Roadmap for Emerging Materials for Spintronic Device Applications

Atsufumi Hirohata1, Hiroaki Sukegawa2, Hideto Yanagihara3, Igor Žutić4, Takeshi Seki5, Shigemi Mizukami6, and Raja Swaminathan7

1Department of Electronics, University of York, York YO10 5DD, U.K.
2Magnetic Materials Unit, National Institute for Materials Science, Tsukuba 305-0047, Japan
3Graduate School of Pure and Applied Sciences, University of Tsukuba, Tsukuba 305-8577, Japan
4Department of Physics, University at Buffalo–The State University of New York, Buffalo, NY 14260 USA
5Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
6WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
7Intel Corporation, Chandler, AZ 85226 USA

The Technical Committee of the IEEE Magnetics Society has selected seven research topics to develop their roadmaps, where major developments should be listed alongside expected timelines: 1) hard disk drives; 2) magnetic random access memories; 3) domain-wall devices; 4) permanent magnets; 5) sensors and actuators; 6) magnetic materials; and 7) organic devices. Among them, magnetic materials for spintronic devices have been surveyed as the first exercise. In this roadmap exercise, we have targeted magnetic tunnel and spin-valve junctions as spintronic devices. These can be used, for example, as a cell for a magnetic random access memory and a spin-torque oscillator in their vertical form as well as a spin transistor and a spin Hall device in their lateral form. In these devices, the critical role of magnetic materials is to inject spin-polarized electrons efficiently into a nonmagnet. We have accordingly identified two key properties to be achieved by developing new magnetic materials for future spintronic devices: 1) half-metallicity at room temperature (RT) and 2) perpendicular anisotropy in nanoscale devices at RT. For the first property, five major magnetic materials are selected for their evaluation for future magnetic/spintronic device applications: 1) Heusler alloys; 2) ferrites; 3) rutiles; 4) perovskites; and 5) dilute magnetic semiconductors. These alloys have been reported or predicted to be half-metallic ferromagnets at RT. They possess a bandgap at the Fermi level $E_F$ only for its minority spins, achieving 100% spin polarization at $E_F$. We have also evaluated L1$_2$ alloys and $D_{0_3}$-Mn alloys for the development of a perpendicularly anisotropic ferromagnet with large spin polarization. We have listed several key milestones for each material on their functionality improvements, property achievements, device implementations, and interdisciplinary applications within 35 years time scale. The individual analyses and the projections are discussed in the following sections.

Index Terms—Half-metallic ferromagnets, magnetic anisotropy, magnetic materials, spintronics.

I. HEUSLER ALLOYS

Heusler alloys are ternary alloys originally discovered by Heusler [1]. He demonstrated the ferromagnetic behavior in an alloy consisting of nonmagnetic (NM) atoms, Cu$_2$MnSn. Since then, these alloys have been investigated due to their properties of shape memory and thermal conductance. In 1983, de Groot et al. [2] reported the half-metallic ferromagnetism in one of the Heusler alloys, half-Heusler NiMnSb alloy. A great deal of effort has accordingly been devoted to achieve the half-metallicity at room temperature (RT) using a Heusler alloy. In particular, Block et al. [3] measured a large tunneling magnetoresistance (TMR) in bulk full-Heusler Co$_2$(Cr, Fe)Si alloy, followed by a similar measurement in a thin-film form [4].

Among these Heusler alloys, Co-based full-Heusler alloys are the most promising candidates to achieve the RT half-metallicity due to their high Curie temperature. (T$_C$ $\gg$ RT), good lattice matching with major substrates, large minority-spin bandgap ($\geq$0.4 eV, see Fig. 1), and large magnetic moments in general ($\geq$4 $\mu_B$ per formula unit (f.u.)) [5], [6]. The main obstacle to achieve the half-metallicity in the Heusler-alloy films is the vulnerability against the crystalline disorder, such as the atomic displacement, misfit dislocation, and symmetry break in the

Fig. 1. Minority-spin bandgap [7] and $L2_1$ phase [6] of the full-Heusler alloys.
vicinity of the surface of the films. For the full-Heusler alloys forming X2YZ, where the X and Y atoms are transition metals, while Z is either a semiconductor or an NM metal, the unit cell of the ideal crystalline structure (L21 phase, see Fig. 2) consists of four face-centered cubic sublattices. When the Y and Z atoms exchange their sites (Y–Z disorder) and eventually occupy their sites at random, the alloy transforms into the B2 phase. In addition, the X–Y and the X–Z disorder finally leads to the formation of the A2 phase. By increasing the disorder, the magnetic properties depart further from the half-metallicity.

Toward the RT half-metallicity, two milestones have been identified as listed in the following:

1) (m1.1): demonstration of >100% giant magnetoresistance (GMR) ratio at RT;

2) (m1.2): demonstration of >1000% TMR ratio at RT.

Here, we have regarded these criteria using the MR as an indicator of the half-metallicity at RT.

