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Effect of Si layer thickness on the structural properties of a Co/Si multilayer system

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Abstract

The nature of the interface involved and the structure characteristics of the as-deposited Co/Si multilayer system have been studied. Using the ion-beam sputtering technique, multilayers, having ten bilayers of Co and Si, were deposited. X-ray reflectivity, x-ray standing wave and wide-angle x-ray diffraction techniques were used to study the interface of the system. The x-ray reflectivity curves obtained confirm the good quality of the Co/Si stack. The presence of a silicide layer at the interface is not distinctly visible. The Co is found to be crystalline in nature and it is highly textured. This texture of the Co layer is lost as the Si layer thickness is increased above a particular value. We have performed some preliminary magneto-optical Kerr effect (MOKE) measurements at room temperature, which show that all the samples have ferromagnetic coupling between the different Co layers and that the nature of ferromagnetism is highly governed by the Si layer thickness.

1. Introduction

Over the last few years, metal–metal multilayers (MLs) have been extensively studied due to their potential application in electronics. Co/Si is one such system that shows interesting magnetic behaviour. This system is reported to show superparamagnetic behaviour [1] for low thickness of the Co layer (∼22 Å), whereas for Co layer thickness of the order of 100 Å, it shows an oscillatory magnetic behaviour between different Co layers as a function of Si layer thickness [2–4]. This type of coupling is well established and understood in the case of metal/metal multilayers [5]. However, the picture is not clear for an insulating or semiconducting spacer layer. In this context the Fe/Si system, which shows similar behaviour, has been well studied [6]. Reports have suggested that the formation of an iron silicide layer at the interface might be responsible for such oscillatory behaviour in the Fe/Si system [6–8].

In the similar way, there is a finite possibility that the origin of this type of interesting magnetic behaviour of the Co/Si system might also depend on a characteristic of the interface

i.e. on the formation of a cobalt silicide layer at the interface. This is because Co and Si are also known to have negative heat of mixing, which encourages mutual solubility at the interface and the formation of intermetallic alloys of varying composition [9–11]. It has also been shown that both evaporated [12] and sputtered [13] Co/Si multilayers have amorphous interlayers. High-resolution electron micrography results of Co and Si show that for a small thickness of Co (∼30 Å), the metal is converted to an amorphous silicide layer whereas thicker Co layers can retain an unalloyed part [13]. For thin Co and Si layers (∼50 Å), elemental Co or Si may not be present, but instead intermetallic alloy gets formed with an average composition. Thus, all these studies suggest that Co and Si have a strong tendency of forming alloys at the interface. However, this system has not been studied extensively and information is still lacking which explains the interfacial characteristics and the structure of the Co/Si multilayer system. Hence, it is still not clear whether the Si layer thickness or the formation of silicide is playing the dominant role in governing the magnetic behaviour of the system.

For this reason, we have undertaken the present investigation and have prepared Co/Si MLs of varying thickness to study the interlayer coupling in the as-deposited samples. X-ray reflectivity (XRR), x-ray standing wave (XSW) measurements and x-ray diffraction (XRD) measurements on the as-deposited Co/Si MLs have been performed. Some preliminary magnetic characterization of samples at room temperature has also been done by magneto-optical Kerr effect measurements (MOKE).

2. Experimental details

On Si(100) substrates, six multilayers, designated as A, B, C, D, E and F, of Co and Si with ten bilayers having varying thickness of Co (∼45–80 Å) and Si (∼20–75 Å) were deposited by the ion-beam sputtering (IBS) deposition technique. Bilayers having the same thickness of Co and Si were also deposited on both Si(100) as well as float glass substrates, designated as BL1 and BL2 respectively. The substrates were cleaned properly by chemical cleaning using nitric acid and ultrasonic cleaning using acetone. Before deposition the rms roughness of the substrate was obtained by XRR measurement. During the deposition, the flow rate of argon gas was kept at 4.5 standard cubic centimetres per minute. The total pressure in the chamber during the deposition was maintained at $\sim 10^{-4}$ Torr. To calibrate the thickness of the Co and Si layers, a number of samples were deposited. The deposition rate was kept at ∼0.5 Å s^{-1}.

