A theoretical study on spin-charge coupled transport and entropy production characteristics in nearly compensated metals

［補償金属におけるスピン-電荷結合輸送およびエントロピー生成に関する理論研究］

A dissertation submitted to the Graduate school of Science and Engineering of Saitama University in partial fulfillment of the requirements for the degree of Doctor of Philosophy

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ABSTRACT

A theoretical aspect of entropy production rate in ambipolar conductors has been presented. As joule heating addresses the issue of entropy production in spin Hall geometry, we have evaluated the role of spin current in the energy dissipation mechanism in ambipolar conductors with identical spin-related characteristics between holes and electrons. Since spin injection induces imbalance between up and down spin chemical potentials, application of the Gibbs–Duhem (GD) relation to ambipolar conductors establishes a thermodynamic relation between the spin-dependent chemical potentials of holes and electrons, inducing an asymmetric spin splitting between the hole and electron chemical potentials. This yields two types of spin relaxation as in ambipolar conductors two modes of spin currents are present namely parallel and antiparallel spin current. The GD relation allow the antiparallel spin current, where hole and electron spins flow in the opposite direction, to have a large spin diffusion length, but to retain that of the parallel spin current at a standard value.

A long lifetime as well as a large distance in spin coherence are desirable for spintronics devices because spatially and temporally large spin coherence makes spin manipulation easier. We propose a Baber-type collision in nearly compensated metals which reveals extraordinarily large spin relaxation time (τ_s) associated with the antiparallel spin current. A theoretical study on the spin and charge transports in nearly compensated metals shows that i) antiparallel spin current satisfies the Onsager reciprocal relation in combination with conventional charge current and ii) both the longitudinal and Hall resistivities are influenced by the enhancement of spin relaxation time (τ_s). The resistivities are characterized in terms of two specific mechanisms, i.e., conventional Hall effect and the resonance Hall effect. It is also shown that the resonance Hall effect is coupled to a sustaining mode of antiparallel spin current. These findings convince that nearly compensated metals have potential for being used as spintronics materials.

Acknowledgement

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LIST OF PUBLICATIONS

Journal papers

1. **Mst Sanjida Aktar**, Masamichi Sakai, Shigehiko Hasegawa, Osamu Nakamura, and Hir oyuki Awano, "Entropy production by thermodynamic currents in ambipolar conductors with identical spin dynamics characteristics between holes and electrons", Journal of App lied Physics Express, Vol.12, No.5, Page 053004-1 to 053004-5 (2019).

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Conference presentations

1. **Mst Sanjida Aktar**, Masamichi Sakai, Shigehiko Hasegawa, and Osamu Nakamura, "En tropy production for charge and spin currents in bipolar conductors", a poster presented i n 65th Japan society of applied physics (JSAP) spring meeting, March 17-20, 2018, Wase da university, Tokyo.

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3. **Mst Sanjida Aktar**, Masamichi Sakai, Osamu Nakamura, Shigehiko Hasegawa, and Hir oyuki Awano, "Entropy production by thermodynamic currents in ambipolar conductors ; the case of antiparallel spin polarization for hole and electron" presented in 66th Japan so ciety of applied physics (JSAP) Spring meeting, March 9-12, 2019, Tokyo Institute of Tec hnology, Tokyo, Japan.

4. **Mst Sanjida Aktar**, Masamichi Sakai, Osamu Nakamura, Shigehiko Hasegawa, and Hir oyuki Awano, " Theoretical evaluation of spin coherence length of ambipolar conductors" presented in 80th Japan society of applied physics (JSAP) Autumn meeting, September 18 -21, 2019, Hokkaido University, Sapporo campus, Hokkaido, Japan.

5. **Mst Sanjida Aktar**, Masamichi Sakai, Toshihiro Yoshizumi, Osamu Nakamura, Shigehiko Hasegawa, and Hiroyuki Awano, " An effective carrier compensation under spin injection to ambipolar conductor with intrinsically different hole and electron densities" submitted to the 67th Japan society of applied physics (JSAP) Spring meeting.

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CHAPTER 1

1.1 General Spintronics

Solid state devices have long been dominated by electronics, which operates by controlling the flow of charge carriers, i.e. electrons and holes, to process and store information. Until 1970s electronic micro-processors and resistances in circuits were compacted into an integrated circuit in a semiconductor chip. Since the chip area is limited by considerations of cost, convenience, and practicality, one must increase the density of devices in a chip to keep abreast of the ever-increasing demands of computing. Gordon Moore, the visionary cofounder of Intel Corporation predicted that the number of transistors on an integrated circuit would double in every 18 months. That prediction, now known as Moore's Law, effectively described a trend that has continued ever since, the density has increased roughly by a factor of 2. What might stop device downscaling in accordance with Moore's law is not so much the difficulty of fabricating smaller and smaller devices, nor is it the fact that classical laws of physics will be defunct when device dimensions approach atomic scales, but it is the unmanageable energy and heat dissipation associated with switching of a device [1-7].

The best known electronic switch is a transistor. In the case of a "field effect transistor" (FET) or a "bipolar junction transistor" (BJT), the device is "on" when the active region (the channel of an FET or the base of a BJT) contains a large amount of mobile charges, and it is "off" when that region is depleted of mobile charges. Therefore, switching between logic bits can only be accomplished by physically moving charges in and out of the active region with an external agency (such as the gate voltage in an FET or the base current in a BJT). This physical motion consumes considerable energy, which is ultimately dissipated as heat [8].

The obvious way to reduce the dynamic dissipation during the switching event is to switch between states without moving charges. Unfortunately, this is virtually impossible in charge based electronics, where the difference in the amount of charge in the active region is used to demarcate logic levels.

Continued scaling of complementary metal-oxide-semiconductor (CMOS)-based transistors over the past few decades has reached the point where further scaling causes uncontrollable increase in the power dissipation. With the physical limits of CMOS technology approaching, it has become clear that the need to develop the new logic technology is inevitable.

Spintronics, or spin-electronics as it is often referred to, is a class of devices that manipulate the quantum-mechanical spin angular momentum degree of freedom of electrons, in addition to the charge degree of freedom, to process and store information. Spintronics-based devices are among the several technologies which could enable low power consumption, faster operation, and higher integration densities.

The "spin" of an electron is a quantum-mechanical property and can be crudely thought of as the tiny magnetic moment associated with the electron spinning about its axis. It is a pseudovector that has a fixed magnitude of $\hbar/2$ ($\hbar$ = reduced Planck's constant) and a variable direction or polarization. If the electron is placed in a magnetic field, only two polarizations are allowed and therefore can be viewed as stable and metastable. The polarization parallel to the field will be stable and that antiparallel to the field will be metastable. These two polarizations can encode the binary bits 0 and 1. Switching between them will involve merely flipping the spin, without moving the electron in space and causing current flow with the minimum energy dissipation of $g\mu_B B$ per bit flip event, where g is the Landé g-factor, μ_B is Bohr magneton, and B is flux density of the magnetic field [8,9].

Spintronics was coined in the 1990s to describe devices that take advantage of "spin". Adding the spin degree of freedom provides new effects, new capabilities, and new functionalities. Spintronics is considered as one of the most important emerging research areas which have the immense potential to provide high speed, low power and high density logic and memory electronic devices. The origin of this research field goes back to the discovery of the "giant magnetoresistance" (GMR) effect in the late 1980s [10,11]. Now a days GMR-based spin valves and magnetic tunnel junctions (MTJs) are rapidly found in large-scale commercial applications as magnetic field sensors in tape and disk read heads, position or proximity sensors in cars, automated industrial tools, and biomedical devices. The discovery of spin-transfer torques (STT)[12], together with the optimization of the tunneling magnetoresistance (TMR) and magnetic anisotropy in MgO-based MTJs [13-15] further enabled the realization of scalable

nonvolatile magnetic random access memories (MRAMs) [16-18]. Owing to their low energy consumption, fast switching, and superior endurance, STT-MRAMs are presently commercialized as a replacement for SRAMs in embedded cache memories, with potential applications also as a persistent DRAM technology.

Despite the great current interest in the basic principles and concepts of spintronics there are still obstacles to overcome in the implementation of such spintronic devices. To operate a spin-based functional device, the primary issue is the efficient injection of spin-polarized electrons from the source ferromagnet into the nonmagnetic channel across the interface. Then the spin orientation is detected using a second ferromagnetic drain electrode. Although the device concept is very simple and similar to that of a conventional FET, the implementation of spin-based devices is not that straightforward. The existing spintronic architectures and the proposed solid-state quantum computing schemes rely on the relatively long spin coherence times of conduction electrons. The farther (longer) the electrons in the nonmagnetic sample carry the spin coherence, the more useful the device is. Similarly, if an electron spin represents a qubit in a solid-state quantum computer, the longer the spin survives, the more reliably it can store information. The question of how spins of mobile electrons (and holes) lose their spin coherence is thus of the utmost importance for spintronic technology and for solid-state quantum computing. This has not yet been accomplished. A great deal of basic fundamental physics research will be needed before spintronics applications become a reality [19-21].

1.2 Motivation of the Research

In particular, for many conceptual spintronic devices it is important that the spin orientation of charge carriers remains unperturbed for a sufficiently long time to transport the spins over relevant distances. Important parameters therefore are the spin lifetime and spin diffusion length, which are measures for how long a spin remains unaffected and how far it can travel without being perturbed. Therefore, large spin diffusion length is a desirable feature in spintronics applications. Up to now the quantitative evaluation gives its magnitude scales ranges from

several nanometers (nm) to several micrometers (μm) for metals and from several nanometers to tens of nanometers for ferromagnetic materials [22].

A recent study by Sakai et. al. on the Hall effect measurement of Yttrium di hydride (YH_2), an ambipolar conductor, suggest that the spin diffusion length of YH_2 is much larger than the typical spin diffusion length of paramagnetic metals [23]. Fig.1.a shows an optical microscopy image of the Van der Pauw type Hall device which is used in this experiment. It consists of a single channel of nonmagnetic YH_2 and four magnetized electrodes of TbFeCo which are located at the corners of the square of YH_2 region. The channel length is designed to be approximately 10 μm. In this experiment spin polarized charge carriers are injected into the YH_2 channel by the TbFeCo electrodes and Hall voltages are also measured by the electrodes. Hall effect measurement of YH_2 (Fig.1.b) shows an anomalous Hall effect like feature at room temperature despite a relatively long channel length. From this experiment it is clear that spin polarized charge carriers can travel a long distance through the YH_2 channel without spin flipping, which certainly proves its large spin diffusion length.

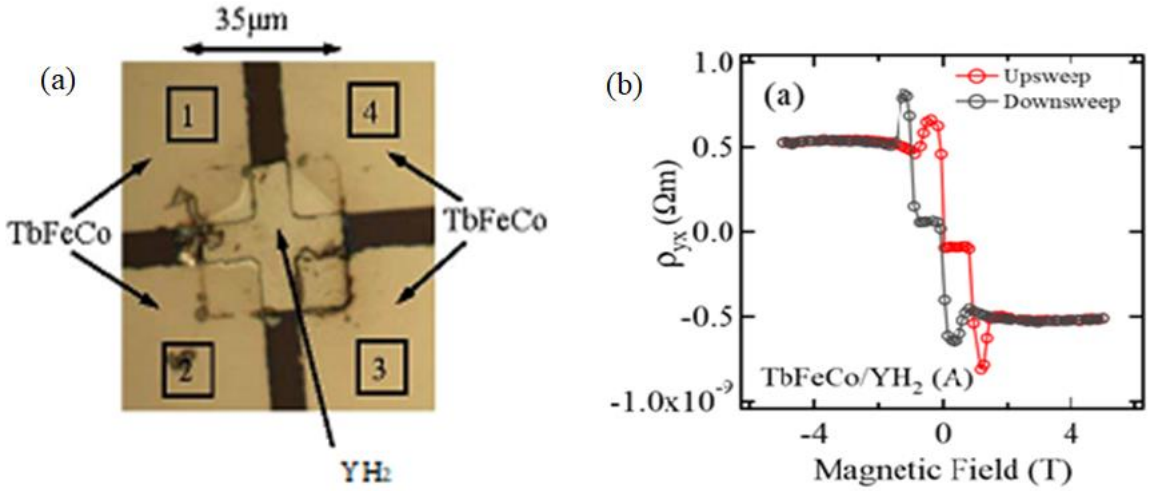

FIG.1.1 (a) Optical microscopy image of Hall device and (b) Room temperature Hall resistivity (ρ_{yx}) curve.

In another report [24] on spin-coherent length in an ambipolar conductor, Lee et.al. extended the spin injection technique to semimetal bismuth samples in a lateral spin valve geometry to study spin injection, diffusion, and detection in a material system where a small change in sample stoichiometry results in a large change in the electronic and spin dependent transport properties of the nonmagnetic material. A schematic diagram of the device is presented in Fig.1.2 (a) where F1, F2, and N denote a CoFe electrode, a NiFe electrode, and a BiPb layer, respectively. The nonlocal voltage between F2 and right Au electrode is measured with dc bias current applied to F1.

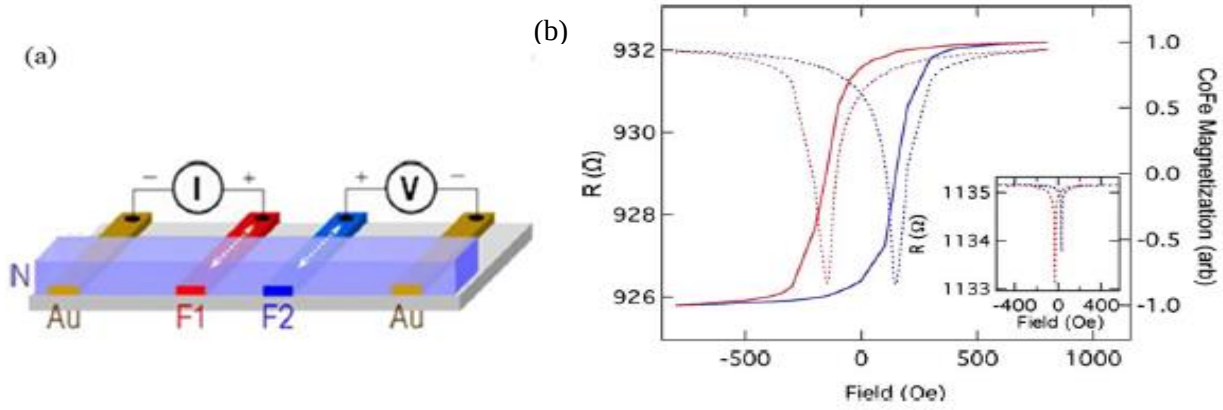

FIG. 1.2 (a) Schematic diagram of a lateral spin valve device and (b) Resistivity and Hall coefficient curve for patterned BiPb sample.

Temperature dependent resistivity, ρ_T and Hall coefficient measurements indicate that both electrons and holes contribute to charge and spin transport in BiPb sample set. Measurements of magnetoresistance (Fig. 1.2 (b)), using a magnetic field applied in the sample plane, as well as the Hanle effect, using a field applied perpendicular to the sample plane, reported a remarkable result of a large spin diffusion length of 230 μm (T=2 K) in a BiPb sample. These long spin diffusion lengths are undoubtedly related to the unusual transport properties of bismuth, but authors acknowledge of no known theory for spin dependent transport in group V materials to explain this extraordinary characteristic of semimetal Bismuth.

These two experimental results on large spin diffusion length of ambipolar conductors stimulate us to investigate the reason for having such a large spin diffusion length by unveiling its spin dependent transport theory.

Another important issue in spintronics devices is the generation of pure spin current still dissipates energy. Dissipation is associated with the production of entropy. Since reducing power consumption remains an important objective in spintronics devices, the development of a theory of energy dissipation due to spin current is indispensable. Theoretical investigation of entropy production rate has been done previously by several authors' using different methodology. For example, the dissipation produced by a spin current has been considered by Johnson and Silsbee[25], J.-E. Wegrowe [26], Sears and Saslow [27] by using irreversible thermodynamics. Tulapurkar and Suzuki [28] used the Boltzmann equation to investigate bulk and interface heating for diffusive flow of spin currents. More recently, Taniguchi [29] has derived a theoretical formula for the entropy production rate in the presence of spin current. Unlike previous theoretical works, his theory is applicable regardless of the source of the spin current, such as nonlocal spin injection [30,31], spin pumping by ferromagnetic resonance [32-34], the spin Hall effect [35,36], or the spin Seebeck effect [37,38], and shows that the energy dissipation of the spin Hall geometry has a contribution proportional to the square of the spin Hall angle. However, the theoretical formula developed by Taniguchi deals with only single charge carrier case either hole or electron take part in conduction. But for ambipolar conductors, where both holes and electrons participate simultaneously in electrical conduction, it is necessary to derive a theoretical formula regarding the energy dissipation for efficient manipulation of spin current in such a multi carrier system.

We have three main purposes for the research on ambipolar conductors:

1. To establish a theoretical formula for the energy dissipation,
2. To investigate the reason for having large spin diffusion length,
3. To formulate spin-charge transport coefficient.

1.3 Introduction to Ambipolar conductors

Ambipolar conductors are in the class of compensated metals or semimetals in which the conduction of holes and electrons take place concurrently. A metal is said to be compensated if the total number of electrons per unit cell of the reciprocal lattice summed over the electron sheets equals the total number of holes summed over the hole sheets [39-40]. The family of compensated metals are widely large. Galvanomagnetic studies in 1960-70s have systematically explored that a number of elemental metals, i.e., Be, Mg, Zn, Cd, Ga, Tl, Sn, Pb, Bi, Sb, Mo, W, Re, Pd, and Pt are compensated. Along with these elemental metals, various metal compounds, i.e., type-II Weyl semimetal WP_2 [41,42] and MoP_2 [43], Fe pnictide/chalcogenide (FePn/Ch) [44], YH_2 [45], and GdH_2 [46] have been revealed for their compensated metal (CM) characteristics. All these systems share a common characteristic of equal concentration of positively and negatively charged quasiparticles. But YH_2, and GdH_2 are defined as a nearly compensated metals because there exists a very small difference in the number densities of holes (n_h) and electrons (n_e) and can be specified by the charge polarization Φ $(= n_h - n_e/n_h + n_e)$ [23]. So, YH_2, and GdH_2 are commonly considered as nearly compensated metals with a very small charge polarization.

In the thesis, we use a term “ambipolar conductor” to represent a nearly compensated metals in which both charge carriers (electrons and holes) simultaneously take part in electrical conduction.

1.4 Orientation of the thesis

The dissertation is organized in the following way:

Chapter 2 contains a brief description about the fundamental concepts of spintronic devices. For using ambipolar conductors in spintronics device application which aims at low power consumption, entropy production rate equation of ambipolar conductors has been derived theoretically in chapter 3. The 4th chapter deals with the spin relaxation mechanism of ambipolar conductors and hence spin diffusion length of ambipolar conductor is calculated. In chapter 5, the knowledge of spin relaxation and spin diffusion length is employed in spin and charge

transport for calculating Hall resistivity and longitudinal resistivity in nearly compensated metals. Finally, the conclusions are drawn in chapter 6 by summarizing the main results.

CHAPTER 2

FUNDAMENTAL ASPECTS OF SPIN TRANSPORT

2.1 Introduction

The spin-dependent transport in hybrid nanostructures is currently of great interest, particularly in the emergence of new phenomena as well as the potential applications to spintronic devices. As spin current is the central to the operation of spin-based nano electronic devices, the efficient manipulation of spin current is essential for developing functional and energy-efficient nano spintronic devices. Recent experimental and theoretical studies have demonstrated that the spin-polarized carriers injected from a ferromagnet (FM) into a nonmagnetic material (NM), such as a normal conducting metal, compensated metal, semiconductor and/or superconductor give rise to nonequilibrium spin accumulation and spin current over the spin diffusion length [47]. In this chapter we will discuss the basic aspects for spin injection, spin transport and spin detection in magnetic nanostructures.

The basic principle of a spintronics device is the ability to generate, control, and detect the spin polarization of charge carriers. In 1980s Johnson and Silsbee was first proposed a device structure F1/NM/F2, consists of a nonmagnetic metal (NM) connected to the two ferromagnets (FMs), for electrical spin injection, manipulation and detection [48, 49]. In these devices, a spin-polarized current is injected from a FM source (F1) into nonmagnetic material (NM) to create nonequilibrium spin accumulation. This spin imbalance diffuses away from the injection point and reaches another FM detector (F2) which measures its local magnitude. This model provides a simple but intuitively correct picture for complete understanding of the phenomenology. Before describing the spin transport mechanism in the hybrid structure, we will briefly discuss about some relevant topics such as the origin of ferromagnetism in transition metal ferromagnets, as it provides the most common methods for generation of spin polarized charge carriers and the concept of electrochemical potential.

