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CONDITIONS FOR STRESS-INDUCED UNIAXIAL ANISOTROPY IN MAGNETIC MATERIALS OF CUBIC SYMMETRY*

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ABSTRACT

Uniaxial magnetic anisotropy may be induced in materials of cubic symmetry by the application of either planar stress or uniaxial stress normal to the plane. It is proven theoretically that either situation will produce the same result provided that the stress is of the same magnitude but opposite sign. From theory, the conditions for effective uniaxial anisotropy are derived for the {001}, {110}, and {111} families of planes, and possible applications are discussed from this standpoint. Of the three families, the {001} is the simplest and most desirable for this effect, while uniaxial anisotropy can be achieved for {111} planes only when the stress effects overwhelm the cubic magnetic anisotropy.

Introduction

The effect of external stress on magnetic anisotropy has been a subject of renewed interest in recent years. Remanent magnetization is affected by uniaxial stress in both microwave ferrites (1) and ferromagnetic metal tapes (2). A theoretical analysis of this effect has revealed that the hard-axis magnetostriction constant controls the changes in remanence properties (3). Another area where stress effects have received attention is in ferrimagnetic films for cylindrical domain device applications. In this case, the stress is biaxial and in the plane of the film. A partial analysis of this situation has been reported (4), and cylindrical domains have been observed in epitaxial films grown on substrates with different thermal expansion coefficients to create the planar stresses necessary for stress-induced uniaxial anisotropy (5).

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Since the uniaxial anisotropy induced by stress is produced by movement of the easy and hard axes of magnetization, the conditions for uniaxial anisotropy may be readily derived from the theory previously outlined (3). The purpose of this paper is to set up the general solution by proving that the planar stress situation may be simplified by treating it as a uniaxial stress along the normal to the plane, and to determine the conditions for stress-induced uniaxial anisotropy with the easy axis perpendicular to the three major families of planes, i.e., {001}, {110}, and {111}.

Theory

The total magnetic anisotropy energy E of a single crystal may be expressed as the sum of three terms which combine the effects of magnetocrystalline anisotropy (E_K), stress anisotropy (E_σ), and shape anisotropy (E_S), such that

$$E = E_K + E_\sigma + E_S \, . \qquad (1)$$

For a material of cubic symmetry with magnetocrystalline anisotropy constant K_1, the associated energy is given by (6)

$$E_K = K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) \qquad (2)$$

where α_i represent the direction cosines of the magnetization vector with reference to the ⟨001⟩ cubic axes. In spherical polar coordinates, Eq. (2) becomes

$$E_K = K_1(\sin^4\theta\sin^2\phi\cos^2\phi + \sin^2\theta\cos^2\theta) \, . \qquad (3)$$

If a uniaxial compressive stress σ is applied to the same material with magnetostriction constants λ_{100} and λ_{111}, the stress anisotropy term in given by (6)

$$E_\sigma = \frac{3}{2}\sigma\lambda_{100}(\alpha_1^2\gamma_1^2 + \alpha_2^2\gamma_2^2 + \alpha_3^2\gamma_3^2) +$$
$$3\sigma\lambda_{100}(\alpha_1\alpha_2\gamma_1\gamma_2 + \alpha_2\alpha_3\gamma_2\gamma_3 + \alpha_3\alpha_1\gamma_3\gamma_1) \, , \qquad (4)$$

where γ_i represent the direction cosines of σ relative to the cubic axes. A biaxial or planar stress may be represented by two orthogonal components of equal magnitude σ having cosines γ_i and γ_i' (7), so that Eq. (4) becomes

$$E_\sigma = \frac{3}{2}\sigma\lambda_{100}\sum_i \alpha_i^2(\gamma_i^2 + \gamma_i'^2) + 3\sigma\lambda_{111}\sum_{i<j} \alpha_i\alpha_j(\gamma_i\gamma_j + \gamma_i'\gamma_j') \, . \qquad (5)$$

