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Permalloy thin films on MgO(001): Epitaxial growth and physical properties

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Permalloy thin films were deposited onto MgO(001) substrates by standard sputtering technique at growth temperatures ranging from 200 to 800 °C. Both reflection high-energy electron diffraction (RHEED) experiments and atomic force microscopy observations reveal that the substrate temperature for two-dimensional epitaxial growth should not exceed 350 °C. A series of permalloy films with thicknesses ranging from 3.5 to 100 nm was prepared at 300 °C. All films were grown as (001) single crystal with the same crystallographic orientations as the MgO(001) substrates. As evidenced by RHEED and x-ray diffraction, films with thicknesses larger than about 20 nm present a structural relaxation. The in-plane magnetization hysteresis loops obtained by longitudinal Kerr loop measurements exhibit a ⟨100⟩ fourfold cubic anisotropy and an enhancement of the magnetic softness with decreasing film thickness, both likely originating from the large interfacial strain configuration. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1520723]

I. INTRODUCTION

NiFe alloys are widely used in magnetic recording media and sensors industry. In particular, permalloy ($Ni_{80}Fe_{20}$) is well known due to its extreme magnetic properties of high permeability, low coercivity, and small magnetic anisotropy. Thin films of this alloy are thus attractive components for spin valve devices.[1–3] The interfacial exchange coupling between ferromagnetic and antiferromagnetic layers is a key element in the spin valve running, and this coupling is deeply correlated to the structural and morphological properties of the layer interface. In addition, epitaxial permalloy structures are essentially obtained with polycrystalline microstructure and a (111) texture,[4–6] or three-dimensional growth.[7,8] Therefore, both fundamental research and technological application call for an understanding and a control of the magnetic as well as the structural properties of single crystal low dimensional permalloy systems.[9]

In this article, we present a preliminary study of the structural and the magnetic properties of epitaxial permalloy thin films on MgO(001) substrates. The bulk permalloy single crystal has a fcc structure with a lattice parameter $a_{100}(Ni_{80}Fe_{20}) = 3.5507$ Å while $a_{100}(MgO) = 4.509$ Å. Assuming an isostructural growth, epitaxial permalloy films on MgO substrates are theoretically in tensile strain in the plane and in compression along the growth direction, with about 17% lattice mismatch. The aim of this work is to underscore a correlation between the strain configuration and the static magnetic properties of the strained films. Single crystal permalloy thin films were epitaxially grown by sputter deposition on MgO(001) substrates by tuning the growth temperature parameter. After optimization, the deposition of a series of different permalloy thicknesses leads to study the influence of the strain evolution on the magnetic behavior.

Structural characterization was carried out by *in situ* reflection high-energy electron diffraction (RHEED) and *ex situ* high-resolution x-ray diffractometry (HRXRD). X-ray reflectometry was also performed to determine the thicknesses and the roughnesses of the permalloy films and the substrate. Both x-ray experiments were carried out on a Seifert diffractometer using the Cu $K_{\alpha 1}$ radiation and a double monochromator. The surface topography was observed by atomic force microscopy (AFM) in tapping mode. Finally, the longitudinal magneto-optical Kerr effect (MOKE) was used at room temperature to determine the static magnetic properties of the permalloy films. No special precautions were taken to shield the Earth's magnetic field.

II. EPITAXIAL GROWTH

Permalloy thin films were deposited on MgO(001) single crystals by a dc magnetron sputter technique in an ultrahigh vacuum chamber. A low plasma pressure of 0.005 mbar was used with 50 sccm flowing argon. The target employed for deposition was made of a Ni(79%)Fe(18%)Cu(1%)Mo polycrystalline alloy and was precleaned by plasma etching. Previous annealing of the substrates was performed at 800 °C at

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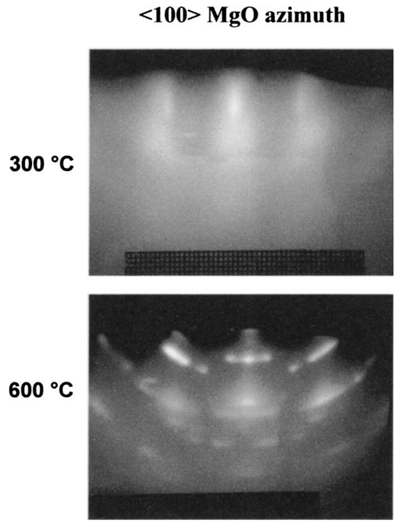

FIG. 1. RHEED images taken from 5 nm thick $Ni_{80}Fe_{20}$ MgO(001) layers (sample B) grown at temperatures of 300 and 600 °C. The RHEED beam was directed along the ⟨100⟩ MgO azimuth.

a base pressure lower than 5×10^{-8} mbar. Thin films (typically 5 nm thick) were prepared with a growth temperature ranging from 200 to 800 °C. The temperature effect on the film properties was investigated using complementary techniques.

