Heusler alloys for metal spintronics

Atsufumi Hirohata* and David C. Lloyd

Heusler alloys have been theoretically predicted and experimentally demonstrated to be an ideal spin source due to their half-metallicity at room temperature. The half-metallicity also offers low Gilbert damping constants for fast magnetization reversal with low switching current density. These intrinsic properties can offer better operationability in spin-transfer-torque-based devices. In addition spin–orbit torque can be exerted using Heusler alloys for spin Hall and caloritronic effects. These properties can be precisely controlled by substituting the constituent elements in a Heusler alloy. We review the recent development on these spintronic devices and summarize their future perspectives.

Introduction to spintronics

Spintronics is one of the emerging fields in nanoelectronics as a post-Moore solution.\textsuperscript{1,2} Metal spintronics has been focusing on the importance of the atomically smooth interfaces by utilizing the device fabrication techniques developed for the Si-based complementary metal oxide semiconductor (CMOS) technology.\textsuperscript{3} In comparison with the semiconductor spintronics with up to a micrometer-long depletion layer at the edge and interfaces of the patterned devices, the metal spintronics can suffer from spin-dependent electron scattering due to atomic roughness,\textsuperscript{4} which may form edge magnetic domains for example. Additional small resistance in metal spintronics by over two orders of magnitude can be advantageous for sustainable development of nanoelectronic devices with nonvolatility in data storage in the form of a magnetic moment in a ferromagnetic layer.

Spintronic devices can be categorized into two types based on the dominant interactions employed: (1) between electron spins and (2) spin–orbit interactions. As shown in Figure 1, the former type uses spin-transfer torque (STT), which is provided from a spin-polarized electron to the others. This torque was formulated by Slonczewski\textsuperscript{5} and Berger\textsuperscript{6} independently. STT can be described as proportional to the cross product of the unit vector of a magnetization and the change in an electrical current density. Because the magnitude of STT can only be increased linearly up to the Walker breakdown, the efficiency of STT can be dependent on the spin polarization of the ferromagnetic material used to generate spin-polarized electrons. For the spin generation, a Heusler alloy has great potential due to the half-metallicity at room temperature (RT).\textsuperscript{7} The generated up- and/or down-spin electrons can be treated as two individual currents as originally proposed by Mott.\textsuperscript{8} This simple model can explain the magnetoresistive (MR) phenomena, such as giant magnetoresistance (GMR)\textsuperscript{10,11} and tunneling magnetoresistance (TMR).\textsuperscript{12} The GMR and TMR junctions consist of two ferromagnetic layers sandwiching a nonmagnetic and insulating layer, respectively. The magnetization of one of the ferromagnets, a free layer, are switched by a magnetic field or spin torques to form parallel and antiparallel configurations to another one, a reference layer, achieving changes in their resistance due to spin scattering. These MR phenomena improved the signal-to-noise ratios in a reading head of a hard disk drive (HDD) and a magnetic sensor, and also used as a cell of a magnetic random-access memory (MRAM).\textsuperscript{3} The latest MRAM commercially available uses a STT-based perpendicularly magnetized cell, which has been used in an aeroplane, a motorbike, and a life tracker due to the robustness under extreme conditions. STT has also been used for the displacement of a magnetic entity formed in a ferromagnetic nanowire (e.g., a...
magnetic domain wall\textsuperscript{13} and a magnetic skyrmion\textsuperscript{14} leading to a racetrack memory (see Heusler Antiskyrmions section in this special issue for details).

The second type of spintronic devices utilizes the spin–orbit interactions (SOI) as formulated by Dirac initially.\textsuperscript{15} SOI is used to predict spin imbalance generation in a paramagnetic metal by introducing an electrical charge current without the application of an external magnetic field, as known as the spin Hall effect\textsuperscript{16,17}. By introducing a spin current instead, a spin Hall voltage can also be generated without a magnetic field, resulting in the inverse spin Hall effect. This is induced by skew and side-jump scattering of conduction electrons at a finite angle for low impurity concentration and with a finite shift by high impurity concentration, respectively, by SOI and is theoretically equivalent to anomalous Hall effect in a ferromagnet. Experimental observation of the spin Hall effect was performed in GaAs using magneto-optical Kerr effect imaging.\textsuperscript{18} The macroscopic scattering angle due to the spin Hall effect can be measured as the spin Hall angle $\theta_{\text{SH}}$. Because SOI is determined as the product of SOI constant, spin and orbital moments, $\theta_{\text{SH}}$ can be maximized by selecting a material with a large SOI constant, which is typically proportional to the spin Hall conductivity. The relationship between the charge and spin currents can be defined as (Spincurrent) $= \theta_{\text{SH}} \times$ (Charge current). A large SOI constant can also modify the spin configuration in a neighboring ferromagnet via the Dzyaloshinskii–Moriya interaction (DMI).\textsuperscript{19,20} The spin Hall conductivity has been reported to be enhanced by the presence of the 4$^d$ and 5$^d$ electrons\textsuperscript{21} as well as the partially filled f-orbitals.\textsuperscript{22} Such interactions can stabilize a magnetic quasi-particle, such as a magnetic skyrmion,\textsuperscript{23} consisting of a vortex configuration of magnetic spins continuously rotating along the radius, tangential and onion-shape from the perpendicularly oriented core to the outer spins antiparallel to the core. The magnetic skyrmion can be used as a magnetic information bit in a racetrack memory because of the advantages in the size and critical current density. SOI also controls the spin-polarized carrier transport in a semiconductor as predicted by Datta and Das\textsuperscript{24} as detailed in Semiconductor Spintronics in this special issue. SOI can exert spin–orbit torque (SOT), which can also switch a magnetization in a ferromagnet.\textsuperscript{25} SOT is known to be more efficient and faster than STT by almost one order of magnitude, which can be implemented in the next-generation MRAM for the power reduction and fast operation.\textsuperscript{3}

