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Full Length Research Paper

Morphology and properties of radio frequency (RF) sputtered cobalt thin films

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Cobalt (Co) films prepared by radio frequency (RF) sputtering for 5 to 55 min were studied by atomic force microscopy (AFM) and vibrating sample magnetometry. The root mean square surface roughness quantified from 1 × 1 μm^2 AFM images tended to increase with increasing sputtering time above 20 min. Compared to hysteresis loops in the case of perpendicular magnetic field, the ferromagnetic Co films of thickness ranging between 120 and 400 nm exhibited smaller saturation field and larger maximum susceptibility when the applied magnetic field was parallel to their surface as a result of the in-plane anisotropy. In the case of thickness over 300 nm, both in-plane and perpendicular magnetizations were markedly increased, whereas the electrical resistance was substantially decreased because of surface oxidation and the geometric alignment of the grains. Since the increase in thickness over 300 nm only enhanced the coercive field in the case of perpendicular magnetization, the coercive field was likely influenced by the elongated grain grown perpendicular to the film surface.

Key words: Radio frequency (RF) sputtering, magnetic thin film, cobalt, atomic force microscopy (AFM), vibrating sample magnetometry (VSM).

INTRODUCTION

Cobalt (Co) thin films are important ferromagnetic materials in sensing and recording. The magnetic properties are manifested by the plot of their magnetization in response to an externally applied magnetic field or the hysteresis loop. Compared to other ferromagnetic materials, Co films which can be prepared by various techniques, including electrodeposition (Manhabosco and Muller, 2009; Lee et al., 2010), metal-organic chemical vapour deposition (Ko et al., 2003; Chioncel et al., 2007), evaporation (Jergel et al., 2009; Szmaja et al., 2010) and sputtering (Bergenti et al., 2007; Kumar and Gupta, 2007) possess a strong magnetic anisotropy. It follows that hysteresis loops measured with in-plane and perpendicular magnetic fields are easily distinguished. The effect of magnetocrystalline anisotropy is reportedly decreased with increase in Co film thickness from 50 and 195 nm (Kharmouche, 2011). Other contributions to anisotropy arise from surface and shape of the structures. Perpendicular anisotropy as well as magnetic stripe domains tends to develop when the films become thicker than 50 nm (Chioncel et al., 2007). In additions, the crystal structure, stress relaxation and grain size are changed as the film thickness is increased (Sharma et al., 2007). For this reason, magnetic properties of electrodeposited Co films on copper substrates were studied by magneto-optic Kerr effect (MOKE) magnetometry in both thick (Bhuiyan et al., 2008) and ultrathin (Mangen et al., 2010) regimes. The magnetic hysteresis is also influenced by other factors, such as the buffer layer (Vahaplar et al., 2009). Hysteresis loops of ultrathin Co films grown by electron-beam evaporation were modified by the addition of a copper overlayer (Chan et al., 2007). For a silicon substrate, the formation of cobalt disilicide at the interface was observed and its effect on magnetic properties was

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revealed by MOKE spectra (Agarwal et al., 2006).

In this work, magnetic properties of Co films prepared by radio frequency (RF) sputtering for various times were studied. Vibrating sample magnetometry (VSM) which measured the flux induced by the oscillating magnetic films was employed instead of conventional use of polarization measurement by MOKE magnetometry. The trends in magnetic as well as electrical properties were then compared with the morphology obtained from atomic force microscopy (AFM) which is a proven technique for investigating organic surfaces (Sowwan et al., 2008; Yap et al., 2011) as well as inorganic Co films (Bergenti et al., 2007; Sharma et al., 2007; Szmaja et al., 2008; Manhabosco and Muller, 2009; Lee et al., 2010; Szmaja et al., 2010).

MATERIALS AND METHODS

A Co disk of 99.99% purity was used as a starting material to prepare Co films by RF sputtering (13.5 MHz, 200 W) onto glass substrates. The RF sputter (Leybold-Heraeus, Univex 300) has a cylindrical chamber of 29.5 cm in diameter and 31.5 cm in height. The base pressure inside the chamber was about 2×10^{-4} mbar and the pressure during deposition was of the order of 10^{-3} mbar by the flow of 32 sccm argon. The time of Co sputtering on the substrate varied from 5 to 55 min. The film thickness was measured after the deposition from side-view micrographs obtained by scanning electron microscopy (SEM).

