Non-local magnon transport in the compensated ferrimagnet GdIG

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We study the diffusive transport of magnons through the compensated ferrimagnetic insulator Gd$_3$Fe$_5$O$_{12}$ (GdIG). The magnons are injected and detected electrically in a non-local measurement configuration via two parallel Pt strips deposited on top of the ferrimagnet. GdIG exhibits a rich magnon spectrum, with several thermally populated magnon bands at room temperature. We observe a strong temperature and field dependence of the non-local voltage in the detector strip. Just below the magnetization compensation temperature we find that the increasing magnetic field causes an unexpected enhancement of the non-local signal. A comparison with GdIG spin wave spectra obtained from atomistic modeling indicates that the thermal magnon population is important for understanding the non-local voltage signal.

Several experimental and theoretical investigations of diffusive magnon transport through the ferrimagnetic insulator yttrium iron garnet (Y$_3$Fe$_5$O$_{12}$, YIG) have recently been conducted, using two electrically isolated Pt strips for electrical injection and detection [1]. Upon running a dc charge current through a Pt injector strip, an electron spin accumulation is generated at the YIG/Pt interface via the spin Hall effect (SHE) [2, 3]. This spin accumulation induces a non-equilibrium magnon accumulation in the magnet beneath the injector which diffuses away from the injector and is detected in a second Pt strip via the inverse spin Hall effect (ISHE). This so-called non-local magnon mediated magnetoresistance (MMR) has been studied as a function of magnetic field [4] and temperature in YIG/Pt bilayers [2, 5], giving insight into the length scales associated with magnon transport. However, the microscopic mechanisms responsible for the MMR are still under investigation. One key question is which magnons or magnon bands provide the most significant contributions to diffusive spin transport and the non-local voltage signal induced in the detector strip. Furthermore, the MMR has only been measured in collinear magnetic systems such as YIG or NiFe$_2$O$_4$ [6] so far. The influence of non-collinear magnetic configurations, such as a canted ferrimagnet, has not yet been investigated.

Here, we study the non-local magnon transport in the compensated ferrimagnet gadolinium iron garnet (Gd$_3$Fe$_5$O$_{12}$, GdIG), where the non-magnetic Y$^{3+}$ in YIG is substituted with magnetic Gd$^{3+}$. GdIG therefore consists of three magnetic sublattices: two antiferromagnetically coupled Fe$^{3+}$ sublattices (FeA and FeD), and a Gd sublattice which is weakly antiferromagnetically coupled to the FeD moments [13, 14]. GdIG exhibits a magnetization compensation temperature $T_{\text{comp}}$, where the remanent magnetization exactly vanishes due to the different temperature dependencies of the antiferromagnetically coupled net Fe and Gd sublattice magnetizations [13]. Far away from the compensation temperature, GdIG is a collinear ferrimagnet and the sublattice magnetizations are all aligned (anti)parallel to the external magnetic field. Close to $T_{\text{comp}}$, a canted phase can be induced with magnetic field magnitudes accessible in experiments, where the magnetic moments on the different sublattices are no longer collinear with the field, or one another. The corresponding magnetic phase diagram can be found in Ref. [14]. Recently it was shown that the spin Hall magnetoresistance effect [17, 20] (SMR) can electrically probe the interface magnetization texture, since the SMR is sensitive to the orientation of the individual sublattice magnetic moments relative to the polarization of the spin accumulation in the adjacent Pt [13, 21]. Non-local magnon transport or spin diffusion has not yet been studied in a non-collinear magnetic structure and will be presented in this Article. Aside from the magnetic compensation point, GdIG also differs from YIG in that the spin wave spectrum is richer in the low THz regime. Several additional modes are present due to the magnetic Gd sublattice. Local spin Seebeck effect (SSE) measurements in GdIG showed strong evidence that two spin wave modes with opposite polarization dominate the thermal and dynamical behaviour of...
the spin system, resulting in a sign change of the SSE voltage \[22, 24\]. GdIG is therefore a good material to investigate the influence of different magnon modes and their polarization on spin diffusion. Our experimental results suggest that the complex magnon spectrum of GdIG indeed qualitatively impacts the field and temperature dependence of the MMR.

