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P. Jiménez-Cavero,[1,2] I. Lucas,[1,2,a)] D. Bugallo,[3] C. López-Bueno,[3] R. Ramos,[3] P. A. Algarabel,[2,1] M. R. Ibarra,[1,2,4] F. Rivadulla,[3] and L. Morellón[1,2]
[1)] *Instituto de Nanociencia y Materiales de Aragón, Universidad de Zaragoza-CSIC, 50018 Zaragoza, Spain*
[2)] *Departamento de Física de la Materia Condensada, Universidad de Zaragoza, 50009 Zaragoza, Spain*
[3)] *Centro de Investigación en Química Biolóxica e Materiais Moleculares (CIQUS), Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain*
[4)] *Laboratorio de Microscopías Avanzadas, Universidad de Zaragoza, 50018 Zaragoza, Spain*

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We report the disentanglement of bulk and interfacial contributions to the thermally excited magnon spin current in the spin Seebeck effect under static heating. For this purpose, we have studied the dependence of the inverse Spin Hall voltage and the thermal conductivity on the magnetic layer thickness. Knowledge of these quantities allows us to take into account the influence of both sources of thermal spin current in the analysis of the voltage dependence. The magnetic layer thickness modulates the relative magnitude of the involved thermal drops for a fixed total thermal difference throughout the sample. In the end, we attain the separate contributions of both sources of thermal spin current —bulk and interfacial— and obtain the value of the thermal magnon accumulation length scale in maghemite, which we find to be 29(1) nm. According to our results, bulk magnon accumulation dominates the spin Seebeck effect in our studied range of thicknesses, but the interfacial component is by no means negligible.

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The spin Seebeck effect (SSE) is a complex phenomenon that lies at the intersection between several spintronics subfields. It is defined as the generation of a spin current in a magnetic material (FM) subjected to a thermal gradient, standing as one of the major spin-caloritronics topics[1]. However, the detection of the SSE makes use of tools from spin-orbitronics, since it is commonly achieved by spin current to charge current conversion by means of the inverse spin Hall effect (ISHE) observed in a nonmagnetic heavy metal (NM) adjacent to the magnetic material (FM)[2]. Furthermore, these thermal spin currents have been proved to be of magnonic origin, which means that magnonics are also involved in SSE[3,4]. This complexity affects both theoretical and experimental aspects and is evidenced, e.g., by the difficulties found in the definition of a standard SSE coefficient[5,6].

Part of this intricacy comes from the fact that there are different physical mechanisms contributing to the SSE[7]. Two main theories have been developed so far which describe two different sources for a magnon spin current in a FM/NM system subjected to a thermal gradient. The first one points to the temperature difference between the metal electrons and the magnetic magnons at the interface, such that the excited spin current is given by[4,8]:

$$J_s^i \propto \Delta T_i^{NM/FM}, \tag{1}$$

where $\Delta T_i^{NM/FM}$ denotes the interfacial thermal drop.

The second origin of the thermal spin current lies on the thermal gradient present in the bulk of the FM layer itself, rather than the temperature difference at the interface[9–12]. This gradient creates a magnon accumulation which acts as a spin potential for the spin current. In this case, the spin current is determined by a finite magnon propagation length scale (Λ_m) and follows the expression[10,12]:

$$J_s^b \propto \frac{\cosh(t_{FM}/\Lambda_m) - 1}{\sinh(t_{FM}/\Lambda_m)} \nabla T_{FM}, \tag{2}$$

where ∇T_{FM} is the temperature gradient across the FM layer and t_{FM} denotes its thickness.

We empashize that both mechanisms arise in the presence of a thermal gradient and thus meet the definition of the SSE, but they actually represent different physical sources for thermally excited magnon spin currents.

Nowadays the logitudinal spin Seebeck effect (LSSE) is widely used because of experimental simplicity, and most theoretical efforts have focused on it[13]. In the LSSE an out-of-plane thermal gradient is applied perpendicularly to a magnetic field. The excited spin current is parallel to the thermal gradient and according to the ISHE phenomenology, the generated voltage can be measured in the transverse direction (perpendicular to both thermal gradient and magnetic field).

Whilst time-resolved optical approaches of SSE measurements have been able to selectively excite only interfacial LSSE (*i*LSSE)[14,15] and the *b*LSSE has been unambiguously detected using a magnon-valve structure[16], the relative contributions in a standard DC measurement in which the whole sample is subjected to a thermal gradient have not been addressed. In these conditions, both thermal spin current sources coexist and contribute to the signal. Notwithstanding, in most of previous works one of the existing models is chosen, as-

[a)] ilucas@unizar.es

suming that only one of the mechanisms is present or relevant. Therefore, a relative quantification of both contributions is still lacking. This is however an important issue, required for a deeper understanding of the LSSE. In addition, it is also desirable from a practical point of view as it will give hints concerning the design of materials and devices to be used in potential applications.

