Manipulation of spin reorientation transition of ultrathin Co films by using an artificially roughened Pd(111) substrate

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The authors present a simple method to control the spin reorientation transition (SRT) in ultrathin Co films using an artificially roughened substrate prepared by ion bombardment with varying the incident angle \(\theta\) from 0° to 80°. The combined study of surface magneto-optical Kerr effects, scanning tunneling microscopy (STM), and scanning tunneling spectroscopy (STS) revealed a drastic increase of the onset thickness of the SRT \(t_c\) by up to 41% for the substrate sputtered at \(\theta=80°\), whereas there is an 18% reduction for the \(\theta=20°\) sample relative to the SRT in the smooth surface. The second- and fourth-order surface anisotropies \(K_{2s}\) and \(K_{4s}\), responsible for the drastic change in the SRT, are determined from a theoretical fit to the magnetization orientation in the spin reorientation region. © 2007 American Institute of Physics. [DOI: 10.1063/1.2431442]

The experiment was performed in an ultrahigh vacuum chamber with a base pressure of \(7\times10^{-11}\) Torr. The chamber was equipped with an in situ surface magneto-optical Kerr effect (SMOKE) measurement system as well as a scanning tunneling microscope (STM). The single crystal Pd(111) substrate was cleaned with cycles of 1 keV Ar\(^+\) (~1.22 \(\times\) \(10^{13}\) ions/cm\(^2\)) ion sputtering and subsequent annealing at 800 °C to obtain a clean and atomically smooth surface. The artificially roughened Pd(111) surface was achieved through 10 min of Ar ion sputtering with the incident angle \(\theta\) varying from 0° to 80°. The angle \(\theta\) was measured from the film normal. Co films were prepared on the roughened Pd(111) substrate of 10 \(\times\) 10 \(\times\) 1 mm(t) at ambient temperature by e-beam evaporation. The pressure was maintained less than 1 \(\times\) \(10^{-10}\) Torr during the deposition and the typical deposition rate, obtained under a Co flux of 25 nA, was 0.8 Å/m. Magnetic hysteresis loops were measured by means of the in situ three-axis SMOKE setup. The structure and cleanliness of the sample were checked with a STM, which also provided the growth morphology of Co films.

After the repeated cycles of Ar\(^+\) ion sputtering and annealing, an atomically flat and smooth Pd(111) surface having large regular terraces of 30–100 nm wide were obtained as shown in Fig. 1(a). This smooth Pd(111) surface was sputtered by Ar\(^+\) ions with the ion incidence angle \(\theta\) varying from 0° to 80° to obtain an artificially roughened surface. Figure 1(b) shows the surface topography of Pd(111) after ion dose sputtering for 10 min at a normal incidence (\(\theta=0°\)). The moundlike surface structure can be observed where the mounds have a height of ~2 nm and a width of ~8 nm. Figure 1(c) shows the Pd(111) surface sputtered at \(\theta=60°\). The moundlike structures are still apparent, however, the height of the mounds is just 0.4 nm which corresponds to the height of two atomic steps, and the width is about 5 nm, i.e., it could be characterized by a regular distribution of a few ML-high clusters. However, as we further increased \(\theta\), the

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surface topography of Pd(111) was modified in a different manner. Figure 1(d) shows the surface sputtered at $\varphi = 80^\circ$. In this case, instead of the isotropic moundlike structure, elongated atomic steplike structures are created along the direction of the ion incidence.\textsuperscript{6} To quantify the observed surface roughness, we determined the roughness $\sigma$, which is the mean value of |$\Delta z$| where $\Delta z$ is the variation of $z$ from the mean flat surface, and the lateral wavelength $L$, the average lateral size of the roughened surface from STM images.\textsuperscript{12}

Figure 2 shows the $\sigma$ and $L$ of the substrate with respect to the ion incident angle. In the inset of Fig. 2, we plot the scaled roughness ($\sigma/L$) as a function of the angle. It can be seen from the figure that, in the region of $0^\circ \leq \varphi \leq 60^\circ$, $\sigma$ and $\sigma/L$ are almost constant or slightly decrease with respect to the sputtering angle. However, in the region of $60^\circ \leq \varphi \leq 90^\circ$, there is a drastic reduction in scaled roughness with increasing the ion incident angle.

Onto these artificially roughened surfaces, we have deposited Co films and carried out in situ SMOKE measurements. In Fig. 3, we present representative polar Kerr hysteresis loops of Co films prepared on roughened Pd(111) substrates. In all cases the initially square polar loops become slanted with increasing the Co thickness. Inverse trends with increasing Co thickness were also observed in the longitudinal Kerr hysteresis loops, which imply that the SRT from normal-to-plane magnetization to in-plane magnetization occurred with increasing the Co thickness. Note that not only the onset thickness of SRT, $t_c$, but also the transition region of spin reorientation changes drastically with the incident angle of ions: SRT occurs in thicker Co films for a substrate prepared at large sputtering angle of $\varphi \geq 60^\circ$ with large transition width, while it occurs at thinner Co films for $\varphi \leq 60^\circ$ with small transition thickness, compared to the smooth substrate. For a quantitative analysis of the SRT on the roughened Pd(111) surface, spin orientations of Co films were determined by a method utilizing both Kerr rotation ($\theta^{(L)}_K$) and ellipticity ($\epsilon^{(L)}_K$). The vector analysis described elsewhere\textsuperscript{13} enables us to determine the magnetization vector $M$ components $M_x$, $M_y$, and $M_z$ normalized by the saturation magnetization $M_S$, where ($x$, $y$, $z$) are the in-plane and normal-to-plane directions, respectively. In Fig. 4(a), we plot the magnetization orientations from the film normal at $H=0$, defined as $\theta = \cos^{-1}(M_y/(M_x^2+M_y^2+M_z^2)^{1/2})$ as a function of Co thickness. It was found that the SRT occurs at $t_c = 4.9$ ML in the case of the smooth surface, while the SRT takes place at $t_c = 5.8$ ML and $t_c = 2.9$ ML for the surface sputtered at $\varphi = 80^\circ$ and $\varphi = 20^\circ$, respectively. It implies an increase of $t_c$ by up to 41% for the sample sputtered.

