論文の内容の要旨

Spin-charge interconversion via Rashba spin-orbit coupling at metal/oxide interfaces

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Spintronics, which is the study based on two fundamental properties of carriers: spins and charges, has attracted great attention in last two decades. Spintronics has several advantages over conventional electronics. For example, flowing of the spins without moving charges cause less energy loss so spintronics devices have less heat dissipation. For making spintronics devices, the generation, manipulation, and detection of spins are the fundamental functions required. One of the most common way to reach these requirements are using the interconversion between charge and spin. So far, two mechanisms for the spin-charge interconversion originating from the spin-orbit interaction have been demonstrated: spin Hall effect (SHE) and Rashba-Edelstein effect (REE). Spin Hall effect appears in any conductive materials with spin-orbit coupling (SOC). An applied charge current converts to a transverse spin current in SOC materials; this charge-to-spin current conversion is called direct spin Hall effect (DSHE). The inverse effect, a process that converts a spin current into a transverse charge current, is called inverse spin Hall effect (ISHE). In contrast, REE originates from the Rashba SOC induced by the broken potential symmetry. At certain interface state in x-y plane without inversion potential symmetry, the Hamiltonian of Rashba SOC can be described as $\boldsymbol{H}_\mathrm{R} = \alpha_\mathrm{R}(\boldsymbol{p} \times \hat{\boldsymbol{z}}) \cdot \boldsymbol{\sigma}$, where $\boldsymbol{\sigma}$ is the vector of Pauli spin matrices, $\boldsymbol{p}$ is the momentum, and α_R is so-called Rashba parameter. Because the $\alpha_\mathrm{R}(\boldsymbol{p} \times \hat{\boldsymbol{z}})$ term acts as a fictitious Rashba field, the conduction electron spins are aligned along $\boldsymbol{p} \times \hat{\boldsymbol{z}}$ direction. This phenomenon is known as spin-momentum locking. When an electric field, the Fermi contour with spin-momentum locking shifts and generates non-equilibrium spin accumulation, whose gradient drives a diffusive spin current into an adjacent conductive layer. This charge-to-spin (C-S) conversion is called the direct Edelstein effect (DEE). In reverse, injecting the spin current into the interface generates charge current; this phenomenon is called the inverse Edelstein effect (IEE). Recently, it has been shown that the conversion efficiency through IEE can be even larger than SHE in typical SHE materials such as β-W and Pt [E. Lesne, *et al. Nat. Mat.* **15**, 1261-1266 (2016)]. Therefore, the spin-charge interconversion via Rashba SOC is expected to have greater potential in spintronics application. So far, the S-C conversion at three types of Rashba interfaces have been studied: metallic interface, metal/oxide interface and oxide/oxide interfaces. The origin of Rashba effect at metallic and oxide/oxide interfaces have been well studied in last few years. However, there are only few experiment reports of metal/oxide Rashba interface. For the possible spintronics application, the further understanding of metal/oxide type Rashba interface is necessary.

In this thesis, we focus on the spin-charge interconversion via Rashba spin-orbit coupling at metal/oxide interface. The spin-to-charge conversion is measured by spin pumping method and the charge-to-spin conversion is detected by spin-torque ferromagnetic resonance method. Firstly, to clarify guiding principles for designing metal/oxide interfaces, we investigated the materials dependence of conversion efficiency at various metal/oxide interfaces. In addition, because the metal/oxide interface is contact with metal layer directly, it is expected that the conversion efficiency at the interface state may be influenced by the adjacent metal bulk state. To understand the relation between metal/oxide interface and metal bulk, we changed the electron momentum relaxation time of metal bulk by varying temperature, and investigated the temperature dependence of S-C conversion at metal/oxide interface.

The major results of the thesis are as follows:

Strong modulation of Rashba spin-splitting due to material dependent electron distribution at metal/Bi_2O_3 interfaces. (Chapter 4)

To understand how to design the metal materials of nonmagnetic metal (NM)/oxide interface with large Rashba spin-spiltting, NM material dependence of spin-to-charge (S-C) conversion efficiency at NM/Bi_2O_3 interfaces is investigated by spin pumping method. Cu, Ag, Au, and Al are used as NM materials. We observed large modulation and sign change in spin-to-charge (S-C) conversion efficiency λ_{IEE} which corresponds to the variation of Rashba spin-splitting. The experimental results together with first-principles calculations indicate that such large variation is caused by material dependent electron distribution near the interface. We found that the work function difference between metal and oxide materials is an important essence for determining electron distribution and Rashba parameter α_{R}. A smaller work function difference corresponds to a stronger localization of electron distribution near NM nuclei which results in a larger α_{R} and λ_{IEE}. In addition, the sign of work function change the direction of interfacial electric field and therefore change the sign of α_{R}. We also found that the SOC of NM layer have almost no contribution to Rashba effect at metal/oxide interface because the SOC of Bi is much larger and becomes a dominant. This study suggests a way to design the metal/oxide interface for large Rashba spin-splitting and efficient spin-to-charge interconversion. [**H. Tsai**, S. Karube, K. Kondou, N. Yamaguchi, F. Ishii and Y. Otani, "Clear variation of spin splitting by changing electron distribution at non-magnetic metal/Bi_2O_3 interfaces", Scientific Reports, vol. 8, no. 1, 2018.]

