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## TOPICAL REVIEW

# Antiferromagnetic Films and Their Applications

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**ABSTRACT** Spintronic devices are expected to replace the recent nanoelectronic memories and sensors due to their efficiency in energy consumption and functionality with scalability. To date, spintronic devices, namely magnetoresistive junctions, employ ferromagnetic materials by storing information bits as their magnetization directions. However, in order to achieve further miniaturization with maintaining and/or improving their efficiency and functionality, new materials development is required: 1) increase in spin polarization of a ferromagnet or 2) replacement of a ferromagnet by an antiferromagnet. Antiferromagnetic materials have been used to induce an exchange bias to the neighboring ferromagnet but they have recently been found to demonstrate a 100% spin-polarized electrical current, up to THz oscillation and topological effects. In this review, the recent development of three types of antiferromagnets is summarized with offering their future perspectives towards device applications.

**INDEX TERMS** Antiferromagnetic materials, Hall effect, magnetoresistance, spintronics, spin polarized transport.

## I. INTRODUCTION

Spintronics is one of the emerging fields in condensed matter physics in the view of replacing the recent nanoelectronic devices by improving their efficiency and functionality [1], [2]. In spintronic devices, further improvements are required to continue miniaturization to be comparable with the Si-based semiconductor technology. With a ferromagnet (FM), the miniaturization may induce edge domains and cross-talk between junctions via stray fields, which may prevent fast and reliable operation. On the other hand, using an antiferromagnet (AF), these obstacles can be avoided.

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Namely for the reduction in power consumption, highly efficient generation and detection of a spin-polarized electrical current need to be developed using the spin-orbit torque, spin caloritronic and topological effects. AF materials and their properties were initially investigated by Néel [3] and have been utilized to exchange couple with the neighboring FM magnetization [4]. This can be measured as a shift in the corresponding magnetization curve as known as an exchange bias (EB) field  $H_{ex}$ .  $H_{ex}$  has been used to pin one of the FM magnetizations in a FM/non-magnet (NM)/FM trilayer, *i.e.*, a spin-valve structure [5]. This is a basic structure for a read head of a hard disk drive (HDD). By replacing the NM layer with an insulating barrier, a magnetic tunnel junction (MTJ) has been fabricated in a similar manner, which has

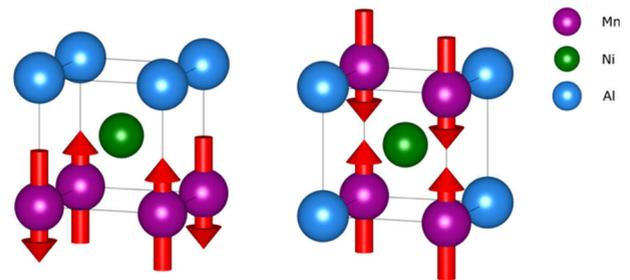
been commonly used as the latest HDD read head and a bit cell of a magnetic random access memory (MRAM). For these applications, an  $\text{IrMn}_3$  alloy has been predominantly employed due to its corrosion resistance and robustness against nanofabrication processes.

Recently, the studies on AF materials and devices have been revitalized after the demonstration of spin polarization by simply flowing an electrical current in an AF layer, which has led to AF spintronics [6], [7], [8]. AF materials have also been demonstrated to generate a spin current at THz frequency [9]. Additionally with the exchange coupling with a FM layer attached, interfacial interactions including Dzyaloshinskii-Moriya interaction (DMI) [10], [11] can be controlled, resulting in the formation of a magnetic skyrmion [12], a semimetal [13] and topological states [14]. These phenomena can offer further improvement in the performance and functionality of spintronic devices.

In parallel, a search for new AF materials which can maintain their AF properties with miniaturisation especially in a film form has been made intensively. There are large variety of AF materials investigated as summarized in Table 1: (i) cubic and (ii) hexagonal structures as shown in Figs. 1 and 2, respectively. The cubic AF includes Group III-VI as appeared in the Periodic Table (e.g.,  $\text{FeO}$ ,  $\text{CoO}$  and  $\text{NiO}$  [15]), II-VI (e.g.,  $\text{MnO}$  [16],  $\text{MnS}_2$ ,  $\text{MnSe}_2$  and  $\text{MnTe}_2$  [16]), Group III (e.g.,  $\text{Cr}$ ,  $\text{FeMn}$  [17] and  $\text{NiMn}$  [18]) and III-V (e.g.,  $\text{FeN}$  [19] and  $\text{MnN}$  [20]) compounds as well as ternary/quaternary Heusler alloys (with some atomic substitutions) [21]. The hexagonal AF contains Group III (e.g.,  $\text{IrMn}_3$  [22]), I-III-VI (e.g.,  $\text{CuFeO}_2$  [23],  $\text{CuFeS}_2$  [24],  $\text{CuFeSe}_2$  [25] and  $\text{CuFeTe}_2$  [26]), tetragonal AF (e.g.,  $\text{CuMnAs}$  [27], [28]), antiperovskite manganites [29], [30], [31], [32], [33] and binary Heusler alloys (with some atomic substitutions).

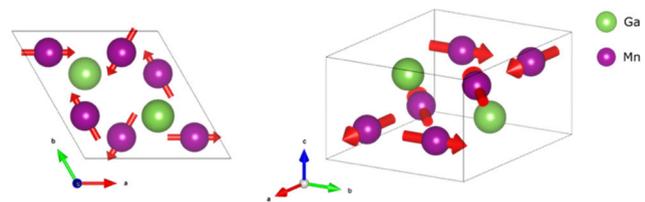
The cubic Heusler alloys crystallize in (i)  $L2_1$  phase with  $X_2YZ$  composition (full-Heusler) and (ii)  $C1_b$  phase with  $XYZ$  composition (half-Heusler) [34]. The half-Heusler alloys have an  $X$ -vacancy in the unit cell, making them susceptible to atomic displacement. Even for the full-Heusler alloys, the perfectly-ordered  $L2_1$  phase can be deformed into the  $B2$  phase by atomically displacing  $Y$ - $Z$  elements, the  $D0_3$  phase by  $X$ - $Y$  displacements and the  $A2$  phase by randomly exchanging  $X$ - $Y$ - $Z$  elements. We have recently found that  $\text{Ru}_2YZ$  [35],  $\text{Ni}_2YZ$  [36], [37] and  $\text{Mn}_2YZ$  [38], [39] Heusler alloys exhibit AF behavior in their  $L2_1$ ,  $B2$  and  $A2$  crystalline ordering phases. By attaching a FM Fe layer to these AF layers,  $H_{\text{ex}}$  of up to 600 Oe at 100 K, 90 Oe at 100 K and 30 Oe at 100 K for  $\text{Ru}_2\text{MnGe}$ ,  $\text{Ni}_2\text{MnAl}$  and  $\text{Mn}_2\text{Val}$ , respectively, have been found.  $\text{Mn}_2\text{Val}$  is found to maintain its AF properties at room temperature (RT). These differences are found to be induced by the AF alignment of spin moments at the  $Y$  site in unique ordered phases. In the ordered  $L2_1$  type  $\text{Ru}_2\text{MnZ}$  ( $Z = \text{Si}$ ,  $\text{Ge}$ ,  $\text{Sn}$  and  $\text{Sb}$ ), the complex AF ordering ( $2^{\text{nd}}$  type) is a consequence of the frustrated exchange interaction between the Mn atoms. It is concluded that Néel temperature  $T_N$  sharply depends on the  $Z$  element and that

