AIP Journal of Applied Physics

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Citation: Journal of Applied Physics **92**, 4541 (2002); doi: 10.1063/1.1506400
View online: http://dx.doi.org/10.1063/1.1506400
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/92/8?ver=pdfcov
Published by the AIP Publishing

JOURNAL OF APPLIED PHYSICS VOLUME 92, NUMBER 8 15 OCTOBER 2002

Effect of growth temperature on the structure and magnetic properties of sputtered biepitaxial (111) permalloy films

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(Received 19 March 2002; accepted for publication 19 July 2002)

Electron backscatter diffraction (EBSD) patterns and coercivity values obtained from magnetization hysteresis loops of several sputtered biepitaxial (111)Py films grown at different temperatures on sputtered biepitaxial (111)Cu buffer layers have been collected. The results show that the coercive field of the Py films systematically increases as the growth temperature decreases, consistent with an increase in the degradation of the corresponding EBSD patterns due to enhancement of strain caused by misfit dislocations, threading defects, and point defects. The magnetization measurements also revealed no magnetocrystalline anisotropy for any of biepitaxial Py films studied. However, strain-induced uniaxial anisotropy was observed in all biepitaxial Py films. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1506400]

I. INTRODUCTION

In recent years there has been rapid progress toward the development of a generation of magnetic devices such as magnetic head readers, magnetic position or velocity sensors, and magnetic random access memory. These magnetic devices typically consist of alternating ferromagnetic such as Co, FeNi [Permalloy™ (''Py'')] and nonmagnetic (such as Cu, Ag) layers. The operation of these devices is based on the large changes in electrical resistance (ΔR) of the magnetic multilayer upon experiencing an external magnetic field, referred to as giant magnetoresistance (GMR). To optimize the performance of such devices, the following are needed: large changes in resistance upon application of a small magnetic field (Oe), small field dependence hysteresis (small coercive field), and small saturation fields. GMR in these devices depends on structural details such as the thin-film growth direction, interfacial roughness and intermixing, and grain boundaries.[1,2] However, the exact effects of these structural details on a given system are not yet well understood. Consequently, it is necessary to further characterize the growth and film structure of these magnetic systems to explore the effect of multilayer structure on the magnetotransport properties.

Permalloy™ (Py) is an alloy of Ni and Fe (typically $Ni_{81}Fe_{19}$) with a fcc derivative $L1_2$ structure. Because, from a magnetic point of view, Py is a ''soft'' ferromagnetic alloy, with only a small external field needed to rotate its magnetic moment. Therefore, it is frequently used as the ferromagnetic material in the earlier mentioned magnetic devices. The magnetic and transport properties of polycrystalline Py thin films (as a single layer or insert layers in a spin valve) have been studied in detail.[2–4] Molecular beam epitaxy grown films have been studied by Schuhl *et al.*,[5] who investigated magnetic and transport properties of epitaxial (001)Py thin films grown on thin Fe/Pd superlattices, and by Huang *et al.*[6] who investigated epitaxial $Ni_{80}Fe_{20}(110)$ on MgO substrates. The structural and magnetic properties of sputtered epitaxial (100)Py thin films grown at room temperature on relaxed Cu/Si(100) has been investigated by Hashim *et al.*[7] However, no intensive studies have been done on the relationship between the film structure, magnetic properties, and growth temperatures of epitaxial (111)Py films.

This work describes the results of crystallographic and magnetic characterization of sputter-deposited (111)Py films grown on (111)Cu films at different substrate temperatures. Electron backscatter diffraction (EBSD) patterns reveal that both the Cu and Py films are biepitaxial, displaying ⟨111⟩ growth directions, but different {111} plane stacking. Magnetization hysteresis loops have been collected from the Py films grown at different temperatures. The results of these measurements have been used to establish correlations between the crystallographic and magnetic properties of epitaxial sputtered Py films.

