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Diffuse Interface and Strain Anisotropy in Co/Pd(111) Multilayers by Polarized EXAFS Study

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Abstract: We examined interface structure of Co/Pd multilayers by quantitative analysis of fluorescence-yield Co *K*-edge EXAFS. To investigate the local atomic structure surrounding Co absorber and its anisotropy in orthogonal directions, two polarization dependent set-ups were used. By means of first shell analysis including distinct nearest neighbors of Co and Pd atoms, it was found that a considerable amount of Co-Pd alloy-like phase near interfaces of thin Co layers is formed by *atomic scaled diffuseness of growing surface*. Moreover, we found Co strain anisotropy depending on Co layer thickness, generally, tensile and compressive strains of in-plane and out-of-plane atoms, respectively. These interface structures might critically affect the tendency of perpendicular magnetization in Co/Pd multilayers.

1. INTRODUCTION

Since an experimental discovery of perpendicular magnetization (PM) in Co/Pd multilayers in 1985 [1], extensive studies have been made to understand the tendency of the PM [2-4]. The magnetic properties of these films might be dominated strongly by the interface structure. The films entirely possess interface widths with anomalous magnetic properties due to repetition of the ultrathin Co layer binding Pd spacers. The presence of interface imperfections is crucial and must be a fundamental factor when explaining strong PM, although clear observation has remained elusive. Thus, we need a detailed interface structure: interfacial mixing and anisotropic Co strains. The polarization-dependent EXAFS technique is an excellent method for anisotropically examining the local structure surrounding specific atoms [5,6]. With this detailed structural information, possible mechanisms corresponding to PM should be studied regarding microscopic origins such as the magnetostriction effect of the Co-Pd alloy-like phase. Therefore, we present the interface structure, namely, intermixing and strain anisotropy for Co/Pd multilayers, investigated by polarized EXAFS technique.

2. EXPERIMENT PROCEDURE

The polarization dependent EXAFS experiments were performed on the EXAFS stage using a Si (111) channel-cut single-crystal monochromator installed on the VEPP-3 storage ring at the SSRC of the BINP, Novosibirsk [6]. The oscillatory absorption coefficient $\mu(E)$ was measured at the Co *K*-edge ($E = 7709$ eV) in the fluorescence-detection mode with a gas electroluminescent detector filled with Xe gas at near-grazing incidence of synchrotron radiation (SR) for both polarization dependent set-ups: electric vectors $\hat{\epsilon}$ are parallel ($\parallel$) and nearly perpendicular ($\perp$) to the film plane. The I_0 ion chamber was filled with 5% Ar + He gases. To suppress undesirable reflection from Si wafer, the samples were rotated around the surface normal. Co and Pd elements were evaporated from two different heating sources in a vacuum of better than 2×10^{-6} mbar (base pressure is 2×10^{-7} mbar). The deposition rate was carefully controlled to less than $0.3 \text{ \AA} \pm 0.04 \text{ \AA}$ during whole the preparation process. Low-angle and high-angle x-ray diffraction profiles, and transmission-electron-microscopy cross-section images indicate that the multilayers have well-ordered atomic and layered stacking with (111)-plane preferred crystallographic orientation.

3. DATA ANALYSIS DETAILS

The SR is strongly linear-polarized in the plane of the electron orbit. The amplitude of the EXAFS signal depends upon an angle between the $\hat{\epsilon}$ and the bonding axis from an absorber to a neighbor. This novel characteristic enables us to get anisotropic structural information of radial distances R and coordination numbers N at two polarization dependent set-ups. There are 4 effective nearest neighbors (NN), $N_{\perp}^* = 4$ in $\hat{\epsilon} \perp$ the film plane, and 8 effective NN's, $N_{\parallel}^* = 8$ in $\hat{\epsilon} \parallel$ the film plane for fcc (111) or hcp (0002)-plane oriented polycrystal where total in-plane atoms of 6 ($N_{\text{tot}}^{\text{in}} = 6$) and total out-of-plane atoms of 6 ($N_{\text{tot}}^{\text{out}} = 6$), 3 above and 3 below. For an analysis of two different polarized EXAFS data, two EXAFS functions should be considered and the total EXAFS's contributed by the first shell of Co and Pd backscatters are given by

