Room Temperature Mott Hopping and Spin pumping
Characterization of Amorphous Gd-alloyed Bi$_2$Se$_3$

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Abstract

Disordered spin-orbit channel films have gained intense interest because of their possibility for spintronics applications by benefiting from other exotic transport properties. Here, we have fabricated disordered Gd-alloyed Bi$_x$Se$_{1-x}$ (BSG) thin films by magnetron sputtering methods and have investigated their magneto-transport and spin-torque properties. Structural characterizations show a mainly amorphous feature for the 8nm thick BSG film, while Bi rich crystallites are developed inside the 16nm thick BSG film. The bulk resistivity of BSG film is found to be relatively high, up to 6×10$^4$μΩ·cm, with respect to the resistivity of the polycrystalline Bi$_x$Se$_{1-x}$ film. Temperature dependent resistivity measurements display the evident character of a variable range hopping transport from 80K to 300K. Spin pumping transport characterizations have been performed on the BSG(t)/CoFeB(5 nm) bilayer structures with different thickness of BSG (t= 6, 8, 12, 16 nm). The possible various origins of the spin-to-charge conversion are related to extrinsic effects. Our study provides a new experimental direction, beyond crystalline solids, to the search for strong SOC systems in amorphous solids and other engineered random systems.
Keywords: spin-orbit torque materials, spin Hall angle; charge to spin conversion; variable range hopping.
Introduction

Bismuth selenide (BiSe) based topologic insulator (TI) materials have recently become extremely interesting in the field of condensed matter physics thanks to their exotic transport properties. Recent interests have garnered from the fact that these materials can exhibit topologically protected surface states.\textsuperscript{[1,2,3,4,5]} These surface states are protected by time reversal symmetry, making them extremely important for various spintronic applications.\textsuperscript{[6,7,8,9]} Many works have demonstrated efficient charge to spin conversion in TI thin films with the aim of magnetization manipulation.\textsuperscript{[10,11,12,13,14]} However, the film growth still remains a major bottleneck since molecular beam epitaxy (MBE) is usually required to grow highly lattice ordered films on the appropriate substrate.\textsuperscript{[15,16,17]} Recently, it was demonstrated the presence of strong spin-orbit coupling (SOC) in sputtered polycrystalline Bi$_x$Se$_{1-x}$,\textsuperscript{[18,19,20]} allowing us to explore novel physics in disordered TI materials for developing industrial applications.

In most of TIs with highly ordered crystallographic structure, the spin current generation is a result of spin-dependent momentum scattering in the surface states.\textsuperscript{[21]} For highly disordered films, there are two major reasons to support the research motivation. Firstly, the scaling effects of the materials beyond the grain size of the films (SHE).\textsuperscript{[22,23]} Disordered films can exhibit interesting new physics like Mott/Variable range hopping which needs exploration.

Furthermore, effects of various dopants in Bismuth Selenide have also been extensively studied. One important advantage is that by embedding atoms with magnetic moment inside the alloys, one can create magnetic topological insulators with time reversal symmetry breaking to observe quantum anomalous Hall effect (QAHE).\textsuperscript{[24,25]} Introduction of such impurities is also interesting for the investigation of the spin-orbit torques (SOT) generated in such disordered materials when in contact with a thin ferromagnetic layer. Relevant figure of merits are used to characterize the SOTs, such as the (effective) spin Hall angle (SHA) when dealing with extrinsic (intrinsic) spin-Hall effect or inverse
Edelstein length ($\lambda_{\text{IEE}}$) for the inverse Edelstein effect (IEE)\cite{26} or its reciprocal quantity $q^{\text{ICS}}$ when dealing with Edelstein effect.\cite{27} Each of the following parameters, effective SHA and $q^{\text{ICS}}$, gives out the ability and efficiency to generate a spin current from an injected charge current in the spin Hall channel. In particular, the use of such spin Hall channels has been suggested for future magnetic random access memory (MRAM) structures that use SOT-assisted switching of the ferromagnetic storage layer.\cite{28,29}

In this work, we have synthesized Gd-alloyed Bi$_x$Se$_{1-x}$ (BSG) TI thin films by DC magnetron sputtering. Structural and chemical characterizations were performed using a high resolution transmission electron microscopy (HRTEM) combined with electron energy loss spectroscopy (EELS) and energy-dispersive spectroscopy (EDS), displaying a mainly amorphous feature in the disordered BSG film. An increase of roughness with the formation of Bi rich nano-crystallites is found in the 16nm thick film. BSG film is highly resistive and the temperature dependent conductivity of BSG film reveals variable range hopping (VRH) process as the dominant transport mechanism in the bulk BSG. Spin pumping transport measurements were performed on BSG($t$=6, 8, 12, 16 nm)/CoFeB(5 nm) bilayer structures in order to analyze the charge to spin SOT properties.\cite{30}

**Results and Discussion**

The multilayer stacks of MgO (2nm)/Bi(20%)-Se(40%)-Gd (40%) (BSG) /Co$_{20}$Fe$_{60}$B$_{20}$ (CFB) (5 nm)/MgO (2nm)/Ta (2nm) were grown by DC magnetron sputtering (with a base pressure 7×10$^{-8}$ torr) on thermally oxidized silicon substrates. Ta (5nm) capping layer was used for TEM characterization samples. Samples with different thickness of BSG ($t$ = 6, 8, 12, 16 nm) were prepared, which are labeled as BSG6, BSG8, BSG12 and BSG16, respectively. After the growth, the samples were processed into Hall bar pattern by UV lithography and devoted to the second harmonic transport measurement of the SHA. We have also prepared a bare BSG (16 nm)/MgO (5 nm) sample for measuring the resistivity at different temperatures. Please see more details in Methods section.

