

RESEARCH ARTICLE | SEPTEMBER 17 2020

Strong interface-induced spin-charge conversion in YIG/Cr heterostructures ^{EP}

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Appl. Phys. Lett. 117, 112402 (2020)

<https://doi.org/10.1063/5.0017745>



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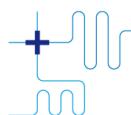
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Cite as: Appl. Phys. Lett. **117**, 112402 (2020); doi: [10.1063/5.0017745](https://doi.org/10.1063/5.0017745)

Submitted: 13 June 2020 · Accepted: 29 August 2020 ·

Published Online: 17 September 2020



View Online



Export Citation



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ABSTRACT

Here, we have investigated the spin pumping effect of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG)/Cu (t_{Cu} nm)/Cr heterostructures at room temperature with the thickness of the Cu interlayer varying from 0.4 nm to 5.0 nm. A huge charge signal $I_c = 0.239 \mu\text{A}$ is observed in a YIG/Cr bilayer with direct contact, whereas I_c drops dramatically by two orders of magnitude when thin Cu interlayers down to 0.4 nm are inserted between YIG and Cr. Meanwhile, the injected spin current J_s stays almost invariant for all the heterostructures. The effective spin Hall angle " θ_{SH} " of the YIG/Cr interface is found to be three orders of magnitude larger than the spin Hall angle θ_{SH} of the bulk Cr layer in YIG/Cu/Cr. The huge spin-charge conversion efficiency at the YIG/Cr interface is attributed to the inverse Rashba–Edelstein effect. Our experimental results demonstrate the dominant role of the interfacial effect in the spin-charge conversion process of the YIG/Cr heterostructures.

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In the last few decades, spintronics has developed rapidly as a new field of electronic science and technology.^{1–7} A great deal of attention is drawn on the generation, detection, and exploitation of pure spin current J_s (or more accurately described as pure spin current density). The conversion between the spin current J_s and the charge current I_c ($=J_c \times S$, where J_c is the charge current density and S is the cross-sectional area of the sample) is exploited to achieve electrical modulation of the magnetic state, which may lead to energy-efficient magnetic storage devices and computing units. The pure J_s can be generated in the systems with large spin–orbit coupling (SOC) by various mechanisms, such as the spin Hall effect (SHE),^{8–10} spin pumping,^{11–13} the spin Seebeck effect (SSE),^{14–16} and the Rashba–Edelstein effect (REE).¹⁷ Correspondingly, the reverse processes can be utilized

to electrically detect the pure J_s , such as the inverse spin Hall effect (ISHE)^{18,19} and inverse Rashba–Edelstein effect (IREE).^{20,21} The spin-charge conversion efficiency is described as spin Hall angle θ_{SH} in the SHE and the inverse Edelstein length λ_{IEE} in REE, which are intimately relevant to the strength of SOC.

It is well known that the nonmagnetic heavy metals (such as Pt and W) are remarkable providers and detectors of J_s because of their large intrinsic SOC, resulting in large θ_{SH} and conspicuous SHE and ISHE. Usually, the conversion of J_s and I_c in metals is a bulk effect occurring within a certain volume determined by the spin-diffusion length λ_{SD} . In previous works, θ_{SH} is generally observed to be in good agreement with the Z^4 trend for metals (Z is the atomic number),^{22,23} indicating a large θ_{SH} in heavy metals such as Pt, W, Ta, and Bi and a

very small θ_{SH} in light metals. However, it has been reported that 3d light metals such as Cr directly grown on $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) also have a large θ_{SH} , as observed by ferromagnetic resonance (FMR) spin pumping²⁴ and spin Seebeck measurements,²⁵ and the values of θ_{SH} are even comparable with those of the heavy metals. Therefore, intriguing questions are raised about what causes the large measured spin Hall angle θ_{SH} in 3d metals and whether there exist some other SOC origins, for example, the interface REE.