In this work, we disentangle the contributions of *i*LSSE and *b*LSSE (bulk LSSE) within a static SSE experiment. We make use of following definitions for the corresponding coefficients (S^{i} and S^{b}):

$$S^{\mathrm{i}} = \frac{\Delta V^{i}_{\mathrm{ISHE}}}{d_y \Delta T^{\mathrm{NM/FM}}} \quad \text{and} \quad S^{\mathrm{b}} = \frac{\Delta V^{b}_{\mathrm{ISHE}}}{d_y \Delta T_{\mathrm{FM}}}, \tag{3}$$

where d_y is the distance between the transverse contacts to measure the transverse ISHE voltage (ΔV_{ISHE}). These definitions make use of the actual temperature difference driving each effect, rather than the overall temperature difference. This normalization is favored by the SSE measurement method known as heat flux method[6,17,18], which measures the heat current flowing through the sample instead of the total temperature difference (used in the temperature difference method).

Our approach to the quantification of *i*LSSE and *b*LSSE contributions is based on the study of the LSSE as a function of t_{FM}. Under a fixed total thermal difference across the whole sample ΔT (typical experimental condition in LSSE static experiments), the thermal drops across the FM layer thickness and at the NM/FM interface change upon changing t_{FM}. Consequently, J_s^b and J_s^i change accordingly, since they are driven by those thermal differences (recall Eqs. 1 and 2). However, this issue has been omitted in previous studies on the t_{FM} dependence of LSSE employing the temperature difference approach, although it represents a source of modulation of the contribution of both LSSE mechanisms. Typically, only the dependence related to the Λ_{m} (prefactor in equation 2) is addressed, whereas the influence of t_{FM} in the temperature drop in FM, ΔT_{FM}, has not been considered so far. To evaluate this, knowledge on the cross-plane thermal conductivities is needed to quantify the gradients. Therefore, a key point in our work is the assessment of the thermal conductivities for thin films of different thicknesses. An alternative to circumvent the thickness variation of ΔT_{FM} is offered by the heat flux method[19]. However, to separate the contributions to the LSSE, knowledge of the thermal conductivities is necessary.

In sum, in a typical experiment of LSSE as a function of t_{FM} in which ΔT is fixed to the same value for all t_{FM}, we consider the modulation of the thermally excited spin current J_s by three different means: (1) the existence of a magnon propagation length scale Λ_{m} comparable to the dimensions of our samples, (2) the change of the involved thermal drops due to the variation of the FM layer thickness in relation to the rest of the stacked layers, and (3) the dependence of the thermal conductivity of the FM layer on its thickness.

The studied samples consisted of epitaxial thin films of insulating ferrimagnetic $\gamma{-}Fe_2O_3$ (maghemite) deposited on (001) oriented MgO substrates of 0.5 mm of thickness, and an additional layer of Pt on top of maghemite for spin-to-charge current conversion. The thickness of the Pt layer is held at 6 nm through all studied samples. Maghemite is an ideal material to study LSSE because of its insulating behavior ruling Anomalous Nernst Effect contribution to the voltage[20–22]. However, proximity magnetism induced in the Pt layer by interdiffusion of Fe may originate an ANE contribution. To avoid this effect, Pt was deposited at room temperature[23,24]. Details about the fabrication of the samples can be found elsewhere, along with the description of the experimental setup for the LSSE measurements[25]. Moreover, the quality of interfaces affects the magnitude of the LSSE[26]; therefore, we assessed the interfacial roughness by X-ray reflectivity, finding comparable values < 1 nm for every t_{FM} (supplementary material).

FIG. 1. Depiction of the thermal differences established in the LSSE experiment through the studied sample, as expressed in Eq. 4. The graded arrow shows the direction of the thermal gradient.

In our setup the temperature difference between the hot and cold baths (i.e. the temperature drop ΔT across the whole sample including MgO substrate) is controlled and measured. The temperature profile will show two main features: (1) a change in its slope from layer to layer because of the different thermal conductivities and (2) a discontinuity at the interfaces due to the interfacial thermal resistance, also known as Kapitza resistance. Therefore, as illustrated in Fig. 1, we can split the total temperature difference into thermal drops along the sample:

$$\Delta T = \Delta T_{\mathrm{Pt}} + \Delta T_{\mathrm{i}}^{\mathrm{Pt/FM}} + \Delta T_{\mathrm{FM}} + \Delta T_{\mathrm{i}}^{\mathrm{FM/S}} + \Delta T_{\mathrm{S}} \tag{4}$$

which accounts for the temperature drop in the Pt layer, at the Pt/FM interface, in the FM layer, at the FM/substrate interface, and across the substrate. In this equation, the thermal drops originated by the thermal contacts of the sample with the baths[27] are disregarded, thus the values obtained in the determination of S^{i} and S^{b} are underestimated. However, the quantification of the relative contributions to ΔV_{ISHE} should be unaffected by this systematic error. The heat flux method[6,17,18] offers another possibility to circumvent this error.

