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On the Magnetic Properties of Sputtered NiFe Films

Abstract: Reproducibility and control of magnetic properties have been significant problems in the preparation of Permalloy films by cathodic sputtering. The data presented in this paper illustrate the reduced sensitivity to gaseous impurities that can be achieved by applying a suitable negative bias to an 81% Ni – 19% Fe Permalloy film that is being deposited by dc sputtering. Results are shown which compare the magnetic properties as a function of deposition rate achieved with and without a bias potential applied to the film during deposition. Data are also presented showing the effect of film bias on magnetic properties when known concentrations of oxygen and nitrogen are present in the atmosphere during deposition. The evidence shows that the effect of the bias is to reduce the quantity of system atmosphere impurity included in the film. The bias technique has permitted, with good reproducibility, accurate experimental determination of the effect of such control parameters as substrate temperature on magnetic properties. (Wall motion threshold H_0, anisotropy field H_{k0}, and dispersion α_{90} are plotted as a function of substrate temperature.) In addition, data are shown which demonstrate the compositional control and simplicity of film thickness and deposition rate control which are inherent in the sputtering process.

Introduction

Thin magnetic films of Permalloy were proposed for memory application several years ago.[1–3] Since that time there have been continuing changes and more rigorous tolerances established in the magnetic properties needed to implement such a use. In this respect, films have been characterized in low frequency measurements, not only by values of anisotropy field H_{k0} and wall coercive force H_c, but also by skew β, dispersion of the easy axis of magnetization α_{90}, and amplitude dispersion of H_k. The full characterization, H_c, H_k, β, α_{90}, is of interest in quickly estimating creep and disturb sensitivity, and memory drive requirements.

Vacuum evaporation,[1] electroplating[4] and, most recently, sputtering[5] have been used for the preparation of thin magnetic films. Each of these methods has certain advantages and, likewise, certain disadvantages. Some advantages of conventional glow-discharge sputtering include: (1) compositional uniformity; i.e., when deposited under properly controlled conditions,[6] the composition of the deposited film is the same as that of the starting cathode material; (2) absence of angle-of-incidence effects, since glow discharge sputtering is a diffusion process; and (3) simplicity of film thickness and deposition rate control inherent in the process. The difficulties associated with sputtering of magnetic film elements are: (1) problems of substrate temperature control which will affect reproducibility; (2) magnetic field distortion due to the unavoidable presence of the ferromagnetic cathode; and (3) impurity sensitivity–particularly, sensitivity to water vapor and oxygen.

This paper presents experimental results which show that the principal problem associated with the preparation of thin ferromagnetic films by cathodic sputtering, i.e., sensitivity to background impurity, can be overcome by use of the technique of bias sputtering. With bias sputtering, the difficulty in depositing 81% Ni – 19% Fe films with reproducible magnetic parameters, previously ascribed to substrate temperature control problems, has not been evidenced. Use of a "thin" foil cathode has previously been shown[7] to be a simple method for minimizing the distorting effect of the cathode on the orienting magnetic field at the substrate.

Also in this paper is experimental evidence further verifying the advantages of compositional uniformity and simplicity of film thickness and deposition rate control.

Experimental procedure

• *Deposition*

The present studies were made with vacuum-melted and rolled, 12.5-cm square 81% Ni – 19% Fe Permalloy sheets of 25 mm thickness as cathodes, using a heat sink cathode assembly in a conventional 45 × 75-cm glass bell jar system. A schematic illustrating the essential features of the sputtering arrangement is shown in Fig. 1. A liquid nitrogen (Meissner) trap was employed in the bell jar. All associated parts of the vacuum system were constructed from nonmagnetic materials. The anode assembly was initially a water-cooled block; a later modification allowed heating of the substrate. Metallographically-polished 5-cm squares of silver-copper with approximately 2μ of vacuum-deposited SiO were used as substrates for depositions where magnetic parameters alone were determined; glass substrates were used when thickness and composition analyses were required.

