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Incoherent Spin Pumping from YIG Single Crystals

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Abstract

Ferromagnetic resonance (FMR) in YIG/Pt with a suitable YIG thickness in the sub-mm range show pure spin current contributions from the uniform FMR (UFMR) mode and various spin wave resonance (SWR) modes, each with a different absorption intensity, inverse spin Hall voltage, and temperature dependence. The higher order mode of SWR with *non-zero moment transferring* along the thickness direction induces a maximum DC voltage among all modes. The spin current driven by UFMR and SWR has a linear T and power-law T dependence respectively. In addition, the mixing conductance is derived for both modes, indicative of power law T-dependences. There is an absence of evidence for coherent spin pumping in YIG/Pt.

The exploration of pure spin current has a long and illustrious history from the pioneering experiments in the 1970's [1-3] to the intense interest in pure spin current phenomena and devices in recent years [4-7]. A pure spin current ($\boldsymbol{j_S}$) transports spin angular momentum with high efficiency using a minimal number of charge carriers in a metal and no carriers at all in an insulator. Only a few mechanisms, including the spin Hall effect (SHE) [1], spin pumping (SP) [2,3], longitudinal spin Seebeck effect (LSSE) [4], and lateral spin valve (LSV) [5], can generate a pure spin current. The detection of a pure spin current is often accomplished via the inverse spin Hall effect (ISHE) [6] in a normal metal (NM) with strong spin-orbit coupling (e.g., Pt) to convert a pure spin current to a charge current or more often as an electric voltage. The ISHE converts a pure spin current $\boldsymbol{j_S}$ with spin polarization $\boldsymbol{\sigma}$ into a charge current $\boldsymbol{j_C}$ in a direction perpendicular to both $\boldsymbol{j_S}$ and $\boldsymbol{\sigma}$, and detected as a DC ISHE voltage [6]. Because the detection of $\boldsymbol{j_S}$ is accommodated indirectly via the ISHE by means of a voltage measurement, it is essential to ascertain the measured voltage is exclusive to spin-to-charge conversion and not other parasitic effects [8,9].

Among the mechanisms for generating pure spin current, SHE, LSSE and LSV are intrinsically incoherent, whereas SP administered by ferromagnetic resonance (FMR) in a ferromagnetic (FM) material, offers the unique prospect of *coherent* SP. In fact, since the theoretical proposal of coherent SP in 2002 [7], the coherent SP formalism has been widely used to analyze experimental SP results at room temperature in many NM/FM systems, both FM metals (e.g., permalloy) [10] and FM insulators (e.g. YIG) [11], to extract various parameters, such as spin Hall angle θ_{SH}, spin diffusion length λ_{SF}, and exclusive to coherent SP, the spin mixing conductance $g^{\uparrow\downarrow}$. However, there are complexities of SP in FM metals due to various parasitic effects (rectification, spin Hall resistance, anomalous Hall effect, and anomalous Nernst effect, etc.) that greatly complicate the analyses as reflected in the large variances among the determined values of θ_{SH} and λ_{SF} of the same NM [8,9]. In contrast, in NM/YIG, most of these complications are absent in FM insulators (e.g. YIG), in which we may address the key question of whether it is coherent spin injection, and explore the relevant experimental evidences.

The coherent SP formalism, a T = 0 K theory [7] predicting a temperature-*independent* spin mixing conductance $g^{\uparrow\downarrow}$[10] , has been widely use to analyze SP experiments conducted mostly at room temperature. Recently, SP results in Pt/YIG covering a wide temperature range from room temperature to low temperatures have been reported [12,13]. Surprisingly, in the SP experiments in YIG(3 μm)/Pt, the value of ISHE voltage V_{ISHE}, due to spin-to-charge conversion from coherent SP, was found to be strongly temperature dependent, and importantly, becomes vanishingly small near T ≈ 0 K. Since the incoherent (e.g., thermal) spin injection contributions are expected to decrease strongly with decreasing temperature and vanish at T = 0 K, one expects substantial coherent SP contributions to remain, but not observed, at low temperatures. These unexpected results have been attributed simply to increased FMR linewidth [12].

