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To cite this article: Yuan-fu Chen *et al* 2008 *J. Phys.: Condens. Matter* **20** 452202

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Photoresist-buffer-enhanced antiferromagnetic coupling and the giant magnetoresistance effect of Co/Cu multilayers

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Received 17 July 2008, in final form 27 August 2008

Published 8 October 2008

Online at stacks.iop.org/JPhysCM/20/452202

Abstract

By introducing a photoresist buffer layer, the enhancement of giant magnetoresistance (GMR) values of Co/Cu multilayers deposited on oxidized Si substrates is up to around 365%. X-ray reflectivity measurements indicate that the interfacial roughness of Co/Cu bilayer stacks buffered with a photoresist layer is lower than that on bare oxidized Si substrates, although their surface roughnesses are similar. Magneto-optical Kerr effect hysteresis loops of $(\text{Co/Cu})_N$ multilayers show that the antiferromagnetic coupling strength and fraction were significantly improved after photoresist buffering for all samples with N ranging from 8 to 50. The interface smoothening of photoresist-buffered multilayers may therefore contribute to such an enhancement, which in turn increases the corresponding GMR values.

(Some figures in this article are in colour only in the electronic version)

The giant magnetoresistance (GMR) effect [1, 2] is widely applied in read heads of hard disk drives or in nonvolatile memory devices [3]. Additionally, the understanding of GMR phenomena has helped to initiate the development of magnetoelectronics (also known as spintronics) [4]. Although different GMR multilayer (ML) systems have been extensively investigated in the past few years, some ambiguity still persists regarding the GMR effect on the interface roughness, preferred crystal orientations of MLs, and the antiferromagnetic coupling [5, 6]. Previous reports showed that the roughness (or interface roughness) would obviously influence the GMR effect and the relation between the roughness and GMR have been investigated in Fe/Cr [7–9], Co/Ag [10], and Co/Cu [11, 12] GMR multilayers systems. Intentionally introducing a buffer layer is another good approach to

investigate the effect of the interface roughness, preferred crystal orientations on the GMR effect. The effects of metallic buffer layers (e.g. Au, Cr, Fe, Cu, Co, Ta, or Al) on structures and magnetotransport properties of GMR MLs have been intensively studied and reported in the literature [6, 13–17]. For example, Bouziane *et al* investigated the buffer effects of Fe, Cr, Cu, etc on the texture and interfacial roughness of grown layers, as well as magnetic and transport properties of Co/Cu MLs [6]; Breidbach *et al* reported the proximity effects of a Au buffer layer on the GMR effect of epitaxial Fe/Cr/Fe(001) trilayers [13]. However, there are few reports that use polymers as buffer layers. Recently, Chen *et al* observed that the photoresist (PR) buffer layer can significantly enhance the GMR effect of Co/Cu MLs on plastic substrates due to the obvious smoothness effect of the photoresist on the rough surface of the plastic substrate [18]. However, further study is necessary to clarify the PR-buffering effect

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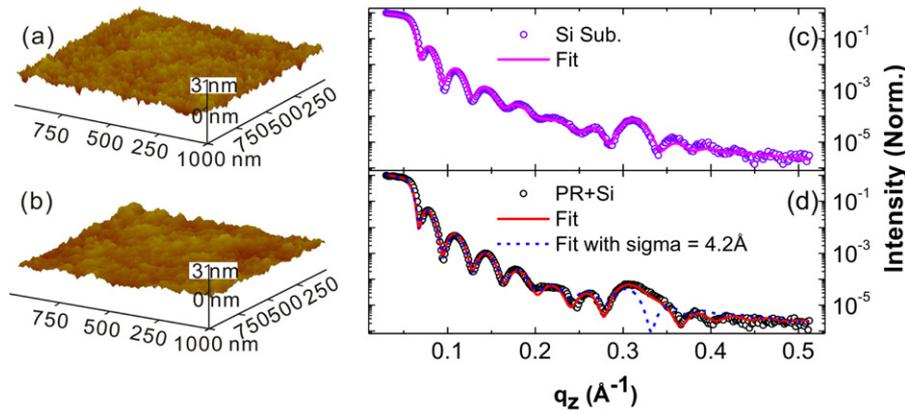


