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Electrodeposition and characterization of Cu/Co multilayers: Effect of individual Co and Cu layers on GMR magnitude and behavior

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Abstract

The present work discusses the successful electrodeposition of Cu/Co multilayers, exhibiting appreciable GMR of 12–14% at room temperature. The effect of individual Cu and Co layers on the magnitude and behavior of GMR has been studied. By varying the thickness of individual layers the field at which saturation in GMR is observed can be controlled. It was observed that for lower thicknesses of Co layer, the saturation fields are reduced below 1 kOe. The Cu layer thickness seems to control the nature of magnetic coupling and the saturation field, with the two showing a correlation.

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Keywords: Magnetic multilayer; Magnetoresistance; Electrodeposition

1. Introduction

Electrodeposition in modern day technology has emerged as a novel economically viable technique with large-scale production capabilities [1]. The technique has generated immense interest worldwide because of its simplicity, low cost, easy deposition irrespective of surface size and area, the deposition being possible even in nanopores [2]. The observation of appreciable GMR in electrodeposited Co/Cu and CoNi/Cu multilayers [3], comparable to that grown by vacuum techniques, further raised the interest in this technique. Co/Cu multilayers exhibiting appreciable GMR have been studied widely and there are some reports on the dependence of GMR on factors like number of bilayers [4], thickness of individual Cu-layers [5,6] and Co-layers [7] and interfacial roughness [8]. However, low sensitivity and high saturation fields have been restricting the application potential of these multilayers. After obtaining high GMR values of 12.6% in Co/Cu multilayer stacks [9,10], in the present paper we report about our efforts of lowering GMR saturation field via control of the thickness of individual Co and Cu layers, and understanding the role of layer thickness on the behavior of GMR.

The multilayers were deposited in a potentiostatic pulse mode, using a computer-interfaced potentiostat (PAR, model 263A) as a potential source. Single sulfate bath comprising of $CoSO_4 \cdot 7H_2O$, $CuSO_4 \cdot 5H_2O$, $Na_3C_6H_5O_7$ and NaCl in de-ionized water was used for the electrodeposition of multilayers. All the depositions were made in a three-electrode cell containing Pt as a counter electrode, saturated calomel electrode (SCE) as reference and ITO coated glass as a working electrode. Cyclic voltammetry (CV) experiments were carried out to optimize the deposition potentials for individual Cu and Co layers. The thicknesses of individual layers, t_{Cu} and t_{Co}, were estimated from in situ *I–t* curves using Faraday's laws. The details have been presented elsewhere [9]. The resistance of the deposited films was measured at room temperature in current in plane/field in plane (CIP/FIP) configuration by making four linear contacts, current being perpendicular to the applied magnetic field. The externally applied magnetic field was varied upto 8 kOe. The GMR was calculated using the usual definition, $\mathrm{GMR\%} = (R_H - R_0)/R_{sat} \times 100$, where R_H is the resistance in presence of applied field, R_0 is

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the resistance in absence of magnetic field and R_{sat} is the resistance at higher field where it almost saturates. The magnetization studies were carried out using vibrating sample magnetometer (VSM), at room temperature by varying the external field up to 8 kOe.

2. Results and discussion

2.1. Effect of Co layer thickness

Figs. 1(a) and (b) show the variation of giant magnetoresistance with applied magnetic field for two different samples having Co layer thickness $t_{Co} = 10$ and 22 nm, respectively. Both the samples had $t_{Cu} = 6$ nm and n (number of bilayers) = 50. The GMR curves for both the samples have been obtained upto 8 kOe, but shown up to 4 kOe for the sake of clarity. Appreciable GMR values have been obtained in both cases. For sample having $t_{Co} = 10$ nm, Fig. 1(a), the GMR saturates to a value of 7.5% at a relatively low magnetic field value of < 1 kOe. For $t_{Co} = 22$ nm sample shown in Fig. 1(b), the GMR obtained was 12.6% which does not show saturation up to a magnetic field of 8 kOe. The measurements were done for both longitudinal ($B \perp I$) transverse ($B \| I$) magnetoresistance. For both, the magnitudes were almost equal and negative, which clearly indicated the presence of prominent GMR component without any AMR component. The attainment of appreciable GMR indicates the presence of appropriate antiferromagnetic alignment between the Co layers.

