Reply to “Comment on ‘Optical detection of transverse spin-Seebeck effect in permalloy film using Sagnac interferometer microscopy’ ”

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Reply to "Comment on 'Optical detection of transverse spin-Seebeck effect in permalloy film using Sagnac interferometer microscopy' "

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Abstract

In the Comment on our original publication [Phys. Rev. B 95, 180401(R)], Kimling *et al* claimed that our optically detected change of Kerr rotation angle in a ferromagnetic slab subjected to a lateral temperature gradient measured by magneto-optical Kerr effect (MOKE) is not, or not only caused by the transvers spin-Seebeck effect. In this Reply, we experimentally confirm the conclusion published in the original paper by measuring MOKE in a striped patterned NiFe slab, thereby refuting the criticism in the Comment.

In their Comment [1], Kimling and Kuschel (hereafter the 'commenters') challenge our original interpretation of the magneto-optical Kerr effect (MOKE) measurements using ultrasensitive Sagnac interferometer [2], claiming that the transverse spin-Seebeck effect (TSSE) is not the only contribution to the measured change in the Kerr rotation angle from a $Ni_{80}Fe_{20}$ (NiFe) ferromagnetic (FM) slab subjected to a lateral temperature gradient in the presence of a magnetic field. The authors assert that: (1) the TSSE, in general, has not been completely proven so far, and that the existing theories, in particular the 'phonon magnon drag' model that we have used in our publication, in fact cannot explain the original work of Uchida *et al* [3]. (2) The commenters based their critique on an estimate of the magnitude of the measured effect, arguing that to observe the TSSE that we originally claimed, the temperature gradient in our measurements should have been much larger than the value we measured.

In this Reply, firstly, for the reader sake, we summarize previous literature reports on the TSSE response in FM metallic systems. Secondly, we dispute the estimate of the necessary large temperature gradient made by the commenters. Importantly, the Kerr effect sensitivity to spin accumulation (via the TSSE response) has not been recognized by the commenters; in fact it was mistakenly assumed that the Kerr effect sensitivity to the spin accumulation by the TSSE is the same as the Kerr sensitivity to the bulk magnetization change due to the temperature change. Thirdly, the spin-dependent Seebeck model used by the commenters is inappropriate for interpreting our experimental results. In addition, we have engineered an experiment which shows that the TSSE is the only viable interpretation of our original measurements, and that the 'phonon-magnon drag model' is indeed capable of explaining our results.

1. Introduction

The elusive phonon-mediated transverse spin Seebeck effect (TSSE) in FM slabs has been previously supported by the so-called 'patterned configuration' [4,5], where the FM slab is divided into thin, isolated stripes. In this case, the spatially distributed SSE may still exist, and, due to the stripe isolation, without any interference of other spurious artefacts such as the spin-dependent Seebeck effect (SDSE). Such measurements were performed before in NiFe [4] and GaMnAs [5] using electrical-based detection, namely via the inverse spin Hall effect (ISHE) in a nonmagnetic overlayer with large spin-orbit coupling, namely Pt. This type of measurement has been considered by many to be a direct evidence of TSSE response, since only TSSE would be generated by the long-range phonons propagation through the substrate via the magnon-phonon coupling (i.e., 'phonon-magnon drag model'). In this Reply (section 3), we show (see below) the continuous-wave (CW) optical detection version of the 'patterned configuration' for the TSSE measurements in the FM metal NiFe stripes, where each FM stripe is separated across the lateral direction of the substrate. Using the Sagnac MOKE optical detection, the patterned configuration provides an artefact-free evidence for the TSSE existence in NiFe ferromagnetic films [6].

