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SPECIALTY SECTION

This article was submitted to Condensed Matter Physics, a section of the journal Frontiers in Physics

RECEIVED 08 January 2023

ACCEPTED 01 March 2023

PUBLISHED 24 March 2023

CITATION

Kondou K and Otani Y (2023), Emergence of spin–charge conversion functionalities due to spatial and time-reversal asymmetries and chiral symmetry. *Front. Phys.* 11:1140286. doi: 10.3389/fphy.2023.1140286

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Emergence of spin–charge conversion functionalities due to spatial and time-reversal asymmetries and chiral symmetry

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Spin–charge conversion (SCC) leads to the driving principle of spintronics devices, such as non-volatile magnetic memory and energy harvesting devices from light, sound, and heat to charge current. Recently, controllable SCCs have emerged in materials with spatial- and time-reversal asymmetry as a new route for efficient manipulation and realization of novel functionalities of future spintronics devices. This study overviews the SCC from the fundamental mechanism to the recent research progress in novel materials, such as topological magnets and atomically layered materials. Additionally, we discuss the chiral organic materials from the viewpoint of a new pathway for the emergence of spin functionalities.

KEYWORDS

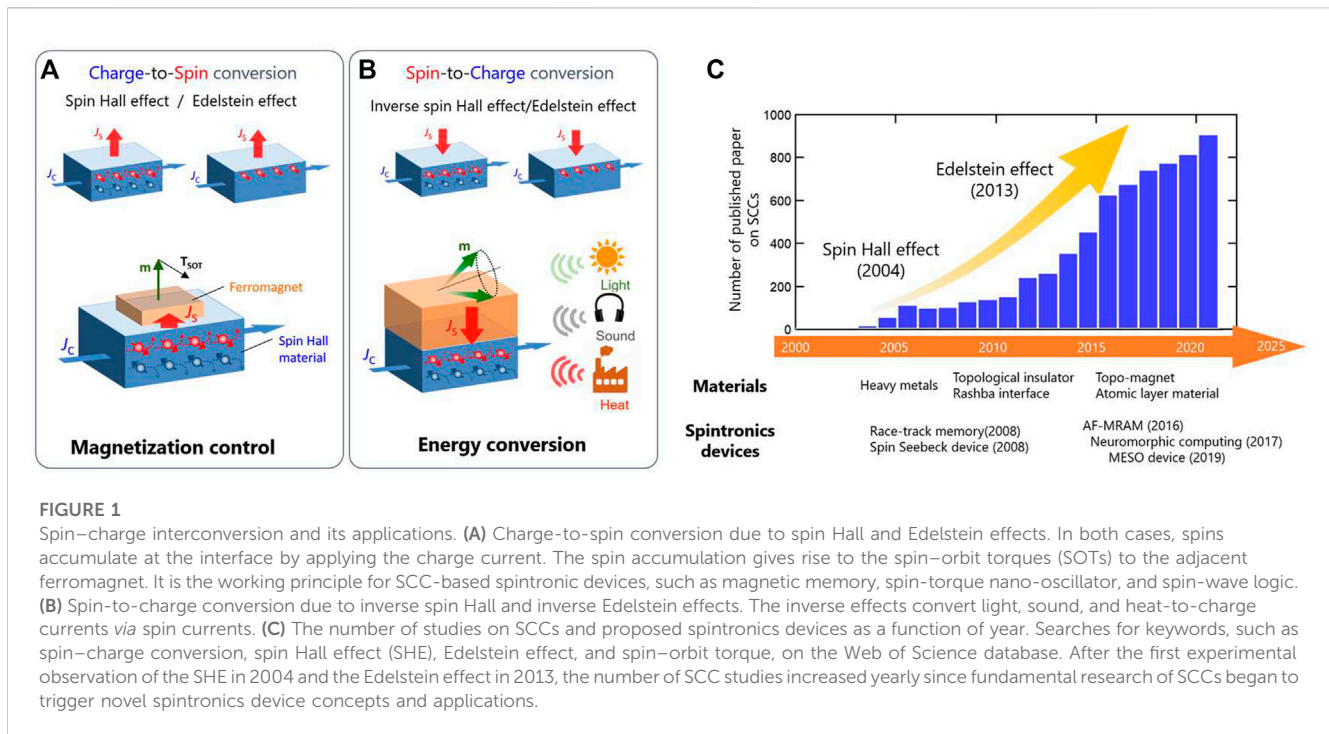
spin-charge conversion, chirality-induced spin selectivity, spatial inversion asymmetry, time-reversal asymmetry, spin Hall effect, Edelstein effect

