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Enhanced spin pumping damping in yttrium iron garnet/Pt bilayers

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Detailed measurements of the magnetic relaxation expressed in the linewidth of the ferromagnetic resonance (FMR) absorption in thick films of yttrium iron garnet (YIG) and in YIG/Pt bilayers carried out at room temperature reveal a very large increase in the relaxation rate with the deposition of a Pt layer. The additional relaxation increases linearly with the microwave frequency characteristics of the spin pumping mechanism. The value of the spin mixing conductance obtained from the data is one order of magnitude larger than the largest possible value determined from measurements of the voltage generated by FMR spin-pumping. © 2013 American Institute of Physics.

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One of the fundamental properties of a magnetic system is the manner by which its magnetization relaxes towards equilibrium. This is governed by the spin interactions and the structure of the magnetic system and its detailed understanding is important from the point of view of basic physics and for technological applications. For several decades, the magnetic relaxation has been investigated experimentally in bulk and thin film materials mainly by measuring the linewidth of the ferromagnetic resonance (FMR) at microwave frequencies. In bulk magnetic insulators, the relaxation occurs through intrinsic mechanisms involving magnon-magnon and magnon-phonon processes as well as extrinsic mechanisms such as scattering by impurities.^{1,2} In bulk metallic materials, the relaxation is dominated by processes involving the conduction electrons.³ In very thin films and multilayers, new physical relaxation processes have been discovered in the last decade, the most important ones being two-magnon scattering off the irregularities at the surfaces or interfaces^{4,5} and the spin pumping mechanism.^{6,7} These processes contribute with additional relaxation rates that increase as the thickness of the ferromagnetic film decreases and are known to dominate the damping in ultra-thin films.⁸

In recent years, structures made of bilayers of ferromagnetic metal (FM)/normal metal (NM) films have attracted considerable interest due to the discoveries of the spin Hall effect (SHE)^{9,10} and the inverse spin Hall effect (ISHE).^{11,12} In a FM/NM bilayer undergoing FMR driven by microwave radiation, it has been found^{11–14} that the precessing spins in the FM inject spins into the adjacent NM layer creating a spin current that is converted into a charge current by means of the ISHE resulting in a spin-pumping dc voltage V_{SP} opening immense possibilities in the field of spintronics.¹⁵ A very important recent development in this field was the demonstration that the ferrimagnetic insulator yttrium iron garnet (YIG) can be used in FM/NM structures to study spin-charge current conversion.¹⁶ Since in YIG the relaxation mecha-

nisms involving conduction electrons are not effective, its FMR linewidth is two orders of magnitude smaller than in FM metals such as permalloy (Py). As a result, the FMR and the V_{SP} spectra exhibit^{17,18} many peaks corresponding to the spin-wave magnetostatic modes.^{19,20} Recently, it has been observed that the deposition of a Pt layer on single crystal YIG films produces an unusually large broadening of the microwave absorption lines.^{17,18}

In this letter, we present a detailed study of the magnetic relaxation in YIG/Pt bilayers measured by the linewidths of the FMR spectra. The experiments were done with many samples consisting of a single-crystal YIG film with thickness in the range 8–28 μm , grown by liquid-phase epitaxy (LPE) on 0.5 mm thick (111) gadolinium gallium garnet (GGG) substrates and cut in an approximate square shape with lateral dimensions of 2 to 4 mm. Measurements were made with bare YIG films and in YIG covered with a Pt layer with thickness in the range 6–20 nm deposited by magnetron sputtering. Two different arrangements were used to measure the FMR linewidths. In one arrangement, the sample is mounted on the tip of a plastic rod inserted in the middle of a shorted rectangular waveguide to allow measurements as a function of the angle. We do not use a microwave cavity in order to avoid nonlinear or heating effects. The setup is placed between the poles of an electromagnet of a homemade FMR spectrometer operating at a fixed X-band microwave frequency so that the static and microwave magnetic fields are kept perpendicular to each other. The applied field is modulated with a 1.1 kHz ac component generated by a pair of Helmholtz coils to allow lock-in detection. Field sweep measurements were made to obtain spectra of the microwave absorption derivative at several values of the angle θ_H between the field \vec{H} and the normal to film plane to investigate the out-of-plane angle dependence of the spectra. In the other arrangement, a high bandwidth coplanar microstrip waveguide setup is used to measure the FMR spectra at several frequencies in the range 1–12 GHz with the field applied in the film plane.

