

Effect of Strain on the Magnetotransport Properties of Co/Ag Multilayer Films Grown by Pulse Electrochemical Deposition

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We report on the magnetoresistance (MR) effect of nano-order scale [Co(t_{Co}) / Ag 1.5 nm]₂₀ ferromagnetic multilayer and alloy films grown with pulse electrochemical deposition on top of a thin conducting layer of copper (15 nm). The copper buffer layer was grown on a polyamide substrate (1 cm²) and annealed. A strain was applied mechanically to study the electrical and magnetic characteristics as a function of the ferromagnetic layer thickness. 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]₂₀ showed a minimum hysteresis loss. The MR ratio was -9% at 1 kOe. A remarkable difference of magnetic field dependence of the 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, strain, magnetic anisotropy, pulse electrodeposition, Co/Ag multilayers

Introduction

Currently, studies on the magnetic and electrical properties of nano-ordered materials created by the special means are being reported actively. Research on metallic multilayer and alloy films is a typical example, which 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 is no concrete explanation for the relationship between the magnetism and the magnetoresistance in ferromagnetic layers. We have attempted to grow many Co/Ag multilayered films, systematically varying the layer thickness of cobalt and silver, by utilizing pulse electrochemical deposition. This method is used to grow thin films of granular alloy or multilayers of commonly immiscible metals exhibiting giant magneto-resistance (GMR) effects. 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.

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 a controlled pulse generator, and induce magnetic anisotropy in the lattice-mismatched ferromagnetic layer by applying strain externally.

The objective of this paper is to 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.

Experimental

The electrolytic bath used to deposit the Co/Ag multilayer film consisted of CoSO₄·7H₂O, AgI and KI with a silver anode, similar to the solution used in Ref. 3. The solution was prepared with 99.9% pure chemicals in doubly-distilled water, and the pH was adjusted to 3.0. In the plating bath, the cobalt concentration was changed while keeping the silver concentration constant, whereas the ratio between cobalt and silver was varied. The present work differs from our past work in that the substrates consisted of 15-nm thin copper films, vapor deposited on a polyamide film. Our earlier work used glass.

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 microprocessor. The pulse current density was switched from 0.01 to 2.5 mA/dm². The deposition time was increased from 1 msec 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. 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.⁴ The details of these measurement methods can be found in our previous work^{4,6} 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 $MR\% = [R_H - R_O] \times 100/R_O$, where R_H is the resistance when all the magnetic moments are aligned parallel and R_O is the resistance when the magnetic moments are anti-parallel in adjacent cobalt layers. Measurements were performed at room temperature with the magnetic field in plane of the sample.

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Results and discussions

Magnetic anisotropy is 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 (in-plane) and perpendicular (in-plane) to the direction of strain using VSM, are shown in Figs. 1a and b.

Figure 1a shows the magnetic field dependence of the magnetization curves for the as-deposited 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 parallel (hard axis) and perpendicular (easy axis) to the directions of strain at room temperature. Fig 1b 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 [Co 1.5 nm / Ag 1.5 nm]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.

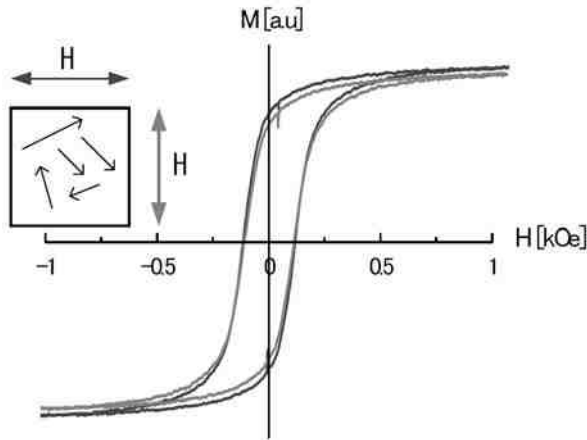


Figure 1a—M-H hysteresis loops for [Co 1.5 nm / Ag 1.5 nm]₂₀ multilayer films curves measured along to the parallel and perpendicular directions to the direction of current flow as-deposited film.

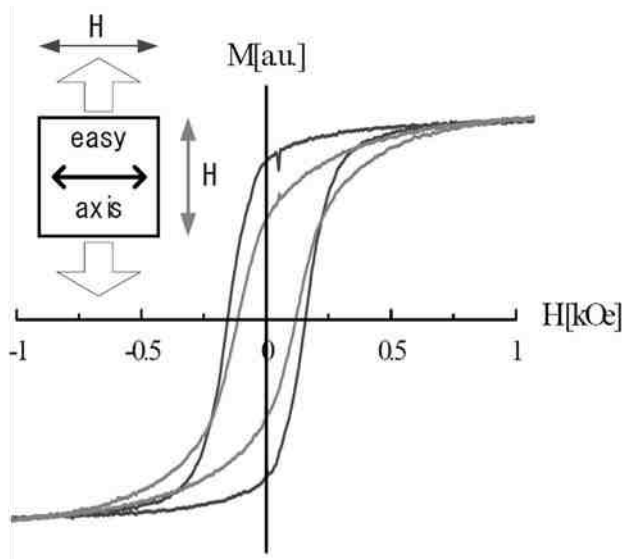


Figure 1b—M-H hysteresis loops for [Co 1.5 nm / Ag 1.5 nm]₂₀ multilayer films curves measured along to the parallel and perpendicular directions to the direction of current flow for the strain-induced films at ε = 1.5%.