II. EXPERIMENTAL DETAILS

250-nm-thick epitaxial Nb film were first deposited at a substrate temperature of 750 °C on $(11\bar{2}0)$ sapphire substrates by sputter deposition.[8] This was followed by deposition of a 20-nm-thick Cu film grown at a substrate temperature of 350 °C. Finally, 200 nm films of Py were deposited at substrate temperatures of 10, 100, and 350 °C onto the Cu layer. Because of relatively small lattice mismatch (1.3%) between Cu and Py, it is expected that these should grow epitaxial on each other. In the growth processes, all of the Py films had identically processed underlayers of Nb and Cu. All films and multilayers were grown in 2.5×10^{-3} Torr Ar,

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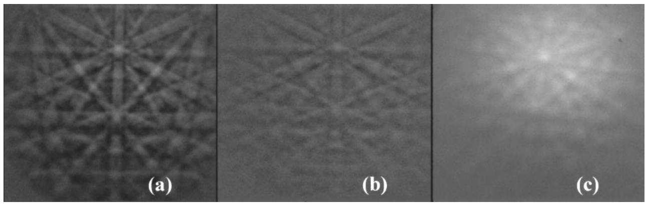

FIG. 1. EBSD patterns obtained from 200 nm epitaxial Py films with fcc crystal structures grown on epitaxial Cu (grown at 350 °C). (a) at a substrate temperature of 350 °C a pattern from single variant. At lower substrate temperatures of 100 °C (b), and 10 °C (c) overlapping pattern are observed. Significant pattern degradation is also noted with decreasing temperature.

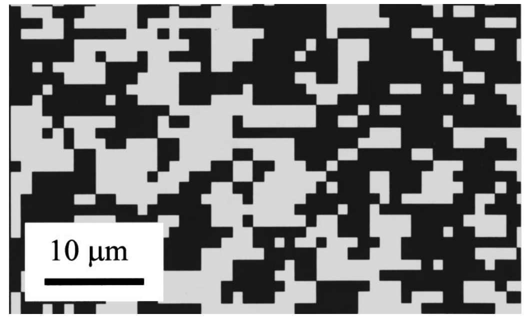

FIG. 2. EBSD orientation distribution map of a 200 nm epitaxial Py film grown at a substrate temperature of 350 °C on a 20 nm epitaxial Cu film. The two Cu twin variants are shown as gray and black areas.

with a chamber base pressure of $\sim 1 \times 10^{-8}$ Torr. The deposition rate was 3 Å/s for Nb and Py; 4 Å/s for Cu. The details of the sputter deposition growth of the epitaxial Nb and the subsequent Cu and Py films are given elsewhere.[8,9]

The coercivity and magnetic anisotropy of the epitaxial Py films were studied by measuring the in-plane magnetization of these Py films in both $[110]$ and $[1\bar{1}\bar{2}]$ directions (90° to each other). Additional magnetization measurements were carried out by continually rotating the sample (in-plane) at zero applied magnetic field. All of the magnetization measurements were carried out at room temperature (300 K) and 12 K in an MPMS Quantum Design magnetometer. At these temperatures the Nb buffer layer is not superconducting and does not affect the magnetization measurements.

EBSD is a scanning electron microscope (SEM) diffraction technique for measuring crystallographic orientations of single-crystal and/or polycrystalline materials. EBSD patterns consist of many intersecting linear features called Kikuchi bands. These bands have slightly higher and lower intensities with respect to a nonuniform background. This contrast is due to the diffraction of backscattered electrons by the crystal.[10] The detection and indexing of these Kikuchi bands determines the crystal orientations, and the data can be displayed in the form of pole figures and inverse pole figures. EBSDs have been used here to examine the orientation and orientation distribution of epitaxial Py films, as outlined below. The EBSD patterns were collected in a CamScan 44FE field emission SEM operated at 25 kV and equipped with a CamScan ORTEX change coupled device. The EBSD patterns were analyzed using the Channel4™ software package that determines crystal orientations and displays these data in the form of pole figures.[11]

