Effects of sputtering pressure on magnetic and magneto-optical properties in compositionally modulated Co/Pd thin films

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We have investigated the effects of sputtering Ar gas pressure on magnetic and magneto-optical properties in compositionally modulated Co/Pd thin films. The samples were prepared by dc magnetron sputtering from 2-in.-diam Co and Pd targets by alternately exposing the substrates to targets. Sputtering Ar gas pressure was varied from 2 to 30 mTorr. All samples had the same bilayer thicknesses composed of 2-Å-thick Co and 9-Å-thick Pd sublayers. It was observed that the intrinsic uniaxial anisotropy energy, magnetization, and polar Kerr rotation were monotonically decreased with increasing Ar gas pressures more than about 10 mTorr. Interestingly enough, the coercivity showed a thirtyfold enhancement as Ar gas pressure varied from 2 to 30 mTorr. We believe that the results are mainly ascribed to the variation of microstructure with sputtering Ar gas pressure.

I. INTRODUCTION

Artificially prepared compositionally modulated (CM) films, consisting of a periodic stacking of two constituents, have been attracting wide attention as a new class of materials because of their novel properties and potential technical applications. Recently, CM Co/Pd thin films have been the subject of numerous studies because these materials provide basic research of magnetism such as surface anisotropy and, in addition, they are promising media for the application to magneto-optical (MO) recording. Superior environmental stability and a larger Kerr effect at shorter wavelengths in these materials are major advantages compared to the current choice of MO media, rare-earth transition-metal alloy films.

In CM Co/Pd films, their magnetic and magneto-optical properties seem to be very sensitive to preparation methods and conditions as well as the sublayer and total film thicknesses. Considering the preparation methods, CM Co/Pd films prepared by vapor deposition have generally better magnetic properties than those prepared by sputtering. For instance, Shin and Palumbo obtained the coercivity of more than 2 kOe for e-beam evaporated Co/Pd films, while Garcia et al. reported the coercivity of a few hundred oersteds for sputtered films. It is well known that much more energetic atoms are involved in sputtering than vapor deposition. These energetic atoms are expected to smear out the interfaces of constituents and to enhance microstructural modification in the CM structure. One of the ways to moderate these energetic atoms is to increase sputtering pressure. In this paper, we report the effects of sputtering Ar gas pressure on magnetic and magneto-optical properties in CM Co/Pd thin films.

II. EXPERIMENT

CM Co/Pd thin films were prepared by dc magnetron sputtering from 2-in.-diam Co and Pd targets onto quartz substrates (for magnetic and magneto-optical measurements) and Si wafer substrates (for structural studies) on a rotatable substrate table. The dwelling time the substrates spent above each target could be controlled by a programmable timer interfaced to a stepping motor which drove the substrate table. A stainless plate with two target-size holes was placed between the targets and the substrate table to prevent cross contamination of their sputtered fluxes. Sputtering Ar gas pressure was varied from 2 to 30 mTorr. All samples had the same bilayer thickness of 11 Å composed of 2-Å-thick Co and 9-Å-thick Pd sublayers.

The CM structure was examined by low- and high-angle x-ray diffraction and the growth morphology of the film was investigated by microfractography. Magnetization was measured using a vibrating sample magnetometer (VSM) calibrated against a Ni standard. The magnetic anisotropy was measured using a torque magnetometer at an applied field of 15 kOe. For both magnetization and anisotropy measurements, the small signal from the sample holder and uncoated substrate was subtracted out. The polar Kerr rotation loops were measured using a 780-nm GaAlAs semiconductor laser in an optical detection system.

III. RESULTS AND DISCUSSION

In Table I we list the characteristic structural and magnetic parameters of the CM Co/Pd films in this study. All samples in this study developed low-angle x-ray diffraction peaks irrespective of sputtering pressure, which suggests the existence of the CM structures in those samples. However, as shown in Table I the full width at half maximum (FWHM) of the first-order low-angle diffraction peak became narrower with increasing the sputtering pressure.

