# Thermally assisted spin transfer torque switching of amorphous GdFeCo for magnetic random access memories

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## Thermally assisted spin transfer torque switching of amorphous GdFeCo for magnetic random access memories

(MRAM 応用に向けたアモルファス GdFeCo 層の熱アシストスピン注入磁化反転の研究)

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By

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#### **Chapter 1 Introduction**

#### 1.1. Development of non-volatile memory

Under the unprecedented growing capacity of information produced in the world by mobile communication, super-computing systems and data centers, which are shown in Fig. 1.1, the traditional memory technologies, e.g. SRAM, DRAM, are facing significant challenges in fabrication process, power consumption and device reliability at 22nm and beyond technology node [1-2]. Therefore, significant efforts have been made on the studies and developments of emerging non-volatile memory (NVM) technologies, that combine the attractive features such as high-density, high-speed, low-power consumption, random-accessibility, non-volatility and unlimited endurance. Some promising NVM candidates include flash memory [3], phase change memory (PCM) [4], spin-transfer torque magnetic memory (STT-RAM) [5], ferro-electric RAM (FRAM) [6] and Resistive random access memory (RRAM) [7] were proposed during the last decades. Among these devices, the flash memory has been the most successful semiconductor technology which sustains the explosion of digital electronic market.

#### **1.1.1. Solid-state memory**

Today's dominant solid-state memory technologies, static RAM, dynamic RAM, and Flash memory, have been existed for a long time. Each technology executes just one single task very well, but many systems need all of the three memories to deliver overall excellent performance at effective cost. However, combination of these memory solutions will largely increase the system complexity.

Among these memory technologies, Static RAM has excellent read and write speeds, and requires little power for data retention. However, its large cell size makes it impractical for main memory of system on chip (SOC). Dynamic RAM uses a single transistor and a storage capacitor per cell and thus provides a denser architecture than static RAM, but dynamic RAM requires continual power input to refresh its bit state every few milliseconds. Therefore, large amounts of dynamic



RAM are impractical for portable electronics with limited battery life.

Fig. 1.1 Historical trend and future prospect of storage capacity needed for information production in the world.

#### **1.1.2.** Conventional non-volatile memory

In contrast to static and dynamic RAM, Flash memory offers non-volatile feature which is highly desirable in portable electronics, because of little power consumption. Flash memory also has high density and moderately fast read time, but its write mode is too slow and write endurance is far limited. There are two gates in flash memory's design. One gate is the control gate (CG) like in metal-oxide-semiconductor (MOS) transistors, but the second is a floating gate (FG) that is insulated with all around by an oxide layer. The FG is located between the CG and the substrate as shown in Fig. 1.2. Because the FG is isolated by its insulating oxide layer, carriers can be trapped there and thus the information is stored.



Fig. 1.2 A typical floating gate memory structure with continual FG.

For storing the information, carriers need to be transferred through the insulation tunnel barrier, which exhibits several disadvantages: the fastest programming times are in the range of µs which is still too slow compared to volatile memories. Additionally, its scalability, working voltage and endurance also needs to be optimized. In 2005, Freescale manufactured the world's first memory array based on silicon nano-crystals (NCs), which is believed to replace conventional floating gate type Flash memory. Nano-crystal memories show better scalability since their tunnel oxide thickness can be reduced without seriously affecting data retention. For the development of the nano-crystal memory, retention and narrow program window has remained to be a main issue for practical applications. From the integration's point of view, uncontrollability of the exact number of Si NCs in each transistor cell could cause threshold voltage shift for individual cells and degrades the performance of chips.

In general, the goal is a universal memory that combines the superior attributes of static RAM, dynamic RAM, and Flash. Such a memory would improve system performance and reliability by avoiding data transfer between multiple memories, and would reduce overall system cost. One of these candidates is the magnetic RAM (MRAM).

#### 1.1.3. Novel non-volatile memory devices

Recently, several kinds of novel non-volatile memories were developed. In this section, other novel non-volatile memories including Phase-change memory (PCM), Ferroelectric RAM (FeRAM) and Resistive random-access memory (RRAM) are briefly introduced.

Phase-change memory (PCM) exploits the unique behavior of chalcogenide glass. In the early development of PCM, heat produced by an electric current through a heating element, generally made of TiN, was used to alter the phase of the glass between amorphous and crystalline. PCMs also have the ability to achieve a number of distinct intermediary states, thereby having the ability to exhibit multiple bits in a

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single cell, but the difficulties in writing these multiple bits has prevented these capabilities from being implemented in devices [25].

Ferroelectric RAM (FeRAM) is a random-access memory similar in architecture with DRAM but uses a ferroelectric layer instead of a dielectric layer to achieve non-volatility. FeRAM is one of some growing alternative non-volatile memory technologies that offer the same functionality as flash memory. FeRAM has the following advantages over flash memory: lower power consumption, faster write performance [26] and a much larger write-erase cycles (exceeding 10<sup>16</sup> for 3.3 V devices). Disadvantages of FeRAM are much lower storage densities than flash devices, storage capacity limitations, and higher cost.

Resistive random-access memory (RRAM) is a non-volatile memory type sharing some similarities with PCM. The basic idea is that an insulating layer can be changed to a conduction layer through a filament or conduction path formed after application of a sufficiently high voltage. Several mechanisms have been proposed to explain the conduction path formation. The conduction path formation can arise from different mechanisms, including defects, metallic atoms migration, etc [7]. Typical materials systems for RRAM include Ge<sub>2</sub>Sb<sub>2</sub>Te<sub>5</sub>, AgInSbTe, NiO or TiO<sub>2</sub>, etc. Compared to PRAM, RRAM operates at a faster timescale (<10ns), while compared to MRAM, it has a simpler, smaller cell structure. Compared to flash memory, a lower voltage is sufficient and hence it can be used in low power applications. A significant hurdle to realize the potential of RRAM is the high requirement of switching endurance caused by destructive readout based on high current measurements.

#### 1.1.4. Magnetic random access memory (MRAM)

Magnetic RAM (MRAM) is currently one of the promising contenders for non-volatile memory with universal characteristics mentioned above. It combines non-volatility with relatively high read and write speeds and unlimited endurance. Furthermore, the MRAM storage element, which is usually a magnetic tunnel junction (MTJ) that consists of insulation tunnel barrier sandwiched by two magnetic electrodes, resides in the metallic interconnect spot. A bias of  $\sim 0.3$  V is applied to read the bit, measuring the amount of current that flows through the bit. Writing is achieved by flowing current through two perpendicular write lines, bit line and word line, as shown in Fig. 1.3. The lines can be cladded with magnetic material to focus the applied field to the bit for improving write selectivity.

The first generation of MRAM is the toggle MRAM in which a full rotation is achieved only if a bit encounters both field pulses [8]. The MTJ long axis (easy axis) is oriented at 45 degrees from the word line-bit line grid, as shown in Fig. 1.4. The free layer of the MTJ is made up of a synthetic antiferromagnetic (SAF) free layer consist of the typical non-magnetic layer ruthenium that can introduce strong Ruderman-Kittel-Kasuya-Yosida (RKKY) interlayer coupling. No net magnetic moment is existed in the SAF free layer. The timing sequence of the applied fields to the selected cell is shown in Fig. 1.5 (a). The word-line writing current is applied before a bit-line writing current which flows with a phase delay. The behaviors of the two magnetic layers with magnetization  $M_1$  and  $M_2$  under different phases of the toggle write are shown in Fig. 1.5 (b). During idling ( $t < t_1$ ),  $M_1$  and  $M_2$  of the SAF free layer are in an antiparallel state and lay in the easy-axis. At  $t_1 < t < t_2$ , spin-flop occurred by the word-line field applied at 90° from horizontal axis when its value is greater than spin-flop field  $H_{SF}$ .



Fig. 1.3 Graphic illustration of scheme of toggle-MRAM.



Fig. 1.4 The MTJ film stack of a toggle-MRAM and the layout of the cell array [8].

During the  $t_2 \le t \le t_3$ , both the word-line and bit-line fields are applied. The resultant field rotates clockwise from 90° to 45°, and so does the net moment of the SAF free layer. During  $t_3 \le t \le t_4$ , the word-line field is off, so only the bit-line field remains in 0° direction. The net moment of the SAF free layer completes a 180° rotation. After  $t_4$ , each *M* of the SAF free layer relaxes to the easy-axis after the bit-line field is removed.

Toggle-mode switching has been proven to be effective in solving the disturbance problem of the half-selected cells in early MRAM. However, thermally assisted writing MRAM has also been proposed as an alternative of toggle MRAM due to its large current consumption. The basic idea of thermally assisted (TA) MRAM is to create an MTJ stack with strong temperature dependence materials such that heating a bit will lower its switching field and thereby reduce the writing current. In the TA-MRAM, the energy barrier for magnetization reversal of the selected bit is required to be reduced dramatically by heating.



Fig. 1.5 (a) the timing sequence of the write field pulse and (b) behavior of the magnetization in a SAF free layer [8].

Methods for heating include use of the write current itself [10,11] and the use of a high current pulse through the MTJ along with the writing field pulses [12], which are shown in Fig. 1.6. The most studied MTJ stack for thermally assisted writing is the one in which the free layer is pinned by an antiferromagnetic (AF) layer so that it does not switch by ambient heat fluctuation. The AF layer has a blocking temperature that is dependent on the layer thickness and choice of AF alloy, above which it is no longer effective to pin the free layer magnetization and then the free layer can be switched by applied field. The thermally assisted approach has a possible additional benefit for lower aspect ratio bits. The stability of bits relies mainly on the exchange coupling strength to the AF layer. Nevertheless, the advanced thermally assisted switching also introduces several new challenges. Additional layers of new materials are needed in the MTJ stack to manage the heat dissipation [13], significantly increasing the complexity of the material design and the micro-fabrication process. Moreover, the new reliability issues will also need to be solved.



Fig. 1.6 (a) Writing procedure of conventional MRAM; (b) TA-MRAM writing procedure [12].

In conclusion, although the commercial success of the 4Mbit MRAM chip, reduction of the bit size associated with the higher magnetic field will require much higher current that may exceed the capability of CMOS circuit when the technological node shrinks to sub-100nm region. The high current will also induce large ohmic heat loss in the metallic wires. Down-size scaling would make this situation worse because of the requirement of higher switching current, while the resistance of the writing wire increases dramatically. To solve these problems, spin transfer torque RAM (STT-RAM) is emerged recently [14].

#### 1.1.5. STT-switching random access memory (STT-RAM)

It has been long understood that the spin-polarized current carries spin angular momentum by which torque is applied to local magnetic moment when the moment interacts with the spin-polarized current. Recently it has been fully understood and studied that this spin torque can cause tiny magnetic bits to precess, and even switch when the device size is on the order of 100 nm or less [13-17]. Analysis shows that the essentials of spin transfer torque (STT) switching can be understood through a simple model with conservation of angular momentum. It can be shown that electrons leaving a ferromagnetic electrode have net electron spin moment parallel to the local moment of that layer, whereas electrons reflected from the surface of the electrode that are oriented antiparallel to that layer. As a result, switching can occur depending on the magnitude and direction of the current as follows and schematic illustration shown in Fig. 1.7:

(1) When a large enough polarized electrons are injected from the pinned layer into the free layer, the spin moment absorbed by the free layer will switch the free-layer moment from the antiparallel state to the parallel state (AP-P switching). If the free-layer moment is already parallel to the pinned-layer moment, it will remain the magnetic state. This process can be referred to writing a "0".

(2) When a large enough polarized electrons are injected from free layer into the pinned layer, electrons reflected from the surface of the pinned layer are polarized antiparallel to the pinned-layer moment and can switch the free layer to the antiparallel state (P-AP switching), which can be regarded as writing a "1".



Fig. 1.7 Schematic demonstration of STT-switching RAM.

This approach clearly solves the write selectivity issues of the conventional MRAM since the write current is flowing only through the selected cell. Another major advantage of the STT switching approach is the down-size scalability. Indeed, from the analysis of the stability of the solutions of the Landau-Lifshitz-Gilbert equation [15] taking into account the spin-transfer term, it can be known that there is a critical current  $I_c$  that must be exceeded for switching. The critical current at zero temperature, with a collinear geometry, and with no externally applied magnetic field, is approximately given by

$$I_{c} = \left(\frac{2e}{\hbar}\right) \left(\frac{\alpha M_{s}V}{\eta}\right) \left(H_{k} + 2\pi M_{s}\right)$$
(1.1)

where the free layer is described by its Gilbert damping constant  $\alpha$ , saturation magnetization  $M_s$ , volume V, and anisotropy field  $H_k$ . The spin-torque efficiency  $\eta$ depends on the polarization of the injected current and the direction of the current, eis the elementary charge,  $\hbar$  is the reduced Planck constant. In general, the goal in current STT-RAM development is to reduce  $I_c$  so that a minimum-size transistor can be used at each technology node to provide the current, thus achieving the highest possible memory density. There has been substantial progress in reducing  $I_c$  over the past several years, but further reductions are still being desired. By optimizing the MTJ stacks, minimum values of switching current density  $J_c$  of the order of 2MA/cm<sup>2</sup> were obtained for 10ns write current pulse width [16,17]. To further reduce of the  $I_c$ , the straightforward way is to minimize the Gilbert damping constant  $\alpha$  because it is proportional to  $I_c$  according to equation (1.1). This requirement restricts the set of materials that can be used as the free layer because  $\alpha$  has minimum limit in common ferromagnetic alloys. Similarly, using thin layers and small devices to minimize the volume of the free layer, and using alloys with low  $M_s$  is available but still within some limits. Additionally, the magnetic bits must be stable enough against thermal agitation and reducing the volume results in unstable bits unless the anisotropy constant  $K_u$  can be increased to compensate that tendency.

The other option to reduce  $I_c$  is to increase the efficiency  $\eta$  by increasing the polarization of the switching current. However, further increases in polarization produce diminishing gain. For example, Sun and Ralph [18] pointed out that increasing the polarization from 66% to 100% would result in only an 8% effective increase in spin torque. Therefore, there is little room for improvement in  $I_c$  through increasing polarization.

One potential approach for lowering  $I_c$  is to introduce a perpendicular anisotropy component to overcome the shape anisotropy, leading to the magnetization perpendicular to the plane of free layer. To achieve current injection switching of free layer other than the spin current induced precession of magnetization, the reference layer should also to be perpendicular magnetized. As depicted in Fig. 1.8, approaches of inducing perpendicular anisotropy include the use of free layer materials with perpendicular anisotropy such as TbCoFe [19] and L1<sub>0</sub>-FePt [20-21], the addition of layers that couple to the free layer to induce the anisotropy [22], and the use of ultrathin multilayered free-layer materials [23-24] with perpendicular anisotropy generated through the interfaces in the material.



Fig. 1.8 (a) Perpendicular TbFeCo in MTJ [19]; (b) Co/Ni multilayer in GMR [24].

In this case, the anisotropy term becomes  $H_{k-perp}-4\pi M_s$ , where  $H_{k-perp}$  is the perpendicular anisotropy and the equation (1. 1) becomes to be equation (1. 2) as follows:

$$I_{c} = \left(\frac{2e}{\hbar}\right) \left(\frac{\alpha M_{s}V}{\eta}\right) \left(H_{k-perp} - 4\pi M_{s}\right)$$
(1.2)

Besides the potential for lowering  $I_c$ , such perpendicular-magnetized devices may also be reduced to smaller cell size than those with in-plane anisotropy, because no in-plane aspect ratio is needed. Although such perpendicular devices appear to have several fundamental advantages, the candidates for perpendicular free layer materials identified so far have lower MR ratio and less ideal magnetic properties than the better established alloys used for in-plane counterparts.

#### **1.2. Spin dependent transport in ferromagnetic metals**

#### 1.2.1. Spin dependent resistivity

The magnetism of materials is carried by electron spin, while electrical transport is caused by the motion of electron charge. The fundamental properties of spin dependent transport are closely related to several parameters describing length scales of electrons motion in metals. The length within which the spin of an electron is conserved is called the spin-flip mean free path  $\lambda_{spin}$  and typically in the 0.1 µm-10 µm range. Due to scattering of electrons, electrical resistivity is governed by the mean free path  $\ell$  of the electrons which is much shorter than the  $\lambda_{spin}$ . Thus, the spin polarization of the current is well defined and the up- and down-spin electrons may be treated independently. This is called Mott's two-current model [27].

Another important length scale is the Fermi wavelength  $\lambda_F$ , which characterizes the electronic states. In general,  $\ell >> \lambda_F$ . This length scale becomes important when interference occurs between wave functions of electrons. The velocity of electrons on the Fermi surface is given by the Fermi velocity  $v_F$  and hence the time scale for an electron with  $v_F$  traveling a distance  $\ell$  is given by  $\tau = \ell / v_F$ , the relaxation time.

The advanced micro-fabrication techniques can make it possible to create artificial structures with scale length *L* shorter than  $\lambda_{spin}$  or  $\ell$  and even close to  $\lambda_{F}$ . In

these cases, novel giant magneto-resistance (GMR), tunnel magneto-resistance (TMR) and ballistic magneto-resistance (BMR) are occurred. GMR occurs when the layer thickness of magnetic multilayers is close to or shorter than  $\ell$ . BMR occurs when the scale of the contact region of two ferromagnets is close to  $\lambda_F$ . TMR is a phenomenon in which the overlap of wave functions of electrons in two separated ferromagnetic metals becomes small but dominates the current flow.

