

# Role of Cu Layer on the Enhancement of Spin-to-Charge Conversion in Py/Cu/Bi<sub>2</sub>Se<sub>3</sub>

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## Research Article

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# Abstract

The enhancement of spin-to-charge conversion in Py/Cu/Bi<sub>2</sub>Se<sub>3</sub> is achieved when increasing the Cu layer thickness up to 7nm. The conversion rate is studied using spin pumping technique. The inverse IEE length  $\lambda_{\text{IEE}}$  is found to increase up to  $\sim 2.7\text{nm}$  when 7nm Cu layer is introduced. Interestingly, maximized  $\lambda_{\text{IEE}}$  is obtained when the effective spin mixing conductance (and thus  $J_s$ ) is decreased due to the Cu insertion. Monotonic increase of  $\lambda_{\text{IEE}}$  with decreasing  $J_s$  suggests that IEE relaxation time  $\tau$  is enhanced due to additional tunnelling barrier (Cu) that limits the interface transmission rate. The results have shown the importance of interface engineering in Py/TI magnetic heterostructure, which is the key factor for optimizing spin-to-charge conversion efficiency.

# Introduction

Manipulation of spin current by electrical charges or voltages is one of the key subjects for new generation of spintronic devices. Three dimensional (3D) topological insulators (TI) are great candidate for spintronic applications, since their spin-momentum locked properties enable spin accumulation on the surface by passing electrical charge current through the TI channel.<sup>1,2</sup> In addition, the degree of spin polarization can be further manipulated by controlling the Fermi level.<sup>3,4</sup> Owing to the spin momentum locking, 3D spin current density  $J_s$  injected onto TI surface will produce 2D charge density  $J_c$  on TI surface states (SS), so-called inverse Edelstein effect (IEE). The inverse IEE length  $\lambda_{\text{IEE}}$  is determined as  $J_c/J_s$  and can be experimentally probed using spin pumping technique.<sup>5,6,7,8</sup>

Numerous studies have been reported for determining the spin-to-charge conversion efficiency in 3D TI.<sup>5,9,10,11</sup> Particularly, giant spin hall angle (SHA) as large as  $\sim 0.43$  was reported in Bi<sub>2</sub>Se<sub>3</sub> that attributed to the enhanced spin current by the surface states and then converted into dc-voltage due to the bulk inverse spin hall effect.<sup>9</sup> However, large variations of SHA was found, which is an order of magnitude difference and the authors ascribed such variation to the non-uniformity of interface quality.<sup>9</sup> On the other hand, Wang et al. observed the dominant role of surface states in spin-to-charge conversion, despite the unavoidable conducting bulk in Bi<sub>2</sub>Se<sub>3</sub>.<sup>5</sup> The effective spin mixing conductance was not increased monotonically although the thickness of Bi<sub>2</sub>Se<sub>3</sub> varied from 2 QL to 60 QL, suggesting surface states dominated mechanism.<sup>5</sup> Obviously, the spin pumping characteristic are important parameters for investigating the spin-to-charge conversion mechanism in 3D TI, where controlling the interfacial properties are necessary steps.

Cu is widely used to control the spin transmissivity in the multilayer devices.<sup>12,13,14</sup> Du et al. demonstrated that the insertion of Cu layer between Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> (YIG) and W substantially improved the spin current injection into W, while similar insertion between YIG and Pt degraded the spin current.<sup>13</sup> The authors reported the quantitative analysis and found that the spin transport efficiency in heterostructures depends on spin conductance of each constituents and their interfaces.<sup>13</sup> Similar results were also

