

CHORUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Quantitative estimation of thermoelectric contributions in spin pumping signals through microwave photoresistance measurements

Jun Cheng, Kang He, Man Yang, Qi Liu, Rui Yu, Liang Sun, Jinjun Ding, Bingfeng Miao, Mingzhong Wu, and H. F. Ding

Phys. Rev. B **103**, 014415 — Published 12 January 2021 DOI: [10.1103/PhysRevB.103.014415](https://dx.doi.org/10.1103/PhysRevB.103.014415)

23 **Introduction**

24 Recently, spintronic research shifts interests from spin-polarized current to pure spin current. In 25 conductors, pure spin current can deliver maximum spin angular momentum with minimum 26 electrons [1,2]. In magnetic insulators, spin information can transfer in the form of collective motion 27 of magnetic moments, i.e., spin waves [3-5], without any moving charge carriers. Utilizing pure spin 28 current generates less Joule heat and thus has less power consumption, as compared to the spin 29 polarized current. Spin Hall effect (SHE) [6,7], spin pumping [8-10] and spin Seebeck effect (SSE) 30 [11,12] based techniques have been developed to generate pure spin current. Among various 31 mechanisms, spin pumping has the unique interface spin current characterizing capability, thus has 32 also been widely used to characterize the spin Hall angle and spin diffusion length of heavy metals 33 [13-16]. Upon the application of the microwave excitation and with an appropriate external magnetic 34 field, the magnetic moments in a ferromagnet can be driven into a coherent precession 35 (ferromagnetic resonance, FMR) [17]. This non-equilibrium magnetization dynamic in a ferromagnet 36 acts as the source for an angular momentum flow, which pumps a spin current into its neighboring 37 non-magnetic layer [8-10]. Due to the lack of net charge current, the detection of pure spin current 38 mainly relies on the inverse spin Hall effect (ISHE) in metals with strong spin-orbit coupling, which 39 converts spin current into charge current with density $J_c = \theta_{\text{SH}} (2e/\hbar) J_s \times \sigma$ [10]. Here, θ_{SH} is the 40 spin Hall angle which characterizes the efficiency of the spin-charge conversion, *e* is the electronic 41 charge, h is the reduced Planck constant, J_S represents the spin current density, and σ denotes the 42 spin direction parallel with the equilibrium magnetization of the ferromagnet. Because of the 43 orthogonal relation, perpendicular flowing of spin current with spin polarization along the *y-*direction 44 results in an in-plane charge current flows along the *x*-direction (see coordinates in Fig. 1). In an

open-circuit, a spin pumping voltage $E_{SP} \propto J_S \times \sigma$ is obtained [Fig. 1(a)].

47 **FIG. 1.** Schematic illustrations of (a) spin pumping (b) longitudinal spin Seebeck effect.

48

49 Spin pumping requires a microwave to excite the precession of the magnetic moments. 50 However, the microwave irradiation may also bring possible thermoelectric artifacts. Both the eddy 51 currents in conductors and magnon–phonon scattering in ferromagnet could heat the samples [18-22]. 52 Typically, for devices with a thin film deposited on a thick substrate, the temperature increase might 53 establish a perpendicular temperature gradient, which gives rise to thermoelectric signals such as the 54 Nernst effect, the longitudinal spin Seebeck effect (LSSE) [12], the anomalous Nernst effect (ANE) 55 [23-25] and the spin-dependent Seebeck effect (SdSE) [26] when the ferromagnetic layer is 56 conducting. Therefore, the spin pumping signals are potentially contaminated with thermoelectric 57 contributions [27,28]. The Nernst effect can be easily excluded since it is independent with the 58 magnetic field, and the SdSE typically is very small [29,30]. However, the separation of the LSSE 59 and ANE contributions from the spin pumping signal is not straightforward. Under an out-of-plane 60 (perpendicular) temperature gradient, the LSSE enables a pure spin current injected vertically from 61 the ferromagnet into the heavy metal and detected as a transverse thermal voltage $E_{LSE} \propto \nabla_z T \times \sigma$ 62 through the ISHE, where σ is parallel with the magnetization *M* of the ferromagnet, as depicted in

Fig. 1(b). When the ferromagnet is conducting, under the same $\nabla_{\tau}T$, the ANE of $E_{\text{ANE}} \propto \nabla_{\tau}T \times \mathbf{M}$ 64 also gives rise to a transverse voltage. One can readily find that the spin pumping, LSSE, and ANE 65 voltages all share the same symmetry with the same angular dependence, hence inseparable and 66 additive. Furthermore, if the thermoelectric contributions are dominating, the measured signal in the 67 ferromagnet/heavy metal structure may even fail to denote the spin Hall angle sign of the heavy 68 metal [31]. Therefore, it is important to develop a quantitative method to separate thermoelectric 69 contributions in the spin pumping experiments.