The REE exists in spin momentum locking systems such as Rashba interfaces or surfaces. It originates from the interaction between the electron's spin and its momentum, which can be used to effectively produce a steady nonequilibrium spin polarization from a charge current. IREE was reported in a two-dimensional electron gas, causing transformation from spin current into charge current.^{26,27} Recently, IREE has been measured using the spin pumping method in different systems with Rashba interfaces, for example, metallic interfaces Bi/Ag,^{28,29} metal/oxide interfaces,³⁰ and oxide/oxide interfaces.³¹ The Hamiltonian of Rashba SOC can be expressed as^{28,32}

$$H_R = \alpha_R(\mathbf{p} \times \hat{z}) \cdot \boldsymbol{\sigma}, \quad (1)$$

where $\boldsymbol{\sigma}$ is the vector of Pauli spin matrices, $\hat{z}$ is the coordinate normal to the interface, $\mathbf{p}$ is the momentum, and α_R is the Rashba coefficient, proportional to the induced internal electric field. The conduction electron spins are aligned to the Rashba field along the $\mathbf{p} \times \hat{z}$ direction, forming a helical spin texture, as shown in Fig. 1(a).

In order to preferably uncover the dominant factor of the large θ_{SH} in the Cr/ $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) system, in this study, we modulated the interface between YIG and Cr by inserting a Cu layer and used the FMR spin pumping method to investigate the corresponding change of I_c signals. Cu has a long spin diffusion length due to its weak SOC; thus, it allows spin transport from YIG to Cr with minimal dissipation. A YIG/Cr bilayer and a series of YIG/Cu (t_{Cu} nm)/Cr (10 nm) trilayers with different Cu thickness varying from 0.4 nm to 5.0 nm were fabricated. A huge spin pumping signal I_c was observed as expected in the

YIG/Cr bilayer. However, I_c dropped sharply by two orders of magnitude when an ultrathin 0.4 nm-Cu interlayer was inserted between YIG and Cr, while the effective spin mixing conductivity $g_{\text{eff}}^{\uparrow\downarrow}$ and the J_s showed no significant change. The results demonstrated that the YIG/Cr interface plays a dominant role in gaining high spin-charge conversion efficiency due to the interface REE.

The YIG films of 1.2 μm were grown on $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (GGG) substrates by liquid-phase epitaxy. The YIG films exhibited high crystalline quality and surface smoothness with a root mean square (RMS) roughness of about 0.4 nm. The GGG/YIG substrates were degreased via ultrasonic bathing in acetone and ethyl alcohol for 12 min and cleaned by de-ionized water before being placed into the sputter growth chamber. Then, the YIG (1.2 μm)/Cu (t_{Cu} nm)/Cr (10 nm) ($t_{\text{Cu}} = 0.0, 0.4, 0.7, 1.0, 2.0, 5.0$ nm) samples were fabricated by ultra-high vacuum sputtering with a base pressure of 3×10^{-8} Torr at room temperature. The working argon pressure was 4.5×10^{-3} Torr for Cu deposition and 5.5×10^{-3} Torr for Cr deposition, respectively. Finally, the samples were patterned into small rectangular bars of 4.0 mm length and 0.5 mm width by combining optical lithography and ion Ar+ plasma etching. The FMR spin pumping measurements of the YIG/Cr and YIG/Cu/Cr samples were performed by using coplanar waveguides (CPW) method at room temperature, and the microwave frequency (f) and power (P_{rf}) were set to be 9 GHz and 20 dBm. As illustrated in Fig. 1(b), the external bias magnetic field H was applied along the x-axis ($\theta_H = 90^\circ$) and voltage measurements were performed along the y-axis. At resonance, the precessing YIG magnetization transferred angular momentum to the conduction electron in the metal layer, resulting in a vertical direct current (DC) spin current J_s injected into the YIG/Cr interface and the metal layers (Cr or Cu/Cr) along the z-axis with the spin polarization along the x-axis. Then, J_s can be converted to a charge current I_c via ISHE or (and) IREE, which is illustrated in the schematic diagrams shown in Figs. 1(c) and 1(d).

