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Changes in Structure of Multilayered Cobalt/Noble Metal Films with Annealing

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We considered that Co/Au and Co/Pd multilayered films will show the different structural changes each, because Co-Au is a eutectic system while Co-Pd is an isomorphous system. Therefore we annealed not only Co/Au and Co/Pd multilayered films but also Co-Au and Co-Pd alloyed films to research the changes of multilayered structures. These samples were analyzed with XRD, extended 3-step model profile fitting, electric resistivity, and AES. After 48h annealing, Au (111) peak appeared and the electric resistivities decreased remarkably in Co-Au alloyed films, whereas the electric resistivities decreased monotonously in Co-Pd alloyed films. For Co/Au and Co/Pd multilayered films, XRD patterns based on multilayered structure such as main peaks or satellite peaks were destroyed with 48h annealing, then Au (111) peak appeared in Co/Au multilayered film, although Co-Pd (111) peak appeared in Co/Pd mulitlayered film. From these results and extended 3-step model profile fittings, we confirmed that the interfaces between Co and Au became sharper with annealing, while the interfaces between Co and Pd formed more alloyed layers. We also found similar results with AES.

Keywords: multilayered cobalt/noble metal film, multilayered structure, X-ray diffraction, extended 3-step model profile fitting

1. INTRODUCTION

Co/Au and Co/Pd multilayered films are known as perpendicular magnetization films. They are researched for applications of the high-density recording media such as magneto optical disks and hard disks.[1] However, the interface effects considered one of the causes of perpendicular magnetization films are not clarified in detail. Accordingly, their studies are very important to create new materials in the near future.

In this paper, we reported the changes in structure of Co/Au and Co/Pd multilayered films with annealing. We considered the differences of interface conditions between Co/Au and Co/Pd multilayered films. Because Co-Au alloyed whose solid solutions hardly be formed is a eutectic system while Co-Pd alloyed is an isomorphous system. Therefore we annealed not only Co/Au and Co/Pd multilayered films but also Co-Au and Co-Pd alloyed films in vacuum, then analyzed the multilayered structural changes in the interfaces with X-ray diffraction (XRD), extended 3-step model profile fitting, electric resistivity, and Auger electron spectroscopy (AES).

2. EXPERIMENT

The samples, multilayered films of Co/Au and Co/Pd, and

Table 1 Structure of film samples

Alloyed films			
Samples	Co (at%)		
Co-Au	25.8%	39.2%	60.9%
Co-Pd	31.4%	39.5%	65.0%
Thickness	2000 Å		
Multilayered films			
Samples	Structures		
Co/Au	$[14.0/53.0\,Å]^{30}$		
	2010 Å		
Co/Pd	$[14.0/44.0\,Å]^{30}$		
	1740 Å		
Multilayered films for AES			
Samples	Structures		
Co/Au	$[100/100\,Å]^{10}$		
Co/Pd	$[100/100\,Å]^{10}$		
Thickness	2000 Å		

alloyed films of Co-Au and Co-Pd, were fabricated onto the rotating glass, polyimide, and silicon substrates with dual-source RF magnetron sputtering methods using Ar as a sputtering gas. Structures of each sample are shown in Table 1. These samples were annealed in vacuum under 1×10^{-4} Pa at 300℃ for Au, at 400℃ for Pd keeping 0.5- 48 h each.

XRD was measured to all samples under 40 kV, 200 mA with Cu-Kα radiation, scanning 1.3-15° in a low angle region and 30-50° in a high angle region. For multilayered films, we performed extended 3-step model profile fittings, and for alloyed films, we measured their resistivities. AES was measured to multilayered films (100/100Å) under 1×10^{-8} Pa and 3-5kV ion beam using Ar as a etching gas.

