Spin Hall effect in topological insulators

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The giant spin Hall effect (SHE) in topological insulators (TIs) is very attractive for applications to various spintronic devices, notably spin-orbit torque magnetoresistive random-access memory (SOT-MRAM). In this paper, we review the recent progress on the giant SHE in TIs, with emphasis on the role of topological surface states. We discuss current challenges and future prospects for TIs as a realistic material in SOT-MRAM.

Key words: spin Hall effect, topological insulator, spin-orbit torque, MRAM

1. Introduction

The spin Hall effect (SHE)\textsuperscript{1, 2} in non-magnetic materials with strong spin-orbit coupling has recently attracted great interest for its possible applications to various spintronic devices, most notably spin-orbit torque (SOT) magnetoresistive random-access memory (MRAM). In the SHE, a perpendicular pure spin current density $J_S$ can be generated by an in-plane charge current density $J_e$ in a non-magnetic layer, whose charge-to-spin conversion efficiency is characterized by the spin Hall angle $\theta_{\text{SH}} = (2e/\hbar)(L/t)J_s/J_e$. This pure spin current can be injected into the free magnetic layer of a magnetic tunnel junction (MTJ), and it consequently exerts a spin torque on the layer for magnetization switching. The relationship between the injected spin current $I_s$ and the charge current $I_e$ in the non-magnetic layer is then given by $I_s = (2e\hbar/\theta_{\text{SH}})I_e$, where $L$ is the length of the magnetic layer, and $t$ the thickness of the non-magnetic layer. Because the factor $(L/t)\theta_{\text{SH}}$ can be much larger than unity, i.e. $I_s/(\theta_{\text{SH}}) >> I_e/e$, the SHE can be used as an effective way of generating a spin current, meaning that a small writing current can be used for magnetization switching. Furthermore, since there is no charge current flowing into the magnetic layer, damage to MTJs can be suppressed. Finally, since the spin-polarization direction of the pure spin current generated by the SHE is perpendicular to the magnetization direction of the free magnetic layer with perpendicular magnetic anisotropy (PMA), the anti-damping-like spin torque is maximized and the magnetization can be switched very fast ($< \text{ns}$).\textsuperscript{3}

Motivated by these prospects, there have been intensive efforts put into the research and development of SOT-MRAM. In particular, very fast switching with low voltage and no in-plane bias field has been demonstrated in SOT-MRAM arrays deposited on 300 mm Si wafers, underlying the feasibility of SOT-MRAM.\textsuperscript{4}

Despite these recent developments, the writing current density in SOT-MRAM is still as large as $10^8$ A/cm², which is too high for implementing SOT-MRAM with Si driving transistors. This is because the heavy metals used for the spin Hall layer in SOT-MRAM, such as Ta, Pt or W, have a limited spin Hall angle. The maximum $\theta_{\text{SH}}$, ~0.4, is obtained with W.\textsuperscript{5} Even if taking into account the $L/t$ factor, the driving current in SOT-MRAM is still one order of magnitude larger than that of the conventional spin transfer torque MRAM. To solve this bottle neck, it is essential to find new materials with spin Hall angles at least one order of magnitude larger than that of heavy metals, which requires $\theta_{\text{SH}} > 1$. Since the extrinsic mechanism of SHE, i.e. side-jump or skew scattering, unlikely yields such a giant $\theta_{\text{SH}}$, we can rely only on its intrinsic mechanism,\textsuperscript{6} which originates from Berry-phase curvature in the momentum space and is an intrinsic quantum mechanical property of the band structure.\textsuperscript{7}

Because Berry-phase curvature has a singularity at Dirac points, we can expect a very large $\theta_{\text{SH}}$ in materials with Dirac-like dispersion. Topological insulators (TIs) are promising since they have metallic surface states with Dirac-point-like dispersion. In fact, very large $\theta_{\text{SH}}$, usually higher than 1, have been routinely observed in many TIs, which is a breakthrough for SOT-MRAM. In this paper, we will briefly introduce some fundamental properties of TIs. Then, we will review recent studies on the giant SHE in TIs, with emphasis on the role of topological surface states. Finally, we will discuss the current challenges and future prospects for TIs as a realistic material in SOT-MRAM.

