JOURNAL OF MATERIALS SCIENCE 26 (1991) 4150–4154

# Magnetic properties of permalloy/permalloy-oxide multilayer thin films

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Magnetic properties of permalloy/permalloy-oxide multilayer thin films are investigated. These thin films are prepared by a repeat of sputter deposition of permalloy thin film, followed by oxidation of the film surface. The total thickness of the permalloy thin films before oxidation is about 100 nm. The number of layers is one to twenty. The oxide layers are formed by oxidation in dry air. The estimated oxide layer thickness is about 2 nm. The oxide $NiFe_2O_4$ is identified by RHEED. The film coercivity decreases linearly with increasing layer numbers. The saturation magnetization and magnetoresistivity decrease as the number of layers increase. The coercivity decrease is due to grain growth suppression and magnetic separation by oxide film of permalloy layer, and magnetoresistivity decrease is due to electrical resistivity increase originating into electron scattering by the oxide layer.

## 1. Introduction

Multilayered thin films are being investigated intensively to develop new materials and improve conventional ones. Magnetic thin films are one material in which multilayering effects are expected. Interface magnetic moment change [1], coercivity decrease [2], magnetostriction control [3], large magnetoresistivity [4], and other effects [5] are observed in multilayered magnetic thin films.

Permalloy thin films have been used as magnetic core materials for inductive magnetic recording heads and sensor films for magnetoresistive heads. To enable these films to be used for magnetic recording heads with higher recording density, their magnetic properties must be improved. The objective of this study is to obtain information on multilayering effects in permalloy very thin films separated by very thin surface oxide films. It is well known that oxide films grown on metal surfaces can destroy crystal lattice continuity between top and bottom thin films and is stable at relatively low temperature. Therefore, stable oxide films can be used as separation layers between deposited thin films.

This paper describes the multilayer microstructure, coercivity, saturation magnetization changes, electrical resistance, magnetoresistivity, and related properties of these films.

## 2. Experimental procedure

Multilayered thin films were prepared by repeated sputter-deposition of permalloy, followed by oxidation of the deposited permalloy thin film surface. The sputtering was performed using a conventional r.f. system. The permalloy targets were Ni-19 wt % Fe. The Ni + Fe purity was 99.9 wt %. The total thickness of the deposited permalloy thin films for the multilayered specimens was about 100 nm. Since the number of layers was one to twenty, the thicknesses of the permalloy layers were 5 to 100 nm. Here, a single-layer contains both permalloy and oxide layers. However, the true thickness of each permalloy layer after oxidation was less than that of the as-deposited film.

The oxidation was performed for 1 h in dried air at 22–24 °C. The dew point of air was below 77 K. The resulting oxide layer thickness was about 2 nm, estimated from a cross-sectional transmission electron micrograph, ellipsometry, and saturation magnetization decrease. The crystal structure of the oxide film was evaluated by reflection high energy electron diffraction (RHEED). The oxygen depth profiles were obtained from a secondary ion mass spectrometry (SIMS).

Coercivity was measured with a B-H looper and saturation magnetization with a vibrating sample magnetometer. Electrical and magnetoresistive properties were measured using the four-probe technique.

## 3. Results and discussion

### 3.1. Microstructure

A cross-sectional transmission electron micrograph of a 100 nm-thick as-sputtered permalloy single-layer thin film is shown in Fig. 1 a. On the single-layer thin film, very fine grains of 10 nm average diameter were observed near the glass substrate. These fine grains grew columnar. The average grain diameter measured parallel to the substrate plane increased with film thickness. The average grain diameter near the surface was 400 nm. A cross-sectional transmission electron micrograph of the six-layer permalloy/oxide multilayer thin film is shown in Fig. 1b, as an example. The arrow indicates the naturally grown oxide layer. The growth of permalloy grains was interrupted by the