70 In this work, we present a universal and quantitative method to obtain the thermoelectric 71 contributions in spin pumping voltage via the assistance of the microwave-photoresistance 72 measurements. We apply this method on two typical systems, i.e., $P_y(N_{180}Fe_{20})/Pt$ and 73 YIG(Y₃Fe₅O₁₂)/Pt bilayers, and find that the microwave radiation indeed can raise the sample 74 temperatures and create a perpendicular temperature gradient. This vertical temperature gradient 75 induces a sizable thermal voltage due to the LSSE and the ANE near zero magnetic field, which acts 76 as a background for spin pumping signals at higher fields. However, the additional heat dissipation 77 due to magnon-phonon scattering at the FMR condition is negligibly small, in consistent with 78 previous findings [27,28]. This conclusion is further supported by the field-dependent microwave 79 absorption measurement using a vector network analyzer. Therefore, we conclude that the 80 thermoelectric contributions are little, if any, as compared with the spin pumping signal in our 81 measurement geometry.

82

83 **Experiments and results**

85 **FIG. 2.** (a) Schematic illustration of the experimental setup for spin pumping measurements of Py/Pt 86 bilayer. (b) Field-dependent voltage for a Py(6 nm)/Pt(3 nm) bilayer stripe with 8.5 GHz microwave 87 irradiation, where the magnetic field is applied along the *y*-direction. The black symbols represent 88 the experimental data, and the red lines are the Lorentz line fittings. The inset presents the zoomed-in 89 feature near zero magnetic field. 2Δ*V*0 denotes the difference of the voltage background for the 90 positive and negative fields. (c) Microwave frequency *f* dependent resonance field *H*r. Black circles 91 are the experimental data, and the red line is the fitting with Kittel equation. (d) Microwave input 92 power dependent *V_r* (black hexagon, left scale) and ΔV_0 (red triangle, right scale). The lines are linear 93 fittings.

94

95 We perform the measurements with two representative ferromagnet/normal metal bilayer 96 structures, Py/Pt and YIG/Pt, where Py is a metal and YIG is an insulator. The Py(6 nm)/Pt(3 nm)

97 bilayer with the length *l*=2 mm and the width *w*=20 μm is deposited on the thermally oxidized Si 98 substrate (total thickness is 0.5 mm, $SiO₂$ is \sim 300 nm) and glass substrate (1 mm thick). For the 99 YIG/Pt system, we first deposit a 25-nm-thick YIG film on a (111) -Gd₃Ga₅O₁₂ (GGG) substrate (0.5 100 mm thick) and perform post-annealing at 800°C at atmosphere for 4 hours. Then, a 5-nm Pt stripe 101 (*l*=1.53 mm and *w*=40 μm) is deposited on the YIG continuous film. A 100-nm copper coplanar 102 waveguide (CPW) with a 50- Ω characteristic impedance is fabricated to introduce the microwaves, 103 with the Pt stripes integrated into the slots between the signal and ground lines of the CPW [Fig. 104 $2(a)$]. In this configuration, the microwave magnetic field h_{rf} is primarily along the *z*-direction. In 105 order to achieve high sensitivity, a lock-in technique is used. We modulate the microwave with a 106 Transistor-Transistor Logic (TTL) signal with a frequency of 8.3 kHz and measure the voltage as the 107 function of an external magnetic field applied in the *xy*-plane with an angle *α* with respect to the 108 *x*-direction, as marked in Fig. 2(a). All films are deposited by magnetron sputtering at room 109 temperature, with the thickness calibrated by x-ray reflection measurements. And all measurements 110 are performed at room temperature except for the *R*-*T* curve. For the measurement of Py/Pt bilayer, 111 the microwave frequency is 8.5 GHz with a 355-mW power unless specified.

112 Figure 2(b) presents the voltage obtained across the two ends of the Py/Pt bilayer stripe, where 113 the magnetic field is applied along the *y*-direction. In this geometry, the spin rectification due to the 114 microwave induction current and oscillating anisotropic magnetoresistance (AMR) is minimized. It 115 shows that a pair of voltage peak and dip appears at ± 1.1 kOe with a symmetric Lorentz line shape, 116 indicating its pure spin current origin. The different amplitudes in the spin pumping signals for $\pm H$ 117 [Fig. 2(b)] in our measurement are caused by different precession angles of the magnetization (*M*) 118 under magnetic fields with opposite directions. After normalizing the measured V_{sp} with their

119 corresponding in-plane and out-of-plane precession angles produce, the normalized signals will 120 become almost identical [32]. Interestingly, the background for the positive and negative magnetic 121 fields has a sizable difference, marked as 2Δ*V*⁰ [see Fig. 2(b)]. The voltage at low fields is 122 asymmetric in *H*, with a field dependence following the magnetization curve of Py. The data show a 123 coercivity smaller than 10 Oe [see Fig. 2(b) inset]. This zero-field step signal may result from 124 non-resonant spin rectification of the Py layer [33], and/or microwave heating induced ANE of the 125 Py layer and LSSE of the Py/Pt bilayer. While the non-resonant spin rectification is proportional with 126 the magnetic field derivative of resistance d*R*/d*H*, it disappears after the magnetization is saturated 127 [33]. Since no discernible difference of voltage background is observed for zero field and high fields, 128 we conclude that the voltage step near zero field here is a thermoelectric contribution.

