Spin caloritronic measurements: a round robin comparison of the longitudinal spin Seebeck effect

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Abstract—The rising field of spin caloritronics focuses on the interactions between spin and heat currents in a magnetic material. The observation of the spin Seebeck effect opened the route to this branch of research making possible the design of thermoelectric devices made of insulating magnetic materials. A round robin test performed by five partners on a single device highlighted the reproducibility problems related to the measurements of the spin Seebeck coefficient, the quantity that describes the strength of the spin Seebeck effect. This work stimulates the search for more reproducible measurement methods.

Index Terms—Ferrimagnetic films, garnets, spintronics, temperature measurements, thermoelectricity.

I. INTRODUCTION

The concept of spintronics gained interest in the last 30 years, since the observations of spin-dependent electron transport phenomena in solids. The idea of handling the spins of the electrons in solids turned out to be very useful thank to phenomena like the spin pumping and the spin Hall effect. These refer to the spin current generation by means of microwave excitation and by an electric current flowing in an adjacent layer of a high spin-orbit coupling material (e.g. platinum). In more recent times since 2008, the research group led by prof. E. Saitoh at Tohoku University followed by others investigated the generation of a spin current in a magnetic material as a consequence of a thermal gradient [1]; this phenomenon is called spin Seebeck effect as reference to the spin counterpart of the Seebeck effect. It is possible to detect electrically the spin current generated by the spin Seebeck effect by means of the inverse spin Hall effect (ISHE) [2]; this rises in a Pt film, deposited on the magnetic material that produces a spin current.

II. MEASUREMENT OF THE SPIN SEEBECK COEFFICIENT

Because of the novelty of the spin Seebeck effect, a straightforward interpretation of the phenomenon was initially lacking, even from the experimental viewpoint. The first debate regarded the geometry of the spin Seebeck effect (longitudinal vs. transverse configuration) [3], [4]. The most studied configuration is the longitudinal spin Seebeck effect (LSSE); this rises in a bilayer system formed by an in-plane magnetized layer, typically a ferrimagnetic Yttrium iron garnet (YIG), covered by a Pt thin film for the ISHE detection. In this configuration, the thermal gradient is set out of plane in order to inject a spin current in the top Pt film. The quantitative determination of the LSSE is represented by the LSSE coefficient $S_{\text{LSSE}}$, defined by the expression

\begin{equation}
S_{\text{LSSE}} = \frac{-E_{\text{ISHE}}}{\nabla T} = \left( \frac{V_{\text{ISHE}}}{L} \right) / \left( \frac{\Delta T}{L_z} \right).
\end{equation}

The voltage $V_{\text{ISHE}}$ represents the electrical observable and is proportional to the charge accumulation along the Pt film of length $L$, as consequence of the ISHE. The thermal observable is the temperature difference $\Delta T$ across the thickness $L_z$. The measurement of the $S_{\text{LSSE}}$ was performed in order to investigate its reproducibility in the framework of a collaboration between five institutions: INRiM, Tohoku University, Bielefeld University, Ohio State University and Argonne National Laboratories. The five groups performed the round robin test according to the measurement method described in Fig. 1 on a single LSSE device fabricated in Tohoku University. This is a 4 $\mu$m-thick YIG film grown on a 0.5 mm-thick GGG substrate. The sample dimensions are 2mm $\times$ 6mm and the thickness of the platinum film on the top of the YIG is 10 nm. The measurement of the voltage $V_{\text{ISHE}}$ has as its only variable the contact between the wire and the film. This can be obtained by means of a bonding machine (Bielefeld University), of a tungsten tip (Tohoku University) or with the use of silver paste (all other institutions). The second observable is the thermal gradient whose measurement is obtained by means of two sensors placed on the two thermal conductor that are clamping the LSSE sample. The thermal measurement has to fulfill the following hypothesis: the temperature drop along the thermal conductor has to be negligible, the thermal gradient
inside the sample is supposed to be constant and the thermal resistance of the contact has to be reproducible. The variables that characterize the thermal measurement for each institution are listed in table I, where T.C. denotes a thermocouple.

