MAGNETOELASTIC EFFECTS IN THIN FERROMAGNETIC FILMS

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ABSTRACT

A method is described for the evaporation of ferromagnetic films onto a very thin water soluble polymer coating on glass substrates and their subsequent removal onto a liquid glycerin film. Determination of the change in magnetic anisotropy on removal provides a measure of the substrate constraint contribution to the anisotropy energy. Data are given for both Ni-Fe and Ni-Co alloys in the entire range 0 to 100% Ni, deposited at 100°C.

Significant changes in anisotropy upon removal were observed for almost all Ni-Fe alloys and for Ni-Co alloys in the range 0 to 40% Co. However, essentially a zero percentage change was observed for pure Fe, 83% Ni-Fe and the entire range 0 to 60% Ni in the Ni-Co alloys. These date are in disagreement with present theories of the constraint energy except for qualitative agreement in very limited composition ranges.

It has been suggested that the discrepancy between experimental and theoretical predictions for the anisotropy energy in thin films may result from the use of bulk material magnetoelastic constants which are inappropriate for thin films. However, this study of the magnetic properties of epitaxial films strongly suggests the equivalence of the magnetoelastic parameters in thin films and bulk materials. In this study a technique was developed for the preparation of step-free epitaxial films of Ni-Fe and Ni-Co alloys deposited at 400°C. The strain sensitivity $\frac{\Delta H_k}{\varepsilon}$ has been measured by ferromagnetic resonance along the [100] and [110] directions in the (001) plane for compositions ranging from 44 to 87% Ni in the Ni-Fe alloys and from 70 to 82% Ni in the Ni-Co

alloys. The data are in good agreement with the theoretical predictions $\frac{3(C_{11}-C_{12})\lambda_{100}}{M} \text{ and } \frac{\frac{6C_{44}\lambda_{111}}{M}}{M} \text{ along the [100] and [110] directions}$ respectively, evaluated using bulk parameters.

In addition, the strain sensitivity of polycrystalline films deposited at 100°C and 400°C for Ni-Fe alloys and 400°C for Ni-Co alloys has been measured. Contrary to previous investigations, a systematic temperature dependence is found. Moreover, it is shown that the isotropic material model used previously by others to calculate the strain sensitivity in polycrystalline films is incorrect. However, the apparent proper formulation does not predict the experimentally determined result. This discrepancy remains unexplained.

New data for the crystalline anisotropy constant K₁ in constrained epitaxial films of Ni-Fe and Ni-Co are presented. Significant deviations from values in bulk material are observed. However, these deviations are believed to result from the substrate constraint and not material differences inherent in thin films.

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Chapter 1

UNIAXIAL ANISOTROPY

1.1 Introduction

The term uniaxial anisotropy energy refers to an energy of the form $K_u \sin^2 \theta$ in which θ is an angle between the magnetization \overline{M} and a fixed material axis determined by previous evaporation or annealing history. The anisotropy is sometimes characterized by the anisotropy field $H_k = \frac{2K}{M}$ required to saturate the film in the hard direction. Films fabricated by vacuum evaporation, sputtering or electroplating exhibit this anisotropy. The anisotropy axis can be oriented to any direction in the plane of the film by application of a magnetic field during deposition, and is surprisingly independent of vacuum, rate of evaporation, film thickness, and substrate material. However, the anisotropy is strongly dependent on the ferromagnetic material, the deposition temperature and the measurement temperature.

The anisotropy field H_k may be measured by the torque magnetometer, ferromagnetic resonance, the method of Kobelev (1962), and the susceptibility exhibited by the hard axis hysteresis loop. Only the first three methods have been used in this research.

It is the purpose of this introduction to discuss the suggested mechanisms for the production of uniaxial anisotropy. Phenomena such as shape anisotropy, angle of incidence effects, and crystalline anisotropy are not included in this discussion, although each is an important aspect of magnetism in thin films. Only films made by evaporation onto smooth substrates at normal incidence are considered. The anisotropy mechanisms discussed most often in the literature are magnetostrictive effects resulting from substrate constraint and directional ordering of atom pairs. In an attempt to explain the observed anisotropy in γ -phase Ni-Fe films, Robinson (1961) suggested that the uniaxial anisotropy could be explained by the combination of the pairorder and substraint constraint models. Using the strain calculation by West (1964) with bulk values for the magneto-elastic constants and the empirical relation for pair-ordering suggested by Ferguson (1958) with a single adjustable constant, good agreement with the experimental results of Wilts (1966) in γ -phase Ni-Fe alloys for room temperature deposition and measurement was obtained. However, studies in the α - α -phase Ni-Fe (Wilts (1965)) and γ -phase Ni-Co alloys (Brownlow and Wilts (1968)) have cast some doubt on the validity of the two-component theory.

1.2 Pair-Ordering Mechanism

The pair-ordering mechanism of anisotropy in bulk Ni-Fe and Ni-Co alloys appears well established. Neél (1953, 1954) and Taniguchi (1955) have interpreted this mechanism as the creation of a short-range directional order. In the pair approximation, this mechanism arises from a hypothetical energy term arising from a magnetic coupling between nearest neighbors that depends on the nature of the neighbors and on the angle between the line joining them and the magnetization. During annealing or deposition at some temperature T' below the Curie temperature T_c , a slight preferential alignment of the various type of pairs occurs by diffusion, and this state may be frozen into the lattice if the material is quenched to a lower temperature T where the diffusion

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rate is negligible. The same interaction energy that gave rise to the preferential alignment of pairs at T' is responsible for the preferential alignment of the magnetization at the new temperature T.

The difficult part of the pair-ordering theory is to calculate the dependence on the concentration of B atoms in an A lattice. For dilute concentrations of B, the number of B-B pairs is proportional to C_B^2 , where C_B is the concentration of the B component. The concept of a simple pair-interaction may not be valid at the higher concentration levels as more than pair-interactions become probable. Because of the difficulty of calculating this dependence, the simple form $C_A^2 C_B^2$ for the concentration dependence is often assumed for calculation purposes. A useful form of the pair-ordering anisotropy energy K_p which provides a reasonable fit to bulk data is given by Ferguson (1958) as

$$K_{p} \circ C (T_{c} - T') C_{A}^{2} C_{B}^{2}$$
 (1.1)

for a two-component alloy.

Although the behavior of bulk Ni-Fe and Ni-Co alloys can be approximated by Eq. (1.1), the situation in thin films is more complicated. In particular the pure metals Ni, Fe, and Co have large values of anisotropy where this model predicts none. In view of this fact, an additional mechanism involving magnetostrictive effects was introduced.