To obtain information about the thickness of the individual layers, surface and interface roughness, interdiffusion and the formation of silicide layer, XRR and XSW measurements were performed in our laboratory using a sealed tube x-ray generator with a Cu target (1.54 Å), operated at 40 kV and 40 mA. The details for the XSW measurements have been described elsewhere [14]. To check the formation of silicide layer and also to check the whether the Co and Si layer that is being deposited is in crystalline or amorphous form, we have performed wide-angle x-ray diffraction (XRD), using a rotating-anode x-ray generator from Rigaku equipped with a Cu target, operated at 50 kV and 200 mA. For MOKE measurements we have used polarized light from He–Ne laser of wavelength 632.8 Å. Applying a magnetic field parallel to the surface of the films, hysteresis loops were recorded up to saturation magnetization.

3. Results and discussion

3.1. X-ray reflectivity

Figure 1 shows the x-ray reflectivity patterns for all the six as-deposited ML samples. Bragg peaks arising due to ML periodicity are distinctly visible up to eighth order. For the theoretical fit to the experimental data, a Parratt recursion formalism is used [15].

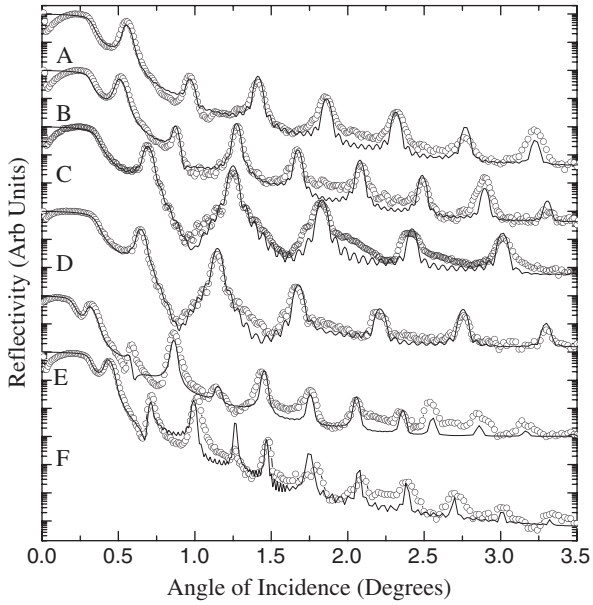

Figure 1. XRR pattern of the Co/Si MLs. Circles are experimental data. For the sake of clarity the data have been shifted along the *y*-axis; continuous lines show the fit to the data.

X-ray reflectivity is a good technique to study the details of the nature of surfaces and interfaces involved. To the best of our knowledge none of the reports on the Co/Si multilayer system has shown a detailed analysis of the x-ray reflectivity data, except for work done by Fallon *et al* [11]. They have fitted the x-ray reflectivity data, considering the formation of a silicide layer at the interface. The quality of fitting shown in the report is not very good, indicating that the model they have chosen might not be correct. Since our aim was to study the nature of the interface in Co/Si ML system, we have made a detailed analysis of the XRR data. Allowing for the variation of various parameters like electron density, layer thickness and surface and interface roughnesses of the layers, the experimental data were fitted. We found that the fitting is quite good when no silicide layer formation at the interface is considered, as shown in figure 1. The slight difference in the fitted and the experimental data, seen in figure 1, might be due to the thickness inhomogeneity in various layers of Co and Si.

In multilayer structures, since a large number of layers are involved, a slight error in parameters like thickness, roughness, or electron density of the individual layers affects the reflectivity pattern. To avoid this, two bilayers of Co and Si of the same thickness on both Si(100) and float glass substrates were prepared. Here in these bilayer samples Co is deposited first on both the substrates.

We have made x-ray reflectivity measurements on both these bilayers. The patterns are shown in figures 2(a) and (b), for the bilayers deposited on Si(100) (BL1) and float glass (BL2) substrates respectively. It can be seen that there is no difference in the x-ray reflectivity pattern of both these bilayers grown on Si(100) and float glass substrates. We have fitted these patterns assuming no silicide layer at the interface. It can be seen that there is an excellent fit with this model. The quality of fit confirms that our model is correct and hence no silicide layer is distinctly visible at the interface. The fitted values of the thickness and roughness of Co and Si for all the multilayers as well as the bilayer samples are given in table 1, and the parameters are found to be physical.

