# Definition and measurement of the surface magnetoelastic coupling coefficients in thin films and multilayers

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The symmetry lowering observed when passing from bulk samples to nearly two-dimensional thin magnetic films requires new expressions of the free energy to be derived. A larger number of coefficients must be included to describe the magnetocrystalline anisotropy and magnetoelastic coupling energies, depending on the symmetries of both the magnet and the surface. Various cases will be discussed, including amorphous and crystalline films. In addition, stresses due to the substrate impose in-plane strains in the film. The experimental methods for investigating stress-induced anisotropy in thin films, e.g., stress-modulated ferromagnetic resonance, or field-induced deflection of a magnetostrictive cantilever, do not permit the determination of all the magnetoelastic coupling coefficients, contrary to the case of bulk materials.

## I. INTRODUCTION

An increasing amount of work has been devoted in the recent past to the magnetocrystalline and magnetoelastic anisotropies in multilayers and very thin films of metals and alloys.[1–7] Following Néel,[8] surface and interface effects have been taken into account by introducing surface contributions to the anisotropy and magnetostriction coefficients, which vary inversely with the thickness of the magnetic films.[2,6,7] However, the number of these surface coefficients varies markedly with the actual symmetry of the surface, as mentioned in most of the previously cited works,[1,3–7] but this fact is actually ignored in a number of experimental papers, which deal with the magnetostriction constant ($\lambda$ or $\lambda_s$) as in isotropic bulk substances.

Even when implications of the symmetry have been recognized, the number of coefficients which appear in the equations does not always correspond to the one predicted by group theory. For instance, only one surface magnetoelastic coupling (SMC) coefficient $B_s$ has been considered in Ref. 3, although the relevant symmetry has been recognized to be uniaxial; only two thickness-dependent magnetostriction coefficients are used in Ref. 4 for describing a (111) thin film of cubic symmetry, while the actual symmetry is hexagonal (four coefficients needed); in a recent paper,[9] the first-order magnetoelastic coupling coefficients $B_1$ and $B_2$ for a fcc cobalt thin film have been expanded as $B_i = B_i^b + B_i^s/h$, which implicitly assumes that the number of SMC coefficients ($B_i^s$) is the same as the number of magnetoelastic coupling coefficients in the bulk ($B_i^b$): This is often wrong except in some specific cases, e.g., the (001) surface for a tetragonal crystal or the (0001) surface for a hexagonal one, where the local symmetry at the surface is the same as in the bulk; note also that a factor of 2 is needed before $B_i^s/h$, because a thin film has two surfaces, while in his original theoretical paper, Néel considered only a semi-infinite medium with one surface.

Even the notion of "surface magnetostriction"[6] is irrelevant to thin films which are necessarily firmly fixed onto a substrate: Only the surface magnetoelastic coupling is meaningful[3,5,9] and must be clearly defined before total confusion occurs.

Callen and Callen have given an illuminating description of the magnetoelastic coupling in single crystals, based on group symmetry considerations,[10] which provides an excellent introduction to the present work. Later on, a slightly different normalization has been adopted, which leads to much more simple and symmetrical equations.[11] Now we shall extend their theory to the case of very thin films and multilayers.

As mentioned above, surface effects do not necessarily lower the symmetry: For instance, in hexagonal crystals, a (0001) plane exhibits the same sixfold symmetry as the bulk; only the intensity of the coefficients will differ at the surface. On the other hand, in a cubic crystal where $a = b = c$, a (001) surface exhibits the tetragonal symmetry with $a = b$: In this case, the cubic coefficients are split into several lower-symmetry coefficients, and additional coefficients appear. The same is true for multilayers. For instance, a (111) multilayer composed of two different metals of cubic symmetry will exhibit hexagonal symmetry at each interface, which increases significantly the number of independent magnetoelastic coupling coefficients.

