

1988

# A study of magnetoresistivity in multilayer thin film

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*Iowa State University*

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**A study of magnetoresistivity in multilayer thin film**

**Hur, Jae Hun, Ph.D.**

**Iowa State University, 1988**

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A study of magnetoresistivity  
in multilayer thin film

by

Jae Hun Hur

A Dissertation Submitted to the  
Graduate Faculty in Partial Fulfillment of the  
Requirements for the Degree of  
DOCTOR OF PHILOSOPHY

Department: Electrical Engineering and Computer Engineering  
Major: Electrical Engineering (Microelectronics)

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1988



## TABLE OF CONTENTS

	PAGE
INTRODUCTION . . . . .	1
BACKGROUND THEORY . . . . .	4
Magnetoresistance Effect . . . . .	4
Characteristics of Double Layer Structure . . . . .	4
NiFeCo for Magnetic Layers . . . . .	10
Tantalum for Nonferromagnetic Middle Layer . . . . .	12
Magnetic Annealing . . . . .	13
R.F. sputtering . . . . .	17
Applications of Multilayer Thin Film Structure . . . . .	20
Memory cells . . . . .	20
Transverse mode . . . . .	20
Longitudinal mode . . . . .	24
Transducers (Magnetoresistive read head) . . . . .	25
EXPERIMENTAL PROCEDURES . . . . .	28
No Pre-substrate Heating . . . . .	28
Film deposition . . . . .	28
Magnetoresistive ratio measurement . . . . .	29
Annealing process . . . . .	30
Grain size measurement . . . . .	31
Films with Elevated Substrate Temperature . . . . .	32
Elevating the substrate temperature . . . . .	32
Film deposition . . . . .	32
Magnetic properties measurement . . . . .	33
RESULTS AND DISCUSSION . . . . .	34
Single Layer versus Double Layer . . . . .	34
Films Without Substrate Heating . . . . .	34
Grains of the films . . . . .	46
Films with Elevated Substrate Temperature . . . . .	55
CONCLUSIONS AND FUTURE STUDIES . . . . .	59
Conclusions . . . . .	59
Future Studies . . . . .	60
BIBLIOGRAPHY . . . . .	61
ACKNOWLEDGEMENTS . . . . .	66

## LIST OF FIGURES

	PAGE
FIGURE 1. Schematic diagram of magnetoresistive element . . . . .	5
FIGURE 2. Double layer thin film structure (cross section) . . . . .	6
FIGURE 3. Magnetization change in Neel wall of single layer film sample . . . . .	6
FIGURE 4. A Neel wall in a single layer film . . . . .	8
FIGURE 5. Schematic diagrams of edge view of double layer Neel walls . . . . .	9
FIGURE 6. The non-magnetostrictive composition line of NiFeCo . . . . .	11
FIGURE 7. Current flow for memory element . . . . .	14
FIGURE 8. Grain size dependence on (a) substrate temperature, (b) deposition rate, (c) annealing temperature, and (d) film thickness ( $T_s$ is substrate temperature) . . . . .	15
FIGURE 9. Formation of a thin film (after Chapman [33]) . . . . .	19
FIGURE 10. Transverse mode operation . . . . .	22
FIGURE 11. Longitudinal mode operation . . . . .	26
FIGURE 12. Deposition of multilayer films . . . . .	28
FIGURE 13. Rectangular four-point probe configuration . . . . .	30
FIGURE 14. Annealing oven configuration (cross section) . . . . .	31
FIGURE 15. B-H loops with easy and hard axis (a) single layer film (b) double layer film (1.8 Oe/div horizontal and 0.8 Oe/div vertical) . . . . .	35

FIGURE 16. MR ratio vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 40 Angstroms thick . . . . .	40
FIGURE 17. Resistance vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 40 Angstroms thick . . . . .	41
FIGURE 18. MR ratio vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 50 Angstroms thick . . . . .	42
FIGURE 19. Resistance vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 50 Angstroms thick . . . . .	43
FIGURE 20. MR ratio vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 60 Angstroms thick . . . . .	44
FIGURE 21. Resistance vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 60 Angstroms thick . . . . .	45
FIGURE 22. Grains of the top magnetic layer . . . . .	46
FIGURE 23. Grains of the top magnetic layer annealed at 200°C for 1 hour . . . . .	47
FIGURE 24. Grains of the top magnetic layer annealed at 200°C for 2 hours . . . . .	48
FIGURE 25. Grains of the top magnetic layer annealed at 200°C for 4 hours . . . . .	49
FIGURE 26. Grains of the top magnetic layer annealed at 200°C for 8 hours . . . . .	50
FIGURE 27. Grains of the top magnetic layer annealed at 300°C for 1 hour . . . . .	51
FIGURE 28. Grains of the top magnetic layer annealed at 300°C for 2 hours . . . . .	52

FIGURE 29. Grains of the top magnetic layer annealed  
at 300°C for 4 hours . . . . . 53

FIGURE 30. Grains of the top magnetic layer annealed  
at 300°C for 8 hours . . . . . 54

FIGURE 31. Grains of the top magnetic layer with  
substrate temperature at 200°C . . . . . 57

FIGURE 32. Grains of the top magnetic layer with  
substrate temperature at 300°C . . . . . 58

## LIST OF TABLES

	PAGE
TABLE 1. Double layer films annealed at 200°C . . . . .	38
TABLE 2. Double layer films annealed at 300°C . . . . .	39
TABLE 3. Data for films with 200°C substrate temperature . . . . .	55
TABLE 4. Data for films with 300°C substrate temperature . . . . .	56

## INTRODUCTION

Ever since William Thomson discovered the anisotropic magnetoresistance effect in ferromagnetic materials, many studies and attempts have been done to fabricate memory cells and transducers using this magnetoresistance effect [1]. Magnetoresistance is observed as a change in the electrical resistivity of a magnetic substance when it is subjected to a magnetic field. It is caused by a quantum mechanical electron spin-orbital interaction. The resistivity is dependent on the angle between the magnetization of the element and the direction of sense current flowing through the element. The magnetoresistive ratio is the percentage change of the resistance of the magnetic substance.

In early 1970s, magnetic core memory, magnetic thin film memory and semiconductor memory technologies were competing with each other, but only semiconductor memory technology prevailed simply because it didn't need lots of connections between elements as other technologies did.

Current advanced fabrication technology enables magnetic thin film memories to be compatible with modern semiconductor processing. Moreover these thin film memories have some special features such as nonvolatility, radiation hardness, high density (comparable to CMOS static RAMs),

high speed (1000 times faster than bubbles), and unlimited Read/Write.

For submicron dimensions of these memory cells and sensor elements, the demagnetizing fields play a major role in determining the performance of magnetoresistive elements. One simple answer for this demagnetizing field problem is a double layer configuration composed of two ferromagnetic layers with a nonferromagnetic layer between the two magnetic layers. This structure results in a decrease of the demagnetizing fields from what they would be for a single layer element. The decrease is due to the magnetic flux closure of the sandwich configuration.

For these applications, it is desirable to have a high magnetoresistive ratio to obtain appropriate signal levels. Permalloy (81 wt.% Ni, 19 wt.% Fe) has previously been used as the ferromagnetic material but a Ternary alloy (65 wt.% Ni, 15 wt.% Fe, 20 wt.% Co) was used in our study since it has some advantages over Permalloy. These advantages are bulk magnetoresistive ratio and a lower limitation in compositional variation which give rise to low easy axis dispersion.

Since previous studies have been done on single layer films, our goal is to study the multilayer structure which we subsequently used to fabricate magnetoresistive device

elements. In this research, a Ternary alloy for the material of the two ferromagnetic layers, and Tantalum for the middle layer material were studied for the multilayer magnetic thin film structures. An investigation correlating grain size, resistivity, and magnetoresistive ratio with after-deposition annealing was performed. Finally, effects of substrate temperature before and after deposition on the magnetoresistive ratio of these multilayer films were also studied.



## BACKGROUND THEORY

## Magnetoresistance Effect

Magnetoresistance is observed as a change in the electrical resistivity of a substance when it is exposed to a magnetic field. Quantum mechanical electron spin-orbital interaction is a cause of this phenomenon [2-3]. As shown in Figure 1, the resistivity which is dependent on the angle,  $\theta$ , between the magnetization and direction of the sense current, can be represented as in equation (1),

$$\begin{aligned}\rho &= \rho_0 + \delta\rho_{\max}\cos^2\theta \\ &= \rho_0 + (\rho_1 - \rho_2)\cos^2\theta\end{aligned}\quad (1)$$

where  $\rho_0$  is inherent in the material,  $\rho_1$  and  $\rho_2$  are the resistivities when the current is flowing parallel and perpendicular to the magnetization respectively. The resistivity is maximum when the angle is  $0^\circ$  and is minimum,  $\rho_0$ , when it is  $90^\circ$ .

## Characteristics of Double Layer Structure

In single layer films the demagnetizing field for submicron dimensions of the magnetoresistive elements is so large that it causes complicated micromagnetic domain structures and noise effects (i.e., Barkhausen noise) which

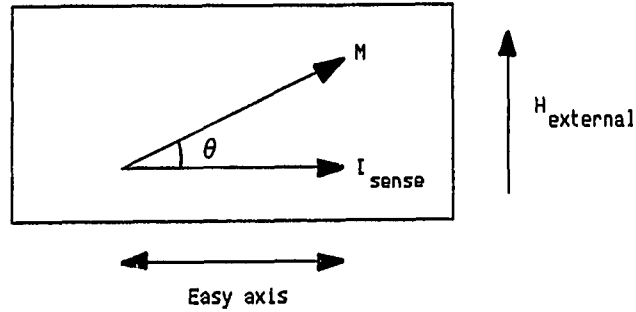


FIGURE 1. Schematic diagram of magnetoresistive element

result in a significant degradation of the magnetoresistive switching characteristics. One simple solution to answer for this demagnetizing field problem is a double layer structure as shown in Figure 2. This configuration results in a decrease of the demagnetizing field due to the magnetic flux closure primarily at its edges.

Many studies have been done to characterize the multilayer thin films [4-8]. For very thin films, as a result of the high stray field energy, the Neel type wall becomes favorable. This is a wall in which the magnetization turns around an axis which is perpendicular to the plane of the film, as shown in Figure 3.

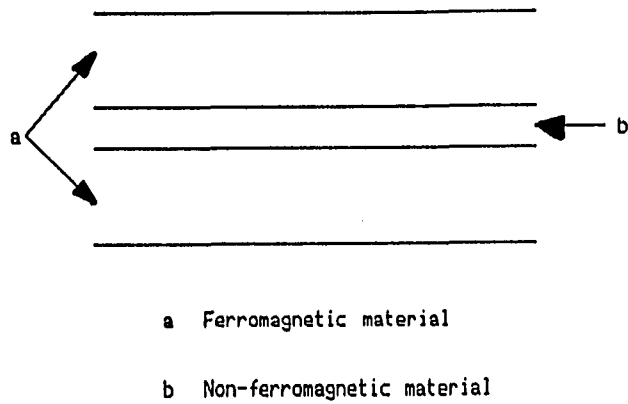


FIGURE 2. Double layer thin film structure (cross section)

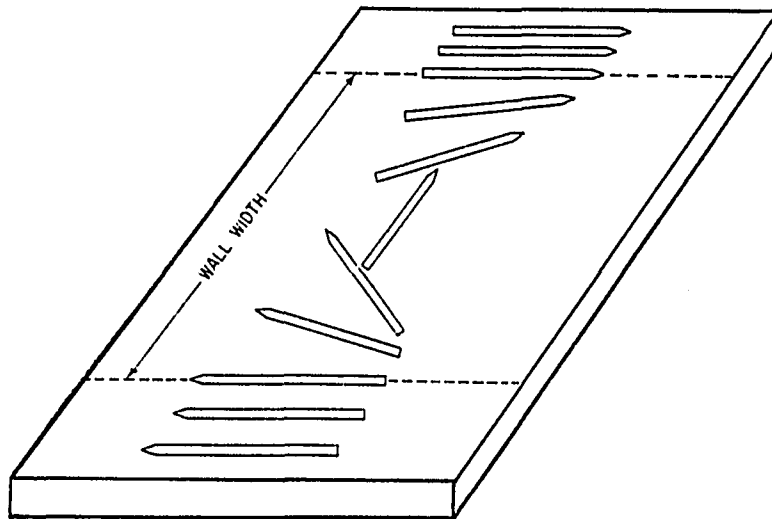


FIGURE 3. Magnetization change in Neel wall of single layer film sample

The total energy of the wall consists primarily of three terms: exchange energy, anisotropy energy, and stray field energy [9]. The surface energy density of the Neel wall is given by

$$E = A \left( \frac{\pi}{b} \right) b + \frac{1}{2} b \frac{M_S H_k}{2} + \frac{\pi T b M_S^2}{b + T} \quad (2)$$

where A : Exchange stiffness constant

b : Wall width

$H_k$  : Anisotropy field

$M_S$  : Saturation magnetization

T : Film thickness

Since a Neel wall lies in the plane of the film as shown in Figure 4, it implies that two superimposed Neel walls of opposite polarity can exist in a nearly closed flux loop configuration [10-12]. This effect lowers the energy of Neel walls in double layer films.

