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Determination of the Copper Layer Thickness in Spin Valves by Grazing Incidence X-ray Fluorescence

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Abstract- We show that at the standard laboratory wavelength of CuKα the scattering factors of Cu and $Ni_{0.8}Fe_{0.2}$ are identical, thereby making it impossible to distinguish the boundary of the Cu spacer layer in a Cu/permalloy spin valve structure from grazing incidence x-ray reflectivity curves. Use of grazing incidence fluorescence, in conjunction with x-ray reflectivity provides sufficient information to control the Cu layer thickness. We demonstrate the technique on two spin valves with Cu spacer layers differing in thickness by a factor of 2.5.

Index Terms - x-ray grazing incidence fluorescence, spin valves

1. INTRODUCTION

Grazing incidence x-ray reflectivity (GIXR) is now well established as a technique for the measurement of the thickness of thin film structures with single atomic layer precision [1,2]. Using specular and diffuse scattering, we have measured the layer thickness, interface roughness, lateral correlation length, and fractal parameter in multilayers of 3d transition metal systems such as Cu/Co and Fe/Cr, and successfully related these to the magnetotransport properties[3,4].

Unfortunately, for the important class of spin valve systems such as Si/Ta/NiFe/Cu/NiFe/FeMn/Ta where there are only a few individual interfaces, there are serious difficulties. The scattering factor for x-rays varies monotonically across the Periodic Table, and the contrast between 3d transition metals is poor. For laboratory measurements with CuKα radiation, the reflectivity curve is determined by the entire stack thickness, the total thickness of the transition metal layers, together with the cap and buffer roughness. Contrary to the claims of Huang *et al* [5] it is not possible to distinguish between the permalloy and copper layers at the CuKα wavelength. Thus it is not possible to measure the Cu spacer layer thickness, a key parameter in quality control for magnetoresistive head manufacture.

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Fig 1 shows reflectivity curves simulated using the Bede Scientific REFS program [6] for a simplified spin valve structure. At 1.5405Å (CuKα_1 wavelength), the dispersion corrections are such that the scattering factor for Cu and permalloy are identical. There is no change in the reflectivity curve when the permalloy/Cu/permalloy sandwich is replaced by a single layer of Cu of the same total thickness. Only very close to the Cu absorption edge (upper curve of Fig 1) can these two cases be distinguished, and then only with difficulty. Although changes to the total thickness of the permalloy and copper layers can be distinguished readily, as illustrated experimentally in Fig 2, it is effectively impossible to determine the relative thickness of the NiFe and Cu layers from the reflectivity curve.

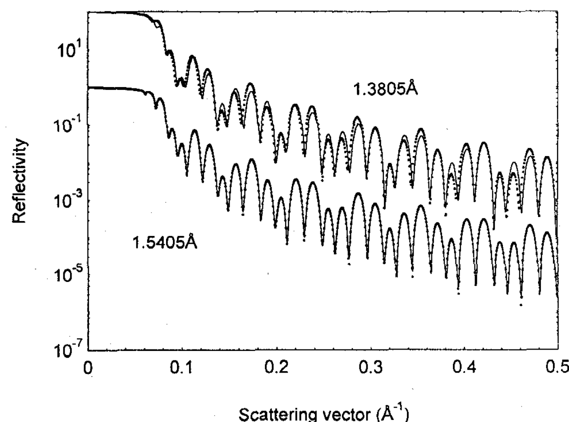

Fig 1: Simulated reflectivity profiles from a Si/Ta(100Å)/$Ni_{0.8}Fe_{0.2}$(60Å)/Cu(50Å)/$Ni_{0.8}Fe_{0.2}$(60Å)/Ta(100Å) system. Upper curves at 1.3805Å (displaced by x100 for clarity), lower curves at 1.5405Å. Points correspond to replacement of all NiFe with Cu in the simulation.

However, a combination of grazing incidence x-ray scattering and fluorescence does, in principle, enable us to determine individual layer thickness and in particular the Cu spacer layer thickness.

2. EXPERIMENTAL TECHNIQUE

While total external reflection is now extensively used for chemical analysis in the very near surface region, there have been relatively few reports of the application of the depth

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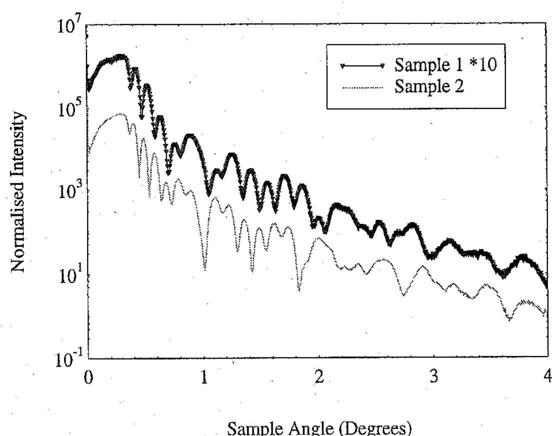