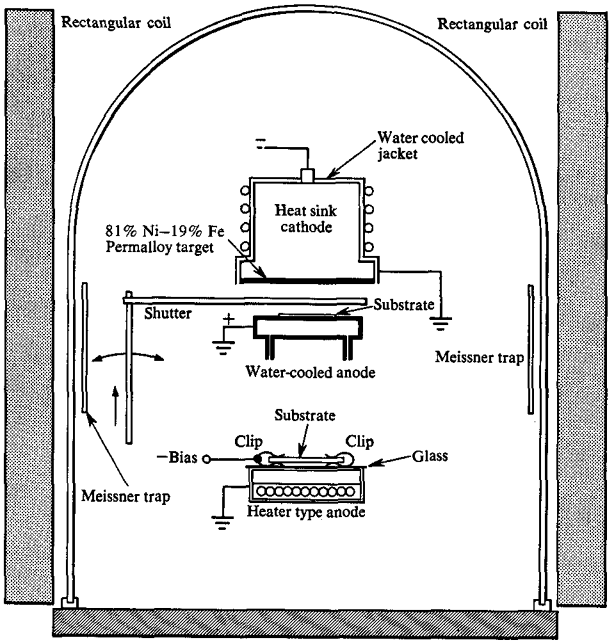

Figure 1 Schematic of sputtering system.

The substrate was placed on the anode surface under the center portion of the cathode at a spacing of 2.5 cm, and the system was evacuated to less than 1×10^{-6} Torr, after which argon was admitted to the desired pressure, usually 0.1 Torr (McLeod gage pressure) with the diffusion pump operating. The substrates were not clamped to the anode, since clamping could introduce stress in the magnetic film and cause variation in the magnetic properties throughout the films. The system's capabilities enabled other gases to be mixed with argon by means of a flow meter system. In addition, a bias potential could be applied to the film being formed through a clip contacting the substrate (Fig. 1).

With a shutter in place over the substrate, the preconditioned cathode (see section on cathode preparation) was pre-sputtered at a potential of 3000V and a current of 110mA for a controlled period of time, in this instance 18 minutes. This permitted removal of contaminants from the cathode surface in addition to providing energy for preheating of the substrate. With the water-cooled anode assembly, the presputtering provided the only source of heat to the substrate. With the later heater-type anode assembly, the presputtering operation was followed by a 30-minute, constant-temperature bakeout without glow discharge. The substrate temperature was continuously monitored by means of a chromel-alumel thermocouple embedded in the side of the actual substrate.

Immediately after completion of the presputtering operation or the baking period, the cathode voltage and current were rapidly adjusted to their operating values for the particular run in progress, and the shutter was removed to initiate deposition. The cathode potentials used were between 1000V and 4000V and the currents were between 40 mA and 270 mA; deposition times of 30 to 600 seconds, correlating well with sputtering power, were found to deposit films of approximately 800Å. Total time for a complete deposition cycle was approximately 90 minutes.

A uniform dc magnetic field of about 25 Oe was applied in the substrate plane during deposition. The magnetic field was generated by a pair of rectangular coils which, in the absence of ferromagnetic material, were designed to give a uniform field having a maximum deviation of $\pm 0.1\%$ over the substrate area.

- *Cathode preparation*

After installation of a new 81% Ni − 19% Fe Permalloy cathode in the sputtering apparatus, a cathode precleaning operation was necessary before films could be deposited whose composition corresponded to that of the bulk cathode and whose magnetic properties on glass substrates were typical for the 25-mm cathodes used, i.e., $H_0 = 2.0 - 2.5$ Oe, $H_{k0} = 3.0 - 3.5$ Oe, $\alpha_{90} \leq 2°$, $\beta \leq \pm 2°$. The operation was performed by sputtering the cathode at a potential of 3000V and a current of 110mA for a total of approximately 3 hours. The precleaning served to remove the surface layers which were found to have compositional variations to a depth of approximately 10^5Å on a sheet of 25-mm thickness. (Compositional variations throughout the thickness were experimentally determined at various stages by means of x-ray fluorescence analysis.) It also served to remove the surface oxide layers.

- *Measurements*

Quasi-static magnetic measurements of wall motion threshold H_0 (H_0 is defined as the field necessary to initiate hysteresis in the easy-direction B–H loop), anisotropy field H_{k0}, dispersion of the easy axis of magnetization α_{90}, and skew of the easy axis of magnetization β were made with a 60-cycle Kerr-effect loop tracer having a light-spot dimension of less than 2 mm in diameter. Measurements were taken at the center and four edges of each specimen, a method which has been found to adequately indicate any magnetic uniformity in thin ferro-magnetic films deposited by sputtering, evaporation, or electroplating.

