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Enhanced room-temperature spin Seebeck effect in a YIG/C_{60}/Pt layered heterostructure

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We report on large enhancement of the longitudinal spin Seebeck effect (LSSE) in the $Y_3Fe_5O_{12}$ (YIG)/Pt system at room temperature due to the addition of a thin layer of organic semiconductor (C_{60}) in between the YIG and the Pt. LSSE measurements show that the LSSE voltage increases significantly, from the initial value of 150 nV for the YIG/Pt structure to 240 nV for the YIG/C_{60}(5nm)/Pt structure. Radio-frequency transverse susceptibility experiments reveal a significant decrease in the surface perpendicular magnetic anisotropy (PMA) of the YIG film when C_{60} is deposited on it. These results suggest that the LSSE enhancement may be attributed to increased spin mixing conductance, the decreased PMA, and the large spin diffusion length of C_{60}. https://doi.org/10.1063/1.5007233

I. INTRODUCTION

The spin Seebeck effect (SSE) has emerged as one of the topical research areas in the last decade mainly due to its potentially revolutionary applications in spintronic devices.[1,2] It is one of the most effective ways to generate pure spin currents, along with spin pumping and the spin Hall effect.[3,4] The observations of SSE in a variety of materials ranging from ferromagnetic metals to magnetic insulators and semiconductors have surprised the scientific community.[1,5,6] Since its observation in a ferrimagnetic insulator $Y_3Fe_5O_{12}$ (YIG),[5] YIG has taken the center stage of SSE research due to its extremely low damping and absence of other spurious effects such as Planar Nernst effect (PNE) and Anomalous Nernst effect (ANE).

One of the limitations in realizing spintronic devices based on the SSE lies in its relatively low magnitude, in nV, of the resulting inverse spin Hall voltage signal. Different approaches have been proposed to improve the SSE voltage.[7–9] One method was to use multi-layers to increase the out-of-plane spin current, resulting in a significantly improved SSE signal.[7] The other method was to enhance the spin mixing conductance (G) at the interface between YIG and Pt.[8] Most recently, it has been shown that adding a very thin layer of antiferromagnetic NiO improves the SSE signal at room temperature as a result of the enhancement of G.[9] However, the shortcoming of using the NiO antiferromagnet lies in its intrinsically small spin diffusion length beyond which the spin transport decays rapidly.

Recently, we have shown that the magnetic anisotropy plays an important role in determining the longitudinal SSE (LSSE) in the YIG/Pt system.[10,11] In addition to the bulk (magnetocrystalline) anisotropy, we demonstrate that the perpendicular magnetic anisotropy (PMA) of the YIG surface

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affects spin current transport and hence the LSSE behavior. The effects of surface and bulk magnetic anisotropies are corroborated with those of thermally excited magnon number and magnon propagation length[12] to satisfactorily explain the temperature dependence of LSSE in the Pt/YIG system. Since the PMA of YIG may be altered when the YIG is interfaced with other materials, it is essential to understand how such an intermediate layer like NiO influences the PMA and the LSSE in Pt/YIG.

In this study, we aim to improve the room temperature LSSE in the Pt/YIG system by adding a thin layer (∼5 nm) of organic semiconductor (C_{60}) in between YIG and Pt. The deposition of this semiconductor between the insulator (YIG) and the metal (Pt) is expected to rectify the spin conductivity mismatch problem. Since C_{60} possesses a long spin diffusion length (∼100 nm),[13] which is substantially longer than that of Pt (∼2 nm), the presence of C_{60} can improve the spin mixing conductance of the YIG/Pt interface. Effects of the added C_{60} layer on the PMA of the YIG film are probed by radio-frequency (RF) transverse susceptibility measurements. We have observed a large enhancement of the LSSE at room temperature in the YIG/C_{60}/Pt heterostructure in comparison to the YIG/Pt system, and attribute this enhancement to the increased spin mixing conductance, the decreased PMA, and the large spin diffusion length of C_{60}.