1 Research background and progress on the spin–charge conversion

Spin and charge degrees of freedom can be converted by the spin Hall effect (SHE), which was theoretically predicted by Dyakonov and Perel [1]. Approximately 3 decades later, Hirsch re-predicted and introduced it as SHE [2]. Since the 2000s, high-quality film preparation and microfabrication technologies for semiconductors and metals have been developed, leading to the first observation of spin accumulation in GaAs by optical technique in 2004 [3]. After the pioneering works, systematic experiments in semiconductors have clarified the fundamental properties of SHE, such as carrier type [4], amount of dopant [5], and intervalley transition dependence [6]. These findings might lead to the establishment of fundamental technology for a novel integration platform that combines photonic, magnetic, and electronic components.

Moreover, intensive experimental research on the SHE in metallic materials [7–10] established the so-called spin–orbit torque (SOT) that enables switching magnetization and driving magnetic domain wall for magnetic memory applications [11, 12] and auto-oscillation for microwave generators [13], among others. Thus, SHE is currently one of the indispensable phenomena in spintronic devices.

An essential characteristic of the SHE is that it can act as a spin source by producing the flow of spin angular momentum (spin-current), perpendicular to the charge current, as shown in [Figures 1A, B](#). Thus, the SHE contributes to simplifying the device structure, for example, ferromagnetic metal/spin Hall material bilayer for magnetization switching [11–13]. It has also been developed to extract electrical charge current from heat, sound,



and light using the inverse effect. Accordingly, this technique draws attention from the standpoint of energy harvesting [14, 15].

The spin-charge conversion (SCC) efficiency, defined as the ratio of the spin current density and the charge current density, called the spin Hall angle, determines the performance of such SHE-based spintronic devices. The SHE relies on spin-orbit interactions and can be generated by intrinsic or extrinsic mechanisms. So far, systematic theoretical and experimental works reveal that SHE in 4d and 5d transition metals comes from the intrinsic mechanism based on the degeneracy of d-orbitals via spin-orbit coupling (SOC) [16–19]. However, the extrinsic SHE relies on scattering, namely, skew scattering [20] and side jump [21] by impurities causing strong spin-orbit interactions. However, the maximum conversion efficiency of these materials is only in the range of a few tens of percent [19] [11, 22, 23]. Thus, it was required to discover and establish the technology for novel conversion phenomena to improve efficiency.

Recently, as a new type of spin conversion principle instead of the SHE, the Edelstein effect (EE) in the two-dimensional electron systems with spin-splitting surface states has been actively studied [24, 25]. Unlike the bulk SHE, this effect generates spin accumulation via spin momentum locking linked to the charge current flow at material interfaces. Such spin-splitting surface and interface states caused by the spatial inversion asymmetry have been observed at the surface of the topological insulator and Rashba interface by means of angle-resolved photoemission spectroscopy (ARPES) [26, 27]. In the surface states, the polarization vector of electron spins depends on the direction of electron flows, called spin-momentum locking [27]. The application of an electric field gives rise to spin accumulation at the interfaces, a behavior known as the Edelstein effect (EE). The large spin splitting at the surface of the topological insulator and Rashba interface realizes more