The use of YIG thin films might have inadvertently introduced other complications due to unresolved contribution from various spin wave modes. It is well established that the FMR microwave excites not only the uniform FMR (UFMR) mode but also various spin wave resonance (SWR) modes, both bulk and surface modes, which also inject pure spin current [11,14,15] and give rise to the ISHE voltage. The resonance fields of these various SWR modes are clearly resolved in YIG of suitable thicknesses but not in YIG thin films, where the UFMR mode and various SWR merge into one resonance peak. The SWR modes in fact can generate pure spin current with a higher efficiency than the uniform UFMR mode in YIG [14], which although may be beneficial for some magnonic devices [11,16], further escalates the complexity in addressing the issue of coherent SP and spin-to-charge conversion. These issues can best be addressed in SP measurement using YIG single-crystals of appropriate thicknesses where all contributions are clearly separated.

We investigate SP in YIG/Pt, using single crystal YIG plates with suitable thicknesses in the sub-mm range. In addition to the UFMR mode, higher order SWR modes can be clearly resolved. Instead of only the modes of in-plane wave vector with $k_z = 0$ as observed in thin films, the higher order SWR modes with $k_z = \pi/d_z$ are clearly resolved and identified in single crystals. This in-depth study of the temperature dependence of each resonance mode and the UFMR mode shed light on the mechanism of SP and the evidence for coherent SP.

The experimental setup for our SP measurements is schematically shown in Fig. 1(a). We use two (111)YIG single crystal plates 1.3 mm x 1.3 mm in size and thickness of d_z = 70 μm and 200 μm. These large thicknesses are necessary to resolve all the SWR modes. The DC magnetic field (*H*) and the microwave field are applied in the *x* and the *y* direction, respectively. A Pt thin film of 10 nm in thickness is deposited on the surface of (111)YIG single crystal plates to measure the ISHE voltage as a result of spin-to-charge conversion of the spin current. Two Platinum wires are attached at the two opposite edges of the Pt thin film in the *y* direction to measure the DC voltage using a KEITHLEY 2182A nanovolt meter. A Bruker EMX system is used to provide the X-band microwave with a cavity of TE_{102}. The microwave frequency for room temperature and low-T experiments are 9.8 and 9.45

GHz respectively. The samples are placed with a 10 mm offset from the center-line of zero electric field of cavity to obtain the optimal signal.

Bulk modes and surface modes are two types of spin waves in a YIG crystal. The bulk (B) modes are the backward-volume magnetostatic spin waves [14,17], whereas the surface (S) modes are the Damon–Eshbach surface spin waves [14,18]. Both the B modes and the S modes depend on the thickness d_z of the sample as shown in Fig. 1(b), where we show the calculated thickness dependence, from thin film to 300 µm, of the internal field H_i using the parameters of d_x = 1.3 mm of our samples, and ν_{MW} = 9.8 GHz and $4\pi M_s$ = 1750 emu/cm^3 for YIG [19]. The experimentally obtained value of H_r may be higher than H_i for a thick plate due to the thickness dependent demagnetization field. The solid blue curves are the solutions for the magnetostatic standing waves with the wavelength $\lambda_x = 2d_x/n$ for n = 1, 3, 5… and k_z = 0, denoted as the B1, B3, B5… modes respectively. The red dash curves are the solutions with $\lambda_x = 2d_x/n$ for n = 1, 3, 5, ... and $k_z = \pi/d_z$, denoted as the L1, L3, L5, … modes. For comparison, a constant H_i value for the UFMR mode is also shown. The solid squares in Fig. 1b are the experimentally obtained H_r values of bulk modes for a 70 µm thick bare YIG plate, showing very good agreement with the calculated values of H_i. The results of the thicker plate (200 µm) are in general agreement but follow less closely with the calculated values due to the substantially non-uniform demagnetization field in a thick crystal.