$T_N$  in  $\text{Ru}_2\text{MnGe}$  can be increased by avoiding the disorder in the Mn- $Z$  sub-lattice. For  $\text{Ni}_2\text{MnAl}$ , the (checkerboard-like) AF order only exists in the chemically disordered  $B2$  phase due to the large AF nearest neighbor Mn-Mn interaction as schematically shown in Fig. 1. Decreasing the atomic disorder in the Mn-Al sublattice leads to non-zero total magnetization (i.e., ferrimagnet; FI). The excess of Mn or Ni does not improve the anisotropy of the AF state. From the device application point of view, Mn-based AF Heusler alloys are ideal due to their robustness against atomic disordering, especially at the interfaces to neighboring layers.



**FIGURE 1.** Schematic crystalline and spin structures of the pseudo- $B2$  (I and II) phases for  $\text{Ni}_2\text{MnAl}$  created by VESTA [40].

As Mn-based AF Heusler alloys, binary Heusler alloys with perpendicular anisotropy, such as the  $D0_{19}$   $\text{Mn}_3Z$  ( $Z = \text{Ga}$ ,  $\text{Ge}$  and  $\text{Sn}$ ) have been studied to determine their structural and magnetic properties. In perpendicularly anisotropic AF films, the effect of AF-coupled “domains” can be minimized in the in-plane electron scattering as shown schematically in Fig. 2. The  $\text{Mn}_3\text{Ge}$  binary alloy is taken as an example. It allows two stable crystalline structures of the tetragonal  $D0_{22}$  and hexagonal  $D0_{19}$  structures which is distorted from the basic full-Heusler  $L2_1$  structures along the  $\langle 001 \rangle$  and  $\langle 111 \rangle$  directions, respectively [41]. As a result of different crystalline structures, the  $D0_{22}$  or  $D0_{19}$  structure exhibits different magnetic anisotropy such as a FI with perpendicular magnetic anisotropy and low saturation magnetization [42] or AF with noncollinear magnetic moments in which the EB effect [43] appears. Recently,  $D0_{19}$   $\text{Mn}_{2.8}\text{Ga}_{1.2}$  films have been grown with exhibiting  $H_{\text{ex}}$  up to 430 Oe at 120 K [44], which is almost comparable with recent reports on  $\text{IrMn}$  with  $H_{\text{ex}} = 688$  Oe [45] and  $\text{MnN}$  ( $H_{\text{ex}} = 3.6$  kOe but with less corrosion resistance) [46].



**FIGURE 2.** Schematic crystalline and spin structures of the  $D0_{19}$  phase for  $\text{Mn}_3\text{Ga}$  from the top and side views created by VESTA [40].

**TABLE 1.** List of major AF materials and their properties. After Refs. [6] and [21]. The bulk and calculate (calc.) values are also included as references.

Group	AF material	Néel temperature [K] (bulk/film/calculation)	Blocking temperature [K] (FM used)	Exchange bias [Oe] (measured temperature)	Magnetic Anisotropy [erg/cm <sup>3</sup> ]	Ref.	
(III)	Cr	311 (bulk)				[21]	
	FeMn	333 (bulk)		375 (RT)		[17] [47]	
	NiMn	797 (bulk)	>698	650 (RT ~ 453 K)	3.2 × 10 <sup>5</sup>	[18] [48] [49] [50]	
	PtMn	702 (bulk)		~800 (RT)		[48] [51]	
	CuMnAs	50 (bulk)				[27] [28]	
	IrMn <sub>3</sub>	960 (bulk)	463~503			[22] [52]	
	MnF <sub>2</sub>	68 (bulk)		1.8k (RT)	~3 × 10 <sup>7</sup>	[53] [54]	
	FeF <sub>2</sub>	78 (bulk)				[55]	
	(II-VI)	MnO	122 (bulk)		~300 (5 K)	2.7 × 10 <sup>4</sup>	[15] [56]
		MnS	152 (bulk)		290 (5 K)		[16] [57]
MnSe		173 (bulk)				[16]	
MnTe		323 (bulk)	70 (CrTe)	~500 (5 K)	~2.0 × 10 <sup>6</sup>	[16] [58]	
EuSe <sub>2</sub> EuTe		8 ± 0.5 (bulk) 7.8 ± 0.5 (bulk)		~10 – 142 (2 K)		[52] [60]	
(III-V)	FeN	100 (bulk)				[19]	
	FeP	119 (bulk)				[61]	
	FeAs	67 (bulk)				[62]	
	FeSb	105 (bulk)				[63]	
	MnN	660 (bulk)			~6 × 10 <sup>6</sup>	[20] [53] [64]	
	GdP	15				[65]	
	GdAs	25				[66]	
	GdSb	25				[66]	
(III-VI)	FeO	198 (bulk)		~80 (RT)		[15] [67]	
	CoO	293 (bulk)	~220 (Co)	1600 (77 K) 400 (170 K)	5.0 × 10 <sup>6</sup>	[4] [15] [68]	
	NiO	525 (bulk)		133 (RT?)		[15] [69]	
	Cr <sub>2</sub> O <sub>3</sub>	310 (bulk)		1080 (250 K)	~1.8 × 10 <sup>7</sup>	[70] [71]	
	BiFeO <sub>3</sub>	653 (bulk)			~1.0 × 10 <sup>6</sup>	[72] [73] [74]	
	(Heusler)	Cr <sub>2</sub> MnP	240 (calc.)				[75]
Cr <sub>2</sub> MnAs		250 (calc.)				[75]	
Cr <sub>2</sub> MnSb		342 (calc.)				[75]	
Cr <sub>2</sub> MnBi		320 (calc.)				[75]	
Fe <sub>2</sub> VAl						[76] [77]	
Fe <sub>2</sub> VSi		193 (film)			16 (100 K)	[77] [78]	
Ni <sub>2</sub> MnAl		313 (bulk) 245 (calc. B2-I) 350 (calc. B2-II)			>55 (10 K)	[79] [80] [36]	

**TABLE 1. (Continued.) List of major AF materials and their properties. After Refs. [6] and [21]. The bulk and calculate (calc.) values are also included as references.**

