Topical Review

Multifunctional oxides for topological magnetic textures by design

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Abstract

Several challenges in designing an operational skyrmion racetrack memory are well known. Among those challenges, a few contradictions can be identified if researchers are to rely only on metallic materials. Hence, expanding the exploration of skyrmion physics into oxide materials is essential to bridge the contradicting gap. In this topical review, we first briefly revise the theories and criteria involved in stabilizing and manipulating skyrmions, followed by studying the behaviors of dipolar-stabilized magnetic bubbles. Next, we explore the properties of multiferroic skyrmions with magnetoelectric coupling, which can only be stabilized in Cu$_2$OSeO$_3$ thus far, as well as the rare bulk Néel-type skyrmions in some polar materials. As an interlude section, we review the theory of the anomalous and topological Hall effect (THE), before going through the recent progress of THE in oxide thin films. The debate about an alternative interpretation is also discussed. Finally, this review ends with a future outlook regarding the promising strategies of using interfacial charge transfer and (111)-orientation of perovskites to benefit the field of skyrmion research.

Keywords: oxide skyrmions, topological Hall effect, thin-film heterostructures

(Some figures may appear in colour only in the online journal)

1. Introduction

1.1. Essential physics of skyrmions

The concept of topological defects as soliton solutions to a continuous field is present in a broad scope of natural physics, including tornadoes, whirlpools, Abrikosov vortices in type-II superconductors, liquid crystals, etc, that involve a non-linear field equation. Skyrme first proposed such a quasiparticle theory to predict several properties of nucleons as excitation in pion fields [1], while skyrmions in magnetism are physically real and potentially useful in spintronics. A magnetic skyrmion is a quasiparticle of collective magnetic moment excitations with a topological winding number, $Q$, that fully wraps around a Bloch sphere for an integer number of times (figures 1(a)–(c)),

$$ Q = \frac{1}{4\pi} \int \hat{m} \cdot \left( \frac{\partial \hat{m}}{\partial x} \times \frac{\partial \hat{m}}{\partial y} \right) \, dx dy = \pm \text{integer}. \quad (1) $$

In a typical magnetic system, the relevant atomic-scale energy landscape is described by an effective Hamiltonian,

$$ \hat{H} = -J_{ex} \sum_{ij} \hat{m}_i \cdot \hat{m}_j - K_u \sum_i m_{z,i}^2 - \sum_{ij} D_{ij} \cdot (\hat{m}_i \times \hat{m}_j) - \mu_o H \cdot \sum_i \hat{m}_i \quad (2) $$

where the first to fourth terms are the isotropic exchange, uniaxial anisotropy, Dzyaloshinskii–Moriya interaction (DMI) and Zeeman energy. There are two types of spin–orbit coupling (SOC): the Rashba type $\alpha_R (k \times \hat{z}) \cdot \hat{\sigma}$ and the Dresselhaus
Skyrmions with topological parameters \((Q, Q_x, Q_y)\): (a) \((1, 1, \pi/2)\), (b) \((1, 1, 0)\) and (c) \((-1, -1, 0)\). The top panel shows Skyrmions in 2D, while the bottom panel shows their respective stereographic projections into 3D Bloch spheres. (d) A typical magnetic phase diagram of bulk MnSi. (e) Vertical DW racetrack memory array proposed by Parkin et al. The DW information storage bits can be replaced by skyrmions. (f) An illustration of a skyrmion racetrack with the heavy metal layer providing STT or SOT, and storage bits can be replaced by skyrmions. (f) From [3], CC BY 4.0. (d) From [4]. Reprinted with permission from AAAS. (e) From [5]. CC BY 4.0.

Type \(\alpha_D [k_x (k^2_x - k^2_y) \sigma_y + k_y (k^2_y - k^2_x) \sigma_x + k_z (k^2_z - k^2_y) \sigma_z]\), which emerge from broken interface and bulk inversion symmetry, respectively, where \(k\), \(\sigma\) and \(z\) are the electron’s momentum, spin and the sharp interface normal vector. Correspondingly, there are two types of DMI that would stabilize the Néel- and Bloch-type skyrmions [6–8]. The DMI term in equation (2) can be rewritten into a linear combination of \(D \sin (\gamma) \hat{m} \times (\nabla \times \hat{m}) + D \cos (\gamma) \hat{m} \cdot (\nabla \times \hat{m})\), where \(\tan (\gamma) = \frac{D}{\mu}\) is a ratio between the Rashba-type and Dresselhaus-type DMI, emerging in crystals with \(C_{av}\) and \(D_{av}\) symmetries, respectively [9, 10]. Hence, the Bloch-type skyrmions can be found in materials with non-centrosymmetric or hexagonal crystal structures [3, 11, 12], while the Néel-type ones exist at sharp interfaces between ferromagnetic (FM) and paramagnetic (PM) heavy metal without specific requirement on crystal structures [13–16]. From equation (1), skyrmions can be nucleated, usually assisted by the application of a small magnetic field, when a criterion is fulfilled [17]:

\[
\frac{D}{\sqrt{J_{ct} K_a}} \geq \frac{4}{\pi}.
\]  

This is also coined as the effective anisotropy model [10]. The resulting skyrmion radius is \(r_{sk} = \sqrt{J_{ct} / 2K_a (1 - D/D_c)}\) where \(D_c = 4\sqrt{J_{ct} K_a}/\pi\). From the Landau–Ginsburg framework, three helicoids merge to form a skyrmion lattice (SKL) assisted by an out-of-plane magnetic field [18, 19]. This hence results in a typical skyrmion phase diagram: at zero/low magnetic field, helical or cycloidal domains exist in labyrinth-like stripes; the SKL phase exists at the intermediate field; at large field, moments are saturated into a collinear FM phase (figure 1(d)). A complete theoretical categorization of \(|Q| = 1\) skyrmions can be done according to three topological parameters: the winding number, vorticity and helicity \((Q, Q_x, Q_y)\), which results in 16 configurations in total, including antiskyrmions [20, 21]. Other topologies such as Merons \((Q = \pm 0.5)\), skyrmionium \((Q = 0)\) and bi-skyrmion \((Q = \pm 2)\) are also possible, though rare.

1.2. Design criteria for skyrmion memory

From a practical application viewpoint, Parkin et al. first proposed the concept of domain wall (DW) racetrack memory in the year 2008 (figure 1(e)), where the DW movements are driven by current via spin-transfer torque (STT) [4, 22, 23]. Subsequent research motivation in this field placed emphasis on reducing the threshold current density \(j_x\) for initiating the DW motion, below which the DWs will be pinned by impurities/defects. Typical \(j_x\) with STT on soft collinear FMs such as permalloys is \(10^{11}–10^{12}\) A m\(^{-2}\). However, Bloch-type skyrmions in MnSi and FeGe single crystals [24–26] have demonstrated an astoundingly low \(j_x\) of just \(\sim 10^6\) A m\(^{-2}\), suggesting that replacing FM DWs with skyrmions as information storage bits is an ideal strategy for building future energy-saving racetrack memories. On the route to achieving an operational skyrmion racetrack memory [5, 27], several challenging aspects need to be overcome as listed below:

(1) Individual skyrmions should be controllably nucleated at the WRITE-head by providing a perturbation to a local FM energy landscape. Thus far, the two successful schemes are: (i) application of an electric field via a sharp, antiferromagnetic (AF) Cr scanning tunneling microscope (STM) probe tip [28, 29], and (ii) injection of a nanosecond pulsed current via a nano-patterned injector electrode [30]. For scheme (i), a near-bi-stable energy landscape between collinear FM and skyrmion is first achieved by applying a small magnetic field, and subsequent injection of spin-polarized tunneling electrons into the sample would excite across the energy barrier for skyrmion nucleation due to the STT effect. Changing the spin-polarized Cr tip to an unpolarized W tip also demonstrated the pure electric field switching effect [29]. For scheme (ii), a charge current injection in the \(x\)-direction \(I_{c, x}\) of a heavy metal (e.g. Pt) would produce a spin current in the \(z\)-direction, carrying \(y\)-direction spin \(J_{c, y}\) by the spin Hall effect (SHE). It then exerts a spin–orbit torque (SOT) onto the adjacent FM with perpendicular magnetic anisotropy (PMA) \(M_z\), assisting to nucleate skyrmions with opposite-moment core with respect to the surrounding magnetization [31, 32]. To enhance the spin accumulation density by SHE, a delicately patterned sharp current injector or constriction is desirable, so that skyrmions always nucleate at a predictable position.
To minimize energy consumption by achieving zero-current skyrmion nucleation, an intrinsic material property—magnetoelastic (ME) coupling [12, 33], allowing electric field control of magnetism in multiferroic skyrmions such as Cu$_2$OSeO$_3$—is valuable but remains very rare. In addition, two more possible skyrmion tuning mechanisms by an electric field gating have gained attention. In the voltage-controlled magnetocrystalline anisotropy (VCMA) design, the electric field is expected to tune the carrier density significantly (typical at a ferromagnet/oxide interface) and induce a change in magnetic anisotropy locally in the nanotrack. Maruyama discussed that a negative voltage may lift the degeneracy of 3d orbitals—raising the energy of $d_{3z^2-r^2}$ ($L_z = 0$) states, populating more electrons into $d_{x^2-y^2}$ ($L_z = 2$) states and hence increasing the PMA [34]. This mechanism can be used to control the skyrmions’ motion by creating a trapping/de-trapping potential beneath the top gate (figure 1(f)) [5, 27], or function as the WRITE-head to create/annihilate skyrmions [35].

A magnetic-field-free scenario of such operation is possible by providing an exchange bias field from an adjacent antiferromagnet. On the other hand, using tight-binding calculation, Bhowal et al predicted a change in intra-orbital hopping amplitude and Rashba-type SOC by electric-field tuning [36], but is nevertheless a very weak (second-order perturbation) effect if ME coupling is absent. In short, we can see that all strategies involved tunings around fulfilling the equation (3) criterion.

