<table>
<thead>
<tr>
<th><strong>Title</strong></th>
<th>Exchange-coupling of c-axis oriented L1₂–FePd and Fe in FePd/Fe thin films</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Author(s)</strong></td>
<td>Ichitsubo, T.; Takashima, S.; Matsubara, E.; Tamada, Y.; Ono, T.</td>
</tr>
<tr>
<td><strong>Citation</strong></td>
<td>Applied Physics Letters (2010), 97(18)</td>
</tr>
<tr>
<td><strong>Issue Date</strong></td>
<td>2010-11</td>
</tr>
<tr>
<td><strong>URL</strong></td>
<td><a href="http://hdl.handle.net/2433/134550">http://hdl.handle.net/2433/134550</a></td>
</tr>
<tr>
<td><strong>Right</strong></td>
<td>© 2010 American Institute of Physics</td>
</tr>
<tr>
<td><strong>Type</strong></td>
<td>Journal Article</td>
</tr>
<tr>
<td><strong>Textversion</strong></td>
<td>publisher</td>
</tr>
</tbody>
</table>

Kyoto University
Exchange-coupling of c-axis oriented $L1_0$–FePd and Fe in FePd/Fe thin films

T. Ichitsubo,1,a) S. Takashima,1 E. Matsubara,1 Y. Tamada,2 and T. Ono2
1Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501, Japan
2Division of Materials Chemistry, Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan

(Received 14 September 2010; accepted 21 October 2010; published online 5 November 2010)

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 investigation1–6 for applications to perpendicular recording media because of their large uniaxial magnetocrystalline anisotropy of $K \approx 10^6$ 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 been reported by Yamamoto et al. and Tamada et al.3,5 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 FePd has been shown to be oriented in the normal direction of the film surface with the aid of FePt layer interlayers.3,9 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 perpendicular recording media because of their large uniaxial magnetocrystalline anisotropy of $K \approx 10^6$ 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 been recently reported by Yamamoto et al. and Tamada et al.3,5 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 FePd has been shown to be oriented in the normal direction of the film surface with the aid of FePt layer interlayers.3,9 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 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 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 Kα 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 $10^{-3}$ 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
largely biaxial-tensile strained state. Thus, under such a condition, the c–axis of $L_1$ FePd comes to be aligned along the normal direction of the film surface. Figure 2 shows the magnetization-field (M–H) curves obtained for (a) Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO$_2$ and (b) Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO$_2$. 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 $H_c$ of each film is about 1 kOe for Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO$_2$ and 1.5 kOe for Fe(20 nm)/FePd(5 nm)/Fe(5 nm)/SiO$_2$. 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)/SiO$_2$, the jump is apparently large. We confirmed that this was not measurement error by checking that the magnetization never became zero during the magnetization process (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)/SiO$_2$. 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-
As the external field is reduced, the magnetization of FePd begins to be reversed by following an domain wall would disappear, returning the system to the state of FePd with Fe. Thus by removing the external field, the domain wall would disappear, returning the system to the state of FePd with 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.

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.

This work was partly supported by Grant-in-Aid for “Wakate Kenkyu A (Grant No. 20686043)” from JSPS, Japan. One of the authors, T.I., was grateful to Ms. Niedziela, Jennifer Lynn, for reading and correcting our manuscript.