2. Brief history and fundamental properties of topological insulators

TIs are quantum materials having insulating bulk states and metallic surface states with spin-momentum locking, as shown in Fig. 1. TIs have had roots in research on the quantum Hall effect since the early 1980s. When two-dimensional electron gas at the interface of a semiconductor heterostructure is subjected to low temperatures and strong magnetic fields, its energies are quantized to multiple Landau levels. If the Fermi level lies in between those Landau levels, the charge conduction inside the sample is suppressed, and the inside of the sample becomes insulating with only one-dimensional conduction at the...
edges of the sample. Thus, electron gas undergoing the quantum Hall effect can be considered as a two-dimensional TI with one-dimensional edge states. It was later found that the quantum Hall effect originates from the non-trivial topology of the two-dimensional electron wavefunctions under strong magnetic fields. This further led to the prediction of a novel quantum Hall effect without Landau levels (quantum anomalous Hall effect) based purely on topology arguments by Haldane in 1988. Then, in 2006, a two-dimensional TI with spin-polarized edge states was predicted in a heterostructure consisting of a HgTe quantum well sandwiched by two (HgCd)Te barriers. For this heterostructure, spin-polarized edge states were predicted even without a magnetic field (quantum spin Hall effect), and immediately confirmed. These theoretical and experimental achievements opened the dawn of TI research. Theories on quantum spin Hall states were further studied for the two-dimensional surface states of three-dimensional TI materials, and BiSb (Sb composition of 72%) was proposed as a three-dimensional TI. The topological surface states of BiSb and their spin-momentum locking were confirmed using angle-resolved photoemission spectroscopy, making it the first experimentally confirmed three-dimensional TI. This sparked a gold rush of both theoretical and experimental searches for new three-dimensional TI materials. Among them, Bi-based chalcogenides, such as Bi2Se3, Bi2Te3, and (BiSb)2Te3, are the most studied for their electronic properties and possible device applications because they have a large band gap and a single surface state with simple Dirac-like band dispersion and because their thin films can be easily grown on various substrates using the molecular beam epitaxy (MBE) technique. Notably, Cr-doped magnetic TI (CrBiSb)2Te3 with PMA was synthesized, in which the quantum anomalous Hall effect was finally confirmed.

From this brief history, we can see that TIs inherit many properties of quantum Hall states and thereby have been the object of many studies on low-temperature physics such as the quantum spin Hall effect or quantum anomalous Hall effect. At the same time, TIs have many different characteristics from quantum Hall states that make them attractive for room-temperature spintronic applications. One feature is that the existence of one (two)-dimensional edge(surface) states of TIs is ensured by the non-trivial topology of their band structure and can emerge without the application of large external magnetic fields. If the band gap energy is much larger than the thermal energy at room temperature and the Fermi level is inside the band gap, the edge(surface) states can be electrically accessed even at room temperature. Another is that the surface states have Dirac-like band dispersion, which promises a large intrinsic SHE. Yet another feature is the unique spin-momentum locking feature of the surface states, which prioritizes pure spin current generation in the direction perpendicular to the film plane. All of these features are very promising for applications to spintronics, especially their expected giant SHE.

3. Spin Hall effect in Bi2Se3

Bi2Se3 is a TI with a large band gap of about 300 meV and has thus become one of the most studied TIs. As a result, the SHE in TIs was first evaluated in Bi2Se3. A.R. Mirlinik et al. cleaned the surface of a MBE-grown Bi2Se3 layer and deposited NiFe on top of it. They then used the spin torque ferromagnetic resonance (ST-FMR) technique to determine the spin Hall angle. The obtained $\theta_{\text{SH}} = 2-3.5$ was larger than that of heavy metals by one order of magnitude. This work immediately generated great interest in the SHE of TIs. Nevertheless, studies on the SHE in Bi2Se3 face a big problem. Since Bi2Se3 is a V-VI group compound, strict control of the stoichiometry is required to obtain the insulating bulk. In reality, due to the existence of anti-site Se or anti-site Bi defects during MBE growth, there are always extrinsic electrons or holes that shift the Fermi level to either the conduction band or the valence band, making the bulk of Bi2Se3 a degenerated semiconductor. Therefore, there can exist parallel conduction in the bulk and on the surfaces of Bi2Se3. In such a situation, it becomes impossible to separate the electric currents flowing in the bulk and on the surfaces of Bi2Se3. The ratio between the surface current and the bulk current may vary from sample to sample, depending on the growth condition and the thickness of the Bi2Se3 thin film. Consequently, there has been a broad distribution of reported $\theta_{\text{SH}}$ values for Bi2Se3, making it difficult to draw a clear conclusion about the origin of the giant SHE in Bi2Se3, i.e. whether the observed giant spin Hall angle is real and originates from the surface states or not. For example, Y. Wang et
al. used the same ST-FMR technique to measure the spin Hall angle in Bi$_2$Se$_3$/CoFeB bilayers, but obtained only $\theta_{SH} = 0.047$ (room temperature) ~ 0.42 (50 K).$^23$ Meanwhile, the same group reported $\theta_{SH} = 1$–1.75 and successfully realized room-temperature SOT magnetization switching in Bi$_2$Se$_3$/NiFe bilayers with a current density as low as $6 \times 10^5$ A/cm$^2$. $^24$