We note, however that the TSSE response measured via electrical-based ISHE detection is still under intense debate, due to the possible interference from various thermoelectric and magneto-thermoelectric artifacts. Specifically when the ISHE is used to measure the TSSE in a FM slab, the additional top contacts might influence the thermal conductivity of the system, and consequently induce an unintentional vertical temperature gradient along the FM/Pt interface that leads to overwhelming artefact signals arising from the magneto-thermoelectrics effect. *This is an unfortunate disadvantage of the electrical detection measurement.* Thus all arguments against the existence of TSSE response in FM slabs have been based on the same type of electrical measurement, namely via ISHE. This suggests that a 'cleaner' and artefact-free detection scheme is required to provide an alternative route of proving and studying the TSSE response. In this respect our optical detection scheme excludes all of the electrical-related artefacts. We thus believe that the TSSE response may be subtle when trying to measure it via an electrical detection scheme, but it might be clearly observable via magneto-optical detection. **This has been the main purpose of our optical measurements in the original paper**. We also note that similar magneto-optical approach has been successfully used to detect the spin accumulation via spin Hall effect in both Pt and W thin films [7].

2. Comparison of Kerr sensitivity to spin accumulation via TSSE and bulk magnetization via 'thermal effect'

The commenters claimed that for observing the TSSE response via magneto-optic measurements the temperature gradient in the FM slab should have been much larger than the measured value. One key parameter used in the commenters' equation to estimate the temperature gradient is the Kerr sensitivity to spin accumulation via TSSE. For this estimate the commenters used an incorrect sensitivity parameter that was taken from the original S.I. figures (Figs. S7-S9) which in fact characterizes the Kerr sensitivity parameter to the reduction of magnetization due to the temperature change upon uniform heating, rather than the Kerr sensitivity to spin accumulation on the FM slab surface. These are **two different types** of Kerr sensitivity, which are orders of magnitude different, and thus should not be confused.

Therefore, when a **small temperature gradient** is applied to the FM slab, there are two different contributions to the change of the Kerr signal along the slab direction (x). These are: (a); heating induced decrease in the FM magnetization, which exhibits a progressive Kerr angle reduction along the slab direction (x), as expected. (b); spin accumulation via TSSE which induces an opposite Kerr signal at each end of the slab. Here the Kerr signal increases at the left end but decreases at the right end, with the same amplitude. Consequently, the measured Kerr signal in the actual experiment is composed of the two Kerr response contributions (i.e. due to heat and TSSE). In this case the contribution to the Kerr signal due to heat may be derived from the asymmetry in the Kerr signals at the two ends of the FM slab, which is less than 5% at low temperature gradient (see below) similar as in our original paper [2].

At large applied temperature gradient, the temperature of the left end can no longer be maintained at room temperature. This leads to a reduction of magnetization (and corresponding Kerr signal) at both ends of the slab. Meanwhile, the Kerr signal due to the TSSE response also increases. However by combining the two Kerr contributions, it is no longer possible to clearly distinguish between the Kerr response due to heating and that related to the TSSE. The Kerr sensitivity to the TSSE claimed by the commenters was probably extracted from this situation, and therefore is erroneous. We note that all the **optical TSSE coefficients extracted** in our original paper were only determined at the small-temperature gradient condition [2].

We now estimate the correct Kerr sensitivity to the heat and TSSE effects from our original data. For this purpose we analyze in more detail the dynamics and magnitude of the Kerr response at the left end, $\Delta\theta_L(t)$, and right end, $\Delta\theta_R(t)$ upon applying a temperature gradient, that were depicted in Fig. 2 of our original paper. We note that at small temperature gradient $\Delta\theta_R(t)$ saturation value is somewhat larger than that of $\Delta\theta_L(t)$. This can be readily explained if there is an additional contribution to $\Delta\theta_R(t)$ that is not related to that of TSSE. We identify the additional contribution to the dcrease in $\theta_R(t)$ as due to a 'thermal effect', as shown in Fig. 1 in our Reply, where the FM magnetization of the right end decreases when its temperature increases. A crude estimate shows that this thermal contribution to $\Delta\theta_R(t)$ is about 5% of the total TSSE-related $\Delta\theta$ at saturation; namely $[|\Delta\theta_R| - |\Delta\theta_L|]/[|\Delta\theta_R| + |\Delta\theta_L|]=5\%$. This shows that in NiFe the Kerr angle sensitivity associated with the TSSE is at least one order of magnitude larger than that associated with the change in magnetization due to the temperature increase, at the conditions set in the experiment, i.e. temperature difference, ΔT=2.0 K between the left and right ends of the FM slab. This argument, based on the experimental results in our original paper refutes the assumption made in the Comment that the TSSE sensitivity to MOKE is the same as that of the bulk magnetization. We speculate that the higher MOKE sensitivity to the TSSE is the accumulation of the excess spins at the upper surface of the NiFe film (or stripes) within the optical skin depth of the laser excitation beam, compare to the spin density reduction in the bulk of the FM film. This observation calls into question the exagerated estimate for the ΔT made in the Comment.