As outlined in the Appendix,

$$\gamma_i^2 + \gamma_i'^2 = 1 - \beta_i^2, \qquad \gamma_i\gamma_j + \gamma_i'\gamma_j' = -\beta_i\beta_j, \tag{6}$$

where β_i represents the direction cosines of the normal to the plane. Substitution of these relations into Eq. (5) and application of the normality condition $\sum_i \alpha_i^2 = 1$ give

$$E_\sigma = \frac{3}{2}\sigma\lambda_{100} - \frac{3}{2}\sigma\lambda_{100}\sum_i \alpha_i^2\beta_i^2 - 3\sigma\lambda_{111}\sum_{i<j}\alpha_i\alpha_j\beta_i\beta_j . \tag{7}$$

When Eq. (7) is compared with Eq. (5), it is evident that a planar stress of magnitude σ produces essentially the same expression for E_σ as a uniaxial stress along the normal, with the same magnitude but opposite sign. The only difference is an isotropic term $\frac{3}{2}\sigma\lambda_{100}$.

The shape anisotropy energy arises from the demagnetizing effects of magnetic poles. For a thin film of magnetization $4\pi M$, the shape energy is given by (8)

$$E_S = 2\pi M^2\cos^2\zeta , \tag{8}$$

where ζ is the angle between $4\pi M$ and the normal to the plane. Of particular interest in this work are the principal crystallographic planes, the {001}, {110}, and {111} families, and the relations between $\cos\zeta$, the direction cosines α_i of $4\pi M$, and β_i of the normal may be determined from the standard relation

$$\cos\zeta = \alpha_1\beta_1 + \alpha_2\beta_2 + \alpha_3\beta_3 . \tag{9}$$

Thus, it may be readily shown that

$$\cos^2\zeta = \cos^2\theta \tag{10}$$

$$\cos^2\zeta = \frac{1}{2}\sin^2\theta + \sin^2\theta\sin\phi\cos\phi \tag{11}$$

$$\cos^2\zeta = \frac{1}{3} + \frac{2}{3}\sin^2\theta\sin\phi\cos\phi + \frac{2}{3}\sin\theta\cos\theta(\sin\phi + \cos\phi) \tag{12}$$

for the (001), (110) and (111) planes, respectively.

For stress in the (001) plane, $\beta_1 = \beta_2 = 0$, $\beta_3 = 1$ and

$$E_\sigma^{001} = \frac{3}{2}\sigma\lambda_{100}(1-\alpha_3^2). \tag{13}$$

In spherical polar coordinates, Eq. (13) becomes

$$E_{\sigma}^{001} = \frac{3}{2}\sigma\lambda_{100}\sin^2\theta. \tag{14}$$

It is convenient at this point to define $E_{\sigma S} = E_{\sigma} + E_S$ and combine Eqs. (14), (8) and (10), with the result that

$$E_{\sigma S}^{001} = \frac{3}{2}(\sigma\lambda_{100} - \frac{4\pi M^2}{3})\sin^2\theta + 2\pi M^2. \tag{15}$$

For stress in the (110) plane, $\beta_1 = \beta_2 = 1/\sqrt{2}$, $\beta_3 = 0$ and the combination of Eqs. (7), (8) and (11) expressed in polar coordinates yields

$$E_{\sigma S}^{110} = \frac{3}{2}\sigma\lambda_{100} - \frac{3}{4}(\sigma\lambda_{100} - \frac{4}{3}\pi M^2)\sin^2\theta$$
$$- \frac{3}{2}(\sigma\lambda_{111} - \frac{4}{3}\pi M^2)\sin^2\theta\sin\phi\cos\phi. \tag{16}$$

For stress in the (111) plane, $\beta_1 = \beta_2 = \beta_3 = 1/\sqrt{3}$ and the combination of Eqs. (7), and (8) and (12) expressed in polar coordinates yields

$$E_{\sigma S}^{111} = \sigma\lambda_{100} - (\sigma\lambda_{111} - \frac{4}{3}\pi M^2)[\sin^2\theta\sin\phi\cos\phi + \sin\theta\cos\theta(\sin\phi + \cos\phi)]$$
$$+ \frac{2}{3}\pi M^2. \tag{17}$$

In the analysis reported previously (4), the stress terms of Eqs. (15), (16), and (17) were obtained by applying Eq. (5) directly after selecting suitable sets of γ_i and γ_i' for the three specific cases. It should be pointed out that a different sign convention for σ was used in that work.

