

Magnon and phonon thermal transport and spin Seebeck effects in magnetic insulators

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Magnon and phonon thermal transport and
spin Seebeck effects in magnetic insulators

(磁性絶縁体におけるマグノン-フォノン熱輸送と
スピンゼーベック効果)

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I. Introduction

The spin Seebeck effect (SSE) refers to the generation of a spin current, i.e., a flow of spin angular momentum, as a result of a temperature gradient in a magnetic material. Here, the thermally generated spin current in a magnet is converted into a conduction-electron spin current in a paramagnetic metal attached to a magnet by the interfacial exchange interaction and detected as a transverse electric voltage via the inverse spin Hall effect (ISHE). The SSE is one of the phenomena discovered in the field of spin caloritronics, a new branch of spintronics, that focuses on the interaction between spin, charge, and heat currents in condensed-matter structures and devices.

This thesis addresses the origin of the SSE in the longitudinal configuration [i.e., longitudinal SSE (LSSE)] for metallic-film/magnetic-insulator heterostructures. The central purpose of this thesis can be divided into two parts: (i) constructing a versatile method to separate the LSSE from the parasitic anomalous Nernst effect (ANE) induced by a static magnetic proximity effect (Chap. 3) and (ii) unraveling the role of magnon and phonon thermal excitations and transport in the SSE (Chaps. 4 and 5).

This thesis consists of 6 chapters and is organized as follows.

- *Chapter 1* shows a brief history of spintronics and spin caloritronics and explains basic concepts and former studies which are necessary to understand the following Chapters. Besides, pioneering works and some research topics that have attracted much attention in spintronics are also introduced.

- *Chapter 2* describes the sample preparation, characterization, and measurement methods. Physical properties of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (yttrium iron garnet: YIG), a ferrimagnetic insulator used in this study, are also overviewed in this Chapter.

- *Chapter 3* shows our proposal and demonstration to separate the LSSE from the parasitic ANE induced by a static magnetic proximity effect.

- *Chapter 4* presents our systematic experimental results on the high-magnetic-field response of the LSSE at various temperatures, clarifying strong coupling of the SSE with thermal magnon excitation and propagation.

- *Chapter 5* demonstrates that the SSE can detect the hybridized magnon-phonon excitations, i.e., magnon-polaron formation, arising from the magnetoelastic coupling at the band (anti-)crossings between the magnon and phonon dispersion curves.

- *Chapter 6* is devoted to summarize our results and comment on their importance.

Below I elaborate on the main results obtained in this study.

II. Separation of longitudinal spin Seebeck effect from anomalous Nernst effect induced by proximity ferromagnetism (Chapter 3)

In the field of spintronics and spin caloritronics, many experimental and theoretical studies have been focused on spin-transport phenomena in paramagnet/ferromagnet junction systems, where a spin current plays a central role. Since the first demonstration of spin-current transport in a magnetic insulator YIG in 2010, the Pt/YIG junction system has become one of the prototype samples; Pt is a paramagnetic metal exhibiting high ISHE efficiency due to its strong spin-orbit interaction, while YIG is a ferrimagnetic insulator with a high Curie temperature (~ 560 K) and extremely high resistivity. Therefore, this structure enables pure detection of spin-current phenomena free from conduction-electrons' contribution in YIG.

Recently, several research papers pointed out the possibility that the LSSE in this Pt/YIG system may be