Figure 1. 3D AFM images of (a) bare oxidized Si and (b) PR-buffered oxidized Si substrates. A comparison of experimental and fitted XRR data for $(\text{Co/Cu})_8$ MLs deposited on (c) bare and (d) PR-buffered oxidized Si substrates.

on other factors like the interfacial roughness, magnetic coupling strength, as well as the GMR. In order to isolate the effects of interface smoothening, conventional oxidized Si substrates were used to study the PR-buffering effect on interfacial roughness, interlayer antiferromagnetic coupling strength and fraction, and magnetoresistive property of Co/Cu MLs. The $(\text{Co}_{1 \text{ nm}}/\text{Cu}_{1 \text{ nm}})_N$ ($N = 8\text{--}50$) ML system was adopted, where the Cu thickness corresponds to the first antiferromagnetic (AFM) coupling maximum and is associated with the strongest magnitudes of GMR.

In this communication, we report that the interfacial roughness of the Co/Cu MLs decreases after PR buffering, although their surface roughnesses on the Si and PR-buffered Si substrates are similar. The enhancement of GMR values of Co/Cu multilayers (MLs) on Si substrates is up to 365% after introducing a photoresist buffer layer. Such strong GMR enhancement might be explained as the decrease in the interfacial roughness of Co/Cu MLs and thus as the increase in the antiferromagnetic coupling strength and fraction as well as GMR.

The $(\text{Co}_{1 \text{ nm}}/\text{Cu}_{1 \text{ nm}})_N$ MLs in this study were deposited by dc magnetron sputtering of Co and Cu targets, respectively. Following a 1 nm Co bottom layer directly deposited on oxidized Si substrates with and without PR buffering, N periods of Co_{1 nm}/Cu_{1 nm} bilayers were deposited. Except where stated differently, the notation $(\text{Co/Cu})_N$ MLs denotes N periods of Co_{1 nm}/Cu_{1 nm} bilayers. The base pressure of the sputter system was 1×10^{-5} Pa. Argon with a purity of 99.998% was introduced as a working gas. The sputter pressure was 0.5 Pa and the deposition rate of Co was fixed at $\sim 0.05 \text{ nm s}^{-1}$, while the deposition rate of Cu was adjusted in the region of $0.02\text{--}0.12 \text{ nm s}^{-1}$ by changing the power for the Cu target. Thermally oxidized Si (Si) and photoresist-buffered thermally oxidized Si (PR + Si) substrates were used. About 2 μm -thick photoresist buffer layer was prepared by spin coating AR-P 3510 positive photoresist (Allresist, Germany) on thermally oxidized Si substrates at a rotation speed of 3500 rpm. The PR-buffered substrates were subsequently put on a hot plate and soft baking was performed at 95 °C for 1 min. Several series of $(\text{Co/Cu})_N$ MLs with various bilayer number N of

8, 10, 15, 20, 30, to 50 were prepared on thermally oxidized Si and photoresist-buffered oxidized Si substrates. In order to allow a direct comparison with the GMR of $(\text{Co/Cu})_N$ MLs, we deposited MLs on oxidized Si substrates with and without PR buffer layers in a single run to exclude the effects from different deposition conditions. AFM images of bare and PR-buffered Si substrates were obtained by a Nanoscope III AFM working in tapping mode, allowing us to directly measure the surface roughness. X-ray reflectivity (XRR) data were measured on a Bruker-AXS D8 discover system equipped with a four-bounce Ge (022) monochromator. Since Co and Cu layers have about the same electronic density, XRR measurements provide an average interface roughness for the whole ML stack. Magneto-optical Kerr effect (MOKE) hysteresis loops were measured in a home-made MOKE system in an in-plane configuration under a maximal magnetic field of ± 0.1 T. The conventional four-point technique and current-in-plane configuration were used to measure the GMR of Co/Cu MLs at room temperature under 0.2 T. The GMR ratio was calculated by $(R_0 - R_H)/R_H$, where R_0 is the maximum resistance near zero magnetic field, and R_H is the resistance at high magnetic field (0.2 T).