III. RESULTS AND DISCUSSION

A. EBSD

The EBSD technique was used to examine the crystallographic nature of the sputtered Py films by collecting data randomly from different locations on the Py films. Figure 1 shows representative EBSD patterns from the three 200-nm-thick epitaxial Py films grown on 20 nm epitaxial Cu at substrate temperatures of 350, 100, and 10 °C. The resulting pole figures obtained from the EBSD data of the 200-nm-thick Py films grown at 350 °C revealed that two different orientation variants were present with the two orientations distinguished by a change in the Kikuchi bands in the patterns.[8] Figure 1(a) shows a representative EBSD pattern from one of the variants. Both variants display $\langle 111 \rangle$ growth normals (two in-plane twin variants), consistent with the growth occurring on the close-packed $\{111\}$ planes in the fcc-derivative Py. As the beam was moved across the Py film sputtered at 350 °C, the pattern was found to change between these two distinct orientations corresponding to two different stacking sequences of $\{111\}$ fcc planes. These results indicate that the Py is actually a biepitaxial film. In Fig. 1(b), and 1(c) the EBSD patterns of the 200 nm Py films grown at 100 and 10 °C are shown. These display two distinct features: (1) degradation of the patterns in comparison to those from the film grown at 350 °C and (2) overlapping of the two variant patterns. The substantial pattern degradation noted for the samples grown at lower substrate temperatures is most likely due to defects. The degradation results from the local elastic strain associated with point defects, dislocations, and planar defects distorting the diffracting planes.[12] It was also observed that unlike the sample grown at 350 °C, the patterns did not change as the electron beam was moved across the samples grown at low temperatures. In all cases, the patterns for samples grown at 100 and 10 °C were found to be a mixture of the two patterns observed in the film grown at 350 °C. This indicates grain sizes in the samples grown at 100 and 10 °C are much smaller (smaller than the effective EBSD interaction volume of approximately 0.8–1 μm at 25 kV) than in the sample grown at 350 °C.

To measure the size and distribution of the variants in the biepitaxial Py films grown at 350 °C, the samples were mapped using stage control scanning. An orientation distribution map of a 200-nm-thick Py film grown on a 20-nm-thick epitaxial Cu film with a substrate temperature of 350 °C is shown in Fig. 2. Orientation distribution maps revealed that the area of the twin variants varies widely and does not display any particular pattern. The average grain size of the twin variants in the biepitaxial Py film grown at 350 °C, as measured by the linear intercept method, was approximately 5 μm, with fractional distribution of the variants varying between 44% and 56%. Note that the underlayer Cu films also have grown biepitaxial with two (111) twin variants.[8] However, structural characterization of similar samples using high-resolution transmission electron micro-

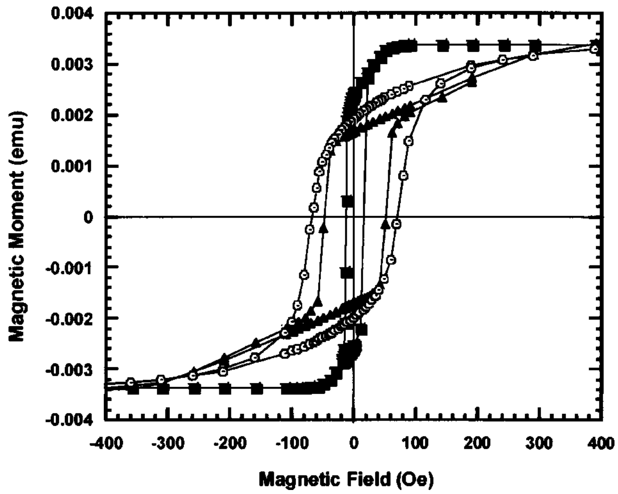

FIG. 3. Magnetization hysteresis loops of 200-nm-thick epitaxial (111)Py films grown on 20 nm thick epitaxial (111)Cu at substrate temperatures of 10 (○), 100 (▲), and 350 °C (■). The coercivity (H_c= half width of magnetization loop) increases from 12 to 75 Oe as the temperature decreases from 350 to 10 °C. All curves show absolute values of the magnetization, with all sample having the same nominal dimension of 5 mm×5 mm ×200 nm.

