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Magnetoresistance Measurements on Electrodeposited Co_xCu_{1-x} Alloy Films

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Abstract

Co_xCu_{1-x} alloy films were prepared by using electrodeposition technique. The variations of Co and Cu contents of the films were investigated as functions of bath pH and Co content. The compositions of the alloy films were determined using an atomic absorption spectrophotometer. The crystal structures of the alloy films were analyzed using a Cu (Kα)-X-ray diffractometer. The diffraction lines observed were only those of copper component in the alloy films. All three films showed negative magnetoresistance and a giant magnetoresistance effect on the order of 1% in $Co_{0.26}Cu_{0.74}$ and 1.7% in $Co_{0.19}Cu_{0.81}$ alloy films at 100K. It was also detected that the magnetoresistance effect at first increased, then decreased with increasing Co content.

Key Words: Giant magnetoresistance; Electrodeposited alloy

1. Introduction

Giant magnetoresistive materials are used in sensor technology and magnetic read-heads. The observation of giant magnetoresistance (GMR) effect in multilayer films, e.g. in Fe/Cr, Co/Cu, has recently restarted the investigation of magnetoresistance in alloy films, although the magnetoresistance effect was observed in metals long ago. The GMR effect in alloy films is usually observed in systems composed of insoluble elements at room temperature. Co in Cu is an example of such a system, which tend not to make solid solution with each other at equilibrium conditions. The GMR effect was

detected in sputtered [1] and evaporated [2] CoCu alloy films. Electrodeposition is an alternative yet easy and relatively cheap method to prepare multilayers and alloy films for the investigation of their magnetoresistance (MR) effect. Most recently Co/Cu multilayer thin films [3,4,5,6], nanowires [7] and granular CoCu alloys [8] have been successfully electrodeposited. However, there is not much study on the electrodeposited CoCu alloy films.

2. Experimental

An acid-citrate bath with the composition $CuSO_4.5H_2O$(30 g/l), $CoCl_2.6H_2O$ (3,3 g/l), H_3BO_3 (6,6 g/l) and $CoSO_4.7H_2O$ (10 to 30 g/l) was used to electrodeposite Co_xCu_{1-x} alloy films. The effect of pH on the Co concentration in film was investigated by keeping the Co sulfate (10 g/l) and the other components (as given above) constant in the bath. The pH was adjusted by using hydrochloric acid. In order to obtain alloy films with different compositions, the cobalt sulfate component in the electrolyte was varied between 10 to 30 g/l while the amounts of the others were kept constant. The deposition was carried out at room temperature with a current density of 5 mA/cm^2. The cathode was an aluminum foil with a thickness of 0.5 mm while a platinum sheet was used as anode. Before each run, the cathode was mechanically polished, washed with distilled water, kept in a 10% solution of NaOH and rinsed in distilled water again. The aluminum substrate was subsequently dissolved away from the alloy film in a 10% NaOH solution. The compositions of the Co_xCu_{1-x} alloy films were determined by using an atomic absorption spectrophotometer. The thicknesses of the films were calculated to be $\sim$2.5 μm by using the film area, the amounts of Co and Cu in the film and the bulk densities of Co and Cu. The crystal structures of films were determined by using a Cu (Kα) X-ray diffractometer. The longitudinal (the current parallel to the magnetic field in the film plane) and transverse (the current perpendicular to the magnetic field in the film plane) magnetoresistance measurements on some selected CoCu alloy films were done with the traditional four point probe method in the temperature range of 100K and 320K. A constant current of 0.1 mA and a magnetic field of 0.8T were applied parallel to the film plane in the magnetoresistance measurements.

3. Results and Discussion

Three series of CoCu alloy films were obtained for the deposition times of 10, 30, 60 minutes by taking the bath components of $CoSO_4.7H_2O$ (10g/l) and the other components same as given in the experimental section above and by varying pH values between 3.5 and 6.5. A bright brown deposit was obtained between the pH values of 3.5-6.5. The pH values lower than 3.5 give mossy deposits. The variation of the Co concentration in deposit with pH is shown in Figure 1. The Co concentration in films is kept constant below pH=4.5 and above pH=5.5 while a gradual increase is observed between pH=4.5 and 5.5 for each deposition time.

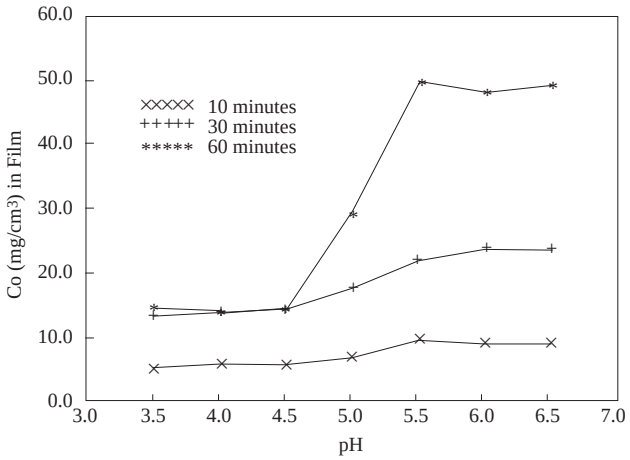

Figure 1. The variation of Co in the deposit as a function of pH for the deposition time of 10, 30 and 60 minutes.