The electrical resistance of the films was measured by a standard 4-point probe technique. The magnetic properties of Co films were measured by vibrating sample magnetometer (Lakeshore, 7400) within ± 800 $kA.m^{-1}$ magnetic field applied in 2 different directions, that is, parallel and perpendicular to the surface of Co films. To increase the accuracy, the signal from a bare glass substrate was also collected and used in the subtraction from each substrate with Co deposits. The coercive field was subsequently determined from the x-intercept of the obtained hysteresis loop whereas the y-intercept represented the remanent magnetization. The magnetization in response to the maximum field was estimated as the saturation magnetization and the changing slope corresponded to the magnetic susceptibility. The surface morphology of each Co film was characterized by atomic force microscopy (Asylum Research, MFP-3D) with 1×1 μm^2 view.

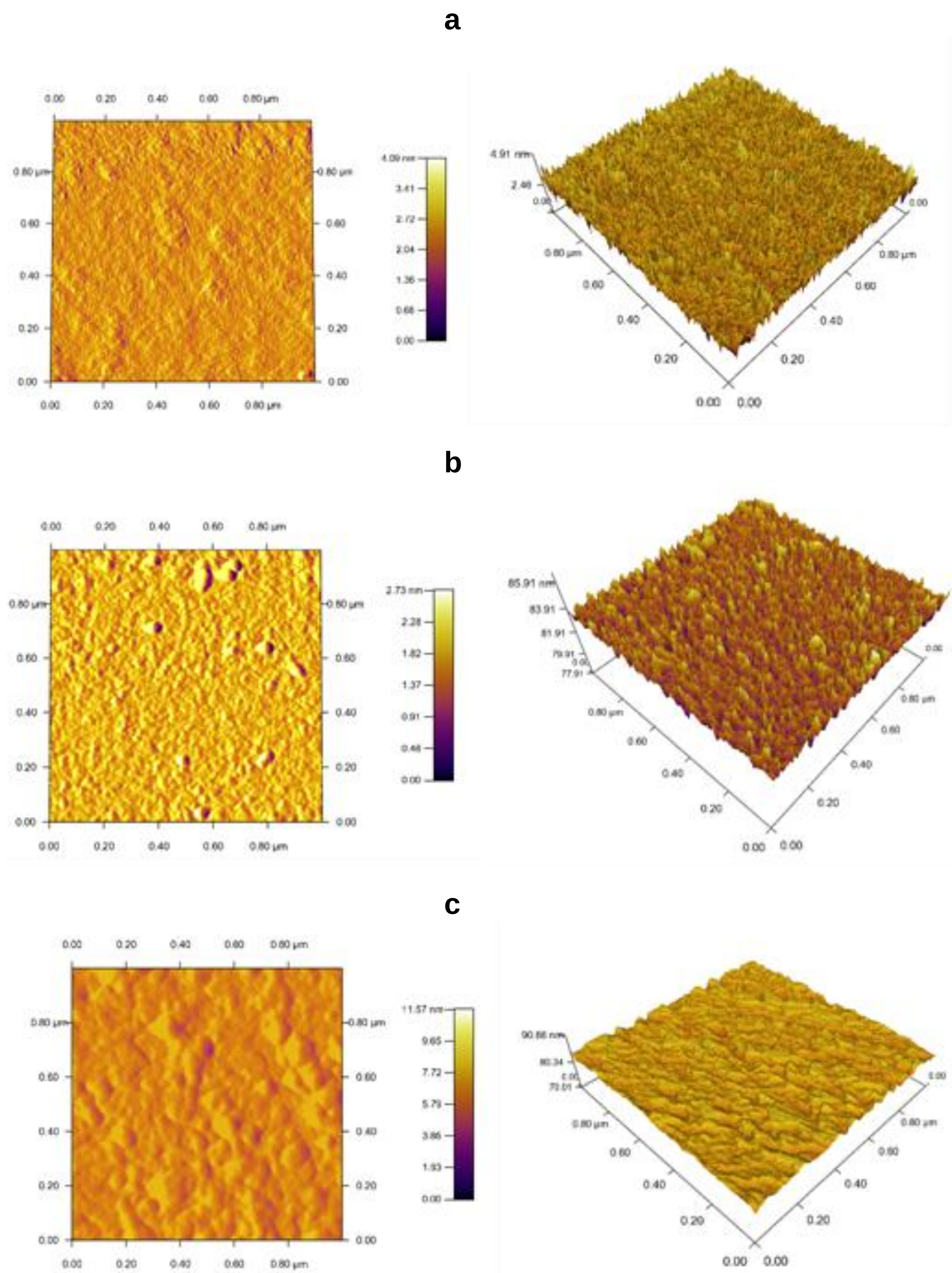
RESULTS AND DISCUSSION

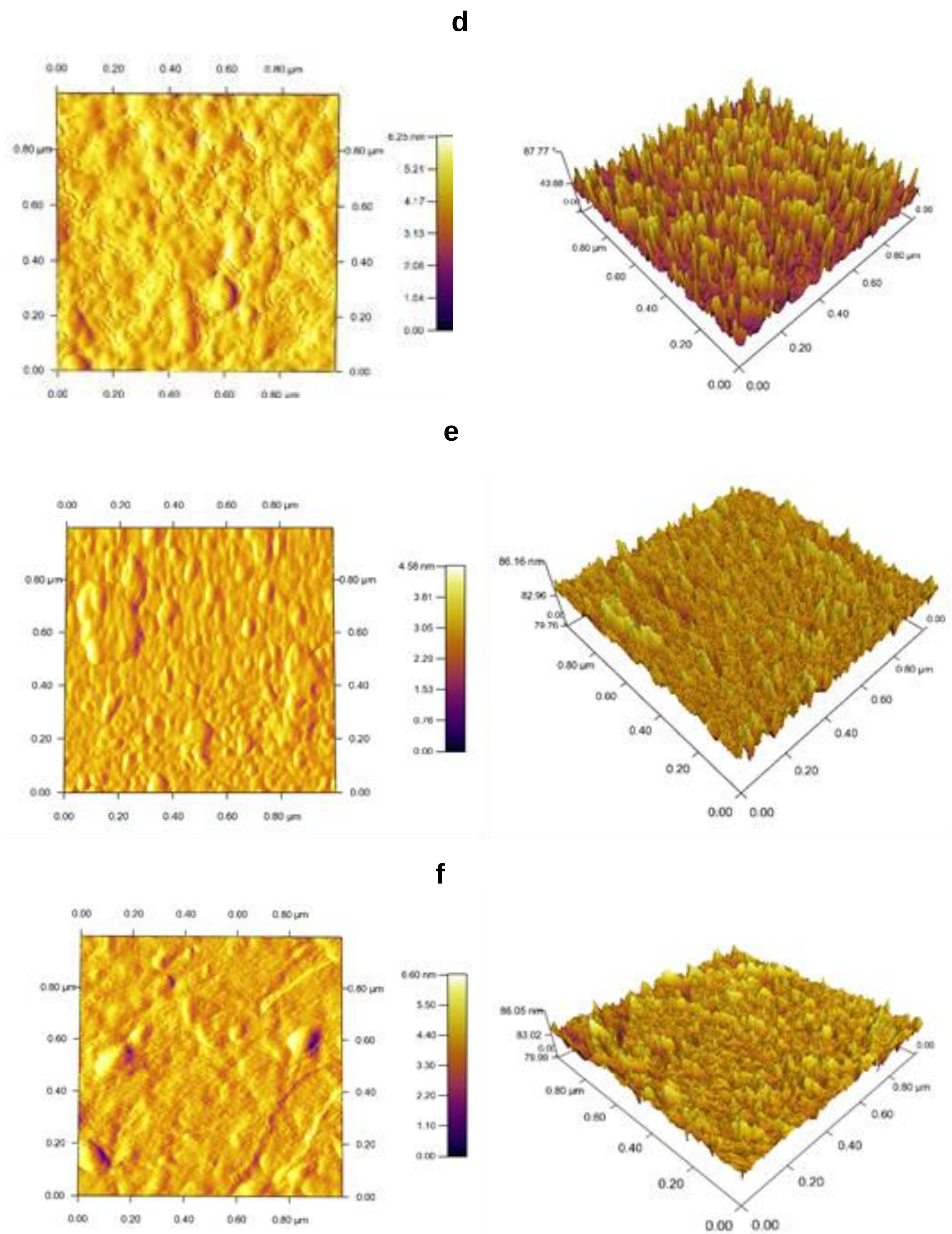
Figure 1 shows 1×1 μm^2 AFM images of Co films. Overall, the film morphology is generally continuous, but microcracks are detected in some areas of the samples. With the shortest sputtering time in Figure 1b, only some grains are randomly deposited on the glass substrate. The grainy morphology is clearly observed after longer sputtering times. Also, it was previously shown by AFM images of evaporated Co films that the grains can be closer together and the film became more continuous with longer deposition time (Szmaja et al., 2008). The films in Figure 1c to i from 10 to 55 min sputtering are clearly composed of many sub-micron polygon grains which are uniform in size in some areas and comparable to the literature (Lee et al., 2010). With increase in sputtering time, the grain growth in the direction parallel to the film surface is still not conclusive, but the homogeneity in deposition is increasingly disrupted by random deposits. It follows that the roughness is inevitably increased after the prolonged sputtering. Figure 2 compares the root mean square surface roughness, quantified from these AFM images, of Co films from 8 different sputtering times. The roughness, comparable to that of Co films prepared by ion-beam sputtering (Kumar and Gupta, 2007), shows a linear increase from 0.36 to 1.08 nm with increase in sputtering time from 20 to 55 min.