2.2 Ferromagnetism: Source of spin polarized current

The characteristic property of ferromagnetism in a solid is the spontaneous long-range order of magnetic moments in absence of an applied magnetic field because of a delicate balance between the exchange interaction and the atomic hybridization. In ferromagnetic 3d metals the d band is exchange split [50, 51]. Due to the localized nature of d-electrons, two d electrons experience a strong Coulomb repulsion provided that they have antiparallel spins and occupy the same orbital. To reduce the energy, it is advantageous for the d electrons to have parallel oriented spins because the Pauli exclusion principle does not permit two electrons with the same spin to approach each other closely (i.e. occupy the same orbital) and hence the Coulomb interaction is reduced. Therefore, the Coulomb repulsion in conjunction with the Pauli principle leads to the ferromagnetic exchange interaction and favors the formation of spontaneous magnetic moment.

However, putting all the electrons into states with the same spin direction increases the total kinetic energy. In ferromagnets like Fe, Ni and Co, the energy gain from the exchange interaction is strong enough to cause a splitting of the d-bands leading to reshuffling of spin-polarized 3d bands. This split results in an imbalance of the concentrations of spin-up and spin-down electrons which leads to ferromagnetism.

The condition that has to be satisfied for the appearance of ferromagnetism is the famous Stoner criterion $\boldsymbol{J}_{ex}N(E_F) > 1$, where $\boldsymbol{J}_{ex}$ is exchange constant and $N(E_F)$ is the density of states for a given spin at the Fermi energy [50]. From the Stoner criterion, it is evident that ferromagnetism will arise in materials which have a strong exchange integral and a large density of states (DOS) at the Fermi level. The DOS of ferromagnetic metal is shown in Fig 2.1 illustrating the splitting of 3d band. By convention the larger amount of spin-polarized carriers are called majority carriers $\left(n_{\uparrow}(E_F)\right)$ and the smaller amount as minority carriers $\left(n_{\downarrow}(E_F)\right)$. This asymmetry in the DOS at the Fermi level can be defined as polarization of ferromagnet and is expressed as

$$P = \frac{n_{\uparrow}(E_F) - n_{\downarrow}(E_F)}{n_{\uparrow}(E_F) + n_{\downarrow}(E_F)}, \tag{2.1}$$

where $n_\uparrow$ and $n_\downarrow$ are the up and down spin carriers' number density. The quantity of spin polarization P in ferromagnets is one of the important parameters for application in spintronics since a ferromagnet having a higher P is able to generate larger various spin-dependent effects such as the magnetoresistance effect, spin transfer torque, spin accumulation, and so on.

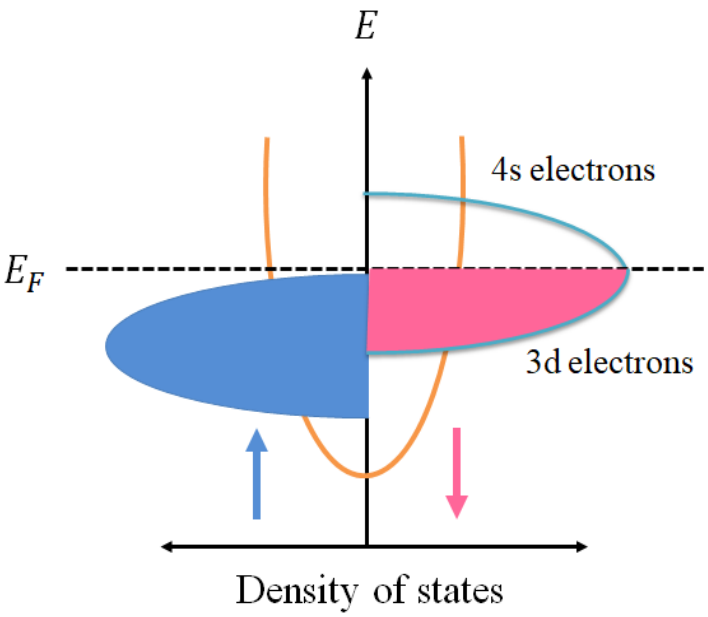

FIG. 2.1 A simplified band structure of a ferromagnetic metal showing splitting of 3d band and net density of states at Fermi level between spin-up and spin-down states. The arrows indicate the spin direction of the carriers.

2.3 Electrochemical potential

To describe the spin polarized transport in hybrid systems such as FM/NM interface, it is necessary to introduce the basic physical concept of chemical potential and electrochemical potential and the difference between them. Chemical potential (μ) is defined as the Gibbs free energy necessary to add a particle in a system. When a large number of particles within a certain volume are in thermodynamic equilibrium with the surroundings, then it will have a constant chemical potential. For the fermionic system the chemical potential is approximately equal to the Fermi energy (E_F). By convention, the chemical potential of electrons at the Fermi surface is often set to zero. If two systems with different chemical potentials are placed in contact, then the gradient in the chemical potential leads to the fundamental driving force for particle transport between the two systems. For small variations from equilibrium ($\Delta\mu \ll E_F$), the chemical potential is related to the excess particle density (n) via the density of states at the Fermi energy $N(E_F)$ which is expressed as [52]

$$\mu = \frac{n}{N(E_F)}\,. \tag{2.2}$$

However, in addition to the kinetic energy, a particle may also have potential energy. If the system of particles is kept at a certain electrostatic potential V, then the relevant quantity in describing the thermodynamic equilibrium will be the electrochemical potential (ε)

$$\varepsilon = \mu \pm eV. \tag{2.3}$$

where e denotes hole/electron charge. From the above equation the electrochemical potential is linearly related to the electron density. Therefore, a gradient in the electrochemical potential provides the driving force that leads to electron transport induced by both applied electric field ($\mathbf{E} = -\mathrm{grad}\, V$) and diffusion of electrons due to spatially varying particle density ($\mathrm{grad}\, n$).

2.4 The model of Spin transport in hybrid nanostructure

To comprehend the underlying phenomenon of spintronics device which encompasses spin injection, accumulation, and detection, we consider a model of three terminal device made up of a nonmagnetic material NM sandwiched between two ferromagnets F1 and F2 [48,49,53] . Schematic diagram of the device is shown in Fig.2.2. A dc current is driven through Fl into NM and returned to the current source from the bottom of NM. The magnetization of F1 and F2 are oriented either parallel or antiparallel. A single voltage probe is attached to F2. The sign of voltage is determined by the relative magnetization orientations of F1 and F2. The working principle of these devices is described in the following three sections:

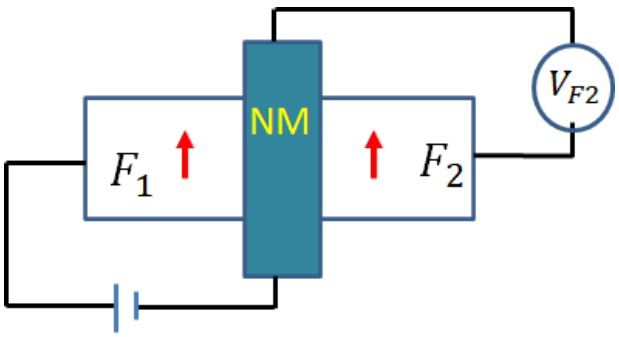

FIG.2.2 Model of three terminal device for spin injection and detection. Arrows in F1 and F2 refer to magnetization orientation.

2.4.1 Spin injection

An electric current in a ferromagnetic metal (F) is spin polarized, because spontaneous magnetization allows the electronic states to become spin-dependent, while for nonmagnetic materials, the electronic states come in pairs with the same energy but with opposite spin, which leads to a density of states independent of spin. When a ferromagnet is in interfacial contact with a nonmagnetic metal, the current crossing the FM/NM interface is spin polarized. This phenomenon is broadly known as spin injection [48].

By flowing an electrical current from F1 which is shown in Fig. 2.2, spin polarized electrons are injected into non-magnetic layer NM [54,55]. As the number density and mobility of the electrons at the Fermi level carrying the electrical current in F1 is different for opposite spin directions, the conductivities for majority spin and minority spin electrons are unequal [56,57]. Here we consider the magnetization direction of the ferromagnetic metal is up, so, we refer to the majority spins as "spin-up" (↑) and the minority spins as "spin-down" (↓). The charge current in F1 is thus $\boldsymbol{I} = (\boldsymbol{I}_{\uparrow} + \boldsymbol{I}_{\downarrow})$, which will contribute a net spin or magnetization current $\boldsymbol{I}_s == (\boldsymbol{I}_{\uparrow} - \boldsymbol{I}_{\downarrow})$ entering NM, with $\boldsymbol{I}_{\uparrow}(\boldsymbol{I}_{\downarrow})$ the current components associated to spin-up (down) electrons.

2.4.2 Spin accumulation

As the conductivities for spin up and spin down electrons are equal in NM, the injection of spin-polarized current from FM to NM induces a chemical potential splitting in the NM layer because of a sudden change in spin-dependent electrical conductivity. The splitting of chemical potential between up and down spin at the interface is known as spin accumulation. This spin accumulation diffuses relatively far from the interface and extends over a distance of the order of spin diffusion length (SDL), the length scale over which spin current flows. There is a broad zone of spin accumulation which extends over characteristic distances λ_s^{FM} and λ_s^{NM} on both sides of the F/N interface as shown in Fig. 2.3

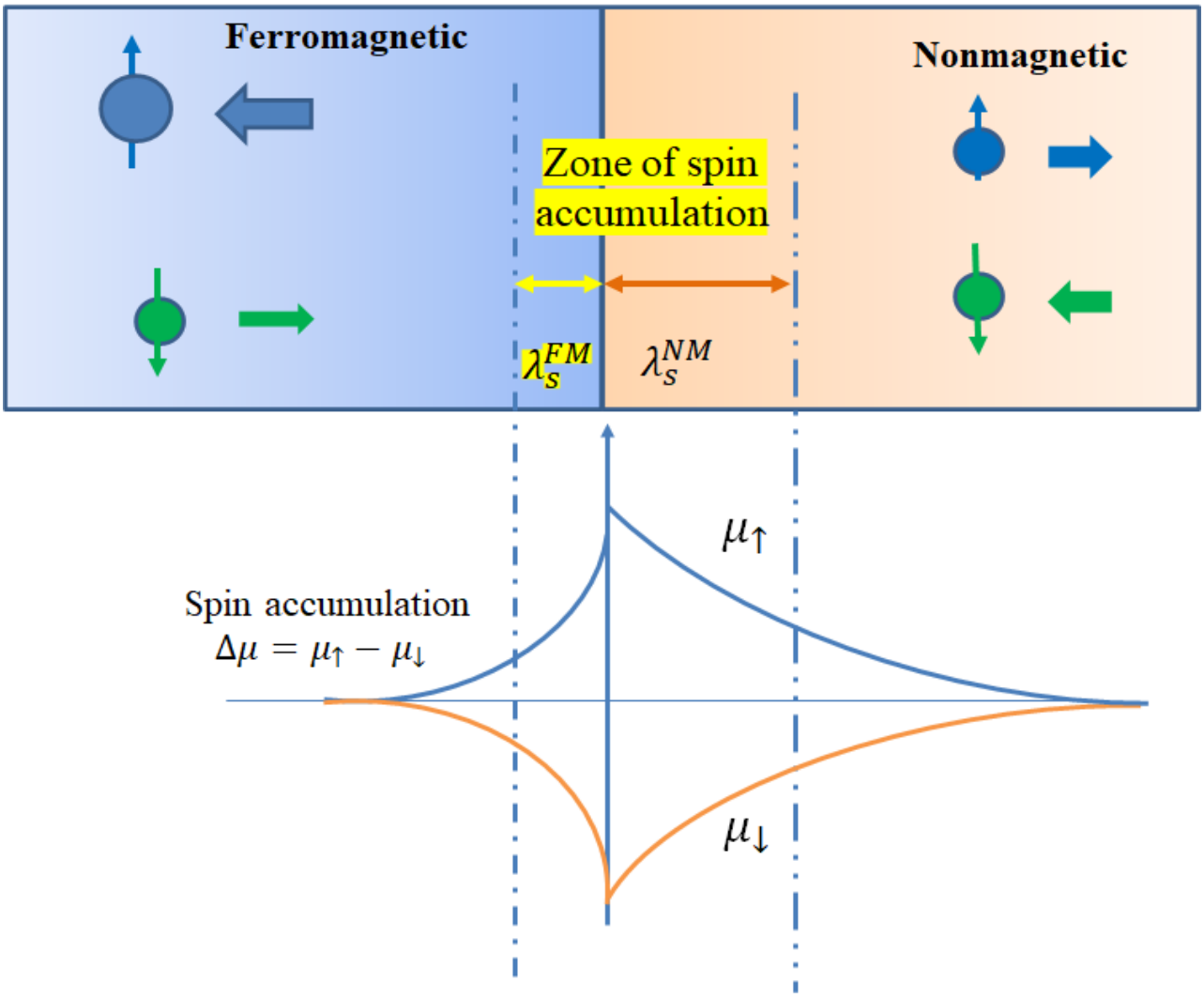

FIG. 2.3 Schematic representation of spin accumulation at an interface between a ferromagnetic metal and a nonmagnetic layer.

The microscopic spin transport of F1/NM/F2 device can be explained by using density of states diagram. In the absence of an imposed current, the Fermi levels E_F of all three films align at E_{F_0}, Fig 2.4 (a). For simplicity we consider F1 and F2 are half metals, so its spin polarization is $P = 1$. When a current is driven from Fl into N, only one spin sub-band in Fl is available to carry the current because transport involves only electrons within the energy range $\pm k_B T$ of E_F, which induces a difference in spin sub-band chemical potential in NM and forces to rise its up spin chemical potential to align with that of up-spin sub-band chemical potential of F1. As the spin carriers diffuses towards the F2, the spin accumulation forces the chemical potential of the F2 to adjust in order to maintain the steady state of no charge flow into F2. When the magnetization of F2 is parallel with that of Fl (Fig 2.4 (b)), its chemical potential will also rise so that the chemical potential of its up-spin sub-band aligns with that of the up-spin sub-band of NM. If the magnetization of F2 is antiparallel with that of Fl, then its chemical potential lowers (Fig. 2.4 (c)). In this case, the chemical potential of down spin in F2 aligns with that of the down-spin sub-band of NM [48,49].

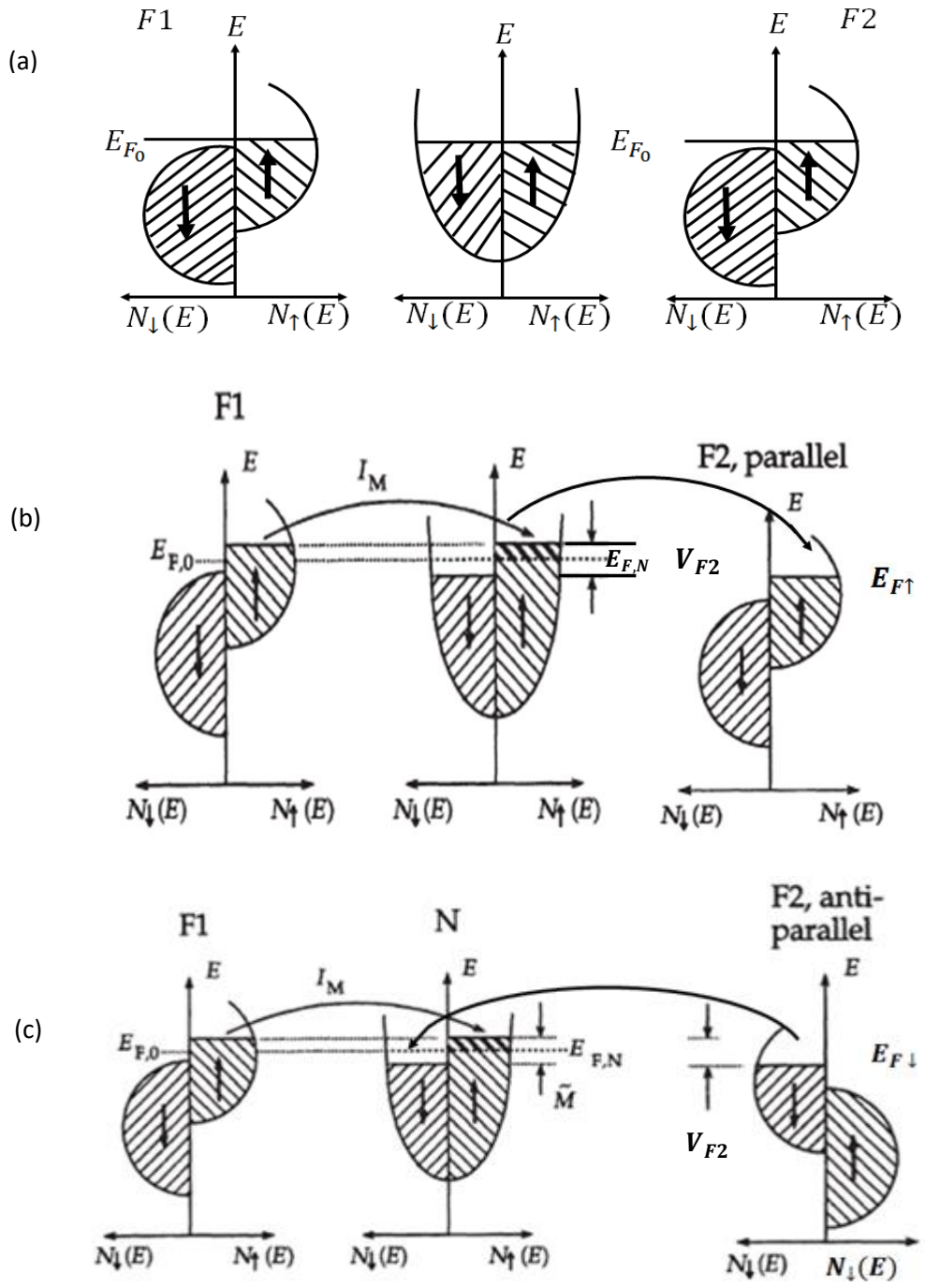

FIG. 2.4 (a) Density of states diagram for F1, NM, and F2 without bias current. Schematic illustration of density of states when bias current is present for parallel magnetization (b) and (c) antiparallel magnetization.

2.4.3 Spin detection

A second ferromagnetic film F2 in Fig. 2.2 that is in interfacial contact with the NM region acts as a spin detector. Spin accumulation in NM can be probed by a voltage labelled as V_{F2} , an output terminal which is attached to F2, in Fig.2.2. When this terminal is connected to ground through a low impedance current meter, a positive current $\boldsymbol{I}_d \propto \left(E_{F,N_\uparrow} - E_{F,N}\right)$ (where $E_{F,N}$ is the average chemical potential of the two spin subbands) is driven across the N/F2 interface and through a current detector when the magnetizations F1 and F2 are parallel [Fig.2.4 b]. When F1 and F2 are antiparallel, the current $\boldsymbol{I}_d \propto \left(E_{F,N} - E_{F,N_\downarrow}\right)$ is negative. A gradient of spin sub band electrochemical potential across the N/F2 interface (a thermodynamic force) causes an interfacial electric field (an emf source) that drives an electric current, either positive or negative depending on the sign of the gradient, across the interface [58].

In order to quantify the magnitude of the spin accumulation and V_{F2} , a common approach is based on a diffusive transport model [25,52,59-61], which is justified by the spin resolved Boltzmann equation [59] when the spin-diffusion length is larger than the mean free path of the electrons. For properly designed devices, the solution for a one dimensional (1D) geometry is in excellent agreement with the experimental results.