All samples investigated exhibited similar behavior. The FMR linewidths in the bare YIG films are less than 1 Oe

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with the external field applied either parallel or perpendicular to the film plane. After deposition of a 6 nm to 20 nm Pt layer, the linewidths increase to several Oe. We present here only some representative results obtained with input microwave power less than 10 mW to avoid nonlinear or heating effects. Figure 1 shows the field derivative dP/dH of the microwave absorption spectra for a 28 μm thick YIG film obtained with the field parallel and perpendicular to the film plane, measured with the rectangular waveguide at a fixed microwave frequency of 9.4 GHz. The spectra in Figs. 1(a) and 1(b) obtained before deposition of the Pt layer allow a clear identification of the absorption lines. They correspond to standing-wave magnetostatic spin-wave modes that have quantized in-plane wave numbers due to the boundary conditions at the edges of the film.^{17–20} The strongest line corresponds to the FMR mode that has frequency given by $\omega_0 \approx \gamma H^{1/2} (H + 4\pi M)^{1/2}$, where γ is the gyromagnetic ratio ($2\pi \times 2.8$ GHz/kOe for YIG) and $4\pi M$ is the saturation magnetization (1.76 kG at room temperature). In Fig. 1(a) with the field in the plane, the lines to the left of the FMR correspond to hybridized surface modes, whereas those to the right are hybridized volume magnetostatic modes.^{17,18}

As shown in Figures 1(c) and 1(d), the deposition of a 6 nm thick Pt layer on the YIG film produces a pronounced broadening of the lines for all modes. They have nearly the same peak-to-peak linewidth $\Delta H_{pp} \approx 5.4$ Oe, corresponding to a half-width at half-maximum (HWHM) $\Delta H = \sqrt{3}\Delta H_{pp}/2 \approx 4.6$ Oe, which is nearly ten times larger

than in bare YIG. In order to investigate the source of the linewidth broadening, we have done measurements of the FMR spectra varying the direction of the applied field out-of-the plane. Figure 2 shows the out-of-plane angle dependence of the FMR linewidths measured in three samples with YIG film thickness 8, 15, and 28 μm , the first two with a 20 nm Pt layer and the third with a 6 nm Pt layer. While the bare YIG films show a small change in the linewidth with variation of the angle between the field and the plane from $\theta_H = 0$ to 90° , the films with a Pt layer exhibit a clear angle dependence with a maximum at $\theta_H \approx 60^\circ$. The error bars in Fig. 2 correspond to the range of the linewidths measured in the three cleanest mode lines. Measurements made with other samples with different thicknesses of the YIG film and the Pt layer exhibit similar behavior. We do not observe any change in the spectra as the microwave power varies below 10 mW.

Figure 3 shows the FMR (HWHM) linewidth for a bare YIG (8 μm) film and a bilayer YIG (8 μm)/Pt (8 nm) as a function of frequency, measured with the broadband microstrip setup. Again the error bars correspond to the range of the linewidths measured in the three cleanest mode lines. While the bare YIG film has linewidth smaller than 1 Oe and with little variation with frequency, the YIG/Pt bilayer has clearly larger linewidth that increases linearly with frequency.