According to a simple thermal model the heat flux is constant through the whole sample:

$$\begin{aligned} J_{\mathrm{Q}} &= \left(\frac{t_{\mathrm{Pt}}}{\kappa_{\mathrm{Pt}}} + \frac{t_{\mathrm{FM}}}{\kappa_{\mathrm{FM}}} + \frac{t_{\mathrm{S}}}{\kappa_{\mathrm{S}}} + R_{\mathrm{i}}^{\mathrm{Pt/FM}} + R_{\mathrm{i}}^{\mathrm{FM/S}} \right)^{-1} \Delta T = \\ &= \frac{1}{R_{\mathrm{T}}} \Delta T = \frac{\kappa_{\mathrm{FM}}}{t_{\mathrm{FM}}} \Delta T_{\mathrm{FM}} = \frac{1}{R_{\mathrm{i}}^{\mathrm{Pt/FM}}} \Delta T_{\mathrm{i}}^{\mathrm{Pt/FM}} \end{aligned} \tag{5}$$

t's denote thicknesses and κ's denote thermal conductivities; $R_{\mathrm{i}}^{\mathrm{Pt/FM}}$($R_{\mathrm{i}}^{\mathrm{FM/S}}$) is the thermal resistance coming from the Pt/FM (FM/substrate) interface; and R_{T} represents the thermal resistance of the system as a whole, i.e., the composition of the thermal resistances of each layer and interface.

As pointed above, the first step is establishing the actual thermal drop across the magnetic layer as well as through the FM/Pt interface. This is especially troublesome when

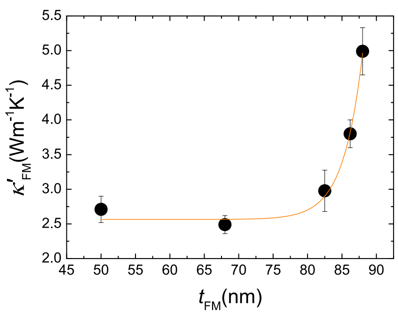

FIG. 2. Effective thermal conductivity of maghemite epitaxial thin films as a function of the thickness. Solid line represents an experimental fit to a sigmoid function used for interpolation of $\kappa'_{\rm FM}$ at the desired $t_{\rm FM}$.

dealing with thin films in which thermal transport properties differ from those of the bulk materials and are not handily characterizable[28]. However, the knowledge of these quantities is paramount for the quantitative analysis of the LSSE. This means that measurement of the thermal conductivity of the thin film is mandatory, since its value must be included into even the simplest thermal model along with that of the substrate.

In this work, the cross-plane thermal conductivity of maghemite thin films as well as MgO substrate were determined by the 3ω method[29] (supplementary material). It is important to note that the determined thin film conductivity is not the intrinsic $\kappa_{\rm FM}$ but an effective $\kappa'_{\rm FM}$ which also accounts for the thermal loss at the $\gamma{-}Fe_2O_3$/MgO interface $R_{\rm i}^{\rm FM/S}$[30].

$$\kappa'_{\rm FM} = \frac{\kappa_{\rm FM}}{1 + R_{\rm i}^{\rm FM/S}\,\kappa_{\rm FM}/t_{\rm FM}} \tag{6}$$

We will use this effective $\kappa'_{\rm FM}$ to calculate $\Delta T_{\rm FM}$. This means that hereafter, $\Delta T_{\rm i}^{\rm FM/S}$ will be contained in $\Delta T_{\rm FM}$.

We measure $\kappa'_{\rm FM}$ at $T = 300$ K for different $t_{\rm FM}$ thicknesses up to the thickness range of the samples we studied in the LSSE experiments. The thereby obtained values of the thermal conductivities at 300 K are depicted in Fig. 2. They follow a rather complex dependence, which may be attributed to the presence of defects rapidly changing with the film thickness; a possible candidate are the antiphase boundaries (APBs) which are usually shown by spinel structures (such as maghemite) and whose density decrease on increasing thickness[31]. A detailed analysis of this dependence is however beyond the scope of this work. From this experimental curve, we interpolate the values of $\kappa'_{\rm FM}$ at the $t_{\rm FM}$ of the samples studied in the LSSE experiments. Concerning MgO substrate, we measured $\kappa_{\rm S} = 44.2(5)\ {\rm W\,m^{-1}K^{-1}}$ at $T = 300$ K, in agreement with reported values for MgO single crystals[32,33].

