Anomalous Hall Effect and Anisotropic Magnetoresistance in Perpendicularly Magnetized FePt$_{1-x}$Pd$_x$ Films

T. Soki, S. Kikushima, and K. Takanashi

‘Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
‘Center for Spintronics Research Network, Tohoku University, Sendai 980-8577, Japan

$L1_0$-ordered alloys with high uniaxial magnetic anisotropy ($K_u$) are key materials for future spintronic devices to realize large-scale integration. In this study, we investigated the anomalous Hall effect (AHE) and anisotropic magnetoresistance (AMR) effect for FePt$_{1-x}$Pd$_x$ (001) epitaxial films. Highly $L1_0$-ordered structures and perpendicular magnetization were obtained for all the compositions. The FePt film exhibited the highest $K_u$ and the largest anomalous Hall angle ($\theta_{AHE}$) among the present FePt$_{1-x}$Pd$_x$ samples, and the values of $K_u$ and $\theta_{AHE}$ were gradually decreased as the Pd concentration increased. In addition, the composition dependence of the AMR ratio showed a similar tendency to those for $K_u$ and AHE. We discuss possible scenarios to explain these composition dependencies.

Key words: $L1_0$-type ordered alloy, anomalous Hall effect, anisotropic magnetoresistance effect

1. Introduction

For the further progress in magnetic storage technology, we need to tackle several essential problems every magnetic material has. One of the inevitable problems is the thermal instability of magnetization appearing as the volume of magnet (V) is reduced. In order to hurdle this problem, high uniaxial magnetic anisotropy ($K_u$) is needed because the thermal stability factor ($\Delta$) of magnetization is given by $\Delta = K_u V / k_B T$, where $k_B$ is the Boltzmann constant and $T$ is the temperature. Thus, high $K_u$ materials are key to maintain sufficient $\Delta$ even for a nanometer-sized magnet, enabling us to realize the large-scale integration of spintronic devices.

$L1_0$-ordered alloys are representative high $K_u$ materials. Thanks to their high $K_u$ along the c-axis of $L1_0$-ordered structure, perpendicularly magnetized layers can be formed when these alloys are grown with the (001) crystal orientation in the thin film form. This characteristic is beneficial for perpendicular magnetic recording and spintronic devices utilizing perpendicular magnetization as a perpendicular spin-polarizer. Among the $L1_0$-ordered alloys, $L1_0$-FePt and $L1_0$-FePd are famous alloys, and are regarded as candidate materials for ultrahigh density magnetic storage. Although both alloys have similar characteristics, such as high $K_u$ values and structures being composed of Fe and Pt or Pd with large spin-orbit interaction, there are also definite differences between them. The most intriguing difference is the magnitude of magnetization damping. $L1_0$-FePd exhibits a much smaller damping constant than that of $L1_0$-FePt. Although that might be partially interpreted by the smaller spin orbit interaction of Pd than that of Pt, the relationship between the transport properties and the spin orbit interaction has not been totally understood yet for the Fe-Pt-Pd system.

In this study, we focus on the anomalous Hall effect (AHE) and anisotropic magnetoresistance (AMR) effect. The magnitude of AHE is characterized by the anomalous Hall angle ($\theta_{AHE}$). The spin-orbit interaction is regarded as an essential factor for AHE, and several mechanisms, such as skew scattering, side jump and intrinsic deflection, are responsible for the appearance of AHE. The expression of AMR also includes the spin-orbit coupling constant. Namely, both AHE and AMR are phenomena related with the spin-orbit interaction although they are often discussed in the different theoretical frameworks.

We prepared FePt$_{1-x}$Pd$_x$ (001) epitaxial films and systematically investigated the structures, magnetic properties and transport properties. The Pd concentration (x) dependencies of $K_u$, AHE and AMR are compared, and possible scenarios to understand those composition dependences are qualitatively discussed.

