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The progression of interface structure through sputtered Co/Cu and Co/Pt multilayer films

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Abstract. The progression of interface structure through some Co/Cu and Co/Pt multilayer systems, grown on silicon and glass substrates respectively, has been studied using grazing incidence x-ray scattering. Simulations of the data shows that the interface characteristics of the Co/Cu system propagate unchanged with increasing numbers of bilayers. The interface roughness, lateral length scale and fractal dimension remain constant across the series of samples. In contrast, a reduction in the interface conformality between top and bottom surfaces with respect to increasing numbers of bilayers is observed in the Co/Pt system; however the roughness correlation between successive layers is largely maintained. Magnetization data for Co/Cu indicate that the Co layer immediately adjacent to the substrate is magnetically 'dead'. This could result from mixing of the cobalt and silicon oxide species at the substrate interface.

1. Introduction

Since the initial discovery of the phenomenon of giant magnetoresistance (GMR) in magnetic multilayers there has been a great deal of interest in determining exactly what factors control the magnitude of the GMR effect. Interface structures within magnetic multilayers are known to have an effect on their physical and magnetic properties. In a previous study [1] we examined the correlation of GMR with the crystallographic texture and interface structure in a similar series of Co/Cu multilayers. The interface roughness was found to be highly correlated in nature and of the same magnitude in all of the samples with a constant lateral correlation length at all of the interfaces in the multilayer. Earlier studies by Egelhoff and Kief [2] have suggested that the oscillatory dependence of the antiferromagnetic coupling between the magnetic layers as a function of the non-magnetic spacer layer thickness can be explained in terms of Fermi surface effects. Our measurements support the proposal [3] that this oscillatory coupling effect in the Co/Cu system should be stronger for ⟨100⟩ and ⟨110⟩ oriented films than those with ⟨111⟩ orientation. The observed reduction in the GMR across the series of samples was attributed to the observed decrease in the volume fraction of ⟨100⟩ oriented grains as the texture was transformed from random polycrystalline to a well oriented ⟨111⟩ polycrystalline texture.

The formation of a columnar growth mode in Co/Pt has been observed directly, by cross sectional transmission electron microscopy (TEM). Zeper *et al* [4] found the grains to grow columnwise, in a close packed arrangement, throughout the entire stack while still retaining the individual layer definition. A fine columnar structure was also observed by Hashimoto *et al* [5]. Other high resolution TEM measurements [6] have indicated the interfaces to be semi-coherent in nature, consistent with a columnar growth mode.

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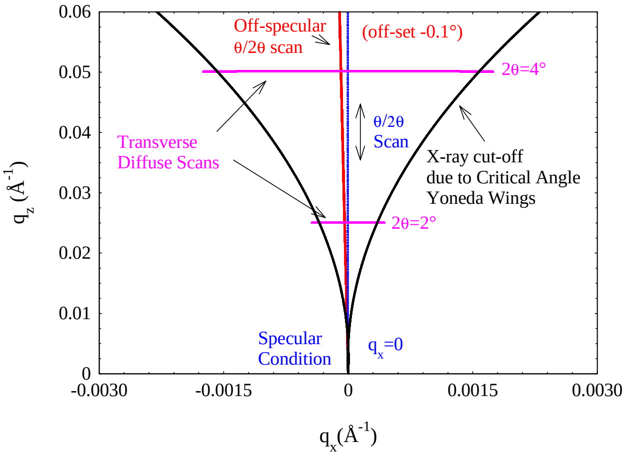

Figure 1. Schematic representation of scan types in reciprocal space showing specular (q_z only, $q_x = 0$), off-specular (sample off-set $-0.1°$), and transverse diffuse (q_x only at fixed q_z).

The role of surface anisotropy is critical in the production of perpendicular magnetization. Perpendicular anisotropy is well known in the Co/Pt system [7] where the interface anisotropy contrives to pull the magnetization out of the plane. The mechanism responsible for producing this anisotropy is believed to be related to the interface structure. The applications of Co/Pt with regard to magneto-optic [8, 9] and thermo-magneto-optic stability have been investigated extensively [10, 11]. Pronounced grain boundaries in columnar structures can act as pinning centres, restricting domain wall motion and enhancing the coercivity [4, 5, 12].