Anisotropy field H_{k0} was measured from the cross-field loop M_x vs. H_y and its approximation to a circle as originally described by Kump.[8] The cross-field loop represents the easy-direction magnetization of a film when the film is driven with a sinusoidally-varying field in the hard direction and, in this case, with an alternating-polarity square pulse applied in the easy direction. The easy-direction field was applied during the time $\pi/2$ to π of the hard-direction drive to reset the film into a saturated easy-axis state prior to measurement time. Dispersion of the easy axis α_{90} was determined by the well-known Crowther technique.[9]

Easy-axis skew β was found by noting the angle between the intended and actual film hard axis. The actual hard axis was determined by observing the direction of minimum net easy-axis magnetization in the cross-field loop when no easy-axis pulse field is applied. The $\Delta\beta$ values result from taking the greatest angular difference between any of the five measured spots.

Magnetostriction was determined by measuring the change in anisotropy field introduced on subjecting the film and substrate to a bending stress in the Kerr apparatus. Experimentally, ΔH_{k0} was taken as the change in anisotropy field produced by the application of a strain of 2.2×10^{-4} along the easy axis of the film; the above figure of elastic strain induced in the film was experimentally determined by strain gage measurements. The weight percent compositional difference (ΔC) from the zero magnetostriction value (assumed to be 19.1% Fe) was determined from the expression for the strain anisotropy field,[10] $H_{ks} = Ae\Delta C$ where e is the strain and A is a material constant having a value of 10.8×10^3 Oe/percent change in composition.

Composition and thickness determinations, assuming bulk density, were made by x-ray fluorescence measurements on glass substrates, the precision of which was ± 15Å in thickness and $< \pm 0.3\%$ by weight in composition. When the film was deposited onto glass substrates, no bias potential was applied.

Experimental results

- *Film composition control*

In the preparation of films for computer application it is desirable to use a material that is insensitive to stress introduced during packaging or handling. This requires materials having zero magnetostriction. Similarly, low magnetocrystalline anisotropy is desirable, to minimize grain orientation effects.[11] Ideally, then, a material having both zero magnetostriction and zero magnetocrystalline anisotropy, with an easily varied uniaxial anisotropy, is desired. Normally, all these parameters possess varying degrees of dependence on film composition, requiring a compromise to achieve usable films. For the Permalloy system these considerations have led investigators to concentrate on the 81% Ni − 19% Fe alloy, which has zero magnetostriction and low magnetocrystalline anisotropy,[12] i.e., $\lambda_s = 0$; $K_1 = 10 \times 10^3$ ergs/cm.

X-ray fluorescence analysis

Because of the importance of film composition control, a study of the effect of sputtering parameters and film thickness on composition was conducted. The results are shown in Figs. 2, 3, 4, and 5. Each point in the figures represents a discrete experiment, the lines being determined by least-squares analysis.

Figures 2 and 3 show the dependence of film composition

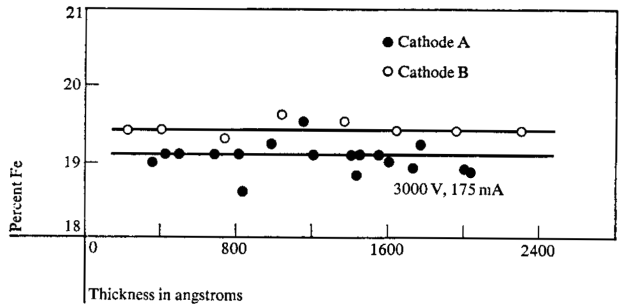

Figure 2 Composition vs. film thickness for films sputtered at constant cathode potential and current.

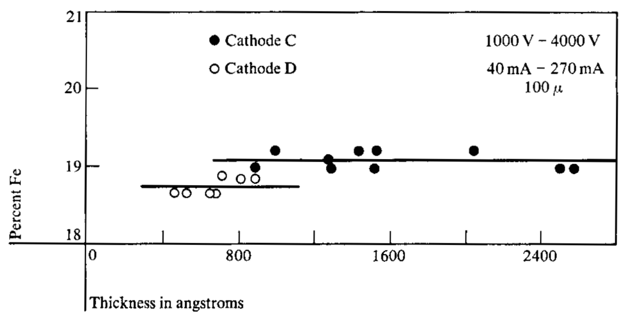

Figure 3 Composition vs. thickness for films sputtered at different cathode potentials (at constant pressure).