3. RESULTS AND DISCUSSION

3.1 Alloyed film structure

The XRD patterns for Co-Au and Co-Pd alloyed films are shown in Fig.1. For Co-Au films, Co-Au (111) broad peaks were observed before annealing, and then Au (111) peaks appeared clearly after 48h annealing. For Co-Pd films, Co-Pd (111) peaks were observed before annealing, and they became more acute after 48h annealing. Moreover, the electric resistivities of Co-Au alloyed films decreased remarkably as the annealing went on. This decrease suggests that Co-Au alloys were separated into Co and Au with the annealing because of a eutectic system, and also their separations are clear from the fact that Au (111) peaks appeared after annealing. For Co-Pd alloyed films, the electric resistivities decreased monotonously, because the strains of lattice spacings were removed with annealing, showing Co-Pd (111) acute peaks in XRD patterns. And this result suggests that Co-Pd alloys were more mixed after annealing because of an isomorphous system.

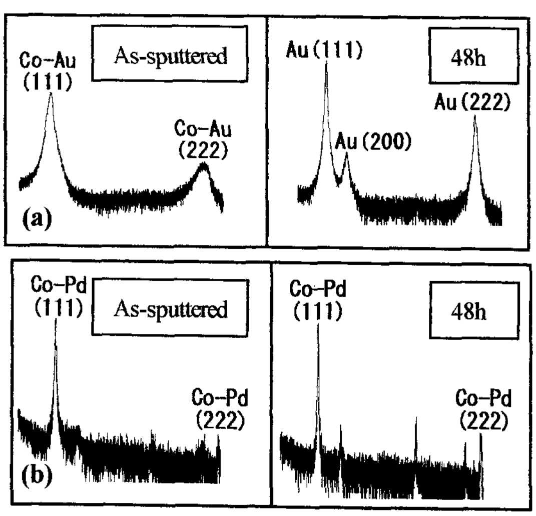

Fig. 1 Alloyed film XRD patterns on the high angle region (a) Co-Au film and (b) Co-Pd film

3.2 Multilayered film structure

The XRD patterns for Co/Au and Co/Pd multilayered films are shown in Fig.2. The XRD peaks based on the periodic structures were observed in the low angle region before annealing in both films, as well as the main peaks (0 order) based on the composite peaks of Co and noble metal and the satellite peaks (±1, ±2, ... order) based on the periodic structures were observed in the high angle region before annealing in both films. For Co/Au multilayered film, the XRD peaks hardly changed until 1.5h annealing, but +4 and +5 order peaks disappeared after 24h annealing in the low angle region. In the high angle region, the XRD peaks hardly changed until 1.5h annealing, but the main peak and the satellite peaks disappeared after 24h annealing, then Au (111) peak appeared. For Co/Pd multilayered film, the XRD peaks disappeared gradually, and all peaks disappeared completely after 24h annealing in the low angle region. In the high angle region, -2 and +2 order peaks disappeared after 1.5h annealing, and then Co-Pd (111) peak appeared after 24h annealing. Above results suggest that the periodic structure was kept to some extent in Co/Au multilayered film, after that, and then the periodic structure was destroyed as the annealing went on, and then Au was separated from Co. On the other hand, the periodic structure was destroyed gradually as the annealing went on, then Co-Pd alloyed layers formed at the interfaces were spread.

3.3 Extended 3-step model profile fitting

A profile fitting, which is a comparison between calculated XRD peaks and experimental peaks, is very useful to analyze multilayered structure. Performing profile fittings, we need to assume calculated models first, and then we calculate their XRD peak intensities. Fig.3 shows the calculated models. 2-step model is an ideal periodic structure that has no alloyed layers between Co layers and noble metal layers, and each atomic layer are crystalline. [2] 3-step model includes alloyed layers at the interfaces. [3] Generally, it is difficult to fabricate the multilayered films that have an ideal periodic structure without fluctuation.

In consequence, we modified 3-step model and assumed extended 3-step model that includes a fluctuation of a periodic thickness and of an alloyed layer. In this study, we analyzed the multilayered structures with extended 3-step model. We describe the XRD peak intensity calculation by extended 3-step model in the following.

