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FMR line shape effect on spin pumping in bilayer structures

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Pure spin current induced by ferromagnetic resonance (FMR) excitation in thin-film heterostructures consisting of ferromagnetic (FM) and normal metal (NM) layers is studied as a function of FMR line shape and width. Experiments were carried out with thin films of ferromagnetic $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) grown epitaxially on $NdGaO_3$ substrate and covered with Pt. The spin current injected into the NM layer was measured using the inverse spin Hall effect (ISHE) in the temperature range of 100-350 K. The samples under study revealed different width of the FMR line, which was attributed to inhomogeneous broadening with a specific Voigt shape of the ISHE signal. To take this effect into account, substantial corrections are proposed to the existing theory of spin pumping.

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Keywords: pure spin current, spin pumping, ferromagnetic resonance, ferromagnetic-normal metal thin-film structure, manganite, inverse spin Hall effect, inhomogeneous broadening.

Preface

This paper is a contribution to the MRSej issue dedicated to the 85 years of Prof. B.I. Kochelaev. His classical works made a great impact on the theory of spin relaxation and physics of strongly correlated systems. Thus, though no references are given here to specific Kochelaev's publications, the authors acknowledge great positive influence of his scientific ideas.

1. Introduction

Generation and control of spin currents flowing across an interface between media with different magnetic properties attract recently considerable interest in both scientific and technical aspects related to spintronics, see the review articles [1, 2]. To create a "pure" spin current (not related to any charge flow), the so-called spin pumping (SP) is used in ferromagnet / normal metal (FM/NM) thin-film bilayers [3-6]. SP is implemented by excitation of ferromagnetic resonance (FMR) in the FM layer and injection of the non-equilibrium spin momentum into the NM film. The density of the pure spin current entering the NM layer is

$$\mathbf{j}_s = \frac{\hbar}{4\pi} g_{\text{mix}} \left[\mathbf{m} \times \frac{d\mathbf{m}}{dt} \right] \tag{1}$$

where $\hbar$ is the Planck constant; $\mathbf{m}$ is the unity vector directed along the precessing magnetic moment $\mathbf{M}$ of the FM layer; and g_{mix} is the so-called spin-mixing conductance [4-6].

The pioneering works on spin pumping driven by FMR excitation were published in Refs. [3-5] and then an effective method of measuring the spin current was suggested and implemented [7, 8]. This technique is based on the inverse spin Hall effect (ISHE) which leads to arising an electric charge current

$$\mathbf{j}_{\text{ISHE}} = \theta_{\text{SH}} \frac{e}{\hbar} \left[\mathbf{n} \times \mathbf{j}_s \right] \tag{2}$$

in the NM layer. Here θ_{SH} is the dimensionless spin Hall angle, e is the elementary charge, and $\mathbf{n}$ is a unit vector in the direction of the spin momentum flow (as a rule, normal to the

FM/NM interface). As a result, a d.c. electric voltage ($U^{\rm SP}$) arises across the NM film and can be measured experimentally.

Studies of spin pumping in various materials and structures were performed by many authors, see, for example, Refs. [9-12]. As follows from the theory [9-11], the SP voltage can be presented in the form

$$U^{\rm SP}(H) = CRw\,\frac{f_{\rm L}(H)}{\delta^2} \qquad (3)$$

where R is the electrical resistance between measuring contacts; w is the width of the NM layer; $f_{\rm L}(H)$ is the Lorentzian FMR line shape as a function of the external magnetic field H (normalized to unity at resonance, $H = H_0$); δ is the half-width of this line; and, finally, the temperature-dependent factor C includes all other relevant parameters, such as $g_{\rm mix}$, the ISHE angle, the microwave pumping frequency and power, the spin diffusion length, etc.

It should be emphasized that the FMR line broadening in Eq. (3) is always supposed to be homogeneous (caused by spin relaxation). As a result, the SP voltage measured at resonance (we denote it as $U_0^{\rm SP}$) is expected to be inversely proportional to the line width squared. Until now, this assumption was accepted in most papers where the SP effect was studied. It is known, however, that in many cases the observed FMR width is considerably greater than the homogeneous value. This may be caused by the influence of inhomogeneous broadening which modifies the line shape and leads to a significant change in Eq. (3) and, especially, to a violation of the δ^{-2} dependence. As a result, significant errors may occur in the estimates of the spin conductivity, the diffusion length, and other important parameters affecting the efficiency of spin pumping.