Finding the highest Co concentration in the CoCu alloy film above pH=5.5, several CoCu alloy films were prepared by varying $CoSO_4.7H_2O$ between 10 to 30 g/l and keeping the other components constant in the bath, and using a current density of 5 mA/cm^2 and pH=6 for 60 minutes. Figure 2 shows the variation of the Co concentration in the deposit as a function of Co concentration in the bath. The Co content in the deposit increases with increasing Co content in the bath and it just reaches around 26% when the Co content in the bath is about 70%. This may be due to the insufficient negative potentials that may not allow the deposition of more Co.

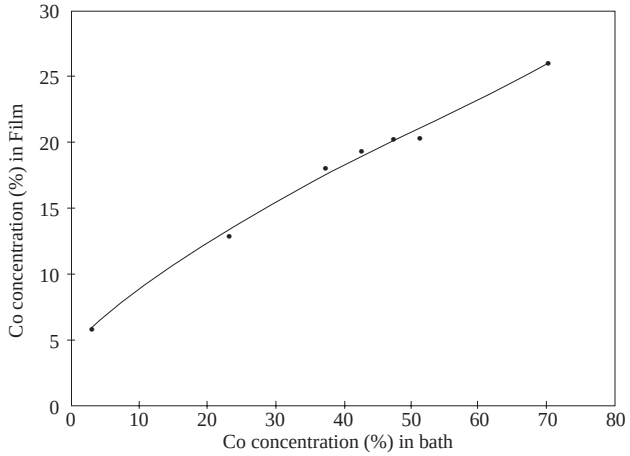

Figure 2. The variation of Co concentration (%) in the deposit with Co concentration (%) in the bath.

Looking at the y-axis, which gives us the Co concentration percentages in the deposit, one might say that the copper content has a preference in the deposition process, even above the Co content of 70% in the bath. The increasing behavior of Co concentration in the deposit with increasing Co concentration in the bath is also reported in some acidic-citrate bath studies [8], with an exception that the curve in the similar graph of [8] is almost parabolic.

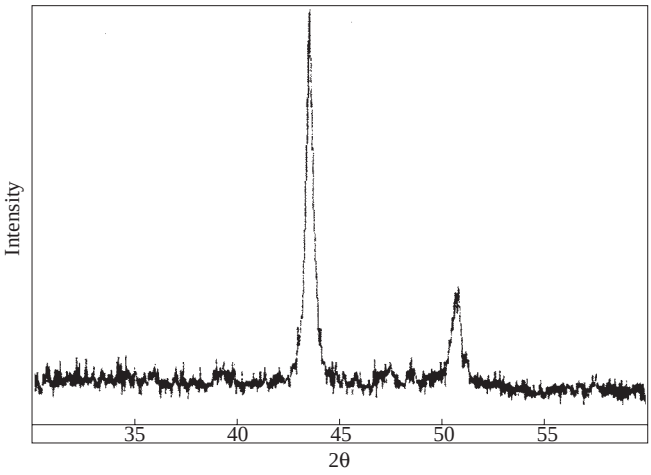

Figure 3. An example of the X-ray diffraction spectrum of the $Co_{0.26}Cu_{0.74}$ alloy film.

Figure 3 shows an example of X-ray diffraction spectrum of $Co_{0.26}Cu_{0.74}$ film. Both lines in the spectrum belong to the diffraction lines of face centered cubic (FCC) structure of copper. It was detected that the diffraction angles of the alloy films shifted toward higher angles as the Co content in the alloy films increased. The crystal lattice constant a for each film was calculated by using the FCC-Cu diffraction angles θ and the Bragg equation. The lattice constants a of the alloy films were determined to be 3.624 Åfor the $Co_{0.06}Cu_{0.94}$ film, 3.611 Å for the $Co_{0.19}Cu_{0.81}$ film, and 3.599 Å for the $Co_{0.26}Cu_{0.74}$ film. The variation of crystal lattice constant with the Co concentration in the films is shown in Figure 4.

As it can be seen from the figure, the lattice constant a decreases with increasing Co percentage in the Co_xCu_{1-x} film. Similar decrease in the lattice constant is also reported in [2,8]. This reduction is in agreement with the Vegard's rule, which expresses that the lattice parameter is proportional to the atomic percent of solute present in the continuous solid solution alloys. This gives us a clue that the alloy films prepared in this study may be regarded as a solid-solution.