RHEED patterns, as given in Fig. 1, reveal a transition of the film structure and morphology with increasing growth temperature from a single-crystalline character and two-dimensional (2D) morphology (vertical lines at 300 °C) to a polycrystalline character and three-dimensional (3D) morphology (rings and points at 600 °C). The evolution of the film morphology was confirmed by AFM measurements. AFM images are presented in Fig. 2 for growth temperature values of 300 and 600 °C. A large enhancement of the surface roughness is observed, with $R_a \approx 5$ Å for 300 °C while $R_a \approx 15$ Å for 600 °C, where R_a is the mean roughness. Finally, in-plane hysteresis loops were measured for the different growth temperatures, as displayed in Fig. 3. For 300 °C, the hysteresis loop exhibits a soft magnetic behavior with a coercive field lower than 0.5 Oe and a planar fourfold cubic anisotropy with a magnetization easy axis parallel to the ⟨100⟩ direction. For films deposited at higher temperature, the coercive field dramatically increases up to 50 Oe for 500 °C and the cubic anisotropy vanishes. Indeed, the structural and morphological disorder strongly affects the magnetization reversal and breaks down the presence of the planar fourfold anisotropy that results in the observed isotropic-like magnetic behavior with a much enhanced coercivity. The hysteresis loop shift on the magnetic axis is due to the Earth's magnetic field.

III. STRUCTURAL PROPERTIES

Films of thicknesses ranging from 3.5 to 100 nm were deposited at 300 °C (see Table I). RHEED patterns confirm that a two-dimensional epitaxial growth occurs along the [001] axis and shows that the MgO substrate and the permalloy films have the same cubic axis orientations. No qualitative change of the RHEED patterns was observed for varying the permalloy thickness, suggesting that the permalloy layer has a fcc structure with a (001) surface when grown on MgO(001). The epitaxial relationship between MgO and permalloy is maintained despite the large lattice mismatch value. Therefore, large epitaxial stress exists in the films.

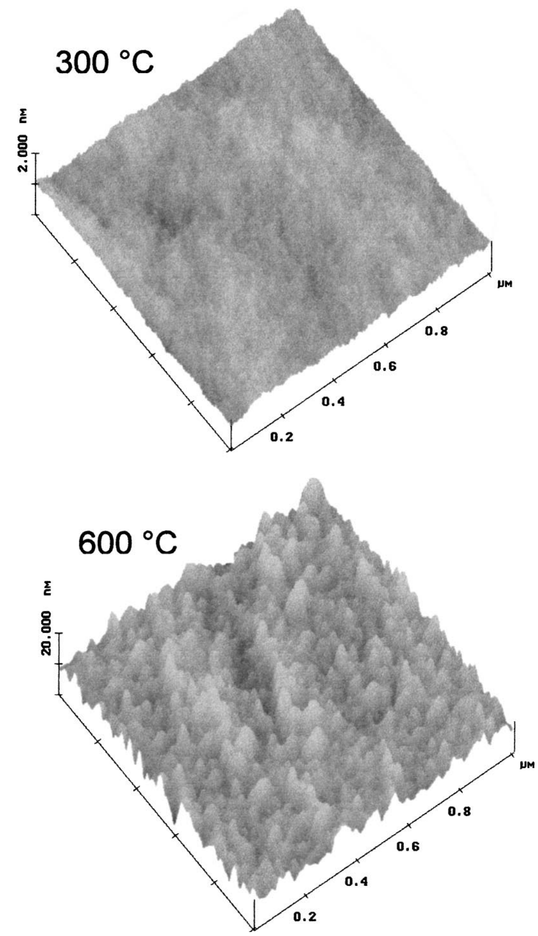

FIG. 2. AFM images of 5 nm thick $Ni_{80}Fe_{20}$/MgO(001) layers grown at temperatures of 300 and 600 °C.