(2) Skyrmions should be driven by low current to drift along the racetrack [24–26]. From a simple power dissipation ($I^2R$) concept, reducing drive current is the key to developing low energy-consumption memory, with contemporary criterion in the atto-joule ($10^{-18}$ J) range. The Thiele-modified Landau–Lifshitz–Gilbert equation is usually employed in simulating skyrmions’ motion [37]:

$$G \times (v_e - v_{sk}) + D \cdot (\beta v_e - \alpha v_{sk}) + F_{pin} = 0$$  

(4)

where the first and second terms are the Magnus and spin-transfer forces, respectively, with the gyromagnetic vector $G = 4\pi Q \mathbf{z}$, and the dissipative tensor $D = \begin{bmatrix} 0 & \int (\partial \mathbf{\hat{m}} \cdot \partial \mathbf{\hat{m}}) d^2r \\ \int (\partial \mathbf{\hat{m}} \cdot \partial \mathbf{\hat{m}}) d^2r & 0 \end{bmatrix}$. $v_e$ and $v_{sk}$ are the drift velocities of conduction electrons and skyrmions, and $\alpha, \beta$ are the adiabatic and non-adiabatic Gilbert damping parameters. $F_{pin}$ in the third term is the pinning force from impurity/defects or control of anisotropy. For simplicity, the SOT term $\zeta_{j_e}$ was understood as the skyrmions’ ability to evade impurity/defect pinning due to their topological protection [40]. A pulsed current injection is a common engineering strategy to reduce Joule heating, by using a low duty cycle = pulse width/interval [30, 32, 41, 42]. Conversely, a DC current would always cause heat accumulation and the continuous rise of device temperature with time, resulting in uncontrollable thermal-induced nucleation, and size change or deletion of magnetic skyrmions. For example, in the thermally driven skyrmion nucleation scheme, the threshold nucleation current can be seen to vary inversely with the duty cycle [41]. Pulsed-current injection also facilitates the quantification of skyrmion/DW velocities by static position imaging [43, 44].

(3) The skyrmions’ motion across the racetrack has to be straight to avoid annihilation at the sidewall. From equation (4), the Magnus force (first) term is expected to produce a transverse skyrmion velocity component $(v_{sk,\perp})$ named as the skyrmion Hall effect (SkHE) [43, 44]. Again, a pulsed current is beneficial to avoid annihilation at sidewalls, since skyrmions would be repelled by sidewalls if the Magnus force is below a certain limit. Ferrimagnetic skyrmions such as GdFeCo have also been shown to mitigate the SkHE angle [45]. To totally eliminate this SkHE, a search for AF skyrmions [46, 47] and skyrmioniums [48] (both have $Q = 0$) was proposed but they remained elusive until recently when a synthetic AF multilayer skyrmion structure bound by Ruderman–Kittel–Kasuya–Yosida interaction [15] was found to be stable.

(4) Skyrmion detection at the READ-head should be via tunneling non-collinear magnetoresistance (TNCMR), which is a tool compatible with practical device miniaturization. A tunnel barrier should be inserted between the skyrmion racetrack and the FM metal. Due to the difference in density of states (DOS) between collinear FM and skyrmion textures, a change in tunneling resistance can be detected [29, 49].

1.3. Why oxides? Motivations and opportunities

- Oxides have various physical interactions—superexchange, strong SOC, strain effect on crystal field splitting, and tunable Mott–Hubbard interaction, etc.
- Varieties of oxides are AF, bringing benefit for aspect (3).
- Oxides also have lower and tunable carrier density than typical metals, and are more susceptible to electric field gating for aspect (1). The ferroelectric or multiferroic ME coupling can be found in various oxides, allowing non-volatile, zero-current tuning of magnetic properties.
- Oxides may have complex non-centrosymmetric crystal structures that contribute Dresselhaus DMI.
- Interfacial or Rashba-type DMI could be controlled via termination engineering in perovskite oxides, such as polar/nonpolar AO and BO$_2$ in (001), or polar AO$_3$ and B in (111) orientations.
- Perovskite oxide thin-film interfaces can be fabricated with high quality for an interesting search of composite Néel- and Bloch-type skyrmions.
- Skyrmions hosted by high-quality crystalline oxides may be driven by a much lower $j_e$ for movement due to low defect pinning. The present records in amorphous metallic heterostructures are $10^{11} - 10^{12}$ A m$^{-2}$ due to high defect pinning, while those in bulk single-crystalline B20 compounds can be as low as $10^5 - 10^6$ A m$^{-2}$. 


• Perovskite oxides with $t_{2g}$ conduction band or in (111) orientation have topologically non-trivial and highly spin-textured band structures.

• When skyrmions driven by large current to move via equation (4), oxides with higher molar heat capacity $C_m$ would produce less of a temperature rise compared to metals, hence the magnetic properties are more stable against thermal fluctuations.

1.4. Popular imaging techniques for magnetic domains/bubbles/skyrmions

The earliest search for dipolar-stabilized bubbles began around the 1970s and was usually performed by the Faraday effect [50–52], which is the transmission version of the modern magnetic optical Kerr effect (MOKE), involving a rotation of linear polarization of incident light by a particular magnetic material. This technique showed contrast between the upward and downward magnetization domains but was not sensitive to the intermediate in-plane moment regions; hence information about $Q_b$ of the bubbles was concealed. Likewise, MOKE is popular among researchers working on Néel skyrmions [43] hosted by metallic ultrathin films where dark nanosized dots are seen in a light background. Although the DMI is widely accepted to be of interfacial Rashba type, no topological parameters can be revealed. Nevertheless it is a convenient and non-destructive measurement tool for skyrmions’ motions.

Lorentz transmission electron microscopy (LTEM) is by far one of the most convincing tools for magnetic bubble and skyrmion imaging, with the moment orientations analyzed by the transport-of-intensity equation (TIE) [53, 54]. The LTEM intensity contrast is $I \propto |(\nabla \times \vec{m}) \cdot \vec{k}|$, implying that it is sensitive to only a curl of moments around the incident electron beam $\vec{k}$. Since $\vec{k}$ is usually parallel to the normal vector of a thin sample plate, the out-of-plane moments would appear dark, their directions can be inferred by knowing the moments of majority surrounding area is parallel to the magnetic field but the skyrmion/bubble core is antiparallel.

This way, LTEM can clearly reveal the topological parameters ($Q$, $Q_0$, $Q_b$) and distinguish among mixed-helicity (type-1) bubbles, trivial (type-2) bubbles, uniform-helicity Bloch-type skyrmions, antiskyrmions [55] as well as some intermediate transformations. Examples are given in table 1. Despite its resolution advantage, LTEM imaging is challenging for Néel skyrmions, since Néel skyrmions have $\nabla \times \vec{m}$ perpendicular to the sample’s normal vector and $\vec{k}$, thus requiring a sample tilt [56], and their typical ultrathin film hosts do not provide adequate interaction volume for LTEM.

On the other hand, small-angle neutron scattering (SANS) is the only imaging technique for magnetic textures in the reciprocal space, and it pioneered the discovery work of a true Bloch-type skyrmion in the year 2009 by Mühlbauer et al [3]. The SANS patterns can reveal long wavelength modulations of magnetic moments along a particular crystal orientation. By analyzing the relationship between scattering patterns with the directions of the magnetic field ($\vec{H}$) and incident neutron ($\vec{k}_0$), the magnetic phases can be deduced. Nevertheless, it is only suitable for dense skyrmion lattices and bulk samples.

In recent years, more varieties of non-destructive imaging techniques were developed, particularly complementing LTEM’s shortcoming on Néel skyrmions. Magnetic force microscopy (MFM) [68], spin-polarized STM (Sp-STM) [16, 28, 29] and nitrogen-vacancy magnetometry (NVM) [69, 70] are the prominent scanning probe techniques. Although Sp-STM offers much higher resolution compared to MFM and has done a great job in imaging atomic-scale Néel skyrmions such as the one in Fe/Ir(111), its reliance on tunneling current limits its usage within the class of metallic ultrathin films. NVM is believed to bring more freedom in imaging scale and was demonstrated to be non-destructive for defining single skyrmion number.

Having established a good understanding of the valuable aspects of skyrmions, the subsequent sections are dedicated to a thorough review on oxide skyrmions, which is a broad platform with numerous interesting physical aspects that offer extra freedoms of control.

2. Magnetic bubbles in centrosymmetric crystals stabilized by dipolar/demagnetization field

2.1. Dipolar or demagnetization contribution

At larger scale, a fifth term for dipolar/demagnetization energy $\sum_{i=x,y,z} \frac{\mu_0 N_i M_i^2}{2}$ can be added into the energy landscape (equation (2)) for materials with negligible DMI due to their centrosymmetric crystal structures, where the demagnetization field $H_{ij} = -N_i M_i$, $N_i$ is the demagnetization factor and $M_i$ is the saturation magnetization. For any thin film/plane [76], we can consider a cylindrical disc with radius $R$ aligning in the $z$-direction of a plate of thickness $L$. We have $N_z = \left( L + R - \sqrt{L^2 + R^2} \right) / L$, while $N_{x,y} = \left( \sqrt{L^2 + R^2} - R \right) / 2L$. Hence it is easy to see that this demagnetization term yields a shape anisotropy that favors in-plane magnetization for thin plates at a large limit of $R/L$ where $N_z \rightarrow 1$ and $N_{x,y} \rightarrow 0$. To produce vortex-shape magnetic bubbles, large uniaxial magnetic anisotropy $K_u$ is needed. In fact, these bubbles have surfaced since the 1970s, and they do not always fulfill equation (1) but may result in a richer and often uncontrollable varieties of topologies; i.e. unlike skyrmions stabilized
by DMI, their helicity $Q_h$ is not uniformly single-valued across the whole sample, but randomly mixed with clockwise (CW) and counterclockwise (CCW) handedness. In oxides, such bubbles have been found in manganites, garnets, hexaferrites and orthoferrites in the past few decades. They are well described by the wall-energy model [51, 52, 78, 79] (note that in Erg. unit: $\mu_0 = 4\pi \times 10^{-7}$, the energy quantities are divided by $10^{-7}$, $\mu_0$ replaced by $4\pi$):

- **Quality factor for bubble emergence**, $q = \frac{2K_u}{\mu_0M_s}$

- **DW energy per unit area**, $\sigma_w = 4\sqrt{J_gK_u}$

- **Characteristic length**, $l = \frac{\sigma_w}{\mu_0M_s}$

- **Optimal plate thickness**, $h < l$

- **DW thickness**, $w \sim \frac{l}{q} = \sqrt{\frac{J_g}{K_u}}$

- **Bubble size** $\approx 8l = 32\sqrt{J_uK_u}$

- **Reducing $h$ reduces $l$ and hence the bubble size.**

- If DMI of a particular sign is present, $\sigma_w = 4\sqrt{J_gK_u} - \pi D$ is modified by DMI [17, 80], causing the helicity $Q_h$ of one type to be more stable than the other.

- If DMI is present together with demagnetization, then reducing $h$ would cause enhancement in SOC, $K_u$ and $q$, such that the bubble phase space is enlarged.

### 2.2. Oxide thin-plate skyrmions with mixed helicities

Several magnetic bubbles stabilized by dipolar/demagnetization were found in the early 1970s. In oxides, these bubbles usually form in the magnetic insulator phases, which limits their use in electronics. Here we present a list of dipolar-stabilized oxide magnetic bubbles with their characteristics in Table 1, which were mainly studied by LTEM.