Fig 2: Experimental reflectivity profiles for a Si/Ta/NiFe/Cu/NiFe/FeMn/Ta structure. Wavelength 1.38Å The lower curve corresponds to a Cu layer three times the thickness of that in the upper curve. (All other layer thicknesses are constant.)

sensitivity inherent in measurements made as a function of incidence angle [7]. In the present experiments, the fluorescence yield was measured simultaneously with the x-ray scattering as the incident beam angle was tuned through the critical angle for total external reflection. Measurements were made at station 2.3 of the Daresbury Laboratory Synchrotron Radiation Source, where the beam is monochromated by a water cooled (111) channel-cut silicon crystal. The specular reflectivity was measured using a slit-collimated scintillation detector. A Si(Li) energy dispersive solid state detector was situated 20 cm above the sample, and the fluorescent spectrum was recorded at each point in the reflectivity curve. All the GIXR measurements were taken at the Cu absorption edge (λ=1.38Å); the excitation wavelength for the fluorescence was 1.3Å, at the peak flux of the station. The samples were grown in a fully automated sputtering system with six DC magnetron sources fitted with a liquid nitrogen cold trap giving a base pressure of ~$2*10^{-8}$ Torr. The Ar pressure was 5.0 mTorr. All the samples were grown in the same vacuum cycle, and the deposition rates for the various materials were all of the order of 4Å/s. The structure of the samples was nominally: Si/Ta(120Å)/$Ni_{0.8}Fe_{0.2}$(40Å)/Cu(x)/$Ni_{0.8}Fe_{0.2}$(30Å)/$Fe_{0.5}Mn_{0.5}$ (40Å)/Ta(20Å). Here we report results for samples 1 and 2, nominally x = 20Å and 60Å respectively.

Normalised fluorescence spectra were fitted by minimising the residuals and taking all peaks to be Gaussian in shape. The centre of the Gaussians were fixed prior to the fit using the values for elemental emission lines taken from the International Tables for X-ray Crystallography Vol. III and an initial ^{55}Fe calibration. As the $K\alpha_1$ and $K\alpha_2$ lines were unresolved, they were weighted in the ratio 2:1 in the fits and the full width at half height maximum was kept at a fixed value for all peaks. The integrated intensity under specific peaks from individual layers was plotted as a function of the incident x-ray beam angle and compared with simulations from the Bede Scientific GIXS software, which was also used for simulation of the specular and diffuse scatter.

3. EXPERIMENTAL RESULTS

Prior to structural analysis, the magneto-transport properties of the samples were also investigated. The room temperature giant magnetoresistance of sample 1 was 3.7%, whilst for the sample with the thicker Cu layer, this fell to 2.2%. The exchange fields were high; 460 Oe for sample 1 and 450 Oe for sample 2. The reflectivity (Fig 2) shows clear differences between the samples which can be attributed to a change in the total stack thickness. A structural model in the form of discrete layers could not be found, although some structural information could be obtained by fitting to a simple tri-layer structure (with the 3d elemental layers being considered as a single layer). There were additional beats which could not be explained in terms of oxide formation. This suggests that the Ta buffer and capping layers may have formed ternary alloys.

As seen in Fig 3, with a high resolution energy dispersive solid state detector we can easily separate out the x-ray fluorescence from elements adjacent in the Periodic Table.

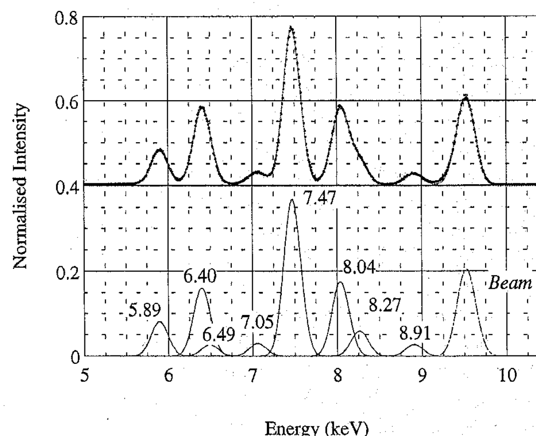

Fig 3: The deconvolution of the fluorescence spectra from sample 1 with the incidence beam angle 0.38° and wavelength λ=1.3Å

Although the reflectivity curves cannot be fitted exactly, simulation of the integrated fluorescence intensity under specific elemental lines has been conducted using the best fit of the total 3d transition metal thickness from the reflectivity (Fig 4). These results show that the fluorescence yield rises to a maximum at the critical angle and then falls rapidly to about 50% of the maximum value, subsequently exhibiting oscillations characteristic of the layer structure. This is in marked contrast to the behaviour for a thick crystal, where the fluorescence yield rises to a maximum and thereafter only falls slowly as absorption losses rise due to the increased depth penetration into the sample. From a measure of the fluorescence yield, together with a knowledge of the layer thickness deduced from the reflectivity it is possible to produce a compositional model of the 3d elemental layer.