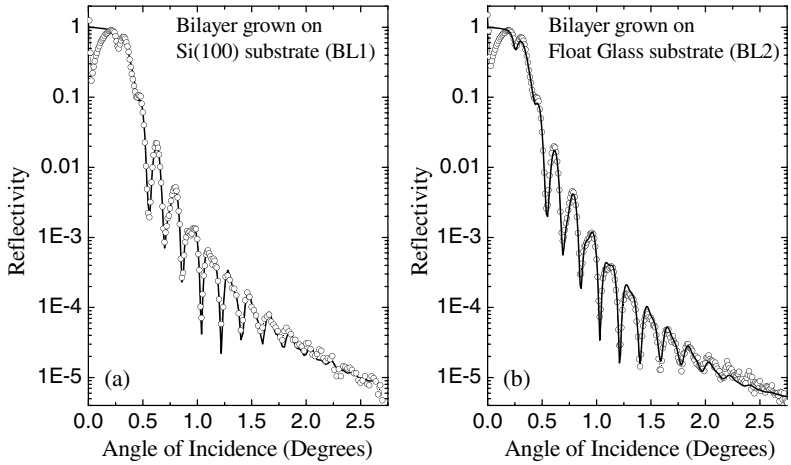

Figure 2. XRR pattern of the Co/Si bilayer samples: (a) on Si(100) substrate, BL1; (b) on float glass substrate, BL2. Circles are experimental data. Continuous lines show fits to the data.

Table 1. Parameters obtained from the reflectivity fitting of multilayers and bilayers.

	Co layer		Si layer	
Samples	Thickness (Å)	Roughness (Å)	Thickness (Å)	Roughness (Å)
A	48	10	48.5	3.6
B	51	9	56	4
C	53.5	4	20.1	8
D	59.5	4	22	7
E	73.8	7.7	70.5	4
F	80	11	60	5.5
BL1	231	8.3	219	7.6
BL2	228.5	8.5	221	8.3

However, Petford-Long *et al* [12] and Ruterana *et al* [13] have shown that both evaporated as well as sputtered Co/Si multilayers have an amorphous interlayer. High-resolution electron micrographs have suggested that for small, nominal Co thicknesses (∼3 nm) the metal is converted to an amorphous silicide layer. Thicker Co layers can retain a part that is unalloyed. For relatively thin Co and Si layers ($\leqslant$50 Å), elemental Co and Si may not be present, and instead, an alloy forms with an average composition. Moreover, Naik *et al* [16] have also demonstrated that, using XRR data, even the formation of a ∼12–15 Å thick silicide layer in the Fe/Si system could be detected. This suggests that in our Co/Si MLs also, if a silicide layer is present, its thickness or volume fraction is so low that it could not be detected distinctly as a layer by XRR measurements. This may also mean that it might be present as a discontinuous layer, which in turn gives a large roughness (as shown in table 1).

3.2. X-ray standing wave

To further confirm the x-ray reflectivity results, we have also performed XSW measurements. It is well known that XSW is an interesting technique to obtain information on the depth distribution of the elements in the layered material. Moreover, for periodic MLs, the shape of the XSW intensity near a Bragg peak is sensitive to a change in the composition [17]. Thus,

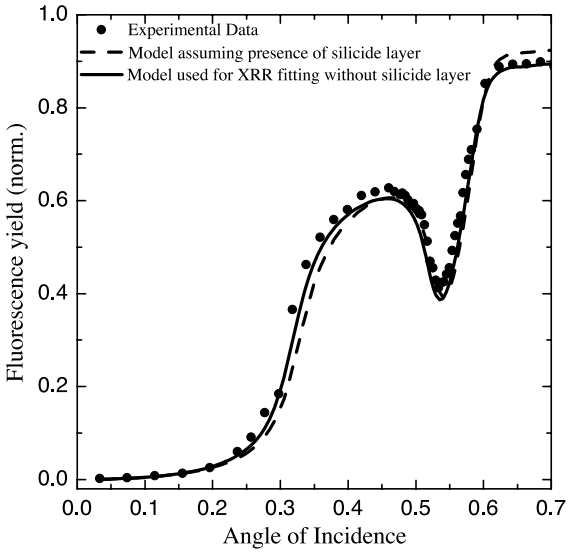

Figure 3. XSW pattern of the Co/Si multilayer system A. Circles are experimental data. The dashed line represents the model assuming the presence of a silicide layer at the interface. The solid line represents the model that contains all the parameters obtained from the fitting of the XRR data (i.e. without a silicide layer).

if any type of segregation or diffusion is taking place at the interface, it can be analysed by the XSW method. A typical curve for XSW measurements is shown in figure 3. Here we have shown the data and analysis only for sample A; all the other ML samples show similar behaviour. Here the Co Kα fluorescence yield has been measured over an angular region containing the first-order Bragg peak. The modulation in Co fluorescence intensity in the Bragg region is due to x-ray standing wave field generated in the ML structure. To the best of our knowledge no reports show XSW measurements on Co/Si system to study the nature of its interface. For the theoretical fitting [18] of this data, we have considered two models.