It is therefore important to not only be able to determine accurately the roughness and texture but also to examine how interface structures propagate through the multilayer. Microscopy techniques, by definition, provide only a localized picture of interface morphology and often require additional sample preparation. Grazing incidence x-ray scattering techniques on the other hand are ideally suited to following the propagation of interface structure through a multilayer, as they provide a global method by which to average laterally over micron length scales in the plane of the film. Measurements of the diffuse scatter provides a quantitative measure of the out-of-plane correlations between the many buried interfaces in a multilayer.

In this paper we report studies of two series of sputtered samples of Co/Cu and Co/Pt multilayers. Within each series the samples are nominally identical in structure but with varying bilayer number, N.

2. Experiment

A series of six Co/Cu multilayer films was grown on silicon by magnetron sputtering. All samples were nominally $N\{10$ Å Co/10 Å Cu$\}$ + 10 Å Pt where N, the bilayer number, was varied as $N = 1, 3, 6, 9, 12, 16$. Using the same apparatus a similar series of five Co/Pt films was grown on glass, with a 50 Å Pt buffer layer, again nominally $N\{4$ Å Co/20 Å Pt$\}$ with $N = 1, 5, 10, 15, 22$. Glass substrates were used for the Co/Pt samples to facilitate Kerr microscopy measurements; however, the use of a Pt buffer layer should avoid any anomalous results between the two systems due to the differing substrate materials. The deposition rates at ambient temperature, under an argon partial pressure of 0.4 Pa, were 0.4, 0.5 and 0.9 Å s^{-1} for Co, Cu and Pt respectively.

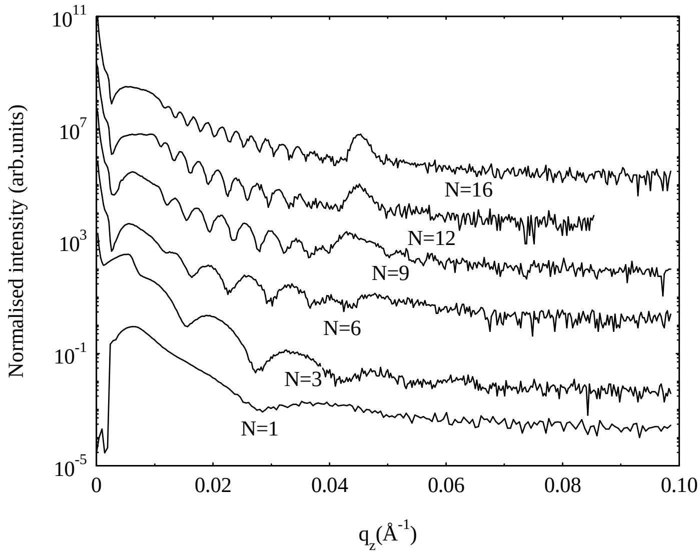

Figure 2. A series of Co/Cu off-specular scans (sample off-set $-0.1°$), for $N = 1$ to $N = 16$, $\lambda = 1.3798$ Å.