*Interfacial structure and chemical properties*
The full film samples of BSG8 and BSG16 were used for the interfacial structure and chemical characterization. **Figure. 1(a) and (b)** shows the large-scale scanning transmission electron microscopy–high angle annular dark field image (STEM-HAADF) and magnified HR-TEM image of the BSG8 sample, respectively. The sample shows a relatively flat and sharp interfaces in the large scale image (Figure. 1(a)). In Figure. 1(b), the BSG and CFB layers appear mainly amorphous in feature. The bottom MgO barrier exhibits textured features characterized by small nano-crystallites, while the top MgO layer appears mainly amorphous. The interface chemical distribution has been characterized by electron energy loss spectroscopy (EELS). Figure. 1(c) displays the different element maps drawn by EELS spectrum images on the region indicated by the red dashed rectangle zone marked in Figure. 1(a). Figure. 1(d) displays the element profile and each data point presents an average of element intensity in a zone with 10nm width and 0.5nm depth. From the chemical maps and profile, several pieces of information can be drawn. The distribution of Gd and Se are not homogenous in the BSG layer. Gd has a tendency to accumulate towards the bottom while, on the contrary, Se tends to segregate to the interface with CFB. Gd, Bi and Se over concentration spots are clearly visible. The mean atomic concentration of this area was evaluated as Bi 20%:Se 40%:Gd 40%. The ratio between Co and Fe in the CFB layer was estimated to be 1:4.

Figure. 2(a) shows the HRTEM image of the sample BSG16. It is found, here, that the interface roughness becomes important. By the guide of black dashed lines shown in Figure. 2(a), it is evident that the increase of interface roughness is mainly attributed to the thick BSG layer. CFB, MgO and Ta layers follow well the morphology of the BSG layer. Some nanocrystals inside the BSG layer can be clearly evidenced, as marked by the red dashed zones. EDS element mapping images are shown in Figure. 2(c-e) with the corresponding STEM-HAADF image in Figure. 2(b). From Figure. 2(b), many zones with brighter contrast can be observed inside the BSG layer, which is due to an inhomogeneous chemical distribution and points out the segregation of some elements. From the chemical maps, the bight contrast
zone (marked with red arrow) is identified to be Bi rich with less Se and Gd, which may also be correlated to the nanocrystals observed in Figure. 2(a). Therefore, during the growth of thicker BSG layer, the enhanced segregation of chemical elements forms the Bi-rich nanocrystals, which results in a pretty rough interface for the BSG16 sample.

**Temperature dependent transport properties**

Figure. 3(a) shows the temperature dependence of the resistivity for the BSG (16 nm) film covered with a MgO (5 nm) capping layer. The resistivity shows very strong temperature dependence and increases by a factor of 20 upon cooling from 300K (65mΩ·cm) to 80K (1.3Ω·cm). Note that the resistivity of the BSG film is found to be much larger than the reported values of amorphous Bi$_2$Se$_3$ film (3mΩ·cm at 300K)\cite{Error! Bookmark not defined.} and disordered Bi$_{0.1}$Sb$_{0.9}$ film (0.4mΩ·cm at 300K)\cite{23}. It is also higher than previous report on the polycrystalline Bi$_x$Se$_{1-x}$ film (1.3mΩ·cm at 300K).\cite{19} Since the coverage by the MgO layer could oxidize the BSG top surface and may kill the surface states,\cite{31} the measured resistivity should be mainly attributed, at this stage, to the bulk BSG resistivity.

We have compared the temperature dependent conductivity with an Arrhenius type dependence of variable range hopping (VRH) conduction.\cite{32} The VRH mechanism is usually dominant in disordered films with strong localization near the Fermi level. This results in electrons hopping from one site to another, which may usually be viewed as a compromise between tunneling and thermal jumps by phonon excitations.\cite{33} In VRH, the hopping length and hopping energy can vary from one site to another. The average value of the hopping energy may be deduced from the corresponding hopping length and density of states at Fermi level.

The relationship between the conductivity and temperature for VRH is given as follows:\cite{32}

$$\sigma = \sigma_0 \exp \left[ -\left(\frac{T_0}{T}\right)^p \right]$$  \hspace{1cm} (1)

Here $\sigma_0$ is the characteristic prefactor of VRH. The exponent $p$ gives information on the type of carrier conduction mechanism and may be divided into three different subsets. In a bulk material, $p=0.25$ is
attributed to a three-dimensional (3D) Mott VRH conduction wherein electron-electron (e-e) interactions are neglected. $p=0.33$ corresponds to a two-dimensional (2D) Mott VRH conduction in a 2D system. On the other hand, Efros-Shklovskii (ES) hopping mechanism includes long-range e-e interactions which results in $p=0.5$. $T_0$ is the characteristic Mott or ES temperature. Figure 3(b-d) display the resulting fits using ES-VRH, 2D-VRH and 3D-VRH, respectively. Rigorous data analyses, based on the residual sum of squares (RSS) for each fit, show a best fit matching with a Mott 3D VRH. This is a strong indication that a 3D Mott hopping takes place dominantly in our BSG film. The value of $\sigma_0$ obtained from the fit is $1.75 \times 10^{10} \Omega^{-1} \cdot \text{cm}^{-1}$. The value of $T_0$ obtained from the fit is $9.7 \times 10^5 \text{K}$, which is quite high, indicating that the amount of disorder in the film should be very high.$^{[33,35,36]}$ This could be due to the inhomogeneous chemical distribution as revealed by EELS element mapping.