## II. MAGNETOELASTIC COUPLING IN AMORPHOUS THIN FILMS

There is no magnetocrystalline anisotropy in bulk amorphous substances, and the magnetoelastic energy is described by only two coupling coefficients, $B^{\alpha,0}$ for the isotropic exchange magnetoelastic coupling and $B^{\gamma,2}$ for the anisotropic Joule magnetoelastic coupling. Extending the symmetrical notations first introduced by Callen and Callen, the free energy density may then be written[11]

 

$$E/V=E_{\rm ms}+\frac{1}{3}B^{\alpha,0}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})+B^{\gamma,2}\left\{\left[\varepsilon_{xx}\left[\alpha_1^2-\frac{1}{3}\right]+2\varepsilon_{yz}\alpha_2\alpha_3\right]+\text{cycl}\right\}$$
$$+\frac{1}{6}C^{\alpha}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})^2+\frac{1}{2}C^{\gamma}\left\{\frac{2}{3}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]^2+\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})^2+2(\varepsilon_{xy}^2+\varepsilon_{yz}^2+\varepsilon_{zx}^2)\right\}, \tag{1}$$

where $V$ is the volume, $E_{\rm ms}$ is the magnetostatic energy, the $\alpha_i$'s are the direction cosines of the magnetization, and the $C^{\mu}$'s are the elastic coefficients defined as

$$C^{\alpha}=c_{11}+2c_{12} \quad \text{and} \quad C^{\gamma}=c_{11}-c_{12}=2c_{44} \quad \text{(condition for isotropic material)} . \tag{2}$$

The spherical symmetry is lowered to the cylindrical one in very thin films deposited onto a thicker planar substrate. Letting $(Ox,Oy)$ be the film plane, one may write the free energy density for such a thin film:

$$E/V=b_1^{\alpha,0}\frac{1}{3}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})+b_2^{\alpha,0}\frac{\sqrt{2}}{3}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]$$
$$+\left\{\frac{1}{2}\mu_0 M_s^2-k_2+b_1^{\alpha,2}\frac{\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}}{\sqrt{2}}+b_2^{\alpha,2}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]\right\}\left[\alpha_3^2-\frac{1}{3}\right]$$
$$+b^{\gamma,2}\left[\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})(\alpha_1^2-\alpha_2^2)+2\varepsilon_{xy}\alpha_1\alpha_2\right]+2b^{\delta,2}(\varepsilon_{yz}\alpha_2\alpha_3+\varepsilon_{zx}\alpha_3\alpha_1)$$
$$+\frac{1}{6}c_{11}^{\alpha}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})^2+c_{12}^{\alpha}\frac{\sqrt{2}}{3}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]$$
$$+\frac{1}{3}c_{22}^{\alpha}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]^2+\frac{1}{2}c^{\gamma}\left[\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})^2+2\varepsilon_{xy}^2\right]+\frac{1}{2}c^{\delta}(2\varepsilon_{yz}^2+2\varepsilon_{zx}^2) . \tag{3}$$

This latter expression must reduce to Eq. (1) when the thickness of the film, $t_f$, increases and becomes very large as compared with the one of a single layer: The only remaining anisotropy will be then the dipolar shape anisotropy.

For the sake of simplicity, we shall assume that a thin protective film of the same nature as the nonmagnetic thick substrate has been deposited onto the magnetic film. As there are two identical faces to the film, we shall write, according to Néel's model,[8]

$$k_2=\frac{2}{t_f}k^s, \quad b_1^{\alpha,0}=B^{\alpha,0}+\frac{2}{t_f}b_0^{\alpha,s}, \quad b_2^{\alpha,0}=\frac{2}{t_f}b_0'^{\alpha,s}, \quad b_1^{\alpha,2}=\frac{2}{t_f}b_1^{\alpha,s} ,$$
$$b_2^{\alpha,2}=B^{\gamma,2}+\frac{2}{t_f}b_2^{\alpha,s}, \quad b^{\gamma,2}=B^{\gamma,2}+\frac{2}{t_f}b^{\gamma,s}, \quad b^{\delta,2}=B^{\gamma,2}+\frac{2}{t_f}b^{\delta,s} . \tag{4}$$

Such relations define a surface magnetocrystalline anisotropy coefficient $k^s$ and a few SMC coefficients $b_i^{\mu,s}$. Rigorously, we should speak of interface effects and of interface magnetoelastic coupling coefficients, since the nature of the atoms in the substrate and the protective film will influence those coefficients; however, we shall retain the widely accepted term of surface effects, keeping in mind that the surface coefficients may differ from one film to another, depending on the nature of the substrate and of the protective film. When comparing thin films with bulk samples, one observes that $B^{\alpha,0}$ is simply modified and $B^{\gamma,2}$ is split into three different coefficients, while $b_2^{\alpha,0}$ and $b_1^{\alpha,2}$ are new coefficients generated by the symmetry lowering, as is $k_2$.