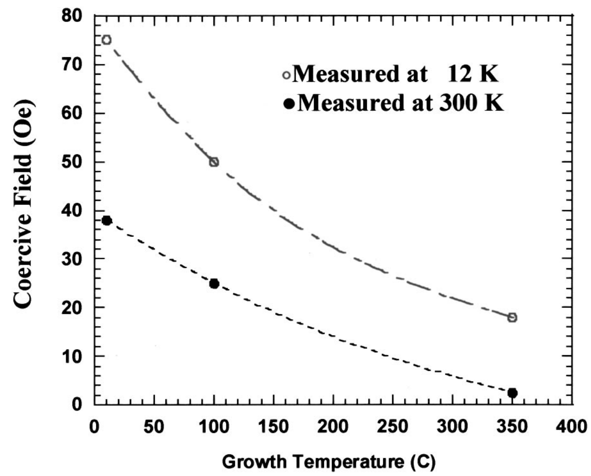

FIG. 4. Variation of coercive field as a function of growth temperatures. (○) Coercive field values collected from magnetization hysteresis loops of epitaxial (111)Py films when measured at 12 K, and (●) when measured at 300K.

scope indicates that the growth of twin variants of Py films do not necessary follow these of underlayer Cu.[13]

B. Magnetization measurements

1. Hysteresis loops

The magnetization hysteresis loops (measured at 12 K) of three sputter deposited biepitaxial Py films (same samples as for EBSD) grown at 350, 100, and 10 °C are shown in Fig. 3. The coercive field, H_c, (half the width of the hysteresis loop at zero magnetization) of the biepitaxial Py film grown at 350 °C is small (approximately 18 Oe), while the H_c of the samples grown at 100 and 10 °C are much larger, about 50 and 75 Oe, respectively. The values of H_c from measurements at room temperature were about 2.5, 25, and 38 Oe, respectively. The variation of H_c with growth temperature for measurement at 12 K and room temperature is shown in Fig. 4. The difference between coercive field values of individual samples measured at room temperature and 12 K is most likely related to elastic strain produced in the samples at low temperature. However, the variation of H_c for samples grown at different substrate temperatures may be related to the decrease in crystalline quality of the biepitaxial Py films as the growth temperature decreases. Following measurement at room temperature, then 12 K, the measurements were repeated again at room temperature. The values of the H_c were fully recovered, indicating no effect of thermal history on H_c, but instead showing that the changes in H_c at room temperature are a direct consequent of the sputtering temperature.

2. Magnetic anisotropy

The magnetic anisotropy (in particular, magnetocrystalline anisotropy) of these biepitaxial Py films was measured by comparing the magnetization of each film for two in-plane orientations. The difference in magnetic properties in different directions in magnetic materials is called magnetic anisotropy, while magnetocrystalline anisotropy is the more specific tendency of the spins in the ferromagnetic materials to align in a particular direction imposed by the crystal lattice. In epitaxial metallic films with a cubic structure and (111) growth direction, threefold symmetry of ⟨110⟩ and ⟨112⟩ directions occurs in the (111) plane. In cubic magnetic materials, there should be no in-plane magnetocrystalline anisotropy (first-order cubic anisotropy) in the (111) plane.[14,15] Therefore, the magnetic properties of a magnetic film with [111] growth direction should not be different if measured in the in-plane [110] and $[1\bar{1}2]$ (90° to each other) directions. The magnetization hysteresis loop of all of the biepitaxial Py films were measured in both [110] and $[1\bar{1}2]$ directions. A result of such measurements is shown in Fig. 5. The overlapping hysteresis loops of Py films grown at 350 °C reveal no magneto anisotropyfor any of these biepitaxial (111) Py

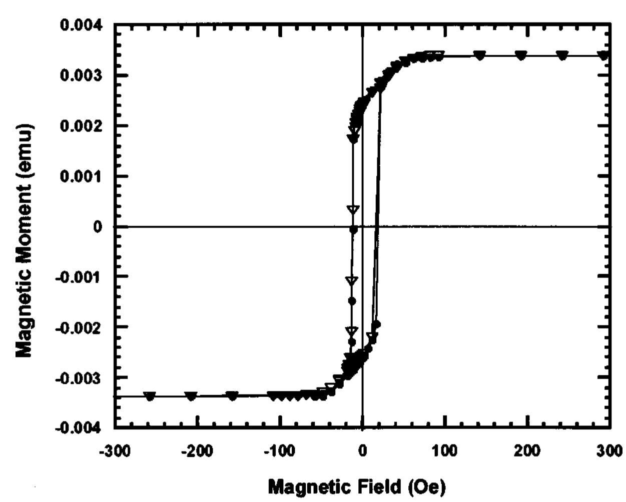

FIG. 5. Hysteresis loops measured with applied magnetic field in the [110] direction (▽) and the $[1\bar{1}2]$ direction (●). The hysteresis loops overlap indicating no magnetocrystalline anisotropy in this film. Similar behavior was found for all (111)Py films studied.