From SEM micrographs as exemplified in the insets of Figure 3, the thickness of Co films is averaged. The thickness plotted in Figure 3 is approximately proportional to the time of sputtering. This linear correlation can be used to extrapolate the thickness in the case of 5 to 15 min sputtering which cannot be determined from SEM micrographs and the deposition rate of 7 to 9 nm/min was estimated from this experiment.

In this experiment, magnetic properties of film thinner than 100 nm cannot be accurately detected because of the instrument limit and the effect from magnetic-nonmagnetic interfaces. Figure 4 compares hysteresis loops of Co films from 25 to 55 min sputtering corresponding to the thickness from 122 to 395 nm in both in-plane and perpendicular magnetizations. The sputtered Co films exhibit ferromagnetic properties with saturation magnetization in response to ± 800 $kA.m^{-1}$ magnetic field. The saturation field is smaller and the maximum susceptibility is larger when the magnetic field is applied in parallel to the surface of each film. Although, the appearances of the loops somewhat deviate from ideal uniaxial anisotropic characteristics of easy and hard axis magnetizations (Kumar and Gupta, 2007), the preference of in-plane anisotropy is evident.

The magnetic parameters are listed in Table 1. In general, the magnetizations may not be proportional to the film thickness, because of the variation in crystal and grain structures with the film thickness (Sharma et al., 2007). In this case, the magnetizations in the 122 and 250 nm-thick Co films are even comparable. This discrepancy can only be described by the effect of oxidation at the surface which is pronounced in the thinner films, but become negligible when the entire film is much thicker than this oxidation layer. The much higher remanent and saturation magnetizations in the 332 and 395 nm-thick Co films are typical signs of grain growth in magnetic films (Fouad, 2008). Interestingly, the coercive fields of these films are also enhanced only in the case of perpendicular magnetization. These magnetic properties imply that longer sputtering times give rise to grains elongated perpendicular to the film surface and the shape anisotropy is developed (Itoh et al., 2002). By contrast, the coercive field of every Co film is relatively constant in


a
0.00 0.20 0.40 0.60 0.80 µm
4.09 nm
4.91 nm
b
0.00 0.20 0.40 0.60 0.80 µm
2.73 nm
85.91 nm
c
0.00 0.20 0.40 0.60 0.80 µm
11.57 nm