reported by Deorani et al. where the effect of Cu interlayer on spin mixing conductance indeed material dependent (Pt versus Ta).<sup>14</sup> Recently Cu layer was deposited on TI film in order to eliminate the proximity induced ferromagnetism in the spin-orbit torque (SOT) devices.<sup>1,15</sup> Despite the fact that Cu is the most common used spacer layer in the spintronic devices, there is still lacking of quantitative studies about the role of Cu insertion on spin-to-charge conversion in TI that measured based on spin pumping mechanism. In this work, we fabricated trilayer structure of Py/Cu/Bi<sub>2</sub>Se<sub>3</sub> (Fig. 1) and studied the spin pumping characteristic by varying the Cu layer thickness. Cu layer is used to protect the TI surface from exchange interaction with Py. Our results imply that Cu also acts as the barrier to spin transmission into TI film. More importantly, spin-to-charge conversion efficiency was enhanced due to the introduction of Cu barrier. The related mechanism is discussed in this work.

## Results And Discussion

Figure 2a shows the dc voltage as a function magnetic field H that measured at excitation frequency of 3GHz for sample Py/Cu(7nm)/Bi<sub>2</sub>Se<sub>3</sub>. The results for other frequencies can be found in Figure S2. The voltage signals consisting of symmetric ( $V_s$ ) and antisymmetric ( $V_{as}$ ) parts, which can be isolated by fitting the measured voltage (data curve) with the form

$$V = \frac{V_s(\Delta H)^2}{((\Delta H)^2 + (H - H_r)^2)} + \frac{V_{as}(\Delta H(H - H_r))}{((\Delta H)^2 + (H - H_r)^2)}$$

Here  $H_r$  is the FMR resonant field and  $\Delta H$  is the line width of the signal.  $V_s$  is attributed to the injected spin current whereas  $V_{as}$  originated from anisotropic magnetoresistance (AMR) or anomalous Hall effect (AHE). Similar fitting is also done for other samples and  $V_s$  is extracted as shown in Figure 2b. It was found that  $V_s$  is larger in the presence of Cu layer. FMR experiments were also conducted as shown in Figures 2c and 2d. Figure 2d presents the frequency dependence of FMR line width for samples with different Cu thickness. The linear fitted slope is larger for bilayer and trilayer samples (Cu0, Cu3 and Cu7 denote  $t_{Cu}=0, 3, 7$ nm respectively) in comparison to single Py, indicating spin current injected into Bi<sub>2</sub>Se<sub>3</sub>, resulting in the broadening of the FMR line width and thus larger damping constant  $a$ . Interestingly, the  $a_{Py/Cu/TI}$  was found decreases from 0.01262 to 0.01185 when increasing the thickness of Cu layer up to 7nm.

The resistance of the multilayer samples  $R_d$  were measured using four probe method.  $J_c$  is determined as  $J_c = I_c/w = V_s/wR_d$ , where  $w$  and  $I_c$  are the width of the sample and charge current as shown in Figure 3a. We used the standard analysis of spin pumping on TI<sup>5,6,7</sup> to evaluate the spin-to-charge conversion  $J_c/J_s$ . Spin mixing conductance  $G_{eff}^-$  which is used to illustrate the efficiency of generating spin current is extracted using Equation (1):

$$G_{eff}^{\uparrow\downarrow} = \frac{4\pi t_{Py} M_s}{g u_B} \Delta\alpha \quad (1)$$

where  $M_s$  is saturation magnetization of Py,  $t_{Py}$  is thickness of Py and  $g$  is Landé factor and  $u_B$  is the Bohr magneton.  $M_s$  is calculated using Kittel formula from  $f$  vs.  $H_r$  (Figure 2c).  $Da = a_{Py/Cu/TI} - a_{Py}$  and is determined by analysing  $DH_{pp}$  vs.  $f$  as shown in Figure 2d. For the spin current densities that injected across the interface due to spin pumping, Equation (2) is utilized as shown below:

$$J_s^{3D} = \frac{G_{eff}^2 \gamma^2 \hbar h_{rf}^2}{8 \pi \alpha^2} \left( \frac{4\pi M_s \gamma + \sqrt{(4\pi M_s \gamma)^2 + 4\omega^2}}{(4\pi M_s \gamma)^2 + 4\omega^2} \right) \frac{2e}{\hbar} \quad (2)$$

in which  $g$  is gyromagnetic ratio,  $w(=2\pi f)$  and  $h_{rf}$  are frequency and amplitude of microwave magnetic field respectively. The calculated  $J_s$  is presented in Figure 3b. By dividing  $J_c$  with  $J_s$ , spin-to-charge conversion efficiency  $J_c/J_s$  can be determined as shown in Figure 3c.

