Magnetoresistance and Magnetic Anisotropy Properties of Strain-Induced Co/Ag Multilayer Films

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We report on the magnetoresistance (MR) properties of [Co(tCo)/Ag 1.5 nm]$_{50}$ multilayer and alloy films grown with the pulse electrochemical deposition on a polyamide substrate (1 cm$^2$). The induced uniaxial magnetic anisotropy was observed due to the effect of strain in all the multilayer films. The multilayer [Co 1.5 nm/Ag 1.5 nm]$_{50}$ showed a minimum hysteresis loss. The maximum MR ratio for Co/Ag was 9.2% at 1 kOe. A remarkable difference of magnetic field dependence of the magnetoresistance ratio was observed, corresponding to the orientation of magnetization curves.

Index Terms—Anisotropy, Co/Ag, magnetoresistance, multilayer, pulse electrodeposition.

I. INTRODUCTION

RECENTLY, reviews on the magnetic and electrical properties of nano-ordered materials created by the special means have been reported. Research on the metallic multilayer and alloy films is a typical example that has attracted much attention in view of the fundamental science and applications [1], [2]. Recent studies on giant magnetoresistance in the thin films are based on films grown mainly from the vapor phase. The phenomena exhibit different properties depending on the growth condition [3]–[7]. However, the relationship between the growth conditions and the properties of the film is still unclear.

Electro-deposition is a convenient method for producing alloys and multilayer by controlling the film composition and thicknesses on an atomic scale by regulating the pulse amplitude and width [8], [9]. The electrolyte metal growth differs from that of the vapor phase principally due to the presence of metal-solution double layer. One of the possible factors expected to cause different properties of the electrodedeposited film is due to the existence of the electric field in the order of $10^6$ V/cm between the electrode and ions in the double layer. Another is the charged nature of the particles arriving at the surface during the growth process. Therefore, it provides the possibility of depositing film structures different from those being produced from the vapor phase. We have attempted to grow many Co/Ag multilayered films, systematically regulating the electrode potential wave (pulse amplitude), current density, and the deposition time (pulse width). In multilayers, 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.

We have attempted to grow multilayer and granular alloy films by a micro-computer with a controlled pulse generator, and induce magnetic anisotropy in the ferromagnetic layers by applying strain externally. We report the results of our investigation of the magnetic and electrical properties of the as-deposited and strained ferromagnetic cobalt layers in the Co/Ag multilayer films.

II. EXPERIMENTAL PROCEDURE

The electrolytic bath was prepared with 99.9% pure chemicals in doubly-distilled water, and the pH was adjusted to 3.0. The bath used to deposit the Co/Ag multilayer film consisted of CoSO$_4$·7H$_2$O, AgI and KI with a silver anode [9]. In the plating bath, the cobalt concentration was changed while keeping the silver concentration constant, whereas the ratio between cobalt and silver was varied. We used glass as a substrate in our earlier work. The present work differs from our past work in that the substrates consisted of 15-nm thin copper layer vapor deposited on a polyamide film.

A multilayer film was grown by means of pulse electrochemical deposition from the specified solution. A single electrolyte containing the salts of the two components (metals) of the multilayer film, i.e., a salt of cobalt and silver was employed to deposit the films by controlling the pulse wave output from the microprocessor. The pulse current density was switched from 0.1 to 25 mA/cm$^2$. The deposition time was increased from 1 ms and varied depending on the individual material and layer thickness desired. The layer thickness and composition were determined experimentally by employing several methods, including microbalance, chemical methods, X-ray diffraction, flame emission spectroscopy, and energy dispersive X-ray analysis (EDAX). On the basis of an average taken from ten samples, the precision range of these multilayer thicknesses were within 3%. These samples were subjected to external fields. We found the composition of the single ferromagnetic layer to be 92 at% Co 8 at% Ag. Magnetic anisotropy was deduced using a vibrating sample magnetometer (VSM). The easy axis of magnetization was along the perpendicular direction (in-plane) 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. The details of these measurement methods can be found in our previous work [10]. The magnetic field dependence of the MR ratio was examined by varying the relative direction between the H field and the current. The MR ratio was defined as $\text{MR} \% = \left[ R_H - R_O \right] \times 100/R_O$, where $R_H$ is the resistance at the magnetic field, $H$, and $R_O$ is the resistance at the zero applied field. Measurements were performed at room temperature with the magnetic field in plane of the sample.

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multilayer films showed a minimum hysteresis loss, i.e., it

Ag15/Co15 multilayer films. In the weak magnetic field (H = 21 kOe), the difference for the magnetic field dependence of MR is clearly observed with depending on the orientation of magnetization, that is, it is corresponding to the shape of the magnetization curves. The field dependence of MR does not significantly depend on the direction of measuring current, but it only has a tendency to depend on the direction of the applied field.

III. RESULTS AND DISCUSSIONS

Magnetic anisotropy is most commonly determined from magnetization measurements along the two orthogonal directions of the magnetic field relative to the direction of strain. Examples of such measurements, with the field parallel (in-plane) and perpendicular (in-plane) to the direction of strain using VSM, are shown in Figs. 1(a) and (b).