d
e
f

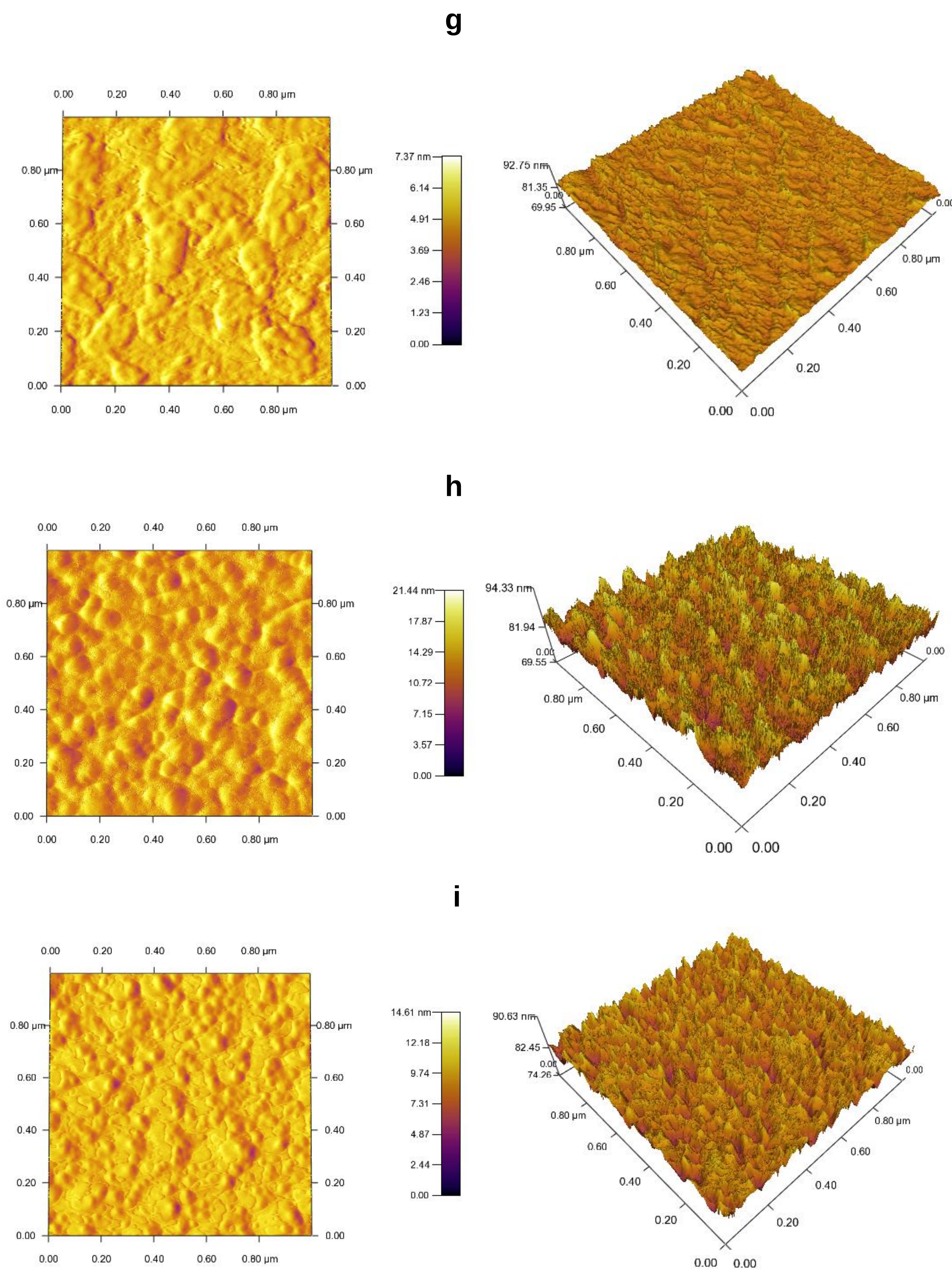


Figure 1. AFM images of (a) a glass substrate and Co films after sputtering for (b) 5, (c) 10, (d) 15 (e) 20, (f) 25, (g) 35, (h) 45 and (i) 55 min.