In Figure 2 (a) and (b) we show the microwave absorption and the DC voltage spectrum, respectively, in the Pt(10 nm)/YIG(200 µm) from 2500 to 3500 Oe covering all the essential resonances. The vertical solid and dashed lines are drawn to indicate the positions of each resonance mode. The UFMR mode commands the most absorption, followed by the S modes on the small field side, and the B and the L modes on the large field side (Fig. 2a). However, not just the UFMR mode, *every* SWR mode *injects* pure spin current and *exhibits* ISHE DC voltages [11,14] as shown in Fig. 2b. Although UFMR commands the most absorption, it does not generate the largest voltage, in agreement with the previous results [14]. As shown in Fig. 1b, for thinner YIG slabs and especially in thin films, all the SWR modes and UFMR mode are so close to each other and merge into a single peak thus indistinguishable. The ISHE voltage for the YIG thin films show only one unresolved peak, but with unspecified contributions. For 200 µm YIG plate, the sufficient separation between modes allows both FMR and DC voltage spectra fitted with multiple Lorentz functions as the solid curves plotted in Fig. 2 (a) and (b). The highest voltage is not that of UFMR but L1 (n = 1 and $k_z = \pi/d_z$), which is hardly resolved in the absorption spectrum with low intensity. The L3 mode is clearly seen in both the absorption and the voltage spectra; while L7 and higher order modes are relatively weaker in both spectra. Note that L5 overlaps with B3, which was also predicted in Fig. 1(b) as the interception of the B1 and the L5 curves at 200 µm. The efficiency of ISHE voltage is clearly k_z dependent.

In Fig. 2b, the measured voltages roughly scale with the microwave power from 10 mW to 20 mW. In Fig. 3a we show the actual dependence of the measured ISHE voltages in Pt(10 nm)/YIG(200 µm) of several main modes on the microwave power. The measured voltages of all modes increase monotonically with increasing microwave power but not with the same dependence. The voltage linearly increases with increasing power for all modes, except L1 and most clearly UFMR, which saturates at high power. According to the previous report on YIG thin films, the saturation behavior is attributed to the spin back-flow [11].

Although the spin rectification induced DC voltage is negligible in insulating

YIG due to its high resistance, the microwave heating effect is inevitable [20]. Therefore, the contribution from microwave heating should be subtracted from the total voltage in order to obtain the actual ISHE voltage V_{ISHE} due to SP. The heating effect can be separated from the total voltage by reversing the field direction because it is a field-independent contribution while the spin current contribution depends on the field direction [21]. Fig. 3(b) shows the power-dependence of the L1-driven voltage with positive (θ = 0°) and negative (θ = 180°) magnetic field. The thermal contribution can be obtained as $(V_0+V_{180})/2$, which is around 20% of the total voltage at the power of 10 mW. It is interesting to note that the power dependence of the heating-induced voltage is different from that of V_{ISHE} which is $(V_0-V_{180})/2$. The heat-induced voltage is a linearly dependent on microwave power, whereas V_{ISHE} is non-linear and saturates at high power. The solid line in Fig. 3(c) displays the ISHE spectrum calculated from V_0 and V_{180} spectra plotted with squares and circle respectively. Lorentz function is applied to decompose the V_{ISHE} for each UFMR and SWR mode. The spin current density, j_s, can be derived from V_{ISHE} with following relationship: [22]

$$j_s = \frac{d_N \sigma_N}{w \theta_{SHE} \lambda_{SD} \tanh(d_N / 2\lambda_N)} \left(\frac{\hbar}{2e} \right) V_{ISHE} \quad (1)$$

Here, w, d_N, σ_N, λ_{SD}, and θ_{SHE} are the separation between electrodes, thickness, conductivity, spin diffusion length, and spin Hall angle of Pt respectively. In this work, w, d_N, and σ_N are the measured values of 1.3 mm, 10 nm, and 1.08×10^6 Ω^{-1}m^{-1} respectively. The values of λ_{SD} and θ_{SHE}, adopted from the literature, are 7.7 nm and 0.012 respectively. Following the result of j_s, the effective spin mixing conductance $g^{\uparrow\downarrow}_r$ at the interface can be estimated as: [22]

$$g_r^{\uparrow\downarrow} = \frac{8\pi\alpha_G{}^2\left[(4\pi M_s\gamma)^2 + 4\omega^2\right]}{\gamma^2 h^2 \hbar\left[4\pi M_s\gamma + \sqrt{(4\pi M_s\gamma)^2 + 4\omega^2}\right]} j_s \quad (2)$$