Figure 3a plots the  $J_c$  against  $t_{Cu}$ . There is an optimized  $J_c$  at thickness of 3 and 7nm. In contrast,  $J_s$  is reduced when 3 and 7nm Cu were incorporated. The trend of  $J_s$  vs  $t_{Cu}$  is consistent with the changes of  $a_{Py/Cu/TI}$ , where effective damping constant is found decreases when 3 and 7nm Cu were introduced. Interestingly, a maximized  $J_c/J_s$  is observed at  $t_{Cu}=7$ nm, where  $J_c/J_s$  reaches  $\sim 2.7$ . This result suggests that the optimization of  $J_c/J_s$  could be related to the decrease of  $J_s$  due to the Cu insertion. To investigate the possible reason for the  $J_c/J_s$  enhancement, we plot  $J_c/J_s$  as a function of effective spin mixing conductance  $G_{eff}^-$  as shown in Figure 4a. Various  $G_{eff}^-$  values were obtained by changing the Cu layer thickness. Large  $J_c/J_s$  is obtained at low value of  $G_{eff}^-$  (and thus minimum  $J_s$  as shown in Figure 3b). We further examined the  $J_c$  vs.  $G_{eff}^-$  as shown in Figure 4b. There is no enhancement of  $J_c$  with increasing  $G_{eff}^-$ , revealing the spin-to-charge mechanism may not be dominated by the bulk spin hall effect (SHE).<sup>14</sup> Hence, we here suggest that the spin-to-charge conversion in our Py/Cu/TI system arises from inverse Edelstein effect, where the origin is the spin-momentum locked surface states of TI layer, same interpretation as other literatures.<sup>5,11</sup>

Low  $G_{eff}^-$  indicates strong spin backflow and spin memory loss (spin absorption) at the high SOC interface.<sup>16,17</sup> Both factors are relevant in this Py/Cu/TI trilayer system. If we examine  $G_{eff}^-$  (Py/Cu/TI) at various  $t_{Cu}$ , as presented in Figure 4c, except for Py/Cu(3nm)/TI and Py/Cu(7nm)/TI, samples Py/TI, Py/Cu(9nm)/TI and Py/Cu(11nm)/TI exhibit  $G_{eff}^- \sim 1.25 \times 10^{19} \text{ m}^{-2}$ , which is the typical value for metal-metal interfaces.<sup>18,19</sup> As reported by Du et al., the effective spin mixing conductance of trilayer system (FM/Cu/NM, FM denotes ferromagnetic while NM denotes nonmagnetic material) is determined by the serial contribution of the two interfaces (FM/Cu and Cu/NM) and the spin resistance of Cu.<sup>13</sup> Here we

refer FM to Py while NM to TI film. One of the reason for the lower  $G_{\text{eff}}^-$  compared with  $G_{\text{Py/TI}}^-$  may be resulted from smaller spin conductance  $g_{\text{Cu/TI}}$  of Cu/TI than that of  $G_{\text{Py/TI}}^-$ , similar to the case in Cu/Pt.<sup>13,14</sup> However, since  $G_{\text{eff}}^- \gg G_{\text{Py/TI}}^-$  at  $t_{\text{Cu}} \approx 9\text{nm}$ , here we assume Cu/TI and Py/Cu present similar quality with  $G_{\text{Py/Cu}}^- \gg g_{\text{Cu/TI}}$ . Thus, by assuming degree of spin absorption at Cu/TI interfaces are similar in all cases, we suggest that the reason for lower  $G_{\text{eff}}^-$  of 3nm and 7nm Cu-based trilayer samples could be due to the strong spin accumulation at this ultrathin regime.<sup>13</sup> Stronger spin backflow occurs, as compared to  $t_{\text{Cu}} \approx 9\text{nm}$ , which eventually leads to reduction of  $G_{\text{eff}}^-$ .