II. EXPERIMENT

A 7 µm thick YIG film was grown on $Gd_3Ga_5O_{12}$ (GGG) substrate using liquid phase epitaxy (LPE).[14,15] The YIG film possesses a very smooth surface (surface roughness, 0.12±0.01 nm), as seen clearly in an AFM image of Fig. 1a. A 5 nm thick C_{60} was deposited on YIG using thermal evaporation in a high vacuum chamber, followed by dc sputtering of Pt of ∼5 nm thickness. Figure 1b illustrates the measurement geometry and the spin transport through the YIG/C_{60}/Pt layer. In the measurement set-up for the longitudinal SSE, the sample was sandwiched between two copper (Cu) plates. A resistive heater was attached to the bottom Cu plate and the temperature of the top Cu plate was controlled through molybdenum (Mo) screws attached to the cryogenic system. Desired temperature gradient between the top and bottom plates is achieved by varying the current in the heater. Silicon diode temperature sensors were used to monitor the temperature of the top and bottom plates. After stepping the system and heater temperature, the system was allowed to stabilize for 2 h before sweeping the magnetic field. Direction of the spin current is dictated by the direction of temperature gradient as shown in Figure 1b. All the measurements were performed in LSSE configuration as depicted. Measurements were performed in a cryogenic probe station which has the operating range up to 0.6 T magnetic fields and a temperature range of 77-340 K. SSE voltage was recorded as the magnetic field was swept between the positive and negative saturation of YIG, using a Keithley 2182 Nano voltmeter.

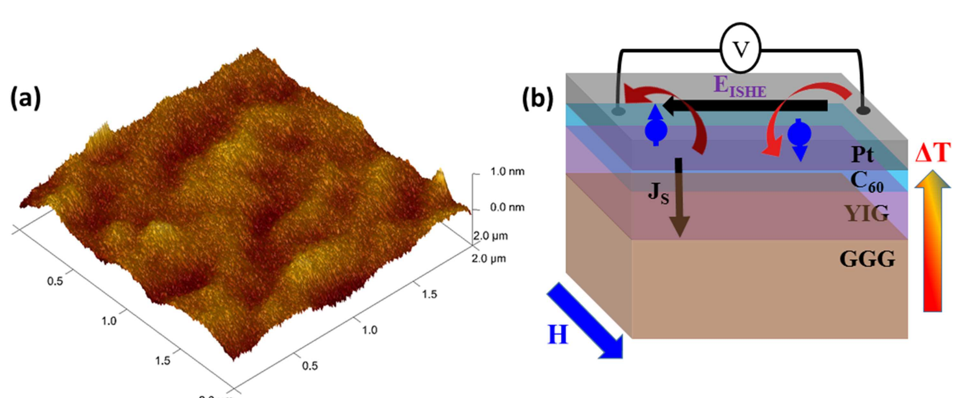

FIG. 1. (a) An AFM image of the YIG film grown on a GGG substrate. (b) A schematic illustration of the experimental configuration for the measurement of LSSE in the YIG/C_{60}/Pt system. ΔT, H, J_S, and E_{ISHE} denote the temperature gradient, magnetic field, spin current, and electric field induced by ISHE, respectively.

Transverse susceptibility measurements were performed using a self-resonant tunnel diode oscillator (TDO) with a resonant frequency of 12 MHz and sensitivity on the order of 10 Hz in 10 MHz.[16] The TDO is integrated with an insert that plugs into a commercial Physical Properties Measurement System (PPMS, Quantum Design), where the applied magnetic field can be varied up to 7T and the variable temperature range is 10 to 350 K.

III. RESULTS AND DISCUSSION

We first present results of the magnetic field dependence of LSSE voltage (V_{LSSE}). Figures 2a and 2b show the magnetic field dependence of V_{LSSE} at 300 K for the YIG/Pt and YIG/C_{60}/Pt structures, respectively. When the YIG film thickness is more than the magnetic domain size of YIG (~5 µm), a low field anomaly is observed in the SSE signal, which is attributed to the difference in bulk and surface magnetic behavior.[17] The absence of these low field plateau behavior in our YIG film could arise from the fact that the thickness of the film (~7 µm) is close to the magnetic domain size of YIG (~5 µm). At 300 K, the YIG/Pt system with no C_{60} layer showed a saturated V_{LSSE} of ~160 nV. When a 5 nm C_{60} film was added in between the YIG and Pt layers, the saturated V_{LSSE} increased significantly to ~240 nV. To check the reproducibility of the obtained results, we have repeated the LSSE experiments several times on the same YIG/Pt sample with adding and removing C_{60}. All the repeated measurements show reproducible values of the LSSE voltage with ± 5% error. The increase in the LSSE signal can thus be attributed to increase of the spin mixing conductance, due to improvement in interface quality with C_{60} addition.