A possible source of the line broadening with Pt deposition is an enhancement in the interface two-magnon scattering relaxation similar to the one observed when an antiferromagnetic layer is deposited on a FM film.⁵ Note that in the range

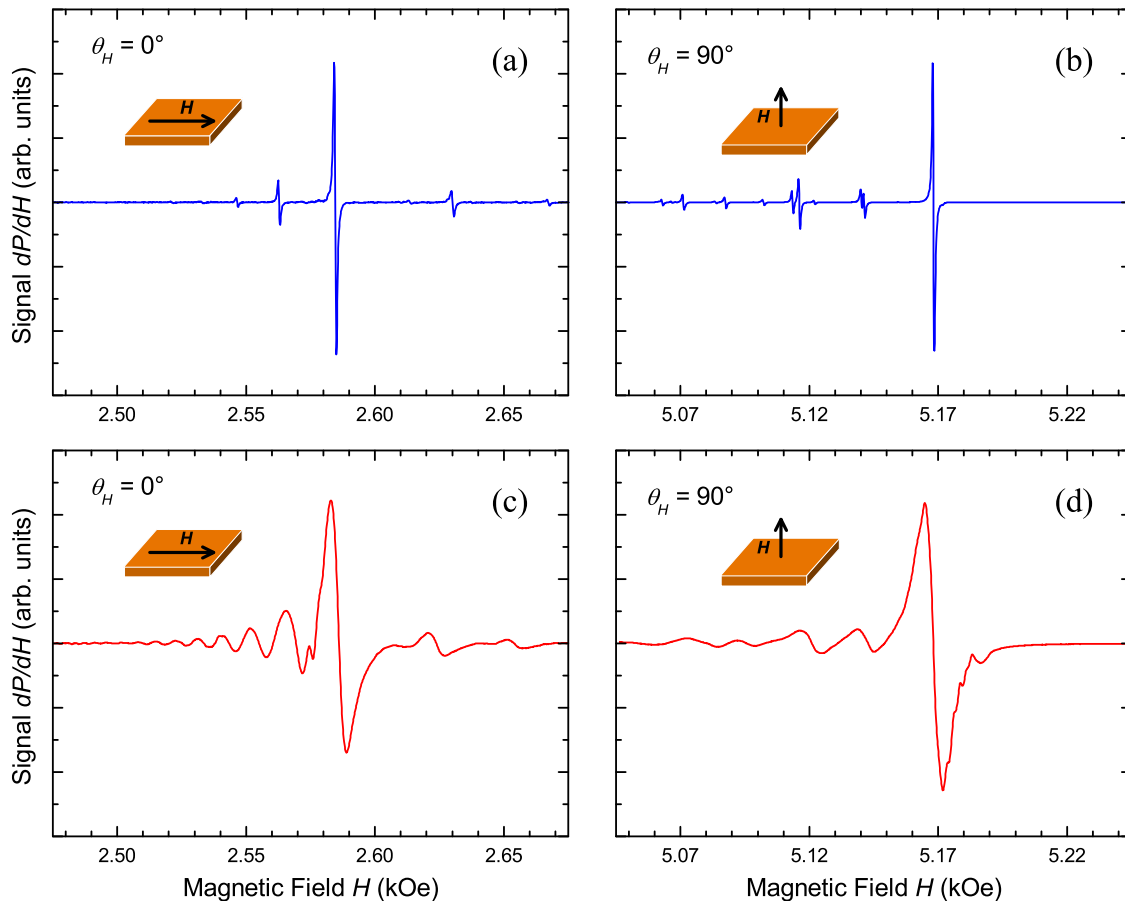


FIG. 1. Field scan microwave absorption spectra at a frequency 9.4 GHz of 28 μm thick YIG film with lateral dimensions 2×3 mm with the magnetic field applied parallel or perpendicular to the film plane as indicated. In (a) and (b), the YIG film is bare, while in (c) and (d), it is covered with a 6 nm Pt layer.