Figs. 2(a) and (b) show the variation in GMR value with increase in Co layer thickness. The thickness of Co layer

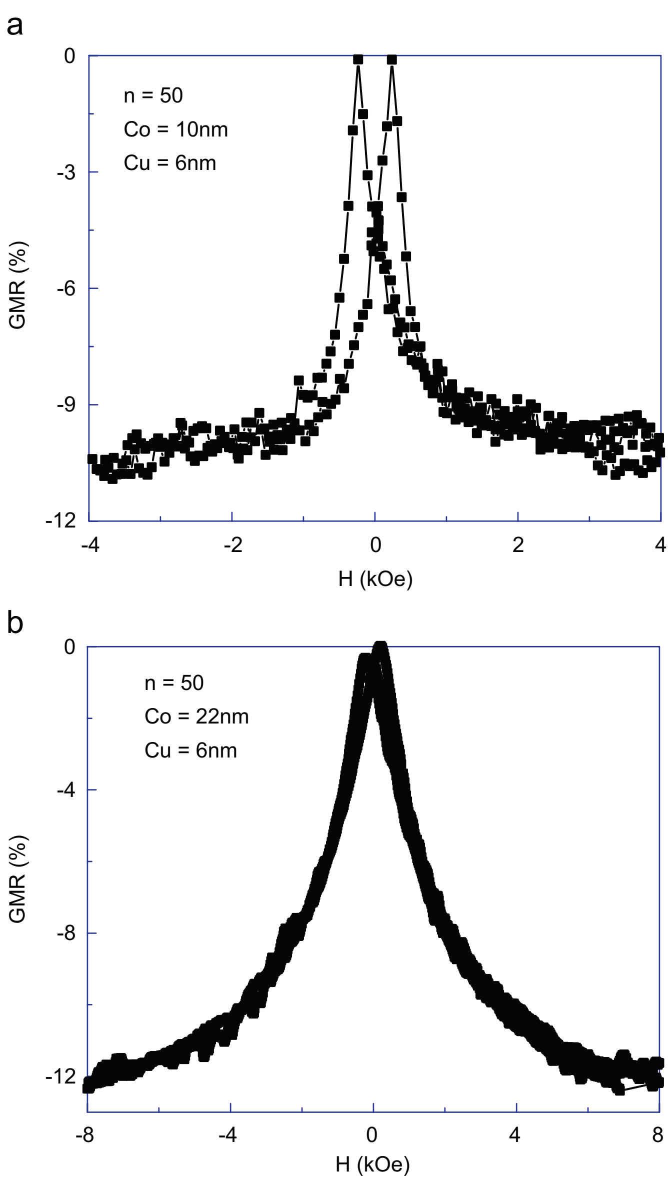

Fig. 1. Variation in magnitude of GMR with applied magnetic field for a 50 bilayer sample on ITO (a) $t_{Co} = 10$ nm and (b) $t_{Co} = 22$ nm. Both the samples have $t_{Cu} = 6$ nm and number of bilayers, $n = 50$.

