



CHORUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Surface-State-Dominated Spin-Charge Current Conversion in Topological-Insulator-Ferromagnetic-Insulator Heterostructures

Hailong Wang, James Kally, Joon Sue Lee, Tao Liu, Houchen Chang, Danielle Reifsnyder Hickey, K. Andre Mkhoyan, Mingzhong Wu, Anthony Richardella, and Nitin Samarth

Phys. Rev. Lett. **117**, 076601 — Published 11 August 2016

DOI: [10.1103/PhysRevLett.117.076601](https://doi.org/10.1103/PhysRevLett.117.076601)

Surface state dominated spin-charge current conversion in topological insulator/ferromagnetic insulator heterostructures

Hailong Wang¹, James Kally¹, Joon Sue Lee,¹ Tao Liu,²
Houchen Chang,² Danielle Reifsnnyder Hickey,³ Andre Mkhoyan,³
Mingzhong Wu,² Anthony Richardella,¹ Nitin Samarth^{1*}

¹*Department of Physics, The Pennsylvania State University,
University Park, Pennsylvania 16802, USA*

²*Department of Physics, Colorado State University, Fort Collins, CO 80523, USA and*

³*Department of Chemical Engineering and Materials Science,
University of Minnesota, Minneapolis, Minnesota 55455, USA*

(Dated: July 13, 2016)

Abstract

We report the observation of ferromagnetic resonance-driven spin pumping signals at room temperature in three-dimensional topological insulator thin films – Bi_2Se_3 and $(\text{Bi,Sb})_2\text{Te}_3$ – deposited by molecular beam epitaxy on $\text{Y}_3\text{Fe}_5\text{O}_{12}$ thin films. By systematically varying the Bi_2Se_3 film thickness, we show that the spin-charge conversion efficiency, characterized by the inverse Rashba-Edelstein effect length (λ_{IREE}), increases dramatically as the film thickness is increased from 2 quintuple layers, saturating above 6 quintuple layers. This suggests a dominant role of surface states in spin and charge interconversion in topological insulator/ferromagnet heterostructures. Our conclusion is further corroborated by studying a series of $\text{Y}_3\text{Fe}_5\text{O}_{12}/(\text{Bi,Sb})_2\text{Te}_3$ heterostructures. Finally, we use the ferromagnetic resonance linewidth broadening and the inverse Rashba-Edelstein signals to determine the effective interfacial spin mixing conductance and λ_{IREE} .

The development of next-generation spintronic devices has driven extensive studies of spin-to-charge conversion through measurements of the inverse spin Hall effect (ISHE) and/or the inverse Rashba-Edelstein effect (IREE) in both three-dimensional (3D) [1–6] and two-dimensional (2D) material systems [7–15]. Topological insulators (TIs) such as the Bi-chalcogenides are naturally relevant in this context due to the large spin-orbit coupling (SOC) strength and the inherent spin-momentum “locking” in their surface states [9, 16, 17] which promise very efficient spin-charge conversion efficiency. Previous studies of spin transfer in TI-based heterostructures have involved ferromagnetic metals that provide a shunting current path, therefore introducing potential artifacts which complicate the picture and analysis [8, 9, 11, 12]. To circumvent these problems, we have grown and characterized bilayers of TIs on ferrimagnetic insulator $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) thin films with an exceptionally low damping constant [19]. Here, we report the ferromagnetic resonance (FMR)-driven spin pumping observed in YIG/ Bi_2Se_3 bilayers, showing robust spin pumping signals at room temperature. Systematic variation of the Bi_2Se_3 thickness allows us to unambiguously demonstrate that the spin-charge conversion efficiency, characterized by the inverse Rashba-Edelstein effect (IREE) length λ_{IREE} in a 2D material system[7], dramatically increases from (1.1 ± 0.13) pm to 35 ± 4 pm as the Bi_2Se_3 thickness varies from 2 to 6 quintuple layers (QL). When the top and bottom surface states with opposite spin polarizations decouple from each other, λ_{IREE} saturates and is constant, providing clear evidence for the dominant role of surface states in inducing spin-charge conversion in 3D TIs.