Three different situations can exist in the demagnetized Neel wall configurations as shown in Figure 5.

In Figure 5 - a, the Neel walls separate parallel domains and the film has positive coupling between domains in each layer. Figure 5 - b shows the paired Neel wall separating antiparallel domains for which there is negative coupling between domains in each layer. Finally in Figure 5

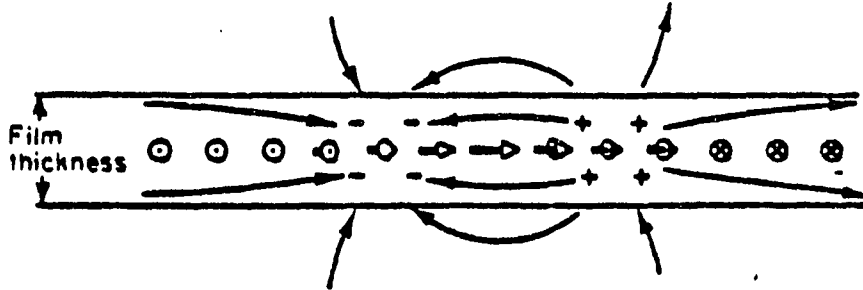


FIGURE 4. A Neel wall in a single layer film

- c, the wall in the bottom layer has induced a quasiwall in the upper film. The quasiwall separates parallel oriented magnetizations, but the wall-quasiwall pair has a lower total energy than a wall in just one layer [13].

For the surface energy density of paired Neel walls, the energy equation becomes [10].

$$E = A \left( \frac{\pi}{b} \right) b + \frac{1}{2} b K + \frac{1}{b} \pi^3 M_S^2 T \left( \frac{1}{2} D + \frac{1}{3} T \right) \quad (3)$$

where  $K$  : Anisotropy constant ( $=1/2 H_k M_S$ )

$D$  : Middle layer thickness

$A$  : Exchange stiffness

$b$  : Wall width

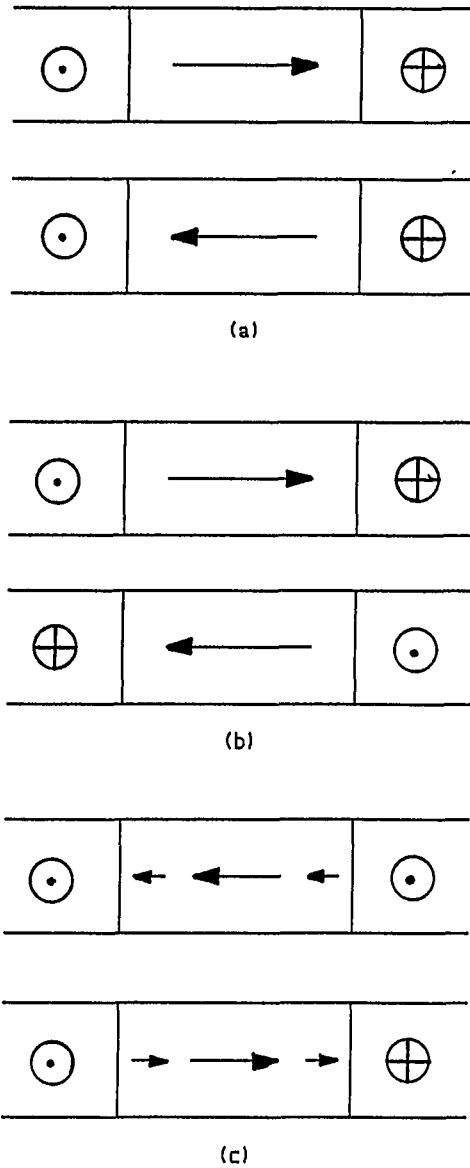


FIGURE 5. Schematic diagrams of edge view of double layer Neel walls

By comparing equation 2 and 3, the only difference is in the third term, the magnetostatic stray field energy. The total energy density of the wall structure of double layer films is much lower than the energy density of the wall structure of a single layer primarily because of the stray field energy term.

#### NiFeCo for Magnetic Layers

Magnetic materials for memory cells and transducers should have properties such as low coercivity, low easy axis dispersion, zero magnetostriction to minimize Barkhausen noise effects caused by localized stress fluctuation, and, finally, a good magnetoresistive ratio to have appropriate signal level output [14]. The low easy axis dispersion is necessary in order to have coherent magnetization rotation. Otherwise there is a undesirable magnetoresistance characteristic which is a cause of poor detector signal.

Permalloy, 81 wt.% Ni, 19 wt.% Fe, has been used by many researchers as the magnetic material for magnetoresistive elements. Unfortunately, Permalloy has deficiencies such as limitations on being able to control the compositional variation. Lower compositional variation is necessary in order to achieve low easy axis dispersion and zero magnetostriction. Permalloy also has a relatively low magnetoresistive ratio of 2 - 3 %.

Asama et al. investigated a NiCo alloy, 70 wt.% Ni, 30 wt.% Co, to improve the magnetoresistive ratio [15]. It exhibits a higher magnetoresistive ratio than Permalloy, almost 3.8 %, but it has coercive force of 14 - 15 Oe, which causes very noisy sensor output signals.

Because of the limited compositional variational control of Permalloy and high coercivity of NiCo, the ternary alloy of NiFeCo has become a promising material for sensor and other applications.

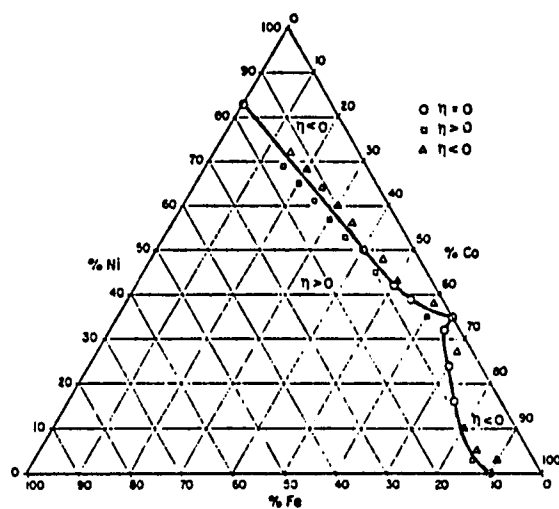


FIGURE 6. The non-magnetostrictive composition line of NiFeCo



As shown in Figure 6, a 65 wt.% Ni, 15 wt.% Fe, 20 wt.% Co ternary alloy is a nonmagnetostrictive composition [16]. It also exhibits low coercivity and low easy axis dispersion. NiFeCo has a magnetoresistive ratio of 3.5 % in contrast to 2.8 % for Permalloy films prepared under similar conditions [14]. A. Collins and I. Sanders [17] found that a maximum in the magnetoresistive ratio occurred for an alloy composition of 60 wt.% Ni, 10 wt.% Fe, 30 wt.% Co. However, that composition is not nonmagnetostrictive and has a relatively high coercive force. Obviously the NiFeCo ternary alloy has desirable magnetic properties compared with many other materials [18-21].

#### Tantalum for Nonferromagnetic Middle Layer

Many different materials, such as SiO, SiO<sub>2</sub>, Pd, Au, Cu, have been used as a nonferromagnetic middle layer to study the magnetic coupling behavior of double layer structures [20].

When SiO<sub>2</sub> is used as a middle layer, since it is an insulator, an electric contact would be required at the end of each of its two magnetic layers. This would be necessary to allow sense current to flow in both magnetic layers. Since the thickness of the magnetic layers is 250 Angstrom each, the process of making these two end contacts is much too difficult.

Tantalum is a conductor and has a relatively high resistivity, 25 ~ 50  $\mu\text{Ohm} - \text{cm}$  for a thin film structure [21]. When it is used as a middle layer, most of the current will flow through the top and bottom magnetic layers parallel to the plane of the film. Because the planar sandwich structure is now conductive, only one electric contact is necessary, either on the top or bottom magnetic layer, to supply sense current to both magnetic layers as shown in Figure 7. A study shows that multilayer films with NiFeCo as the two ferromagnetic layers and a 20 Angstrom Tantalum intermediate layer exhibit good magnetostatic coupling [22].

Tantalum was also used as protection layers to prevent the magnetic layers from oxidizing since Tantalum forms tough self-protective oxides through heat treatment in oxygen or anodic oxidation.

#### Magnetic Annealing

Graham defined magnetic annealing as the heat treatment of a material in a magnetic field in order to change or improve its magnetic properties [23]. In general a thin film will possess more grain boundaries than bulk material since the average grain size will generally be smaller. As shown in Figure 8, the grain size depends on deposition

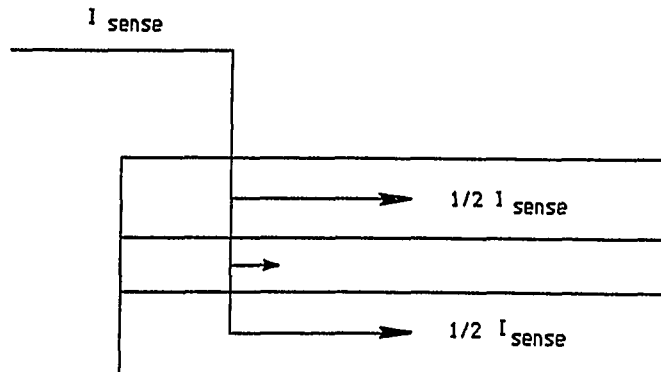


FIGURE 7. Current flow for memory element

conditions such as substrate temperature, deposition rate, and film thickness. Grain size also depends on post deposition annealing time and temperature.

As seen in Figure 8 - a, larger grain size can be obtained by increasing the substrate temperature during

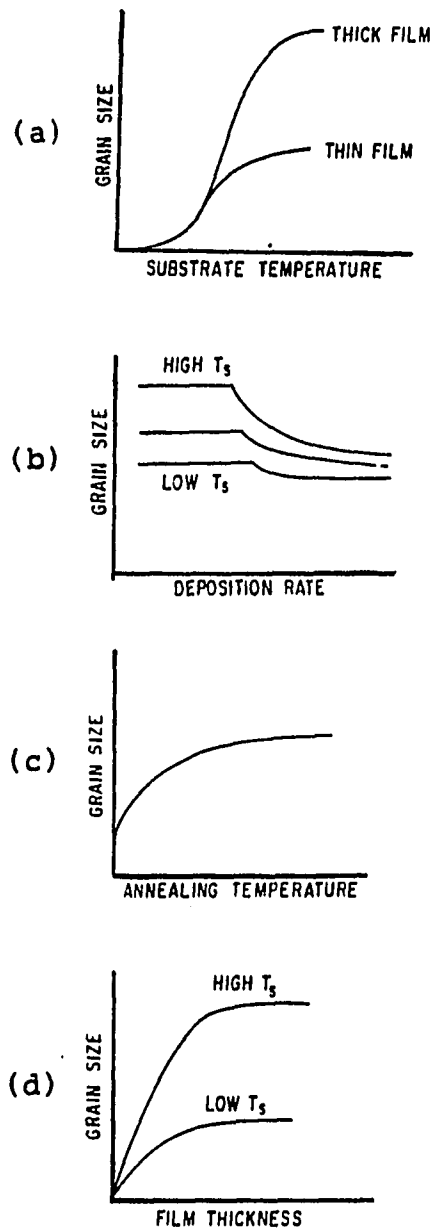


FIGURE 8. Grain size dependence on (a) substrate temperature, (b) deposition rate, (c) annealing temperature, and (d) film thickness ( $T_s$  is substrate temperature)

deposition. This is due to a corresponding increase of the mobility of (target) atoms condensing on the surface of the substrate, which allows the film to decrease its total energy by growing larger grains [24]. There is difference in the grain growth effect caused by annealing at a temperature which is higher than the substrate temperature during deposition and the growth caused by annealing at the deposition temperature. The difference originates from the high activation energy process of thermal diffusion of surface atom of a condensate in the former case as compared with the orderly process of condensation of mobile atoms in the latter case [25].