The first attempt to account for this effect was presented in our previous paper [13]. Here we will develop this idea with new experimental material.

2. Materials and methods

Our experiments were carried out with two samples of the bilayer thin-film structure FM/NM where the manganite $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) and Pt were used as FM and NM, respectively. LSMO is known as ferromagnetic metal with Curie temperature $T_{\rm C}$ about 360 K; this material attracts considerable interest due to its unusual properties such as colossal magnetoresistance [14, 15]. Platinum is commonly used for the ISHE detection due to relatively strong spin-orbit coupling typical of heavy elements [1, 16]. LSMO films with the thicknesses of 45 nm (sample S1) and 20 nm (sample S2) were epitaxially grown by RF magnetron sputtering on the (110) surface of the orthorhombic $NdGaO_3$ (NGO) substrate (for technical details see [13]). The Pt layer with the thickness of 10 nm (for both samples) was deposited over LSMO ex situ.

The sample S1 was used previously in our study of temperature dependence of the SP effect [13]. The half-width of the FMR line in this sample was found to be about 70-100 G in the temperature range from 300 to 100 K; these values exceed strongly the homogeneous (spin relaxation) broadening (about 10-12 G [17]), thus evidencing for a considerable inhomogeneous contribution. To suppress the inhomogeneity, the modified fabrication technology was implemented for the sample S2. Namely, the grown epitaxial LSMO film (before the Pt deposition) was additionally annealed at 820°C in the oxygen atmosphere. As a result, a dramatic narrowing of the FMR line was achieved, see the next Section.

It is known [18, 19] that the basic plane of the LSMO films grown on the (110) NGO surface is (001), and an additional in-plane uniaxial magnetic anisotropy with the easy axis directed along [010] LSMO is created due to orthorhombic distortions of the LSMO crystal structure.

This was confirmed in our samples by the the measurements of X-ray diffraction and the FMR spectra anisotropy [13].

To measure U^{SP} under conditions of spin pumping, the samples were prepared as strips with the length of 5 mm, the substrate thickness of 0.5 mm and the width $w = 1$ mm and 0.4 mm for the samples S1 and S2, respectively. The strips were cut along the easy axis (for S1) and along the hard axis (for S2) of the uniaxial in-plane anisotropy. The voltage U^{SP} was measured along the strip direction using the leads at the ends of the platinum layer. The technique is the same as described previously [13]. The sample was placed in the central plane of the rectangular TE_{102} microwave cavity. The strip was directed parallel to the microwave magnetic field $\mathbf{h}_{\mathrm{rf}}$. The external static magnetic field $\mathbf{H}$ can be rotated in the film plane and, as a rule, was fixed perpendicularly to the strip length (and so to $\mathbf{h}_{\mathrm{rf}}$ and U^{SP} directions). This geometry provides maximum effectiveness of SP and, at the same time, the absence of additional voltage caused by anisotropic magnetoresistance [9, 10]. The microwave power up to 130 mW was supplied by two Gunn diodes working at the frequency of $\omega/2\pi = 9$ GHz. The $U^{\mathrm{SP}}(H)$ signals were detected upon sweeping the field H across the resonance.

3. Results and discussion

A typical SP signal $U^{\mathrm{SP}}(H)$ registered at the S2 sample is shown in Fig. 1, together with the corresponding FMR spectrum. In Fig. 2, the SP magnitudes U_0^{SP} and half-widths δ measured in both S1 and S2 samples are plotted in the temperature range of 100-350 K. To facilitate comparison and reveal the dependence on δ (see Eq. 3), the magnitudes for the two samples are normalized (divided by the corresponding R values) and reduced to the same value of the strip width, namely, to $w = 1$ mm characteristic of the sample S1.