The decrease of  $G_{\text{eff}}^-$  seems have strong correlation to the spin-to-charge conversion efficiency. The next question is how such condition could increase the  $J_c/J_s$ ? Here we defined  $J_c/J_s$  as  $I_{\text{IEE}} = n_f \lambda$  where  $n_f$  is Fermi velocity of TI surface states and  $t$  is IEE relaxation time. As shown in the Figure 4d,  $t$  is modified due to the tunnelling current into TI, which is determined by momentum relaxation time  $t_p$  and interface tunnelling time  $t_t$  as shown in Equation (3):<sup>20</sup>

$$\begin{aligned} \lambda_{\text{IEE}} &= v_f \tau \\ &= v_f \frac{\tau_p}{\left(1 + \frac{2\tau_p}{\tau_t}\right)} \end{aligned} \quad (3)$$

where  $l_{\text{mf}} = n_f t_p =$  mean free path TI. According to this model, we proposed that the monotonic increase of  $I_{\text{IEE}}$  with decreasing  $G_{\text{eff}}^-$  attributed to the modification of IEE relaxation time  $t$  due to additional tunnelling barrier (Cu) that limits the interface transmission rate ( $1/t_t$ ).<sup>20,21</sup>  $I_{\text{IEE}}$  is always lower than  $l_{\text{mf}}$  due to the correction factor of  $(1 + 2t_p/t_t)$ . It becomes clear that one can increase  $I_{\text{IEE}}$  by reducing  $1/t_t$ , which can be done by introducing tunnelling barrier in between Py and TI layer. Using  $I_{\text{IEE}}(t_{\text{Cu}}=7\text{nm}) = 2.7\text{nm}$  and based on our previous ARPES result,  $n_f = 5.7 \times 10^5 \text{ m/s}$ ,<sup>22</sup> we find  $t \sim 4.7\text{fs}$ , which is same order of magnitude as Bi/Ag<sup>23</sup> and a-Sn/Ag<sup>6</sup> interfaces. Our extracted  $I_{\text{IEE}} (= 2.7\text{nm})$  is higher than 0.1-0.4nm in Bi/Ag Rashba interface,<sup>23</sup> 2.1nm and 2nm in TI SS of a-Sn/Ag<sup>6</sup> and HgTe/HgCdTe<sup>7</sup> respectively. We attribute the enhancement to the insertion of Cu tunnelling barrier. Although more theoretical calculation might be needed, our works indicate the importance of interface engineering to enhance the spin-to-charge conversion. This method could also be applied to other high SOC interfaces for obtaining high spin-to-charge conversion based on the inverse Edelstein effect, which is essential for spin current detector and other novel application such as broadband terahertz emitter.<sup>24,25</sup>

## Conclusion

In conclusion, we investigated the spin-to-charge conversion in Py/Cu/Bi<sub>2</sub>Se<sub>3</sub> using spin pumping technique. Enhancement of  $J_c/J_s$  with increasing  $t_{Cu}$  is observed at room temperature, where  $J_c/J_s \sim 2.7$  is achieved when 7nm of Cu layer is inserted. We proposed that the enhancement is attributed to the additional Cu interlayer as tunnelling barrier that modified the relaxation time at the interface. This work has provided a viable route for enhancing spin-to-charge conversion efficiency of TI, which is crucial for spin functional device applications.