4. Spin Hall effect in (BiSb)$_2$Te$_3$

(BiSb)$_2$Te$_3$ is a ternary TI that can avoid the bulk current problem in Bi$_2$Se$_3$. Since Bi$_2$Te$_3$ tends to be n-type while Sb$_2$Te$_3$ tends to be p-type, it is possible to prepare alloy (BiSb)$_2$Te$_3$ thin films with insulating bulk by adjusting the Sb composition (88–96%).$^{25}$ The spin Hall angle of (BiSb)$_2$Te$_3$ was first evaluated in (Bi$_{0.5}$Sb$_{0.5}$)$_2$Te$_3$/(Cr$_{0.08}$Bi$_{0.54}$Sb$_{0.38}$)$_2$Te$_3$ bilayers.$^{26}$ Because (Cr$_{0.08}$Bi$_{0.54}$Sb$_{0.38}$)$_2$Te$_3$ is a diluted magnetic TI with a low Curie temperature, the experiment was performed at 1.9 K. SOT magnetization switching was realized at a very low current density of $8.9 \times 10^4$ A/cm$^2$. The spin Hall angle was investigated with second harmonic Hall measurements. However, the obtained $\theta_{SH} = 140$–425 value may have been significantly overestimated due to an artifact from the asymmetric magnon scattering on the surfaces of (Cr$_{0.08}$Bi$_{0.54}$Sb$_{0.38}$)$_2$Te$_3$.$^{27}$ The corrected value was obtained later in (BiSb)$_2$Te$_3$/Ti/CoFeB junctions, which yielded $\theta_{SH} = 2.5$ at room temperature.

5. Room temperature SOT magnetization switching by topological insulators

Room temperature SOT magnetization switching and its advantage compared with heavy metals was first demonstrated by J. Han et al. using Bi$_2$Se$_3$, (BiSb)$_2$Te$_3$, Ta, and Pt/CoTb bilayers.$^{28}$ Figures 2(a) and 2(b) show the SOT magnetization switching loops of the Bi$_2$Se$_3$/CoTb (4.6 nm) bilayer, while Figs. 2(c) and 2(d) show that of the reference Pt/CoTb (2 nm) and Ta/CoTb (2 nm) bilayer, respectively. The threshold switching current density in the Bi$_2$Se$_3$/CoTb (4.6 nm) bilayer was $3 \times 10^6$ A/cm$^2$, which was much smaller than of Pt/CoTb (2 nm) ($40 \times 10^6$ A/cm$^2$) and Ta/CoTb (2 nm) ($10 \times 10^6$ A/cm$^2$).

Figure 3(a) shows the effective spin Hall angle of (BiSb)$_2$Te$_3$, Bi$_2$Se$_3$, Pt, and Ta in junctions with CoTb measured by the loop shift method. The effective spin Hall angles of (BiSb)$_2$Te$_3$, Bi$_2$Se$_3$, Pt and Ta were 0.4, 0.16, 0.017, and -0.031, respectively. These values were smaller than their intrinsic values, which is explained by the small spin transmissivity of CoTb. Nevertheless, the effective spin Hall angles of (BiSb)$_2$Te$_3$ and Bi$_2$Se$_3$ are clearly larger than those of Pt and Ta. Figure 3(b) compares the power consumption of SOT switching using (BiSb)$_2$Te$_3$, Bi$_2$Se$_3$, Pt, and Ta, which confirmed the advantage of TIs.

