Fabrication of ferrimagnetic Co/Gd/Pt multilayers with structural inversion symmetry breaking

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We developed ferrimagnetic $[\text{Co}(0.5)/\text{Gd}(1)/\text{Pt}(1)]_N$ (unit: nm) multilayers with structural inversion symmetry breaking and investigated the dependence of magnetic properties and magnetic domain structures on the repetition number ($N = 1 - 50$). The magnetization compensation temperature increases as N increases, and saturates at around 210 K for $N \geq 20$. All films with various repetition number possess the out-of-plane magnetic easy axis, and multi domain structure at the remanence state was observed in the film with $N = 50$. These results show that the magnetic properties of the ferrimagnetic Co/Gd/Pt multilayers can be tuned by the repetition number.

Key words: ferrimagnets, structural inversion symmetry breaking, multilayers, magnetic properties, perpendicular magnetic anisotropy, thin film

1. Introduction

In rare earth (RE) - transition metal (TM) ferrimagnets, the magnetic moments of two inequivalent sublattices are antiferromagnetically coupled. Because RE and TM elements possess different temperature-dependences of spin density as well as different Landé g -factors, RE-TM ferrimagnets exhibit two special temperatures, the magnetization compensation temperature T_M and the angular momentum compensation temperature T_A . At T_M (T_A), RE and TM magnetic moments (angular moments) are canceled each

other, resulting in no net magnetic moment (angular moment) ¹⁻³). Because the net magnetization and the net angular momentum of the ferrimagnets are dependent on the temperature and the composition, many interesting studies have been reported recently; for example, ultra-high-speed magnetization switching faster than the time scale of the exchange interaction ⁴), all optical magnetization switching ⁵), current-induced dynamics of bubble domains near T_M ⁶) and fast domain wall motion due to antiferromagnetic spin dynamics at T_A ⁷). Therefore, RE-TM ferrimagnets are potential

Fig. 1 (a) Schematic image of $[\text{Co/Gd/Pt}]_N$ multilayers. (b) X-ray diffraction $2\theta/\theta$ patterns for $[\text{Co/Gd/Pt}]_N$ multilayers with various N ($N = 1, 5, 10$, and 30). (c) X-ray reflectivity patterns for $[\text{Co/Gd/Pt}]_N$ multilayers with various N ($N = 5, 10, 20, 30, 40$ and 50), respectively. The black lines are best fits.

candidates for the next generation of high-speed spintronic devices. Most studies on RE-TM ferrimagnets have focused on amorphous alloys, and their magnetic properties have been controlled mainly by tuning element composition⁸⁾⁻¹⁷⁾. However, recent studies have shown that breaking of the inversion symmetry leads to novel phenomena such as spin-orbit torque and Dzyaloshinskii-Moriya interaction^{18), 19)}, motivating us to explore ferrimagnets with structural inversion symmetry breaking. In this study, we prepared Co/Gd/Pt multilayers to develop RE-TM ferrimagnetic multilayers with structural inversion symmetry breaking, and investigated their structure and magnetic properties.

2. Sample fabrication and structural analysis

Figure 1(a) shows a schematic illustration of Co/Gd/Pt multilayers investigated in this study. The repetition of the sequence of Co/Gd/Pt breaks the inversion symmetry of the multilayers. The Pt/Co interfaces induce the interfacial perpendicular magnetic anisotropy (PMA)^{20), 21)}, and the interlayer antiferromagnetic exchange coupling at Co/Gd

interfaces^{22), 23)} results in ferrimagnetic nature of multilayers. Multilayers composed of Ta(5)/Pt(3)/[Co(0.5)/Gd(1)/Pt(1)]_N/Ta(3) (unit : nm) were deposited on thermally oxidized silicon substrates at room temperature by using direct current magnetron sputtering. Here, *N* denotes the repetition number of Co/Gd/Pt trilayers, and was varied from 1 to 50. Structures of the deposited films were identified by X-ray diffraction (XRD) using a conventional four-circle diffractometer. Figure 1(b) shows $2\theta/\theta$ XRD patterns of [Co/Gd/Pt]_N multilayers with *N* = 1, 5, 10, and 30. For all films, only reflections from Si substrate, buffer layer of Ta/Pt and oxidized capping Ta layer are observed, indicating no crystallization of [Co/Gd/Pt]_N multilayers. Figure 1(c) shows X-ray reflectivity (XRR) data of [Co/Gd/Pt]_N multilayers with *N* = 5, 10, 20, 30, 40, and 50 together with fitting lines by the software X'Pert Reflectivity (black solid lines). The procedure of the fitting is described in the next paragraph together with obtained parameters.

Figure 2(a) shows high-angle annular dark-field scanning transmission electron microscopy

Fig. 2 (a) High-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) images of [Co/Gd/Pt]₅ multilayer and each elemental map of the Co *L*_{2,3}-edge, of the Gd *M*_{4,5}-edge, of the Pt *M*_{4,5}-edge, and of the Ta *M*_{4,5}-edge obtained by electron energy-loss spectroscopy (EELS). (b) Intensity profiles of each element along the growth direction. (c) Intensity profile of Pt/Gd compositional ratio along the growth direction. The orange and red boxes are regions of Co and Ta layers, respectively.