1.3 Strain Mechanism

The strain model has been discussed by Bozorth and Dillinger (1935), MacDonald (1957), Ignatchenko (1961), Robinson (1961), and West (1964). It assumes that each crystallite forms during evaporation or annealing

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at temperature T' under conditions of zero stress but magnetostrictively strained. For an isotropic medium the strain is simply the magnetostrictive extension $\lambda(T')$ parallel to M during evaporation and contractions $-\frac{1}{2}\lambda(T')$ perpendicular to \overline{M} . The temperature is now changed to a new value of T and it is assumed that the strains induced at T' are frozen into the film by means of the substrate constraint. Since the crystallites are no longer free to deform under the rotation of \overline{M} , a strain anisotropy energy K_c is developed given by

$$K_{s} = \frac{9 \ge \lambda(T) \ \lambda(T')}{4 \ (1+\nu)}$$
(1.2)

where E is Young's modulus, v is Poisson's ratio and $\lambda(T)$ is the magnetostriction constant at the new temperature T. Robinson first suggested the isotropic material model as an approximation to the constraint energy in thin films. In his derivation Robinson neglected the effects of the Poisson and transverse magnetostrictive contractions.

West first pointed out the fact that the isotropic material approximation to the constraint energy of a constrained, polycrystalline film was in principle not correct. West averaged the single-crystal magnetoelastic energy over an ensemble of randomly oriented crystallites. Implicit in his averaging process are the assumptions that the strain within each crystallite is that associated with an isolated singlecrystal, and that all components of the strain are frozen into the film at T'. West finds the strain anisotropy energy for this case to be

$$K_{s} = \frac{9}{10} \left[(C_{11} - C_{12}) \lambda_{100}(T) \lambda_{100}(T') + 3C_{44}\lambda_{111}(T) \lambda_{111}(T') \right] (1.3)$$

for cubic crystallites. Here the C's and λ 's are the standard elastic

and magnetostriction constants. It is particularly important to note that K_s due to West is small only when the λ 's are small, if it is assumed that sign changes with temperature are absent. The isotropic approximation predicts small magnitudes when the average magnetostriction is small, which is possible even when the single crystal constants are large provided they are opposite in sign.

The result of West reduces to the isotropic material model for the case of isotropic constants in the cubic case, i.e.

$$C_{11} - C_{12} = \frac{E}{(1 + v)}$$

$$C_{44} = \frac{1}{2} (C_{11} - C_{12}) = \frac{E}{2(1 + v)}$$

$$\lambda_{100}(T) = \lambda_{111}(T) = \lambda(T). \qquad (1.4)$$

The West calculation was reasonably successful in predicting the anisotropy energy at the pure metals. Here there is no possibility of pair-ordering, so the comparison is particularly simple. As shown in Table (1.1), the anisotropy energy K_u measured by Wilts (1965) and the predicted strain anisotropy K_s by West are in excellent agreement, whereas K_s predicted by the isotropic material model is nearly an order of magnitude lower than observed experimentally. The predictions of both models are nearly a factor of two greater than K_u observed in this experiment. For the theoretical predictions the data of West were used.



Table 1.1. Comparison of the measured value of the anisotropy energy K with the predicted values K on the basis of the isotropic material model and that proposed by West. All data are for films deposited at 100°C and measured at 25°C. Units are in 10³ erg/cc.

TABLE 1.1

1.4 Removal of Constraint

According to the two-component model, the substrate constraint should be removed if the film is detached from the substrate and supported in a relatively stress-free environment. The only remaining component would then be the pair-ordering mechanism. In pure Ni, Fe, and Co there is no possibility of atom pairs. Here it is expected that the anisotropy would vanish upon removal from the substrate. Contrary to this, Wako et al. (1963) found only a small change in a 7000 A film of Ni removed from a glass substrate. Similarly Pugh et al. (1960) found little change in Fe films removed from NaCl substrates. More recently Krukover (1967) has reported little change in Co and Fe films removed from NaCl and NaCl-onglass substrates. However, for Ni films, Krukover found that the anisotropy vanished upon removal. Attempts to verify these experiments in this study proved difficult as the agreement between films on glass and NaCl before removal was poor. Moreover, no systematic results could be obtained from films removed from NaCl substrates. In particular the anisotropy of Ni films never decreased to zero. Individual samples were widely scattered, but on the average the anisotropy decreased to about 25% of the original value.

In view of these results, and because of their relevance in understanding anisotropy, a new systematic study was attempted.

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A major portion of this thesis is concerned with new experimental methods and results that permit detailed comparison with theory.

Chapter 2

Measurement of Substrate Constraint Contributions to Uniaxial Anisotropy

2.1 Introduction

As indicated in the previous chapter, Robinson (1961) first suggested that uniaxial anisotropy in thin films is composed of two principal components; a magnetostrictive component resulting from substrate constraint should be removed if the film is detached from the substrate and supported in a relatively stress-free environment. In the present investigation, thin magnetic films of Ni-Fe and Ni-Co were evaporated onto a thin water soluble polymer coating on glass substrates and subsequently removed onto liquid glycerin films. If one assumes that residual constraints among crystallites in the free film are negligible, the change in the anisotropy field upon removal should provide a measure of the anisotropy energy due to substrate constraint. For purposes of comparison, the prediction of constraint energy by West (1964) which accounts for the polycrystalline nature of the film is included.

2.2 Experimental Method

Films 7 mm in diameter were deposited by evaporation simultaneously on coated and uncoated glass substrates 0.5 mm thick and 12 mm square. The slides were coated with a 4% solution of polyvinyl pyrrolidone in ethanol and spun on a rotating table until the ethanol evaporated, leaving a very thin uniform layer of the polymer covering the glass. Films on coated substrates were floated off by means of a water drop placed on the edge of the slide which migrated under the film. The water was carefully replaced by glycerin by drawing off the water at one substrate edge with an absorbent tissue and adding glycerin at the opposite edge. The glycerin provided sufficient viscosity to permit torquemeter measurements.

The experimental results appear independent of magnetic film thickness (300 to 1200 Å), pressure during evaporation (less than 10^{-6} Torr), and rate of evaporation (10 to 100 Å/sec). Although sensitivity to substrate temperature is expected, substrate temperature was limited by the stability of the organic polymer to below 150°C. A deposition temperature of 100°C was chosen to afford some measure of "bake-out". Additionally, this also provided a slightly wider γ -phase region in the Ni-Co alloys as can be seen from the crystal structure data by Suzuki (1969) in Figs. (2.1) and (2.2). Since the substrate temperature was close to room temperature, reliable estimates of the bulk magnetoelastic constants could be made from available data.