CHAPTER 3

THEORETICAL EVALUATION OF ENTROPY PRODUCTION RATE IN AMBIPOLAR CONDUCTORS

3.1 Introduction

Spintronics uses the spin degree of freedom to demonstrate new functionality in ferromagnetic/nonmagnetic hybrid devices. The basic principle of spintronics is to utilize spin current to transport information. Various mechanisms such as spin field effect transistor (spin-FET), spin-transfer torque, spin pumping, and (inverse) spin Hall effect have been proposed to generate, control, and detect the spin current, which made designing practical devices a reality. The most imperative and primary issue associated with these generic spintronics device is the production of current-generated Joule heat in both ferromagnetic (FM) injector and NM channel, as the spin diffusion length and the spin polarization strongly deteriorate with increasing charge current, resulting in a drastic decrease of the spin generation efficiency. So, Joule heating is naively considered to be disadvantageous to meet the future need for low-power operated, ultrasmall, and multifunctional electronic devices and can be a limiting factor in the performance of spintronic devices.

Spin current consists of antiparallel flows of up ($\hbar/2$) and down ($-\hbar/2$) spin angular moments with equal magnitude particle current density, i.e., $|\mathbf{j}_\uparrow| = |\mathbf{j}_\downarrow|$ where $2\pi\hbar$ is Planck's constant [62]. Since the same type of charge is accompanied by antiparallel flows of up and down spins, a spatial gradient of scalar potential cannot couple with the spin current, so no Joule heat is generated. Instead, a spatial gradient of nonequilibrium chemical potential difference between up and down spin particles is required for a continuous flow of the spin current [63-65]. As a result, the spin current is not precluded from entropy generation due to spin relaxation, i.e., intermixing of up and down spins. The extra heat associated with the spin degree of freedom is usually attributed to the mechanisms of spin accumulation, spin relaxation or the spin-flip scattering leading to a change of the entropy. So, dissipation produced by a spin-polarized current or pure spin-current is an important issue and associated with the production of entropy.

3.2 Derivation of entropy production rate equation

To derive a theoretical formula for the entropy production rate of ambipolar conductors where both holes and electrons simultaneously participate in electronic conduction, we start with the spin dependent transport of holes and electrons which can be written using the local form of conservation law as the continuity equations for holes and electrons:

$$\begin{aligned} \operatorname{div} \mathbf{j}_{\nu}^{(\mathbf{h})} &= -\frac{\partial n_{\nu}^{(h)}}{\partial t} - \varphi_{\nu}^{(h)}, \\ \operatorname{div} \mathbf{j}_{\nu}^{(\mathbf{e})} &= -\frac{\partial n_{\nu}^{(e)}}{\partial t} - \varphi_{\nu}^{(e)} \end{aligned} \tag{3.1}$$

where n_ν and $\mathbf{j}_\nu$ are the spin-ν particle density and spin-ν particle current density respectively. The spin relaxation rate φ_ν is given as [47,52]

$$\begin{aligned} \varphi_{\uparrow}^{(h)} &= \frac{n_{\uparrow}^{(h)}}{\tau_{\uparrow\downarrow}^{(h)}} - \frac{n_{\downarrow}^{(h)}}{\tau_{\downarrow\uparrow}^{(h)}}, \qquad \varphi_{\downarrow}^{(h)} = -\frac{n_{\uparrow}^{(h)}}{\tau_{\uparrow\downarrow}^{(h)}} + \frac{n_{\downarrow}^{(h)}}{\tau_{\downarrow\uparrow}^{(h)}} \\ \varphi_{\uparrow}^{(e)} &= \frac{n_{\uparrow}^{(e)}}{\tau_{\uparrow\downarrow}^{(e)}} - \frac{n_{\downarrow}^{(e)}}{\tau_{\downarrow\uparrow}^{(e)}}, \qquad \varphi_{\downarrow}^{(e)} = -\frac{n_{\uparrow}^{(e)}}{\tau_{\uparrow\downarrow}^{(e)}} + \frac{n_{\downarrow}^{(e)}}{\tau_{\downarrow\uparrow}^{(e)}}. \end{aligned} \tag{3.2}$$

In equation (3.2) $\tau_{\nu\bar{\nu}}$ represents the average time for flipping a spin-ν to a spin-$\bar{\nu}$ and $\varphi_{\uparrow}^{(e/h)} + \varphi_{\downarrow}^{(e/h)} = 0$ indicates electron (hole) number conservation. Since our system is embedded in an external field, the total energy consists of the internal energy and potential energy due to the external field [66]. Therefore, the total energy current density $\mathbf{J}_E$ can be written in terms of internal energy current density $\mathbf{J}_u$ and the potential energy current density due to particle flows of the spin-ν holes and electrons as

$$\mathbf{J}_{\mathrm{E}} = \mathbf{J}_{\mathrm{u}} + \sum_{\nu=\uparrow,\downarrow} (eV)\, \mathbf{j}_{\nu}^{(\mathrm{h})} + \sum_{\nu=\uparrow,\downarrow} (-eV)\, \mathbf{j}_{\nu}^{(\mathrm{e})}, \tag{3.3}$$

where e and V are the hole charge and scalar potentials, respectively.

On the other hand, $\mathbf{J}_{\mathrm{u}}$ is thermodynamically expressed in terms of the heat current density $\mathbf{J}_Q$ and the flow densities of the spin-ν electron chemical potential $\mu_{\nu}^{(e)}$ and the spin-ν hole chemical potential $\mu_{\nu}^{(h)}$ as [67]

$$\mathbf{J}_{\mathbf{u}} = \mathbf{J}_{\mathrm{Q}} + \sum_{\nu=\uparrow,\downarrow} \mu_{\nu}^{(h)} \mathbf{j}_{\boldsymbol{\nu}}^{(\mathbf{h})} + \sum_{\nu=\uparrow,\downarrow} \mu_{\nu}^{(e)} \mathbf{j}_{\boldsymbol{\nu}}^{(\mathbf{e})}, \tag{3.4}$$

because the Gibbs free energy is given by the sum of the chemical potentials of constituent particles. Substitution of Eq. (3.4) into Eq. (3.3) yields

$$\mathbf{J}_{\mathrm{E}} = \mathbf{J}_{\mathrm{Q}} + \sum_{\nu} \left(\mu_{\nu}^{(h)} + eV\right) \mathbf{j}_{\boldsymbol{\nu}}^{(\mathbf{h})} + \sum_{\nu} \left(\mu_{\nu}^{(e)} - eV\right) \mathbf{j}_{\boldsymbol{\nu}}^{(\mathbf{e})}, \tag{3.5}$$

Equation (3.5) indicates that the total energy current density of the ambipolar conductor consists of heat current density and the electrochemical potential current densities from hole and electron contributions.

Next, we take advantage of the energy conservation law $\partial E/\partial t + \mathrm{div}\, \mathbf{J}_{\mathbf{E}} = 0$, where E is the total energy density. The definition of Gibbs free energy density yields

$$\sum_{\nu=\uparrow,\downarrow} \left(\mu_{\nu}^{(h)} n_{\nu}^{(h)} + \mu_{\nu}^{(e)} n_{\nu}^{(e)}\right) = E - eV \sum_{\nu=\uparrow,\downarrow} \left(n_{\nu}^{(h)} - n_{\nu}^{(e)}\right) + P - TS\,, \tag{3.6}$$

where S is the entropy density and T is temperature. We obtain from Eq. (3.6) under constant pressure P

$$\frac{\partial E}{\partial t} = \sum_{\nu=\uparrow,\downarrow} \left(\mu_\nu^{(h)} \frac{\partial n_\nu^{(h)}}{\partial t} + \mu_\nu^{(e)} \frac{\partial n_\nu^{(e)}}{\partial t} \right) + eV \sum_{\nu=\uparrow,\downarrow} \frac{\partial n_\nu^{(h)}}{\partial t} - eV \sum_{\nu=\uparrow,\downarrow} \frac{\partial n_\nu^{(e)}}{\partial t} + T \frac{\partial S}{\partial t} . \tag{3.7}$$

Substitution of Eq. (3.5) and (3.7) into the energy conservation law yields:

$$\begin{aligned} T \frac{\partial S}{\partial t} + \mathrm{div}\mathbf{J}_\mathrm{Q} + \sum_{\nu=\uparrow,\downarrow} \mathbf{j}_\nu^{(\mathrm{h})} \cdot \mathrm{grad}\left(\mu_\nu^{(h)} + eV\right) \\ + \sum_{\nu=\uparrow,\downarrow} \mathbf{j}_\nu^{(\mathrm{e})} \cdot \mathrm{grad}\left(\mu_\nu^{(e)} - eV\right) \\ - \sum_{\nu=\uparrow,\downarrow} \left(\mu_\nu^{(h)} + eV\right)\varphi_\nu^{(h)} - \sum_{\nu=\uparrow,\downarrow} \left(\mu_\nu^{(e)} - eV\right)\varphi_\nu^{(e)} = 0 . \end{aligned} \tag{3.8}$$

In Eq. (3.8) entropy production rate is expressed with the spin dependent electrochemical potential of holes and electrons. For further calculation of entropy production rate, two types of spin dependent chemical potential of holes and electrons is defined for ambipolar conductors.

In the present situation, we consider a system in which ferromagnetic material is used as the source of spin polarized charge carriers and injected into the ambipolar conductor. Application of an external electric field to the ferromagnetic material produces spin accumulation at the interface between ferromagnet and ambipolar conductor as shown in Fig. 2.3. Spin accumulation refers to the imbalance of number density of charge carriers between the two spin states (up and down). This imbalance of the system can be measured in terms of chemical potential as it defines as the average energy requires to add a particle in a system. So, at the interface chemical potential of charge carriers split into the up ($\mu_\uparrow$) and down ($\mu_\downarrow$) spin chemical potential from it's average value ($\bar{\mu}$) which is defined by the following equations:

$$\mu_{\uparrow}^{(h)} = \bar{\mu}^{(h)} + \Delta\mu^{(h)}, \qquad \mu_{\downarrow}^{(h)} = \bar{\mu}^{(h)} - \Delta\mu^{(h)},$$

$$\mu_{\uparrow}^{(e)} = \bar{\mu}^{(e)} + \Delta\mu^{(e)}, \qquad \mu_{\downarrow}^{(e)} = \bar{\mu}^{(e)} - \Delta\mu^{(e)}, \tag{3.9}$$

where $\Delta\mu^{(h/e)}$ is the spin splitting chemical potential of charge carriers which explains that if up spin chemical potential is increased by $\Delta\mu^{(h/e)}$ then down spin chemical potential will be decreased by the same amount or vice versa. Using the above definition, $\bar{\mu}^{(h/e)}$and $\Delta\mu^{(h/e)}$ are expressed as $\frac{1}{2}\left(\mu_{\uparrow}^{(h/e)} + \mu_{\downarrow}^{(h/e)}\right)$ and $\frac{1}{2}\left(\mu_{\uparrow}^{(h/e)} - \mu_{\downarrow}^{(h/e)}\right)$ respectively. Although $\bar{\mu}^{(h/e)}$ are spatially homogenous in our ambipolar conductors, and $\Delta\mu^{(h/e)}$are spatially inhomogeneous owing to spin injection and/or accumulation. Figure 3.1 shows a schematic diagram of the chemical potentials of holes and electrons.

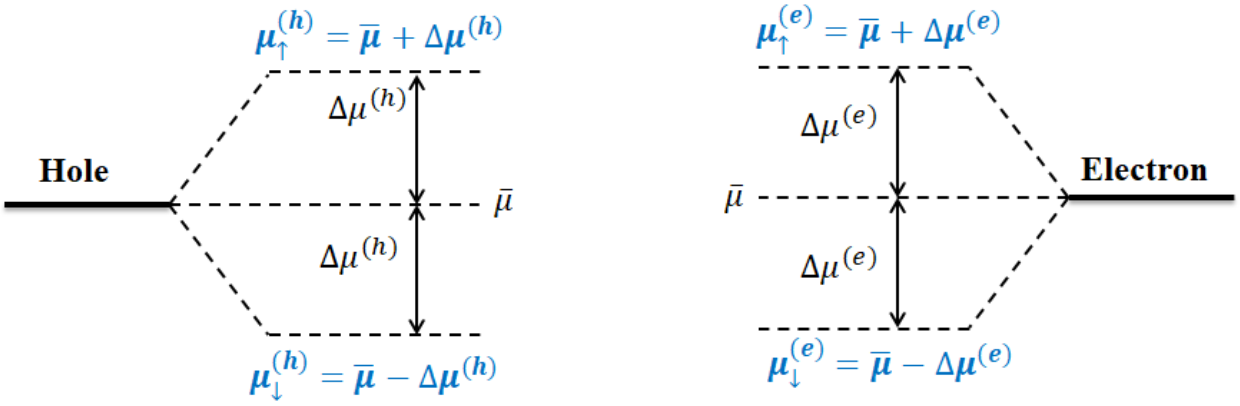

FIG. 3.1 Schematic of the definitions of $\bar{\mu}^{(h)}$ and $\Delta\mu^{(h)}$ for holes (a) and $\bar{\mu}^{(e)}$ and $\Delta\mu^{(e)}$ for electrons (b)

We cannot determine which spin (up or down) has a large chemical potential at this stage of calculation. Substituting Eq. (3.9) into Eq. (3.8), we temporally arrive at an equation for the entropy production rate of ambipolar conductors:

$$\frac{\partial S}{\partial t}+\operatorname{div}\left(\frac{\mathbf{J_Q}}{T}\right)=\mathbf{J_Q}.\operatorname{grad}\left(\frac{1}{T}\right)-\frac{1}{T}\mathbf{J_C}.\operatorname{grad}V-\frac{1}{\hbar T}\left(\mathbf{J_S^{(h)}}+\mathbf{J_S^{(e)}}\right).\operatorname{grad}\left(\Delta\mu^{(h)}+\Delta\mu^{(e)}\right)$$
$$-\frac{1}{\hbar T}\left(\mathbf{J_S^{(h)}}-\mathbf{J_S^{(e)}}\right).\operatorname{grad}\left(\Delta\mu^{(h)}-\Delta\mu^{(e)}\right)-\frac{1}{\hbar T}\left(\Delta\mu^{(h)}+\Delta\mu^{(e)}\right)\operatorname{div}\left(\mathbf{J_S^{(h)}}+\mathbf{J_S^{(e)}}\right)$$
$$-\frac{1}{\hbar T}\left(\Delta\mu^{(h)}-\Delta\mu^{(e)}\right)\operatorname{div}\left(\mathbf{J_S^{(h)}}-\mathbf{J_S^{(e)}}\right) \qquad (3.10)$$

In deriving Eq. (3.10), electron spin current and hole spin current have been expressed as $\mathbf{J_S^{(e)}}=\hbar/2\left(\mathbf{j}_{\uparrow}^{(e)}-\mathbf{j}_{\downarrow}^{(e)}\right)$ and $\mathbf{J}_{\mathrm{S}}^{(h)}=\hbar/2\left(\mathbf{j}_{\uparrow}^{(h)}-\mathbf{j}_{\downarrow}^{(h)}\right)$, respectively. Charge current densities for hole $\mathbf{J}_{\mathrm{C}}^{(h)}=e\left(\mathbf{j}_{\uparrow}^{(h)}+\mathbf{j}_{\downarrow}^{(h)}\right)$ and for electron $\mathbf{J}_{\mathrm{C}}^{(e)}=-e(\mathbf{J}_{\uparrow}^{(e)}+\mathbf{J}_{\downarrow}^{(e)})$ are defined in such way that both contribute to the total charge current as $\mathbf{J}_{\mathrm{C}}=\mathbf{J}_{\mathrm{C}}^{(h)}+\mathbf{J}_{\mathrm{C}}^{(e)}$. Since the total charge current is given by $\mathbf{J_C^{(h)}}+\mathbf{J_C^{(e)}}$, it is natural to define the total spin current as $\mathbf{J_S^{(h)}}+\mathbf{J_S^{(e)}}$, which is called parallel spin current in this study, while we call $\mathbf{J_S^{(h)}}-\mathbf{J_S^{(e)}}$ antiparallel spin current.

The entropy production rate equation (3.10) involves spin splitting of the chemical potential of holes and electrons in the form of $\Delta\mu^{(h)}\pm\Delta\mu^{(e)}$. To determine a relationship between spin splitting chemical potential of holes and electrons, we employ the Gibbs-Duhem (GD) relation [68] because this relation deals with thermodynamic change in the chemical potential in a mixer of different components. Since ambipolar conductors have up and down spin of electrons and holes one can write the Gibbs free energy density as

$$G=n_{\uparrow}^{(h)}\mu_{\uparrow}^{(h)}+n_{\downarrow}^{(h)}\mu_{\downarrow}^{(h)}+n_{\uparrow}^{(e)}\mu_{\uparrow}^{(e)}+n_{\downarrow}^{(e)}\mu_{\downarrow}^{(e)}. \qquad (3.11)$$

Spin injection and/or accumulation in ambipolar conductor alters these carrier concentrations and chemical potentials. When the spin injection and accumulation are achieved under constant temperature and pressure, the GD relation for ambipolar conductor takes the following form:

$$n_{\uparrow}^{(h)} d\mu_{\uparrow}^{(h)} + n_{\downarrow}^{(h)} d\mu_{\downarrow}^{(h)} + n_{\uparrow}^{(e)} d\mu_{\uparrow}^{(e)} + n_{\downarrow}^{(e)} d\mu_{\downarrow}^{(e)} = 0\,, \tag{3.12}$$

where $n_{\nu}^{(h/e)}$ are spin-ν particle densities before spin injection/accumulation. From the definition of carrier spin polarization $P_S^{(h/e)} = \left(n_{\uparrow}^{(h/e)} - n_{\downarrow}^{(h/e)}\right)/\left(n_{\uparrow}^{(h/e)} + n_{\downarrow}^{(h/e)}\right)$, the density of up and down spin charge carriers are represented in terms of spin polarization as

$$\begin{aligned} n_{\uparrow}^{(h)} &= \tfrac{1}{2} n^{(h)}\left(1 + P_S^{(h)}\right), \qquad n_{\downarrow}^{(h)} = \tfrac{1}{2} n^{(h)}\left(1 - P_S^{(h)}\right), \\ n_{\uparrow}^{(e)} &= \tfrac{1}{2} n^{(e)}\left(1 + P_S^{(e)}\right), \qquad n_{\downarrow}^{(e)} = \tfrac{1}{2} n^{(e)}\left(1 - P_S^{(e)}\right). \end{aligned} \tag{3.13}$$

Similarly, charge polarization definition $\Phi = \left(n^{(h)} - n^{(e)}\right)/\left(n^{(h)} + n^{(e)}\right)$ gives us hole and electron concentrations in terms of Φ as $n^{(h/e)} = \frac{1}{2} n_{total}(1 \pm \Phi)$. By assuming equal carrier spin polarization between electrons and holes with nonzero value ($P^{(h)} = P^{(e)} \equiv P \neq 0$), the GD relation can be represented in terms of P and Φ:

$$\begin{aligned} &(d\mu_{\uparrow}^{(h)} + d\mu_{\downarrow}^{(h)} + d\mu_{\uparrow}^{(e)} + d\mu_{\downarrow}^{(e)}) + \Phi(d\mu_{\uparrow}^{(h)} + d\mu_{\downarrow}^{(h)} - d\mu_{\uparrow}^{(e)} - d\mu_{\downarrow}^{(e)}) + P(d\mu_{\uparrow}^{(h)} - d\mu_{\downarrow}^{(h)} + d\mu_{\uparrow}^{(e)} - d\mu_{\downarrow}^{(e)}) \\ &+ \Phi P(d\mu_{\uparrow}^{(h)} - d\mu_{\downarrow}^{(h)} - d\mu_{\uparrow}^{(e)} + d\mu_{\downarrow}^{(e)}) = 0. \end{aligned} \tag{3.14}$$

If we assume that the averaged values of $\bar{\mu}^{(h/e)}$ are not influenced by spin injection/accumulation and go back to our chemical potential relation (Eq. (3.9) and Fig. 3.1), we

can substitute the relations $d\mu_{\uparrow}^{(h)} = -\,d\mu_{\downarrow}^{(h)} (\equiv -\Delta\mu^{(h)})$ and $d\mu_{\uparrow}^{(e)} = -\,d\mu_{\downarrow}^{(e)} (\equiv -\Delta\mu^{(e)})$ in Eq. (3.14). We therefore obtain a very simplified form

$$(1+\Phi)\Delta\mu^{(h)} + (1-\Phi)\Delta\mu^{(e)} = 0, \tag{3.15}$$

where we cannot use $\Phi = 1$ and $\Phi = -1$, which yield $\Delta\mu^{(h)} = 0$ and $\Delta\mu^{(e)} = 0$, respectively. In the ambipolar conductors with very small charge polarization value ($\Phi \ll 1$) [69], Eq. (3.15) reduces to $\Delta\mu^{(h)} + \Delta\mu^{(e)} = 0$. This equation states that if the chemical potential of spin up hole increases then the chemical potential of spin up electron decreases accordingly, as indicated in Fig. 3.2.