Regarding (m1.1), in 2011, 74.8% GMR ratio was reported by Sato et al. [8] using a junction consisting of Co2Fe0.4Mn0.6Si/Ag/Co2Fe0.4Mn0.6Si. This is a significant improvement from 41.7% reported in [26]. Using such a GMR junction as a read head, the GMR ratio of ~0.17 Ω·μm2 satisfies the requirement for 2 Tb/in2 areal density in a hard disk drive (HDD). Fig. 2 shows the requirement and recent major efforts toward the Tb/in2 areal density. It is clear that the Heusler-alloy GMR junctions are the only candidates satisfying the requirement to date. By reflecting on the development over the last five years, one can expect that the Heusler-alloy GMR junctions can achieve 100% GMR ratios within three years. This will satisfy (m1.1) and will lead to device applications as HDD read heads.

For (m1.2), Fig. 3 shows the development of the TMR ratios using amorphous and MgO barriers with both the conventional ferromagnets and the Heusler alloys as electrodes. As shown here, the largest TMR reported to date is 604% at RT using a magnetic tunnel junction (MTJ) of CoFeB/MgO/CoFeB [10]. In 2005, an MTJ with an epitaxial L21 Co2MnSi film has been reported to show very high TMR ratios of 70% at RT [11]. These are the largest TMR ratios obtained in an MTJ with a Heusler alloy film and an Al–O barrier. The TMR is purely induced by the intrinsic spin polarization of the Heusler electrodes, which is different from an MTJ with an oriented MgO barrier, where a TMR ratio of 386% has been achieved at RT (832% at 9 K) for Co2FeAl0.5Si0.5 [12]. The TMR ratio reported here is the highest ever in an MTJ with a Heusler alloy film but with the assistance of coherent tunneling through an oriented MgO barrier. By taking a moderate extrapolation, one can estimate that 1000% TMR ratios (m1.2) can be achieved within ten-year time period, i.e., the RT half-metallicity by 2024.

The other device application expected is to fabricate all Heusler junctions consisting of antiferromagnetic/ferromagnetic/NM/ferromagnetic Heusler-alloy layers. Such junctions can offer a template to avoid any crystalline disorder at the interfaces as the lattice matching and symmetry can precisely be controlled by atom substitution in these alloy layers. As a first step, Nayak et al. [13] reported an antiferromagnetic Heusler alloy of Mn3PtGa for the first time but at low temperature (<160 K). One can anticipate that RT antiferromagnetism can be demonstrated within 5 years, leading to all Heusler-alloy junctions in 20 years.

By summarizing the above consideration, one can anticipate a roadmap on the Heusler-alloy films, as shown in Fig. 4. The Heusler-alloy films are expected to be used in GMR read heads and sensors within 3 years. These films are also to be combined with antiferromagnetic and/or NM Heusler-alloy films to form all Heusler junctions. Such junctions may be used in a magnetic random access memory subject to their perpendicular magnetic anisotropy, which is still in the infant stage in research.

II. OXIDES

Ferromagnetic oxide thin films have intensively been studied for more than last two decades due to their large
variety and tunability of physical properties, such as the ferro,
ferri, antiferromagnetism, ferroelectricity, superconductivity,
optical properties, and colossal MR (CMR) effect [14], [15].
In particular, some of ferromagnetic oxides are predicted as
promising candidates of a half-metal and a spin filter,
which directly lead to a large MR, as discussed in Section I.
In addition, due to a high compatibility with other oxides and
organic materials, the establishment of high-quality all-oxide
heterostructure beyond CMOS device is highly expected.
In this section, milestones and their associated roadmaps for
three half-metal oxide ferromagnets, (A) spinel ferrites,
(B) rutiles, and (C) perovskites are discussed.

A. Spinel Ferrites

The most commonly studied oxides of Fe is Fe$_3$O$_4$, which
has an inverse spinel structure and a magnetic moment of 4.1 $\mu_B$/f.u. [16]. Among various spinel-type ferrites, Fe$_3$O$_4$
珊 a major conductive oxide at RT. The Curie temperature $T_C$
珊 ~850 K, and the characteristic magnetic transition point
(Verwey temperature) is 123 K. According to a band
calculation, half-metallicity has been predicted [18], [19], and
spin-resolved photoemission experiments show that Fe$_3$O$_4$
exhibits the spin polarization of up to $\sim 80\%$ [20]. A very high
spin polarization has also been suggested by the measurement
of an MR ratio of over 500% through a nanocontact [21].

Epitaxial Fe$_3$O$_4$ films have been grown by various tech-
niques, including molecular beam epitaxy under an oxygen
atmosphere, magnetron sputtering, and laser ablation [20].
By replacing one of the Fe ions with a divalent metal ion,
e.g., Mn, Co, Ni, and so on, a ferrite can be formed [20].
Siratori and Iida [22] have predicted half-metallicity in Mn,
Co, and Ni ferrites, although the bulk materials are insulators
except Fe$_3$O$_4$. In particular, NiFe$_2$O$_4$ shows a bandgap in
the majority band, indicating that this compound can become an
insulator or semimetallic half-metal. The discrepancy of the
bandgap structure between the ab initio calculation results and
the experimental results suggests that the treatment of electron
correlation is significant.