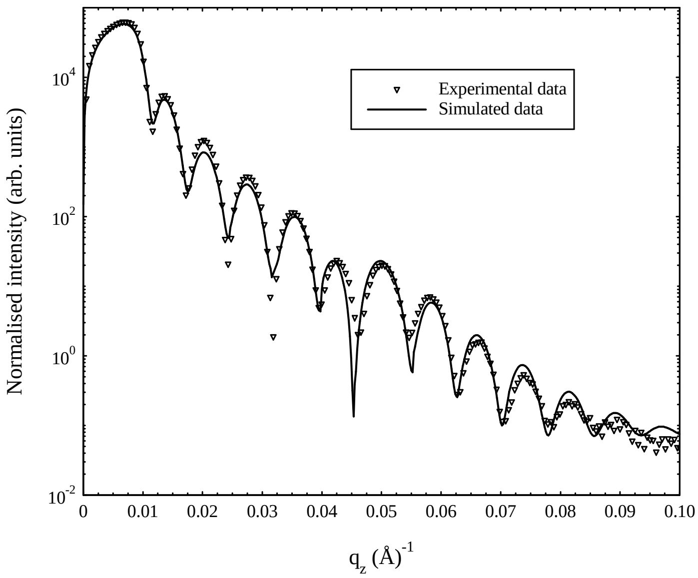

Figure 3. Specular (q_z only) scan showing data (symbols) and simulation (solid line) for Co/Cu $N = 9$. $\lambda = 1.3798$ Å.

Grazing incidence x-ray scattering measurements were undertaken on a Bede GXR1 laboratory reflectometer and on station 2.3 at the Daresbury Synchrotron Radiation Source. A schematic representation of the scan types in reciprocal space is shown in figure 1. Specular scans (q_z scan only), in which the detector is scanned at twice the rate of the sample, provide information on near surface electron density and total interface width. The bilayer and total stack thickness is determined from the Bragg peak and Kiessig fringe period.

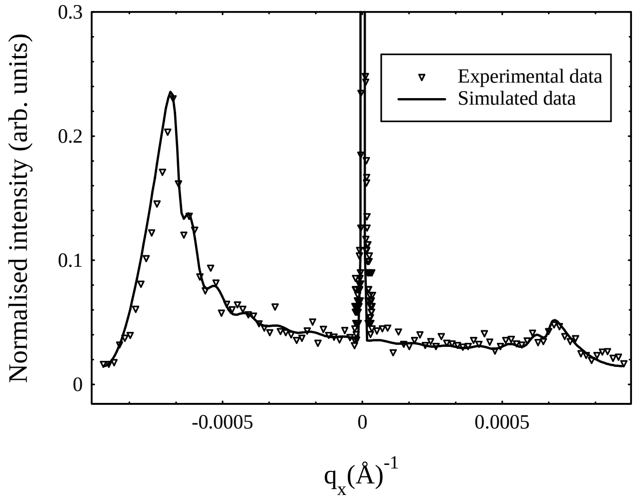

Figure 4. Transverse diffuse (q_x only) scan showing data (symbols) and simulation (solid line) for $N = 9$. $\lambda = 1.3798$ Å.

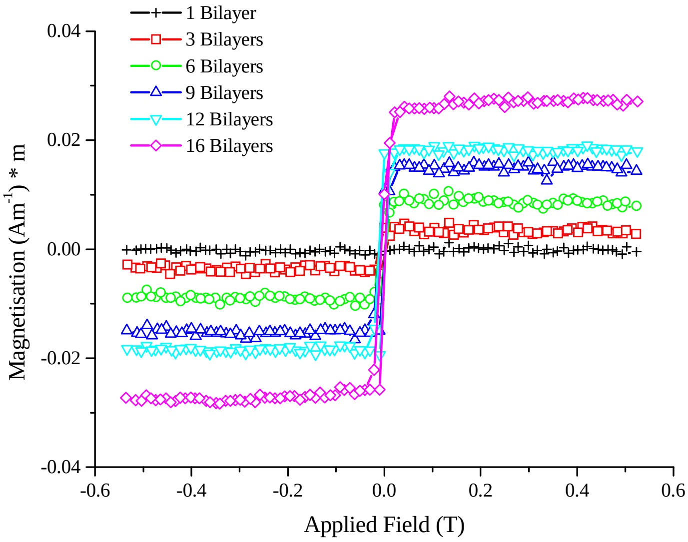

Figure 5. Magnetization loops normalized to sample area.