For $La_{1-x}Sr_xMnO_3$ (LSMO) at low Sr doping, the Jahn–Teller distortion of $e_g^2$ configuration from the parent $LaMnO_3$ is still partially active, promoting lifting of Kramer’s degeneracy and orbital ordering (OO), creating the FM insulating OO phase. This OO involving double-exchange among the $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals possibly contributes to large $K_u$ along $[001]_o$ [57]. In general, the well-known phase diagram for LSMO consists of:

- $0 < x < 0.1$: Canted AF for $T < T_N$, PM insulator for $T > T_N$

- $0.1 < x < 0.15$: FM insulator for $T < T_M = 150$ K, FM metal for $T_M < T < T_C$, PM insulator for $T > T_C$.

- $0.15 < x < 0.3$: FM metal at $T < T_C$, PM insulator at $T > T_C$.

A structural phase transition from orthorhombic ($T < T_S$) to rhombohedral ($T > T_S$) exists, where $T_S < T_C$.

- $0.3 < x < 0.6$: FM metal for $T < T_C$, PM metal for $T > T_C$

- $x > 0.6$: Canted AF for $T < T_N$, PM insulator for $T > T_N$

Generally, the external magnetic field ($H_{ext}$) and imaging electron beam $k$ vector should be aligned along the uniaxial easy axis with the largest $K_u$ to realize stripe domains (at zero field) and bubble formations (with small mT fields); otherwise, large in-plane collinear domains with narrow DWs will be found. Increasing the value of $H_{ext}$ too much along the easy axis also causes bubbles to shrink and vanish; this is intuitive since $H_{ext}$ is typically parallel to the circumference moments but antiparallel to the core moments. In LSMO ($x = 0.175$), rising above the structural phase transition temperature ($T_S = 190$ K), the rhombohedral structure is unable to provide sufficient $K_u$ for bubble formation, and crossing the phase boundary causes abrupt (first order) disappearance of bubbles. In the orthorhombic phase ($T < T_S$) at $[001]_o$ orientation, besides showing coexistence of mixed $Q_h = \pm \pi/2$ (CW and CCW) due to their degeneracy (figures 2(a)–(c)), type-2 bubbles with two counter-propagating Bloch walls (which are not skyrmion, $Q = 0$) can also be randomly found (figures 2(d) and (e)). By tilting the magnetic field slightly away from the easy axis towards the in-plane ($2^\circ$–$3^\circ$), the majority of the proper Bloch-type skyrmion-like bubbles ($Q = 1$) will transform into type-2 bubbles following some distortion of Bloch lines [58, 59, 64] (figure 2(j)). On the other hand, composite bubbles similar to biskyrmions also formed but one of the pairs has $Q = 0$ [58].

In $BaFe_{10.35}Sc_{1.6}Mg_{0.05}O_{3\delta}$ hexaferrite [64], although the stripe domains at zero field generally follow the expected sequence in a Bloch sphere, some repetitions in the form of 'oscillation' are spotted around $\pm M_{z}$ (figures 2(f)–(g)). Such complexity yielded three concentric CW and CCW rings with $M_{B}$ in one bubble when a field of 150 mT was applied (figures 2(h) and (i)). However, it still has $Q = 1$ but not a concentric Bloch skyrmion of $Q = 3$, since overall the moments

### Table 1. List of oxides hosting dipolar-stabilized bubbles. The abbreviations 'Orient.' = crystal plane normal to magnetic field, 'Thick.' = plate thickness, and 'Dia.' = bubble diameter. Here 'Bloch-type' refers to proper skyrmion-like bubbles with $Q = 1$. Type-2 bubbles have no net topological charge. Data adapted from respective journals with permissions.

| Name | Crystal structure | Orient. | Thick. (µm) | $T_{Curie}$ (K) | $T_{Bubblle}$ (K) | $\mu_0H$ (mT) | Bubble type | Dia. (µm) | Ref. |
|------|------------------|---------|------------|---------------|-----------------|--------------|-------------|-----------|-----|
| $La_{0.875}Sr_{0.125}MnO_3$ | Perovskite | (011)$_o$ | 0.05–0.25 | 180 | 100 | 280–360 | Bloch | 0.1 | [57] |
| $La_{0.825}Sr_{0.175}MnO_3$ | Perovskite | (001)$_o$ | 0.05–0.15 | 280 | 90–100 | 185–470 | Bloch, type-2 | 0.3 | [58–60] |
| $La_0.5 Ba_0.5 MnO_3$ | Poly-crystals | 0.07 | 300 | 300–360 | 0–150 | Bloch, dumbbell | 0.2 | [61] |
| $La_{1.37}Sr_{0.63}MnO_3$ | Layered perovskite | (001) | 0.1–5 | 100 | 20–46 | 350 | biskyrmion | 0.2 | [62, 63] |
| $Doped BaFe_{12}O_{19}$ | Hexagonal | (001) | 0.03 | 453 | 300 | 100–200 | Bloch, type-2 | 0.15 | [64, 65] |
| $R_2Fe_{12}O_{19}$ | Garnet | various | 11–25 | 570 | RT | 1–37 | No info | 2.5–20 | [66, 67] |
| $RFeO_3$ | Orthoferrites | (001)$_o$ | 28–76 | 645 (T$_N$) | RT | ~0.3–6 | No info | 19–190 | [50] |
only wrap around the Bloch sphere once. Such oscillation in the Bloch sphere is also encountered again in LSMO ($x = 0.175$) [58].

In the reported La$_{0.5}$Ba$_{0.5}$MnO$_3$ case [61], the polycrystalline random orientation could be the root cause for the persistence of skyrmion-like bubbles up to 360 K which is above $T_{Curie}$. One would note that the moment versus temperature ($M$–$T$) curve shows a significant non-vanishing moment above $T_{Curie}$, similar to the magnetic polaron or chirality fluctuation as observed before in La$_{1-x}$Cu$_x$MnO$_3$ [81, 82] as well as recently in SrRuO$_3$ [83] thin films, via the topological Hall effect (THE). The author suggested that there should be another $T_{Curie}$ (~350 K) accounting for inherent random potential by quenched disorder. In particular, the typical stripe domains expected at zero field in the other aforementioned system is absent, but random bubbles appear in a contrast-less FM background. Another interesting feature is the coexistence of bubbles at intermediate out-of-plane fields $H_z$, with some $M_z$ oscillations analyzed in (i) according to the number labels. (j) Tilting the magnetic field by an angle $\theta$ away from the out-of-plane causes topological transformation into type-2 bubbles. Reprinted with permission from [73]. Copyright (2016) American Physical Society.

We speculate that such spontaneous $Q$ sign reversal at a fixed field could be very common due to thermal fluctuation and low coercive field ($H_c$) near $T_{Curie}$, but should be absent at $T = T_{Curie}$ and large $H_c$.

In La$_{1.37}$Sr$_{1.63}$Mn$_2$O$_7$ with tetragonal structure and easy axis [001] studied by Yu [63], biskyrmions were unambiguously spotted (figures 3(g) and (h)). In biskyrmions, two $Q = 1$ but opposite-sign $Q_h = \pm \pi/2$ bubbles are merged together into a $Q = 2$ pair, sharing a common Bloch wall. In fact this is only possible when DMI is absent, and the dipolar field ensures that $Q_h = \pm \pi/2$ are degenerate. As a counterexample, two bubbles with $Q_h$ of the same sign cannot share an oppositely oriented Bloch wall. In addition, type-2 ($Q = 0$) bubbles also emerge at zero field if they are first subjected to a field-cooling procedure with a field of 150 mT. An attraction force between the Bloch walls of nearby CW and CCW helicity will align the biskyrmions in forming a polar single-domain lattice everywhere. One interesting feature occurs when the field is swept back to zero from 0.35 mT without reaching the annihilation field of 400 mT, when the biskyrmions will co-exist with stripes. Another amazing work in that paper demonstrated biskyrmion flow at an encouragingly low current density of $7 \times 10^7$ A m$^{-2}$, where the LTEM images for biskyrmions were blurred, but the stripe image still remained sharp and did not move. Hence, this again verified the intrinsic property of skyrmions having less pinning and lower $J_C$ compared to the helical stripes, consistent with equation (4) and the finding of [40].

3. Multiferroic skyrmions in Cu$_2$OSeO$_3$

3.1. Basic properties

The multiferroic Cu$_2$OSeO$_3$ single crystals are usually grown by chemical vapor transport with HCl carrier gas [84]. Cu$^{2+}$ with electronic configuration 3$d^9$5$e_g^3$ ($S = 1/2$) is the only species that contributes a magnetic moment with 0.5 $\mu_B$/Cu$^{2+}$ at (high-field) saturation. In a Cu$_2$OSeO$_3$ unit cell, Cu$^{2+}$ ions are surrounded by square-pyramidal or trigonal bi-pyramidal oxygen cages at a ratio of 3:1, yielding a three-up-one-down ferrimagnetic system (figure 5(a)) [12]. The crystal structure is non-centrosymmetric and contributes a strong Dresselhaus-type DMI.

Using SANS on bulk Cu$_2$OSeO$_3$ crystal, Adams et al realized that its magnetic ground state is not simply ferrimagnetic, but helimagnetic [85]. In SANS (figure 4(a)), the multi-$q$ helical phase produces a fourfold or twofold symmetric scattering pattern, with the modulation vector perpendicular to the magnetic field ($q \perp \mathbf{H}$). The single-$q$ conical phase produces a twofold symmetric pattern, but its distinct feature is $q||\mathbf{H}$, whereas a triple-$q$ skyrmion lattice would produce a unique sixfold symmetric pattern when the field and incident neutron are parallel but perpendicular to the modulation vectors ($H || k_0 \perp q$). Generally, by subjecting a magnetic field onto any crystallographic direction of Cu$_2$OSeO$_3$, a common phase diagram would be obtained (figure 4(b)); i.e. at a temperature far below $T_{Curie}$, helical stripes (with modulation wavelength, $\lambda_h$, of 50–60 nm) dominate the zero-
Correspondingly, the AC susceptibility at 58 K and kinetic. In the skyrmion A-phase within the phase space of 56–20 mT, applying a small \( \partial \) CCW and CW bubbles sharing a common Bloch wall, due to lack of DMI. (a)–(d) Reprinted by permission from Springer Nature Customer Service Centre GmbH.