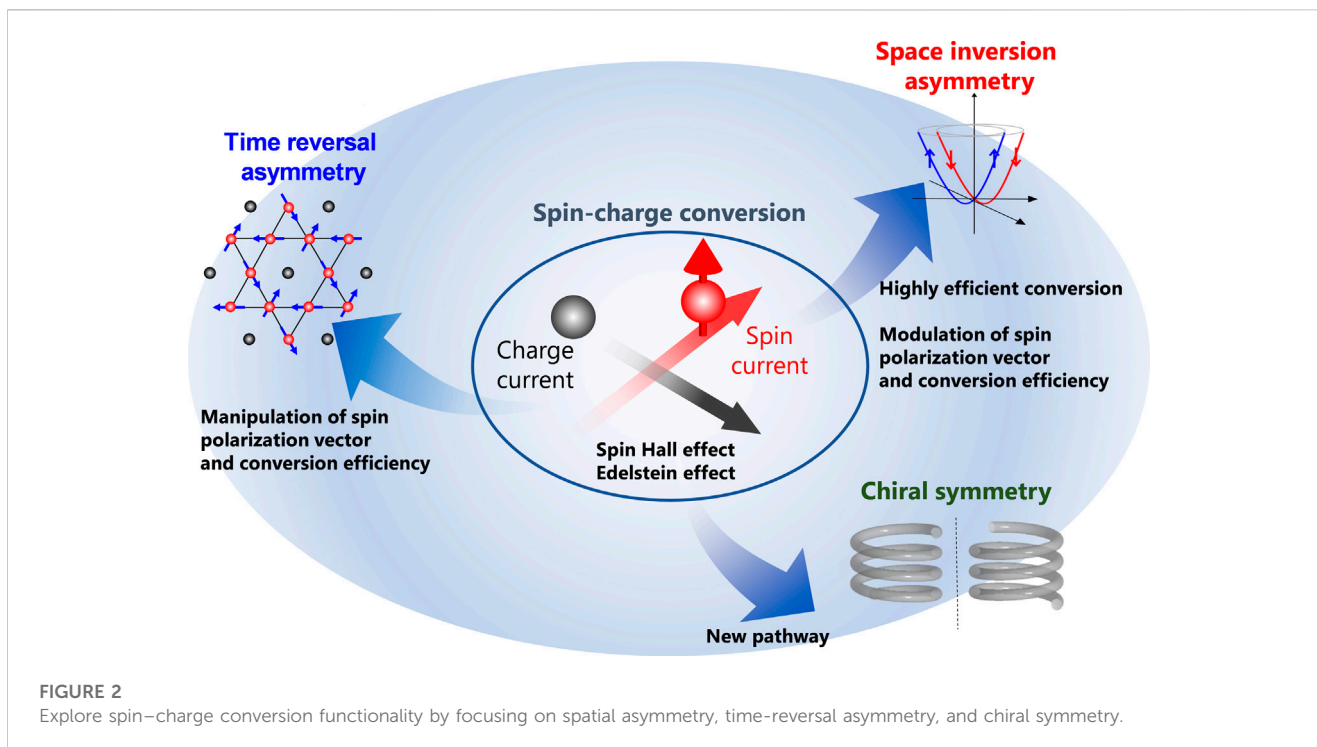
efficient SCC than transition metals, at several orders of magnitude smaller current densities than for transition metals [28–31].

These experimental demonstrations of SCCs via SHE and EE led to the novel concepts of spintronics devices [32–36], such as a racetrack memory driven by high-speed magnetic domain wall motion [32] and a magnetoelectric spin-orbit (MESO) device based on interfacial SCCs [33]. Since the first observation of SHE in 2004 and EE in 2013, the number of studies on SCCs has been increasing every year, as shown in Figure 1C. Thus, not only are SCCs interesting as a fundamental science, but they also attract attention in terms of applications and are expected to have practical applications.

From such research backgrounds, to realize lower power consumption and higher speed operation of spintronics devices, it is desired to develop a method to control the spin polarization vector and the conversion efficiency freely. This review article overviews the recent research trend of the emergence of SCC functionalities utilizing symmetry, such as space, time, and chirality, as shown in Figure 2.