Additionally, a torque magnetometer was designed to measure anisotropy. Figure 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 was derived as a function of the measurement angle. The torque *T*, which is a derivative of the anisotropy energy, corresponding to $E_A = K_u \sin^2 \theta$ is given by:

$$T = \frac{\delta E_A}{\delta \theta} = -K_u \sin 2\theta \quad (1)$$

This measurement confirms the anisotropic behavior of these multilayer films.

The MR ratio as a function of t_{Co} reported elsewhere^{6,7} 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 ($\Delta R = R_H - R_0$), *i.e.*, the difference between resistance during anti-parallel and parallel alignments of the magnetic moment of the adjacent cobalt layers indicated that the change in resistance value was 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. This result is in agreement with the results reported elsewhere.⁷

Field dependent curves have been found to be very sensitive to both the cobalt layer thickness (t_{Co}) and silver layer thickness (t_{Ag}). Figure 3 shows typical field dependent of magnetoresistance curves for [Co 3.0 nm / Ag 1.5 nm]₂₀. The maximum MR ratio for this film is 7.3%. The field dependence of the MR ratio for the oriented films with *H* parallel to the current is indicated by open circles. Similarly, the relation with *H* perpendicular to the current is 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. The negative field dependence tendency seems to be related to the GMR effect. It seems to be that the orientation of magnetization in the film has an effect on the GMR. For the anisotropic film, there is a difference between the values of field dependence measured by changing the direction of applied field against the measuring current.

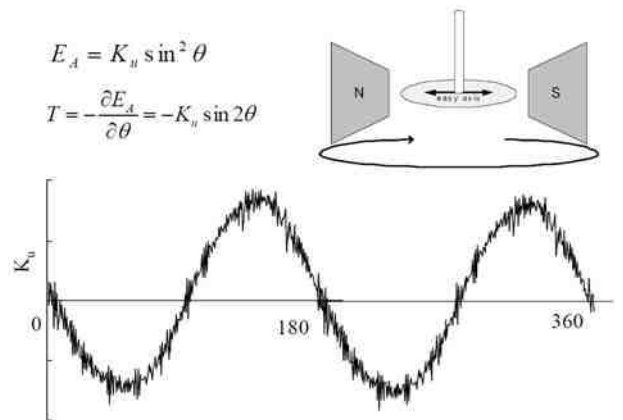


Figure 2—Torque measurement and anisotropic constant as a function of rotation obtained from the torque measurement to deduce magnetic anisotropy in [Co 1.5 nm / Ag 1.5 nm]₂₀ films.

Figure 4 shows the magnetization as a function of the field for [Co 1.5 nm / Ag 1.5 nm]₂₀ multilayers with the strain varied in the range of 0 to 1.5%. The 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⊥* of the magnetization curves were reported previously.⁴

Figure 5 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 to 3.0 nm, 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-Wohlfarth (S-W) model,⁸ 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, $\chi \frac{dM}{dH}$ at *H* = 0 for polycrystalline film, is given by:

$$\frac{M_s \sin 2\theta_0}{H_k} \tag{2}$$

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

$$H_k - M_s \times \frac{\sin 2\theta_0}{\chi_{H=0}} \tag{3}$$

When the angular distribution of domain magnetization is isotropic, $\sin 2\theta_0 = 2/3$ and *H_k* can be evaluated by using experimental values of *M_s* and $\chi_{H=0}$ obtained by a slope of *H_c*. The increase of the MR ratio owing to the increase in anisotropy, *K_u*, is calculated by using

$$K_u = \frac{M_s H_k}{2} \tag{4}$$

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. Figure 6 shows the magnetoresistance effect plotted as a function *K_u*. The MR ratio of [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 reason for showing the smaller MR ratio in the anisotropic sample seems to be 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 *K_u*. 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.

In conclusion, we have presented series of studies on the electrochemically grown as-deposited and strained Co/Ag multilayers and the effect of strain on electrical and magnetic anisotropy properties. These multilayer structures with remarkable anisotropy properties are useful in the application as a magnetic sensor.

