京都大学
KYOTO UNIVERSITY

Title	Exchange-coupling of c-axis oriented L1[sub 0]–FePd and Fe in FePd/Fe thin films
Author(s)	Ichitsubo, T.; Takashima, S.; Matsubara, E.; Tamada, Y.; Ono, T.
Citation	Applied Physics Letters (2010), 97(18)
Issue Date	2010-11
URL	http://hdl.handle.net/2433/134550
Right	© 2010 American Institute of Physics
Type	Journal Article
Textversion	publisher

Exchange-coupling of *c*-axis oriented *L*1₀−FePd and Fe in FePd/Fe thin films

T. Ichitsubo,[1,a)] S. Takashima,[1] E. Matsubara,[1] Y. Tamada,[2] and T. Ono[2]

[1]*Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501, Japan*
[2]*Division of Materials Chemistry, Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan*

This letter reports the control of the *c*-axis orientation and the magnetic properties of $L1_0$ FePd in the well-annealed FePd/Fe thin films Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂-substrate and Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂-substrate, with the compositions of dual phase region in phase equilibrium. After annealing at 550 °C for 24 h, the *c*-axis of $L1_0$ FePd is controlled to be oriented in the normal direction in the soft Fe matrix. These thin films exhibit the exchange-coupling of hard FePd with soft Fe, and a characteristic spring-magnet behavior showed up reversibly as a significant jump from positive to negative in the magnetization reversal. © *2010 American Institute of Physics*. [doi:10.1063/1.3514582]

Ferromagnetic $L1_0$ ordered alloys FePt, CoPt, and FePd, are under extensive investigation[1–6] for applications to perpendicular recording media because of their large uniaxial magnetocrystalline anisotropy of $K \sim 10^6$ J/m³, where the *c*-axis is the easy axis of magnetization.[7] However, for magnetic recording media applications, well-ordered $L1_0$ particles with the *c*-axis orientation in the normal direction in the thin film are required. To overcome this problem, a method to synthesize fully ordered FePt nanoparticles has recently been reported by Yamamoto *et al.* and Tamada *et al.*[4,5] In this method, high-melting-temperature SiO₂ covers the surface of isolated FePt nanoparticles prepared by Sun's method,[1] and subsequently the SiO₂ covered nanoparticles are annealed at a sufficiently high temperature for fully ordering reaction and extracting its high coercivity. The *c*-axis of FePt has been shown to be oriented in the normal direction of the film surface with the aid of B₂O₃ interlayers.[8,9] In our previous paper,[10] we tried to elucidate the mechanism of the *c*-axis orientation of FePt particles experimentally and phenomenologically, and proposed a mechanism of the *c*-axis orientation of $L1_0$ FePt or CoPt, in which the biaxial strain field due to the difference in the thermal contraction plays a significant role in making *c*-axis aligned in the normal direction of thin films. That is, this biaxial strain field can be regarded as one of the effects of applying an external field, which has been intensively investigated by the one of the present authors.[11–14] Here we deal with the well-ordered $L1_0$ FePd alloy embedded in a soft magnetic Fe matrix, which can be easily attained because the FePd/Fe dual-phase region exists in thermal equilibrium only in the Fe–Pd alloy system. The focus of this work is the control of the *c*-axis orientation and the occurrence of exchange-coupling of hard FePd with soft Fe. In this letter, we report the intriguing magnetic behavior of exchange-coupled FePd/Fe magnetic thin films.

Thin films were deposited on SiO₂ glass substrates by using a three-cathode magnetron sputtering apparatus with 2 in. diameter Fe and Pd targets; rf (13.56 MHz) sputter depositions were performed at room temperature in an Ar (purity: 99.99%) atmosphere of about 0.3 Pa, after the base pressure reached about 10^{-5} Pa. The distance between the cathodes and a substrate was set at 200 mm (as long as possible). We have prepared two types of thin films; Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂ and Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂, the Fe compositions of which are about 80–90 at. %, being within the range of the dual phase region of $L1_0$−FePd/α-Fe in phase equilibrium. After the depositions, to attain a fully ordered structure of FePd without oxidation, the thin films were annealed at 550 °C for 24 h under H₂ gas flow. The crystallographic orientation of the post-annealed thin films was analyzed by x-ray diffraction (XRD) using Cr $K\alpha$ radiation and the surface topographic morphology was observed by atomic-force microscope (AFM). A superconducting quantum interference device (SQUID: Quantum Design) with a field of up to 30 kOe (3 T) was used to measure the hysteresis curves at room temperature.