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$$\chi(k)_{\text{tot}}^{\perp} = \chi(k)_{\text{Co}}^{\text{out}} + \chi(k)_{\text{Pd}}^{\text{out}} \quad (1)$$

$$\chi(k)_{\text{tot}}^{\parallel} = \chi(k)_{\text{Co}}^{\text{in}} + \chi(k)_{\text{Pd}}^{\text{in}} + \chi(k)_{\text{Co}}^{\text{out}} + \chi(k)_{\text{Pd}}^{\text{out}} \quad (2)$$

For quantitative analysis of the first shell, we fit both Co-Co and Co-Pd peaks in real space simultaneously to the model standards generated by FEFF6 [7] and using UWXA3.0 code [8], as shown in Fig. 1. It should be noted that the minor peak of Pd first shell is overlapped with the main peak of the Co first shell. Here we first extracted $\bar{R}_{\text{Co(Pd)}}^{\text{out}}$ and $N_{\text{Co(Pd)}}^{\text{out}}$ as shown in Fig. 1 (a) with Eq. (1), because in-plane atoms do not contribute to EXAFS signals observed in $\hat{\varepsilon} \perp$ the film plane. Bar above symbols means average value over period, i.e., depth z . As a subsequent step, four contributions coming from both out-of-plane and in-plane backscatters were fit using measured $\bar{N}_{\text{Co(Pd)}}^{\text{out}}$ and $\bar{R}_{\text{Co(Pd)}}^{\text{out}}$ with Eq. (2), as shown in Fig. 1(b). Thus, $\bar{N}_{\text{Co(Pd)}}^{\text{in}}$ and $\bar{R}_{\text{Co(Pd)}}^{\text{in}}$ can be extracted from radial distribution functions (RDF's) observed in $\hat{\varepsilon} \parallel$ the film plane. Fourier filtered $k^3 \chi(k)$ are also shown together with each contribution in Fig. 1.

Fig. 2 shows Fourier-transforms (FT's) of Co K -edge EXAFS data for $[\text{Co}(x\text{ \AA})/\text{Pd}(24\text{ \AA})]_{13}$ multilayers, where subscript means the number of bilayers. The same quantitative analysis procedures were employed for those multilayers. The numerical results are shown in Table I.

4. RESULTS AND DISCUSSION

4.1 Interface Broadness

In RDF's at $x = 3.2$ and 5.0 \AA in Fig. 2, considerable Pd and Co neighbor peaks appear in both polarizations, which are similar to RDF (see open circles) of Co-Pd alloy film seen in Fig. 1 [9], indicating Co atoms binding Pd spacer layers are scattered into the Pd matrix. Measured values of $\bar{N}_{\text{Co}}^{\text{in}} < 6$ and $\bar{N}_{\text{Co}}^{\text{out}} > 0$ in Table I clearly indicate a presence of atomically diffuse heterointerfaces formed during film growth at slightly increasing room temperature. If one monolayer thick Co atoms were layered on a flat interface, we could observe $\bar{N}_{\text{Co}}^{\text{in}} = 6$ and $\bar{N}_{\text{Co}}^{\text{out}} = 0$. By comparing measured $\bar{N}_{\text{Co}}^{\text{in}}$ with a calculated one according to the intermixing evolution, the Co concentration C_{Co} profiles within the unit period can be estimated. Assuming a random mixed solid solution, thus $\bar{N}_{\text{Co}}^{\text{in}}$ is calculated with respect to the

evolution of intermixing at the interface to be, $\bar{N}_{\text{Co}}^{\text{in}} = n^{\text{in}} \int_{\text{period}} C_{\text{Co}}^2 dz / \int_{\text{period}} C_{\text{Co}} dz$ with $C_{\text{Co}} = \frac{1}{2} C_{\text{max}} [1 + \text{erf}(z/\Gamma)]$, where $n^{\text{in}} = 6$ is the total in-plane atoms. The Γ is the interface broadness and C_{max} is the maximum of C_{Co} obeying mass

Figure 1: Fourier transform $[FT(r)]$ of $k^3 \chi(k)$ for the Co K -edge EXAFS of $[\text{Co}(2\text{ \AA})/\text{Pd}(15\text{ \AA})]_{13}$ multilayer, compared to Co_1Pd_3 alloy film (open circles), and Fourier filtered $k^3 \chi(k)$.

