

Experimental report

07/09/2022

Proposal: 4-01-1610

Council: 10/2018

Title: Revealing the Origin of the Spin Seebeck Effect: Polarised Neutron Inelastic Scattering Studies of Dy₃Fe₅O₁₂ on IN20

Research area: Physics

This proposal is a continuation of 4-01-1560

Main proposer: Daniel MANNIX

Experimental team: Daniel MANNIX
Mechthild ENDERLE
Stephan GEPRAGS
Maxim DIETLEIN
Thomas THIES

Local contacts: Tobias WEBER
Mechthild ENDERLE

Samples: Dy₃Fe₅O₁₂
Tb₃Fe₅O₁₂

Instrument	Requested days	Allocated days	From	To
IN20	7	7	22/07/2019	29/07/2019

Abstract:

During our previous experiment on IN20 we obtained beautiful magnon data with full polarisation analysis on the Spin Seebeck Effect (SSE) material Tb₃Fe₅O₁₂. The high energy (3-10meV) optical magnons agree quantitatively well with our theoretical model of the origin of the Spin Seebeck Effect. However, the lower energy acoustic modes (0-3meV) do not appear to agree with our theoretical model of the SSE. Here we propose to extend these measurements to Dy₃Fe₅O₁₂ SSE material on IN20. In particular, the Dy₃Fe₅O₁₂ compound shows an interesting magnetic field dependence of the SSE not observed in Tb₃Fe₅O₁₂ (fig 1c). This field effect occurs at low temperatures (<120K) where the SSE becomes quenched. By investigating the magnon behavior in magnetic fields of 0-4T available in the horizontal magnet on IN20 will provide more information on the magnon mechanism involved at low temperatures. These results will enable us to further refine our model on the origin of the SSE in the compensated rare-earth iron garnet systems. The SSE The unique energy resolution of neutron methods and bulk single crystals provide an idea tool for investigating origin of the technologically important SSE.

Revealing the Magnon Contribution to the Spin Seebeck Effect in Tb₃Fe₅O₁₂

Aim of Experiment

In experiment 4-01-1609 on IN20, we proposed to use Polarised Inelastic Neutron Scattering (PINS) to investigate the chirality of the magnon contribution giving rise to the so-called Spin Seebeck Effect (SSE) [1-4]. The aim of the experiment on IN20 was to investigate chirality of the magnon modes in Tb₃Fe₅O₁₂ (TbIG). The ReIG (Re=Gd,Tb,Y etc) are important to spintronics applications such as SEE, due to their long magnon lifetimes. The work is motivated by theoretical analysis first applied to Gd₃Fe₅O₁₂ [5], in which the thermal response of the SSE is proposed to be due to the interplay of low energy magnon modes of opposite chirality. This can be shown in figure 1(a), where the first optical mode (lowest blue parabolic curve), is shifted up in energy as temperature is lowered and eventually shifts above the thermal energy (horizontal flat line) around 40K (bottom panel). At low temperatures the magnons are dominated by acoustic Goldstone modes of opposite Chirality. This should then be responsible for an observed change in sign in the SSE voltage generated [5] at low temperatures.

In our experiments on IN20, we find clear evidence for the optical blue magnon mode as shown in figure 1b. In a PINS experiment the chirality of a magnon band is evident by an asymmetry in the spin-flip intensities $+$ - & $-$ +. From the data in fig 1b we can conclude that the first optical mode has 100% chirality because the intensity is 100% in one of the $-$ + spin-flip channel.

However in the experiments on IN20 and later on IN12 (fig 1c) with higher energy resolution cold neutrons, we failed to observe any evidence of low energy dispersive Goldstone acoustic modes in TbIG. After a careful analysis of the data, we failed to find any clear evidence of a low energy acoustic mode as proposed in the theory for GdIG [5].

These results have puzzled us for a while and motivated new theory in collaboration with the ILL theory group and the university of Kent. In our theory, we use a simplified model to investigate the effects of the strong crystal-field levels of Tb³⁺ to the chirality dependent magnon modes involved in driving the SSE in TbIG [6]. Our simplified model is general for ReIG, and we obtain a measure of the energy as a function of the canting angles (θ_R, θ_{Fe}) of the Re and of the Fe magnetic moments

$$\begin{aligned} \epsilon(\theta_R, \theta_{Fe}) = & -K_e S^2 \cos^2(\alpha_0 - \theta_R) + J_{cd} S s_d \cos(\theta_R - \theta_{Fe}) \\ & + J_{dd} s_d^2 (2 \cos^2 \theta_{Fe} - \sin^2 \theta_{Fe}) / 2 \\ & - 2 J_{ad} s_d s_a \cos^2 \theta_{Fe} . \end{aligned}$$

α_0 is a constant, K_e is the anisotropy constant for the easy-axis of the Re magnetic moment S . The constants J_{cd} , J_{dd} , J_{ad} account for the exchange couplings between the magnetic moments of the rare-earth (S , c -sites) and of the iron ions (s_d , d -sites; s_a , a -sites). This study has proven that the acoustic mode can become hybridised with the crystal field levels and is shifted to higher energies with mixed chirality [6]. These effects are expected to be negligible for GdIG, since Gd³⁺-ions have a spin-only ground multiplet and should exhibit a negligible single-ion anisotropy. We are currently working towards a more detailed theory with the aim to fully reproduce our PNIS on TbIG, and to know more of ReIG as a class of materials.

