Multilayer Permalloy films grown by molecular-beam epitaxy

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Single and multilayer [111]-textured films of Permalloy and Cu were grown by molecular-beam epitaxy (MBE) on (111) Si substrates. The magnetic properties of the films were measured by ferromagnetic resonance and M-H loop tracer. The microstructure was observed by transmission electron microscopy, reflection high-energy electron diffraction, and x-ray diffraction. Even the single-layer films had lower easy axis coercivity H_{ce} ($\approx$0.6 Oe) and a lower in-plane anisotropy field ($\approx$1.1 Oe) than permalloy films of similar thickness ($\approx$80 nm) deposited by sputtering. In these single-layer films, the grain size and H_{ce} both increased with improved pre-MBE cleaning of the Si substrate. The multilayers consisted of five identical thickness Permalloy layers separated by Cu interlayers. Multilayers with total magnetic thickness greater than 100 nm exhibited lower H_{ce} than equivalent single-layer films. $4\pi M_s$, measured by a combination of ferromagnetic resonance and M-H loop tracer in very thin (<5 nm) permalloy layers was found to drop off relative to the bulk value of 9.5 kG; a value of 5.0 kG was measured for a multilayer with 1-nm-thick Permalloy layers.

I. INTRODUCTION

Sputtered multilayer films of Permalloy separated by nonmagnetic interlayers are known to exhibit low coercivity.[1] The coercivity reduction has been associated with the magnetostatic coupling of domain walls between the individual magnetic layers.[2,3] The microstructure and the purity of films deposited by molecular-beam epitaxy (MBE) can be controlled more carefully; also the layers can be deposited more slowly. We anticipated, therefore, that improved soft magnetic properties could be achieved with MBE. In this paper we present results on the magnetic properties and microstructure of single-layer Permalloy and multilayer Permalloy/Cu films grown by MBE on single-crystal Cu and Si substrates. The magnetic properties of the films were measured by M-H loop tracer (MHL) and ferromagnetic resonance (FMR). Structural characterization was done by reflection high-energy electron diffraction (RHEED), x-ray diffraction (XRD), and transmission electron microscopy (TEM).

II. EXPERIMENT

A. Film preparation

The MBE system used for film deposition was a three chamber ultra-high-vacuum apparatus built by VG Scientific. The chamber pressure during deposition was about 10^{-9} Torr. The films were grown on Cu (111) and Si (111) substrates. Our standard pre-MBE treatment of the Si surface consisted of an HF clean intended to remove the native oxide layer. However, as described in the next section, we also investigated the effect of different Si surface treatments.

The Cu and the 83% Ni-17% Fe alloy sources were in electron-beam evaporators. The deposition rate was 0.1 nm/s. The substrate surface temperature during deposition was 25 °C. The substrates were rotated at 0.4 rev/s in the presence of a 13-Oe in-plane dc magnetic field.

The single-layer Permalloy films had thicknesses of 5–200 nm. Each Permalloy/Cu multilayer film consisted of five identical thickness Permalloy layers separated by four identical thickness Cu interlayers. The initial permalloy layer was deposited directly on the Si substrate; the final permalloy layer was covered by a 10-nm-thick Cu cap. Films were grown with 1–40-nm-thick Permalloy layers, and with Cu interlayers of 1–5 nm in thickness.

B. Film characterization

In-plane easy (H_{ce}) and hard (H_{ch}) axis coercivities of the films were measured at 10 Hz by MHL. The error in the measured coercivity values is 0.1 Oe. The growth-induced in-plane uniaxial anisotropy field H_{ku} was also measured by MHL from the initial slope of the hard axis hysteresis loop.

The anisotropy fields were measured by FMR with 9 and 33 GHz resonant cavity spectrometers. Resonance field positions could be determined to an accuracy of 1 Oe. If shape anisotropy was the only contribution to the perpendicular anisotropy, we infer a value for $4\pi M_s$. The $4\pi M_s$ values were confirmed by MHL.