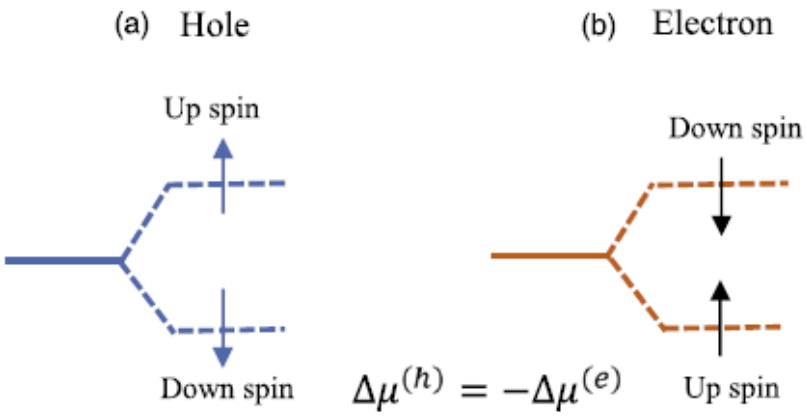

FIG. 3.2. (a) A hole has a large chemical potential for up spin and (b) an electron has a large chemical potential for down spin. This feature results from the Gibbs-Duhem relation with the condition $P_S^{(h)} = P_S^{(e)}$.

Substitution of $\Phi = -1$ into Eq. (3.14) yields $(1+P)d\mu_{\uparrow}^{(e)} + (1-P)d\mu_{\downarrow}^{(e)} = 0$, which thermodynamically explains the electron spin splitting caused by spin injection/accumulation in single carrier type conductors. Substitution of Eq. (3.15) into Eq. (3.10) yields

$$\frac{\partial S}{\partial t} + \mathrm{div}\left(\frac{\mathbf{J_Q}}{T}\right) = \mathbf{J_Q}.\,\mathrm{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J_C}.\,\mathrm{grad}V + \frac{2\Phi}{\hbar T(1-\Phi)}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right).\,\mathrm{grad}\,\Delta\mu^{(h)}$$
$$- \frac{2}{\hbar T(1-\Phi)}\left(\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}}\right).\,\mathrm{grad}\,\Delta\mu^{(h)} + \frac{2\Phi}{\hbar T(1-\Phi)}\Delta\mu^{(h)}\mathrm{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right)$$
$$- \frac{2}{\hbar T(1-\Phi)}\Delta\mu^{(h)}\mathrm{div}\left(\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}}\right). \qquad (3.16)$$

When the third, fourth, fifth and sixth terms on the right hand side of Eq. (3.16) are omitted, it becomes the usual expression in the presence of heat and charge currents [70].

Now we will simplify the divergence of parallel and antiparallel spin current which are present in the fifth and sixth terms on the right-hand side of Eq. (3.16). Using the definitions of spin current of holes and electrons, we can write the divergence of parallel spin current as

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right) = \frac{\hbar}{2}\mathrm{div}\left(\mathbf{j}_{\uparrow}^{(\mathbf{h})} - \mathbf{j}_{\downarrow}^{(\mathbf{h})} + \mathbf{j}_{\uparrow}^{(\mathbf{e})} - \mathbf{j}_{\downarrow}^{(\mathbf{e})}\right). \qquad (3.17)$$

Again, the divergence of up and down spin particle current densities of holes and electrons can be replaced with the continuity equation (3.1) and spin relaxation equation (3.2) then the above equation takes the following form

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right) = \frac{\hbar}{2}\left\{-\frac{\partial}{\partial t}\left(n_\uparrow^{(h)} - n_\downarrow^{(h)}\right) - \frac{\partial}{\partial t}\left(n_\uparrow^{(e)} - n_\downarrow^{(e)}\right) - 2\left(\frac{n_\uparrow^{(h)}}{\tau_{\uparrow\downarrow}^{(h)}} - \frac{n_\downarrow^{(h)}}{\tau_{\downarrow\uparrow}^{(h)}}\right) - 2\left(\frac{n_\uparrow^{(e)}}{\tau_{\uparrow\downarrow}^{(e)}} - \frac{n_\downarrow^{(e)}}{\tau_{\downarrow\uparrow}^{(e)}}\right)\right\} \tag{3.18}$$

Now we assume that $\tau_{\uparrow\downarrow}^{(h)} = \tau_{\uparrow\downarrow}^{(e)} = \tau_{\uparrow\downarrow}$ and $\tau_{\downarrow\uparrow}^{(h)} = \tau_{\downarrow\uparrow}^{(e)} = \tau_{\downarrow\uparrow}$. Applying this assumption and substituting the relations for spin dependent carrier number densities derived from spin and charge polarization definition in equation (3.18),we get

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right) = -\frac{\hbar}{2} n_0 \left[\frac{1}{\tau_{\uparrow\downarrow}} - \frac{1}{\tau_{\downarrow\uparrow}} + \frac{1}{2}\left(P_S^{(h)} + P_S^{(e)} + \Phi\left(P_S^{(h)} - P_S^{(e)}\right)\right)\left(\frac{1}{\tau_{\uparrow\downarrow}} + \frac{1}{\tau_{\downarrow\uparrow}}\right)\right] \tag{3.19}$$

similarly, the divergence of antiparallel spin current can be written in terms of spin relaxation

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}}\right) = -\frac{\hbar}{2} n_0 \left[\Phi\left(\frac{1}{\tau_{\uparrow\downarrow}} - \frac{1}{\tau_{\downarrow\uparrow}}\right) + \frac{1}{2}\left(\Phi\left(P_S^{(h)} + P_S^{(e)}\right) + P_S^{(h)} - P_S^{(e)}\right)\left(\frac{1}{\tau_{\uparrow\downarrow}} + \frac{1}{\tau_{\downarrow\uparrow}}\right)\right] \tag{3.20}$$

Since we assumed $P_S^{(h)} = P_S^{(e)} \equiv P_S \neq 0$ when obtaining Eq. (3.15), we also apply this spin polarization condition to Eq. (3.19) and (3.20) so that we obtain

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right) = -\hbar\left(\frac{n_\uparrow^{(h)} + n_\uparrow^{(e)}}{\tau_{\uparrow\downarrow}} - \frac{n_\downarrow^{(h)} + n_\downarrow^{(e)}}{\tau_{\downarrow\uparrow}}\right), \tag{3.21}$$

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}}\right) = -\hbar\Phi\left(\frac{n_\uparrow^{(h)} + n_\uparrow^{(e)}}{\tau_{\uparrow\downarrow}} - \frac{n_\downarrow^{(h)} + n_\downarrow^{(e)}}{\tau_{\downarrow\uparrow}}\right). \tag{3.22}$$

The detailed balance condition $N_{\uparrow}^{(h/e)}/\tau_{\uparrow\downarrow}^{(h/e)} = N_{\downarrow}^{(h/e)}/\tau_{\downarrow\uparrow}^{(h/e)}$, where $N_{\uparrow/\downarrow}^{(h/e)}$ represents density of states of charge carriers at the Fermi level, we employ it into Eq. (3.21) and (3.22) so that it yields

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right) = -\frac{2\hbar}{\tau_s}\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right) \times \Delta\mu^{(h)} \qquad (\Phi \neq \pm 1), \tag{3.23}$$

$$\mathrm{div}\left(\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}}\right) = -\frac{2\hbar}{\tau_s}\Phi\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right) \times \Delta\mu^{(h)} \qquad (\Phi \neq \pm 1). \tag{3.24}$$

where $N^{(h/e)}$ in the above equations represents this relation $\left(N_{\uparrow}^{(h/e)}N_{\downarrow}^{(h/e)}\right)/\left(N_{\uparrow}^{(h/e)} + N_{\downarrow}^{(h/e)}\right)$ and τ_s is used in terms of $(\tau_{\uparrow\downarrow}\tau_{\downarrow\uparrow})/(\tau_{\uparrow\downarrow} + \tau_{\downarrow\uparrow})$.

Now we are going to express parallel and antiparallel spin currents in terms of its spin dependent conductivities. For doing so , first we will write parallel and antiparallel spin currents in terms of charge current unit such as

$$\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}} = \frac{\hbar}{2e}\left\{\left(\mathbf{j}_{\uparrow}^{(\mathbf{h})} - \mathbf{j}_{\downarrow}^{(\mathbf{h})}\right) - \left(\mathbf{j}_{\uparrow}^{(\mathbf{e})} - \mathbf{j}_{\downarrow}^{(\mathbf{e})}\right)\right\} \tag{3.25}$$

$$\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}} = \frac{\hbar}{2e}\left\{\left(\mathbf{j}_{\uparrow}^{(\mathbf{h})} - \mathbf{j}_{\downarrow}^{(\mathbf{h})}\right) + \left(\mathbf{j}_{\uparrow}^{(\mathbf{e})} - \mathbf{j}_{\downarrow}^{(\mathbf{e})}\right)\right\} \tag{3.26}$$

Using Ohm's law, spin dependent electrical current densities can be written in terms of the electrical conductivity $\sigma_{\uparrow/\downarrow}$as

$$\mathbf{j}_{\uparrow/\downarrow} = -\frac{1}{q}\sigma_{\uparrow/\downarrow}\ \mathrm{grad}\ \varepsilon_{\uparrow/\downarrow}\ , \tag{3.27}$$

where $\varepsilon_{\uparrow/\downarrow}$ are the spin up (spin -down) electrochemical potentials consisting of the scalar potential ϕ_c due to external charges and the spin dependent chemical potential $\mu_{\uparrow/\downarrow}$caused by spin accumulation ; $\varepsilon_{\uparrow/\downarrow} = -q\phi_c + \mu_{\uparrow/\downarrow}$. Substituting this relation in Ohm's law gives

$$\mathbf{j}_{\uparrow/\downarrow} = \sigma_{\uparrow/\downarrow}(-\text{grad}\,\phi_c) + \sigma_{\uparrow/\downarrow}\left(-\frac{1}{q}\,\text{grad}\,\mu_{\uparrow/\downarrow}\right), \tag{3.28}$$

$$\mathbf{j}_{\uparrow/\downarrow} = \sigma_{\uparrow/\downarrow}\mathbf{E} + \sigma_{\uparrow/\downarrow}\Delta\mathbf{E} \tag{3.29}$$

where $-\text{grad}\,\phi_c$ gives a spin dependent electric field ($\mathbf{E}$) and $-\frac{1}{q}\,\text{grad}\,\mu_{\uparrow/\downarrow}$ gives a spin dependent effective field ($\Delta\mathbf{E}$). By using Ohm's law (3.29), spin dependent particle current densities of holes and electrons can be written in terms of electric field and effective field such as:

$$\mathbf{j}^{(h)}_{\uparrow/\downarrow} = \sigma^{(h)}_{\uparrow/\downarrow}\mathbf{E} \pm \sigma^{(h)}_{\uparrow/\downarrow}\Delta\mathbf{E}^{(h)} \tag{3.30}$$

$$\mathbf{j}^{(e)}_{\uparrow/\downarrow} = \sigma^{(e)}_{\uparrow/\downarrow}\mathbf{E} \pm \sigma^{(e)}_{\uparrow/\downarrow}\Delta\mathbf{E}^{(e)} \tag{3.31}$$

where $\Delta\mathbf{E}^{(h)} = -\left(\frac{1}{e}\,\text{grad}\,\Delta\mu^{(h)}\right)$ and $\Delta\mathbf{E}^{(e)} = \left(\frac{1}{e}\,\text{grad}\,\Delta\mu^{(e)}\right)$ represents the effective field for hole and electron respectively. Substituting Eqs. (3.30) and (3.31) in Eqs. (3.25) and (3.26) gives

$$\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}} = \frac{\hbar}{2e}\left\{\left(\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)}\right)\left(-\frac{1}{e}\,\text{grad}\,\Delta\mu^{(h)}\right) - \left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\left(\frac{1}{e}\,\text{grad}\,\Delta\mu^{(e)}\right)\right\} \tag{3.32}$$

$$\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}} = \frac{\hbar}{2e}\left\{\left(\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)}\right)\left(-\frac{1}{e}\,\text{grad}\,\Delta\mu^{(h)}\right) + \left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\left(\frac{1}{e}\,\text{grad}\,\Delta\mu^{(e)}\right)\right\} \tag{3.33}$$

Using Gibbs-Duhem relation $\Delta\mu^{(e)} = -\frac{1+\Phi}{1-\Phi}\Delta\mu^{(h)}$ in Equs. (3.32) and (3.33) yields parallel and antiparallel spin currents in terms of spin-dependent longitudinal conductivities as

$$\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}} = \frac{\hbar}{2e}\left\{\left(\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)}\right) - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right\}\left(-\frac{1}{e}\,\text{grad}\,\Delta\mu^{(h)}\right) \tag{3.34}$$

$$\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}} = \frac{\hbar}{2e}\left\{\left(\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)}\right) + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right\}\left(-\frac{1}{e}\,\text{grad}\,\Delta\mu^{(h)}\right) \qquad (3.35)$$

Substituting Eqs. (3.34) and (3.35) into Eqs. (3.23) and (3.24) gives

$$\nabla^2\left(\Delta\mu^{(h)}\right) = \frac{4e^2}{\tau_s}\,\frac{N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}}{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)} \times \Delta\mu^{(h)} \qquad (\Phi \neq \pm 1), \qquad (3.36)$$

$$\nabla^2\left(\Delta\mu^{(h)}\right) = \Phi\frac{4e^2}{\tau_s}\,\frac{N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}}{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)} \times \Delta\mu^{(h)} \qquad (\Phi \neq \pm 1) \qquad (3.37)$$

Equations (3.36) and (3.37) imply the presence of two types of spin relaxation channels, respectively associated with parallel and antiparallel spin currents. The spin diffusion lengths for the parallel and antiparallel spin currents are therefore given respectively by

$$l_P = \left[\frac{\tau_s}{4e^2}\,\frac{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)}{N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}}\right]^{1/2} \qquad (\Phi \neq \pm 1), \qquad (3.38)$$

$$l_{AP} = \left[\frac{\tau_s}{4e^2}\,\frac{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)}{\Phi\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right)}\right]^{1/2} \qquad (\Phi \neq \pm 1) \qquad (3.39)$$

The spin diffusion length characteristic associated with the parallel and antiparallel spin currents can be intuitively explained using Fig. 3.3. The condition of Eq. (3.15) indicates that one type of carrier (e.g., hole) has a higher chemical potential for up spin and other type of carrier (electron) has a higher chemical potential for down spin. The parallel spin current makes both the spin-up hole and the spin-up electron densities large, enhancing the relaxation of spin splitting given by

Eq. (3.15). On the other hand, the antiparallel spin current makes both the spin-up hole and the spin-down electron densities large, suppressing the relaxation of spin splitting given by Eq. (3.15).

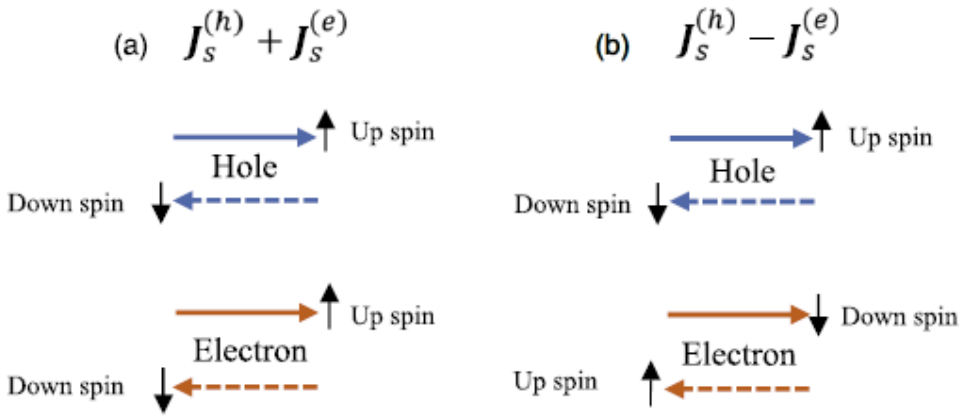

FIG. 3.3 (a) Hole and electron spin currents are in the same direction (parallel spin current) and (b) in the opposite direction (antiparallel spin current).

Entropy production rate equation given in Eq. (3.16) can be expressed in terms of one dimensional model by

$$\frac{\partial S}{\partial t} + \mathrm{div}\left(\frac{\mathbf{J_Q}}{T}\right) = \mathbf{J_Q}.\,\mathrm{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J_C}.\,\mathrm{grad}V$$
$$- \frac{\Phi}{(1-\Phi)e^2T}\left[\sigma_\uparrow^{(h)} + \sigma_\downarrow^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_\uparrow^{(e)} + \sigma_\downarrow^{(e)}\right)\right] \times \left(\frac{\Delta\mu_0^{(h)}}{l_P}\right)^2 \exp\left(-\frac{2x}{l_P}\right)$$
$$+ \frac{1}{(1-\Phi)e^2T}\left[\sigma_\uparrow^{(h)} + \sigma_\downarrow^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_\uparrow^{(e)} + \sigma_\downarrow^{(e)}\right)\right] \times \left(\frac{\Delta\mu_0^{(h)}}{l_{AP}}\right)^2 \exp\left(-\frac{2x}{l_{AP}}\right)$$
$$- \frac{4\Phi}{(1-\Phi)T\tau_s}\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right)\left(\Delta\mu_0^{(h)}\right)^2 \times \left[\exp\left(-\frac{2x}{l_P}\right) - \exp\left(-\frac{2x}{l_{AP}}\right)\right] \quad (3.40)$$

By simplifying Eq. (3.40), we get the entropy production rate equation in ambipolar conductor such as

$$\frac{\partial S}{\partial t} + \operatorname{div}\left(\frac{\mathbf{J_Q}}{T}\right) = \mathbf{J_Q}.\operatorname{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J_C}.\operatorname{grad}V - \frac{8\Phi}{(1-\Phi)T\tau_s}\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right)\left(\Delta\mu_0^{(h)}\right)^2 \times \left[\exp\left(-\frac{2x}{l_P}\right) - \exp\left(-\frac{2x}{l_{AP}}\right)\right] \tag{3.41}$$

3.3 Application of spherical Fermi surface to entropy production rate equation

Spherical Fermi surface yields a relation between the number density of electrons and/or holes with the density of states at the Fermi level which is expressed as [71]:

$$n^{(e)} = \frac{4}{3}E_F^{(e)}N^{(e)}, \tag{3.42}$$

$$n^{(h)} = \frac{4}{3}E_F^{(h)}N^{(h)}. \tag{3.43}$$

Now, if we assume, the Fermi energy for hole and electron is equal i.e. $E_F^{(h)} = E_F^{(e)}$, then the spherical Fermi surface model appears as the density of states of electrons and holes at the Fermi level is proportional to the density of electron and hole which yield the following result:

$$\frac{N^{(h)}(E_F) - N^{(e)}(E_F)}{N^{(h)}(E_F) + N^{(e)}(E_F)} = \frac{n^{(h)} - n^{(e)}}{n^{(h)} + n^{(e)}} = \Phi. \tag{3.44}$$

So, by substituting Eq.(3.44) into Eq. (3.41), it is found that the last term in Eq. (3.41) is zero. Finally, the entropy production rate equation for ambipolar conductor takes the usual expression as [70]

$$\frac{\partial S}{\partial t} + \mathrm{div}\left(\frac{\mathbf{J_Q}}{T}\right) = \mathbf{J_Q}.\,\mathrm{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J_C}.\,\mathrm{grad}V. \tag{3.45}$$

3.4 Significance of entropy production rate equation in ambipolar conductors

Our derived formula for entropy production rate $\left(\frac{\partial S}{\partial t}\right)$ in ambipolar conductor Eq. (3.45) reveals the usual expression in which the entropy is accompanied by the production of Joule heat due to the flow of charge current density $(\mathbf{J_C})$ and heat current density $(\mathbf{J_Q})$ and it is completely free from both parallel and antiparallel spin current term. It implies that spin currents in ambipolar conductor do not take part in entropy production and hence no dissipation is produced due to the transportation of spin current. From the derivation of entropy production rate equation it is observed that dissipationless spin current is emerged as the characteristic feature of ambipolar conductor which is the most desirable property for materials using in spintronics devices application.