It was observed that the saturation magnetization was monotonically decreased with increasing sputtering Ar gas pressure as shown in Fig. 1. The saturation magnetization of the figure was measured at a field of 10 kOe. To understand a decrease of $M_s$ with increasing sputtering pressure $P_{Ar}$, the density of a deposited film was deter-
TABLE I. Some structural and magnetic parameters of compositionally modulated Co/Pd films. Each parameter is denoted as follows: $P_{Ar}$ is the sputtering Ar gas pressure, FWHM is the full width at half maximum of the first-order low-angle diffraction peak, $\tau_p$ is the peak torque value, $\phi_0$ is the angle where the torque is zero in the first quadrant, and $M_r/M_s$ is the ratio of the remnant magnetization to the saturation magnetization.

<table>
<thead>
<tr>
<th>$P_{Ar}$ (mTorr)</th>
<th>FWHM (deg)</th>
<th>$\tau_p$ ($10^6$ dyn cm/cm$^3$)</th>
<th>$\phi_0$ (deg)</th>
<th>$M_r/M_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>1.2</td>
<td>-2.06</td>
<td>0</td>
<td>1.0</td>
</tr>
<tr>
<td>5</td>
<td>1.0</td>
<td>-2.27</td>
<td>0</td>
<td>1.0</td>
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<tr>
<td>10</td>
<td>0.8</td>
<td>-1.90</td>
<td>0</td>
<td>1.0</td>
</tr>
<tr>
<td>15</td>
<td>0.8</td>
<td>-1.64</td>
<td>0</td>
<td>0.9</td>
</tr>
<tr>
<td>20</td>
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</tr>
<tr>
<td>30</td>
<td>0.3</td>
<td>-1.22</td>
<td>6</td>
<td>0.7</td>
</tr>
</tbody>
</table>

Depended via independent measurements of weight and thickness. The density of a film prepared at $P_{Ar} = 2$ mTorr, $\rho (2$ mT), was 11.22 g/cm$^3$, which is 98% of a bulk density. In Fig. 1 we plot the relative density of a film normalized by $\rho (2$ mT). It can be seen that the density of the film decreases with $P_{Ar}$, which reflects an increase of the porous region due to the fact that less energetic atoms are associated with increasing sputtering pressure. Indeed, it was observed that columnar structure, where columnar regions were surrounded by a network of low density or void regions, was more distinctively developed with increasing $P_{Ar}$. Therefore, it could be concluded that the decrease of the film density with $P_{Ar}$ mainly accounts for the behavior of $M_r$. However, we speculate that an increasing oxidation of Co with $P_{Ar}$ also plays a role, which might explain slightly different dependencies of $M_r$ and $\rho$ on $P_{Ar}$.

In Fig. 1, it is worthwhile to mention that the magnetization per volume of Co exceeds the value of crystalline Co (1422 emu/cm$^3$), except for the film prepared at 30 mTorr. Enhanced magnetization must be due to polarization of the Pd, as already reported by several investigators.$^4$ In Fig. 2, we show the dependence of the intrinsic anisotropy energy $K_u$ and the easy-axis orientation $\delta$ on sputtering Ar gas pressure $P_{Ar}$.

Dependence of the intrinsic uniaxial anisotropy energy $K_u$ and the easy-axis orientation $\delta$ on sputtering Ar gas pressure were investigated. $K_u$ and $\delta$ were obtained by analyzing the torque curves according to the method of Shin and Agarwala reported elsewhere.$^{11}$ Using the shape anisotropy energy $K_s (2\pi M_s^2)$, the peak value $\tau_p$ in the measured torque curve, and the angle $\phi_0$ where the torque is zero, $K_u$ and $\delta$ are given$^{11}$ in Eqs. (1) and (2), respectively:

$$K_u = (K_s + \tau_p^2 + 2K_s \tau_p \cos 2\phi_0)^{1/2},$$

$$\delta = \phi_0 - \frac{1}{2} \arctan \left( \frac{K_s \sin 2\phi_0}{K_s \cos 2\phi_0 \pm \tau_p} \right),$$