The investigated sample is a 2.6 \( \mu \)m thick GdIG film grown on top of a (111)-oriented Gd\(_3\)Ga\(_5\)O\(_{12}\) substrate via liquid phase epitaxy (LPE). After cleaning in Piranha solution and annealing at 500\(^{\circ}\)C in an oxygen atmosphere of 75\(\mu\)bar for 40 min, a 10nm thick Pt film was deposited onto the GdIG via electron beam evaporation (see Refs. [2] and [23] for details). Two Pt strips with edge-to-edge separation \(d = 200\) nm and strip width \(w = 500\) nm (Fig. 1(a)) were defined by electron beam lithography followed by Ar ion etching. The sample was mounted in the variable temperature insert of a superconducting magnet cryostat allowing dc transport measurements at temperatures between 5 \(K\) \(< T \) \(< 300 \)K and magnetic fields up to 15 \(T\). For the electric transport measurements a charge current \(I_c = 100 \mu\)A (corresponding to a current density of \(2 \times 10^{10}\) A m\(^{-2}\)) was applied to the left strip using a Keithley 2400 sourcemeter. The local (\(V_{loc}\)) and non-local voltage drop (\(V_{nl}\)) in the injector and detector strip were simultaneously measured with two Keithley 2182 nanovoltmeters (see Fig. 1(a)). To eliminate thermal signals due to Joule heating in the local strip (e.g. spin Seebeck effect [25]) and to increase the signal-to-noise ratio, a current switching method was used, as described in Ref. [2]. To obtain the SMR and MMR amplitude, angle dependent magnetoresistance (ADMR) measurements were carried out by rotating the external magnetic field of constant magnitude in the thin film plane, while recording \(V_{loc}\) and \(V_{nl}\).

The local response \(V_{loc}\) measured at 300 K as a function of the angle \(\alpha\) between \(I_c\) and \(H\) is shown in Fig. (b) for \(\mu_0H = 1\) \(T\) (grey) and 7 \(T\) (green). From these ADMR measurements, the SMR amplitude defined as \(\Delta V_{loc}/V_{loc, min} = (V_{loc}(0^\circ) - V_{loc}(90^\circ))/V_{loc}(90^\circ)\) was extracted and plotted as a function of temperature for magnetic field strengths \(\mu_0H = 1, 3, 5\) and 7 \(T\) in Fig. (d). Cooling to lower temperatures, the SMR amplitude decreases by a factor 2 compared to room temperature, consistent with measurements in YIG/Pt [20] and (In, Y) doped GdIG/Pt bilayers [14]. In a narrow temperature range around the compensation temperature (\(T_{comp} = 268\) K determined via SQUID magnetometry), the SMR decreases to zero. A similar behaviour has been observed in (In, Y) doped GdIG [14] and was attributed to the formation of the canting phase in GdIG close to the compensation temperature, where the sublattice magnetizations are no longer collinear. In Ref. [14] a sign change of the SMR was observed and interpreted as a perpendicular alignment of the sublattice moments with respect to the external field. The absence of such a sign change in the present data may be attributed to magnetic domain formation for temperatures close to \(T_{comp}\), such that on average the SMR modulation vanishes. However the broadening of the dip in the SMR vs. temper-
ature curves with increasing magnetic field (see inset of Fig. 1 (d)) is in good agreement with the observations in Ref. [14] and is expected for the magnetic canting phase in GdIG [15]. As reported in Ref. [14], in the collinear phase the SMR in Fig. 1 (d) hardly depends on the applied magnetic field. This is consistent with the established SMR model [18, 27], where only the orientation of the sublattice moments is relevant [14, 28].

The angular dependence of the non-local response $V_{nl}$ at 300 K is shown in Fig. 1 (c) for different magnetic field strengths. The non-local voltage signature has the same dependence on the external magnetic field orientation as in previous MMR measurements in YIG/Pt nanostructures for the same wiring scheme [2]. The MMR effect amplitude extracted as $\Delta V_{nl} = V_{nl}(0^\circ) - V_{nl}(90^\circ) = -V_{nl}(90^\circ) > 0$ is plotted as a function of temperature in Fig. 1 (e) for fields up to 7 T. In contrast to the local SMR, where no substantial field dependence is observed in the collinear phase, the non-local MMR signal displays a more complex field and temperature dependence. We have therefore taken additional non-local data in fields up to 15 T for temperatures between 150 K and 300 K, as shown in Fig. 1 (e). We first focus on the data taken at 1 T (grey open squares in Fig. 1 (e)). At low temperatures $\Delta V_{nl}$ vanishes, similar to recent observations in YIG/Pt [2] where a $T^{3/2}$ dependence of $\Delta V_{nl}$ was observed. This temperature dependence in YIG is in agreement with theoretical expectations based on the magnon density of states and distribution function [3, 4, 29], i.e. general properties of the magnonic system. At low temperatures, we therefore expect GdIG to behave very similar to YIG.