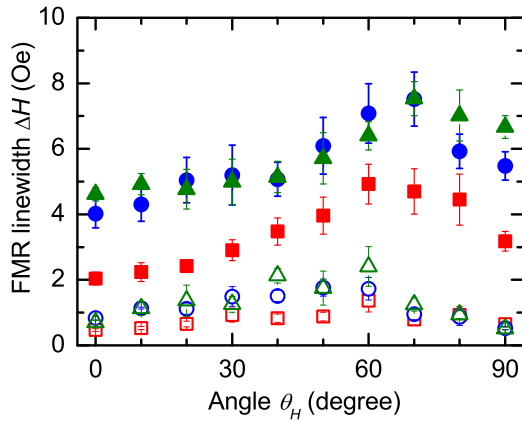


FIG. 2. Variation of the ferromagnetic resonance linewidth with the out-of-plane angle of the applied magnetic field in YIG films (open symbols) and in YIG/Pt bilayers (solid symbols). Open squares, circles, and triangles are the data for 8, 15, and 28 μm thick YIG films, respectively. Solid squares, circles, and triangles are the data for 8, 15, and 28 μm thick YIG/Pt with 20, 20, and 6 nm thick Pt layers, respectively.

of frequencies investigated the two-magnon linewidth varies linearly with frequency,²² as observed here. However, the shape of the measured out-of-plane angle dependence in Fig. 2 is very different from the one arising from two-magnon scattering. With the field normal to the plane, the FMR ($k=0$) mode can decay, so that the relaxation due to two-magnon scattering should be small as predicted theoretically²¹ and observed experimentally.²² As it is clear in Figs. 1 and 2, the linewidths in YIG/Pt at $\theta_H = 0$ and 90° are very similar, ruling out the two-magnon scattering as the mechanism responsible for the line broadening.

The only other possible source for the additional damping due to the deposition of a Pt layer on the YIG film is the spin pumping mechanism that relaxes the magnetization by producing a flow of angular momentum out of the FM film into the NM layer.^{6,7} The additional linewidth due to the spin pumping mechanism is given by^{6,7}

$$\Delta H_{SP} = \frac{\hbar \omega g_{eff}^{\uparrow\downarrow}}{4\pi M d_{FM}}, \quad (1)$$

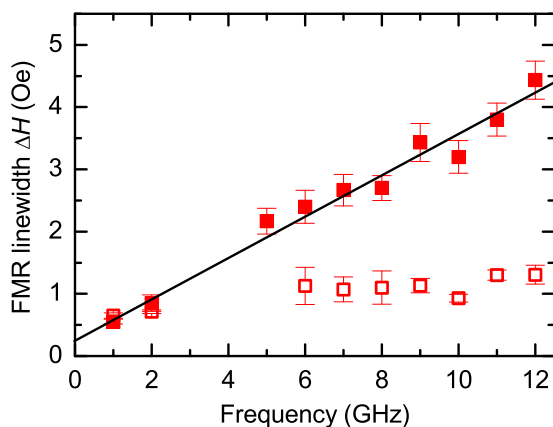


FIG. 3. Variation of the ferromagnetic resonance linewidth with frequency in a 8 μm thick bare YIG film (open square symbols) and in a YIG (8 μm)/Pt (8 nm) bilayer (solid square symbols).