The Mott characteristic temperature $T_0$ can be related to the localization length as $T_0 = \frac{\lambda \alpha^3}{k_B N(E_F)}$ where $\alpha$ is the inverse localization length. $N(E_F)$ is the 3D density of states for the bulk material at the Fermi level, which is approximately in the range of $10^{21} \text{eV}^{-1} \cdot \text{cm}^{-1}$. $\lambda$ is a dimensionless constant with a value of 18.1.$^{[33]}$ By injecting the fitted value of $T_0$ ($9.7 \times 10^5 \text{K}$) into the formula, we obtain a typical localization length of $\alpha^{-1} = 1.36 \text{ nm}$. The hopping distance ($R_{\text{hop}}$) can be calculated from the localization length in the Mott’s case as $R_{\text{hop}} = 0.4 \left( \frac{T_0}{T} \right)^{0.25} \alpha$. To obtain respectively $R_{\text{hop}} \approx 5.6 \text{ nm}$ ($T=80 \text{K}$) and $R_{\text{hop}} \approx 4.1 \text{ nm}$ ($T=300 \text{K}$).

In addition to the temperature dependent resistance analyses, we have also measured the magnetoresistance of another 30 nm thick BSG sample (covered with 5 nm thick MgO) (see SI note 1). These measurements were done at different temperature (100-300K) by sweeping the in-plane and out-of-plane field between $\pm 5 \text{ T}$. The magnetoresistances for both in-plane and out-of-plane field configurations show a characteristic parabolic shape. The absence of sharp dip feature linked to the weak anti-localization confirms the strong 3D Mott hopping in the bulk BSG.$^{[\text{Error! Bookmark not defined.}]}$
Spin pumping characterization

We performed spin pumping measurements to characterize spin-to-charge conversion (ISHE) in CFB(5nm)/BSG(t = 6,8,12,16 nm). In spin pumping, an AC magnetic field is applied to excite the FM layer in the presence of a DC magnetic field sweep. For each AC frequency there is a specific DC field where the precession frequency of the magnetization matches with the AC frequency. This point is called the ferromagnetic resonance (FMR). At FMR, the spins from the FM layer are highly excited and pumped into the BSG layer. The spin current is converted into charge current by the virtue of ISHE which can then be measured by a DC voltage generated across the device. Fig. 4(a) shows the schematic of spin pumping measurement.

The spin current injected from the FM layer into the spin Hall channel [40].

\[ J_s = \frac{G\gamma^2\hbar^2}{8\pi\alpha^2\sqrt{(4\pi M_S\gamma)^2 + (2\omega)^2}} \left\{ 4\pi M_S \gamma + \sqrt{(4\pi M_S\gamma)^2 + (2\omega)^2} \right\} \]

(2)

Here \( \gamma \) is the gyromagnetic ratio, \( \alpha \) is damping constant, \( M_S \) is saturation magnetization, \( \omega \) is angular frequency at FMR, \( G \) is real part of the spin-mixing conductance, This is essentially the key quantity that one derives from the spin pumping experimental results. The other one being the charge current density which is created from the device as a result of spin-to-charge conversion. This can be very simply derived from Ohm’s law:

\[ J_c = IA = \frac{V}{RA} \]

(3)

This is then combined with the spin current density derived from the spin pumping results to obtain spin-to-charge conversion efficiency: SCE (~ \( J_c/J_s \)).

The oscillations from AC and DC component will reach a resonance, given by Kittel formula. Under these circumstances, we get the maximum excitation of the electrons in the CFB layer which are then injected into the FM layer. The key output is the DC voltage measured across the device created due to
ISHE. Fig. 4 shows the experimental results of spin pumping on BSG6 sample with an AC excitation voltage of 2V (19.03 dBm).

As we can see from fig 4(b), for a given excitation frequency, there is a specific DC field, where the output voltage is maximized. The point of maxima corresponds to the resonance field for that given frequency. We can then extract this resonance field by doing a combination of symmetric and anti-symmetric Lorentzian fit into these peaks. The fit equation is given by [40]:

\[
V = \frac{V_S \Delta H^2}{\Delta H^2 + (H_{\text{ext}} - H_0)^2} + \frac{V_A (H_{\text{ext}} - H_0)}{\Delta H (\Delta H^2 + (H_{\text{ext}} - H_0)^2)}
\]  

(4)

\(V_S\) is symmetric Voltage, \(V_A\) is anti-symmetric Voltage, \(\Delta H\) is linewidth, \(H_0\) is Resonance field, \(H_{\text{ext}}\) is external DC field. Upon doing the Lorentzian fits, we get the symmetric and anti-symmetric voltage, which corresponds to the output DC voltage. And, we also get the linewidth and the resonance field, which will allow us to calculate the spin current injected into the BSG layer. The key difference between the symmetric and anti-symmetric voltage is that the symmetric voltage is maximized at resonance whereas the anti-symmetric voltage reaches zero at resonance. Fig. 4(d) shows the spin pumping voltage obtained for different excitation frequencies. We can see an increase in the voltage with decreasing frequency. This is a well-known behavior in spin pumping, attributed to the compensation between frequency changes during spin pumping and the dynamic magnetic susceptibility. The anti-symmetric component of the spin pumping voltage mostly corresponds to anisotropic magnetoresistance (AMR) and Anomalous Hall effect (AHE). In a pure FM layer, only the anti-symmetric component will dominate.