The morphologies of the surface of bare Si and PR-buffered Si substrates were measured by AFM, and corresponding three dimensional (3D) AFM images are shown in figures 1(a) and (b), respectively, leading to very close roughness values of ~ 2.2 and ~ 2.1 Å, respectively. The interfacial roughness of Co/Cu MLs can be extracted by fitting the XRR experimental data using the Parratt formalism [19]. Experimental and fitted XRR data of $(\text{Co/Cu})_8$ MLs deposited on bare oxidized Si and PR-buffered oxidized Si substrates are shown in figures 1(c) and (d) as a function of $q_z = (4\pi/\lambda)[\sin\theta]$, where λ is the x-ray wavelength (copper $K\alpha$, $\lambda = 1.541$ Å) and θ is the sample tilt angle in specular conditions. From the experimental XRR data, one can find that well defined deep minima are observed for both samples which indicate good qualities of Co/Cu MLs on both substrates.

The total layer thickness extracted from the fits of figures 1(c) and (d) correspond to the nominal deposition within an error bar smaller than 3%. The most remarkable difference between both measured curves is in the

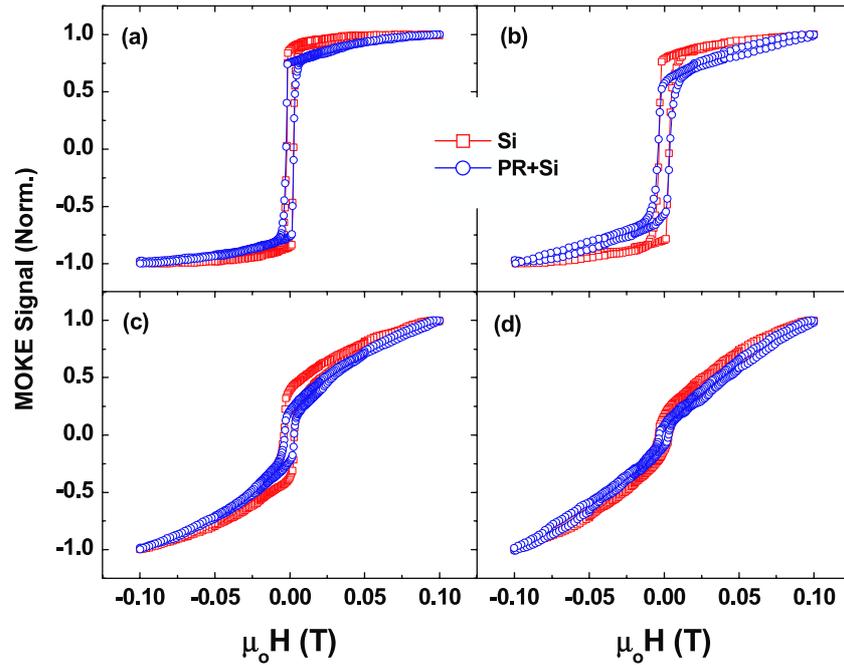


Figure 2. MOKE hysteresis loops for $(\text{Co/Cu})_N$ deposited on bare (squares) and PR-buffered (circles) oxidized Si substrates with $N = 8$ (a), 10 (b), 15 (c), and 20 (d).

superstructure peak observed at $q_z = 0.31 \text{ \AA}^{-1}$. A box-shaped peak is observed at the PR-buffered sample, while a symmetric peak is seen at the SiO_2 sample. From the simulations performed for the samples with and without PR we found an average interfacial roughness of 2.8 \AA and 4.2 \AA , respectively. The box-shaped superstructure peak can only be retrieved by assuming a smaller roughness of Cu/Co interfaces for the PR-buffered layer stack. Similar differences between the superstructure peaks for different N (not shown here) indicate that PR buffering leads to a decrease in the interfacial roughness of the Co/Cu stack. For a cross-check and to confirm our interpretation, we assume an interfacial roughness of 4.2 \AA to model the XRR curve from the sample with PR buffer layer (dashed blue curve in figure 1(d)), which obviously cannot fit our XRR data as well as the curve with 2.8 \AA roughness.