The thickness and coercive force H_c of the samples were measured by the hysteresis-looptracer. The anisotropy field H_k was measured for all samples before and after removal by the high-field torque method (Humphrey and Johnston (1963)). Measurements of the anisotropy and coercive force for films on uncoated glass and polymer coated substrates were in excellent agreement for the hcp and fcc alloys. In these regions the mean deviation of H_k between films coevaporated onto coated and uncoated substrates were less than 1%. The reproducibility of H_c , H_k , and ΔH_k for films evaporated onto the polymer in the α -phase alloy









(0 to 30% Ni in Fe) was poor, possibly because of the susceptibility of the bcc lattice to absorb the small interstitial atoms of the polymer. The percentage change in H_k upon removal of the films from the substrate is given in Figs. (2.3) and (2.4) for Ni-Fe and Ni-Co respectively.

2.3 Results and Discussion

The degree of correlation between ΔK_{μ} , the anisotropy energy change upon removal, and K_s , the predicted constraint energy by West, permits assessment of the validity of the proposed model. Briefly, West assumes randomly oriented, non-interacting crystallites, which reach an equilibrium condition during deposition or subsequent anneal at temperature T' where stress relief mechanisms are operative. The magnetostrictive strain in each crystallite is then supposed to be that associated with an unstressed single crystal at T' with the same crystallographic orien-In addition, it is assumed that the crystallites are rigidly tation. attached to the substrate at T' and that all components of strain are preserved after quenching to the measurement temperature T. Isotropic strains such as film-substrate thermal mismatch do not alter the West prediction. Since magnetoelastic data are not available for thin films, the assumption that bulk constants are appropriate for thin films will be made for purposes of comparison.

If it is assumed that all constraints are removed when the film is detached from the substrate, the comparison becomes particularly simple at the pure metals where there is no possibility of pair ordering. Accordingly, it is expected that K_u is initially equal to K_s and that K_u should vanish if the substrate constraint is removed. The comparison

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Fig. 2-3. Percentage change of anisotropy field ${\rm H}_{\rm k}$ for Ni-Fe alloys upon removal from substrates.



Fig. 2-4. Percentage change of anisotropy field ${\rm H}_{\rm k}$ for Ni-Co alloys upon removal from substrates.



	(K _u) _{exp} .	(ΔK _u) _{exp} .	(K _s) _{West}	(K ₁) _{Bulk}
Ni	2.1	-1.6	3.7	52
Fe	1.5	-0.06	1.4	400
Со	18.3	-0.5	38.0	2,000

Units are in 10^3 ergs/cc.

Table 2.1. Comparison of the measured value of the anisotropy energy K with the predicted values K on the basis of the isotropic material model and that proposed by West. All data are for films deposited at 100°C and measured at 25°C. The crystalline anisotropy for bulk materials (Bozorth (1951)) is also tabulated. between the theory and experimental data at the pure metals is given in Table (2.1). Although the values of K_u are in reasonable agreement with the West theory, the values of ΔK_u are not equal to $-K_u$. The anisotropy of Ni did not vanish, but decreased to about 20% of the original value. Furthermore, even worse agreement was observed at Co and Fe where only small changes were observed upon removal. For alloys the comparison is in Figs. (2.5) and (2.6). For the theoretical predictions the data of West were interpolated for the Ni-Fe alloys. For Co-Ni alloys the values of magnetostriction constants were obtained from Ref.37, and the elastic constants from Ref. 17. The most noteworthy features are the very small values of $\Delta K_u/K_u$ for the Co-Ni alloys between 0% and 60% Ni, the extreme disagreement with West for Co-Ni alloys, and the good qualitative agreement between West and the experimental results in the γ -phase Ni-Fe.

West, in noting that K_u at Ni was bounded above by the predicted K_s , has suggested that incomplete adherence of the film to the substrate may exist. It is expected that this would simply scale down the value of K_s predicted on basis of the theory by roughly the percentage of the film that is not constrained. Although this concept would be consistent with the γ -phase Ni-Fe data where qualitative agreement exists with West's theory, the extreme disagreement between the theory and the γ -phase Ni-Co alloy data does not support this hypothesis.

While the likely origin of constraint was thought to have been filmsubstrate adhesion, West suggested the possibility that part of the constraint arises from local stresses among adjacent crystal grains of a continuous film in a manner similar to that proposed by Bozorth and



_, and calculation Fig. 2-5. Anisotropy energy, ..., change in anisotropy energy, _______, after West, _______, as a function of composition for Ni-Fe.





Dillinger (1935) for bulk material. If the constraint were due in part to interactions among crystallites, the anisotropy due to strain would not be expected to entirely disappear upon removal of the film from the substrate. This is consistent with the observation that ΔK_u is less than K_s . However, it does not seem reasonable that Ni should differ so greatly from Fe, Co, and Ni-Co alloys.

One would expect this same mechanism to be operative in bulk material, particularly at pure Ni or Fe. The expected effect was not observed in earlier work, and the idea was rejected by Bozorth (1956) for bulk materials. In any case, the argument cannot be made independent of alloys for thin films and appears inconsistent with the experimental results.

Small amounts of preferential alignment among crystallites in the film plane would provide a large component of anisotropy through the mechanism of the crystalline anisotropy (in which case K_u would correlate with K_1). However, removal of the film from the substrate would not affect that component, and the change in anisotropy ΔK_u would still be expected to correlate with K_s while the residual ($K_u + \Delta K_u$) might correlate with K_1 . Examination of Table (2.1) shows that neither of these correlations is found. Moreover, such preferential alignment has not been observed in many careful studies by transmission electron diffraction.

The assumption that the film is deposited in equilibrium, magnetostrictively strained but not stressed is strongly challenged by the observation that large isotropic stresses not related to substrate-film thermal expansion mismatch and as yet not adequately explained are known.

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to exist even in films deposited at room temperature. In a recent investigation by Johnson and Wilts (1970), utilizing the film removal technique described here, they found that this stress increases roughly linearly with thickness to a maximum of 10^{10} dynes/cm² at 600 Å and that it remains constant for further increasing thickness. Although the microstresses within crystallites may not strictly mirror the magnitude of the macrostress measured in their experiment, it is known that H_k is surprisingly constant down to 100 Å. It appears then that no correlation exists between the isotropic stress and the deviation of the constraint theory from the experimental observations.

The small crystallite size (100 A) in room temperature films resulting in a large fraction of the atoms to be near surfaces and the high density of defects known to exist in thin films may result in elastic and magnetostriction constants substantially different from bulk material. If in fact this were the case, it is difficult to imagine what form the deviation might take. The expected zero in ΔK_u on the basis of bulk data near 80-20% Ni-Fe was observed. However, particularly near Co the expected change was large but the experimental value was near zero. Since no systematic assumption can be made for the change in the magnetoelastic constants independent of alloy, the only alternative for this theory is to shown the magnetostriction and elastic constants are different for thin films. It is the purpose of the remainder of this thesis to determine if in fact the magnetoelastic constants of a single crystallite in a thin film resemble those of a large single crystal.