We first discuss the structural and interfacial characterization of the YIG/ Bi_2Se_3 heterostructure using high-resolution scanning transmission electron microscopy (HR-STEM). Figure 1(a) shows an atomically ordered 6 QL Bi_2Se_3 layer grown on an epitaxial 30-nm YIG thin film. We note that an amorphous layer of about 1 nm in thickness is observed at the YIG/ Bi_2Se_3 interface, most likely due to the nucleation of the template layer in the two-step growth process (see Supplementary Material at [link to be added] for more details about the growth method). The atomic force microscopy (AFM) image in Fig. 1(b) shows a smooth surface with a roughness of about 0.71 nm. A representative $\theta - 2\theta$ x-ray diffraction (XRD) scan of a 40 QL Bi_2Se_3 film shown in Fig. 1(c) indicates a phase-pure Bi_2Se_3 layer. Figure 1(d) shows a representative FMR derivative absorption spectrum for a 30-nm YIG film used in this study taken at a radio-frequency (rf) $f = 3$ GHz with a magnetic field H applied in the film plane. The peak-to-peak line width (ΔH_{PP}) obtained from the spectrum is 9.2 Oe,

and an effective saturation induction of 1.76 kOe is extracted from fitting the frequency dependence of the resonance field [19]. The spin pumping measurements are performed using a microwave transmission line on the YIG/TI bilayers at room temperature (approximate sample dimensions of 1 mm \times 5 mm). During the measurements, a DC magnetic field H is applied in the x - z -plane and the spin pumping voltage V_{SP} is measured across the ~ 5 mm long TI layer along the y -axis, as illustrated in Fig. 2(a). At the resonance condition, the YIG magnetization M precesses around the equilibrium position and transfers angular momentum to the conduction electrons in the TI films through interfacial exchange coupling [4]. The resulting pure spin current is injected along the z -axis with spin polarization σ parallel to M , and then converted to a charge current leading to the spin pumping signals.

Figure 2(b) shows the temperature dependence of the resistivity of 6 QL Bi_2Se_3 and $(\text{Bi,Sb})_2\text{Te}_3$ thin films grown on YIG. The metallic behavior (decrease in resistivity at low temperature) is the typical behavior of Bi_2Se_3 due to Se vacancies [20]. For $(\text{Bi,Sb})_2\text{Te}_3$, the resistivity increases by $\sim 50\%$ from room temperature to 2 K, consistent with surface state dominated transport in this thin film [21]. The carrier concentrations obtained from Hall effect measurements at room temperature are $4 \times 10^{13} \text{ cm}^{-2}$ and $9.8 \times 10^{12} \text{ cm}^{-2}$ for 6 QL Bi_2Se_3 and $(\text{Bi,Sb})_2\text{Te}_3$, respectively.

Figure 2(c) shows the observed V_{SP} vs. H spectra of the YIG/ Bi_2Se_3 (6QL) bilayers at $f = 2, 3,$ and 4 GHz using 100 mW microwave power. The observed spin pumping signals change sign when the magnetic field H is reversed from $\theta_H = 90^\circ$ to 270° , as expected from either IREE or ISHE. At 2 and 3 GHz, the observed signal is about $40 \mu\text{V}$, and for 4 GHz, the signal decreases to about $20 \mu\text{V}$, which results from the variation of the microwave transmission line performance at different frequencies. Figure 2(d) shows the spin pumping spectra of a YIG/ Bi_2Se_3 (6 QL) sample at microwave powers of 18, 32, 56, and 100 mW and an excitation frequency of 3 GHz. The upper inset shows the rf-power dependence of V_{SP} at $\theta_H = 90^\circ$, indicating that the observed spin pumping signals are in the linear regime.

To probe the spin to charge conversion mechanism in TI layers, we systematically vary the Bi_2Se_3 thickness from 2 to 60 QL. Figure 3(a) shows the spin pumping spectra when $\theta_H = 90^\circ$ for 4, 6, 24, and 40 QL thicknesses of Bi_2Se_3 grown on YIG, respectively. The significant enhancement of the spin pumping signal in the low Bi_2Se_3 thickness regime mainly results from the increased resistivity. For a 2D material system, such as the TI surface states, the spin to charge conversion is dominated by IREE [22, 23] and the injected spin current is

converted into a 2D charge current, $J_c = \lambda_{\text{IREE}} J_s$. The spin current density J_s is in units of A m^{-2} , and the 2D charge current density J_c is in units of A m^{-1} ; the parameter λ_{IREE} has the dimension of length and is introduced to characterize the spin to charge conversion efficiency in 2D material systems [7, 23]. The observed spin pumping voltages V_{SP} dominated by IREE depend on several material parameters [7]:

$$V_{\text{SP}} = -wR\lambda_{\text{IREE}}J_s, \quad (1)$$

where w and R are the sample width and resistance, respectively. J_s is the spin current density at the YIG/TI interface which can be expressed as [3, 5, 6]:

$$J_s = \frac{2e}{\hbar} \frac{g_{\uparrow\downarrow} h_{\text{rf}}^2 \hbar \omega^2 \left[\gamma 4\pi M_s + \sqrt{(\gamma 4\pi M_s)^2 + 4\omega^2} \right]}{2\pi (\Delta H_{\text{pp}})^2 [(\gamma 4\pi M_s)^2 + 4\omega^2]}, \quad (2)$$