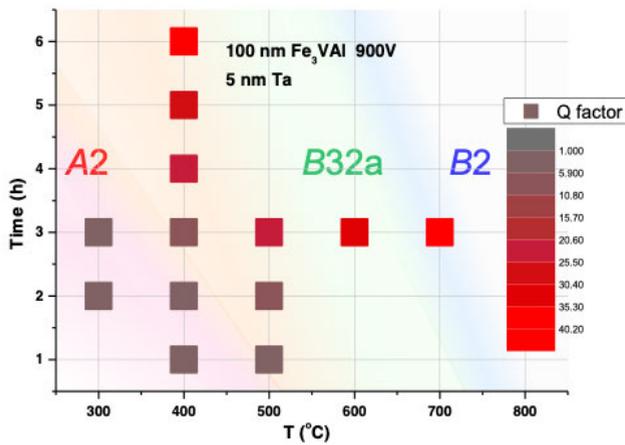
			≤100 (Fe)		[37]
	Mn <sub>2.4</sub> Pt <sub>0.6</sub> Ga	120 (bulk)		33k (2 K)	[81]
	Mn <sub>2</sub> VAl	>600	~75 (Fe)		[39]
	Mn <sub>2</sub> VSi		<100 (CoFe)	34 (100 K)	[38]
	Ru <sub>2</sub> MnGe	300 (bulk)			[82]
	Ru <sub>2</sub> MnSi	313 (calc.)	126 (Fe)	680 (10 K)	[35]
	Ru <sub>2</sub> MnSn	296			[21]
	Ru <sub>2</sub> MnSb	195			[21]
	Pt <sub>2</sub> MnGa	350 (bulk)			[83]
(Heusler)	Mn <sub>3</sub> Ge		390		[84]
	Mn <sub>3</sub> Ga	470 (bulk)			[85]
		648 (film)	~400	1.5k (RT)	[42]
			235	430 (230 K)	[44]
	Mn <sub>3</sub> Sn	430 (bulk)			[86]
				2.8 × 10 <sup>3</sup>	[87]
	Mn <sub>2</sub> FeGa		235 (CoFe, in-plane)	446 (120 K)	[88]
			240 ([Co/Pt] <sub>3</sub> , perp.)	163 (120 K)	
	Mn <sub>2.4</sub> Pt <sub>0.6</sub> Ga		~90	33k (2 K)	[81]
(Antiperovskite)	Mn <sub>2.5</sub> Co <sub>0.3</sub> Ga <sub>1.2</sub>		>350	250 (RT)	[21]
	Mn <sub>3</sub> GaN	380 (bulk)			[89]
		345 (film)			[90]
	Mn <sub>3</sub> NiN	266 (bulk)			[91]
		250 (film)			[92]
	Mn <sub>3</sub> ZnN	183 (bulk)			[91]
	Mn <sub>3</sub> AgN	290 (bulk)			[91]
	Mn <sub>3</sub> SnN	475 (bulk)			[91]
(Manganite)	La <sub>0.45</sub> Sr <sub>0.55</sub> MnO <sub>3</sub>	220 (film)	~50	98	[29]
					[30]
	SrMnO <sub>3</sub>	260 (bulk)	~150	1.0 – 2.0 × 10 <sup>5</sup>	[31]
					[32]
	CaMnO <sub>3</sub>	122 (bulk)			[33]
(I-II-V)	LiMnSb	>RT(bulk)			[93]
	LiMnAs	374(bulk)			[94]
(I-III-VI)	CuFeO <sub>2</sub>	11 (bulk)			[23]
	CuFeS <sub>2</sub>	825 (bulk)			[24]
	CuFeSe <sub>2</sub>	70 (bulk)			[25]
	CuFeTe <sub>2</sub>	254 (bulk)			[26]
(II-IV-V)	MnSiN <sub>2</sub>	490(bulk)			[95]

The major challenge in the development of AF Heusler alloys is that there are over 3000 known Heusler alloy compositions [96]. The key parameter in an AF order is the spacing between planes where the magnetic spins are ordered ferromagnetically in the (001) plane as shown in Figs. 1 and 2 (with slight canting from the plane). Hence it is critical to engineer the composition, so that the right spacing is achieved with typically  $B2$  or  $D0_{19}$  ordering. This can be confirmed using our recently developed Q-factor analysis as shown in Fig. 3 [37]. The Q-factor is defined as the peak intensity

measured by X-ray diffraction (XRD) divided by full width at half maximum, which offers a very simple measure to evaluate the crystallinity of Heusler alloys and beyond.

## II. ELECTROMAGNETIC CHARACTERISATIONS

For the characterization of AF materials, the following techniques have been traditionally used: Magnetization measurements with and without a FM layer attached, transport measurements with and without a magnetic field, and synchrotron-based measurements. The former two



**FIGURE 3.** Calculated Q-factors and the corresponding crystallization of  $\text{Fe}_{72.1}\text{V}_{14.4}\text{Al}_{13.5}/\text{Ru}$  samples annealed at elevating temperatures.

techniques are relevant for macroscopic analysis, while the last one is sensitive to microscopic characterization as detailed below and Fig. 4.

#### A. MAGNETIC SUSCEPTIBILITY

In a simplified picture, AF can be treated as two sets of FM-coupled magnetic moments antiparallely aligned with each other,  $M_A = -M_B$  (see Fig. 1). The temperature dependence of this antiparallel alignment can be calculated similar to that of a ferromagnet with the parallel alignment. The antiparallel alignment is stable up to the characteristic temperature,  $T_N$ , above which the alignment becomes random due to thermal fluctuation, typically leading to paramagnetic phase [97]. This magnetic phase transition becomes apparent by plotting the temperature dependence of magnetic susceptibility  $\chi$ . For a single crystal, by applying a magnetic field along the moments,  $\chi$  increases linearly with increasing temperature  $T$ . By applying a field perpendicular to the moments,  $\chi$  stays constant. For polycrystalline AF,  $\chi(T)$  follows between these two cases. Above  $T_N$ ,  $\chi$  decreases almost inversely proportional with  $T$ , forming a kink in  $\chi(T)$  at  $T_N$  (see Fig. 4). This method can be useful for a bulk material to generate sufficient change in the magnetic moments, especially in a single phase.

#### B. EXCHANGE BIAS

As AF does not produce intrinsic magnetization macroscopically, no signal can be detected by a magnetization measurement. Instead, by attaching a FM layer to induce an EB at the interface, a shift of the corresponding FM magnetization curve ( $H_{ex}$ ) can be measured, the amplitude of which is proportional to the interfacial exchange coupling between AF and FM. This is particularly useful for AF films.  $H_{ex}$  can be evaluated by the York Protocol [98]. In an AF/FM bilayer, AF is first set at the setting temperature  $T_{SET}$  for 90 min., which is above  $T_N$  of AF but below the Curie temperature  $T_C$  of the FM film. The bilayer is then cooled to the thermally activated temperature  $T_{NA}$ , followed by the

heating to the activation temperature  $T_{ACT}$  for 30 min. and the magnetization measurement at  $T_{NA}$ . In the activation period any activated FM grains reverse their magnetic moments to their originally set direction. This procedure removes the first loop training effect and any thermal activation that may occur during the temperature rise and fall. In polycrystalline bilayers, individual grains have their own blocking temperature  $T_B$ , which can be determined by increasing  $T_{ACT}$  until the loop shift becomes zero, which represents the median value of  $T_B$  ( $<T_B>$ ).  $<T_B>$  satisfies the reversed AF volume to be the same with that of the initially set volume [98], which can be an indicative measure of  $T_N$ .