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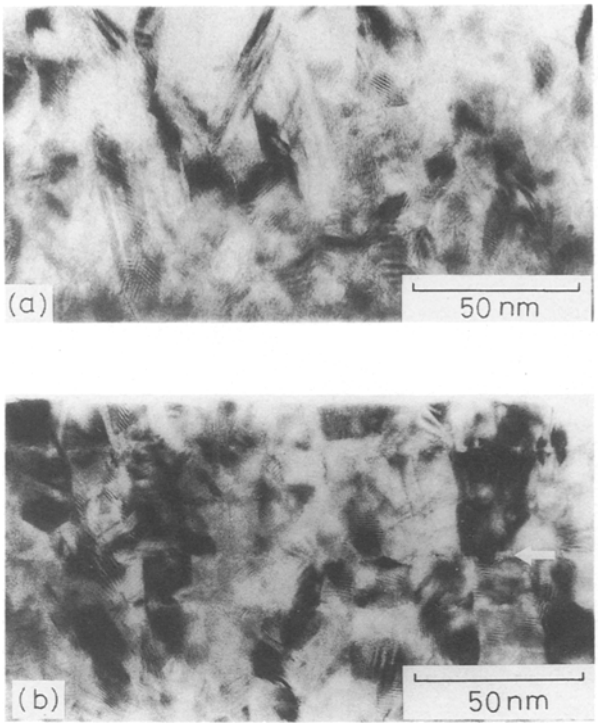


*Figure 1* Cross-sectional transmission electron micrographs of as-sputtered permalloy (a) and permalloy/permalloy-oxide (b) multilayer thin films. Arrows indicate oxide layers.

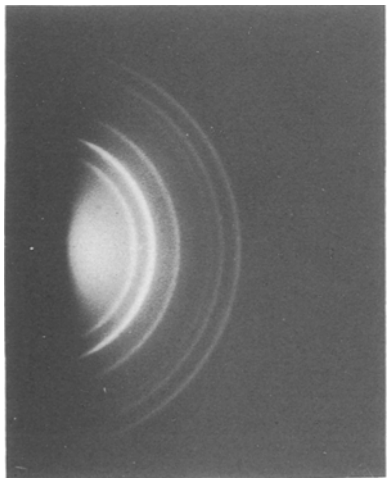

*Figure 2* Reflection high energy electron diffraction pattern of surface oxide of permalloy thin films.

oxide layers. This caused the average grain diameter to decrease with increasing layer number. However, a part of moiré pattern and grain contrast continues beyond the oxide layer. It is therefore thought that the separation between the permalloy layers may be incomplete. This may be caused by the difference in the oxide film thicknesses between crystal planes. A RHEED pattern obtained from the oxidized surface is shown in Fig. 2. The diffraction pattern shows rings, indicating that the oxide layer is polycrystalline. The obtained lattice spacings and relative strengths are shown in Table I. These data are in good agreement with the $NiFe_2O_4$ phase shown in JCPDS card No. 10-325. Since the permalloy composition is Ni-19 wt % Fe (approximately composition $Ni_4Fe$), the concentration of Fe in the oxide film is higher than that of permalloy. It is therefore thought that Fe species oxidize preferentially near the film surface.

## 3.2. Depth composition profile

Oxygen depth-composition profiles for single-, four-, and eight-layer thin films are shown in Fig. 3. On the single-layer profile, only oxygen species were observed near both the surface and the substrate. The four-layer profile shows three independent oxygen peaks. The eight-layer profile shows linked oxygen peaks. The layer number dependence of electric and magnetic properties for the multilayer thin films was linear to the eight-layer specimen as described later. Thus the duplication of oxygen peaks may be due to resolution limit. SIMS measurement profiles for twelve- and sixteen-layer specimens did not show oxygen peaks.

## 3.3. Coercivity and saturation magnetization

The influence of layer number on the coercivity of permalloy multilayer thin films is shown in Fig. 4. The total thickness of deposited permalloy thin films is 100 nm for all specimens. Although the coercivity of the single layer specimen was 200 A $m^{-1}$, those of the multilayered specimens decreased linearly with increasing layer number, reaching a minimum of approximately 110 A $cm^{-1}$ for the eight-layer specimen. The coercivities then increased for the twelve- and sixteen-layer specimens.