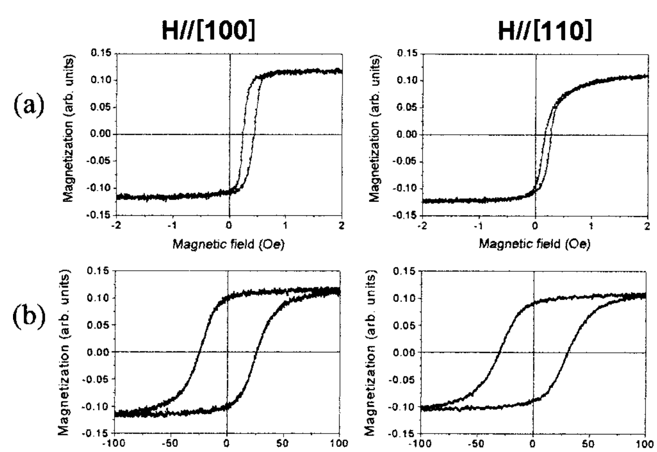

FIG. 3. Longitudinal MOKE loops for 5 nm thick $Ni_{80}Fe_{20}$/MgO (001) layers grown at temperatures of (a) 300 °C and (b) 500 °C. The magnetic field is applied along the [100] direction for the left-hand panel, and along the [110] direction for the right-hand panel.

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TABLE I. Sample notations.

Sample	A	B	C	D	E	F	G
Thickness (nm)	3.5	5	10	20	35	50	100

Simulations of the RHEED diagrams provide the structural evolution of the strained films versus the layer thickness. Figure 4(a) shows the fit of the in-plane surface parameter and Fig. 4(b) the crystalline coherence, defined by $a/\delta a$ where a is the lattice parameter and δa is the diffraction peak width. The in-plane lattice parameter decreases with increasing layer thickness while the crystalline coherence is enhanced. Comparing with the permalloy bulk value, this evolution shows a full structural relaxation of the films with a critical thickness about 20 – 30 nm.

The structural properties of the films were further investigated by HRXRD and reflectivity measurements. Classical $\theta-2\theta$ scanning modes were performed in order to determine the out-of-plane crystal parameter of the permalloy films by diffraction. Table II summarizes the obtained results for three samples. As expected, the out-of-plane crystal parameter increases with increasing layer thicknesses, which confirms the stress relaxation with increasing layer thickness. Reflectivity experiments confirm the accuracy of the layer thicknesses and reveal that sample G presents a much stronger roughness than samples C and F.

IV. MAGNETIC PROPERTIES

In-plane MOKE hysteresis loops observed for varying layer thickness are shown in Fig. 5. As already pointed out in Sec. I, the epitaxial films exhibit an in-plane fourfold cubic anisotropy with a magnetic easy axis parallel to the $\langle 100\rangle$ direction. Bulk permalloy exhibits a weak magnetocrystalline anisotropy with a magnetic easy axis parallel to the $\langle 111\rangle$ direction, given by fcc crystal structure. A $\langle 110\rangle$ in-plane cubic anisotropy is therefore expected in single crystalline permalloy. The easy axis switching is assumed to originate from the magnetoelastic contribution induced by the epitaxial strain, according to the large increase of magnetostriction observed in permalloy thin films.[10–12] Highly soft behavior is also observed with a coercive field lower than 0.25 Oe for thin films (<10 nm) and smaller than ~ 1 Oe in every case. No significant change of the hysteresis loops is observed as far the relaxation occurs, i.e., for films thicker than 30 nm.