where the positive sign is taken when $\pi/4 < \phi_0 < \pi/2$ and the negative sign is taken when $0 < \phi_0 < \pi/4$. The easy-axis orientation $\delta$ in Eq. (2) is defined from the film normal. Figure 2 shows the dependence of the intrinsic anisotropy energy $K_u$ and the easy-axis orientation $\delta$ on sputtering Ar gas pressure $P_{Ar}$. As seen in Fig. 2, the film has perpendicular magnetic anisotropy until $P_{Ar}$ is increased up to 15 mTorr. Then, the easy-axis orientation deviates from the film normal at higher pressures. (These results are consistent with the VSM measurements, where squareness, defined by the ratio of the remnant magnetization to the saturation magnetization, becomes smaller with higher $P_{Ar}$ as seen in Table I.) Therefore, for the application to MO recording it is not desirable to prepare a film at more than a 15-mTorr sputtering Ar gas pressure since perpendicular magnetic anisotropy is an essential requirement to be a suitable MO medium.$^{12}$

Dependence of $K_u$ on $P_{Ar}$ in Fig. 2 is qualitatively similar to the result reported by Hashimoto et al.$^4$ $K_u$ associated with an interface can be described as $K_u = 2K_s / t_{Co} \perp K_{in}$ where $K_s$ is the anisotropy originating from the interface per unit area, $t_{Co}$ is the Co thickness, and $K_{in}$ is the volume anisotropy consisting of demagnetization energy, magnetocrystalline anisotropy, and magnetoelastic energy.$^3$ CoPd alloy films and CM Co/Pd films.
FIG. 3. Dependence of the coercivity $H_c$ and the polar Kerr rotation $\theta_k$ on sputtering Ar gas pressure $P_{Ar}$.

are known to exhibit large magnetostriction. We believe that an increase of $K_u$ up to 5-mTorr $P_{Ar}$ in Fig. 2 is ascribed to magnetostriction as the stress of a film changes from compressive to tensile with $P_{Ar}$. High-angle x-ray measurements revealed that the texture of a film became poor with $P_{Ar}$. Thus, decrease of magnetocrystalline anisotropy with $P_{Ar}$ seems to be responsible for dependence of $K_u$ on $P_{Ar}$ at high sputtering pressures.

Figure 3 shows the effects of $P_{Ar}$ on the coercivity $H_c$ and the polar Kerr rotation angle $\theta_k$ obtained from the measurement of the Kerr hysteresis loop. As seen in Fig. 3, the coercivity of a film shows a thirtyfold enhancement as $P_{Ar}$ goes from 2 mTorr to 30 mTorr. The coercivity at 30 mTorr is close to that of an evaporated film. As pointed out by Hashimoto et al., a large coercivity at a high Ar pressure is believed to be caused by the pinning of domain wall at the boundary of a column as colomnar structure exists for the high Ar pressure.

Whereas, the Kerr rotation in Fig. 3 is relatively less sensitive to $P_{Ar}$. It is reasonable to assume that the Kerr rotation is proportional to the magnetization. Therefore, it can be concluded that a decreasing behavior of the Kerr rotation with $P_{Ar}$ is originated from a corresponding decrease of the magnetization as already demonstrated in Fig. 1.

IV. CONCLUSIONS

We have studied the effects of sputtering Ar gas pressure $P_{Ar}$ on magnetic and magneto-optical properties of compositionally modulated Co/Pd thin films. Variations of uniaxial intrinsic anisotropy, magnetization, coercivity, and Kerr rotation with $P_{Ar}$ were observed. We have found that the results were closely correlated with variation of the microstructure of a film with $P_{Ar}$.

This work suggests that suitable sputtering Ar gas pressure is between 10 and 15 mTorr to achieve a desirable magneto-optical recording medium by sputtering.

ACKNOWLEDGMENTS

This work was supported in part by the Korea Science and Engineering Foundation under Grant No. 901-0206-022-2 and a GoldStar Research Grant.

1See, for example, articles and references cited in Synthetic Modulated Structures, edited by L. L. Chang and B. C. Glessen (Academic, Orlando, 1985).