The surface quality of the film and substrate was characterized by RHEED in the MBE apparatus. Film texture was investigated by XRD. Microstructural characterization was carried out with a Philips 420 analytical scanning transmission electron microscope operating at 120 kV. The following techniques were employed: bright and dark field TEM, selected area diffraction (SAD), and microdiffraction. The planar and cross-sectional electron transparent specimens were prepared using a Gatan ion mill.

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TABLE I. Grain size, measured by TEM, easy axis coercivity, and in-plane anisotropy field in 80-nm NiFe films grown on Si substrates with different pre-MBE surface treatments.

Pre-MBE substrate surface treatment	Grain size (nm)	Easy axis coercivity (Oe)	In-plane uniaxial anisotropy field (Oe)
Not cleaned	15	0.6	1.3
HF cleaned	25	1.0	1.1
HF cleaned + 800 °C heat treatment	30	5.4	10.4

III. RESULTS AND DISCUSSION

A. Single-layer Permalloy films

Initially Cu (111) was used as the substrate in order to grow single-crystal Permalloy and thus achieve low coercivity; the (111) orientation was expected to provide low in-plane anisotropy. By electron diffraction (TEM and RHEED) the Permalloy films appeared to be single-crystal fcc, with their (111) planes parallel to the (111) Cu substrate. However, dark field TEM and microdiffraction revealed that the films contained twin related regions (≈ 100 nm in size) separated by incoherent double positioning boundaries.[4] Contrary to expectations, the films had consistently high coercivity ($H_{ce} \approx 5$ Oe) and high uniaxial in-plane anisotropy ($H_{ku} \approx 4$ Oe), possibly due to the presence of these incoherent boundaries. Permalloy films grown on (111) Si substrates were found to be polycrystalline fcc with [111] texture perpendicular to the plane of the film. In Table I the easy axis coercivity values and the grain sizes for 80-nm single-layer films grown on various Si surfaces are presented.

The easy axis coercivity lay in the 0.6–6-Oe range. The films with the best soft magnetic properties have H_{ce} of 0.6 Oe, H_{ch} of 0.2 Oe, and H_{ku} of 1.1 Oe. The lowest coercivity and smallest grain size (≈ 15 nm) were seen in films grown on substrates that were not HF cleaned prior to deposition. The highest coercivity and largest grain size (≈ 30 nm) were seen in films grown on substrates that, in addition to the HF cleaning, were heat treated at 800 °C in the MBE apparatus. Substrates cleaned in this manner are assumed to have fewer nucleation sites; this can account for the larger grain size. Under appropriate circumstances, films with larger grains are predicted to have higher coercivity.[5] In Fig. 1 we present bright field TEM images and selected area diffraction (SAD) patterns of the first and third entries in Table I. Note the larger size and less random orientation of the grains in the film on the heat-treated substrate. Also note the stronger δ-Ni_2Si phase rings (first and fourth rings) in the film deposited on uncleaned Si.

The variation of coercivity with thickness of the single-layer films, grown on the standard HF-cleaned substrates, is shown in Fig. 2. (The multilayer coercivity data are also shown.) The coercivity of the single-layer films increases slightly with increasing film thickness, for thicknesses up to 100 nm.

The perpendicular anisotropy field is $\approx(-)9.5$ kOe from FMR; this agrees well with the bulk Permalloy $4\pi M_s$. We expect a minimal stress contribution to the an-