3. Optical detection of TSSE response in a striped patterned NiFe slab

The second assumption of the commenters in estimating the temperature gradient in the FM slab of our original experiment is the use of an equation taken from the spin-dependent Seebeck (SDSE). This is an inappropriate model for interpreting our experimental results. In the supplementary sections of our original paper (namely Figs. S10 to S12), we carefully checked all artifacts and excluded the contribution of SDSE. In order to further refute the commenters' claim we present here an additional experiment showing that TSSE is the only viable interpretation of our original measurements; it adds to the argument that the SDSE equation in the Comment is inapplicable to our case.

For the present MOKE measurements, we have utilized the same optical set up as in our original report [2]. In brief, we used a modified fiber Sagnac interferometer operating at 1.3 μm having a Kerr rotation angle resolution of ~20 nanoRadians (nRad) and spatial resolution of ~1 μm. For the TSSE measurements, the Sagnac interferometer was adapted into a two dimensional (2D) scanning confocal microscope with a diffraction-limited spot size of ~1 μm (see Fig. 1(a)). The samples investigated were NiFe stripes of 50 nm thick and planar dimension of 0.6mm × 2mm grown by e-beam deposition on clean silicon nitride (SiN)/Si substrate, which was capped by 100 nm SiO_2 overlayer to prevent oxidation. The sample was connected to two copper blocks with a separation of 5 mm of which temperatures were controlled using Peltier heaters. The temperature difference, ΔT, applied along the substrate x-direction, which is perpendicular to the NiFe stripes, was monitored by two thermocouples. The sample was exposed to an out-of-plane magnetic field (B ~ 800 mT) in a configuration suitable for probing the polar MOKE effect ($TSSE_{PM}$, $\boldsymbol{B}\perp\Delta\boldsymbol{T}$) as shown in Figure 1(a) [6].

For measuring the MOKE response of the NiFe stripes we fixed the Sagnac beam on the right or left NiFe stripes and recorded the transient response, $\Delta\theta_R(t)$ and $\Delta\theta_L(t)$, respectively, where t is

