Longitudinal spin Seebeck effect contribution in transverse spin Seebeck effect experiments in Pt/YIG and Pt/NFO

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The spin Seebeck effect, the generation of a spin current by a temperature gradient, has attracted great attention, but the interplay over a millimetre range along a thin ferromagnetic film as well as unintended side effects which hinder an unambiguous detection have evoked controversial discussions. Here, we investigate the inverse spin Hall voltage of a 10 nm thin Pt strip deposited on the magnetic insulators Y3Fe5O12 and NiFe2O4 with a temperature gradient in the film plane. We show characteristics typical of the spin Seebeck effect, although we do not observe the most striking features of the transverse spin Seebeck effect. Instead, we attribute the observed voltages to the longitudinal spin Seebeck effect generated by a contact tip induced parasitic out-of-plane temperature gradient, which depends on material, diameter and temperature of the tip.
Spin caloritronics is an active branch in spintronics\textsuperscript{1,2}. The interplay between heat, charge and spin transport opens a new area of fascinating issues involving the use of waste heat in electronic devices. One potentially useful effect for heat collecting\textsuperscript{2} is the spin Seebeck effect (SSE)\textsuperscript{3} which was observed in 2008. It was reported that a spin current perpendicular to an applied temperature gradient can be generated in a ferromagnetic metal (FMM) by the transverse SSE (TSSE)\textsuperscript{4}. An adjacent normal metal (NM) converts the spin current via the inverse spin Hall effect (ISHE)\textsuperscript{5} into a transverse voltage, which is antisymmetric with respect to the external magnetic field \(H\). For pure \(\nabla T_z\) in a ferromagnetic or ferrimagnetic insulator (FMI) there is no PNE, since there are no free charge carriers available. However, if the NM material is close to the Stoner criterion, a static magnetic proximity effect could induce a so-called proximity PNE, which in general is present in spin polarized NM adjacent to a FMM and could also occur in a NM–FMI contact (Fig. 1a). Reports of previous experiments with pure \(\nabla T_z\) can be found for NM/FMM\textsuperscript{6,8} and for NM/FMI\textsuperscript{9}.

For the longitudinal SSE (LSSE)\textsuperscript{10} the spin current flows directly from the FM into an adjacent NM parallel to the temperature gradient (Fig. 1b), which is typically aligned out-of-plane \((\nabla T_x)\). In NM/FMM bilayers the anomalous Nernst effect (ANE) can occur, but is absent in the FMI. In semiconducting materials the ANE contributes to the LSSE as already shown for Pt/\(\text{NiFe}_2\text{O}_4\) at room temperature\textsuperscript{11}. In addition, if the NM would be spin polarized by the proximity to the FM, an additional proximity ANE could occur\textsuperscript{12} (Fig. 1b). Reports of previous experiments with pure \(\nabla T_z\) can be found for NM/FMM\textsuperscript{14} and for NM/FMI\textsuperscript{10,13–19}. As summarized in Fig. 1c an unintended \(\nabla T_x\) can hamper the evaluation of TSSE experiments with applied \(\nabla T_z\). Heat flow into the surrounding area or through the electrical contacts can induce an additional ANE in NM/FMM bilayers and NM/magnetic semiconductors as discussed in literature\textsuperscript{6,20–24}. But since, in principle, all the effects of an LSSE experiment can be present in the TSSE experiment with unintended \(\nabla T_x\), proximity Nernst effects and especially parasitic LSSE can also be present in NM/FMI bilayers as already mentioned recently\textsuperscript{25}. This leads to four possible effects which are antisymmetric with respect to the external magnetic field, when the temperature gradient is not controlled very carefully (see Fig. 1c). The prevalence of multiple thermoelectric effects in conducting ferromagnets poses a significant challenge for identifying any individual contribution to the total voltage signal. Previous reports on this matter\textsuperscript{6,8,21–23} are thus limited in significance and could not unambiguously prove or disprove the existence of a TSSE.