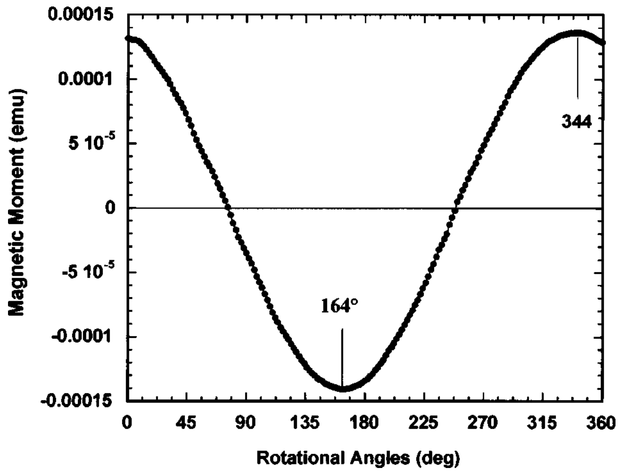

FIG. 6. In-plane angular dependence of magnetization at zero applied magnetic field of a biepitaxial (111)Py film when measured at room temperature. The maximum and minimums in the magnetic moments occur at 180° to one another.

films. Similarly, the overlapped magnetization hysteresis loops of all other biepitaxial Py films deposited onto biepitaxial Cu buffer layer at lower temperatures (100 and 10 °C), revealed no magnetoanisotropy as well. Rezende *et al.*[15] have investigated the in-plane anisotropy of epitaxial Fe(111) thin films, using the ferromagnetic resonance technique. This approach allows measurements of the second-order cubic anisotropy, strain-induced in-plane uniaxial anisotropy, and misorientations from the (111) plane. In the present study, we have used the superconducting quantum interference device magnetometer and performed full angular variation (0°–360°) magnetization measurements at zero applied magnetic field to study such higher order anisotropies. Figure 6 shows an example of one of these rotational magnetization measurements. Because the maximum and minimum magnetization occur 180° to each other, and because the magnitudes are the same for each of these first derivative zeros, it is clear that sample only display a strong strain-induced in-plane uniaxial anisotropy. Similar behavior was observed for all three of the sputter deposition temperatures. However, this approach may not be sensitive to detect subtle second-order effects.

IV. SUMMARY

The crystallographic characterization of sputter deposited biepitaxial Py films has been correlated to their magnetic properties.

(a) The results of EBSD characterization revealed that Py grew as biepitaxial fcc films on $Cu(111)_{fcc}/Nb(110)_{bcc}/Al_2O_3\ (11\bar{2}0)_{hcp}$. The crystallographic characterization of Py films grown at high temperature (⩾350 °C) revealed two in-plane twin variants corresponding to two different stacking sequences of {111} fcc planes. Orientation distribution maps revealed that the area of the twin variants varied widely and did not display any particular patterns. The crystallographic characterization results of Py samples grown at 100 and 10 °C revealed that two different orientation variants were also present, but in sizes smaller than the EBSD interaction volume of 0.8–1 μm. These films also displayed substantial EBDS pattern degradation, indicating significant strain in the films due to crystalline defects.

(b) Growth temperature is an important factor that can affect the magnetic properties of single magnetic Py films. The Py samples grown at 100 and 10 °C had much larger coercive fields than those grown at an elevated temperature (350 °C). This is likely due to the presence of the significant strains in the epitaxial Py films grown at low temperature. Regardless of the growth temperature, no magnetic anisotropy was observed in single epitaxial Py films; however, strain-induced uniaxial anisotropy was observed in all biepitaxial Py films at all sputter deposition temperatures.

ACKNOWLEDGMENTS

This work was supported in part by the MSU CFMR and the U.S. NSF under Grant Nos. DMR 98-20135 and MRSEC DMR 98-09688. The authors would like to thank Professor W. P. Pratt, Jr. for useful discussions.

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