Figures 2(a)–2(f) show the representative results for the derivative of the ferromagnetic resonance absorption of the YIG/Cu (t_{Cu} nm)/Cr samples with t_{Cu} of 0.0 nm, 0.4 nm, and 5.0 nm at $f = 9$ GHz under the FMR of YIG and the corresponding I_c vs $H - H_{\text{res}}$ (H_{res} is the FMR resonance field) spectra, respectively. Interestingly, I_c at the FMR point drops sharply by two orders of magnitude when an ultrathin Cu interlayer of 0.4 nm was inserted between the YIG and Cr, while the difference of linewidth between FMR spectra is trivial. To further understand the effect of the inserted Cu spacer on I_c , the same measurements were performed on three additional YIG/Cu (t_{Cu} nm)/Cr ($t_{\text{Cu}} = 0.7, 1.0$, and 2.0 nm) trilayers, and the maximum values of I_c at the FMR for all samples are displayed in Fig. 3(a) and Table S1 (see the supplementary material). As shown in Fig. 3(a), the values of I_c generated at the FMR point are 0.2390, 0.0013, and 0.0049 μA for the samples with $t_{\text{Cu}} = 0.0, 0.4$, and 5.0 nm, respectively. There is a huge I_c in YIG/Cr (10 nm) bilayer with direct contact, which is comparable to YIG/heavy metals. Compared to YIG/Cr, I_c is reduced significantly by a factor of 185 for YIG/Cu (0.4 nm)/Cr sample, and this factor gradually decreases to ~ 50 even when t_{Cu} reaches to 5.0 nm. Hence, the observed dramatic change in I_c suggests that the YIG/Cr interface may play a crucial role in obtaining high efficiency of spin-to-charge conversion.

In order to explain the observed phenomenon and reveal the roles of the YIG/Cr interface and the Cu interlayer in spin-to-charge conversion, it is necessary to analyze the J_s value. The absolute

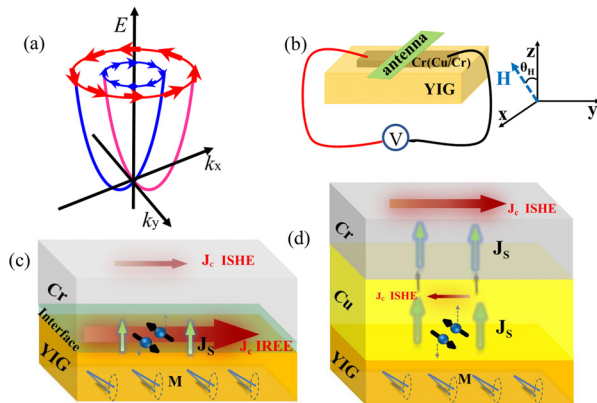


FIG. 1. (a) Typical spin-split dispersion and Fermi contours at the Rashba interface. (b) Schematic illustration of the YIG/Cr (YIG/Cu/Cr) samples for FMR spin pumping measurement. The radio frequency (rf) field is along the y-axis, and the DC field is along the x-axis ($\theta_H = 90^\circ$). (c) Spin to charge conversion in the YIG/Cr heterostructure: J_s (back flow included) converted into a horizontal DC charge current I_c due to IREE at the YIG/Cr interface and ISHE in the Cr layer. (d) In YIG/Cu/Cr trilayer, J_s converted into I_c by ISHE in Cu and Cr layers, respectively.