Coercivity decrease with multilayering of permalloy thin films has been reported previously after Clow [2]. Magnetic interaction normal to the film surface is decreased by inserting nonmagnetic thin films. Therefore, since anisotropic dispersion, magnetic wall energy, and static magnetic energy of multilayered thin films decrease, coercivity decreases [6]. Coercivity is thought to be decreased by suppression of grain growth [5, 7, 8]. This is considered to cause the coercivity decrease obtained in the present study.

On the other hand, it is assumed that the coercivity increase in the twelve- and sixteen-layer specimens is

TABLE I RHEED data for permalloy surface oxide

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| JCPDS No. | *d* (nm) | 0.482 | 0.2948 | 0.2513 | 0.2408 | 0.2085 | 0.1913 | 0.1703 | 0.1605 | 0.1476 |
| 10-325 | s* | 20 | 30 | 100 | 7 | 25 | 3 | 7 | 30 | 40 |
| | (h k l) | 111 | 220 | 311 | 222 | 400 | 331 | 422 | 511 | 440 |
| surface oxide | *d* (nm) | $0.48_1$ | $0.29_5$ | $0.25_0$ | – | $0.20_8$ | – | $0.17_0$ | $0.15_9$ | $0.14_5$ |
| | s** | w | s | vs | | s | | w | s | s |

*,**: relative strength; vs: very strong; s: strong; w: weak.

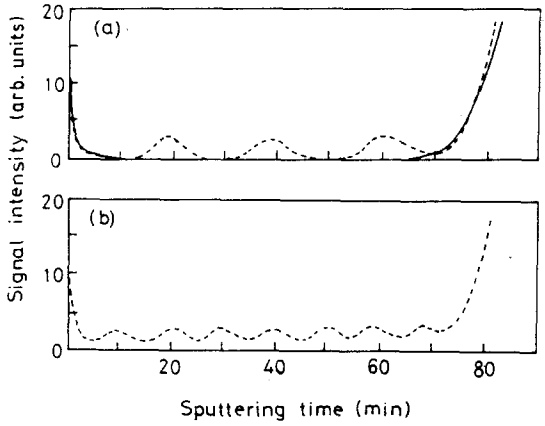


*Figure 3* Oxygen depth profiles for single and four layered (a) and eight layered (b) thin permalloy films measured by SIMS.

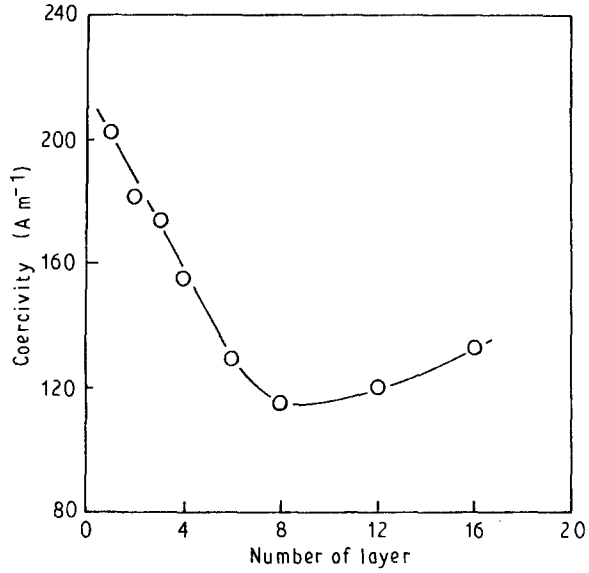


*Figure 4* Influence of number of layers on coercivity of permalloy/permalloy-oxide multilayered thin films.