Here we report on the investigation of the TSSE on the magnetic insulators \(\text{Y}_3\text{Fe}_5\text{O}_{12}\) (YIG) and \(\text{NiFe}_2\text{O}_4\) (NFO). We observe the influence of out-of-plane temperature gradients when an in-plane temperature gradient is applied intentionally by varying the material, diameter and temperature of the electrical contacts. In NM/FMM systems a PNE due to magnetic anisotropy in the FMM or a proximity PNE due to the spin polarization in the NM can exist. In NM/FMI a PNE is absent due to the lack of charge carriers in the FMI. In NM/FMM systems the ANE exists. In both NM/FMM and NM/FMI the proximity ANE can appear if the magnetic proximity effect generates a spin polarization at the interface. The LSSE is proved for NM/FMI systems and should also be present in NM/FMM systems. A combination of all effects from a and b has to be considered.
contact tips. We circumvent all of the above issues and are, for the first time, able to demonstrate that the LSSE alone accounts for the observed voltage signals. Thus, we fill a gap in the controversial discussion about TSSE investigations.

Results
Sample structure and characterization. Magnetic insulators are of increasing importance for spintronics, magnonics and spin caloritronics. The phenomena presented in Fig. 1 and the discussions of side effects in TSSE experiments have not been treated systematically in the literature for NM/FMI bilayers so far (Table 1). For the measurements in this work we used YIG films with a thickness of $t_{\text{YIG}} = 180$ nm. The films show a coercive field of about 100 Oe and a saturation magnetization of $M_s = 120$ kA m$^{-1}$. As common for many thin films these values deviate slightly from bulk properties. Previous studies demonstrated, however, that the LSSE is very robust to variations in the magnetic properties. Our results should thus also be applicable to bulk samples and other thin films. YIG films with similar properties are also relevant for further spin transport phenomena, for example, the recently observed spin Hall magnetoresistance. For reference measurements on another magnetic insulator system we used NFO films with a thickness of $t_{\text{NFO}} = 1$ μm and a coercive field of about 160 Oe. On top of the magnetic insulator systems a thin Pt strip with a thickness of $t_{\text{Pt}} = 10$ nm was deposited on one side as a spin detector material.

Experimental setup. If not mentioned otherwise the experiments are performed under ambient conditions as described in (ref. 8) mirroring those of the very first TSSE measurements on FMIs by Uchida et al.9. The ends of the Pt strip ($l_{\text{Pt}} = 5$ mm) were contacted with a microprobe system with Au and W tips of different diameters. Furthermore, one Au tip was equipped with a 1.5-kΩ resistor for heating the tip (only thermally connected to the tip, not electrically). Intentionally induce a $\nabla T_z$ (Fig. 2a)8. Figure 2b shows the nearly linear relation between the power $P_{\text{needle}}$ dissipated in the resistor and the tip temperature $T_{\text{needle}}$ as determined with a type-K thermocouple glued to the tip in a calibration measurement. The heated Au needle is labelled $T_{\text{needle}} = RT_x + x$ with room temperature $RT_x = 296$ K. The voltage $V$ at the Pt strip was measured with a Keithley 2182A nanovoltmeter. An external magnetic field $H$ in $x$ direction was applied in a range of ±600 Oe for YIG and of ±1,000 Oe for NFO films. The voltage values $V_{\text{sat}}$ quoted throughout the remainder of the manuscript are the respective averaged voltages levels at saturation.

Measurements in vacuum. Before measurements under ambient conditions were performed we did reference measurements on Pt/YIG under vacuum. The Pt strip was contacted by Au bonding wires with 25 μm in diameter. The results are shown in Fig. 3 for Pt strip on the hot side (Fig. 3a) and Pt strip on the cold side of the YIG film (Fig. 3b) for various in-plane temperature gradients $\nabla T_x$. For both sample sides no significant effect within the measurement sensitivity limit below ±20 nV can be observed. The same behaviour was obtained for Pt/NFO where no significant effect could be observed for Pt strip on both sample ends (Supplementary Fig. 1).