129 Now we turn to the signal at the resonance field, *H*r. As depicted in Fig. 2c, the *f*-dependent *H*^r 130 can be well described by the Kittle equation, which yields the saturation magnetization $4\pi M_0(Py)$ to 131 be 7.53 kOe. We further define the amplitude of the Lorentzian line fitting at the positive resonance 132 field as V_r , which is typically attributed to the spin pumping signal only. However, if the temperature 133 enhancement due to magnon-phonon scattering under the resonance condition is non-negligible, 134 thermoelectric signals from the ANE and the LSSE will also be involved. As mentioned above, the 135 signals from ANE, LSSE, and spin pumping all share the same symmetry, it is difficult to distinguish 136 them by routing methods. Moreover, we further find that both ΔV_0 and V_r are linearly proportional to 137 the input microwave power [Fig. 2(d)]. Thus, it is not possible to distinguish them from their power 138 dependences, either.

As discussed above, ΔV_0 is of pure thermoelectric origin, while V_r at the resonance state may 140 have both spin pumping and thermoelectric contributions. Thus, it is pivotal to find a parameter 141 which links zero-field ΔV_0 and possible thermoelectric contributions in V_r . A natural option is the 142 resistance of the Py/Pt bilayer, which relates to the sample temperature as well as the temperature 143 gradient. Figure 3(a) shows the temperature-dependent resistance change with respect to the 144 resistance at 300 K. Since the sample is a metallic bilayer, its resistance shows a linear increase with 145 the temperature. The fitting yields a slope of $7.12(\pm 0.02)$ Ω/K . Thus, it could serve as a sensitive tool 146 to probe the temperature change.

148 **FIG. 3.** (a) *R*-*T* curve of a Py(6 nm)/Pt(3 nm) bilayer near room temperature, with a slope of 7.12(\pm 149 0.02) Ω/K. R_{300K} = 8.86 kΩ. (b) Magnetic field-dependent voltages with dc current + I_0 (+0.9 mA, red 150 curve) and −*I*0 (−0.9 mA, black curve) for the Py/Pt sample, respectively. (c) The resistance 151 difference Δ*R* of the Py/Pt bilayer between microwave on and off states. (d) Linear relation between 152 the thermoelectric ground ∆*V*0 and the resistance difference background Δ*R*b. (e) Magnetic field 153 dependent ΔR for different α . The curves are shifted for clarity. (f) Magnetic field-dependent S₂₁ 154 parameter data for a "7 mm×7 mm" Py(6 nm)/Pt(3 nm) film.

155

156 We further investigate the change of the sample resistance during the spin pumping 157 measurement. In order to obtain this, we feed the sample with a small current and measure the 158 voltage change as a function of the external field. We note that the microwave is modulated with an 159 8.3-kHz TTL signal, and the lock-in detection picks up the voltage difference between the 160 microwave on and off states with the same frequency. Therefore, the resistance change reflects the 161 temperature difference between the microwave on and off states (in ms), instead of the real 162 temperature of the Py/Pt bilayer. Figure 3(b) presents the *H* dependent voltages with dc current +*I*⁰ 163 (+0.9 mA, red curve) and −*I*0 (−0.9 mA, black curve), respectively. The magnetic field is applied 164 along the *y*-axis ($\alpha = 90^{\circ}$). We obtain the resistance difference of the Py/Pt bilayer between the 165 microwave on and off states by $\Delta R = [V(+I_0) - V(-I_0)]/2I_0$, as presented in Fig. 3(c). The ΔR curve 166 has a non-zero background ΔR_b and a peak with the amplitude ΔR_r coincide with the resonance field *H_r*. At the magnetic field away from H_r , ΔR_b comes from the heating due to the microwave only, and 168 increases with the power. Thus, we find a linear relation between ΔV_0 [the thermoelectric background 169 signal depicted in Fig. 2(b)] and ΔR_b (the resistance increase background value), with a slope 0.162 170 *μ*V/mΩ [Fig. 3(d)]. If we further obtain the additional heating-induced resistance increase at the 171 FMR condition, the thermoelectric contributions in the spin pumping signal can be calculated. 172 However, aside from the temperature increase via magnon–phonon scattering at FMR, the resistance 173 change Δ*R* has another origin which also needs to be addressed. As a magnetic material, Py has AMR 174 with $R_{\parallel} > R_{\perp}$, where R_{\parallel} and R_{\perp} are the longitudinal (*M*||*I*) and transverse (*M*⊥*I*) magnetoresistance, 175 respectively. At the FMR, the magnetization precession alters the angle of the magnetization with 176 respect to the dc current, resulting in a change of the time-averaged AMR. This is termed as the 177 microwave photoresistance ΔR_{MW} , and its angular dependence is given by: [34]

$$
\Delta R_{\text{MW}} = R_{\text{A}}(-\alpha_{1}^{2}\cos 2\alpha - \beta_{1}^{2}\cos^{2}\alpha)/2
$$
 (1)