#### C. ELECTRICAL RESISTIVITY AND MAGNETORESISTANCE

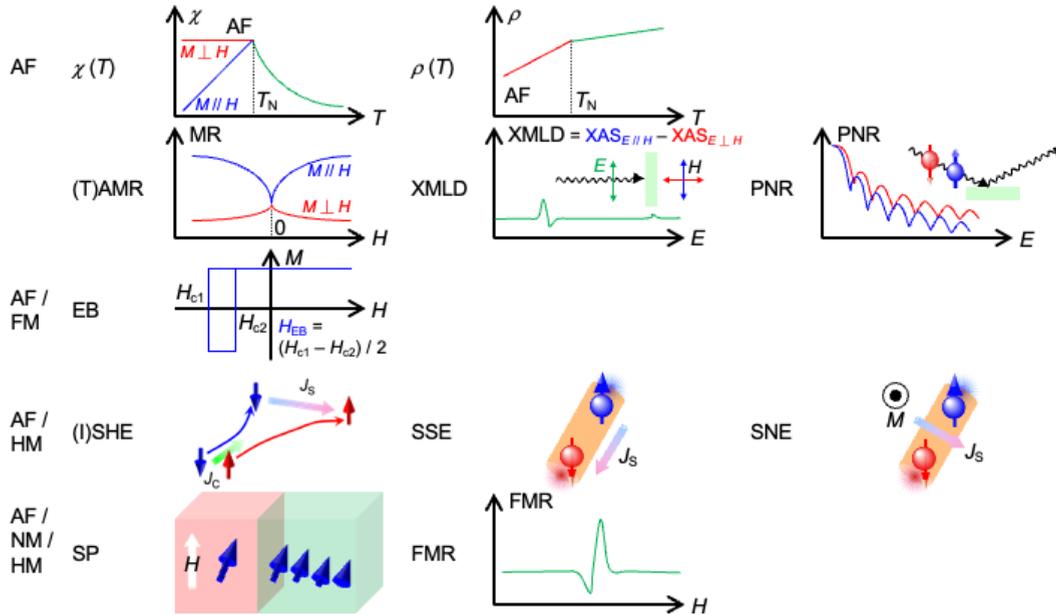
Similarly, the temperature dependence of electrical resistivity  $\rho(T)$  exhibits a kink in the gradient [21]. Below  $T_N$ , antiparallely-coupled magnetic moments in single-crystal AF can suppress electron scattering. Above  $T_N$ , however, the moment alignment becomes random and changes the corresponding resistivity. It should be noted that the changes in the resistivity are found to be 11% maximum, which can be smaller than that due to electron scattering at grain boundaries by over three orders. This is a powerful technique to determine  $T_N$  for epitaxial or highly-textured films as well. By applying an external magnetic field during the transport measurements, similar changes in anisotropic magnetoresistance (AMR) [99] and tunneling AMR (TAMR) [100] can be used to determine  $T_N$  due to the magnetic phase transformation. TAMR was experimentally demonstrated by Gould et al. with a junction consisting of  $\text{Ga}_{0.94}\text{Mn}_{0.06}\text{As}/\text{Al-O}/\text{Ti}$  [101]. A TAMR ratio was found to increase using an AF layer up to 0.15% with a  $\text{IrMn}/\text{MgO}/\text{Pt}$  multilayer at room temperature [102]. Recently, AF materials has also been used for TAMR, demonstrating 10% TAMR in  $\text{IrMn}/\text{MgO}/\text{Ta}$  junctions [103] and 20% TAMR using CsCl-ordered  $\text{FeRh}$  magnetic phase transition in  $\text{FeRh}/\text{MgO}/\text{FeRh}$  junctions [100].

#### D. X-RAY MAGNETIC LINEAR DICHROISM

For microscopic evaluation, synchrotron radiation can be used. X-ray magnetic linear dichroism (XMLD) [104] utilizes a pair of linearly polarized soft X-ray with perpendicular polarization. XMLD signals are proportional to the average value of the magnetic moment squared in a domain  $\langle M^2 \rangle$ . For an AF material,  $\langle M \rangle$  is zero as  $M_A = -M_B$  within an AF domain but  $\langle M^2 \rangle$  is a finite value, allowing AF domain imaging. Such domain imaging requires a relatively large uniform domain ( $> \text{a few } \mu\text{m}$ ) due to the spatial resolution, which makes it difficult to be used for AF films. Recently, XMLD has been combined with photoemission electron microscopy (PEEM) imaging to achieve sub- $\mu\text{m}$  resolution [105].

#### E. POLARISED NEUTRON REFLECTIVITY

Another synchrotron-based technique for the characterization of AF materials is polarized neutron reflectivity (PNR). PNR can determine magnetic properties of bulk and layered



**FIGURE 4.** List of characterization techniques for AF materials and devices. For AF only, magnetic susceptibility, electrical resistivity and (tunneling) anisotropic magnetoresistance, X-ray magnetic linear dichroism, and polarized neutron reflectivity can be used for characterization as detailed in Sections II-A, C, D and E. By attaching FM and a heavy metal (HM), exchange bias, and (inverse) spin Hall and spin caloritronic effect can indirectly show the AF properties as discussed in Sections II-B and III, respectively. Using a trilayer consisting of AF/NM/HM, spin pumping and ferromagnetic resonance can be used for characterization as described in Section IV.

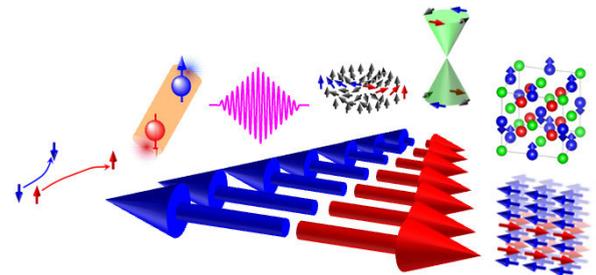
materials [106]. Due to the magnetic moment of neutron beam interacting with magnetic materials to be imaged, not only layer structures, such as thickness, density, composition and interfacial roughness as similar to X-ray reflectivity (XRR), but also in-plane magnetic moments can be determined. PNR has higher accuracy in a shorter scanning period (<1 min.). The latter magnetic information can be obtained by detecting the neutron reflection with its spins interacted with those in an AF and/or FM layers.

### III. SPIN-ORBIT TORQUE

In AF spintronics, three key phenomena can be used for device applications: spin-orbit torque (SOT), spin dynamics and interfacial effects as schematically listed in Fig. 5. These phenomena are discussed in the following sections. In this section, (inverse) spin Hall effects and spin caloritronic effects are reviewed, both of which are induced by SOT.