The thermal resistance of the Pt layer is lower than the rest of the terms by two orders of magnitude, due to its small thickness $t_{\rm Pt} = 6$ nm and large thermal conductivity $\kappa_{\rm Pt} = 64$ ${\rm Wm^{-1}K^{-1}}$[34]. The thermal resistances of the stacked layers are $R_{\rm MgO} \sim 10^{-5}\ {\rm W^{-1}m^2K}$, $R_{\rm FM}$ ranges from $\sim 10^{-9}$ to 10^{-8} ${\rm W^{-1}m^2K}$, and from literature $R_{\rm i} \sim 10^{-9}\ {\rm W^{-1}m^2K}$[34,35]; all of them exceed $R_{\rm Pt} \sim 10^{-11}\ {\rm W^{-1}m^2K}$ by at least two orders of magnitude. This means that we can hence neglect the temperature drop in the Pt layer in Eq. 4 and its resistance contribution to Eq. 5.

Eq. 5 allows us to write every temperature drop in terms of the known ΔT:

$$\begin{aligned} \Delta T_{\rm FM} &\approx \frac{\kappa_{\rm S} t_{\rm FM}}{\kappa_{\rm S} t_{\rm FM} + \kappa'_{\rm FM} t_{\rm S}} \Delta T \\ \Delta T_{\rm i}^{\rm Pt/FM} &\approx \frac{\kappa_{\rm S}\,\kappa'_{\rm FM} R_{\rm i}^{\rm Pt/FM}}{\kappa_{\rm S} t_{\rm FM} + \kappa'_{\rm FM} t_{\rm S}} \Delta T \end{aligned} \tag{7}$$

where we have taken a second approximation: $\kappa_{\rm S}\,\kappa_{\rm FM} R_{\rm i}^{\rm Pt/FM} \ll \kappa_{\rm FM} t_{\rm S}$, which is reasonable, given the substrate thickness as well as the typical values of $R_{\rm i}$.

Once the thermal conductivities $\kappa'_{\rm FM}$ and $\kappa_{\rm S}$ have been estimated in this way, we proceed to the LSSE experiments for different FM thicknesses. It has been shown that variations in the thermal contacts between the sample and the baths are source of error[17,27]; care was taken to minimize these errors by ensuring that the same heat power sustained a similar ΔT for all samples. Alternatively, other approaches measure the heat flux instead[6,17,36].

In Fig. 3 the measured dependence of transverse voltage $\Delta V_{\rm ISHE}$ on $t_{\rm FM}$ is shown. Following a widespread practice, these quantities have been normalized by the total thermal difference ΔT[13,37], using the slopes of the linear fits of $\Delta V_{\rm ISHE}$ as a function of different ΔT (see the inset of Fig. 3).

However, this means that, according to Eq. 7, $\Delta T_{\rm FM}$ and $\Delta T_{\rm i}$ depend on $t_{\rm FM}$ in Fig. 3. Hence, the observed behavior comprises not only the effect of $\Lambda_{\rm m}$ (as assumed by previous works) but also the variation of the thermal differences driving the LSSE (bulk and interfacial). To rigorously take into account both effects, the scaling with the corresponding thermal drops should be used instead[17,18], as proposed in Eq. 3. For this, we rewrite the spin Seebeck coefficients as a function of the measured total ΔT, using Eq. 7:

$$\begin{aligned} \Delta V_{\rm ISHE}^{\rm i} &\approx S^{\rm i} \cdot d_y \cdot \frac{\kappa_{\rm S}\,\kappa'_{\rm FM} R_{\rm i}^{\rm Pt/FM}}{\kappa_{\rm S} t_{\rm FM} + \kappa'_{\rm FM} t_{\rm S}} \Delta T \\ \Delta V_{\rm ISHE}^{\rm b} &\approx S^{\rm b} \cdot d_y \frac{\kappa_{\rm S} t_{\rm FM}}{\kappa_{\rm S} t_{\rm FM} + \kappa'_{\rm FM} t_{\rm S}} \Delta T \end{aligned} \tag{8}$$