where $g_{\uparrow\downarrow}$ is the effective interfacial spin mixing conductance [24], ΔH_{pp} is the FMR peak-to-peak linewidth, h_{rf} is the radio frequency field, ω is the FMR angular frequency, and M_s is the saturation induction of the YIG thin films. We can determine the effective spin mixing conductance $g_{\uparrow\downarrow}$ from the FMR linewidth broadening of the YIG thin film [2, 3, 24]:

$$g_{\uparrow\downarrow} = \frac{2\pi\sqrt{3}M_s\gamma t_{\text{YIG}}}{g\mu_B\omega} (\Delta H_{\text{YIG/TI}} - \Delta H_{\text{YIG}}), \quad (3)$$

where γ is the absolute gyromagnetic ratio, t_{YIG} denotes the thickness of the YIG thin films, g is the Landé factor, and μ_B is the Bohr magnetron.

If the spin pumping signal is dominated by the ISHE, spin diffusion should be taken into account according to $J_c = \theta_{\text{SH}} \lambda_{\text{SD}} \tanh\left(\frac{t_{\text{TI}}}{2\lambda_{\text{SD}}}\right) J_s$, and the spin pumping signal will follow [2, 3, 6]:

$$V_{\text{SP}} = -wR\theta_{\text{SH}}\lambda_{\text{SD}} \tanh\left(\frac{t_{\text{TI}}}{2\lambda_{\text{SD}}}\right) J_s, \quad (4)$$

where λ_{SD} is the spin diffusion length, t_{TI} is the thickness of the TI thin film and θ_{SH} is the spin Hall angle. The distinct difference between Eqs. (1) and (4) is whether the observed spin pumping signal is dominated by the spin momentum “locking” in the surface states [25–27] or by the SOC interaction.

To answer this question, Fig. 3(b) shows the Bi_2Se_3 thickness dependence of V_{SP} (blue points) and λ_{IREE} (or J_c/J_s) (red points), where we define $J_c = \frac{V_{\text{SP}}}{wR}$. Above 6 QL, J_c/J_s almost follows a constant value of about 35 pm. Below 6 QL, J_c/J_s dramatically decays by a factor of 30 from 35 ± 4 pm to 1.1 ± 0.13 pm when at 2 QL thickness. Earlier studies have

reported that the thickness of the Bi_2Se_3 surface states is approximately 2-3 nm [28, 29]. Above 6 QL, the top and bottom Bi_2Se_3 surface states decouple from each other; below 6 QL, the interaction of the two surface states with opposite spin polarizations can decrease the interfacial spin momentum “locking” efficiency. This is consistent with angle-resolved photoemission spectroscopy (ARPES) studies that show the opening of a gap in the Dirac cone when the Bi_2Se_3 thickness is below 6 QL, accompanied by a decrease in the spin polarization of the surface states [28, 29]. Qualitatively, our data shown in Fig. 3(b) follow this trend and strongly indicate the key role played by the surface states in spin-charge conversion in Bi_2Se_3 . If we try to interpret the data in Fig. 3(b) with the spin diffusion model (Eq. 4), the fit yields a value of $\lambda_{\text{SD}} \sim 1.6$ nm and also requires the presence of a “dead” layer at the interface (see Supplementary Material at [link to be added] for detailed analysis using the spin diffusion model). This short vertical spin diffusion length suggests that the spin polarized electron current is restricted to the bottom surface of the TI. Thus, while we cannot definitively rule out the spin diffusion model, a more physically meaningful picture at this stage is that the surface states probably play a dominant role in the spin-charge conversion. We note that the value we obtain for (J_c/J_s) (or λ_{FREE}) is approximately two orders of magnitude smaller than the spin Hall angle reported using a spin torque FMR study at room temperature [9]. One possible reason for this discrepancy is the amorphous layer at the interface shown in the HR-STEM figure, which potentially decreases the spin injection efficiency. Another reason may be the difference in the fundamental measurement mechanism between these two probing techniques. In a spin torque FMR experiment, as the charge current flows through the TI layers, the electrons can potentially have multiple scattering processes to transfer the spins to the ferromagnetic layers. However, in an FMR spin pumping measurement, this multiple scattering process may not be valid.