Some ferrites are expected as a good candidate of a spin
filter because of their ferromagnetic insulator properties and
high $T_C$. The spin-filtering device consists of a ferromagnetic
insulator layer sandwiched between an NM metallic (NMM)
layer and a ferromagnetic metallic (FMM) layer (or a super-
conductive layer). Due to the exchange splitting of the energy
levels in the conduction band of the ferromagnetic insulator,
the effective barrier height for the up-spin electron differs from
that for the down-spin one, leading to a large difference in
the tunneling probabilities between the two spin orientations.
Therefore, ideally, an almost perfectly spin-polarized current
is generated and this results in an infinite MR if a ferromagnetic
insulator with a large exchange splitting is used. Here, the MR
ratio is defined as $2P_{SF}P/(1 - P_{SF}P)$, where $P_{SF}$ is the spin-
fILTERING efficiency $\text{[}I_{up} - I_{down}]/(I_{up} + I_{down})$, $I_{ap(\text{down})}$ $\propto 
\text{exp}(-d/d_{up(down)})$, where $I$ is the tunneling current, $d$
is the thickness of the spin filter, and $\phi$ is the effective barrier
height and $P$ is the spin polarization of the FMM layer.
The RT spin-filtering effect has been demonstrated using
CoFe$_2$O$_4$-based spin-filter devices [23], [24]. However, $|P_{SF}|$
at RT is <5%.

Related to Section IV, the perpendicular magnetization
behavior with a high uniaxial magnetic anisotropy of $K_u = 1.47 \times 10^6$ J/m$^3$ in CoFe$_2$O$_4$ ferrite [25] has been reported.
In addition to the ferromagnetic spinel ferrites, NM
spinel, MgAl$_2$O$_4$ has also attracted much attention as a new
spintronics material, because an ultrathin MgAl$_2$O$_4$ layer
shows coherent tunneling properties (symmetry selective
tunneling) and high MR ratios, such as an MgO tunnel
barrier. Using an epitaxial CoFe/MgAl$_2$O$_4$ (with cation-site
disordered)/CoFe structure, an MR ratio of >300% at RT
was reported [26].

Toward the magnetic ferrites as a spintronic material, the
following milestones have been recognized:
1) (m2.1.1): half-metallic behavior and high MR by
improving the microstructure and the control of interface
states;
2) (m2.1.2): high spin-filtering effects at RT by reducing
structural and chemical defects;
3) (m2.1.3): tuning of perpendicular magnetic anisotropy;
4) (m2.1.4): development of new NM spinel-based mate-
rials to tune the transport properties and the coherent
magneto-tunneling effect.

Regarding (m2.1.1) and (m2.1.2), ferrite films with a very
high-quality crystalline structure, i.e., without any crystal
imperfections, such as antiphase boundaries (APBs), atomic-
site disorder, and dislocations, are necessary to obtain high
saturation magnetization, high squareness of the hysteresis
loops, and high $T_C$. The presence of APBs within a ferrite
film, for instance, significantly degrades the saturation magne-
tization under a high magnetic field and the remanence. It also
increases the resistivity of the film, since the APBs induce the
electron-scattering center. Consequently, high-quality films
are indispensable to the achievement of stable half-metallic
characteristics and a spin-filtering effect at RT. In addition,
the realization of a perfect and an abrupt ferrite/NM interface
is required to preserve high effective spin polarization at the
interface states. Therefore, the establishment of the growth
method and procedures for the high-quality ferrite films, as
well as a high-quality interface with the FMM layer and
the NMM layer, are strongly desired. The development of an
advanced growth process will lead to RT half-metallicity
using ferrite family materials, such as Fe$_3$O$_4$, $\gamma$-Fe$_2$O$_3$,
CoFe$_2$O$_4$, NiFe$_2$O$_4$, MnFe$_2$O$_4$, and ZnFe$_2$O$_4$.

The milestone of (m2.1.3) is important to ensure the high
thermal stability for nanoscale structures using CoFe$_2$O$_4$-based
ferrites for future spin-filtering devices and other spintronics
uses at RT. In particular, strong perpendicular magnetic
anisotropy in a very thin region (below several nanometers)
is desirable to control the tunneling resistance for device
applications.

For (m2.1.4), providing the new NM tunnel barrier is now
considered as an important issue to establish novel spintronic
heterostructures, since only a limited tunnel barrier material
(Al$_2$O$_3$ and MgO) is currently available to obtain high RT MR
ratios. In particular, the ability to tune the physical properties
is required to achieve higher performance, multifunctionality,
and better compatibility to ferromagnetic electrodes. For instance, MR enhancement by crystalline barrier (coherent tunneling), a perfect lattice matching (lattice constant tuning), a low tunneling resistance (barrier height tuning), and applicability of high electric fields to a ferromagnetic layer facing the barrier (dielectric constant tuning) are presumably possible in spinel-based NM barrier with tailored compositions.

In summary, one can propose a roadmap on spinel ferrite films, as shown in Fig. 5. Using spinel ferrite-based MTJs consisting of ferrite/NM barrier/ferrite (or FMM) structure, >100% RT TMR (corresponding \(|P|\) is ∼0.7 according to the Julliere model) is expected within 10 years through the development of high-quality spinel ferrite thin films and the selection of a proper NM barrier. Further improvement of an MTJ structure and suppression of a rapid TMR reduction with increasing temperature will lead to a giant TMR over 1000% (corresponding \(|P|\) is ∼0.9) within 25 years.

To construct spin-filtering devices, one can use the techniques for the MTJ fabrication; a typical stacking structure is NMM/ferrite spin-filter/NM barrier/FMM, where the NM barrier is used to weaken the exchange coupling between the ferrite and the FMM layers. Recently, a high \(P\) of ∼8% at RT (MR ∼6%) has been demonstrated using an epitaxial Pt/CoFe\(_2\)O\(_4\)/Al\(_2\)O\(_3\)/Co nanocontact junction [27]. Thus, the improvement of the junction structure as well as the ferrite film quality can enhance the MR ratio. More than 100% RT MR ratio due to the spin-filtering effect is expected within 10 years by reducing structural and chemical defects in spin-filter junctions.