Chapter 3

Measurement of the Magnetoelastic Constants in Thin Films

3.1 Introduction

As discussed in the last chapter, it is of practical as well as theoretical interest to determine the magnetoelastic constants for thin films. In particular the theory of West (1964) cannot be correct if in fact the magnetostriction and elastic constants are in reasonable agreement with bulk data. Although these constants are readily measured in bulk crystals, the techniques used do not appear immediately applicable to the thin films constrained by substrates. However, the magnetic properties of most ferromagnetic materials change with the application of stress to such an extent that stress may be ranked with field strength and temperature as a primary factor affecting magnetic change. It has been predicted and indirectly verified that the induced anisotropy in bulk polycrystalline material due to a uniaxial stress σ when applied at angle ϕ with respect to the magnetization M in the single domain state can be represented by

$$K_{\sigma} = \frac{3}{2} \lambda_{s} \sigma \sin^{2} \phi$$
(3.1)

where λ_{λ} is the saturation magnetostriction constant.

If a thin film of isotropic material is rigidly attached to a substrate, a uniaxial strain may be applied in the plane of the film by bending the substrate. For thin enough substrates, the lateral strain is negligible compared to the longitudinal strain and it is only necessary to account for the fact that the magnetic material has zero additional strain in the transverse directions and zero stress in the direction perpendicular to the film plane. Under these conditions the additional stresses induced into the films are

$$\sigma_{x} = \frac{E \varepsilon_{x}}{1 + v^{2}} \qquad \sigma_{y} = \frac{v E \varepsilon_{x}}{1 - v^{2}} \qquad (3.2)$$

where E is Young's modulus, v is the Poisson's ratio, and ε_x is the applied strain. The anisotropy energy induced by the strain ε_y is therefore

$$K_{\varepsilon} = \frac{3}{2} \frac{E \lambda_{s}}{1 + \nu} \varepsilon_{x} \sin^{2} \phi.$$
 (3.3)

Among those who have investigated the strain sensitivity $\frac{\Delta H_k}{\epsilon}$ of thin films are Smith (1959), Mitchell et al. (1963), Monnier (1967), Krukover (1967), Reekstin (1969), and Tolmon (1967). In general qualitative agreement with the isotropic material approximation is reported, except by Krukover at pure Co where the average magnetostriction constant is known to be large in bulk materials but the strain sensitivity was small. Smith and Mitchell reported the strain sensitivity to be independent of deposition temperature. This result is not confirmed in the present investigation.

Equation (3.3) is often given incorrectly in the literature with the Poisson term omitted. Moreover, the uniform stress approximation that has been used is incorrect in principle for polycrystalline films where the constraint exists at the crystallite level. The apparent proper formulation would subject each crystallite to a uniform uniaxial strain in the plane of the film. Although the two methods agree qualitatively in their predictions, the quantitative results do not. A simpler problem to treat theoretically, although much more difficult to achieve experimentally is the thin film single-crystal case, approximated by epitaxial films grown on single crystal substrates. Particularly useful is the (001) plane of the film grown parallel to the (001) plane of the substrate. Under this condition, the thermal mismatch which exists between the film and substrate is isotropic and cannot contribute to any components of anisotropy in the film plane according to the first order theory. The (001) plane offers the possibility of determining the quantities $(C_{11}-C_{12})\lambda_{100}$ and $C_{44}\lambda_{111}$ by independent measurement, where the C's and λ 's are the elastic and magnetostrictive constants respectively. Although the C's are in general slow functions of composition, the λ 's are usually rapidly varying with frequent sign changes. This should permit a rapid assessment of the similarity of thin film and bulk constants.

3.2 Cubic Symmetry

It is the purpose of this section to calculate the expected anisotropy field resulting from a uniaxial strain in the plane of a thin film. The case of cubic crystallites will be considered first.

Following Kittel (1949), the contributions to the anisotropy energy density due to deformation from cubic symmetry is given by the approximation

$$K_{\varepsilon} = -\frac{3}{2} (C_{11} - C_{12}) \lambda_{100} (\alpha_1^2 \varepsilon_{11} + \alpha_2^2 \varepsilon_{22} + \alpha_3^2 \varepsilon_{33})$$

- 3 $C_{44} \lambda_{111} (\alpha_1 \alpha_2 \varepsilon_{12} + \alpha_2 \alpha_3 \varepsilon_{23} + \alpha_3 \alpha_1 \varepsilon_{31}).$ (3.4)

In this expression the α 's are the direction cosines of the magnetization relative to the cubic axes and ϵ 's are components of the strain tensor defined by Love. This result follows from a Taylor series expansion of the anisotropy energy linear in strain and quadratic in the direction cosines and includes the lowest order terms consistent with cubic symmetry. A brief outline of this derivation is included in Appendix 1.

If a uniform strain e exists with direction cosines β_{i} measured from the cubic axes, then the tensor components of strain are given by

$$\varepsilon_{ii} = e \beta_i^2 \qquad i = j$$

$$\varepsilon_{ii} = 2e \beta_i \beta_i \qquad i \neq j \qquad (3.5)$$

Substituting (3.5) into (3.4), the induced anisotropy resulting from a uniform strain in a single cubic crystal is given by

$$K_{e} = -\frac{3}{2} (C_{11} - C_{12}) \lambda_{100} e(\alpha_{1}^{2} \beta_{1}^{2} + \alpha_{2}^{2} \beta_{2}^{2} + \alpha_{3}^{2} \beta_{3}^{2})$$

- 6 $C_{44} \lambda_{111} e(\alpha_{1} \alpha_{2} \beta_{1} \beta_{2} + \alpha_{2} \alpha_{3} \beta_{2} \beta_{3} + \alpha_{3} \alpha_{1} \beta_{3} \beta_{1})$ (3.6)

3.2.1 Single Crystal; Thin Film Geometry

In this case, it is assumed that the strain induced by the substrate is uniform throughout the thickness of the film and that the film is rigidly attached to the substrate. Further, it is assumed that the magnetization is confined to the plane of the film by the shape anisotropy inherent in this geometry. Consider first a single-crystal thinfilm with the [001] direction perpendicular to the film plane and a uniform strain e parallel to the [100] direction. In this case $\beta_2 = \beta_3 = \alpha_3 = 0$, so Eq. (3.6) reduces to

$$K_{100} = \frac{3}{2} (C_{11} - C_{12}) \lambda_{100} e \sin^2 \phi$$
 (3.7)

where ϕ is the angle between the magnetization and the applied strain. Next, consider a strain applied parallel to [110] axis in the (001) plane. Here $\beta_1 = \beta_2 = \frac{1}{\sqrt{2}}$; $\beta_3 = \alpha_3 = 0$. Eq. (3.6) yields

$$K_{110} = 3 C_{44} \lambda_{111} e \sin^2 \phi.$$
 (3.8)

It can be seen that application of a unidirectional strain in [100] and [110] directions in the (001) plane induces a uniaxial anisotropy aligned with these directions and involving simple products of the single crystal elastic and magnetostriction constants.