2 Spin conversion due to spatial inversion asymmetry

At material interfaces, spin-splitting surface states appear due to spatial inversion asymmetry. When an electric field is applied to such an interface, non-equilibrium spin accumulation occurs [24, 25]. Edelstein predicted such spin accumulation in 1990 [24]. In the 2000s, by utilizing the spin-resolved ARPES technique, the surface states were investigated intensively. In 2007, a large spin splitting of ~200 meV was observed in Ag/Bi surface alloys. Interestingly, it is much larger than the Bi surface (~14 meV) [26, 37, 38]. Around the



same time, it was experimentally discovered that the linear Dirac dispersion exhibits 100% spin polarization in the topological insulator surface [27].

The spin accumulation at the surface state can exert SOTs consisting of field-like and spin-transfer (anti-damping) torques on adjacent magnets. Thus, effective utilization of EE-induced spin accumulation has been desired for spintronics study. High-quality thin film preparation technologies, including molecular beam epitaxy and sputter deposition, are now ready to fabricate devices for quantitative evaluation of spin generation at the interface. These achievements have enabled low-power efficient magnetization reversal [28, 31, 39, 40].

The amplitude of spin splitting, proportional to Rashba parameter α_R , characterizes the efficiency of the EE-induced SCCs. Thus, the charge-to-spin conversion efficiency q and spin-to-charge conversion λ are described by $q = \alpha_R / (\hbar \tau_s v_F^2)$ and $\lambda = \alpha_R \tau_s / \hbar$, where \hbar and τ_s are plank constant and spin relaxation time at the surface state, respectively [25, 28, 31]. There is a trade-off relation between conversion efficiencies q and λ through τ_s , which is determined by the strength of the hybridization between bulk and interface states [41]. Indeed, highly efficient spin-to-charge conversion has been observed at the oxide interface of $\text{LaAlO}_3/\text{SrTiO}_3$, as its interface exhibits a spin relaxation time several orders of magnitude longer than metal interfaces due to weak hybridization between bulk and interface states [42].