3.5 Antiparallel spin polarization between hole and electron

We also investigate the circumstances of equal but opposite spin polarization between electrons and holes i.e., $P_S^{(h)} = -P_S^{(e)} = P_S \neq 0$, in this case Gibbs-Duhem equation turns out to be

$$\Delta\mu^{(e)} = \frac{(1+\Phi)}{(1-\Phi)}\Delta\mu^{(h)}, \tag{3.46}$$

the above relation clearly shows that spin splitting chemical potential between electron and hole is equal ($\Phi \ll 1$), if chemical potential of holes increases (decrease) then chemical potential of electrons will also increase (decrease). Using this relation, we can derive the divergence form of parallel and antiparallel spin currents in the similar way of preceding section as

$$\mathrm{div}\left(\vec{\mathbf{J}}_{\boldsymbol{S}}^{(\boldsymbol{h})} + \vec{\mathbf{J}}_{S}^{(e)}\right) = -\frac{2\hbar}{\tau_S}\left[N^{(h)} + \frac{1+\Phi}{1-\Phi}N^{(e)}\right]\Delta\mu^{(h)}, \tag{3.47}$$

$$\mathrm{div}\left(\vec{\mathbf{J}}_{S}^{(h)} - \vec{\mathbf{J}}_{S}^{(e)}\right) = -\frac{2\hbar}{\tau_S}\left[N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right]\Delta\mu^{(h)}. \tag{3.48}$$

Comparing Eqs.(3.47) and (3.48) with Eqs. (3.23) and (3.24), we see that in the present condition the charge polarization term does not appear in antiparallel spin current equation. In previous case, antiparallel spin current equation was accompanied with the charge polarization which enhanced its spin coherence. But antiparallel spin polarization condition yields no significant consequence for parallel and antiparallel spin current.

We can write entropy production rate equation for antiparallel spin polarization case

$$\frac{\partial S}{\partial t} + \mathrm{div}\left(\frac{\mathbf{J}_Q}{T}\right) = \mathbf{J}_Q.\mathrm{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J}_C.\mathrm{grad\ V}$$

$$-\frac{2}{\hbar T(1-\Phi)}\left(\mathbf{J}_S^{(h)} + \mathbf{J}_S^{(e)}\right).\mathrm{grad}\Delta\mu^{(h)} + \frac{2\Phi}{\hbar T(1-\Phi)}\left(\mathbf{J}_S^{(h)} - \mathbf{J}_S^{(e)}\right).\mathrm{grad}\Delta\mu^{(h)}$$

$$-\frac{2}{\hbar T(1-\Phi)}\Delta\mu^{(h)}\mathrm{div}\left(\mathbf{J}_S^{(h)} + \mathbf{J}_S^{(e)}\right) + \frac{2\Phi}{\hbar T(1-\Phi)}\Delta\mu^{(h)}\mathrm{div}\left(\mathbf{J}_S^{(h)} - \mathbf{J}_S^{(e)}\right). \tag{3.49}$$

The spin diffusion lengths for the parallel and antiparallel spin currents are therefore given respectively by

$$l_P = \left[\frac{\tau_s}{4e^2}\frac{\sigma_\uparrow^{(h)} + \sigma_\downarrow^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_\uparrow^{(e)} + \sigma_\downarrow^{(e)}\right)}{N^{(h)} + \frac{1+\Phi}{1-\Phi}N^{(e)}}\right]^{1/2} \qquad (\Phi \neq \pm 1), \tag{3.50}$$

$$l_{AP} = \left[\frac{\tau_s}{4e^2}\frac{\sigma_\uparrow^{(h)} + \sigma_\downarrow^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_\uparrow^{(e)} + \sigma_\downarrow^{(e)}\right)}{N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}}\right]^{1/2} \qquad (\Phi \neq \pm 1) \tag{3.51}$$

Substituting Eqs. (3.50 and (3.51) in Eq. (3.49), entropy production rate equation can be written as

$$\begin{aligned}\frac{\partial S}{\partial t} + \text{div}\left(\frac{\mathbf{J}_Q}{T}\right) = \mathbf{J}_Q.\text{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J}_C.\text{grad V} \\ + \frac{8}{T\tau_s(1-\Phi)}\left[N^{(h)} + \frac{1+\Phi}{1-\Phi}N^{(e)}\right]\left(\Delta\mu_0^h\right)^2 \exp\left(-\frac{2x}{l_P}\right) \\ - \frac{8\Phi}{T\tau_s(1-\Phi)}\left[N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right]\left(\Delta\mu_0^h\right)^2 \exp\left(-\frac{2x}{l_{AP}}\right).\end{aligned}$$

Now, we consider the spherical Fermi surface which results $N^{(h)} = \frac{1+\Phi}{1-\Phi}N^{(e)}$, makes the last term of the above equation zero. Finally, the entropy production rate equation for antiparallel spin polarization case takes the following form:

$$\frac{\partial S}{\partial t} + \text{div}\left(\frac{\mathbf{J}_Q}{T}\right) = \mathbf{J}_Q.\text{grad}\left(\frac{1}{T}\right) - \frac{1}{T}\mathbf{J}_C.\text{grad V} + \frac{16\,N^{(h)}}{T\tau_s(1-\Phi)}\left(\Delta\mu_0^h\right)^2 \exp\left(-\frac{2x}{l_P}\right)$$

This equation states that the parallel spin current is accompanied with the entropy production when antiparallel spin polarization is prevalent between electron and hole.

3.6 Summary

We have developed a formula for the entropy production rate in ambipolar conductors by assuming identical spin relaxation times and carrier spin polarization between holes and electrons, but no assumption is made on the carrier transport characteristics. The presence of two types of spin currents namely parallel and antiparallel spin currents are observed in ambipolar conductor which is defined as the vector sum of hole and electron spin currents and the vector subtraction of hole and electron spin currents, respectively. Application of the GD relation to spin thermodynamics gives a specific relationship between the spin splitting in the chemical potential of electrons and holes, which increases (decreases) the spin-up chemical potential for holes (electrons) due to spin injection/ accumulation. This asymmetric spin splitting between the hole and electron chemical potentials yields two types of spin relaxation, allowing the antiparallel spin current to flow with a large spin diffusion length, but keeping the spin diffusion length of the parallel spin current at a standard value. Finally, theoretical evaluation reveals no entropy production by the spin currents in ambipolar conductor. On the other hand, antiparallel spin polarization between electron and hole gives rise to another aspect. In this case, parallel spin current takes part in entropy generation.

CHAPTER 4

SPIN RELAXATION MECHANISM AND ENHANCEMENT OF SPIN COHERENCE IN NEARLY COMPENSATED METALS

In the previous chapter, we observed an enhancement factor of spin coherence length in ambipolar conductor by using several assumptions [72]. One of the most important assumptions for this enhancement is the identical nature of spin relaxation times between electrons and holes, i.e., $\tau_{\nu\bar{\nu}}^{(e)} = \tau_{\nu\bar{\nu}}^{(h)}$, where $\tau_{\nu\bar{\nu}}$ represents the average time for flipping a spin-ν to a spin-$\bar{\nu}$. Feasibility of this assumption, however, remains unclear. Here we show that the Baber-type collision [73] accompanied with spin flipping and the detailed balance conditions supports the validity of $\tau_{\nu\bar{\nu}}^{(e)} = \tau_{\nu\bar{\nu}}^{(h)}$, from which the enhancement factor of the spin flipping time is derived to be $1/\Phi$.

4.1 Effect of the Baber-type collision on spin relaxation time

In what follows, we incorporate degree of spin freedom to the Baber-type collision. We assume that the presence of two Brillouin zones, which are referred as the s- and d-zones, responsible for the conduction. The energy E near the surface of the occupied s-zone is given in terms of the wave number $\mathbf{k}$ as $E(k) = \hbar^2 k^2/2m_1$, while the energy near the surface of the unoccupied d-zone is given by $E(j) = E_0 - (\hbar^2 j^2/2m_2)$. The unoccupied states in the d-band are referred to as positive holes. If the s-electron jumps from a state $\mathbf{k}$ to a state $\mathbf{k}'$, the d-electron jumps from a state $\mathbf{j}$ to a state $\mathbf{j}'$, shown in Fig. 4.1 (a) and (b) . If the s- and d-electrons have the same spin quantum numbers ν before collision, no spin flipping occurs via the collision because of the conservation of angular momentum (Fig. 4.1 (c)). On the other hand, if the s- and d-electrons have the opposite spin quantum numbers, ν and $\bar{\nu}$, before collision, spin flipping occurs via the exchange type interaction [74,75] (Fig. 4.1 (d)).

The present spin flipping rate $(1/\tau_{\nu\bar{\nu}})$ can be evaluated from the carrier scattering probability $\left(W_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'}\right)$ of the Baber-type collision accompanied with spin flipping. If electrons interact via a Coulomb potential V, the scattering probability for a carrier with spin α is given by [76]

$$W^{12}_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'} = W^{21}_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'} = \frac{2\pi}{\hbar}\sum_{\beta\gamma\delta}\left|V(\mathbf{q})\delta_{\alpha\gamma}\delta_{\beta\delta} - V(\mathbf{k}-\mathbf{j}-\mathbf{q})\delta_{\alpha\delta}\delta_{\beta\gamma}\right|^2, \tag{4.1}$$

where indices 1 and 2 denote the electron and hole bands, respectively, indicating that identical scattering probability between the electron and hole. A wave number $\mathbf{q}$ is a momentum transfer defined by $\mathbf{q} = \mathbf{j}' - \mathbf{j}$. The first and second terms under $|...|$ is the direct and exchange interaction, respectively. If $\alpha = \uparrow$ and $\beta = \downarrow$, i.e., a spin-up electron has a collision with a spin-down hole, then $\gamma = \uparrow$ and $\delta = \downarrow$ for the first term while $\gamma = \downarrow$ and $\delta = \uparrow$ for the second term, indicating that no spin flipping occurs via the direct interaction while spin flipping occurs via the exchange interaction.

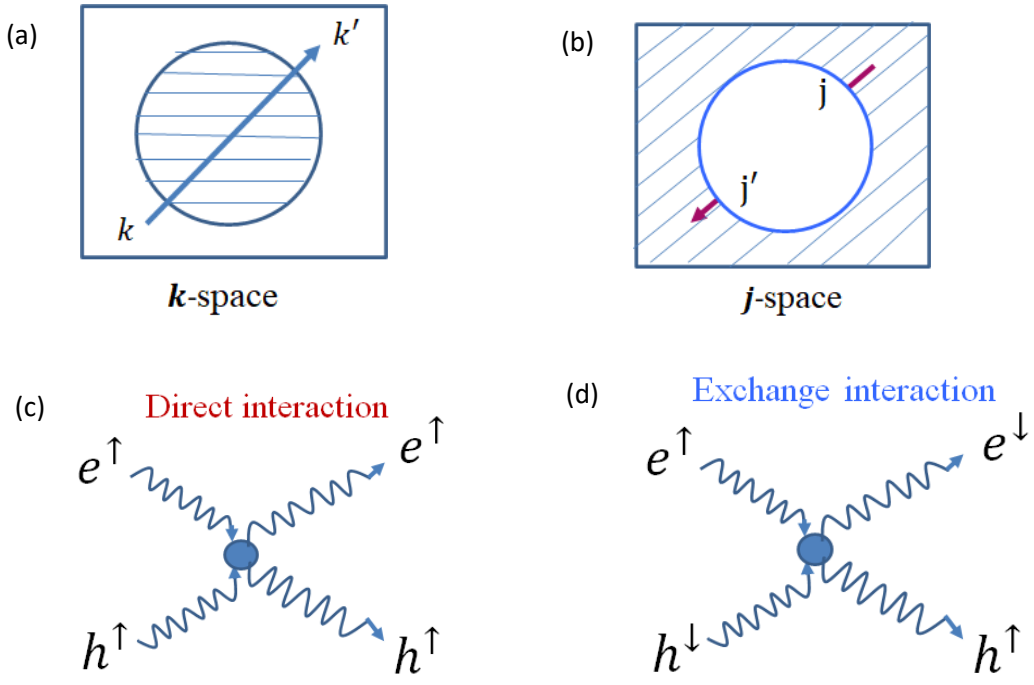

FIG. 4.1. Schematic diagram for ***k*** and ***j*** space (a) and (b). Shaded area indicates occupied states of electrons. (c) and (d) represents two types of collisions between holes and electrons.

We omit the direct interaction term in Eq. (4.1) for evaluation of $1/\tau_{\nu\bar{\nu}}$ because the term does not contribute to spin relaxation, so that we obtain

$$W^{12}_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'} = W^{21}_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'} = \frac{2\pi}{\hbar}\sum_{\beta\gamma\delta}\left|V(\mathbf{k}-\mathbf{j}-\mathbf{q})\delta_{\alpha\delta}\delta_{\beta\gamma}\right|^2 . \tag{4.2}$$

Although the scattering probability due to the exchange interaction depends on the initial momenta **k** and **j** , it is still used for both the electron and hole participating in the collision. Therefore, the spin relaxation time of the electron with spin ν is given by summing over all final wave vectors $\mathbf{k}'$and $\mathbf{j}'$[77]:

$$\frac{1}{\tau^{(e)}_{\nu\bar{\nu}}} = \frac{1}{(2\pi)^3}\int d\mathbf{j}'\big(1-f(\mathbf{j}')\big)\frac{1}{(2\pi)^3}\int d\mathbf{k}'\, W^{12}_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'}\big(1-f(\mathbf{k}')\big), \tag{4.3}$$

where f is the Fermi-Dirac distribution function. Similarly, the spin relaxation time of the hole with spin $\bar{\nu}$ is given by

$$\frac{1}{\tau^{(h)}_{\bar{\nu}\nu}} = \frac{1}{(2\pi)^3}\int d\mathbf{j}'\big(1-f(\mathbf{j}')\big)\frac{1}{(2\pi)^3}\int d\mathbf{k}'\, W^{21}_{\mathbf{kj}\to\mathbf{k}'\mathbf{j}'}\big(1-f(\mathbf{k}')\big). \tag{4.4}$$

It follows from Eqs. (4.1)-(4.4) we obtain

$$\frac{1}{\tau^{(e)}_{\nu\bar{\nu}}} = \frac{1}{\tau^{(h)}_{\bar{\nu}\nu}} \cong (k_BT)^2 N^{(e)}(E_F)N^{(h)}(E_F)W(E_F) \qquad (\nu = \uparrow,\downarrow), \tag{4.5}$$

where $N^{(e)}(E_F)$ and $N^{(h)}(E_F)$ are the electron and hole density of states at the Fermi level (E_F), $W(E_F)$ is the scattering probability at E_F, T is temperature, and k_B is Boltzmann's constant.

We introduce the detailed balance conditions in CMs. The Baber-type collision, where the s-electron jumps from a state **k** to a state $\mathbf{k}'$ in the s-zone and simultaneously the d-electron jumps from a state **j** to a state $\mathbf{j}'$ in the d-zone, does not allow the s-electron to jump to the d-zone and vice versa. In addition, the energy transferred between the electron and hole is an order of k_BT as indicated in Eq. (4.5), which is small by a factor of approximately 10^4 compared to E_F. As a result, the detailed balance conditions for the electron and hole may be separately given as

$$\frac{N^{(e)}_{\uparrow}}{\tau^{(e)}_{\uparrow\downarrow}} = \frac{N^{(e)}_{\downarrow}}{\tau^{(e)}_{\downarrow\uparrow}} \quad , \quad \frac{N^{(h)}_{\uparrow}}{\tau^{(h)}_{\uparrow\downarrow}} = \frac{N^{(h)}_{\downarrow}}{\tau^{(h)}_{\downarrow\uparrow}} \tag{4.6}$$

where $N_\nu^{(e)}$ and $N_\nu^{(h)}$ are respectively the electron and hole density of states at the Fermi level for spin ν. Substitution of Eq. (4.5) into Eq. (4.6) yields

$$\tau_{\uparrow\downarrow}^{(e)} = \frac{N_\downarrow^{(h)}}{N_\uparrow^{(h)}}\tau_{\uparrow\downarrow}^{(h)} \ , \ \tau_{\downarrow\uparrow}^{(e)} = \frac{N_\downarrow^{(e)}}{N_\uparrow^{(e)}}\tau_{\downarrow\uparrow}^{(h)} \tag{4.7}$$

Since our CMs are nonmagnetic, i.e., $N_\uparrow^{(e)} = N_\downarrow^{(e)}$ and $N_\uparrow^{(h)} = N_\downarrow^{(h)}$, we obtain from Eq. (4.7)

$$\tau_{\uparrow\downarrow}^{(e)} = \tau_{\uparrow\downarrow}^{(h)} , \qquad \tau_{\downarrow\uparrow}^{(e)} = \tau_{\downarrow\uparrow}^{(h)} \tag{4.8}$$

as shown in Eqs. (3.21) and (3.22). The identical nature $\left(\text{Eq. (4.8)}\right)$ of spin relaxation times between electrons and holes yields

$$\text{div}\left(\mathbf{J_S^{(h)}} + \mathbf{J_S^{(e)}}\right) = -\hbar\left(\frac{n_\uparrow}{\tau_{\uparrow\downarrow}} - \frac{n_\downarrow}{\tau_{\downarrow\uparrow}}\right) \ , \ \text{div}\left(\mathbf{J_S^{(h)}} - \mathbf{J_S^{(e)}}\right) = -\hbar\Phi\left(\frac{n_\uparrow}{\tau_{\uparrow\downarrow}} - \frac{n_\downarrow}{\tau_{\downarrow\uparrow}}\right) \tag{4.9}$$

where $n_\uparrow$ and $n_\downarrow$ are total spin-up and spin-down carrier densities consisting of the electron and hole, and $\mathbf{J}_s^{(h)}$ and $\mathbf{J_S^{(e)}}$ are electron and hole spin currents (SCs), respectively, given in J/m^2. Eq. (4.9) implies that the spin flipping time $\tau_{\nu\bar{\nu}}$ associated with antiparallel spin current is enhanced by a factor of $1/\Phi$ while that for parallel spin current remains unchanged. In particular, the divergence of antiparallel spin current is zero when charge polarization is zero $(\Phi = 0)$, implying the generation of a steady flow of antiparallel spin current, which is not accompanied with any creation and annihilation of spin angular moment.

We discuss as to how the enhancement of $\tau_{\nu\bar{\nu}}$ can be observed. We cannot observe the enhancement effect under equilibrium condition because the number of spin-up and spin down electrons/holes are equal and spatially homogeneous under equilibrium condition so that no spin relaxation occurs.

Spin injection is one of perturbations, which enables us to observe phenomena relevant to spin relaxation. Usual spin injection causes unbalance between spin-up and spin-down carrier numbers. On the other hand, it follows from an above argument on the Baber-type collision that the numbers of spin-up electrons and spin-down holes, which are injected to CMs, must be equal for achievement of the enhancement mechanism of spin relaxation time based on the electron-hole collision.

The GD relation is used as a useful equation for analyzing chemical potentials not only in Bose and Fermi gases [78,79] but also in 2D Fermi liquid systems [80]. So, in dealing with multicarrier systems such as nearly CMs, the GD relation is the ultimate choice for manipulating the change in chemical potential of the system. Application of the GD relation to spin thermodynamics of ambipolar conductor gives a specific relationship between the spin splitting in the chemical potential of electrons and holes, as [72]

$$\Delta\mu^{(h)} = -\frac{(1-\Phi)}{(1+\Phi)}\Delta\mu^{(e)}, \qquad (\Phi \neq \pm 1) \tag{4.10}$$

where $\Delta\mu^{(h)}$ and $\Delta\mu^{(e)}$ are chemical potential changes caused by both electron and hole spin injection. This equation states that if the chemical potential of a spin-up electron increases then the chemical potential of a spin-down hole increases with the increase ratio of $(1+\Phi)/(1-\Phi)$. This implies that if the density of a spin-up electron increases then the density of a spin-down hole increases such that the number of the injected spin-up electrons is equal to that of the injected spin-down holes, allowing the Baber-type collision with spin flipping. As a result, the enhancement of $\tau_{v\bar{v}}$ is assured when electron and hole spins are simultaneously injected to a nearly CM. In what follows, $(1+\Phi)/(1-\Phi)$ is called the GD factor.