New PINS experiments on a low absorption isotope of GdIG are required to test our new theory and provide a richer understanding of the ReIG SSE class of materials.

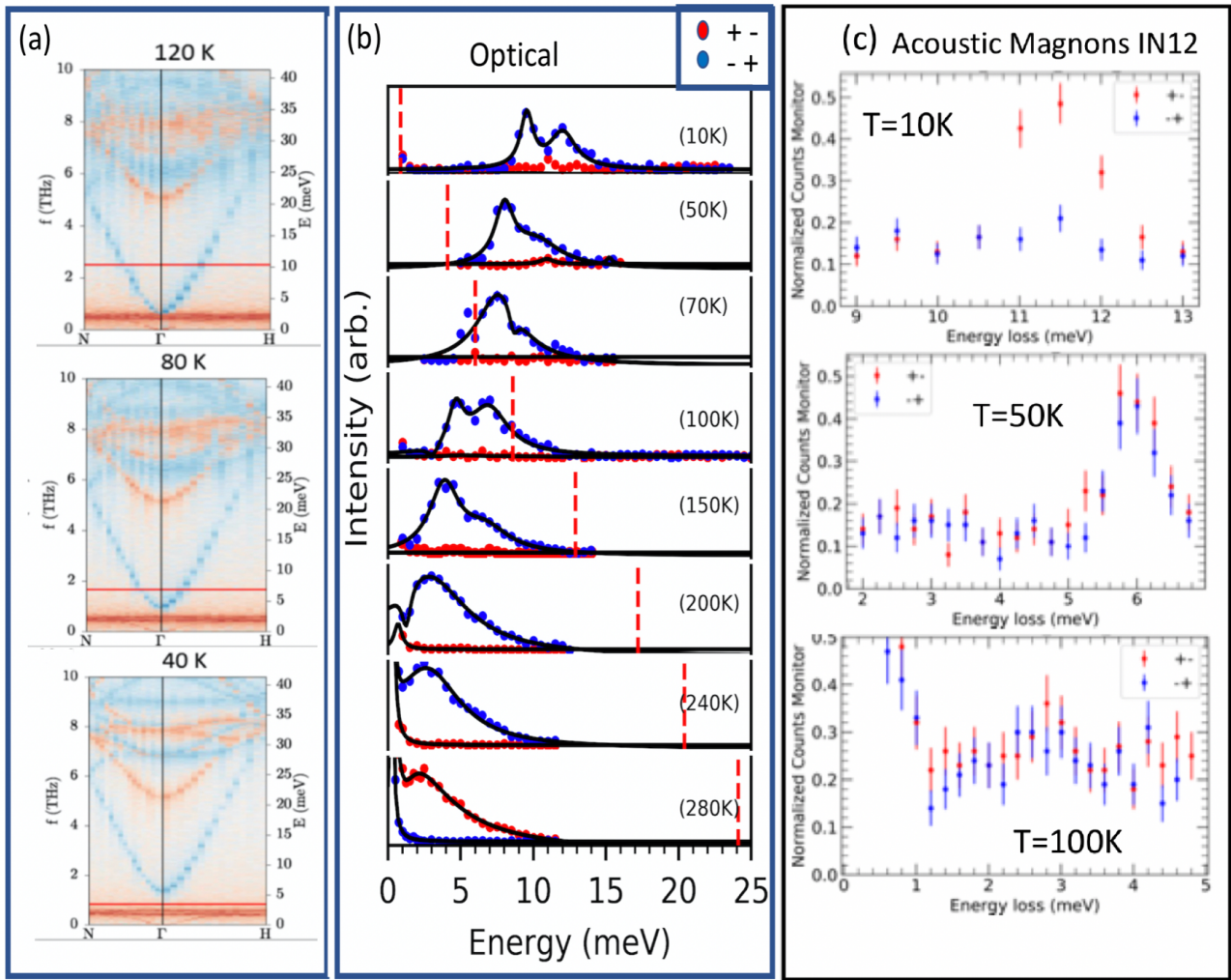


Figure 2 (a) left. Theoretical model of SSE [5]. The first parabolic (blue) mode is the optical mode of opposite chirality to the lower energy acoustic (flat-red) magnons. At high temperature $>120K$ SSE response is dominated by the optical magnons. Below $T < 120K$ the optical modes become frozen out (horizontal line is thermal energy) as the band gap opens and the SSE response is dominated by acoustic magnons of opposite chirality (red). The model proposes that this effect causes an change in $V(SSE)$ shown in fig 1b for $T < 100K$. (b) middle. The measured optical magnons in SF channels $+-$ (red) and $-+$ (blue) at Γ -point (444) on IN20, in qualitative agreement with theory (fig 2a). (c) right. The measured PINS in the spin-flip channels on IN12 at low energy loss. These magnon modes are not clear are not consistent with the model proposed for GdIG [5]: They are not purely chiral with both $+-$ and $-+$ SF scattering and do not switch chirality above T_{comp} .

References:

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<https://arxiv.org/abs/2207.00017>