3.2.2 Polycrystalline; Thin Film Geometry

This case will first be considered under the assumptions that the crystallites are non-interacting and of such aspect ratios that the strain induced by the substrate is nearly uniform throughout the thickness of the crystallite. As before it is assumed that all components of strain in the plane of the substrates are fixed by the substrate and that the magnetization is confined to the film plane.

The total strain in each crystallite is the combination of a uniform strain in the plane imposed by bending the substrate, a Poisson contraction perpendicular to the film due to the in-plane strain through elastic effects and a variable component perpendicular to the film plane sensitive to the position of M in the plane due to magnetostrictive effects. To the order of approximation in this analysis, the magnetostrictive elongations perpendicular to the film plane are due only to the presence of the substrate constraint and are independent of the in-plane strain. Therefore, if only the change in the anisotropy as a function of applied strain is desired, the magnetostriction component may be neglected.

The Poisson contraction perpendicular to the plane cannot contribute to the in-plane anisotropy because its axis of symmetry is perpendicular to the film plane. For any particular crystallite orientation the induced anisotropy due to the Poisson contraction perpendicular to the film plane is not necessarily parallel to the Poisson strain axis. However, in a random film there will exist another crystallite with its easy axis in the plane determined by the strain and easy axis of the first crystallite which cancels any in-plane component of anisotropy. The net contribution to the anisotropy due to the Poisson strain can only be a perpendicular anisotropy. Hence, the Poisson contractions may be neglected if only the magnitude of the in-plane strain-sensitivity is of interest.

In order to calculate K_{poly} , $K_e(\alpha_i, \beta_i)$ must be averaged over all possible directions of the saturation magnetization and induced strain relative to the axes of an individual crystallite and subject to the constraint that the angle between the strain and magnetization is ϕ . These directions are assumed randomly distributed. The averages of the terms in α_i and β_i have been given by Birss (1960) as

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$$<\alpha_{1}^{2}\beta_{1}^{2} + \alpha_{2}^{2}\beta_{2}^{2} + \alpha_{3}^{2}\beta_{3}^{2} > = \frac{2}{5}\cos^{2}\phi + \frac{1}{5}$$
$$<\alpha_{1}\alpha_{2}\beta_{1}\beta_{2} + \alpha_{2}\alpha_{3}\beta_{2}\beta_{3} + \alpha_{3}\alpha_{1}\beta_{3}\beta_{1} > = \frac{3}{10}\cos^{2}\phi - \frac{1}{10}$$
(3.9)

Substituting (3.9) into (3.6), the result is

$$K_{\text{poly}} = \frac{3}{10} \quad \text{e} \quad \{2(C_{11} - C_{12})\lambda_{100} + 6 C_{44}\lambda_{111}\} \sin^2 \phi$$
$$= (\frac{2}{5}K_{100} + \frac{3}{5}K_{100}) \sin^2 \phi \qquad (3.10)$$

where the terms independent of ϕ have been dropped.

Alternatively, it might be assumed that as before all components of strain in the plane of the film are fixed by the substrate and that M is confined to the film plane. However, because of the interactions among crystallites and their small size, it is possible that the Poisson contraction normal to the film would be roughly constant throughout the film. If this is the case, the Poisson terms cannot contribute to the strain sensitivity in the plane. The result is again Eq. (3.10).

These assumptions are made in order to simplify the problem, but are certainly open to question. In very thin films made at high temperatures, the approximation of noninteracting particles may be valid. In thick films the assumption of a uniform contraction may be better. It is interesting that both pictures yield the same result which is supported by the experimental observation that the strain induced anisotropy is independent of film thickness (200 to 2000 Å).

3.3 Hexagonal Symmetry

The case of a polycrystalline sample composed of crystallites having hexagonal symmetry is somewhat more complex. However, the calculation, details of which are given in Appendix 2, proceeds in the same manner as for the cubic case. The result for the strain sensitivity is

$$K_{\text{poly}} = \{ \frac{7}{15} (C_{11} - C_{12}) (\lambda_a - \lambda_b) - \frac{2}{15} [C_{13} (\lambda_a + \lambda_b) + C_{33} \lambda_c] \\ + \frac{2}{15} [C_{12} \lambda_a + C_{11} \lambda_b + C_{13} \lambda_c] \\ - \frac{2}{5} C_{44} (\lambda_a + \lambda_c - 4 \lambda_d) \} \text{ e sin}^2 \phi \qquad (3.11)$$

where λ_a , λ_b , λ_c , and λ_d are the four magnetostriction constants and C_{11} , C_{12} , C_{13} , C_{33} , and C_{44} the five elastic constants which are necessary to describe the hexagonal case. As for the cubic case, an anisotropy is induced by application of a uniaxial strain.

3.4 Experimental Methods

Films 7 mm in diameter were deposited by evaporation simultaneously onto 12 mm square substrates of glass 0.6 mm thick and single-crystal MgO, 0.4 mm thick, at 400°C. The reasons for using MgO substrates are discussed later. The MgO substrates had [001] directions perpendicular to the large surfaces and edges parallel to [100] or [110] directions depending on the particular strain axis desired. The MgO substrates were supplied commercially (Semi-Elements, Inc.) with one of the large surfaces finished to 1µ. This surface was further polished by dilute solutions of $.3\mu \ Al_2O_3$ particles in water over silk and $.05\mu$, Al_2O_3 particles over felt and finally in water over felt. The duration of each operation in this sequence was approximately 15 minutes. The last trace of visible scratches was eliminated by annealing the substrates in triple distilled water at 35° to 45°C for 1 hour. For temperatures below this range the scratches persisted; for temperatures above this range new defect structures developed and the scratches were enhanced. The substrates were then rinsed in electronic grade acetone and placed immediately in vacuum for deposition. Substrates were re-used by lapping off the deposited film in the $.3\mu$ solution (this was necessary to maintain the surface flatness) and repeating the cycle.

Films were deposited with the applied field in the direction of the strain axis. The induced uniaxial anisotropy field was then parallel to the strain axis during the resonance measurement.