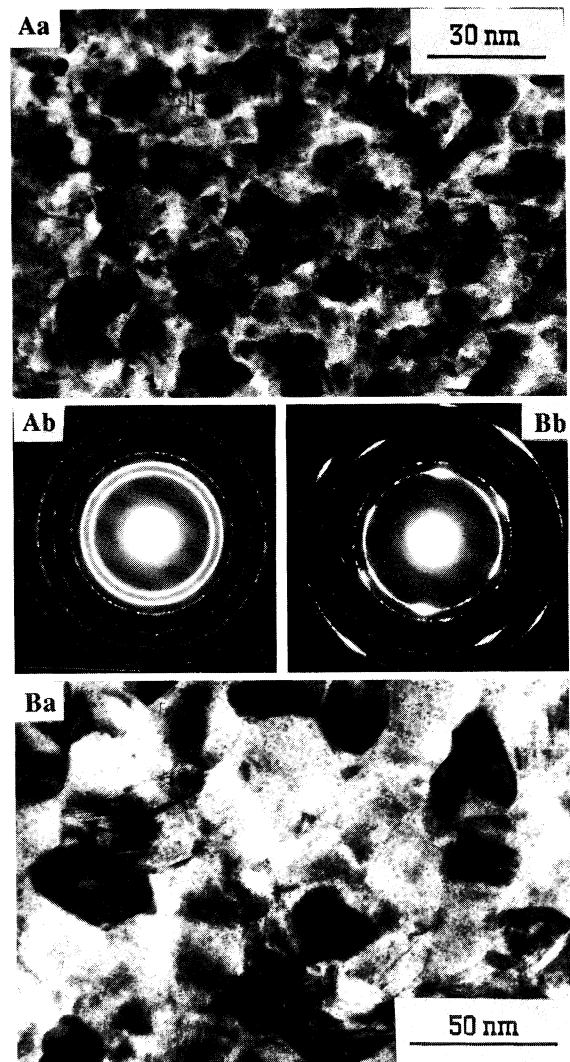

FIG. 1. TEM views and SAD patterns of single-layer 80-nm NiFe films. A: Uncleaned substrate: (a) bright field planar view and (b) SAD pattern. B: HF-cleaned and heat-treated substrate: (a) bright field planar view and (b) SAD pattern.

isotropy field, since the magnetostriction constant λ is ≈ 0 for these alloys. H_{ku} was not detectable by our FMR experiment, implying a value less than 4 Oe. The H_{ku} values measured by MHL lay in the 0.7–2.4-Oe range.

B. Multilayer Permalloy/Cu films

We also show in Fig. 2 data on the variation of H_{ce} with total magnetic thickness for multilayer films with various interlayer thicknesses. In the multilayer films, unlike the single-layer films, the coercivity decreases with increasing total magnetic thickness. The variation of H_{ce} with interlayer thickness is shown in Fig. 3 for total magnetic thicknesses of 10 nm (5×2 nm) and 100 nm (5×20 nm). In each multilayer series "zero" interlayer thickness refers to a single-layer film of thickness equal to the total magnetic thickness. In the case of the 5×20-nm multilayer films, the coercivity is decreased by multilayering. However, in the 5×2-nm multilayers the coercivity is increased.

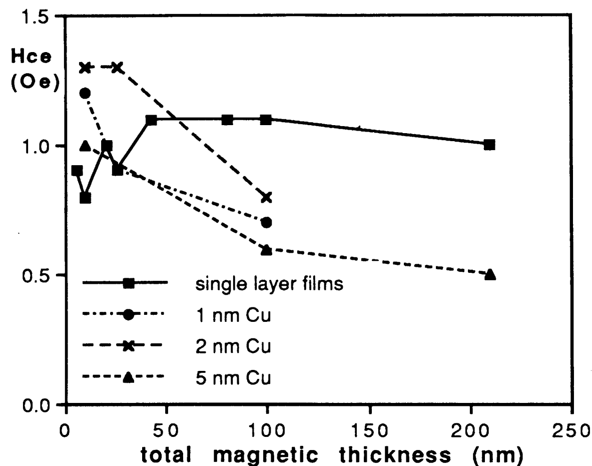

FIG. 2. Easy axis coercivity H_{ce}, vs the total magnetic thickness for single-layer and multilayer NiFe films.

We believe that this coercivity increase is due to inhomogeneous strain present in the very thin Permalloy layers. (Strain-induced anisotropy fields in the Oersted range would not be sensed by our FMR technique.) Sufficiently thick multilayers have good soft magnetic properties; for example a five-layer multilayer of 200 nm total magnetic thickness has H_{ce} of 0.5 Oe, H_{ch} of 0.4 Oe, and H_{ku} of 1.2 Oe.