In the first model we incorporated all the parameters extracted from the reflectivity fit, whereas in the second model we considered a silicide layer at the interface. The fit to these models is shown by the solid and dashed line, respectively, in figure 3. It can be seen that the fitting for the first model, in which we have incorporated all the parameters of the XRR analysis, is better than that for the model, which assumes a silicide layer at the interface. This result is consistent with the results of our reflectivity fitting and indicates that a silicide layer is not present at the interface, and even if one is present, then its thickness is very small.

3.3. X-ray diffraction

We have also performed wide-angle XRD investigations of all six samples. Figure 4 shows the XRD patterns of the as-deposited Co/Si MLs and the bare Si(100) substrate. A strong peak at around $2\theta = 33^\circ$ corresponds to the Si substrate. Other very small and fine peaks which are observed are due to the substrate. It can also be observed that the XRD pattern of the as-deposited Co/Si multilayers A, B, C and D shows a broad and well-defined peak at around $2\theta = 44^\circ$. This intense and broad peak corresponds to the (002) hcp planes of Co. The presence of this peak indicates that the cobalt in these MLs is crystalline in nature and is highly oriented in the [001] direction. This result is consistent with reports by

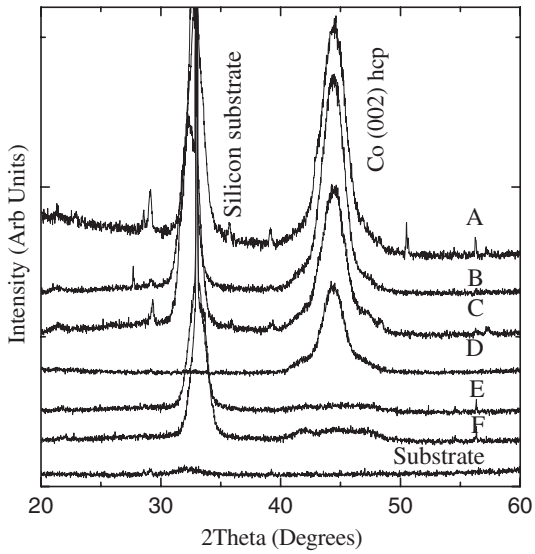

Figure 4. Wide-angle XRD patterns of all the MLs along with the Si(100) substrate. For the sake of clarity the data have been shifted along the y-axis. The presence of a broad peak at $2\theta \sim 44^\circ$ for samples A, B, C and D indicates that the Co is crystalline in nature and that the films are highly textured.

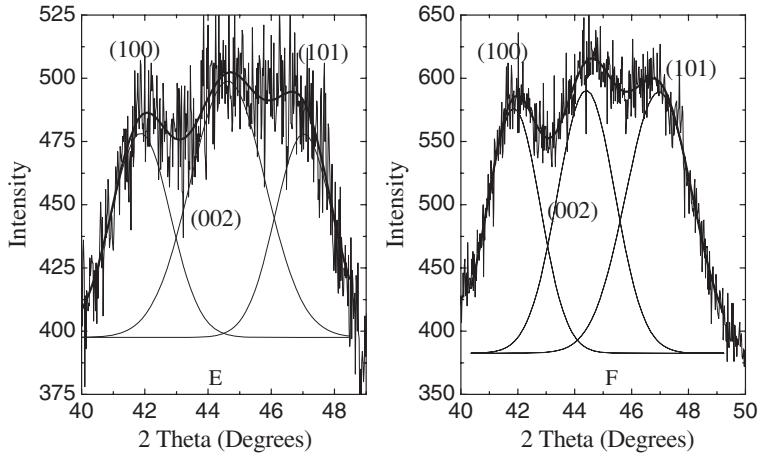

Figure 5. Magnified wide-angle XRD patterns for samples E and F. The single peak corresponding to the (002) plane of Co, as seen in figure 4, has been broken into three different peaks. These three peaks correspond to the (100), (002) and (101) planes of Co.