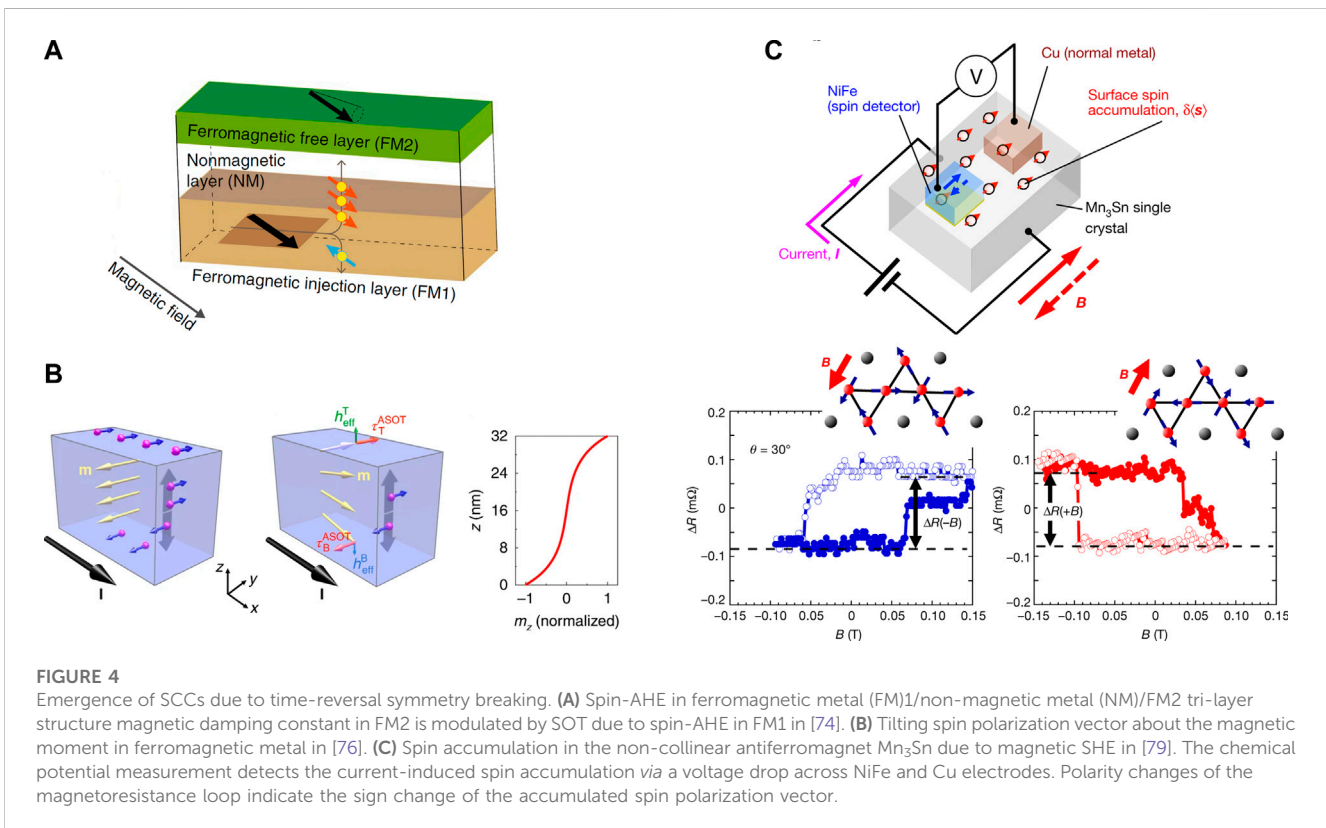
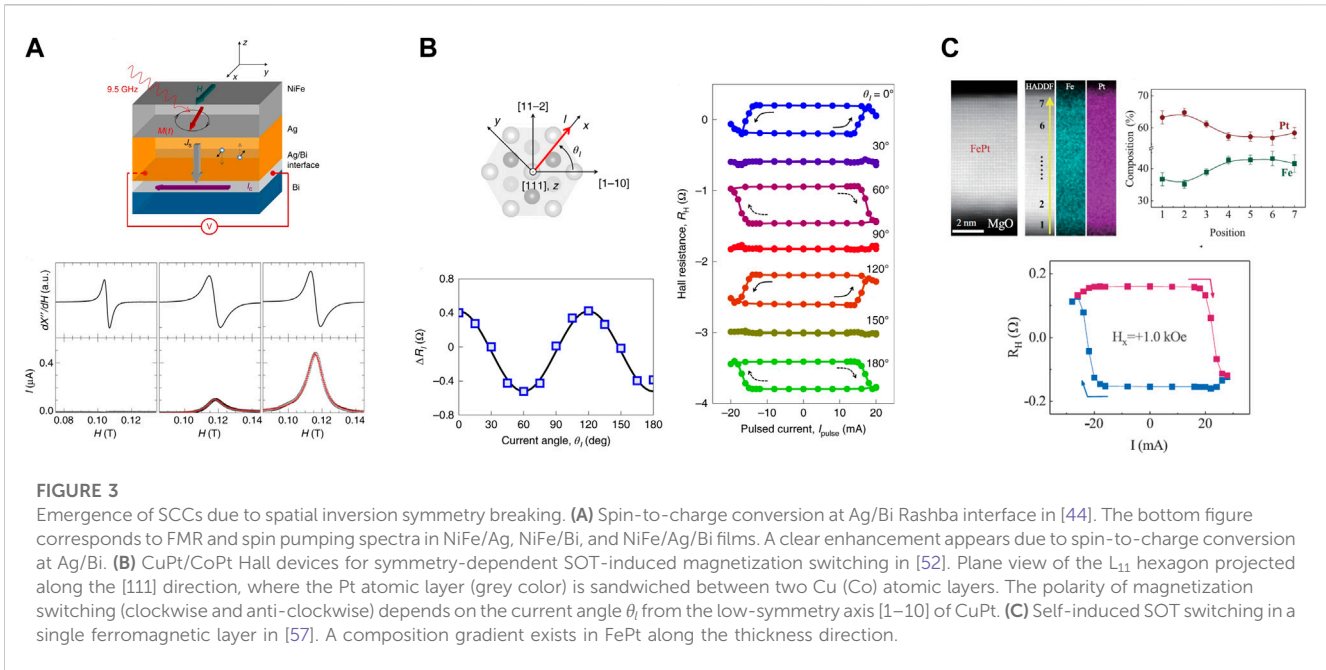
Since one of the origins of a large α_R is known as the asymmetric electron distribution at the interface [37, 43], the large α_R can be induced not only by the surface alloy such as Ag/Bi but also by many kinds of material interfaces [41, 44–47]. From this point of view, recently, novel SCCs have emerged by modifying the interfacial state of metal/organic molecules depending on the molecular structure, polarity, and arrangement. By utilizing the highly spin-to-charge