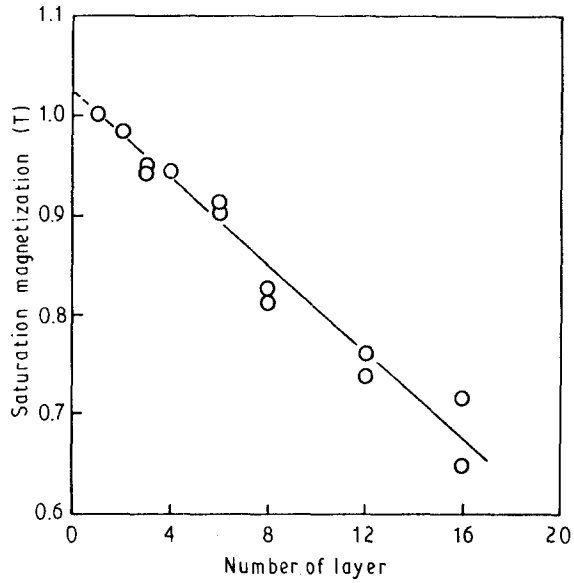


*Figure 5* Relationship between saturation magnetization and number of layers of permalloy/permalloy-oxide multilayer thin films.

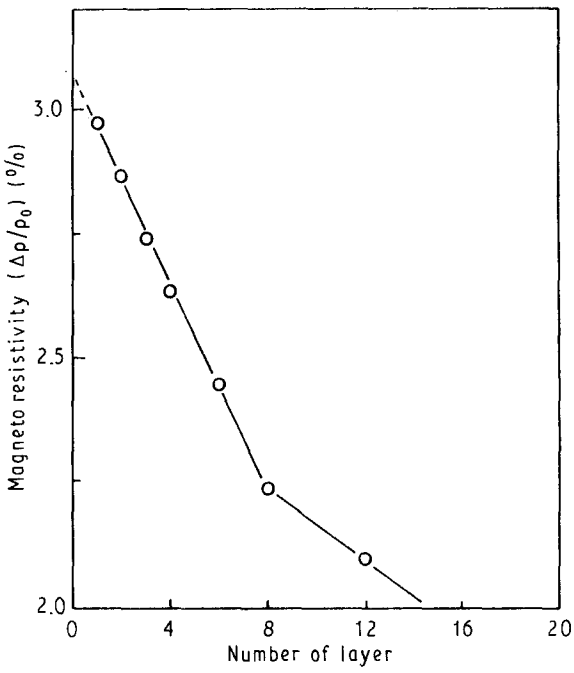


*Figure 6* Influence of number of layers on magnetoresistivity of permalloy/permalloy-oxide multilayer thin films.

related to solute oxygen in the permalloy [9, 10]. Generally, oxygen atoms diffuse into metal layers before oxide growth. Since the permalloy's total thickness is constant, the solute oxygen content increases with increasing layer number. Saturation magnetization of the multilayer specimen is shown in Fig. 5. The saturation magnetization decreases linearly with increasing layer number to the sixteen-layer specimen. The decrease became notable over 16-layer specimen. The saturation magnetization decrease is thought to be due primarily to oxidation of the permalloy surface and secondarily to the solute oxygen. Details will be reported elsewhere.

## 3.4. Magnetoresistivity

The magnetoresistivity as a function of the number of layer specimen is shown in Fig. 6. The magnetoresistivity first decreased linearly with increasing numbers of layers up to eight layers. This decrease is due to the cross-sectional area decrease of the permalloy layer of the specimen, electron scattering increase at the oxide layers, solute oxygen content increase, and other factors.

The magnetoresistivity is conveniently shown as the ratio $\Delta\rho/\rho_0$ of electrical resistance under zero magnetic field $\rho_0$ and saturation magnetoresistance change $\Delta\rho$ when a magnetic field is applied. Values of $\rho_0$, $\Delta\rho$ and $\Delta\rho/\rho_0$ of multilayer films having one, three, and six layers are shown in Table II. $\rho_0$ increased and $\Delta\rho$ decreased, respectively, with increasing numbers of layers. The percentages of $\rho_0$ increase and $\Delta\rho$ decrease of the six-layer specimen were 15.2 and 7.1%, respectively. Therefore, the contribution of $\Delta\rho$ is smaller than that of $\rho_0$ for the $\Delta\rho/\rho_0$ decrease.

