

# Experimental report

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**Proposal:** 4-01-1560

**Council:** 4/2017

**Title:** Revealing the Magnon Contribution to the Spin Seebeck Effect in the Prototype Compound Gd<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> by Inelastic Neutron Scattering

**Research area:** Physics

**This proposal is a resubmission of 4-01-1541**

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**Samples:** Gd<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>

Instrument	Requested days	Allocated days	From	To
IN8	7	0		
IN20	7	5	21/06/2018	27/06/2018

## Abstract:

The Spin Seebeck Effect (SSE) has attracted significant interest due to its potential in spintronics applications. In ferrimagnetic insulators, the SSE has been proposed to be a consequence of magnonic spin currents, the prototypical example being Gd<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> (GdIG). We propose to measure the low energy magnon spectrum from GdIG using Inelastic neutron scattering (INS) and compare these results to our theory. This model suggests that an observed reversal in the SSE signal at low temperature is a consequence of low energy magnon band dynamics. We will use a proven method [7] to measure the INS from high neutron absorbing Gd, by using a large area (3x3cm) thin (1-micron) film of epitaxial GdIG on a Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> substrate (proof of feasibility!) We will use IN8 or IN20 to investigate the INS close to the gamma-point at (004) corresponding to a momentum transfer of 2.5 Inverse Angstroms and perform energy scans out to 16meV as a function of temperature. We expect to observe the low energy modes to merge as the temperature is raised from 20 to 100K. Confirmation of the magnon contribution to the SSE will provide a substantial step in understanding this technological class of materials.

# Revealing the Magnon Contribution to the Spin Seebeck Effect in the Prototype Compound $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ by Inelastic Neutron Scattering

## Experimental Report: 4-01-1560

Thermoelectric generation is one of the promising technologies for green energy harvesting since it enables direct conversion from thermal energy to electrical power. A magnetic or *spintronic* analogue to thermoelectric generation, the ‘Spin Seebeck Effect’ (SSE), has attracted recent science interest due to the possibility for new technology innovations in the renewable energies sector. The SSE refers to the generation of a spin current, a flow of spin angular momentum, as a result of a temperature gradient in a magnetic material (figure 1a). In normal metal / oxide insulator interfaces, the spin current is transformed into a charge current by the inverse Spin Hall effect in the metal surface layer. While SSE results from the interface between the magnetic insulator and metal surface layer, the oxide magnon characteristics determine the magnitude and sign of the resulting SSE current generation. Therefore, a precise characterisation of magnon behaviour is crucial in understanding the SSE. The goal of experiment 4-01-1560 was to investigate if inelastic neutron scattering could be used to reveal the magnonic contribution in known SSE compensated garnet systems  $\text{Re}_3\text{Fe}_5\text{O}_{12}$ .

Thin film epitaxial nanostructures of  $\text{Gd}_3\text{Fe}_5\text{O}_{12}/\text{Pt}$  (GdIG) bilayers (figure 1a), are well known prototypal systems that demonstrate the SSE effect. In the original proposal, we proposed to investigate large area thin films of  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$  in order to reduce the problem of high neutron absorption of Gd. However, due to the problem of fabricating high quality films and following an official request to ILL for a sample change, we decided to switch samples to single crystal of  $\text{Tb}_3\text{Fe}_5\text{O}_{12}$  (TbIG) in which we find similar SSE characteristics to  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ . (see figure 1).

**Figure 1.** (a) Left: Measurement of SSE in TbIG, the current source is used to generate a thermal gradient and the SSE signal is measured in Pt electrodes. (b) Centre: The measured SSE in TbIG, the sign change in  $V_{\text{SSE}}$  at  $T=250\text{K}$  is due to the magnetic compensation temperature. (c) Right: The measured magnetisation in TbIG showing  $T_{\text{comp}}=250\text{K}$ .

Figure 1b, shows the temperature dependence of the measured SSE voltage in TbIG. The change in sign at of  $V_{\text{SSE}}$  at  $250\text{K}$  can be attributed to the compensation temperature, where the magnetisation of the Tb ions and Fe ions are equal. However, the increase in  $V_{\text{SSE}}$  observed below  $T\sim 100\text{K}$  (and sign change at very low temperature) cannot be readily understood from the magnetisation response. In our experiment we wished to characterise the magnon modes in TbIG and correlate them with this response. At IN20 we used polarised neutrons with polarisation analysis and horizontal applied field, to distinguish between the magnon and phonon excitations.

In our experiment, we focussed on measuring the optical magnon modes close to the  $\Gamma$ -point  $Q=(444)$  and lower energy acoustic modes at  $Q=(3.5\ 3.5\ 3.5)$  see figure 2a and 2b. The polarisation analysis of the optical modes (figure 2b) reveals a large asymmetry in the spin-flip intensities (+- & -+), which indicates that these magnon modes are entirely chiral. Furthermore, we observe a cross-over in intensity asymmetry above and below  $T_{\text{comp}}=250\text{K}$ , which is consistent with a change in chirality of these magnon modes, accompanying the change in sign of  $V_{\text{SSE}}$ . On the other hand, the acoustic magnon modes show a much less pronounced asymmetry in spin-flip intensities, which might indicate that these modes are less chiral in character. The results demonstrate that the high temperature SSE response (in sign and magnitude) are largely due to the chirality of the optical magnons. As temperature is lowered, a gap starts to increase

between the acoustic and optical modes and becomes quite pronounced below  $T=100\text{K}$ . The red vertical lines indicate the thermal energy and below  $T=100\text{K}$ , the optical mode becomes frozen out, leaving only the acoustic modes to dominate. The freezing out of the optical modes coincides with the sharp increase in  $V_{\text{SSE}}$  that occurs below  $T=100\text{K}$  and eventually changing sign at  $T=20\text{K}$ . While the optical modes are consistent with our theoretical model, the acoustic modes are not. In particular our model predicts a single chiral state also for the acoustic modes. We required better resolution magnon data on the acoustic modes using cold neutrons to better understand this issue. Also the extension of these measurements to other Rare earth iron garnet systems such as DyIG will provide a better overview on the magnon characteristics in this class of materials. A more quantitative model, directly related the magnitude and sign of the magnon characteristics to SSE generation is still to this class of materials.

**Figure 2.** (a) left: The temperature dependence of the acoustic magnons measured at  $Q=(3.5\ 3.5\ 3.5)$  and  $+-$  (red) and  $-+$  (blue) refers to spin-up (down) flipped to spin-down(up). (b) The temperature dependence of the optical phonons measured at  $Q=(444)$ .