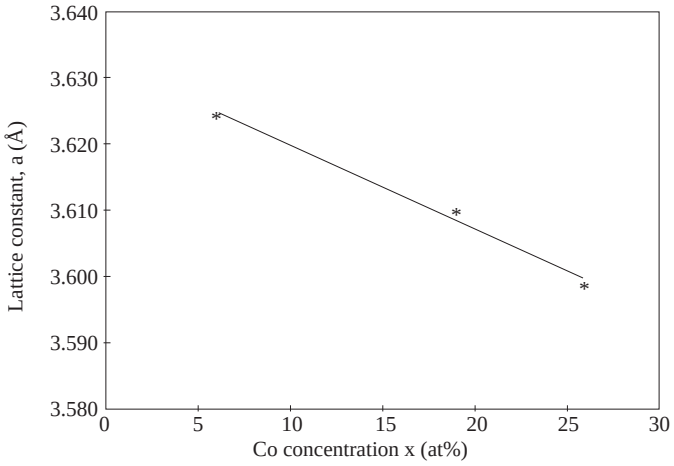

Figure 4. The variation of crystal lattice constant with Co concentration (%) in the CoCu alloy films.

The resistivity of an alloy exhibiting magnetoresistance effect has three components: i) temperature independent contribution from defects; ii) temperature dependent phonon contribution; and iii) temperature dependent contribution due to the magnetoresistance (MR) effect. The relative magnitude of contribution (iii) to resistivity may be deduced by measuring magnetoresistance effect at different temperatures [9]. The magnetoresistance ratio (MRR) is described as ΔR/R=[R(0)-R(H=0.8T)] /R(H=0.8T), where R(H=0.8T) and R(0) are the electrical resistances of the Co_xCu_{1-x} films measured with and without the existence of magnetic field, respectively. A negative isotropic MR was detected in both measurements taken with a current parallel and perpendicular to the magnetic field for each film. Similar MR results were reported for CoCu films [8] and for the granular CoAg films [9]. The variation of the MRR of the three alloys with temperature is shown in Figure 5. The MRR of $Co_{0.06}Cu_{0.94}$ alloy at 320K is around 0.1%. It does not change much as the temperature of the sample reduces toward 100K. The MRR values of $Co_{0.26}Cu_{0.74}$ and $Co_{0.19}Cu_{0.81}$ films increase as the sample temperature decreases toward 100K, and the MRR reaches to about 1% for $Co_{0.26}Cu_{0.74}$ and about 1.7% for $Co_{0.19}Cu_{0.81}$ at 100K.

The MRR values of 1% and 1.7% for $Co_{0.26}Cu_{0.74}$ and $Co_{0.19}Cu_{0.81}$ films at 100K, respectively, are the values in the range of so called Giant Magnetoresistance Ratio. The magnetisation of a ferromagnetic sample increases with decreasing temperature. This means that the ferromagnetic coupling between the magnetic moments of magnetic entities becomes stronger with decreasing temperature, which results in decreasing the number of spin waves. The electron-magnon scattering becomes less effective as the temperature decreases [10]. The increase in the MRR as the temperature decreases in Figure 5 is therefore supposed to be due to the decreasing electron-magnon scattering in our samples.

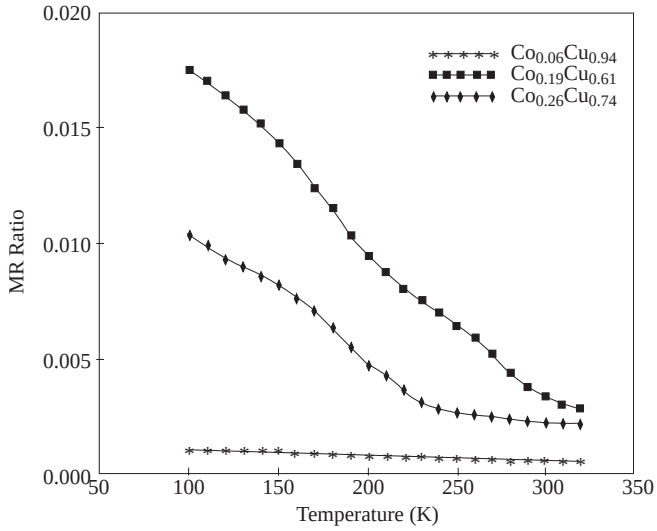

Figure 5. The variation of the magnetoresistance ratio of the Co_xCu_{1-x} alloy films with temperature.

The MRRs of the three films also change differently with decreasing temperature. The MRR of the $Co_{0.19}Cu_{0.81}$ film has the highest value among the three films studied at temperatures between 320K and 100K. In other words, Figure-5 indicates that the MRR values initially increase and then decrease with increasing Co percentage in the films. It is reported that the grain size in an alloy film affects the MRR, and the annealing process changes the grain size and the MRR of alloy films [8,9,11]. The ferromagnetic or antiferromagnetic coupling among the magnetic entities in magnetic films also plays the major role in the magnitude of MR. The conduction electron scattering is more at the antiferromagnetic coupling environment than the ferromagnetic coupling environment. The more antiferromagnetic coupling in the magnetic films results in the bigger spin scattering and therefore the bigger magnetoresistance effect occurs because the conduction electrons often find themselves in very different potentials in the antiferromagnetic coupling environment. The grain size and the magnetic couplings in our films may be different from each other. This may be the reason for the different MRR observed at the same temperature in our three samples. Our next investigations are therefore going to be on finding out the ferromagnetic/antiferromagnetic ratio in our films and the MR properties of the annealed CuCo alloy films with the variation of applied field.

Acknowledgments

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