#### A. ANOMALOUS HALL AND SPIN HALL EFFECTS

The Hall effect is induced by the Lorentz force under an external magnetic field (ordinary Hall effect). In a magnetic material, anomalous Hall effect (AHE) can be induced due to the spin-orbit interaction, where an effective magnetic field exists by the presence of an intrinsic magnetization. Large AHE was reported in  $Mn_3Sn$  [107]. This is due to a weak ferromagnetism induced in the non-collinear AF alignment ( $\sim 0.002 \mu_B/Mn$  [87]). Note that additional topological contribution to the Hall signals may need to be considered in a noncollinear magnet [108]. This was employed to



**FIGURE 5.** Concept of antiferromagnetic spintronics, showing spin Hall effects, spin caloritronics, THz oscillation, magnetic skyrmions, topological effects, Heusler alloys and exchange bias (from left to right).

develop an AF memory with the writing capability at THz frequency [109].

Spin accumulation in a semiconductor was theoretically predicted by Averieiev and Dyakonov under the flow of an electron charge current, introducing the resultant spin current perpendicular to the charge current [110]. This is induced by spin-dependent scattering by an impurity and intrinsic spin-orbit interactions of the material. Averieiev and Dyakonov also proposed the inverse effect by aligning the spins by an electromagnetic wave to generate a charge current [111]. These predictions were revisited by Hirsch and named as spin Hall and inverse spin Hall effects (SHE and ISHE), respectively (see Fig. 6) [112]. The relationship between the charge and spin currents can be defined as

$$(\text{Spin current}) = \theta_{SH} \times (\text{Charge current}), \quad (1)$$

where the coefficient  $\theta_{SH}$  is the spin Hall angle specific to a material used. The corresponding Hamiltonian can be determined as

$$H = \frac{\hbar^2 k^2}{2m} + \lambda_{SO} (\boldsymbol{\sigma} \times \mathbf{k}), \quad (2)$$

where  $\hbar$ : Planck constant divided by  $2\pi$ ,  $k$ : wave vector,  $m$ : electron mass,  $\lambda_{SO}$ : spin-orbit coupling constant and  $\boldsymbol{\sigma}$ : spin matrix.

Experimentally, magneto-optical Kerr effect (MOKE) imaging was used to detect the spin accumulation at the edges of GaAs at 30 K, resulting in a spin current of the order of  $10 \text{ nA}/\mu\text{m}^2$  [113]. In a similar system, a spin current was also electrically detected [114].

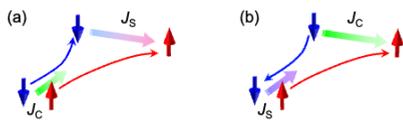


FIGURE 6. Schematic diagram of the (a) SHE and (b) ISHE measurements.

SHE and ISHE do not require an external magnetic field unlike the original Hall effect. When a large magnetic field is applied perpendicular to the material, the accumulated spins start to precess and diminish SHE and ISHE. This induces the corresponding resistance changes with respect to the field, spin Hall magnetoresistance (SHMR) [115]. SHMR was experimentally measured in  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (YIG)/Pt bilayer [116].

In AF materials, large (I)SHE signal has been shown, e.g.,  $(5.3 \pm 2.4)\%$  for  $\text{Mn}_3\text{Sn}$  [117], as listed in Table 2. These signals are induced by the spin Hall angles  $\theta_{SH}$  as listed in Table 2, which are almost one order of magnitude smaller than those for heavy-metals, e.g.,  $-35$  ( $-50$ )% for W [118] (WOx [119]) and  $5.6\%$  for Pt [120].

**B. SPIN CALORITRONIC EFFECTS**

Spin caloritronic effects, namely spin Seebeck and Nernst effects (SSE and SNE, respectively), can induce a spin current as originally demonstrated in  $\text{Ni}_{80}\text{Fe}_{20}/\text{Pt}$  [133] and YIG/Pt (see Fig. 7) [134]. In AF materials, a pioneering work was performed with a  $\text{Cr}_2\text{O}_3/\text{Pt}$  bilayer by Seki et al. [135] as listed in Table 3. These effects have been intensively investigated for energy harvesting. For SSE, the figure of merit  $ZT$  can be determined as [136]

$$ZT \text{ (SSE)} = \frac{\sigma S^2}{\kappa} T, \quad (3)$$

where  $T$ : temperature,  $\sigma$ : electrical conductivity,  $S$ : Seebeck coefficient and  $\kappa$ : thermal conductivity.  $ZT > 1$  is needed for practical device applications. Similarly for SNE,  $ZT$  can be obtained using the Nernst coefficient  $N$  as follows:

$$ZT \text{ (SNE)} = \frac{\sigma N^2}{\kappa} T. \quad (4)$$

An anomalous Nernst effect (ANE) is normally proportional to the intrinsic magnetization of the material under

TABLE 2. List of spin Hall angle and SMR reported for AF and Weyl materials. After Ref. [7]. SP, SSE and FMR represent spin pumping, spin Seebeck effect and ferromagnetic resonance measurements, respectively.

Group	AF material	Spin diffusion length at RT [nm]	Spin Hall angle $\theta_{SH}$ [%]	Ref.
Cubic (III-VI)	NiO	~3.8		[121]
	Cr		$-5.1 \pm 0.5$ (SP, YIG/Cr)	[122]
(III)			$-9$ (SSE, YIG/Cr)	[123]
			$\sim 0.7$ ( $\text{Co}_2\text{FeAl}/\text{Cr}$ )	[124]
	FeMn		$0.8 \pm 0.2$ (SP, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{FeMn}$ )	[125]
			$2.2\sim 2.8$ (FMR, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{FeMn}$ )	[126]
	PtMn		$6 \pm 1$ (SP, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{PtMn}$ )	[125]
			$6.4\sim 8.1$ (FMR, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{PtMn}$ )	[126]
		2.3	$24$ (SP, $\text{Fe}_{50}\text{Co}_{30}\text{B}_{20}/\text{Hf}/\text{PtMn}$ )	[127]
	IrMn		$2.2 \pm 0.5$ (SP, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{IrMn}$ )	[125]
			$5.3\sim 9.7$ (FMR, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{IrMn}$ )	[126]
	PdMn		$1.5 \pm 0.5$ (SP, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{PdMn}$ )	[125]
			$2.8\sim 4.9$ (FMR, $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}/\text{PdMn}$ )	[126]
(Heusler)	YPtBi		$4.1$ (planar Hall, YPtBi/ $c\text{-Al}_2\text{O}_3$ )	[128]
	$\text{Co}_2\text{MnGa}$ (FM)		$-19\pm 4$ (planar Hall, $\text{Co}_2\text{MnGa}/\text{MgO}(001)$ )	[129]
(III)	$\text{IrMn}_3$		$\sim 9, \sim 11(111), \sim 20(100)$ (FMR, $\text{Ni}_{80}\text{Fe}_{20}/\text{IrMn}_3$ )	[130]
	$\text{Mn}_2\text{Au}$		$0.22$ (SP, $\text{Mn}_3\text{Ga}/\text{Co}_{0.2}\text{Fe}_{0.6}\text{B}_{0.2}$ )	[131]
Hex.	$\text{Mn}_3\text{Sn}$	$0.75 \pm 0.67$	$5.3 \pm 2.4$	[117]
	$\text{Mn}_3\text{Ga}$		$0.31 \pm 0.01$ (SP, $\text{W}/\text{Mn}_3\text{Ga}/\text{Co}_{0.2}\text{Fe}_{0.6}\text{B}_{0.2}$ )	[132]

investigation as similar to AHE. Even so, a Nernst signal of  $\sim 0.35 \mu\text{V}/\text{K}$  was reported at RT [135], which is over two orders of magnitude greater than that expected from the weak ferromagnetism [87]. This increase is due to the fact that the transverse thermoelectric conductivity is determined by the Berry curvature in the vicinity of the Fermi level ( $E_F$ ) offering adiabatic electron motion, while the anomalous Hall

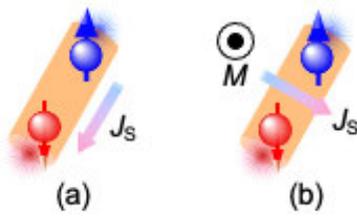


FIGURE 7. Schematic diagram of the (a) SSE and (b) SNE measurements.