Here, R_A is the difference between R_{\parallel} and R_{\perp} , which is about 36.19 Ω for our Py/Pt sample, and $α_1$, 180 β_1 are the amplitudes of in-plane and out-of-plane precession angles of the magnetization, 181 respectively. According to the FMR theory, the in-plane and out-of-plane precession angles have a 182 relation of $\alpha_1/\beta_1 = \sqrt{1 + M_0/H_r}$ [16,34], with M_0 being the saturation magnetization of the 183 ferromagnet. Equation (1) shows that ΔR_{MW} is α -dependent, and disappears at

184
$$
|\cos \alpha| = \sqrt{\frac{H_{\rm r} + M_0}{3H_{\rm r} + 2M_0}}
$$
 (2)

185 For the Py/Pt bilayer, we find that ∆*R*_{MW} equals to zero when *α*=46.7°. Thus, the residual resistance 186 enhancement at the FMR condition with α =46.7° can be attributed to the temperature increase only.

Figure 3(e) presents the ∆*R* versus *H* curves in the vicinity of the resonance field *H*^r 187 for 188 different *α*. The curves are shifted for clarity. When *α* is varied from 90° to 36°, ∆*R*r changes from 189 positive to negative and disappears at around 46.7°. For *α*=46.7°, ∆*R* is almost a flat curve. The 190 fitting yields ∆*R*r = −0.05(±0.008) mΩ. Combined with the calibration curve presented in Fig. 3(d), 191 we estimate the thermoelectric signal to be $\langle 9.4 \times 10^{-3} \mu V \rangle$ [the product of the slope in Fig. 3(d) and 192 the measure ∆*R*r]. Thus, thermal contributions in spin pumping voltage for Py/Pt are < 0.09% [0.009 193 μ V/10.20 μ V, with 10.20 μ V is the value of the symmetrical line fitted by the positive magnetic field 194 part in Fig. 2(b)], which can be safely neglected. The slope of *R*-*T* curve is 7.12 Ω/K [Fig. 3(a)], and 195 the microwave on-off resistance change ∆*R*b is 6.01 mΩ [Fig. 3(d)] for 355-mW microwave power, thus we estimate the ΔT due to off-resonance microwave heating to be $8.43(\pm 0.48) \times 10^{-4}$ K [the ratio 197 of the measured ∆*R*_b and the slope in Fig. 3(a)], and the additional ∆*T* at FMR condition due to 198 magnon-phonon scattering is $\leq 8.15 \times 10^{-6}$ K.

199 It is important to emphasize that both ∆*R* and ∆*T* are not the resistance and temperature

200 differences compared with room temperature after the microwave irradiation. Instead, they 201 correspond to the quasi-steady resistance and temperature differences between microwave on and off 202 state, which is modulated by lock-in amplifier with 8.3 kHz. It is also interesting to note that the 203 thermoelectric signal at FMR state is about two orders' magnitude smaller than that of the 204 off-resonance state. In order to understand this, we perform S parameter S_{21} using a vector network 205 analyzer (VNA). S_{21} characterizes the transmission insertion loss of the whole devices, obtained 206 through the ratio of transmitted and input microwave. Due to the small volume of stripe line sample, 207 the FMR absorption dip is not observed. Therefore, a Py(6nm) /Pt(3nm) bilayer film of lateral 208 dimension 7 mm×7 mm is grown to achieve a reasonable signal-to-noise ratio. In consistent with the 209 spin pumping measurements, microwave absorption due to FMR occurs at ± 1.1 kOe [Fig. 3(f)]. We 210 note that the additional absorption due to FMR is relatively small compared with the S_{21} parameter 211 background, around 0.1%. This explains why thermoelectric signal for ANE/LSSE has sizable 212 contribution near zero magnetic field, while it is negligibly small at FMR condition for Py/Pt bilayer 213 system. And we expect that thermoelectric contribution plays important role only if the magnetic 214 contrast in S_{21} parameter is comparable with non-magnetic background.

215 Recently, aside from magnetic metals, magnetic insulators also attract growing interests from 216 spintronics community. Due to its ability to accommodate pure spin currents without charge carriers, 217 magnetic insulators have great potential for low-power spintronics application. Among various 218 magnetic insulators, YIG has the unique attributes of ultra-low damping [35], long spin diffusion 219 length [3]. It has been widely investigated in spin Hall magnetoresistance (SMR) [36,37], spin 220 Seebeck effect [12], spin pumping [38,39] and photon-magnon coupling [40,41] etc. Therefore, it is 221 intriguing to study the thermoelectric contributions in spin pumping experiment of YIG/Pt system.

