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Electrodeposition and Characterization of Co/Cu Multilayers

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Co/Cu multilayers having different bilayer number (total thickness) were electrodeposited on polycrystalline Cu substrates with a strong [100] texture from an electrolyte including Co and Cu ions under potentiostatic control. The structural data from X-ray diffraction (XRD) revealed that all films have face-centered cubic (fcc) structure, but their crystal textures change from [100] to [111] as the bilayer number increases. The magnetic analysis by vibrating sample magnetometer (VSM) showed that the magnetic moment per volume decreases as the bilayer number increases. Magnetoresistance (MR) measurements were made at room temperature in the magnetic fields of ± 12 kOe using the Van der Pauw (VDP) method with four probes. The samples with the bilayer number less than 111 exhibited giant magnetoresistance (GMR) with a negligible amount of anisotropic magnetoresistance (AMR), while the ones with the bilayer number larger than 111 have pure GMR effect.

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1. Introduction

After the discovery of giant magnetoresistance (GMR), magnetic multilayers have attracted much attention due to their potential applications in magnetoresistive sensors and read heads [1–3]. One of the most common techniques to grow such multilayered structures is electrodeposition, which is a simple and low cost one. The properties of electrodeposited multilayers can be affected by chemical parameters such as the electrolyte pH, concentration and the deposition potentials as well as physical parameters such as layer thicknesses, total thickness (bilayer number, N) and substrate [4]. Co/Cu multilayers are one of the systems that exhibit high GMR effect, since there is coherency of the electronic band and lattices structure of Co and Cu [5]. The similarity of the electronic band structure of majority spins in Co to that of Cu allows high transmission of majority spins at the interface of Co and Cu layers. The large band mismatch between minority spin in Co and Cu causes poor transmission of minority-spin electrons across the Co/Cu interface [5]. However, for electrodeposited Co/Cu multilayers, the widespread problem is the dissolution of Co atoms during Cu deposition. Preferred dendritic or columnar growth of Co causes dissolution [6]. Thus Co layers are thinner than expected, and Cu layers become thicker. Mostly, it leads to GMR values lower than those in structures produced by vacuum techniques. In this study, structural, magnetic and magnetoresistance (MR) properties of Co/Cu multilayers were investigated as a function of the bilayer number. It was observed that the bilayer number has a significant effect on structural magnetic and MR properties of the multilayers.

2. Materials and method

Co/Cu multilayers were electrodeposited on polycrystalline Cu sheets of the face-centered cubic (fcc) structure, having a preferred [100] orientation. Experimental details were reported elsewhere [7]. The electrolyte used to grow multilayers was composed of 0.27 M CoSO_4 , 0.022 M CuSO₄, 0.022 M H₃BO₃ and 0.01 M NH₂SO₃H. The pH value of the electrolyte was 2.5 ± 0.1 at room temperature. The Cu layers were deposited at -0.4 V with respect to saturated calomel electrode (SCE). The Co layers were grown at -1.5 V versus SCE. The nominal thicknesses of Cu and Co layers were adjusted to be 1 and 8 nm respectively. The bilayer number for the multilayers with a N[Co(8 nm)/Cu(1 nm)] form was varied between 33 and 1111. It means that the total thicknesses of the samples changes from 0.3 μ m to 10 μ m. After growth, magnetic properties of the samples were analyzed on their substrate. For structural and MR measurements, they were peeled off from their substrates electrochemically and mounted on glass. For the structural characterizations, X-ray diffraction (XRD) technique was used. In order to determine the preferential orientation (PO), the texture coefficients were found from the experimental and theoretical relative integral intensities of the (hkl) reflections [8]. Magnetic analyses were performed by vibrating sample magnetometer (VSM). The hysteresis curves were obtained with magnetic field applied both parallel and perpendicular to the film plane. MR was measured using Van der Pauw (VDP) method at room temperature. The magnetic field in the range of ± 12 kOe was applied both parallel and perpendicular to the current flowing through the film plane, in order to find longitudinal magnetoresistance (LMR) and transverse magnetoresistance (TMR) respectively.