## Materials And Methods

Bi<sub>2</sub>Se<sub>3</sub> films with 10nm thickness were synthesized using molecular beam epitaxy (MBE) method. The as-grown Bi<sub>2</sub>Se<sub>3</sub> were *in situ* capped with 2nm Se film that used as protecting layer. The sample was then transferred to pulse laser deposition (PLD) chamber for depositing Cu and subsequently NiFe (Py) layer at room temperature. Before the depositions, Se layer was decapped in PLD chamber at 183°C for 1 hour. A series of trilayer samples were prepared by varying Cu thickness, ranging from 3 to 11nm. Bilayer of Py/Bi<sub>2</sub>Se<sub>3</sub> was also prepared for comparison study. Py thickness was fixed at 17 nm. 1 nm of Al film was deposited on Py as capping layer. To evaluate spin-to-charge conversion, spin pumping technique was utilized (Fig. 1). Spin current was generated in Py via its ferromagnetic resonance (FMR) condition and injected into Bi<sub>2</sub>Se<sub>3</sub>, either with or without passing through the Cu layer (-z direction) (Fig. 1b). DC voltage was measured in x-direction and the produced charge current could be evaluated. All the measurements were done at room temperature.

## Declarations

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### Author contributions

S. H. S. and C.-W. C. designed the experiment flow, analyzed data, and wrote the main manuscript. Y.-X. H. grew the samples and performed the measurement. Y.-C. C. and V. V. M. provided useful discussion. J.-C.-A. H. financed the funding for this research.

**Competing financial interests:** The authors declare no competing financial interests.

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## Figures

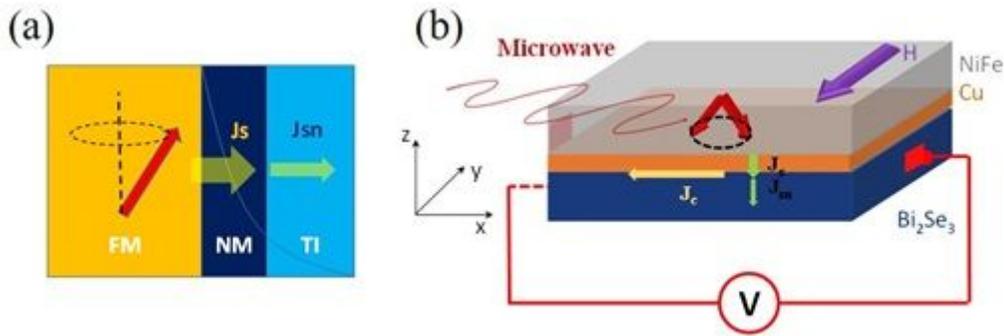


Figure 1

(a) Tri-layer samples for spin pumping study, where nonmagnetic (NM) spacer was introduced in between FM and TI layer; (b) schematic illustrates the spin pumping experiment.