Fallon *et al* [9–11]. However, it can also be noted from the figure that the intensity of this peak decreases for samples E and F, which have a high thickness of Co as well as Si layers. It is almost seen as a flat background in the figure. However, if we magnify only the portion containing this peak for samples E and F (as shown in figure 5), we will find that instead of one peak there are three different peaks. These three peaks are identified as reflections from the hcp (100), (002) and (101) planes at d-spacing close to those of hcp cobalt. This indicates that the films, which were otherwise strongly textured for low thickness of Co and Si layers,

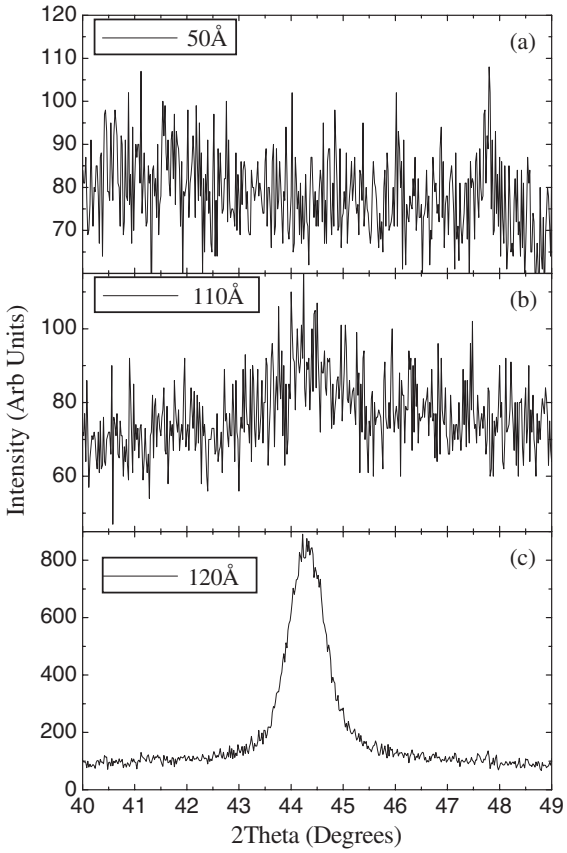

Figure 6. XRD pattern of Co thin-film samples deposited on Si(100) substrates of thickness (a) 50 Å, (b) 100 Å and (c) 120 Å.

lose their texture for higher thickness of Co and Si layers. Since in these two samples, E and F, the thickness of both the Co and Si layers is rather high, it is difficult to say if the Co or the Si layer is forcing the Co layer to lose its texture. To check this we performed x-ray diffraction measurements on pure Co films of various thickness grown on Si(100) substrate. The XRD patterns for Co films having different thickness are shown in figure 6. It can be seen that up to a thickness of 120 Å for the Co layer, it remains textured. It is therefore clear that Co is losing its texture not due to any increase in its thickness beyond a certain limit. To further confirm this we performed x-ray diffraction measurements on the both the bilayer samples BL1 and BL2, where the Si layer is 220 Å thick and the Co layer is 230 Å thick. We have chosen such high thicknesses since we needed thicknesses above those we have in the ML samples E and F. The x-ray diffraction data for both these bilayers are shown in figure 7.

It can be seen from figure 7 that the bilayer grown on Si(100) substrate has lost its texture. This is evident from the three peaks that are distinctly present. A similar effect is seen for the bilayer sample grown on float glass substrate. But in this sample it can be seen that although the texture of the Co layer is lost the three peaks are also not distinctly visible. This might be because in this particular case the Co layer is grown on an amorphous substrate, i.e. float glass, and hence it might initially not be textured, and the deposition of the Si layer above the Co layer must have enhanced this effect.