Transverse diffuse scans (q_x scan only at fixed q_z), in which the detector is fixed and only the sample scanned, allow the effective interface roughness determined from the specular scans to be subdivided into correlated, uncorrelated and compositional grading components [13]. Correlated roughness is defined as that having spatial frequencies that replicate from one layer to another through a stack. The average roughness measured in the specular scan is equal to the sum, in quadrature, of the correlated, uncorrelated and compositional grading components measured in the diffuse scan. The transverse diffuse scan is also sensitive to the in-plane structure of the interfaces. By matching the experimental data to simulations, based on a self-affine fractal model of the interface, we can determine the lateral correlation length and the fractal Hirst parameter [14–16]. Off-specular (longitudinal diffuse) scans have also

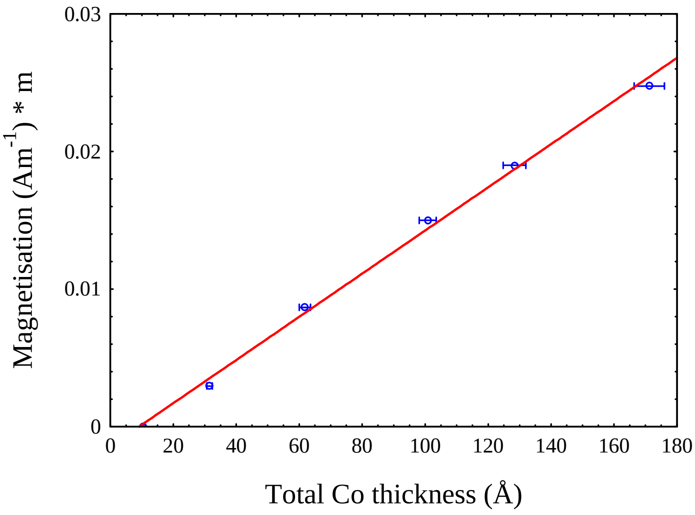

Figure 6. Magnetization, normalized to sample area, as a function of the total Co thickness.

been performed in which the scattering geometry is the same as that of the specular scan but with a small, initial offset in the sample angle. These longitudinal diffuse scans allow us to probe the diffuse scatter close to the specular ridge and when subtracted from the specular data allows the genuinely specular scatter to be obtained.

The x-ray scattering factor of an element scales monotonically with atomic number, Z, which can prove problematic when trying to achieve scattering contrast between close neighbours in the periodic table, such as Co and Cu. The tunability of synchrotron radiation allows the x-ray wavelength to be set to the absorption edge of one of the constituent elements, in this case Cu, thereby greatly increasing the scattering factor difference between the copper and cobalt layers by exploiting the phenomenon of anomalous dispersion [14].

Measurements of the magnetization were made on a vibrating sample magnetometer (VSM) in order to study how the volume magnetization of the samples, once normalized to sample area, changed with increasing bilayer number. By assuming that the magnetic volume of the Co/Cu multilayer scales with the component layer thickness the experimental values can be directly compared against standard bulk values. Calculation of the magnetically active volume of the sample is thus possible from the x-ray data.

3. Results

3.1. Co/Cu multilayers

A series of off-specular scans for the Co/Cu system can be seen in figure 2. These scans, normalized to the beam monitor, illustrate the growth of the multilayer stack as the bilayer number is varied from 1 to 16. The presence of Kiessig fringes in these off-specular scans is strongly indicative of a high proportion of the interface roughness being correlated in nature [17]. This means that the spatial frequency of the roughness is replicated between successive layers from the substrate upwards, indicating very uniform growth as more bilayers are added. The presence of an off-specular Bragg peak confirms correlation within the bilayer repeats and off-specular Kiessig fringes indicate a high degree of conformality between the top capping layer and the substrate. The way in which these periodic features exist in the off-specular scans