The simplest formula to describe the electrical conductivity  $\sigma$  is given by the Drude formula:

$$\sigma = \frac{e^2 n\tau}{m} \tag{1.3}$$

where *e*, *n*,  $\tau$  and *m* are the electrical charge, carrier density, relaxation time and effective mass of carrier electrons, respectively. For ferromagnets, basically, *n*, *m* and  $\tau$  are all spin dependent and the most important one is the spin dependent relaxation time, since it affects electron scattering strongly. The relaxation time is related to the mean free path  $\ell$  via the relation  $\tau = \ell / v_F$  as mentioned above. For typical ferromagnetic metals,  $\ell$  is much shorter than the spin-diffusion length  $\lambda_{spin}$ , and the spins of the carrier electrons are well conserved in the time scale  $\tau$ .

The spin dependence of  $\tau$  caused by impurity scattering of electrons in ferromagnetic metals may be evaluated by using the formula

$$\tau_s^{-1} = \left(\frac{2\pi}{\hbar}\right) N_i V_s^2 D_s(E_F)$$
(1.4)

which is given by the Born approximation, where  $N_i$ ,  $V_s$  and  $D_s(E_F)$  are the impurity density, scattering potential and density of states (DOS) at the Fermi energy  $E_F$ , respectively. Here, both  $V_s$  and  $D_s(E_F)$  are spin dependent. Equation (1.4) indicates that the relaxation time becomes short as the scattering potential becomes large and the number of final states of the scattering process increases.

The material dependent  $\alpha$ -parameter, defined as  $\rho_{\downarrow}/\rho_{\uparrow}$ , is a factor to characterize the spin dependent  $\tau$  caused by impurity scattering of electrons in ferromagnetic metals, which is given by experiments (Fig. 1.9(a)) and that estimated theoretically (Fig. 1.9 (b)) for TM impurities in Fe and in Cu may be understood by adopting a two-band model [28].  $\rho_{\downarrow}(\rho_{\uparrow})$  is defined as the resistivity for the electrons with spin-down (spin-up) orientation. The model consists of a broad s-like band and narrow d-like bands with a mixing between the s- and d-bands. The results shown in Fig. 1.9 (b) reproduce the experimental tendency rather well. The calculated results can be easily understood in terms of matching/mismatching of the d-electronic states between impurities and host atoms.



Fig. 1.9 (a) Experimental *α*-parameters for 3d, 4d and 5d TM impurities in Fe [29];(b) Calculated *a*-parameters for TM and noble metal impurities in Fe [28].

#### **1.2.2. Giant Magneto-Resistance (GMR)**

Magnetic multilayers are composed of an alternating stack of thin magnetic and non-magnetic layers. The MR is called giant MR (GMR) when the non-magnetic layers are metals and tunneling MR (TMR) for the non-magnetic layer is an insulator. The first observation of antiparallel coupling between magnetic layers was reported by Grünberg et al. [30], for Fe/Cr trilayers. Two years later, Fert's group [31] reported MR as large as 40% for Fe/Cr multilayers, which is shown in Fig. 1.10.



Fig. 1.10 Resistivity change at an external magnetic field for Fe/Cr multilayers [31].

The resistivity decreases with increasing magnetic field due to the change in the alignment of the magnetization of Fe layers. The resistivity is high when the alignment is antiparallel (AP) and is low when parallel (P). The magnitude of the MR is expressed by the so-called MR ratio, defined as

$$MR = \frac{\rho_{AP} - \rho_P}{\rho_P} \tag{1.5}$$

where  $\rho_{AP}$  and  $\rho_{P}$  are the resistivity in AP and P alignments of the magnetizations of the magnetic layers.

AP alignment of the magnetizations is a prerequisite for GMR. A detailed study of the alignment of magnetization has found that the coupling of magnetization in magnetic layers changes as a function of the non-magnetic layer thickness [32]. The coupling between magnetic layers is called interlayer exchange coupling. Fig. 1.11 shows an experimentally determined oscillation of coupling energy as a function of non-magnetic spacer layer thickness [33]. The period of the oscillation is rather long and the magnitude decays as the thickness of the non-magnetic layer increases. GMR with the current flowing parallel to the layer planes is called current-in-plane GMR (CIP-GMR) while CPP-GMR refers to that the current flowing perpendicular to the planes.



Fig. 1.11 Oscillation of coupling energy as a function of spacer layer thickness [33].

CPP-GMR is usually much larger than CIP-GMR in the same architecture, which is a general trend for GMR that can be interpreted as the results of the combination of the geometry of multilayers effects and the spin-dependent resistivity of ferromagnetic metals. Regarding to the mechanism of GMR, both random scattering potentials caused by intermixing of magnetic atom A and non-magnetic atom B at the A/B interfaces and the bulk magnetoresistance effect in the magnetic layers in multilayers cause spin dependent resistivity. Both are crucial for the spin dependence of the resistivity of CPP-GMR and CIP-GMR, while the spin-dependent random potential at the interface likely contributes more in CPP-GMR than CIP-GMR.

#### **1.2.3.** Tunneling Magneto-Resistance (TMR)

Since TMR is caused by tunneling current through an insulating barrier, MTJs are usually made of a thin non-magnetic insulator sandwiched between two ferromagnetic electrodes. A schematic figure of such a junction is shown in Fig. 1.12. The ferromagnetic metals are predominantly Fe, Co and their alloys, while amorphous Al<sub>2</sub>O<sub>3</sub> and single-crystal MgO are the most stable materials for the insulating barrier.



Fig. 1.12 A schematic figure of the ferromagnetic tunnel junction.

An intuitive picture of the tunneling process explained above is shown in Fig. 1.13. In P alignment, majority and minority spin electrons in the left (L) electrode tunnel through the barrier into the majority and minority spin states in the right (R) electrode, respectively. In AP alignment, however, the majority and minority spin electrons in L electrode tunnel into the minority and majority spin states in R electrode, respectively. The difference between the conductances of the P and AP alignments gives rise to TMR. In this scheme, the definition of MR ratio is slightly modified as follows:

$$MR = \frac{2P_L P_R}{1 + P_L P_R} \qquad \text{where} \qquad P_{L(R)} = \frac{D_{L(R)\downarrow} - D_{L(R)\uparrow}}{D_{L(R)\downarrow} + D_{L(R)\uparrow}}$$

and  $D_{L(R)\uparrow}$  and  $D_{L(R)\downarrow}$  correspond to the densities of states in left and right electrodes for spin up and spin down, respectively.



Fig. 1.13 A schematic figure of the tunneling process in magnetic tunnel junctions.

The typical experimental demonstrations of MTJs are based on Fe/AlO<sub>x</sub>/Fe [34] and Fe/MgO(100)/Fe [35] junctions and were shown in Fig. 1.14. Recently, the junction using CoFeB as electrodes in MgO-MTJs make important progress in enhancing the MR ratio, which reaches to 600% at room temperature as shown in Fig. 1.15.



Fig. 1.14 Experimental results for tunneling resistance in (a) Fe/AlO<sub>x</sub>/Fe [34]; (b) Fe/MgO(100)/Fe [35] junctions.

#### 1.2.4. Other MR effects: AMR, BMR and CMR

There are other interesting MR in bulk systems including the anisotropic MR (AMR), ballistic MR (BMR) and colossal MR (CMR). This section, briefly describes these MR phenomena.

Anisotropic MR is a phenomenon that occurs in ferromagnets in which the resistivity depends on the angle between the current and magnetization directions. Fig. 1.16 shows the experimental results obtained for the change in resistivity of Ni when a magnetic field is applied parallel and perpendicular to the current direction [36]. The rapid change in the resistivity at low magnetic fields is due to magnetization rotation. The magnitude of AMR is obtained by extrapolating the resistivity change to H=0.



Fig. 1.15 Summarizes the progress of MR ratio in AlOx-and MgO-based MTJs.



Fig. 1.16 Experimental results for AMR in Ni [36].

When a current flows in a region that has a length scale shorter than the mean free path, the conductance becomes quantized and the effect of scattering may be neglected. Such transport is referred to as ballistic transport and the related MR effect is called BMR. One of the theoretical interpretations of BMR is to attribute the large MR by vanishing of the domain walls (DWs) at the ferromagnetic constriction [37]. Bruno [38,39] has pointed out the narrow width of a DW can sufficiently produce a large MR effect.

Colossal MR is a property of mostly manganese-based perovskite oxides, that enables them to dramatically change their electrical resistance in the presence of a magnetic field. The MR of conventional materials enables changes in resistance of up to 5%, but materials featuring CMR may show resistance changes of several hundred percent. It is believed that CMR is related to insulator-metal transitions in the presence of magnetic field.

#### 1.2.5. Application of MR effects

One of the main applications of the GMR and TMR is magnetic field sensors, e.g., in hard disk drives and biosensors, as well as detectors of oscillations in MEMS. Another important application is MRAM in which a cell has a structure similar to the spin-valve sensor. The value of the stored bits can be encoded via the magnetization direction in the sensor layer.

#### 1.3. Fundamentals and applications of spin injection

#### 1.3.1. Basis of spin injection and torque

When an electron moves into a material, it carries not only a charge but also an angular momentum, spin. In Fig. 1.17, a schematic of electrons transfer from a ferromagnetic material (FM) to a non-magnetic material (NM) through an interface is shown. In the ferromagnetic material, an electric current accompanies the net flow of spins traveling as the spin polarized current in the ferromagnet. The spin polarized current is then injected into the non-magnetic material through the interface. The injected spins are subjected to spin relaxation because of spin-orbit interaction, and they lose their spin orientations as they move away from the interface. It is well known that the magneto-resistance effect occurs at the interface as a current flow through two ferromagnetic materials with different magnetization orientations. In addition, the electron spins injected from the ferromagnetic material on the left-hand side (FM1) into the ferromagnetic material on the right-hand side (FM2) interact with the electron spins in FM2 through exchange interaction and exert torque to local spins in the FM2. When the exerted torque is large enough, a precession of the FM2 local spins can be excited and the magnetization can be switched [40] in FM2.



Fig. 1.17 Spin injection across an interface. (A) Spin injection from a ferromagnetic material (FM) to a non-magnetic material (NM); Spin injection from a FM1 to another FM2 with different magnetization direction [40].

#### 1.3.2. Spin injection magnetization reversal

An electric current passing through a magnetic nano-pillar composed of magnetic multilayers transfers spin angular momentum to the magnetic free layer and changes the direction of the local spins in the layer. The dynamic properties of the local spins can be expressed by the following Landau-Lifshitz-Gilbert (LLG) equation, which includes a spin-transfer torque term [40]:

$$\frac{d\vec{S}_2}{dt} = \gamma \vec{S}_2 \times \vec{H}_{eff} - \alpha \vec{e}_2 \times \frac{d\vec{S}_2}{dt} + g(\theta) \frac{J}{-e} \times \frac{\hbar}{2} \vec{e}_2 \times \left(\vec{e}_1 \times \vec{e}_2\right)$$
(1.6)

The first term is the effective field torque; the second, Gilbert damping torque; and the third, the spin-transfer torque.  $\vec{S}_2 = S_2 \vec{e}_2$  is the total spin angular momentum of the free layer and is opposite to its magnetic moment,  $\vec{e}_2(\vec{e}_1)$  is a unit vector that expresses the direction of the spin angular momentum of the free layer (fixed layer).

The first term determines the precessional motion of  $\vec{S}_2$ . In the second term,  $\alpha$  is the Gilbert damping factor ( $\alpha > 0$ ) which is inversely proportional to the decay time of the precession.  $g(\theta)$  is a coefficient that expresses the efficiency of the spin-transfer process as a function of the relative angle,  $\theta$ , between  $\vec{S}_1$  and  $\vec{S}_2$ . Explicit expressions for  $g(\theta)$  of the GMR structure was expressed as below[40]:

$$g(\theta) = \left[-4 + \left(P^{-1/2} + P^{1/2}\right)^3 \left(3 + \cos\theta\right)/4\right]^{-1}$$
(1.7)

, where P is polarization of the current, the directions of each torque are illustrated in Fig. 1.18. The effective field torque promotes a precession motion of  $\vec{S}_2$  around  $-H_{\text{eff}}$ , while the damping torque tends to reduce the opening angle of the precession. By the effective field and damping torques,  $\vec{S}_2$  exhibits a spiral trajectory and finally aligns antiparallel to the effective field if a junction current J is absent. It must be noted that the direction of  $\vec{S}_2$  is opposite to that of its magnetic moment. Direction of the spin-transfer torque is also illustrated in Fig. 1.18 for the case where both  $g(\theta)$ and J are positive. If the current J is sufficiently large, the spin-transfer torque overcomes the damping torque, resulting in an increase in the opening angle of the precession motion. Due to the angular dependence of the effective damping, the amplification of the precession motion leads to a limit cycle (spin-transfer oscillation, STO) or to a total magnetization reversal (current induced magnetization switching, CIMS) [41].



Fig. 1.18 The directions of each torque and trajectory of the free layer [41].

The CIMS is an important phenomenon in the magnetization reversal process. The conventional approach in magnetization reversal process is magnetic field -induced magnetization reversal. To distinguish CIMS and magnetic field-induced magnetization switching processes, a trajectory for CIMS (a) is compared with that for a magnetic field-induced magnetization switching (b) in a nano-pillar with in-plane magnetization in Fig. 1.19. In the absence of a current and an external magnetic field, the potential shows a double minimum for parallel (P) and antiparallel (AP) configurations of the local spins. In the case of CIMS, the spin-transfer torque does not affect the shape of the magnetic potential but amplifies the precession thereby providing energy to the local spin system. It rapidly turns to opposite direction once the trajectory crosses the critical point. In contrast to this process, the external magnetic field deforms the shape of the magnetic potential and the minimum on the P side is flatted which induces reversal from P state to AP state.



Fig. 1.19 Comparison of the magnetization processes driven by (A) spin-transfer torque; (B) external magnetic field for in-plane magnetized nano-pillars.

#### 1.3.3. Thermal effects in spin transfer torque (STT) switching

The critical current density  $J_{c0}$  required at zero temperature [42] for current driven magnetization reversal in a MTJ with in-plane magnetization can be analytically expressed as:

$$J_{c0} = \frac{2e\alpha M_s t_F (H + H_k + 2\pi M_s)}{\hbar \eta}$$
(1.8)

where *H* is the field applied along the easy axis,  $M_s$  and  $t_F$  are the magnetization and thickness of the free layer respectively,  $\alpha$  is the damping constant, and  $H_k$  is the effective anisotropy field including magneto-crystalline anisotropy and shape anisotropy. The spin transfer efficiency  $\eta$ , is a function of the current polarization (related to the MR ratio) and the relative angle between the free and pinned layer. The global picture of current of STT switching vs. the current pulse width shown in Fig. 1.20 can be categorized as three switching modes including thermal activation, dynamic reversal and precessional switching [43]. For fast precessional switching in a few nano-second regime (<3ns), the switching current is much larger than the intrinsic switching current density [42] which can be described with the equation of

 $J_{c0}(\tau) \propto J_{c0} + \left[\frac{\ln(\pi/2\theta)}{\tau}\right]$ , where  $\tau$  is the pulse width of switching current and  $\theta$  being

the initial angle between the magnetization vector of the free layer and the easy axis. Thermal effect plays an central role in reducing the switching current at long current pulses (>10ns) at specific temperature in the thermal activation regime in which the switching current density is mainly dependent on the current pulse width  $\tau$  and thermal stability factor  $\Delta = K_u V/k_B T$  of the free layer (assuming *H*=0) [42]:

$$J_c(\tau) = J_{c0} \left[ 1 - \frac{k_B T}{K_u V} \ln(\frac{\tau}{\tau_0}) \right]$$
(1.9)

where  $\tau_0 \sim 1$  ns is the inverse of the attempt frequency.  $K_u V$  is the anisotropy energy. It is believed that dynamic reversal is a combination of precessional switching and thermal activation switching in the time scale of 3ns ~10ns between the other two distinct regimes.



Fig. 1.20 STT switching current as a function of current pulse width.

#### **1.3.4.** Applications: STT-RAM and beyonds

The intensive studies have been focused on the reduction of  $J_{c0}$ . The main challenge is to lower  $J_{c0}$  while maintaining the thermal stability  $\Delta$  sufficiently large ( $\Delta > 60$ ). Based on Eq. (1. 8), there may be three approaches to reduce the  $J_{c0}$ : (1) material engineering of the free layer; (2) improvement of the MTJ structure; (3) adoption of MTJ with perpendicular anisotropy.

For material engineering of the free layer, as seen from Eq. (1. 8), the intrinsic current density  $J_{c0}$  can be reduced by using materials with low  $M_s$  and low  $\alpha$ . The replacement of Co<sub>90</sub>Fe<sub>10</sub> with Co<sub>40</sub>Fe<sub>40</sub>B<sub>20</sub> induces a reduction of  $J_{c0}$  by a factor of about 2 due to the reduction of  $M_s$ . Co and Fe are known to exhibit smaller  $\alpha$  than Ni, and the free layer consisting of Co and Fe may be beneficial to keep  $\alpha$  low.