6. Role of topological surface states

Although giant spin Hall angles and low SOT switching current densities have been confirmed in various TI/ferromagnet bilayers, the origin of the giant SHE in TIs is still not clear due to the fact that the current may flow on both the surfaces and in the bulk of TIs. To definitely determine the origin of the giant SHE in TIs, H. Wu et al. investigated the SHE in (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ thin films with varying Sb compositions.$^{29}$ As shown in Fig. 4(a), the Fermi level of (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ can be tuned by changing the Sb composition. When the Sb composition is about 93%, the Fermi level approaches the Dirac point. Figure 4(b) shows the sheet carrier density and the resistivity of (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ as a function of Sb composition. Near the Dirac point, the electron sheet density decreases and the resistivity reaches the maximum at $x = 93$%.
indicates that the Fermi level is inside the band gap and approaches the Dirac point. Figure 4(c) shows the threshold switching current density and the SOT-induced effective field as a function of Sb composition in (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$/Ti/CoFeB junctions. Near the Dirac point, the switching current density $J_c$ is at the minimum, and the SOT-induced effective field is maximized. This unambiguously demonstrates the surface state origin of the giant SHE. The $J_c$ of the (Bi$_{0.07}$Sb$_{0.93}$)$_2$Te$_3$/Ti/CoFeB junction is reduced to $5.2 \times 10^5$ A cm$^{-2}$, which is one to two orders of magnitude smaller than that of the reference Pt,Ta,W/Ti/CoFeB junctions. Furthermore, $\theta_{SH} = 2.5$ was obtained for the (Bi$_{0.07}$Sb$_{0.93}$)$_2$Te$_3$/Ti/CoFeB junction from second harmonic Hall measurements.

7. High electrical conductivity and giant spin Hall effect in BiSb

One disadvantage of using TIs as the spin Hall material in SOT-MRAM is their low electrical conductivity $\sigma$ of $\sim 10^4$ $\Omega^{-1}$m$^{-1}$. For example, the $\sigma$ of (Bi$_{0.07}$Sb$_{0.93}$)$_2$Te$_3$ discussed in the previous section is only $1.8 \times 10^4$ $\Omega^{-1}$m$^{-1}$, which is significantly smaller than the $6 \times 10^5$ $\Omega^{-1}$m$^{-1}$ of CoFeB. Therefore, in the junctions of Tl/metallic ferromagnets, most of the current will be shunted by the ferromagnetic layer and does not contribute to the generation of a pure spin current. For example, in a typical (Bi$_{0.07}$Sb$_{0.93}$)$_2$Te$_3$ (six quintuple layers)/CoFeB (1.5 nm) bilayer, 87% of the current will flow into the CoFeB layer. Therefore, finding a TI material with both high conductivity and a giant spin Hall effect is essential. Thus, we have been concentrating on BiSb.$^{30}$ Although BiSb is the first experimentally confirmed three-dimensional TI (Sb of 7–22%), it has a small bulk band gap of $\sim 20$ meV and complex surface states. Therefore, not much attention has been paid to BiSb since the discovery of Bi$_2$Se$_3$ and related compounds with a much larger band gap of $\sim 300$ meV and much simpler surface states. However, as a pure spin current source for SOT-MRAM, BiSb is very attractive because it has multiple surface states and high mobility as evidenced by the high bulk electrical conductivity of $4\sim 6.4 \times 10^5$ $\Omega^{-1}$m$^{-1}$. Furthermore, because Bi and Sb are in the same V-group, deviation of the composition or existence of anti-site defects does not result in any donors/acceptors that would generate free carriers and shift the Fermi level to the conduction band or to the valence band as in the case of Bi$_2$Se$_3$. As a result, the Fermi level of BiSb is always in the band gap, making it easier to investigate the origin of the SHE. Motivated by these promising prospects, we grew and characterized Bi$_{1-x}$Sb$_x$ thin films on semi-insulating GaAs(111)A substrates by MBE, with various Sb concentration ranging from 0 to 100%. By optimizing the growth condition, we were able to grow single crystalline Bi$_{1-x}$Sb$_x$ thin films on GaAs(111)A substrates, despite the large and changing lattice mismatch between Bi$_{1-x}$Sb$_x$ and GaAs(111) as the Sb concentration changes. For thick enough thin films, their conductivity approached those of bulk values, indicating that the crystal quality was high. From the temperature dependence of the electrical conductivity, we confirmed the existence of the metallic surface states of Bi$_{1-x}$Sb$_x$.31)
various Bi$_{1-x}$Sb$_x$ samples with different thickness and Sb concentrations $x$. We found that Bi$_{1-x}$Sb$_x$ thin films show electrical conductivity higher than $1 \times 10^5 \Omega^{-1}\text{m}^{-1}$ with an average of $2.5 \times 10^5 \Omega^{-1}\text{m}^{-1}$, which is higher than other Bi-based chalcogenides by one order of magnitude. Figure 5(b) shows the temperature dependence of the normalized resistivity of Bi$_{0.88}$Sb$_{0.11}$ thin films with thicknesses of 10 nm, 41 nm, and 92 nm. For the 92 nm-thick thin film, the resistivity exponentially increased as the temperature decreased, as expected for an intrinsic semiconductor, but it approached a constant value at low temperatures. This behavior can be explained by the parallel conduction model. Although the bulk Bi$_{1-x}$Sb$_x$ only has a band gap $\sim 15\text{ meV}$, the band gap increased to 200 meV, which is large enough so that most of the current flowed on the surfaces. The strong quantum confinement effect reflects the high crystal quality of BiSb thin films.