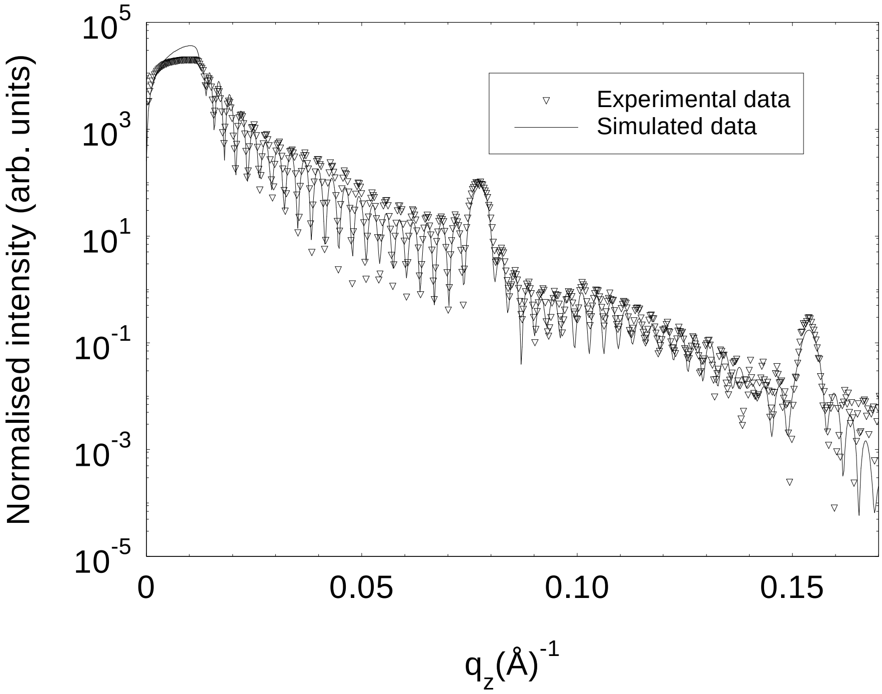

Figure 7. Specular (q_z only) scan showing data (symbols) and simulation (solid line) for Co/Pt $N = 20$. $\lambda = 1.3$ Å.

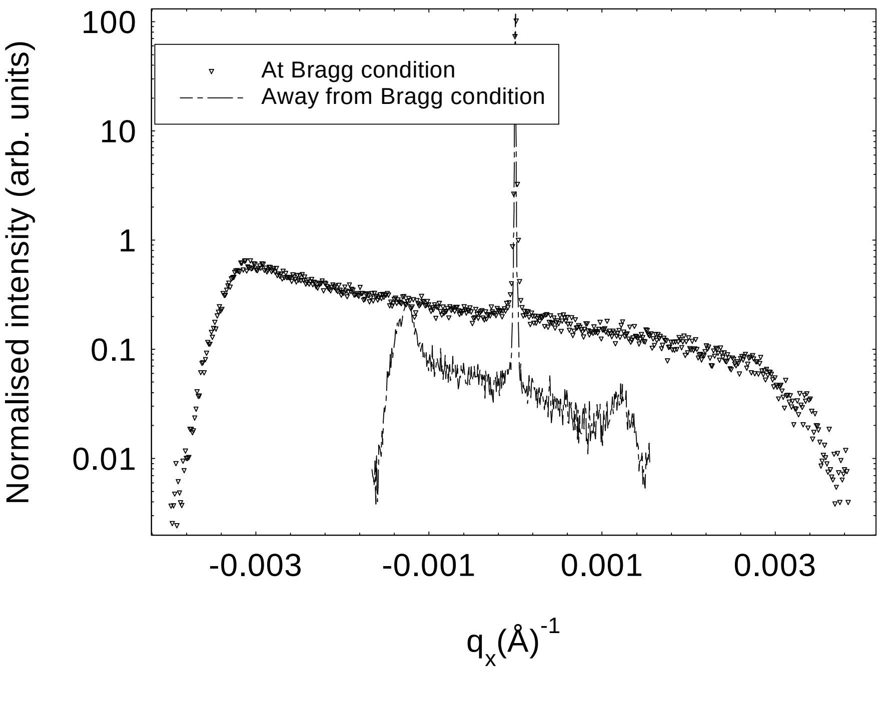

Figure 8. Transverse diffuse scan (q_x only) taken at and away from the Bragg condition, $\lambda = 1.3$ Å.