222 Figure 4(a) illustrates our setup of YIG/Pt measurements. Figure 4(b) presents the spin pumping 223 curve under the irradiation of 5 GHz microwave with 355 mW, where the magnetic field is applied 224 along the *y*-axis (α =90°). A 20-*μV* voltage with opposite polarity is observed at \pm 1.2 kOe. Fitting the resonance magnetic field H_r dependent microwave frequency [Fig. 4(c)] with Kittel equation yields 226 the saturation magnetization $4\pi M_0 = 1.40$ kOe, which is similar as the reported value for YIG film 227 [42]. Because of the small half-line width of YIG film, the peak of spin pumping curve here is much 228 sharper than that of Py/Pt system. Although there may exist magnetic proximity effect in the YIG/Pt 229 bilayer system [43], the possible ANE has been shown to be negligible [44]. Thus, the voltage step 230 near zero-field ∆*V*₀ mainly comes from the LSSE of YIG/Pt bilayer.

232 **FIG. 4.** (a) Schematic illustration of the experimental setup for spin pumping measurement of 233 YIG/Pt bilayer. (b) Field-dependent voltage for a YIG/Pt(5 nm) bilayer stripe with 5 GHz microwave 234 irradiation, where the magnetic field is applied along the *y*-direction. (c) Ferromagnetic resonance 235 field dependent microwave frequency. (d) ΔV_0 as the function of ΔR_b . The red line is the linear fitting

236 with a slope of $2.8 \times 10^{-3} \mu V/m\Omega$. (e) *H*-dependent ΔR for different *α*. The curves are shifted for 237 clarity. (f) The *R*-*T* curve of YIG/Pt(5 nm) near room temperature, where the slope is around 238 $3.48(\pm 0.04)$ Ω/K . $R_{300K} = 3.72$ k Ω .

239

240 By measuring the spin pumping curve with 0 mA, and ± 0.3 mA under microwave with 241 different power, we obtain the corresponding zero-field voltage step ∆*V*0 and resistance difference 242 ∆*R*_b at off-resonance state for the YIG/Pt bilayer. The calibration curve for ∆*V*₀ versus ∆*R*_b for the 243 YIG/Pt bilayer is presented at Fig. 4(d). ∆*V*0 is linearly proportional with ∆*R*b, with a slope of 2.8×10⁻³ μV/mΩ. Although YIG is insulating, the resistance in the YIG/Pt depends on the direction 245 of the magnetization of the underlying YIG with respect to the current due to the SMR effect [36,37]. 246 When the magnetization of YIG rotates within the *xy*-plane, SMR has exactly the same angular 247 dependence as AMR. Thus, YIG/Pt also has microwave photoresistance ΔR_{MW} with the same 248 symmetry as that of Py/Pt, described by Eq. (1). With the measured parameters of our YIG/Pt sample, 249 we calculate that ΔR_{MW} disappears at $\alpha = 39.6^{\circ}$. Any detected resistance change at this specific angle 250 can be attributed to the heating due to magnon-phonon scattering at the FMR condition. We present 251 the ∆*R*r of YIG/Pt at the vicinity of *H*r of YIG for different magnetic field directions in Fig. 4(e). The 252 polarity of ΔR_r changes from positive at *α* = 90.0° to negative at *α* = 29.6°, vanishing at *α* = 39.6° 253 with a noise level < 0.3 mΩ. We note the small deviation of resonance field H_r (< 8 Oe) at different 254 angles is due to the misalignment between magnetic field and sample plane. In combination with the calibration curve in Fig. 4(d), we obtain the thermoelectric contributions of YIG/Pt to be $\leq 8.4 \times 10^{-4}$ 256 μ V, which is around 4~5 order's smaller than the spin pumping voltage. Therefore, in our geometry, 257 thermoelectric contributions in spin pumping signal of YIG/Pt are also negligibly small. The

258 resistance difference background ∆*R*b is 1170 mΩ for YIG/Pt under 355-mW microwave irradiation 259 [same condition for Fig. 4(b)], in combination with the slope of R -*T* curve, 3.48(\pm 0.04) Ω /K [Fig. 260 4(f)], we estimate the temperature difference is 0.34 K near zero magnetic field. Meanwhile, we estimate the additional temperature increase at the FMR condition is less than 8.62×10^{-5} K.

262

263 **FIG. 5.** Lock-in modulation frequency dependent ΔR_b (a), and ΔV_0 and V_r (b) for Py/Pt deposited on 264 Si and glass substrates. All voltages have been normalized to the value with $f_{\text{mod}} = 8.3$ kHz. The 265 applied microwave is 8.5 GHz in frequency and 355-mW in power.