TABLE 3. List of spin caloritronic properties reported for AF materials.

Group	AF material	Spin Seebeck coefficient [ $\mu\text{V/K}$ ]	Spin Nernst coefficient [ $\mu\text{V/K}$ ]	Figure of merit $ZT$	Ref.
Cubic	NiO	$6.5 \pm 0.5$ (20 K)			[138]
(III-VI)	$\text{Cr}_2\text{O}_3$	0.015 (40 K)			[135]
(Heusler)	NiMnSb (FM)	$< -2$ (100 K)			[139]
	NiTiSn	-155 (300 K)			[140]
	ZrNiSn	$\sim 300$ (300 K)			[141]
	$\text{Co}_2\text{TiAl}$	$-55$ (300 K)			[140]
(III)	$\text{Co}_2\text{TiSn}$	50 (300 K)			[142]
	$\text{IrMn}_3$	$390 \pm 10$ (RT)		2.2 (RT)	[143]
Hex.	$\text{MnF}_2$	4.5 (35 K)			[144]
	$\text{Mn}_3\text{Sn}$		$\sim 0.35$ (RT)		[136]

conductivity is defined as the sum of the Berry curvature for all the occupied bands. Hence, a Weyl metal can be advantageous for spin caloritronic applications due to the unique Berry curvature at Weyl points near  $E_F$  [see Fig. 10(b)] The detailed model to calculate the corresponding spin current can be found in Ref. [137].

#### IV. DYNAMICS

##### A. SPIN PUMPING

In a FM/AF bilayer, a spin current can be introduced by spin pumping (SP) from FM to AF by precessing the FM magnetization (see Fig. 8). The spin current may be damped in AF and may reduce the reflected spin current into FM, which accordingly increases the damping constant of FM. Using ISHE, the spin current  $J_S^{\text{SP}}$  satisfies the following relationship [145].

$$V_{\text{ISHE}} = wR\theta_{\text{SH}} \left( \frac{2e}{\hbar} \right) J_S^{\text{SP}}, \quad (5)$$

where  $w$  and  $R$  are the width and resistance of the bilayered Hall bar and  $e$  is the electron charge.  $V_{\text{ISHE}}$  takes the maximum at the resonant magnetic field, where the maximum precession is achieved and the resulting maximum  $J_S^{\text{SP}}$  is introduced to AF. Using this condition,  $\theta_{\text{SH}}$  can be estimated as listed in Table 2. This is sensitive to a small spin current

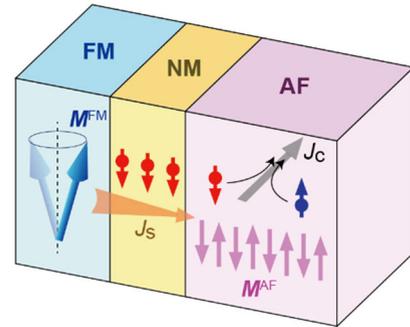


FIGURE 8. Schematic diagram of the spin pumping (SP) measurement in a trilayered structure with ferromagnet (FM)/nonmagnet (NM)/antiferromagnet (AF). Spin current ( $J_S$ ) generated by magnetization precession is converted to charge current ( $J_C$ ) via inverse spin Hall effect in the AF.

to be introduced by increasing  $R$  of the Hall bar sample. This technique can also be applied to characterize a spin current generated optically and thermally.

##### B. FERROMAGNETIC RESONANCE

Similar to SP, a FM/AF bilayer is used for ferromagnetic resonance (FMR) as schematically shown in Fig. 9(a). A high-frequency current  $I$  (typically at 10 GHz) is applied to a FM/AF bilayer to generate an in-plane radio-frequency (rf) magnetic field perpendicular to the current in the AF layer accompanying with the spin current from the AF layer. This experimental technique is called ‘‘spin-torque FMR (ST-FMR)’’, which was originally employed to evaluate  $\theta_{\text{SH}}$  in the bilayer consisting of FM and NM [146], and has widely been used for a variety of materials [147], [148]. The in-plane rf magnetic field exerts a torque and induces the magnetization precession in the FM layer. At the same time, a spin current can be generated by SHE in the AF and/or at the FM/AF interface, which diffusively flows into the FM layer and exerts a torque perpendicular to the layer. Here, the in-plane torque is in-phase with the high-frequency current, while the perpendicular spin-current torque is shifted by  $\pi/2$  from the current frequency. By fitting a FMR spectrum to antisymmetric and symmetric contributions due to the in-plane and perpendicular torques, respectively,  $J_S^{\text{FMR}}$  can be estimated from the latter contribution.  $V_{\text{FMR}}^{\text{sym}}$  and  $V_{\text{FMR}}^{\text{antisym}}$  are given as follows [146] and [149]:

$$V_{\text{FMR}}^{\text{sym}} = \frac{1}{4} \frac{dR}{d\theta} \frac{\gamma I \cos \theta}{2\pi (df/dH)_{H=H_0}} \times \left[ \frac{\hbar J_S^{\text{FMR}} \Delta}{2e\mu_0 M_S t_{\text{FM}} (H - H_0)^2} \right] \quad (6)$$

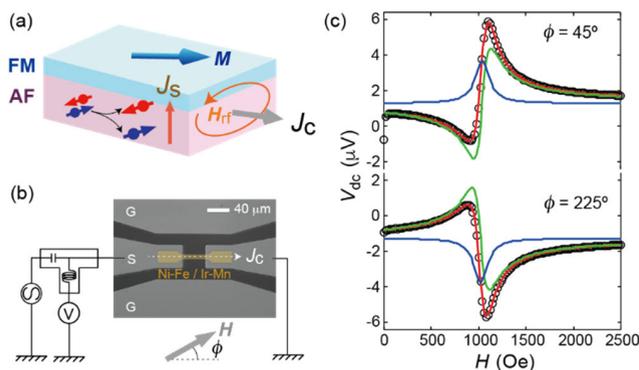
$$V_{\text{FMR}}^{\text{antisym}} = \frac{1}{4} \frac{dR}{d\theta} \frac{\gamma I \cos \theta}{2\pi (df/dH)_{H=H_0}} \times \left[ \frac{J_C t_{\text{AF}}}{2} \left( 1 + \frac{e\mu_0 M_S}{\hbar} \right)^{1/2} \frac{(H - H_0)}{\Delta^2 + (H - H_0)^2} \right] \quad (7)$$

where  $dR/d\theta$ : resistance change by the precession,  $\theta$ : angle between the external magnetic field ( $H$ ) and  $I$ ,  $\gamma$ : gyromagnetic constant,  $f$ : frequency of  $I$ ,  $H_0$ : FMR field,  $\mu_0$ : magnetic permeability in a vacuum ( $4\pi \times 10^{-7}$  H/m),  $M_S$ : saturation magnetisation of FM,  $t_{\text{FM}}$ : FM thickness,  $\Delta$  is half width of half maximum of FMR spectrum,  $t_{\text{AF}}$ : AF thickness and  $J_C$ : charge current.