In Fig. 4, $4\pi M_s$, inferred from FMR data, is plotted against the individual layer thickness; data from both single and multilayer films are included. The $4\pi M_s$ values of very thin layers (<5 nm) drop off relative to the bulk value. The same trend in M_s was found by MHL in these thin films.

From cross-sectional TEM on the 5×20-nm multilayer sequence we observed that, for 1-nm Cu interlayers, the columnar growth was fairly continuous throughout the composite film; however, as the Cu interlayer thickness increased, the continuity decreased. Consequently, in multilayer films fabricated with thicker Cu interlayers we anticipate a smaller grain size than in the single-layer films

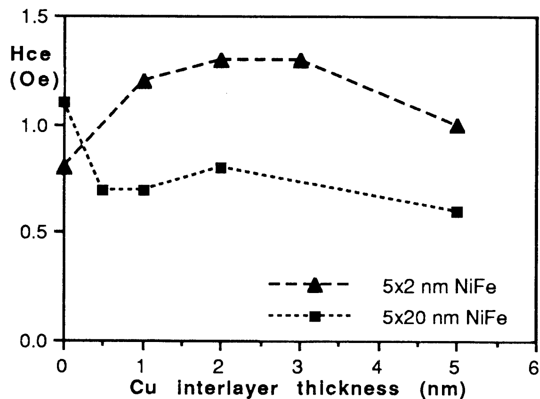

FIG. 3. Easy axis coercivity H_{ce}, vs the Cu interlayer thickness in multilayer films.

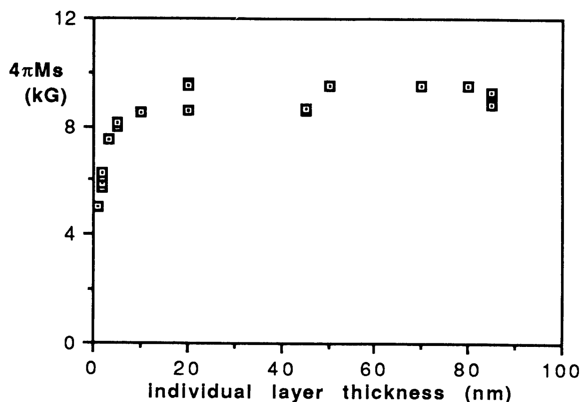

FIG. 4. $4\pi M_s$, measured by FMR vs the individual NiFe layer thickness in multilayer and single-layer films.

(of equivalent total magnetic thickness). For example, in a film consisting of two 40-nm Permalloy layers separated by a 5-nm Cu interlayer, the average grain size was 17 nm, lower than that (≈25 nm) in an 80-nm single-layer film grown on the same type of HF-cleaned substrate.

Hence, the magnetostatic domain-wall coupling and the smaller grain size are both expected to cause a lowering of H_{ce} in multilayers as compared to "equivalent" single-layer films.

IV. SUMMARY

MBE-grown single-crystal Permalloy on Cu substrates exhibits high coercivity and in-plane anisotropy; [111]-textured polycrystalline Permalloy on Si exhibits lower coercivity and anisotropy than typically achieved in randomly oriented polycrystalline films grown by sputtering. Our single-layer films show an increase in H_{ce} with increasing grain size. In the lowest grain size (≈15 nm) films we achieved $H_{ce} = 0.6$ Oe and $H_{ku} = 1.1$ Oe. Values reported for sputtered films are at least twice these. The improvement in soft magnetic properties may be due to the slow deposition rate used in MBE. Slow deposition is assumed to favor the observed [111] texture and to give lower stress, both of which should reduce the coercivity. In sufficiently thick multilayer films both H_{ce} and grain size are lower than in single-layer films of equivalent thickness. The smaller grain size and the magnetostatic domain-wall coupling are both expected to cause the decrease in H_{ce} seen in the multilayers. When the individual Permalloy layer thicknesses are too low, the multilayer coercivity increases relative to the single layer; this is probably due to strain-related effects. We find also that $4\pi M_s$ in these thin layers decreases from the bulk value.

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