Since each of the above equations may be added to Eq. (3) to form the total E, the energy extrema may be determined in the usual manner from $\partial E/\partial\theta = 0$ and $\partial E/\partial\phi = 0$. As in the problem of determining the effects of stress on remanence ratios (3), the major attention is focused on the movement of these axes of extreme energy, i.e., the easy and hard axes of magnetization.

Uniaxial anisotropy induced from the application of stress will be defined as any situation in which the principal easy axis is directed along the normal and all other major extrema are in the plane. In the following section, the relations that control the movement of the pertinent extrema are given, and the conditions for stress-induced uniaxial anisotropy are determined for the specific cases of interest.

Results

For the simplest case of σ in the (001) plane, the energy extrema along the three ⟨100⟩ axes do not rotate. However, the extrema initially (if shape anisotropy is neglected) along the ⟨111⟩ axes may be rotated either towards the normal or into the plane as depicted in Fig. 1. Although only one of these axes is shown it should be understood that all four behave identically because of the fourfold symmetry of the [001] axis. For the [111] extremum, which moves in the (110) plane, the sum of Eqs. (3) and (15) with $\phi = \pi/4$ gives

$$E^{001} = K_1(\tfrac{1}{4}\sin^4\theta + \sin^2\theta\cos^2\theta) + \tfrac{3}{2}(\sigma\lambda_{100} - \tfrac{4}{3}\pi M^2)\sin^2\theta + 2\pi M^2. \quad (18)$$

By setting $\partial E^{001}/\partial\theta = 0$, it follows directly that the movement of this extremum is described by

$$\cos^2\theta = \tfrac{1}{3} - (\sigma\lambda_{100} - \tfrac{4}{3}\pi M^2)/K_1. \quad (19)$$

For uniaxial anisotropy with $K_1 < 0$, the extremum is a minimum or easy axis and must be rotated to the normal ($\theta = 0$) and $(\sigma\lambda_{100} - \frac{4}{3}\pi M^2)/K_1 \leq -\frac{2}{3}$. With $K_1 > 0$, the extremum is a hard axis and must be rotated into the plane ($\theta = \pi/2$) and $(\sigma\lambda_{100} - \frac{4}{3}\pi M^2)/K_1 \geq \frac{1}{3}$.

For σ in the (110) plane, the sum of Eqs. (3) and (16) gives

$$E^{110} = K_1(\sin^4\theta\sin^2\phi\cos^2\phi + \sin^2\theta\cos^2\theta) + \tfrac{3}{2}\sigma\lambda_{100} - \tfrac{3}{4}(\sigma\lambda_{100} - \tfrac{4}{3}\pi M^2)\sin^2\theta - \tfrac{3}{2}(\sigma\lambda_{111} - \tfrac{4}{3}\pi M^2)\sin^2\theta\sin\phi\cos\phi. \quad (20)$$

By setting $\partial E^{110}/\partial\theta = \partial E^{110}/\partial\phi = 0$, it may be readily shown that the equations controlling the movement of the pertinent extrema are

$$\sin 2\phi = 3(\sigma\lambda_{111} - \tfrac{4}{3}\pi M^2)/2K_1 \text{ for the [100] and [010] axes,} \quad (21)$$

and

$$\cos^2\theta = \tfrac{1}{3} + (\sigma\lambda_{100} + \sigma\lambda_{111} - \tfrac{8}{3}\pi M^2)/2K_1 \text{ for the [111] axis.} \quad (22)$$

In Fig. 2, the conditions for uniaxial anisotropy determined as in the (001) plane case are presented as follows:

for $K_1 < 0$,

$$(\sigma\lambda_{111} - \tfrac{4}{3}\pi M^2)/K_1 \leq -\tfrac{2}{3} \qquad (\phi = -\tfrac{\pi}{4})$$
$$(\sigma\lambda_{100} + \sigma\lambda_{111} - \tfrac{8}{3}\pi M^2)/K_1 \leq -\tfrac{2}{3} \qquad (\theta = \tfrac{\pi}{2}) \quad (23)$$

for $K_1 > 0$,

$$(\sigma\lambda_{111} - \tfrac{4}{3}\pi M^2)/K_1 \geq \tfrac{2}{3} \qquad (\phi = \tfrac{\pi}{4})$$
$$(\sigma\lambda_{100} + \sigma\lambda_{111} - \tfrac{8}{3}\pi M^2)/K_1 \geq \tfrac{4}{3} . \qquad (\theta = 0) \quad (24)$$