For improvement of the MTJ structure, use of a crystalline MgO barrier instead

of amorphous Al<sub>2</sub>O<sub>3</sub>, increases TMR ratio from 30-70% to 300% at room temperature. The  $J_{c0}$  is expected to be lowered by about two times due to much higher spin transfer efficiency (represented as enhanced TMR ratio). Fig. 1.21 and Fig. 1.22 show the STT switching results for MTJ cells with Al<sub>2</sub>O<sub>3</sub> and MgO barriers [45, 46]. The structures are similar, otherwise the MR ratio in MgO MTJ is 155% compared to that of 42% in Al<sub>2</sub>O<sub>3</sub> MTJ. The average  $J_{c0}$  was obtained to be 2.2 × 10<sup>6</sup>A/cm<sup>2</sup> in MgO MTJs, which is about one third of that (6 × 10<sup>6</sup> A/cm<sup>2</sup>) obtained in Al<sub>2</sub>O<sub>3</sub> junctions. Dual magnetic tunnel junction (MTJ) structures have also been developed consisting of two MgO insulating barriers of different resistances. Two pinned reference layers aligned anti-parallel to one another, and a free layer sandwiched between the two insulating barriers. Diao et al. [47] have successfully built such dual MgO barrier MTJ devices with TMR = 70%, which exhibited a significant reduction in  $J_{c0}$  (= 1.1 × 10<sup>6</sup> A/cm<sup>2</sup>) compared to simple MgO stacks.



Fig. 1.21 Typical magnetic field (a) and current (b) driven magnetization switching for an MTJ sample with an MgO barrier, and field (c) and current (d) driven magnetization switching for an MTJ sample with an AlO<sub>x</sub> barrier. Current pulse width of 30 ms was used in obtaining (b) and (d) [45].

The synthetic free layer  $Co_{40}Fe_{40}B_{20}/Ru/Co_{40}Fe_{40}B_{20}$  employed in MgO MTJs was demonstrated to enable high thermal stability ( $\Delta = 67$ ) and low switching current

density ( $J_{c0} = 8.7 \times 10^6 \text{ A/cm}^2$ ), as compared to the equivalent single free layer  $Co_{40}Fe_{40}B_{20}$  MTJ with ( $\Delta = 56$  and  $J_{c0} = 2.0 \times 10^7 \text{ A/cm}^2$ ) [48].

The perpendicular STT switching was first demonstrated in metallic spin valve systems with out-of-plane anisotropy [24]. In 2007, Toshiba presented first STT switching in MTJs with perpendicular anisotropy composed of TbCoFe / CoFeB / MgO / CoFeB / TbCoFe. The critical current density  $4.67 \times 10^6$  A/cm<sup>2</sup> with a pulse duration of 100ns has been achieved with a large thermal stability  $\Delta = 107$  [49]. More recently, lower  $J_{c0} = 2.7 \times 10^6$  A/cm<sup>2</sup> and higher TMR of 60% have also been demonstrated in MgO based perpendicular-MTJs (p-MTJs) consisting of CoFeB / [Pd/Co]<sub>2</sub> / Pd free layer and FePt / CoFeB reference layer [50].



Fig. 1.22 Spin current switching in TbFeCo / CoFeB based MTJ [49].

#### 1.4. Rare earth-transition metal (RE-TM) layer for spin injection

#### 1.4.1. Conventional materials for spin injection switching

According to Equation (1. 8), reduction of  $J_{c0}$  can be achieved by optimizing the parameters of the material or selecting the material with optimal combination of parameters. For the in-plane magnetization, Fe [51], CoFe [52] and CoFeB [22] were used as free layer in the MTJs. These free layers provide high spin polarization, thus high efficiency  $\eta$ , and low damping  $\alpha$ , which are suitable to lower  $J_{c0}$  of STT switching. However, in-plane MTJs will not be practical to achieve high density STT-RAM. The cell size of STT-RAM has to keep shrinking in order to meet the need of high density. On the other hand, the thermal stability factor  $\Delta$  of each cell, defined by  $K_u V/k_BT$ , should be above a value of 60 to beat thermal fluctuation and secure 10 years data safety. Since V is reduced with size shrinking,  $K_u$  has to be increased to maintain the value of  $\Delta$ . For an MTJ with a diameter of 20nm, if the free layer thickness is 2nm for example, the required  $K_u$  should be larger than  $2.5 \times 10^6$  erg/cc to secure  $\Delta$  above 60. Such a large  $K_u$  will not be obtained by the shape anisotropy of the in-plane free layer. Moreover, the  $K_u$  will dramatically increase to  $10 \times 10^6$  erg/cc for a diameter of 10 nm cell and further exceed  $40 \times 10^6$  erg/cc for 5nm diameter cell.

In contrast, materials with PMA can be used to obtain high  $K_u$  in GMR or TMR devices. Multilayers based on transition metal (TM) like Co, Fe and CoFe and noble metal such as Pt, Pd and Au show perpendicular magnetic anisotropy for a certain range of thickness. The magnetic properties of these materials such as  $M_s$  and  $K_u$  can be easily tailored by adjusting the thicknesses of each layer. The conventional multilayers so far are Co/Pd, Co/Pt and Co/Ni exhibiting relatively large PMA. First principle calculations predicted that the hybridization of Fe-3d and O-2p orbitals may introduce PMA in an MgO/Fe/MgO system [53] and has been confirmed in CoFeB-MgO based MTJ devices [22]. Highly oriented ordered alloys with large PMA have been also studied for p-MTJs. One of the material candidates is L1<sub>0</sub>-FePt. In 2006, Seki et al. integrated L10-FePt in a GMR device and achieved a Ku value of  $3.4 \times 10^7$  erg/cc at room temperature [54]. However, growing FePt with L1<sub>0</sub> ordered phase requires a substrate temperature as high as 500°C, which brings a process integration issue with CMOS transistors. In addition, since a special seed layer / substrate such as MgO (001) is required to introduce texture growth before FePt deposition, the material and structure options are very limited. The same group further demonstrated the STT switching in an FePt/Au/FePt GMR structure, where  $J_c$ was up to  $10^8$  A/cm<sup>2</sup> due to the high  $K_u$  value, weak spin polarization and large damping constant of FePt [54].

#### 1.4.2. Properties of RE-TM layer

The rare-earth – transition metal (RE-TM) alloys form an important class of materials that are applied in permanent magnets, magnetostrictive devices and magneto-optical (MO) recording. The intrinsic properties of the RE-TM alloys can be understood in terms of exchange interactions and one ion anisotropy of RE

atoms/ions. The exchange interactions take place between all unpaired RE-4f and TM-3d electrons. It is generally understood that there are three types of interactions, the RE-RE interaction, TM-TM interaction and RE-TM inter-sub-lattice interaction. Some of the RE-TM intermetallic alloys, such as Sm<sub>2</sub>Co<sub>17</sub>, Nd<sub>2</sub>Fe<sub>14</sub>B, exhibit huge magneto-crystalline anisotropy and they are used as a high performance permanent magnet. Even in amorphous structure, the RE-TM films usually have perpendicular magnetic anisotropy larger than 10<sup>6</sup> erg/cc so that they are practically used as magneto-optical (MO) recording media. The other parameter of amorphous RE-TM materials is the temperature profile of the magnetization. Fig. 1.23 shows the temperature dependence of the magnetization for a GdCoMo amorphous alloy film, whose magnetization disappears at a compensation temperature far below the Curie point.

The constituent element's content is another major factor affecting the magnetic properties in RE-TM alloys. In RE-FeCo alloys, the Curie temperature is mostly influenced by the kind of RE elements. As shown in Fig. 1.24, the Curie temperature is also controlled by varying the Co content. Usually, the RE-TM alloys with various content exhibits two distinct regions: TM-rich and RE-rich regions.



Fig. 1.23 Temperature dependence of the magnetization for a GdCoMo film [55].



Fig. 1.24 Curie temperature as a function of Co content for RE-FeCo alloy.

Even when the Curie temperatures of both types of film are almost the same, the temperature profile of  $M_s$  is much different. It should be noted here that a wide variety of temperature profiles can be easily obtained in the amorphous RE-TM system by adjusting TM and/or RE contents, which results in various unique properties.

In general, RE-TM materials exhibit the following unique properties which is suitable for the STT-RAM application:

- large perpendicular magnetic anisotropy (PMA). The RE-TM materials with large PMA can keep the thermal stability  $\varDelta$  sufficiently large (>60) to retain the information stable for greater than 10 years.

- High resistivity. The high resistivity of RE-TM materials holds the promise for easy generation of Joule heat, which is essential for thermally-assisted STT-RAM which is a main topic of this thesis.

- Adjustable magnetic properties. The magnetic properties such as  $M_s$ ,  $K_{eff}$  and  $H_c$  can be easily adjusted by varying the RE and/or TM element's content to achieve the desired  $M_s$ ,  $K_{eff}$  which is crucial for reduction of current density in STT-RAM.

- Sufficient heat-induced reduction of  $K_{\text{eff}}$ . For the thermally assisted scheme, heat-induced reduction of  $K_{\text{eff}}$  needs to be large enough.

#### 1.4.3. Applications: RE-TM layer for spin injection-switching

STT switching in the MTJ with PMA is important in the application point of view and a RE-TM alloy possessing unique magnetic properties was described in previous section, which in combination inspire the development of STT switching in MTJ with RE-TM materials. This kind of MTJ has been demonstrated in Fig. 1.22. In this MTJ, The TbCoFe / CoFeB free layer with a large coercive field of 1.2 kOe and a large thermal stability factor of 107 at room temperature was switched by a 100 ns pulse current with a current density of  $4.7 \times 10^6$  A/cm<sup>2</sup>.

#### 1.5. Scope of this study for STT-RAM

Rare earth-transition metal (RE-TM) ferrimagnetic amorphous alloy films, such as TbFeCo, TbFe and GdFeCo, etc., have attracted intensive interest for their use in ferrimagnetic physics [56-58] and application in ultrahigh areal density [59] and ultrafast [60-62] magneto-optical (MO) recordings. Due to the unique magnetic properties and high resistivity of the RE-TM alloys, the exploration of RE-TM alloys for spin transfer torque switching is desirable but rarely studied. Therefore, one goal of this thesis is to develop a spin-valve device with GdFeCo as memory layer and to clarify the effect of thermally assisted STT switching. The utilization of GdFeCo memory layer can not avoid the problem of poor thermal stability of GdFeCo layer due to its low effective perpendicular anisotropy constant. So the information stored in the GdFeCo cell is not stable against thermal agitation. To overcome this drawback, the second creative aim of this thesis is to introduce GdFeCo/TbFe bilayer as the memory layer, the thermal stability is considered to be improved largely due to large perpendicular anisotropy and coercivity of the TbFe layer. The TbFe and GdFeCo can be exchange coupled with each other. When the injection current heats the TbFe layer up to Curie temperature, the magnetization of the constituent GdFeCo layer can also be switched by the polarized current simultaneously. So, the information stored in GdFeCo/TbFe bilayer can achieve two advantages for the STT switching devices: (1) enhanced thermal stability at room temperature; (2) writing the bit by polarized injection current.

To investigate the validity of GdFeCo as memory layer for STT switching, GMR structure with 10nm-thick GdFeCo layer will be fabricated. The STT switching will be confirmed by measuring resistance-current curve obtained by applying pulse current. It is well believed that the magnetic bias field can modulate  $K_{\text{eff}}$  that induce the lower  $J_c$  to switch the magnetic bits. In this sense, the magnetic bias field dependence of switching current will also be investigated. Pulse width dependence of  $J_c$  will be measured to discuss the relationship between  $K_{\text{eff}}$  of un-patterned film and  $\Delta$  of patterned junctions. Besides the magnetic bias field and pulse width dependences, the temperature is another critical factor that can affect the  $K_{\text{eff}}$  and  $J_c$ , so the temperature dependences of  $K_{\text{eff}}$  and  $J_c$  are important issues for STT switching of GdFeCo and the thermal effect on the STT switching.

Single GdFeCo memory layer is found to exhibit low  $K_{eff}$  which does not guarantee the thermal stability in the small bit size for high density STT-RAM. One solution is to introduce exchange-coupled bilayer structure consisting of TbFe and GdFeCo layers. It is noted that TbFe exhibits high  $K_{eff}$  and large slope of  $K_{eff}$ -*T* curve which is beneficial for the thermally assisted STT-RAM. The second part of this thesis is therefore about the STT switching of TbFe/GdFeCo bilayer. There are some similar characters compared to that of the single GdFeCo layer, such as magnetic bias field dependence of  $J_c$ , temperature dependence of  $K_{eff}$  and  $J_c$ . All of these measurements are necessary for the systematic study on the thermally assisted STT switching.

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# **Chapter 2 Experimental Methods**

#### **2.1. Samples preparation**

Magnetron sputtering is a well exploited physical vapor deposition method for thin film fabrication. The magnetron sputtering method has several advantages over the conventional deposition methods such as evaporation, in which one important aspect is that even the materials with high melting point can be easily deposited. Another advantage is that the composition of the deposited films has almost the same content with that of the target. Additionally, the films deposited by the sputtering method usually have excellent adhesion with substrates compared to evaporated films [1].

Based on the advantages, all of my samples were fabricated by RF magnetron sputtering systems. The basic process of magnetron sputtering deposition is described as follows. The material of the target is stroked with energetic ions, typically inert gas ions such as Argon  $(Ar^+)$ . The collisions of these ions with the atoms on the target expel or sputter the target atoms into the chamber and then the sputtered atoms are deposited on the substrate, leading to the formation of the complete layer. The quality of the deposited films is dependent on several parameters, such as sputtering gas pressure, substrate temperature, distance between the target and substrate, etc. In sputtering, electrons in the plasma are confined by the strong magnetic field close to the target surface, and move in helical paths in the magnetic field. This effectively produces argon ions accelerated to the target surface, which results in a higher deposition rate even at low Ar pressure. The most sputtered atoms from the target are neutral, so that they are not influenced by the electric and magnetic fields near the target. Insulating targets can be sputtered by the use of RF sputtering power source where the sign of bias voltage is varied at commonly 13.56 MHz. RF sputtering shows some advantages in depositing insulating oxide films but with extra circuits for impedance matching. In addition, stray magnetic fields leaking out of ferromagnetic targets disturb normal magnetic field, which may cause some inhomogeneous deposition of films. To solve this kind of problem, strong permanent magnets are

desirable.

#### 2.1.1. Deposition of samples with GMR structure

#### 2.1.1.1. 8-sources magnetron-sputtering system

The magnetron sputtering system in my experiments is based on the basic principle of magnetron sputtering mentioned above, and some components were customized to meet specific requirements. The schematic illustration of the system is shown in Fig. 2.1.



Fig. 2.1 Schematic illustration of the 8-sources magnetron-sputtering system.

All of the samples were deposited on thermally oxidized silicon wafers. Argon gas of purity (99.9999%) was used as the sputtering gas and the working pressure was constantly kept to be around 0.4 Pa. Before inserting Si substrates into the deposition chamber, they are cleaned using isopropyl alcohol (IPA) in ultrasonic bath for 5 minutes. Usually, the base pressure of the chamber is maintained in ultra-low level of 10<sup>-7</sup> Pa to ensure the quality of the deposited films, and thus the system has a sample exchange chamber for inserting and removing the sample without breaking the vacuum of the deposition chamber. It is also a pre-requisite to etching the substrates

using moderate energetic  $Ar^+$  ions to remove contamination on the substrate surface before the deposition. The typical parameters of deposition for my experiments were listed in Table 2.1.

Base pressure (Pa)						
1.0×10 <sup>-7</sup> Pa						
Ar <sup>+</sup> ions etching						
Accelerating voltag	ge Ar Pressure	I	Etching time			
(kV)	(Pa)	ion current (µA)	(min)			
1	1.2 × 10 <sup>-4</sup> Pa	40	5			
Sputtering conditions						
Targets	$\mathbf{D}_{1}$	Ar pressure	Deposition rate			
	Power (w)	(Pa)	(nm / s)			
Та	80		0.0411			
Cu	60		0.0432			
Al	120		0.0401			
Gd	20~60		0.0194~0.0623			
Fe <sub>90</sub> Co <sub>10</sub>	120	0.4	0.0395			
Tb	20~60	0.4	0.0181~0.0597			
Fe	120		0.0437			
Co40Fe40B20	120		0.0384			
Со	100		0.0376			
Pd	50		0.0382			

Table 2.1 Parameters of the deposition for 8 sources sputtering system
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# 2.1.1.2. 5-sources magnetron-sputtering system

A 5-sources magnetron-sputtering system was used to deposit the metallic aluminum electrodes and the insulating SiN layers for micro-fabrication of the current perpendicular to the plane structure. Unlike the Ar ion etching for the substrate cleaning in the 8-sources sputtering system, a process called reverse-sputtering, where RF voltage was applied to the substrate, was carried out to clean the substrate surface before the deposition. The parameters of the reverse sputtering were at the pressure of 2 Pa, power of 100 W and for the sputtering time of 60 sec. The base pressure of the 5-sources sputtering system is usually around  $8.0 \times 10^{-6}$  Pa. The typical parameters in the 5-sources sputtering system are shown in Table 2.2. The schematic structure of the 5-sources magnetron sputtering system used for this experiments is shown in Fig. 2.2.