Measurements under ambient conditions. Afterwards, the samples were mounted in the setup described in (ref. 8) performed under ambient conditions. First, the ISHE voltage from the Pt/YIG sample was measured for various $\Delta T_{\text{z}}$. The Pt strip was on the hot side and contacted with W tips carefully. Again, the voltage shows no significant variation within the sensitivity limit of about ±40 nV when $H$ is varied (Fig. 4a). No Nernst effects are observed due to the insulating magnetic layer and no evidence for an additional $\nabla T_z$ can be detected. Therefore, the clamping and heating of the sample in our setup and the contacting with thin W tips results in a pure $\nabla T_z$ as already shown in (ref. 8). TSSE is not observable although the Pt strip is located on the hot side of the YIG film and far away from the center where the TSSE should vanish. The comparison with Pt strip at the cold sample end is shown in Fig. 4b for the thinnest W tips and different temperature gradients $\nabla T_x$. For the largest $\nabla T_x$ we can observe a noticeable effect given by a difference between the voltages in saturation for positive and negative magnetic fields.

Contact area dependence. In the next step we contacted again and pushed the thin W tips with more pressure into the Pt strip, which increases the effective electrical contact area $A$ (blue curve in Fig. 4c). Afterwards, the tips were changed by other W tips with a larger contact area. The observed voltages are plotted in Fig. 4c with Pt strip at the hot sample end and $\Delta T_z = 15$ K. The voltages vary significantly when $H$ is changed. $V$ is antisymmetric
with respect to $H$ and the voltage in saturation increases for larger contact areas. In Fig. 4d the same increase of antisymmetric effect is obtained for different W tips with increasing contact area at the cold sample side, but the magnitude of effect is generally smaller compared with the hot side. However, the sign of $V$ in saturation is the same for Pt strip at the hot and the cold side. This behaviour could be verified for Pt/NFO films (Supplementary Fig. 2) and excludes the TSSE which has to show a sign change in $V$ between hot and cold side (Supplementary Note 1).

In Fig. 5 the magnitudes $V_{\text{sat}}$ obtained for all W tips with different contact areas (Fig. 4) are plotted against the contact area $A$ of the tip for Pt strip on the hot and cold side for a temperature difference $\Delta T_x = 15$ K. Furthermore, a variation of Au tips with different contact areas were added which show the same behaviour of $V_{\text{sat}}$ depending on $A$ for both sample sides (Supplementary Fig. 3). Due to the fact that the real attached contact area could not be reproduced we estimated the average between the possible maximum contact area and a sufficiently low area (Supplementary Fig. 4). This leads to relatively large error bars. However, for Pt strip on the hot and the cold side the sign of $V_{\text{sat}}$ is the same. Furthermore, the absolute value of $V_{\text{sat}}$ decreases for smaller contact areas of the used tips for both materials (W and Au). The effect size for Pt strip on the cold side is generally smaller compared with the hot side when the same temperature difference of $\Delta T_x = 15$ K is applied. We explain this behaviour of $V_{\text{sat}}$ by an unintended heat flux through the tips leading to a vertical temperature gradient $\nabla T_z$ and, therefore, to a LSSE induced spin current into the Pt.

Furthermore, in Fig. 5, $V_{\text{sat}}$ does not tend to zero when $A$ gets sufficiently small, which can be explained by an additional influence of $\nabla T_z$ contributions due to different thermal conductivities of the investigated film and substrate materials.

**Influence of contact tip heating.** In the next step we used the thickest Au tips (Fig. 6) with a resistor glued to the tip. The contact area $A$ was about 0.28 mm². It can be seen that the measured voltage is antisymmetric with respect to the magnetic field (Fig. 6a–d). Next, we applied a voltage to the Au tip resistor to increase the temperature of the tip and change the out-of-plane heat flow.