According to ISHE phenomenology, ISHE transverse voltage $\Delta V_{\rm ISHE}$ and the exciting spin current density $J_{\rm s}$ are related by $\Delta V_{\rm ISHE} \propto \theta_{\rm SH} \rho_{\rm c} J_{\rm s}$[38], where $\theta_{\rm SH}$ is the so-called spin Hall angle accounting for the efficiency of the spin-to-charge conversion, and $\rho_{\rm c}$ is the longitudinal electrical resistivity of the NM ISHE medium (here, Pt). Depending on the mechanism governing ISHE, $\theta_{\rm SH}$ may depend on $\rho_{\rm c}$[39–41]. However, in this experiment, Pt layer thickness is the same for all samples, resulting in constant $\rho_{\rm c}$. Therefore, $\Delta V_{\rm ISHE} \propto J_{\rm s}$. In sight of this fact and Eqs. 1, 2 and 8, we note that, unlike $S^{\rm i}$, $S^{\rm b}$ depends on t_{FM} and can be expressed as:

$$S^{b} = A \cdot \frac{1}{t_{FM}} \frac{\cosh(t_{FM}/\Lambda_{m}) - 1}{\sinh(t_{FM}/\Lambda_{m})} \quad (9)$$

where the coefficient A describes the thickness-independent term.

Altogether, the measured transverse voltage ΔV_{ISHE} will be

$$\Delta V_{ISHE} = \Delta V^{i}_{ISHE} + \Delta V^{b}_{ISHE} = S^{i} \cdot d_y \cdot \Delta T^{Pt/FM}_{i} + S^{b} \cdot d_y \cdot \Delta T_{FM} \approx \approx \left(S^{i} \cdot d_y \cdot \frac{\kappa_S \kappa'_{FM} R^{Pt/FM}_{i}}{\kappa_S t_{FM} + \kappa'_{FM} t_S} + A \cdot d_y \cdot \frac{\cosh(t_{FM}/\Lambda_m) - 1}{\sinh(t_{FM}/\Lambda_m)} \frac{\kappa_S}{\kappa_S t_{FM} + \kappa'_{FM} t_S} \right) \Delta T \quad (10)$$

A fit of this expression to the experimental data, shown in Fig. 3, provides (supplementary material) $\Lambda_m = 29(1)$ nm, $S^{i} R^{Pt/FM}_{i} = 1.83(6) \times 10^{-9}$ $(Vm^{-1}K^{-1})\cdot(W^{-1}m^{2}K)$ and $A = 40.8(9) \times 10^{-9}$ $V \cdot K^{-1}$. Given the reported order of magnitude of interfacial thermal resistances $R_i \sim 10^{-9} - 10^{-8}$ $W^{-1}m^{2}K$, we can also establish that S^{i} order of magnitude must lie between 0.1 and 1 $Vm^{-1}K^{-1}$, in agreement with previous estimations in other systems[7,15,26]. Concerning S^{b}, the value of A provides from $S^{b} = 0.65(6)$ $Vm^{-1}K^{-1}$ for the thinnest sample down to $S^{b} = 0.46(4)$ $Vm^{-1}K^{-1}$ for the thickest one.

We can now compute the relative contribution of each source —*b*LSSE and *i*LSSE— to the total ΔV_{ISHE} output in our range of FM thicknesses for an experiment like ours, in which a total ΔT is established. For this, we calculate ΔV^{i}_{ISHE} and ΔV^{b}_{ISHE} generated per Kelvin of total thermal drop ΔT from Eqs. 3 and 7 (i.e., the two addends between brackets in Eq. 10). The result is shown in Fig. 4.As one could expect, the influence of $R^{Pt/FM}_{i}$ is greater for thinner samples and consequently *i*LSSE contribution to the overall signal reduces as t_{FM} is increased, and reversely for *b*LSSE.

In summary, we have experimentally separated the bulk magnon accumulation and purely interfacial contributions to the LSSE in static heating conditions. We have done so by studying the LSSE as a function of the FM layer thickness in $\gamma-Fe_2O_3$/Pt bilayers, and taking into account the relative change of the present thermal drops along the sample on varying the FM thickness. To that end, we have also measured the thickness dependence of the thermal conductivity of $\gamma-Fe_2O_3$ thin films by means of the 3ω method. With this, we managed to fit the addition of theoretical models for both sources to our experimental data. This allowed us to in the end compute the separate contribution of each source of thermal spin current. We found that although bulk component dominates in the range of thickness of our samples, the interfacial contribution is not negligible against it, as it represents from $\approx 33\%$ to $\approx 12\%$ of the total voltage. Besides, *b*LSSE coefficient values were determined: it varies between $S^{b} = 0.68(6)$ $Vm^{-1}K^{-1}$ for the thinnest sample ($t_{FM} = 14.5$ nm) and $S^{b} = 0.46(4)$ $Vm^{-1}K^{-1}$ for the thickest one ($t_{FM} = 77$ nm). *i*LSSE coefficient order of magnitude was estimated to lie between $S^{i} \sim 0.1$ and 1 $Vm^{-1}K^{-1}$, in agreement with the values reported in literature for other systems[7,15,26]. The fit also provided a thermal magnon accumulation length in $\gamma-Fe_2O_3$ of $\Lambda_m = 29(1)$ nm. In literature, Λ_m at room temperature corresponding to other ferrimganetic oxides can be found: $\Lambda_m = 17$ nm for half-metal Fe_3O_4[42] or $\Lambda_m \sim 100$ nm for insulating YIG[26] were reported using the temperature difference method. However, one must be cautious before comparing our result, because those works only considered the influence of the thermally excited magnon propagation length on the thickness dependence of LSSE. More recent works using different approaches to investigate this dependence circumvent the change of ΔT_{FM}. For example, Noack *et al.* studied the LSSE time evolution in YIG thin films heating with microwave pulses, finding $\Lambda_m \sim 425$ nm[43]; and Venkat *et al.*, using the heat flux method, reported $\Lambda_m = 19(2)$ nm for Fe_3O_4 thin films[19]. Still, only the bulk effect is considered to explain the results.