The electrical resistance change originates in both electron scattering in the oxide films and solute oxygen atoms. If the insulator oxide-layer thickness is

TABLE II Magnetoresistive properties of permalloy/permalloy-oxide multilayered thin films. $\Delta\rho$, $\rho_0$, indicate magnetoresistive change and electrical resistivity containing all layers. $\Delta\rho_1$ and $\rho_1$ indicate magnetoresistive change and electrical resistivity, respectively, without an oxide layer. $\Delta\rho/\rho_0$ and $\Delta\rho_1/\rho_1$ are shown as percentages.

| | Multilayer film | | | Estimated* | | |
|---|---|---|---|---|---|---|
| $N$ | $\Delta\rho$ | $\rho_0$ | $\Delta\rho/\rho_0$ | $\Delta\rho_1$ | $\rho_1$ | $\Delta\rho_1/\rho_1$ |
| 1 | 0.70 | 23.7 | 2.95 | 0.71 | 23.4 | $3.0_3$ |
| 3 | 0.68 | 24.9 | 2.73 | 0.71 | 23.6 | $3.0_1$ |
| 6 | 0.65 | 27.3 | 2.38 | 0.72 | 24.0 | $3.0_0$ |

$N$: number of layers; *: only permalloy layers.

20 nm, the electrical resistivities of only the permalloy layers of the multilayer specimens is shown by $\rho_1$ in Table II. The estimated $\rho_1$ increases are very small. It is therefore thought that the electrical resistivity increase is mainly due to the electron scattering in the oxide films. Generally, the electrical resistivities of thin metallic films increase very rapidly when the thickness approaches the mean free path of electrons. From the above estimation, the electrical resistivity increase caused by multilayering is mainly due to electron scattering in the oxide layer. On the other hand, estimated $\Delta\rho_1$ of only permalloy layers are also shown in Table II. These values are independent of the number of layers. From these results, it is considered that the effect of the solute oxygen on the $\rho_0$ and $\Delta\rho$ is neglegible. The effect of the solute oxygen on the magnetic and electrical properties of the films are continuously investigated.

## 3.5. Magnetic domain pattern

The coercivities of the multilayered permalloy specimens decreased with increasing the numbers of layers. It is therefore thought that magnetic domain patterns are changed by multilayering effect. Bitter patterns of the multilayered permalloy specimens are shown in Fig. 7. The pattern of the single layer specimen consisted of cross-tie walls. However, the linear density of cross-tie walls decreased with increasing numbers of layers and then the cross-tie walls were eliminated.

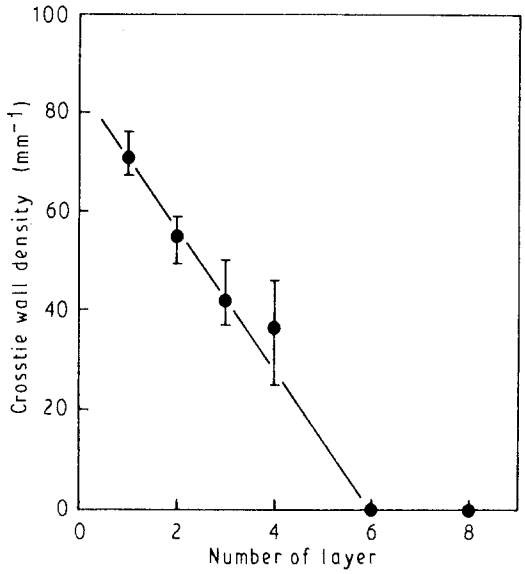


*Figure 8* Cross-tie wall density vs layer number relation for permalloy/permalloy-oxide multilayered thin films.