Base pressure (Pa)						
8.0 × 10 <sup>-6</sup> Pa						
Ar <sup>+</sup> ions reverse etching						
Power (W)		Ar Pressure (Pa)	Time (sec)			
100		2.0 Pa	60			
Sputtering conditions						
Targets	Power (W)	Ar pressure (Pa)	Deposition rate ( nm / s)			
Al	150	0.4	0.0741			
Al <sub>2</sub> O <sub>3</sub>	150	0.1	0.0264			
SiN	150		0.0281			

Table 2.2 Parameters of the deposition for 5 sources sputtering system



Fig. 2.2 Schematic illustration of the 5-source magnetron-sputtering system.

# 2.1.2. Micro-fabrication of current perpendicular to the plane (CPP) structure

# 2.1.2.1. Photo-lithography (PL)

Photo-lithography is a process used in micro-fabrication to selectively remove parts of a thin film or the bulk of a substrate [2]. It uses light to transfer a geometric pattern from a photomask to a photo-sensitive chemical, photoresist, on the substrate. The photoresist patterns are transferred to the sample either by deposition of other materials followed by the lift-off process or by  $Ar^+$  ion etching to remove the materials. The typical steps involved in the photo-lithographic process are wafer cleaning; photoresist coating; soft baking; mask alignment; exposure and development; and hard-baking. The general steps of photo-lithography are shown in Fig. 2.3.



Fig. 2.3 Schematic diagram of typical process of photo-lithography.

The photo-lithography process must be performed in a clean room with a yellow light, since dust particles on the sample can result in defects of the micro-fabrication and since the photoresist is generally sensitive to blue light. Each step of photolithography process in this experiment is described below:

Step 1: Substrate cleaning and dehydration

Before photoresist coating, the Si substrates were rinsed with isopropyl alcohol (IPA) in an ultrasonic bath for 5 minutes to remove contaminants, then the substrates were blew with dry  $N_2$  to eliminate any unfavorable adsorbates.

Step 2: photoresist coating

The photoresist (Rohm & Haas MICROPOSIT Photo Resist S1813G) was coated by a spin-coater (MIKASA spin-coater 1H-D7), which produces a uniform, adherent, and defect-free photoresist coating on the sample. The sample was fixed by vacuum chuck connected to the spin-coater. The spin-coating process consists of two distinct steps. The first one is to fling off the excess resist; the second step is designed to make the resist smoothly coated on the surface of the sample. The program set in the spin-coater is as follow:

1000 rpm  $\times$  10 sec + 5000 rpm  $\times$  35 sec (resist thickness : 1.0  $\sim$  1.5  $\mu m)$  Step 3: Soft baking

The coated photoresist was pre-baked at 110°C for 20 min in the oven

(YAMATO Constant Temperature Oven DKM300) to drive off solvents in the photoresist coating. Soft-baking plays a very critical role in photo-lithography. The photoresist coatings become photosensitive only after soft-baking. Over-baking will degrade the photo-sensitivity of resists by either reducing the developer solubility or actually destroying a portion of the sensitizer. Under-baking will prevent light from reaching the sensitizer. Positive resists are incompletely exposed if considerable solvent remains in the coating.

# Step 4: Exposure

Once the mask has been accurately aligned with the pattern on the wafer's surface (MIKASA, M-1S type mask alignment tool), the photoresist S1813G is exposed through the pattern on the mask with a high intensity ultraviolet light emitted from ultra-high pressure mercury-vapor lamp (MIKASA M-10) with power of 250 W. The mask patterns (50~70nm Cr) for bottom electrodes, top electrodes, and insulating layers to electrically isolate top and bottom electrodes are shown in Fig. 2.4.



Fig. 2.4 photo-mask for (a)bottom electrode, (b)intermediate insulating layer, and (c) top electrode.

Step 5: Post exposure baking

Post exposure baking was done to reduce an effect of standing waves in the exposed resist, and the condition of the baking in this experiment was 110 °C for 10 min in the oven. The post exposure baking is effective to form rectangle resist patterns after the development described below.

### Step 6: Development and striping

After the post exposure baking process, the sample was developed in Rohm & Haas MFCD-26 DEVELOPER to remove the exposed regions in the resist. The sample was dipped for 15 sec and agitated gently for 45 sec in the developer, and followed by rinsing in pure water about 1 minute. After the etching by Ar<sup>+</sup> ion bombardment or deposition of other materials, the remaining resist was removed by using the Rohm & Haas MICROPOSIT REMOVER 1165 heated up to 80°C in an ultrasonic bath for 20 minutes and followed by IPA cleaning for 5 minutes.

### 2.1.2.2. Electron beam lithography

Electron beam lithography (EB lithography) is the lithographic process using a focused electron beam to modify the chemical property of the coated resist, which can selectively removes either exposed or non-exposed regions of the resist [3]. The purpose, as for photolithography, is to create very small structures in the resist that can subsequently be transferred to the sample. The primary advantage of electron beam lithography is that it is one of the ways to beat the diffraction limit of photolithography and fabricate nano-structure. The key limitation of EB lithography is that this process is a time-inefficient process. The each step of micro-fabrication using EB lithography is described below:

Step 1: substrate cleaning

To clean and dry the substrates, the following steps are performed.

- Acetone cleaning: 5min (in ultrasonic bath, at room temperature)
- IPA cleaning: 5min (in ultrasonic bath, at room temperature)
- N<sub>2</sub> blowing

- drying (hotplate, 2min)

Step 2: resist coating

The resist used for EB lithography is Nihon ZEON ZEP520A. The coating process is very similar to that in photolithography. The program was varied slightly

 $500 \text{ rpm} \times 5 \text{ sec} + 3000 \text{ rpm} \times 30 \text{ sec}$ 

Step 3: pre-baking

To drive off the solvent in the resist, pre-baking is needed, which is also similar to that in the photolithography but at the temperature of 150°C for 30min.

### Step 4: exposure

This step is completed by JEOL JBX-6000FS EB lithography system which is illustrated in Fig. 2.5. The JBX-6000FS generates a high density electron beam using a thermal field emission gun with a ZrO/W emitter at 50 kV voltage. It also incorporates two different objective lenses for fine and ultra fine pattern exposures with a beam diameter range of 200 nm to 5 nm. Typical beam currents are set to be 2 nA and 100 pA, where the former is used to expose the larger area and the latter is responsible for exposure of small region. The typical SEM image of exposed nano-structure is shown in Fig. 2.6.



Fig. 2.5 Schematic diagram of the JEOL JBX-6000FS EBL system.



Fig. 2.6 SEM image of the EB lithographed resist for GMR cell with a size of  $120 \times 180 \text{ nm}^2$ .

Step 5: developing

The differences in developing in EB lithography from photolithography is the developer chemical, xylene. For the precise control of the patterned shape, the temperature control during the developing is crucial, and in this experiment the xylene was kept at 25°C by using temperature thermostat. The following is the process for developing:

- xylene, the sample was gently agitated for 45 sec at 25°C

- IPA rinsing for 30sec

- N<sub>2</sub> blowing, for IPA removal and drying

Step 6: resist removal (striping)

The remover used in EB lithography is Nihon ZEON ZDMAC and the steps is as follows:

- ZDMAC stripping (1st) : for 20 min (in ultrasonic bath, at 70 ~80°C)

- ZDMAC stripping (2nd) : for 20 min (in ultrasonic bath, at 70 ~80°C)

- IPA cleaning: for 10 min (in ultrasonic bath, at room temperature)

- N2 blowing, for IPA removal and drying

### 2.1.2.3. Electron Cyclotron Resonance (ECR) Ar<sup>+</sup> plasma etching

ECR  $Ar^+$  plasma etching was used to transfer resist pattern profile to the sample. Ar<sup>+</sup> ion etching is a dry etching technique in which  $Ar^+$  ions generated in plasma were accelerated and incident on the sample to remove the regions without resist coverage after the development. The apparatus exploited in this etching process is schematically illustrated in Fig. 2.7 and the parameters specified for this experiments were also shown in Table 2.3.



Fig. 2.7. Schematic diagram of ECR system.

Table 2.3 Experimental parameters for ECR etching

Base pressure(Pa)	$2.0 imes10^{-6}$
Work pressure (Pa)	1.2~1.5 × 10 <sup>-2</sup>
Forward/reflect current (mA)	0.5/0.02
Accelerating/forward voltage (V)	600/-100

The ECR ion source exploits the electron cyclotron resonance to induce ionized plasma [4]. Microwave is guided into a volume containing a low pressure gas. The alternating electric field of the microwave is synchronous with the gyration period of the free electrons under a magnetic field applied inside the volume, which is called the electron cyclotron resonance.

Microwave at the frequency of 2.45 GHz was injected through a co-axial cable into the plasma source and a resonant magnetic field (about 875 G) was applied by a cylindrical shaped permanent magnet. The plasma source produces the ion flux is extracted into the etching chamber where the sample is mounted. Due to the low operating pressure for ECR plasmas (about  $1.2 \times 10^{-2}$  Pa in this work), the anisotropic etching is featured in this technique. The etching process can be monitored by the analysis of emitted ions from the sample surface during the etching by a secondary ion mass spectrometry (SIMS; PFEIFFER EPD 400) [4], which is widely used to detect the end point of the ion etching of the sample with a layered structure.

#### 2.2. Measurement methods

#### 2.2.1. Alternating gradient-field magnetometer (AGM)

Alternating gradient-field magnetometer (AGM) is a typical system to measure magnetic moments of thin films with high sensitivity in the order of  $10^{-8}$  emu [5]. The underlying principle of the AGM is that the magnetic field with alternating gradient exerts an alternating force on the magnetic sample and the force is proportional to the amplitude of the field gradient and the magnetic moment of the sample. The alternating force applied on the sample induces oscillation of the piezoelectric element connected to the sample, and then the magnetic moment of the sample can be determined by measuring the oscillated voltage signal. The diagram of the AGM set-up is shown in Fig. 2.9. The amplitude of this voltage signal is first calibrated by measuring the standard sample, Ni foil with 528 µemu. The AGM used in this experiment is Princeton AGM 2900 and the maximum field of the AGM is 22 kOe and the measurement frequency is from 0.1 kHz to 1kHz.



Fig. 2.9 Schematic diagram of alternating gradient-field magnetometer.

#### 2.2.2. Torque magnetometer

A magnetic element will produce a torque when it is placed in a uniform magnetic field H, The torque, denoted by L, can be expressed as

$$L = M \times H = MH \sin \varphi$$

 $\varphi$  is the angle between the magnetic field and the direction of magnetization of the sample [6].

The torque is proportional to the derivative of the magnetic energy with respect to the angle of rotation:

$$L(\varphi) = -\frac{\partial E(\varphi)}{\partial \varphi}$$
(2.1)

The magnetic energy of the magnetic element with an uniaxial magnetic anisotropy is expressed as the sum of the effective anisotropy and the Zeeman terms:

$$E = K_{eff} \sin^2 \theta - MH \cos(\varphi - \theta)$$
(2.2)

where  $K_{\text{eff}}$  is effective anisotropy constant, M is saturated magnetization, H is the applied magnetic field,  $\theta$ ,  $\varphi$  are the angles of magnetization and H with respect to the easy-axis, respectively. When the applied field H is high enough and  $\theta$  is approximated to be  $\varphi$ , the torque  $L(\varphi)$  can be easily obtained, and expressed as the following term:

$$L(\varphi) = -K_{eff} \sin 2\varphi \tag{2.3}$$

The above equation can be simply graphically illustrated in Fig. 2.10.



Fig. 2.10 Typical torque curve of magnetic materials with an uniaxial anisotropy.

In the torque measurement system, the sample together with the holder sticking to a feedback coil which is suspended using a thin gold wire as illustrated in Fig. 2.11. The torques acting on the samples or on the coil can be compensated with the torque produced by a current flow through this coil. The rotation of the sample can be detected using a sub-system consisting of a small mirror, a light source and a photo-potentiometer. It is noted that the current through the coil is proportional to the torque exerted on the sample [6].



Fig. 2.11 Schematic drawing of torque measurement system.

# 2.2.3. Magneto-optical Kerr spectrometer

The magneto-optical Kerr effect (MOKE) describes the changes to light

reflected from a magnetized material which can change both in polarization and reflected intensity [7]. Physically, MOKE results from the off-diagonal component of the dielectric tensor  $\varepsilon$  which induces the variation of speed of light depending on the orientation of magnetization vector of the magnetic material.



Fig. 2.12 Illustration of the polar Kerr effect.

In the polar Kerr effect, the magnetization vector is parallel the plane of incidence of the light and perpendicular to the reflecting surface. The schematic polar setup shown in Fig. 2.12 involves light reflected at an angle from the reflecting surface other than normal to it. In this manner, linearly polarized light incident on the surface becomes elliptically polarized, with the change in polarization directly proportional to the component of magnetization that is perpendicular to the reflecting surface and parallel to the plane of incidence.

A typical setup for magneto-optical Kerr spectrometer is shown schematically in Fig. 2.13. The light emitted from Xe lamp can be transformed to linear polarized light through a polarizing prism after passing a monochromator. The incident angle of the polarized light onto the surface of samples is almost 5°. After passing the Faraday cell, the reflected light goes through an analyzer and is detected by a photo-detector. The Faraday cell consists of paramagnetic glass tube or SiO<sub>2</sub> tube which was wounded by Cu wire to flow DC and AC currents in it. The magnetic field generated by the DC current can induce shift of the polarization plane and compensate the Kerr rotation angle  $\theta_{\rm K}$  of the reflected light. The DC current necessary to minimize the light detector signal is proportional to the Kerr rotation  $\theta_{K}$ , and thus the  $\theta_{K}$  is measured by measuring the DC current flowing the cell. Besides, an AC current with frequency of 510 Hz is superimposed on the DC current to modulate the polarization of the light through the Faraday cell. The shift of the polarization is detected as the 510 Hz output signal, which is filtered by a lock-in amplifier. By sweeping the applied magnetic field and recording the DC feedback current to minimize the 510 Hz signal, Kerr loop of the sample can be easily obtained.



Fig. 2.13 Schematic illustration of the setup of of a Kerr spectrometer.

### 2.2.4. Scanning electron microscope (SEM)

A scanning electron microscope (SEM) is a type of electron microscope that produces images of a sample surface by scanning with a focused beam of electrons. The electrons interact with atoms in the sample, producing various signals which are useful to get information about the topography of the sample surface and composition of the film.

The principle of the topography imaging by the SEM is to collect secondary electrons emitted from atoms excited by the primarily electron beam [8]. The intensity of secondary electrons is dependent on the angle between the surface of the sample and the primarily beam. It is easy to distinguish the flat and tilted surface in

the microscopy. By scanning the sample with the primary electron beam and detecting the amount of secondary electrons, an image of the sample surface can be simply displayed. The schematic set-up of the SEM (Hitachi S-5200) used in this experiment is shown in Fig. 2.14.



Fig. 2.14 Schematic set-up of SEM

#### 2.2.5. Magneto-resistance measurement

Generally, the electrical impedance of GMR multilayer structures was measured using 4-point probe method as shown in Fig. 2.15, which is an accurate measurement to evaluate the sample resistance. The key advantage of 4-point probe method is that the separate configuration of current carrying and voltage sensing electrodes can eliminate the resistances of contact and metallic wiring.



Fig. 2.15 Schematic diagram of the 4-point probe method.

For I-V measurements of the junctions, an electrical current was applied through two current terminals, I- and  $I_+$ , and the resulting voltage across the junction was measured by two voltage ends, V- and  $V_+$ . The current was provided by YOKOGAWA 7651 programmable current source and the voltage was sensed using Keithley 2182 nano-voltmeter. The *I-V* curve was normally obtained by sweeping the current from a negative value to a positive one. The magnetic field in this system was controlled by a DC power current source. During the magneto-resistance measurement, the samples were placed between two magnetic coils and the resistance of the junction was measured by sweeping the magnetic field. In the measurement of R-I curves, the positive current direction is defined as that current flows from top aluminum electrode to bottom electrodes. The samples are usually attached to the circuit board by double sided tapes, then they are placed between the gap of electromagnet. The four terminals are simply bonded using thin metallic wires by ultrasonic wire bonder, and measured by sending current in the current ends, Iand  $I_{+}$ . Usually, 1 mA current was applied for the MR measurement, while 0.5 mA sense current were adopted for the spin injection switching measurement. Such a relatively low sense current was applied after the application of the pulse current with a pulse width of 0.1 sec, and it can avoid the unwanted switching of the cell by the sense current itself. For the experiment of the temperature dependence of switching current density, the samples were in touch with a copper plate placed on a

specified heater. To eliminate the unstable air flow near the surface of the samples, a heat-insulating cover box was used to cover the sample. Specially, in the experiment of pulse width dependence of current density, a pulse generator was connected to the wires which bonded to the current ends. The pulse current generated by the pulse generator was applied ahead of the sense current in each measurement period. A in-house made CMOS switch circuit was used to select the input current between DC sense current and pulse current.

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# **Chapter 3 Spin injection switching of GdFeCo layer**

In this chapter, the spin injection switching of a perpendicularly magnetized GdFeCo single layer will be introduced and discussed. The motivation of selecting GdFeCo as memory layer in the spin injection switching is based on the following considerations:

First, the comparatively low damping constant  $\alpha$  of GdFeCo layer holds the promise of low critical current.