266

267 **Discussion**

268 We further study the influence of substrate thermal conductivity and the lock-in modulation 269 frequency on the thermoelectric effect. In addition to thermally oxidized Si, we also deposit a Py(6 270 nm)/Pt(3 nm) bilayer onto a glass substrate, whose thermal conductivity is about 2 orders of 271 magnitude smaller than that of Si. Figure. 5(a) presents the background resistance difference ΔR_b as 272 function of lock-in frequency *f*mod on both the Si and the glass substrate. With decreasing *f*mod, Δ*R*^b 273 increases sharply at low frequency. This can be explained by the relative slow bulk thermal 274 relaxation, similar feature had been reported in Ref. [45]. For the Si substrate, we estimate the ∆*T* 275 due to off-resonance microwave heating to be 56.4 mK for 11.6 Hz of f_{mod} , almost 67 times large as 276 that of 8.3 kHz. Although the microwave power applied on Py layer deposited on the glass substrate 277 is smaller due to the low microwave transmission efficiency, ΔR_b for glass substrate is larger than 278 that of thermally oxidized Si substrate. Therefore, the global temperature enhancement is larger for 279 low thermal conductivity substrate. In addition, we expect that temperature increase for thicker 280 ferromagnet should be larger as the absorbed microwave enhances thus producing more heating. The 281 much higher lock-in frequency and thinner ferromagnet qualitatively explain the observed smaller 282 temperature increase in this study as compared with those reported in previous works [22,46]. 283 Interestingly, we find the thermoelectric background Δ*V*0 for Py/Pt bilayer are almost *f*mod 284 in-dependent on both the Si and the glass substrates [Fig. 5(b)]. This implies that interfacial 285 temperature gradient can be established with a fast speed. This observation is consistent with 286 temporal evolution study of spin Seebeck effect, where the interfacial temperature gradient is found 287 to be stable within 1 μs, while the global temperature itself needs several ms to saturate [47]. And the 288 lock-in frequency ($\leq 10^8$ Hz) independent spin Seebeck effect for Pt/YIG (thin film) was also 289 reported [48].

290 Although we focus our study on the thermoelectric contributions of the measured spin 291 pumping signal with out-of-plane microwave magnetic field in this manuscript, our method is not 292 limited to this specific geometry. As long as the angular dependence of the microwave 293 photoresistance ΔR_{MW} and microwave absorption at FMR condition (proportional with the square of 294 microwave magnetic field component that is perpendicular to the magnetization of ferromagnet) is different, our method will be effective. For instance, when h_{rf} is along the *y*-direction, the microwave absorption at FMR condition is proportional to $\cos^2 \alpha$, while the angular dependence of $\Delta R_{\rm MW}$ is still

$$
\Delta R_{\text{MW}} = R_{\text{A}}(-\alpha_{1}^{2} \cos 2\alpha - \beta_{1}^{2} \cos^{2} \alpha)/2
$$
 [34]. We note that the in-plane and out-of-plane precession
angles of the magnetization α_{1} , β_{1} are external field direction dependent in this geometry. However,
the relation $\alpha_{1}/\beta_{1} = \sqrt{1 + M_{0}/H_{\text{r}}}$ is always maintained. Thus, ΔR_{MW} disappears at
 $\alpha = \arccos \sqrt{\frac{H_{\text{r}} + M_{0}}{3H_{\text{r}} + 2M_{0}}}$, where the additional resistance increase at the FMR condition can be
attributed to the thermal effect. With the calibrated curve for voltage background at non-resonant
condition, one can obtain the thermoelectric contributions in spin pumping signal as well. In addition,
it is also very interesting to apply our approach to investigate thermoelectric contributions in the
spin-torque ferromagnetic resonance technique [49,50], where the microwave current is directly
injected into the sample and the thermal effect might be stronger.

306

307 **Summary**

308 In this work, we present a quantitative method to obtain the thermoelectric contributions in 309 spin pumping signals. Benefiting from their different angular dependence on the magnetization 310 direction, we can isolate the resistance increase due to magnon-phonon scattering at the FMR 311 condition from the microwave photoresistance. In combination with the calibrated curve for 312 non-resonant voltage background, we further quantitatively obtain the thermoelectric contributions at 313 the FMR condition. Although sizable LSSE/ANE are observed near zero magnetic field for Py/Pt and 314 YIG/Pt, they are negligible in resonant spin pumping signals. The influence of the substrate thermal 315 conductivity and the lock-in modulation frequency are also discussed. Our work also demonstrates 316 that spin pumping is a reliable technique to investigate pure spin current behavior, no matter the 317 ferromagnet is a conductor or an insulator.

318 **Acknowledgement**

- 319 This work was supported by the National Key R&D Program of China (Grant No.
- 320 2018YFA0306004 and 2017YFA0303202), the National Natural Science Foundation of China
- 321 (Grants No. 51971110, No. 11974165, No. 11734006, No. 11727808), and the Natural Science
- 322 Foundation of Jiangsu Province (Grant No. BK20190057). Work at CSU was supported by the U.S.
- 323 National Science Foundation under Grants No. EFMA-1641989 and No. ECCS-1915849.
- 324