(HAADF-STEM) image of $[\text{Co}/\text{Gd}/\text{Pt}]_5$ multilayer with each elemental map of the Co $L_{2,3}$ -edge, of the Gd $M_{4,5}$ -edge, of the Pt $M_{4,5}$ -edge, and of the Ta $M_{4,5}$ -edge obtained by electron energy-loss spectroscopy (EELS). The intensity profiles of each element along the growth direction are shown in Fig. 2(b), confirming that Gd and Pt are severely intermixed and form an alloyed layer. Based on this observation, we fit the XRR results by assuming that $[\text{Co}/\text{Gd}/\text{Pt}]$ multilayers are composed of Co and GdPt alloy layers, and the results are shown in Fig. 1(c). Table 1 shows thicknesses of Co and GdPt layers estimated from the fitting, indicating that the multilayers have the designed period of $[\text{Co}/\text{Gd}/\text{Pt}]$ unit. It should be noted that the Pt/Gd ratio is different between the lower and the upper part in each GdPt layer as shown in Fig. 2(c), indicating that the Pt/Gd ratio is not uniform in the GdPt layer and the composition gradually changes. Accordingly, the structural inversion symmetry breaking of the whole film still exists irrespective of the alloying of Gd and Pt.

Table 1 Summary of thicknesses of Co and GdPt layers estimated from XRR measurements. Target thicknesses of Co, Gd, and Pt layers are 0.5 nm, 1.0 nm, and 1.0 nm, respectively. (unit : nm)

N	Co	GdPt
5	0.52	1.96
10	0.54	1.99
20	0.53	1.99
30	0.52	2.00
40	0.53	1.99

3. Magnetic properties

Out-of-plane (OOP) and in-plane (IP) hysteresis loops of these films were also measured by SQUID magnetometer at 300 K. Typical OOP and IP hysteresis loops are shown in Fig. 3. It should be noted that the square hysteresis loops in OOP magnetic field were observed at small N (Fig. 3(a)), whereas the gradual magnetization reversal becomes noticeable as increasing N (Fig. 3(b)), indicating the effect of demagnetization field in the multilayer with larger N . M_s and the anisotropy field (H_K) were determined from OOP and IP hysteresis loops. As shown in Fig. 4(a), M_s decreases as N increases for small N owing to T_M being close to 300 K. The magnetic anisotropy energy K_{eff} ($= 1/2 M_s H_K$) as a function of the repetition number are calculated, and the results are shown in Fig. 4(b). Larger K_{eff} for smaller N suggests a strong influence from the interface between 3-nm-buffer Pt layer and 0.5-nm-Co layer^{24), 25)}.

Temperature dependences of the saturation magnetizations (M_s) in $[\text{Co}/\text{Gd}/\text{Pt}]_N$ multilayers were examined under an out-of-plane magnetic field of 200 mT using a superconducting quantum interference

device (SQUID) magnetometer. As shown in Fig. 5(a), the magnetization compensation temperatures T_M , where the Gd and Co magnetic moments are canceled

Fig. 3 Magnetic hysteresis loops of $[\text{Co}/\text{Gd}/\text{Pt}]_N$ multilayers under (a),(b) out-of-plane magnetic field and (c),(d) in-plane magnetic field. The repetition numbers are $N=3$ for (a),(c) and $N=30$ for (b),(d). The measurements were performed at 300 K.

Fig. 4 (a) The saturation magnetization M_s with respect to N . The black broken line is the best fit. (b) The magnetic anisotropy energy K_{eff} with respect to N .

each other, were observed for all films, confirming the ferrimagnetic property of these films. Figure 5(b) shows T_M with respect to N . T_M increases with N , and saturates at around 210 K for $N \geq 20$. We discuss this T_M dependence on N in the following.

The relationship between M_s and N of the $[\text{Co}/\text{Gd}/\text{Pt}]_N$ multilayers in which Co and Gd are coupled antiferromagnetically is written as

$$M_{s,\text{total}}(N) = \frac{M_{s,\text{Pt/Co}} + (N-1)M_{s,\text{Co}} - NM_{s,\text{Gd}}}{N}, \quad (1)$$

where $M_{s,\text{total}}(N)$ is the total saturation magnetization, $M_{s,\text{Pt/Co}}$ is the magnetization of Co with the buffer Pt layer, $M_{s,\text{Co}}$ is the magnetization of Co in $[\text{Co}/\text{Gd}/\text{Pt}]$ unit, and $M_{s,\text{Gd}}$ is the magnetization of Gd in $[\text{Co}/\text{Gd}/\text{Pt}]$ unit.