As seen from Fig. 2*b*, the half-width of the $U^{\mathrm{SP}}(H)$ signals at S1 is noticeably larger than that at S2, the latter being close to the homogeneous limit (10-12 G [17] near 250-300 K), with gradual increasing on either sides of the minimum. To verify the validity of the δ^{-2} dependence of the SP magnitude predicted by Eq. (3), we multiplied all the $U_0^{\mathrm{SP}}(T)$ data shown in Fig. 2*a* by the correspondent values of δ^2. The results are presented in Fig. 3*a*.

It is evident that the scaling done in this way does not provide any satisfactory match for the two samples. So, the inhomogeneous broadening should be taken into account. To do

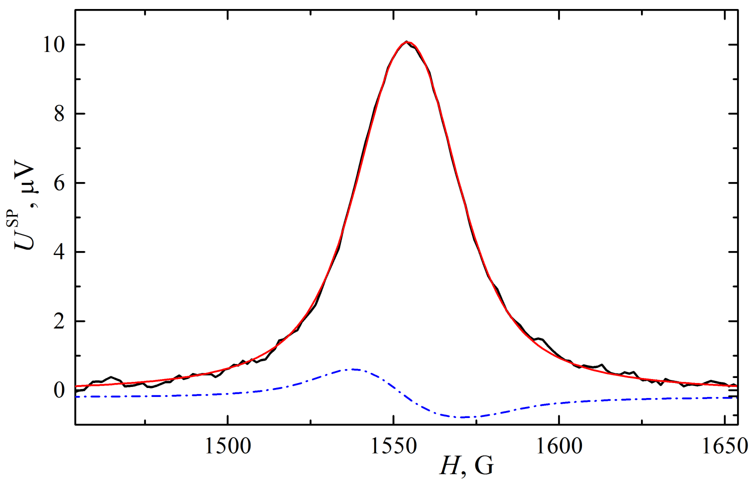

Figure 1. The spin pumping signal (black solid line) registered at the sample S2 at $T = 248$ K. The best fit with Eqs. (4), (5) with $\delta_{\mathrm{L}} = 12.5$ G and $\delta_{\mathrm{G}} = 10.2$ G is shown with the red curve. The bottom shows the corresponding FMR line.

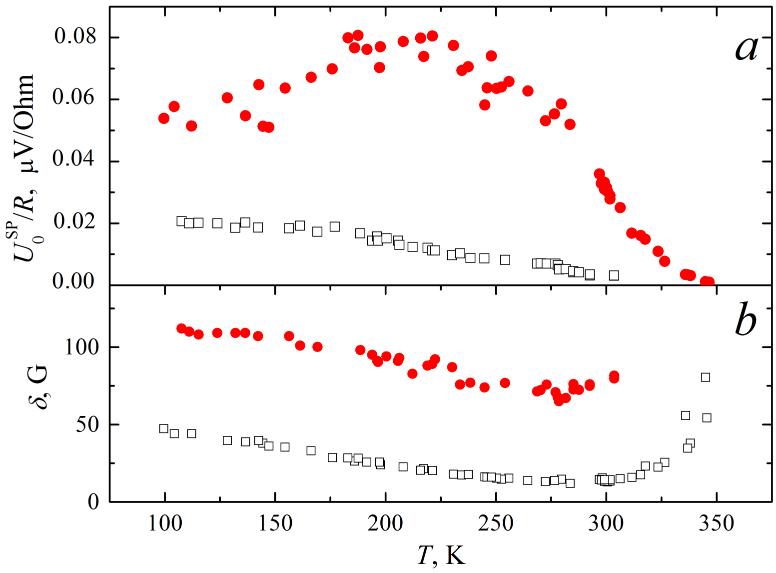

Figure 2. Normalized magnitudes (a) and half-widths (b) of the SP signals for the samples S1 (black empty squares) and S2 (filled red circles) measured in the temperature range of 100-350 K.