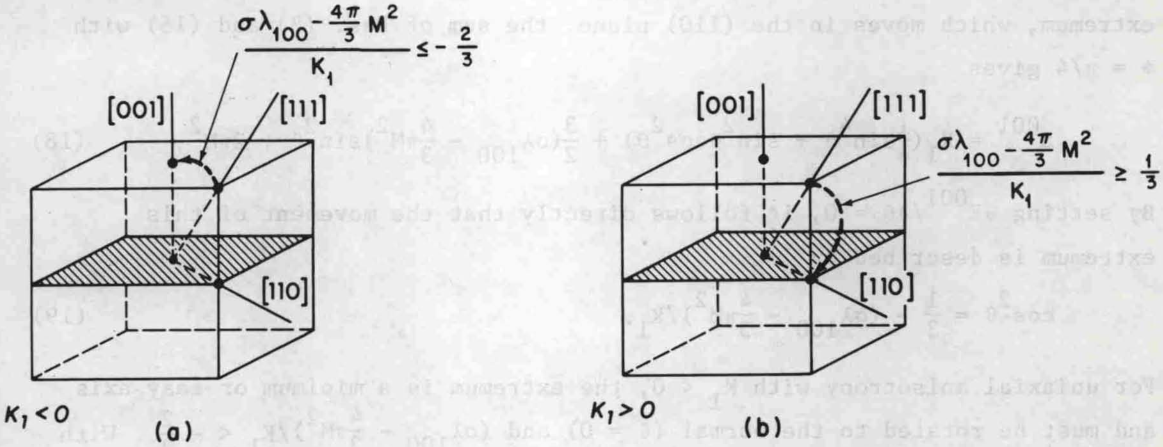

FIG. 1

Movements of important energy extrema and conditions for uniaxial anisotropy with stress in the (001) plane, for (a) $K_1 < 0$ and (b) $K_1 > 0$. Only one of the four is shown with its initial position along the $[1\bar{1}1]$ axis (for negligible shape anisotropy).

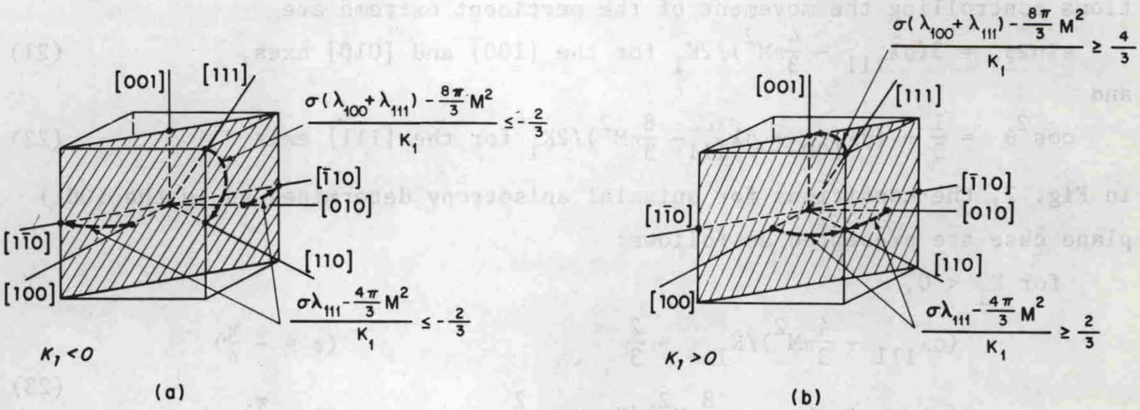

FIG.2

Movements of important energy extrema and conditions for uniaxial anisotropy with stress in the (110) plane, for (a) $K_1 < 0$ and (b) $K_1 > 0$. Initial positions of extrema are shown for the case of neglibible shape anisotropy.

It is evident from Eqs. (23) and (24) that the conditions for anisotropy are more complicated because of the dependence on both magnetostriction constants. If shape anisotropy is ignored, uniaxial anisotropy will be obtained only if either $\lambda_{100}/\lambda_{111} > 0$ or $\lambda_{100}/\lambda_{111} < 0$ with $|\lambda_{111}| > |\lambda_{100}|$. With compressive stress, both cases would require $\lambda_{111} > 0$ in addition to the above.