FIG. 2. Dependence of roughness and lateral wavelength on the ion sputtering angle. The inset shows the value of the calculated scaled roughness.
at $\varphi=80^\circ$, whereas there is an 18% reduction for the $\varphi=20^\circ$ sample compared to the smooth substrate. The shape anisotropy of magnetic thin films energetically favors in-plane magnetization, and therefore it contributes negatively to perpendicular magnetic anisotropy always.\textsuperscript{14} Thus, the increased $t_s$ in the range of $\varphi>60^\circ$ might be ascribed to the reduced magnetic shape anisotropy caused from the rough Co surface. However, the reduced $t_s$ in the region of $\varphi<60^\circ$ could not be attributed to the shape anisotropy even though the roughness of this region is larger than the region $\varphi>60^\circ$. Symmetry breaking could also directly influence the magnetic anisotropy at the step edges which determine the macroscopic uniaxial anisotropy.\textsuperscript{15} Therefore, in the case of $\varphi=80^\circ$ where the step density is high enough and step edges are nearly parallel with each other, the step-induced in-plane anisotropy might exist with the easy axis parallel to the direction of the step edges. However, it also could not be adopted to explain the reduced $t_s$ in the region of $\varphi<60^\circ$ since the anisotropic step density is negligible in this region.

Therefore, in order to elucidate the variation of the SRTs on artificially roughened substrates, we determined the second-order anisotropy ($K_2$) and fourth-order surface anisotropy ($K_4$) through a theoretical fit of the magnetization orientation in the spin reorientation region. Figure 4(a) shows that the magnetic easy axis is switched from normal to in plane via a smooth transition rather than an abrupt one, i.e., from normal to in plane via canted phase. Thus, we consider a uniaxial anisotropy system in the second-order approximation for the determination of $K_2$ and $K_4$.\textsuperscript{15} The free energy density in the absence of an applied magnetic field is given by

$$E = K_2 \sin^2(\theta) + K_4 \sin^4(\theta) + \frac{1}{2} \mu_0 M^2 \cos^2(\theta),$$  \hspace{1cm} (1)

where $M$ is the magnetization, $\theta$ is the polar angle measured from the film normal, and the last term is the demagnetization energy. Minimization of the total energy with respect to $\theta$ results in three possible stable phases: perpendicular ($\theta = 0^\circ$), in plane ($\theta = 90^\circ$), and canted [$\sin^2(\theta) = (\frac{1}{2} \mu_0 M^2 - K_2)/2K_4$]. Note that the transition thickness from the perpendicular phase to the canted phase corresponds to the $t_s$ mentioned above. We can also determine the $K_{2s}$ and $K_{4s}$ from a fit to the observed canted angle in Fig. 4(a) by combining Eq. (1) and the phenomenological models of $K_2 = K_{2b} + K_{2s}/t$ and $K_4 = K_{4b} + K_{4s}/t$, which consider bulk and surface (or interface) contributions. The solid lines in Fig. 4(a) represent the best fit by taking into account the Co bulk values of $M = 14400$ kA/m, $K_{2b} = 453$ kJ/m$^3$, and $K_{4b} = 144$ kJ/m$^3$.\textsuperscript{14} We obtained $K_{2s} = +0.78$ mJ/m$^2$ and $K_{4s} = -0.04$ mJ/m$^2$, $K_{2s} = +0.62$ mJ/m$^2$ and $K_{4s} = -0.05$ mJ/m$^2$, and $K_{2s} = +1.04$ mJ/m$^2$ and $K_{4s} = -0.13$ mJ/m$^2$ for the smooth, $\varphi = 0^\circ$, and $\varphi = 80^\circ$ sputtering angles, respectively. In Fig. 4(b), we demonstrate the variation of $K_{2s}$ and $K_{4s}$ with respect to the sputtering angle. The value of $K_{2s}$ is increased up to 33% and decreased down to 32% in magnitude. The $K_{4s}$, which plays an important role in determining whether the transition proceeds via a continuous canted phase or an irreversible coexistence phase, varies from $-0.02$ to $-0.13$ mJ/m$^2$ depending on the substrate roughness. The increased value for $K_{2s}$ favors perpendicular magnetic anisotropy persisting up to larger thickness of Co film and vice versa. Therefore we claim that the large variation of $K_{2s}$ and $K_{4s}$ is responsible for the contrasting onset thicknesses as well as transition range for artificially roughened substrates. This work was supported by the Korean Ministry of Science and Technology through the Cavendish-KAIST Cooperation Program.