In Fig. 6a for $T_{\text{needle}} = RT$ a small antisymmetric effect of about $V_{\text{sat}} = -50$ nV is obtained when the Pt strip is at the cold side of the YIG film. When the needle is heated to $T_{\text{needle}} = RT + 12$ K
at the hot side for the data in various T its sign with increasing W tips or Au bonding wires generate an additional out-of-plane The Au needles with larger contact areas compared with thinner T increase further to D when the Au tip is heated on the hot sample side the amount of heating power is larger to reverse the direction of the out-of-plane temperature gradient than on the cold sample side. Therefore, the slope of the curves in Fig. 7 for Au tip at the cold sample side is larger compared with the slope of the curves for Au tip at the hot sample side.

The ISHE voltage changes its sign at a value of $V_{sat} = +95 \text{nV}$ and changes further to $V_{sat} = +590 \text{nV}$ for $T_{needle} = RT + 24 \text{ K}$. The Au needles with larger contact areas compared with thinner W tips or Au bonding wires generate an additional out-of-plane heat flow across the YIG film, for which the experimentally measured $V_{sat}$ describes the deviation between high and low temperature. When the Au tip heating compensates the out-of-plane heat flux induced by the ISHE voltage, it changes its sign to a value of $V_{sat} = -170 \text{nV}$ for $T_{needle} = RT$ and therefore three times larger than for $T_{RT} = 10 \text{ K}$. The ISHE voltage again increases with increasing $T_{needle}$ and changes sign.

For a Pt strip at the hot side $V_{sat}$ without tip heating is larger than at the cold side. For $T_{RT} = 5 \text{ K}$ the magnitude is about $V_{sat} = -130 \text{nV}$ (Fig. 6c) and can be decreased to $V_{sat} = -300 \text{nV}$ for $T_{RT} = 10 \text{ K}$ (Fig. 6d). The sign and the magnitude of $V_{sat}$ can also be controlled by $T_{needle}$ and, therefore, by $\nabla T_x$. When $T_{needle}$ is fixed at $RT + 31 \text{ K}$, $V_{sat}$ is about +180 nV for $T_{RT} = 5 \text{ K}$ and +90 nV for $T_{RT} = 10 \text{ K}$.

$V_{sat}$ measured for Pt/YIG at the hot and cold side is plotted as a function of $H$ in Fig. 6b) the ISHE voltage at the Pt is $V_{sat} = -120 \text{nV}$ (Fig. 6c) and can be decreased to $300 \text{nV}$ (Fig. 6d) the corresponding $\Delta T_{RT}$ is $5 \text{ K}$ and +90 nV for $T_{RT} = 10 \text{ K}$. We simulate the phonon and magnon temperatures assuming one-dimensional transport in our films and disregard the influence of thermal contact resistance other than the coupling between magnons and electrons. This yields a value $\Delta T_p$ the phonon temperature drop across the YIG film, for which the experimentally measured $V_{sat}$ is obtained due to the LSSE. We assume $\rho_p = 40 \mu\Omega \text{m}$.

We calculated $\Delta T_{me}$ for the largest and smallest $V_{sat}$ taken from Fig. 7 at RT (red curves in Fig. 6a,d) and the corresponding $\Delta T_p$. For $|V_{sat}| = 50 \text{nV}$ (Fig. 6a) we obtain $\Delta T_{me} = 0.5 \mu\text{K}$ and a corresponding $\Delta T_p = 2 \text{ mK}$. For $|V_{sat}| = 300 \text{nV}$ (Fig. 6d) the corresponding values are $\Delta T_{me} = 3.2 \mu\text{K}$ and $\Delta T_p = 12 \text{ mK}$. The obtained $\Delta T_p$ are in the order of a few millikelvins. It is reasonable to assume that such values can be induced by for example, thick contact tips, especially considering that our initial simplifications should lead to an overestimation of $\Delta T_p$. These calculations for the TSSE configuration were investigated in detail in (ref. 25). It was found that for $T_{RT} = 20 \text{ K}$ the obtained $\Delta T_{me}$ is well below 1 $\mu\text{K}$, even at the very edge of the sample where $\Delta T_{me}$ is maximized. This further supports the notion that spurious out-of-plane temperature gradients are responsible for the voltages observed in our samples.