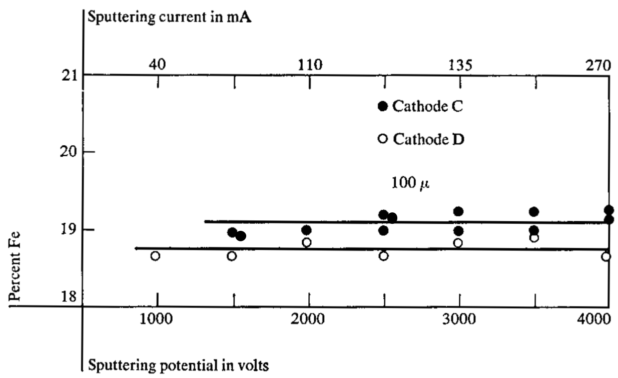

Figure 4 Composition vs. sputtering potential (constant pressure).

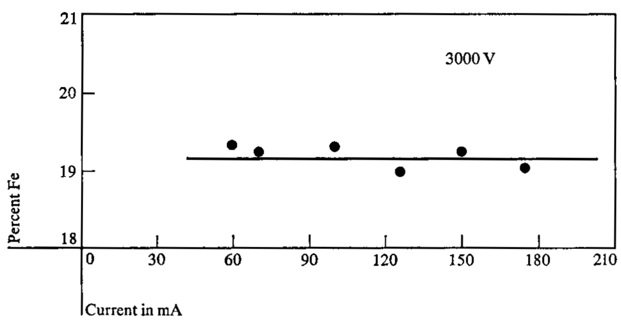

Figure 5 Composition dependence on current (pressure).

on thickness. Figure 2 shows dependence at constant voltage, current density and pressure with time as the variable and Fig. 3 shows dependence under a number of different sputtering conditions. Figures 4 and 5 show film composition as a function of cathode potential (at constant pressure) and cathode current density (at constant voltage), respectively.

Statistical analysis of the data finds no compositional dependence on the cathode potential, the cathode current, or the thickness of film deposited, the resulting film composition being dependent only on the cathode from which the film was sputtered. The slight compositional difference seen in films prepared from different cathodes is to be expected, since the vendor claimed a $\pm 0.3\%$ variance from the nominal composition of 81% Ni − 19% Fe throughout the melt from which the cathodes were rolled.

Magnetostriction measurements

The results of the x-ray fluorescence analysis show that the major constituent composition, i.e., NiFe content, of sputtered 81% Ni − 19% Fe Permalloy films is determined only by the composition of the starting cathode. The fluorescence method of compositional analysis, however, cannot distinguish between the presence of Fe or Ni in element form and the same materials in the form of minor nonmetallic constituents, such as oxides, nitrides, etc., making it possible to have an effective magnetic composition that is different from the analytical composition. This effective composition, relative to the non-magnetostrictive composition, was determined by measuring the change in H_{k0} produced by the application of a known strain along the easy axis of the film.[10]

Magnetostriction measurements were made on films both with and without a bias potential applied during deposition;[13] the results are shown in Table 1. From these results it can be seen that (1) the films prepared without bias exhibit an effective magnetic composition that is iron rich, relative to the composition of 19.1% Fe determined by fluorescence analysis on glass substrates deposited under identical conditions; (2) the films prepared with bias exhibit essentially the same effective magnetic and assumed chemical composition of 19.1% Fe; and (3) there does not appear to be any effect of initial substrate temperature or sputtering voltage on the magnetostrictive behavior of films with and without bias applied during deposition.

