Epitaxial growth and characterization of (100) and (110) permalloy films

J.C.A. Huang *, T.E. Wang, C.C. Yu, Y.M. Hu, P.B. Lee, M.S. Yang

Physics Department, National Cheng-Kung University, Tainan, Taiwan, ROC

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Abstract

High-quality, single-crystal fcc (100) and (110) permalloy films were epitaxially grown on MgO(100) and MgO(110) substrates, respectively, while polycrystalline structures were established on Si(111) substrates. The excellent crystal growth of the permalloy films on the MgO substrates was evidenced by fine streaks in reflection high energy electron diffraction and X-ray diffraction. Low-temperature magnetic hysteresis measurements show that the polycrystalline permalloy films grown on the silicon substrates are magnetically hard with coercive fields of ~ 15–20 Oe. On the other hand, (100) and (110) permalloy films are magnetically softer with coercive fields of about 1 and 6 Oe, respectively.

1. Introduction

Permalloy (Ni$_{1-x}$Fe$_x$, $x \approx 19\%$) is known as a material with high permeability, low coercivity and small magnetic anisotropy [1–4]. It has been studied for years from fundamental research as well as technological application points of view. Recently, permalloy related multilayers [5–9] and spin-valve heterostructures [10–12] attract considerable interest due to the discovery of the giant magnetoresistance effect with a low saturation field, which can be applied for advanced magnetic sensors and read-head. It has been noted that the crystal structure may play an important role upon the magnetic behavior. Most of the previous studies on permalloy films have been restricted to polycrystalline or textured structures. In this investigation, we study the epitaxial growth and characterization of single-crystal fcc (100) and (110) permalloy films on MgO(100) and MgO(110) substrates. For comparison, polycrystalline permalloys were prepared on silicon (111) substrates.

2. Experimental procedure

The crystal growth of permalloy films was carried out by vacuum product molecular beam epitaxy (MBE-930), as schematically shown in Fig. 1. The MBE system consists of three chambers: an introduction chamber, an analysis chamber and a growth chamber. Both the introduction chamber and the analysis chamber are connected to the growth chamber by a gate valve. This allows sample transfer under ultra-high vacuum (UHV) conditions for film deposition or surface characterization by reflection high-energy electron diffraction (RHEED), atomic force microscope (AFM) and scanning tunneling microscope (STM).
The permalloy (100) and (110) films were grown on epitaxial grade MgO (100) and MgO (110) substrates (purchased from Kyocera Co.), respectively. The miscut of the MgO (100) and (110) substrates are within 1.5°. To enable the growth of high-quality permalloy films, the MgO substrates were chemically precleaned in acetone and isopropyl alcohol for at least 15 min each in an ultrasonic bath. They were then introduced into the growth chamber and outgassed at \( \sim 1000^\circ C \) for 1 h under UHV conditions before initial deposition. Pure (99.99%) permalloy material (81% Ni + 19% Fe) was evaporated from an e-beam source. The growth pressure was below \( 5 \times 10^{-9} \) Torr, the deposition rates at about 0.1 Å/s, and the substrate temperatures at \( \sim 350^\circ C \) (0.35 \( T_m \)). The substrate temperature was measured by a W–Re thermocouple. The deposition rate and sample thickness were calibrated by a quartz crystal monitor located very close to the sample holder. To retain the sample uniformity the sample holder was rotated with a constant speed of about 30 rpm.

The surface structure and the epitaxial orientation of the permalloy films were determined in situ by RHEED. The energy of the RHEED beam is 20 keV, and the incidence angle \( \sim 1^\circ - 3^\circ \). The RHEED patterns were displayed on a phosphorus screen and recorded by a polaroid camera. The bulk structure of the permalloy films were measured by X-ray diffraction (XRD) using Cu Kα₁ radiation. Magnetic properties were investigated by using a commercial (Quantum Design made) superconducting quantum interference device (SQUID) magnetometer with a measuring temperature from 10 to 350 K.

3. Results and discussion

For permalloy (Py) films grown on MgO (100) and (110) substrates we have obtained the simple orientational relationships: Py(100)||MgO(100) with Py[010]|MgO[010] and Py[001]|MgO[001];
Py(110)∥MgO(110) with Py[001]∥MgO[001] and Py[110]∥MgO[110]. In spite of the large lattice mismatch, high-quality permalloy (Ni$_{81}$Fe$_{19}$) lattice spacing $a = 3.548$ Å) films can be established on MgO ($a_{\text{MgO}} = 4.2$ Å) substrates for a film thickness above 50 Å, as confirmed by RHEED and XRD studies. By XRD analysis of the permalloy films with thicknesses varying from 10 to 250 Å, we have observed that the permalloy films were almost strain-relieved for a thickness above $\sim 50$ Å [13]. Similar results were found by Hashim and Atwater using RHEED investigation [4].