Many studies have been done on temperature dependence of the magnetoresistance effect in single layer Permalloy thin films [26-29]. They all found that the resistivity decreased because of the larger grain size caused by heat treatment either during or after deposition. The thin film resistivity model of Mayadas and Shatzkes predicts an increase in the measured resistivity with decreasing grain size. Grain size, in turn, is primarily determined by substrate temperature [30].

The effects of substrate temperature on the magnetoresistive ratio for NiFeCo ternary single layer films deposited by electron beam evaporation at a dynamic pressure

of  $10^{-5}$  Torr was studied by A. Collins and I. Sanders [17]. They found that the magnetoresistive ratio increased with increasing substrate temperature and the increase was mainly due to the decrease in resistivity. But they couldn't find any overall correlation between resistivity and grain size. Later V. Chapman et al. studied the variation in resistivity as a function of substrate temperature for single layer Permalloy films prepared by an ultra high vacuum evaporation process [31]. They found that the grain size remained constant over the temperature range considered,  $0^{\circ} \sim 300^{\circ}\text{C}$ , with a film thickness of 400 Angstroms. They claimed that their result was consistent with Mayadas and Shatzkes model because the resistivity remained constant and the magnetoresistive ratio didn't change over the temperature range considered. This contradicted their earlier experiment with NiFeCo.

#### R.F. sputtering

Sputtering is a momentum-transfer process on an atomic scale. The source (target) is bombarded by energetic positive ions, here Argon, and some of the surface atoms are ejected from the target as a result of momentum transfer between incident ions and the target. The target-ejected particles traverse the vacuum chamber and are deposited on the substrate as a thin film.

An electrical insulator can be sputtered using RF sputtering by placing the insulator target material directly on a metal cathode electrode. The electrode is biased with an RF power supply to ion bombard the target during the negative part of the cycle and electron wash it during the positive part of the cycle. It prevents the build-up of positive charge on the target material so that sputtering can continue. The RF sputtering technique has some advantages such as being able to deposit very wide range of metals and insulators with a high film adhesion to the substrate. It also has some drawbacks such as relatively slow speed, damage to growing thin films and substrate materials because of bombardment by energetic secondary electrons emitted from the target and accelerated by the bias potential [32].

In sputter deposition, an atom arrives at the substrate and scatters around on the surface with a motion determined by its binding energy to the substrate and the temperature of the substrate. As shown in Figure 9, after a certain time, the atom will either evaporate from the surface or will join with another single atom to form a doublet which is less mobile than the single atom. These doublets will be joined by other single atoms to form triplets, quadruplets and so on. This is the nucleation stage of thin film

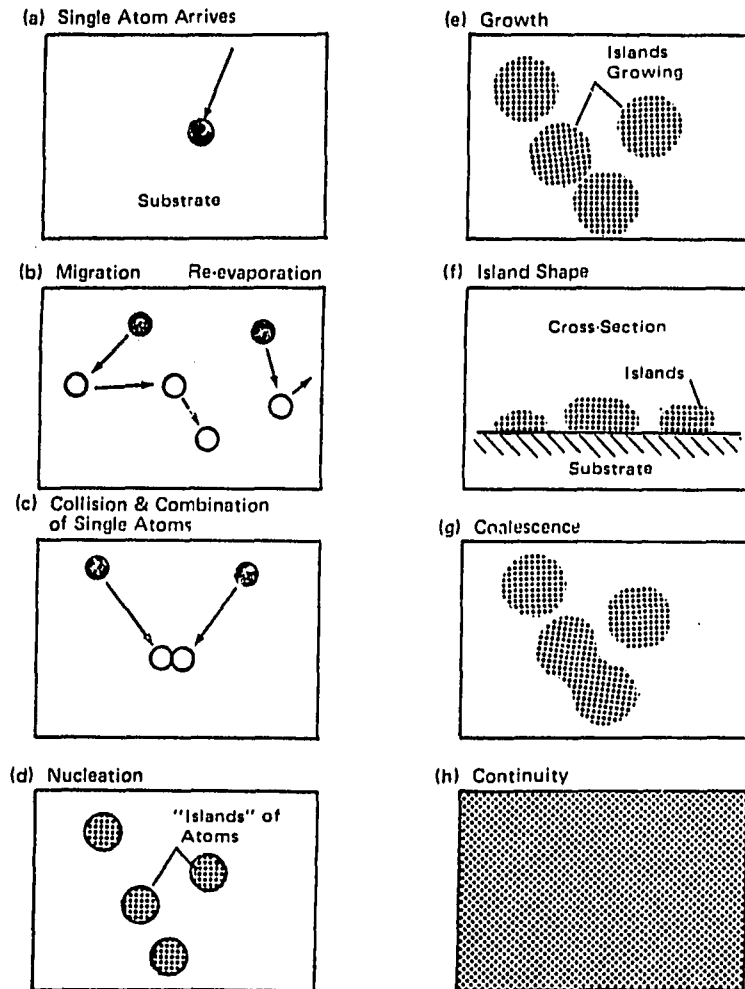


FIGURE 9. Formation of a thin film (after Chapman [33])



growth, leading to the formation of quasi-stable islands which contains tens or hundreds of atoms. Eventually these islands grow large enough to touch. Coalescence of islands proceeds until the film reaches continuity [33].

#### Applications of Multilayer Thin Film Structure

A tremendous amount of work have been done on multilayer thin films in engineering applications [34-38]. Magnetic properties of sandwich structures have been investigated using the magnetoresistance effect of thin Permalloy films [39-40]. As expected, this structure shows strong magnetostatic coupling which decreases the demagnetizing field and improves the switching properties.

#### Memory cells

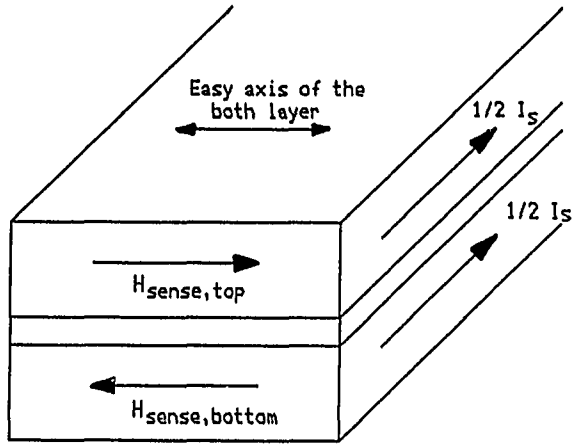
Memory cells, 2 by 20 microns, have been fabricated and studied and there are two different modes of operation [41].

Transverse mode In this mode, the easy axis of the material is perpendicular to the long axis of the element as shown in Figure 10. As shown in Figure 10 - a, an electric current, called the sense current, is passed through the element parallel to the long (longitudinal) axis, which produces a magnetic field, called the sense or transverse field, parallel to the easy axis of the top and bottom magnetic layers. Depending upon the magnetic state of the

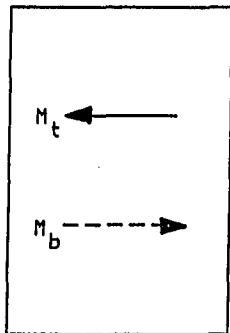
element this transverse field can either be in the same or opposite direction to the easy magnetic directions of the layers. Another magnetic field is applied externally. It is called the word or longitudinal field and is applied parallel to the long axis of the element. This field is produced by a current through a conductor which is directly above and the perpendicular to the element.

The switching process of the transverse memory mode can be described as follows, assuming the word field is variable from zero in this process. Consider initially that the word field is zero, sense current is applied, and both magnetic layers are in their easy direction state. This means the magnetization of the top magnetic layer is oriented the left and that of the bottom layer oriented to the right, antiparallel to the corresponding top and bottom sense field respectively. This is shown in Figure 10 - a and b. The resistance of the element is at a minimum ( $\theta = 90^\circ$ ).

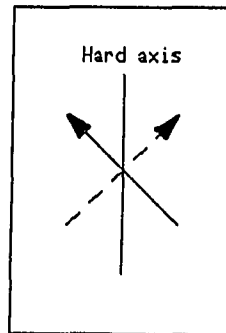
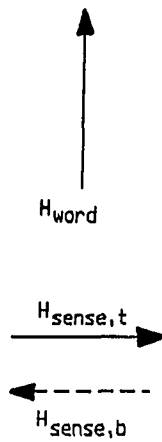
When the word field increased from zero, the magnetization of each layer starts to rotate (scissor) as shown in Figure 10 - c (due to the torque exerted by the word field). This increases the resistance across the element since the angle between the sense current and the magnetization in each layer decreases. This resistance change can be detected electronically on the sense current



(a)  
Current flow in the element



(b)  
Initial state



(c)  
Scissored state

FIGURE 10. Transverse mode operation

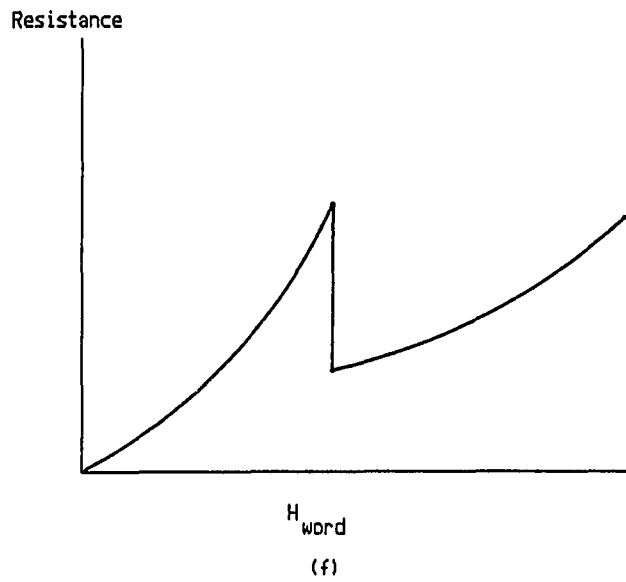
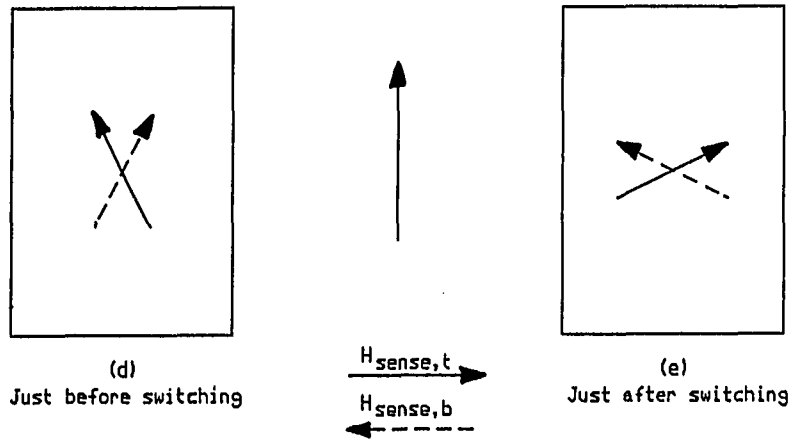


FIGURE 10. (continued)

as a voltage change across the element. As the word current continues to increase, both magnetizations rotate toward each other, and the resistance continues to increase (see Figure 10 - a and f). At a particular threshold value of  $\theta$ , the combined effect of the sense and word fields will be such that the magnetization of each layer will suddenly cross the hard axis of the element. This is called switching. The switching process can be observed by an abrupt decrease in resistance as shown in the sequence from Figure 10 - c to Figure 10 - f. After switching, the sense fields force the magnetization closer to the easy axis, which reduces the resistance as shown in Figure 10 - f. As word field is increased above the value at which switching took place, the resistance (after the sudden decrease at switching) begins to increase as shown in Figure 10 - f. This is due to the magnetization in each layer rotating towards the hard axis of the element. These two states, before and after switching, can represent the two binary states of the memory element. For nondestructive read operation, the word field should not be applied high enough to switch the magnetization of the element.