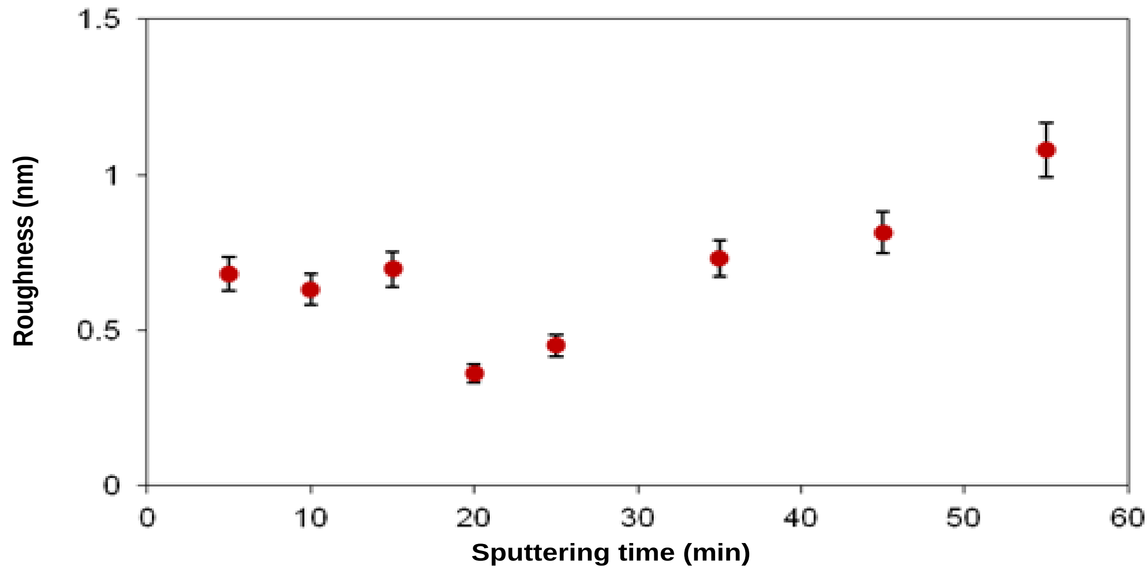


Figure 2. Variations of root mean square surface roughness with the sputtering time of Co films.