Using new NM barriers, one can highly expect a giant TMR ratio exceeding 500% at RT within 5 years. Furthermore, the tuning of physical properties will be achieved by searching for new candidate barrier materials within 10 years.

B. Rutiles

Using Andreev reflection, CrO\(_2\) has been proven to show a half-metallic nature at low temperature, as suggested by the \textit{ab initio} calculations [16], [17]. The high spin polarization of 90% has been confirmed at low temperature using the point-contact Andreev reflection method [18], [19], and high powder MR has been reported [20]. However, RT half-metallicity has not been demonstrated yet. CrO\(_2\) has a tetragonal unit cell with a magnetic moment of 2.03 \(\mu\text{B}/\text{f.u.}\) at 0 K [21]. The ferromagnetism of CrO\(_2\) appears <391 K [22]. Above this temperature another phase of Cr\(_2\)O\(_3\) is known to show antiferromagnetism, which is the major cause of the reduction of the half-metallicity. Highly ordered CrO\(_2\) films are predominantly grown by chemical vapor deposition [23]. However, obtaining the CrO\(_2\) single phase as a thin film is not easy, and thus MR properties steeply decrease below RT.

In order to utilize the rutiles in a spintronic device, the following milestones have been identified:

1) (m2.2.1): development of a high-quality CrO\(_2\) thin film with a single rutile phase and achievement of a clean interface structure with tunnel junctions;
2) (m2.2.2): search for new rutile-based materials with higher \(T_C\) and robust half-metallicity by tailoring their composition.

Regarding (m2.2.1), the undesirable reduction in MR ratio below \(T_C\) could be suppressed by the improvement of the crystal structure and the interface state. The optimization of an epitaxial growth process for a single rutile phase and the use of a suitable NM barrier, which does not invade the interface of CrO\(_2\), will be effective. In addition, the elimination of the NM Cr\(_2\)O\(_3\) phase, which generally forms on the surface of the CrO\(_2\) film, using sophisticated deposition and treatment processes will enhance the magnetic and half-metallic properties.

For (m2.2.2), to obtain a more stable half-metallic phase with high \(T_C\), doping of other elements to CrO\(_2\) or searching ternary or quaternary rutile-based ferromagnetic materials would be necessary. Such a new composition and a new material will lead to stable half-metallic properties and higher MR at RT.

In summary, one can anticipate a roadmap on the half-metallic rutile films, as shown in Fig. 6. Obtaining epitaxial thin films with a single CrO\(_2\) phase will lead to the observation...
of RT TMR ratios within 10 years. To demonstrate high TMR ratios (>100%) at RT is still challenging. Searching new rutile-type ferromagnetic oxides and a sophisticated MTJ structure might yield a technological breakthrough toward a higher TMR ratio in the future.

C. Perovskites

Perovskites, such as (La, Sr)MnO₃, exhibit both strong ferromagnetism and metallic conductivity with a partial substitution of La³⁺ ions with ²⁺ ions, such as Ca, Ba, Sr, Pb, and Cd [28], [29]. Since only one spin band exists at E_F in these films, 100% spin polarization can be achieved. Using these materials instead of a conventional ferromagnet, a very high MR of ~150% at RT has been observed [30]. This is known as CMR. Using Mn–perovskite thin films and SrTiO₃ oxide tunnel barrier, a TMR ratio of up to 1850% has been reported but only below TC [31]. CMR can be induced either by breaking the insulating symmetry of Mn³⁺ and Mn⁴⁺ alternating chains or by suppressing spin fluctuation near TC. Even so, it is unlikely to achieve the RT half-metallicity in the conceivable future.

Much effort has been spent to search for new high TC perovskites for an RT half-metallicity. The family of double perovskites with a chemical composition of A₂BB′O₆ (A is an alkaline earth or rare-earth ion, B and B’ are transition metal ions) has been focused for more than 15 years, since some of the double perovskites exhibit high TC above RT and half-metallic band structures [32]. Sr₂FeMoO₆ (SFMO) has high TC of 420 K and has been predicted to be a half-metal [33], indicating the double perovskites are a promising oxide family for high MR at RT. At low temperature, high P ~ −80% in an SFMO film has been demonstrated using a Co/SrTiO₃/SFMO MTJ. Much higher TC of 635 K is reported in Sr₂CrReO₆ [34].

Recently, 2-D electron gas (2-DEG) at the interface of an NM perovskite heterostructure consisting of LaAlO₃/SrTiO₃ has intensively been investigated due to a high mobility in the 2-DEG. Highly efficient spin transport in the 2-DEG could be usable to establish the new type spin transistors in the future.

The following milestones have been established toward the perovskites as a spintronic material:

1) (m2.3.1): search for new perovskite-based materials with TC > RT;
2) (m2.3.2): development of a high MR at RT.

Regarding (m2.3.1), the double perovskites with A₂FeMoO₆ or A₂FeReO₆ series are promising due to their high TC. However, a high MR using an MTJ structure has not been achieved, since there are some considerable obstacles against (m2.3.2): 1) site disorder of magnetic ions deteriorates the magnetic properties and the spin polarization and 2) their high reactivity to water, which restricts the use of common microfabrication techniques.

