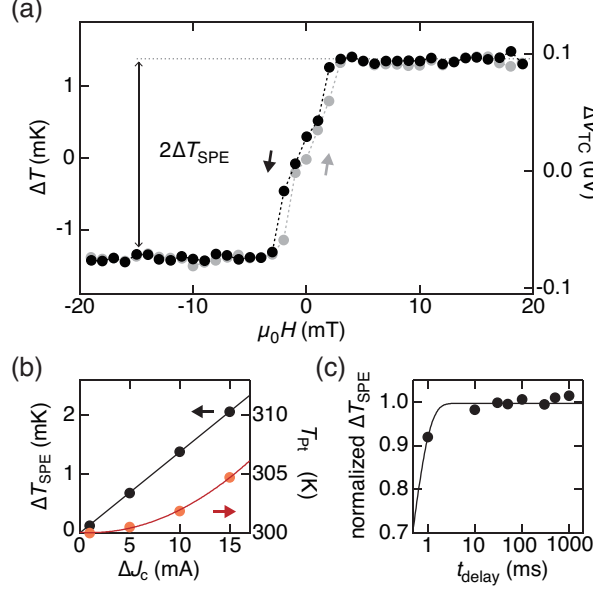


FIG. 2. (a) Field magnitude  $H$  dependence of  $\Delta V_{TC}$  (the right axis) and  $\Delta T$  without offset (the left axis), measured at  $\Delta J_c = 10$  mA and  $t_{delay} = 50$  ms. (b) Current  $J_c$  dependence of  $\Delta T_{SPE}$  at  $\mu_0 |H| = 0.1$  T, where  $\mu_0$  represents the permeability of vacuum. (c) Delay-time  $t_{delay}$  dependence of  $\Delta T_{SPE}$ . Plotted data are estimated from the results measured at  $\Delta J_c = 10$  mA and  $5 \text{ mT} < \mu_0 |H| < 20 \text{ mT}$  and normalized based on the fitting with  $B[1 - \exp(-t_{delay}/\tau)]$ , where  $B$  denotes the proportional constant and  $\tau = 0.4$  ms denotes the characteristic time scale. The acquisition time of 20 ms is used for measurements.

to small uncanceled current offsets, and possible electrical leakage of the applied current from the sample to the TC. Since the Peltier and resistance effects are of even functions of the magnetization cosines though the SPE is of an odd function, the SPE-induced temperature modulation  $\Delta T_{SPE}$  can be extracted by subtracting the symmetric response to the magnetization:  $\Delta T_{SPE} = [\Delta T(+H) - \Delta T(-H)]/2^5$ . Figure 2(b) shows the  $\Delta J_c$  dependence of  $\Delta T_{SPE}$  and the temperature ( $T_{Pt}$ ) of the Pt strip, estimated from the resistance of the strip. While  $T_{Pt}$  increases parabolically with the magnitude of  $\Delta J_c$  for Joule heating,  $\Delta T_{SPE}$  increases linearly as is expected from the characteristic of the SPE. This distinct dependencies show negligibly small contribution to  $\Delta T_{SPE}$  from the Joule heating in this study<sup>39</sup>. The magnitude of the SPE signal is estimated to be  $\Delta T_{SPE}/\Delta j_c = 3.4 \times 10^{-13} \text{ Km}^2/\text{A}$ , where  $\Delta j_c$  is the difference in  $j_c$ . This value is almost same as the value obtained in the thermographic experiments<sup>17,19</sup>; since in Ref.<sup>19</sup> the sine-wave amplitude  $A$  of  $\Delta T_{SPE}$  is di-