Second, the perpendicular magnetization induces relative low effective anisotropy constant in the GdFeCo layer that ensures low value of critical current.

Final, the combination of high resistivity (amorphous state) and relative low Curie temperature compared to conventional FeCo layer is considered to be suitable for the thermally assisted spin injection switching scheme.

In addition, it is difficulty to demonstrate thermally assisted spin injection switching effect in  $AIO_x$  based MTJs in which its high resistance limits the increase of current density provided by power source. Alternatively, GMR structure with Cu spacer is potentially expected to be competent for the thermally assisted spin injection switching because of low resistance of the junction therefore the large current supplied by the power source.

Accordingly, to verify the Gd compositional dependence of various magnetic properties and STT switching, giant magneto resistance (GMR) devices having amorphous GdFeCo memory layers with various Gd compositions were prepared by magnetron sputtering and subsequent micro-fabrication processes. Their spin transfer torque (STT) switching was investigated by applying a current pulse to the GMR devices. In addition, pulse width and temperature dependences of the critical current density for the STT switching were also measured.

### **3.1. Sample preparation**

The giant magneto-resistance (GMR) film stack is Si / Ta (10) / CuAl (150) / [Pd (1.6) / Co (0.4)]<sub>6</sub> / Co<sub>40</sub>Fe<sub>40</sub>B<sub>20</sub> (0.5) / Cu (3) / Gd<sub>x</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>100-x</sub> (10) / Cu (5) /

Ta (5) (thickness is in nanometer). The samples were deposited using an 8-sources magnetron sputtering system with a base pressure around  $1 \times 10^{-7}$  Pa. The multilayer [Co/Pd]<sub>6</sub> acts as a reference layer and the perpendicular easy axis was achieved by varying the thicknesses of both Co and Pd layers. The Gd<sub>x</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>100-x</sub> layer was deposited by a co-sputtering of Gd and  $Fe_{90}Co_{10}$  targets, and the Gd composition x was controlled by varying sputtering power of the Gd target. The magnetic properties of the as-deposited films were characterized by an alternating gradient magnetometer (AGM) and a torque magnetometer. GMR devices with various dimensional elliptical patterns ranging from 120×180 nm<sup>2</sup> to 300×450 nm<sup>2</sup> were micro-fabricated by electron-beam (EB) lithography and subsequent Ar<sup>+</sup> etching from an electron cyclotron resonance (ECR) plasma source. After the etching, a SiN insulation layer was deposited by a 5-sources magnetron sputtering system. The use of SiN as an insulation layer is to avoid oxidation of the Gd atoms in the memory layer. The lift-off process was carried out after the SiN deposition using EB resist remover (ZDMAC) at about 80°C to open the window and connect the GMR element to the top electrode. Then, a top aluminum electrode was formed by photolithography. The magneto-resistive (MR) effect of the GMR devices was measured by four-point probe method. The STT switching property of these devices was characterized by the resistance measurement after applying current pulses. Typical structure of the sample investigated is illustrated in Fig. 3.1.



Fig. 3.1 typical structure of the sample before deposition of top aluminum electrode.

### 3.2. Magnetic properties of GdFeCo layer

### 3.2.1. Saturation magnetization (M<sub>s</sub>)

*M-H* loops in normal direction of TM-rich ferrimagnetic  $Gd_{21}(Fe_{90}Co_{10})_{79}$  layers with various thickness were shown in Fig. 3.2.



Fig. 3.2 *M*-*H* loops in normal direction of single  $Gd_{21}(Fe_{90}Co_{10})_{79}$  layer with thickness of (a) 15nm; (b) 10nm; (c) 5nm and (d) 5nm GdFeCo layer coupled with 0.5nm  $Co_{40}Fe_{40}B_{20}$  layer.

It is noted that the single GdFeCo layer is sandwiched by two 10nm-thick tantalum (Ta). The shape of the loops varies with the thickness of GdFeCo layer. The 15nm and 10nm-thick GdFeCo layers show similar shape due to their similarity to the sufficient thick GdFeCo films in terms of magnetic properties. While, in the GdFeCo layer with 5nm thickness, the contribution of interface on the variation on magnetic properties tends to be dominant and the shape of *M*-*H* loop varies. For the requirement of spin injection switching, thinner layer is believed to be benefit for the reduction of switching current density, which is a key issue of this study. Meanwhile, the thickness of the layer should be sufficient to keep magnetization of the GdFeCo layer in perpendicular direction. In addition, the material of adjacent layers on both

sides of the GdFeCo layer is found to be crucial for the magnetic properties [1]. To verify and optimize the configuration of the memory layer consisting of GdFeCo, the adjacent material of Ta in Fig. 3.2 is switched to Cu in the GMR structure of Ta /Cu /GdFeCo / Cu / [Co/Pd] /Ta /Si substrate, and the *M-H* loops of this GMR structure with GdFeCo memory layer, having various Gd content, were measured. The major loops taken by sweeping the field of  $\pm 15$  kOe and the minor loops by sweeping the field of  $\pm 1$  kOe are shown in Fig. 3.3.

Because of the antiparallel configuration of sub-lattices of TM and RE atoms in the ferrimagnetic GdFeCo, it can be easily concluded that a compositional compensation point exists in the range of 18~28at.% of Gd content, that can be also confirmed in Fig. 3.3 (a)-(j). For Gd content below that point, the magnetization of constituent TM atoms dominates the net-magnetization while RE atoms dominate for the Gd content larger than that point. The net-magnetization almost vanishes in the vicinity of that point, which is around 23~24at.% as shown in Figs. (f) and (g). Interestingly, the direction of the coupling between the magnetizations of GdFeCo and Co/Pd depends on the Gd content in the memory layer. The negative loop shift in the minor loops was observed for TM-rich GdFeCo (See Figs. (a) - (e)) samples, while positive for RE-rich samples. This is due to the parallel exchange coupling between Co moment in the Co/Pd and TM moment in the GdFeCo through the Cu spacer. This suggests that the direction of the loop shift is important information to determine which moment dominates the net moment in the GdFeCo layer, which is difficult to measure in the micro-fabricated GMR samples.



(a) Gd<sub>18</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>82</sub>



(e) Gd<sub>22</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78</sub>



(h) Gd<sub>25</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>75</sub>



(j) Gd<sub>28</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>72</sub>

Fig. 3.3 Variation of major (right side) and minor (left side) *M-H* loops with Gd content from 18at.% ~ 28at.% in the GMR films. The minor loops was taken after the saturation of Co/Pd magnetization by applying +15 kOe.

The temperature is another crucial parameter that can affect the magnetic properties of GdFeCo layer. As described in Sec. 2.1.2, several heat treatments are required for the micro-fabrication of CPP-GMR structure. To verify the effect of the heat cycle on magnetic properties of GdFeCo layer, the magnetic properties of typical GdFeCo layer after the heat treatment similar to the baking during the micro-fabrication were investigated.

As shown in Fig. 3.4, the minor loops of the GMR film with TM-rich  $Gd_{21}(Fe_{90}Co_{10})_{79}$  memory layer were measured after the heat treatment at various temperature in the oven for 30 min (the oven was not evacuated to low pressure), which is a standard process in the micro-fabrication. The measurements were done at

room temperature. There was no significant difference in the minor loops between as-prepared and heat cycled samples up to 160°C. However, above 160°C the magnetic properties, such as coercivity  $H_c$  and saturated magnetization  $M_s$ , are significantly changed.

The RE-rich  $Gd_{26}(Fe_{90}Co_{10})_{74}$  layer is believed to behave differently on the magnetic properties. For comparison, the minor loops of typical RE-rich  $Gd_{26}(Fe_{90}Co_{10})_{74}$  layer after the heat cycle at various temperature in the oven was investigated (See Fig. 3.5).





Fig. 3.4 (a)-(e) Minor loops of GMR film with  $Gd_{21}(Fe_{90}Co_{10})_{79}$  memory layer after the heat cycle at various temperature in oven for 30 min. The loops were taken at room temperature and after the saturation of Co/Pd magnetization by applying +15 kOe.

It can be seen that the RE-rich layer definitely exhibits unique magnetic performance compared to that of TM-rich layer. The maximum temperature above which the magnetic properties will be seriously altered is about 150°C which is smaller than that of the TM-rich layer. The possible reason may be due to the more Gd content in the RE-rich layer and the Gd's weaker resistance to oxidization. From these results, the maximum temperature for the micro-fabrication is chosen to be around 150°C for my CPP-GMR micro-fabrication process. In this case, the variation of magnetic properties in all the samples from RE-rich to TM-rich GdFeCo is considered to be avoided.



Fig. 3.5 (a)-(d) Minor loops of GMR film with  $Gd_{26}(Fe_{90}Co_{10})_{74}$  memory layer after the heat cycle at various temperature in oven for 30 min. The loops were taken at room temperature and after the saturation of Co/Pd magnetization by applying +15 kOe.

The major hysteresis loops for the GMR films with memory layers of  $Gd_{21}(Fe_{90}Co_{10})_{79}$  and  $Gd_{26}(Fe_{90}Co_{10})_{74}$  taken applying a magnetic field in the film normal direction are shown in Fig. 3.6, the coercivity of the Co/Pd reference layer is around 2 kOe for the both films. The squareness of the loops indicates that the samples have a large PMA. The switching fields of the GdFeCo layers depend both on the Gd content *x* and the magnetization direction of the Co/Pd layers, i.e., parallel coupling of the magnetizations between TM-rich  $Gd_{21}(Fe_{90}Co_{10})_{79}$  and Co/Pd while anti-parallel between RE-rich  $Gd_{26}(Fe_{90}Co_{10})_{74}$  and Co/Pd. As discussed above, this can be understood by the exchange coupling between Fe<sub>90</sub>Co<sub>10</sub> and Co/Pd moments through the Cu spacer.



Fig. 3.6 Hysteresis loops of GMR films with Gd<sub>21.4</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78.6</sub> and Gd<sub>26.5</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>73.5</sub> memory layers.

### **3.2.2. Effective anisotropy constant** (*K*<sub>eff</sub>)

Effective anisotropy constant ( $K_{eff}$ ) is another key parameter to determine the spin injection switching current density.  $K_{eff}$  should be adjusted to achieve low switching current density and sufficiently high thermal stability simultaneously. There are several parameters to tailor the value of  $K_{eff}$ , among which composition and temperature are two key factors to modulate the  $K_{eff}$  for the RE-TM memory layer. Fig. 3.7 (a)-(d) shows that the Gd compositional dependence of  $K_{eff}$ . Especially, Fig. 3.7 (e) shows *M-H* loops for the 10 nm-thick single  $Gd_{22}(Fe_{90}Co_{10})_{78}$  layer. The anisotropy constant  $K_{eff}$  was estimated to be  $6 \times 10^4$  erg/cc from the area surrounded by the perpendicular and in-plane loops. The perpendicular anisotropy constant  $K_u$  can be obtained by Eq. (3. 1)

$$K_{u} = K_{eff} + 2\pi M_{s}^{2}$$
(3. 1)

and it became to be  $4.5 \times 10^5$  erg/cc.





Fig. 3.7 Gd compositional dependence of  $K_{\text{eff}}$  of single  $\text{Gd}_x(\text{Fe}_{90}\text{Co}_{10})_{100-x}$  (10 nm) layers in (a)-(d); *M*-*H* loop of 10-nm thick single  $\text{Gd}_{22.0}(\text{Fe}_{90}\text{Co}_{10})_{78.0}$  film in (e).

It can be concluded that the  $K_{\text{eff}}$  of  $\text{Gd}_x(\text{Fe}_{90}\text{Co}_{10})_{100-x}$  layer increases with the Gd content in the range of 21~25at.%. This tendency can be explained as follows: the uniaxial anisotropy constant  $K_u$  is almost constant in this range, while the magnetization decreases with Gd content, therefore, the overall  $K_{\text{eff}}$  increases with the Gd content. In the following part, the temperature dependence of  $K_{\text{eff}}$  of GdFeCo layer is discussed. The effective anisotropy constant  $K_{\text{eff}}$  for 10 nm-thick  $\text{Gd}_{23}(\text{Fe}_{90}\text{Co}_{10})_{77}$  single layer at various temperatures was estimated from the torque measurement and was shown in Fig. 3.8. The  $K_{\text{eff}}$  seems to be constant in the relatively low temperature range and shows obvious decrease when the temperature exceeds about 80°C. However, the saturation of  $K_{\text{eff}}$  at < 50 °C is considered to be due to the limited external field of 15 kOe in the torque measurement. Thus we tried 45° method [4] to estimate the actual  $K_{\text{eff}}$  at temperatures < 50°C and the results are shown in the inset. From the 45° method, we confirmed roughly 5% decrease of  $K_{\text{eff}}$  when the temperature increased from 30°C to 50°C.



Fig. 3.8 Temperature dependence of effective anisotropy constant  $K_{\text{eff}}$  of the  $Gd_{23}(Fe_{90}Co_{10})_{77}$  single layer obtained by torque curves. Inset shows  $K_{\text{eff}}$  data taken by  $45^{\circ}$  method.

The effective anisotropy constant of the memory layer shown in Fig. 3.8 is more than  $5 \times 10^5$  erg/cc even at temperature of 90°C and this temperature is believed to emerge in the STT-MRAM cells during the writing process [5]. The large value of  $K_{\text{eff}}$ suggests that the GdFeCo memory layer may has enough thermal stability even in the memory cell micro-fabricated down to 30 nm × 30 nm ×10 nm size. As for the temperature dependence of the  $K_{\text{eff}}$ , one can clearly see the decrease of the  $K_{\text{eff}}$  with elevating temperature. We believe this decrease of  $K_{\text{eff}}$  is benefit for the reduction of critical current density  $J_c$  of current injection switching which will be discussed later.

#### 3.2.3. Magneto-resistance (MR) properties

Reasonable magneto-resistance (MR) is critical criterion to demonstrate the STT-RAM. MR ratio can reveal the qualities of several intrinsic magnetic properties, such as spin polarization  $\eta$  of the constituent magnetic layers in the GMR and/or MTJs, which is crucial for the reduction of switching current density  $J_c$  according to equation (1.1). In Fig. 3.9, the Gd compositional variation of MR curves of the non-micro-fabricated CPP-GMR samples with  $Gd_x(Fe_{90}Co_{10})_{100-x}$  memory layer was

shown. For reference, the corresponding M-H loops (note that the vertical axis is magnetic moment per unit area) of the GMR films were also shown.



(a) Gd<sub>18</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>82</sub> 10nm GMR structure



(b) Gd<sub>19</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>81</sub> 10nm GMR structure



(c) Gd<sub>20</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>80</sub> 10nm GMR structure


(d) Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> 10nm GMR structure



(e) Gd<sub>22</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78</sub> 10nm GMR structure



(f) Gd<sub>23</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>77</sub> 10nm GMR structure



(g) Gd<sub>24</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>76</sub> 10nm GMR structure



(h) Gd<sub>25</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>75</sub> 10nm GMR structure



(i) Gd<sub>27</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>73</sub> 10nm GMR structure

Fig. 3.9 Gd compositional variation of the MR curve for the CPP-GMR structure with  $Gd_x(Fe_{90}Co_{10})_{100-x}$  memory layer. For reference, corresponding *M*-*H* loops (note that the vertical axis is magnetic moment per unit area) of the GMR films were also shown.

In MR loops, there exists a shift of the average resistance when magnetic field sweep direction is changed. This is due to the electromotive force induced in the sensing wires during the field sweeping, and independent of the switching of the magnetizations of memory and reference layers. The MR change due to the switching should be appeared near the field where the switching was observed in the hysteresis loop. From Fig. 3.9, the MR loops due to the magnetization switching show different features in the TM-rich and RE-rich samples. The resistance of the antiparallel state (AP) state for the TM-rich samples is usually higher than that of parallel state (P), while for the RE-rich samples, the AP state becomes low resistance and P state exhibits high resistance. It is noted that the MR ratio almost vanishes near the Gd compensation point and this is possibly due to the increase of the coercivity of GdFeCo memory layer.

The temperature dependences of the MR ratio and the resistance change  $\Delta R = R_{high}-R_{low}$  were shown in Fig. 3.10, and temperature dependence of the resistance  $R_{low}$  (resistance of parallel configuration) for the typical Gd<sub>23</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>77</sub> memory layer was shown in the inset. From Fig. 3.10, the MR ratio decreased with increasing temperature. This typical temperature dependence of MR ratio is well known in most GMR structures [6]. However, the value of MR ratio is not only dependent on the resistance change  $\Delta R$  but also the absolute value of resistance  $R_{low}$ , which is also influenced by temperature increase as shown in the inset. It is obviously shown in Fig. 3.10, that the resistance change  $\Delta R$  has the similar decreasing tendency with that of MR ratio when the temperature rises. From general viewpoint, the decrease of the resistance change  $\Delta R$  indicates the decrease of the spin polarization near the Fermi level of the magnetic atoms [7]. The reduction of critical current density  $J_c$  that will be argued in detail later.



Fig. 3.10 Temperature dependence of MR ratio and resistance change  $\Delta R = R_{\text{high}}-R_{\text{low}}$  of the GMR device with Gd<sub>23</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>77</sub> memory layer. Temperature dependence of the resistance of parallel configuration,  $R_{\text{low}}$ , is shown in the inset.