A remarkable difference in spin relaxation time between parallel and antiparallel SCs can be intuitively explained in terms of the total number of a specific spin component. Figure 4.1 shows schematics of (i) $\mathbf{J}_P = 0$, $\mathbf{J}_{AP} \neq 0$ (pure antiparallel SC) and (ii) $\mathbf{J}_P \neq 0$, $\mathbf{J}_{AP} = 0$ (pure parallel SC). In case of pure parallel SC, a region where spin-up hole density is increased is spatially overlapped with that where spin-up electron density is increased. This situation allows the spin-up hole and spin-up electron to jump to spin-down hole and spin-down electron, respectively, via the Baber type collision with spin flipping. As a result, the total number of spin-up component is decreased, implying the presence of spin relaxation. In case of pure antiparallel SC, a region where spin-up hole density is increased is spatially overlapped with that where spin-down electron density is increased. This situation allows the spin-up hole and spin-down electron to jump to spin-down hole and spin-up electron, respectively, via the Baber type collision with spin flipping. Contrary to pure parallel spin current case, the total number of spin-up component remains unchanged, implying that effectively no spin relaxation occurs.

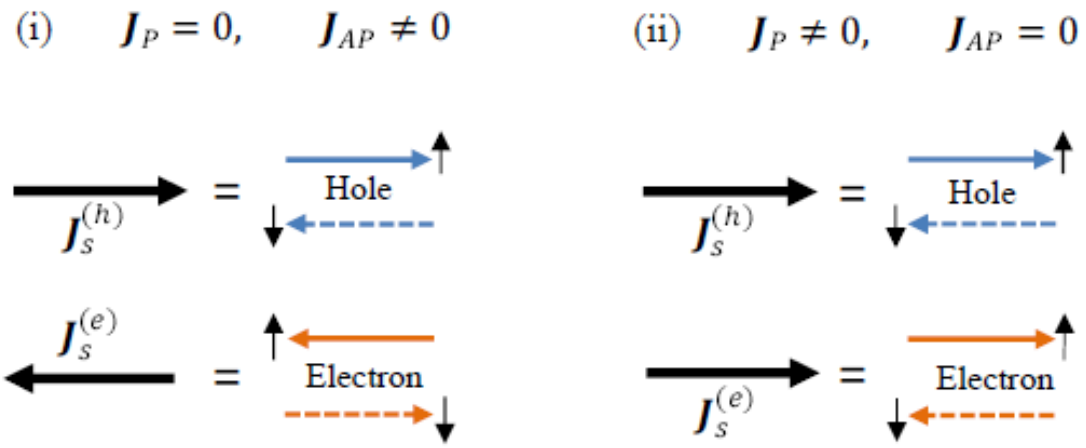

FIG. 4.2. Schematics of (i) pure antiparallel spin current ($\mathbf{J}_P = 0$, $\mathbf{J}_{AP} \neq 0$) and (ii) pure parallel spin current ($\mathbf{J}_P \neq 0$, $\mathbf{J}_{AP} = 0$).

4.2 Quantitative evaluation of spin diffusion length of ambipolar conductors

The difference in spin relaxation time between parallel and antiparallel spin currents affect the spin diffusion lengths associated with the parallel and antiparallel spin currents in ambipolar conductors. The spin diffusion length (SDL) is defined as the length scale over which the travelling electron spin keeps the memory of its initial orientation. It is a characteristic length and a key parameter to describe the spatial decay of the spin polarization which plays an important role for the analysis of the spin transport.

Simultaneous injection of electron and hole spins to the ambipolar conductor produces nonequilibrium spin population at the interface which diffuse through two different types of spin relaxation channel and hence two spin diffusion lengths in ambipolar conductor, one for the parallel spin current (l_P) and another is responsible for antiparallel spin currents (l_{AP}) which can be expressed by the following equations [72]:

$$l_P = \left[\frac{\tau_s}{4e^2}\frac{\sigma_\uparrow^{(h)} + \sigma_\downarrow^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_\uparrow^{(e)} + \sigma_\downarrow^{(e)}\right)}{N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}}\right]^{1/2} \qquad (\Phi \neq \pm 1), \qquad (4.11)$$

$$l_{AP} = \left[\frac{\tau_s}{4e^2}\frac{\sigma_\uparrow^{(h)} + \sigma_\downarrow^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_\uparrow^{(e)} + \sigma_\downarrow^{(e)}\right)}{\Phi\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right)}\right]^{1/2} \qquad (\Phi \neq \pm 1) \qquad (4.12)$$

The spin diffusion length characteristic associated with the parallel and antiparallel spin currents can be explained by the spin relaxation such as the suppression of spin relaxation allow the antiparallel spin current to flow with a large spin diffusion length while keeping the spin diffusion length of the parallel spin current at a standard value due to the enhancement of spin relaxation.

Using Eq. (4.11) and (4.12), we can directly measure the spin diffusion lengths in ambipolar conductors. But due to the scarcity of experimental results on the density of states of holes and electrons at the Fermi level for ambipolar conductors, we perform an indirect assessment of spin diffusion lengths of ambipolar conductor by comparing it with the spin diffusion length of single carrier type paramagnetic metals. The idea is, if the spin relaxation time (τ_s) and the spin diffusion coefficient of the ambipolar metals are approximately the same as those of single-carrier-type paramagnetic metals then the spin diffusion length of the parallel spin current in ambipolar conductor will resemble the spin diffusion length of single carrier paramagnetic metal. In this way, we can measure the spin diffusion length of antiparallel spin current (l_{AP}) by the following equation:

$$l_{AP} = \frac{1}{\sqrt{\Phi}} \times \text{spin diffusion length of paramagnetic metal} \qquad (4.13)$$

This simplified equation tells that l_{AP} in an ambipolar metal has a value larger than the spin diffusion lengths in conventional paramagnetic metals by a factor of $1/\sqrt{\Phi}$ if the above consideration is satisfied.

For quantitative assessment of spin diffusion length of ambipolar conductor such as YH_2 , whose charge polarization value is $(\Phi)\sim 0.15\%$ [81], we consider Silver (Ag) as the single carrier paramagnetic metal. The spin diffusion length of Ag is ~ 162 nm [82]. By substituting the value of charge polarization of YH_2 and spin diffusion length of Ag in our simplified model equation (4.13), l_{AP} of YH_2 is determined to be approximately 4 μm, which probably explains the observation of the anomalous Hall effect in YH_2 with approximately 10 μm channel length [23] because the spin-polarized hole and electron currents are injected from the source and drain magnetic electrodes, respectively, so that an antiparallel flow configuration is achieved.

Another example of large spin diffusion length is in semi-metal Bismuth $(Bi_{0.95}Pb_{0.05})$ [24]. The holes and electrons in $Bi_{0.95}Pb_{0.05}$ have the approximately same density of 3×10^{23} m^{-3} which yields the charge polarization $\Phi \sim 0$. Substitution of $\Phi \sim 0$ into Eq.(4.13) gives an extremely large spin diffusion length, which possibly explains the spin diffusion length of 230 μm observed in $Bi_{0.95}Pb_{0.05}$.

4.3 External control of charge polarization

From Eq. (4.11) and (4.12), we have seen that the most significant difference between the spin diffusion length of parallel and antiparallel spin current is the presence of charge polarization term in antiparallel spin current. And Eq. (4.13) shows the charge polarization plays an important role in making large spin diffusion length of ambipolar conductor. As the ambipolar conductors are characterized by their charge polarization, a large number of compensated metals [39] can be used as ambipolar conductors by varying its number density of holes and electrons to produce very small charge polarization. For doing so, alloying, or solid solutions of one metal in another are favorable. For solid solutions to occur, there are some general requirements which were discussed by Hume-Rothery [83] and suggested that the solid solution range is very restricted if the atomic diameters of solvent and solute differ by more than 15%. By following Hume-Rothery rule, charge polarization can be generated and controlled in compensated metals.

4.4 Summary

We have proposed a mechanism responsible for an extraordinarily large spin coherence length associated with the antiparallel spin current, which is defined as the vector subtraction of hole and electron spin currents in nearly CMs. Its principal reason, identical character between electron and hole spin relaxation times, is a consequence of the exchange interaction between electron and hole subjected to a long-range Coulomb interaction, i.e., the Baber type collision with spin flipping. The GD relation provides the equal density condition between nonequilibrium spin-ν electrons and spin-$\bar{\nu}$ holes, driving the Baber type collision.

CHAPTER 5

SPIN AND CHARGE TRANSPORT IN NEARLY COMPENSATED METALS

A long lifetime as well as a large distance in spin coherence are desirable features for realizing spintronics devices such as magnetic pn junctions [84], spin transistors [85,86], spin logic gates [87-91], and spin qubits for quantum information processing [92]. The nearly CM having an extraordinarily large spin coherence length is a potential candidate for spintronics materials because spatially and temporally large spin coherence make spin manipulation easier. The purpose of this chapter is to theoretically investigate how the enhancement of spin coherence length affects the Hall resistivity and longitudinal resistivity in nearly CMs under simultaneous injection of electron and hole spins.

5.1 Relationships between charge-spin flows and their thermodynamic forces in nearly CM

This section is devoted to a formulation of relationships between charge and spin flows and their thermodynamic forces under simultaneous consideration of electron and hole contributions to charge and SCs. The present formulation consists of (i) introducing two types of SCs, i.e., parallel and antiparallel SCs, (ii) specifying spin dependent particle current densities in terms of the spin dependent chemical potentials each specific to parallel and antiparallel SCs, and (iii) reconfiguration of charge and SCs with consideration of the Onsager reciprocal relations [70,93].

In our previous study[72], we have theoretically characterized $\mathbf{J}_P$ and $\mathbf{J}_{AP}$ in terms of spin splitting of chemical potentials $\Delta\mu_P$ and $\Delta\mu_{AP}$ each specific to $\mathbf{J}_P$ and $\mathbf{J}_{AP}$, respectively, and given by

$$\mathbf{J}_{\boldsymbol{P}} = \frac{\hbar}{2e}\left[\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right]\left(-\frac{1}{e}\right)\text{grad}\,\Delta\mu_{\text{p}} \qquad (\Phi \neq \pm 1)\,, \tag{5.1}$$

$$\mathbf{J}_{\boldsymbol{AP}} = \frac{\hbar}{2e}\left[\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right]\left(-\frac{1}{e}\right)\text{grad}\,\Delta\mu_{\text{Ap}} \qquad (\Phi \neq \pm 1)\,, \tag{5.2}$$

where $\sigma_{\uparrow/\downarrow}^{(h/e)}$ the spin dependent conductivities for hole and electron. The GD factor allows the electron to apparently have the same density as the holes because $(1+\Phi/1-\Phi) = n_h/n_e$. The spatially inhomogeneous quantities $\Delta\mu_{\text{p}}$ and $\Delta\mu_{\text{Ap}}$ are characterized by specific spin coherence lengths l_p and l_{Ap} each given as

$$l_P = \left[\frac{\tau_s}{4e^2}\frac{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)}{N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}}\right]^{1/2} \qquad (\Phi \neq \pm 1), \tag{5.3}$$

$$l_{AP} = \left[\frac{\tau_s}{4e^2}\frac{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)}{\Phi\left(N^{(h)} - \frac{1+\Phi}{1-\Phi}N^{(e)}\right)}\right]^{1/2} \qquad (\Phi \neq \pm 1) \tag{5.4}$$

where $N^{(h/e)} = N_{\uparrow}^{(h/e)}N_{\downarrow}^{(h/e)}/\left(N_{\uparrow}^{(h/e)} + N_{\downarrow}^{(h/e)}\right)$ and $\tau_s = \tau_{\uparrow\downarrow}\tau_{\downarrow\uparrow}/(\tau_{\uparrow\downarrow} + \tau_{\downarrow\uparrow})$. In deriving Eqs. (5.1)-(5.4), we used the identical nature of spin relaxation times between electrons and holes such as $\tau_{\uparrow\downarrow}^{(e)} = \tau_{\uparrow\downarrow}^{(h)}$, $\tau_{\downarrow\uparrow}^{(e)} = \tau_{\downarrow\uparrow}^{(h)}$. It follows from Eqs. (5.3) and (5.4) that the reduced spin relaxation time τ_s associated with $\mathbf{J}_{\boldsymbol{AP}}$ is enhanced by a factor of $1/\Phi$ while that for $\mathbf{J}_{\boldsymbol{P}}$ remains unchanged. The definition of $\mathbf{J}_{\boldsymbol{P}}$ and $\mathbf{J}_{\boldsymbol{AP}}$ allows us to write hole and electron contributions to SCs in terms of $\Delta\mu_{\text{p}}$ and $\Delta\mu_{\text{Ap}}$

$$\begin{aligned}\mathbf{J}_{\text{s}}^{(h)} = \left(-\frac{\hbar}{4e^2}\right)\Bigg[&\left\{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right\}\text{grad}\,\Delta\mu_{\text{p}} \\ &+ \left\{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right\}\text{grad}\,\Delta\mu_{\text{Ap}}\Bigg]\end{aligned} \tag{5.5}$$

$$\mathbf{J}_{\mathrm{s}}^{(e)} = \left(-\frac{\hbar}{4e^2}\right)\left[\left\{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right\}\mathrm{grad}\,\Delta\mu_{\mathrm{p}} - \left\{\sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\right\}\mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right], \quad (5.6)$$

Equations (5.5) and (5.6) indicate that electron and hole SCs are no longer characterized in terms only of their original characteristics, instead, they are mutually influenced owing to the GD relation, which thermodynamically constrains the chemical potentials of constituents in nearly CMs.

Next mission is to express individual particle current densities in terms of $\Delta\mu_{\mathrm{p}}$and $\Delta\mu_{\mathrm{Ap}}$. This mission is later required for expressing the total charge current and SCs both given in A/m^2, e.g., $\mathbf{J}_C = e\mathbf{j}_{\uparrow}^{(h)} + e\mathbf{j}_{\downarrow}^{(h)} - e\mathbf{j}_{\uparrow}^{(e)} - e\mathbf{j}_{\downarrow}^{(h)}$ and $\mathbf{J}_S = e\mathbf{j}_{\uparrow}^{(h)} - e\mathbf{j}_{\downarrow}^{(h)} - (e\mathbf{j}_{\uparrow}^{(e)} - e\mathbf{j}_{\downarrow}^{(e)})$, the latter of which is the expression corresponding to $\mathbf{J}_P$ given in J/m^2. Although the hole and electron spin currents $\mathbf{J}_S^{(h)}$and $\mathbf{J}_S^{(e)}$ which are related to particle current densities as $\mathbf{J}_S^{(h)} = (\hbar/2)\mathbf{j}_{\uparrow}^{(h)} - (\hbar/2)\mathbf{j}_{\downarrow}^{(h)}$ and $\mathbf{J}_S^{(e)} = (\hbar/2)\mathbf{j}_{\uparrow}^{(e)} - (\hbar/2)\mathbf{j}_{\downarrow}^{(e)}$, respectively, are given in terms of $\Delta\mu_P$ and $\Delta\mu_{AP}$, no formulae corresponding to $(\hbar/2)\mathbf{j}_{\uparrow}^{(h)} + (\hbar/2)\mathbf{j}_{\downarrow}^{(h)}$ and $(\hbar/2)\mathbf{j}_{\uparrow}^{(e)} + (\hbar/2)\mathbf{j}_{\downarrow}^{(e)}$are known in terms of $\Delta\mu_P$ and $\Delta\mu_{AP}$, so that we cannot a priori obtain an unique expression for $\mathbf{j}_{\uparrow}^{(h)}$, $\mathbf{j}_{\downarrow}^{(h)}$, $\mathbf{j}_{\uparrow}^{(e)}$, and $\mathbf{j}_{\downarrow}^{(e)}$. Instead, we seek their expressions in an ad hoc manner such that a special condition, $\Delta\mu_P = \Delta\mu_{AP}$ yields a unique set of expressions. Therefore, we obtain:

$$\mathbf{j}_{\uparrow}^{(h)} = -\frac{1}{2e^2}\left[\left(\sigma_{\uparrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{p}} + \left(\sigma_{\uparrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right], \quad (5.7)$$

$$\mathbf{j}_{\downarrow}^{(h)} = \frac{1}{2e^2}\left[\left(\sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{p}} + \left(\sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right], \quad (5.8)$$

$$\mathbf{j}_{\uparrow}^{(e)} = -\frac{1}{2e^2}\left[\left(\sigma_{\uparrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{p}} - \left(\sigma_{\uparrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right], \quad (5.9)$$

$$\mathbf{j}_{\downarrow}^{(e)} = \frac{1}{2e^2}\left[\left(\sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{p}} - \left(\sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right]. \quad (5.10)$$

Since substituting of $\Delta\mu_P = \Delta\mu_{AP}$ into Eqs. (5.7)−(5.10) yields the original expressions, which are mutually independent, our ad hoc derivation is naturally acceptable.

If we closely examine Eqs. (5.7)-(5.10), we see that the spin dependent particle current densities of holes and electrons are lacking for drift current term, which is essential for generation of spin Hall effect as well as anomalous Hall effect. So, we add drift current terms along with diffusion terms in the following way

$$\mathbf{j}_{\uparrow}^{(h)} = -\frac{1}{2e^2}\left\{\begin{array}{l}\left(\sigma_{\uparrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\left(e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{p}}\right) \\ +\left(\sigma_{\uparrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\left(e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right)\end{array}\right\}\ , \tag{5.11}$$

$$\mathbf{j}_{\downarrow}^{(h)} = \frac{1}{2e^2}\left\{\begin{array}{l}\left(\sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\left(-e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{p}}\right) \\ +\left(\sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\left(-e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right)\end{array}\right\}\ , \tag{5.12}$$

$$\mathbf{j}_{\uparrow}^{(e)} = -\frac{1}{2e^2}\left\{\begin{array}{l}\left(\sigma_{\uparrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\left(e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{p}}\right) \\ -\left(\sigma_{\uparrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\uparrow}^{(e)}\right)\left(e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right)\end{array}\right\}\ , \tag{5.13}$$

$$\mathbf{j}_{\downarrow}^{(e)} = \frac{1}{2e^2}\left\{\begin{array}{l}\left(\sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\left(-e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{p}}\right) \\ -\left(\sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\sigma_{\downarrow}^{(e)}\right)\left(-e\,\mathrm{grad}\,V + \mathrm{grad}\,\Delta\mu_{\mathrm{Ap}}\right)\end{array}\right\}\ . \tag{5.14}$$

When adding the drift current terms, we still considered the GD factor for electric potential term V such that the Onsager reciprocal relations are still retained.

Substitution of Eqs. (5.11)-(5.14) into the definitions of $\mathbf{J}_C$ and $\mathbf{J}_S$ yields four types of current densities as

$$\mathbf{J}_{\mathrm{C}} = -\bar{a}\,\mathrm{grad}\,V - \frac{1}{e}\bar{d}\,\mathrm{grad}\,\Delta\mu_{\mathrm{AP}}\ , \tag{5.15}$$

$$\mathbf{J}_{\mathrm{C}}^{*} = -\bar{c}\ \mathrm{grad}\ V - \frac{1}{e}\bar{b}\ \mathrm{grad}\ \Delta\mu_{\mathrm{P}}\ , \tag{5.16}$$

$$\mathbf{J}_{\mathrm{S}} = -\bar{b}\ \mathrm{grad}\ V - \frac{1}{e}\bar{c}\ \mathrm{grad}\ \Delta\mu_{\mathrm{AP}}\ , \tag{5.17}$$

$$\mathbf{J}_{\mathrm{S}}^{*} = -\bar{d}\ \ \mathrm{grad}\ V - \frac{1}{e}\bar{a}\ \mathrm{grad}\ \Delta\mu_{\mathrm{P}}\ \ . \tag{5.18}$$

In the above equations, $\mathbf{J}_{\mathrm{c}}^{*}$ and $\mathbf{J}_{\mathrm{s}}^{*}$ are the conjugate of charge and spin current densities, the definitions of which are given as $\mathbf{J}_{c}^{*} = e\mathbf{j}_{\uparrow}^{(h)} + e\mathbf{j}_{\downarrow}^{(h)} + e\mathbf{j}_{\uparrow}^{(e)} + e\mathbf{j}_{\downarrow}^{(h)}$ and $\mathbf{J}_{s}^{*} = e\mathbf{j}_{\uparrow}^{(h)} - e\mathbf{j}_{\downarrow}^{(h)} + e\mathbf{j}_{\uparrow}^{(e)} - e\mathbf{j}_{\downarrow}^{(h)}$, respectively. $\mathbf{J}_{\mathrm{s}}^{*}$ is given in A/m^2 and corresponds to $\mathbf{J}_{AP}$ given in J/m^2. Matrices $\bar{a}$, $\bar{b}$, $\bar{c}$ and $\bar{d}$ have been written as the shorthand notation for the relations below

$$\bar{a} = \sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\ , \tag{5.19}$$

$$\bar{b} = \sigma_{\uparrow}^{(h)} - \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} - \sigma_{\downarrow}^{(e)}\right)\ \ , \tag{5.20}$$

$$\bar{c} = \sigma_{\uparrow}^{(h)} + \sigma_{\downarrow}^{(h)} - \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} + \sigma_{\downarrow}^{(e)}\right)\ \ , \tag{5.21}$$

$$\bar{d} = \sigma_{\uparrow}^{(h)} - \sigma_{\downarrow}^{(h)} + \frac{1+\Phi}{1-\Phi}\left(\sigma_{\uparrow}^{(e)} - \sigma_{\downarrow}^{(e)}\right)\ \ , \tag{5.22}$$

It follows from Eqs. (5.15) and (5.18) that the Onsager reciprocal relations are retained between $\mathbf{J}_{c}$ and $\mathbf{J}_{\mathrm{s}}^{*}$; the $\mathbf{J}_{c}$ per unit of chemical potential difference and the $\mathbf{J}_{\mathrm{S}}^{*}$ per unit of electric potential difference are equal when both chemical and electric potential vary. Similarly, it follows from Eqs. (5.16) and (5.17) that the Onsager reciprocal relations are retained between $\mathbf{J}_{C}^{*}$ and $\mathbf{J}_{S}$. Again, we address that the GD factor effectively makes nearly CMs complete CMs with exactly equal carrier density between electrons and holes.