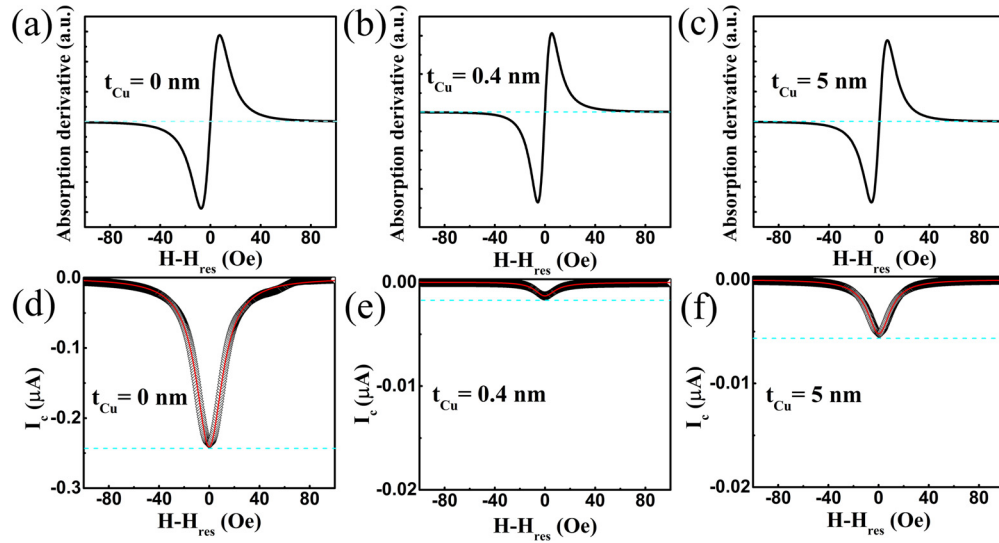


FIG. 2. (a)–(c) are the FMR signals of the YIG/Cu(t_{Cu})/Cr samples with t_{Cu} of 0.0 nm, 0.4 nm, and 5.0 nm at $f = 9$ GHz, respectively, and correspondingly, (d)–(f) are the I_c vs $H-H_{\text{res}}$ spectra derived from FMR spin pumping voltage measurements for the samples with t_{Cu} of 0.0 nm, 0.4 nm, and 5.0 nm, respectively. The red lines are Lorentzian fits.

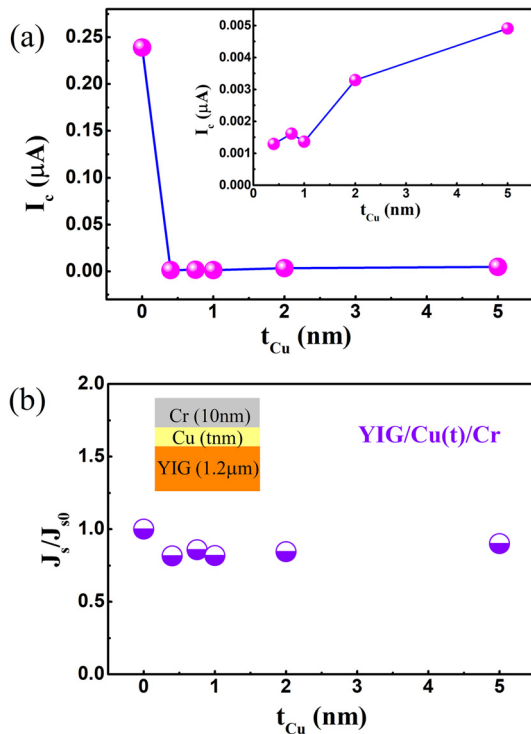


FIG. 3. (a) Cu thickness dependence of I_c at the FMR point for the YIG/Cu (t_{Cu} nm)/Cr samples with t_{Cu} from 0.0 to 5.0 nm, respectively. The inset in (a) shows the detail of I_c at the FMR point for the samples with t_{Cu} from 0.4 to 5.0 nm. (b) Spin current J_s in YIG/Cu/Cr trilayers normalized to J_{s0} in YIG/Cr bilayer, as a function of the Cu thickness.

magnitude of J_s cannot be determined accurately from the spin pumping experiment because of the lack of calibration of the actual value of h_{rf} in our measurement system. However, the normalized spin current J_s/J_{s0} can be calculated (see the [supplementary material](#)), where J_{s0} is the spin current detected in the YIG/Cr bilayer without the Cu interlayer. [Figure 3\(b\)](#) shows that J_s/J_{s0} stays almost invariant for all heterostructures with different t_{Cu} . The invariant J_s/J_{s0} confirms that the Cu interlayer has a negligible influence on the J_s transporting into the Cr layer from YIG. However, with similar J_s , the I_c in YIG/Cr bilayer is significantly larger than that in the YIG/Cu/Cr trilayer; even the thickness of Cu interlayer is only 0.4 nm. Therefore, the small values of I_c observed in YIG/Cu/Cr trilayers reveal the low spin-to-charge conversion efficiency in the bulk Cr layer; this is not surprising since the 3d metals with low Z have small θ_{SH} . The interface of YIG/Cr should be responsive for the observed huge I_c .

Besides, there is a subordinate phenomenon that the maximum value of I_c increases slightly from $0.0013 \mu\text{A}$ to $0.0049 \mu\text{A}$ with increasing thickness of Cu as shown in the inset of [Fig. 3\(a\)](#). This weak increment may be related to factors, such as optimized interfaces and improved quality of Cu layer for better transmitting J_s with increasing thickness of a Cu spacer. Furthermore, I_c increasing with Cu thickness also confirms that the Cu inserted layer indeed acts as a well spin transmitter with minimal spin dissipation.

