Vol. 91 (1997) *ACTA PHYSICA POLONICA A* No. 5

Proceedings of the 3rd ISSSRNS Jaszowiec '96

GRAZING INCIDENCE X-RAY REFLECTIVITY STUDY OF MBE-GROWN Co/Cu MULTILAYERS

J.B. PELKA, L.T. BACZEWSKI, A. WAWRO AND J. DOMAGAŁA

Institute of Physics, Polish Academy of Sciences
Al. Lotników 32/46, 02-668 Warsaw, Poland

In this work we report preliminary grazing-incidence X-ray reflectometry studies of multilayer structures composed of 3*d* metals Co and Cu deposited in the ultra-high vacuum molecular beam epitaxy system. The multilayers of different modulation period were deposited on glass substrate directly, or on 3*d*-metallic buffers of various thicknesses. The experimental specular reflectivity spectra were analyzed by a comparison with a theoretical model calculated from a recursive algorithm based on the Fresnel formula [1, 2]. It enabled us to estimate the structural parameters concerning layer thickness and roughness. The results obtained are correlated with magnetization measurements of the layered structures, as a function of modulation period, buffer type and thickness. A special attention to influence of interfacial roughness on magnetization results is paid.

PACS numbers: 75.30.–m, 75.30.Gw, 75.70.–i

1. Introduction

The Co/Cu multilayer structures grown by molecular beam epitaxy (MBE) belong to a class of layered materials with a giant magnetoresisitivity effect (GMR). It enables an application of the multilayers as magnetic spin valves or recording heads of high sensitivity and resolution [3]. The giant magnetoresistivity effect is widely considered as due to a difference in electron scattering cross sections between the electrons with spin up and spin down in magnetic layers. However, it has not yet been clear, whether this scattering takes place in the layer itself or at phase boundaries [4]. The conditions of the Co/Cu multilayer deposition and resulting the final structure of samples strongly influence magnetic properties of the layers. Among others, the most important are thickness and interfacial roughness of the Co/Cu layers, as well as analogous parameters describing the substrate and buffer layer which has the principal influence on the structural and magnetic properties of the Co/Cu structure. The samples studied in this work, due to a deposition on amorphous glass substrate are expected to be polycrystalline. Such films have obviously more structural defects than epitaxial samples but still GMR is clearly present, what gives arguments for some theoretical models of the effect [5, 6].

Obtaining precise information about the sample structure is therefore of primary importance to understand the correlations between the structural parameters and magnetic properties.

The grazing-incidence X-ray reflectometry (GIXR), among other methods, seems to be especially suitable to provide the necessary information. It has widely been used to study multilayer structures, produced with various techniques mainly as optical elements for X-ray optics [7]. In the last years, due to a substantial effort in construction of X-ray devices it is getting to become a standard tool in laboratories dealing with thin film structures. The X-ray reflectivity at grazing incidence for a multilayer system was first described by Parratt [1]. He formulated a recursive algorithm based on the Fresnel equation and showed that it is possible to calculate also the reflectivity from a layer of arbitrary electron density profile by "slicing" it onto a set of sublayers with fixed electron densities. Any differences in specular reflectivity are caused by changes in electron density of the material independently of its crystalline structure. Therefore it can serve as an excellent, non-destructive tool to study in-depth electron density profiles in crystalline or amorphous layers with a thickness in the range of 1 nm – few hundreds nm. In addition, it can provide information about surface and interfacial roughness [8]. The application of the technique to the study of the multilayer system is limited only by the sample size and the flatness of its surface.