**Spin-transfer torque**

*Magnetoresistance*

A key measure for a STT-based device is a magnetoresistance (MR) resistance, for example, GMR and TMR ratios, which can be defined as

$$\text{MR ratio} = \frac{\Delta R}{R} = \frac{R_{\text{AP}} - R_{\text{P}}}{R_{\text{P}}}. \quad (1)$$

Here, $R_{\text{AP}}$ and $R_{\text{P}}$ represent the resistance measured with the antiparallel and parallel configurations of the magnetizations of the two ferromagnetic layers sandwiching a non-magnetic or insulating layer in a GMR or TMR junction. The GMR junctions generally show less than 100% MR ratios at RT, but with low resistance-area products ($RA$, below 1 $\Omega \ \mu m^2$) for the reduction of power consumption to be competitive with major RAMs due to their all-metallic nature as shown in Figure 2. To date, the maximum GMR ratio is reported to be 82% at RT with $RA$ of 31 m$\Omega \cdot \mu m^2$ for Co$_2$FeGa$_{0.5}$Ge$_{0.5}$ (10)/NiAl (0.21)/Ag (5)/NiAl (0.21)/Co$_2$FeGa$_{0.5}$Ge$_{0.5}$ (10) (thickness in nm),\textsuperscript{26} where NiAl is used to improve lattice matching between the ferromagnetic Co$_2$FeGa$_{0.5}$Ge$_{0.5}$ Heusler alloy layer and the nonmagnetic Ag spacer. The TMR junctions, on the other hand, show much

![Figure 1. List of spintronic devices.\textsuperscript{1,9} MRAM, magnetic random-access memory; HDD, hard disk drive; GMR, giant magnetoresistance; TMR, tunneling magnetoresistance](image-url)
larger MR ratios up to 604% at RT for a magnetic tunnel junction (MTJ) consisting of Co$_2$Fe$_{0.4}$B$_{0.6}$ (6)/MgO (2.1)/Co$_2$Fe$_{0.4}$B$_{0.6}$ (4) (thickness in nm) at RT using the $\Delta$-band matching between the ferromagnet and MgO layers for coherent tunneling as theoretically predicted, 28-30 followed by experimental demonstration. 30, 31 For both junctions, further improvement in the MR junctions is necessary to satisfy the requirements for the set goals of 10 Gbit MRAM and 2 Tbit/in$^2$ HDD as shown in Figure 2. Recently perpendicularly magnetized CoFeB/MgO/CoFeB MTJs have successfully met the requirement for the 1 Gbit MRAM with RA of 18 $\Omega$ m$^2$ and TMR ratio of 124% at RT. 32