For σ in the (111) plane, the sum of Eqs. (3) and (17) gives

$$E^{111} = K_1(\sin^4\theta\sin^2\phi\cos^2\phi + \sin^2\theta\cos^2\theta) + \sigma\lambda_{100}$$
$$- (\sigma\lambda_{111} - \tfrac{4}{3}\pi M^2)[\sin^2\theta\sin\phi\cos\phi + \sin\theta\cos\theta(\sin\phi + \cos\phi)]$$
$$+ \tfrac{2}{3}\pi M^2 . \qquad (25)$$

By setting $\partial E^{111}/\partial\theta = \partial E^{111}/\partial\phi = 0$, it may be shown that the pertinent extrema move in the three {110} planes which contain the [111] axis. Since these three situations are equivalent, the problem may be solved for only one of them (as with σ in the (001) plane) by setting $\phi = \frac{\pi}{4}$. The first result is that the extremum along the [111] axis does not move. However, the movements of the other extrema in the (110) plane (i.e., $\phi = \frac{\pi}{4}$) are given by

$$(\sigma\lambda_{111} - \tfrac{4}{3}\pi M^2)/K_1 = \frac{\sin\theta\cos\theta(3\cos^2\theta - 1)}{\sin\theta\cos\theta + \sqrt{2}(\cos^2\theta - \sin^2\theta)} \qquad (26)$$

which is plotted in Fig. 3 as θ is varied from zero to π. As the ordinate increases in a positive sense, the extremum at [001] rotates towards the normal [111] direction until it meets the extremum from the [110] axis and both disappear for $(\sigma\lambda_{111} - \frac{4}{3}\pi M^2)/K_1 \geq 0.51$. At the same time, the $[11\bar{1}]$ extremum rotates into the plane at $\theta \simeq 145°$, which is a pole in Eq. (26), thus requiring that $(\sigma\lambda_{111} - \frac{4}{3}\pi M^2)/K_1 \to \infty$ in order to reach the plane. Where $(\sigma\lambda_{111} - \frac{4}{3}\pi M^2)/K_1$ increases in a negative direction,the [001] extremum rotates toward the pole at $\theta \simeq 145°$, and the [110] and $[11\bar{1}]$ extrema merge and vanish at $\theta \simeq 110°$ for $(\sigma\lambda_{111} - \frac{4}{3}\pi M^2)/K_1 \leq -0.15$. These conditions are depicted in Fig. 4.

For convenience,all of the conditions for uniaxial anisotropy discussed above are listed in Table 1. In Table 2, the expressions for E along the normal and different directions in the plane are given together with the differences in energy which represent the effective uniaxial anisotropy constants K_u. These results may be used to calculate K_u once the uniaxial anisotropy conditions have been satisfied.

