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Research on a pure spin current source using YPtBi topological semimetal and its device applications

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Spin-orbit torque (SOT) is one of the promising technologies for next generation spintronic devices because it can manipulate magnetization with low power consumption and ultra-high speed. In SOT-based spintronic devices, the charge-to-spin conversion efficiency is determined by the strength of the spin Hall effect (SHE), so-called, spin Hall angle θ_{SH}. To realize ultra-low power SOT devices, it is essential to explore spin Hall materials with large θ_{SH}. Because the SHE is originated from the spin orbit interaction, large θ_{SH} has been reported in materials with strong spin orbit interaction such as heavy metals (HMs) and topological materials. Although both kinds of materials have successfully demonstrated large θ_{SH}, topological materials, especially topological insulators (TIs) are most attractive thanks to their giant θ_{SH} larger than unity, which is originated form the strong intrinsic Berry phase mechanism from their topological surface states (TSSs). However, because conventional chalcogenide-based TIs having giant θ_{SH}

> 1 are composed of only V and VI group elements, device integration is still challenging due to their toxic elements and low melting points. Therefore, a new class of topological material which has both giant $\theta_{SH} > 1$ and high affinity of the silicon Back-End-of-Line (BEOL) process is strongly required. In this thesis, we focus on a half-Heusler alloy topological semimetal (HHA-TSM), YPtBi, and evaluated the SHE in YPtBi, to overcome this difficulty.

In Chapter 1, we introduced the background for this thesis. The spin current generation method and spintronic devices were reviewed, and the potential of a pure spin current generated by the SHE was emphasized.

In Chapter 2, we explained the fundamental phenomena in spin Hall materials such as the spin orbit interaction and the SHE. We then emphasized the importance of the Berry phase on the SHE, and explained that TSSs in topological materials play an important role because their Dirac points are hot spots for Berry phase. We also explained the magnetization dynamics and magnetoresistance in ferromagnetic materials.

In Chapter 3, we generalized the angle resolved second harmonic technique to include the ordinary Nernst effect (ONE), and demonstrated that it can disentangle the contribution from SOT, ONE, anomalous Hall effect, and spin Seebeck effect. These results provide an accurate way to precisely estimate SOT in a sample with large effective perpendicular magnetic anisotropy field. The generalized angle resolved second harmonic was applied to CoPt/YPtBi films with strong perpendicular magnetic anisotropy in following chapters.

In Chapter 4, we investigated the SHE and its mechanism in YPt alloy thin films with various resistivity and thickness. It was found that the effective spin Hall angle $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ of YPt increased linearly but drastically from 0.045 to 0.081 for a slight increase of its resistivity ρ_{YPt} from 299 to 331 μΩcm. This strong sensitivity of $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ to ρ_{YPt} was found to be governed by both the intrinsic mechanism and the extrinsic side-jump mechanism with opposite polarity. By deconvoluting the contribution of the intrinsic and extrinsic side-jump mechanisms, the large intrinsic spin Hall conductivity of 700±100 $\hbar/2e$ $\Omega^{-1}\mathrm{cm}^{-1}$ was obtained despite its amorphous structure. Furthermore, we found that spin relaxation length of 0.9 nm for YPt is larger than that expected for a crystallized Pt-based alloy with the same resistivity. Although these unique results are very different from the conventional Pt-based alloys, YPt also cannot realize $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ larger than unity. This result highlighted the importance of the contribution of a TSS to the giant SHE in YPtBi.

In Chapter 5, we successfully realized an efficient spin source by using a HHA-TSM, YPtBi, having both giant $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ (> 1) and high thermal stability (> 400°C). The YPtBi layers were deposited using the co-sputtering technique, which makes it much easier for industrial adoption. Existence of a TSS in our sputtered YPtBi films was confirmed by using the planar Hall technique. We revealed that the SHE in YPtBi is dominated by the intrinsic SHE from its TSS by evaluating conductivity and thickness dependence of $\theta_{\mathrm{SH}}^{\mathrm{eff}}$. We also performed current induced SOT switching by using CoPt/YPtBi heterostructure. Thanks to its giant $\theta_{\mathrm{SH}}^{\mathrm{eff}}$, full switching with ultra-low pulse current density of 10^6 A/cm^2 was demonstrated. Although we focused on YPtBi in this thesis, there are a lot of material combinations since HHA-TSMs include a group of three-element topological materials, and thus, we call for further investigation of the spin Hall performance of this family. Indeed, first principle calculations have suggested at least ten materials in this group. Although the YPtBi thin films in this thesis were deposited using multi target sputtering, bulk single crystal of some bismuthides have been synthesized by the Bi-flux technique, indicating that it is possible to make large single crystal targets of HHA-TSM for mass production. Contrasting to the case of TIs, we found that there is no obvious physical nor technical difficulty for HHA-TSMs to be used in realistic spintronic applications.