contaminated by the proximity-induced ANE, since Pt is near the Stoner ferromagnetic instability and proximity ferromagnetism may be induced in the Pt layer in the vicinity of the Pt/YIG interface. These reports raise the question whether or not the LSSE is merely the ANE: this is an essential issue in the fields of spintronics and spin caloritronics. This Chapter provides a clear answer to this question; we show conclusive evidence that the LSSE is an intrinsic phenomenon and completely different from the conventional ANE. To this end, we performed systematic experiments and analyses on the LSSE and Nernst effects in Pt/Y₃Fe₅O₁₂ (YIG), Pt/Gd₃Ga₅O₁₂ (GGG), Au/YIG, Au/GGG, Ni₈₁Fe₁₉/YIG, and Ni₈₁Fe₁₉/GGG samples in different temperature-gradient and magnetization configurations: the in-plane magnetized (IM) and perpendicularly magnetized (PM) configurations, where the LSSE appears only in the IM configuration while the ANE appears both in the IM and PM configurations. Using these samples and configurations, we have demonstrated the clear separation of the SSE from the ANE induced by the static magnetic proximity; the possible ANE contamination in the Pt/YIG sample is experimentally estimated to be less than 0.1 %, which is supported by our phenomenological model calculation. Finally, using the Ni₈₁Fe₁₉/YIG sample, we have realized the hybrid thermoelectric generation based on the SSE and ANE. Since the experimental methods demonstrated here provide a simple and versatile way to separate the SSE from the ANE and to detect the pure SSE signal, it will be useful for determining the origin of transverse thermoelectric voltage in various metal/insulator junction systems.

III. Critical suppression of spin Seebeck effect by high magnetic fields (Chapter 4)

The observation of the SSE in a ferrimagnetic insulator YIG in 2010 suggested that this phenomenon is attributed to nonequilibrium magnon dynamics driven by a temperature gradient and led to the development of SSE theories, in which its mechanism is explained by the thermally induced spin pumping by magnons in a ferromagnetic layer. However, microscopic understanding of the relation between the magnon excitation and thermally generated spin currents is yet to be established, and more systematic experimental studies are necessary. Here we focus on the high-magnetic-field response of the LSSE. We showed that the LSSE signal in YIG-bulk based systems gradually decreases with increasing magnetic field and at low temperatures converges to zero. The field-induced SSE suppression is interpreted in terms of the Zeeman gap $\hbar\gamma\mu_0H$ in magnon excitation, indicating that the SSE originates from thermally excited magnon dynamics. Interestingly, the field-induced reduction of the SSE signal turned out to appear even at room temperature (i.e., $\hbar\gamma\mu_0H \ll k_B T$). The result suggests that low-frequency magnons with energy comparable to or less than the Zeeman energy provide a dominant contribution to the LSSE rather than higher-frequency magnons. This spectral nonuniformity of the thermospin conversion is associated with the characteristic lengths of the LSSE since the SSE signal and its magnetic field dependence are strongly affected by the thickness of YIG.

IV. Magnon-phonon hybridization in spin Seebeck effect (Chapter 5)

One of the interesting features of the SSE is that this phenomenon provides a sensitive probe for spin correlations in magnetic materials as shown in Chap. 4. Here, we show that the SSE can probe not only spin dynamics but also phonon dynamics. A high-resolution field scan reveals the emergence of asymmetric peaks in the magnetic field

dependence of the LSSE in Pt/YIG systems. Interestingly, the amplitude of the structures turns out to reflect the number of crossing points between magnon- and phonon-dispersion curves in momentum space at which magnon polarons are formed by the magnetoelastic interaction; the SSE anomalies coincide with magnetic fields tuned to the threshold of magnon-polaron formation. Experimental results are well reproduced by a Boltzmann theory in which the magnetoelastic coupling as well as an acoustic quality much better than a magnetic quality are taken into consideration. The sharp peak structures of the SSE are thereby attributed to the spin current carried by magnon polarons exhibiting longer lifetimes than pure magnons. Our finding unveils an unknown role of phonons in spintronics and spin caloritronics and demonstrates the power of the SSE as a spectroscopic tool for coupled magnon-phonon dynamics.

V. Conclusion

The results obtained in this study are an important step towards a complete physical picture of the SSE in magnetic insulators. The measurement scheme established here provides a novel platform to exclusively detect the SSE and elucidate its underlying physics. This indeed contributed to the first observation of the effect of magnon excitations on thermally induced spin currents and led to the discovery of hybridized magnon-phonon excitations in the SSE signals. The crucial roles of magnons and phonons in thermal spin transport revealed here will provide new insights into spintronics and spin caloritronics and will open new avenues for developing future spintronic devices that utilize coupled magnon-phonon modes as efficient spin information carriers.