To further verify that the spin-charge conversion efficiency is dominated by the surface states of TIs, we grew five different TI heterostructures on YIG as control samples and measured their spin pumping signals. The five control samples are sample A: YIG/ $(\text{Bi,Sb})_2\text{Te}_3$ (6 QL); sample B: YIG/ Bi_2Se_3 (1 QL)/ $(\text{Bi,Sb})_2\text{Te}_3$ (6 QL); sample C: YIG/ Bi_2Se_3 (6 QL)/ $(\text{Bi,Sb})_2\text{Te}_3$ (6 QL); sample D: YIG/ Bi_2Se_3 (1 QL)/ $\text{Cr}_{0.2}(\text{Bi}_{0.5}\text{Sb}_{0.5})_{1.8}\text{Te}_3$ (6 QL); and sample E: YIG/ Bi_2Se_3 (6 QL)/ $\text{Cr}_{0.2}(\text{Bi}_{0.5}\text{Sb}_{0.5})_{1.8}\text{Te}_3$ (6 QL). Figure 3(c) shows the spin pumping spectra of control samples B, C, D and E at 3 GHz radio-frequency and 100 mW power. The enhancement of the spin pumping signal of samples D and E mainly results

from the larger resistivity of $\text{Cr}_{0.2}(\text{Bi}_{0.5}\text{Sb}_{0.5})_{1.8}\text{Te}_3$ compared to $(\text{Bi,Sb})_2\text{Te}_3$. Normalizing by the resistance and sample width, we obtained the spin charge conversion ratio of the five control samples and compared them with the values for $\text{YIG}/\text{Bi}_2\text{Se}_3$ in Fig. 3(d). First, the values of λ_{IREE} obtained for sample C and sample E are 37 ± 4 pm and 34 ± 4 pm, respectively. Both the values are quite close to 35 ± 4 pm measured for $\text{YIG}/\text{Bi}_2\text{Se}_3$ (6QL), indicating that as long as the Bi_2Se_3 thickness is above 6 QL, the spin-charge conversion efficiency is roughly constant and does not depend on the bulk properties: Cr doping and different band structures do not change the values. Second, for sample A, $(\text{Bi,Sb})_2\text{Te}_3$ directly grown on YIG, $\lambda_{\text{IREE}} = 17 \pm 2$ pm, about half of the value of Bi_2Se_3 . This is in sharp contrast with earlier results which reported a much larger spin Hall angle of the $(\text{Bi,Sb})_2\text{Te}_3$ compared with Bi_2Se_3 using a spin-polarized tunneling study [23]. This most likely results from the different interfacial quality and conditions that determine the spin momentum “locking” efficiency. We expect that the bottom surface state condition at the $\text{YIG}/(\text{Bi,Sb})_2\text{Te}_3$ interface [28] is not as good as the $\text{CoFeB}/\text{MgO}/(\text{Bi,Sb})_2\text{Te}_3$ interface [23] for which TI was grown on the commercial InP substrates with minimal lattice mismatch and the highest sample quality. In the end, we compare the values in samples B and D that both have 1 QL Bi_2Se_3 seed layers. For sample D, we intentionally dope the $(\text{Bi,Sb})_2\text{Te}_3$ with Cr, which can induce ferromagnetism at low temperature [30]. At room temperature, the Cr doping mainly changes the transport properties and the SOC strength of the bulk states. The values for samples B and D are $\lambda_{\text{IREE}} = 20 \pm 2$ pm and 22 ± 3 pm, respectively. Their similar spin-charge conversion efficiencies demonstrate that the properties of the TI bulk state do not play a significant role here, confirming the interface-dominated spin pumping phenomena. It is also important to note that values of λ_{IREE} for samples B and D are lower than the value for $\text{YIG}/\text{Bi}_2\text{Se}_3$ (6QL). As in other studies of spin pumping into TIs, the interfacial condition presents a critical challenge for controlling the spin conversion efficiency [8, 11, 12]; in sample B, both $\text{YIG}/\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Se}_3 / (\text{Bi,Sb})_2\text{Te}_3$ interfaces will contribute to the formation of the surface states. Thus, structural defects and/or strain induced dislocations in the trilayer heterostructures can potentially result in the observed lower values. A thorough understanding about the correlation of the interfacial conditions of TI surfaces states and the spin-charge conversion efficiency requires further investigation.

Finally, we compare the spin transfer efficiency at $\text{YIG}/\text{Bi}_2\text{Se}_3$ to that at YIG/Pt . Note that Pt is an ideal spin sink and a well-studied non-magnetic material with large SOC [3, 6].