Discussion and Conclusions

From the results summarized in Table 1, it is evident that the effect of

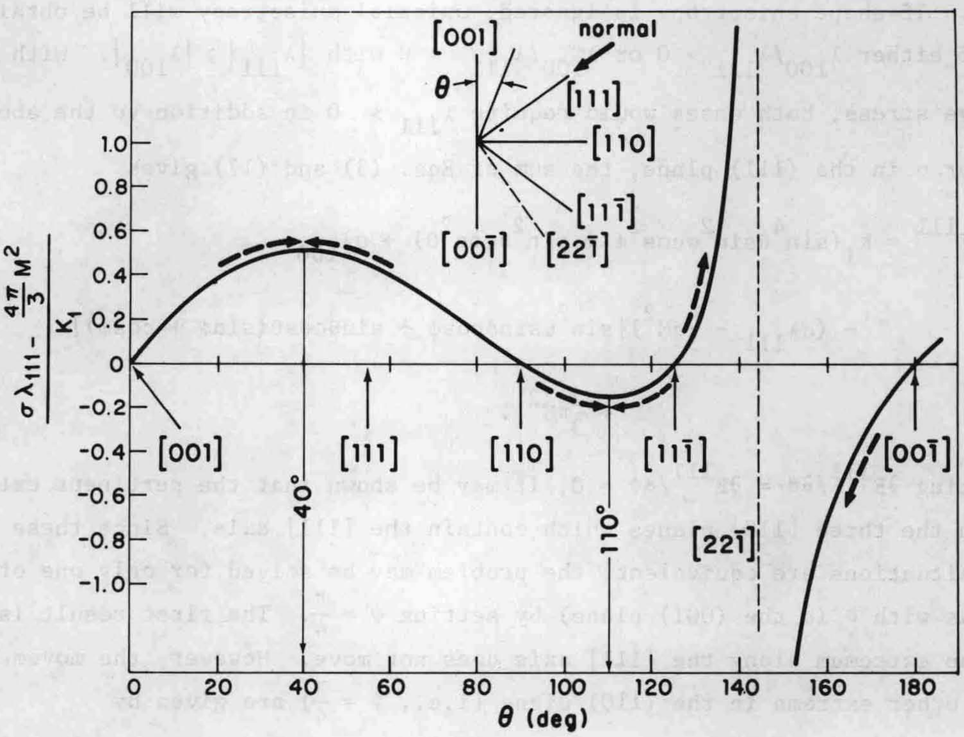

FIG. 3

Variation of $(\sigma\lambda_{111} - \frac{4}{3}\pi M^2)/K_1$ with the polar angle θ in the (110) plane for compressive stress σ in the (111) plane. This curve is a plot of Eq. (26) for $0 \leq \theta \leq \pi$.

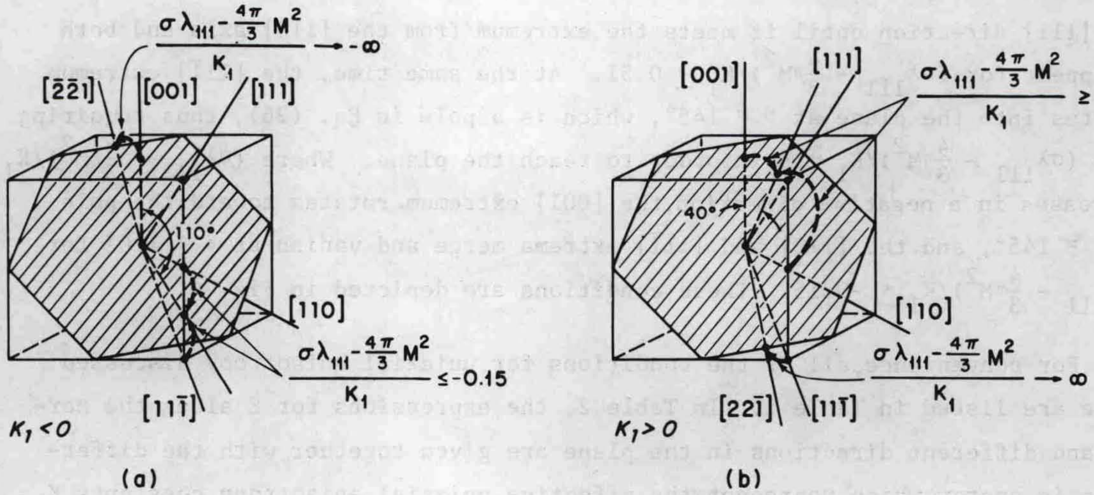

FIG. 4

Movements of important energy extrema and conditions for uniaxial anisotropy with stress in the (111) plane, for (a) $K_1 < 0$ and (b) $K_1 > 0$. Activity in only one of the three pertinent {110} planes is shown and shape anisotropy is assumed to be neglibible.

TABLE 1

Conditions for Stress-Induced Uniaxial Anisotropy

Plane	$K_1 < 0$	$K_1 > 0$
(001)	$\frac{\sigma\lambda_{100} - \frac{4}{3}\pi M^2}{K_1} \leq -2/3$	$\frac{\sigma\lambda_{100} - \frac{4}{3}\pi M^2}{K_1} \geq 1/3$
(110)	$\frac{\sigma(\lambda_{100} + \lambda_{111}) - \frac{8}{3}\pi M^2}{K_1} \leq -2/3$	$\frac{\sigma(\lambda_{100} + \lambda_{111}) - \frac{8}{3}\pi M^2}{K_1} \geq 4/3$
	$\frac{\sigma\lambda_{111} - \frac{4}{3}\pi M^2}{K_1} \leq -2/3$	$\frac{\sigma\lambda_{111} - \frac{4}{3}\pi M^2}{K_1} \geq 2/3$
(111)	$\frac{\sigma\lambda_{111} - \frac{4}{3}\pi M^2}{K_1} \to -\infty$	$\frac{\sigma\lambda_{111} - \frac{4}{3}\pi M^2}{K_1} \geq 0.51$
	$\frac{\sigma\lambda_{111} - \frac{4}{3}\pi M^2}{K_1} \leq -0.15$	$\frac{\sigma\lambda_{111} - \frac{4}{3}\pi M^2}{K_1} \to \infty$