5.2 Derivation of diagonal and off-diagonal parts of resistivity

It follows from Sec.5.1 that $\Delta\mu_{AP}$, the spin dependent chemical potential specific to $\mathbf{J}_{AP}$, plays an important role in $\mathbf{J}_C$. This section is devoted to derivations of longitudinal resistivity ρ_{xx} and Hall resistivity ρ_{yx} using the charge and spin currents reconfigured in terms of V and $\Delta\mu_{AP}$.

Again, from Eq. (5.15) we see that $\mathbf{J}_C$ is composed of electric field $\mathbf{E}$ ($=-\mathrm{grad}\,V$) and effective field $\Delta\mathbf{E}_{\mathrm{AP}}$ ($=-(1/e)\mathrm{grad}\Delta\mu_{AP}$) . Since ρ_{xx} and ρ_{yx} are the coefficients relating between $\mathbf{J}_C$ and $\mathbf{E}$, i.e., $E_x = \rho_{xx}J_{\mathrm{c,x}} + \rho_{xy}J_{\mathrm{c,y}}$ and $E_y = \rho_{yx}J_{c,x} + \rho_{yy}J_{c,y}$, it is required to effectively include $\Delta\mathbf{E}_{\mathrm{AP}}$ into these relations. For this purpose, we introduce phenomenological variables x_{AP} and y_{AP} defined as $\Delta E_{AP,x} = x_{AP}E_x$ and $\Delta E_{AP,y} = y_{AP}E_y$. Quantities x_{AP} and y_{AP} cannot be a prior determined but are empirically determined from observed values of ρ_{xx} and ρ_{yx}. Therefore, we obtain from Eq. (5.15)

$$J_{\mathrm{c},x} = \left(\bar{a}'_{xx} + \bar{d}'_{xx}x_{AP}\right)E_x + \left(\bar{a}'_{xy} + \bar{d}'_{xy}y_{AP}\right)E_y\,, \tag{5.23}$$

$$J_{\mathrm{c},y} = -\left(\bar{a}'_{xy} + \bar{d}'_{xy}x_{AP}\right)E_x + \left(\bar{a}'_{xx} + \bar{d}'_{xx}y_{AP}\right)E_y\,. \tag{5.24}$$

Conventional Hall resistivity measurement as well as our measurements are performed under transverse current free condition $J_{\mathrm{c,y}} = 0$, while a bias current is injected from a source electrode and is ejected from an ambipolar conductor at a drain electrode. The source and drain electrodes must be specific materials such that the bias current be spin polarized. Substituting $J_{\mathrm{c,y}} = 0$ into Eqs. (5.23) and (5.24) yields

$$\binom{1}{0} = \begin{pmatrix} \bar{a}'_{xx} + \bar{d}'_{xx}x_{\mathrm{AP}} & \bar{a}'_{xy} + \bar{d}'_{xy}y_{\mathrm{AP}} \\ -\bar{a}'_{xy} - \bar{d}'_{xy}x_{\mathrm{AP}} & \bar{a}'_{xx} + \bar{d}'_{xx}y_{\mathrm{AP}} \end{pmatrix}\binom{\rho_{xx}}{\rho_{yx}}\,, \tag{5.25}$$

from which we obtain

$$\rho_{xx} = \frac{\bar{a}'_{xx} + \bar{d}'_{xx}y_{\mathrm{AP}}}{\left(\bar{a}'_{xx} + \bar{d}'_{xx}x_{\mathrm{AP}}\right)\left(\bar{a}'_{xx} + \bar{d}'_{xx}y_{\mathrm{AP}}\right) + \left(\bar{a}'_{xy} + \bar{d}'_{xy}x_{\mathrm{AP}}\right)\left(\bar{a}'_{xy} + \bar{d}'_{xy}y_{\mathrm{AP}}\right)} \tag{5.26}$$

$$\rho_{yx} = \frac{\bar{a}'_{xy} + \bar{d}'_{xy} x_{\mathrm{AP}}}{\left(\bar{a}'_{xx} + \bar{d}'_{xx} x_{\mathrm{AP}}\right)\left(\bar{a}'_{xx} + \bar{d}'_{xx} y_{\mathrm{AP}}\right) + \left(\bar{a}'_{xy} + \bar{d}'_{xy} x_{\mathrm{AP}}\right)\left(\bar{a}'_{xy} + \bar{d}'_{xy} y_{\mathrm{AP}}\right)} \tag{5.27}$$

Equations (5.26) and (5.27) are general expressions independent of microscopic mechanism of the spin dependent conductivities. They yield conventional formulae of ρ_{xx} and ρ_{yx} when $x_{AP} = 0$ and $y_{AP} = 0$. Supposed $\bar{a}$ and $\bar{d}$ are given, it follows from Eqs. (5.26) and (5.27) that x_{AP} and y_{AP} can be determined from observed values of ρ_{xx} and ρ_{yx}.

For further assessment of the Hall effect, we need the diagonal and off-diagonal components of $\bar{a}$ and $\bar{d}$ in terms of spin dependent electrical conductivities of holes and electrons, which were derived from the Drude model with hole density (n_h), electron densities (n_e), hole mobility (μ_h), electron mobility (μ_e), effective magnetic fields due to spin orbit interactions (SOI) for hole and electron (S_h and S_e) [89,91] are as follows:

$$\bar{\sigma}_{\uparrow}^{(h)} = \frac{e n_{\uparrow}^{(h)} \mu_h}{1 + \mu_h^2 (B - S_h)^2} \begin{pmatrix} 1 & \mu_h (B - S_h) & 0 \\ -\mu_h (B - S_h) & 1 & 0 \\ 0 & 0 & 1 + \mu_h^2 (B - S_h)^2 \end{pmatrix}, \tag{5.28}$$

$$\bar{\sigma}_{\downarrow}^{(h)} = \frac{e n_{\downarrow}^{(h)} \mu_h}{1 + \mu_h^2 (B + S_h)^2} \begin{pmatrix} 1 & \mu_h (B + S_h) & 0 \\ -\mu_h (B + S_h) & 1 & 0 \\ 0 & 0 & 1 + \mu_h^2 (B + S_h)^2 \end{pmatrix}, \tag{5.29}$$

$$\bar{\sigma}_{\uparrow}^{(e)} = -\frac{e n_{\uparrow}^{(e)} \mu_e}{1 + \mu_e^2 (B - S_e)^2} \begin{pmatrix} 1 & \mu_e (B - S_e) & 0 \\ -\mu_e (B - S_e) & 1 & 0 \\ 0 & 0 & 1 + \mu_e^2 (B - S_e)^2 \end{pmatrix}, \tag{5.30}$$

$$\bar{\sigma}_{\downarrow}^{(e)} = -\frac{e n_{\downarrow}^{(e)} \mu_e}{1 + \mu_e^2 (B + S_e)^2} \begin{pmatrix} 1 & \mu_e (B + S_e) & 0 \\ -\mu_e (B + S_e) & 1 & 0 \\ 0 & 0 & 1 + \mu_e^2 (B + S_e)^2 \end{pmatrix}. \tag{5.31}$$

In the present study, two type of cases are considered: (case I) low mobility and/or weak SOI case: $\mu_h^2 S_h^2 \ll 1$, $\mu_e^2 S_e^2 \ll 1$ and (case II) high mobility and/or strong SOI case: $\mu_h^2 S_h^2 \gg 1$,

$\mu_e^2 S_e^2 \gg 1$. A weak external magnetic field, the magnitude of which is significantly smaller than S_h and S_e, is considered for both cases.

The scattering probability given by Eq. (4.2) has a symmetric character showing that the probability from $(\mathbf{k}, \mathbf{j})$ to $(\mathbf{k}', \mathbf{j}')$ is equal to that from $(\mathbf{k}', \mathbf{j}')$ to $(\mathbf{k}, \mathbf{j})$. Therefore, the Baber type transition probability cannot play a role similar to SOI with an asymmetric transition characteristic [94]. Since SOI includes spin flipping process [95], we have to assume that SOI used in the present assessment of Hall resistivity has a less contribution to spin relaxation in nearly CMs compared to the Baber-type collision given by Eq. (4.2).

Case I condition, $\mu_h^2 S_h^2 \ll 1$ and $\mu_e^2 S_e^2 \ll 1$, gives $\bar{a}_{xx} \cong en(\mu_h + |\mu_e|)$; $\bar{a}_{xy} \cong -en(\mu_e^2 S_e - \mu_h^2 S_h)P$; $\bar{d}_{xx} \cong en(\mu_h + |\mu_e|)P$; $\bar{d}_{xy} \cong -en(\mu_e^2 S_e - \mu_h^2 S_h)$, where the GD factor $(1+\Phi)/(1-\Phi)$, in Eqs. (5.15) and (5.18) is renormalized into electron density such that n is newly defined as $n = (1+\Phi)/(1-\Phi)n_e + n_h$, where n_e and n_h are intrinsic electron and hole densities, respectively. Quantity *P* is an equilibrium paramagnetic spin polarization of electron and hole before spin injection: we assume equal spin polarization between electron and hole. Substituting these conditions into Eqs. (5.26) and (5.27) yields approximate expressions

$$\rho_{xx} = \frac{(\mu_h + |\mu_e|)(1 + Py_{\mathrm{AP}})}{en\{(\mu_h + |\mu_e|)^2(1 + Px_{\mathrm{AP}})(1 + Py_{\mathrm{AP}}) + (\mu_h^2 S_h - \mu_e^2 S_e)^2(P + x_{\mathrm{AP}})(P + y_{\mathrm{AP}})\}}, \tag{5.32}$$

$$\rho_{yx} = -\frac{(\mu_h^2 S_h - \mu_e^2 S_e)(P + x_{\mathrm{AP}})}{en\{(\mu_h + |\mu_e|)^2(1 + Px_{\mathrm{AP}})(1 + Py_{\mathrm{AP}}) + (\mu_h^2 S_h - \mu_e^2 S_e)^2(P + x_{\mathrm{AP}})(P + y_{\mathrm{AP}})\}} \tag{5.33}$$

Provided that $P \ll x_{AP} < 1$ and $P \ll y_{AP} < 1$, we obtain from Eqs. (5.32) and (5.33)

$$\rho_{xx} = \frac{\mu_h + |\mu_e|}{en(\mu_h + |\mu_e|)^2 + en(\mu_h^2 S_h - \mu_e^2 S_e)^2 x_{\mathrm{AP}} y_{\mathrm{AP}}} \tag{5.34}$$

$$\rho_{yx} = -\frac{(\mu_h^2 S_h - \mu_e^2 S_e) x_{\mathrm{AP}}}{en(\mu_h + |\mu_e|)^2 + en(\mu_h^2 S_h - \mu_e^2 S_e)^2 x_{\mathrm{AP}} y_{\mathrm{AP}}} \tag{5.35}$$

After substitution of the definitions of x_{AP} and y_{AP} and several algebraic manipulations, we obtain closed formulae as

$$\rho_{xx} = \frac{1}{en(\mu_h + |\mu_e|)} + \left(\frac{\mu_h^2 S_h - \mu_e^2 S_e}{\mu_h + |\mu_e|}\right)\frac{\Delta E_{AP,y}}{J_{c,x}} \tag{5.36}$$

$$\rho_{yx} = \left(\frac{\mu_e^2 S_e - \mu_h^2 S_h}{\mu_h + |\mu_e|}\right)\frac{\Delta E_{AP,x}}{J_{c,x}} \tag{5.37}$$

When $n, \mu_e, \mu_h, S_e, S_h$ and $J_{c,x}$ are given , $\Delta E_{AP,x}$ and $\Delta E_{AP,y}$ can be assessed from observed values of ρ_{yx} and ρ_{xx}, respectively.

Case II condition, $\mu_h^2 S_h^2 \gg 1$ and $\mu_e^2 S_e^2 \gg 1$, gives $\bar{a}_{xx} \cong en[1/(\mu_h S_h^2) + 1/(|\mu_e| S_e^2)]$; $\bar{a}_{xy} \cong en(1/S_e - 1/S_h)P$; $\bar{d}_{xx} \cong en[1/(\mu_h S_h^2) + 1/(|\mu_e| S_e^2)]P$; $\bar{d}_{xy} \cong en(1/S_e - 1/S_h)$ Substituting these conditions into Eqs. (5.26) and (5.27) yields approximate expressions

$$\rho_{xx} = \frac{en\left(\frac{1}{\mu_h S_h^2} + \frac{1}{|\mu_e| S_e^2}\right)(1 + P y_{\mathrm{AP}})}{en\left(\frac{1}{\mu_h S_h^2} + \frac{1}{|\mu_e| S_e^2}\right)^2 (1 + P x_{\mathrm{AP}})(1 + P y_{\mathrm{AP}}) + en\left(\frac{1}{S_e} - \frac{1}{S_h}\right)^2 (P + x_{\mathrm{AP}})(P + y_{\mathrm{AP}})}, \tag{5.38}$$

$$\rho_{yx} = \frac{\left(\frac{1}{S_e} - \frac{1}{S_h}\right)(P + x_{\mathrm{AP}})}{en\left(\frac{1}{\mu_h S_h^2} + \frac{1}{|\mu_e| S_e^2}\right)^2 (1 + P x_{\mathrm{AP}})(1 + P y_{\mathrm{AP}}) + en\left(\frac{1}{S_e} - \frac{1}{S_h}\right)^2 (P + x_{\mathrm{AP}})(P + y_{\mathrm{AP}})}. \tag{5.39}$$

Provided that $P \ll x_{AP} < 1$ and $P \ll y_{AP} < 1$, we obtain closed formulae from Eqs. (5.38) and (5.39)

$$\rho_{xx} = \frac{1}{en[1/(\mu_h S_h^2) + 1/(\mu_e S_e^2)]} + \left[\frac{1/S_h - 1/S_e}{1/(\mu_h S_h^2) + 1/(\mu_e S_e^2)}\right]\frac{\Delta E_{AP,y}}{J_{c,x}}, \tag{5.40}$$

$$\rho_{yx} = -\left[\frac{1/S_h - 1/S_e}{1/(\mu_h S_h^2) + 1/(\mu_e S_e^2)}\right]\frac{\Delta E_{AP,x}}{J_{c,x}} \tag{5.41}$$

Therefore, $\Delta E_{AP,x}$ and $\Delta E_{AP,y}$ can be assessed from observed values of ρ_{xx} and ρ_{yx}, respectively.

It follows from Eqs. (5.36), (5.37), (5.40), and (5.41) that spin dependent field ΔE_{AP} influences ρ_{xx} and ρ_{yx} in the same way between cases I and II, indicating that ρ_{xx} is affected by $\Delta E_{AP,y}$ while ρ_{yx} is affected by $\Delta E_{AP,x}$.

Qualitative assessment of ρ_{yx} is given in Fig. 5.1. When assessing ρ_{yx}, we assumed that $\Delta E_{AP,x}$ is in proportion to the carrier spin polarization of spin injector used as source and drain electrodes of the Hall device. On the other hand, $\Delta E_{AP,y}$ is probably insensitive to magnetic fields when the voltage detection electrodes used in the Hall device are nonmagnetic metals. Therefore, ρ_{xx} hardly depends on magnetic fields. A schematic diagram of Hall bar device is shown in Fig. 5.2. When hole and electron SCs are injected from magnetic source and drain electrodes, respectively, the antiparallel SC flows from the source towards the drain electrodes. This configuration allows us to approximately express $\Delta\mu_{AP}$ in terms of spin diffusion length l as $\Delta\mu_{AP} \propto \exp(-x/l) - \exp(x - L/l)$[95], where L is current channel length in the Hall device. Therefore, spin-dependent field $\Delta E_{AP,x}$ is given as $\Delta E_{AP,x} \propto (1/l)[\exp(-x/l) + \exp(x - L/l)]$.Since $\Delta\mu_{AP}$, and hence, $\Delta E_{AP,x}$ is in proportion with the magnetization of the source and drain electrodes, ρ_{yx} given by Eq. (5.41) is also in proportion with the magnetization of the source and drain electrodes, as schematically shown in Fig. 5.1.

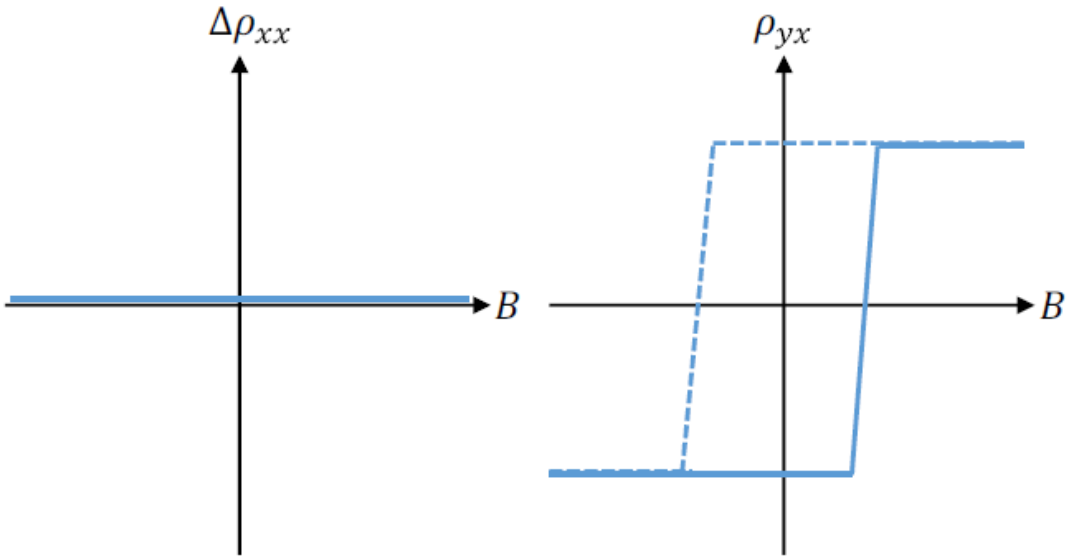

Fig.5.1 Qualitative assessments of magnetic field dependences of $\Delta\rho_{xx}$, which is given by Eq. (5.36) (low mobility and/or weak SOI case) and Eq. (5.40) (high mobility and/or strong SOI case), and ρ_{yx} given by Eq. (5.37) (low mobility and/or weak SOI case) and Eq. (5.41) (high mobility and/or strong SOI case) under assumption of $\Delta E_{AP,y} = 0$. $\Delta\rho_{xx}$ is defined as $\Delta\rho_{xx} = \rho_{xx} - \rho(B = 0)$. Solid and dashed lines indicate field-upsweep and field-downsweep, respectively.