3. Results and discussion

Figure 1 shows the XRD diffraction patterns for N[Co(8 nm)/Cu(1 nm)] multilayers with different bilayer

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numbers between 33 and 1111. All peaks for each multilayer are observed at around 2θ positions of the main Bragg peaks of the face-centered cubic (fcc) structure, that is, multilayers have a polycrystalline fcc structure. For the multilayer with the smallest bilayer number, 33[Co(8 nm)/Cu(1 nm)], the [200] reflection appeared at $2\theta \approx 51^{\circ}$ was only detected. As the bilayer number increases, the detected peak number increases, that is, following the [200], first, the [111] peak begins to appear, and then the [220] and [311] reflections occur. Furthermore, the [111] peak becomes strong with increasing bilayer number. This suggests that thin multilayers adopt the orientation of Cu substrate while the orientation of the sample shifts to the PO of the bulk Cu as the sample gets thicker. The texture coefficients of the multilayers were calculated to determine the PO of the multilayers. The samples with the bilayer number less than 333 have the [100] PO. So these multilayers have the same orientation as in their substrates. In the multilayers having more than 333 bilayers, the orientation of [100] weakens and the [111] becomes strong.



Fig. 1. XRD patterns of multilayers with N[Co(8 nm)/Cu(1 nm)] as a function of bilayer number N.

The hysteresis curves of all multilayers were obtained. It was found that, the easy magnetization axes of the samples are in the film plane since the curves in the film plane saturate at a smaller magnetic field than in those with magnetic field perpendicular to the film. Figure 2a displays parallel hysteresis curves of N[Co(8 nm)/Cu(1 nm)] multilayers as a function of the bilayer number. As the bilayer number increases, the amount of the magnetic moment per volume decreases. In order to see clearly, the change from rectangular to sigmoid shape of the hysteresis curves, the hysteresis curves for some multilayers are given at low magnetic field, in the inset of Fig. 2a. For example, for 33[Co(8 nm)/Cu(1 nm)] multilayer, the shape of the curve is rectangular, but as the bilayer number increases the shape of the curve becomes sigmoid. It was reported that, the sigmoid shape arises from superparamagnetic (SPM) regions in the ferromagnetic layers, while rectangular hysteresis curves reflect effective ferromagnetic reactions [9]. Figure 2b shows the coercivity (H_c) of the samples as a function of the bilayer number. It increases up to bilayer number of 444, but beyond this it decreases slightly. The H_c values of the multilayers are larger than that of the bulk Co (20 Oe).



Fig. 2. (a) Hysteresis curves of N[Co(8 nm)/Cu(1 nm)] multilayers deposited with different bilayer number (N), (b) coercivity (H_c) values of the multilayers.

Figure 3 shows the evolution of the MR curves for N[Co(8 nm)/Cu(1 nm)] multilayers depending on the In Fig. 3a, for a multilayer with bilayer number. 33[Co(8 nm)/Cu(1 nm)], TMR and LMR decreases as the magnetic field increases, but LMR shows a little increase at low magnetic fields. This means that the multilayer exhibits GMR with a negligible amount of anisotropic magnetoresistance (AMR). The multilayers with the bilayer number more than 111 have the GMR behavior (Fig. 3b and 3c). As seen from the figures, the MR curves do not saturate fully even in large magnetic fields because of occurring SPM regions in ferromagnetic layers. The dependence of the MR ratio on the bilayer number is given in Fig. 3d. For the multilayers with the bilayer number more than 222, GMR decreases and then remains nearly constant.



Fig. 3. TMR and LMR curves of multilayer having (a) N = 33, (b) N = 222, (c) N = 667, (d) variation of the GMR in N[Co(8 nm)/Cu(1 nm)] multilayers with bilayer number.

4. Conclusions

A series of CoCu/Cu multilayers with different bilayer numbers was electrodeposited on polycrystalline Cu [100] substrates. The bilayer number was changed from 33 to 1111. The XRD results revealed that the number of the appeared reflections increases as the bilayer number increase. Also, it was found that the preferential orientation of the multilayers varies from [100] to [111]. From the magnetic data, it was determined that the easy magnetization axis of the samples is in the film plane. The multilayers with the bilayer number smaller than 111 exhibited the GMR behavior with a negligible amount of AMR, while the multilayers with larger bilayer numbers had predominantly GMR.

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