325 **References**

- 326 [1] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A.
- 327 Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
- 328 [2] I. Zutic, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76** (2004).
- 329 [3] Y. Kajiwara, K. Harii, S. Takahashi, J. Ohe, K. Uchida, M. Mizuguchi, H. Umezawa, H. Kawai,
- 330 K. Ando, K. Takanashi, S. Maekawa, and E. Saitoh, Nature **464**, 262 (2010).
- 331 [4] A. V. Chumak, V. I. Vasyuchka, A. A. Serga, and B. Hillebrands, Nat. Phys. **11**, 453 (2015).
- 332 [5] M. Collet, X. de Milly, O. D. Kelly, V. V. Naletov, R. Bernard, P. Bortolotti, J. Ben Youssef, V. E.
- 333 Demidov, S. O. Demokritov, J. L. Prieto, M. Munoz, V. Cros, A. Anane, G. de Loubens, and O. Klein,
- 334 Nat. Commun. **7**, 10377 (2016).
- 335 [6] J. E. Hirsch, Phys. Rev. Lett. **83**, 1834 (1999).
- 336 [7] S. F. Zhang, Phys. Rev. Lett. **85**, 393 (2000).
- 337 [8] Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, Phys. Rev. Lett. **88**, 117601 (2002).
- 338 [9] Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, Phys. Rev. B **66**, 224403 (2002).
- 339 [10] E. Saitoh, M. Ueda, H. Miyajima, and G. Tatara, Appl. Phys. Lett. **88**, 182509 (2006).
- 340 [11] K. Uchida, S. Takahashi, K. Harii, J. Ieda, W. Koshibae, K. Ando, S. Maekawa, and E. Saitoh, 341 Nature **455**, 778 (2008).
- 342 [12] K. Uchida, H. Adachi, T. Ota, H. Nakayama, S. Maekawa, and E. Saitoh, Appl. Phys. Lett. **97**, 343 172505 (2010).
- 344 [13] O. Mosendz, V. Vlaminck, J. E. Pearson, F. Y. Fradin, G. E. W. Bauer, S. D. Bader, and A.
- 345 Hoffmann, Phys. Rev. B **82**, 214403 (2010).
- 346 [14] A. Azevedo, L. H. Vilela-Leao, R. L. Rodriguez-Suarez, A. F. L. Santos, and S. M. Rezende, 347 Phys. Rev. B **83**, 144402 (2011).
- 348 [15] H. L. Wang, C. H. Du, Y. Pu, R. Adur, P. C. Hammel, and F. Y. Yang, Phys. Rev. Lett. **112**,
- 349 197201 (2014).
- 350 [16] Z. Feng, J. Hu, L. Sun, B. You, D. Wu, J. Du, W. Zhang, A. Hu, Y. Yang, D. M. Tang, B. S.
- 351 Zhang, and H. F. Ding, Phys. Rev. B **85**, 214423 (2012).
- 352 [17] C. Kittel, Phys. Rev. **73**, 155 (1948).
- 353 [18] N. Yoshikawa and T. Kato, J. Phys. D: Appl. Phys. **43**, 425403 (2010).
- 354 [19] F. L. Bakker, J. Flipse, A. Slachter, D. Wagenaar, and B. J. van Wees, Phys. Rev. Lett. **108**, 355 167602 (2012).
- 356 [20] Z. H. Zhang, Y. S. Gui, L. Fu, X. L. Fan, J. W. Cao, D. S. Xue, P. P. Freitas, D. Houssameddine,
- 357 S. Hemour, K. Wu, and C.-M. Hu, Phys. Rev. Lett. **109**, 037206 (2012).
- 358 [21] T. An, V. I. Vasyuchka, K. Uchida, A. V. Chumak, K. Yamaguchi, K. Harii, J. Ohe, M. B.
- 359 Jungfleisch, Y. Kajiwara, H. Adachi, B. Hillebrands, S. Maekawa, and E. Saitoh, Nat. Mater. **12**, 549 360 (2013).
- 361 [22] K. Yamanoi, Y. Yokotani, and T. Kimura, Appl. Phys. Lett. **107**, 182410 (2015).
- 362 [23] T. Miyasato, N. Abe, T. Fujii, A. Asamitsu, S. Onoda, Y. Onose, N. Nagaosa, and Y. Tokura,
- 363 Phys. Rev. Lett. **99**, 086602 (2007).
- 364 [24] K. Uchida, T. Kikkawa, T. Seki, T. Oyake, J. Shiomi, Z. Y. Qiu, K. Takanashi, and E. Saitoh,
- 365 Phys. Rev. B **92**, 094414 (2015).
- 366 [25] Z. H. Duan, B. F. Miao, L. Suni, D. Wu, J. Du, and H. F. Ding, Ieee Magn Lett **10**, 4501805 367 (2019).
- 368 [26] A. Slachter, F. L. Bakker, J. P. Adam, and B. J. van Wees, Nat. Phys. **6**, 879 (2010).