Figure 4(a) shows the inverse spin Hall spectrum of a YIG(30nm)/Pt(5nm) bilayer sample under 3 GHz and 100 mW microwave power when the H field is in plane. The observed sign change of the spin pumping signal with field reversal is expected for the ISHE in a 3D material system [2, 3]. From the FMR linewidth broadening, the obtained YIG/Pt effective spin mixing conductance is $(5.19 \pm 0.6) \times 10^{18} \text{m}^{-2}$, which lies in the range of the values reported by other groups using spin pumping [5, 6]. We compare this value with the obtained spin mixing conductance at various Bi_2Se_3 thicknesses in Fig. 4(b). When Bi_2Se_3 is 6 QL thick, the spin mixing conductance at the YIG/ Bi_2Se_3 interface is $(4.13 \pm 0.5) \times 10^{18} \text{m}^{-2}$. Although there are some variations, the reported values are in the range of $3 - 7 \times 10^{18} \text{m}^{-2}$ when the Bi_2Se_3 thickness varies from 2 to 60 QL, which is essentially comparable to the determined value at the YIG/Pt interface, demonstrating an efficient spin transfer in YIG/TI heterostructures. It is important to note that in the large Bi_2Se_3 thickness regime, we do not observe an enhancement of $g_{\uparrow\downarrow}$, which is typically observed in the YIG/transition metal bilayers due to the decrease in backflow spin current caused by the spin diffusion in the bulk [24, 32]. This also confirms the TI surface states dominated spin-charge conversion mechanism.

In conclusion, we report robust spin pumping at room temperature in YIG/ Bi_2Se_3 bilayers and other YIG/TI heterostructures. By measuring IREE voltages and interfacial spin current density, we determine the value of λ_{IREE} and reveal its systematic behavior with Bi_2Se_3 thickness, demonstrating the dominant role of surface states in spin-charge conversion. The inferred IREE length indicates the important role of interface conditions in spin Hall physics in topological insulators. Further investigation is required for a thorough understanding of the correlation between the formation of the surface states and the variation of spin-charge conversion efficiency at the interfaces.

The work at Penn State, Colorado State and University of Minnesota is supported by the Center for Spintronic Materials, Interfaces, and Novel Architectures (C-SPIN), a funded center of STARnet, a Semiconductor Research Corporation (SRC) program sponsored by MARCO and DARPA. NS and AR acknowledge additional support from ONR- N00014-15-1-2364. TL, HC, and MW acknowledge additional support from NSF-ECCS-1231598 and ARO-W911NF-14-1-0501. This work utilized (1) the College of Science and Engineering (CSE) Characterization Facility, University of Minnesota (UM), supported in part by NSF through the UMN MRSEC program (No. DMR-1420013); and (2) the CSE Minnesota Nano

Center, UM, supported in part by NSF through the NNIN program.

* nsamarth@psu.edu

- [1] Y. Kajiwara, K. Harii, S. Takahashi, J. Ohe, K. Uchida, M. Mizuguchi, H. Umezawa, H. Kawai, K. Ando, K. Takanashi, S. Maekawa, and E. Saitoh, *Nature* **464**, 262 (2010).
- [2] B. Heinrich, C. Burrowes, E. Montoya, B. Kardasz, E. Girt, Y.-Y. Song, Y. Y. Sun, and M. Z. Wu, *Phys. Rev. Lett.* **107**, 066604 (2011).
- [3] O. Mosendz, V. Vlaminck, J. E. Pearson, F. Y. Fradin, G. E. W. Bauer, S. D. Bader, and A. Hoffmann, *Phys. Rev. B* **82**, 214403 (2010).
- [4] C. H. Du, H. L. Wang, Y. Pu, T. L. Meyer, P. M. Woodward, F. Y. Yang, and P. C. Hammel, *Phys. Rev. Lett.* **111**, 247202 (2013).
- [5] C. Hahn, G. de Loubens, O. Klein, M. Viret, V. V. Naletov, and J. Ben Youssef, *Phys. Rev. B* **87**, 174417 (2013).
- [6] H. L. Wang, C. H. Du, Y. Pu, R. Adur, P. C. Hammel, and F. Y. Yang, *Phys. Rev. Lett.* **112**, 197201 (2014).
- [7] J.-C. Rojas-Snchez, L. Vila, G. Desfonds, S. Gambarelli, J. P. Attan, J. M. De Teresa, C. Magn, and A. Fert, *Nat. Commun.* **4**, 2944 (2013).
- [8] Y. Shiomi, K. Nomura, Y. Kajiwara, K. Eto, M. Novak, K. Segawa, Y. Ando, and E. Saitoh, *Phys. Rev. Lett.* **113**, 196601 (2014).
- [9] A. R. Mellnik, J. S. Lee, A. Richardella, J. L. Grab, P. J. Mintun, M. H. Fischer, A. Vaezi, A. Manchon, E.-A. Kim, N. Samarth, and D. C. Ralph, *Nature* **511**, 449 (2014).
- [10] Y. Fan, P. Upadhyaya, X. Kou, M. Lang, S. Takei, Z. Wang, J. Tang, L. He, L.-T. Chang, M. Montazeri, G. Yu, W. Jiang, T. Nie, R. N. Schwartz, Y. Tserkovnyak, and K. L. Wang, *Nat. Mater.* **13**, 699 (2014).
- [11] P. Deorani, J. Son, K. Banerjee, N. Koirala, M. Brahlek, S. Oh, and H. Yang, *Phys. Rev. B* **90**, 094403 (2014).
- [12] M. Jamali, J. S. Lee, J. S. Jeong, F. Mahfouzi, Y. Lv, Z. Zhao, B. K. Nikolic, K. A. Mkhoyan, N. Samarth, and J.-P. Wang, *Nano Lett.* **15**, 7126 (2015).
- [13] A. A. Baker, A. I. Figueroa, L. J. Collins-McIntyre, G. van der Laan, and T. Hesjeda, *Scientific Reports* **5**, 7907 (2015).