2. Experimental

The Co/Cu multilayers were deposited in MBE EVA 32 Riber system on the substrates formed from float glass. During the whole process the substrate temperature was kept at 40°C, and the vacuum was better than 5×10^{-9} Pa. The glass substrates were cleaned by a chemical method in a standard way. In a loading chamber they were annealed in 200°C for 60 min. The evaporation of Co and Cu was from electron guns. The deposition rates of the evaporated layers were controlled by Leybold Heraeus electron emission spectrometer Sentinel III. The evaporation rates were kept at 0.03–0.05 nm/s. Few series of samples were produced of configuration defined as: glass/buffer/(CoXCuY)n, where $10 < n < 50$ — a number of Co/Cu bilayers, $5 < X, Y < 100$ — thicknesses in Å of Co and Cu individual layers. Cobalt or copper buffer layers of different thicknesses between 50 and 300 Å were used. In-situ characterization was performed by RHEED and Auger spectroscopy-textured, a polycrystalline structure was found with no sign of oxygen contamination.

A VSM magnetometer was used for magnetization measurements. The magneto-resistance measurements were carried out in the temperature range of 4.2–300 K. The magnetic field of 3.6 kOe was applied in-plane of the sample in two directions: parallel and perpendicularly with respect to the current flow. The resistance of the samples was measured with the standard four-probe method.

The reflectivity measurements were recorded with the conventional Cu K_α radiation source by a high performance MRD Philips spectrometer equipped with a Si (440) Bartels monochromator and a Ge (1, −1) analyzer. An additional slit of 50 μm wide was applied between the sample and monochromator to limit the beam dimension, and the spot size at low angles of incidence. Most of the spectra were measured in the angular range of 0–2.5°. The overall horizontal resolution of the spectrometer setting was about 12 arc seconds (0.0033°). The experimental reflectivity of all samples were compared with a theoretical model calculated with the above-mentioned algorithm [1, 2].

3. Results and discussion

An example of the typical experimental reflectivity for a sample with nominal composition glass/Cu100/(Co26/Cu22)10 is shown in Fig. 1 (full circles) together with the theoretical fit (solid line). The intensity scale is logarithmic. The excellent agreement between the experimental and theoretical curves in this case was, however, exceptional. The parameters found from the fitting for this sample showed that the thickness of Co layers is 6% lower and of Cu layers 2% higher than assumed; also the buffer thickness was found to be lower about 5%. On the top of the sample an additional CuO_x layer about 3 nm thick was added to the model to fit the experimental data. This illustrates sensitivity of the GIXR measurements on even small changes of various parameters determining the multilayer structure. It should be stressed, however, that there is a standard problem of multiparameter curve fitting in the case of multilayer structures. Each layer is described by three parameters: the thickness, layer composition (we assume here the layer to be monoatomic and homogeneous), and roughness of its top interface. For the reflectivity shown in Fig. 1; it gives 66 parameters (with an additional oxide layer). Without reducing the number of free parameters the fitting problem does not lead to a unique solution. In the case of the multilayers regarded here, the thickness of each individual Co (or Cu) layer was found to be the same (we assumed that also for a composition), what significantly reduced the number of free parameters in the model. The overall thickness of the multilayer structure, as well as the thickness of component layers were always verified by the comparison with the model; the other parameters were fitted less accurately. To determine the thickness it is usually enough to fit exactly to the positions of maxima and minima; for other parameters, like roughness we have to fit precisely the intensities as well.

This procedure also permitted to do determination of surface and interfacial roughness, which was as low as 0.6–0.7 nm in our best samples. In some cases, however, the roughness reached values of the order of layer thickness. It can suggest that the roughness in the neighboring interfaces is laterally correlated, otherwise it can lead to strong bridging across neighboring layers, what should be reflected in the magnetic measurements. Surface roughness of copper layer deposited at room temperature was lower than that deposited at 200°C. The measurements revealed that the surface roughness of Cu is considerably higher than that of Co layer, therefore cobalt seems to be more suitable as a buffer layer. Buffer roughness deteriorated the quality of modulated structure and it was reflected by low GMR values. However it seems that the GMR effect is more sensitive on the interface quality than antiferromagnetic coupling (AF). Detailed results of GMR will be published elsewhere.