Increasing stripe domains are randomly aligned and cancel out (\( T \)). Hence, the skyrmion phase exists at the intermediate-field regime, housed within the conical phase. Adams et al further discussed that although the temperature dependence of the helical phase in Cu$_2$OSeO$_3$ shows a second-order phase transition, its specific heat capacity shows a first-order phase transition with a latent heat peak (figure 4(c)). This indicates the onset of helical spin fluctuation slightly above \( T_{Curie} \), and is likely the reason for the enhanced magneto-dielectric response, thus ME coupling.

The seminal work of LTEM imaging on Cu$_2$OSeO$_3$ was performed by Seki et al with \( H||[110] \) and [111] (figures 5(b)–(e)). Since DMI is active, these skyrmions have uniform \( Q_0 \), unlike the dipolar-stabilized ones discussed in section 2. Cu$_2$OSeO$_3$ in thin-film form was also found to exhibit skyrmion phase space enlargement behavior (figure 5(f)) consistent with other B20 compounds [86]. The P$_{213}$ space-group (threefold rotation around [111]) should be non-polar, but the presence of \( M_{[111]} \) removes this symmetry and creates a net \( P_{[111]} \). Hence, the ME coupling of Cu$_2$OSeO$_3$ can be characterized by AC magnetometry, which is able to detect frequency-dependent resonances. As seen in figures 5(g) and (h), for the bulk Cu$_2$OSeO$_3$ at any temperature below \( T_{Curie} \) and zero field, the electric polarizations in the multi-\( q \) stripe domains are randomly aligned and cancel out (\( P = 0 \)). Increasing \( H||[111] \) causes \( P_{[111]}(H) \) to increase in a negative direction and reverse the sign to positive, which can be fitted by a parabolic \( P \propto M^2 \) trend into the conical phase. Finally, \( P_{[111]} \) saturates when \( M_{[111]} \) saturates into collinear ferrimagnetic. In the skyrmion A-phase within the phase space of 56–58 K and \( \sim 20–40 \) mT, \( P_{[111]}(H) \) shows a hump anomaly. Correspondingly, the AC susceptibility \( \chi' = \frac{\partial M}{\partial H} \) also shows a dip anomaly in both the helical and skyrmion phase spaces (figure 5(h)).

### 3.2. Magnetoelectric coupling, vibrational modes and metastable skyrmion phase

The mentioned parabolic P–M relationship agrees with the d-p hybridization model [87, 88]—at a cation–anion pair \( i-j \) where the moment resides at the cation site \( i \), and the polarization is related to the moment by \( P_j \propto (e_j \cdot M_i)^2 e_j \), where \( e_j \) is the bonding direction. Using angle-dependent \( M \) and \( P \) measurements, Seki et al verified that \( H||[111] \), \( H||[110] \) induces \( P_{[001]} \), while \( H||[001] \) causes \( P = 0 \) in all directions (figure 6(a)), which is consistent with \( (P_3, P_y, P_x) \propto (m_{010}m_{001}, m_{001}m_{100}, m_{100}m_{010}) \) [89]. Such ME coupling rule by symmetry as well as the parabolic relationship is applicable for all the helical, conical, skyrmion and collinear ferrimagnetic phases, but small differences in the coefficient exist in the parabolic relationship between the phases. Seki et al also calculated detailed mappings of the polarization and charge density of a Bloch-type skyrmion in different crystallographic orientations (figures 6(b)–(h)), which is beneficial for designing electric-field tunable skyrmions [87].

Next, the skyrmion vibrational modes (figures 7(a)–(c)) in Cu$_2$OSeO$_3$ can be probed by using FM resonance (FMR). The geometry used by Onose et al was a thin plate with surface normal at [110] and an oscillating AC magnetic field \( (H_{AC}) \) of the microwave aligned with the in-plane [110] direction [90]. A static DC field was then applied either onto [110] or
[110], hence forming the \( H_{\text{DC}} \perp H_{\text{AC}} \) or \( H_{\text{DC}} \parallel H_{\text{AC}} \) configurations. At out-of-plane \( H_{\text{DC}} \parallel [110] \), 40 mT is needed for saturation into the collinear phase, but only 20 mT is needed for saturation with in-plane \( H_{\text{DC}} \parallel [110] \), because a thin plate has shape anisotropy that favors in-plane easy axis and stronger demagnetization field at out-of-plane. At 57.5 K, SKLs are created at \( H_{\text{DC}} \parallel [110] \) within 14–32 mT and \( H_{\text{DC}} \parallel [110] \) with 5–15 mT, as judged from \( \chi'(H) \) dip anomalies. Correspondingly, the \( H_{\text{DC}} \perp H_{\text{AC}} \) configuration showed resonance peaks at 1.8 GHz for the multi-\( \varphi \) helical stripe domains (near \( H_{\text{DC}} = 0 \)) and conical phase (\( H_{\text{DC}} = 34–40 \) mT), 1 GHz in the skyrmion phase (\( H_{\text{DC}} = 14–32 \) mT), and 2.5 GHz in the collinear phase (\( H_{\text{DC}} = 100 \) mT) (figure 7(e)). For the \( H_{\text{DC}} \parallel H_{\text{AC}} \) configuration, the only resonance is at 1.5 GHz in the skyrmion phase (\( H_{\text{DC}} = 5–15 \) mT) (figure 7(f)). The 1 GHz resonance at \( H_{\text{DC}} \perp H_{\text{AC}} \) and 1.5 GHz at \( H_{\text{AC}} \parallel H_{\text{DC}} \) are the CCW sloshing rotation and breathing modes of skyrmion excitations, respectively.

Okamura et al employed a coplanar waveguide (CPW) (figure 7(d)) that was able to access both the

\[ k_{[110]}(\omega) \parallel (P_{[001]} \times M_{[110]}) \] and \( k_{[001]}(\omega) \perp (P_{[001]} \times M_{[110]}) \) setups, where \( k \) is the microwave propagation vector, while the \( H_{\text{DC}} \parallel [110] \) is fixed [91]. Within each setup, both \( H_{\text{AC}} \parallel [110] \) and \( H_{\text{AC}} \parallel [001] \) exist, which is an advantage of the compact CPW. Particularly, Okamura investigated the non-reciprocal directional dichroism (NDD) \( \Delta S_{ij} \), which is an advantage of the compact CPW. Particularly, Okamura investigated the non-reciprocal directional dichroism (NDD) \( \Delta S_{ij} \) when \( \Delta S_{ij} \) and \( \Delta S_{ji} \) are the absorption coefficients at opposite \( k \)-vectors. This is based on the concept that ME coupling causes the absorption intensity of linearly polarized light to depend on constructive or destructive interference. Firstly, in the collinear phase at large \( H_{\text{DC}} \), the NDD peak for the \( k_{[110]}(\omega) \parallel (P_{[001]} \times M_{[110]}) \) setup at 1.8 GHz shows sign reversal from positive to negative upon reversing the direction of \( H_{\text{DC}} \) from positive to negative. This is because upon reversing the signs of \( H_{\text{DC}} \) and \( M \), \( (P \times M) \) should have sign reversal, which can be understood from the fact that \( P_{[111]}(H_{\text{DC}}) \propto M^2 \) discussed earlier is a parabolic even function and does not reverse sign. However, the NDD for \( k_{[001]}(\omega) \perp (P_{[001]} \times M_{[110]}) \) is always zero and unresponsive to \( H_{\text{DC}} \) sign reversal, which is expected from the selection rule of microwave absorption. Secondly, in the skyrmion phase, absorption for the \( k_{[110]}(\omega) \parallel (P_{[001]} \times M_{[110]}) \) setup showed two close broad peaks at 1 GHz (NDD > 0) and 1.5 GHz (NDD < 0). These two peaks correspond to the CCW sloshing rotation and
breathing modes of skyrmions, respectively (figure 7(g)), and their co-detection is because both $\mathbf{H}_{\text{AC}}||[\text{110}]$ and $\mathbf{H}_{\text{DC}}||[\text{001}]$ components exist in CPW. Probing such NDD response (figures 7(h) and (k)) is direct evidence of ME coupling.

After confirming ME coupling, Okamura et al attempted ME cooling with both a magnetic and electric field, and induced a metastable skyrmion phase in bulk Cu$_2$OSeO$_3$ [92]. The setup was $\mathbf{H}_{\text{DC}}||\mathbf{E}_{\text{DE}}||[\text{111}]$ to access the strongest ME coupling, which is expected to tune the uniaxial anisotropy by $\Delta K_u \propto -P_{[\text{111}]}.E_{[\text{111}]}.m_{[\text{111}]},E_{[\text{111}]},$, which can be a form of VCMA. Hence a positive electric field will enhance $K_u$, and thus enlarge the skyrmion phase space (figure 8(a)). A suitable ME cooling scheme (figure 8(b)) is achieved by using a small magnetic field (35 mT) within the bulk A-phase skyrmion phase and electric field of $+30$ kV cm$^{-1}$, and the metastable skyrmion phase was shown to persist down to a low temperature of $\sim 10$ K (figure 8(c)). This is evidenced from the presence (and absence) of the 1.6 GHz microwave absorption peak corresponding to CCW skyrmion sloshing rotation mode when ME cooling is applied (and removed), while the other absorption peak at 2.8 GHz corresponds to the conical phase. After creating the metastable phase, the phase continuity between the metastable and normal skyrmions can be broken, starting from $\sim 54.5$ K, by switching off or reversing the polarity of the electric field (figure 8(d)). The phase space overlapped between the metastable skyrmion at zero electric field and normal skyrmion at $+30$ kV cm$^{-1}$ at $\sim 55$ K is thus identified as the region where non-volatile switching between the skyrmion and conical phases can be demonstrated (figure 8(e)).

As a closing remark for this section, Cu$_2$OSeO$_3$ remains the only host for multiferroic skyrmions thus far, and its complex magnetic phase diagram has continued to produce new physics discoveries. However, its low $T_{\text{Curie}}$ and the difficulty in making device-integrable thin films would limit its practical application. On the other hand, multiferroic BiFeO$_3$ with cycloidal AF modulation wavelength of $\sim 64$ nm, high Néel temperature of $\sim 643$ K and high ferroelectric Curie temperature of $\sim 1083$ K would likely be the next candidate in the search for multiferroic skyrmions [93–95].