These results reassure maghemite's potential in spin-caloritronic devices and bring insight into the global picture of the LSSE. In particular, the relative quantification of bulk and

FIG. 3. Transverse voltage ΔV_{ISHE} measured for every different t_{FM}, normalized by ΔT and fit of Eq. 10 to the experimental data (solid line).

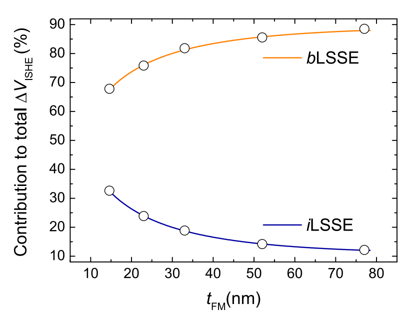

FIG. 4. Relative contribution ΔV^{i}_{ISHE} and ΔV^{b}_{ISHE} to the total ΔV_{ISHE}. Note that these percentages cannot be directly extended to S^{i} and S^{b}, given that they do not share the voltages dependences on t_{FM} (see Eq. 8).

interfacial contributions improves the precision of its description, therefore assisting a deeper understanding of the effect.

SUPPLEMENTARY MATERIAL

See supplementary material for details of the 3ω method, the structural characterization of samples, and the fit parameters initialization.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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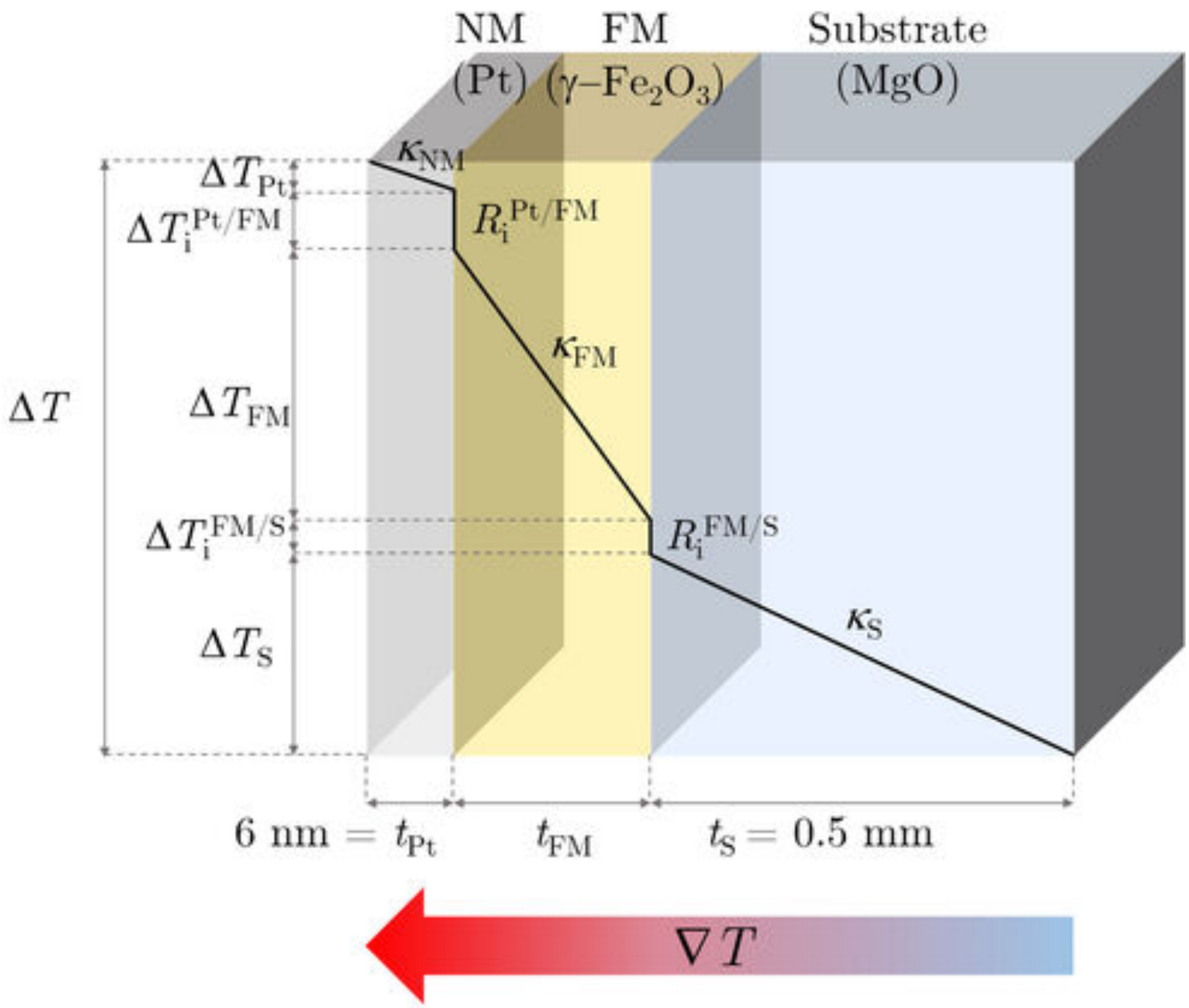