A sensitive indication of the quality of epitaxy is the magnitude of K_1 , the crystalline anisotropy. The general behavior of K_1 of Ni as a function of substrate temperature is shown in Fig. (3.1) as measured by the high-field torque method. At low temperatures the interaction between the substrate and film is weak. However, beyond a critical temperature, epitaxial growth occurs which under suitable conditions can cause the deposited material to assume a single-crystal structure generally with the same orientation as the substrate. Neither the crystal structure nor the lattice constants of the substrate or deposited material need be the same. In this experiment the fcc Ni-Fe and Ni-Co alloys were deposited on MgO which has a NaCl structure and lattice constant of 4.20 Å compared to Ni, 3.52 Å. The choice of MgO is a



Fig. 3-1. The crystalline anisotropy K_1 of epitaxial Ni films deposited on MgO substrates as a function of substrate temperature.

logical one because it nearly matches the thermal expansion properties of Ni and is not hygroscopic at room temperature. A lower bound on substrate temperature was 350°C required for reliable epitaxy and an upper bound was 450°C set by contamination of substrate surfaces by Cu evaporated from the cavity used to determine the substrate temperature.

The strain sensitivity can be obtained experimentally from ferromagnetic resonance measurements of the induced anisotropy field. The dispersion relation for parallel resonance (DC magnetic field parallel to film plane) is given by Bozorth (1951) for the case of a thin cubic crystal cut parallel to the (001) plane in the presence of crystalline anisotropy as

$$\frac{\omega^2}{\gamma^2} = \{H + 4\pi M + \frac{K_1}{2M} (3 + \cos 4\phi) + \frac{K_2}{4M} (1 - \cos 4\phi) + \frac{K_u}{M} [1 + \cos 2(\phi - \psi)] \}$$

$$\times \{H + \frac{2K_1}{M} \cos 4\phi + \frac{2K_u}{M} \cos 2(\phi - \psi)\}$$
(3.12)

Here ϕ is the angle between the magnetization M and the crystallographic [100] axis, H is the applied field, and ψ is the angle between the easy axis of a uniaxial anisotropy K_u, and the same [110] axis. The first and second order crystalline anisotropy constants are K₁ and K₂ respectively.

The only experimental situations of interest here are resonance with M and the strain axis in the [100] direction in which the resonance condition is

$$\frac{\omega^2}{\gamma^2} = \left[H + 4\pi M + \frac{2K_1}{M} + \frac{2K_u}{M}\right] \times \left[H + \frac{2K_1}{M} + \frac{2K_u}{M}\right]$$
(3.13)

and the case with M and the strain axis in the [110] direction for which resonance occurs with

$$\frac{\omega^2}{\gamma^2} = \left[H + 4\pi M + \frac{K_1}{M} + \frac{K_2}{M} + \frac{2K_u}{M}\right] \times \left[H - \frac{2K_1}{M} + \frac{2K_u}{M}\right]. \quad (3.14)$$

For all Ni-Fe-Co alloys, $4\pi M >> \frac{2K_1}{M}$ and $\frac{K_2}{2M}$, in this experiment $4\pi M >> H$, and $\frac{2K}{M}$, and the resonance frequency is fixed at ω , so the shift in the resonance field $|\Delta H|$ is very nearly equal to the shift in the anisotropy field, $|\Delta H_k| = |\frac{2\Delta K}{M}|$.

A stripline for the resonance measurements was constructed that permitted strains to be introduced by simply supporting the substrates along two parallel edges and deflecting it by means of a wide plunger which contacted the substrate near the edges of the deposited film. This induced a nearly uniform, uniaxial strain in the deposited film. The deflection was related to the induced strain by means of strain gages applied to dummy substrates of each type. The maximum strain in the MgO substrates was 2×10^{-4} ; in the glass substrates 4×10^{-4} . The resonance frequency was held fixed at 3.5 or 5.5 GHz. The shift in the resonance field when the induced strain was parallel to the applied field along [100] or [110] direction gave the strain sensitivity directly. Photographs of the stripline are in Fig. (3.2).

Although the measurement of K_1 , the crystalline anisotropy, was primarily used to establish the existence of epitaxy, the actual values are of some interest because it is believed that these data are the first systematic studies of K_1 in constrained, epitaxial films deposited



Fig. 3-2. Photographs of stripline.

by evaporation. Experimental values of K_1 in Ni-Fe and Ni-Co films are shown in Figs. (3.3) and (3.4). Bulk data for unconstrained film are given from Refs. 7 and 28.

3.5 Experimental Measurement of Crystalline Anisotropy

Previous work in ferromagnetic films has centered on materials with large K_1 grown on cleaved NaCl. However, some work has been done on alloys with low K_1 after the films were removed from substrates. The limited use of epitaxial films in the investigation of thin film properties is believed to result from inconsistent data resulting from substrate imperfections such as oriented cleavage steps resulting in shape anisotropies, the high thermal mismatch inherent with metal films on NaCl or the susceptibility of NaCl to water vapor.

In general, the reported magnitude of K_1 for Ni films on substrates is significantly higher than that of bulk crystals, while most workers report near bulk values for Ni films floated off NaCl. Part of this difference arises from the conditions of measurement. Films deposited on substrates are measured at constant strain while bulk samples are generally measured under conditions best approximated by zero stress. The presence of magnetostriction gives rise to an apparent contribution to the anisotropy energy as the lattice is allowed to deform under the action of magnetoelastic forces. This contribution is easily calculated for a cubic crystal by substitution of the magnetostrictive strains measured under conditions of constant stress into the expression for the total energy density found in the Appendix 1, Eq. (A-1.5). This result may be written

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of film composition.



Fig. 3-4. K_1 of Ni-Co films deposited at 400°C as a function of film composition.

$$K_1^{\sigma} = (K_1 + \Delta)$$

where K_1^{σ} is the apparent anisotropy energy measured at constant stress, K_1 is the anisotropy energy at constant strain and Δ is given by

$$\Delta = \frac{9}{4} \left[(C_{11} - C_{12}) \lambda_{100}^2 - 2 C_{44}^2 \lambda_{111}^2 \right]$$

where constant terms and uniaxial components that may exist are neglected. For Ni, $\Delta = +2 \times 10^3$ erg/cc which is negligible compared to the observed discrepancy of 6 x 10^4 erg/cc between bulk and epitaxial films evaporated onto MgO at 400°C.