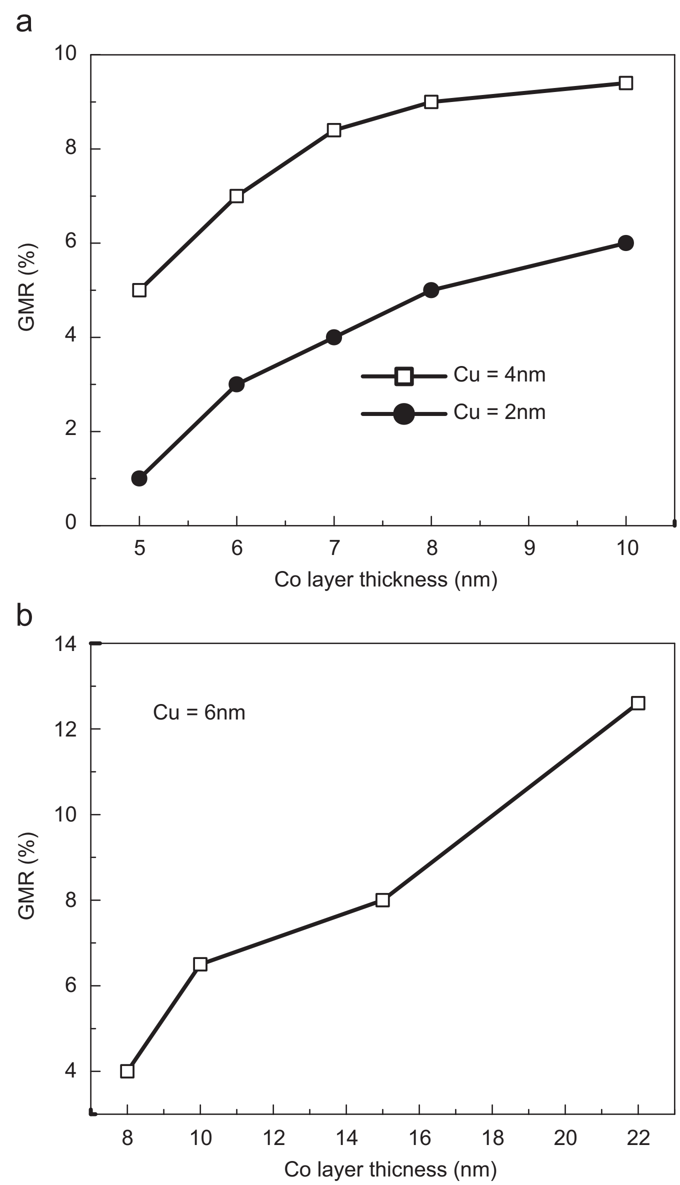

Fig. 2. Variation in magnitude of GMR with thickness of Co layer: (a) $t_{Co} = 5$–10 nm and $t_{Cu} = 2$ and 4 nm and (b) $t_{Co} = 8$–22 nm and $t_{Cu} = 6$ nm.

was varied in two different ranges, 5–10 nm for lower t_{Cu} of 2 and 4 nm and 8–22 nm for higher t_{Cu} of 6 nm, respectively. An increase in GMR with increasing thickness of Co layer was observed in all the sets of multilayers, where the thickness of Cu layer was kept fixed at 2, 4 and 6 nm. This achievement of appreciable GMR and its increase with Co layer thickness indicates that there is no bulk or FM contribution from the Co layers. Even for thicker Cu layers there exists sufficient AF coupling between the adjacent Co layers. It also indicates that the intermediate Cu spacer layer is continuous to the extent of giving high enough GMR. If the Cu layer was not uniform or was discontinuous, Co layers would have exhibited bulk behavior, with negligible or no GMR.

Though both the samples exhibited appreciable GMR, a remarkable difference was observed in their behavior regarding the achievement of saturation in magnetoresistance and the saturation magnetic field. It is observed from Fig. 2(a) that for the samples having t_{Cu} of 2 and 4 nm, the GMR tends to saturate as t_{Co} increases to about 10 nm, whereas Fig. 2(b) shows no saturation tendency for GMR even as the t_{Co} values approach 22 nm and magnetic field approaches till 8 kOe, though after 3 kOe the change in GMR was quite gradual. Figs. 3(a) and (b) show the variation in saturation field with Co layer thickness, for two set of multilayers having $t_{Cu} = 4$ and 6 nm. It is observed that saturation magnetic field initially increases in all cases with increasing Co layer thickness. But for thinner Co (5–10 nm) and Cu (2–4 nm) layers the saturation fields as low as 0.5–1.2 kOe are achieved. All the ML samples having Co layer thickness in this range exhibited saturating behavior of GMR. For the case of $t_{Cu} = 6$ nm, the saturation field continues to increase beyond 1 kOe with Co layer thicknesses increasing beyond 10 nm. The ML samples having thicker Co layers in the range 8–22 nm had higher saturation fields and exhibited a non-saturating behavior of GMR. The attainment of higher saturation fields, with increase in Co layer thickness can be explained on the basis of surface roughness of individual layers. It was observed that with increase in layer thicknesses, the roughness of individual layers and hence the average roughness of ML increases. This surface roughness seems to induce granular like behavior in the MLs, exhibiting superparamagnetic (SPM) behavior and in turn causing them to either show a non-saturating behavior or exhibit higher saturation fields. Moreover, it can also be said that, larger is the thickness of Co layer; stronger will be the coupling between two adjacent Co layers. Thus higher fields will be required to saturate the GMR values. But at present we do not have any further support or explanation for this reasoning.