Once, we know the ISHE voltage, we can simply calculate the charge current density, from the resistance of the device. The key part is to now derive the spin current density injected into the BSG layer. We use the frequency dependence of resonance field and the linewidth to derive the saturation magnetization and the damping constant (\(\alpha\)) of the FM layer respectively. Fig.4 (e,f) shows this dependence. The dependence of resonance field with frequency is fitted with Kittel formula. This is used
to derive the saturation magnetization of the FM layer. The dependence of linewidth with frequency is fitted by a linear curve, given as [40]:

\[ \Delta H = \Delta_0 + \frac{4\pi}{\sqrt{3}Y} \alpha f \]  

(5)

Fig. (5 a,b) shows the obtained values of saturation magnetization and damping constant for different thicknesses of the BSG layer. The values of saturation magnetization and damping constant are then combined to obtain the spin mixing conductance.

\[ G = \frac{4\pi M_S t_{FM}}{g \mu_B} (\alpha - \alpha_0) \]  

(6)

Once, we have the spin mixing conductance, we have all the required values to calculate the spin current density injected. The spin current density is then combined with charge current density to obtain the spin-to-charge conversion efficiency (SCE).

Fig. 5(d) shows the final SCE obtained for different thicknesses of the BSG layer at 9 GHz excitation frequency. As we can see, the SCE shows a steady increase with decreasing thickness. We also performed spin pumping measurements on a single FM layer.

Discussion

We start by looking at possible artifacts that can contribute to the spin pumping signal. One concern is the self spin pumping of the CoFeB layer. The self spin pumping of CoFeB will create four major trends: 1) It will create a significant anti-symmetric signal due to AMR [44] 2) The damping constant will be very similar to the intrinsic damping constant of single layer CFB 3) The spin mixing conductance will be negligible. 4) There will be no major BSG thickness dependence. We don’t observe any of the above things. We observe good symmetric signals in our samples. The damping constant of the samples are larger than the intrinsic damping constant and the spin mixing conductance is reliably positive. Furthermore, we also observe a strong BSG thickness dependence. All these results show that the self spin pumping of CFB cannot explain our spin pumping signal.
Now, we look at possible thermal origins. There are several thermal effects that can come into play. The origins of the thermal effects arise from the temperature gradient created across the thickness of the ferromagnet and/or spin hall channel. However, the thermal effects that do contribute to the spin pumping signal can be of 3 major kinds: 1) Spin Seebeck effect (SSE) 2) Nernst effect (NE) 3) Anomalous Nernst effect (ANE). Thermal gradients are created due to Joule heating and increase quadratically with current. We can write the voltage-current relationship as:

\[ V = IR_1 + I^2R_2 \] (7)

The first term is the traditional Ohm’s law and the second term shows a quadratic current relation, which is the source of thermal effects. To characterize these thermal effects, we did second harmonic measurements with an AC current injection of 1 mA (channel width \( \sim 10 \mu m \)) RMS, which is much greater than the current density we obtain in spin pumping (few \( \mu As \), channel width \( \sim 620 \mu m \)) and measure the first (linear) and second (quadratic) harmonic part of the voltage (Supplementary S4). The first harmonic voltage will come from the AMR of the FM which is connected directly to the anti-symmetric voltage of spin pumping and the second harmonic is the thermal voltages. If the thermal effects dominate due to Joule heating during spin pumping, we should see a large and dominant quadratic current term [42, 43].

SSE is the simplest and causes a spin current to arise under a thermal gradient. SSE doesn’t change sign like ISHE does and causes a simple voltage shift. NE and ANE are two major thermal artifacts which follow the same behavior as ISHE during spin pumping and hence are causes of concern. The NE is created when we have a temperature gradient perpendicular to the magnetic field. One of the major signatures of NE is the linear dependence of second harmonic voltage on the magnetic field. Fortunately, we don’t observe any such dependence on the magnetic field. Anomalous Nernst effect (ANE) is the generation of a non-zero voltage across the dimensions of a ferromagnet under a thermal gradient. Anomalous Nernst effect (ANE) will create a voltage proportional to the cross product between
the magnetization and the thermal gradient: $V_{ANE} \sim \Delta T \times M$. Typically, the second harmonic signals for bilayers with resistive spin Hall channels will be devoid of any spin-orbit torque (due to dominant thermal contributions and current shunting) leaving us with almost pure thermal effects ($NE + ANE + SSE$). The key thing to look at is which harmonic (first or second) dominates the voltage. Fig. S4.5 and S4-3 shows that the first harmonic voltage (AMR, PHE) is much higher than the second harmonic (thermal). Furthermore, the thermal voltage doesn’t show any appreciable thickness dependence unlike spin-pumping, which depends very heavily on the thickness of Gd-BiSe layer. This shows that the temperature gradients don’t vary too much with Gd-BiSe thickness. The spin pumping voltage increases by almost an order of magnitude at lower thicknesses. These differences hint to the fact that the effects of these thermal voltages are not a major contributor to the measured DC spin pumping voltage.