Figure 2: Fourier transforms of Co K -edge EXAFS for $[\text{Co}(x\text{ \AA})/\text{Pd}(24\text{ \AA})]_{13}$ for both polarization set-ups. $x = 3.2, 5.0, 12.5, 16.5, 23$, and 29 \AA are in ascending order from the bottom.

Table I. Best-fit values of Co-Co pairs. Symbol (*) notes that $\bar{N}_{\text{Co}}^{\text{in}}$ is fixed at 6 when fit.

$x(\text{\AA})$	$\bar{N}_{\text{Co}}^{\text{in}}$	$\bar{N}_{\text{Co}}^{\text{out}}$	$\bar{R}_{\text{Co}}^{\text{in}}(\text{\AA})$	$\bar{R}_{\text{Co}}^{\text{out}}(\text{\AA})$
3.2	1.6 ± 0.2	2.0 ± 0.4	2.57 ± 0.02	2.37 ± 0.04
5.0	2.8 ± 0.1	3.5 ± 0.5	2.60 ± 0.02	2.36 ± 0.03
12.5	4.6 ± 0.3	4.6 ± 0.2	2.51 ± 0.01	2.46 ± 0.01
16.5	5.3 ± 0.3	5.3 ± 0.2	2.501 ± 0.009	2.48 ± 0.01
23	6*	5.4 ± 0.2	2.508 ± 0.006	2.486 ± 0.007
29	6*	6 ± 0.2	2.514 ± 0.006	2.48 ± 0.02

conservation law. The plots of calculated $\bar{N}_{Co}^{in}$ vs. Co layer thickness t_{Co} are seen depending on Γ in Fig. 3, compared to the experimental data (solid circles). The inset shows the C_{Co} within a period, determined from estimated Γ . Fig. 3 shows the quantitative degrees of intermixing and that the Co/Pd multilayers deposited at a rate of even less than 0.3 Å have not abrupt interfaces, but rather diffuse ones.

4.2. Strain Anisotropy

From the curve-fitting results shown in Table I, the change in $\bar{R}_{Co}^{in(out)}$ is plotted as a function of t_{Co} in Fig. 4. Strong strain anisotropy, generally, in-plane tensile and out-of-plane compressive strains are observed at thinner t_{Co} . Remarkable relaxation of Co atoms toward the bulk value at the thicker t_{Co} might be due to strain relief introduced by misfit dislocations.[6]

4.3. Correlated Magnetic Anisotropy

The observed interface broadnesses indicate that Co and Pd atoms are mixed in one or two monolayers width. This demonstrates that the Co atoms do not have broken symmetry by Pd spacers, but have a characteristic Co-Pd alloy-like. This kind of interfacial mixing on atomic scale mainly leads to strong PM together with large strain anisotropy of Co atoms, usually tensile in-plane via magnetostriction effect through its much more negative magnetostriction coefficient than that of bulk Co by a relation of $K_{\sigma}^{\lambda} = -\frac{3}{2}\lambda_{111}\sigma_{\parallel}$ where $\sigma_{\parallel}$ is the stress applied to Co atoms in the film plane, and λ_{111} is the magnetostriction coefficient of [111] crystallographic direction. The microscopic origin of magnetoelastic anisotropy is similar to that of magnetocrystalline anisotropy coming from spin-orbital coupling. When the stress is applied to the magnetic film, the orientation of magnetization varies according to total energy valence. Detailed description of magnetic anisotropy was made elsewhere [10].

5. SUMMARY

Polarization dependent EXAFS is an excellent technique for examining anisotropic local structures such as interfacial mixing and strain anisotropy in multilayers with ultrathin layers. These detailed interface structures in the Co/Pd multilayers should be considered in experiments and theories, which might lead to strong PM via magnetostrictive effect. Finally, the importance of interface structure necessitates further development of depth-resolving EXAFS technique to directly differentiate interface structure from the interior of thicker layers.

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Figure 3: Calculated $\bar{N}_{Co}^{in}$ (solid lines) depending on Γ in Å. Solid circles show experimental data. Inset shows C_{Co} within unit period using determined Γ .

Figure 4: Co-Co radial distance versus Co layer thickness