Table 1 Magnetostriction measurements on films deposited (a) without and (b) with a bias potential applied to the film during deposition

Initial substrate temperature	*Sputtering potential,* volts	H_{ko} *(unstrained),* Oe	H_{ko}, Oe	*Effective composition,* % Fe
		(a) No bias		
325°C	1500	3.4	+1.2	19.6
325°C	1500	3.6	+1.0	19.5
325°C	3000	3.0	+1.0	19.5
325°C	3000	2.9	+2.0	19.9
400°C	1500	2.8	+1.5	19.7
400°C	1500	3.2	+1.2	19.6
		(b) Negative bias (− 100V)		
325°C	2000	3.1	−0.2	19.0
325°C	1500	3.6	0	19.1
325°C	3000	2.1	0	19.1
325°C	3000	2.4	−0.8	18.8
350°C	2000	2.6	+0.2	19.2
360°C	2000	2.9	−0.6	18.9
370°C	2000	2.6	+0.3	19.2
370°C	2000	2.3	−0.1	19.1
400°C	2000	2.6	0	19.1
405°C	2000	2.2	+0.1	19.1
415°C	2000	2.4	−0.2	19.0
415°C	2000	2.7	−0.5	18.9

• *Thickness control*

The foremost effect of film thickness is on the demagnetizing field in discrete film storage elements. Thickness also has a strong influence on domain wall structure which influences the creep and disturb sensitivity of the magnetic elements.[14,15] Because of these effects, it is important that the thickness of the deposited films be controlled as accurately as possible. In films prepared by vacuum evaporation, elaborate and costly rate monitors are necessary. Inherent in the sputtering process, however, is simplicity in the control of film thickness and deposition rate. This is demonstrated in Fig. 6, which is a plot of deposition rate vs. time of deposition. Data points in the figure were obtained by depositing a series of films under constant sputtering conditions for varying periods of time. Film thicknesses were then determined by x-ray fluorescence analysis, and the deposition rate was calculated. The line, determined by the least-squares method, corresponds to a deposition rate of 16 ± 0.7 Å/sec.

• *Impurity sensitivity − bias sputtering*

One of the principal difficulties present in the preparation of thin ferromagnetic films is the tendency to trap and retain large amounts of impurities during deposition. This is a problem usually more severe in sputtering than in evaporation, for two reasons. First, the presence of reactive gases

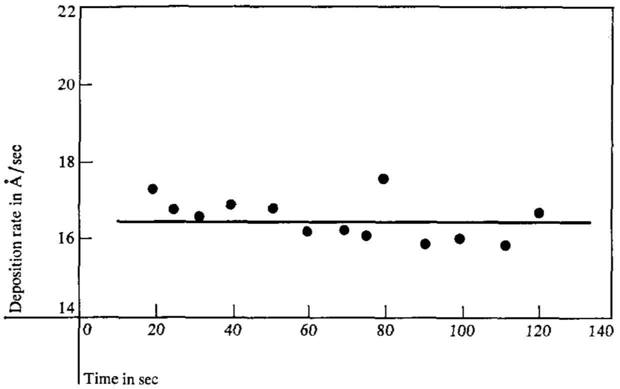

Figure 6 Deposition rate vs. deposition time.

produced by the bombardment of fixtures and inner surfaces of the vacuum system by high energy ions of the sputtering gas will not normally be detected because of the masking effect of the primary sputtering gas. Second, the ionized species present during sputtering are generally much more reactive than their neutral counterparts present during evaporation. The difficulty encountered in reproducibly depositing uniform ferromagnetic films by sputtering, using conventional vacuum systems and techniques, is evidence of this problem.

Recently, it has been shown by Maissel and Schaible[13] that the application of a suitable negative bias to a film during deposition by dc sputtering results in films of higher purity than those deposited without bias. These results were obtained on films of tantalum, niobium, aluminum, molybdenum and tungsten. The action of the bias was shown to be the preferential removal of absorbed impurities due to steady ion bombardment of the film throughout its growth. Thus, the application of a suitable negative potential to films of Permalloy being deposited by dc sputtering would be expected to minimize the sensitivity to impurities, resulting in more uniform and reproducible films.

Table 2 presents data comparing the magnetic properties obtained from two sets of films that were sputtered under identical conditions except that, in one case, the film was at anode potential, whereas in the other it was at a potential of 100 volts negative relative to the anode. The films were sputtered at different rates onto substrates with the initial substrate temperature maintained constant at approximately 275°C using the water-cooled anode; the temperature to which the substrate rose during deposition was a function of the deposition rate and time, as has previously been shown by Francombe and Noreika.[16] The sputtering rate was varied by changing the sputtering potential (and current) at a given pressure. Thickness was held constant by adjusting the sputtering time in accordance with the rates of sputtering at the different potentials.