Longitudinal mode      In this mode, the easy axis of the element is parallel to the long axis of the element as shown in Figure 11 - a. In this mode, the remnant (zero

applied field) magnetizations of each layer are in the same direction because of the shape anisotropy of the elements. By the same token as the transverse mode, the sense field is produced by the sense current and the word field by the word current.

When a constant sense current is applied, the sense field scissors the magnetizations of both top and bottom layers, but the magnetizations of the edges are pinned as shown in Figure 11 - b. When the word current is applied, the word field will rotate the magnetization further and eventually switches the centers first. As the word field increases, the pinned edges will rotate by the wall motion as shown in Figure 11 - c. The final switched state is shown in Figure 11 - d.

#### Transducers (Magnetoresistive read head)

Many studies have been done using the magnetoresistance effect for read head transducers for magnetic recording systems [42-46]. For conventional inductive read heads for disk storage systems a magnetic flux change induces a voltage in a coil that is proportional to the time rate of the flux change. As media track and bit densities increase, signals become weaker, so the sensor needs more turns on the coil or lower head flying height to obtain a proper output signal.

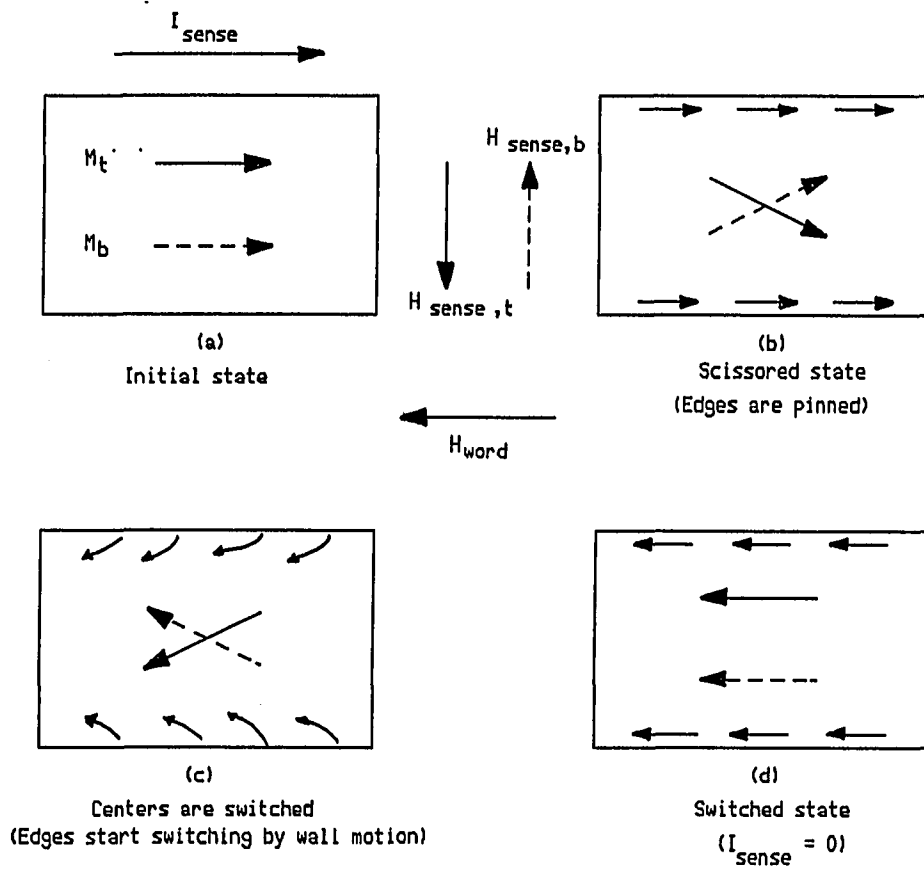


FIGURE 11. Longitudinal mode operation

Magnetoresistive sensors are direct flux sensors, which means that they detect stationary magnetic field signals through direct changes in the resistivity of a magnetoresistive material (e.g., Ternary material). The sensor output is read as a voltage change across the element with a constant sense current flowing through the element. The magnetoresistive outputs are independent of the linear velocity of the flux transition. A coupled multilayer magnetoresistive head having a separate read and write transducer has an ability to write wide read narrow. This head was studied by G. Kelley et al. and has a track density of more than 2500 tracks per inch [47].



## EXPERIMENTAL PROCEDURES

## No Pre-substrate Heating

Film deposition

Five layers of films were deposited by R.F. sputtering on an oxidized 3-inch silicon wafer.

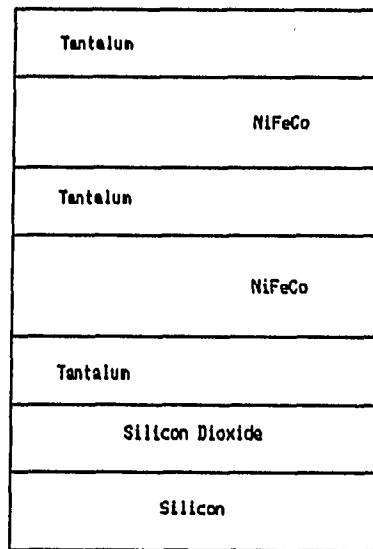


FIGURE 12. Deposition of multilayer films

As shown in Figure 12, 100 Angstroms of Tantalum was deposited first to prevent silicon dioxide from diffusing into the first Ternary alloy. A nonferromagnetic Tantalum middle layer, between 40 and 60 Angstrom thick, was deposited between, the nonmagnetostrictive NiFeCo Ternary

layers. Each Ternary layers was 250 Angstroms thick. Finally, 100 Angstrom of Tantalum was deposited on top to protect the second ferromagnetic layer from oxidation. All five layers were deposited consecutively without breaking a vacuum. All the films were deposited at Iowa State University by R.F. sputtering with Argon gas at 5 to 6 milli-Torr. The sputtering rates were 1 Angstrom per second for the ferromagnetic layers and 0.5 Angstrom per second for Tantalum layers. A 6 Oe external field was applied to induce the anisotropy of the films during deposition.

Thicknesses of the films during the sputtering process were measured by an Inficon model XTM thickness monitor. The B-H hysteresis loop method was used to measure the coercivity ( $H_C$ ), the anisotropy field ( $H_K$ ), and the saturation magnetization ( $M_S$ ). After deposition a wafer was divided into four pieces. Each piece (from the same deposition run) could then be annealed at different temperatures and different lengths of time.

#### Magnetoresistive ratio measurement

Magnetoresistive ratios were measured after deposition. Simple isotropic resistivity of an unpatterned film can be easily measured with a conventional four-point probe. Since the magnetoresistance has an anisotropic nature itself, the in-line four-point probe cannot be used for the

magnetoresistance measurement because the current flow is not uniform.

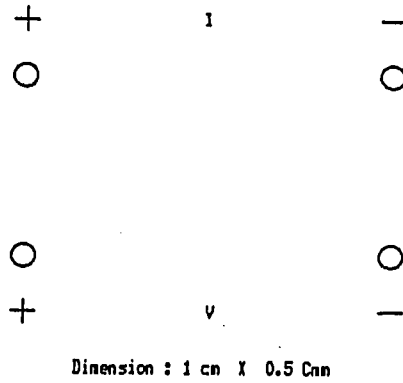


FIGURE 13. Rectangular four-point probe configuration

R. Norton designed a modified geometry four-point probe, as shown in Figure 13, which works very well for magnetoresistivity measurement [48]. The aspect ratio of the probe is 2 : 1 with spring loaded Tungsten tips. Magnetoresistive ratios were measured automatically by using a Zenith Z-100 PC with a rotating magnetic field of 900 Gauss to saturate the film in any direction.

#### Annealing process

After the initial magnetoresistive ratio measurement, films were annealed at temperatures from 200°C up to 300°C at atmospheric pressure. An annealing oven was made as shown in Figure 14.

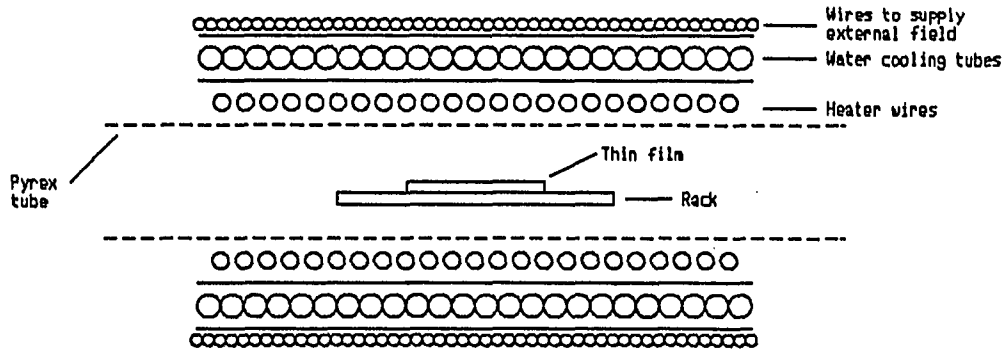


FIGURE 14. Annealing oven configuration (cross section)

An external field of 100 Oe was applied parallel to the easy axis of the films during annealing. Since annealing was not performed in vacuum, the films were easily oxidized. Consequently, a forming gas, 15 % Hydrogen in Nitrogen, was used to prevent oxidation throughout the annealing process. Each wafer was annealed for a different length of time at a different temperature. After annealing, magnetoresistive ratios were measured again on the same film.

#### Grain size measurement

In order to measure grain sizes of the Ternary layer before and after annealing, using a Scanning Electron Microscope, it is necessary to etch off the top Tantalum

layer. J. Grossman and D. S. Herman [49] found an etchant for thin films of Tantalum and Tantalum compounds such as  $Ta_2O_5$  and TaN. It consists of 30 % diluted (with water) sodium hydroxide (NaOH) and 35 % diluted hydrogen peroxide ( $H_2O_2$ ). The proportion is about 9 or 10 to 1 of NaOH to  $H_2O_2$ . The solution was heated to  $90^\circ C$  and then the  $H_2O_2$  was added. The film was then etched in the mixture. The etch rate is 1000 - 2000 Angstroms per minute. After etching off the top Tantalum layer, the films were observed by SEM. Pictures for different annealing times and different annealing temperatures were investigated.

#### Films with Elevated Substrate Temperature

##### Elevating the substrate temperature

The substrates were heated to  $200^\circ C$  or  $300^\circ C$  before depositing the layers of the film. Before deposition began the heater was turned off.

##### Film deposition

Five layers of films, as shown in Figure 12, were deposited on the heated substrate by RF sputtering with Argon gas at 5 to 6 milli - Torr. A 6 Oe external field was applied to induce the anisotropy of the films during deposition. The thicknesses of the magnetic layers were 250 Angstroms thick and the Tantalum middle layer was between 40

to 60 Angstroms thick. The outer Tantalum layers were 100 Angstrom thick.

#### Magnetic properties measurement

The coercivity, the anisotropy field, and the saturation magnetization were measured by the B-H looper method after the deposition. Magnetoresistive ratios were also measured by the rectangular four-point probe. Finally, grains sizes of the films were observed by Scanning Electron Microscopy after etching off the Tantalum top layer. There was no post annealing process for these films.