Figure 1 shows the microstructure by AFM and XRD profile for each thin film after annealing. As seen in the AFM photographs, the particles of about 100 nm appear aggregated. According to the XRD profiles, in both cases, the *c*-axis is well oriented in the normal direction of the film surface. Interestingly, it is also found that the crystallographic [200] axis of Fe is cooperatively oriented along the normal direction. This was the typical feature when the *c*-axis of FePd can be along the normal direction, but in contrast, the [110] axis of Fe is oriented in the normal direction when the [111] axis of FePd is oriented in the normal direction. Furthermore, it should be noted that the tetragonal distortion is very much enhanced, especially for the Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂ film; the axial ratios, 0.93 in the former and 0.87 in the latter film, were considerably smaller than usual (about 0.96). The reason for the *c*-axis orientation and the small axial ratios can be understood in the light of the thermal-contraction effect proposed by the previous paper.[10] The thermal expansion coefficient of SiO₂ is quite small (of the order 10^{-7} 1/K), compared to that of metals (of the order 10^{-5} 1/K). Since the SiO₂ substrate temperature could be increased during the sputter-deposition, the deposited FePd/Fe thin film is considered to be in a
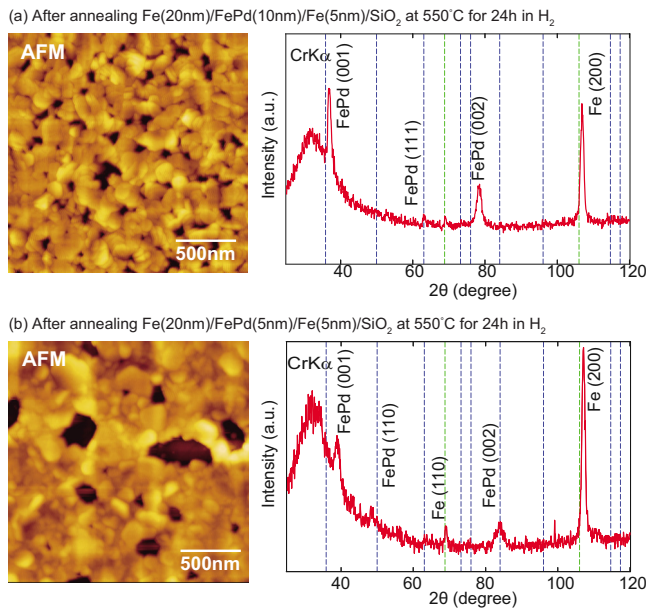
a)Electronic mail: tichi@mtl.kyoto-u.ac.jp.

(a) After annealing Fe(20nm)/FePd(10nm)/Fe(5nm)/SiO₂ at 550˚C for 24h in H₂

(b) After annealing Fe(20nm)/FePd(5nm)/Fe(5nm)/SiO₂ at 550˚C for 24h in H₂

FIG. 1. (Color online) AFM microstructures and XRD profiles of the films of Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂ and Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂. The dashed lines are also drawn referring each powder diffraction file, Fe: no. 06-0696 and $L1_0$-FePd: no. 02-1440.

largely biaxial-tensile strained state. Thus, under such a condition, the c-axis of $L1_0$ FePd comes to be aligned along the normal direction of the film surface.
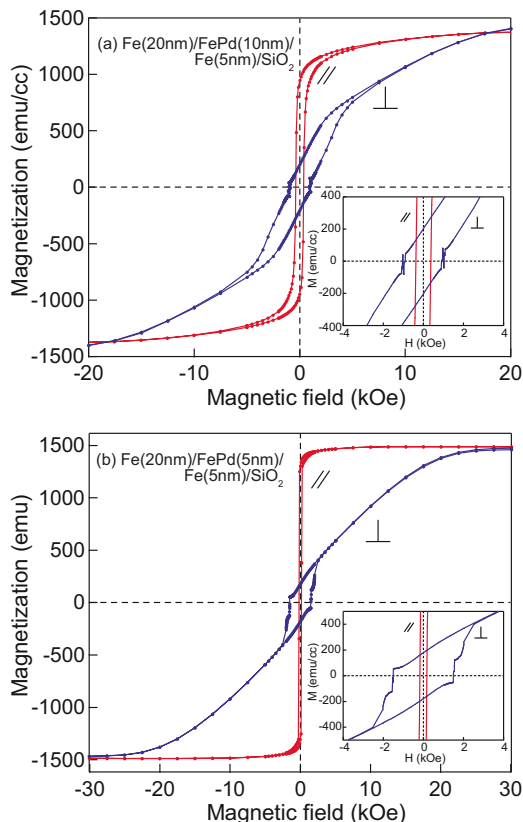
Figure 2 shows the magnetization-field (M-H) curves