The temperature dependence $M_s(T)$ can be written as ^{7), 26)}

$$\begin{aligned} M_s(T) &= \frac{(M_{s,\text{Co/Pt}}(T) + (N-1)M_{s,\text{Co}}(T))}{N} - M_{s,\text{Gd}}(T) \\ &= \frac{(M_{s,\text{Pt/Co}}(0) + (N-1)M_{s,\text{Co}}(0))}{N} \left(1 - \frac{T}{T_C}\right)^{\beta_{\text{Co}}} \\ &\quad - M_{s,\text{Gd}}(0) \left(1 - \frac{T}{T_C}\right)^{\beta_{\text{Gd}}} \end{aligned} \quad (2)$$

Fig. 5 (a) Temperature dependence of M_s for $[\text{Co}/\text{Gd}/\text{Pt}]_N$ multilayers with $N = 5, 8, 10$, and 30 . (b) The magnetization compensation temperature T_M with respect to N . The black dots are calculated values.

with $M_{s,\text{Co/Pt}}(T) = M_{s,\text{Co/Pt}}(0) \left(1 - \frac{T}{T_C}\right)^{\beta_{\text{Co/Pt}}}$, $M_{s,\text{Co}}(T) = M_{s,\text{Co}}(0) \left(1 - \frac{T}{T_C}\right)^{\beta_{\text{Co}}}$, and $M_{s,\text{Gd}}(T) = M_{s,\text{Gd}}(0) \left(1 - \frac{T}{T_C}\right)^{\beta_{\text{Gd}}}$. Here, we assume $\beta_{\text{Pt/Co}} = \beta_{\text{Co}}$.

By fitting the experimentally obtained $M_s(T)$ with Eq. (2), we obtain $\frac{(M_{s,\text{Pt/Co}}(0) + (N-1)M_{s,\text{Co}}(0))}{N} = 1.40 \pm 0.01$ MA/m², $M_{s,\text{Gd}}(0) = 1.57 \pm 0.02$ MA/m², $\beta_{\text{Co}} = 0.501 \pm 0.002$, and $\beta_{\text{Gd}} = 0.700 \pm 0.002$. $M_{s,\text{Gd}}(300\text{K}) = 0.73 \pm 0.01$ MA/m² is calculated using these parameters. Then, we fit the experimentally obtained M_s dependence on N by Eq. (1) with two fitting parameters of $M_{s,\text{Co/Pt}}(300\text{K})$ and $M_{s,\text{Co}}(300\text{K})$. The black broken line in Fig. 4(a) is the best fit, and $M_{s,\text{Pt/Co}}(300\text{K}) = 0.93 \pm 0.02$ MA/m², $M_{s,\text{Co}}(300\text{K}) = 0.81 \pm 0.01$ MA/m² are determined.

In addition, T_M as a function of N can be deduced from Eq. (2) as

$$\begin{aligned} T_M = -T_C \left[\frac{1}{M_{s,\text{Gd}}(0)} \left\{ \frac{1}{N} (M_{s,\text{Pt/Co}}(0) - M_{s,\text{Co}}(0)) \right\} \right. \\ \left. + M_{s,\text{Co}}(0) \right]^{\frac{1}{\beta_{\text{Gd}} - \beta_{\text{Co}}}} - 1 \end{aligned} \quad (3)$$

Calculated T_M by Eq. (3) using the determined parameters by the fittings described above are plotted as black dots in Fig. 5(b), which reproduces the experimental trend that T_M increases with N , and saturates for $N \geq 20$. These results indicate that both M_s and T_M dependences on N can be explained by the simple idea that only the Co layer neighboring the buffer Pt layer has different magnetization from other Co layers.

To understand the relationship between the magnetic properties and the domain structure, the OOP hysteresis loop and the magnetic domain images of $[\text{Co}/\text{Gd}/\text{Pt}]_{50}$ multilayers were observed using magneto-optical Kerr effect (MOKE) microscope at room temperature. Figure 6(a) shows the OOP hysteresis loop of $[\text{Co}/\text{Gd}/\text{Pt}]_{50}$ multilayers. The magnetic domain images of $[\text{Co}/\text{Gd}/\text{Pt}]_{50}$ multilayers under the corresponding OOP magnetic field are shown in Figs. 6(b) – (f). The gradual reversal in the hysteresis loop was attributed to the formation of multi-domain structure to reduce the magnetostatic energy due to the strong demagnetization field in the multilayer with large N .

4. Conclusion

In summary, we developed the ferrimagnetic $[\text{Co}(0.5)/\text{Gd}(1)/\text{Pt}(1)]_N$ (unit : nm) multilayers and investigated the dependence of magnetic properties on the repetition number ($N = 1 - 50$). Our results show that the magnetic properties in the multilayered films can be controlled by the repetition number. Since the ferrimagnetic multilayers developed in this study inherit the inversion symmetry breaking, they are promising candidates for the investigation of novel phenomena such as Dzyaloshinskii-Moriya interaction (DMI) [27 - 29], spin orbit torque (SOT) [30, 31], and skyrmion motion [32-34]. Combination of those phenomena and

Fig. 6 (a) The out-of-plane hysteresis loop of $[\text{Co}/\text{Gd}/\text{Pt}]_{50}$ multilayers measured by Magneto-optical Kerr effect (MOKE) microscope at room temperature. (b) – (f) Magnetic domain images of $[\text{Co}/\text{Gd}/\text{Pt}]_{50}$ multilayers obtained at the corresponding magnitude of magnetic field.

novel properties of ferrimagnets will open a new avenue in the field of spintronics.

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