Magnetic moments of Fe site in $ThMn_{12}$-type $Sm(Fe_{1-x}Co_x)_{12}$ compounds and their temperature dependence

With increasing demand for "green technology" to reduce CO_2 emissions, the development of highly efficient electric motors and generators is required. For this, high-performance permanent magnets, especially for traction motors of EVs, HEVs, and turbines of wind power generation, are necessary. Because the working temperature of EV/HEV motors is approximately 150°C, a high coercivity ($\mu_0 H_c$) larger than 0.8 T is necessary at this temperature. Although Nd–Fe–B is the most important permanent magnet material for "green technology," one drawback is its low thermal resistance to $\mu_0 H_c$ owing to its low Curie temperature (T_c) [1]. Therefore, there is a strong demand for the development of new permanent magnets whose magnetic properties are superior to those of Nd–Fe–B, especially at elevated temperatures. One of the candidate materials for the next-generation permanent magnets is an RT_{12}-based compound with a $ThMn_{12}$ structure, where R and T represent rare-earth and transition elements, respectively. The RT_{12}-based compound is expected to show high saturation magnetization (M_s) owing to its highest Fe composition among rare-earth magnet materials. In addition to the high M_s, it has high Curie temperature (T_c) and high anisotropy (K).

Hirayama *et al.* reported that $Sm(Fe_{0.8}Co_{0.2})_{12}$ has the intrinsic magnetic properties of M_s~1.78 T, anisotropy field of 12 T and T_c~859 K, which are superior to those of $Nd_2Fe_{14}B$ [2]. Thus, $Sm(Fe_{0.8}Co_{0.2})_{12}$ is attracting attention as a new permanent magnetic material. Since these magnets are used in various applications at various temperatures, this study was conducted to clarify the internal magnetic field of $Sm(Fe_{0.8}Co_{0.2})_{12}$ thin films and its temperature dependence.

Figure 1 shows (a) magnetization curves, (b) the temperature dependence of the magnetization and (c) the temperature dependence of the normalized magnetization in $Sm(Fe_{1-x}Co_x)_{12}$ (x = 0, 0.07, 0.2) thin films. All samples show strong perpendicular anisotropy. Saturation magnetization and Curie temperature increase with Co content. The thermal resistance in the saturation magnetization is the highest in the sample with x = 0.2. Figure 2 shows the (a) Mössbauer spectrum and fitting curves, and (b) temperature and (c) Co composition dependences of the Mössbauer spectrum of $SmFe_{12}$ thin films at 300 K. The ^{57}Fe Mössbauer spectra were measured in the temperature range of 80–523 K at SPring-8 **BL11XU**. The Mössbauer spectra were nicely fitted by three different Fe sites of 8i, 8j and 8f. The disappearance of 2nd and 5th absorptions corresponds to the strong (001) texture of $SmFe_{12}$, which shows good agreement with the XRD result [3]. The fitting parameters are summarized in the inset. The hyperfine field decreases with increasing temperature and increases with Co content. Figure 3 shows the temperature dependence of (a) average and (b) normalized magnetic moments in $Sm(Fe_{1-x}Co_x)_{12}$ (x = 0, 0.07, 0.2) thin films. These temperature dependences well match with those of the magnetization of the sample shown in Fig. 1. Figure 3(c) shows the change in hyperfine field as a function of the Fe–Fe distance. The Fe–Fe distance was estimated by XRD analysis. The magnetic moment of Fe linearly increases with the Fe–Fe distance, and the effect is similar for all Co

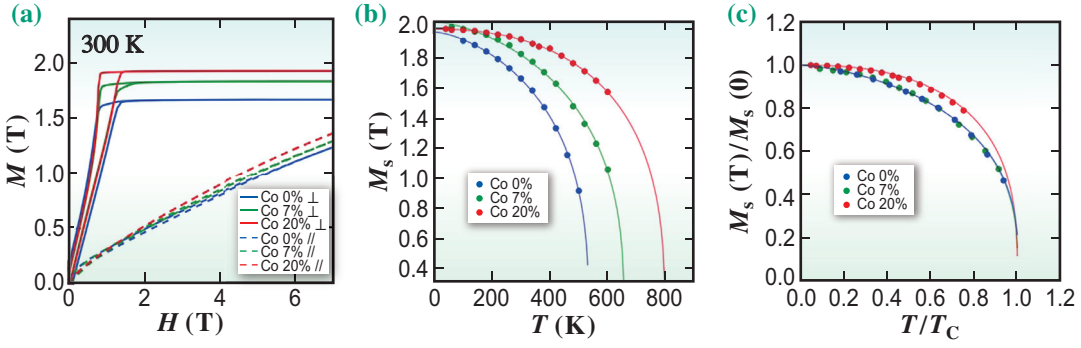
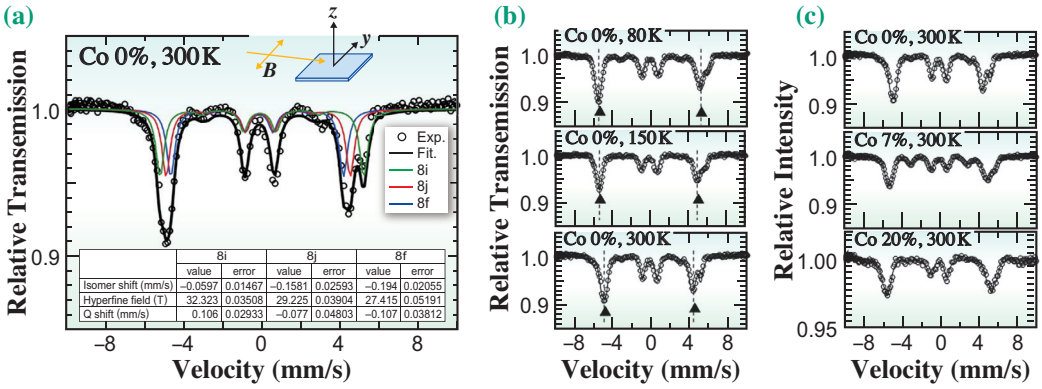