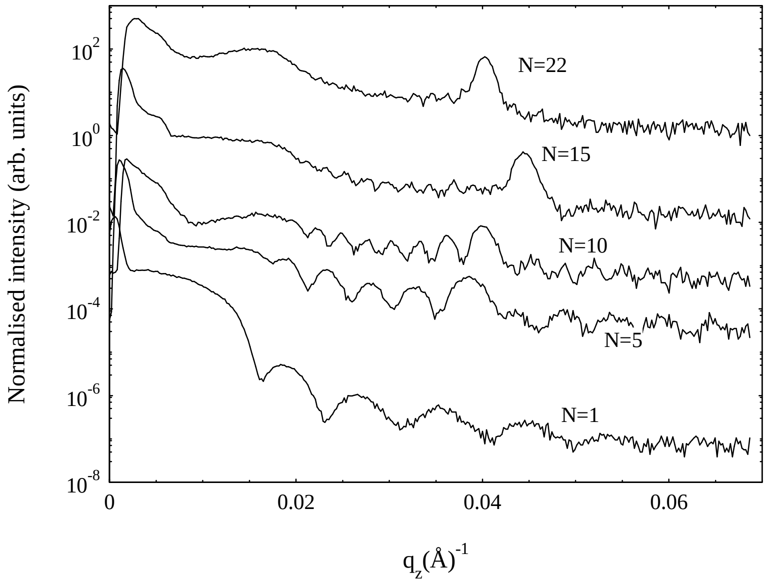

Figure 9. A series of Co/Pt off-specular scans (sample off-set $-0.1°$) for $N = 1$ to $N = 22$, $\lambda = 1.3798$ Å.

Table 1. Multilayer structure parameters, such as interface roughness (σ), lateral roughness correlation length (ξ) and fractal parameter (h) determined from x-ray scattering simulations.

Bilayers (N)	Co thickness (±0.3 Å)	Cu thickness (±0.3 Å)	Pt thickness (±0.3 Å)	$\sigma_{correlated}$ (±0.3 Å)	$\sigma_{uncorrelated}$ (±0.3 Å)	ζ (±5 Å)	h (±0.05)
1	10.4	10.4	4.3	3.3	1.8	135	0.50
3	10.5	10.6	4.2	3.4	1.8	135	0.50
6	10.3	10.5	4.4	3.3	1.8	135	0.50
9	11.3	11.4	4.4	3.3	1.8	135	0.50
12	10.7	10.9	4.1	3.5	1.8	135	0.50
16	10.7	10	4.1	3.5	1.8	135	0.50

for all values of N up to $N = 16$, corresponding to a total stack thickness of 330 Å, suggests that the out-of-plane correlation length scale is appreciably greater than 330 Å.

An example of a specular scan and simulation, in this case for the nine bilayer Co/Cu sample, is shown in figure 3. Figure 4 shows an example of a transverse diffuse scan taken for this nine bilayer system. For each sample transverse scans were taken through a Bragg peak and a Kiessig maximum and minimum, both at and away from the copper absorption edge. The specular and transverse diffuse data were modelled using the Bede REFS code developed by M Wormington. This uses a fractal model within the distorted wave Born approximation [13, 16]. Best fits to the experimental data are included in the figures.

In all cases we were able to obtain good fits between the simulated and experimental data, the results for the Co/Cu system being shown in table 1. As indicated by the off-specular data, the roughness at the interfaces is highly correlated in nature with a correlated to uncorrelated roughness ratio of approximately 2:1 and an average interface width of 3.8 Å. There was found to be no interdiffusion contribution to the average interface width in any of the samples.

Table 2. Multilayer structure parameters for Co/Pt.