In order to overcome these obstacles, the improvement of film quality and the preparation of a clean interface are necessary to achieve the high MR ratios at RT. In particular, the specific microfabrication method should newly be developed to reduce the damage during the processes. In addition, a new barrier material that matches with the perovskites will be needed to compose a high-quality perovskite-based MTJ.

In summary, one can expect a roadmap on the perovskite films, as shown in Fig. 7. RT TMR ratios will be obtained using the MTJs with a high TC perovskite layer within 5 years. Less than 100% TMR at RT will be expected in the future after the demonstration of high TMR ratios at low temperatures.

III. Dilute Magnetic Semiconductors

Unlike metals, semiconductors have a relatively low carrier density that can drastically be changed by doping, electrical gates, or photoexcitations, to control their transport and optical properties. This versatility makes them the materials of choice for information processing and charge-based electronics.

In magnetically doped semiconductors, such as (Cd, Mn)Te, (In, Mn)As, or (Ga, Mn)As, these changes of carrier density also enable novel opportunities to control the magnetic properties and lead to applications that are not available or ineffective with ferromagnetic metals [35]. For example, a carrier-mediated magnetism in semiconductors offers a tunable control of the exchange interaction between the carriers and the magnetic impurities. The onset of ferromagnetism and the corresponding change in the TC can be achieved by increasing the carrier density using an applied electric field, photoexcitations, or even heating. Two milestones for the research on novel magnetic semiconductors are identified:

1) (m3.1): search for tunable ferromagnetism in semiconductors with TC > RT.
2) (m3.2): demonstrating RT devices that are not limited to magnetoresistive effects.

Considering (m3.1), despite numerous reports for TC > 300 K in many semiconductors, a reliable RT ferromagnetic semiconductor remains elusive [36], [37]. However, even the existing low-TC magnetic semiconductors have provided the demonstrations of novel magnetic effects and ideas that have also subsequently been transferred to ferromagnetic metals, for example, electric-field modulation of coercivity and magnetocrystalline anisotropy at RT [37].
An early work on ferromagnetic semiconductors dates back to CrB$_3$ in 1960 [38]. Typically studied were concentrated magnetic semiconductors, having a large fraction of magnetic elements that form a periodic array in the crystal structure. Important examples are Eu-based materials, in which the solid-state spin-filtering effect was demonstrated for the first time [39]. However, the complicated growth and the modest $T_C$ (up to $\sim150$ K) limited these materials to fundamental research. Starting with the mid-1970s, the dilute magnetic semiconductors (DMS), alloys of NM semiconductor and magnetic elements (typically, Mn) [40], became intensely explored first in II–IV, and later in III–V NM hosts. In II–IVs, Mn$^{2+}$ is isovalent with group II providing only spin doping, but not carriers and thus making robust ferromagnetism elusive. In III–Vs, Mn yields both spin and carrier doping, but low-Mn solubility limit complicates their growth and can lead to an extrinsic magnetic response due to nanoscale clustering of metallic inclusions. This complex dual role of Mn doping in III–Vs possess both: 1) challenges to establish the universal behavior among different NM III–V hosts. (Ga, Mn)N predicted to have $T_C > 300$ K [41], but shown to only have $T_C \sim 10$ K [42] and 2) makes the ab initio studies less reliable, requiring careful considerations of secondary phases and magnetic nanoclustering—a source of many reports for an apparent high-$T_C$ in the DMS.

An important breakthrough came with the growth of III–V DMS: (In, Mn)As in 1989 and (Ga, Mn)As in 1996 [43], [44], responsible for demonstrating tunable $T_C$, coercivity, magnetocrystalline anisotropy, as well as the discovery of tunneling anisotropic MR [37]. However, even if the low-Mn solubility is overcome (maximum $\sim10\%$), the upper $T_C$ limit is given MnAs with $T_C \sim 330$ K. This suggests that (Ga, Mn)As, with the current record $T_C \sim 190$ K [41], is not a viable candidate for RT ferromagnetism in DMS. Influential mean-field calculations [39] for DMS with 5% Mn in Fig. 8(a) show a strong correlation with an inverse unit cell volume [45]. However, the ab initio studies reveal a more complex, material-dependent situation [46].

Instead of III–V compounds, more promising is recently discovered II–IV–V DMS [47]. They are isosctructural to both II–IV class of high-temperature Fe-based superconductors and antiferromagnetic $\text{BaMn}_2\text{As}_2$, offering intriguing possibilities to study their multilayers with different types of ordering. In (Ba,K)(Zn,Mn)$_2$As$_2$ with an independent carrier (K replacing Ba) and spin doping (Mn replacing Zn), some of the previous limitations are overcome: the absence of carriers in II–IVs and the low-Mn solubility in III–Vs. With 30% K and 15% Mn doping, their $T_C \sim 230$ K [48] exceeds the value in (Ga, Mn)As. Selected highest reliable experimental $T_C$ reported for the Mn-doped DMS is shown in Fig. 8(b). Circles are given for GaN, which has about 30 times smaller $T_C$ than predicted in Fig. 8(a), and (Ba, K)Zn$_2$As$_2$, a current record for DMS. The ab initio studies predict a further increase in $T_C$ [49]. We expect that tunable RT carrier-mediated ferromagnetism will be realized in II–IV–V DMS within 5 years.