Here the damping constant of YIG α_G is $1.7\text{x}10^{-3}$[22], magnetic magnetization $4\pi M_s$ is 0.176 Tesla, γ is 1.76×10^{-11} $T^{-1}s^{-1}$. The microwave field at 20 mW is $h = 5.29\text{x}10^{-3}$ mT. Substituting eq. (1) into (2), the effective spin mixing conductance at room temperature for our sample is 2.1×10^{18} m^{-2} which is quantitatively comparable to the value obtained for thin film bilayer of YIG/Pt with well controlled interface (1.3×10^{18} m^{-2}).[22]

We next address the temperature dependence. The voltage spectra at various temperatures from 106 K to 300 K are shown in Fig. 4a. With decreasing temperature, the locations of all modes shift to lower fields due to a larger YIG magnetization. The dashed lines mark the positions of the five main resonance modes (S1, FMR, L1, L3, and B1) at various temperatures, which are corresponding to the microwave absorption spectra in Fig. 4(b). Immediately apparent is the strongly decreasing resonance intensity for all modes. The ISHE signal induced by the B and L modes becomes so weak what they are barely observable at 150 K, while the UFMR induced voltage persists to somewhat lower temperatures. It is also apparent that the rapid decrease in intensity is *not* accompanied by a proportionally large increase in linewidth because the mechanism of line broadening of YIG in low temperature is impurity driven scattering [23] and inhomogeneity[24], instead of spin pumping enhanced damping.

To compensate for the T-dependent resistivity of Pt as shown in the inset of Fig.4 (d), we show the T-dependences of j_s, instead of V_{ISHE}. Apparently, the UFMR-driven j_s decreases linearly with decreasing temperature in contrast with the power-low T-dependence in other three SWR modes (L1, L3, and B1) that can be well described by a power law of T^a with a = 2.0 ~ 2.5, indicating a different SP mechanism for SWR than that in UFMR. The linear T-dependence of j_s from UFMR mode is qualitatively consistent with what was observed in YIG(3 μm)/Pt [12] and YIG(40nm)/Pt bilayers with the measurement down to 20 K and 50 K respectively. The fact that V_{ISHE} approaches zero at low temperature might be attributed to the T-dependence of magnetization or magnetic damping in YIG.[13] Therefore, the T-dependencies of spin current should be converted to T-dependences of mixing conductance to remove the contributions from the changes of magnetization and damping constant. However, the T-dependency of spin mixing conductance is hard to derive in most YIG/Pt bilayers since the linewidth of YIG is highly contributed by impurity scattering [23] and inhomogeneity[24]. And the intrinsic damping constant, α_G, is implicit in the spin pumping experiments with fixed microwave frequency. Recently, the broadband FMR experiment on a 300μm YIG sphere revealed a linear decrease of intrinsic damping constant with cooling, which is related to magnon-phonon process [24] and provides a reference for T-dependent intrinsic damping. Applying the obtained T-dependent magnetization (M_s) as seen in the inset of Fig 4(e), the T-dependent spin mixing conductance can be derived with Eq. (2) as plotted in Fig. 4(e) and Fig. 4(f) for UFMR and SWR respectively. Both show a power-law relationship with T, indicating the non-coherent spin pumping.

It is noted that spin Seebeck effect can also generate inverse spin Hall voltage in Pt, where a temperature gradient injects spin current. However, the T-dependent SSE in YIG/Pt has a critical transition near 40 K for YIG bulk crystal, [24] which is defined as the cutoff temperature dividing the thermal magnons (T > 40 K) and the subthermal magnons (T < 40 K). For thermal magnons (T > 40 K), SSE signal enhances with decreasing T which is opposite to the trend of ISHE voltage. In addition, the heating power changes as the absorption power P decreases with decreasing T. Since the SSE does not increase significantly above 100 K, when P decreases, the ISHE voltage may decrease correspondingly. However, the reduction of microwave absorption intensity is only around 50%, which is much smaller than reduction rate of ISHE voltage. Therefore, we consider the SSE effect does not play a significant role in the T-dependence of spin pumping effect of YIG/Pt.