- 369 [27] Y. Huo, F. L. Zeng, C. Zhou, and Y. Z. Wu, Phys. Rev. Appl. **8**, 014022 (2017).
- 370 [28] P. Noel, M. Cosset-Cheneau, V. Haspot, V. Maurel, C. Lombard, M. Bibes, A. Barthelemy, L.
- 371 Vila, and J. P. Attane, J. Appl. Phys. **127**, 163907 (2020).
- 372 [29] M. Beens, J. P. Heremans, Y. Tserkovnyak, and R. A. Duine, J. Phys. D: Appl. Phys. **51**, 394002 373 (2018).
- 374 [30] R. Iguchi, A. Yagmur, Y. C. Lau, S. Daimon, E. Saitoh, M. Hayashi, and K. Uchida, Phys. Rev. B 375 **98**, 014402 (2018).
- 376 [31] W. W. Lin and C. L. Chien, arXiv :1804.01392.
- 377 [32] X. D. Tao, Q. Liu, B. F. Miao, R. Yu, Z. Feng, L. Sun, B. You, J. Du, K. Chen, S. F. Zhang, L.
- 378 Zhang, Z. Yuan, D. Wu, and H. F. Ding, Sci. Adv. **4**, eaat1670 (2018).
- 379 [33] X. F. Zhu, M. Harder, J. Tayler, A. Wirthmann, B. Zhang, W. Lu, Y. S. Gui, and C. M. Hu, Phys. 380 Rev. B **83**, 140402 (2011).
- 381 [34] N. Mecking, Y. S. Gui, and C.-M. Hu, Phys. Rev. B **76**, 224430 (2007).
- 382 [35] H. C. Chang, P. Li, W. Zhang, T. Liu, A. Hoffmann, L. J. Deng, and M. Z. Wu, IEEE Magn. Lett. 383 **5**, 6700104 (2014).
- 384 [36] Y. T. Chen, S. Takahashi, H. Nakayama, M. Althammer, S. T. B. Goennenwein, E. Saitoh, and G. 385 E. W. Bauer, Phys. Rev. B **87**, 144411 (2013).
- 386 [37] H. Nakayama, M. Althammer, Y. T. Chen, K. Uchida, Y. Kajiwara, D. Kikuchi, T. Ohtani, S.
- 387 Geprägs, M. Opel, S. Takahashi, R. Gross, G. E. W. Bauer, S. T. B. Goennenwein, and E. Saitoh, 388 Phys. Rev. Lett. **110**, 206601 (2013).
- 389 [38]J. C. Rojas-Sanchez, N. Reyren, P. Laczkowski, W. Savero, J. P. Attane, C. Deranlot, M. Jamet, J.
- 390 M. George, L. Vila, and H. Jaffres, Phys. Rev. Lett. **112**, 106602 (2014).
- 391 [39] C. Hahn, G. de Loubens, O. Klein, M. Viret, V. V. Naletov, and J. Ben Youssef, Phys. Rev. B **87**, 392 174417 (2013).
- 393 [40] L. H. Bai, M. Harder, Y. P. Chen, X. Fan, J. Q. Xiao, and C.-M. Hu, Phys. Rev. Lett. **114**, 227201 394 (2015).
- 395 [41] Y.-P. Wang, J. W. Rao, Y. Yang, P.-C. Xu, Y. S. Gui, B. M. Yao, J. Q. You, and C.-M. Hu, Phys.
- 396 Rev. Lett. **123**, 127202 (2019).
- 397 [42]Y.-M. Kang, S.-H. Wee, S.-I. Baik, S.-G. Min, S.-C. Yu, S.-H. Moon, Y.-W. Kim, and S.-I. Yoo, J.
- 398 Appl. Phys. **97**, 10A319 (2005).
- 399 [43] Y. M. Lu, Y. Choi, C. M. Ortega, X. M. Cheng, J. W. Cai, S. Y. Huang, L. Sun, and C. L. Chien,
- 400 Phys. Rev. Lett. **110**, 147207 (2013).
- 401 [44] T. Kikkawa, K. Uchida, Y. Shiomi, Z. Qiu, D. Hou, D. Tian, H. Nakayama, X. F. Jin, and E.
- 402 Saitoh, Phys. Rev. Lett. **110**, 067207 (2013).
- 403 [45] Y. S. Gui, N. Mecking, A. Wirthmann, L. H. Bai, and C. M. Hu, Appl. Phys. Lett. **91**, 082503 404 (2007).
- 405 [46] N. Vlietstra, B. J. van Wees, and F. K. Dejene, Phys. Rev. B **94**, 035407 (2016).
- 406 [47]M. Agrawal, V. I. Vasyuchka, A. A. Serga, A. Kirihara, P. Pirro, T. Langner, M. B. Jungfleisch, A.
- 407 V. Chumak, E. T. Papaioannou, and B. Hillebrands, Phys. Rev. B **89**, 224414 (2014).
- 408 [48] M. Schreier, F. Kramer, H. Huebl, S. Geprägs, R. Gross, S. T. B. Goennenwein, T. Noack, T.
- 409 Langner, A. A. Serga, B. Hillebrands, and V. I. Vasyuchka, Phys. Rev. B **93**, 224430 (2016).
- 410 [49] L. Q. Liu, T. Moriyama, D. C. Ralph, and R. A. Buhrman, Phys. Rev. Lett. **106**, 036601 (2011).
- 411 [50] L. Q. Liu, C. F. Pai, Y. Li, H. W. Tseng, D. C. Ralph, and R. A. Buhrman, Science **336**, 555
- 412 (2012).
- 413