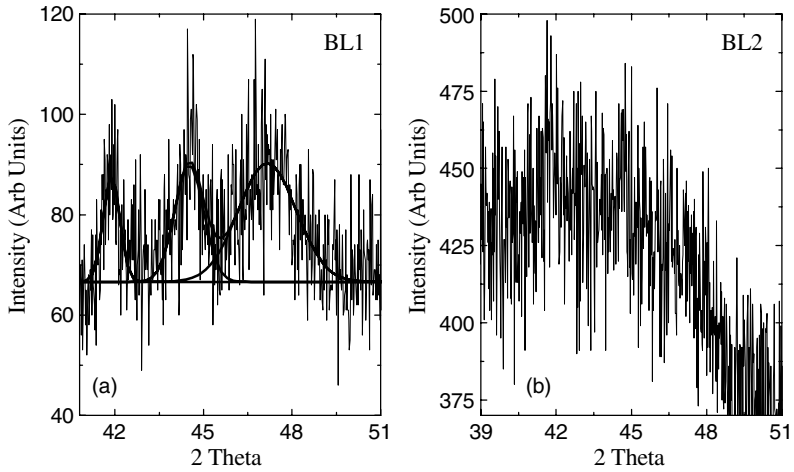

Figure 7. XRD patterns of the bilayer samples: (a) BL1 on Si(100) substrate, and (b) BL2 on float glass substrate.

Table 2. Correlation length calculated using Scherrer's formula and the value of coercivity and F_{AF} parameter as obtained from the hysteresis loops, for all the as-deposited ML samples.

Samples	Correlation length (ξ)	Coercivity (H_c)	F_{AF} parameter
A	38	27.3	0.2
B	39	21.1	0.11
C	40	25.3	0.19
D	41	22.2	0.10
E	32	50	0.2
F	34	37.7	0.33

The crystalline correlation length, ξ, can be estimated from the full width at half maximum (FWHM) of the Co (002) diffraction peak. The correlation length for the samples A, B, C, D, E and F calculated using the Scherrer formula is given in table 2. The absence of Si peaks in the XRD pattern suggests that the Si, a spacer layer, in all these multilayers is amorphous in nature. Thus, we have found that in all the multilayers the Co is crystalline in nature whereas the Si is amorphous. Fallon *et al* [11] have shown that Co in the Co/Si ML system is highly oriented, and the correlation length increases as the thickness of the Co layer is increased. Fallon *et al* [9] have also a reported that Co–Si alloy films shows (002) hcp texture for low concentration of Si, and this texture is lost when the concentration of Si is increased. But none of them have shown the effect of increase in the thickness of the Si layer in the Co/Si ML system. Thus, we have found that the Co layer in Co/Si multilayer system is highly oriented only for a multilayer containing a low thickness of Si layers, and that this texture is lost as we increase the thickness of the Si layer: this thickness is ∼56 Å. No peak due to silicide phase formation in the as-deposited Co/Si ML is seen in the XRD pattern, suggesting that the silicide phase is not present and even if it is present its volume fraction is so low that is presence is not seen in the XRD pattern. The result is consistent with the XRR and XSW results.

3.4. Magneto-optical Kerr effect

MOKE measurements at ambient temperature were carried out on all as-deposited Co/Si multilayers. The MOKE data normalized with respect to the saturation magnetization (M_S)

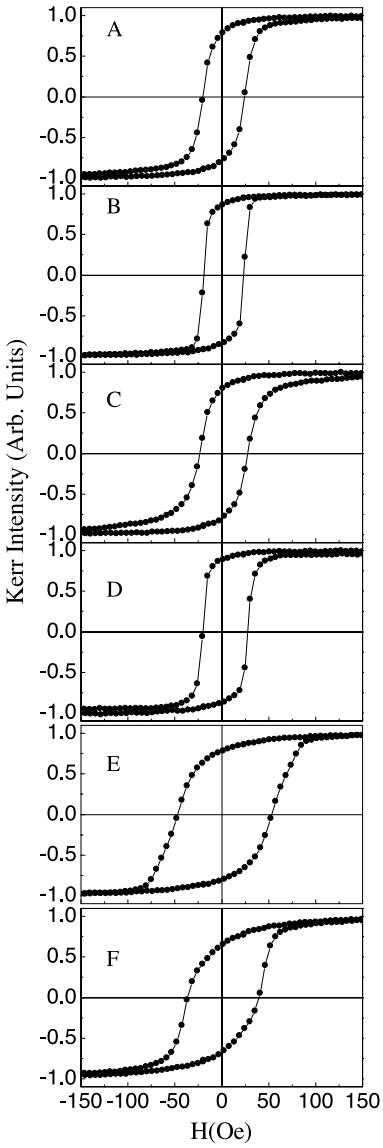

Figure 8. Hysteresis loops for all the as-deposited Co/Si multilayers.