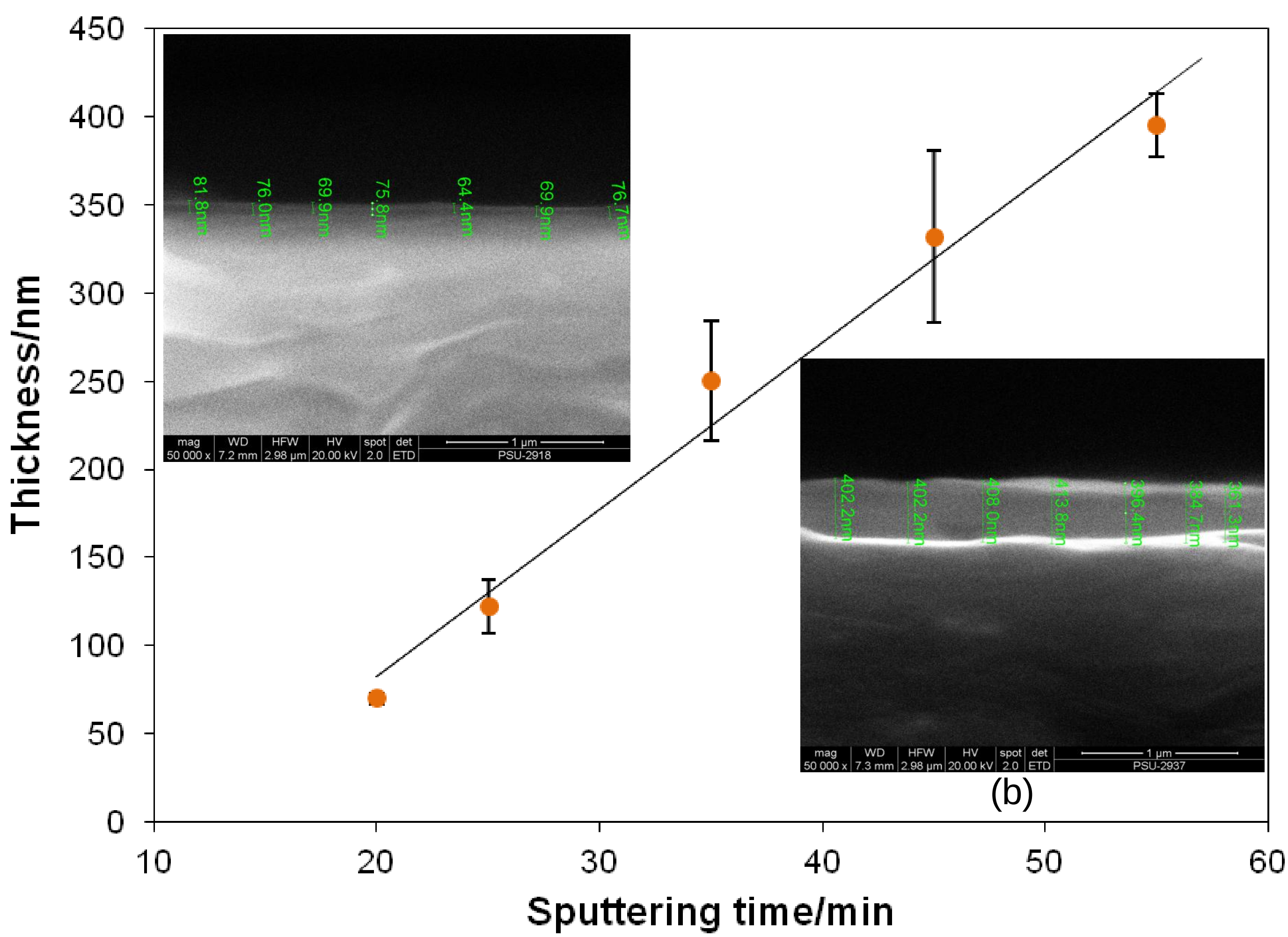


Figure 3. Variations of thickness with the sputtering time of Co films. Insets are examples of side-view SEM micrographs of Co films from (a) 20 min and (b) 55 min sputtering.

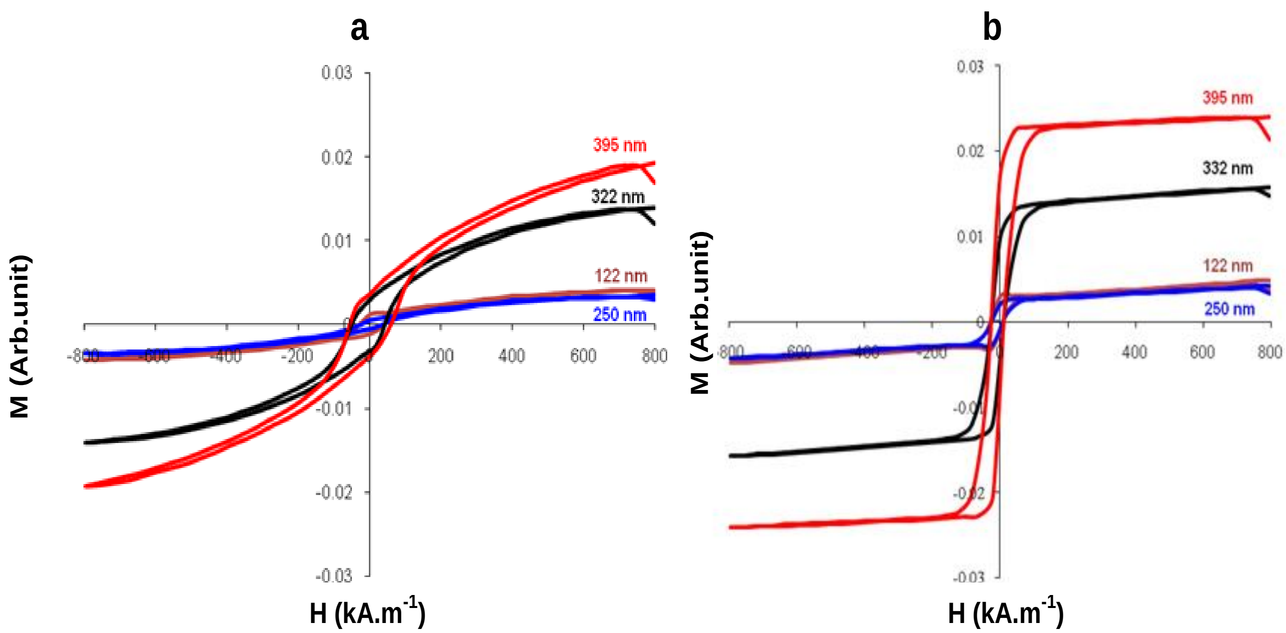


Figure 4. Hysteresis loops in the case of the magnetic field is (a) perpendicular and (b) parallel to the surface of Co films of varying thickness.