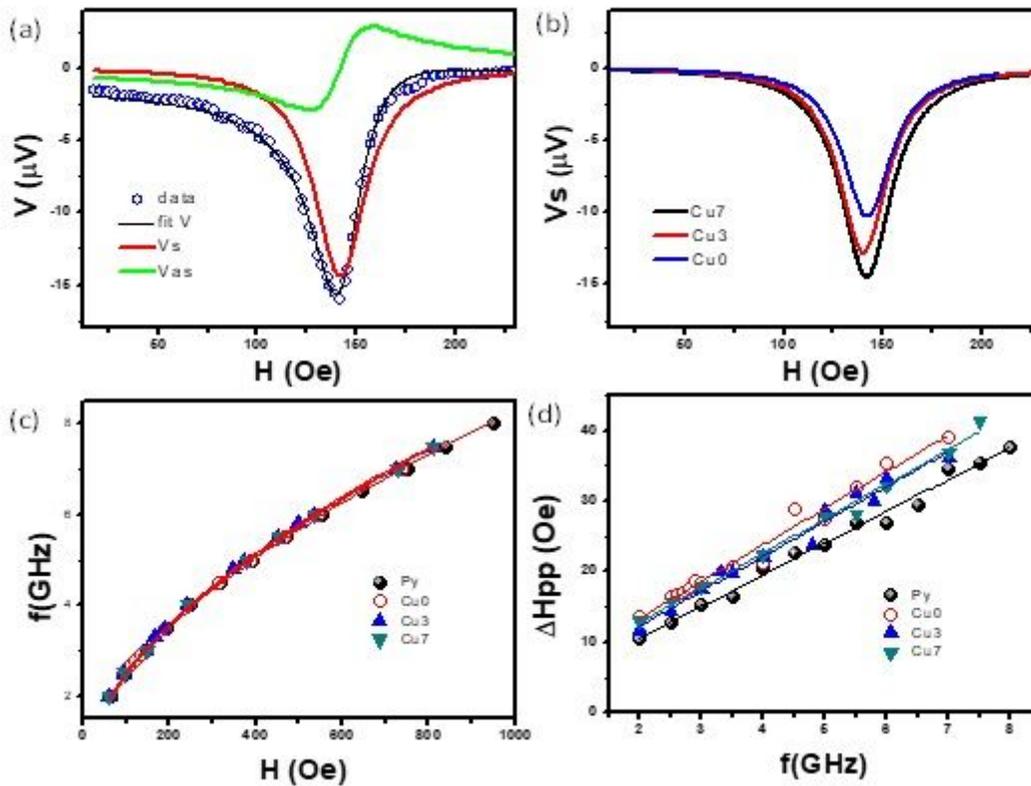


Figure 2

(a) Measured dc voltage at 3GHz for Py/Cu(7nm)/Bi<sub>2</sub>Se<sub>3</sub>; (b) extracted  $V_s$  for various samples; (c) excitation frequency as a function of resonant field. The solid lines are the fitted curves using Kittel formula; (d) frequency dependence of FMR line width for samples with different Cu thickness. The solid lines show the linear fitting, from which the damping factor( $\alpha$ ) of each sample is derived.

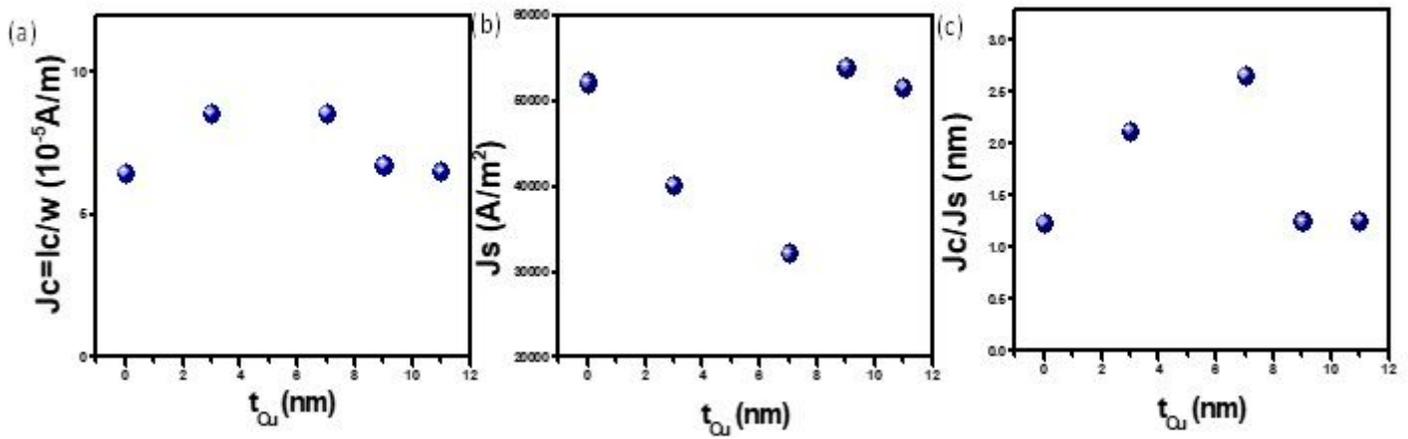


Figure 3

(a)  $J_c$  vs  $t_{\text{Cu}}$ ; (b)  $J_s$  vs  $t_{\text{Cu}}$ ; (c)  $J_c/J_s$  vs  $t_{\text{Cu}}$  measured at 3GHz excitation frequency.