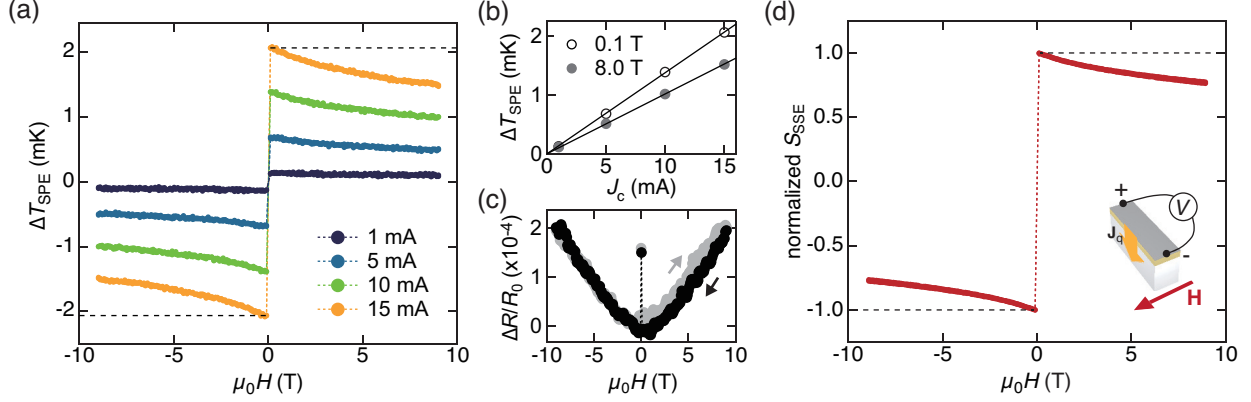


FIG. 3. (a) Field dependence of  $\Delta T_{\text{SPE}}$  including the high-magnetic-field range measured at various  $\Delta J_c$  values. (b)  $J_c$  dependence of  $\Delta T_{\text{SPE}}$  at  $\mu_0 |H| = 0.1$  T and 8.0 T. The solid lines represent the linear fitting. (c) Field dependence of the resistance change  $\Delta R = R(H) - R_0$ , where  $R$  denotes the resistance and  $R_0$  that at  $\mu_0 H = 0.1$  T. (d) Field dependence of the spin Seebeck voltage  $V_{\text{SSE}}$  normalized at  $\mu_0 H = 0.1$  T, where  $V_{\text{SSE}}$  is obtained by measuring the voltage under heater power of 100 mW and subtracting the component symmetric to  $H$ .

vided by the rectangular-wave amplitude of  $\Delta j_c$ , a correction factor of  $\pi/4$  is necessary, i.e.  $\Delta T_{\text{SPE}}/\Delta j_c = \pi A/(4\Delta j_c) = 3.7 \times 10^{-13}$   $\text{Km}^2/\text{A}$  in the previous study. We note that, in the above and following measurements,  $t_{\text{delay}} = 50$  ms is chosen based on the  $t_{\text{delay}}$  dependence of  $\Delta T_{\text{SPE}}$  [Fig. 2(c)], where  $\Delta T_{\text{SPE}}$  is almost saturated at  $t_{\text{delay}} > 10$  ms. Such finite but small thermal-stabilization time can be explained by the thermal diffusion from the junction to the TC and rapid thermal stabilization of the SPE-induced temperature modulation<sup>17</sup>.

Next, we measured the field dependence of the SPE at higher fields up to  $\mu_0 H = 9.0$  T. Figure 3(a) shows  $\Delta T_{\text{SPE}}$  as a function of  $H$ , where  $\Delta J_c$  is changed from 1 to 15 mA. The decrease of the  $\Delta T_{\text{SPE}}$  signal at higher fields is clearly observed for all the  $\Delta J_c$  values. As shown in Fig. 3(b), the signal shows a linear variation with  $J_c$  both at  $\mu_0 H = 0.1$  T and 8.0 T, demonstrating a constant decreasing rate. Since the resistance of the sample varies only 1 % at most [Fig. 3(c)], the junction temperature keeps constant during the field scan. The field dependence of the thermal conductivity of YIG is also irrelevant to the  $\Delta T_{\text{SPE}}$  suppression as it is known to be negligibly small at room temperature<sup>40</sup>. Thus we can conclude that the decrease is attributed to the nature of the SPE. By calculating the



magnitude of the decrease as

$$\delta_{\text{SPE}} = 1 - \frac{\Delta T_{\text{SPE}}(\mu_0 H = 8.0 \text{ T})}{\Delta T_{\text{SPE}}(\mu_0 H = 0.1 \text{ T})},$$

we obtained  $\delta_{\text{SPE}} = 0.26$ .

To compare  $\delta_{\text{SPE}}$  to the field-induced decrease of the SSE, we performed SSE measurements in a longitudinal configuration using a Pt/YIG junction system fabricated at the same time as the SPE sample. The SSE sample has the lateral dimension  $2.0 \times 6.0 \text{ mm}^2$  and the same vertical configuration as the SPE sample except for the absence of the  $\text{Al}_2\text{O}_3$  layer. The detailed method of the SSE measurement is available elsewhere<sup>28,41,42</sup>. Figure 3(d) shows the field dependence of the SSE thermopower in the Pt/YIG junction. The clear decrease of the SSE thermopower is observed. Importantly, the high-field response of the SSE is quite similar to that of the SPE in the Pt/YIG system. The decreased magnitude of the SSE  $\delta_{\text{SSE}}$ , defined in the same manner as the SPE, is estimated to be  $\sim 0.22$ , consistent with the previously reported values<sup>22–24</sup>.