To evaluate the SHE in BiSb, we prepared Mn$_{0.45}$Ga$_{0.55}$ (3 nm)/Bi$_{0.9}$Sb$_{0.1}$ (10 nm) bilayers grown on GaAs(001) substrates. Because MnGa on GaAs(001) has a cubic crystal structure, BiSb grown on top of MnGa has a pseudocubic (012) orientation. MnGa has a very large perpendicular anisotropy field of 40–50 kOe. However, at the Mn concentration of 45%, the magnetization of MnGa is not perfectly perpendicular, but tilts toward the in-plane direction. The spin Hall angle was measured by the loop-shift technique with the experimental setup shown in Fig. 6(a). The pure spin current injected from BiSb to MnGa generated an effective anti-damping-like SOT field $H_{SO}$ that can either increase or decrease the coercive force of MnGa, depending on the current direction. Figure 6(b) shows the perpendicular hysteresis loops of MnGa measured under different current densities and polarities. Figure 6(c) plots the coercive force of MnGa and its change, which is equal to the effective SOT field, as a function of the current density inside the BiSb layer. This data indicates that the effective SOT field was as large as 2.3 kOe per (10$^6$ Acm$^{-2}$), which yielded $\theta H = 52$ for the BiSb(012) orientation. The giant spin Hall angle for the BiSb(012) surfaces can be understood by the fact that there are multiple Dirac points for the BiSb(012) surface states, compared with only one Dirac point for the (001) surface states.

Figure 7 shows SOT magnetization switching loops for a Mn$_{0.45}$Ga$_{0.55}$ (3 nm)/Bi$_{0.9}$Sb$_{0.1}$ (5 nm) bilayer. The threshold switching current density is only $1.5 \times 10^6$ Acm$^{-2}$, which is smaller than that of MnGa/Pt,Ta,IrMn bilayers by two orders of magnitude.
8. Current challenges and future prospects

Figure 8 shows the expected writing current and writing energy of a realistic SOT-MRAM device using various type of materials for the pure spin current source, including heavy metals, two-dimensional materials, and topological insulators. It can be seen that our MBE-grown BiSb outperformed all other materials by several orders of magnitude, because BiSb shows not only high electrical conductivity but also a giant spin Hall angle. However, most of the TIs thin films studied so far, including BiSb, were epitaxially grown on dedicated III-V semiconductor substrates by the MBE technique, which is not suitable for mass-production. Therefore, it is essential to investigate the performance of non-epitaxial TIs thin films deposited on silicon substrates with an industry-friendly technique, such as sputtering deposition. Recently, there was an attempt to investigate the performance of sputtered non-epitaxial Bi₁₋ₓSbₓ TI thin films by Mahendra DC et al., who found a promisingly large \( \theta_{\text{SH}} = 8.7 - 18.6 \) but very low \( \sigma = 7.8 \times 10^3 \) Ω⁻¹cm⁻¹. On the other hand, we have demonstrated that it is possible to deposit BiSb thin films on sapphire substrates by sputtering deposition with a high crystal quality and high electrical conductivity approaching those of MBE-grown thin films. Furthermore, we found that even non-epitaxial BiSb thin films deposited on Si substrates by MBE or...
sputtering deposition could show a high spin Hall performance. Further work is needed to demonstrate BiSb thin films with a high crystal quality and optimized spin Hall performance on Si substrates with sputtering deposition for ultralow power SOT-MRAM.

Fig. 8. (a) Switching current and (b) switching energy of SOT-MRAM using various materials as pure spin current source, plotted against their sheet resistance.

Acknowledgements The work on MBE-grown BiSb thin films was conducted in collaboration with Y. Ueda, N.H. D. Khang, and K. Yao and was supported by a Grant-in-Aid for Challenging Exploratory Research from MEXT (No. 16K14228).

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Received May. 31, 2020; Revised Sep. 2, 2020; Accepted Sep. 3, 2020