Effect of strain on the magnetotransport properties of Co/Ag multilayer films grown by pulse electrochemical deposition

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Abstract:

We report magnetoresistance effect of nano-order scale $[Co1.0 \text{ nm/Ag} (t_{Ag})]_{20}$ ferromagnetic multilayer and alloy films grown with pulse electrochemical deposition on top of a thin conducting layer of Cu (15 nm). The Cu-buffer layer was grown on a polyamide substrate (1cm²) and annealed. A strain was applied mechanically to study the electrical and magnetic characteristics as a function of the ferromagnetic layer thickness. An induced magnetic anisotropy was observed in the entire films. The magnetoresistance ratio $[R_H-R_0]/R_0$ was ~9 % at 1 kOe. A remarkable difference of magnetic field dependence of magnetoresistance ratio was observed corresponding to the orientation of magnetization curves. We suggest that these effects are due to the spin-dependent scattering at interface and/or domain boundaries.

Keywords: magnetotransport properties, magnetic anisotropy, pulse electrodeposition, Co/Ag multilayer

1. Introduction

Currently, studies on the magnetic and electrical properties of nano-ordered materials created by the special means are being reported actively. Research metallic multilayer and alloy films is a typical example, and has attracted much attention not only as fundamental science but also in applications [1-2].

Pulse electrochemical deposition is a convenient method that makes it possible to control the film composition and thicknesses of the multilayers on an atomic scale by regulating the pulse amplitude and width [3–5]. There are no concrete explanation for the relationships between the magnetism and the magnetoresistance in ferromagnetic layers. We attempted to grow many Co/Ag multilayered films, systematically varying the layer thickness of Co and Ag, by utilizing pulse electrochemical deposition. This method is in use to grow thin films of granular alloy or multilayer of commonly immiscible metal exhibiting giant magneto-resistance (GMR) effects. In multilayer, by varying the thicknesses of the individual layers, choosing appropriate material composition, and inducing strain, it appears to be possible to tailor the magnetic anisotropy.

The anisotropy of the ferromagnetic layers depends on the strain and lattice mismatch between adjacent layers. We have attempted to grow electrodeposited multilayer and granular alloy films by a micro-computer with controlled pulse generator, and induce

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magnetic anisotropy in the lattice mismatched ferromagnetic layer by applying strain externally.

The objective of this paper is to report in detail, the results of our investigation of the magnetic and electrical properties of the as-deposited and strained ferromagnetic Co layer in the Co/Ag multilayer films.

2. Experimental

The electrolytic bath used to deposit Co/Ag multilayer film was CoSO₄.7H₂O, AgI, KI with Ag anode having pH of 3.0. The present work differs from our past in that substrates were constructed of 15 nm thin copper films, vapor deposited on the polyamide film as compared to glass. A multilayer film was grown by means of pulse electrochemical deposition from the specific solution. A single electrolyte containing the salts of two components (metals) of the multilayer film i.e., a salt of Co and Ag was employed to deposit the films by controlling the pulse wave output from microprocessor. The pulse current density was varied from 0.1 to 25 mA/cm². The layer thickness and composition were determined experimentally by employing several methods. These were microbalance, chemical methods, X-ray diffraction, flame emission spectroscopy, and energy dispersive X-ray analysis (EDAX). On average taken out of 10 samples, the precision range these multilayer thicknesses were within 3 %. These samples were subjected to external fields. We found the composition of the single ferromagnetic layer to be 92at%Co-8at%Ag. Magnetic anisotropy was deduced using a vibrating sample magnetometer. The easy axis of magnetization was along the perpendicular direction (inplane) of applied force or strain. The strain was applied mechanically by stretching the polyamide and its value was measured by using a strain gauge. Samples were measured in all four configurations [4]. The details of these measurement methods can be found in our previous paper reported elsewhere [4, 6]. The magnetic field dependence of MR ratio was examined by varying the relative direction between the H field and the current. The MR ratio was defined as MR% = $[(\rho(H) - \rho(H = 0)) \times 100\%]/\rho(H = 0)$. Measurements were performed at room temperature with the magnetic field in plane of the sample.

3. **Results and Discussions**

The magnetic anisotropy is the most frequently determined from magnetization measurements using a vibrating sample magnetometer or superconducting quantum interface device (SQUID), along two orthogonal directions of the magnetic field relative to the direction of strain. Examples of such measurements, with the field parallel (inplane) and perpendicular (in-plane) to the direction of strain using VSM, are shown in figures 1(a) and (b).

Fig. 1(a) shows the magnetic field dependence of the magnetization curves for the asdeposited film. The measured results of magnetization illustrate that the magnetization is randomly oriented, that is, the magnetism of the film is isotropic. Magnetization curves were measured along to the parallel (hard axis) and perpendicular (easy axis) relative to the directions of strain at room temperature. Fig 1(b) shows the magnetization curves following the introduction of strain. The induced uniaxial magnetic anisotropy was observed in all the multilayer films. The multilayer [Co 15 Å /Ag 15 Å]₄₀ showed a minimum hysteresis loss i.e., it showed the uniaxial anisotropy. The orientation of the induced anisotropy is perpendicular to the direction of strain.