4. Unconventional skyrmions in bulk crystals

This section introduces two peculiar examples that challenge the usual belief that all bulk non-centrosymmetric crystals should host Bloch-type skyrmions. Firstly, in general, crystal structures with $C_{nv}$ symmetry are polar and would satisfy the criterion on hosting Néel-type skyrmions in bulk crystals, such as GaV$_4$S$_8$ ($C_{nv}$) [96] with $T_{\text{Curie}} \sim 13$ K and skyrmion phase at 11.6 K, as well as VOSe$_2$O$_5$ ($C_{nv}$) [97] with $T_{\text{Curie}} \sim 8$ K...
and skyrmion phase at 7.5 K. Their magnetic phase evolutions in \(H-T\) phase space are similar to the phase diagrams of commonly known Bloch-type skyrmions, but with ‘cylindrical’ replacing ‘helical’, while the conical phase is absent. In SANS for VOSe\(_2\)O\(_3\), the incident neutron direction is \(k_\perp l\perp c\)-axis. The polar \(c\)-axis of the material has DMI vector \(D_{1c}\) causing orientation confinement of the magnetic moment modulation vector \(q\) onto the in-plane. At zero-field cooling (ZFC), the SANS pattern is fourfold symmetric but slightly blurred, implying multi-\(g\)-domains. After field training \(H_{\parallel l\perp c} = 12\) mT and SANS measurement at 0 mT, a twofold symmetric sharp SANS pattern appeared, which is understood as ordered cylindrical stripes. Both the cylindrical stripes at zero-field and Néel skyrmion phase at \(H_{\parallel l\perp c} = 1.5\) mT show valleys/depressions in AC magnetometry \(\chi'(H)\) similar to Cu\(_2\)OSeO\(_3\), as well as the imaginary part \(\chi''(H)\). A slight difference in SANS patterns between the Néel skyrmion phase in GaV\(_4\)S\(_8\) and VOSe\(_2\)O\(_3\) can be noted: in GaV\(_4\)S\(_8\), clear fourfold and sixfold patterns are separated at \(H||[001]\) and \(H<111>\), respectively (figure 9(a)), but in VOSe\(_2\)O\(_3\), a superposition of one set of fourfold-symmetric, and two sets of sixfold-symmetric (30°-offsetted) patterns can all be found at \(H||[001]\) (figures 9(c) and (d)).

Secondly, the broken inversion symmetry at the surface of a non-centrosymmetric crystal structure would contribute an additional \(C_{nv}\) symmetry and create a chiral surface twist/reconstruction, resulting in a gradual transformation or intermediate state between Néel- \((Q_h = 0, \pi)\) and Bloch-type \((Q_h = \pm \pi/2)\) skyrmion along its depth profile. This is true if the sample is made thinner than the critical thickness of 8.17 \(\lambda_h\), and is also another mechanism of skyrmion phase-space enlargement in thin films of B20 compounds. This was first discussed by Rybakov et al [99] and was observed directly by Zhang et al in Cu\(_2\)OSeO\(_3\) using circular dichroism in resonant elastic x-ray scattering (CD-REXS) [98]. The resulting CD-REXS pattern on a skyrmion lattice is also sixfold symmetric, reminiscent of that of SANS, but the CD bisects the pattern into \(\pm\) signs, enclosing a zero-CD vector (extinction direction) that may rotate following the skyrmion’s \(Q_h\).

Zhang et al first used theoretical calculations to predict that \(Q_h\) approaches 134° and 46°, respectively, at the top and bottom surfaces of the Cu\(_2\)OSeO\(_3\) thin plate for a particular out-of-plane magnetic field direction \(B_z\) (figure 9(e)). This agrees very well with the observed CD-REXS result, where upon the reversal of \(B_z\), the surface \(Q_h\) changed from 129° to \(- 51.7°\) (figures 9(f) and (g), top and bottom panels). The \(B_z\) sign reversal is equivalent to flipping the crystal plate upside down, since in reality the CD-REXS signal is averaged over their limited x-ray penetration depth \((L_z \sim 98\) nm) and is unable to detect \(Q_h\) from purely the bottom surface. Zhang et al also managed to extract and verify the expected bulk \(Q_h = 91°\) (figure 9(g), middle panel), which is located far away from both surfaces. This is achieved by first recognizing that \(L_Z \gg L_p\) where \(L_p \sim 7.1\) nm is the decay length of the chiral surface twist, and tilting the magnetic field slightly away (15°) from the surface normal while keeping the incident x-ray at normal direction. This way, the skyrmion cylinder is tilted, and a higher fraction of the bulk signal will be picked up by the same \(L_Z\). Finally, a subtraction between the tilted and normal incidence signals yielded the pure bulk \(Q_h\).

Figure 9. SANS pattern (a) and MFM image (b) of skyrmion phase in GaV\(_4\)S\(_8\), where the color scale of (b) represents magnetic interaction energy. (c) SANS pattern of skyrmion phase in VOSe\(_2\)O\(_3\), interpreted as a superposition of three magnetic textures in (d), where tri, SKL, IC-1 and IC-2 denote trigonal, skyrmion lattice, cylindrical and square skyrmion lattice, respectively. (e) Calculated depth profile of helicity in left- and right-handed Cu\(_2\)OSeO\(_3\). CD-REXS data (f) and their fitting analyses (g) for the right-handed Cu\(_2\)OSeO\(_3\) skyrmion, where the field inversion can reveal the surface twist at the top and bottom surfaces (top and bottom panels). Middle panel: the bulk state is represented by the cyan curve after subtraction. Inset of (g) indicates the length of skyrmion cylinders \((d_z > d_i)\) being probed. (a), (b) Reprinted by permission from Springer Nature Customer Service Centre GmbH: Nature Materials [96] (2015). (c), (d) Reprinted figure with permission from [97]. Copyright (2017) by the American Physical Society. (e), (f) Reprinted figure with permission from [98]. Copyright (2018) by the American Physical Society.

5. Hall effects in magnetic materials (Interlude)

THE arises from spin-polarized conduction electrons deflected off non-coplanar magnetic moment textures. A simple picture of this process is that during the hopping process across non-coplanar magnetic textures, conduction electrons have to constantly adjust and align their spin reference frame to the local magnetic moment due to strong Hund coupling between the free spin and moments \((- J_{\parallel \hat{\sigma}_i \cdot \hat{m}_i})\), hence gaining a net
real-space geometric phase. In the low hole-doped manganites, at the phase transition region from FM metal to PM insulator, a hump-shaped Hall effect emerged. Ye et al. [100], Chun et al. [82] and Calderon and Brey [101] postulated the existence of skyrmion strings or chirality fluctuation, using the language of second quantization for the tight-binding model:

$$\hat{H} = -t \sum_{i,j} \cos \left( \frac{\theta_{ij}}{2} \right) e^{\left( \frac{\pi}{\theta} + \pi A \right)} c_i^\dagger c_j + \lambda_{\text{soc}} (k \times \nabla V) \cdot \hat{\sigma} + g \mu_B H \cdot \sum_i m_i$$

(5)

where $c_i^\dagger$ and $c_i$ are creation and annihilation operators, $\cos (\theta_{ij}) = \mathbf{m}_i \cdot \mathbf{m}_j$, $B = \nabla \times A$ is the external magnetic field, and the phase factor $\phi = 2\tan^{-1} \left( \frac{\hat{a}_i \cdot (\hat{a}_i \times \hat{a}_j)}{1 - \hat{a}_i \cdot \hat{a}_j} \right)$ contains the Pontryagin charge. The two phase terms $\cos (\theta_{ij}) + \frac{\pi}{\theta} A_{ij}$ correspond to the THE and ordinary Hall effect (OHE), while the second and third terms account for the SOC and Zeman effect, respectively. We can understand that an effective magnetic field \( 9 \times \hat{V} \hat{\phi} = \nabla \times \nabla \hat{\phi} = \frac{\mu}{\pi} \hat{m} \cdot (\hat{\partial} \hat{m} \times \hat{\partial} \hat{m}) \) emanates from the magnetic textures. This produces the total Hall resistivity $\rho_{\text{THE}} = (B + b_{\text{eff}}) / n_e$, showing the equivalent form of OHE and THE. On the other hand, Bruno et al. [102] and Everschor-Sitte and Sitter [103] used a simpler Schrodinger Hamiltonian for electrons on a topological magnetic texture and unitary transformation for the strong Hund coupling, and arrived at the same insights for THE. A spin-polarization $P_s = \frac{D_{\text{OHE}}(\varepsilon_F) - D_{\text{OHE}}(\varepsilon_F)}{D_{\text{OHE}}(\varepsilon_F)}$ correction factor was also added into $\rho_{\text{THE}}$, where $D_{\text{OHE}}(\varepsilon_F)$ is the spin-dependent DOS at Fermi level.

For the anomalous Hall effect (AHE) that requires out-of-plane magnetization, three distinct mechanisms have been discussed [104–108]. The intrinsic k-space Berry curvature effect can be understood from the semiclassical approach $v_{\text{sc}} = \frac{1}{\hbar} [\hat{E} x, y] = \frac{1}{\hbar} [E_z \times \Omega_{\text{loc}}, k]$), where $\Omega_{\text{loc}}(k) = \nabla V \times u_\alpha(k) \nabla u_\alpha(k)$ is the Berry curvature, $u_\alpha(k)$ is the Bloch wave function, and the x- and y-directions are parallel and perpendicular to the applied electric field $E$, respectively. For the side-jump mechanism, Nozières and Lewiner [109] described how SOC-assisted electron scattering off an impurity potential $V$ yields a transverse velocity $v_\perp = \frac{\hbar}{2 \pi} (\nabla V \times \hat{\phi})$. Meanwhile, the skew scattering across the Fermi golden rule—where the transition probability between $|k, s\rangle$ and $|k', s'\rangle$ states is asymmetric for left and right deflections when SOC is taken account: $W_{k,s,k',s'} = \frac{\hbar}{2 \pi} [k, \mathbf{V} \mathbf{m} k', s''] \delta (\varepsilon_{k,s} - \varepsilon_{k',s'})$, where $k, \mathbf{V} \mathbf{m} k', s' = V_{k,k'} \left( \delta_{s,s'} + \frac{\hbar}{2m} \frac{\delta}{\delta \varepsilon_{k,s'}} \right) (\mathbf{\sigma} \delta s' \times k') \cdot k$. Typical experiments scale AHE conductivity with mean free time $\tau$. For both intrinsic and side-jumps, no dependence on $\tau$ is expected, hence $\rho_{\text{sc}} \approx \frac{\hbar}{2 \pi} \proportional \rho_{\text{AHE}}^2$; yet for skew scattering, $\rho_{\text{sc}} \propto \tau^{-1}$ leads to $\rho_{\text{sc}} \propto \rho_{\text{AHE}}$.