Fig. 2a and Fig. 2b show the XRD spectra of permalloy films grown on MgO(110) and (110) substrates, respectively, indicating a good bulk structure of the films. On the other hand, polycrystalline permalloy films, mainly with (111), (200) and (220) domains, were formed for samples grown on the Si(111) substrates, as shown by the XRD spectrum in Fig. 2c. As calculated from the XRD spectra, the lattice spacing of the grown permalloy films were $\sim 3.546$ Å. Assuming that the lattice parameter of the observed permalloy films (Ni$_{1-x}$Fe$_x$) is equal to $a = (1-x)a_{\text{Ni}} + xa_{\text{Fe}}$ for a small variation of $x$ (around 19%), we have determined that the alloy composition was Ni$_{82}$Fe$_{18}$ ($x = 18\%$), very close to the source composition ($x = 19\%$). Here $a_{\text{Ni}}$ (3.5236 Å) and $a_{\text{Fe}}$ (3.6468 Å) denote the bulk lattice spacing of fcc Ni and fcc Fe.

Fig. 3a–Fig. 3d show typical RHEED patterns on the surfaces of the permalloy films (250 Å) showing: (a) Py(100) along [010], (b) Py(100) along [110], (c) Py(110) along [110] and (d) Py(110) along [111].
(250 Å). Note that the permalloy (100) surface possesses a 2 × 1 superstructure. For a RHEED beam directed along the [010] and [001] directions on the (100) surface, superstructure peaks (see Fig. 3a) were observed. Since the alloy composition (Ni$_{35}$Fe$_{18}$) is quiet close to that of Ni$_{75}$Fe$_{25}$ which holds an ordered L1$_2$ structure (see Fig. 4) below the critical temperature of ~ 500°C [14], the observed superstructure peak may correspond to superlattice satellites of an ordered NiFe (100) surface. For the same growth temperature (350°C), no superstructure peak was observed on the (110) permalloy films. Details of the surface (and bulk) structures and the order–disorder transformation of (100) and (110) Ni$_{1-x}$Fe$_x$ (x ≈ 18–25%) films are under intensive study.

Fig. 5a, Fig. 5b and Fig. 5c show the low-temperature (10 K) magnetic hysteresis loops of the permalloy films grown on MgO(100), MgO(110) and Si(111) substrates, respectively. Interestingly, (100) permalloy films are magnetically soft with coercive fields ($H_c$) of only ~ 1–2 Oe. The $H_c$ are about 5–6 Oe for the (110) permalloy films. The high-temperature (350 K) $H_c$ are less than 1 Oe for both (100) and (110) permalloy films. Permalloy films deposited on the silicon substrates are, on the other hand, magnetically harder with $H_c$ (10–350 K) of ~ 15–20 Oe. Indeed, for all our epitaxial polycrystalline permalloy films (Ni$_{1-x}$Fe$_x$, x ≈ 19%) grown on silicon substrates, we have systematically found that they possess an unusual high coercivity. Since nickel reacts easily with silicon, the observed large coercivity is possibly due to the formation of silicide (a much harder ferromagnet compared to permalloys) in the silicon–permalloy interface.

In summary, we have prepared single-crystal fcc (100), (110) and polycrystalline permalloy films on MgO(100), MgO(110) and Si(111) substrates, respectively. Excellent epitaxial growth of permalloy films on the MgO substrates was evidenced by fine peaks in RHEED and XRD. Magnetic hysteresis measurements show that permalloy films grown on MgO substrates are magnetically softer than samples on the silicon substrates. The magnetic anisotropic behavior and magnetoresistance of the permalloy films, multilayers and related spin–valve heterostructure are under study and will be published elsewhere.

(a)

\[ H_c=1.5 \text{ Oe} \]

(b)

\[ H_c=6 \text{ Oe} \]

(c)

\[ H_c=16 \text{ Oe} \]

Fig. 5. The magnetization as a function of applied magnetic field at 10 K for permalloy (Py) films (250 Å) showing (a) Py(100) on MgO(100), (b) Py(110) on MgO(110) and (c) polycrystalline Py on Si(111).
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References