It was expected that the films sputtered at lower potentials (lower sputtering rates) would contain more entrapped impurities resulting in higher values of the magnetic parameters. Note that all magnetic parameters increased with decrease in cathode potential when no bias was used, as anticipated, but when a negative bias was applied, little change was observed. It can be seen that if a bias is used, the properties of the films become essentially independent of the deposition rate, the effect of the bias on the magnetic properties at the lower potentials being very marked. In addition, the uniformity of magnetic properties over the substrate area was markedly superior for the films deposited under bias, especially at the lower potentials. The decrease in H_{k0} observed on the bias sputtered films with increasing rate of deposition (sputtering potential) can be attributed to the higher substrate temperature that results.

It should be noted that no systematic study of film properties as a function of bias voltage was performed on 81% Ni — 19% Fe films. Such a study, however, has been reported[17] on manganese containing Permalloy, where it was found that there is a minimum film bias potential at which uniform films exhibiting low dispersion of the easy axis of magnetization are obtained.

Table 2 Magnetic properties as a function of cathode potential (a) without and (b) with a bias potential applied to the film during deposition

Sputtering		*Rate of*	*No bias*				*Negative bias (−100V)*			
Potential, volts	*Current,* mA	*deposition,* Å/sec	H_0, Oe	H_{k0}, Oe	α_{90}, deg	$\Delta\beta$, deg	H_0, Oe	H_{k0}, Oe	α_{90}, deg	$\Delta\beta$, deg
4000	250	27.0	1.9	3.1	<1	2.0	2.3	2.5	4	6.2
3000	175	16.5	2.5	3.9	1	2.5	1.5	3.3	<1	2.8
2500	130	12.0	3.3	4.3	5	3.6	2.3	3.9	1	2.2
2000	110	7.5	6.8	6.0	18	3.9	2.0	3.6	2	3.3
1500	70	4.0	16.0	—	—	—	1.5	4.3	<1	1.3

Table 3 Magnetic properties of films deposited with known amounts of oxygen and nitrogen present in the sputtering gas.

	No bias				*Bias (−100V)*			
Impurity, molecular %	H_0, Oe	H_{k0}, Oe	α_{90}, deg	$\Delta\beta$, deg	H_0, Oe	H_{k0}, Oe	α_{90}, deg	$\Delta\beta$, deg
O_2								
0.0	2.1	2.5	2	4	—	—	—	—
0.25		(brownish deposit)			2.6	3.6	3	3.2
1.0		(brownish deposit)				(brownish deposit)		
N_2								
0.0	2.1	2.5	2	4	—	—	—	—
0.25	—	—	—	—	1.6	2.9	2	2.5
0.40	—	—	—	—	6.0*	3.2	10	—
0.40	—	—	—	—	7.0*	4.5	15	—
1.0	>15				>15			
1.0	—	—	—	—	>15			
					Bias (−200V)			
0.40	—	—	—	—	2.2	3.2	2.5	6

* Discolored deposit, extremely nonuniform over substrate area.

• *Effectiveness of bias against impurities in the sputtering gas*

Further experiments were performed in which the effect of bias on magnetic films sputtered in atmospheres containing known traces of impurities was examined. Films were sputtered in argon containing varying amounts of oxygen and nitrogen, both with and without potential applied to the film during deposition. The effect of oxygen (or water vapor) was of particular interest since it is one of the most likely contaminants in the sputtering system. The results are summarized in Table 3. Depositions were performed at a substrate temperature of 325°C in an argon atmosphere containing one of the following trace impurities: 0%, 0.25%, 0.40% and 1% of nitrogen and 0%, 0.25% and 1% of oxygen. In these particular examples, the potential between cathode and anode was maintained at 2000V with a current of 110mA.

It can be seen from Table 3 that, without the application of a bias, very small traces of either nitrogen or oxygen caused a marked deterioration in the magnetic properties of the deposited films. The effectiveness in removing the consequences of these impurities by application of a suitable bias to the films during deposition can be seen. The data also show that larger impurity concentrations require a greater bias potential to eliminate adverse effects and bring about the desired magnetic properties. These results further indicate that sensitivity to impurity contamination is decreased, and the control over the resultant magnetic properties is improved, by application of suitable bias during deposition.