TABLE II. Results of the reflectivity measurements ($\lambda_{xr}=1.5406$ Å: layer thickness, roughness $\sigma_{\text{substrate layer}}$ and $\sigma_{\text{layer vacuum}}$) and diffraction measurements (out-of-plane lattice parameter a_{001}). The simulation parameters are: mass per unit volume of MgO$=3.60\ \text{g/cm}^3$, mass per unit volume of permalloy films $=8.4\ \text{g/cm}^3$, and film composition $=Fe_{20}Ni_{80}$

Sample	Thickness (nm)	$\sigma_{\text{substrate layer}}$ (Å)	$\sigma_{\text{layer vacuum}}$ (Å)	a_{001} (Å)
C	9	2.5	7.0	3.529
F	48.8	3.0	7.0	3.5448
G	99.0	5.0	15.0	3.5474

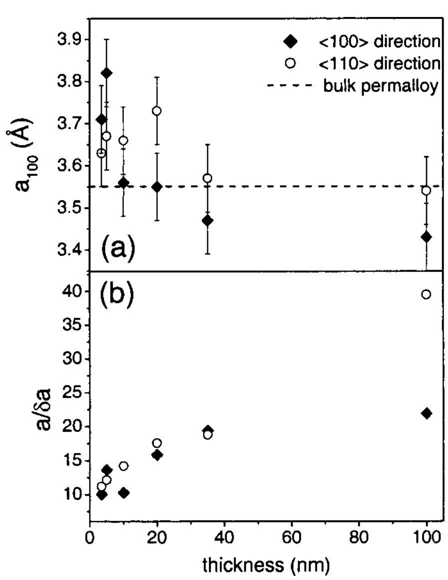

FIG. 4. (a) In-plane lattice parameter and (b) crystalline coherence vs the layer thickness.

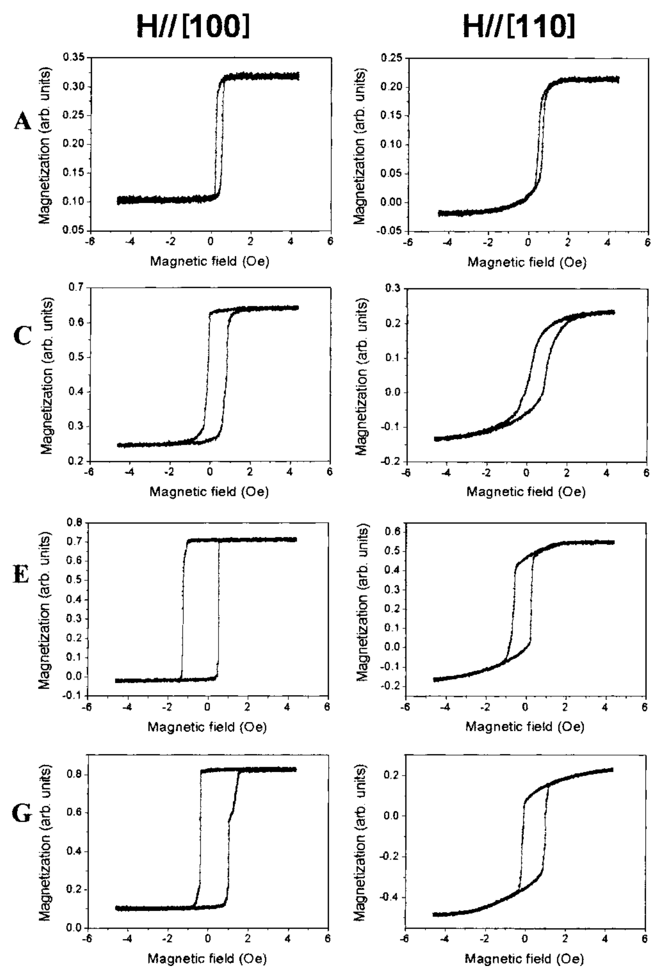

FIG. 5. Longitudinal MOKE loops for samples A (3.5 nm), C (10 nm), E (35 nm), and G (100 nm). The magnetic field is applied along the [100] direction for the left-hand side panel, and along the [110] direction for the right-hand side panel.

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V. CONCLUSION

We investigated the influence of the strain configuration on the magnetic properties of epitaxial permalloy thin films on MgO(001) substrates. The films were prepared by standard sputtering technique with control of the growth temperature. As the two-dimensional growth occurs in the low temperature regime ($<350\,°C$), the permalloy thin films were prepared at $300\,°C$. The permalloy films were grown as (001)-oriented single crystal films with the in-plane azimuth relation $Ni_{80}Fe_{20}[100]//MgO[100]$. Films thicker than about 20–30 nm show a full structural relaxation. The epitaxial strain is concluded to reduce the coercivity with a minimum value of 0.2 Oe at a thickness of 3.5 nm and to induce a $\langle 100 \rangle$ fourfold cubic anisotropy in the plane of the films.

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