These observations point to the fact that the source of the signal should be mostly ISHE. We now attempt to explain the possible physics/mechanisms behind the ISHE. The increase in SCE with decreasing thickness is expected. This comes from the fact that spin pumping is inherently an interfacial effect which dominates at lower thicknesses. However, the ideal drift-diffusion model doesn’t give a good fit for our case. This means that there might be other factors that might be affecting the thickness dependence of SCE. Since our material is amorphous, devoid of any interesting band structure or density of states, the possible mechanisms for ISHE must be from extrinsic effects. The presence of disorder in the film could be a strong scattering source, which would explain the presence of ISHE in the film. The lower thickness shows a higher SCE, which corresponds to a more disordered film as evident from the TEM characterization of the films. This would increase the extrinsic ISHE for lower thicknesses. Furthermore, the TEM suggests that the interface for 16 nm film is rougher than 8 nm film. Rough interfaces can cause higher spin loss at the interface and thereby decrease the spin injection efficiency. This can result in a lower SCE for higher thickness.
The highest SCE, we observe is around 0.01 for the 6 nm film. As mentioned in section 1.2, we are unable to attain the spin hall angle since the spin diffusion length is unknown for this material. However, we expect the spin diffusion length to be small in this material due to two major reason: 1) The material is highly disordered (amorphous) 2) The material is resistive. Both these reasons can contribute to lowering of the spin diffusion length in the film.

**Conclusions**

In conclusion, we have synthesized Gd alloyed BiSe thin films and performed a detailed structural and magneto-transport characterization of those BSG films. The TEM characterization reveals the BSG film has a mostly amorphous feature. The inhomogeneous of chemical distribution is enhanced when BSG thickness increases, resulting in the appearance of Bi-rich nanocrystals inside BSG layer and the increase of BSG surface roughness. Temperature dependent resistivity measurements indicate a dominant 3D VRH transport mechanism in the BSG film from 80 to 300K. This hopping transport and the high resistivity of BSG film could be a consequence of extra disorder in the Gd alloyed BiSe films compared to the pure BiSe films. Spin pumping measurements were done to characterize spin-to-charge conversion in the film. The highest spin-to-charge conversion efficiency is 0.01 obtained for 6 nm BSG film. The thermal effects were characterized and removed using the second harmonic method.
Methods

Sample preparation:

The films were grown by magnetron sputtering on thermally oxidized silicon substrate. The stacks are: MgO (2 nm)/ BSG (t nm)/ CFB (5 nm)/ MgO (2 nm)/ Ta (2 nm). BSG was grown by co-sputtering. The Ar flow during sputtering was 40 sccm and anode bias was 60 V. Cathode power for BiSe was 30 W and for Gd was 40 W respectively. The deposition rate was 0.7 Å/s. These films were then patterned into Hall bars by standard photolithography process followed by Ar ion milling. Second step of photolithography involved exposing the contact areas and followed by metal contact deposition of Ti (10 nm) and Au (120 nm) by using e-beam evaporation.

TEM characterization:

HRTEM and STEM were performed to characterize the interfacial structure using a probe corrected JEOL ARM 200 CF operated at 200kV. Thin lamella was extracted by focused ion beam (FIB) milling using an FEI Helios Nanolab dual beam. EELS spectrum images (SI) were recorded in STEM mode with a Gatan Quantum Imaging filter. In order to correct energy drift and estimate the local thickness, the zero-loss and the core-loss spectra were simultaneously recorded for a dispersion of 1eV (Dual EELS method). The core loss spectra were registered in the range 670-2700 eV in order to record FeL, CoL, GdM, MgK, SeL, TaM and BiM edges. BK and OK signals with edges respectively near 188 eV and 532eV, were not recorded. The pixel size of the SI was fixed at 0.4 nm for a dwell time of 0.5 s/pixel for the core loss spectra. After energy drift correction, the SI were denoised using a principal component analysis method before quantitative analysis. EDS spectrum images were recorded in STEM mode with a JEOL JED2300T silicon-drift detector. SI were obtained by integrating 170 frames of 256×256 pixels The pixel size was fixed at 0.23 nm for a dwell time of 0.2 msec.
Transport measurement:

For spin pumping measurement, the samples were patterned into stripes, with a width and length of 620 μm and 1500 μm, respectively, using ultra-violet photolithography and ion milling. On top of that, we deposited a 55 nm thick silicon dioxide layer in order to insulate the wave guide from the film. Following that, the contact pads and waveguides were patterned using ultra-violet photolithography. The contact layer was deposited using e-beam evaporation of 10 nm of Titanium layer followed by 150 nm of Gold layer. The wave guide of the spin pumping devices consists of a signal linewidth of width 75 μm, a ground linewidth of width 225 μm, and separation between the lines of 37.5 μm. Temperature dependent resistance were done with Keithley 6221 as current source and nanovoltmeter to probe the DC voltage. These DC transport measurements were carried out on Quantum design PPMS 3000 which provides good temperature control, external field and a rotating stage.
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Figure 1: Structural and chemical characterization of BSG8 sample by TEM. (a) STEM HAADF image on the BSG8 sample at middle magnification. The red rectangle shows the region where the EELS spectrum images were recorded. (b) HRTEM image in magnified scale on the BSG8 sample. (c) Elemental maps of the stacks drawn from EELS spectrum images after quantitative processing with all the essential elements from the substrate to the capping layer. The tiny silicon signal visible in the Ta capping layer and the Mg signal inside BSG layer are due to an artefact of processing because of the overlapping of different element edge peaks. (d) Elemental profiles across the multilayer structure drawn from EELS maps.
Figure 2: Structural and chemical characterization of BSG16 sample by TEM. (a) HRTEM image in magnified scale on the BSG16 sample. The black dashed lines guide the eyes to show the interface roughness due to the BSG layer. The red cycles show the zone where we can find nanocrystals. (b) STEM HAADF image and corresponding EDS elemental maps for (c) Bi, (d) Se and (e) Gd. The red arrows indicate that the white contrast zone is due to the inhomogeneous element distribution with a Bi-rich character.
Figure 3: Temperature dependent transport properties of a bare 16nm BSG sample covered by MgO protection layer. (a) Temperature dependent resistivity. (b-d) Temperature vs log(conductivity) with various fits with known theoretical models: (b) ES VRH, (c) 2D VRH and (d) 3D VRH, respectively.
Figure 4: (a) Schematic of spin pumping measurement (b) DC voltage measured as a function of DC magnetic field for different frequencies of AC magnetic field. (c) Lorentzian fit of the 9 GHz peak. The red curve is the symmetric component and the blue curve is the anti-symmetric component. (d,e) symmetric and anti-symmetric voltage vs excitation frequency for different thicknesses. (f) Dependence of resonance field on the excitation frequency (g) Dependence of linewidth on excitation frequency.
Figure 5: Saturation magnetization (a), damping constant (b), spin mixing conductance (c), Spin-to-charge conversion efficiency (d) for different thicknesses of BSG layer
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Supplementary Information

For

Room Temperature Mott Hopping and Second Harmonic Characterization of Amorphous Gd-alloyed Bi$_2$Se$_3$

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S1. Magnetoresistance measurements

We have performed magnetoresistance measurements on a 30 nm thick BSG sample. These measurements were done at different temperatures (100K-300K) by sweeping the in-plane and out-of-plane field between ±5T. The magnetoresistance for both in-plane and out-of-plane field shows parabolic magnetoresistance. Fig. S1(a,b) show the magnetoresistance measurements at 100 K. The absence of sharp dip feature linked to the weak anti-localization confirms the strong 3D Mott hopping in the bulk BSG.

**Fig. S1:** Measurement of magnetoresistance at 100K on the bare 30 nm BSG sample with (a) in-plane and (b) out-of-plane magnetic field.

S2. Magnetic characterization of BSG film

We have performed magnetic characterizations by SQUID for the bare 16 nm BSG sample at different temperature, as shown in Fig. S2. The curves show only linear variation behavior due to the paramagnetic character of Si/SiO₂ substrate. The signal from BSG layer can be extracted after subtracting the linear background. However, we found the signal from BSG is very small, which indicates that the BSG layer is almost non-magnetic although the Gd atom is magnetic.
S3. Current shunting model

We have applied the current shunting model to calculate the current flowing through the BSG layer, as schematically shown in Fig. S3 for the room temperature condition.

\[ I_2 = \frac{I_1 \cdot R_1}{R_1 + R_2} = \frac{I_1 \cdot \rho_{\text{CFB}}}{t_{\text{CFB}}} \cdot \frac{t_{\text{BSG}} + \rho_{\text{CFB}}}{\rho_{\text{CFB}}} \]

(S1)

Fig. S3: Schematic of the current shunting model to calculate the current flowing through the BSG layer.

S4. Thermal effects

We now discuss the thermal properties of our CoFeB(5nm)/BSG bilayer structures acquired by the second-harmonic Hall technique [1, 2]. Typically, the current voltage relationship can be extended to higher order terms: \( V = IR_1 + I^2R_2 \). The linear term is the simple Ohm’s law, the quadratic term is the
Second order term that usually encompasses all the thermal effects. Separating the linear term from the quadratic term is done by injecting an AC current of frequency $\omega$ and then measuring its first harmonic (frequency = $\omega$) and second harmonic voltage (frequency = $2\omega$). The first harmonic signal will give us an estimate of linear term and the second harmonic voltage will give us an estimate of the quadratic term and how they compare.

Second harmonic method also allows us to calculate the charge-to-spin conversion and systematically separate out any thermal effects like ANE, SSE etc. However, with resistive spin hall channels, second harmonic doesn’t give us a good value due to the dominance of the thermal terms to the second harmonic signal and the current shunting through the FM layer. This is one of the reason why spin pumping is preferred for characterizing resistive spin hall channels. Hence, in these cases, the second harmonic signal is almost exclusively dominated by thermal effects and devoid of any SOT effects [3]. These thermal effects are created due to a temperature gradient across the thickness of the FM and spin Hall channel due to Joule heating. This temperature gradient is a consequence of Joule heating and increases with the magnitude of injected current. Therefore, second harmonic voltage gives us a ballpark of what kind of signal levels we should expect emanating from these thermal effects and whether they should interfere with our spin pumping signals. The second harmonic voltages are then compared with the first harmonic voltages to see which term dominates the voltage.