for all the MLs are shown in figure 8. Lucinski *et al* [3, 4] have also presented MOKE results on Co/Si MLs. They have clearly shown [3] that for thickness of Si layer greater than or equal to 15 Å, the hysteresis loop gets modified from a single hysteresis loop to a step-like loop, which is characterized by two coercive fields H_{C1} and H_{C2}. They have even observed an oscillatory behaviour in the F_{AF} $(=1 - M_r/M_S)$ parameter as a function of Si layer thickness, where M_r and M_S are remanent and saturation magnetization respectively. According to them the oscillatory behaviour is observed since the Co–Si nonmagnetic metallic phases replace the nominally pure Si-spacer layer. We have also calculated the F_{AF} parameter as given in table 1. But as can be seen, it shows no oscillatory behaviour as a function of Si layer thickness, though the thickness of the Si layer in our MLs is comparable to what they have reported. However,

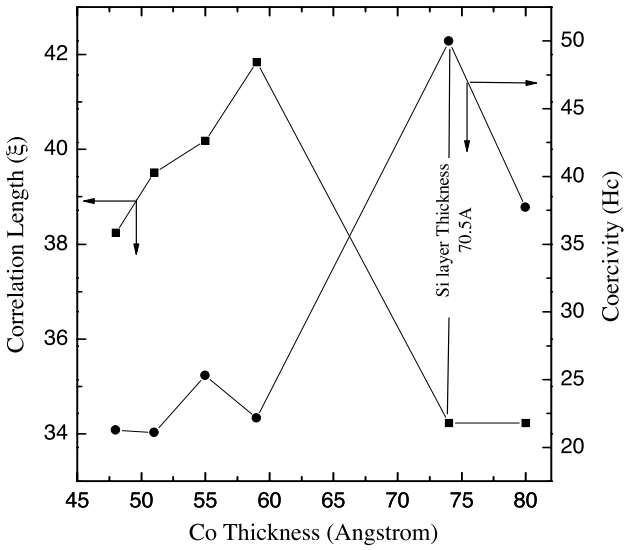

Figure 9. Dependence of correlation length and coercivity on the thickness of the Co layer.

the Co layer thickness as reported by Lucinski *et al* [3] is about 30 Å. In our samples the Co layer thickness is ∼50–80 Å, suggesting that the coupling is different in the samples containing thicker Co layers. It can be even be seen in figure 8 that none of the hysteresis loops show any step-like behaviour, even up to a Si layer thickness of 70 Å, and the shape of the loops and the F_{AF} parameter clearly indicate that the coupling between various Co layers is ferromagnetic.

It can be seen that the recorded hysteresis loops for samples A, B, C and D are almost square in shape; the values of the saturation magnetization and the remanent magnetization are almost equal. This indicates the presence of strong anisotropy in these MLs. Here it should be noted that the magnetic field is applied parallel to the plane of the film. Therefore the shape of the hysteresis loop and the presence of well-defined saturation magnetization with applied magnetic field indicate that the easy axis of magnetization lies in the plane parallel to the plane of the film. But for hcp bulk Co, it is known that the easy axis of magnetization is along the [0001] direction [19]. This direction in our MLs is perpendicular to the plane of the film, as seen from XRD data. It is well known [19] that the magnetization in thin films lies in the plane of the film, because of the huge demagnetization fields would act normal to the plane of the film, if M_S were turned in that direction. The domain in the film extends completely through the film thickness and the walls between them are mainly of the 180° kind, roughly parallel to the easy axis of the films. Thus, in the present case the easy axis for the Co/Si MLs, which show strong texturing, is in the plane parallel to the plane of the film instead of in the direction perpendicular to the plane of the film.

It can also be observed that all these samples (A, B, C and D) have almost equal coercivity, as seen in the figure 8. But samples E and F have large coercivity. Along with large coercivity, these samples even get saturated at higher fields and even the shape of the hysteresis loop has changed from square to smoother ones. This result can be well supported by our XRD data, which show that the samples with low Si thickness (A, B, C and D) are textured and the samples with large Si thickness (E and F) have lost their texture. Loss of texture means that the sample has broken into smaller grains, which is supported by a decrease in correlation length. Now, for these samples the response towards the applied magnetic field for each grain will be different and that is why the coercivity for these samples has increased; also they have large saturation

fields. This effect can also be seen in figure 9, where the correlation lengths of the Co layer and coercivity have been compared. It can be seen that with increase in the thickness of Co layer, the correlation length also increases up to (∼63 Å), whereas the coercivity remains almost equal up to this thickness of Co layer. After this, since the thickness of the Si layer has increased and the Co layer has broken into smaller grains, the correlation length decreases whereas the coercivity has increased. The decrease in coercivity for sample F is seen since for this sample the thickness of the Si layer is less than that for sample E.