2. Experimental Procedure

50 nm-thick FePt$_{1-x}$Pd$_x$ layers were directly grown on an MgO (001) single crystal substrate by employing the ultrahigh vacuum compatible magnetron sputtering apparatus. The base pressure of main growth chamber was below $2 \times 10^{-7}$ Pa. The main growth chamber was equipped with three independent sputtering cathodes, where Fe, Pt, and Pd targets were installed. This cathode arrangement enabled us to vary the alloy composition by tuning the input powers for sputtering cathodes. The growth temperature for the FePt$_{1-x}$Pd$_x$ layers was set at 500°C. This high temperature process is indispensable to promote the $L1_0$-ordering. We monitored the film growth by employing reflection high-energy electron diffraction, which exhibited streak patterns for all the samples. This indicated that flat surfaces were obtained. The thin films were patterned into a Hall cross shape in order to measure AHE and AMR through the use of photolithography and Ar ion milling. The width of Hall cross was designed to be 20
The structure was characterized using the x-ray diffraction (XRD) with Cu-Kα radiation. Magnetization measurements were carried out at room temperature using a superconducting quantum interference device magnetometer. Transport properties were measured at room temperature using a physical property measurement system (PPMS) with the maximum magnetic field of 90 kOe.

In this study, we set the Pd concentration $x$ in Pt$_{1-x}$Pd$_x$ to 0, 0.25, 0.5, 0.75, and 1, where $x \times 100$ corresponds to the atomic percent of Pd concentration. The actual alloy compositions were determined to be Fe$_{38}$Pt$_{62}$ (for $x =$ 0), Fe$_{38}$Pt$_{32}$Pd$_2$ (for $x =$ 0.25), Fe$_{38}$Pt$_{32}$Pd$_{25}$ (for $x =$ 0.5), Fe$_{53}$Pt$_{22}$Pd$_{25}$ (for $x =$ 0.75), and Fe$_{51}$Pd$_{49}$ (for $x =$ 1) by electron probe x-ray microanalysis. Hereafter, the designed Pd concentration $x$ refers to the sample name instead of the measured composition for simplicity.

3. Results and Discussion

First, we explain the structure and the magnetic properties for the FePt$_{1-x}$Pd$_x$ films. Figure 1 shows the XRD profiles for (a) $x =$ 0, (b) 0.25, (c) 0.5, (d) 0.75, and (e) 1. The unlabeled sharp peaks come from the reflections of the MgO substrate. All the peaks, denoted by the arrows, are 00l reflections of FePt$_{1-x}$Pd$_x$, which indicates that all the FePt$_{1-x}$Pd$_x$ layers show the (001) epitaxial growth on the MgO (001) substrate. In addition to the 002 and 004 fundamental peaks, the 001 and 003 superlattice peaks are clearly observed, suggesting the formation of $L1_0$-ordered structure.

The lattice constants of $c$-axis and $a$-axis, tetragonality ($c/a$), and the degree of long-range order ($S$) are summarized in Figs. 2 (a), (b), (c) and (d), respectively. The lattice constants of $c$-axis were obtained from the (002) peaks in the out-of-plane XRD profile (Fig. 1) whereas those of $a$-axis were obtained from the 200 peaks in the in-plane XRD profiles (not shown here). $S$ is defined as

$$S^2 = \frac{I_{\text{super}}}{I_{\text{fund}}},$$

where $I_{\text{fund}}$ and $I_{\text{super}}$ are the integrated intensities of fundamental and superlattice peaks, respectively, and the subscripts of obs and calc represent the experimentally observed values and the calculated values, respectively. The intensities for the fully ordered state ($S = 1$) were calculated taking into account atomic scattering factors, Debye-Waller factors, Lorentz factor, polarization factor, and structure factors.

![Fig. 1 Out-of-plane x-ray diffraction profiles for FePt$_{1-x}$Pd$_x$ films with (a) $x =$ 0, (b) 0.25, (c) 0.5, (d) 0.75, and (e) 1.](image-url)

![Fig. 2 Lattice constants of c-axis and a-axis, tetragonality (c/a), and the degree of long-range order (S) as a function of Pd concentration (x). Dashed lines are guides for eyes.](image-url)
Magnetization curves for FePt$_{1-x}$Pd$_x$ films with (a) $x = 0$, (b) 0.25, (c) 0.5, (d) 0.75, and (e) 1. Solid (dashed) curves are results with field ($H$) applied perpendicular (parallel) to the film plane.