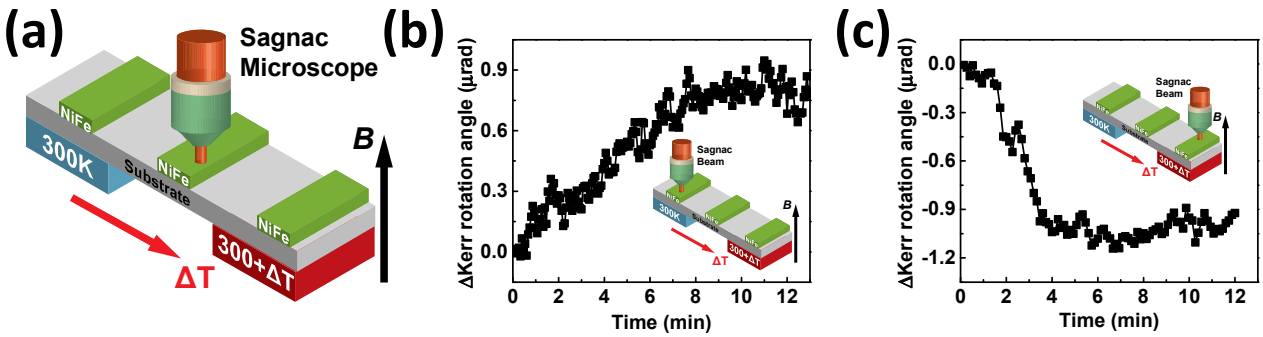

Figure 1. *(a) Schematic illustration of the Sagnac microscope in the polar configuration for measuring $TSSE_{PM}$ of NiFe stripes deposited on SiN substrate. (b) and (c) Time evolution of the TSSE-related Kerr rotation angle in the NiFe stripes located, respectively at the left and right NiFe stripes under a temperature difference of ΔT=3.2K and field of 800 mT The insets show schematically the measurement geometry where the position of the Sagnac MOKE microscope beam spot, temperature gradient direction, and orientation of the magnetic field are denoted.*

the time after the heater on the right stripe is turned on. Figures 1(b) and 2(c) show the change, $\Delta\theta$ of the Kerr rotation angle, θ measured from the NiFe stripe at each side of the SiN substrate, when subjected to a thermal gradient of 0.64 K/mm, that corresponds to a temperature difference ΔT=3.2 K between the opposite substrate sides. By monitoring the temperature increase measured by the thermocouples when the heating power is turned on at the right copper block, we conclude that the sample holder reaches thermal equilibrium in about 10 minutes. Importantly, when the right NiFe stripe was heated by a relatively small ΔT, its temperature, T_R **increases**; but the temperature, T_L of the left stripe remains approximately at room temperature (RT) as monitored by the thermocouple (see Fig. 1(a)).

It is clear from Figs. 1(b) and 1(c) that $\Delta\theta(t)$ reaches saturation roughly at the same time that the the temperature reaches equilibrium condition. Importantly we see that $\Delta\theta_R(t)<0$ as measured on the right stripe (Fig. 1(c)), but $\Delta\theta_L(t)>0$ on the left stripe (Fig. 1(b)), where the temperature is still close to RT. One may be tempted to explain $\Delta\theta_R(t)<0$ as due to the magnetization decrease that is caused by the temperature increase, ΔT of the right stripe. However the obtained $\Delta\theta_L(t)>0$ on the left stripe, even that its temperature is maintained at room temperature, cannot be explained by a similar argument, since T_L cannot *decrease below RT upon the heating the stripe*. We therefore conclude that a process other than a temperature increase dominates both $\Delta\theta_L(t)$ and $\Delta\theta_R(t)$. We may reasonably explain $\Delta\theta_L(t)>0$ as due to the TSSE process, where excess spin accumulation is generated at the unheated stripe by the SSE. The excess spins cannot be accumulated on the left stripe without spin depletion somewhere else along the slab, such as on the right stripe. This explains the existing similarity between the left and right stripe dynamics and saturation magnitudes. We therefore conclude that $\Delta\theta_R(t)<0$ response is also mainly due to TSSE and not due to the temperature increase, ΔT on the right stripe.

Our finding using the isolated stripes is similar to our original observation in a continuous NiFe film deposited on SiN substrate that is generated by the applied in-plane thermal gradient in a magnetic field. Except that here the TSSE is generated even when the NiFe stripes are isolated from each other on the SiN substrate. This behavior mimics the reported TSSE response in isolated NiFe stripes measured by electrical methods [4]. The TSSE response does not require physical connection between the NiFe stripes, since their magnetic response may couple to each other via the acoustic phonon flow in the substrate that is generated upon the application of the thermal gradient. The magnon-phonon coupling produces spin accumulation on the top surface of the individual NiFe stripes that is detected by the Sagnac MOKE.

In conclusion; our additional 'control experiment' refutes the arguments made in the Comment. We conclude that the MOKE sensitivity to spin accumulation on the NiFe surface vastly exceeds that of the magnetization variation in the NiFe bulk, and therefore is possible to measure TSSE by optical means. *We thus find the critique in the Comment unjustified.*

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