Fig. 1. **(a)** Magnetization curves of $Sm(Fe_{1-x}Co_x)_{12}$ thin films (x = 0, 0.07, 0.2) measured perpendicular and parallel to film plane at 300 K, **(b)** temperature dependence of spontaneous magnetization, $\mu_0 M_s$, and **(c)** normalized saturation magnetization $\mu_0 M_s/\mu_0 M_s(0)$ *versus* normalized temperature T/T_C of $Sm(Fe_{1-x}Co_x)_{12}$ (x = 0, 0.07, 0.2) thin films. [3]



	8i		8j		8f	
	value	error	value	error	value	error
Isomer shift (mm/s)	−0.0597	0.01467	−0.1581	0.02593	−0.194	0.02055
Hyperfine field (T)	32.323	0.03508	29.225	0.03904	27.415	0.05191
Q shift (mm/s)	0.106	0.02933	−0.077	0.04803	−0.107	0.03812

Fig. 2. **(a)** Mössbauer spectrum of $SmFe_{12}$ thin films measured at 300 K with fitting curves using three independent spectra for 8i, 8j, and 8f Fe sites, **(b)** temperature dependence of Mössbauer spectrum of $SmFe_{12}$ thin films, and **(c)** Co content dependence of Mössbauer spectrum of $Sm(Fe_{1-x}Co_x)_{12}$ thin films at 300 K. The dash line and arrows in (b) show the peak positions of 1st and 6th lines at each temperature. [3]

contents; the interdependence of the Fe–Fe distance and Fe magnetic moment is caused by the magneto-volume effect associated with *d*-orbital electrons. The increase in Fe magnetic moment upon Co addition is independent of the Fe–Fe interstitial distance and is similar to the trend shown in Fig. 1.

In conclusion, we first investigated the dependences of temperature and Co composition on the magnetic moment of crystallographically different Fe sites in $Sm(Fe_{1-x}Co_x)_{12}$ (x = 0, 0.07, 0.2) thin films by *in situ* synchrotron Mössbauer spectroscopy. The microscopic origin of the magnetism and temperature dependence of $Sm(Fe_{1-x}Co_x)_{12}$ (x = 0, 0.07, 0.2) thin films is clarified by experiments and theoretical calculations.

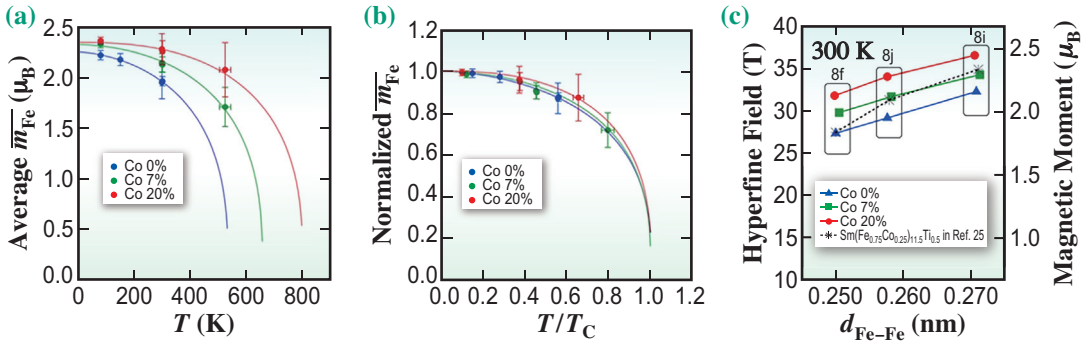


Fig. 3. **(a)** Temperature dependence of magnetic moments at various Co contents and **(b)** normalized magnetic moments at various Co contents site *versus* normalized temperature for $Sm(Fe_{1-x}Co_x)_{12}$ thin films (x = 0, 0.07, 0.2). **(c)** Calculated results of normalized magnetic moments at each Fe site *versus* reduced temperature for $Sm(Fe_{1-x}Co_x)_{12}$ compounds (x = 0, 0.07, 0.2).

Yukiko K. Takahashi

National Institute for Materials Science

Email: TAKAHASHI.Yukiko@nims.go.jp

References

[1] M. Sagawa *et al.*: J. Appl. Phys. **55** (1984) 2083.
[2] Y. Hirayama *et al.*: Scr. Mater. **138** (2017) 62.
[3] *D. Ogawa, T. Fukazawa, S. Li, T. Ueno, S. Sakai, T. Mitsui, T. Miyake, S. Okamoto, S. Hirosawa, Y. K. Takahashi: J. Magn. Magn. Mater.* ***552*** *(2022) 169188.*