The intensity of XRD peak I (Q) is given by

$$I(Q) = L(Q)\,|F(Q)|^2 \qquad (1)$$

Here L(Q) is a Laue function and F(Q) is a layer structure factor. We assumed that Co layer is a, noble metal layer is b, and alloyed layer is c with each bulk lattice spacings d_a, d_b, d_c, with each element n_a, n_b, n_c atomic planes. When this periodic thickness Λ is stacked by N times, L(Q) and F(Q)F*(Q) are expressed as follows

$$L(Q)=\frac{1+\exp(-N\sigma^2Q^2/2)-2\exp(-N\sigma^2Q^2/4)\cos(N\Lambda'Q)}{1+\exp(-\sigma^2Q^2/2)-2\exp(-\sigma^2Q^2/4)\cos(\Lambda'Q)} \quad (2)$$

$$\begin{aligned}
|F(Q)|^2 = {} & 4f_c^2(Q)\frac{1+\exp[-n_c\sigma^2Q^2]-2\exp[-n_c\sigma^2Q^2/2]\cos(n_cd_cQ)}{1+\exp[-\sigma^2Q^2]-2\exp[-\sigma^2Q^2/2]\cos(d_cQ)} \\
& \times\cos^2(\lambda_bQ/2)\exp(-\sigma^2Q^2/4) \\
& +2f_a(Q)f_b(Q)L_a^{1/2}(Q)L_b^{1/2}(Q)\cos(\Lambda Q/2) \\
& +\frac{2f_a(Q)f_c(Q)L_a^{1/2}}{1+\exp[-\sigma^2Q^2]-2\exp[\sigma^2Q^2/2]\cos(d_cQ)}\cos(\lambda_bQ/2)\exp(-8\sigma^2Q^2) \\
& \times\{\exp[-(n_c+1)\sigma^2Q^2/2]\cos[(n_c-1)d_cQ/2] \\
& -\exp[-n_c\sigma^2Q^2/2]\cos[\Lambda Q/2+(n_c+1)d_cQ/2] \\
& -\exp[-n_c\sigma^2Q^2/2]\cos[(n_c+1)d_cQ/2] \\
& +\cos[\Lambda Q/2-(n_c-1)d_cQ/2]\} \\
& +\frac{2f_b(Q)f_c(Q)L_b^{1/2}}{1+\exp[-\sigma^2Q^2]-2\exp[\sigma^2Q^2/2]\cos(d_cQ)}\cos(\lambda_b/Q)\exp(-\sigma^2Q^2/8) \\
& \times\{\exp[-(n_c+1)\sigma^2Q^2/2]\cos[\Lambda Q/2+(n_c+1)d_cQ/2] \\
& -\exp[\sigma^2Q^2/2]\cos[\Lambda Q/2-(n_c+1)d_cQ/2] \\
& +\cos[(n_c-1)d_cQ/2]\} \\
& -\exp[-\sigma^2Q^2/2]\cos[(n_c+1)d_cQ/2]
\end{aligned} \quad (3)$$