**Fig.: S4.1: The thermal gradient generated across the FM layer**

To this end, the multilayer thin films were patterned into cross Hall bar structure with dimensions ~ 10 µm wide and 30-50 µm long, as schematically shown in Figure.S4.2(a). Since the
magnetization for the BSG layer is negligible, the measured anomalous Hall resistance should mainly be attributed to the CoFeB layer. From the shape of the curve, we can conclude an in-plane magnetic anisotropy for the CoFeB layer with an out-of-plane saturation field of about 1.5T. Figure. S4.2(a) shows the schematic of the second harmonic measurement setup. For this system, the current flowing through the spin Hall channel can produce two different torque components: a field-like torque ($\vec{m} \times \vec{\sigma}$) and a damping-like ($\vec{m} \times (\vec{\sigma} \times \vec{m})$) torque. Both components depend on the relative orientation between the injected spins and the local magnetization. The respective effective fields giving rise to the Hall voltage ($V_{xy}^{2\omega}$) can be expressed by the second harmonic Hall response model [1,3]

$$V_{xy}^{2\omega} = \left(\frac{H_{FL}+H_{oe}}{H_{A}-H_{ext}} R_p \cos 2\theta \cos \theta + \frac{1}{2} \frac{H_{DL}}{H_{K}-H_{ext}} R_A \cos \theta + V_t \cos \theta + V_{NE} H_{ext} \cos \theta \right) I$$

(S4.1),

$$V_{DL} = \left(\frac{1}{2} \frac{H_{DL}}{H_{K}-H_{ext}} R_A + V_t + V_{NE} H_{ext} \right) I \cos \theta$$

(S4.2),

$$V_{FL} = \frac{H_{FL}+H_{oe}}{H_{A}-H_{ext}} R_p \cos 2\theta \cos \theta$$

(S4.3),

where $H_A$ and $H_K$ are the out-of-plane and in-plane anisotropy, respectively. $H_{FL}$ and $H_{DL}$ are the effective fields due to the field-like and damping-like torque, respectively. $H_{oe}$ is the Oersted field generated from the current in the channel and $H_{ext}$ is the external applied magnetic field. $R_p$ and $R_A$ are the planar Hall and anomalous Hall coefficients, respectively. $V_t$ is the thermal voltage (ANE and SSE) generated by the perpendicular temperature gradient created across the ferromagnetic layer. $V_{NE}$ is the voltage created due to Nernst effect, which will be linear to the external field. $\theta$ is the angle between the current and the in-plane field and $I$ is the current in the spin Hall channel (Figure. S4.2 (a)). $V_{DL}$ is defined as the part of the second harmonic voltage that shows $\cos(\theta)$ dependence. This voltage is a combination of the damping-like torque and thermal voltage. The injected RMS value of the current is 1 mA with a frequency of 33 Hz. By measuring the second harmonic Hall resistance ($R_{xy}^{2\omega} = V_{xy}^{2\omega} / I$) and by varying the angle between the in-plane field and the channel at different
external fields, we can separate the different contributions of the field-like, damping-like and thermal voltages from their specific angular signatures.

Figure. S4.2(b) shows the second order Hall signal at 3T external field at RT. As it can be seen, the data shows a pretty fair cosine shape that can be fitted to obtain the amplitude. Indeed, the field-like torque often manifests as a small depression in the second harmonic Hall voltage around 90 and 270 degrees. The fitted amplitudes are then plotted against \( \frac{1}{H_{\text{ext}} - H_K} \) and reported in Figure. S4.2(d), further processed by a linear fitting. We notice that there is no second harmonic voltage that is directly linear to the magnetic field indicating that the NE voltage is negligible. Hence, in order to obtain better fits and lower the number of variables, we ignore this NE effect from equation S4.1. The slope of the linear fit allows us to determine the damping-like/field-like field and the intercept gives out the thermal voltage contribution. The damping-like/field-like field is related to the spin current generated from the spin Hall channel: \( H_{\text{DL/FL}} = \frac{h J_S}{2 e M_S t_{\text{FM}}} \) [1]. Here \( J_S \) is the spin current generated by the BSG layer. Using this relationship, we can calculate, in a model of a 3D (bulk) conduction for SHE (\( \zeta \)), the equivalent spin Hall angle which is defined as the ratio of the spin current generated from spin Hall channel over the charge current injected according to [1]:

\[
\zeta = \frac{J_S}{I_c} = \frac{2 e M_S t_{\text{FM}} H_{\text{DL/FL}}}{h J_{\text{SH}}} \quad \text{(S4.4)},
\]

where \( M_S \) is the saturation magnetization of CoFeB layer and \( J_{\text{SH}} \) is the current density through the spin Hall channel calculated by the current shunting method (see SI note 3). We have performed the angular dependent second order Hall measurement on the BSG/CFB bilayers with different thickness of BSG (6, 8, 12, 16nm). The effective SHAs are deduced from the obtained damping-like and field-like torque \( H_{\text{DL/FL}} \) and injecting into the Equation. (S4.4). Fig. S4.2 (f) shows the field sweep of the second harmonic Hall signal under transverse in-plane magnetic field. We can see a jump which corresponds to the magnetization switching of the in-plane CoFeB layer. The low point of this voltage
is the same as the voltage at $\theta \sim 0^\circ$ for sample rotation and the high point is the same as $\theta \sim 180^\circ$. As we can see, we don’t observe any linear magnetic field dependence of the second harmonic voltage during field sweep. This points to the fact that NE is quite small in the device and can be ignored [53].