Bilayers (N)	Buffer thickness (±0.3 Å)	Co thickness (±0.3 Å)	Pt thickness (±0.3 Å)	$\sigma_{r.m.s}$ (±0.3 Å)
1	47.0	4.5	10.1	3.3
5	50.2	3.2	10.2	3.9
10	49.1	3.4	11.6	3.8
15	43.6	4.0	12.6	4.0
22	53.5	3.8	11.9	3.1

The fractal parameter was found to be 0.5, which although a little smaller than a typical value for a sputtered sample remained consistent across the series. Layer thickness for both the Cu and Co layer remained constant throughout the series of samples apart from slightly higher thickness in the nine bilayer sample.

Magnetization data normalized to sample area are shown in figure 5. The one bilayer system trace is flat due to it being below the sensitivity threshold of the VSM. The magnetization, normalized to sample area, versus the total Co layer thickness, determined from the x-ray measurements, is shown in figure 6. This shows a linear increase in magnetization with Co thickness corresponding to a constant increase in the magnetic bulk of the multilayer as further bilayers are added (the x-ray measurements show the individual Co layers to be thicker in the nine bilayer sample which is matched by a proportional increase in the magnetization of that sample). The zero magnetization intercept for a Co thickness of 10 ± 0.5 Å suggests that the Co layers immediately adjacent to the substrate are magnetically ‘dead’. We tentatively attribute this to a mixing of the cobalt, silicon and oxygen species at the interface.

3.2. Co/Pt multilayers

An example of a specular scan, with simulation, for Co/Pt grown on silicon is shown in figure 7 with a transverse diffuse scan taken at and away from the Bragg condition shown in figure 8. The way in which the diffuse scatter is concentrated around the Bragg condition is a consequence of the roughness in these samples being highly correlated in nature. The corresponding structure parameters are summarized in table 2. In contrast to the Co/Cu system, the Co/Pt layers show a very different propagation of interface structure through the multilayer stack. The series of off-specular scans shown in figure 9 displays notable differences from the scans taken for the Co/Cu multilayers. As the bilayer number is increased, the off-specular Bragg peak remains, indicating that out-of-plane correlation is retained within the bilayers. However, the off-specular Kiessig fringes disappear as the stack increases, the out-of-plane correlation between the interfaces at the substrate and cap being lost. This conformality between layers within the bilayer but not between successive bilayers can be explained by the presence of the columnar type growth mode in the Co/Pt system [9]. In such a structure the lateral correlation length on the surface increases with increasing N. Over the thickness of a bilayer this effect is small and the majority of the conformality remains. However, over the entire stack, the loss of conformal growth is appreciable leading to the disappearance of the off-specular Kiessig fringes which are determined by the overall degree of conformality between the top and bottom interfaces. The off-specular fringes are lost between 15 and 22 bilayers indicating that within the stack the out-of-plane roughness correlation length is of the order of 350 to 400 Å and the surface is sensitive to features at a distance of no greater than 400 Å. The off-specular Bragg peak for the $N = 15$ sample is off-set due to the bilayer being slightly thinner than nominal

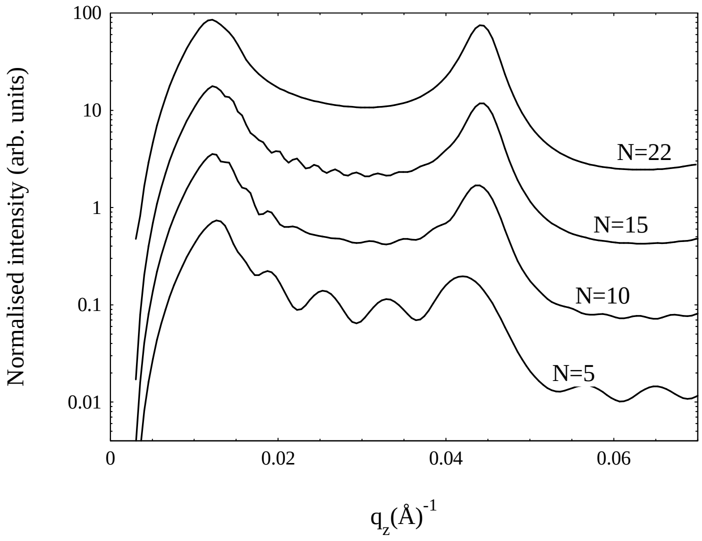

Figure 10. A series of off-specular (sample off-set -0.1°) simulations for Co/Pt in which the vertical roughness correlation length is fixed at 350 Å, $\lambda = 1.3926$ Å.