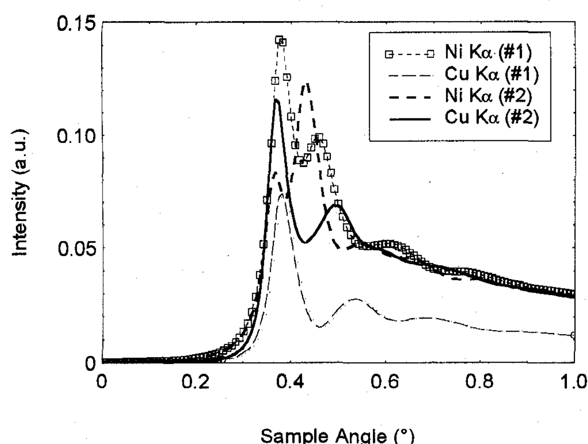

Fig 4: Simulation of the integrated intensity under the Cu and Ni Kα lines with an incident wavelength of 1.3Å for a spin valve structure using the best fit for the 3d elemental layer and the compositions shown in table 1. The lines correspond to a sample with a copper layer 2.5 times that of the points.

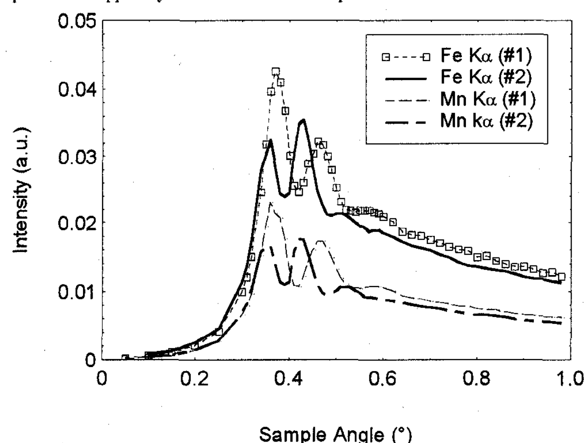

Fig 5: The integrated intensity under the Fe and Mn Kα lines for sample 1 (points) and sample 2 (lines)

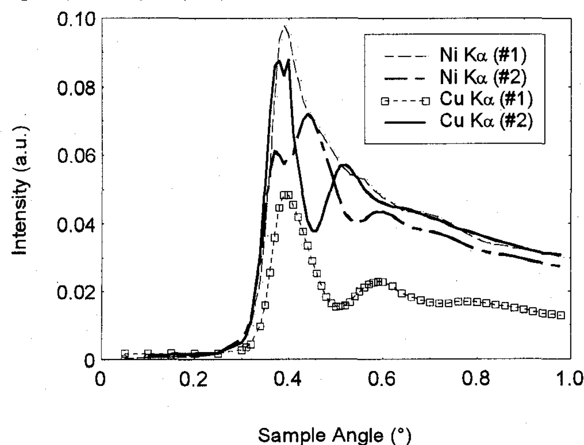

Fig 6: The integrated intensity under the Ni and Cu Kα lines for sample 1 (points) and sample 2 (lines)

It is clear from fig. 5 that the period increases for sample 2; this is due to the different film thicknesses. Sample 2 has 10% less Mn and Ni than sample 1, suggesting that the NiFe and FeMn layers are slightly thinner in the former sample. Far greater differences are observed in Fig. 6. There is again 10% less Ni in sample 2, confirming the trend observed from the Mn and Fe peaks, but there is approximately 2.5 times more Cu in sample 2 compared with sample 1.

The table below shows the percentage composition of the 3d elemental layer calculated from the fluorescence yields at an incidence angle of 0.98°:

	Sample 1	Sample 2
$Fe_{0.5}Mn_{0.5}$	19.4 ± 1%	14.2 ± 1%
$Ni_{0.8}Fe_{0.2}$	60.3 ± 1%	45.4 ± 1%
Cu	20.3 ± 1%	40.4 ± 1%

Table 1: Compositions of the 3d layer

The best fit of the reflectivity from sample 1, estimates the thickness of the total 3d elemental layers to be 130 ± 10Å; this implies that the NiFe layer is 10% thicker than the nominal structure in sample 1. It also shows that the FeMn layer is much thinner than expected for both samples (25Å for sample 1 and 22.5Å for sample 2). From the magneto-transport measurements, a much thicker FeMn layer was expected to account for the high biasing fields, but this does not seem to be the case here.

4. CONCLUSIONS

We have shown that the combined technique of GIXR and GIXF is a powerful tool in understanding the structure of spin valve systems. For industrial quality control purposes, the relative ratios of the fluorescence peaks may suffice for the measure of the Cu layer thickness. Except for longer counting times and the need to place the detector close to the sample, there is no difficulty in making such measurements with a laboratory source.

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