Using the symmetric and antisymmetric FMR spectrum components,  $\theta_{\text{SH}}$  can be calculated as follows [146]:

$$\theta_{\text{SH}} = \frac{J_S^{\text{FMR}}}{J_C} = t_{\text{FM}} t_{\text{AF}} \frac{V_{\text{FMR}}^{\text{sym}}}{2V_{\text{FMR}}^{\text{antisym}}} \frac{e\mu_0 M_S}{\hbar} \sqrt{1 + \left(\frac{4\pi M_{\text{eff}}}{H}\right)^2} \quad (8)$$

The literature values of  $\theta_{\text{SH}}$  measured by ST-FMR are listed in Table 2. An example of ST-FMR for the FM/AF bilayer is shown in Fig. 9, in which the  $\text{Ni}_{81}\text{Fe}_{19}$  and  $\text{Ir}_{22}\text{Mn}_{78}$  ( $\text{IrMn}_{3.55}$ ) were chosen as FM and AF materials, respectively. Fig. 9(b) displays the optical microscope image of the coplanar-waveguide-shaped device with the sputter deposited  $\text{Ni}_{81}\text{Fe}_{19}$  (3 nm)/ $\text{IrMn}_{3.55}$  (10 nm) bilayer together with the measurement setup for ST-FMR. The representative ST-FMR spectra are shown in Fig. 9(c), in which the rf current with the frequency of 8 GHz was applied and the in-plane  $H$  angles were set at  $45^\circ$  and  $225^\circ$ . The spectra were well fitted with the summation of antisymmetric and symmetric Lorentzian functions. Using Eq. (8),  $\theta_{\text{SH}}$  was evaluated to be 3% for the  $\text{IrMn}_{3.55}$  alloy.



**FIGURE 9.** (a) Schematic diagram of the spin torque ferromagnetic resonance (ST-FMR) measurement in a bilayer with ferromagnet (FM)/antiferromagnet (AF). (b) optical microscope image of the coplanar-waveguide-shaped device together with the measurement setup. (c) ST-FMR spectra for the  $\text{Ni}_{81}\text{Fe}_{19}$  (3 nm)/ $\text{IrMn}_{3.55}$  (10 nm) bilayer. The rf current with the frequency of 8 GHz was applied, and the in-plane magnetic field angles were set at  $45^\circ$  and  $225^\circ$ .

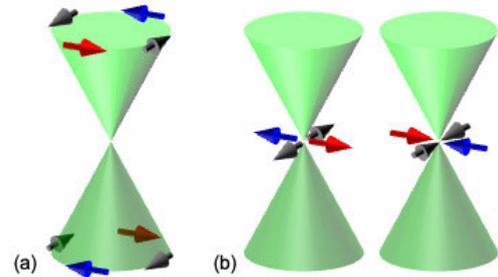
### C. THz OSCILLATION

By overlaying a direct current (dc) on  $I$  in the FMR technique as described in Section IV-B, an effective damping constant of FM  $M_{\text{eff}}$  can be modified as

$$\Delta\alpha_{\text{eff}} = \frac{\sin\theta}{(H + M_{\text{eff}}/2)} \frac{\hbar J_S^{\text{FMR}}}{2e\mu_0 M_S t_{\text{FM}}}. \quad (9)$$

**TABLE 4.** List of spin dynamics reported for AF materials.

Group	AF material	Gilbert damping constant	Resonant frequency [THz]	Ref.
Cubic (III-VI)	NiO	$(2.1 \pm 0.1) \times 10^{-4}$	1	[152]
	$\text{Cr}_2\text{O}_3$		0.240	[152]
	$\text{FeBO}_3$		$\sim 0.45$	[9]
	$\text{TmFeO}_3$		$\sim 0.8$	[9]
	$\text{YFeO}_3$		$\sim 0.55$	[9]
(III)	$\text{IrMn}_3$		0.22	[153]



**FIGURE 10.** Schematic band diagram of the (a) Dirac and (b) Weyl semimetals.

By controlling  $I$ , the in-plane and perpendicular torques can be cancelled out, resulting in  $\Delta\alpha_{\text{eff}} = 0$ . This allows oscillation of the FM magnetisation.

THz oscillation can be observed in AF due to strong exchange interactions between two sublattices with  $M_A$  and  $M_B$  [150], [151]. In NiO, the first demonstration of THz oscillation was achieved [151]. Since then, a great deal of research has been made to achieve higher oscillation frequency in AF materials as listed in Table 4.

### V. TOPOLOGICAL EFFECTS AND BEYOND

Topological and interfacial phenomena in AF materials have been intensively investigated recently [154]. For example, Dirac and Weyl semimetals can be formed with AF nature as schematically shown in Fig. 10. The Dirac semimetals form a point connection between the valence and conduction bands at  $E_F$ , which is known as the Dirac point. The Dirac point can demonstrate the ideal conductance of  $2G_0$  ( $G_0 = e^2/h$ , where  $e$  is the electron charge and  $h$  is the Planck constant). The Weyl semimetals contain the chirality at the Dirac point as schematically shown in Fig. 10(b). Weyl semimetals show large AHE, e.g., anomalous Hall angle and conductivity of 0.23 and  $60 \Omega^{-1}\text{cm}^{-1}$  for  $\text{GdPtBi}$  [155], 0.11 and 1258.9 for  $\text{Co}_2\text{MnGa}$ , and 0.08 and 1421.6 for  $\text{Co}_2\text{MnAl}$ , respectively [156]. The chiral topological semimetal  $\text{CoSi}$  shows a small spin Hall angle of  $\sim 0.03$  due to the unique electronic structure [157].

### A. MAGNETIC SKYRMIONS

Néel-type magnetic skyrmions were stabilized by attaching  $\text{IrMn}_3$  underneath  $\text{Co}_{20}\text{Fe}_{60}\text{B}_{20}$  at RT [12], which has been supported by theoretical calculations [158]. According to

theoretical prediction, skyrmions in AF can be displaced faster by a smaller critical current density ( $10^6 \sim 10^7$  A/cm<sup>2</sup>) than those in conventional FM materials [159], [160]. Although AF skyrmions were stabilized and imaged in a synthetic AF [161] and FI [162], no report has been made to date in AF materials.