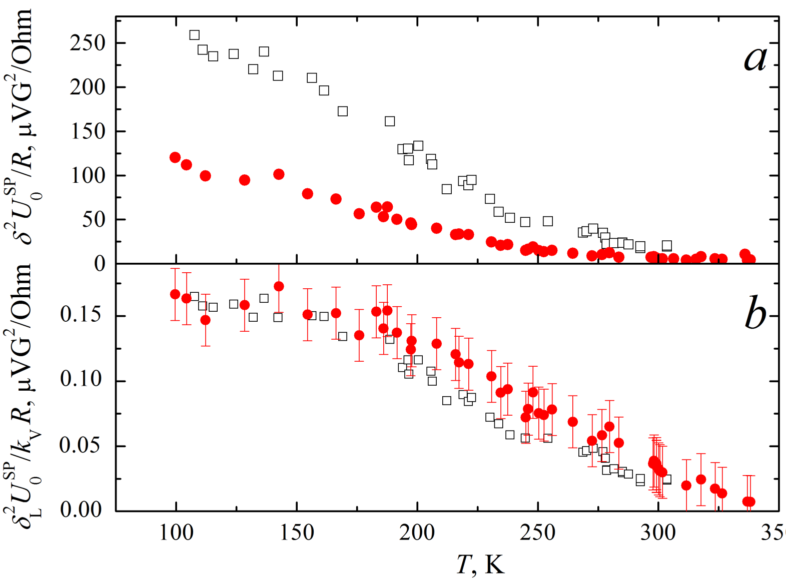

Figure 3. Comparison of the normalized SP magnitudes for the samples S1 (empty black squares) and S2 (filled red circles) scaled by δ^2 (a) and δ_L^2/k_V (b).

this, we represent the SP signal (as well as the FMR line) as a set of homogeneous Lorentzian "spin packets" with the half-width δ_L distributed around H_0 with amplitudes obeying Gaussian envelope with a half-width δ_G.

The resulted convolution is known as the Voigt shape and has the form [20]:

$$f_V(H) = \frac{b^2}{\sqrt{\pi}} \int_{-\infty}^{\infty} \frac{e^{-x^2}}{b^2 + (x-\nu)^2}\, dx \tag{4}$$

where

$$b = \frac{\delta_L \sqrt{\ln 2}}{\delta_G}, \qquad \nu = \frac{(H - H_0)\sqrt{\ln 2}}{\delta_G}. \tag{5}$$

An example of the SP signal approximation with the Voigt shape is shown in Fig. 1. From the best fitting, the values of δ_{L} and δ_{G} were determined for the sample S2 with the accuracy of 10%. It was found that $\delta_{\mathrm{L}} = (12.5 \pm 1.5)$ G in all temperature range, in accordance with the published data for homogeneous broadening in LSMO [17]. As to the sample S1, the observed large half-width seems to be mostly inhomogeneous and was identified as δ_{G}. Thus, Eq. (3) should be modified with account made for the Voigt shape. As a result, the SP magnitude U_0^{SP} is multiplied by an additional factor

$$k_{\mathrm{V}} = \frac{f_{\mathrm{V}}(H_0)}{f_{\mathrm{L}}(H_0)} = b^2 \int\limits_{-\infty}^{\infty} \frac{e^{-x^2}}{b^2 + x^2}\, dx \tag{6}$$

which determines a decrease in the SP magnitude due to inhomogeneous broadening. As a result, one has:

$$U_0^{\mathrm{SP}} = CR\, w\, \delta_{\mathrm{L}}^{-2}\, k_{\mathrm{V}}. \tag{7}$$

Note that in the homogeneous limit ($b \gg 1$) the Voigt factor k_{V} tends to unity, whereas at strong inhomogeneous broadening ($b \ll 1$), k_{V} tends to b. In the latter case, one gets $U_0^{\mathrm{SP}} \propto \delta^{-1}$, as it was suggested in our previous work [13]. Evidently, this case is applicable to the sample S1.

Now we are able to compare the results for the two samples using new renormalization according to Eqs. (6) and (7). To do this, we divide both the S1 and S2 magnitudes of Fig. 2*a* by the corresponding values of $\delta_{\mathrm{L}}^{-2} k_{\mathrm{V}}$. The resulted data are presented in Fig. 3*b*. It can be seen that the normalized values for the two samples coincide within the experimental and fitting accuracy.

In conclusion, our approach to the effect of inhomogeneous broadening in the spin pumping has been confirmed experimentally. This result indicates the limited applicability of the quadratic dependence of spin current on the line width and forces one to reconsider some estimates of such parameters as spin conductivity and spin diffusion length.

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