# 3. 3. Spin injection switching of GdFeCo in GMR structure

Figure 3.11 shows typical major (left side) and minor (right side) MR curves taken for the CPP-GMR structures with GdFeCo memory layer having different Gd compositions. The MR changes in high and low fields correspond to the switching of Co/Pd reference layer and GdFeCo memory layer, respectively. The minor loops were taken after applying a large positive field to saturate the sample, and thus the loops reflect the switching of GdFeCo memory layer. The MR ratio did not depend systematically on the Gd composition varied from 21 at.% to 24 at.%. and the maximum MR ratio about 0.24% was observed for the Gd<sub>21.4</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78.6</sub> memory layer. The MR ratio is considered to originate from the spin polarization at Fermi level of the GdFeCo memory layer which is mainly dominated by FeCo atoms [7]. The MR ratio of 0.24% is reasonable for CPP-GMR structure but still small compared to that of CPP-GMR structure with a Co/Ni memory layer [8]. The low MR ratio is possibly due to the low spin polarization and/or high resistance of the amorphous GdFeCo memory layer. The coercivities of both reference and memory layers are larger than those of the corresponding as-deposited layers shown in Fig. 3.3 (d). This is mainly caused by the size effects on the coercivity of ferromagnetic materials of the sample after micro-fabrication. Similar increases of coercivity are

also found in the Gd<sub>22.3</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>77.7</sub> and Gd<sub>23.2</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>76.8</sub> memory layers whose hysteresis loops can been seen in Fig. 3.3 (e) and (f), respectively. The coercivities of the GdFeCo memory layers increase with the Gd composition as shown in Fig. 3.11 (a)-(d). We attribute this to the reduction of the magnetization of the GdFeCo layers in the TM-rich regime because of the anti-parallel configuration of Gd and FeCo sub-lattice moments. The decrease of the magnetization of GdFeCo layer causes the increase of  $K_{eff}$  when the anisotropy constant  $K_u$  is assumed to keep constant within the narrow Gd composition range of 21 ~ 24 at.%.



(c)  $Gd_{22.3}(Fe_{90}Co_{10})_{77.7}$ 



(d)  $Gd_{21.4}(Fe_{90}Co_{10})_{78.6}$ 

Fig. 3.11 Major and minor MR loops for GMR structure with  $Gd_x(Fe_{90}Co_{10})_{100-x}$ memory layer of  $120 \times 180$  nm<sup>2</sup>.

### 3.3.1. Resistance-current (R-I) measurement

Among the four samples in Fig. 3.11, the CPP-GMR structure with  $Gd_{21.4}(Fe_{90}Co_{10})_{78.6}$  memory layer was selected to be checked for the spin injection switching since it exhibited slightly higher MR ratio than the others. The *R-I* loops to confirm the STT switching for  $120 \times 180$  nm<sup>2</sup> cell at various bias magnetic field are shown in Fig. 3.12, where the external magnetic field was applied during the measurement to cancel the exchange coupling through the Cu spacer. The loops were taken by measuring the resistance after applying the current pulse with a pulse width of 100 msec.









Fig. 3.12 *R-I* loops for the CPP-GMR structure with  $Gd_{21.4}(Fe_{90}Co_{10})_{78.6}$  memory layer at various bias magnetic field.

The distinct resistance changes correspond to the full reversal of the GdFeCo layers by the STT switching were confirmed. A positive current means that electrons flow from the bottom Co/Pd multilayer to GdFeCo memory layer which tend to align the magnetization of the GdFeCo parallel (P) to the Co/Pd as seen in Fig. 3.12, and a negative current tends to be antiparallel (AP) vice versa. Fig. 3.13 shows the dependence of the critical current densities  $J_c$  to switch P to AP and AP to P configurations on the external magnetic field,  $H_{ext}$ , for the sample with Gd<sub>21.4</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78.6</sub> memory layer. Both P-AP and AP-P current densities, J<sub>c P-AP</sub> and  $J_{c AP-P}$ , respectively, depend on the external field, but their average value,  $(J_{c P-AP} + J_{c}$  $_{AP-P}$ ) / 2, was almost independent of the field, which is estimated to be 1.6  $\times$  10<sup>7</sup> A/cm<sup>2</sup>. It is noted that the current density for P to AP was lower than that for AP to P within the coercivity of the GdFeCo layer:  $H_{\text{ext}} = -1100$  Oe to -700 Oe . From Fig. 3.13, relatively low current densities were found even at the conditions of low MR ratio and relatively thick GdFeCo memory layer (10 nm). The current density in the order of a few 10<sup>7</sup> A/cm<sup>2</sup> is comparable to that of the conventional MTJs with much larger MR ratio and thickness of the free layer about 1~2 nm [9].



Fig. 3.13 Bias magnetic field dependence of critical current density  $J_c$  of the CPP-GMR structure with Gd<sub>21.4</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78.6</sub> memory layer.

## **3.3.2.** Compositional dependence of critical current density $(J_c)$

The Gd compositional dependence of  $J_c$  was also systematically investigated due to the reason described in the previous section. Figs. 3.14 (a) – (c) show three typical R-I loops for the CPP-GMR structure with GdFeCo memory layer having different Gd contents. The distinct resistance changes correspond to the full reversal of the GdFeCo layers by the STT switching are also confirmed in all the samples. In Fig. 3.15, Gd compositional dependence of the average critical current density  $J_{\rm c}$ was drawn from the data of the STT switching shown in Fig. 3.14. The current density  $J_c$  increased with increasing the Gd composition, which is considered to be mainly due to the increase of the effective perpendicular anisotropy since the demagnetizing energy of GdFeCo layers reduces with increasing Gd content in the range of Gd 21.4~24.1 at.%. The effective anisotropy  $K_{\rm eff}$  as a function of the Gd content is also shown in Fig. 3.15. The  $K_{eff}$  was estimated from the *M*-H loops of the GdFeCo single layers with various Gd compositions as described in Fig. 3.7 (e). The increase of the  $K_{\rm eff}$  is coincident well with the increase of  $J_{\rm c}$  with the Gd content. A similar tendency is reported recently in GdFe memory layer by Aoshima et al. [10]. To estimate the intrinsic current density  $J_{c0}$ , one needs to investigate the current pulse width dependence of the critical current density  $J_c$  according to the Eq. (3.2) [11]

$$J_{c} = J_{c0} \left[ 1 - \frac{k_{B}T}{K_{eff}V} \ln\left(\frac{\tau}{\tau_{0}}\right) \right]$$
(3.2)

where  $\tau_0 \sim 1$  ns is the inverse of the attempt frequency and  $\tau$  is the pulse width for the STT switching.  $K_{\text{eff}}V$  and  $k_{\text{B}}T$  are the anisotropy energy of the memory layer and the thermal energy, respectively.



Fig. 3.14 *R-I* loops measured for GMR devices with (a)  $Gd_{21.4}(Fe_{90}Co_{10})_{78.6}$ , (b)  $Gd_{22.3}(Fe_{90}Co_{10})_{77.7}$ , and (c)  $Gd_{24.1}(Fe_{90}Co_{10})_{75.9}$  memory layers. The external field  $H_{ext}$  ((a) -788Oe, (b) -917Oe, (c) -193Oe) was applied to cancel the exchange coupling between the GdFeCo and [Co/Pd] multilayer.

To roughly estimate the intrinsic current density  $J_{c0}$ , we calculate the value of thermal stability factor  $\Delta = K_{eff}V/k_BT$  from  $K_{eff}$  estimated for the single layers. For the case of sample with  $Gd_{21.4}(Fe_{90}Co_{10})_{78.6}$  memory layer, the effective anisotropy constant  $K_{eff}$  of  $6.0 \times 10^4$  erg/cc and subsequently  $\Delta = 210$  were obtained. Finally, the value of  $J_{c0}$  of  $1.7 \times 10^7$  A/cm<sup>2</sup> was obtained. The values of  $J_{c0}$  estimated from Eq. (3.2) are also plotted in Fig. 3.15, and the  $J_{c0}$  is quite comparable to  $J_c$  because of the large  $\Delta$  for the present GdFeCo memory layers. The  $J_{c0}$  is known to be expressed as

$$J_{c0} = \alpha \frac{\gamma e}{\mu_B \eta} K_{eff}$$
(3.3)

Where  $\alpha$  is the Gilbert damping constant,  $\gamma$  the gyromagnetic ratio, *e* the elementary charge,  $\mu_{\text{B}}$  the Bohr magnetron, and  $\eta$  the STT efficiency. The Eq. (3.3) roughly describes the result shown in Fig. 3.15;  $J_{c0}$  is proportional to the  $K_{\text{eff}}$ . However, Eq. (3.3) means that the  $J_{c0}$  is also influenced by  $\alpha$  and  $\gamma$  of the memory layer. The  $\alpha$  and  $\gamma$  are known to be also dependent on both temperature and Gd composition in GdFeCo [12, 13]. The increase of  $J_c$  with the Gd composition shown in Fig. 3.15 may be partly due to the increase of  $\alpha$  and  $\gamma$ , since both parameters will increase when the Gd content approaches to the angular momentum compensation point (Gd 24 at.%) [13]. Further increase of Gd content will drastically change  $\alpha$  and  $\gamma$ , and thus more systematic study will be needed to understand the compositional dependence of  $J_c$  in GdFeCo.



Fig. 3.15 Dependence of critical current density  $J_c$ ,  $J_{c0}$  (estimated from Eq. (3.2)) and effective anisotropy  $K_{eff}$  on the Gd composition in GMR devices with GdFeCo memory layers.

The conventional method to obtain the thermal stability  $\Delta$  is to investigate the pulse width dependence of critical current density  $J_c$ . In Fig. 3.16, the pulse width dependence of  $J_c$  was measured by checking the resistance after the pulse applications with pulse width in the range of 100ns~100ms. To carry out this experiment the pulse generator (Agilent 81104A) was connected parallel to the DC current source for the resistance measurement. The electrical connection to the CPP-GMR was switched from one power source to another before and after the pulse application by using in-house made CMOS switch circuit. In Fig. 3.16, the slope of the  $J_c$  vs  $\ln(t/t_0)$  curve corresponding to the reverse of the thermal stability  $\Delta$ according to the Eq. (3.2). The slope can be obtained by fitting the experimental data of  $J_c$  at different pulse width and subsequently the  $\Delta$  was achieved by simple calculation. The sample investigated in Fig. 3.16 was chosen to be GMR structure with memory layer of Gd<sub>23.4</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>76.6</sub>. From the calculation of the slopes of two  $J_{\rm c} - \ln(t/t_0)$  curves for the AP-P and P-AP reversals respectively, the average thermal stability  $\Delta$  of about 141 was obtained which was shown in Fig. 3.15 as  $K_{\rm eff}(STT)$  for comparison. The large difference of  $K_{\rm eff}$  obtained by two alternative methods may be due to the small nucleation volume in the STT switching of the GdFeCo. In the STT switching, the current is considered to switch first the GdFeCo much smaller than the cell size, which is called as nucleation site, and the reversed nucleation site will be expanded resulting in the full reversal of GdFeCo. In this case,  $\Delta$  will be independent on the cell size until the size will approach to the nucleation volume. Another possibility is the shift of Gd composition after the micro-fabrication. Even in RE-rich sample, positive MR (high resistance in AP state) was observed after the micro-fabrication. As shown in Fig. 3.9, the GMR films with RE-rich GdFeCo exhibit the negative MR (high resistance in P state), but CPP-GMR after the fabrication exhibited the positive MR, which may be due to the shift of the composition to TM-rich side after the micro-fabrication. From Fig. 3.15, the shift of the composition to TM-rich side will result in the decrease of  $\Delta$  due to the increase of the GdFeCo magnetization.



Fig. 3.16 Pulse width dependence of critical current density of GMR structure with  $Gd_{23.4}(Fe_{90}Co_{10})_{76.6}$  memory layer.

#### **3.3.3.** Temperature dependence of critical current density $(J_c)$

Temperature is another crucial factor to affect the current density  $J_c$ . Therefore, *R-I* loops of the sample with Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> memory layer were measured at various temperatures and the data are shown in Figs. 3.17 (a)-(f). The critical currents to switch the magnetization from parallel (P) state to antiparallel (AP) state and vice versa gradually decreased by elevating the temperature from room temperature (17°C) to about 70°C at a constant bias magnetic field  $H_{\text{bias}} = -828.7$  Oe. The resistance of sample at parallel state (or antiparallel state) gradually increased with the temperature, which obeys the resistance-temperature relationship shown in the inset of Fig. 3.10. The noise-like fluctuation of the resistance becomes significant above 50°C. Fig. 3.17 (g) shows the temperature dependence of the average critical current density  $J_c$  obtained from the results of Figs. (a)–(f). The  $J_c$  clearly decreased with temperature. It is noted that the  $J_c$  was reduced by 16% by elevating the temperature. It is well known that  $J_c$  is proportional to the  $K_{\text{eff}}$  [5] and such distinct reduction of  $J_c$  shown in Fig. 3.17 was considered to be partly due to the decrease of  $K_{\text{eff}}$  with increasing temperature.







Fig. 3.17 Temperature dependence of  $J_c$  of GMR structure with  $Gd_{21}(Fe_{90}Co_{10})_{79}$  memory layer.

The temperature dependence of  $K_{\text{eff}}$  of single  $Gd_{21}(Fe_{90}Co_{10})_{79}$  memory layer was shown in Fig. 3.18. However, this will not fully explain the 16%  $J_c$  reduction for the temperature increase of ~50°C as shown in Fig. 3.17 (g).



Fig. 3.18 K<sub>eff</sub> dependence on temperature increase of Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> memory layer.

Another possible origin may be related to the fluctuation of magnetization direction of memory layer when the temperature is increasing. The efficiency of the STT torque is known to be dependent on the relative angle of the directions between the injected spins and local magnetic moment, and it will vanish at relative angles of  $0^{\circ}$  and 180<sup>°</sup> [15]. Thus the fluctuations of the magnetization direction in the memory layer may enhance the efficiency of the STT torque, which contributes to the reduction of  $J_c$ . We have to state here that the fluctuation of the magnetization at elevated temperature does not indicate the thermal instability at room temperature. The GdFeCo is considered to have sufficient thermal stability factor  $\Delta$  at room temperature, but the  $\Delta$  of this material can be modified by moderate temperature increase, which is critical parameter in our scheme of thermally assisted STT-RAM. Besides, the temperature variation can additionally affect other parameters, e.g., g-factor and damping constant  $\alpha$ , and finally influence the  $J_c$  value [12, 13]. To fully understand all the related parameters to determine the critical current density  $J_c$ , further systematical investigation on the parameters is needed.

#### **3.4.** Conclusions

Samples with Gd content in the range of 18~27at.% were measured in terms of magnetic properties. There is a compensation point at which the magnetization of GdFeCo layer vanishes, that was confirmed both in *M-H* and MR loops in that range of Gd content. The perpendicular magnetization was affected after the heat cycleat the temperature of about 170°C for TM-rich GdFeCo layer and 150°C for RE-rich GdFeCo layer.

GMR devices with ferrimagnetic amorphous GdFeCo memory layers were prepared by magnetron sputtering and subsequent micro-fabrications by EB- and photo-lithography. The magnetic properties and magneto-resistance measurements show that GdFeCo memory layers with various Gd contents have sufficiently large PMA. The *R-I* loops indicate the GdFeCo memory layer can be fully reversed by a pulse current due to STT switching. A relatively low current density of  $1.6 \times 10^7$ A/cm<sup>2</sup> while high thermal stability factor  $\Delta = 210$  (estimated from *M-H* loop) was obtained for the sample with Gd<sub>21.4</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>78.6</sub> memory layer. The critical current density *J*<sub>c</sub> is found to increase with increasing Gd content, mainly due to the increase of the effective perpendicular anisotropy of the memory layer.

R-I loops of the sample with GdFeCo memory layer were measured under

varying the current pulse width and temperature. The  $K_{\text{eff}}$  of the GdFeCo memory layer estimated from the pulse width dependence of  $J_c$  was much smaller than that estimated from the *M-H* loop. This may be due to the small nucleation volume in STT switching and/or shift of Gd composition after the micro-fabrication. For temperature dependence, the critical current density  $J_c$  is found to decrease with increasing temperature, which will be related to the reduction of  $K_{\text{eff}}$  and fluctuation of the magnetization direction of the memory layer with elevating temperature. Further investigation is necessary to understand the compositional and temperature dependences of the  $J_c$  in the GdFeCo memory layers obtained in this chapter. Finally, the present results show that the  $J_c$  is clearly reduced with elevating temperature, and the effective perpendicular anisotropy  $K_{\text{eff}}$ , one of the key parameter to determine  $J_c$ , is easily tuned by the temperature increase for the GdFeCo memory layer. Thus, the amorphous GdFeCo is considered to be a promising candidate used for the memory layer of the thermally assisted STT-RAM.