- [14] Y. Wang, P. Deorani, K. Banerjee, N. Koirala, M. Brahlek, S. Oh, and H. Yang, *Phys. Rev. Lett.* **114**, 257202 (2015).
- [15] K. Kondou, R. Yoshimi, A. Tsukazaki, Y. Fukuma, J. Matsuno, K. S. Takahashi, M. Kawasaki, Y. Tokura, and Y. Otani, arXiv:1510.03572.
- [16] D. Hsieh *et al.*, *Nature* **460**, 1101 (2009).
- [17] M. Z. Hasan and C. L. Kane, *Rev. Mod. Phys.* **82**, 3045 (2010).
- [18] X. -L. Qi and S. -C. Zhang, *Rev. Mod. Phys.* **83**, 1057 (2011).
- [19] H. Chang, P. Li, W. Zhang, T. Liu, A. Hoffmann, L. Deng, and M. Z. Wu, *IEEE Magn. Lett.* **5**, 6700104 (2014).
- [20] A. Kandala, A. Richardella, D. W. Rench, D. M. Zhang, T. C. Flanagan, and N. Samarth, *Appl. Phys. Lett.* **103**, 202409 (2013).
- [21] J. S. Lee, A. Richardella, D. Reifsnnyder Hickey, K. A. Mkhoyan, and N. Samarth, *Phys. Rev. B* **92**, 155312 (2015).
- [22] V. M. Edelstein, *Solid State Commun.* **73**, 225 (1990).
- [23] K. Shen, G. Vignale, and R. Raimondi, *Phys. Rev. Lett.* **112**, 096601(2014).
- [24] Y. Tserkovnyak, A. Brataas, G. E.W. Bauer, and B. I. Halperin, *Rev. Mod. Phys.* **77**, 1375 (2005).
- [25] L. Liu, A. Richardella, I. Garate, Y. Zhu, N. Samarth, and C.-T. Chen, *Phys. Rev. B* **91**, 235437 (2015).
- [26] J. Tang, L.-T. Chang, X. Kou, K. Murata, E. S. Choi, M. Lang, Y. Fan, Y. Jiang, M. Montazeri, W. Jiang, Y. Wang, L. He, and K. L. Wang, *Nano Lett.* **14**, 5423 (2014).
- [27] C. H. Li, O. M. J. van't Erve, J. T. Robinson, Y. Liu, L. Li, and B. T. Jonker, *Nat. Nanotechnol.* **9**, 218 (2014).
- [28] Y. Zhang *et al.*, *Nat. Phys.* **6**, 584 (2010).
- [29] M. Neupane *et al.*, *Nat. Commun.* **5**, 3841 (2014).
- [30] Z. Jiang, C.-Z. Chang, C. Tang, P. Wei, J. S. Moodera, and J. Shi, *Nano Lett.* **15**, 5835 (2015).
- [31] A. Kandala, A. Richardella, S. Kempinger, C.-X. Liu, and N. Samarth, *Nat. Commun.* **6**, 7434 (2015).
- [32] H. J. Jiao and G. E. W. Bauer, *Phys. Rev. Lett.* **110**, 217602 (2013).