With increasing temperature, the non-local signal measured at 1 T increases up to 150 nV just below the compensation temperature. The MMR amplitude then drops below the experimental resolution of about 5 nV [2] at $T_{comp}$ and recovers a finite value above $T_{comp}$, similar to what is observed in the local SMR (see Fig. 1 (d)). We attribute the vanishing MMR signal at $T_{comp}$ to the change of the magnetic structure of GdIG into the canted phase. A vanishing non-local signal in the canting phase suggests two possible scenarios, either (i) the magnon injection/detection becomes inefficient due to the non-collinear alignment of the magnetic sublattice moments and/or (ii) the damping close to the compensation point is enhanced [30] leading to shorter magnon lifetimes and thus shorter diffusion lengths. Additional magnon scattering effects may arise due to magnetic domain formation in the canting phase as discussed above, which further suppress magnon transport.

We now turn to the magnetic field dependence of the MMR effect. The relative field dependence $\delta(\mu_0 H)$ of the MMR amplitude $\Delta V_{nl}$ is defined as

$$\delta(\mu_0 H) = \frac{\Delta V_{nl}(\mu_0 H) - \Delta V_{nl}(1 \text{ T})}{\Delta V_{nl}(1 \text{ T})}$$

and plotted as a function of temperature in Fig. 2 for $\mu_0 H = 7$ T (green triangles). A negative (positive) value corresponds to a suppression (enhancement) with increasing field. The grey shaded region indicates the temperature range around $T_{comp}$ in which the MMR drops to values close to or even below the noise level, impeding a reliable quantification of $\delta$.

![FIG. 2. Relative magnetic field dependence $\delta(7\text{ T})$ (green triangles) and $\delta(15\text{ T})$ (purple hexagons) of the MMR effect defined by Eq. (1). A negative (positive) value corresponds to a suppression (enhancement) with increasing field. The grey shaded region indicates the temperature range around $T_{comp}$ in which the MMR drops to values close to or even below the noise level, impeding a reliable quantification of $\delta$.](image-url)
is much stronger. Furthermore, the enhancement of the MMR with magnetic field in the temperature range 200 K to 250 K as shown in Fig. 1 and 2 is also not observed in YIG/Pt.

In the following we discuss our experimental observations in terms of the magnon injection efficiency which depends on the available magnon states in the ferrimagnet [29] and is thus related to the thermal magnon population. In YIG, only the lowest energy ferromagnetic-like mode is thermally populated within the studied temperature range [31, 32] (only near room temperature the high energy exchange mode starts to become populated and is expected to affect the SSE in YIG [32]). An external magnetic field shifts the ferromagnetic mode to higher frequencies as the Zeeman gap opens, thereby freezing out the lowest energy magnons. In this picture, the magnetic field suppression observed in SSE experiments in YIG/Pt was interpreted in terms of the importance of low energy magnons at elevated temperatures [33, 34]. The suppression of the detected MMR signal in YIG when increasing the external magnetic field at a fixed injector-detector separation [10] is also consistent with a freeze out of magnons from the ferromagnetic mode.