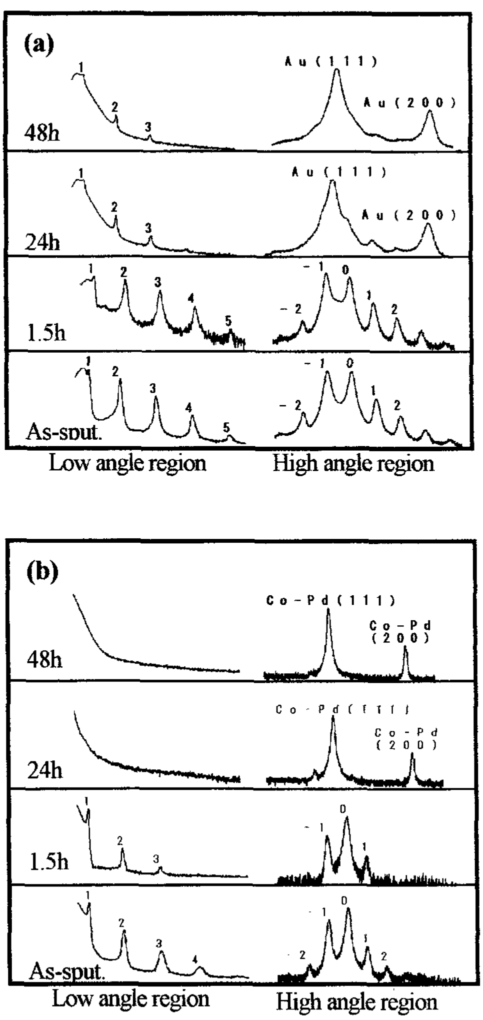

Fig. 2 Multilayered film XRD patterns
(a) Co/Au film and (b) Co/Pd film

Next, we performed the multilayered structural analyses making a comparison between experimental XRD peaks and calculated peaks given by eq. (1) on the high angle region. Fig.4 shows extended 3-step model profile fittings. For Co/Au multilayered film, the periodic thickness is 27 atomic planes, and the alloyed layer thickness decreased from 2 to 0 atomic planes after 1.5h annealing. This result suggests that the alloyed layers formed in the fabrication process disappeared with annealing, that is, the stacking interfaces became sharper since Co-Au is a eutectic system. For Co/Pd multilayered films, the periodic thickness is 27 atomic planes, and the alloyed layers increased from 2 to 5 atomic planes after 12h annealing. This result suggests that the alloyed layers formed in fabrication process were more spread with annealing since Co-Pd is an isomorphous system.

In the above discussions, we described structural analyses of multilayered films with extended 3-step model profile fittings. Moreover, we performed AES depth profiles to analyze their structural changes in detail. Fig.5 shows AES depth profiles for Co/Au and Co/Pd films (100/100 Å in both films). For Co/Au

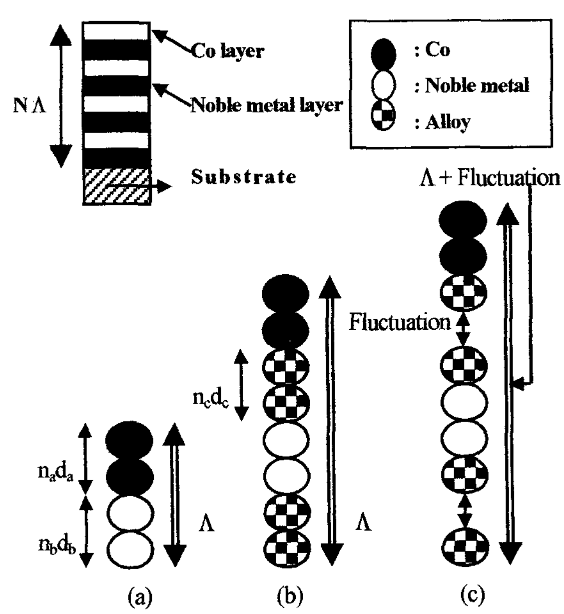

Fig. 3 Schematic view of multilayered films
(a) 2-step model, (b) 3-step model
,and (c) extended 3-step model

multilayered film, it is clear that the stacking interfaces were much the same after 48h annealing. In this case, Co and Au did not separate since Co and Au layers were more thick than usual. For Co/Pd multilayered film, the stacking interfaces were more mixed after 48h annealing.

4. CONCLUSION

We annealed not only Co/Au and Co/Pd multilayered films but also Co-Au and Co-Pd alloyed films in vacuum to analyze multilayered structures. For Co-Au alloyed films, appearance of Au (111) peaks and decrease of electric resistivities after 48h annealing proved separations of Co and Au based on a eutectic system. For Co-Pd films, it was confirmed that Co-Pd alloys were more mixed after 48h annealing because of an isomorphous system.

Next, XRD peaks based on the multilayered structures were observed in both Co/Au and Co/Pd multilayered films before annealing. For Co/Au multilayered film, the periodic structure was kept to some extent, and then it was destroyed with annealing owing to the separation of Co and Au. For Co/Pd multilayered film, the periodic structure was destroyed gradually with annealing, then Co-Pd alloyed layers at the interfaces were spread. In addition, results from profile fittings with extended 3-step model, alloyed layers at the interfaces formed in the fabrication process decreased in Co/Au films after annealing although increased in Co/Pd films.