where $g_{eff}^{\uparrow\downarrow}$ is the real part of the effective spin-mixing conductance taking into account the back flow spin current from the spin accumulation and the reflection at the surface of the NM layer, ω is the driving frequency, and d_{FM} is the thickness of the FM layer. The measured linear dependence of the linewidth on the frequency, shown in Fig. 3, is consistent with Eq. (1) and is a well known characteristics of the spin pumping damping. The measured dependence of the linewidth on the YIG film thickness does not follow the $1/d_{FM}$ prediction of Eq. (1). However, we note that the range of thickness investigated, 8–28 μm , is quite small to determine the experimental thickness dependence, considering that the value of the spin-mixing conductance is very sensitive to the interface quality^{23,24} which may vary from one sample to another. With the measured 2.3 Oe increase in the linewidth at 10 GHz (Fig. 3) due to the Pt layer deposition on the 8 μm thick YIG film, we find from Eq. (1) a value for the effective spin-mixing conductance in YIG/Pt of $g_{eff}^{\uparrow\downarrow} = 4.8 \times 10^{16} \text{ cm}^{-2}$. This value is two orders of magnitude larger than the one determined for YIG/Au interfaces from the FMR linewidth measurements,^{23,24} $g_{eff}^{\uparrow\downarrow} = 5.2 \times 10^{14} \text{ cm}^{-2}$, which is consistent with theoretical prediction.²⁵ One may argue that the YIG films investigated in Refs. 23 and 24] were grown by pulsed laser deposition and had a surface roughness of 0.5 nm while our LPE films are atomically flat and it is known^{24,25} that the spin-mixing conductance decreases with interface roughness. On the other hand, the values for $g_{eff}^{\uparrow\downarrow}$ previously reported for YIG/Pt are very discrepant. They were determined from the spin-pumping/ISHE voltage generated by FMR,^{12–14} $V_{SP} \propto g_{eff}^{\uparrow\downarrow} \lambda_{SD} \theta_{SH}$, where λ_{SD} is the spin-diffusion length and θ_{SH} is the spin-Hall angle of the Pt layer. Thus, the determination of $g_{eff}^{\uparrow\downarrow}$ depends on knowledge of the parameters for Pt, which have reported results that disagree by factors larger than 20.²⁶ The value reported in Ref. 16 is $g_{eff}^{\uparrow\downarrow} = 1.8 \times 10^{12} \text{ cm}^{-2}$ whereas the value obtained from measurements of V_{SP} in Ref. 18 using¹⁴ $\theta_{SH} = 0.045$ and $\lambda_N = 3.7 \text{ nm}$ is $g_{eff}^{\uparrow\downarrow} = 8.4 \times 10^{13} \text{ cm}^{-2}$. However, if one uses the parameters $\theta_{SH} = 0.0037$ from Ref. 16 and $\lambda_N = 1.7 \text{ nm}$ from Ref. 26, the V_{SP} data of Ref. 18 yield a value $g_{eff}^{\uparrow\downarrow} = 2.2 \times 10^{15} \text{ cm}^{-2}$. This is the largest possible value of $g_{eff}^{\uparrow\downarrow}$ for YIG/Pt determined from the spin-pumping/ISHE voltage, and it is still one order of magnitude smaller than the value obtained here. Perhaps a more serious problem with the determination of $g_{eff}^{\uparrow\downarrow}$ for YIG/Pt from the V_{SP} measurements is one recently pointed out,²⁷ that when in contact with a thick YIG film, the Pt layer acquires strong ferromagnetic characteristics due to the proximity effect. As a result, its suitability for detecting pure spin currents produced by the spin pumping due to FMR might be compromised due to contamination with the anomalous Hall effect.²⁷ The fact is that the direct determination of the spin-mixing conductance from the measurement of the frequency dependence of the FMR linewidth in thick YIG films covered with a Pt layer leads to a surprisingly large value for $g_{eff}^{\uparrow\downarrow}$. Thus, either YIG/Pt has a much larger spin-mixing conductance than previously determined or there is an enhancement of the spin pumping relaxation when a Pt is in contact with a thick YIG film not present in the usual damping given by Eq. (1).

In conclusion, we have used ferromagnetic resonance linewidth measurements to study the magnetic damping in YIG/Pt bilayers. The deposition of Pt produces a large

increase in the FMR linewidth with an increment that varies linearly with frequency. This should characterize the spin-pumping mechanism as the source of the additional damping. However, the value of the spin mixing conductance obtained from the data is one order of magnitude larger than the largest possible value determined from spin-pumping-ISHE voltage, raising questions about the suitability of the usual spin pumping mechanism in describing the damping YIG/Pt bilayers.

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