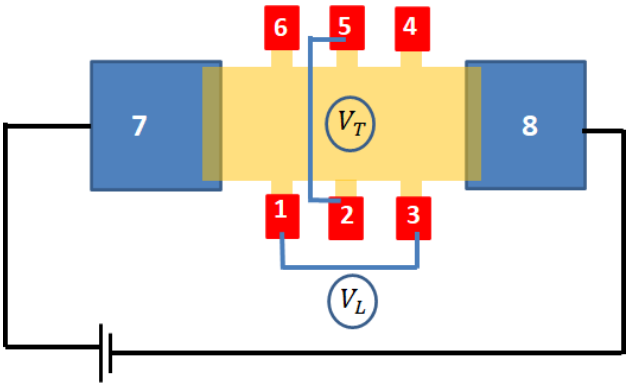

FIG. 5.2. A Hall bar device which contains six voltage detection electrodes, labeled as 1 to 6. 7 and 8 represents two ferromagnetic electrodes act as source and drain respectively, including a channel made with ambipolar conductor in between. V_T and V_L characterized as transverse and longitudinal voltage.

5.3 Mechanism of resonance Hall effect

An intriguing implication of the present Hall effect can be found out as we closely examine Eqs. (5.24) and (5.27). In the absence of spin dependent chemical potential, $x_{AP} = 0$ and $y_{AP} = 0$, condition $J_{c,y} = 0$ yields $\rho_{yx} = 0$ when $\bar{a}_{xy} = 0$ because $\bar{a}_{xx} \neq 0$.On the other hand, the presence of spin dependent chemical potential, $x_{AP} \neq 0$ and $y_{AP} \neq 0$, gives rise to a different aspect. Provided $\bar{a}_{xy} + \bar{d}_{xy}x_{AP} = 0$, it follows from Eq. (5.24) that condition $J_{c,y} = 0$ yields $\bar{a}_{xx} + \bar{d}_{xx}y_{AP} = 0$, indicating that both numerator and denominator of Eq. (5.27) are zero, so we cannot definitely infer $\rho_{yx} = 0$. This aspect is essentially different from $x_{AP} = 0$ and $y_{AP} = 0$ case, and is named a resonance Hall effect, the mechanism of which will be described below. The mechanism is essentially different from the enhancement mechanism of Hall effect signal in two-dimensional electron gas with Rashba spin orbit interaction (SOI) [96] and Dresselhaus SOI [97].

It is $\mathbf{J}_S^*$ that satisfies the Onsager reciprocal relation in combination with $\mathbf{J}_c$. Similarly, Eqs. (5.23) and (5.24), we obtain from Eq. (5.18)

$$J_{S,x}^* = \left(\bar{d}'_{xx} + \bar{a}'_{xx}x_{AP}\right)E_x + \left(\bar{d}'_{xy} + \bar{a}'_{xy}y_{AP}\right)E_y\,, \tag{5.42}$$

$$J_{S,y}^* = -\left(\bar{d}'_{xy} + \bar{a}'_{xy}x_{AP}\right)E_x + \left(\bar{d}'_{xx} + \bar{a}'_{xx}y_{AP}\right)E_y\,. \tag{5.43}$$

Elimination of **E** from Eqs. (5.23), (5.24), (5.42), and (5.43) yields

$$\mathbf{J}_c = \begin{pmatrix} \bar{a}_{xx} + \bar{d}_{xx}x_{AP} & \bar{a}_{xy} + \bar{d}_{xy}y_{AP} \\ -\bar{a}_{xy} - \bar{d}_{xy}x_{AP} & \bar{a}_{xx} + \bar{d}_{xx}y_{AP} \end{pmatrix} \begin{pmatrix} \bar{d}_{xx} + \bar{a}_{xx}x_{AP} & \bar{d}_{xy} + \bar{a}_{xy}y_{AP} \\ -\bar{d}_{xy} - \bar{a}_{xy}x_{AP} & \bar{d}_{xx} + \bar{a}_{xx}x_{AP} \end{pmatrix}^{-1} \mathbf{J}_{\mathrm{s}}^*. \tag{5.44}$$

The generation condition of self-sustaining mode of $\mathbf{J}_{\mathrm{s}}^*$, which can flow under completely current free condition $\mathbf{J}_c = 0$, is obtained from Eq. (5.44) as

$$\left(\bar{a}_{xx}+\bar{d}_{xx}x_{\mathrm{AP}}\right)\left(\bar{a}_{xx}+\bar{d}_{xx}y_{\mathrm{AP}}\right)+\left(\bar{a}_{xy}+\bar{d}_{xy}x_{\mathrm{AP}}\right)\left(\bar{a}_{xy}+\bar{d}_{xy}y_{\mathrm{AP}}\right)=0\ . \tag{5.45}$$

which makes the denominator of Eqs. (5.26) and (5.27) zero. The resonance conditions, $\bar{a}_{xy}+\bar{d}_{xy}x_{\mathrm{AP}}=0$ and $\bar{a}_{xx}+\bar{d}_{xx}y_{\mathrm{AP}}=0$, satisfies Eq. (5.45), indicating that the present resonance Hall effect may be accompanied with a self-sustaining mode of $\mathbf{J}_{\mathrm{s}}^{*}$. Condition $\bar{a}_{xy}+\bar{d}_{xy}x_{\mathrm{AP}}=0$ is a necessary and sufficient for the resonant Hall effect because the other condition $\bar{a}_{xx}+\bar{d}_{xx}y_{\mathrm{AP}}=0$ is derived when substituting $J_{c,y}=0$ into Eq. (5.24).

We cannot use Eqs. (5.26) and (5.27) for the resonance Hall effect because their denominator is zero. Instead we start with Eqs. (5.23) and (5.24). Provided $\bar{a}_{xy}=0$ and $\bar{d}_{xy}=0$, the necessary and sufficient condition for the resonant Hall effect is assured for an arbitral value of x_{AP}. Therefore, we obtain from Eqs. (5.23) and (5.24) using $J_{c,y}=0$

$$J_{c,x}=\left(\bar{a}_{xx}+\bar{d}_{xx}x_{\mathrm{AP}}\right)E_x\,,\qquad \left(\bar{a}_{xx}+\bar{d}_{xx}y_{\mathrm{AP}}\right)E_y\ =0. \tag{5.46}$$

If we still use $E_x\,/J_{c,x}$ and $E_y\,/J_{c,x}$ for the definition of ρ_{xx} and ρ_{yx}, respectively, then we obtain from Eq. (5.46)

$$\rho_{xx}=\frac{1}{\bar{a}_{xx}}-\left(\frac{\bar{d}_{xx}}{\bar{a}_{xx}}\right)\frac{\Delta E_{AP,x}}{J_{c,x}}\,, \tag{5.47}$$

$$\rho_{yx}=-\left(\frac{\bar{d}_{xx}}{\bar{a}_{xx}}\right)\frac{\Delta E_{AP,y}}{J_{c,x}} \tag{5.48}$$

For further assessment of the resonance mechanism of Hall effect, we again consider case I ($\mu_h^2S_h^2\ll 1,\mu_e^2S_e^2\ll 1$) and case II ($\mu_h^2S_h^2\gg 1,\mu_e^2S_e^2\gg 1$) . A weak external magnetic field, the magnitude of which is significantly smaller than S_h and S_e, is indispensable for both cases because paramagnetic spin polarization (P) plays an important role in the resonance mechanism.

Case I enables us to find out that a condition, $\mu_h^2S_h=\mu_e^2S_e$, gives the necessary and sufficient condition for the resonant Hall effect, $\bar{a}_{xy}+\bar{d}_{xy}x_{\mathrm{AP}}=0$, for an arbitral value of x_{AP} because

$\bar{a}_{xy} \cong en(\mu_e^2 S_e - \mu_h^2 S_h)P$ and $\bar{d}_{xy} \cong en(\mu_e^2 S_e - \mu_h^2 S_h)$. As a result, substitution of $\bar{a}_{xx} \cong en(\mu_h + |\mu_e|)$ and $\bar{d}_{xx} \cong en(\mu_h + |\mu_e|)P$ into Eqs. (5.47) and (5.48) yields

$$\rho_{xx} = \frac{1}{en(\mu_h + |\mu_e|)} - P\frac{\Delta E_{AP,x}}{J_{c,x}}, \tag{5.49}$$

$$\rho_{yx} = -P\frac{\Delta E_{AP,y}}{J_{c,x}}. \tag{5.50}$$

When n, μ_e, μ_h and $J_{c,x}$ are given, $\Delta E_{AP,x}$ and $\Delta E_{AP,y}$ can be assessed from observed values of ρ_{xx} and ρ_{yx}, respectively.

Case II enables us to find out that a condition, $S_h = S_e$, gives the necessary and sufficient condition for the resonant Hall effect, $\bar{a}_{xy} + \bar{d}_{xy} x_{\mathrm{AP}} = 0$, for an arbitral value of x_{AP} because $\bar{a}_{xy} \cong en(1/S_e - 1/S_h)P$ and $\bar{d}_{xy} \cong en(1/S_e - 1/S_h)$. As a result, substitution $\bar{a}_{xx} \cong en[1/(\mu_h S_h^2) + 1/(|\mu_e| S_e^2)]$ and $\bar{d}_{xx} \cong en[1/(\mu_h S_h^2) + 1/(|\mu_e| S_e^2)]P$ into Eqs. (5.47) and (5.48) yields

$$\rho_{xx} = \frac{1}{en[1/(\mu_h S_h^2) + 1/(\mu_e S_e^2)]} - P\frac{\Delta E_{AP,x}}{J_{c,x}}, \tag{5.51}$$

$$\rho_{yx} = -P\frac{\Delta E_{AP,y}}{J_{c,x}}. \tag{5.52}$$

When $n, S_e, S_h, \mu_e, \mu_h$ and $J_{c,x}$ are given, $\Delta E_{AP,x}$ and $\Delta E_{AP,y}$ can be assessed from observed values of ρ_{xx} and ρ_{yx}, respectively.

It follows from Eqs. (5.49), (5.50), (5.51), and (5.52) that spin dependent field $\Delta \mathbf{E}_{AP}$ influences ρ_{xx} and ρ_{yx} in the same way between cases I and II, indicating that ρ_{xx} is affected by $\Delta E_{AP,x}$ while ρ_{yx} is affected by $\Delta E_{AP,y}$. The roles of $\Delta E_{AP,x}$ and $\Delta E_{AP,y}$ are, however,

complemental to the non-resonance case in which ρ_{xx} is affected by $\Delta E_{AP,y}$ while ρ_{yx} is affected by $\Delta E_{AP,x}$ as described in Sec.5.2. This complemental difference is useful for experimentally specifying with either the resonance or non-resonance cases. This complemental nature allows us to assess magnetic field dependences of ρ_{xx} and ρ_{yx} in the resonance case, as schematically shown in Fig. 5.3. In contrast to the non-resonance argued in Sec. 5.2, ρ_{yx} shows no field dependence while ρ_{xx} is influenced by magnetic field through the field dependences of $\Delta E_{AP,x}$ and P.

The $\Delta E_{AP,y} = 0$ nature, which is caused when nonmagnetic metals are used for Hall voltage detection, is responsible for $\rho_{yx} = 0$ in the resonance case. Using magnetic metals for the voltage detection, however, makes $\Delta E_{AP,y}$ nonzero values because spin accumulation is generated in the transverse direction (y-direction). Figure.5.4 schematically shows ρ_{yx} for $\Delta E_{AP,y} \neq 0$ case, provided that $\Delta E_{AP,y}$ has an even function characteristic as shown in the inset of Fig.5.4 Using magnetic metals as the Hall voltage detection electrode is indispensable for observation of the resonance Hall effect.

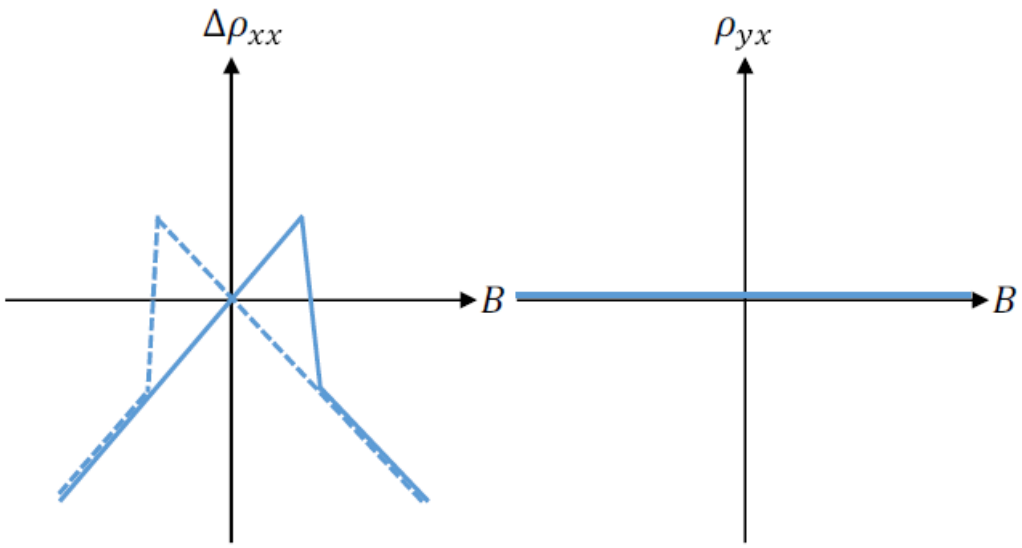

FIG. 5.3. Qualitative assessments of magnetic field dependences of resonance case for $\Delta\rho_{xx}$, which is given by Eq. (5.49) (low mobility and/or weak SOI case) and Eq. (5.51) (high mobility and/or strong SOI case), and ρyx given by Eq. (5.50) (low mobility and/or weak SOI case) and Eq. (5.52) (high mobility and/or strong SOI case) under assumption of $\Delta E_{AP,y} = 0$. $\Delta\rho_{xx}$ is defined as $\Delta\rho_{xx} = \rho_{xx} - \rho(B = 0)$. Solid and dashed lines indicate field-upsweep and field-downsweep, respectively.

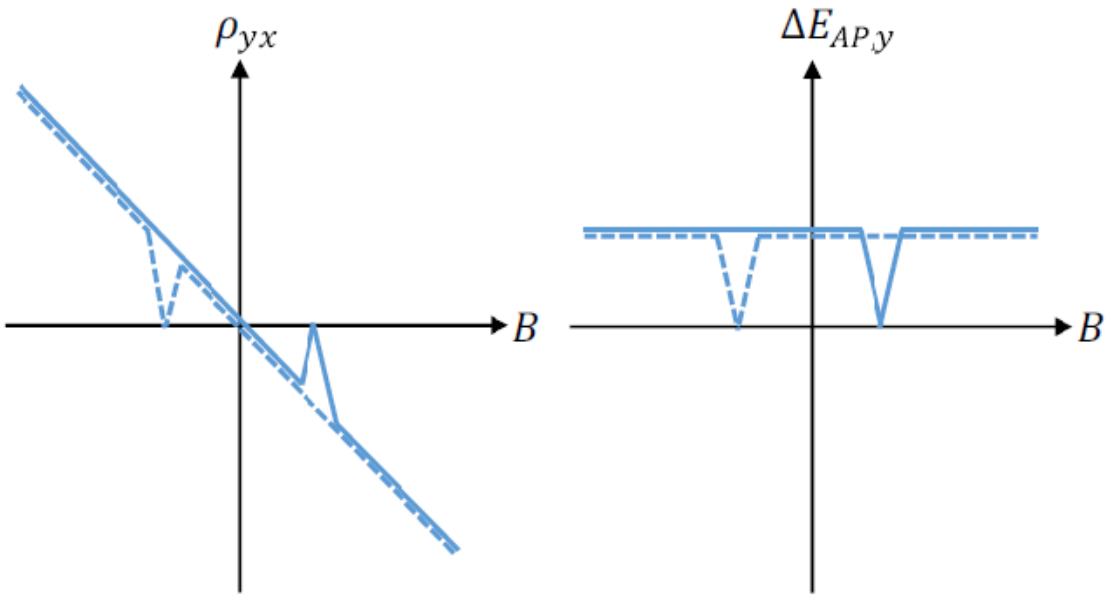

FIG. 5.4. Qualitative assessment of magnetic field dependences of resonance case for ρ_{yx} given by Eq. (5.50) (low mobility and/or weak SOI case) and Eq. (5.52) (high mobility and/or strong SOI case) under consideration of $\Delta E_{AP,y}$, which is also schematically indicated. Solid and dashed lines indicate field-upsweep and field-downsweep, respectively.

5.4 Summary

We theoretically investigated how the enhancement of spin coherence length affects the Hall and longitudinal resistivities in nearly CMs under simultaneous injection of electron and hole spins. Proper specifications of spin dependent particle current densities were given in terms of the electrochemical potentials associated with the parallel and antiparallel SCs, showing that the antiparallel SC keeps the Onsager reciprocal relations when it is in combination with conventional charge current.

After derivation of general formulae for Hall and longitudinal resistivities, two types of Hall effects, non-resonance and resonance types, are described considering two cases, small mobility and/or weak SOI case and large mobility and/or strong SOI case. Both types of Hall effects are influenced by the spin dependent chemical potential with the extraordinarily large spin coherence length but the roles of longitudinal and transverse components of spin dependent field are complementally interchanged between the two types of Hall effects. In addition to the spin dependent chemical potential field, paramagnetic spin polarizations of electrons and holes are indispensable for the resonance type, which is accompanied with self-sustaining mode of antiparallel SC.

CHAPTER 6

CONCLUSION

Low power consumption and less heat generation is the ultimate goal of spintronics devices for altering conventional charge based electronics. By taking this matter in great importance, we have selected ambipolar conductors for being used as spintronics material. Because experimental results reveal that ambipolar conductors have large spin diffusion length which is an essential feature of spintronics devices to become in reality.

Heat dissipation is formulated with the production of entropy. A dissipation formula of spin current is derived by Taniguchi which is independent of the explicit form of source term of spin current. But this dissipation formula of spin current cannot be applicable in ambipolar conductors because of two types of charge carriers are present simultaneously whereas Taniguchi's formula deals with only single charge carrier conductors. Due to the unavailability of the theory of dissipation of double carrier conductors such as ambipolar conductors, we have performed a theoretical evaluation for the entropy production rate equation of ambipolar conductors. Our derived theoretical formula yields several significant outcomes. Firstly, it reveals the presence of two types of spin currents in ambipolar conductors, such as parallel and antiparallel spin current. Secondly these spin currents do not take part in entropy production which implies that spin currents in ambipolar conductors are dissipationless. Finally, it ends up with an enhancement factor $(\frac{1}{\Phi})$ associated with the spin diffusion length of antiparallel spin current in ambipolar conductors.

After getting the idea of antiparallel spin current, which is specific to the ambipolar conductors, we focus our attention to the spin relaxation mechanism as the spin lifetime is another important aspect in spintronics. Realizing Baber-type collision and application of Gibbs-Duhem relation in ambipolar conductors justify the identical nature of spin relaxation times between electrons and holes. From these observations we propose that the nearly compensated metals with electron-hole collision have a potential mechanism making the spin relaxation time (τ_s) associated with the antiparallel spin current extraordinarily large and hence an enhancement

factor for spin coherence length of ambipolar conductor is derived to be $\frac{1}{\Phi}$. For instance, the enhancement factor predicted in YH_2 is approximately 670 because of $\Phi = 0.15\%$. With this result it is possible to explain the reason behind the ambipolar conductors for having a large spin diffusion length found in Ref (23)and (24) .

Finally, another theoretical study have performed on the spin and charge transport in nearly compensated metals to investigate how the enhancement of spin relaxation time, and hence, the enhancement of spin coherence length, affects the Hall resistivity and longitudinal resistivity in nearly compensated metals under simultaneous injection of electron and hole spins. A key finding of this study is the antiparallel spin current which comply with the Onsager reciprocal relation in combination with the conventional charge current. Derived formulae for Hall resistivity and longitudinal resistivity shows that enhanced spin relaxation time has important impact on both resistivities. It is found out that in some special cases, when resonance Hall effect condition is achieved, antiparallel spin current is accompanied with the self-sustaining mode which means that the antiparallel spin current can flow without any external current source.

In conclusion, we would like to state that our theoretical investigations on ambipolar conductors reveal i) the presence of dissipationless spin current ii) validate its long spin coherence and iii) self-sustaining mode of spin current in resonance condition. All of these characteristics make ambipolar conductors a promising material for realizing future spintronics devices.

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