Figure 7 | Influence of contact tip heating. $V_{sat}$ as a function of the Au needle temperature $T_{needle}$ for various in-plane temperature differences $\Delta T_x$ in Pt/YIG and Pt strip on the hot (red triangles, yellow squares) and on the cold sample side (green circles, blue rhombuses). The experimental error of $T_{needle}$ was estimated in the reference measurement and describes the deviation from the average value over several measurements. The experimental error of $V_{sat}$ describes the deviation between high and low contact forces.

The ISHE voltage changes its sign to a value of $V_{sat} = +95 \text{nV}$ and changes further to $V_{sat} = +590 \text{nV}$ for $T_{needle} = RT + 24 \text{ K}$. For a heated Au needle $V_{sat}$ increases and crosses zero (Fig. 7). A non-heated Au tip, which is at room temperature, is larger compared with the temperature difference between the colder sample side, which is closer to room temperature, and the non-heated Au tip, which is at room temperature.
Measurements on Pt/NFO. For Pt/NFO a similar behaviour of \( V_{\text{sat}} \) could be observed for various combinations of in-plane temperature gradient, Au needle temperature and Pt strip location (hot or cold side) (Supplementary Fig. 5, Supplementary Note 2). The magnitudes of \( V_{\text{sat}} \) obtained for Pt/NFO are about an order of magnitude larger when compared with \( V_{\text{sat}} \) in Pt/YIG (see Supplementary Fig. 2 and Supplementary Fig. 5) for similar values \( \Delta T_\text{r} \) and \( T_\text{needle} \). This difference can be explained by different film thicknesses and thermal conductivities of NFO and YIG as well as by different spin mixing conductances\(^{36,37}\).

**Discussion**

For all experiments we performed in vacuum and using thin bonding wires for the electrical contacts, we could not observe any evidence for a TSSE as well as any other transport phenomena shown in Fig. 1. For the most experiments we performed under ambient conditions and using contact tips, we can clearly observe an antisymmetric behaviour of the voltage \( V \) with respect to the external magnetic field \( H \) which can be addressed to the LSSE due to an unintended out-of-plane temperature gradient \( \Delta T_z \). For all remaining experiments, there is no evidence for any transport phenomena given in Fig. 1. For both investigated sample systems the spin mixing conductance is large enough to observe a thermally driven spin current across the NM/FMI interface as proven by the results of experiments with LSSE configuration for Pt/YIG (Supplementary Fig. 6) and Pt/NFO\(^{11}\), respectively.

In addition to the LSSE, we now discuss other parasitic effects like the ANE and proximity ANE, which can be produced by an unintended \( \Delta T_z \) (see Fig. 1). We can exclude an ANE for YIG due to the lack of charge carriers. For NFO we observed an ANE which is one order of magnitude smaller at RT than the LSSE\(^{11}\). This ANE can be explained by the weak conductance of NFO at RT due to thermal activation energies of a few hundred meV depending on the preparation technique\(^{11,29}\). Absence of proximity ANE in Pt/NFO is ensured by X-ray resonant magnetic reflectivity measurements finding no interface spin polarization with the experimental limit of 0.02\( \mu_\text{B} \) per Pt atom\(^{38}\).

In case of Pt/YIG Geprägs et al. presented X-ray magnetic circular dichroism measurements (XMCD) with no evidence for any spin polarization in Pt\(^{39}\) while Lu et al. could show XMCD measurements indicating magnetic moments in Pt on their YIG samples\(^{40}\). Future investigations with X-ray resonant magnetic reflectivity can give more insight to this discrepancy. However, Kikkawa et al.\(^{41}\) could show that a potential contribution of a proximity ANE additional to the LSSE is negligibly small. This supports our conclusion that the main antisymmetric contribution in our measurements on both Pt/YIG and Pt/NFO is the LSSE, which is driven by an out-of-plane temperature gradient.