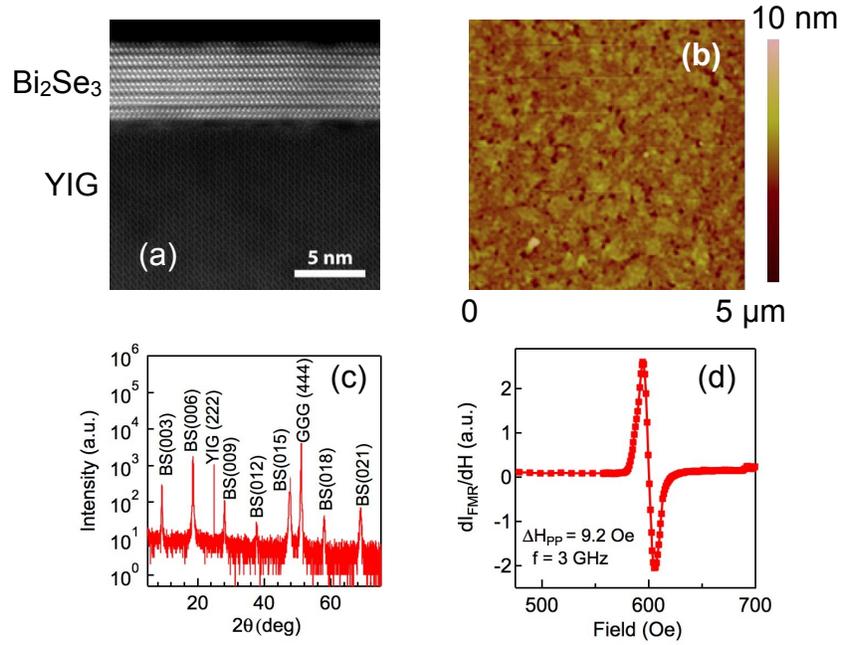


FIG. 1. (Color online) (a) Cross-sectional high-angle annular dark-field scanning TEM image of the YIG/Bi₂Se₃ interface. (b) Atomic force microscopy image of the YIG/Bi₂Se₃ (6QL) sample with a surface roughness of 0.71 nm. (c) Semi-log $\theta - 2\theta$ XRD scan of a YIG/Bi₂Se₃ (40QL) sample which exhibits clear x-ray scattering peaks from the (003) to (0021) planes of Bi₂Se₃. (d) A representative room-temperature FMR derivative spectrum of a 30-nm YIG film with an in-plane field, which gives a peak-to-peak line width of 9.2 Oe at $f = 3$ GHz.

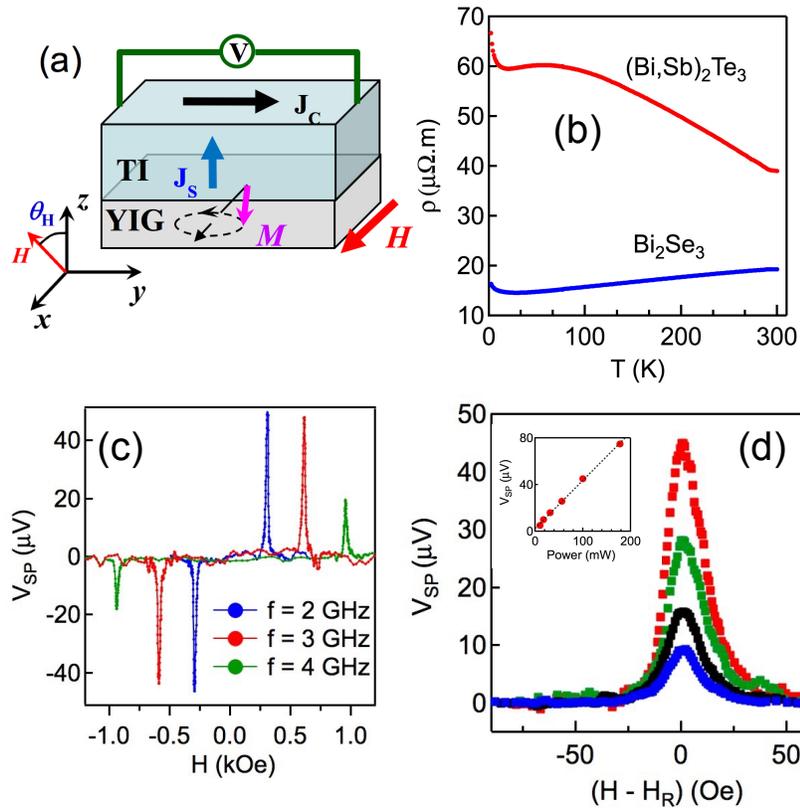


FIG. 2. (Color online) (a) Schematic of the experimental setup for FMR spin pumping measurements. (b) Resistivity of 6 QL Bi_2Se_3 and $(\text{Bi,Sb})_2\text{Te}_3$ thin films grown on YIG as a function of temperature. (c) V_{SP} vs. H spectra of YIG/ Bi_2Se_3 (6QL) at $f = 2, 3,$ and 4 GHz using 100 mW microwave power. (d) V_{SP} vs. H spectra of the YIG/ Bi_2Se_3 (6QL) sample for the microwave power of $18, 32, 56,$ and 100 mW at $f = 3\text{GHz}$. Inset: rf-power dependence of the corresponding V_{SP} at $\theta_H = 90^\circ$.