Fig. 1(a) shows the magnetic field dependence of the magnetization curves for the as-deposited film. The measured results of the magnetization illustrate that the magnetism of the film is magnetically isotropic. Magnetization curves were measured parallel (hard axis) and perpendicular (easy axis) 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 because of the effect of strain in all the multilayer films. The multilayer [Co15/Ag15]20 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.

The MR ratio as a function of tCo, reported elsewhere [8], [11] suggested that adequate thicknesses of cobalt and silver layers were required to create the high sensitivity and the large MR ratio. The measured value of change of resistance (ΔR = RH − RO), i.e., the difference between the resistance values indicated that the change in resistance seems to be caused by the spin-dependent scattering at the interface of cobalt and silver layers due to the orientation of magnetic moment above the silver layer thickness of 1.5 nm. Field dependent curves have been found to be very sensitive to both the cobalt layer thickness (tCo) and silver layer thickness (tAg). Fig. 2 shows the magnetic field dependence of MR ratio and corresponding magnetization curves for the randomly oriented and uniaxially oriented [Co15/Ag15]20 multilayer films. In the weak magnetic field (<1 kOe), the difference for the magnetic field dependence of MR is clearly observed with depending on the orientation of magnetization, that is, it is corresponding to the shape of the magnetization curves. The field dependence of MR does not significantly depend on the direction of measuring current, but it only has a tendency to depend on the direction of the applied field.

Fig. 3 shows the Co layer thickness dependence of the MR ratio for the Ag thickness of 15 Å: (a) randomly oriented film (b) field/hard axis, and (c) field/easy axis at an external field of 21 kOe.

a) As the ferromagnetic layer adjacent to the non-magnetic layer becomes to be discontinuous for the multilayer having thin Co layer, the MR ratio increases due to the increase in the region of antiparallel alignment.

b) On the other hand, when the ferromagnetic layer adjacent to the non-magnetic layer becomes to be continuous, the MR ratio decreases due to the increase in the ferromagnetic region not showing antiparallel alignment of the spin.
The Ag layer thickness of 1.5 nm showing the maximum MR ratio is different from that of sputtered or MBE grown films [3], [5]. We have attributed this to the composition of the ferromagnetic layer in the multilayer films. The ferromagnetic layer produced by the pulse electrodeposition is not 100% Co. It contains certain amount of Ag atoms. It was earlier observed that the peak value of MR appears at a lower Ag layer thickness when increasing the Ag concentration in the ferromagnetic films [12]. Compared with the MR ratio (19–21% at 50 kOe) values for the sputter deposited films, our electrodeposited films show smaller MR ratio (at 21 kOe). However, it is expected that the films deposited at higher current densities will produce smaller ferromagnetic particles [12]–[14] in the ferromagnetic layers and show higher MR ratio at higher magnetic fields.

Fig. 4 shows $K_u$ as a function of $t_{Co}$ for a constant silver layer thickness of 1.5 nm and a cobalt layer thickness varied in the range of 0.5 to 3.0 nm, at room temperature with the $H$ field present. The reason for showing the lower anisotropy constant at the thinner Co layers at room temperature can be attributed to the discontinuous Co layers and crystal structures. The as-deposited films were magnetically isotropic and the uniaxial anisotropy was induced by introducing strain. An anisotropy constant was determined first by evaluating susceptibility, using Stoner-Wohlfarth (S-W) model [15] assuming that the magnetization process of Co/Ag proceeds via a coherent rotation of magnetization at the field above 0.1 kOe. In Stoner-Wohlfarth (S-W) model, the susceptibility, $\chi_{H=0}$, for a polycrystalline film is given by

$$\chi_{H=0} = M_s \frac{\sin 2\theta_O}{H_k}$$

(1)

where $\theta_O$ is the angle between induced uniaxial axis and the external applied field, $M_s$ is the saturation magnetization. The anisotropic field is therefore expressed as

$$H_k = M_s \times \frac{\sin 2\theta_O}{\chi_{H=0}}$$

(2)

When the angular distribution of domain magnetization is isotropic, $\sin 2\theta_O = 2/3$ and $H_k$ can be evaluated by using experimental values of $M_s$ and $\chi_{H=0}$ obtained by the slope of $H_C$. The increase of the MR ratio owing to the increase in anisotropy constant, $K_u$, is calculated by using the following:

$$K_u = M_s \frac{H_k}{2}$$

(3)

The correlation of the MR ratio and $K_u$ for (a) $H$ parallel to the magnetic hard direction and (b) $H$ parallel to the magnetic easy direction was studied. Fig. 5 shows the magnetoresistance effect plotted as a function of $K_u$ for the [Co 1.5 nm/Ag 1.5 nm]$_{20}$ film for the magnetic field parallel to the hard axis is about 9% and larger than that of the magnetic field parallel to the easy axis. The overall MR ratio with $H$ parallel to the easy axis (perpendicular to the strain) is less significant than the field parallel to the magnetic hard axis (parallel to the direction of strain) for all the samples. The reason for showing the smaller MR ratio in the anisotropic sample seems to be due to the following: The number of anti-parallel alignments of the magnetic spin between the ferromagnetic layers adjacent to the 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 difference in the slopes of the MR ratio. The result suggests that magnetoresistance decreases with increasing anisotropic constant. These Co/Ag multilayer films with the remarkable anisotropy properties are considered useful in the application as a magnetic sensor.

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