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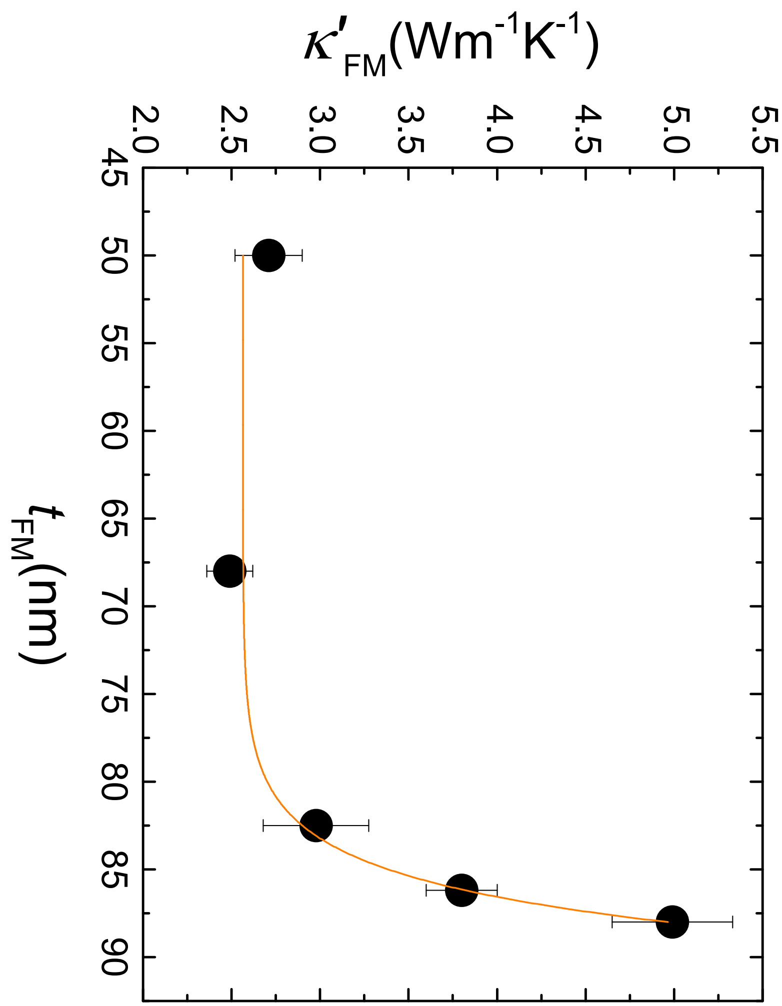