Table 1. Coercive field (H_c), saturation magnetization (M_s) and remanent magnetization (M_r) of Co films from various sputtering times.

Sputtering time (min)	Film thickness (nm)	Perpendicular			In-plane		
		H_c (kA.m^{-1})	M_s (Arb.unit)	M_r (Arb.unit)	H_c (kA.m^{-1})	M_s (Arb.unit)	M_r (Arb.unit)
25	122 ± 15	39.0	0.0041	0.0013	9.6	0.0048	0.0025
35	250 ± 34	36.6	0.0035	0.0005	10.0	0.0043	0.0015
45	332 ± 49	43.0	0.0140	0.0030	11.9	0.0157	0.0075
55	395 ± 18	59.7	0.0192	0.0037	12.7	0.0240	0.0130

the case of in-plane magnetization, because the grain growth is not clearly observed in the direction parallel to the film surface. The strong dependence of hysteresis loops on the direction of applied magnetic field was also observed in other sputtered Co thin films (Bergenti et al., 2007; Kumar and Gupta, 2007; Sirisathitkul et al., 2012) because of the strong anisotropy of Co. Such anisotropy is not observed in sputtered nickel films of comparable thickness (100 to 300 nm) whose coercive fields are constant regardless of the magnetic field direction (Rattanasakulthong et al., 2011).

Figure 5 shows a large reduction in electrical resistance when the thickness of Co films is above 300 nm. Like the substantial variation in magnetizations, this nonlinear trend can also be described by the effect of surface oxidation. This surface oxidation and the geometric alignment of the grains have pronounced effects on the electrical conduction and magnetizations. On the other hand, change in roughness has no correlation with the magnetization. For an example, the 332 and 395 nm-thick Co films have much higher magnetizations, but AFM images indicate roughness values marginally higher than those of the thinner films. However, the effect of surface roughness on the coercive field cannot be ruled out and it has been shown that coercive field is increased by enhanced roughness of magnetic films' surface, because the domain wall motion is impeded (Jergel et al., 2009; Chuang et al., 2011).

Conclusions

The sputtered Co films of thickness between 120 and 400 nm exhibited the preference of in-plane anisotropy. In this thickness regime, the root mean square surface roughness was increased with increase in sputtering time from 25 to 55 min. When the thickness of these films is over 300 nm, the magnetizations substantially rose and

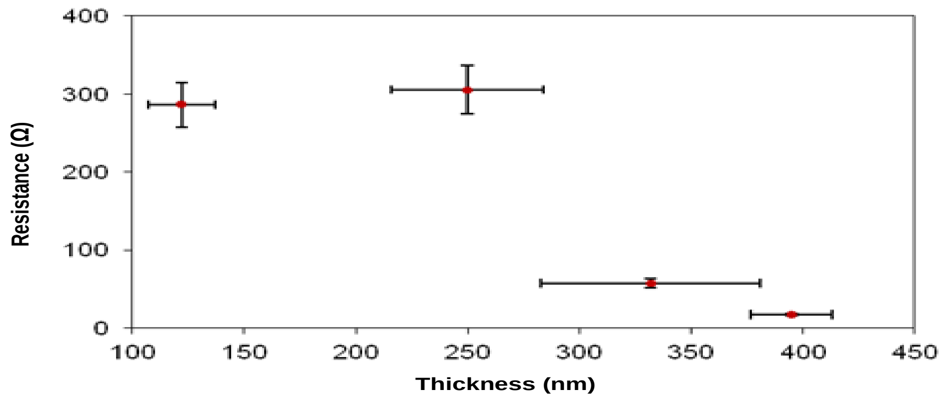


Figure 5. Variations of resistance with the thickness of Co films.

the electrical resistance was significantly decreased. These sudden changes are likely attributable to the surface oxidation and the shape anisotropy of grains elongated perpendicular to the films surface. The latter was also the origin of the enhanced coercive field exclusively in the case of perpendicular magnetization as the thickness was increased.

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