conversion at the molecule/metal interfaces, electrical detection of ultra-thin molecules absorption less than a single layer has been reported [41]. These research directions have drawn much attention as a new aspect of molecular spintronics [41, 47].

Unique features of these interfacial SCCs are the realization of highly efficient conversion, improvement of material selectivity, and the ability to modulate the conversion efficiency by the external electric field significantly. So far, several methods by utilizing an external electric field have been reported for 1) Fermi level tuning [48], 2) lattice strain [49], 3) oxygen transfer [50], and 4) electric polarity reversal in ferroelectric materials [51]. Especially for the latter two methods, it is possible to remain in the state even after the external electric field is turned off (i.e., non-volatile control). These modulation techniques are essential for future applications, such as magnetoelectric spin-orbit (MESO) devices based on interfacial SCCs [33].

Furthermore, spatial inversion asymmetry in crystals is also important for the emergence of SCC functions. Figure 3B shows the symmetry-dependent SOT magnetization switching in low-symmetry crystals, such as CuPt. Notably, out-of-plane spin polarization can be generated in such low-symmetry crystals by breaking lateral mirror symmetry [52–55] but not the conventional SHE in transition metals. In the inverse conversion (i.e., spin-to-charge conversion), the spin polarization and the charge current are parallel. In addition, the conventional conversion where the spin polarization is orthogonal to the charge current has been observed [56].

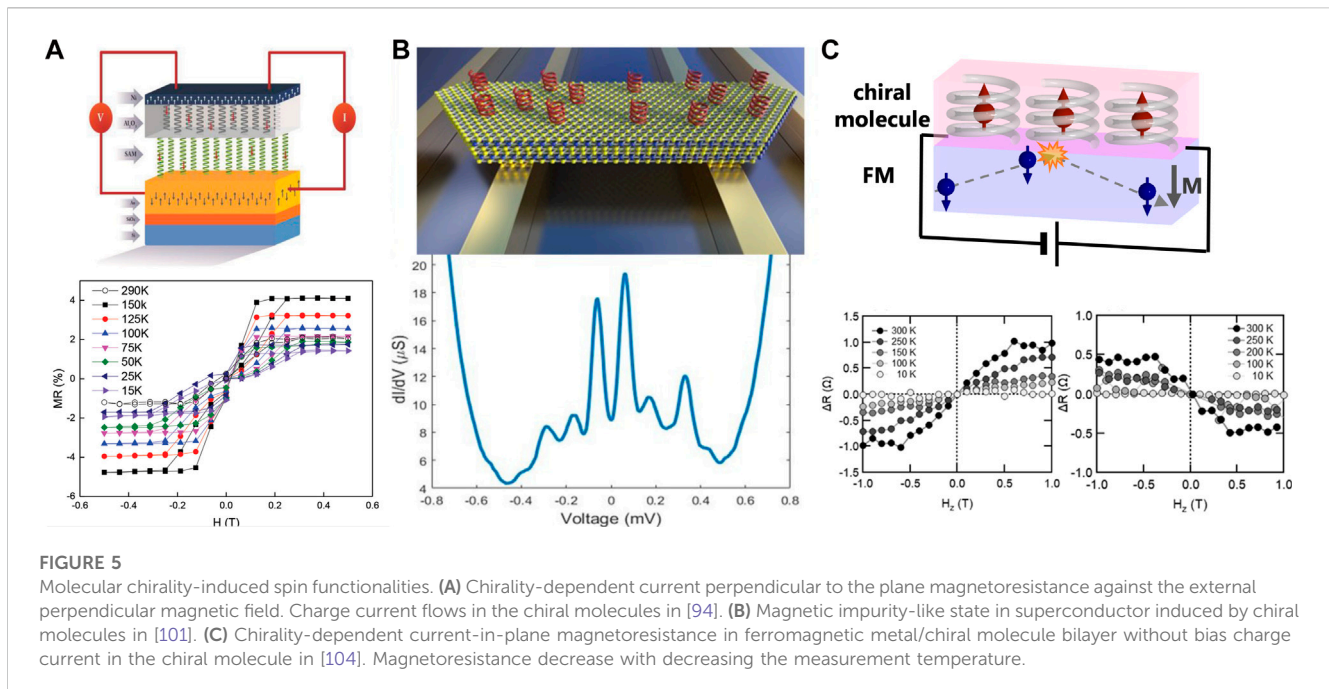
Additionally, spatial inversion asymmetry in the magnetic materials is also interesting because current-induced magnetization reversal has been observed in a single magnetic layer i.e., a spin source layer like Pt is unnecessary for magnetization control [57–61]. It might be caused by “self-