For all three mechanisms of AHE, SOC is directly involved in electron deflection, but is not necessarily involved in producing THE when magnetic textures are present [110]. Ideally, for a magnetic material with out-of-plane uniaxial anisotropy and hosting hexagonal-closed packed magnetic SKL, the total Hall resistivity is

$$\rho_{\text{THE}}(B) = R_0 B + (\alpha \rho_{\text{AHE}} + \beta \rho_{\text{M}}) \lambda_{\text{soc}} M + R_a P \frac{\Omega_{\text{L}}} {\varepsilon \tau_{\text{int}}}$$

(6)

where the first, second and third terms are OHE, AHE and THE, respectively, $R_a = 1 / n_e$, and $\alpha, \beta$ are fitting parameters. The above picture for THE is accurate when $J_H$ is strong and electron hopping is adiabatic (slow), providing adequate time for the fast $\hat{\sigma} \cdot \mathbf{m}$ interaction. Nakazawa et al. recently improvised the THE theory by accounting for non-adiabatic and/or non-local $\hat{\sigma} \cdot \mathbf{m}$ interactions [111]. This way, THE behavior at strong electron-electron correlation and high effective mass (especially true in oxides) can be more accurately modeled. It is customary to understand the field dependence of $\rho_{\text{OHE}}, \rho_{\text{AHE}}$ and $\rho_{\text{THE}}$. For $\rho_{\text{OHE}}(H)$, hysteretic square loops can be fitted with the Langevin function: $L(H) \propto \left\{ \cos [\frac{\mu}{\Omega_{\text{OHE}}} (H \pm H_c)] - \frac{H_c \Omega_{\text{OHE}}}{\mu} \right\}$ which usually follows the shape of magnetization field loops. Meanwhile $\rho_{\text{THE}}(H)$ should follow the phase evolution of skyrmions or bubbles—typically stripe domains exist and no skyrmion at zero field; skyrmions would reach the smallest size and densest lattice packing at the intermediate field $H_{\text{peak}}$, and finally vanish into the collinear ferromagnet at large field. Hence $\rho_{\text{THE}}(H) \sim \frac{\varepsilon[H]}{(1 + H/H_{\text{peak}})^\alpha}$ showing the hump/dome feature is a good phenomenological fit [83, 112].

THE also appears in pyrochlore frustrated antiferromagnets [113–115]. Theoretical advancements in this field have enabled several insightful Monte Carlo simulations [112, 116, 117]. In the next section, we see that the hump-shaped Hall features in SrRuO$_3$ ultrathin films have caused heated debates over its representation of the true THE signal or partial cancellation of two oppositely signed AHE loops. Nevertheless, without proper imaging, Hall effects do not distinguish between the skyrmion/bubble types, if any of them exist at all.

6. Topological Hall effect and skyrmion-like bubbles in perovskite oxide thin films

6.1. Basic properties of SrRuO$_3$ and SrIrO$_3$

Bulk SrRuO$_3$ (SRO) perovskite has a FM Fermi liquid ground state by minority-spin double-exchange (figure 10(a)) between the electrons in the degenerate 4$d_{xy}$ bands, with $T_C = 150$ K [118] and pseudocubic lattice parameter ($a_{pc}$) of ~3.93 Å. Due to the high crystal field splitting $\Delta_{\text{rya}} \approx 2.62$ eV [119] between 4$d_e$ and $4d_g$ bands of Ru$^{4+}$, the $e_g$ band is empty, and the $t_{2g}$ band has two unpaired spins and two paired spins, contributing to $S = 1$. With SOC between $S = 1$ and $L = 1$, a final magnetic moment of ~1.6 $\mu_B$/Ru$^{4+}$ is obtained [120], together with large PMA for SRO thin film that is stronger than the in-plane shape anisotropy. Each $t_{2g}$ ($n^{th}$) band with non-trivial topology in k-space also carries a non-zero Chern number [121] $Z_n = \frac{1}{\pi B} \int \Omega_{\text{loc}}(k) \mathbf{d}^2 \mathbf{k}$, therefore endowing large intrinsic AHE [122, 123]. To be specific, the AHE...
loop has a negative sign due to the minority-spin double-exchange, with a $P_s$ value of around $-9.5\%$ as measured by point-contact Andreev reflection [124]. For the monoclinic (m-) phase of SRO thick films, the optimized growth process is usually via pulsed laser deposition (PLD) at standard 100 mTorr (13 Pa) O$_2$ pressure, 600 °C–680 °C temperature and cooled down at >200 Torr of oxygen ambiance to remove oxygen vacancies (V$_O$). Hence, its properties could be bulk-like, with octahedral tilt described by Glazer notation a ’b+c’. In the ultra-thin regime of SRO along the (001) orientation, lifting of degeneracy among $t_{2g}$ bands and preferential occupation of the energetically lower $d_{xy}$ orbital will promote AF superexchange among Ru$^{4+}$ species (figure 10(b)) [125, 126]. Tight-binding calculations with the SRO$^{2g}$ band structure have predicted the intrinsic AHE sign reversal due to reduction of the Ru moment [121]. In addition, reducing the growth pressure and introducing $V_O$ will expand the $e_g$-axis [127], suppress the octahedral tilt and form the tetragonal (t-) phase SRO with a Glazer notation of a’c’c’. Correspondingly, a positive-sign AHE loop will dominate up to even large SRO thickness, which could be associated with extrinsic AHE mechanisms by a careful scaling analysis but is not yet done.

For bulk SrIrO$_3$ (SIO) with $\theta_{xc} \sim 3.94$ Å, the Ir$^{4+}$ (5d $t_{2g}$) $e_g$-orbitals in the perovskite crystal field also has empty $e_g$ due to the high $\Delta_{xyy}$ of $\sim 3$ eV. Strong SOC $\sim 0.5$ eV splits the $t_{2g}$ band into the upper $J_{\text{eff}} = 3/2$ and lower $J_{\text{eff}} = 1/2$ bands, which are entangled states of the $t_{2g}$ orbitals (figure 10(c)). The three pairs of undisorted $t_{2g}$ wavefunctions ($J, \pm m_f$) are thus ($\sigma$ denotes spin up/down):

$$\frac{1}{2}, \pm \frac{1}{2} = \sqrt{\frac{1}{3}}(|d_{xz}, \sigma_\pm \pm i|d_{yz}, \sigma_\pm \pm \sigma_{xy}, \sigma_\pm )$$

$$\frac{3}{2}, \pm \frac{3}{2} = \sqrt{\frac{1}{2}}(|d_{yz}, \sigma_\pm \pm i|d_{xz}, \sigma_\pm )$$

$$\frac{3}{2}, \pm \frac{1}{2} = \sqrt{\frac{1}{6}}(|d_{xz}, \sigma_\pm \pm i|d_{yz}, \sigma_\pm - 2|d_{xy}, \sigma_\pm ) .$$

The five electrons Ir$^{4+}$ thus fully fill the $J_{\text{eff}} = 3/2$ band by four electrons, and half-fill the $J_{\text{eff}} = 1/2$ Kramer’s doublet by one electron [128, 129], and can be equivalently represented by a one-hole state with the $|\frac{1}{2}, \pm \frac{1}{2}\rangle$ wavefunction. This way, bulk SIO perovskite is a PM semimetal with no magnetic long-range order at ground state [130, 131]. The SIOhases a Dirac-like (massless) electron dispersion but heavy hole dispersion coinciding at the $\Gamma$-point. The half-filled $J_{\text{eff}} = 1/2$ band of Ir$^{4+}$ is sensitive to bandwidth (W) tuning, and a large Mott–Hubbard correlation factor $U/W$ would open a Mott gap in the $J_{\text{eff}} = 1/2$ band into upper and lower bands, akin to Sr$_2$IrO$_4$. If a tetragonal distortion is present, the degeneracy among the $d_{xz}, d_{yz}$ and $d_{xy}$ orbitals in superposition will be lifted, according to the usual crystal-field (repulsion between $B$-site $d$-orbital and anion) concept [132]. Particularly, SIOand CaIrO$_3$ ultrathin films with suppressed inter-plane coupling may easily transform to weakly ordered quasi-two-dimensional chanted antiferromagnets [133–135]. Such magnetic phase transition is of higher order, such that a clear $T_C$ cannot be clearly identified according to the Mermin–Wagner theorem [134, 135]. In addition, according to Zhong’s calculation, Ir$^{4+}$ is a potent electron donor while interacting with another perovskite B-site cation species [136]. When such electron transfer occurs, Ir$^{4+}$ becomes hole-doped and a weakly FM spin-liquid phase may arise assisted by the proximity effect with another magnetic material that co-forms the interface [137].

6.2. Analyses and tuning of Hall effects in SRO/SIO bilayers

Matsuno et al were the first who paid attention to the THE signal of SrRuO$_3$ (2uc)/SrRuO$_3$ (m = 4–7 uc) grown epitaxially on SrTiO$_3$ (001), with tuning of SrRuO$_3$ thickness (m-uc) [138]. The expected trends with reducing SRO thickness are carrier localization, reducing $T_{\text{Curie}}$, and an AHE sign reversal from negative (bulk-like) to positive, which scales with diminishing magnetization. Appreciable humps resembling THE emerged only within 4–5 uc SRO, from 5 K to 80 K (figure 11(a)). Decomposition of the total Hall effect into OHE, AHE and THE components following equation (6) was done, by fitting OHE to a high field background, and AHE to follow the $H_T$ and square loop shape extracted from magnetometry. THE is thus showing an antisymmetric hump shape. Using values for $R_s$ from the high-field Hall slope, literature-reported $P_s = -9.5\%$ and $Q = 1$, the skyrmon diameter was estimated to be around 5–15 nm from THE. The diminishing THE trend with increasing SRO thickness is consistent with their micromagnetic simulation using the Hamiltonian in equation (2), by considering the weakening effective DMI by $D_{\text{eff}} = D/m$. The authors provided real-space imaging by low-temperature MFM to support the presence of skyrmon-like bubbles near $H_T$.

Subsequently, electric field tuning of THE signals became a highly pursued aspect. Ohuchi et al attempted back-gating via STO(001) substrate (figure 11(b)), and STO is suitable due to

![Figure 10](image-url)

Figure 10. (a) FM minority-spin double-exchange and (b) AF superexchange in SRO. (c) Effect of crystal field $\Delta_{xyy} = 10$ Dq and SOC splittings on electronic band structures for 5d B-site perovskites. (c) Reprinted figure with permission from [128]. Copyright (2008) by the American Physical Society.
its moderately high dielectric constant. We would expect the electric field tuning to be effective on material with low carrier density. Hence, Ohuchi’s result showed that it is effective when an ultrathin (2 uc) SIO is inserted between STO and SRO. The MFM data in the inset show the appearance of bubbles at THE emergence. (b) The back-gating method used by Ohuchi et al with silver paste as bottom electrode and SRO as top electrode. (c), (d) Gating tuning effect is absent in the structures with SRO adjacent to STO(001) where no THE humps could be discerned, but is successful in SRO/SIO/STO(001) (e) for both AHE and THE. From [139]. Reprinted with permission from AAAS. (b), (e) Reproduced from [138]. Reprinted with permission from Elsevier.