Thus, we have observed only the ferromagnetic coupling between the Co layers. The hardness or softness of this ferromagnetic behaviour is highly governed by Si layer thickness. We have not observed any oscillatory behaviour with respect to the Si layer thickness in the interlayer coupling as observed by Lucinski *et al* [3] and superparamagnetic behaviour as observed by Grundy *et al* and Lucinski *et al* [1, 3, 4] in the preliminary MOKE measurements at room temperature.

All these suggest that it is the texture of the Co layer, which depends on the thickness of the Si layer, that affects the magnetic properties of Co/Si system. Since no silicide layer is distinctly visible at the interface, its possible effect on the magnetic behaviour of the system is not clear. Thus, the structural and in turn the magnetic properties of the Co/Si ML system are highly governed by thickness of the Si layer and not by a silicide layer.

4. Conclusions

In contrast to earlier studies of the Co/Si ML system, the presence of a silicide layer at the interface of as-deposited Co/Si samples is not distinctly visible from detailed analysis of XRD, GIXRR and XSW data. The XRD data show that the Co layer is crystalline in nature whereas the Si is amorphous. The Co present in the samples is highly textured up to a particular thickness of the Si layer, and beyond this thickness of the Si layer the Co loses its texture. MOKE measurements show that the interlayer coupling is ferromagnetic in nature and that the nature of ferromagnetism is highly governed by the Si layer thickness. Since no silicide layer is distinctly visible at the interface, its possible effect on the magnetic behaviour of the system is not clear.

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References

[1] Grundy P J, Fallon J M and Blythe H J 2000 *Phys. Rev.* B **62** 9566
[2] Inomata K and Saito Y 1997 *J. Appl. Phys.* **81** 5334
[3] Lucinski T, Kopcewicz M, Hutten A, Bruckl H, Heitmann S, Hempel T and Reiss G 2003 *Mater. Sci.* **21** 25
[4] Lucinski T, Wandziuk P, Stobiecki F, Andrzejewski B, Kopcewicz M, Hutten A, Reiss G and Szuszkiewicz W 2004 *J. Magn. Magn. Mater.* **282** 248
[5] Vega A, Rubio A, Balbas L C, Dorantes-Davila J, Bouarab S, Demangeat C, Mokrani A and Dreysse H 1991 *J. Appl. Phys.* **69** 4544
[6] Endo Y, Kitakami O and Shimada Y 1999 *Phys. Rev.* B **59** 4279
[7] de Vries J J, Kohlhepp J, de Broeder F J A, Jungblut R, Coehoorn R, Reinders A and de Jonge W J M 1997 *Phys. Rev. Lett.* **78** 3023
[8] Endo Y, Kitakami O and Shimada Y 1998 *IEEE Trans. Magn.* **34** 906
[9] Fallon J M, Faunce C A and Grundy P J 2000 *J. Phys.: Condens. Matter* **12** 4075

[10] Fallon J M, Faunce C A, Grundy P J and Blythe H J 2000 *J. Appl. Phys.* **87** 6833
[11] Fallon J M, Faunce C A and Grundy P J 2000 *J. Appl. Phys.* **88** 2400
[12] Petford-Long A K, Sterns M B, Lee C H, Nutt S R, Ceglio M and Hawryluk A M 1987 *J. Appl. Phys.* **61** 1422
[13] Ruterana P, Haudy P and Boher P 1990 *J. Appl. Phys.* **68** 1033
[14] Tiwari M K, Sawhney K J S, Gowri Shankar B, Raghuvanshi V K and Nandedkar R V 2004 *Spectrochem. Acta* B **59** 1141
[15] Parratt L G 1954 *Phys. Rev.* **95** 359
[16] Naik S R, Rai S, Lodha G S and Brajpuria R 2006 *J. Appl. Phys.* **100** 013514
[17] de Boer D K G 1991 *Phys. Rev.* B **44** 48
[18] Tiwari M K, Naik S R, Lodha G S and Nandedkar R V 2005 *Anal. Sci.* **21** 757
[19] Cullity B D 1972 *Introduction to Magnetic Materials* (Reading, MA: Addison-Wesley)