The Co layers can be considered to have both ferromagnetic and SPM regions within it. So, a conduction electron crossing through the Cu spacer layer can travel from one FM region to another FM or SPM region or between both SPM regions. In case, the conduction electron encounters only FM regions, it is expected that the GMR magnitude will be low with lower saturation fields. For MLs having thin Co layers, if the layers are continuous, this is expected. Also because of lower surface roughness, FM regions are more prominent than SPM regions, giving rise to lower GMR and low saturation fields. For the other two paths where SPM regions are also involved, the MLs are expected to have prominent granular behavior, thus exhibiting either a non-saturating behavior or having high fields of saturation. Thicker Co layers are expected to have some SPM regions along with FM regions, because in electrodeposited MLs due to dissolution and exchange reactions during deposition, we cannot expect to have very smooth and continuous films as we go for higher thicknesses, i.e. deposition done for larger times.

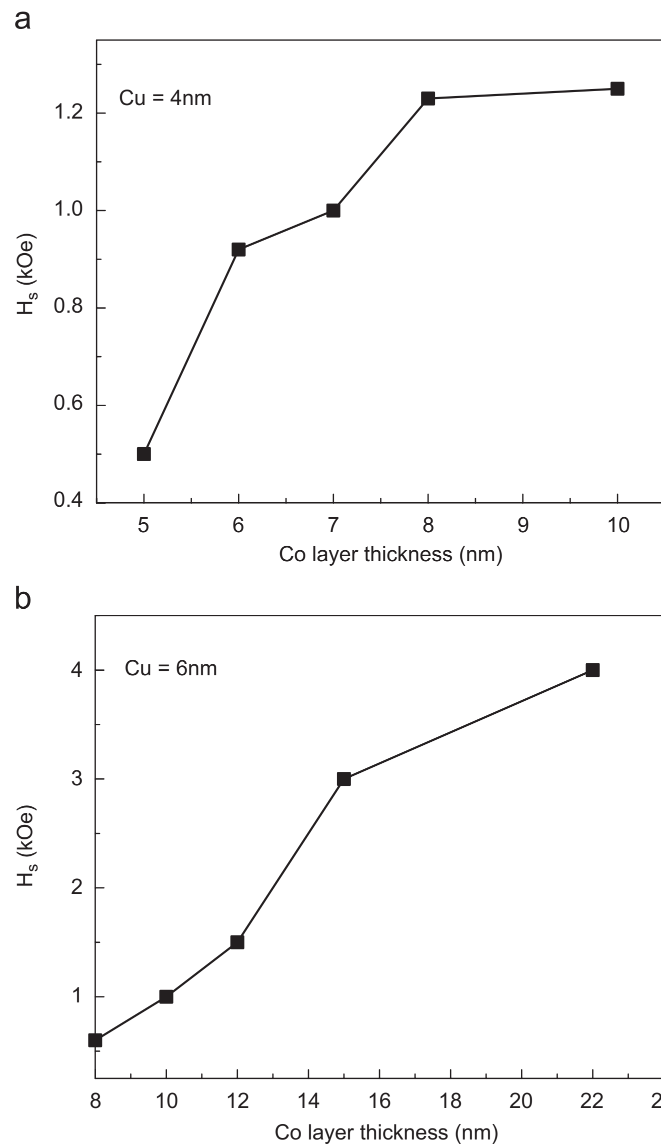

Fig. 3. Variation in the saturation field values for two sets of samples. (a) $t_{Cu} = 4$ nm and t_{Co} lying in the range of 5–10 nm and (b) $t_{Cu} = 6$ nm and t_{Co} lying in the range of 8–22 nm.

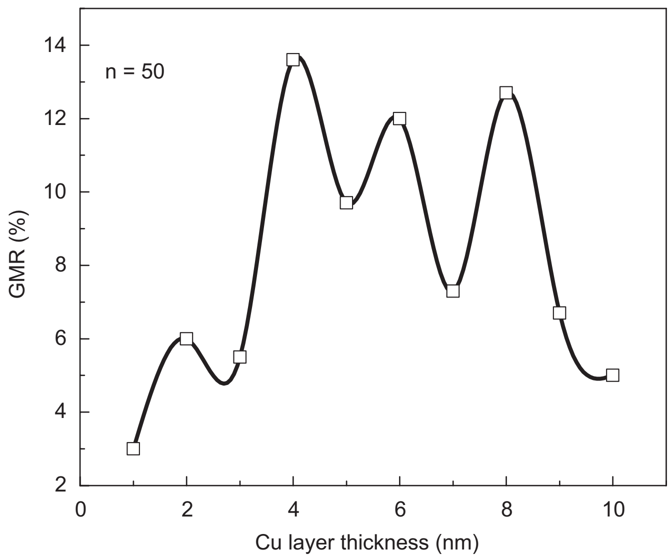

Fig. 4. Variation in the magnitude of GMR with thickness of Cu layer for $n = 50$.