To explain this more quantitatively, we have considered the ratio of spin currents between YIG/C_{60}/Pt and YIG/Pt via the following expression[9,18]

$$\frac{J_s^{Pt/C60/YIG}}{J_s^{Pt/YIG}} = \left(1 + \frac{\left(\left(\frac{G_{\frac{Pt}{C60}}}{G_{\frac{Pt}{YIG}}}\right) - 1\right) G_{Pt}}{G_{\frac{Pt}{C60}} + G_{Pt}}\right) \frac{1}{\cosh\left(\frac{t_{c60}}{\lambda_{c60}}\right) + G_{c60}\left(\frac{1}{G_{YIG}} + \frac{1}{G_{\frac{C60}{YIG}}}\right)\left(\sinh\left(\frac{t_{c60}}{\lambda_{c60}}\right)\right)}, \tag{1}$$

where J_s is the spin current, G is the spin current conductance, λ is the spin diffusion length of the corresponding material and t_{C60} is the thickness of the C_{60} layer. As the spin diffusion length of C_{60} ($\lambda_{C_{60}}$~100 nm)[13] is greater than that of Pt (~2 nm), more spin current is expected to reach the Pt layer. According to Eq. (1), the enhancement in the LSSE voltage for the YIG/C_{60}/Pt system can be related to the improvement of G at the interface and the high value of $\lambda_{C_{60}}$.

Now we turn to investigate how the addition of C_{60} affects the magnetic anisotropy of the YIG film and hence the LSSE. As described in the Experiment section, the radio frequency transverse susceptibility (TS) technique, which is based on a tunnel diode oscillator with a resonant frequency around 12 MHz, has been employed to determine magnetic anisotropy of the YIG film with and

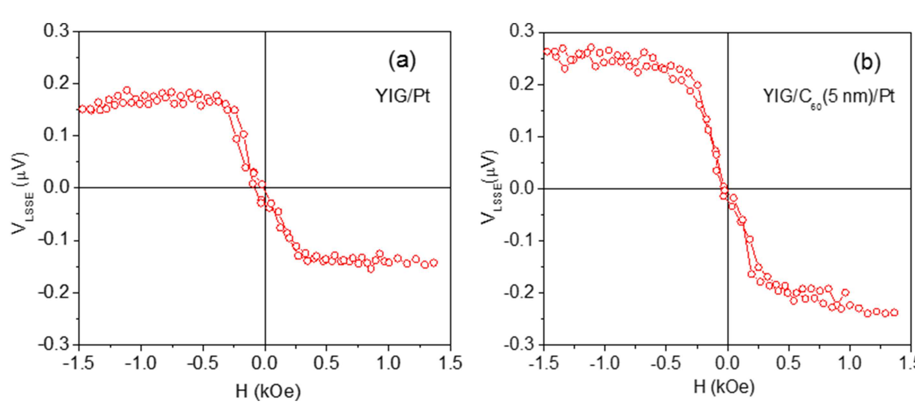

FIG. 2. The magnetic field dependence of LSSE voltage taken at 300 K for the temperature gradient of 1 K in the (a) YIG/Pt and (b) YIG/C_{60}/Pt systems.