Figure 12. (a) Hall effect showing strong THE hump signal, and (b) the MFM image corresponding to the THE hump emergence. (c) MFM image of random stripes at 10 K and zero field after zero-field cooling. Simulated (d) CCW and (e) CW Neél walls with a fixed value 1.288 μB Ru⁻¹ of magnetic moment. Simulated image for Bloch/Ising walls is not shown here but is available in the original paper. (f) A summary of the MFM contrasts at the cross-sectional lines of the respective images. Reprinted with permission from [140]. Copyright (2019) American Chemical Society.
positive gate voltage led to sign reversal from negative to positive AHE. Qin et al also noted that the single-layer t-phase SRO showed wider THE temperature range and electric field tunability, as compared to m-phase SRO.

Wang et al also fabricated BaTiO$_3$ (BTO) thin film on a single-layer SRO Hall bar to demonstrate the non-volatile switching of THE [144]. The ferroelectric polarization ($P$) of BTO is switched by piezoresponse force microscopy (PFM) at room temperature, followed by cooling down the sample to low temperature for Hall effect or MFM measurement (figure 13(d)). Firstly, their result showed that there is an extra BTO/SRO interface contribution to the observed THE signal even without polarization switching (figure 13(c)). Secondly, the area density of magnetic bubble evolution with magnetic field can be quantified by MFM imaging, particularly reaching the densest at THE peaks. Using the same approach in equation (6), the skyrmion diameter was estimated to be $\sim 55$ nm. Thirdly, by increasing the ratio of $P_{\text{DOWN}}$ to $P_{\text{UP}}$ domains within the Hall bar channel, both AHE and THE can be reduced (figure 13(e)). Using a density-functional theory (DFT) calculation supported by high-resolution scanning transmission electron microscopy (HR-STEM), the authors claimed that the Ru–O bond angle and length at the BTO/SRO interface is slightly changed by the $P_{\text{BTO}}$ switching. In other words, interface SRO becomes polar where the Ru$^{4+}$ is displaced away from the oxygen cage center in the same direction as Ti$^{4+}$ displacement. In a bent Ru$_1$–O–Ru$_2$ bond, we can understand that the DMI coefficient vector $D_{12} \propto (r_1 \times r_2)$, where $r_1$ and $r_2$ are the Ru–O bond vectors and are perpendicular to $D_{12}$. In addition, since the increased bond bending only happens at the interface, the DMI does not cancel out as in centrosymmetric bulk perovskites. Hence, $P_{\text{UP}}$ ($P_{\text{DOWN}}$) will increase (decrease) the bond bending and magnitude of $D$, thus tuning the skyrmion density and size. In addition, since Ru–O–Ru bond bending cannot be inverted due to the intrinsic octahedral tilts/rotation, the $D$ sign reversal does not occur. Conversely, if $D$ were to invert sign, we could expect changes only in the skyrmion’s helicity $Q_h$ but the topological charge $Q$ and thus $R_{\text{THE}}$ should maintain the same sign at a fixed magnetic field. We believe that this electric field gating approach via ferroelectric switching is closer to true atomic-scale DMI tuning compared to the earlier back-gating and ionic liquid gating approaches.

Almost concurrently, Wang et al provided an insight into THE from a quite different phase space, which is near and above $T_{\text{Curie}}$ in a single-layer SRO and V-doped Sb$_2$Te$_3$ topological insulator (TI) [83]. This corresponds to the random chirality fluctuation regime similar to the earlier...
La$_{1-x}$Ca$_x$MnO$_3$ cases [82] as discussed in the previous section (figures 14(a) and (b)). Judging from the PLD growth parameter for SRO of 650 °C and 0.25 mbar, the structural phase should be m-phase. Due to an additional capping layer of 2 uc STO on the 6 uc SRO (figure 14(c)), out-of-plane $K_u$ is enhanced, and no THE hump signal could be discerned at low temperatures; however, the THE signals spotted around $T_{Curie} \sim 116$ K are non-hysteretic since the $H_c$ at that regime is vanishingly small (figures 14(e) and (f)). The THE antisymmetric humps can be fitted well with $\rho_{\text{THE}} (H) \sim \frac{M(H)}{1 + (H/H_{\text{eff}})^2}$ where $M(H)$ should take the form of the Langevin function, and a $D_{\text{eff}}$ of $\sim 0.2$ meV was extracted. With similar Hall data processing techniques, the THE signal is maximum at $T_{Curie}$ but decays fast around it. A similar result was also obtained in the V:Sn:Te$_3$/STO(111) by tuning the temperature and electric field gating voltage, although with an opposite-sign $\rho_{\text{THE}}$. The author supported these trends with Monte Carlo simulation for chirality fluctuation by: exp $\left( -\frac{\phi_{ijk}}{K} \right) = \frac{1}{\sqrt{2(1 + \phi_{ijk})(1 + \phi_{jik})(1 + \phi_{kji})}}$ and $Q = \frac{1}{2}\sum \phi_{ijk}$ where i,j,k are indices for neighboring moments in a triangle (figure 14(d)).

Most recently, hydrogen doping was injected into SRO thin film by liquid ion-doping, as performed by Li et al, causing structural lattice expansion and weakened ferromagnetism [145]. Surprisingly, THE signals also emerged at 2–100 K by an optimal gate voltage.

6.4. Emergence of THE in Ce-doped CaMnO$_3$ with strong electron correlations

On the other hand, Vistoli et al found strong THE signals in perovskite Ce$_x$Ca$_{1-x}$MnO$_3$ with Ce$^{3+}$ doping of $0 < x < 5\%$ [146]. Such doping creates a weakly FM metallic phase with $T_{Curie} \sim 100$ K (figures 15(a) and (b)), which has been a lattice-matched bottom electrode material widely used for earlier BiFeO$_3$-based ferroelectric tunneling junctions (FTJ) [147]. Particularly, for $x = 4\%$, the THE signal spanned across a wide temperature range of 15–75 K (figure 15(c)). Their low-temperature MFM images support the presence of magnetic bubbles and have almost the same quality as those described by Matsuno and Meng et al mentioned above. Since there is no obvious sharp interface with a heavy metal providing SOC and Rashba-type DMI, nor a non-centrosymmetric crystal structure, the author used the earlier approach in dipolar/demagnetization-stabilized bubbles (as discussed in section 2). Namely, the bubble diameter was estimated by $32\sqrt{\mu_{\text{M}}} T_{\text{eff}}$ to be $\sim 250$ nm, agreeing with the MFM measurement, where $J_{so}$, $K_u$ and $M_s$ were calculated from $T_N$, magnetometry loops and the moment-canting angle, respectively. A particular strong selling point of this paper is the superfast scaling of the THE signal with carrier density ($n$) (figure 15(d)), which was found to deviate significantly from the simple Bruno model of strong $\hat{\sigma}$-$\hat{n}$ (adiabatic) interaction regime (equation (6)). Particularly at low Ce doping, $\rho_{\text{THE}}$ is enhanced by $\propto n^{-8/3}$ together with the enhanced electron effective mass $m^*$ due to strong electron correlation, consistent with the ‘weak coupling, localized magnetic texture’ regime as theorized by Nakazawa et al [111] (as well as the supplementary information of [146]).

6.5. The alternative Bi-AHE interpretation

Several other groups have provided an alternative interpretation that the hump signals are artifacts of partial cancelation between two AHE loops, which can be modeled by two pairs of overlapping Langevin functions $L_{\alpha\beta}(H)$ with opposite sign of coefficients and different $H_{\alpha,\beta}$ (figure 16(a)). This way, at the hump emergence, only trivial Ising-like (collinear) DWs exist instead of non-coplanar textures. In particular, Kan et al [148] and Groenendijk et al [121] performed such Bi-AHE analyses on the hump-shaped Hall features of the SRO-based heterostructures. Using a DFT calculation combined with a tight-binding model for Wannier function interpolation, the possible AHE sign reversal at interfaces between Ru/Ir, Ru/Ru and Ru/Ti was calculated, as shown in figure 16(b). In addition, they performed minor Hall loop measurement (figures 16(c) and (d)) and revealed that upon reaching the hump peaks and without reaching high-field saturation, the peak $\rho_{\text{xy}}$ value can be retained by sweeping back to zero field. We opine that while such behavior looks very supportive of the picture of partial cancelation of two AHE loops, it could also be hysteretic behavior of skyrmion-like bubbles, i.e. skyrmions can stabilize at the phase space of multi-$q$ helical/cycloidal stripes.

This debate points to the importance of demanding a reliable magnetic imaging technique. In parallel to the Bi-AHE
between the two AHE contributions cause incomplete cancellation (figure 16(e)) when the two different magnetic domains are antiparallel.

As a concluding remark for this sub-section, notably, obtaining high-resolution MFM images of skyrmions is very challenging, and deducing the actual skyrmion type (Néel/Bloch) remains unclear or indirect. To probe deeper, the diamond NVM capable of single-spin sensitivity in constructing a smoothly varying magnetic texture, as achieved by Dovzhenko et al. [69], would be very useful. Alternatively, if oxide thin films can be detached from the thick substrates by a suitable epitaxial lift-off technique [150], LTEM would be again useful, either with or without slight tilting of the electron beam vector $k$ away from the sample’s surface normal for imaging Néel- [56] or Bloch-type skyrmions, respectively. The possibility of measuring the interface DMI strength using Brillouin light scattering (BLS) [151] in oxide heterostructures is also almost ruled out since the reflective surface as required in BLS is absent, unlike in true metals. Furthermore, in the work on electric-field tuning of Hall effect signals by Ohuchi et al., Qin et al and Wang et al. [139, 143, 144], the possibility of $V_0$ movement in SRO that contributes to AHE tuning has not been ruled out. Nevertheless, tuning the surface bond-bending angle of a non-multiferroic FM material via polarization switching of an adjacent ferroelectric film is certainly a good strategy of modulating the interface DMI strength. In short, we believe the THE phenomenon in oxide thin films remains as an emergent field at infancy stage, with more challenging physics to be explored.

6.6. Skyrmions at ferrimagnetic oxide/heavy metal bilayers

Here, we review iron-based ferrimagnets bringing together two valuable properties—having vanishingly small $M_s$ near its compensation temperatures as well as high (>300 K) magnetic ordering temperatures. Assisted by an adjacent ultrathin heavy metal layer, such properties are good candidates for stabilizing ultra-small skyrmions (<10 nm diameters) at room temperature [152], which are also highly mobile [153] with reduced SkHE [45]. These properties are superior compared to the THE signals observed in the Ru-, Ir- and Mn-based oxide perovskites discussed in the earlier sub-sections.