Because only antiferromagnetically coupled regions of the MLs can contribute to the magnetoresistance, the antiferromagnetic coupling fraction (AFF) is generally used to quantify the fraction of MLs with antiparallel alignment of adjacent film magnetizations at a zero external magnetic field. The AFF is given by [20, 21]

$$\text{AFF} = 1 - \frac{M_R}{M_S}, \quad (1)$$

where M_R and M_S are the remnant and saturation magnetizations, respectively. Generally, M_R and M_S can be obtained from the magnetic hysteresis loops or MOKE hysteresis loops [20].

In order to investigate the interlayer antiferromagnetic coupling strength and fraction, MOKE hysteresis loops were measured for $(\text{Co/Cu})_N$ MLs with N ranging 8–50. Figures 2(a)–(d) show a comparison of selected normalized

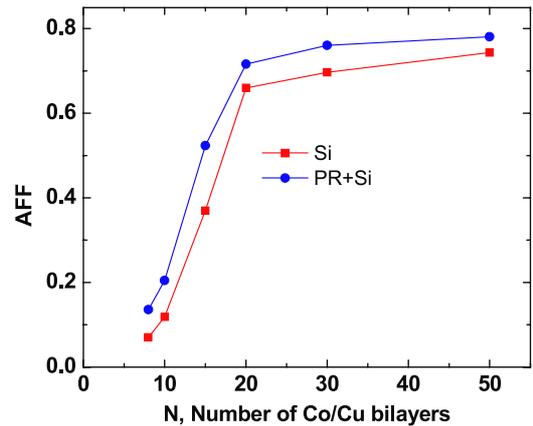


Figure 3. Bilayer number dependence of antiferromagnetic coupling fraction of $(\text{Co/Cu})_N$ MLs deposited on bare (squares) and PR-buffered (circles) oxidized Si substrates.

MOKE hysteresis loops for $(\text{Co/Cu})_N$ MLs deposited on bare and PR-buffered Si substrates with N of 8, 10, 15, and 20, respectively. The remarkable feature in figures 2(a)–(d) is that the remnant magnetization of MLs with PR buffering is obviously lower than that of corresponding MLs without PR buffering. It directly indicates that the stronger antiferromagnetic coupling occurs in MLs with PR buffering. In addition, the antiferromagnetic coupling is stronger with increasing bilayer number for both cases with and without PR buffering. Furthermore, the AFF values can be calculated from MOKE hysteresis loops using equation (1).

The AFF as a function of the bilayer number N for Co/Cu MLs deposited on bare and PR-buffered Si substrates was calculated (shown in figure 3). With N ranging from 8 to 50, the AFF increases with the bilayer number. Additionally,