Taking above results into consideration, we would like to emphasize that alloyed interlayers at the interfaces of Co/Au film become sharper as annealing went on, and then Co precipitate in Au after 48h annealing since Co-Au is a eutectic system. On the other hand, alloyed interlayers at the interfaces of Co/Pd film were spread as annealing went on since Co-Pd is an isomorphous system. These interface changes were also made certain from AES depth profiles.

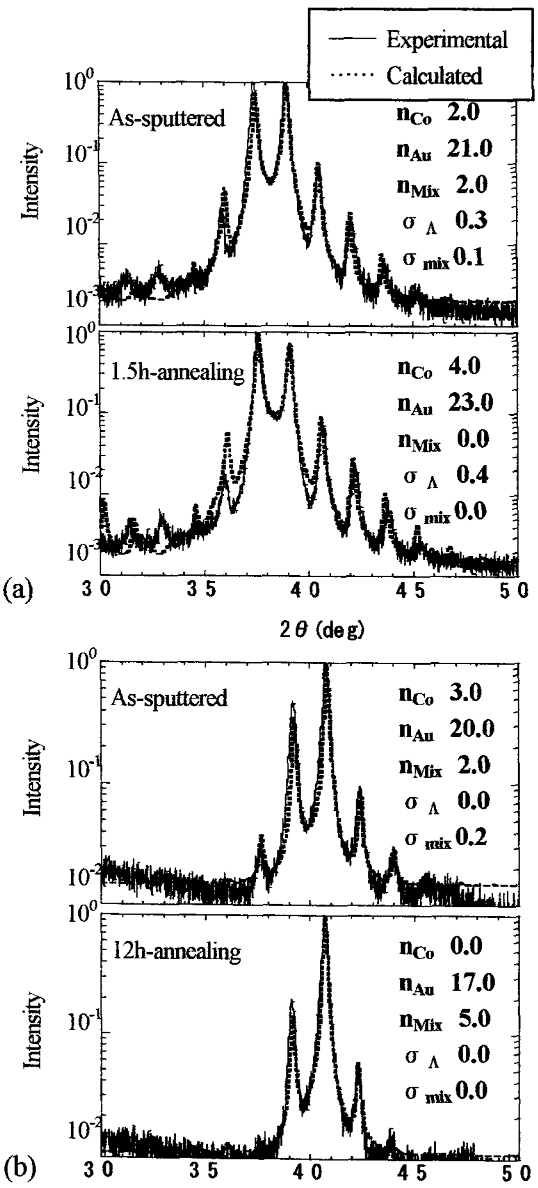

Fig. 4 Extended 3-step model profile fittings
(a) Co/Au film and (b) Co/Pd films

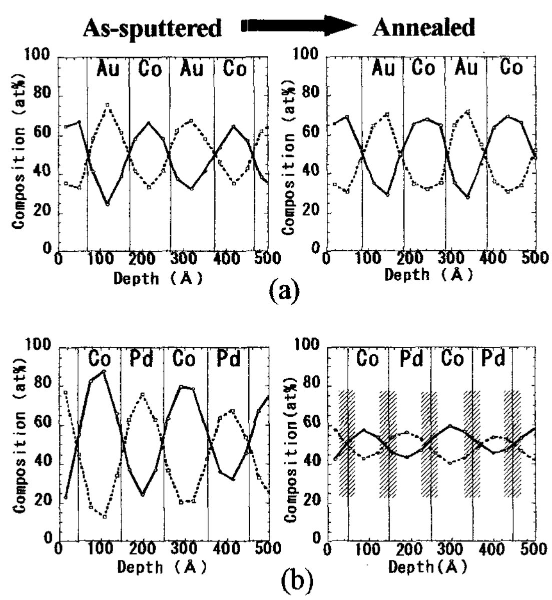

Fig. 5 AES depth profiles
(a) Co/Au film and (b) Co/Pd film

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