## RESULTS AND DISCUSSION

## Single Layer versus Double Layer

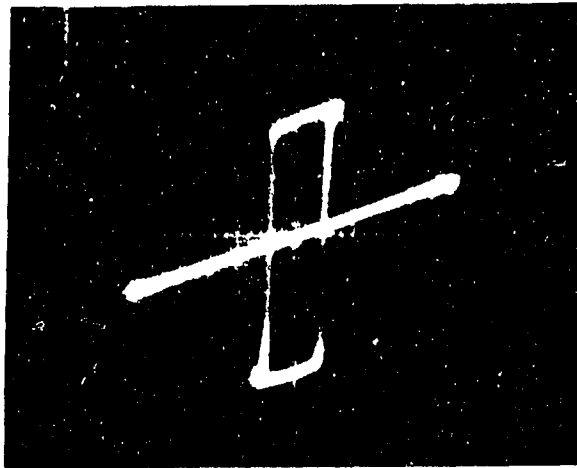
In Figure 15 - a, a single magnetic layer film is composed of 100 Angstroms Tantalum, 250 Angstroms NiFeCo, and 100 Angstroms Tantalum. In Figure 15- b, a double layer structure is composed of 100 Angstroms Tantalum, 250 Angstroms NiFeCo, 60 Angstroms Tantalum, 250 Angstroms NiFeCo, and 100 Angstroms Tantalum. The coercivity of the single layer film is higher than that of the double layer film. The double layer film reduces the demagnetizing fields through magnetostatic coupling between magnetic layers. This magnetostatic coupling also decreases the coercive force of the film by reducing the wall energy of each layer. There was not noticeable difference in  $H_k$  between the single and double magnetic layer films.

## Films Without Substrate Heating

Tables 1 and 2 show the magnetic properties of the films with double magnetic layers before and after annealing at 200°C and 300°C respectively. There were noticeable changes in the magnetoresistive ratio and the resistance as shown in Figures 16 to 21. As shown in Figure 16, four different wafers were divided into four pieces.



(a)



(b)

FIGURE 15. B-H loops with easy and hard axis (a) single layer film (b) double layer film (1.8 Oe/div horizontal and 0.8 Oe/div vertical)



One piece of each wafer was annealed at 200°C and another piece was annealed at 300°C. Each piece (even from the same film) had different values of magnetoresistive ratio and resistance because the film thickness was not uniform across the film. Each wafer was annealed for 1, 2, 4, and 8 hours.

Figures 16, 18, and 20, pertaining to films annealed at 200°C and 300°C, demonstrate the relationship between magnetoresistive ratio and annealing time. At 200°C, magnetoresistive ratios increased by an average of 15 % and, at 300°C, magnetoresistive ratios increased by an average of 22 %. The maximum magnetoresistive ratio was 3.1 % for films annealed at 300°C for 1 hour. Figures 17, 19, and 21 show the resistance change before and after annealing with respect to annealing time. As shown in those three figures, the main reason for the increase in magnetoresistive ratio was due to a decrease in resistance of the films.

A film, annealed at 400°C, had no change in magnetoresistive ratio but there was a decrease in amount of magnetic material evidenced by decrease in saturation magnetization ( $M_s$ ). Also for this film, there was an increase in resistance. These two factors are evidences that there was severe oxidation in the film.

Increases in magnetoresistive ratio correlate well with increased annealing temperatures. For a given temperature,

however, the percentage increase of the magnetoresistive ratio drops off as annealing time is increased. It is believed that this is due to a corresponding decrease in the rate of grain growth. The larger grains would tend to increase the easy axis dispersion of the magnetic material thereby causing an increase in the coercivity ( $H_C$ ). This was observed in Table 2. It was noticed that the 200°C annealing process didn't cause a change in  $M_S$ . The interpretation of this is no oxidation of the magnetic layers took place. After annealing at 300°C,  $M_S$  didn't seem to change much either, indicating that oxidation was again very small.

TABLE 1. Double layer films annealed at 200°C

Middle layer (Å)	Annealing Time (Hour)	B <sup>a</sup> H <sub>C</sub> A <sup>b</sup> (Oe)	B H <sub>k</sub> A (Oe)	M <sub>S</sub> B A (Oe)
40	1	0.756	0.72	10 10 0.198 0.198
40	2	0.54	0.36	10 10 0.198 0.18
40	4	0.648	0.684	10 10 0.207 0.198
40	8	0.432	0.468	10 10 0.18 0.18
50	1	0.9	0.9	10 10 0.198 0.198
50	2	0.9	0.64	10 10 0.18 0.18
50	4	0.45	0.504	10 10 0.189 0.198
50	8	0.594	0.576	10 10 0.198 0.198
60	1	0.936	0.936	10 10 0.198 0.198
60	2	0.9	0.9	10 10 0.198 0.18
60	4	0.936	0.936	10 10 0.198 0.198
60	8	0.792	0.72	10 10 0.198 0.198

<sup>a</sup>B stands for before annealing.

<sup>b</sup>A stands for after annealing.

TABLE 2. Double layer films annealed at 300°C

Middle layer (Å)	Annealing Time (Hour)	B <sup>a</sup> H <sub>C</sub> A <sup>b</sup> (Oe)	B H <sub>k</sub> A (Oe)	B M <sub>S</sub> A (Oe)
40	1	0.792 0.792	10 10	0.171 0.171
40	2	0.484 0.648	10 10	0.189 0.18
40	4	0.432 1.224	10 10	0.18 0.18
40	8	0.594 0.828	10 10	0.198 0.162
50	1	0.504 0.72	9 9	0.198 0.198
50	2	0.648 0.72	10 10	0.18 0.18
50	4	0.648 0.828	9.3 9.6	0.144 0.144
50	8	0.576 0.72	11.3 11.3	0.198 0.216
60	1	0.792 0.9	9 9	0.189 0.189
60	2	0.864 1.08	11 11	0.198 0.216
60	4	0.9 0.9	9 11	0.18 0.18
60	8	0.792 1.36	11.3 12.3	0.216 0.234

<sup>a</sup>B stands for before annealing.

<sup>b</sup>A stands for after annealing.

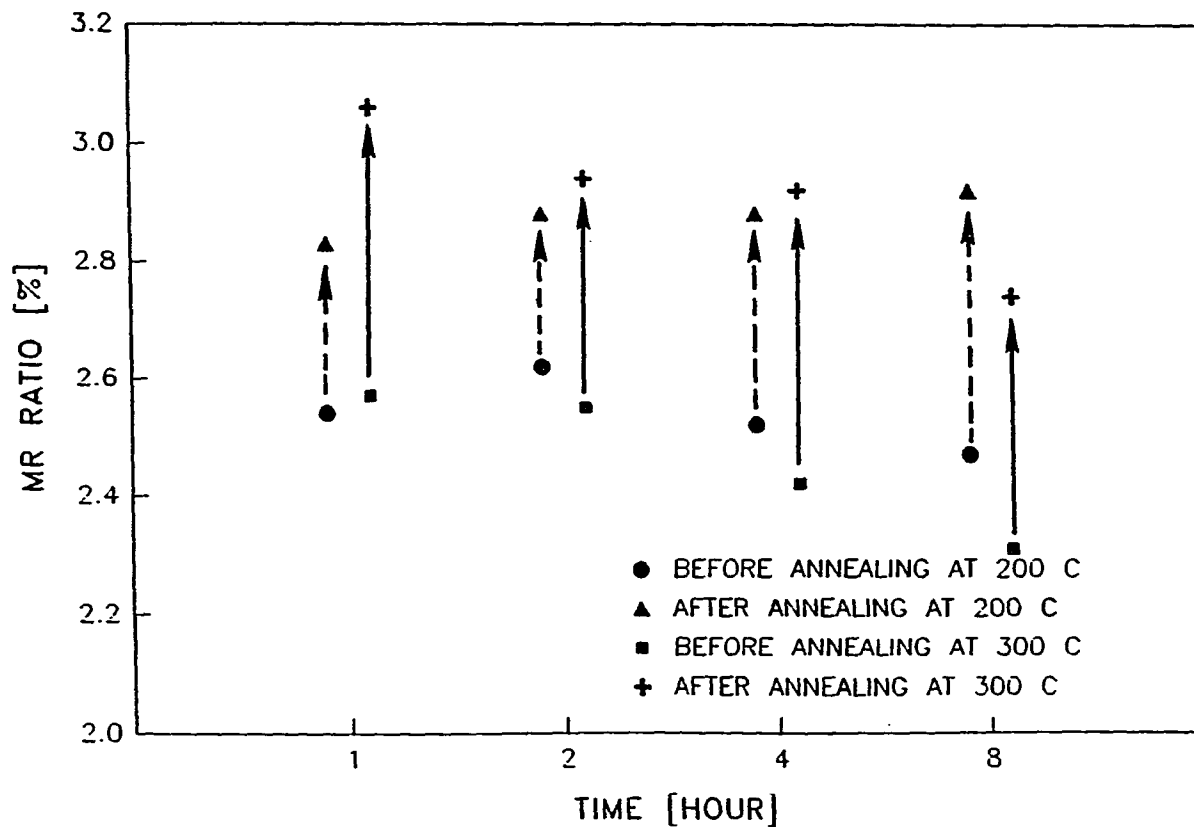


FIGURE 16. MR ratio vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 40 Angstroms thick

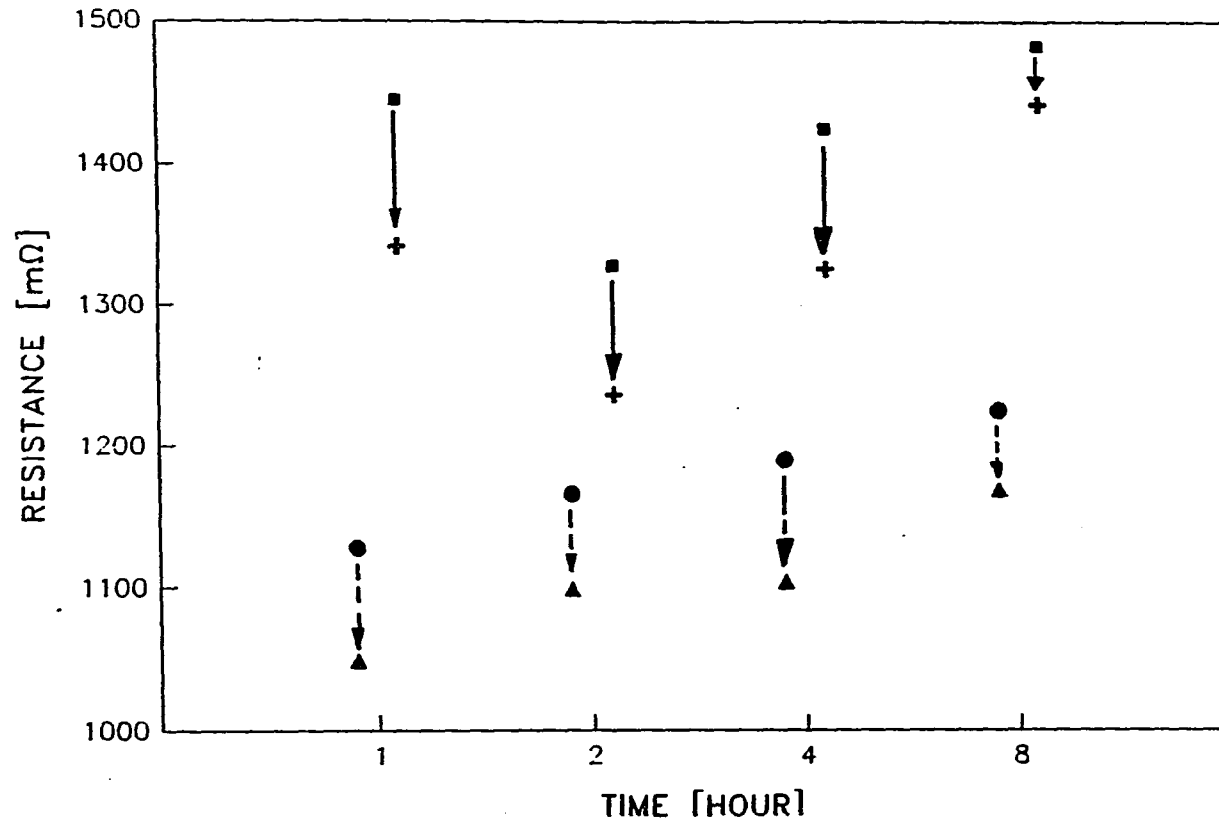


FIGURE 17. Resistance vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 40 Angstroms thick