In Chapter 6, we evaluated the SHE in YPtBi while changing the Y/Pt composition ratio r to investigate the effect of stoichiometry on the SHE. We successfully grew highly ordered YPtBi films at r = 0.5– 1.5. Furthermore, the extra carrier density was minimized, and hole mobility was maximized at the exact stoichiometry of r = 1.0, resulting in the minimum σ_{YPtBi}. We achieved a large $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ of 1.7 at r = 1.0, where the power consumption of magnetization manipulation was minimized. We also found that the $\theta_{\mathrm{SH}}^{\mathrm{eff}}$–$\sigma_{\mathrm{YPtBi}}$ relationship is robust against the change of its stoichiometry unlike the conventional TIs, which is a strong advantage of this alloy for spintronic applications.

In Chapter 7, we investigated the SHE of YPtBi films grown at lower temperature. When we reduced the growth temperature from 600ºC to 300ºC, both $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ and effective spin Hall conductivity $\sigma_{\mathrm{SH}}^{\mathrm{eff}}$ decrease due to degradation of crystallinity of YPtBi and spin transparency at the interface between CoPt and YPtBi. We then reduced the Ar gas pressure to increase the migration energy of atoms arrived on the surface of YPtBi for 300ºC growth. By improving spin transparency via recovery of interfacial quality, we successfully achieved giant $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ up to 8.2, which is larger than those of most TIs. We demonstrated current-induced SOT magnetization switching with ultralow current density of 10^5 A/cm^2 in the YPtBi layer grown at 300ºC by both DC and pulse currents.

In Chapter 8, we investigated the SHE in YPtBi deposited by the sputtering technique on SiO_2/Si substrates. Although the crystallinity of YPtBi on a SiO_2/Si substrate was degraded by comparing with that on a c-Sapphire substrate, the spin Hall conductivity was maintained, which may be attributed to the robustness of the TSS. By using the 1 nm-thick Ta buffer layer, we successfully obtained not only large $\theta_{\mathrm{SH}}^{\mathrm{eff}}$ of 1.3 but also high conductivity of $1.4\times10^{5}\ \Omega^{-1}\mathrm{m}^{-1}$ which is desirable feature for low power SOT-MRAM. Our results successfully demonstrate the feasibility of YPtBi-based SOT devices such as SOT-MRAM and racetrack memory on SiO_2/Si substrate.

In Chapter 9, we proposed out-of-plane precession spin Hall oscillators (OPP-SHOs) which can oscillate without any bias-fields by controlling the hard axis. The effective oscillation can be excited at arbitrary situation N'_{x}, $N'_{\mathrm{z}} < N'_{\mathrm{y}}$, by applying an initial short pulse current I_{cri} followed by a small constant current $I_{\mathrm{max}} > I_{\mathrm{C}} > I_{\mathrm{min}}$, where N'_{i} (i = x, y, z) is the demagnetizing constant along the i-axis, I_{max} is the minimum current at which magnetization is fully relaxed along the $-y$-direction, I_{min} is the minimum current for sustainable oscillation around the y-axis. We derived analytical equations for I_{cri}, I_{max}, I_{min}, and maximum oscillation frequency f_{max}, and showed that they agreed well with numerical simulation results, which indicates that these equations are useful for design of bias-field-free SHOs. Furthermore, we showed how to improve the performance of our SHOs in term of its frequency and drive current. Oscillation frequency can be increased by controlling the magnetic anisotropy, or by applying a magnetic field along the $+y$-direction. Although HMs can reduce driving currents, topological materials, especially

YPtBi, can significantly reduce the driving current.

In Chapter 10, we summarized the achievements in this thesis. Our work opens the door to the next generation spintronic devices with YPtBi having both giant θ_{SH} and high thermal stability.