TABLE 2

General Anisotropy Energy Relations for Planar Stress Situations

PLANE	E^n (normal)	E^p (in plane)	Axis (in plane)	$K_u = E^n - E^p$
(001)	$2\pi M^2$	$(3/2)\sigma\lambda_{100}$	[100]	$-(3/2)\sigma\lambda_{100} + 2\pi M^2$
		$K_1/4 + (3/2)\sigma\lambda_{100}$	[110]	$-K_1/4 - (3/2)\sigma\lambda_{100} + 2\pi M^2$
(110)	$K_1/4 + (3/4)\sigma(\lambda_{100} - \lambda_{111}) + 2\pi M^2$	$(3/2)\sigma\lambda_{100}$	[001]	$K_1/4 - (3/4)\sigma(\lambda_{100} + \lambda_{111}) + 2\pi M^2$
		$K_1/4 + (3/4)\sigma(\lambda_{100} + \lambda_{111})$	$[1\bar{1}0]$	$-(3/2)\sigma\lambda_{111} + 2\pi M^2$
		$K_1/3 + \sigma(\lambda_{100} + \lambda_{111}/2)$	$[1\bar{1}1]$	$-K_1/12 - (1/4)\sigma(\lambda_{100} + 5\lambda_{111}) + 2\pi M^2$
(111)	$\sigma(\lambda_{100} - \lambda_{111}) + 2\pi M^2$	$\sigma(\lambda_{100} + \lambda_{111}/2)$	$[1\bar{1}0]$	$-(3/2)\sigma\lambda_{111} + 2\pi M^2$
		$\sigma(\lambda_{100} + \lambda_{111}/2)$	$[22\bar{1}]$	$-(3/2)\sigma\lambda_{111} + 2\pi M^2$

shape anisotropy in films must be overcome by applied stress regardless of the crystallographic orientation or the sign of K_1. From a quantitative standpoint, the effect may be ignored only when $\left|\frac{4}{3}\pi M^2/K_1\right| << 1$. In some cases of nearly compensated ferrimagnetism, shape anisotropy may be small in comparison with magnetocrystalline anisotropy and the effect may be neglected. The following discussion will be focused on these situations, although the conclusions apply qualitatively even where shape effects are significant and the actual values of $\frac{4}{3}\pi M^2/K_1$ must be taken into account in the uniaxial anisotropy conditions.

Based on the above theory and results, the {001} family of planes appears to be the most desirable for applications requiring uniaxial anisotropy induced by planar stress. It is dependent on only the λ_{100} magnetostriction constant and can provide almost uniform energy in the plane if $\sigma\lambda_{100} >> K_1$. As discussed in an earlier section, the requirements for uniaxial anisotropy for {110} planes are complicated because either $\lambda_{100}/\lambda_{111} > 0$ or $\lambda_{100}/\lambda_{111} < 0$ with $|\lambda_{111}| > |\lambda_{100}|$. In addition, it may be seen from Table 2 that although the basic uniaxial conditions can be satisfied, significant anisotropy will usually remain within the plane. For the {111} planes, the uniaxial conditions can be approached only if $\sigma\lambda_{111} >> K_1$. This fact could place restrictions on the use of this common family of planes.

For applications involving ferrimagnetic oxides, some general observations may be made. Most iron garnets have K_1, λ_{100}, and λ_{111} negative at room temperature. According to the relations for K_u listed in Table 2, the required uniaxial anisotropy must be obtained by tensile stress, as was recently reported for Ga^{3+} - substituted $Y_3Fe_5O_{12}$ (5). For spinels, the situation is almost the same except that λ_{111} is positive and compressive stress could be used in {111} planes provided $\sigma\lambda_{111} >> K_1$. Since magnetic oxides are not noted for their tensile strength, one is led to consider methods of altering the signs of anisotropy and magnetostriction constants.