• *Elevated temperature properties*

With the improved uniformity and reproducibility attainable due to the bias technique, it was possible to determine, with confidence, the effect of substrate temperature on the magnetic properties of sputtered films. Plots of H_0, H_{k0} and α_{90} vs. initial substrate temperature are shown in Figs. 7, 8, and 9, respectively. The drawn curves represent approximate fits to the actual data points shown. All depositions

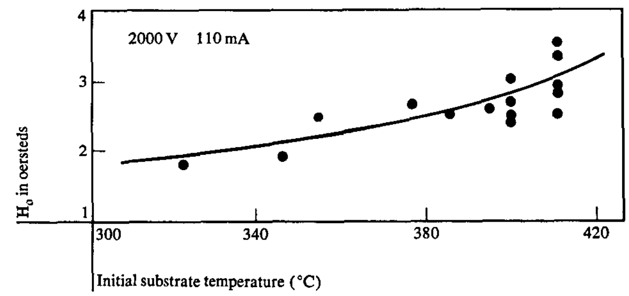

Figure 7 Coercive force vs. initial substrate temperature.

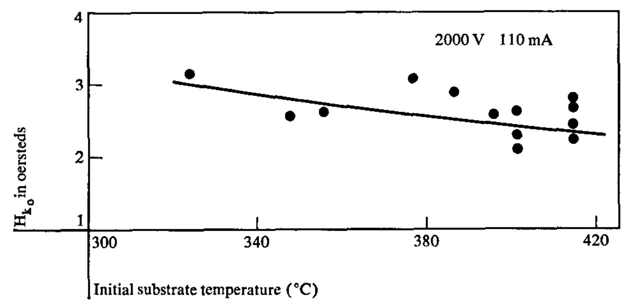

Figure 8 Anisotropy field vs. initial substrate temperature.

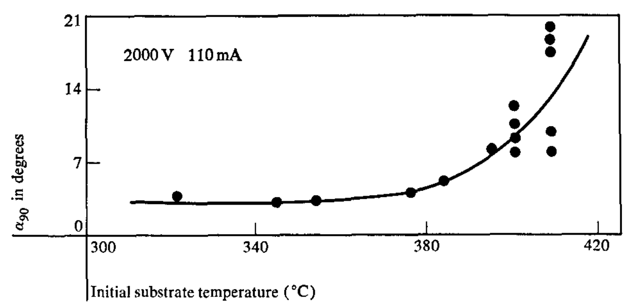

Figure 9 Dispersion vs. initial substrate temperature.

were performed at a cathode potential of 2000V, cathode current of 110mA, and film potential of 100V negative relative to the anode. From the plots, it can be seen that H_0 increases slightly with increasing temperature; H_{k0} decreases very slightly with increasing temperature; and α_{90} is relatively insensitive to temperature over the range 325–380°C, but it shows a marked increase with further increase in temperature (at 415°C, the films appear almost isotropic). The properties of bias-sputtered magnetic films as a function of temperature are similar to those found in this laboratory for evaporated NiFe films.

Conclusions

The experimental results presented here have shown that the primary problem remaining in the preparation of thin 81% Ni — 19% Fe permalloy films by cathodic sputtering, i.e., sensitivity to background impurity and substrate temperature control, can be either eliminated or overcome. It has previously been shown[7] that the problem resulting from magnetic field distortion due to the unavoidable presence of the ferromagnetic cathode can be minimized. The data presented in this paper have also demonstrated the compositional control and simplicity of film-thickness and deposition-rate control inherent in the conventional sputtering process.

The technique of sputtering allows for the preparation, in a reproducible manner, of thin ferromagnetic films having a high degree of magnetic uniformity. By proper choice of cathode composition, films having properties compatible with present memory design can likewise be prepared, giving an alternate method of preparation to the vacuum deposition and electrodeposition processes.

The use of the technique of bias sputtering has also provided a unique tool for the investigation of multicomponent alloy systems,[17] whose preparation by vacuum evaporation is prohibited by the differences in volatility and reactivity of many of the alloying elements of interest.

Acknowledgments

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