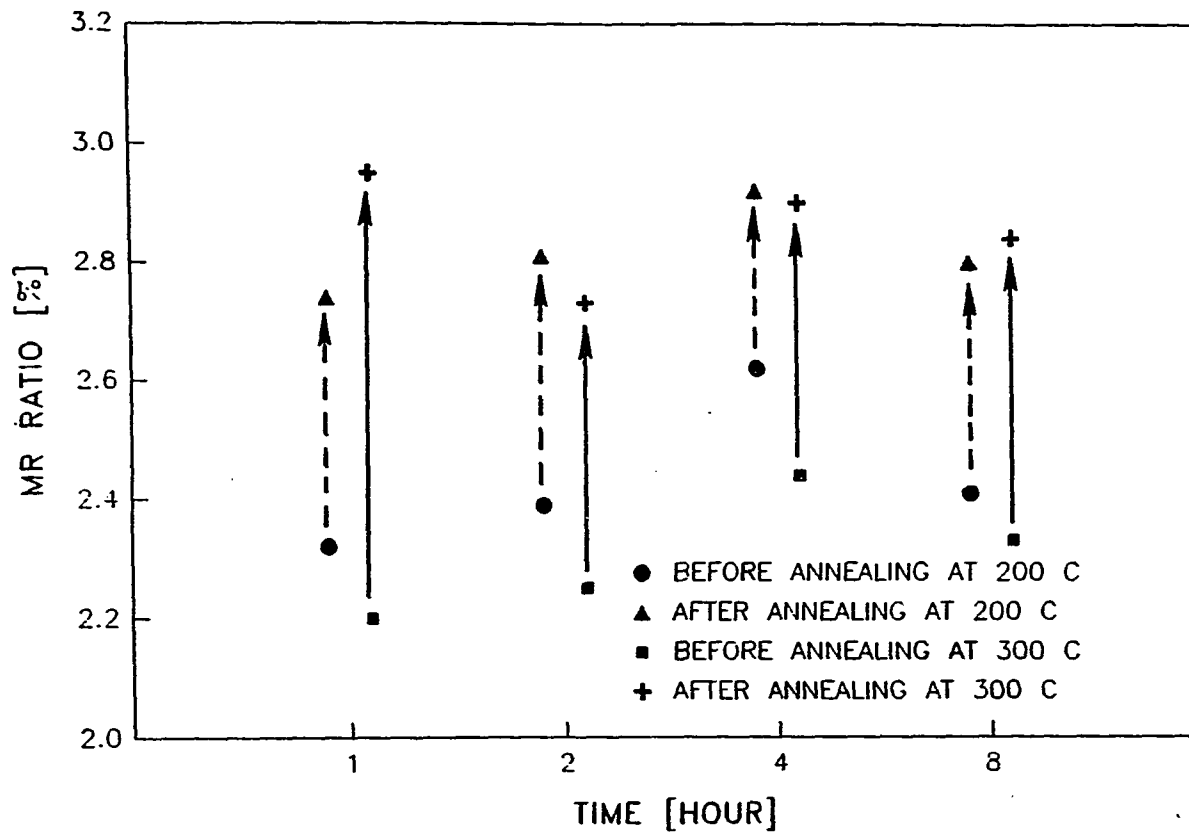


FIGURE 18. MR ratio vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 50 Angstroms thick

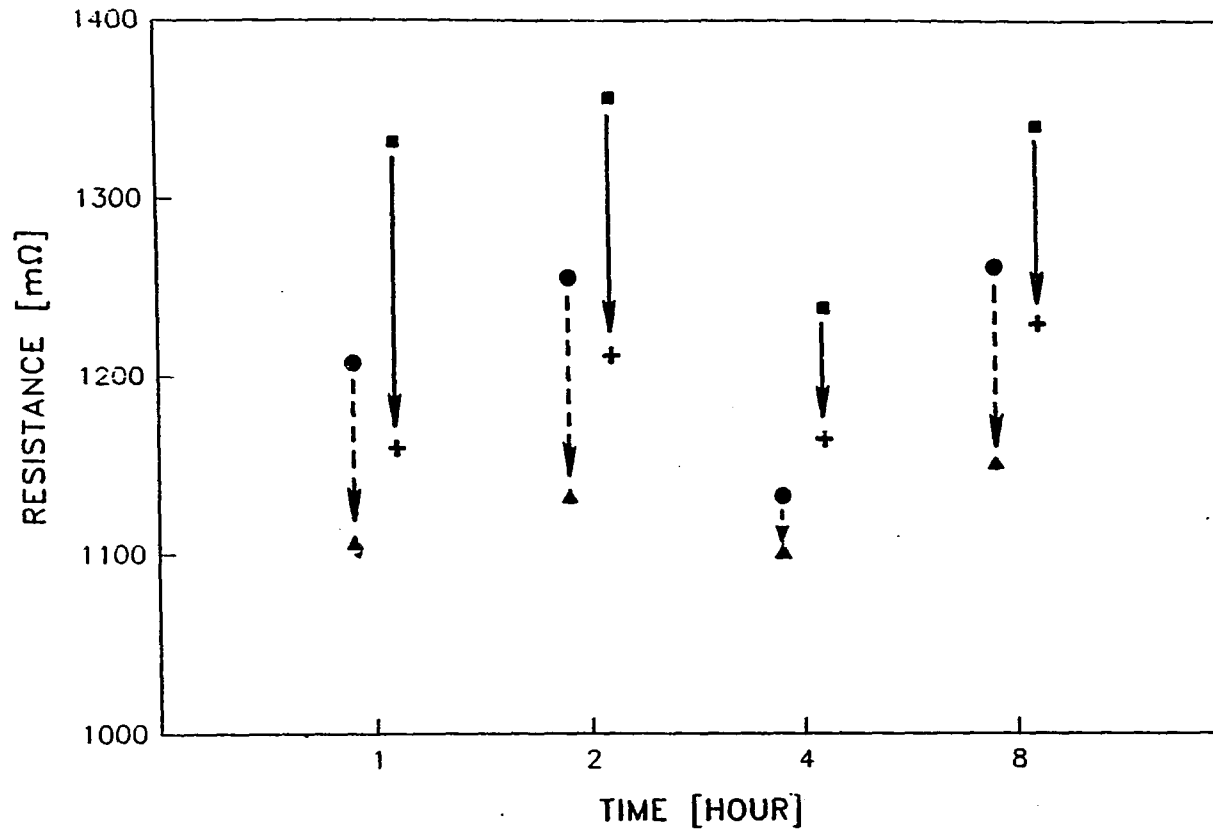


FIGURE 19. Resistance vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 50 Angstroms thick



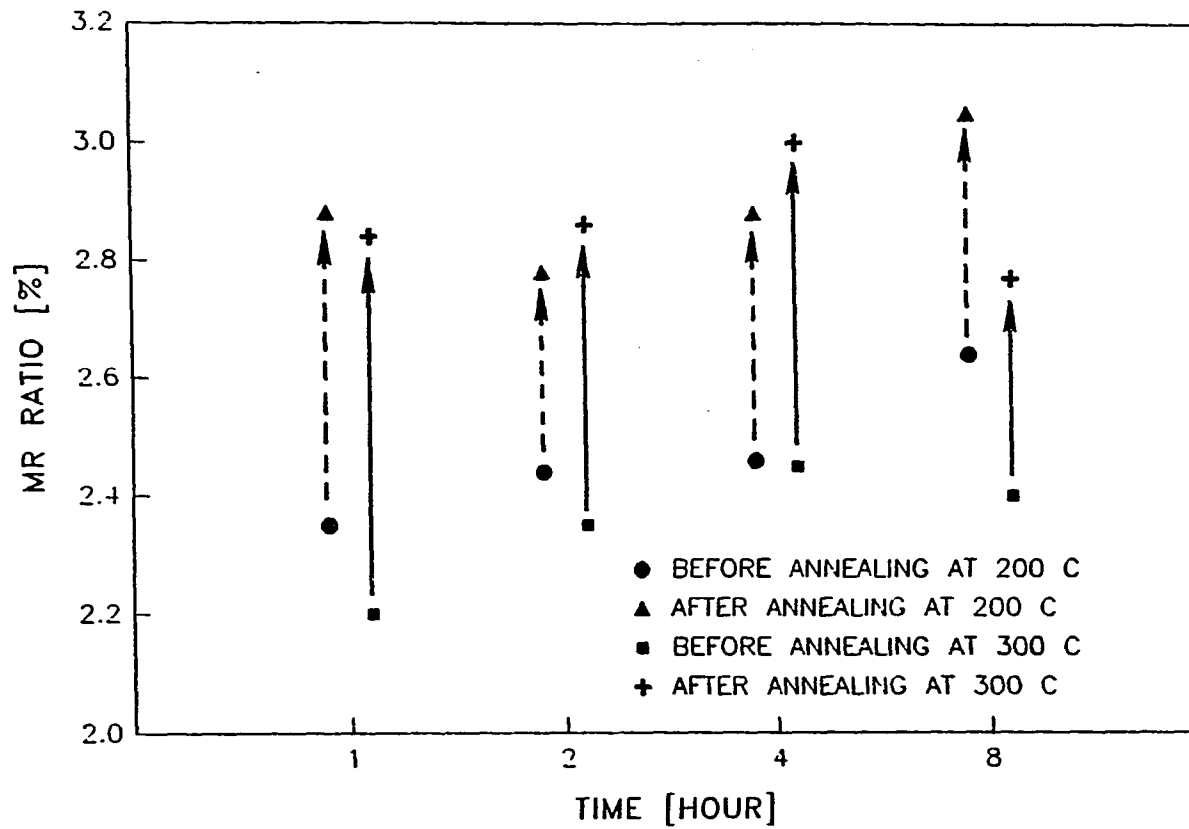


FIGURE 20. MR ratio vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 60 Angstroms thick