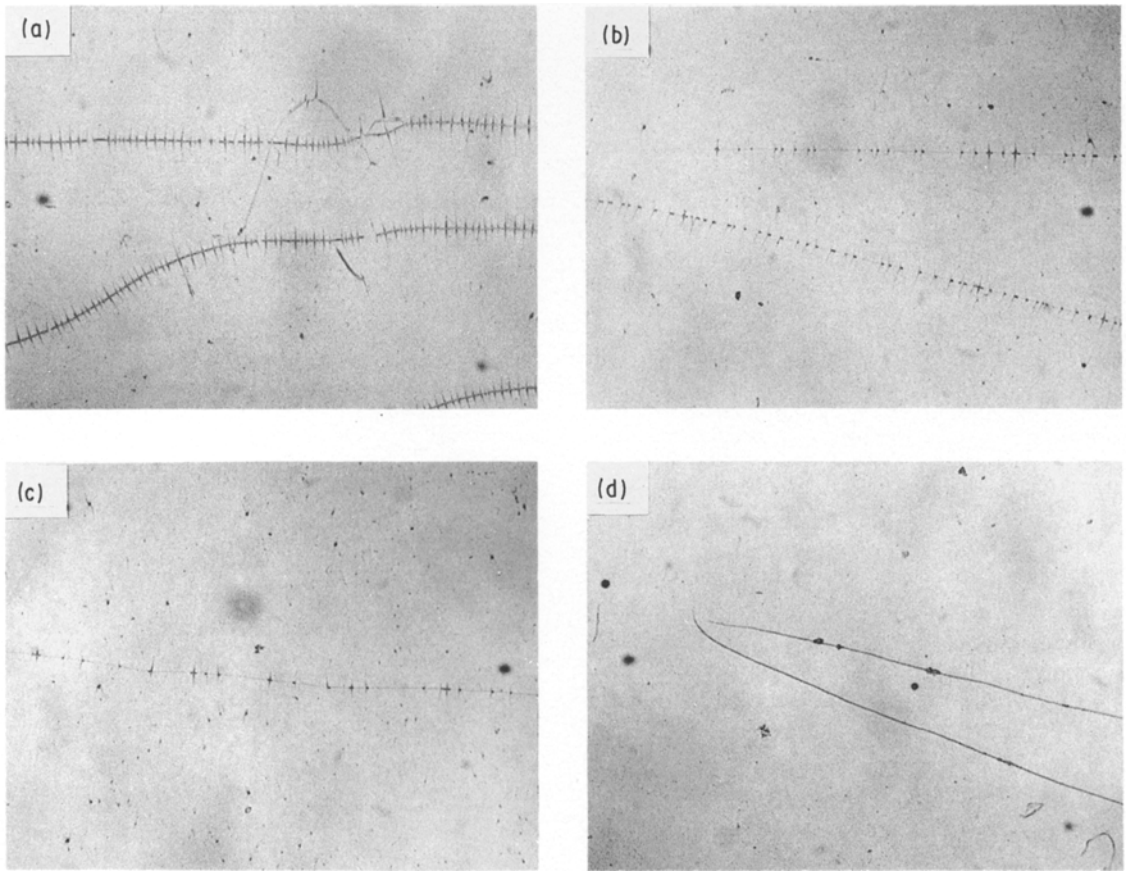


*Figure 7* Magnetic domain patterns of permalloy/permalloy-oxide multilayer thin films, (a) single, (b) two, (c) four, (d) eight-layer.

This was due to a decrease in magnetic interaction normal to the film plane. The interaction decrease is due to separation of the permalloy layers by nonmagnetic oxide layers.

## 4. Conclusion

Permalloy multilayer thin films were prepared using a surface oxidation process. The estimated surface oxide layer thickness was approximately 20 nm. The oxide determined by RHEED was $NiFe_2O_4$. The coercivity and magnetic wall density decreased with increasing numbers of layers. These were mainly due to a decrease in magnetic interaction normal to the film plane and obstruction of crystal grain growth by the oxide layer. Electron scattering by the oxide layer is an important factor in decreasing magnetoresistivity.


## Acknowledgement

We are grateful to Mr. N. Shimizu and Mr. T. Shimotsu for analysing the specimens.


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*Received 14 March*
*and accepted 3 December 1990*