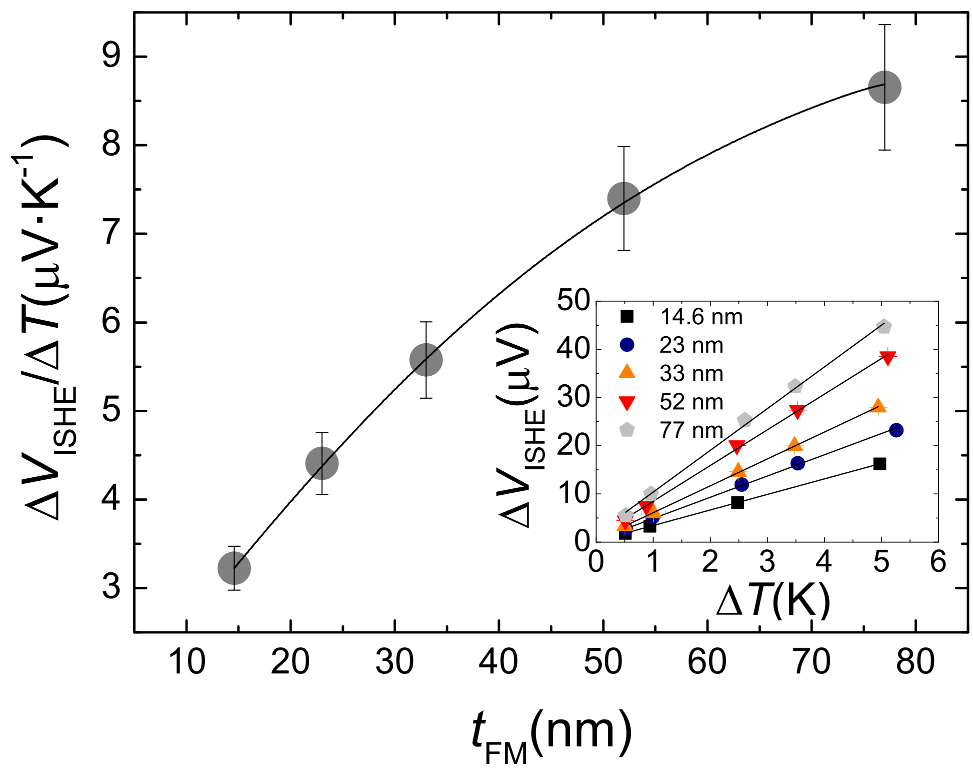
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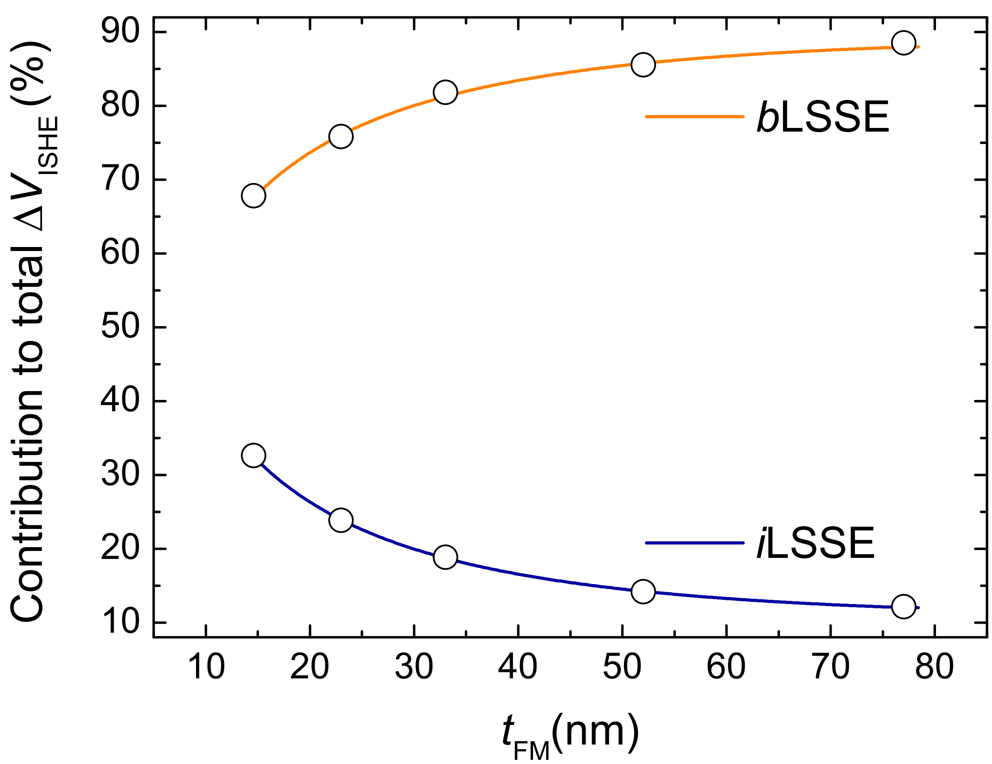
33 nm
52 nm
77 nm
ΔV_ISHE(μV)
ΔT(K)


Contribution to total ΔV_ISHE (%)
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bLSSE
iLSSE
10
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80
t_FM(nm)