FIG. 2. (Color online) The magnetization-field (M-H) curves obtained for the films of Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂ and Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂. The symbols ∥ and ⊥ mean, respectively, "in-plane" and "out-of–plane" directions of applying the magnetic field during the M-H curve measurement.
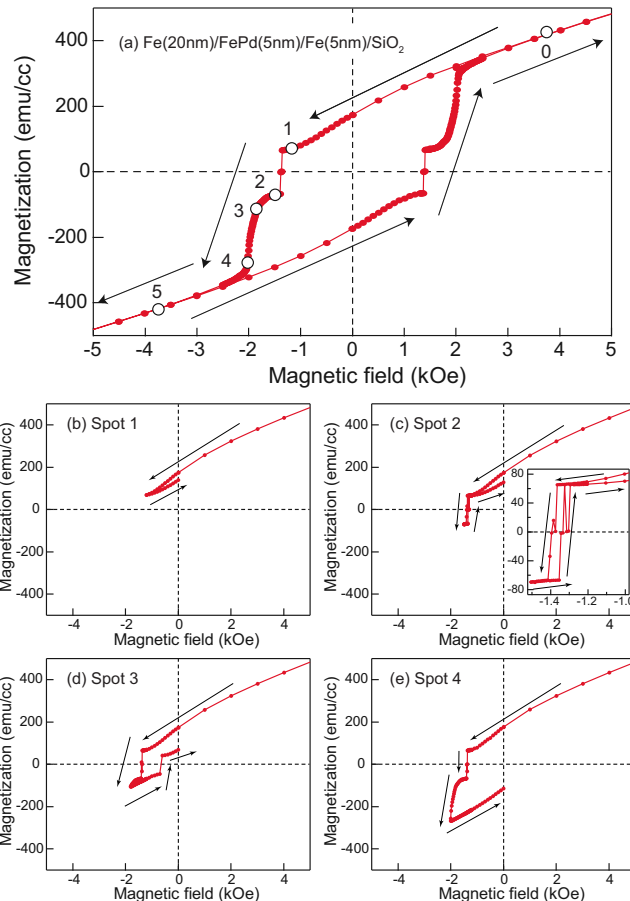

FIG. 3. (Color online) The detailed magnetization-field (M-H) curves for the film of Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂, in which the measurement has been done along the arrows in (a). The figures [(b)–(e)] display respective M-H curves which were measured down to the numbered spot in (a) and subsequently up to 0 T (after the measurement, it returned back to the initial magnetic field, 30 kOe) to obtain each segment of the whole M-H curve in (a).

obtained for (a) Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂ and (b) Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂. In both curves, we can clearly see there is fairly large magnetic anisotropy, taking account of the fact that the films contain mainly Fe. This anisotropy is considered to be caused by the well oriented FePd particles in the film. The coercivity $H_{c\perp}$ of each film is about 1 kOe for Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂ and 1.5 kOe for Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂. Most importantly, we observe that there are jumps (gaps) in each M-H curve when the magnetization turns to be reversed in both films. For Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO₂, the jump is smaller, but for Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂ the jump is apparently large. We confirmed that this was not measurement error by checking that the magnetization never became zero during the magnetization flipping (the current induced by passing through the three solenoidal SQUID coils was appreciably large).

In order to observe this magnetization flipping behavior more in detail, we have further examined the segmental M-H curves for Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO₂. Figure 3 shows the magnetization processes of the film and Fig. 4 illustrates the conjecture of the magnetic reversal processes. As shown in Fig. 4, most of the magnetization process is dominated by the planar shape of soft Fe (which is magneti-
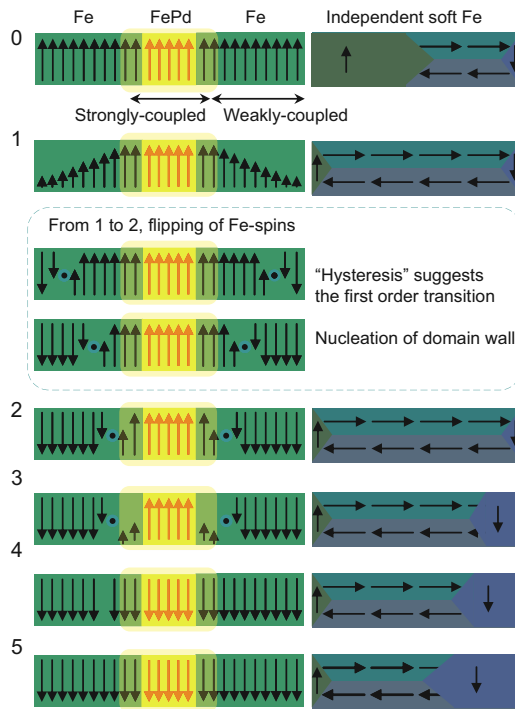
FIG. 4. (Color online) Schematic illustration showing the magnetization processes of the spin-flipping of Fe that is weakly exchange-coupled with the *c*-axis oriented FePd, and the magnetically independent soft Fe. Each number corresponds to the numbered in the M-H curve in Fig. 3. The hysteresis shown in the spot 2 of Fig. 3 strongly indicates the nucleation of the domain walls. In most of soft Fe regions, the magnetization is done by the motion of the domain walls.