Cape and Lehman and Lemesh et al. developed a full magnetic stray-field model that bridges the gap between the DW model [78, 79] (equation 5) and the effective anisotropy model [10] (equation 3). They noticed that in an $M_s$-versus-$K_u$ phase diagram (figures 17(a) and (b)), conventional metallic ferromagnets or their multilayers are located in the large $M_s$ but small-to-moderate $K_u$ regime. Hence, they do not satisfy the condition of small skyrmions stabilized at room temperature and zero/weak magnetic field [152], but produce the stripe phase instead. To possibly reach the small skyrmion (~10 nm) regime at room temperature, low $M_s$ and high $K_u$ conditions are required, and ferromagnets are better candidates for this purpose.

In addition, Caretta et al. demonstrated the benefit of using ferrimagnetic Gd$_4$Co$_5$ to achieve ultrafast DW or skyrmion speed [153] up to 1 km s$^{-1}$. Ferrimagnets typically
Newtonian particles without velocity saturation even at large net magnetic quasiparticles (such as DW or skyrmions) behave like Hall angle $\theta_H$, and DW width ($T$) the effective field of damping-like SOT should diverge at $T_c$. The coefficients of the two sub-lattices $S_1$ and $S_2$ of the two antiferromagnetically coupled sub-lattices can vanish at $T_c$, resulting in divergence well with experimental data. (d) Temperature-dependent $H_c$ and $M_c$ variations across $T_M$ and $T_A$ for Co$_3$Ge$_5$. The calculated SOT-driven DW velocity in Pt/Co$_3$Ge$_5$ varies with (d) temperature and (e) current density agrees well with experimental data. (f) Hall effect of Pt/TmIG/NGG(111) with SHE signals shaded varying with (d) $\gamma$ and (g) with the dotted lines marking the threshold below which skyrmions are expected to emerge. (h) Hall effect from various heavy metal/TmIG (111) bilayers. (a), (b) Reproduced from J. Phys. D: Appl. Phys. (2018). (c) Reprinted by permission from Springer Nature Customer Service Centre GmbH: Nature Nanotechnology (2019). (f), (g) Reprinted with permission from [155], Copyright (2020) by the American Physical Society.

Figure 17. (a) $M_3$-$K_u$ phase diagram from Buttner’s full strain-field model built with near-realistic parameters with color bar showing skyrmion diameters. (b) The same strain-field model overlapped with experimental data (circles and triangles), with color bar showing their stabilizing energy. (c) Temperature-dependent $H_c$ and $M_c$ variations. At $T_M$, $M_{1,2}$ of the two antiferromagnetically coupled sub-lattices cancel, i.e. $M(T) = M_1 + M_2 = 0$, resulting in divergence of $H_c$. Meanwhile at $T_A$, their spin angular momentum $S(T) = M_1/\gamma_1 + M_2/\gamma_2 = 0$, resulting in divergence of DW velocity. The difference between the gyromagnetic coefficients of the two sub-lattices $\gamma_{1,2} = g_1,2\mu_B/\hbar$ implies $T_A$ and $T_M$ are separated but near. From the SHE-driven DW velocity, the effective field of damping-like SOT should diverge at $T_M$ (near $T_A$). It also becomes very efficient when $S(T)$ vanishes at $T_A$, resulting in the huge increase in SHE-driven DW velocity, for ferrimagnets, where is related to the DMI, effective spin-hall angle $\gamma$, and DW width $\Delta$. Hence at $T_A$, the ferrimagnetic quasiparticles (such as DW or skyrmions) behave like Newtonian particles without velocity saturation even at large current densities (figures 17(d) and (e)). Conversely, the DW velocity of a conventional ferromagnet, $v_{FM} = \frac{\gamma}{\mu} \frac{\partial H}{\partial \alpha}$, is limited by the large $M_3$ and would saturate when $j_{HM} \gg \alpha \gamma$, where $\alpha$ is the Gilbert damping constant. Insulating ferromagnetic oxides also bring the advantage of having lower $\alpha (\sim 10^{-4})$ compared to metallic ferromagnets ($\sim 10^{-2}$), implying that lower $j_{HM}$ is needed for driving DWs or skyrmions.

Shao et al first observed the SHE signal in Pt/TmFe$_2$O$_5$ (TmIG)/Nd$_2$Ga$_3$O$_{12}$ (111) [154] (figures 17(f) and (g)). The THE signal appears above 300 K where $K_u < \frac{\pi^2}{6} \mu$, justifying the presence of DMI-stabilized skyrmions. The SHE signal is also distinct from those observed in SrRuO$_3$ systems reported earlier, since it is not hysteretic—Hall peak features appear at small fields when the field is swept towards both positive and negative directions. Such non-hysteretic behavior could be understood as the result of the low coercive field $\mu_0H_c < 10$ mT of typical ferrimagnetic garnets. Furthermore, such non-hysteretic THE signal is also convincing since it avoids the debated interpretation arising from partial cancelation of two oppositely-signed AHE loops. Yang’s group in Ohio University subsequently coined this effect as the ‘spin-Hall topological Hall effect (SH-THE)’, describing how the SHE in ultrathin Pt causes (vertical) spin accumulation at the interface, which is able to detect the presence of topological magnetic textures at the Pt/TmIG interface [156]. Comparing between the strain effect from Gd$_3$Ga$_5$O$_{12}$ (GGG) and substituted GGG (s-GGG) substrates on TmIG film, the larger compressive strain on TmIG from s-GGG (but not GGG) is realized to induce PMA and hence the SH-THE signal. It is known that the spin-Hall conductivity flips sign across the 5d heavy metals from Ta to Au [157]. Lee et al further replaced ultrathin Pt with Ta, W and Au on TmIG films [155] (figure 17(h)). While W showed similar behavior to Pt, Ta was unable to create skyrmions in TmIG due to its weak interfacial DMI contribution. For the Au case, a strong DMI exists to create skyrmions but its spin accumulation and detection of SH-THE are ineffective due to the long spin diffusion length in Au. Lastly, the 4 f electrons in the rare-earth cations of ferrimagnetic garnets are found to contribute no obvious role in stabilizing SH-THE [158].

7. Future outlook

In this final section, we briefly direct the readers to some interesting related work. Note that there is a slight contradiction from the design of the skyrmion racetrack memory in section 1. The criterion (1) for skyrmion nucleation by dissipation-less electric field requires an insulator with low carrier density to avoid screening of the field effect, yet criterion (2) for current-driving skyrmions along the racetrack requires a metallic body. Such contradiction would raise the question of whether an interface skyrmion residing at an insulator–metallic material interface would be useful, i.e. the insulating film provides the necessary magnetic properties $(M, K_u)$ that can be tuned by the electric field, while the metallic film provides the SOC and Rashba-type DMI via the sharp interface, and also functions as an electrode for the electric field gating process. Hence, a good example is an FM
interface arising from charge transfer between a heavy metal and AF insulator, such as the SrMnO$_3$/SrIrO$_3$ interface, which received much attention. Following Zhong’s calculation based on the concept of O2p band alignment before contact [136], Ir$^{3+}$ with a larger ($E_{\text{O2p}}$–$E_F$) gap will readily transfer electrons to Mn$^{4+}$. Nichols et al fabricated such a superlattice (SL) and found strong negative-sign AHE but no THE hump features [159]. The similarity of AHE between SMO/SIO-SL and SRO suggests that they share similar $t_{2g}$ band structures with large amounts of Weyl nodes. Using a tight-binding Hamiltonian $\tilde{H} = -t \cos \left( \frac{\pi}{a} \right) \sum_{\langle ij \rangle} \left( a^\dagger_{i \uparrow} a_{j \uparrow} + \text{h.c.} \right) + \sum_{\langle ij \rangle} J_{SE} (m_i \cdot m_j)$, Bhowal et al [137] explained that there exists a threshold charge-transfer fraction $x_c = \frac{2SE}{\Delta}$ above which the system is FM but below which the system is canted AF. Okamoto calculated that it is possible to stabilize SK in LSMO/SIO-SL [117], and Bhowal suggested that the Rashba-type SOC and AHE in SIO/SMO-SL can be tuned by electric field on modulating the hopping integral between non-overlapping orbitals [36]. Hence such charge-transfer-induced interface ferromagnetism can be a good platform for skyrmion searches, though a careful strategy is needed to avoid mutual cancelation of interfacial DMI. Another very promising structure is Pt/Tm$_2$Fe$_{12}$O$_{32}$ interfaces where non-hysteretic THE signals were found at above room temperature [154, 156]. The THE data are similar to those in figure 9(j), due to the low $H_F$. For the aspect of electrically tuning THE on such structure, the electric field could be applied on the insulating FM garnet via back-gating through the substrate with Pt as the top electrode, but this has not been demonstrated yet.

Several theorists have investigated confined heavy metal oxide perovskite systems in (111) orientation with honeycomb lattice that are potential candidates of TI [160–162]. However, ideal candidates such as LaAuO$_3$ may be difficult to synthesize. Si et al considered the concept that confined ultrathin SRO(111) can avoid degeneracy lifting among $t_{2g}$ orbitals, and thus can avoid a phase transition to canted AF but remain FM [163], but will open a small Mott gap near $E_F$. This way a Chern insulator hosting quantum AHE (QAHE) could be stabilized in SRO(111). It would also be interesting to explore SRO/SIO/STO(111) or SMO/SIO-SL/STO(111) in the search for THE, since the single terminations of (111)-oriented perovskite are always polar and would confine moment modulation and DMI vectors towards the in-plane (perpendicular to the polar direction).

In addition, a coexistence of QAHE and THE may also be formed at low temperature such as in the recent case of Cr$_{1-x}$(Bi,Sb)$_x$Te$_3$—although the AHE part must be quantized, the THE signal and possible magnetic skyrmions can still emerge around the $H_F$ field range of magnetization reversal [164]. This way, it might be possible to utilize the recent finding of topological ME effect (TME) for achieving electric field control of skyrmion nucleation or annihilation. In the seminal work by Marguerite et al [165], a nanoSQUID-on-tip [166] was employed to apply a localized electric field and image the circulating surface current around mirror magnetic monopoles [167] that are induced by TME. Such mirror magnetic monopole may potentially interact with the emergent magnetic field $B_{\text{eff}} = \frac{\partial \psi}{\partial \theta}$ of the topological charges of non-multiferroic skyrmions in general, therefore offering an alternative opportunity to achieve skyrmion size tuning, creation and annihilation by electric field.

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