Regarding (m3.2), while DMS is often viewed as the materials for multifunctional devices to seamlessly integrate nonvolatile memory and logic [35], other device opportunities could be more viable. DMS-based optical isolators [50], [51] were already commercialized by Tokin Corporation [52]. Such devices, relying on large magnetooptical effects (Faraday and Kerr) that are proportional to the giant Zeeman splitting in DMS, are used to prevent feedback into laser cavities and provide a one-way transmission of light. Even without demonstrating $T_C >$ RT, enhancing RT Zeeman splitting is important for DMS (exceeding a large $g$-factor $\sim50$ for InSb).

Spin lasers [53], [54] are another example of devices not limited to MR effects. They can outperform [55], [56] conventional lasers with injected spin-unpolarized carriers. For spin lasers, electrical spin injection is desirable, currently limited up to $\sim230$ K [57]. $T_C >$ RT in DMS would be beneficial to such spin lasers, both as an efficient spin injector and possibly a tunable active region that could alter the laser operation through the tunable exchange interaction. To remove the need for an applied $B$-field, the perpendicular anisotropy of the spin injector is suitable. We expect RT electrical spin injection in spin lasers by 2020. It is important to critically assess if extrinsic $T_C >$ RT in DMS, from magnetic metallic nanostructures and secondary phases (having GaAs+MnAs, rather than (Ga, Mn)As, a true DMS) is a viable path for RT spintronic devices. RT magnetooamplification was demonstrated in (In, Mn)As-based magnetic bipolar transistor, operating above $T_C < 100$ K of a single-phase (In, Mn)As [58]. Another test for useful extrinsic (multiphase) DMS is a robust RT electrical spin injection. A road map for DMS is shown in Fig. 9.

IV. PERPENDICULARLY ANISOTROPIC FERROMAGNETS

A perpendicularly magnetized system is currently an important building block in spintronic devices, since it enables us to shrink the size of memory bits and to reduce the electric current density required for spin-transfer switching. There are several ways to obtain the perpendicular magnetic anisotropy in a thin film. To use an ordered alloy showing high magnetocrystalline anisotropy is one possible way. If its easy magnetization axis is aligned along the normal direction to the film plane, and the magnetocrystalline anisotropy field overcomes the demagnetization field, the film shows the perpendicular magnetization. Another way is to use the
interface magnetic anisotropy between a ferromagnetic layer and an NM layer. In addition, multilayered structures are useful to obtain perpendicular magnetization.

Toward the perpendicularly anisotropic ferromagnet as a spintronic material, the following milestones have been established:

1) (m4.1): high thermal stability of perpendicular magnetization;
2) (m4.2): structural stability against the thermal process;
3) (m4.3): demonstration of the high spin polarization;
4) (m4.4): reduction of the magnetic damping constant.

(m4.1) means the stability of magnetization at a nanometer scale overcoming the magnetization fluctuation due to the thermal energy. Considering several thermal treatments in device fabrication processes, (m4.2) should be satisfied. (m4.3) is a key determining the performance of MTJ and GMR devices. In terms of spin-transfer torque (STT) magnetization switching, as indicated in (m4.4), the magnetic damping should be small to reduce the electric current density for switching.

An L10-ordered structure exists in the thermodynamically stable phase and is composed of the alternative stacking of two kinds of atomic planes along the c-axis. Thus, L10-ordered alloys, such as FePt, FePd, CoPt, MnAl, and MnGa, exhibit uniaxial magnetic anisotropy along the c-axis direction. When one aligns the c-axis of L10-ordered structure in the normal direction to the film plane, a perpendicular magnetic anisotropy is obtained. Since the L10-ordered structure is thermally stable, L10-ordered alloys have an advantage from the viewpoint of (m4.2). Among the L10-ordered alloys, L10–FePt shows the largest uniaxial anisotropy (Ku) of $7 \times 10^6$ J/m^3 [59], which leads to the excellent thermal stability of magnetization at a reduced dimension, e.g., 4 nm diameter in L10–FePt nanoparticles. This property satisfies (m4.1). Because of its perpendicular magnetization for FePt (001) films, L10–FePt has been regarded as an ideal material for perpendicular recording media in an HDD. In addition, the spin polarization of FePt is theoretically predicted to be approximately 70% [24], which is a good characteristic for a spintronic material. L10-ordered FePt films have already been implemented in both the MTJ [60] and the GMR [24] junctions. In the case of GMR nanopillars consisting of two FePt layers separated by NM Au, the STT phenomena have systematically been examined by tuning the crystalline order of the FePt layer [24]. However, the observed TMR and the GMR ratios are still low for L10–FePt.