**TABLE I**

**EXPERIMENTAL VARIABLES OF THE THERMAL MEASUREMENTS OF LSSE**

| Institution               | Sensor   | Thermal conductor | Area | Thermal grease | μm | \(Wm^{-1}K^{-1}\) |
|---------------------------|----------|-------------------|------|----------------|----|-----------------|
| INRIM                     | T-type   | AIN               | 10   | Rs silicone    | 3.6| 1.85            |
| Tohoku University         | T-type   | AIN               | 10   | Chemtronics Boron Nitride | 1.85|
| Bielefeld University      | K-type   | Al₂O₃             | 10   | Silicone       | 0.194|
| Ohio State University     | Cernox   | c-BN              | 8.76 | Apiezon N      | 2.5|
| Argonne National Labs.    | K-type   | OFHC-Cu           | 8    | Wakefield 122 silicone | 2.5|

The last two columns of table I are reporting the characteristics of the thermal contact that is responsible for the last hypothesis of the thermal measurement. The results of this round robin experiment in V/K units exhibit a large variation as reported in Fig 2.

**III. SOURCES OF SYSTEMATIC ERRORS**

As proven by the results reported in Fig. 2, the measurements of the LSSE coefficient \(S_{LSSE}\) are affected by systematic errors. For what concerns the electrical measurement, we analysed how the characteristics of the electrical contact influence the \(S_{LSSE}\), by keeping constant the thermal contribution and changing the size of a silver paste contact. We observed a first improvement of the reproducibility as a consequence of the controlled shape of the electric contacts, i.e. by depositing a gold electrode. The largest source of errors concerns with the resistance of the thermal contacts. Contextually to the round robin test, it was shown that the different conditions of contacts yield to variations of their thermal resistance as large as the one of the LSSE sample [5]. In order to overcome this problem, some groups proposed a method based on the use of Pt films deposited on the two faces of the sample for the measurements of the thermal gradients [6]. A second method, based on the measurement of heat fluxes was proposed; this allows to neglect the contribution of the thermal resistance of the contacts on a sample with known thermal conductivity. In order to validate this method, a second comparison was performed between INRIM and Bielefeld University; the \(S_{LSSE}\) coefficients of another LSSE device obtained by the two institutions was 

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(9.716 \pm 0.060) \times 10^{-7} \text{ V/K and (9.350} \pm 0.128) \times 10^{-7} \text{ V/K with the heat flux method and (2.313} \pm 0.017) \times 10^{-7} \text{ V/K and (4.956} \pm 0.005) \times 10^{-7} \text{ V/K with the temperature difference method [7].}
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**IV. CONCLUSIONS**

A round robin test for the measurement of the \(S_{LSSE}\) pointed out the experimental problems; this caused a considerable effort in the search for a method of reproducible measurements, which has been identified as the heat flux method. This approach is also promising for the quantitative characterization of different LSSE materials and other spin caloritronics effects as the spin Peltier effect.

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**REFERENCES**

[1] K. Uchida et al., Observation of the spin Seebeck effect, Nature 455, 778–781, 2008.
[2] E. Saitoh et al., Conversion of spin current into charge current at room temperature: Inverse spin-Hall effect, Appl. Phys. Lett. 88, 182509, 2006.
[3] K. Uchida et al., Observation of longitudinal spin-Seebeck effect in magnetic insulators, Appl. Phys. Lett. 97, 172505, 2010.
[4] D. Meier et al., Longitudinal spin Seebeck effect contribution in transverse spin Seebeck effect experiments in Pt/YIG and Pt/NFO, Nature Communications 6, 2015.
[5] A. Sola et al., Evaluation of thermal gradients in longitudinal spin Seebeck effect measurements, J. Appl. Phys. 117, 17C510, 2015.
[6] K. Uchida et al., Quantitative temperature dependence of longitudinal spin Seebeck effect at high temperatures, Phys. Rev. X 4, 041023, 2014.
[7] A. Sola et al., Longitudinal spin Seebeck coefficient: heat flux vs. temperature difference method, Sci. Rep. 7, 46752, 2017.