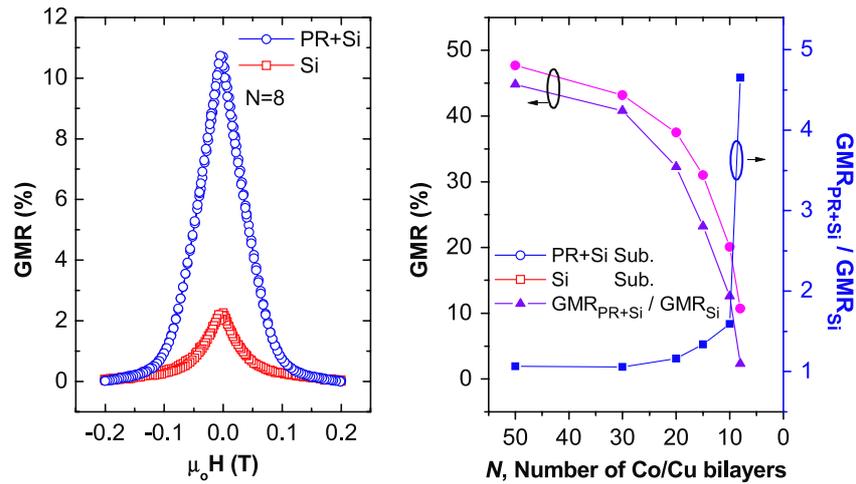


Figure 4. (a) GMR of $(\text{Co}_{1 \text{ nm}}/\text{Cu}_{1 \text{ nm}})_8$ multilayers deposited on bare and PR-buffered oxidized Si substrates; (b) effect of the number of (Co/Cu) bilayers N on GMR values and GMR ratios of Co/Cu MLs on Si and PR + Si substrates.

the AFF values of MLs with PR buffering are systematically larger than those without PR buffering. We think that such AFF enhancement may originate from the decrease in the interfacial roughness.

Figure 4(a) shows GMR values of $(\text{Co}/\text{Cu})_8$ MLs on Si and PR + Si substrates, respectively. It was found that $(\text{Co}/\text{Cu})_8$ MLs on Si have a smaller GMR (2.3%) compared to MLs on PR + Si (10.7%), which is about 365% larger than that of MLs deposited on bare oxidized Si substrate. Figure 4(b) presents the effect of the number of (Co/Cu) bilayers N on GMR values and GMR ratios. We find that for each N , GMR values of Co/Cu MLs deposited on bare oxidized Si substrates are systematically lower than those deposited on PR-buffered oxidized Si substrates. The ratio between GMR values for Si and PR + Si substrates is also shown in figure 4(b) as a function of the bilayer number N . With increasing N , the ratio decreases and approaches one for 50 bilayers deposited. The larger ratio observed for the low N values suggests that the Co/Cu ML quality is related to the reduced interfacial roughness in MLs with PR buffering. Such phenomena may be related to the PR buffer surface tension and softer elastic properties, which allows for a smoother accommodation of the initial sputtered layers. As N gets large, the effect of the Co/Cu interfaces for the overall roughness becomes increasingly important with respect to the substrate roughness from the starting Si or Si + PR interface. While in samples with lower N the average interface roughness could be mainly tied to the quality of the initial substrate interface (SiO_2 or PR), for larger N values it depends on the intrinsic roughness of Cu/Co interfaces obtained by sputtering that may converge to a saturation value. Therefore, MLs with large N have $\text{GMR}_{\text{PR+Si}}/\text{GMR}_{\text{Si}}$ ratios closer to unity. Another possible explanation of the PR-buffer-enhanced GMR might be attributed to the effect of strain-induced interface improvement after PR buffering: such an improvement effect would be dominant for low N samples while becoming faint with N increasing. A similar buffer-enhanced GMR effect was also observed on flexible plastic substrates [18]. Based on the discussion above, we consider that the PR buffer layer can

decrease the interfacial roughness of Co/Cu MLs and thus might improve the antiferromagnetic coupling strength and fraction of Co/Cu MLs, which leads to the increase of the GMR effect.

In summary, the enhancement of GMR values of Co/Cu MLs is up to around 365% by introducing a photoresist buffer layer. We find an improved interfacial roughness of Co/Cu MLs after PR buffering compared to that in the MLs without PR buffer layers, while MOKE hysteresis loops prove that the antiferromagnetic coupling strength and fraction are significantly enhanced. Such GMR enhancement of Co/Cu MLs can possibly be explained by the decrease in the interfacial roughness and the increase in the antiferromagnetic coupling strength and fraction.

The authors would like to thank I Fiering, B Eichler, and C Krien for their technical assistance.

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