Fig. S4.2: Magneto-transport characterization of spin-orbit torque. (a) Schematics of cross Hall bar device and second harmonic transport measurement setup. (b) Second harmonic Hall resistance as a function of angle for BSG8 at room temperature and external 3T magnetic field. (c) Amplitude of second harmonic field-like signal as a function of $\frac{1}{H_{\text{ext}} - H_k}$. (d) Amplitude of second harmonic damping-like signal as a function of $\frac{1}{H_{\text{ext}} - H_k}$. The linear fitting allows to obtain the amplitude of damping like torque. (e) Spin Hall efficiencies as a function of thickness of BSG layer. (f) Second harmonic transverse magnetoresistance response with magnetic field at room temperature.

Firstly, the SOT contribution in the device is quite negligible. One of the reason this happens is because the thermal effects are quite significant in this experimental structure since we are injecting a charge current of 1 mA into a channel width of ~ 10 µm. Typically, the thermal effects are created via Joule heating and scale parabolically with the current. The second reason is that, the charge current is mostly shunted through the ferromagnetic layer which is more conductive than the spin hall channel. This makes the SOT contribution quite negligible. This is seen in fig. S4.2 (e). This means that almost
all the measured voltage in the second harmonic setup is the consequence of thermal effects like SSE and ANE. As one can see from fig. S4.3, the thermal voltage (SSE and ANE) is around 1 µV.

The thermal voltages also don’t show any proper thickness dependence unlike our spin pumping results which increases quite heavily with decreasing thickness. This shows that temperature gradients don’t vary too much with the thickness of Gd-BiSe, which means that Gd-BiSe layer doesn’t contribute a lot to Joule heating. This is expected, since due to resistivity difference most of the current will be confined to the FM layer. And hence FM layer is the only one that contributes to Joule heating. Using parallel-resistor model, with constant total injected current \( \sim I \), Joule heating created due to Gd-BiSe layer is:

\[
I_{BSG}^2 R_{BSG} \sim I^2 R_{CFB}^2 / R_{BSG}
\]

Similarly, the Joule heating for CFB is:

\[
I_{CFB}^2 R_{CFB} \sim I^2 R_{CFB}
\]

Here we make the assumption that \( R_{BSG} \ll R_{CFB} \). This shows that the Joule heating from CFB layer is dominant and the BSG layer doesn’t play a major role hence showing the thickness independence. Next step is to look at the corresponding first harmonic voltage and compare to see which one the more dominant term (first or

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**Fig. S4.3:** Thermal voltage obtained via second harmonic at 1mA injected current.
second harmonic). Fig. S4.4 shows the first harmonic voltage of the same device and conditions as in fig. S4.2.

\[ \Delta V_{xy}^{1\omega} = \frac{\text{max}(V_{xy}^{1\omega}) - \text{min}(V_{xy}^{2\omega})}{2} \]  

(S4.4)

**Fig. S4.4: First harmonic resistance of the same device and conditions as fig. S4-2**

As one can observe, the thermal effects (ONE, ANE, SSE etc.) are absent in the first harmonic signal, as it doesn’t show any of their characteristic angle dependence. The first harmonic voltage is typically a consequence of the planar hall effect (PHE) present in the bilayer. This signal is very sensitive to the sample orientation and tilt and will usually vary due to human error caused during sample mounting. That’s why it can be difficult to obtain a perfect sinusoidal-type signal from the first harmonic. Nevertheless, the difference between the highest and lowest first harmonic voltage will give us a good sign of how the first and second harmonic voltages compare.
Fig. S4.5 shows the difference between maximum and minimum first harmonic voltage for different thicknesses. As one can see, it is much larger than the obtained thermal second harmonic voltage (1 µV) at 1 mA RMS current. This basically tells us that the first harmonic signal dominates the DC voltage. In spin pumping, the current density is much lower than this (few µAs, channel width ~ 620 µm). That’s why it is reasonable to assume that the effects of thermal voltage in the measured DC voltage should be quite negligible.

Fig. S4.5: The difference in maximum and minimum first harmonic voltage for different thicknesses of BSG

So far, we have looked at the Hall signals. We also look at the longitudinal first and second harmonic since that would be a better comparison with spin pumping signal. Fig. S4.6 shows the first and second harmonic longitudinal signal. The first harmonic is the AMR signal and is much larger than second harmonic (thermal effects). Here, first the harmonic ($V_{xx}^{1\omega}$) is essentially what we measure from the anti-
symmetric part of the spin pumping and overwhelms the thermal part. The absence of thickness
dependence in thermal voltage and dominance of first harmonic, indicates that the spin pumping should
be devoid of many of these thermal effects.

Fig. S4.6: First and second harmonic longitudinal signals for the same device under same conditions as
fig. S4-2

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