In the cases of ternary alloys, the above factors for the Pt-Pd site were calculated assuming the averaged values with the atomic fractions of Pt and Pd.

The lattice constant of $c$-axis is gradually reduced as $x$ is increased while $a$-axis is slightly increased. Consequently, $c/a$ shows a slight decrease with $x$. The literature values$^{15}$ are $c = 0.3702$ nm and $a = 0.3842$ nm for FePt, and $c = 0.3715$ nm and $a = 0.3850$ nm for FePd, which lead to $c/a = 0.964$ for FePt and 0.965 for FePd. These reported values do not explain the Pd concentration dependence of lattice constants for the present FePt$_{1-x}$Pd$_x$ films. The epitaxial strain may be related with the changes in the lattice constants, which depends on the composition and the kind of substrate, although the detailed mechanism has not been understood yet. It is noted that the continuous change in the lattice constants suggests the formation of Fe-Pt-Pd ternary alloy without any phase separation.

From Fig. 2 (d), it is confirmed that highly ordered FePt$_{1-x}$Pd$_x$ layers are perpendicularly magnetized. Figure 3 shows the magnetization curves for (a) $x = 0$, (b) 0.25, (c) 0.5, (d) 0.75, and (e) 1, where the solid (dashed) curves are the results with field ($H$) applied perpendicular (parallel) to the film plane. One sees that the easy magnetization axis is in the normal direction to the film plane for all the films. Namely, all the FePt$_{1-x}$Pd$_x$ layers are perpendicularly magnetized. However, the saturation field in the in-plane magnetization curve is decreased as $x$ is increased. For example, for $x = 0$ (FePt), $H = 50$ kOe is insufficient to saturate the magnetization along the in-plane direction while the magnetization of $x = 1$ (FePd) saturates even at the in-plane $H = 25$ kOe. This indicates that the perpendicular magnetic anisotropy is reduced with $x$.

Figure 4 displays (a) saturation magnetization ($M_s$) and (b) $K_u$ as a function of $x$. The value of $K_u$ was calculated from the effective magnetic anisotropy ($K_{eff}$) with the correction of shape anisotropy energy ($2\pi M_s^2$), i.e. $K_u = K_{eff} + 2\pi M_s^2$. $K_{eff}$ was determined from the area enclosed between the perpendicular and in-plane magnetization curves. In Fig. 4(a), $M_s$ does not show the remarkable composition dependence. A previous paper$^{11}$ reported $M_s = 1140$ emu/cm$^3$ and 1100 emu/cm$^3$ for FePt and FePd, respectively, indicating that no remarkable difference in $M_s$ between FePt and FePd.

This explains why $M_s$ keeps almost constant regardless of $x$. On the other hand, $K_u$ shows the significant composition dependence. The largest $K_u$ among the present samples is obtained at $x = 0$, which is $K_u = 3.5 \times 10^7$ erg/cm$^3$, and $K_u$ is drastically reduced as $x$ is varied from 0 to 1. The larger $K_u$ for $x = 0$ than that for $x = 1$ is consistent with the fact that the bulk FePt shows the larger $K_u$ than the bulk FePd$^{11}$. Our finding is that $K_u$ is monotonically decreased with $x$ for the Fe-Pt-Pd ternary alloys and does not show a local minimum or maximum value.