In Fig. 2 typical examples of hysteresis loops for two (Co26/Cu22)10 multilayers deposited on Cu buffers of 200 Å (solid line) and 100 Å (dashed line) are shown; the buffer roughness was found to be of a similar value in both cases. The

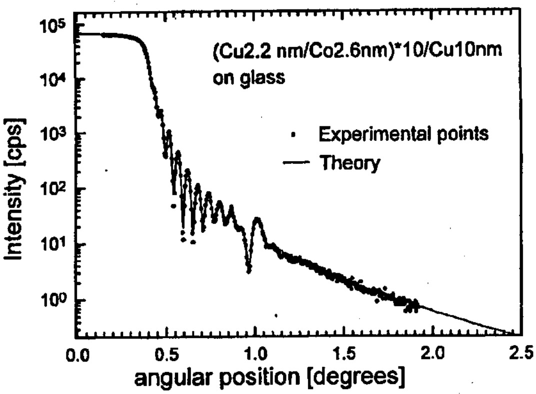

Fig. 1. GIXR reflectivity for multilayer composed of 10 Co 2.6 nm/Cu 2.2 nm bilayers deposited on 10 nm of Cu buffer/glass substrate. Points — experimental data; solid line — theory.

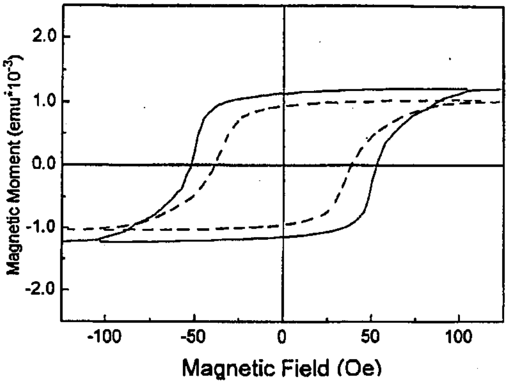

Fig. 2. Hysteresis loops for Cu200/(Co26/Cu22)10 (solid line) and Cu100/(Co26/Cu22)10 (dashed line) multilayers. Measurements performed at room temperature with the applied field in the film plane.

sample with 100 Å (Cu buffer layer has smaller remanence magnetization and coercivity than the one with 200 Å Cu buffer. A similar situation was observed in samples with Co buffer. Different thicknesses of Co buffer layers were tried (50, 100 and 200 Å) for Co25/Cu18 multilayer. Co buffer thickness had clear influence on a shape of hysteresis loop, changing coercivity, remanence and coupling. The sample with 200 Å Co buffer showed almost a rectangular hysteresis loop while for the samples with 50 Å and 100 Å Co buffer AF coupling features were present. However, the sample with 50 Å Co buffer layer had smaller remanence magnetization and coercivity than for the one with 100 Å Co buffer. Also magnetoresistance measurements have given the highest value for the sample Co25/Cu18 with 100 Å Co buffer layer (5.2% at room temperature). For the same multilayer on 50 and

200 Å Co buffer layer we obtained the values of 3.6% and 2.1%, respectively, for magnetoresistive effect. It seems that a too thin buffer layer did not smooth enough the substrate roughness and a too thick buffer introduced an important shunting effect to the measured value.

4. Conclusions

The structural and magnetic correlations were studied in polycrystalline Co/Cu multilayers deposited by MBE technique on glass substrates. It was found that the magnitude of the GMR effect is closely related to the roughness of the interfaces; it decreases with an increase in roughness. An influence of different buffer layers with different thicknesses was investigated. The best results were obtained for multilayers grown on 100 Å cobalt buffer layer for which the highest value of GMR was measured. The same sample exhibited the lowest interface roughness. The correlation of the interfacial roughness with magnetic characterization of the Co/Cu multilayers can be connected with the bridging effect across the neighboring layers; however, from our present study we are not able to conclude what a particular model of interfacial roughness correlations is realized in the studied samples. To solve this question, a further study, using, e.g., diffuse scattering and grazing incidence X-ray diffraction (GIXD) methods, is necessary.

Acknowledgments

This work was supported in part by the Committee for Scientific Research grant No. 2P03B 021 09 and No. 8T11B 053 09. The Phillips MRD diffractometer used in this work was founded by the Foundation for Polish Science (within the program SEZAM 94).

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