induced SOT,” which is induced by current-induced spin accumulation inside the magnetic material due to spatial asymmetry. For example, such self-induced SOTs have been observed in the gradient composition magnet in Figure 3C [57–59] and low symmetry van der Waals magnetic crystal Fe_3GeTe_2 [60]. In particular, in the F_3GeTe_2 , the critical current

density for magnetization reversal is approximately two orders of magnitude smaller than that in the conventional transition metal/ferromagnetic metal bilayer. Thus, the elucidation of the mechanism has attracted attention [61].

These fundamental research developments regarding the SCC functionalities induced by spatial inversion asymmetry are



promising to contribute to more efficient and faster magnetization control in spintronics devices.

3 Manipulation of spin conversion due to time-reversal asymmetry

This study focuses on SCCs in magnetic materials with time-reversal asymmetry. Magnetic materials have also been studied using the same experimental methods for non-magnetic metal such as non-local spin valves, spin-torque ferromagnetic resonance (FMR), and spin pumping, among others [62, 63]. So far, SHE and ISHE on magnetic materials have been investigated regarding a correlation with the anomalous Hall effect (AHE) because SHE shares the scattering mechanism, such as skew scattering and side jump with AHE [64, 65]. From this point of view, utilizing scattering enhancement due to spin fluctuation near the magnetic phase transition temperature is also a promising way to attribute the amplitude of SHE [66–71].

Recently, there have been remarkable developments in experimental research on the magnetization direction-dependent SHE [70–82]. Figure 4A shows the SHE caused by AHE in a ferromagnet, which was first predicted by theoretical study and demonstrated experimentally later [72, 73, 75], called spin-AHE. In addition, spin precession along a ferromagnetic moment was found to tilt a spin polarization vector inside the ferromagnet shown in Figure 4B [76] and at the ferromagnetic/non-magnetic interface [77]. The spin polarization vector can be controlled by adjusting the magnetic moment in these phenomena.

Furthermore, this concept has been extended to SHE in antiferromagnets [78–82]. Notably, the non-collinear antiferromagnets Mn_3X ($X = Sn, Ge$) draw much attention as a candidate material for next-generation ultra-fast spintronic devices. It is a *Weyl* magnet displaying a huge response comparable to

ferromagnets at room temperature [83]. Such a non-collinear antiferromagnet has a tiny magnetization, approximately 1/1,000 of a ferromagnet, so the time-reversal symmetry is broken, and the spin structure of the Mn_3X can be reversed by a small external magnetic field or electric current *via* SOT [83–85].

In contrast to the ferromagnetic dipole, the spin structure of the Mn_3X generates a magnetic octupole moment consisting of multiple Mn spins. In the Mn_3X , octupole direction-dependent SHE and ISHE caused by the momentum-dependent spin polarization produced by the non-collinear magnetic order were discovered and named magnetic SHE (MSHE) and magnetic ISHE, as shown in Figure 4C [79].