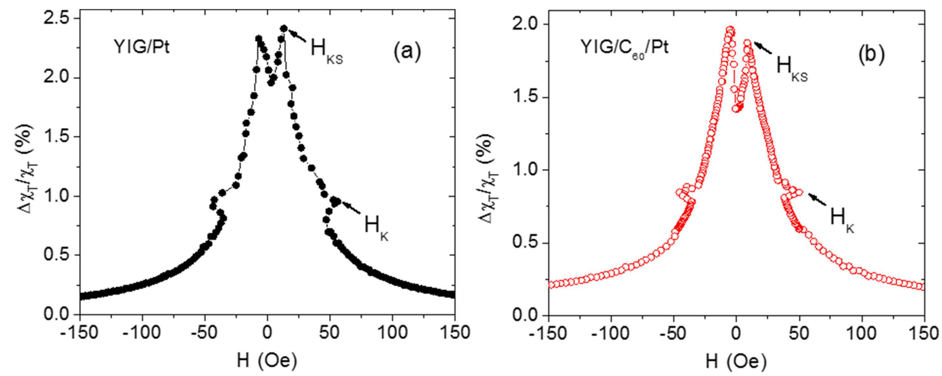

FIG. 3. Transverse susceptibility (TS) scans taken at 300 K for (a) YIG/Pt and (b) YIG/C_{60}/Pt in the out-of-plane direction of the film.

without C_{60}. Over the years, we have established the TS technique as a direct, accurate probe of effective magnetic anisotropy field (H_K) for a wide range of magnetic materials.[19–21] In this method, sample is placed inside an inductive coil which is part of the TDO circuit. An RF perturbation field is applied perpendicular to the DC field. As the DC magnetic field is varying, the resonant frequency of the TDO alters, and the change in resonant frequency is directly proportional to that in TS. As the DC field is swept from positive saturation to negative saturation, different peaks corresponding to anisotropic field (H_K) and switching field (H_S) appear in a TS plot. A detailed description of the TS measurements can be found in our earlier papers.[19–21]

Figures 3a and 3b show the TS spectra of the YIG/Pt and YIG/C_{60}/Pt samples at room temperature. As the DC magnetic field was swept from positive to negative saturation, two peaks were observed in the positive quadrant. As we reported in our recent work,[10] the first peak corresponds to the bulk anisotropy field (H_K) of YIG and the second peak represents the perpendicular magnetic anisotropy field (H_{KS}) of the YIG surface. The observation of such a distinct PMA peak validates the recent observation of a non-collinear magnetization at the YIG surface by Wu *et al.*[22] While the bulk magnetic anisotropy seems to have little influence on the saturated LSSE signal, the PMA at the interface could affect both the low-field LSSE behavior and spin current transport across the interface.[10] It is obvious from Fig. 3b that the addition of C_{60} has reduced the PMA field (H_{KS}) of YIG. The H_{KS} of the YIG sample is ~13 Oe, which decreased to ~ 8 Oe upon the deposition of C_{60}. The decrease of H_{KS} in YIG/C_{60}/Pt with respect to YIG/Pt can be due to the hybridization between the $d_z{}^2$ orbital of Fe and C atom, resulting in the net increase of spin moments at the YIG surface.[23] It has been suggested that the spin mixing conductance increases with increase in the surface magnetic moment density.[24] In case of the YIG/C_{60}/Pt system, the decrease in the PMA field and the increase in the spin mixing conductance are believed to give rise to the enhancement of the LSSE. Once the spin current crosses the interface, the long spin diffusion length of C_{60} facilitates it to reach the Pt layer without much degradation. Nevertheless, a detailed investigation into effects of C_{60} thickness on the LSEE voltage and the interface properties of YIG/C_{60}/Pt would be essential to fully understand the thermal spin transport mechanism in this system, which is beyond the scope of the present work.

IV. CONCLUSIONS

We have shown that the LSSE signal of the YIG/Pt structure can be improved by inserting a thin layer of C_{60} in between the YIG and the Pt. The significant improvement of the LSSE is attributed to a combination of increase of the spin mixing conductance at the interface, the decrease of the surface perpendicular magnetic anisotropy of the YIG film, and the inherently long spin diffusion length of C_{60}. Our study not only paves a new pathway for the design of novel spincaloric materials, but also highlights fundamental issues about spin interface effects in an insulator/semiconductor/metal based SSE system, stimulating further theoretical and experimental studies.

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