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Fig.1a M-H hysteresis loops for [Co15 Å /15Ag Å] $_{50}$ multilayer films curves measured along to the parallel and perpendicular directions to the direction of current flow as-deposited film



Fig.1b M-H hysteresis loops for [Co15 Å /15Ag Å] $_{50}$ multilayer films curves measured along to the parallel and perpendicular directions to the direction of current flow for the strain-induced films at ϵ =1.5%

Additionally, a torque magnetometer was designed to measure anisotropy. Fig. 2 illustrates the anisotropic energy measurement method. To measure the anisotropic energy, a sample in the form of a disc was suspended in the magnetic field and a torque curve as a function of the measurement angle was derived. Torque T, which is a derivative of the anisotropy energy, corresponding to $E_A = K_u \sin^2\theta$ is given by, $T = -\delta E_A/\delta\theta = -K_u \sin^2\theta$. This measurement confirms the anisotropic behavior of these multilayer films.



Fig 2 Torque measurement and anisotropic constant as a function of rotation obtained from the torque measurement to deduce magnetic anisotropy in [Co15 Å/Ag 15Å] $_{40}$ films



Fig 3 Field dependence of mangetoresistance ratio at the range of $0 \sim 21$ kOe; Solid curves are drawn as a guide for the eyes.

Field dependent curves have been found to be very sensitive to both the Co layer thickness (t_{Co}) and Ag layer thickness (t_{Ag}) each at 15 Å with the stacking number of 40. Fig. 3 shows typical field dependent of magnetoresistance curves for [Co15 Å /Ag15 Å]₄₀. The field dependence of MR ratio for the oriented films with H parallel to the current are indicated by open circles. Similarly, and H perpendicular to the current are indicated by closed squares. For the isotropic film, the field dependence MR ratio does not depend on the orientation of current. It is indicated by open triangles, as labeled in the diagram. Solid lines are guide for the eyes. The negative field dependence tendency seems to be due to the GMR effect. It seems to be that the orientation of magnetization in the film has an effect on the GMR. For the anisotropy film, there is a difference between the values of field dependence measured by changing the direction of applied field against the measuring current



Fig. 4 Field dependence of set of magnetization curves for the Co layer thickness after strain is impressed from $0 \sim 1.5\%$ at room temperature.



Fig. 5 The relationship between anisotropy constant and Co layer thickness

Fig. 4 shows the magnetization as a function of the field for $[Co15 \text{ Å} /Ag15]_{40}$ multilayers with the strain varied from 0 to 1. 5%. Field dependence of the series of magnetization curves and the relationship between strain and the degree of anisotropy, β that was calculated using the experimental values of parallel and perpendicular remnant fields M_r // and $M_r \perp$ of the magnetization curves were reported previously [4].

Figure 5 shows K_u as a function of t_{Co} for a constant Ag layer thickness of 15 Å and Co layer thickness varied in the range of 5 ~ 30 Å, at room temperature with the H field present. The as-deposited films were magnetically isotropic and developed anisotropy upon introducing strain. An anisotropy constant was determined first by evaluating susceptibility using Stoner-Wolhfarth (S-W) model [7] under the assumption that the magnetization process of Co/Ag proceeds via a coherent rotation of magnetization at the magnetic field above 0.1 kOe. In the S-W model, the susceptibility, χ (dM/dH) at H=0 for polycrystalline film is given by Ms*sin² θ_0 /H_k where θ_0 is the angle between induced anisotropic uniaxial axis and external applied field. M_s is the saturation magnetization. The anisotropic field is therefore expressed as H_k= Ms*sin² θ_0 / $\chi_{H=0}$ when the angular distribution of domain magnetization is isotropic, sin² θ_0 = 2/3 and can be evaluated by using experimental values of M_s and $\lambda_{H=0}$ obtained by a slope of H_c. The MR ratio increase owing to the increase in anisotropy, K_u, which is calculated by using K_u= M_s H_k/2.



Fig. 6 Magnetoresistance effect against the anisotropy constant, K_u for the (a) as deposited and (b) strain-induced (unaxially-oriented) films

The correlation of MR ratio and K_u for the (a) H parallel to the magnetic hard direction and (b) H parallel to the magnetic easy direction is studied. Fig. 6 shows the magnetoresistance effect plotted as a function K_u . The MR ratio for the magnetic field parallel to the hard axis is larger than the magnetic field parallel to the easy axis. The reason for the showing the smaller MR ratio in the anisotropic sample is seems to be the

following. The number of antiparallel alignments of the magnetic spin between the ferromagnetic layers adjacent to non-magnetic layers seemed to be the major factor for showing the smaller MR ratio for the anisotropic (i.e., oriented) sample. As the orientation characteristic of magnetization in the film becomes strong, with easy axial direction and hard axial direction, there is a differencen in the slopes of MR ratio. The result suggests that magnetoresistance decreases with increasing K_u . The over all MR ratio with H parallel to easy axis (perpendicular to strain) is less significant than the field parallel to the magnetic hard axis (parallel to strain) for all the samples.

In summary we have presented series of studies on the electrochemically grown asdeposited and strained Co/Ag multilayers on electrical and magnetic anisotropy properties.

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