We do not observe any symmetric contribution for \( \nabla T_z \) without tip heating. Therefore, PNE and proximity PNE contributions can also be excluded. Nevertheless, we find a small symmetric contribution for strong tip heating as demonstrated in Fig. 6b for \( \Delta T_\text{needle} = RT + 31 \text{K} \). In the region of \( H_z \) small peaks are visible under symmetrization of the voltage. This hints at the existence of an additional magnetothermopower effect potentially induced by a temperature gradient \( \nabla T_z \) along the Pt strip and will be part of future investigations.

Recently, Wegrowe et al.\(^{42}\) used anisotropic heat transport as an interpretation for the measured voltages using in-plane temperature gradients. In their work, they derived the anisotropic field-dependent temperature gradient in FMM and FMI from the Onsager reciprocity relations. Therefore, the thermocouple effect between the FM, the NM and the contacting tips can generate field-dependent voltages if there is a difference in the Seebeck coefficients. In our investigated systems, the Seebeck coefficients are indeed different for FM, Pt and the contact tips. However, since we do not observe a significant field-dependent variation of the ISHE voltage when the samples are bonded or carefully contacted with W tips, any anisotropic field-dependent heat-transport can be excluded as the reason for the observed voltages.

In summary, we investigated the relevance of TSSE in Pt/YIG and Pt/NFO systems. We found no significant ISHE voltages upon applying an in-plane temperature gradient and using 25-\( \mu \text{m} \) thin Au bonding wires or sharp W tips (0.003-mm\(^2\) contact area) as electric contacts. Increasing the contact area (up to 0.28 mm\(^2\)), however, induces an additional out-of-plane heat flux accompanied by a LSSE ISHE voltage. This antisymmetric effect can be identified as LSSE which was verified by controlling the needle temperature, or changing the tip diameter accompanied by the contact area and therefore varying the out-of-plane temperature gradient. Taken together, in all our experiments, we thus only observe LSSE-type signatures. These LSSE voltages can be considered a remnant of a TSSE-type response if an unintentional (or intentional) \( \nabla T_z \) is present. This shows that utmost care is required if one is to interpret magnetothermopower effects in terms of the TSSE.

**Methods**

**Sample fabrication.** The YIG films were deposited on gadolinium gallium garnet (Gd\(_3\)Ga\(_5\)O\(_{12}\)) (111)-oriented single crystal substrates with width and length \( w = 1 \times 5 \text{mm} \) by pulsed laser deposition from a stoichiometric polycrystalline target. The NFO films with a thickness of about \( t_{\text{NFO}} = 1 \mu\text{m} \) were deposited on \( 10 \times 5 \text{mm}^2 \) MgAl\(_2\)O\(_4\) (100)-oriented substrates by direct liquid injection-chemical vapour deposition (DLJ-CVD)\(^{11,43}\). After a vacuum break and cleaning with ethanol in an ultrasonic bath a \( t_{\text{Pt}} = 10 \text{nm} \) thin Pt strip was deposited by dc magnetron sputtering in an Ar atmosphere of 1.5 \( \times 10^{-3} \text{ mbar} \) through a 100-\( \mu \text{m} \) wide split-mask on one sample side of the YIG and NFO films with a length of \( t_{\text{Pt}} = 5 \text{mm} \).

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Author contributions
D.M. designed the experiment with the input of M.Schm., J.-M.S., T.K., G.R. and analyzed the data; D.M. and M.A. prepared and characterized the samples in collaboration with A.G. and S.T.B.G.; D.M. and M.Schr. carried out the calculations; D.M. and T.K. wrote the manuscript with the input of all authors; T.K. conceived and sketched the tables of effects in figure 1 with the input of D.M., C.H.B. and G.R.; all authors discussed the results and the manuscript.

Additional information
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