Based on the model of magnon-mediated non-coherent SP, both the magnon-to-charge convertibility at the interface of YIG/Pt [25,26] and the magnon spin conductivity in YIG [27] are temperature dependent. The former has a linear temperature dependency (a =1), but the latter has been estimated as a ~ 2/3. Therefore, the linear T-dependence for UFMR-driven SP may be mainly due to the temperature dependence of magnon-to-charge convertibility for the uniform mode at the interface. However, the fitted experimental result of a = 2.0 – 2.5 for SWR-driven SP is more difficult to account. Theoretically, a ~ 1.7 seems to be a reasonable expectation for the SWR-driven SP since the thermal effect on magnon transport may be a product of $T \cdot T^{2/3}$, which gives $a \approx 1.7$. The experimental result of $\alpha > 1.7$ suggests the existence of other contributions to the temperature dependence of SWR-driven SP, and warrant further investigation.

Since the theoretical proposal in 2002, coherent SP has been widely used to analyze experimental SP results at room and other temperatures. Coherent spin pumping, a T = 0 K phenomenon, should exist at all temperatures. However, SP also excites large contributions from other magnon modes. When thin YIG samples are used in SP as in previous studies, various spin wave modes and the potential coherent spin pumping signals are inseparable. Since the spin wave modes are overwhelming, the question of coherent SP cannot be addressed. We show in this work, when YIG sample of an appropriate thickness d_z has been used, all the spin wave resonance (SWR) modes and the UFMR can be resolved and their temperature dependences separately followed. Our results show all SWR modes *and* UFMR vanish at low temperatures. We have found *no* evidence of coherent SP in YIG.

In conclusion, the SWR and UFMR-driven spin pumping effects in YIG/Pt using thick YIG single-crystal slabs of 70 and 200 µm have been investigated with a Pt thin layer as a spin current detector. The series of SWR modes are clearly observed in the resonance spectra. It is a new finding that the SWR mode with $k_z = \pi/d_z$ has much higher efficiency of spin injection in comparison with UFMR mode and other SWR modes with $k_z = 0$. The UFMR mode shows the largest microwave absorption, generates a modest ISHE voltage, but has the lowest efficiency in spin current pumping. Furthermore, the experimental results show that the spin current driven by both UFMR and SWR has strong temperature dependencies, reflecting the nature of incoherent spin pumping. We have found no detectable evidence of coherent SP. The result of distinct T-dependencies of mixing conductance derived from UFMR and SWR spin pumping further suggests that the routes of spin current transport in YIG may be different for these two modes.

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Caption

Fig.1 (a) Schematic description of the spin pumping experiment in Pt/YIG showing the surface and bulk modes standing waves and Pt electrodes for the voltage measurement. (b) The analytical solutions of the thickness dependence of the bulk standing wave modes (B and L) in a YIG thin plate. Lines indicates the solutions of $k_y = k_z = 0$ (blue solid lines) and $k_y = 0$ with $k_z = \pi/d_z$ (red dashed lines). The squares denote the experimentally obtained resonance fields for the B modes in the YIG crystal with $d_z = 70$ μm.

Fig. 2 (a) Absorption spectrum and (b) DC voltage in Pt(10 nm)/YIG(200 μm) in the field range of 2500 to 3500 Oe. Solid curves are Lorentz functions to decompose the spectrum.

Fig. 3 (a) DC voltage obtained from FMR (open squares) and various S1, L1, L3, B1, B3 SWR modes (closed symbols) a function of microwave power. (b) Voltages of the L1 mode with positive H (V_0, closed triangles) and negative H (V_{180}, open triangles), and calculated results for ISHE [$(V_0 - V_{180})/2$] and heating effect [$(V_0 + V_{180})/2$]. (c) V-spectra for V_0 (closed squares), V_{180} (open circles) and the calculated ISHE voltage (solid line). The dash lines are the Lorentz fitting for ISHE curve.

Fig. 4 (a) and (b) are ISHE and microwave spectra respectively at temperatures between 110 K and 300 K with dashed lines linking the position of each mode; Temperature dependence of spin current from (c) the FMR mode and (d) the L1, L3, B2 SWR modes in the range of 106 to 300 K. T-dependent resistance of Pt is shown in the inset of (c). (d) and (e) the calculated T-dependency of spin mixing conductance of UFMR and SWR modes, respectively. Inset of (e) is the T-dependent saturated magnetization.