In the scheme of thermally assisted STT-RAM, sufficiently large reduction of  $K_{\text{eff}}$  with elevating temperature is desired especially to achieve the density of 10Gb. For this requirement,  $K_{\text{eff}}$  of single GdFeCo layer will not be high enough to reduce the cell size for high density memories. In addition, the slope of  $K_{\text{eff}}$ -T curve is not so sharp for the thermally assisted STT-RAM. Thus we propose an unique bilayer structure consisting of TbFe and GdFeCo, in which TbFe and GdFeCo are exchenge-coupled to increase  $K_{\text{eff}}$ . The  $K_{\text{eff}}$  of TbFe can rapidly vanish when temperature increases, the remaining part GdFeCo can then be easily switched by spin polarized current. This bilayer structure will be discussed in detail in next chapter.

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# Chapter 4 Spin injection switching of TbFe/GdFeCo bilayer

In the scheme of thermally assisted STT-RAM, sufficiently large reduction of  $K_{\text{eff}}$  with elevating temperature is desired especially to achieve the memory capacity of Giga bit. For this requirement,  $K_{\text{eff}}$  of single GdFeCo layer will not be high enough to reduce the cell size for high density memories. In addition, the slope of  $K_{\text{eff}}$ -T curve is not so sharp for the thermally assisted STT-RAM. Thus we propose an unique bilayer structure consisting of TbFe and GdFeCo, in which TbFe and GdFeCo coupled as an exchange-coupled bilayer to increase  $K_{\text{eff}}$ . The  $K_{\text{eff}}$  of TbFe can rapidly vanish when temperature increase, the remaining part GdFeCo can then be easily switched by spin polarized current.

Thus, in this chapter, the thermally assisted spin injection switching of TbFe/GdFeCo bilayer structure will be discussed. The Tb compositional dependence of various magnetic properties of the bilayer structure was confirmed to obtain the optimum configuration of constituent TbFe and GdFeCo layers. The STT switching at the room temperature and elevating temperature were both measured. It is noted that the thickness of the TbFe layer has important influence on the STT switching and therefore the dependence of spin injection switching current density on the thickness of TbFe was also investigated. The mechanism of the STT switching in the CPP-GMR with single GdFeCo memory layer. Finally, TbFe/GdFeCo bilayer structure is demonstrated as a potential candidate of the memory layer for the thermally assisted STT switching.

## 4.1. Sample preparation

The GMR structure with the bilayer memory layer is Si / Ta (10) / CuAl (150) / Tb<sub>15</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>85</sub> (20) / Co<sub>40</sub>Fe<sub>40</sub>B<sub>20</sub> (1) / Cu (6) / Gd<sub>x</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>100-x</sub> (10 - t) / Tb<sub>x</sub>Fe<sub>100-x</sub> (t) / Cu (5) / Ta (5) (thickness is in nanometer). The deposition of the samples was carried out in the same way as described in the previous chapter by using an UHV magnetron sputtering system with a base pressure around  $1 \times 10^{-7}$  Pa.

The Tb<sub>15</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>85</sub> reference layer was deposited by co-sputtering of Tb and Fe<sub>90</sub>Co<sub>10</sub> targets. The reference Co/Pd multilayer was replaced with TbFeCo layer, As described later, the introduction of the TbFe/GdFeCo bilayer results in high critical current density J<sub>c</sub> for the switching compared to that of the single GdFeCo layer, and the high  $J_c$  may disturb the magnetization of Co/Pd since the large spin torque is exerted not only to the memory layer but to the reference layer. The magnetization direction of TbFeCo is considered to be stable against the application of the spin torque because of the large damping constant of Tb. The perpendicular easy axis of the TbFeCo reference layer is achieved by varying the content of Tb similar to that of the GdFeCo layer. The  $Tb_{15}(Fe_{90}Co_{10})_{85}$  or  $Gd_x(Fe_{90}Co_{10})_{100-x}$  layer was deposited by a co-sputtering of Tb or Gd and Fe90Co10 targets. Tb or Gd composition was controlled by varying sputtering power of the Tb or Gd target. The magnetic properties of the samples were characterized by AGM and torque magnetometer. GMR devices with various dimensional elliptical patterns ranging from  $120 \times 180 \text{ nm}^2$  to  $300 \times 450 \text{ nm}^2$  were micro-fabricated by EB lithography and subsequent Ar<sup>+</sup> etching from the ECR plasma source. After the etching, a SiN insulation layer was deposited by the RF-magnetron sputtering system to avoid oxidation of the RE elements, such as Gd and Tb atoms. The lift-off process was carried out after the SiN deposition using EB resist remover at about 80°C. The electrodes of aluminum were formed by the deposition of Al and the lift-off process. Four-point probe method was exploited to measure the MR effect, and STT switching property of these devices was characterized by the resistance measurement after flowing pulse current with 100 msec pulse duration. The typical structures of the samples are illustrated in Fig. 4.1.

Tb <sub>16</sub> Fe-5nm	Tb <sub>16</sub> Fe-1nm
Gd <sub>21</sub> (Fe <sub>90</sub> Co <sub>10</sub> ) <sub>79</sub> -5nm	Gd <sub>21</sub> (Fe <sub>90</sub> Co <sub>10</sub> ) <sub>79</sub> -9nm
Cu-6nm	Cu-6nm
CoFeB-1nm	CoFeB-1nm
$Tb_{15}(Fe_{90}Co_{10})_{85}$ -20nm	Tb <sub>15</sub> (Fe <sub>90</sub> Co <sub>10</sub> ) <sub>85</sub> -20nm

Fig. 4.1 Typical GMR structures with TbFe/GdFeCo bilayer.

#### 4.2. Magnetic properties of TbFe/ GdFeCo layer

### 4.2.1. Saturation magnetization (M<sub>s</sub>)

Magnetic properties of bilayer structures of Ta (5)/Cu (5)/Tb<sub>x</sub>Fe<sub>100-x</sub> (10-*t*) /  $Gd_y(Fe_{90}Co_{10})_{100-y}$  (*t*)/Cu (6)/ Tb<sub>15</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>85</sub> (20) (thickness in nanometers) with various Gd and Tb contents and various thickness ratios were investigated. Typical *M-H* loops of the bilayers are shown in Fig. 4.2, and the *M-H* loop of the Tb<sub>15</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>85</sub> (20 nm) single layer is also shown to confirm the loop of the reference layer in the GMR film.

It is noted that the TbFe / GdFeCo bilayers shown in Fig. 4.2 are all sandwiched by Cu (5 nm) layers to simulate the memory layer in the CPP-GMR structure. The shape of the loop varies with the Tb content and thickness ratio in the TbFe / GdFeCo bilayers. For the requirement of spin injection switching, thinner TbFe layer in the bilayer structure and low Tb composition are believed to be benefit for the reduction of switching current density because of the reduced  $K_{\rm eff}$  in the bilayer structure, which will be discussed in the following section. Meanwhile, sufficiently large  $K_{\rm eff}$  by adjusting the Tb content and thickness ratio of the bilayer are necessary to achieve enough thermal stability factor  $\Delta$  at room temperature. The 20 nm-thick  $Tb_{15}(Fe_{90}Co_{10})_{85}$  layer shown in Fig.4.2 (a) exhibits high magnetization, coercivity, high Curie temperature (not shown in the figure), and also high damping constant (expected), whereby it is an ideal candidate for the reference layer under the scheme of thermally assisted STT-RAM. For the TbFe / GdFeCo bilayers, the coercivity is reduced when the Tb content varies from 20 at.% to 16 at.% as shown in Figs. 4.2 (b) - (d). For comparison between TbFe / GdFeCo bilayers with different thickness ratio, Fig. 4. 2 (d) and (e) shows M-H loops of the Tb<sub>16</sub>Fe<sub>84</sub> (5nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5nm) and Tb<sub>16</sub>Fe<sub>84</sub> (3nm) / Gd<sub>19</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>81</sub> (7nm) bilayers, respectively. It can be seen that the coercivity of the bilayer decreases with reducing the thickness of the constituent TbFe and decreasing the Gd or Tb content in the bilayer.



Fig. 4.2 *M*-*H* loops of Tb<sub>x</sub>Fe<sub>100-x</sub> (10–*t*) / Gd<sub>y</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>100-y</sub> (*t*) bilayers with various Gd and Tb contents and thickness ratios. *M*-*H* loop of Tb<sub>15</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>85</sub> (20nm) single layer is shown in Fig. (a) to confirm the magnetic property of the reference layer in the GMR film. (b) x = 20 at.%, t = 5 nm; (c) x = 18 at.%, t = 5 nm; (d) x = 16 at.%, t= 5 nm and (e) x = 16 at.%, t = 7 nm, y = 19 at.%.

To compare the single GdFeCo layer with the bilayer, two GMR structures with memory layers of GdFeCo (10nm) single layer and  $Tb_{16}Fe_{84}$  (5nm) /  $Gd_{21}(Fe_{90}Co_{10})_{79}$  (5nm) bilayer are shown in Fig. 4.3.



Fig. 4.3 Comparison of *M*-*H* loops for GMR structures with GdFeCo single layer and TbFe / GdFeCo bilayer.

From Fig. 4.3, it is clearly shown that the bilayer has much larger coercivity than that of single GdFeCo layer even though the magnetizations are comparable. This feature is believed to be suitable for thermally assisted STT-RAM with memory capacity of a few Giga bit.

The feature of temperature dependence of magnetic properties was investigated by taking Kerr loops for the Tb<sub>16</sub>Fe<sub>84</sub> (3 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (7 nm) bilayer and Tb<sub>16</sub>Fe<sub>84</sub> (5 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) bilayer at various temperatures, which are shown in Figs. 4. 4 (a) – (c) and Figs. 4. 5 (a) – (e) respectively. The coercivities of these two types of bilayers were plotted as a function of temperature as shown in Fig. 4. 4 (d) and Fig. 4. 5 (f) respectively.





Fig. 4.4 Kerr loops of Tb<sub>16</sub>Fe<sub>84</sub> (3 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (7 nm) bilayer taken at (a) 25.1°C, (b) 57°C, (c) 84°C, and (d) temperature dependence of  $H_c$  of the Tb<sub>16</sub>Fe<sub>84</sub> (3 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (7 nm) bilayer.





Fig. 4.5 Kerr loops of Tb<sub>16</sub>Fe<sub>84</sub> (5 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) bilayer taken at (a)
25.1°C, (b) 40°C, (c) 83°C, (d) 140°C, (e) 176°C, and (f) temperature dependence of *H*<sub>c</sub> of the Tb<sub>16</sub>Fe<sub>84</sub> (5 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) bilayer.

The coercivities  $H_c$  of these bilayers were rapidly decreased with increasing temperature, which is suitable for the proposed thermally assisted STT-RAM. The

slope of the  $H_c$ -T curve is somehow the indication of the magnetic properties related to temperature variation, for example, the temperature dependence of  $K_{eff}$ . From the slope of the  $H_c$  - T curve shown in Fig. 4. 4 (d) and Fig. 4. 5 (f), it can be calculated that the temperature at which  $H_c$  becomes zero for the bilayers structure are roughly 100°C.

## **4.2.2.** Effective anisotropy constant (*K*<sub>eff</sub>)

Similar to the case of the single GdFeCo layer, the effective anisotropy constant  $K_{\text{eff}}$  of the bilayer structure is also a crucial parameter to determine switching current density  $J_c$ . The temperature dependences of  $K_{\text{eff}}$  of bilayers with various thickness ratio of Tb<sub>16</sub>Fe<sub>84</sub> (10-*t*)/ Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (*t*) (thickness in nanometer) layers are shown in Fig.4.6. In all the structures with different thickness of TbFe, the  $K_{\text{eff}}$  was decreased with elevating the temperature from room temperature to 160°C. At a certain temperature, the bilayer structure with thicker TbFe exhibits lower ratio of  $K_{\text{eff}}(T) / K_{\text{eff,RT}}$ , in which  $K_{\text{eff}}(T)$  is defined as the  $K_{\text{eff}}$  at specific temperature, T,  $K_{\text{eff,RT}}$  corresponding to  $K_{\text{eff}}$  at room temperature of the identical bilayer structure. From the data shown in Fig. 4.6, the bilayer of TbFe (5 nm) / GdFeCo (5 nm) is considered to be suitable for the thermally assisted STT-RAM due to the significant reduction of the  $K_{\text{eff}}$  by slight increase of temperature. The STT switching experiment will be conducted in the next section to discuss how to achieve the high thermal stability factor and efficient STT switching simultaneously.



Fig. 4.6 Temperature dependence of  $K_{\rm eff}$  for bilayer with various TbFe thickness.

## 4.2.3. Magneto-resistance (MR) properties

Temperature dependence of the MR property of the non-micro-fabricated sample with GMR structure of Ta (5nm)/ Cu (5nm)/ Tb<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) / Cu (6nm)/ [Co (0.4nm)/Pd (1.6nm)]<sub>6</sub> /Si is shown in Fig. 4.7 ~ Fig. 4.11.



Fig. 4.7 MR curve of the GMR sample taken at room temperature.



Fig. 4.8 MR curve of the GMR sample taken at 27°C.



Fig. 4.9 MR curve of the GMR sample taken at 36°C.



Fig. 4.10 MR curve of the GMR sample taken at 47°C.



Fig. 4.11 MR curve of the GMR sample taken at 63°C.

At room temperature, the magnetization of the memory layer did not switch in the maximum field of 10 kOe, and the MR changes at  $\pm$  2.5 kOe are due to the switching of the reference layer. The reference layer in this GMR film is Co/Pd multilayer not TbFeCo layer. The Co/Pd multilayer has a high Curie temperature and its coercivity keeps constant for the temperature range of room temperature to 63°C. For the temperature > 36°C, the MR change due to the switching of TbFe / GdFeCo memory layer was confirmed in the range of  $\pm$  10 kOe external field, and the coercivity *H*<sub>c</sub> of the memory layer was significantly decreased with a slight temperature increase. The MR ratio is slightly decreased with the temperature.

# 4.3. Spin injection switching of TbFe / GdFeCo in GMR structure

#### 4.3.1. Jc of TbFe (5nm) / GdFeCo (5nm) memory bilayer

The spin injection switching of bilayer was first carried out in the CPP-GMR

structure with a Tb<sub>16</sub>Fe<sub>84</sub> (5 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) memory layer. In this experiment, Tb<sub>15</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>85</sub> (20 nm) was used as a reference layer. The area of junction is  $120 \times 180$  nm<sup>2</sup>. The typical minor MR curve of the memory layer is shown in Fig. 4. 12. Before taking the minor loop shown in Fig. 4.12, sufficiently large positive field of +20 kOe was applied to saturate the reference layer. In the minor loop, we can clearly see that the coercivity of the memory layer is much larger than that of the single GdFeCo layer shown in Fig. 3.2 (c) and Fig. 3.3 (d), which is mainly attributed to the introduction of TbFe layer. There is no step in the minor loop that indicates the strong coupling between the TbFe and GdFeCo layers resulting in the simultaneous switching of the two layers at room temperature.



Fig. 4.12 MR minor loops of the CPP-GMR structure with  $Tb_{16}Fe_{84}$  (5 nm) /  $Gd_{21}(Fe_{90}Co_{10})_{79}$  (5 nm) memory layer.



Fig. 4.13 Typical P-AP and AP-P reversals by a current pulse observed in the CPP-GMR structure with the TbFe / GdFeCo memory layer at different bias magnetic field.

Two typical *R-I* curves are shown in Fig. 4.13, where the reference TbFeCo layer was first saturated by a sufficiently large positive field. The parallel (P) state was switched to antiparallel (AP) state by applying a negative current of -12 mA under application of negative field of -1.4 kOe, while AP to P switching was confirmed by applying a positive current of 10 mA under +1.3 kOe. It is noted that the P–AP and AP–P switching of the memory layer by the current pulse was not observed simultaneously in a certain external field. We attribute these phenomena to the very large  $K_{\text{eff}}$  of the bilayer described above. The temperature increase would reduce the  $K_{\text{eff}}$  effectively whereby the switching current is expected to reduce.



Fig. 4.14 Bias magnetic field dependence of critical current density.

A set of data of the current densities of P–AP and AP–P switching were obtained at various bias magnetic field, which are shown as open circles in Fig. 4.14. To get the average current density, the method used in the previous chapter will not be valid because the AP-P and P-AP reversals did not occur at identical bias magnetic field. Here we propose an alternative way to calculate the average current density by the following steps. First,  $J_c$ -H experimental data were linearly extrapolated to H = 0 to obtain  $J_{c,AP-P}$  and  $J_{c,P-AP}$  at H = 0, and then, average  $J_c =$  $(J_{c,AP-P} + J_{c,P-AP}) / 2$  was simply calculated. The average current density of this memory layer is estimated to be  $1.8 \times 10^8$  A/cm<sup>2</sup>, which is much larger than that of CPP-GMR structure with GdFeCo single memory layer. Such a large current may cause significant temperature increase in the memory layer resulting in thermally assisted magnetic field switching instead of STT switching.

To distinguish these two reversal modes; thermally assisted magnetic field switching and STT switching, temperature and bias field dependences of the switching current density were investigated. Several typical reversals, either AP-P or P-AP reversal modes, at various temperatures are shown in Fig. 4.15. The measurements were carried out for the CPP-GMR structure with the Tb<sub>16</sub>Fe<sub>84</sub> (5 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (5 nm) memory layer, and all the measurements were done after the saturation of TbFeCo memory layer by sufficiently large positive magnetic field of 20 kOe. In general, the resistance change due to the switching of the memory layer at low temperature is larger than that at high temperature. Moreover, the large bias field was necessary to observe the switching by applying a current pulse (up to 10 mA) at low temperature, however the required bias magnetic field for the current pulse switching was obviously reduced by increasing temperature.