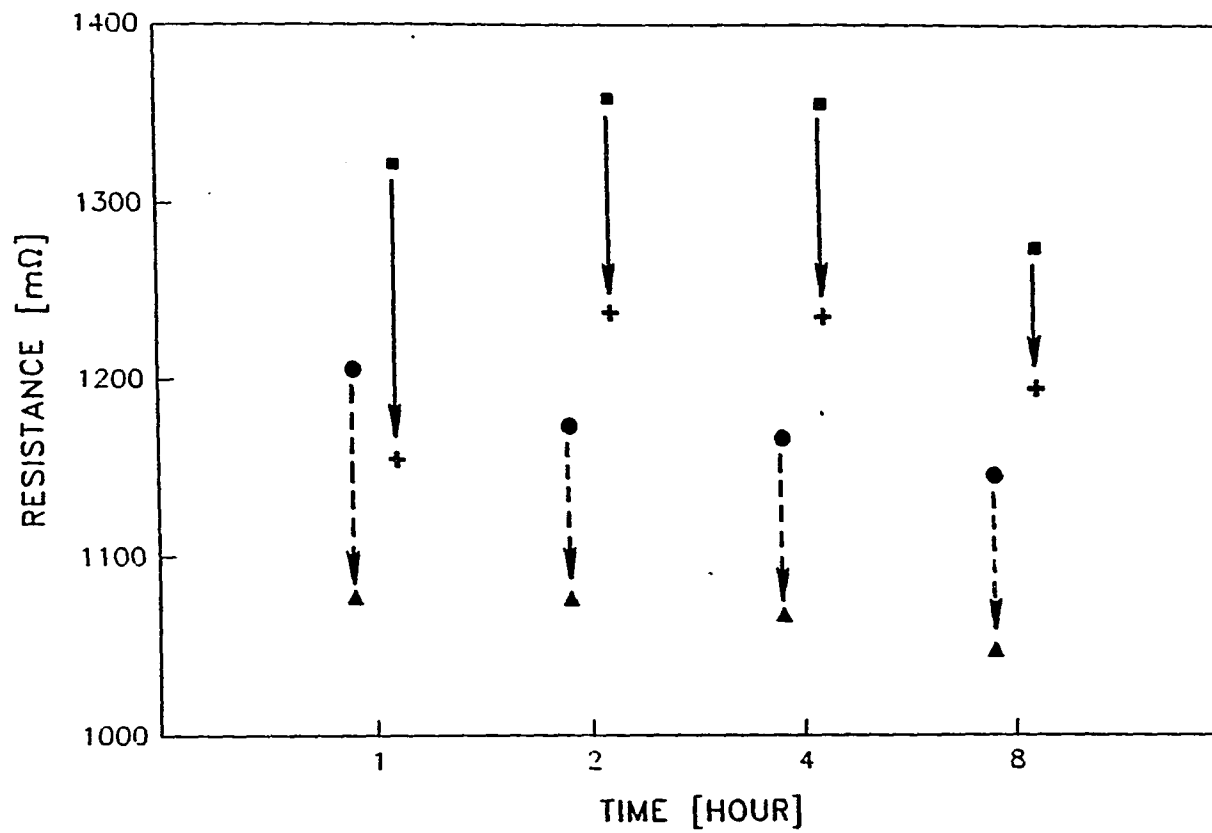


FIGURE 21. Resistance vs. annealing time. Magnetic layers are 250 Angstroms thick and Tantalum middle layer is 60 Angstroms thick

Grains of the films

Pictures of the grains were taken by SEM to investigate the correlation between grain size, resistivity, and magnetoresistive ratio. Figure 22 shows grains of a film not annealed.

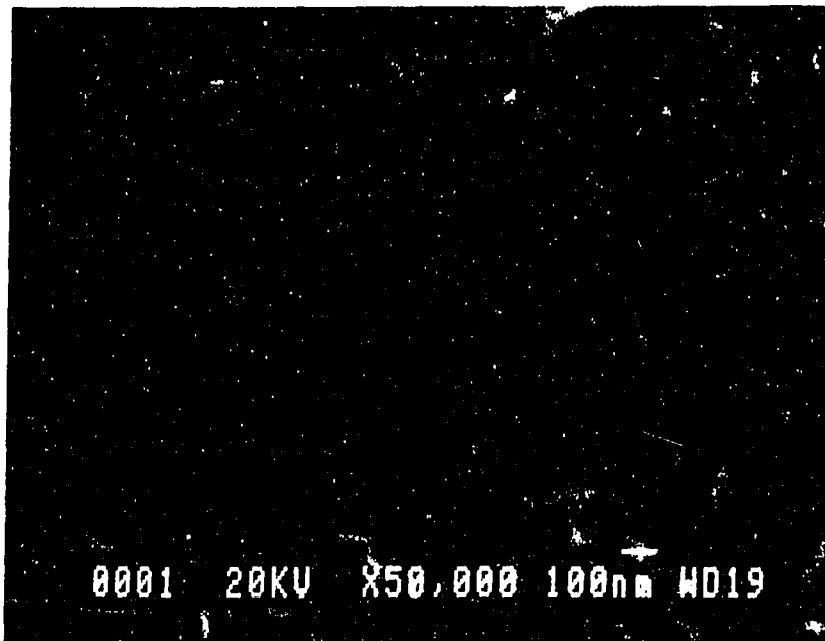


FIGURE 22. Grains of the top magnetic layer

Figures from 23 to 26 show grains of the films annealed at 200°C for different amount of time. Likewise, Figures from 27 to 30 were for 300°C.