To better understand the observed complex field and temperature dependence of the MMR in GdIG we therefore study the characteristic changes in the GdIG spin wave spectrum as a function of temperature and magnetic field using atomic spin dynamics simulations. The calculations are based on a classical Heisenberg model where the Landau-Lifshitz-Gilbert equation is used to solve the spin dynamics and a Langevin thermostat introduces temperature. Details of the model including the parameters used for GdIG can be found in Refs. [14, 22]. The spin wave spectrum is calculated from the space-time Fourier transform of the spin fluctuations. The spin wave modes in ferrimagnets have a polarization which describes the sense of rotation with respect to an applied field [22], i.e. counterclockwise/clockwise, which is encoded by a red/blue coloring in the figures. We label the modes α - the lowest frequency dispersive mode at 75 K and β - the parabolic optical mode with opposite polarization (see Fig. 3(a)). The modes move in frequency space with temperature and field and change polarization across the compensation point at $T_{\text{comp}} = 310 \text{ K}$, but we retain the same α, β designations throughout. Note that $T_{\text{comp}}^\text{sim} = 310 \text{ K}$ is higher than the value $T_{\text{comp}} = 268 \text{ K}$ experimentally observed in our sample. The flat, broadened bands around 1 THz do not contribute significantly to transport because of their small group velocity and large linewidth. This was confirmed by the temperature dependence of the spin Seebeck effect in GdIG/Pt bilayers [22]. As detailed in Ref. [22], the SSE can be understood considering that the transport of thermal magnons in GdIG is dominated by the α and β magnon modes. At low temperatures only the α-mode is thermally populated (red mode in Fig. 3 (a)), while with increasing temperature the β-mode shifts below $k_B T$ (blue mode in Fig. 3 (c)) and dominates the SSE, leading to a low temperature SSE sign change [22]. At $T_{\text{comp}}$ the orientation of the magnetic sublattices is inverted and consequently the two magnon modes exchange roles (and polarization), as shown in Fig. 3(c) and (e), leading to an abrupt second sign change of the SSE signal at the compensation point [22]. These sign changes can be understood considering that magnons with opposite polarization carry opposite angular momentum. The FMR-like magnons with positive polarization (red modes) reduce the net magnetization. The gapped magnons at higher frequencies arise from the precession of magnetic sublattice moments in the exchange field provided by the other sublattices, similar to the excitations in an antiferromagnet [35, 36]. Magnons with negative polarization (blue modes) correspond to the precession of sublattice moments aligned antiparallel to the external field, such that their excitation increases the net magnetization. The MMR arises
from transfer of angular momentum from the electron to the magnon system (and vice versa)\cite{20}, implying that magnons with opposite polarization (carrying opposite angular momentum) may also lead to an opposite sign of the detected MMR signal. Since we observe no such sign change in the MMR in Fig. 1(e), we conclude that either both modes contribute with the same sign or that the red mode dominates the MMR for the Pt strip separation of 200 nm and the temperatures studied here.

The contributions from different modes can be distinguished by studying the field and temperature dependence of the MMR. The frequency values at \(k = 0\) for the \(\alpha\) and \(\beta\) modes from Fig. 3 are compiled in Fig. 4. The orientation of the sublattice magnetizations with respect to the external field \(H\) is depicted by arrows for temperatures above and below the compensation point \(T_{\text{comp}}^{\text{sim}} = 310\) K.

At low temperatures (Fig. 3(a) and (b)), the \(\beta\)-mode is not populated and does not contribute to the spin transport. The applied magnetic field freezes out the \(\alpha\)-magnons, thereby reducing the MMR signal (see Fig. 1(e) and 2). With increasing temperature, the exchange gap of the \(\beta\)-mode decreases (see Fig. 3(c) and 4) due to the thermally induced disorder in the Gd system, leading to a shift of the exchange mode to lower frequencies \[22\]. Below \(T_{\text{comp}}\), this trend is partly reverted by a magnetic field that increases the Gd order and thereby increases the \(\beta\)-mode frequencies again (see field dependence of the blue lines in Fig. 4). The spin current carried by magnons with opposite polarization reduces the detected MMR amplitude. A magnetic field induced depopulation of the blue \(\beta\) magnons below \(T_{\text{comp}}\) should therefore enhance the MMR signal. Although the frequency shift of the beta-mode predicted by simulations is small compared to the Zeeman shift of the alpha-mode, such a trend is indeed observed in the experimental MMR data displayed in Fig. 1 between \(T_{\text{cross}}\) and \(T_{\text{comp}}\).

Above the compensation temperature, the Gd moments are oriented antiparallel to the external field and depolarize when the latter is increased \[14\], which implies a frequency reduction by magnetic field. In other words, the red (blue) mode shifts to higher (lower) frequencies and is depopulated (populated) with magnetic field. The observed strong suppression of the MMR above \(T_{\text{comp}}\) in Fig. 1 and 2 is consistent with this picture.

The study of the magnon spectrum therefore helps to understand the field and temperature dependence of the MMR in GdIG. While for a quantitative analysis additional factors such as the magnon transport properties need to be carefully considered, our results suggest that the magnon population plays an important role for the MMR effect.

In summary, we measured the non-local magnon mediated magnetoresistance effect (MMR) in the collinear as well as in the canted phase of the compensated ferrimagnet GdIG. The data taken close to the compensation temperature suggest that the MMR is suppressed in the canted phase, either due to inefficient magnon injection or to magnetic domain formation. The MMR signal is furthermore suppressed by magnetic field at low temperatures and above \(T_{\text{comp}}\), but is enhanced just below compensation. We relate these changes to the magnon spectrum of GdIG by comparison with atomistic simulations. We can qualitatively explain the field and temperature dependence of the MMR in terms of a competition between magnon modes with opposite polarization, confirming the importance of the magnon modes and their thermal population for spin transport in magnetic insulators.

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