(a) P-AP reversal at H = -1498Oe and  $T = 16^{\circ}$ C



(b) P-AP reversal at H = -1498Oe and  $T = 45^{\circ}$ C


(c) P-AP reversal at H = -548Oe and T = 148°C





Fig. 4.15 Current induced AP-P or P-AP reversal observed in the CPP-GMR structure with TbFe (5 nm) / GdFeCo (5 nm) memory layer at various temperatures and bias magnetic fields.

All the data of the current induced switching obtained for the CPP-GMR structure at various temperatures and bias magnetic fields were summarized in Fig. 4. 16. The typical full reversal loop was not obtained at specific identical bias field, and P-AP (AP-P) reversal was only achieved at negative (positive) field. If the bias field dependence of  $J_c$  is extrapolated linearly to H = 0, average  $J_c$  can be calculated just as the method described above. In the STT switching, the  $J_c$  in Fig. 4.16 should be proportional to  $K_{eff} - M_s H$  which is easily deduced from eq. (3. 3), since the energy barrier height becomes roughly  $K_{eff} - M_s H$  under the external field, H. This

suggests that the slope in Fig. 4.16 will decrease with increasing temperature because of the monotonic reduction of  $M_s$  of the memory layer with temperature. Therefore, the feature observed in Fig. 4.16 is not explained simply by STT switching.



Fig. 4.16 External field dependence of *J*<sub>c</sub> (P–AP or AP–P) of the CPP-GMR with the TbFe (5 nm) / GdFeCo (5 nm) memory layer obtained at various temperatures.

The average  $J_c$  at H = 0 estimated from the simple linear extrapolation of the data in Fig. 4.16 was plotted as a function of temperature in Fig. 4.17 (a). As a reference, temperature dependence of  $K_{eff}$  of TbFe (5 nm) / GdFeCo (5 nm) bilayer film which was measured by torque magnetometer is shown in Fig. 4. 17 (b). It is clearly seen that there is a peak in the  $J_c-T$  curve, which is hardly understood by a STT switching picture. The  $J_c$  should be proportional to  $K_{eff}$  at H = 0 as in eq. (3. 3), and the  $K_{eff}$  decrease monotonously with elevating temperature as in Fig. 4.17 (b). If the STT switching is valid to explain the data shown in Fig. 4.16, the  $J_c-T$  curve should show a similar tendency with  $K_{eff}-T$  curve.



(a) Temperature dependence of average  $J_c$ .



(b) Temperature dependence of  $K_{\rm eff}$ 

Fig. 4.17 Temperature dependences of  $J_c$  of the CPP-GMR structure with TbFe (5 nm) / GdFeCo (5 nm) memory layer and  $K_{eff}$  for the corresponding bilayer film. The  $J_c$ , average values of  $J_{c AP-P}$  and  $J_{c P-AP}$  at H = 0, was estimated by a linear extrapolation of the data in Fig. 4.16 to H = 0.

To verify the switching mode, the data in Fig. 4.16 were re-plotted as  $J_c^2-H$  curves, which were shown in Fig. 4.18. Even in  $J_c^2-H$  curves, we can find linear relationship, and the slope was independent of the measurement temperature. This tendency is consistent with the result that the magnetization (or Kerr rotation) of the bilayer was almost constant in the temperature range from room temperature to 100°C as shown in Fig. 4.5. If we extrapolate  $J_c^2-H$  curves linearly, it is possible to estimate average  $J_c^2$  at H = 0 similar to the previous discussion. Temperature

dependences of the average  $J_c^2$  at H = 0 and  $K_{eff}$  estimated from torque curves are shown in Fig. 4. 19 (a) and (b), respectively. As shown in the figure, the  $J_c^2-T$  curve shares similar tendency to the  $K_{eff}-T$  curve. This means that the barrier height of the current induced switching is scaled with  $J_c^2$  not  $J_c$ , suggesting thermally assisted field switching feature.



Fig. 4.18 External field dependence of  $J_c^2$  of the CPP-GMR with TbFe (5 nm) / GdFeCo (5 nm) memory layer obtained at various temperatures.



Fig. 4.19 Temperature dependences of (a) average  $J_c^2$  of the CPP-GMR structure with TbFe (5 nm) / GdFeCo (5 nm) memory layer and (b)  $K_{eff}$  of the corresponding bilayer film. The  $J_c^2$ , average values of  $J_c^2_{AP-P}$  and  $J_c^2_{P-AP}$  at H = 0, was estimated by a linear extrapolation of the data in Fig. 4. 18 to H = 0.

#### 4.3.2. Jc of TbFe (1nm) / GdFeCo (9nm) memory bilayer

To achieve full reversal loop at specific bias magnetic field, the reduction of the

thickness of TbFe in the bilayer structure which lowers the  $K_{eff}$  of bilayer is considered to be necessary. Therefore, the bilayer structure of Tb<sub>16</sub>Fe<sub>84</sub> (1 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (9 nm) was used as a memory layer to check the STT switching feature in the CPP-GMR with a bilayer memory layer. Two typical *R-I* loops obtained in the CPP-GMR structure with TbFe (1 nm) / GdFeCo (9 nm) memory layer at low and high temperatures were shown in Fig. 4.20 (a) and (b).



(a) *R-I* loop at *H*=-568.3Oe and 28°C;
(b) *R-I* loop at *H*=-568.3Oe and 83°C
Fig. 4.20 *R-I* loops measured for the CPP-GMR with TbFe (1 nm) / GdFeCo (9 nm) memory layer at different temperatures at a bias magnetic field-568.3 Oe.

By reducing the thickness of TbFe layer to 1 nm, both AP–P and P–AP reversals of the memory layer were achieved in a certain bias magnetic field. The average current density  $J_c$  estimated for the bilayer structure of TbFe (1 nm) / GdFeCo (9 nm) was slightly larger than that obtained in the CPP-GMR with GdFeCo single memory layer. The increase of the temperature was confirmed to be effective to reduce  $J_c$  as shown in Fig. 4.20 (b). Fig. 4.21 shows temperature dependences of (a) average  $J_c$  measured for the CPP-GMR structure with TbFe (1 nm) / GdFeCo (9 nm) memory layer and (b)  $K_{eff}$  measured for the corresponding bilayer film.



(a) Temperature dependence of  $J_c$  (b) Temperature dependence of  $K_{eff}$ Fig. 4.21 Temperature dependences of (a) average  $J_c$  measured for the CPP-GMR structure with TbFe (1 nm) / GdFeCo (9 nm) memory layer and (b)  $K_{eff}$  measured for the corresponding bilayer film.

It is clearly shown that  $J_c$  reduced with temperature, which is similar to the reduction of  $K_{eff}$  with temperature. However, the reduction of  $J_c$  was 20% with temperature increase to 82°C while only 19%  $K_{eff}$  reduction was observed with the same temperature increase.

It should be useful to compare the STT switching feature in the single GdFeCo layer and the Tb<sub>16</sub>Fe<sub>84</sub> (1 nm) / Gd<sub>21</sub> (Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (9 nm) bilayer to verify the effect of introduction of TbFe layer onto the GdFeCo layer. Fig. 4.22 shows the dependence of  $J_c$  on the thickness of TbFe in the bilayer memory layer.



Fig. 4.22 Dependences of  $J_c$  and  $K_{eff}$  on the thickness of TbFe in the memory bilayer.

From the figure, the  $J_c$  is estimated to increase about 70 % and the  $K_{eff}$  with a 100 % enhancement by the introduction of 1 nm thick TbFe layer. These unequal gains reveal an intriguing feature for the effective increase of thermal stability factor  $\Delta$  while keeping low switching current density which is crucial to achieve high density STT-RAM.

#### 4.4. Conclusions

The STT switching of the CPP-GMR structures with TbFe / GdFeCo bilayer memory layer were discussed to keep the sufficiently large thermal stability factor even in the cell size for STT-RAM having a memory capacity higher than 10 Gb. The *R-I* loops of two bilayer structure of  $Tb_{16}Fe_{84}$  (5 nm) /  $Gd_{21}(Fe_{90}Co_{10})_{79}$  (5 nm) and Tb<sub>16</sub>Fe<sub>84</sub> (1 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (9 nm) were measured to estimate the  $J_c$  for switching the bilayer. Generally, the  $K_{\rm eff}$  of the bilayer increases with the thickness ratio of the TbFe to the GdFeCo. In the CPP-GMR with a bilayer having 1 nm-thick TbFe, the STT switching was confirmed to be dominant the current induced reversal. The  $J_c$  for the switching increased slightly compared to that of the CPP-GMR with single GdFeCo layer, while K<sub>eff</sub> of the bilayer became twice compared to the single layer. This suggests the bilayer structure is beneficial as a memory layer for STT-RAM beyond a few Giga bit capacity (corresponding to cell area of 120×180  $\rm nm^2$  and the  $K_{\rm eff}$  of 7.5  $\times$  10<sup>5</sup> erg/cc). In the CPP-GMR with a bilayer having 5 nm-thick TbFe, the switching characteristic changed to thermally assisted magnetic field switching. The reasons of the change of the switching mode are due to large  $K_{\rm eff}$  in the TbFe (5 nm) / GdFeCo (5 nm) and large damping constant expected in the TbFe layer. The current work achieved in the GMR structure with TbFe (1nm) / GdFeCo (9nm) memory bilayer shows advantage in the  $K_{eff}$  in the demonstration of Gb memory capacity but a challenge of further reducing the current density is still existed and needs further improvements.

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#### **Chapter 5 General conclusion**

This study centralizes the efforts on the study of thermally assisted STT switching of amorphous rare earth - transition metal (RE-TM) alloys with perpendicular magnetic anisotropy (PMA) which we believe it is a potential solution for STT-RAM with memory capacity of a few Gb. In the dissertation, two types of memory layer incorporated into the CPP-GMR structure were studied: 1) the single GdFeCo memory layer with perpendicular easy axis, and 2) TbFe / GdFeCo memory bilayer in which TbFe acts as high PMA and low Curie temperature layer exchange coupling with the GdFeCo layer. The bilayer structure was adopted to tailor the large PMA and low critical current density  $J_c$  for the STT switching of the memory bilayer.

In chapter 1, various non-volatile memories (NVM) such as flash memory, MRAM, STT-RAM and phase change memory (PCM) were reviewed in terms of technological advantages and current challenges. Moreover, the underlying principle of each NVM technology was briefly introduced. Particularly, the STT-RAM technologies: principle, developments, and prospects, were explained in detail. The magneto-resistance effects, including anisotropic magneto-resistance effect, giant magneto-resistance effect and tunnel magneto-resistance effect, have been briefly described. The underlying mechanism for STT-RAM, spin transfer torque switching phenomenon has been discussed in detail.

In chapter 2, the deposition and micro-fabrication of the current perpendicular to the plane - giant magneto-resistance (CPP-GMR) structure with GdFeCo and TbFe / GdFeCo memory layers were introduced. The GMR film stack is Si / Ta (10) / CuAl (150) / [Pd (1.6) / Co (0.4)]<sub>6</sub> / Co<sub>40</sub>Fe<sub>40</sub>B<sub>20</sub> (0.5) / Cu (3) / Gd<sub>x</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>100-x</sub> (10) / Cu (5) / Ta (5) (thickness is in nanometer). For the GMR films with the TbFe / GdFeCo memory layer, the memory layer replaced from GdFeCo single layer to TbFe / GdFeCo bilayers, and the reference layer [Co/Pd]<sub>6</sub> was also changed to TbFeCo to avoid the unwanted switching of the reference layer. CPP-GMR devices were micro-fabricated by electron-beam (EB) lithography and subsequent Ar<sup>+</sup> etching from an electron cyclotron resonance (ECR) plasma source. The measurement apparatuses and their basic principles were also explained in this chapter. The magnetic properties of the as-deposited films were characterized by alternating gradient magnetometer (AGM) and torque magnetometer. The magneto-resistive (MR) effect of the GMR devices was measured by four-point probe method. The STT switching property of these devices was characterized by the resistance measurement after applying current pulses.

In chapter 3, spin transfer torque switching of single amorphous GdFeCo with perpendicular magnetic anisotropy was investigated. From the characterization of magnetic properties of GdFeCo single layer with various Gd content, the films exhibit large PMA and two distinct compositional regions, rare earth (RE) rich and transition metal (TM) rich, were found. In TM rich samples, parallel coupling between memory and reference layers was confirmed while antiparallel coupling in RE rich samples. The magneto-resistance (MR) ratio of the CPP-GMR with GdFeCo was not so large, typically 0.1 %, which will be due to the low spin polarization at Fermi level of the GdFeCo memory layer and the high resistance of the amorphous GdFeCo. In spite of the small MR ratio, STT switching of GdFeCo for the 120  $\times$ 180 nm<sup>2</sup> cell with Gd content in the range of 21.4~24.1at.% were confirmed in their *R-I* curves, where external magnetic field was applied during the measurement to cancel the exchange coupling through the Cu spacer. The dependence of the critical current densities  $J_c$  of the STT switching from parallel (P) to antiparallel (AP) states and AP to P states on the external magnetic field, Hext, was also investigated, where "parallel" means the magnetization of memory and reference layers aligns parallel. Both current densities, J<sub>c P-AP</sub> and J<sub>c AP-P</sub>, depend on the external field, but their average value,  $(J_{c P-AP} + J_{c AP-P}) / 2$ , was almost independent of the field, which is estimated to be  $1.6 \times 10^7$  A/cm<sup>2</sup>. Considering the low MR ratio and thick GdFeCo memory layer (10 nm) in our samples, the obtained  $J_c$  of 1.6  $\times$  10<sup>7</sup> A/cm<sup>2</sup> is considered to be relatively low, which suggests the efficient STT switching by using GdFeCo. From the Gd compositional dependence of the average  $J_c$ , the current density  $J_c$  increased with increasing the Gd composition, which is considered to be mainly due to the increase of the effective perpendicular anisotropy in the range of Gd 21.4~24.1at. %. *R-I* loops of the CPP-GMR sample with  $Gd_{22}(Fe_{90}Co_{10})_{78}$  memory layer were measured at various temperatures to obtain the temperature dependence of  $J_c$ . The  $J_c$  gradually decreased by elevating temperature from room temperature (16°C) to 70°C. The similar trend was confirmed on  $K_{eff}$  of GdFeCo, which is believed to be responsible for the temperature dependence of  $J_c$ .

The  $K_{\text{eff}}$  of the single GdFeCo memory layer discussed in chapter 3 is not sufficiently large for the STT-RAM with memory capacity of a few Gb. Thus, introduction of TbFe in the memory layer to enhance  $K_{\rm eff}$  of the GdFeCo was investigated in chapter 4. The [Co/Pd] reference layer was replaced by TbFeCo layer to avoid the unwanted current induced switching of the reference layer. In the STT switching process, K<sub>eff</sub> of TbFe will rapidly vanished when temperature is increased by Joule heating while the GdFeCo can be easily switched by spin polarized current. The dependence of the  $K_{\text{eff}}$  on temperature showed that the slope of the  $K_{\text{eff}}$ -T curve became sharp when the thickness of TbFe is increased, which is essentially beneficial for the thermally assisted STT switching. From the measurement of STT switching in the CPP-GMR with the bilayer having 1nm-thick TbFe, the STT switching was confirmed to dominate the current induced reversal of the memory layer. The  $J_c$  was increased 70% compared to that of the single  $Gd_{21}(Fe_{90}Co_{10})_{79}$ layer while  $K_{eff}$  became twice in Tb<sub>16</sub>Fe<sub>84</sub> (1 nm) / Gd<sub>21</sub>(Fe<sub>90</sub>Co<sub>10</sub>)<sub>79</sub> (9 nm) bilayer. Moreover, from temperature dependences of  $J_c$  and  $K_{eff}$  of this bilayer,  $J_c$  reduced 20% while  $K_{\rm eff}$  reduced 19% in the temperature range of room temperature (16°C) to 80°C. In bilayer with 5nm TbFe, the switching characteristic changed from the STT switching to the thermally assisted magnetic field switching due to the large anisotropy of TbFe and/or high damping constant expected in TbFe, which should be studied in detail in the future.

Finally, general conclusions of this dissertation are addressed here. In this dissertation, STT switching in GMR samples with GdFeCo single layers and TbFe / GdFeCo bilayers with various compositions and temperatures were studied. For the GdFeCo single memory layer, relatively low  $J_c$  of  $1.6 \times 10^7$  A/cm<sup>2</sup> was achieved considering the low MR ratio and the thick memory layer. The STT switching of the

TbFe / GdFeCo bilayer was successfully confirmed by this study. The bilayer structure was confirmed to be effective to increases  $K_{\text{eff}}$  while keeping  $J_c$  low, which is attractive to develop future high density STT-RAM. However, underlying mechanism of the present results should be unveiled by the future study. The bilayer structures with various compositions and thickness ratios should be studied in more detail, and the temperature and pulse width dependences of  $J_c$  should be investigated to open the route how to realize high density STT-RAM. Damping constants and gyromagnetic ratio of the bilayer, both of which are closely related with STT switching, and their temperature dependences should be also studied in detail.

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# List of publications

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