Two possible underlying mechanisms are considered to account for the huge I_c at the YIG/Cr interface. One is the magnetic proximity effect (MPE) at the YIG/Cr interface, which could induce ferromagnetic ordering in the Cr atomic layers adjacent to the ferrimagnetic YIG layer. And there probably exists an additional J_s pumped from interfacial ferromagnetic Cr into the top paramagnetic Cr at room temperature, which leads to an increase in I_c . However, [Qu et al.](#)²⁵ reported that MPE is not observed in the YIG/Cr system using Hall measurements, ruling out the possibility of MPE. The other mechanism is the IREE at YIG/Cr interface. Several previous works found

that the efficiency of spin-charge conversion occurring at the Rashba interface could be substantially larger than the bulk SHE in Bi, Pt, and β -W.^{29,31} Except for ISHE of the bulk Cr layer, IREE of YIG/Cr interface should also be considered to analyze the huge I_c in the YIG/Cr heterostructure. A quantitative comparison of spin-charge conversion efficiency between IREE at YIG/Cr interface and ISHE in Cr metal is present in the following discussion.

We notice that the signs of I_c detected in both the YIG/Cr bilayer and YIG/Cu/Cr trilayer are negative, which is the same as W but opposite to Pt.^{13,25} It implies that the sign of θ_{SH} in the metal Cr is negative, owing to the less than half-filled 3d shell in Cr. Moreover, it also shows that IREE at the YIG/Cr interface is in the same direction as the ISHE in Cr film. Besides, considering the positive θ_{SH} of Cu and the observed negative sign of I_c in YIG/Cu/Cr samples, one can find out that Cr itself has a bigger θ_{SH} than that of the metal Cu.

To better illustrate the mechanism of procedure behind the different spin pumping behavior, we construct two simple models including the generations of I_c based on IREE and ISHE discussed above, as shown in Fig. 4. It needs to be emphasized that J_s here, as shown in Fig. 3(b), is an effective J_s including the reflected J_s . Figure 4(a) shows that for the YIG/Cr bilayer, the large fraction of the J_s from the YIG layer, is dissipated first at the YIG/Cr interface and converted into a larger I_c due to the giant IREE-induced spin-to-charge conversion efficiency. Then, the remaining J_s transports into the top Cr film, where a small part I_c is generated due to a weak bulk ISHE in the Cr film. Since the ISHE in Cr has the same sign as the IREE at the YIG/Cr interface as mentioned above, the total I_c of YIG/Cr sample is their summation and exhibits a larger observed value. As plotted in Fig. 4(b), for the YIG/Cu/Cr trilayer, however, the IREE at YIG/Cr interface is removed by inserting the spacer Cu layer, the J_s generated from the YIG layer directly passes through the spacer Cu layer with a negligible loss into the top Cr layer due to the long spin-diffusion length of Cu and is absorbed and converted into I_c due to weak ISHE in the top Cr layer with a low spin-to-charge conversion efficiency. Besides, we should note that the Cu layer generates a negligible charge current I_c and the sign of I_c in Cu is opposite to that in Cr.^{22,24} Thus, the top Cr layer dominates the spin-to-charge current conversion process and the total I_c shows a negative sign.

Finally, we further discuss quantitatively a comparison of spin-charge conversion efficiency between the YIG/Cr interface and Cr film. The I_c induced by IREE is generally expressed as³⁰

$$I_c = \lambda_{IREE} w J_s \frac{2e}{\hbar}, \quad (2)$$

where w and λ_{IREE} are the rectangular bar width of the films and the inverse Rashba–Edelstein length. The I_c in the ISHE layer can be determined by²⁸

$$I_c = w \theta_{SH} \lambda_{SD} \tanh\left(\frac{t}{2\lambda_{SD}}\right) J_s \frac{2e}{\hbar}, \quad (3)$$

where θ_{SH} , λ_{SD} , and t are the spin Hall angle, the spin diffusion length, and the thickness of the nonmagnetic layer, respectively. When $t \ll \lambda_{SD}$, Eq. (3) can also be given by