Another important issue is that the major L10-ordered alloys contain the heavy transition metals, such as Pt. The Pt atom shows strong spin-orbit coupling, which leads to the significant enhancement of magnetization damping. This feature is an opposite trend to (m4.4). L10–FePd exhibits a large Ku and rather smaller damping constant compared with that of L10–FePt, probably because Pd is lighter element than Pt [61]. However, the usage of such noble metals as Pt and Pd is not suitable from the viewpoint of element strategic trend. Considering these recent demands, a new kind of L10 alloy is eagerly desired, which possesses a large Ku and a small damping constant. One of the candidates is L10–FeNi. Since a paper reported that an L10–FeNi bulk alloy exhibited high uniaxial magnetic anisotropy of $K_u = 1.3 \times 10^6$ J/m^3 [62], L10–FeNi is a future material having a possibility to substitute high Ku materials containing the noble metals and rare earths. Kojima et al. [63] reported the preparation of L10–FeNi thin films with a relatively high Ku of $0.7 \times 10^6$ J/m^3, and also the small damping constant has been reported in L10–FeNi [64].

Another candidate material showing perpendicular magnetization is an Mn-based alloy system, such as L10–MnAl. Recently, epitaxial Mn–Ga films, including L10- and D022-ordered phases, have also been found to exhibit strong perpendicular magnetic anisotropy (Ku = 1.2–1.5 $\times 10^6$ J/m^3) with small saturation magnetization ($M_s = 250–500$ emu/cm^3) and small magnetic damping ($\alpha = 0.0075–0.015$) at RT [65], [66]. Moreover, it has been found that D022-Mn3Ge epitaxial films exhibited Ku of $0.91 \times 10^6$ J/m^3 [67] and $1.18 \times 10^6$ J/m^3 [68]. These Mn-based alloy systems can also be used as a perpendicular magnetized layer for STT application, because the ab initio calculations predicted the high spin polarization of 88% for Mn3Ga [69] and a half-metallic band dispersion for MnGe that leads a high TMR, such as Fe/MgO–MTJs [70], [71]. However, the observed TMR ratios are also still low for L10–Mn–Ga and D022–Mn–Ga [72]. Experimental realization of the high spin polarization is essential for all the ordered alloys to achieve (m4.3).

Multilayered structures, such as Co/Pt, Co/Pd, Co/Ni, and so on, also show the perpendicular magnetization. The main origins for perpendicular magnetic anisotropy in the multilayered structures are as follows: 1) breaking the crystal symmetry at the interface, which leads to the interface magnetocrystalline anisotropy; 2) the effect of magnetostriction due to the interface between different atomic planes; and 3) interface alloying. Although the multilayered films show high magnetic anisotropy, we need to consider the stability of the layered structure against a thermal process. In some cases, the high temperature annealing degrades the layered structure.
and its magnetic properties, which should be improved for (m4.2). Mangin et al. [73] and Meng and Wang [74] also demonstrated the STT switching in CPP-GMR nanopillars with perpendicularly magnetized CoNi and CoPt multilayers, respectively. As in the case of the ordered alloys, however, increasing MR effect and lowering magnetization damping are inevitable issues for the multilayered structures to achieve (m4.3) and (m4.4). To explore the adequate materials combination is one of the ways for the multilayered structure to solve the current problems.

One of the new types of multilayering films is an artificial superlattice grown using nearly monoatomic layer alternation of Co and Pt or Pd. Such ultrathin superlattice films had an annealing stability higher than that of the conventional multilayering films [75].

It has also been reported that the CoFeB/MgO junction shows perpendicular magnetic anisotropy [76]. The perpendicular magnetization components of the CoFeB are induced at the MgO interface, which originates from the interface magnetic anisotropy. The perpendicularly magnetized CoFeB/MgO layers have a significant advantage, because MgO-based tunnel junctions show a high TMR ratio. Actually, it has also been demonstrated that a CoFeB/MgO/CoFeB stack with perpendicular magnetization shows the TMR ratio over 120% and the low STT switching current of 49 μA at a 40 nm-diameter junction. This is promising candidate as a building block for the MRAM cell. However, because the interfacial magnetic anisotropy constant is not large enough, and a thin ferromagnetic layer is required to exploit the interface effect, the small volume of the magnetic layer may give rise to the thermal instability of magnetization in a deeper subnanometer region. (m4.1) is an important step for the perpendicular anisotropic ferromagnets using the interface magnetic anisotropy. In addition, perpendicularly magnetized Heusler alloy layers, where interface magnetic anisotropy is used, are attracting attention as an alternative perpendicularly magnetized system, which may lead to the high spin polarization (m4.3) and a low damping constant (m4.4). Recently, the perpendicular magnetization and the TMR ratio of 132% at RT have been demonstrated using an ultrathin Co2FeAl Heusler alloy/MgO/CoFeB MTJ [77]. These are summarized in Fig. 10.

V. OVERVIEW

In this roadmap, we have identified two key properties to develop new (and/or improved) spintronic devices. The first one is the half-metallicity at RT, which can be achieved by clearing milestones to realize large MR and resulting large spin polarization. The second one is the perpendicular anisotropy in nanoscale devices at RT. This is based on milestones, including large perpendicular magnetic anisotropy and small damping constant. Such development is expected to be achieved not only by the development of these alloys but also by the fundamental understanding on these properties using a well-studied test system, i.e., zincblenides. As summarized in Fig. 11, we anticipate these materials investigated here to realize all Heusler and all oxides junctions. These can be implemented in the next-generation MRAM and high-frequency devices within 35 years.

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Atsufumi Hirohata (M’01–SM’10) was born in Tokyo, Japan, in 1971. He received the B.Sc. and M.Sc. degrees from Keio University, Minato, Japan, in 1995 and 1997, respectively, and the Ph.D. degree from the University of Cambridge, Cambridge, U.K., all in physics.