For both GMR and TMR junctions, a half-metallic Heusler alloys should be able to achieve almost infinite MR ratios at RT. However, as shown in Figure 3a, the TMR ratios have not been increased over 10 years since the previously mentioned report with coherent tunneling. 27 Great efforts have been devoted using a half-metallic Heusler alloy film as a ferromagnetic layer in MTJs. The maximum TMR ratio is reported to be 342% at RT (616% at 4 K) with $\Delta m = 8 \times 10^3 \Omega$ m$^2$ for epitaxial $\mbox{B}_2$Co$_2$FeAl (5)/Co$_{0.75}$Fe$_{0.25}$ (0.5)/MgAl$_2$O$_4$ (1.2)/$\mbox{B}_2$Co$_2$FeAl (5) (thickness in nm). 33 Here, the MgAl$_2$O$_4$ tunneling barrier is used instead of MgO to improve the lattice matching of the junction with maintaining the coherent tunneling nature with the $\Delta$-band connection. By improving the quality of the Heusler alloy films by using $\mbox{L}_2\mbox{-CoFeAl}_{0.5}$Si$_{0.5}$ in MTJ consisting of $\mbox{L}_2\mbox{-CoFeAl}_{0.5}$Si$_{0.5}$Si$_{0.5}$ (30)/MgO (1.8)/$\mbox{L}_2\mbox{-CoFeAl}_{0.5}$Si$_{0.5}$Si$_{0.5}$ (5), the TMR ratios are reported to become 386% at RT (832% at 9 K) with $\Delta m = 8 \times 10^3 \Omega$ m$^2$. 34 For polycrystalline sputtered full Heusler MTJs, on the other hand, much smaller TMR ratios have been reported due to the electron scattering at grain boundaries, for example, TMR of 175% at RT using a Co$_2$FeAl (2)/MgO (1.95)/Co$_{0.75}$Fe$_{0.25}$ (5) (thickness in nm) structure, 35 confirming the quality of the Heusler alloy films and the interfacial smoothness in MTJs control the TMR ratios.

In these junctions, the decrease in the TMR ratio with increasing temperature is much faster than the temperature dependence of the magnetization as described by the empirical law of $T^2$. For example, for MTJ consisting of $\mbox{L}_2\mbox{-CoFeSi}$ (3)/MgO ($-2.5$)/$\mbox{L}_2\mbox{-CoFeSi}$ (3) (thickness in nm), TMR ratio at 4 K is reported to be 1910% but that is reduced down to 354% at RT. 36 The steep reduction in the TMR ratio with increasing temperature below ~ 100 K is found to be a unique feature of the coherent tunneling as shown in Figure 3b. According to Shang’s model, 37 the conductance for the parallel and antiparallel configurations, $G_P$ and $G_{AP}$, respectively, at a finite temperature $T$ can be described as

$$G_{P(AP)} = G_0(T) \left[1 + (-)P^3 \right] + G_{IS}(T),$$

where $G_0$ is the mean conductance defined as $G_0(T) = G_0CT/\sinh CT$, $P$ is the tunneling spin polarization, $m$ is the reduced magnetization due to the empirical law as $m(T) = AT^2$, and $G_{IS}$ is the spin-independent conductance due to the two-step hopping via defect states in a MgO barrier expressed as $G_{IS}(T) = BT^4$. Here $C = 1.387 \times 10^{-3} d/\sqrt{\phi}$ with the barrier width $d$ in nm and height $\phi$ in eV. $G_0$ and $B$ are constants. $A$ is the parameter characterizing spin fluctuation at the interface between a ferromagnet and a barrier. In the coherent MTJs, the temperature dependence of the TMR ratios is mainly induced by the spin fluctuation at the ferromagnet/MgO interfaces and the spin-independent hopping within the MgO barrier. 41 In the Heusler alloy-based MTJs, the spin fluctuation at the Heusler alloy/MgO interfaces may be the dominant cause to reduce the TMR ratios faster than the empirical law, possibly due to the formation of a small fraction of atomically disordered phases in the vicinity of the MgO barrier. The elimination of such disordered interfacial phases is critical to improve the TMR ratios at RT further to utilize the half-metallicity of the Heusler alloys. With such interfacial phases, the interfacial resonant states within the half-metallic bandgap are calculated to be formed at the half-metal/insulator interfaces under the presence of atomic mixing, breaking the half-metallicity. 42 Here, the tunneling rate is slower than the spin-flip rate, resulting in the interfacial states for the minority spins to be effectively coupled to the metallic spin reservoir of the majority spin states.

Additionally, the spin polarization of a half-metallic Heusler alloy is calculated to be reduced significantly at temperatures much lower than their Curie temperatures even in a bulk form due to the change in hybridization by spin fluctuation. 43 The other theoretical calculations also revealed that an exchange stiffness constant can be reduced significantly at the interface between a Co-based Heusler alloy and a MgO barrier as
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compared with that in a bulk form due to the instability of the Co moment. These conditions unique to a Heusler alloy-based MTJ can enhance the thermal fluctuation of spin moments in the Heusler alloy layer, inducing the large temperature dependence of a TMR ratio. By eliminating such interfacial states, the temperature dependence of the TMR ratios can be minimized as observed for the conventional Al-O-based MTJs as shown in Figure 3b.