$$I_c = w \theta_{SH} t J_s \frac{e}{\hbar}. \quad (4)$$

The spin-charge conversion coefficient λ_{IREE} and θ_{SH} cannot be directly compared owing to the different origins. Here, we simply assume that the YIG/Cr interface is 0.4 nm thick.²⁸ To directly compare Eqs. (2) and (4), one can get $\lambda_{IREE} = \frac{1}{2} \theta_{SH} t$; thus, for the YIG/Cr interface, λ_{IREE} of IREE can be quantitatively estimated by using an effective spin angle θ_{SH} as used in ISHE with $t = 0.4$ nm. According to the proposed model in Fig. 4, the small spin absorptions in the Cr layer, compared to the strong IREE at the YIG/Cr interface, for YIG/Cr and the Cu layer for YIG/Cu/Cr can be neglected. Based on Eq. (4) (where $e/\hbar$ and w are both constants) and the obtained experimental data: $I_c = 0.2390 \mu A$, $t = 0.4$ nm and $J_s/J_{s0} = 1$ in YIG/Cr; $I_c = 0.0013 \mu A$, $t = 10$ nm and $J_s/J_{s0} = 0.9$ in YIG/Cu (0.4 nm)/Cr, the calculated ratio between the effective SHE angle “ θ_{SH} ” of the YIG/Cr interface and the θ_{SH} of the bulk Cr layer in the YIG/Cu (0.4 nm)/Cr sample reaches $\sim 4.14 \times 10^3$, confirming that the spin-charge conversion efficiency of the YIG/Cr interface is thousands of times higher than that of the bulk Cr layer.

In summary, we have experimentally carried out a comparative study of the spin pumping process in two types of samples: a YIG/Cr bilayer and YIG/Cu/Cr trilayers with different Cu thicknesses. By inserting a thin Cu layer into the YIG/Cr interface, we observe that charge current I_c falls dramatically. The I_c of YIG/Cr bilayer is 185 times higher than that of the YIG/Cu(0.4 nm)/Cr trilayer, which can be explained by combining a high spin-charge conversion efficiency due to strong IREE at the YIG/Cr interface and a relatively small spin-charge conversion efficiency due to weak ISHE in the 3d light metal Cr layer. Our results suggest that tailoring the interface of magnetic heterostructures provides an alternative strategy to achieve the higher spin-charge conversion efficiency for the development of low-power consumption spintronic devices.

See the [supplementary material](#) for the normalized spin current J_s/J_{s0} , which is calculated for all heterostructures with different t_{Cu} , where J_{s0} is the spin current detected in the YIG/Cr bilayer without the Cu interlayer. Spin pumping parameters of YIG (1.2 μm)/Cu (t_{Cu} nm)/Cr (10 nm) ($t_{Cu} = 0.0, 0.4, 0.7, 1.0, 2.0, 5.0$ nm) samples are listed: Gilbert damping constant, effective spin mixing conductance, injected spin current J_s in YIG/Cu/Cr trilayers normalized to J_{s0} in YIG/Cr bilayer, and the electric current magnitude I_c .

AUTHORS' CONTRIBUTIONS

L.N. and Z.C. contributed equally to this work.

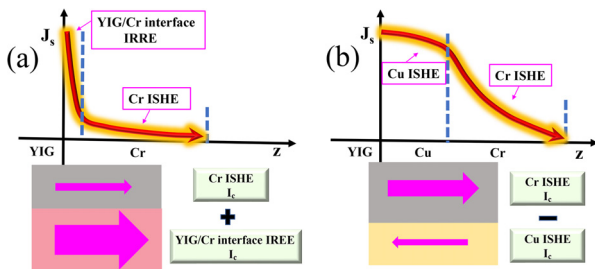


FIG. 4. Schematic illustrations of the J_s (back flow included) converted into I_c for YIG/Cr and YIG/Cu/Cr samples. The I_c (bottom) corresponds to the spin absorption (top) shown in (a) and (b). ISHE in Cr and Cu layers and IREE at the YIG/Cr interface are both taken into consideration. (a) YIG/Cr system and (b) YIG/Cu/Cr system.

This work was supported by the National Key Research and Development Program of China (Grant No. 2016YFA0300803), the National Natural Science Foundation of China (Grant No. 11774160), the Natural Science Foundation of Jiangsu Province of China (Grant No. BK20192006), the National Key Research and Development Program of China (Grant Nos. 2016YFA0300801 and 2018YFA0209002), the National Natural Science Foundation of China (Grant Nos. 61427812, 11774150, and 51702042), and the Fundamental Research Funds for the Central Universities (Grant No. 021014380113).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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