For garnet materials, both λ_{100} and λ_{111} may be changed to positive by substitutions of Mn^{3+} ions in octahedral sites. This type of substitution is particularly effective for λ_{100}, which is the constant that controls uniaxial anisotropy in the {001} planes (9,10). In addition, rare-earth ions such as Tb^{3+}, Eu^{3+}, and Er^{3+} substituted into the dodecahedral sites can affect the signs of the magnetostriction constants and could also permit compressive stress to be employed in some cases (11). Combinations of both Mn^{3+} and rare earths may allow a wide range of magnetostriction constants. Another possibility would be to use Co^{2+} substitutions in octahedral sites (usually with Si^{4+} in tetrahedral sites for charge compensation) to alter the cubic anisotropy constant (12) and provide either $K_1 = 0$ for {111} plane applications or $K_1 > 0$ for the {001} planes.

For spinels, the λ_{100} constant is normally large and negative ($\sim -20 \times 10^{-6}$), while λ_{111} is small and positive ($\sim +0.5 \times 10^{-6}$). Trivalent manganese may be used to change the signs of both constants (13,14) and was effective in producing uniaxial anisotropy by compressive stress for a $Mn_{.7}Fe_{2.3}O_4$ epitaxial film deposited on an MgO substrate with a (110) orientation (4). Divalent cobalt is known to be extremely effective in altering K_1 in spinels (13) and could be a useful additive in designing compositions with zero or positive K_1 values for possible compressive stress applications.

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Appendix

For three mutually orthogonal sets of direction cosines, γ_i, γ_i', and β_i,

$$\sum_i \gamma_i \gamma_i' = \sum_i \gamma_i \gamma_i = \sum_i \gamma_i' \beta_i = 0 \; . \tag{A1}$$

From basic analytical geometry theory, these cosines are related by

$$\begin{aligned} \gamma_1 &= \beta_2\gamma_3' - \beta_3\gamma_2' & \gamma_1' &= \beta_2\gamma_3 - \beta_3\gamma_2 \\ \gamma_2 &= \beta_3\gamma_1' - \beta_1\gamma_3' & \gamma_2' &= \beta_3\gamma_1 - \beta_1\gamma_3 \\ \gamma_3 &= \beta_1\gamma_2' - \beta_2\gamma_1' & \gamma_3' &= \beta_1\gamma_2 - \beta_2\gamma_1 \; . \end{aligned} \tag{A2}$$

By appropriate substitution from among the relations of Eq. (A2), it may be readily shown that

$$\gamma_1^2 = \beta_2^2(\beta_1\gamma_2 - \beta_2\gamma_1)^2 - \beta_3^2(\beta_3\gamma_1 - \beta_1\gamma_3)^2 \tag{A3}$$

and

$$\gamma_1'^2 = (\beta_2\gamma_3 - \beta_3\gamma_2)^2 \; . \tag{A4}$$

By adding Eqs. (A3) and (A4) and simplifying by means of Eq. (A1) in addition to the normality conditions of the cosines, it may be easily shown that the generalized result is given by

$$\gamma_1^2 + \gamma_i'^2 = 1 - \beta_i^2 \; . \tag{A5}$$

By similar applications of the relations in Eq. (A2),

$$\gamma_1\gamma_2 = \gamma_2^2\beta_1\beta_2 + \gamma_2\gamma_3\beta_1\beta_3 - \gamma_1\gamma_2(\beta_2^2 + \beta_3^2) \tag{A6}$$

and

$$\gamma_1'\gamma_2' = \gamma_1\gamma_3\beta_2\beta_3 - \gamma_3^2\beta_1\beta_2 - \gamma_1\gamma_2\beta_3^2 + \gamma_2\gamma_3\beta_1\beta_3 . \tag{A7}$$

After reductions and simplifications of the type employed in the derivation of Eq. (A5), the sum of Eqs. (A6) and (A7) becomes, after generalization,

$$\gamma_i\gamma_j + \gamma_i'\gamma_j' = -\beta_i\beta_j \; . \tag{A8}$$