cally independent, it is done by the motion of the domain walls), but our focus here is on the magnetization of *c*-axis oriented FePd coupled with soft Fe. The magnetization process up to spot 1 is almost reversible; there the downward convex shape seems to be a characteristic feature of the exchange coupling of hard FePd and soft Fe, as seen in the Hard/Soft thin films.[15] It is interesting to see that the magnetization process is almost reversible even when the magnetization flipping occurs at spot 2 but a slightly small hysteresis is present in the backward process. This hysteresis means the transition of the magnetization reversal is first order, suggesting the nucleation of the domain wall, as illustrated in Fig. 4. The flipping could be due to the magnetization of Fe, which may be caused when the Zeeman energy under a relatively strong magnetic field exceeds the exchange-coupling energy of FePd with Fe. Thus by removing the external field, the domain wall would disappear, returning the system to the exchange-coupling state. From positions 3 to 4, the magnetization of hard FePd begins to be reversed by following an

increasing external magnetic field. At spot 4, the magnetization reversal of FePd has been completed, and hereafter the domain-wall motion in the planar Fe would enhance the magnetization of the film. The spring magnetic property like this is very rare and not well known to date. Thus, we expect it to be applied to switching or magnetic resistance devices, etc.[15]

In summary, we have investigated the structure and magnetic properties of FePd/Fe thin films in the dual phase region in thermal equilibrium. The present thin films consist of the soft/hard magnetic regions, and the thin films show the controlled *c*-axis orientation of FePd in the soft Fe matrix. The obtained M-H curves can be interpreted by considering the weak magnetic-exchange coupling of hard FePd with soft Fe. When an external magnetic field is large, the weak coupling could be destroyed and the domain walls nucleated; as a result, the magnetization reversal from positive to negative (and vice versa) is observed reversibly as a significant jump, i.e., as a remarkable spring-magnet behavior.

[1]S. Sun, C. B. Murray, D. Weller, L. Folks, and A. Moser, Science **287**, 1989 (2000).
[2]D. H. Ping, M. Ohnuma, K. Hono, M. Watanabe, T. Iwasa, and T. Masumoto, J. Appl. Phys. **90**, 4708 (2001).
[3]T. Shima, K. Takanashi, Y. K. Takahashi, and K. Hono, Appl. Phys. Lett. **85**, 2571 (2004).
[4]S. Yamamoto, Y. Morimoto, T. Ono, and M. Takano, Appl. Phys. Lett. **87**, 032503 (2005).
[5]Y. Tamada, S. Yamamoto, S. Nasu, and T. Ono, Phys. Rev. B **78**, 214428 (2008).
[6]Y. Tamada, S. Yamamoto, S. Nasu, and T. Ono, Appl. Phys. Express **2**, 123001 (2009).
[7]T. Klemmer, D. Hoydick, H. Okumura, B. Zhang, and W. A. Soffa, Scr. Metall. Mater. **33**, 1793 (1995).
[8]C. P. Luo, S. H. Liou, L. Gao, Y. Liu, and D. J. Sellmyer, Appl. Phys. Lett. **77**, 2225 (2000).
[9]M. L. Yan, H. Zeng, N. Powers, and D. J. Sellmyer, J. Appl. Phys. **91**, 8471 (2002).
[10]T. Ichitsubo, S. Tojo, T. Uchihara, E. Matsubara, A. Fujita, K. Takahashi, and K. Watanabe, Phys. Rev. B **77**, 094114 (2008).
[11]K. Tanaka, T. Ichitsubo, M. Amano, M. Koiwa, and K. Watanabe, Mater. Trans., JIM **41**, 917 (2000).
[12]T. Ichitsubo, M. Nakamoto, K. Tanaka, and M. Koiwa Mater. Trans., JIM **39**, 24 (1998).
[13]T. Ichitsubo, K. Tanaka, M. Koiwa, and Y. Yamazaki, Phys. Rev. B **62**, 5435 (2000).
[14]K. Tanaka, T. Ichitsubo, and M. Koiwa, Mater. Sci. Eng., A **312**, 118 (2001).
[15]K. Mibu, T, Nagahama, T. Shinjo, and T. Ono, Phys. Rev. B **58**, 6442 (1998).