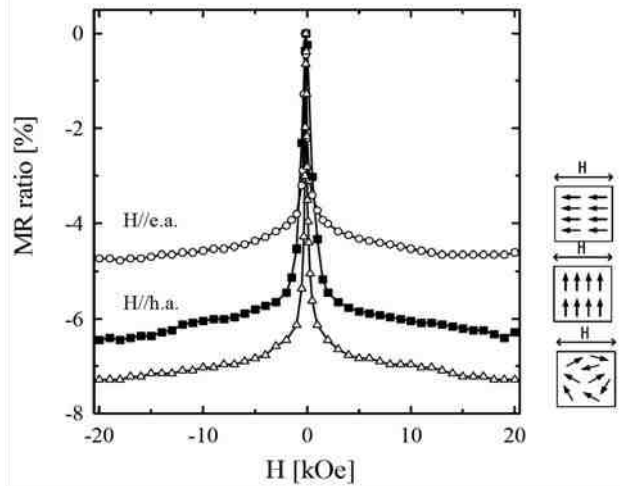


Figure 3—Field dependence of magnetoresistance ratio of [Co 3.0 nm / Ag 1.5 nm]₂₀ multilayer film at the range of 0 to 21 kOe; solid curves are drawn as a guide for the eyes.

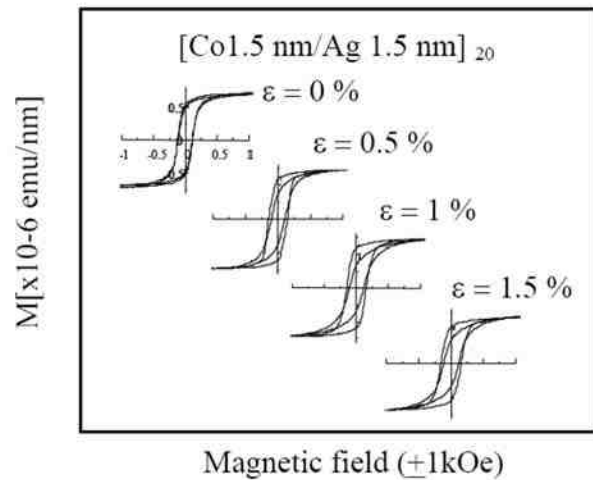


Figure 4—Field dependence of set of magnetization curves for the cobalt layer thickness after strain is impressed in the range of 0 to 1.5% at room temperature.

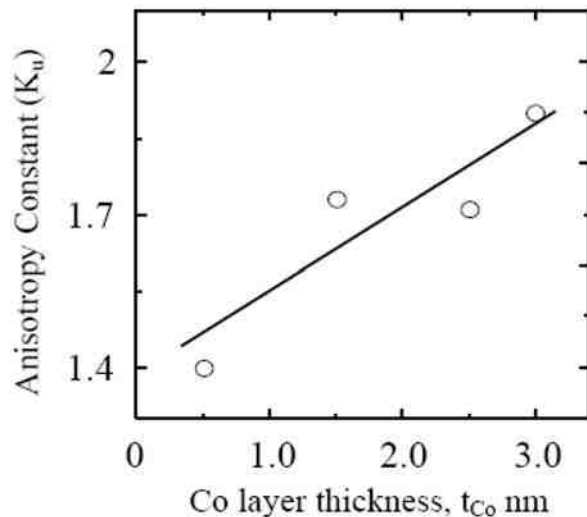


Figure 5—The relationship between anisotropy constant and cobalt layer thickness.

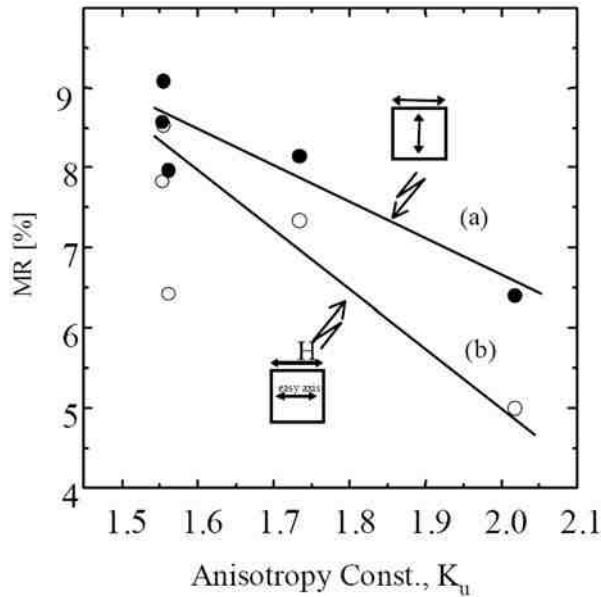


Figure 6—MR ratio against the anisotropy constant, K_u for (a) field parallel to the hard axis and (b) field parallel to the easy axis of the strain-induced [Co 1.5 nm / Ag 1.5 nm]₂₀ multilayer.

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About the Author



Dr. Chhabilal S. Rizal is a Graduate of Tribhuwan University, Kathmandu, Nepal. He is a Chartered Electrical Engineer (C.Eng.), in the U.K. His specialties are in the areas of nano-fabrication and the characterization of ferromagnetic and semiconductor multilayer and granular alloy films. He has to his credit six years of research experience in nano-electronics in Nepal, Qatar, Canada and Japan. He holds a Dr. Eng. degree from the National Institute of Japan.

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