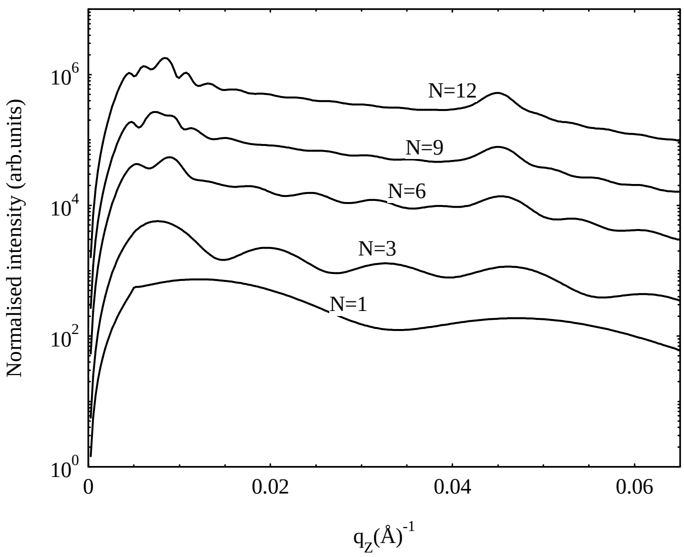

Figure 11. A series of off-specular simulations (sample off-set -0.1°) for Co/Cu in which the vertical roughness correlation length is fixed at 150 Å in order to simulate columnar growth, $\lambda = 1.3798$ Å.

thickness. A series of off-specular simulations, with a fixed out-of-plane roughness correlation length of 350 Å and nominal growth thickness, is shown in figure 10. The loss of off-specular Kiessig fringes for a stack thickness between $N = 15$ and $N = 22$, observed in the actual data, is also observed in these simulations.

4. Discussion

A series of off-specular simulations for Co/Cu was performed in which the out-of-plane roughness correlation length was fixed at 150 Å. In all simulations the layer thickness and interface roughness were kept constant. Figure 11 shows a series of these off-specular simulations for Co/Cu. They show the same disappearance of Kiessig fringes as observed in Co/Pt once the stack thickness exceeds the vertical correlation length. This is in marked contrast to the actual experimental data for Co/Cu in figure 2, giving further support for no columnar type growth mode occuring in Co/Cu. Defocused TEM measurements, performed for Co/Cu, in which the contrast between layers is enhanced to reveal the interface structure, also indicate no columnar growth mode in Co/Cu [1].

The occurrence of columnar growth in Co/Pt but not Co/Cu is most likely due to the lower surface temperature at deposition for the Co/Pt system. Although the sputtering yield for Pt is lower than that for Co at similar energies the deposition rate for Pt is almost twice that of Co. This greater sticking coefficient for Pt indicates a lower surface mobility, and hence a lower surface temperature during Pt deposition, which can favour the formation of a columnar system.

5. Conclusion

The progression of structure through multilayer stacks of Co/Cu and Co/Pt has been examined and significant differences between the two systems have been observed. The growth of Co/Cu multilayers can be seen to be highly uniform in nature as the bilayer number is increased with interface roughness and lateral roughness correlation length remaining constant. In agreement with previous studies the Co/Cu system was found to consist of interfaces with a high degree of correlated roughness which propagate with high conformality through the multilayer stack. In contrast, the Co/Pt system displays conformal growth only over an out-of-plane length scale of the order of 400 Å, approximately 15 bilayers. Beyond this thickness, all correlations between the top and bottom surface are lost consistent with a columnar structure resulting from the lower surface mobility of Pt during deposition. Grazing incidence x-ray scattering is shown to be an effective method by which to follow the progression of interface structure in a multilayer film without the need for specialized sample preparation.

Acknowledgment

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