In the MSHE, the spin-polarization vector is perpendicular to the direction of the magnetic octupole moment. Very recently, unconventional SOT and field-free magnetization switching of adjacent ferromagnets due to the MSHE in non-collinear antiferromagnets have been reported [80–82].

Compared with the spatial asymmetry-induced SCC functionalities, the SCCs caused by the time-reversal asymmetry can be tuned more freely by controlling the magnetization direction by an external magnetic field. This section focused on SCCs generating a steady spin state in magnetic materials. The emergence of novel functionalities of SHE under magnetization dynamics might be an attractive research topic in the near future [66, 86].

4 New pathway for the emergence of spin functionalities due to chiral symmetry

Chirality is a common property in wide branches of science, such as biology, chemistry, physics, and cosmology, for the emergence of any functionality. Molecular chirality is essential to

induce spin functionalities in organic materials. Indeed, the spin polarization of electrons passing through chiral molecules such as DNA chains has been observed [87], implying an interaction between chiral structure and passing electron spins. This property is called chiral-induced spin selectivity (CISS), of which phenomena have been investigated using various experimental techniques, such as photoelectron spectroscopy, conductive atomic force microscope (AFM), and magnetoresistance measurement [87–96]. Surprisingly, a considerably large spin polarization [90, 91] comparable to that of ferromagnets such as Fe (~0.4) appears at room temperature despite the weak SOC of light elements in chiral organic molecules.

However, the physical origin of the large spin polarization due to CISS remains elusive. In particular, significantly large SOC of approximately several 100 meV, which is several orders higher than that of organic materials such as graphene (~10 μ eV [97]), is necessary to realize such large spin polarization [90–92]. Recent theoretical work revealed that the geometric SOC due to molecule structure, such as curvature, gives rise to large SOC even in organic materials such as DNA [98]. Furthermore, as another degree of freedom, the contribution of orbital texture in chiral molecules has also been discussed [99, 100]. Quantitative and systematic experimental and theoretical works are indispensable to reveal the microscopic origin of large spin polarization due to CISS in molecules.

Conversely, several experimental studies have recently reported the CISS-like effect without bias current in chiral molecules [101–104]. For instance, a chiral molecule adsorbed on a superconductor surface exhibits Shiba states similar to the magnetic impurity-induced state in the tunneling spectra, as shown in Figure 5B [103]. Moreover, a chirality-dependent effective magnetic field has occurred at room temperature [101, 102]. Remarkably, no bias charge current flows through the chiral molecules, implying that spontaneous spin polarization may emerge in the chiral molecules. More recently, current-in-plane MR (CIP-MR) effects have been observed in chiral molecule/ferromagnetic metal bilayers at room temperature, as shown in Figure 5C [104]. The temperature dependence of the MR suggests the existence of thermally driven spontaneous spin polarization in the chiral molecules [104–107].

Recently, in the chiral inorganic crystals, systematic experiments regarding the CISS effect have been reported [108, 109]. Spin polarization due to the CISS effect has been observed by applying a charge current along the *c*-axis in chiral crystal CrNb₃S₆. The inverse CISS has also been detected in the same device, confirming the CISS effect's reciprocal relationship [108]. Furthermore, exciting phenomena beyond conventional spintronics physics, such as

chirality-induced spin polarization over millimeters, have been reported in the chiral crystals despite the strong SOC (chiral) materials [109]. It is highly desirable to comprehensively understand such chirality-induced spin phenomena across a range of materials and scales.

5 Conclusion

We have reviewed recent SCCs research regarding the emergence of novel functionality caused by spatial asymmetry, time-reversal asymmetry, and chiral symmetry. These symmetries enable us to control the conversion efficiency of the SCCs and the spin polarization vector. Importantly, such a concept for the emergence of SCCs functionality incorporates the viewpoint of symmetry will progress remarkably the design of the material, device, and its function [104].

Author contributions

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

Funding

This work was supported by JST-CREST (no. JPMJCR18T3) and JST-Mirai (no. JPMJMI20A1) and by Grants-in-Aid for Scientific Research (19H02586 and 19H05629).

Conflict of interest

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