He was a Post-Doctoral Research Associate with the University of Cambridge and the Massachusetts Institute of Technology, Cambridge, MA, USA. He served as a Researcher with Tohoku University, Sendai, Japan, and RIKEN, Wako, Japan. He became a Lecturer with the University of York, Heslington, U.K., in 2007, where he was promoted to Reader in 2011, and has held a Personal Chair appointment since 2014. His major research interests include spintronic devices and magnetic materials. He has edited the books entitled Epitaxial Ferromagnetic Films and Spintronic Applications (Kerela, India: Research Signpost, 2009) and Heusler Alloys (Berlin, Germany: Springer, 2015). His current research interests include spin injection in ferromagnet/semiconductor hybrid structures, lateral spin-valve devices, magnetic tunnel junctions and Heusler alloys.

Prof. Hirohata is a member of the American Physical Society, the Materials Research Society, the Institute of Physics, the Magnetics Society of Japan, the Physical Society of Japan, and the Japan Society of Applied Physics. He served as a member of the Administrative Committee of the IEEE Magnetics Society from 2012 to 2014, and has been a member of the Technical Committee since 2010.

Hideto Yanagihara received the B.Sc. and M.Sc. degrees in materials science from Tohoku University, Sendai, Japan, in 2004 and 2007, respectively.

He became a Researcher with the National Institute for Materials Science, Tsukuba, Japan, in 2007, where he is currently a Senior Researcher with the Magnetic Materials Unit. His current research interests include magnetic thin films and spintronics devices.

Igor Žutić was born in Zagreb, Croatia, in 1967. He received the B.Sc. degree in physics from the University of Zagreb, Zagreb, in 1992, and the Ph.D. degree in physics from the University of Minnesota, Minneapolis, MN, USA, in 1998.

He held a post-doctoral position with the University of Maryland, College Park, MD, USA, and the Naval Research Laboratory. In 2005, he joined the University at Buffalo, The State University of New York, Buffalo, NY, USA, as an Assistant Professor, where he was promoted to Associate Professor in 2009 and Full Professor in 2013. With E. Tsymbal, he co-edited a book entitled Handbook of Spin Transport and Magnetism (New York: Chapman and Hall/CRC Press, 2011). His current research interests include superconductivity, magnetism, and spintronic devices.

Dr. Žutić is a member of the American Physical Society, and has been a member of the Technical Committee of the IEEE Magnetics Society since 2013. He was a recipient of the National Science Foundation CAREER Award in 2006, the National Research Council/American Society for Engineering Education Post-Doctoral Research Award in 2005, and the National Research Council Fellowship from 2003 to 2005. Following the success of Spintronics 2001: International Conference on Novel Aspects of Spin-Polarized Transport and Spin Dynamics, Washington, DC, USA, which he proposed and chaired, he was invited to write a comprehensive review titled Spintronics: Fundamentals and Applications for the Reviews of Modern Physics. The review written with J. Fabian and S. D. Sarma is currently among the most cited articles in spintronics and magnetism.
Takeshi Seki was born in Shizuoka, Japan, in 1980. He received the B.Eng., M.Eng., and Ph.D. degrees in materials science from Tohoku University, Sendai, Japan, in 2002, 2003, and 2006, respectively.

He was a Post-Doctoral Researcher with Tohoku University and Osaka University, Osaka, Japan. He then became an Assistant Professor with Tohoku University in 2010. His major research interests include the materials development for spintronic devices. His current research interests include spin transfer phenomena, magnetization dynamics in a nanosized region, and magnetization reversal mechanism.

Shigemi Mizukami was born in Sendai, Japan, in 1973. He received the B.Sc., M.Sc., and Ph.D. degrees in applied physics from Tohoku University, Sendai, in 1996 and 1998, respectively.

He was a Research Associate with Nihon University, Tokyo, Japan, where he was promoted to Lecturer in 2005. He became an Assistant Professor with Tohoku University in 2008, where he was promoted to Associate Professor in 2011, and also Professor in 2014. His major research interests include spintronic devices, high frequency magnetism, and magnetic materials. His current research interests include ultrahigh-frequency magnetization dynamics, low damping Heusler materials, and perpendicular magnetic tunnel junctions based on Mn-based tetragonal Heusler-like alloys.

Prof. Mizukami is a member of the Magnetics Society of Japan, the Physical Society of Japan, the Japan Society of Applied Physics, and the Japan Institute of Metals and Materials. He was one of the guest editors of the Special Issues: Advancement in Heusler compounds and other spintronics material designs and applications (Journal of Physics D: Applied Physics in 2015).

Raja Swaminathan (SM’10) received the Ph.D. degree in materials science and engineering from Carnegie Mellon University, Pittsburgh, PA, USA.

He is currently a Package Architect with Intel, Santa Clara, CA, USA, for next-generation server, client, and system on a chip (SOC) products. His primary expertise is on delivering integrated hardware virtual machine (HVM) friendly package architectures with optimized electrical, mechanical, and thermal solutions. He is also an expert in magnetic materials synthesis, structure, and property characterizations, and has seminal papers in this field. He has authored 18 peer-reviewed publications, and holds 13 patents.

Dr. Swaminathan, is an ITRS Author and iNEMI Technical WG Chair on packaging and design. He has served on the IEEE Microelectronics and Magnetics Technical Committees.