The observed remarkable field-induced decrease of the SPE at room temperature shows that the SPE is likely dominated by low-energy magnons because the energy scale of the applied field is less than 10 K and thus much lower than the thermal energy of 300 K<sup>22</sup>. The origin of the strong contribution of the low-energy magnons in the SPE can be (i) stronger coupling of the spin torque to the low-energy (sub-thermal) magnons and (ii) greater propagation length of the low-energy magnons than those of high-energy (thermal) magnons<sup>34</sup>. While (i) is not well experimentally investigated, the existence of the  $\mu\text{m}$ -range length scale in the SPE<sup>19</sup> and the similarity between  $\delta_{\text{SPE}}$  and  $\delta_{\text{SSE}}$  suggest the dominant contribution from (ii) as in the case of the SSE<sup>24,34</sup>. In fact, recently, it has been demonstrated that the high magnetic fields reduce the propagation length of magnons contributing to the SSE<sup>34</sup>. This length-scale scenario can qualitatively explain the decrease in the SPE. Recalling that a heat current density ( $j_q$ ) existing over a distance ( $l$ ) generates the temperature difference  $\Delta T = \kappa^{-1} j_q l$  in an isolated system,  $\Delta T$  should decrease when  $l$  decreases, where  $\kappa$  is the thermal conductivity of the system. In the SPE,  $l$  corresponds to the magnon propagation length<sup>43</sup>, and a flow of magnons accompanies a heat current<sup>40</sup>. Consequently, when the high magnetic field is applied and the magnons with longer propagation length are suppressed by the Zeeman gap, the averaged magnon propagation length decreases and thus results in the reduced  $\Delta T$ . To further investigate the microscopic mechanism of the SPE, consideration

of the spectral non-uniformity may be vital both in experiments and theories.

#### IV. SUMMARY

In this study, we showed the magnetic field dependence of the spin Peltier effect (SPE) up to 9.0 T at 300 K in a Pt/YIG junction system. We established a simple but sensitive detection method of the SPE using a commonly-available thermocouple wire. The SPE signals were observed to be suppressed at high magnetic fields, highlighting the stronger contribution of low-energy magnons in the SPE. The similar decreasing rate of the SPE-induced temperature modulation to that of the SSE-induced thermopower suggests that the suppression originates the decrease in the magnon propagation length as in the case of the SSE. We anticipate that the experimental results and the method reported here will be useful for systematic investigation of the SPE.

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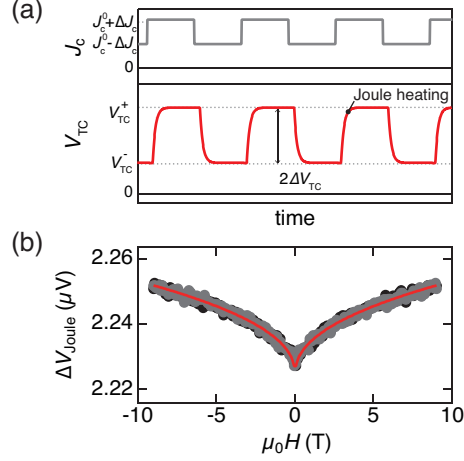


FIG. 4. (a) Expected responses due to the Joule heating at the finite offset current  $J_c^0 \neq 0$ . (b) Field dependence of  $\Delta V_{Joule}$  at  $\Delta J_c = 0.5$  mA and  $J_c^0 = 5$  mA. The solid curve represents the calibration line determined by the fitting.

### Appendix A: Calibration of Thermo Couple at High Magnetic Fields

To measure the field dependence of  $S_{TC}$ , we used the Joule-heating-induced signal as a reference. By adding a non-zero offset ( $J_c^0$ ) to the applied current, we obtained the temperature modulation induced by the Joule heating, of which the power  $P$  changes from  $P(H) = R(H)(J_c^0 - \Delta J_c)^2$  to  $P(H) = R(H)(J_c^0 + \Delta J_c)^2$ , where  $R$  denotes the resistance of the strip [Fig. 4(a)]. Figure 4(b) shows the magnetic field dependence of the component of  $\Delta V_{TC}$  symmetric to the field ( $\Delta V_{Joule} = [\Delta T_{TC}(+H) + \Delta T_{TC}(-H)]/2$ ). As the change in  $R$ , due to the ordinary, spin Hall, and Hanle magnetoresistance effects<sup>44,45</sup>, is in the order of 0.02 % [Fig.3(c)], its contribution to  $P$  can be neglected. Similarly, the field dependence of the thermal conductivity of YIG is negligibly small<sup>40</sup>, ensuring the constant temperature change. Accordingly, the field dependence of  $\Delta V_{Joule}$  directly reflects  $S_{TC}(H)$ . It increases by a factor of  $\sim 1$  % when the field magnitude increases up to 9.0 T. We approximated the field dependence of  $S_{TC}(H)$  as  $S_{TC}(H) = 61 (1 + 4.54 \times 10^{-3} |\mu_0 H \text{ [T]}|^{0.453}) \text{ } [\mu V/K]$  by determining the relative change from the measurement results ( $\Delta V_{Joule}(H) / \Delta V_{Joule}|_{H=0}$ ) and the absolute value from the reference value.

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of the Pt strip to the surface ( $\sim 30 \mu\text{m}$ ) is smaller than the width ( $500 \mu\text{m}$ ).

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