Magnetic anisotropy

A Heusler alloy typically has a cubic crystalline structure, which may not induce strong magnetic anisotropy. For the device implementation, however, perpendicular anisotropy needs to be induced by modifying the crystalline structures of the alloys or introducing interfacial coupling via a layer below, such as MgO and W. For the former structural modification, binary Heusler alloys in the tetragonal D022-phase have been reported to induce perpendicular magnetic anisotropy. For example, ferrimagnetic Mn3Ga has been reported to possess a large uniaxial anisotropy of $1 \times 10^6 \text{erg/cm}^2$ and high Curie temperature of ~770 K. Mn$_3$Ga MTJ has then been reported to show the TMR ratio of 40% at RT for Mn$_{0.62}$Ga$_{0.38}$ (30)/MgO (1.8)/CoFeB (1.2) (thickness in nm). For the latter interfacial coupling, similar to the Fe/MgO/Fe and CoFeB/MgO/CoFeB systems, perpendicular anisotropy has been induced by attaching a MgO tunnel barrier to a Heusler alloy layer. For example, MTJ with Co$_{0.25}$Fe$_{0.75}$/MgO (1.8)/Co$_{0.3}$Fe$_{0.7}$/B$_{0.3}$ (1.4) (thickness in nm) was reported to exhibit a large perpendicular anisotropy of $1 \times 10^6 \text{erg/cm}^2$ and high Curie temperature to the temperature to the reduction in the crystallization temperature to the B2 phase at 335 K within 5 min. deposition time for 10~15-nm thick Heusler alloy film. By implementing in a GMR junction, the GMR ratio of 0.03% at RT was reported for W (10)/Co$_2$FeAl$_{0.5}$Si$_{0.5}$ (12.5)/W (1.2) or Ag (3)/Co$_2$FeAl$_{0.5}$Si$_{0.5}$ (2.5) (thickness in nm). By optimizing the interfacial conditions further, larger perpendicular magnetic anisotropy and MR ratios can be induced.

Magnetic dynamics

For the improvement of data access time in magnetic memories to replace the level caches and temporary memories in the current CMOS-based computation, faster magnetization reversal needs to be achieved, requiring a ferromagnet with a low Gilbert damping constant $\alpha$. $\alpha$ is an important parameter for the reduction of a switching current. To date, Ni$_{0.8}$Fe$_{0.2}$ has been used as a low damping material as shown in Figure 4. Among the conventional ferromagnets, Co$_{0.25}$Fe$_{0.75}$ has been reported to exhibit the smallest $\alpha$ of $(5 \pm 1.8) \times 10^{-4}$. $\alpha$ can theoretically be proportional to the density of states (DOS) filled by electrons at the Fermi level. This means the

Figure 3. (a) Development of magnetoresistance (MR) ratios at room temperature and (b) temperature dependence of MR ratios.
half-metallic ferromagnets can be an ideal alternative for the improvement due to their limited number of DOS in nature. As shown in Figure 4, the Heusler alloys typically exhibit $\alpha < 0.01$. For example, $\alpha$ of 0.0005 was reported for Co$_2$MnSi and Co$_2$MnGe.

For the integration of such spintronic devices, large perpendicularly magnetic anisotropy is also essential. For the conventional ferromagnets, FePt shows the largest $K_u$ in the vicinity of the Fermi level.

Spin–orbit torque

Spin Hall magnetoresistance

The spin Hall effect was predicted to induce the corresponding resistance changes with respect to the field (i.e., spin Hall magnetoresistance [SMR]). SMR was experimentally measured in Y$_3$Fe$_5$O$_{12}$ (YIG)/Pt bilayer. By using antiferromagnetic Heusler alloys, a large anomalous Hall effect and a large $\theta_{SH}$ of $(5.3 \pm 2.4)\%$ was reported for Mn$_3$Sn. This is due to a weak ferromagnetism induced in the noncollinear antiferromagnetic alignment $(\sim 0.002\mu_B$/Mn). Such Heusler alloys were employed to develop an antiferromagnetic memory with the writing capability at THz frequency. The SMR ratios are dependent on the spin Hall angles $\theta_{SH}$, which is listed in Figure 5. For Mn$_3$Sn, $\theta_{SH}$ is reported to be $(5.3 \pm 2.4)\%$ with the spin diffusion length of $(0.75 \pm 0.67)$ nm at RT.

Spin caloritronics

SOT also induces spin caloritronic effects, such as spin Seebeck and Nernst effects (SSE and SNE, respectively). A spin current was originally generated under thermal gradient formed in Ni$_{0.8}$Fe$_{0.2}$/Pt. For SSE and SNE, the figure of merit $ZT$ can be determined as

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

and

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

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Further development of Heusler alloys is essential for the SOT-based device applications.
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Figure 5. Correlations between the magnetic anisotropy constant and the spin Hall angle $\theta_{\text{SH}}$ (closed symbols) as well as the spin Seebeck/Nernst coefficient (open symbols). Single films, multilayers with conventional, and half-metallic Heusler alloys are shown in blue and red symbols, respectively. Data are taken from References 64, 70–72.

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Conflict of interest
The authors confirm there is no conflict of interest.

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