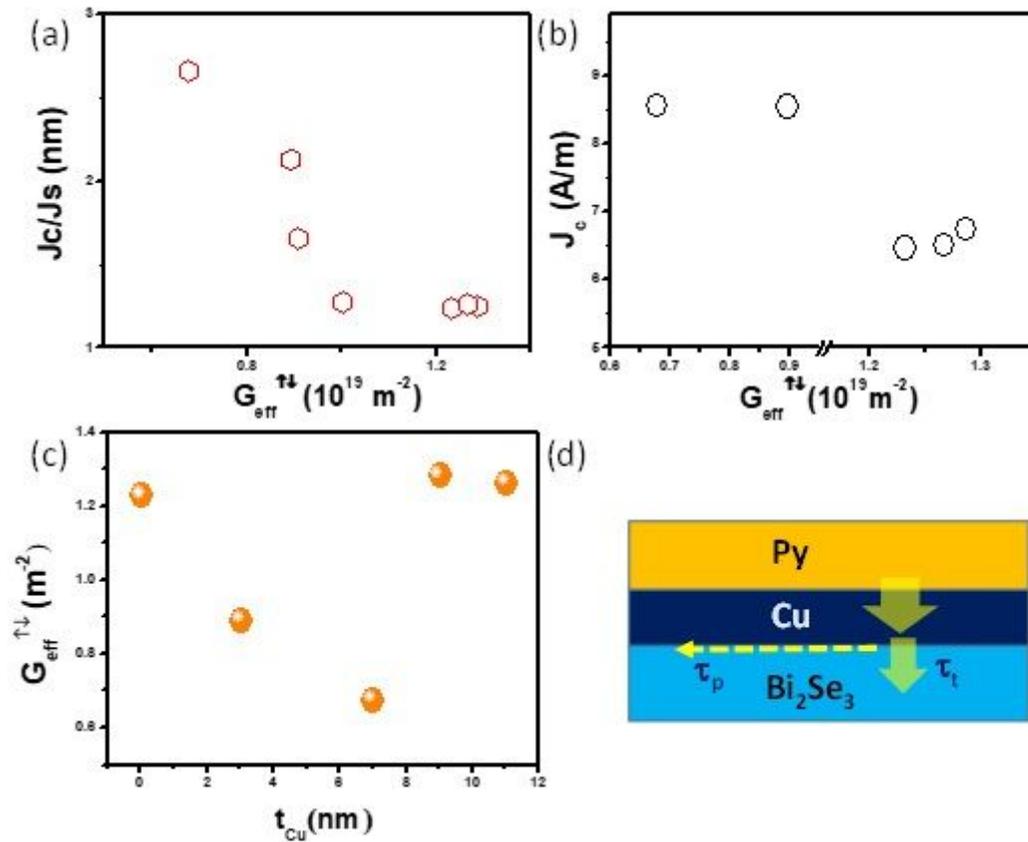


Figure 4

(a)  $J_c/J_s$  at various  $G_{\text{eff}}^{\uparrow\downarrow}$ ; (b)  $J_c$  at various  $G_{\text{eff}}^{\uparrow\downarrow}$ ; (c)  $G_{\text{eff}}^{\uparrow\downarrow}$  vs.  $t_{\text{Cu}}$ ; (d) schematic illustrates the spin transport in Py/Cu/TI.

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