Efficient spin-charge interconversion at Cu(Ag)/oxide interfaces without heavy elements. (Chapter 5)

In chapter 4, Bi_2O_3 is used as the oxide layer due to the large spin-orbit coupling of Bi. To improve the materials selections for oxide materials at metal/oxide interface, the oxide materials dependence of S-C conversion is investigated. SnO_2, HfO_2, Al_2O_3, SiO_2, ITO (Indium Tin oxide) are used as oxide layer. From spin pumping measurement, only Cu/ITO interface shows notable S-C conversion signal which is 5 times larger than others; this results cannot be explained by strength of SOC. We further investigate both S-C conversion and charge-to-spin conversion at Cu(Ag)/ITO interfaces. Both the conversion efficiency and α_{R} of Cu(Ag)/ITO are comparable to Cu/Bi_2O_3 despite of the 5-6 times smaller SOC of In and Sn than Bi. In addition, estimated spin current conductivity at Cu(Ag)/ITO interface can be even larger than typical spin Hall materials Pt and β–Ta. These results indicate that heavy element is not necessary for efficient spin-charge interconversion at metal/oxide interfaces. Such large α_{R} should originate from some special features of the electron distribution at Cu(Ag)/ITO interfaces though the exact shape of electrons distribution is out of our understanding. One hypothesis is that the conductive feature of ITO may enable more electrons locate near In or Sn nuclei than Bi of insulating Bi_2O_3. Further studies are required to understand whether the conductive oxide layer can really enhance the Rashba effect at metal/oxide interface. [K. Kondou, **H. Tsai**, H. Isshiki, and Y. Otani "Efficient spin current generation and suppression of magnetic damping due to fast spin ejection from nonmagnetic metal/Indium-tin-oxide interface", APL Materials 6, 101105, 2018.]

Enhancement of S-C conversion efficiency due to the increasing momentum relaxation time of metal layer at low temperature (Chapter6)

Spin-to-charge conversion coefficient λ_{IEE} can be described by $\lambda_{\mathrm{IEE}} = \alpha_{\mathrm{R}}\tau_{IEE}/\hbar$, where τ_{IEE} is the spin relaxation time at interface. Therefore, increasing τ_{IEE} is a way to enhance λ_{IEE} without changing α_{R}. Because τ_{IEE} is strongly influenced by the momentum relaxation time of adjacent metal layer, it is possible to modulated λ_{IEE} by changing τ_p^{NM}. We increases conductivity, which is proportional to τ_p^{NM}, of metal layer by decreasing temperature from 290 K to 10 K at Cu(Ag)/Bi_2O_3 interfaces. 40(17) percentages enhancement of λ_{IEE} at 10 K due to the increased τ_p^{NM} at Cu(Ag)/ Bi_2O_3 interface are observed. From the experiment results, we found that the spin relaxation time τ_{IEE} at metal/oxide interface is proportional to τ_p^{NM} of metal layer. This relation can be explained by the spin-momentum locking at interface and the additional momentum relaxation process induced by the hybridization between 2D interface state and 3D metallic state. This study indicates that λ_{IEE} at metal/oxide interface can be enhanced by increasing conductivity, i.e. making high conductivity metal layer, and also provides a further understanding of the mechanism of the spin-to-charge conversion at metal/oxide interfaces. [**H. Tsai**, K. Kondou, and Y. Otani in preparation.]

This thesis presents a systematic study on the spin-charge interconversion phenomenon at metal/oxide interfaces. The thesis consists of three studies, the metal materials dependence, the oxide materials dependence and the temperature dependence of the spin conversion at metal/oxide interfaces. The first two studies focus on the relation between Rashba spin-splitting and different species of metal or oxide materials. The third study focus on the contribution of the metal layer of the spin and momentum relaxation time in spin-to-charge conversion process. These results demonstrate how the materials dependent electron distribution modulates Rashba spin-splitting and also explain the additional spin relaxation process due to the metal layer in S-C conversion process. Several guidelines for designing metal/oxide interfaces with efficient spin conversion are provided: (i) Larger Rashba spin-splitting can be obtained by choosing metal materials with close work function with oxide layer. (ii) The sign of spin-to-charge conversion is related to the sign of work function difference. (iii) The heavy elements are not necessary for the oxide layer. (iv) The larger spin-to-charge conversion efficiency can be obtained by increasing conductivity of metal layer.

These understandings of spin conversion phenomenon at metal/oxide interfaces could be useful to design and propose the new spintronics devices utilizing effectively the interface.