**B. TOPOLOGICAL EFFECTS**

SHE can be induced in an individual layer in two-dimensional MnBi<sub>2</sub>Te<sub>4</sub>, achieving layer Hall effect with up and down spins to be generated at the edges of the top and bottom layers at 1.7 K [163]. This is induced by the layer-locked Berry curvature, which can open a new research field of topological AF spintronics.

**C. ORBITAL FERRIMAGNETISM**

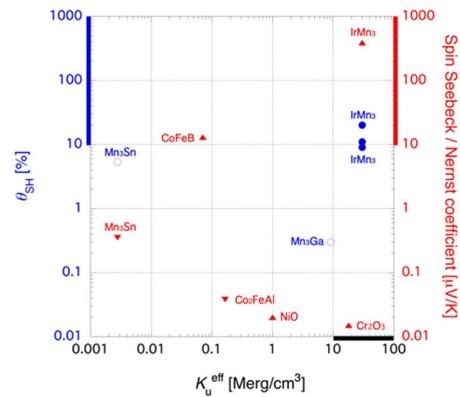
“Orbital Ferrimagnetism” was firstly termed for FI CoMnO<sub>3</sub> by Bozorth et al. [164]. Orbital FI is defined as a system where the net magnetic moment is only attributed to the orbital magnetic momentum. To date, CoMnO<sub>3</sub> is the only material known to exhibit orbital FI, consisting of Co<sup>2+</sup> and Mn<sup>4+</sup>, which possess  $S = 3/2$ , respectively. Since these cations are antiferromagnetically coupled, the spin angular momenta cancel each other out. This causes the spin momenta to be compensated, while the orbital angular momentum of Co<sup>2+</sup> in the crystal field to be conserved. Consequently, the net magnetic moment is proportional to the orbital angular momentum [165], [166]. In other words, an orbital FI has the properties of antiferroic-spin momenta and ferroic-orbital momentum. Therefore, one can expect that the orbital FI would become a bridge between AF spintronics and orbitronics.

**D. ALTERMAGNETISM**

Recently, a new class of collinear antiferromagnet is theoretically predicted by classifying magnetic material based on spin-group formalism, which is termed as altermagnetism [167], [168]. Altermagnetism is induced by the anisotropic band structure that non-degenerate but equally populated spin-up and spin-down energy isosurfaces. Altermagnetism is predicted to exhibit various spintronics phenomena [169], [170], [171] such as AHE and TMR similar to AF, and is expected to be a functional material in a novel region of antiferromagnetic spintronics. For example, field-free switching was demonstrated in the heterostructure using RuO<sub>2</sub> [172], [173]. Some other altermagnets, e.g., MnTe [174], [175] and Mn<sub>5</sub>Si<sub>3</sub> [176], have also been predicted and characterized.

**VI. CONCLUSION AND FUTURE PERSPECTIVES**

We have reviewed the recent development and characterization of AF materials and devices. In general, hexagonal (or non-collinear) AF exhibits larger magnetic anisotropy constants of the order of 10 Merg/cm<sup>3</sup>, while cubic AF shows smaller constants as summarized in Fig. 11. They correspond to the spin Hall angle  $\theta_{SH}$  and spin Seebeck/Nernst



**FIGURE 11. Correlations between the magnetic anisotropy constant and the spin Hall angle  $\theta_{SH}$  (closed symbols) as well as the spin Seebeck/Nernst coefficient (open symbols). After [190], new data added on Mn<sub>3</sub>Ga [132] and NiO [137]. Open and closed symbols represent cubic and non-collinear spin configurations, respectively. Blue and red data show  $\theta_{SH}$  and spin Seebeck and Nernst coefficients, respectively. Target ranges are highlighted as broad lines in the corresponding axes.**

**TABLE 5. List of abbreviations used in this review.**

Abbreviation	Full form
AF	antiferromagnet
AHE	anomalous Hall effect
AMR	Anisotropic magnetoresistance
ANE	anomalous Nernst effect
DMI	Dzyaloshinskii-Moriya interaction
EB	exchange bias
FI	ferrimagnet
FM	ferromagnet
FMR	ferromagnetic resonance
HDD	hard disk drive
HM	heavy metal
ISHE	inverse spin Hall effect
MOKE	magneto-optical Kerr effect
MRAM	magnetic random access memory
MTJ	magnetic tunnel junction
NM	non-magnet
PEEM	photoemission electron microscopy
PNR	polarized neutron reflectivity
RT	room temperature
SHE	spin Hall effect
SHMR	spin Hall magnetoresistance
SNE	spin Nernst effect
SOT	spin-orbit torque
SP	spin pumping
SSE	spin Seebeck effect
ST	spin-torque
Sy	synthetic
TAMR	tunneling anisotropic magnetoresistance
XMLD	X-ray magnetic linear dichroism
XRD	X-ray diffraction
XRR	X-ray reflectivity

coefficients. In order to develop efficient AF spintronic devices, higher anisotropy without atomic disordering is needed as highlighted by broad lines. Such materials can be used for an electric-field controlled device [177], SOT-MRAM [178] and energy harvesting [136]. Hall memory concept has also demonstrated with CuMnAs [109] and Mn<sub>2</sub>Au [179].

By antiferromagnetically coupling two FM layers through a non-magnetic spacer, a synthetic AF (SyAF) can be formed, which has been commonly used to pin the magnetization of the neighboring FM layer in perpendicularly-magnetized MRAM [180]. Such SyAF has also been used to demonstrate memory operation [181], [182], [183] similar to AF as discussed in Sec. III-A. SyAF can offer broad controllability in the quantization axis in the system, offering design flexibility in AF spintronic devices.

Recently, AF-based magnetic tunnel junctions (MTJs) have been fabricated by showing about a 100% TMR ratio with  $\text{Mn}_3\text{Pt}/\text{MgO}/\text{Mn}_3\text{Pt}$  [184] and 2% ratio with  $\text{Mn}_3\text{Sn}/\text{MgO}/\text{Mn}_3\text{Sn}$  [185]. By enhancing the TMR ratio, such an AF-based MTJ may offer a new architecture for spintronic devices. Such a large TMR ratio is predicted using a noncolinear antiferromagnet,  $\text{Mn}_3\text{Sn}$  [186], and even larger ratio of 500% is predicted with a  $\text{RuO}_2/\text{TiO}_2/\text{RuO}_2$  junction [170].

FI can also be used in a similar manner to induce AMR [187], SHE and laser-induced magnetization reversal [188]. The magnetization of FI can be controlled by engineering the composition in the vicinity of the compensation point. This can minimize the corresponding stray field used as an alternative FM in MRAM and magnetic sensors. These AF and FI (as well as a new class of symmetry, altermagnetism [189])-based devices are anticipated to improve the efficiency of their spintronic devices.

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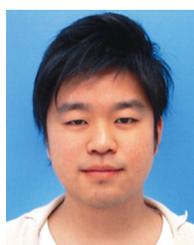


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