Next, let us show the results of AHE measurement for the Hall devices with the FePt$_{1-x}$Pd$_x$ layers. Figure 5 (a) is the transverse resistance ($R_{xy}$) as a function of $H$ for $x = 0$, 0.5, and 1, where $H$ was applied along the perpendicular ($z$) direction to the device plane. The Hall resistivity ($\rho_{H}$) is composed of two terms$^{12}$:

$$\rho_{H} = R_{0} + R_{1}M(T,H).$$

where $M(T,H)$ is the magnetization averaged over the sample. $R_{0}$ and $R_{1}$ represent the ordinary and anomalous Hall coefficients, respectively. The shapes of $R_{xy} \cdot H$ coincide with those of the perpendicular magnetization curves. This means the $R_{xy}$ change originates from the AHE. Figures 5 (b), (c) and (d) summarize the longitudinal conductivity ($\sigma_{xx}$), the transverse conductivity ($\sigma_{xy}$), and the anomalous Hall conductivity as a function of $x$. $\theta_{H}$ is defined as $\theta_{H} = \sigma_{xy} / \sigma_{xx}$.

$\sigma_{xx}$ increases

$\sigma_{xx}$ increases
(decreases) as \(x\) is increased. As a result, \(x = 0\) exhibits the largest \(\theta_{\text{AHE}} = 0.04\), and \(\theta_{\text{AHE}}\) shows the monotonic reduction down to 0.01 at \(x = 1\). This tendency might be explained by the fact that FePt possesses the spin orbit interaction larger than FePd\(^{16}\).

Figure 6 displays the AMR effect for the FePt\(_1-x\)Pd\(_x\) layers. The longitudinal resistance \(R_{xx}\) versus \(H\) for \(x = 0\) is shown in Fig. 6(a), where the linear background due to the force effect was subtracted. The solid curve represents the result with \(H\) applied along the in-plane current \((\hat{I}, \text{i.e. } I \parallel H)\), while the dashed curve represents that with the perpendicular \(H, \text{i.e. } I \perp H\). Thus, the shapes of AMR curves for \(I \parallel H\) and \(I \perp H\) correspond to the in-plane and perpendicular magnetization curves, respectively. In this study, \(R_{xx}\) for \(I \parallel H\) (\(I \perp H\)) are defined as \(R_{xx,l}\) (\(R_{xx,t}\)). As shown in Fig. 6(a), \(R_{xx,l}\) is larger than \(R_{xx,t}\) for \(x = 0\), which means that the sign of AMR for the FePt is positive. The AMR ratio as a function of \(x\) is shown in Fig. 6(b). The definition of AMR ratio in this study is AMR ratio (\%) = \([R_{xx,l} - R_{xx,t}] \times 100 / R_{xx,t}\). As \(x\) is increased, the AMR ratio is decreased, however a sign reversal of AMR does not occur. The FePt and FePd exhibit 0.8 % and 0.4 % AMR ratios, respectively.

Here we discuss the composition dependence of \(\theta_{\text{AHE}}\) and AMR ratio for the present FePt\(_1-x\)Pd\(_x\) layers. Seemann and co-workers reported the different scattering mechanisms in AHE between FePt and FePd\(^{10}\). According to their experimental study\(^{10}\), the AHE in FePt is dominated by the intrinsic deflection whereas the side-jump is a dominant mechanism for the FePd. He \textit{et al.} investigated the correlation between the AHE and the spin-orbit coupling parameter\(^{11}\). They mentioned that there existed a clear positive correlation between the intrinsic term and the spin-orbit coupling parameter. Since the present composition dependence of \(\theta_{\text{AHE}}\) is similar to the composition dependence of intrinsic term reported in the previous paper\(^{11}\), the scenario that the spin-orbit coupling dominates the magnitude of AHE is applicable for the present result.

In contrast to AHE, the magnitude of AMR is affected by complicated several factors: spin-orbit coupling constant \((\lambda)\), exchange field \((H_e)\), and \(s\)-\(d\) scattering process. According the Campbell-Fert-Jaoul (CFJ) model\(^{17}\), the AMR of a strong ferromagnet is related with proportional to \((\lambda / H_e)^2\), suggesting that large spin-orbit interaction gives rise to the increase of AMR. This may be a simple scenario explaining the present composition dependence of AMR ratio. However, Kokado \textit{et al.} derived the more rigorous model for AMR taking into account the \(s\)-\(d\) scattering process\(^{14}\). In order to completely and quantitatively explain the present result, we need the further consideration.
4. Conclusion

We investigated the composition dependence of AHE and AMR for the FePt_{1-x}Pd_{x} (001) epitaxial films with highly ordered structures and perpendicular magnetization. Among the present samples, the FePt film exhibited the highest $K_u$, the largest $\theta_{AHE}$, and the largest AMR ratio. These values were gradually decreased as $x$ was increased. We discussed possible scenarios to explain those composition dependences, suggesting that the spin-orbit interaction plays an important role to determine the magnitude of these values.