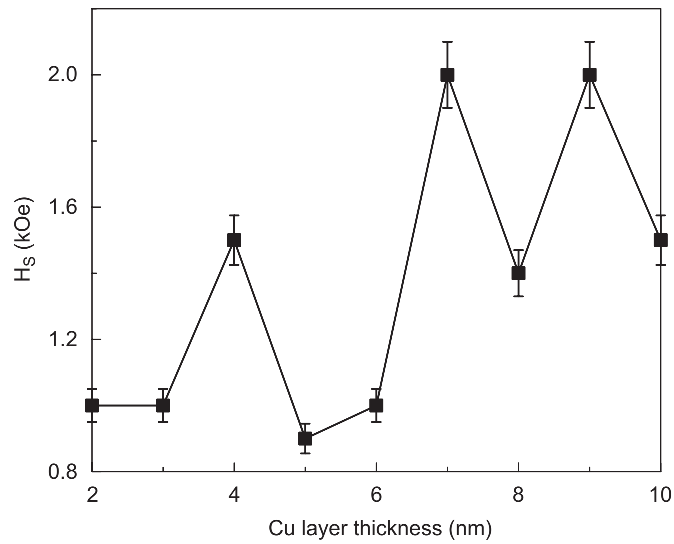

Fig. 5. Variation in the saturation field H_S with thickness of Cu layer for $n = 50$.

2.2. Effect of Cu layer thickness

Fig. 4 shows the variation in GMR as the thickness of Cu layer is changed from 1 to 10 nm, keeping the Co layer of approximately same thickness (20 nm). The number of bilayers was kept 50. It was observed that GMR exhibits oscillations in its magnitude as the thickness of Cu layer was varied. The peak in GMR values were seen for Cu layer thickness of about 2, 4, 6 and 8 nm; though at 4, 6 and 8 nm the GMR values were high in the range of 12–14% and appeared to be saturated. At intermediate values of t_{Cu} lower values of GMR were obtained, suggesting an oscillatory behavior of GMR. In electrodeposited films, much of the previous work shows absence of oscillatory behavior, due to structural imperfections [4,11–13], whereas some reports [14,15] show the presence of oscillatory behavior. Even in our case, the observed oscillatory trend is quite different from that observed by other workers [5,15–17]. The bilayer period obtained in our case is 2 nm, unlike to the 1 nm bilayer period obtained by other workers. This can be attributed to the higher surface roughness induced in the MLs by the initial surface roughness of ITO glass substrate. Also, we were unable to investigate the effect at intermediate Cu layer thicknesses. The oscillations obtained in GMR with Cu spacer layer variation, confirms that even at higher Cu thicknesses of the range of 10 nm, the Co layers are not entirely decoupled. Parkin et al. [5] have stated that Co/Cu structures show the evidence of AF coupling for thicker Co layers ranging from 0.25 to more than 20 nm, and also large GMR values are possible for Cu layers more than 20 nm thick. Actually from our magnetization studies of these samples we find that for Cu spacer layer thickness of 4, 6 and 8 nm the *M–H* loop shows signatures of AF coupling and at all other Cu layer thicknesses *M–H* loop is FM in nature. Fig. 5 shows the variation in saturation field value with changing Cu layer thickness. The oscillations in the values of saturation fields indicate the switching in the coupling from antiferro to ferromagnetic. It can be seen that the oscillations in GMR and saturation field are correlated.

3. Conclusions

Electrodeposited stacks of alternate Co and Cu layers, of thicknesses ranging from 2–20 nm, comprising of 50 bilayers exhibited appreciable high GMR up to 12–14%. GMR was found to increase with increase in Co layer thickness in the range 5–20 nm. The saturation field values were also found to increase with the increase in Co layer thickness. However, stacks with thinner Co and Cu layers exhibited low saturating fields that saturate to a value of about 1 kOe as Co thickness reaches 10 nm. For thicker Co and Cu layers a non-saturating behavior in H_S and GMR is seen for fields up to 4 kOe. The GMR and saturation field values were found to exhibit oscillatory behavior with Cu layer thickness in a correlated manner. The *M–H* behavior clearly shows antiferro coupling for Cu thicknesses of 4–8 nm. Thus it is successfully established that by controlling the thicknesses of individual Cu and Co layers, the magnitude of GMR as well as saturation field values can be tuned in a controlled manner.

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