Freedman has ascribed this large discrepancy to magnetoelastic effects resulting from thermal mismatch between the film and substrate. These effects result from higher order terms in the approximation for the magnetoelastic energy which include products of strain and fourth order angular dependence on the magnetization. Thermal mismatch then gives rise to strains which give energy terms that have the same form as the anisotropy energy. This theory is difficult to test as the higher order magnetostriction constants are unknown. In any case, the view that the substrate constraint is responsible for the abnormal K_1 observed rather than material differences between thin films and bulk is consistent with the near bulk values observed for unconstrained films and the magnetoelastic data discussed later.

3.6 Strain Sensitivity - Experiment vs. Theory

The theoretical predictions given in Eqs. (3.7-8) and experimental results for the strain sensitivity of epitaxial films of Ni-Fe alloys

for both the [100] and [110] directions of applied strain are given in Figs. (3.5) and (3.6). The data for epitaxial films of Ni-Co are in Figs. (3.7) and (3.8). In general the agreement is good with the predictions of section (3.2.1), and it is reasonable to conclude that thin, epitaxial films deposited at 400°C onto MgO substrates have magnetoelastic constants in good agreement with bulk data.

The data for polycrystalline films evaporated at 100°C and 400°C and the results from the calculation of the strain sensitivity of polycrystalline films given in Eq. (3.10) are included in Figs. (3.9) for the Ni-Fe alloys. For comparison, the data by Mitchell for Ni-Fe are also plotted in Fig. (3.8). The results for 400°C Ni-Co films are in Fig. (3.10). Contrary to earlier workers a significant, systematic temperature dependence is found in the Ni-Fe alloys. The magnitude of the strain sensitivity of films deposited at 100°C was approximately 35% lower than in films deposited at 400°C. This was also confirmed for one composition in the Ni-Co alloys (82% Ni). At neither temperature do the experimental data agree with theoretical predictions, although the discrepancy is smaller for the higher temperature deposition. Nevertheless, the dependence on alloy composition is qualitatively correct except for room temperature films of hexagonal Co.

A number of possible mechanisms suggest themselves as sources for the discrepancy between the experimental and calculated strain sensitivity. Stresses which result in deformations into the nonlinear region would seriously alter the prediction of section (3.2) for the strain sensitivity. Large isotropic stresses are known to exist in

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Fig. 3-6. Strain sensitivity for [110] direction of applied strain in epitaxial Ni-Fe films deposited at 400°C.



Fig. 3-7. Strain sensitivity for [100] direction of applied strain in epitaxial Ni-Co films deposited at 400°C.



Fig. 3-8. Strain sensitivity for [110] direction of applied strain in epitaxial Ni-Co films deposited at 400°C.



Fig. 3-9. Strain sensitivity of polycrystalline Ni-Fe films. The solid line corresponds to the predicted value on the basis of bulk data.



Fig. 3-10. Strain sensitivity of polycrystalline Ni-Co films. The solid line corresponds to the predicted value on the basis of bulk data. The experimental value for hcp Co films deposited at room temperature is indicated by the triangle and the predicted value is denoted by the arrow.

films deposited at room temperature. In order to evaluate this effect a single film (51% Ni-Fe; 450 Å thick) was deposited onto a polymer coated substrate at 25°C by methods described in section (2.2). The film was then floated off the substrate with water which was then allowed to evaporate with the result that the film was again attached to the substrate. It was observed that an isotropic strain of 6 x 10^{-3} existed originally in the film due to the substrate constraint (Johnson and Wilts (1970)). The strain sensitivity was measured before and after the film had been floated with less than 3% difference between the two measurements. This isotropic stress (1.6 x 10^{10} dynes/cm²) apparently does not result in nonlinear or plastic deformation and is not responsible for the disagreement between the predicted strain sensitivity and that experimentally determined for polycrystalline films near room temperature.

As mentioned previously, the strain sensitivity was found to be independent of film thickness (200-2000 Å). This result tends to diminish the importance of surface phenomena which would be a strong function of film thickness.

Crystallite size may be responsible for the observed temperature dependence of the strain sensitivity in polycrystalline films with larger deviations from the calculated result expected for the smaller crystallites. Crystallite size is known to be a strong function of substrate temperature and is reported to be independent of the occurrence of epitaxy at least for films deposited on NaCl substrates (Suzuki (1969)). The observation that polycrystalline films deposited at 400°C are in better agreement with the predicted value of strain sensitivity than are films deposited near room temperature may seem to support this hypothesis. However, it does not explain the difference between epitaxial films deposited at 400°C which agree with the predicted result and polycrystalline films deposited at 400°C which do not.

The role of lattice defects is not easily evaluated. However, the relative insensitivity of film properties to vacuum $(10^{-5} \text{ to } 10^{-10} \text{ Torr})$ and the smooth variation in film properties with small changes in alloy do not support this argument unless the defect density is very high and large decreases in defect density occur with the development of epitaxy. Such data for these films are unavailable.

In summary, no explanation has been found for the discrepancy between the experimental and calculated strain sensitivity in polycrystalline films. Nevertheless, the excellent agreement found for epitaxial films demonstrates the essential validity of the theory for strain sensitivity in cubic crystals, and that the elastic and magnetostriction constants of thin films (200 to 2000 Å) deposited at 400°C in good vacuum (10⁻⁷ Torr) are in good agreement with values found in bulk materials.

Appendix 1

Magnetoelastic Energy

Following Kittel (1949), the elastic energy density in a cubic crystal is

$$E_{e1} = \frac{1}{2} C_{11} (e_{11}^{2} + e_{22}^{2} + e_{33}^{2}) + \frac{1}{2} C_{44} (e_{12}^{2} + e_{23}^{2} + e_{31}^{2}) + C_{12} (e_{11}^{2}e_{22}^{2} + e_{22}^{2}e_{33}^{2} + e_{33}^{2}e_{11})$$
(A-1.1)

where C_{ij} are the elastic constants and e_{ij} are strains as defined by Love (1944) and are all zero for the unstressed, unmagnetized crystal. This relation is assumed to hold for a magnetized crystal. However, the strain is not zero in the absence of applied stresses but the crystal is deformed from cubic symmetry by internal magnetoelastic stresses which result in magnetostriction, e.g.

$$e_{11} = S_{11}\sigma_{11} + S_{12}(\sigma_{22} + \sigma_{33}) + \frac{3}{2}\lambda_{100}(\alpha_1^2 - \frac{1}{3})$$

$$e_{12} = S_{44}\sigma_{12} + 3\lambda_{111}\alpha_1\alpha_2 \qquad (A-1.2)$$

where λ 's are the magnetostriction constants, σ 's are the applied stress, α 's are the direction cosines of the magnetization referred to the cubic axes, and the [S] matrix is the inverse of the [C] matrix. The twoconstant approximation for the magnetostriction in cubic crystals is usually found to be satisfactory although higher order constants consistent with the symmetry requirements of the crystals are sometimes required. Here it is assumed that there is no volume change in the crystal as it proceeds from a hypothetical unmagnetized to a magnetized state, and that the volume of the crystal is unaffected by the position of the magnetization.