Acknowledgements The authors thank Mr. Y. Murakami and Mr. I. Narita for their technical assistance. This work was partially supported by the Grant-in-Aid for Scientific Research S (23226001) and Scientific Research B (16H04487) as well as the Research Grant from the TEPCO Memorial Foundation. The device fabrication and structural characterization were partly performed at Cooperative Research and Development Center for Advanced Materials, IMR, Tohoku University.

References

1) D. Weller, A. Moser, L. Folks, M. E. Best, W. Lee, M. F. Toney, M. Schwickert, J. U. Thiele, and M. F. Doerner. IEEE Trans. Magn. 36, 10 (2000)
2) A. Cebeillada, D. Weller, J. Sticht, G. R. Harp, R. F. C. Farrow, R. F. Marks, R. Savoy, and J. C. Scott, Phys. Rev. B 60, 3419 (1994)
3) T. Shima, K. Takanashi, Y. K. Takahashi, and K. Hono, Appl. Phys. Lett. 81, 1050 (2002)
4) S. Okamoto, N. Kikuchi, O. Kitakami, T. Miyazaki, Y. Shimada, and K. Fukamichi, Phys. Rev. B 66, 024413 (2002)
5) M. Ohtake, S. Ouchi, F. Kirino, and M. Futamoto, J. Appl. Phys. 111, 07A708 (2012)
6) T. Seki, S. Mitani, K. Yakushiji, and K. Takanashi, Appl. Phys. Lett. 88, 172504 (2006)
7) S. Mizukami, S. Iihama, N. Inami, T. Hiratsuka, G. Kim, H. Naganuma, M. Oogane, and Y. Ando, Appl. Phys. Lett. 98, 052501 (2011)
8) S. Iihama, A. Sakuma, H. Naganuma, M. Oogane, T. Miyazaki, S. Mizukami, and Y. Ando, Appl. Phys. Lett. 105, 142403 (2014)
9) S. D. Willoughby, J. M. MacLaren, T. Ohkubo, S. Jeong, M. McHenry, D. E. Laughlin, S.-J. Choi, and S.-J. Kwon, J. Appl. Phys. 91, 8822 (2002)
10) K. M. Seemann, Y. Mokrousov, A. Aziz, J. Miguel, F. Kronast, W. Kuch, M. G. Blamire, A. T. Hindmarch, B. J. Hickey, I. Souza, and C. H. Marrows, Phys. Rev. Lett. 104, 076402 (2010)
11) P. He L. Ma, Z. Shi, G.Y. Guo, J.-G. Zheng, Y. Xin, and S. M. Zhou, Phys. Rev. Lett. 109, 086402 (2012)
12) S. Kikushima, T. Seki, K. Uchida, E. Saitoh, and K. Takanashi, AIP Adv. 7, 085210 (2017)
13) N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, Rev. Mod. Phys. 82, 1539 (2010)
14) S. Kokado, M. Tsunoda, K. Harigaya, and A. Sakuma, J. Phys. Soc. Jpn. 81, 024705 (2012)
15) M. Futamoto, M. Nakamura, M. Ohtake, N. Inaba, and T. Shimotatu, AIP Adv. 6, 085302 (2016)
16) Y. Kota, and A. Sakuma, J. Phys. Soc. Jpn. 81, 084705 (2012)
17) I. A. Campbell, A. Fert, and O. Jaoul, J. Phys. C 3, S95 (1970)

Received Oct. 19, 2018; Accepted Dec. 25, 2018