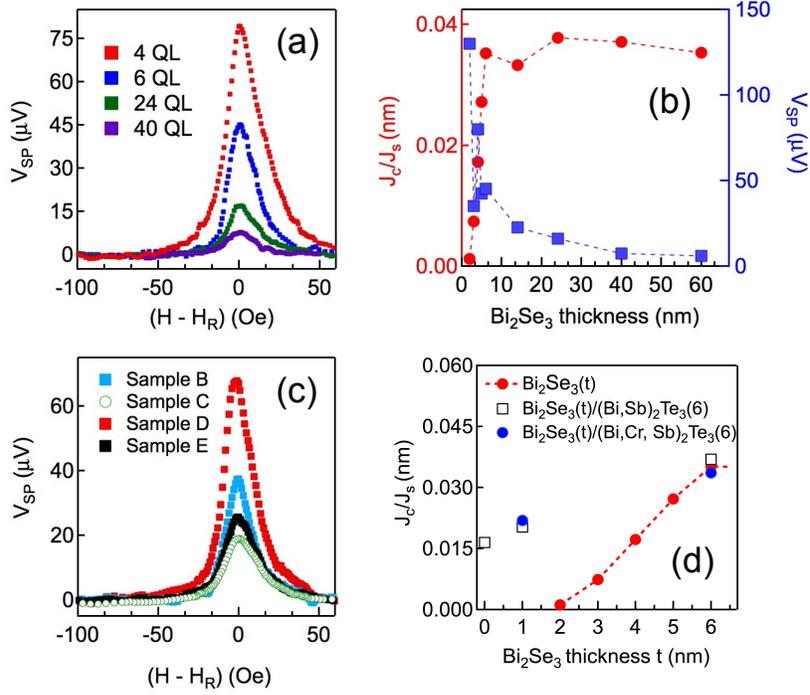


FIG. 3. (Color online) (a) V_{SP} vs. H spectra of YIG/Bi₂Se₃ (4QL), YIG/Bi₂Se₃ (6QL), YIG/Bi₂Se₃ (24QL), and YIG/Bi₂Se₃ (40QL) at $f = 3$ GHz using 100 mW microwave power. The x -axis is shifted by the resonance field (H_R) for clarity. (b) Dependence of V_{SP} (blue points) and the spin-to-charge conversion efficiency J_c/J_s determined by λ_{IREE} (red points) on the Bi₂Se₃ thickness. (c) V_{SP} vs. H spectra of control sample B: YIG/Bi₂Se₃(1QL)/(Bi,Sb)₂Te₃(6QL) (blue curve); sample C: YIG/Bi₂Se₃(6QL)/(Bi,Sb)₂Te₃(6QL) (green curve); sample D: YIG/Bi₂Se₃(1QL)/Cr_{0.2}(Bi_{0.5}Sb_{0.5})_{1.8}Te₃(6QL) (red curve), and sample E: YIG/Bi₂Se₃(6QL)/Cr_{0.2}(Bi_{0.5}Sb_{0.5})_{1.8}Te₃(6QL) (black curve) at 3 GHz and 100 mW. (d) Comparison of J_c/J_s for the control samples with the corresponding values for YIG/Bi₂Se₃.

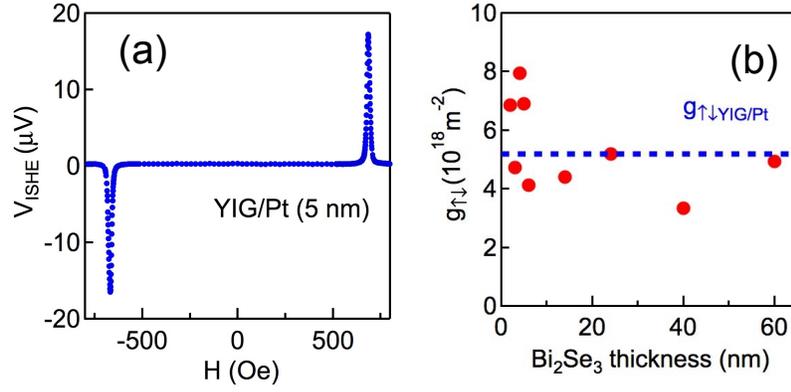


FIG. 4. (Color online) (a) V_{ISHE} vs. H spectra of YIG/Pt (5nm) bilayer at radio-frequency of 3 GHz and 100 mW microwave power. (b) Dependence of the YIG/ Bi_2Se_3 interfacial spin mixing conductance $g_{\uparrow\downarrow}$ on Bi_2Se_3 thickness. The blue dashed line indicates the value of $g_{\uparrow\downarrow}$ at the YIG/Pt interface.