The magnetic anisotropy energy density in an unstrained cubic crystal is to terms of the 6th order

$$E_{k} = K_{1}(\alpha_{1}^{2}\alpha_{2}^{2} + \alpha_{2}^{2}\alpha_{3}^{2} + \alpha_{3}^{2}\alpha_{1}^{2}) + K_{2}(\alpha_{1}^{2}\alpha_{2}^{2}\alpha_{3}^{2})$$
(A-1.3)

where the lowest order anisotropy constants are K_1 and K_2 . To express the dependence of the anisotropy energy on strain, the energy is expanded in a Taylor series in the strains. Keeping only lowest order terms dependent on orientation yields

$$E_{mag-el} = B_{1}[e_{11}(\alpha_{1}^{2} - \frac{1}{3}) + e_{22}(\alpha_{2}^{2} - \frac{1}{3}) + e_{33}(\alpha_{3}^{2} - \frac{1}{3})] + B_{2}[e_{12}\alpha_{1}\alpha_{2} + e_{23}\alpha_{2}\alpha_{3} + e_{31}\alpha_{3}\alpha_{1}]$$
(A-1.4)

where B_1 and B_2 are the magnetoelastic coupling constants.

The equilibrium configuration of the crystal, i.e. the stable state of strain when magnetized in the direction $\vec{\alpha}$ in the absence of external stresses, may be found by minimizing the total energy density

$$E = E_{e1} + E_{mag-e1} + E_k$$
 (A-1.5)

with respect to e ... The solutions are:

$$e_{ii} = \frac{-B_1}{C_{11} - C_{12}} \quad (\alpha_i^2 - \frac{1}{3})$$

$$e_{ij} = \frac{-B_2}{C_{44}} \alpha_i \alpha_j \quad (i \neq j) \quad (A-1.6)$$

The magnetoelastic constants are found to be

$$B_{1} = -\frac{3}{2} (C_{11} - C_{12}) \lambda_{100}$$

$$B_{2} = -3 C_{44} \lambda_{111}$$
(A-1.7)

Thus the magnetoelastic energy is

$$E_{mag-el} = -\frac{3}{2} (C_{11} - C_{12}) \lambda_{100} [e_{11}(\alpha_1^2 - \frac{1}{3}) + e_{22}(\alpha_2^2 - \frac{1}{3}) + e_{33}(\alpha_3^2 - \frac{1}{3}] - 3 C_{44}\lambda_{111}[e_{12}\alpha_1\alpha_2 + e_{23}\alpha_2\alpha_3 + e_{31}\alpha_3\alpha_1]$$
(A-1.8)

For cubic crystals, consistent with the two constant approximation for the magnetostriction.

Appendix 2

Strain Sensitivity, Polycrystalline Film, Hexagonal Crystallites

Following Mason (1954), the contributions to the anisotropy energy due to deformation from hexagonal symmetry is given by the approximation

$$K_{\varepsilon} = - (C_{11} - C_{12}) (\lambda_{a} - \lambda_{b}) (\alpha_{1}^{2} \varepsilon_{11} + \alpha_{2}^{2} \varepsilon_{22} + \alpha_{1} \alpha_{2} \varepsilon_{12})$$

$$- [C_{13} (\lambda_{a} + \lambda_{b}) + C_{33} \lambda_{c}] (1 - \alpha_{3}^{2}) \varepsilon_{33}$$

$$- [C_{12} \lambda_{a} + C_{11} \lambda_{b} + C_{13} \lambda_{c}] (1 - \alpha_{3}^{2}) (\varepsilon_{11} + \varepsilon_{22})$$

$$+ C_{44} (\lambda_{a} + \lambda_{c} - 4\lambda_{d}) (\alpha_{2} \alpha_{3} \varepsilon_{23} + \alpha_{3} \alpha_{1} \varepsilon_{31})$$
(A-2.1)

In this expression the α 's are the direction cosines of the magnetization with the 3 axis coinciding with the c hexagonal axis and ϵ 's are components of the strain tensor. The λ 's and C's are the magnetostriction and elastic constants respectively.

If a uniform strain e exists with direction cosines β_i , the resulting induced anisotropy is

$$K_{e} = - (C_{11} - C_{12})(\lambda_{a} - \lambda_{b}) e (\alpha_{1}\beta_{1} + \alpha_{2}\beta_{2})^{2}$$

$$+ [C_{13}(\lambda_{a} + \lambda_{b}) + C_{33}\lambda_{c}] e (1 - \alpha_{3}^{2})(1 - \beta_{3}^{2})$$

$$- [C_{12}\lambda_{a} + C_{11}\lambda_{b} + C_{13}\lambda_{c}] e(1 - \alpha_{3}^{2})(1 - \beta_{3}^{2})$$

$$+ 2 C_{44}(\lambda_{a} + \lambda_{c} - 4\lambda_{d}) e(\alpha_{1}\beta_{1} + \alpha_{2}\beta_{2}) \alpha_{3}\beta_{3} \qquad (A-2.2)$$

To determine K_{poly} , $k_e(\alpha_i, \beta_i)$ must be averaged over all possible directions of the magnetization and strain relative to the axes of an individual crystallite and subject to the constraint that angle between the strain and magnetization is ξ . The average for the first term has been given by West:

$$<(\alpha_1\beta_1 + \alpha_2\beta_2)^2 > = \frac{1}{15} + \frac{7}{15}\cos^2 \xi$$
 (A-2.3)

The average for the remaining terms have been given by Birss:

$$<(1 - \alpha_3^2)(1 - \beta_3^2) > = \frac{2}{5} + \frac{2}{15}\cos^2 \xi$$

$$<(\alpha_1\beta_1 + \alpha_2\beta_2) \alpha_3\beta_3 > = -\frac{1}{15} + \frac{1}{5}\cos^2 \xi.$$
 (A-2.4)

Substituting the averages (A-2.3) and (A-2.4) into (A-2.2) gives the result

$$K_{\text{poly}} = \{\frac{7}{15} (C_{11} - C_{12}) (\lambda_a - \lambda_b) - \frac{2}{15} [C_{13} (\lambda_a + \lambda_b) + C_{33} \lambda_c] \\ + \frac{2}{15} [C_{12} \lambda_a + C_{11} \lambda_b + C_{13} \lambda_c] \\ - \frac{2}{5} C_{44} (\lambda_a + \lambda_c - 4 \lambda_d)\} \quad e \sin^2 \phi$$
 (A-2.5)

where constant terms have been dropped.

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