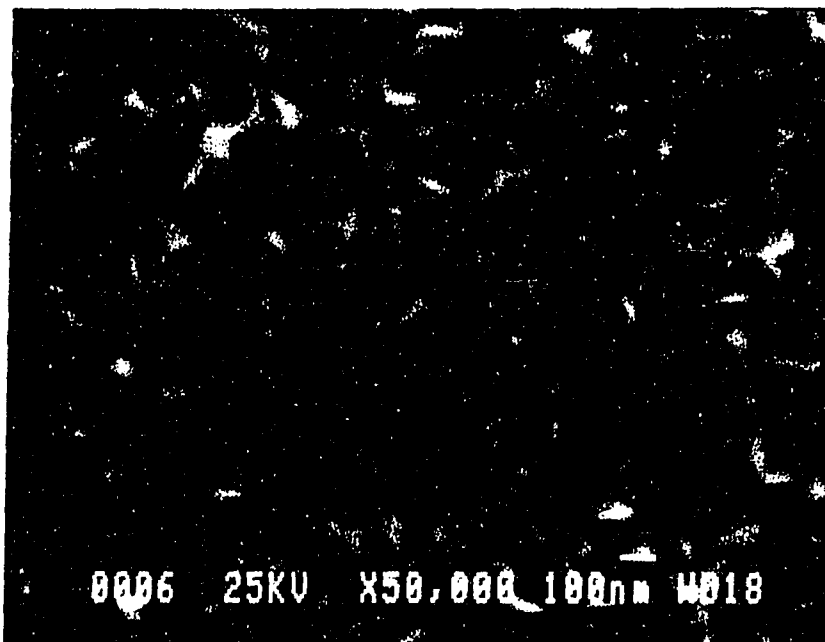


FIGURE 23. Grains of the top magnetic layer annealed at 200°C for 1 hour



FIGURE 24. Grains of the top magnetic layer annealed at 200°C for 2 hours

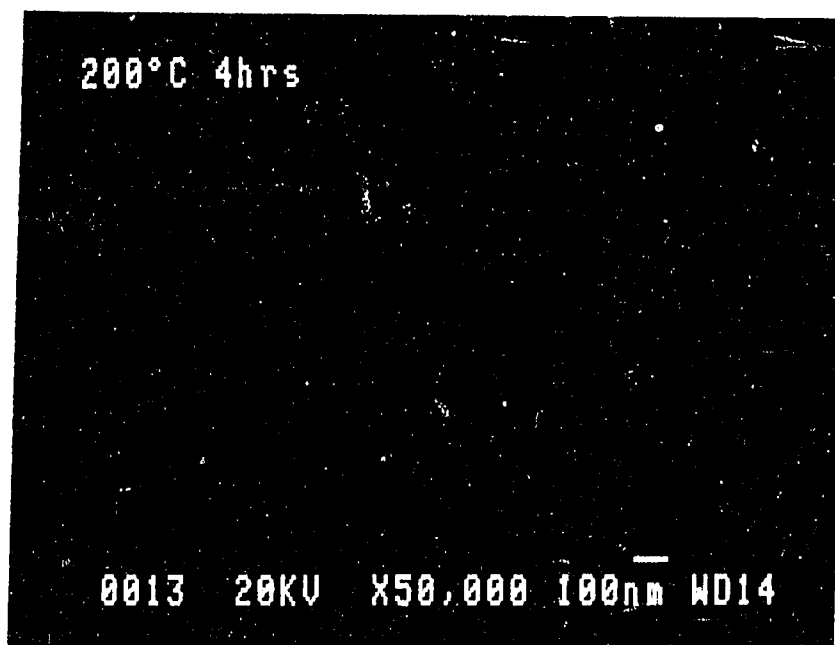


FIGURE 25. Grains of the top magnetic layer annealed at 200°C for 4 hours

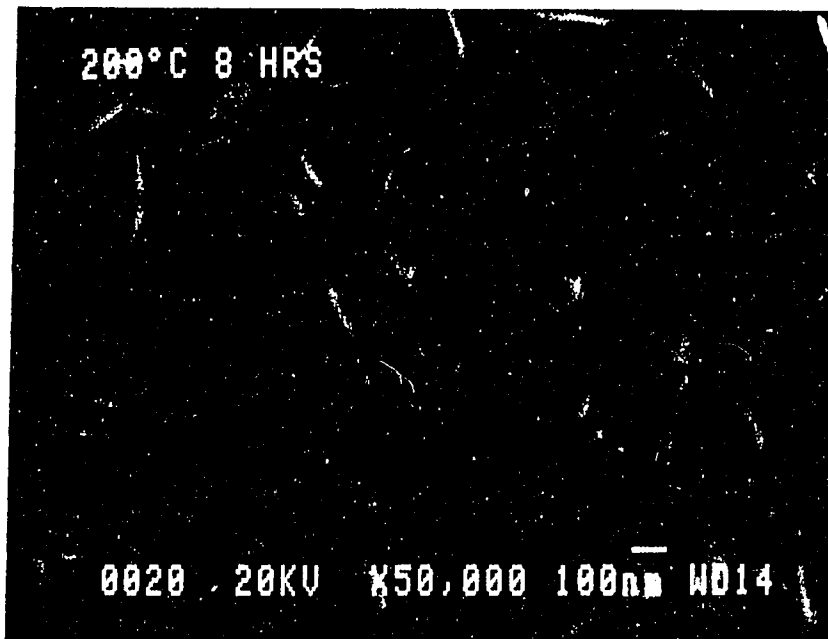


FIGURE 26. Grains of the top magnetic layer annealed at 200°C for 8 hours

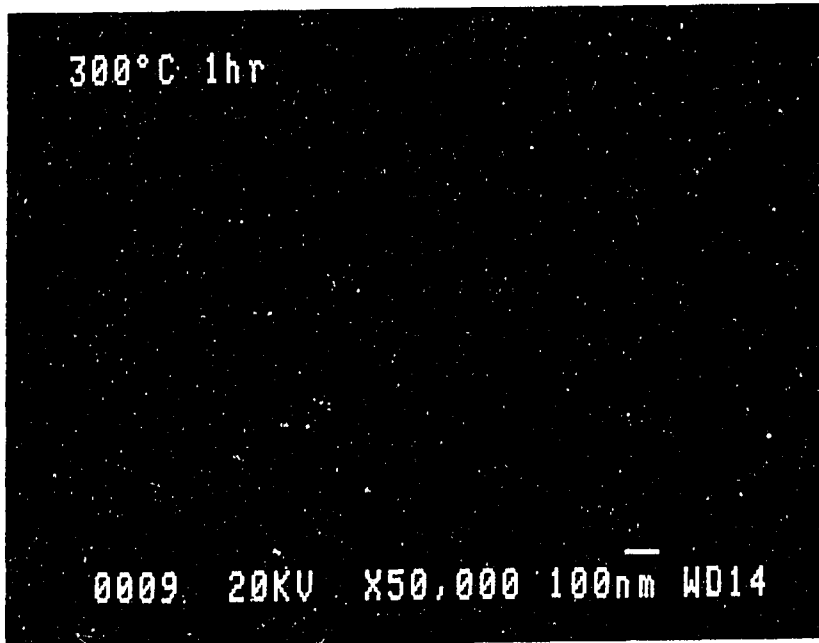


FIGURE 27. Grains of the top magnetic layer annealed at 300°C for 1 hour



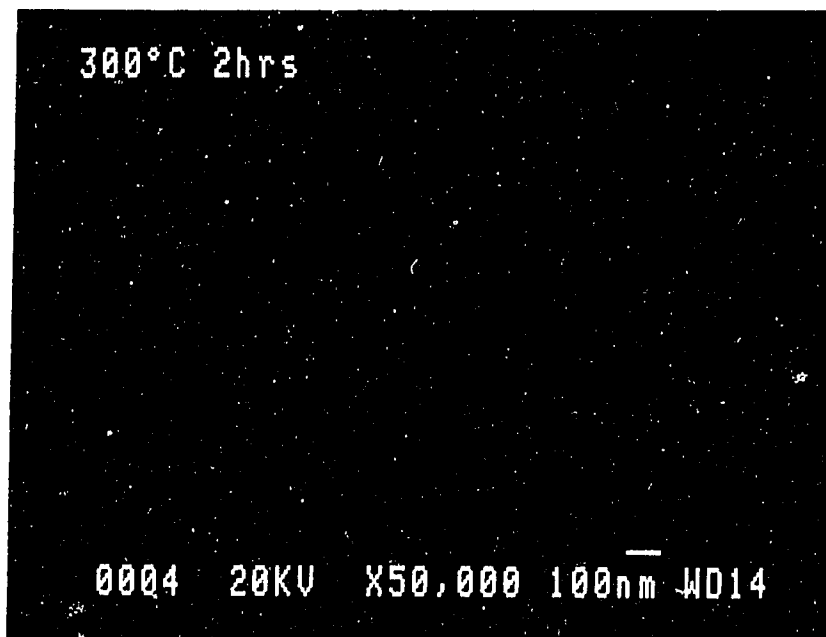


FIGURE 28. Grains of the top magnetic layer annealed at 300°C for 2 hours

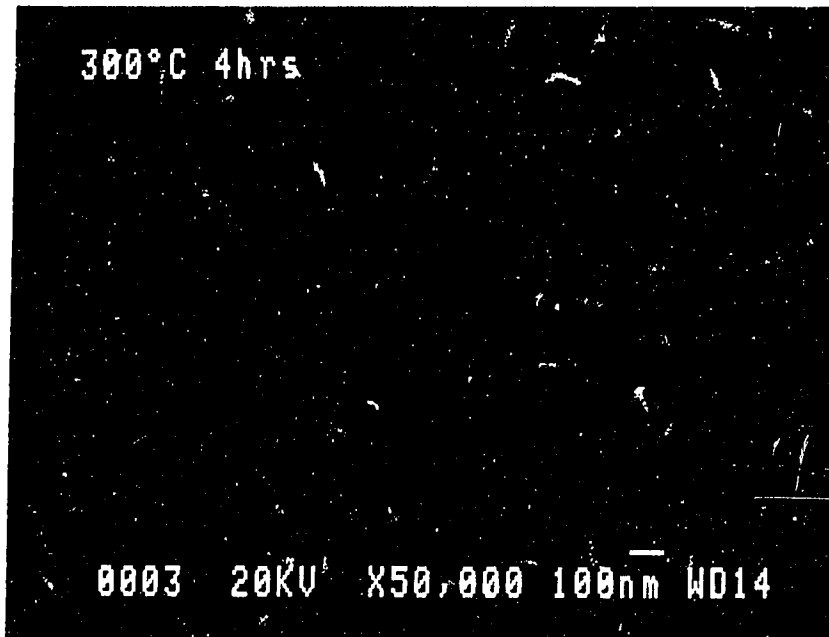


FIGURE 29. Grains of the top magnetic layer annealed at 300°C for 4 hours

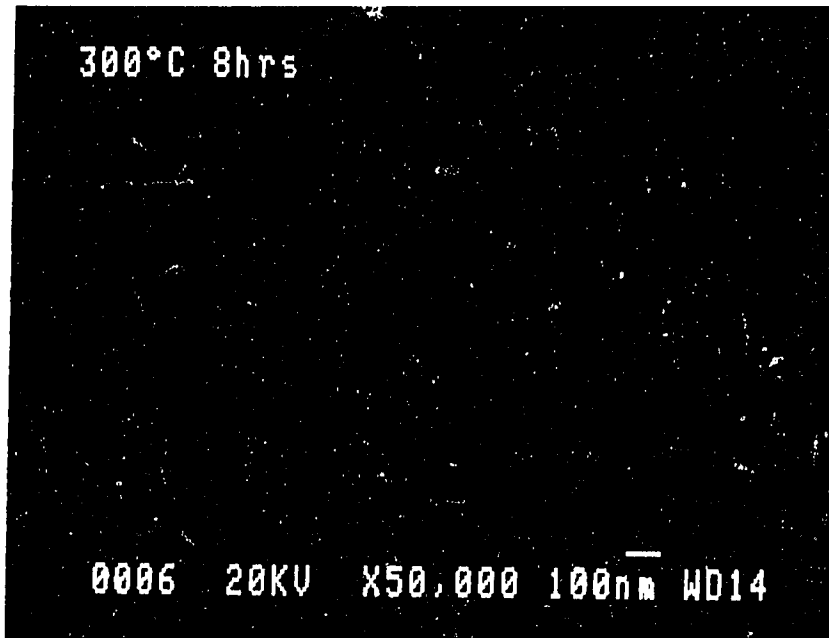


FIGURE 30. Grains of the top magnetic layer annealed at 300°C for 8 hours

### Films with Elevated Substrate Temperature

Films were deposited, after heating up the substrates to 200°C and 300°C, with 250 Angstroms of magnetic layers and different thickness of the middle layer. Tables 3 and 4 show data for substrate temperatures of 200°C and 300°C respectively.

TABLE 3. Data for films with 200°C substrate temperature

Middle layer (Å)	H <sub>C</sub> (Oe)	H <sub>k</sub> (Oe)	M <sub>S</sub> (Oe)	R (mOhm)	MR ratio (%)
40	0.72	9.6	0.396	1140	2.45
40	0.684	9.6	0.396	1085	2.58
40	0.72	9.6	0.396	1150	2.45
50	0.612	9.6	0.432	1125	2.40
50	0.684	9.6	0.396	1120	2.47
50	0.576	11.52	0.396	1126	2.38
60	0.792	10.8	0.432	1080	2.55
60	0.72	9.6	0.432	1120	2.50
60	0.72	9.6	0.414	1120	2.55

As shown in Tables 3 and 4, magnetoresistive ratios were increased by an average of 5 % at 200°C and 7.7 % at 300°C as compared to the films deposited on unheated wafers. The maximum magnetoresistive ratio was 2.7 % at 300°C substrate temperature.

TABLE 4. Data for films with 300°C substrate temperature

Middle layer (Å)	H <sub>c</sub> (Oe)	H <sub>k</sub> (Oe)	M <sub>s</sub> (Oe)	R (mOhm)	MR ratio (%)
40	0.648	9.6	0.396	1076	2.70
40	0.576	11.52	0.378	1090	2.55
40	0.612	9.6	0.396	1092	2.58
50	0.504	9.6	0.396	1120	2.48
50	0.756	11.52	0.414	1115	2.57
50	0.684	9.6	0.378	1110	2.45
60	0.648	9.6	0.396	1087	2.53
60	0.648	9.6	0.396	1160	2.50
60	0.702	9.6	0.396	1120	2.50

The resistance of the films with elevated substrate temperatures were almost the same as the films annealed after deposition. The magnetoresistive ratios of the films with elevated substrate temperature were not as high as expected. It is believed that the bottom Tantalum protection layer didn't prevent oxidation of the first magnetic layer from the silicon dioxide. It is believed that an increase of the grain size compensate for a decrease in magnetic material, which would explain the reason why the resistance didn't change as much as expected. Elevating the substrate temperature obviously didn't have as much of an effect as annealing the film after deposition on the magnetoresistive ratio.

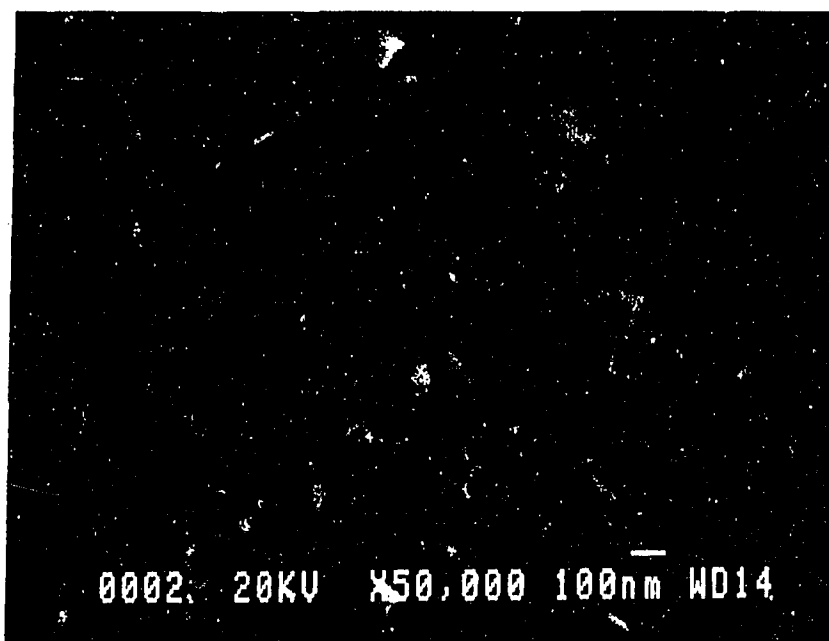


FIGURE 31. Grains of the top magnetic layer with substrate temperature at 200°C

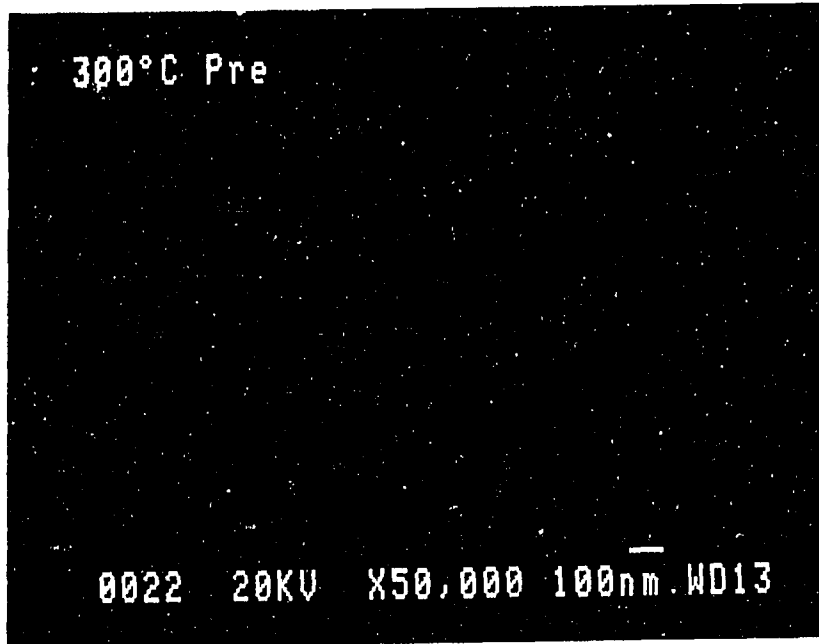


FIGURE 32. Grains of the top magnetic layer with substrate temperature at 300°C

## CONCLUSIONS AND FUTURE STUDIES

## Conclusions

An investigation correlating grain size, resistivity, and magnetoresistive ratio with annealing of NiFeCo magnetic multilayer thin films was performed. Magnetic properties of the films were improved through elevated substrate temperature during deposition and annealing after deposition.

It was observed that the magnetoresistive ratio increased by annealing after deposition to as high as 3 percent. The magnetoresistive ratio increased to 2.8 percent with elevated substrate temperature. Those ratios were desirable to obtain good signal levels for memory cells and transducers. No attempt was made to correct the resultant magnetoresistive ratios for current shunting effect by the Tantalum layers (particularly top and bottom protection layers). The actual magnetoresistive ratios of the NiFeCo material should be higher than what was measured. An increase in magnetoresistive ratio correlates well with a decrease in resistivity for both types of annealing. A decrease in resistivity correlates well with an increased grain size for the top magnetic layer of the film. Only the top layer was studied by scanning electron microscopy



because of the difficulty of etching a single layer without damaging the rest of the film.

In conclusion, the improvement of the magnetoresistive ratio of the NiFeCo enables the multilayer thin film structure to become an excellent material for magnetic memory cells and transducers.

#### Future Studies

Additional studies are needed to improve and characterize the magnetic properties of the multilayer films as follows:

- Post annealing of the films which were deposited with elevated substrate temperature
- Higher deposition rate by RF sputtering
- Compositional variation of the NiFeCo to increase the magnetoresistive ratio
- Reactive sputtering with Tantalum target to have more stable protection layers such as Tantalum nitride
- Grain size measurement by Transmission Electron Microscopy

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