The same kind of relations may be derived from the coefficients of elasticity:

$$c_{11}^{\alpha}=C^{\alpha}+\frac{2}{t_f}c_{11}^{\alpha,s}, \quad c_{12}^{\alpha}=\frac{2}{t_f}c_{12}^{\alpha,s}, \quad c_{22}^{\alpha}=C^{\gamma}+\frac{2}{t_f}c_{22}^{\alpha,s} ,$$
$$c^{\gamma}=C^{\gamma}+\frac{2}{t_f}c^{\gamma,s}, \quad c^{\delta}=C^{\gamma}+\frac{2}{t_f}c^{\delta,s} . \tag{5}$$

The bulk magnetoelastic coupling coefficients are written $B_i^{\mu,l}$ where $l=2,4$, labels the degree of the harmonic polynomial in the direction cosines of the magnetization, $\mu$ indicates the irreducible representation, and $i$ appears when the same representation occurs more than once. The SMC coefficients should have been written $(b_i^{\mu,l})^s$ ($s$ for surface), but it seems reasonable to simplify the full notation, assuming the second-order terms ($l=2$) will dominate higher-order terms ($l=4,6$) in surface, as is the case for magnetocrystalline anisotropy. It is then no

longer necessary to recall the value of $l=2$.

The $1/t_f$ dependence of the total magnetoelastic coupling coefficients shown in Eqs. (4) has been already predicted theoretically and experimentally observed.[6,7,12] It must be mentioned here that elastic coefficients exhibit also this $1/t_f$ dependence, and we shall see that the thickness dependence of any magnetoelastic effect may arise from the combined thickness dependence of both the magnetoelastic and elastic coefficients.

## III. ENERGY DENSITY FOR A STRAINED AMORPHOUS THIN FILM

The free energy (3) is no longer the relevant potential energy since the film is magnetized and rigidly fixed to its substrate. As opposed to the case of bulk samples which work at constant stress, in-plane strains ($\varepsilon_{xx}$, $\varepsilon_{yy}$, and $\varepsilon_{xy}$) are imposed on the film by the thicker substrate. Minimizing Eq. (3) with respect to $\varepsilon_{yz}$, $\varepsilon_{zx}$, and $\varepsilon_{zz}$ provides

$$\varepsilon_{yz}=-(b^{\delta,2}/c^{\delta})\alpha_2\alpha_3,\quad \varepsilon_{zx}=-(b^{\delta,2}/c^{\delta})\alpha_3\alpha_1\ ,$$
$$\varepsilon_{zz}=-\frac{b_1^{\alpha,0}+\sqrt{2}b_2^{\alpha,0}+(3/\sqrt{2})(b_1^{\alpha,2}+\sqrt{2}b_2^{\alpha,2})(\alpha_3^2-\frac{1}{3})+(\varepsilon_{xx}+\varepsilon_{yy})[c_{11}^{\alpha}+(1/\sqrt{2})c_{12}^{\alpha}-c_{22}^{\alpha}]}{c_{11}^{\alpha}+2\sqrt{2}c_{12}^{\alpha}+2c_{22}^{\alpha}}\ . \tag{6}$$

Substituting these equilibrium values in Eq. (3), one gets the following expression for the film energy density:

$$\begin{aligned}E/V=&-\mu_0\mathbf{M}\cdot\mathbf{H}+\left\{\frac{1}{2}\mu_0M_s^2-k_2\right\}\left[\alpha_3^2-\frac{1}{3}\right]-\frac{1}{6}\frac{[b_1^{\alpha,0}+\sqrt{2}b_2^{\alpha,0}+(3/\sqrt{2})(b_1^{\alpha,2}+\sqrt{2}b_2^{\alpha,2})(\alpha_3^2-\frac{1}{3})]^2}{c_{11}^{\alpha}+2\sqrt{2}c_{12}^{\alpha}+2c_{22}^{\alpha}}\\
&-\frac{[b^{\delta,2}]^2}{c^{\delta}}(\alpha_3^2-\alpha_3^4)+\frac{3}{2}(\varepsilon_{xx}+\varepsilon_{yy})\frac{b_1^{\alpha,2}(c_{12}^{\alpha}+\sqrt{2}c_{22}^{\alpha})-b_2^{\alpha,2}(c_{11}^{\alpha}+\sqrt{2}c_{12}^{\alpha})}{c_{11}^{\alpha}+2\sqrt{2}c_{12}^{\alpha}+2c_{22}^{\alpha}}\left[\alpha_3^2-\frac{1}{3}\right]\\
&+b^{\gamma,2}\left\{\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})(\alpha_1^2-\alpha_2^2)+2\varepsilon_{xy}\alpha_1\alpha_2\right\}+\frac{\varepsilon_{xx}+\varepsilon_{yy}}{\sqrt{2}}\frac{b_1^{\alpha,0}(c_{12}^{\alpha}+\sqrt{2}c_{22}^{\alpha})-b_2^{\alpha,0}(c_{11}^{\alpha}+\sqrt{2}c_{12}^{\alpha})}{c_{11}^{\alpha}+2\sqrt{2}c_{12}^{\alpha}+2c_{22}^{\alpha}}\\
&+\frac{3}{4}(\varepsilon_{xx}+\varepsilon_{yy})^2\frac{c_{11}^{\alpha}c_{22}^{\alpha}-[c_{12}^{\alpha}]^2}{c_{11}^{\alpha}+2\sqrt{2}c_{12}^{\alpha}+2c_{22}^{\alpha}}+\frac{1}{2}c^{\gamma}\left[\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})^2+2\varepsilon_{xy}^2\right]\ .\end{aligned} \tag{7}$$

Apart from the last three terms, which do not depend on the orientation of the magnetization, this equation describes four contributions to the magnetic anisotropy in thin films.

(i) The shape anisotropy ($\mu_0M_s^2/2$) favors the in-plane magnetization; its origin is the long-range dipolar interaction.

(ii) The magnetic anisotropy of an unstrained body ($k_2$) arises, as for the magnetoelastic coupling, from short-range interactions, but it appears only when the symmetry is broken at the interfaces of the film; it may be markedly modified by the nature of the substrate and of the protective film, and by the roughness of the surfaces.[13]

(iii) The third contribution (in $\alpha_3^2$ and $\alpha_3^4$) is a consequence of the spontaneous magnetostrictive strains $\varepsilon_{yz}$, $\varepsilon_{zx}$, and $\varepsilon_{zz}$ of the sample which are observed when $\varepsilon_{xx}$, $\varepsilon_{yy}$, and $\varepsilon_{xy}$ are imposed. It is similar to the well-known magnetoelastic contribution to the first anisotropy constant in cubic crystals, but there is a difference: The crystals were allowed to expand freely, while in the present case the interface is clamped. The $\alpha$ contribution is somewhat complicated, and apart from a constant which we neglect, it provides two terms in $(\alpha_3^2-\frac{1}{3})$ and $(\alpha_3^2-\frac{1}{3})^2$, while the $\delta$ one gives $(\alpha_3^2-\alpha_3^4)$. Actually, this third contribution is usually very small compared with the shape anisotropy for $3d$ metals and alloys. In the case of nickel and neglecting surface effects, we have $b_1^{\alpha,0}\approx B^{\alpha,0}\approx 7\times10^8$ J m$^{-3}$, $b_2^{\alpha,2}\approx b^{\gamma,2}\approx b^{\delta,2}\approx B^{\gamma,2}\approx 8\times10^6$ J m$^{-3}$, $c_{11}^{\alpha}\approx C^{\alpha}\approx 5\times10^{11}$ Pa, and $c_{22}^{\alpha}\approx c^{\gamma}\approx c^{\delta}\approx C^{\gamma}\approx 1.5\times10^{11}$ Pa, the other coefficients being zero. For a thick nickel film, this gives a magnetoelastic contribution of about $-7\times10^3$ J m$^{-3}$ for the $\alpha_3^2$ term and $+3\times10^2$ J m$^{-3}$ for the $\alpha_3^4$ one, compared with the dipolar anisotropy, about $1.5\times10^6$ J m$^{-3}$. The inclusion of surface effects would not change these orders of magnitude.

(iv) Last, the stress-induced anisotropy also contributes to the uniaxial $(\alpha_3^2-\frac{1}{3})$ term and may give an additional in-plane anisotropy $(\alpha_1^2-\alpha_2^2)$. Note that in-plane strains may be related to the stresses through elastic coefficients $s_{ij}^s$ of the substrate, *not of the film:*

$$\varepsilon_{xx}=s_{11}^s\tau_{xx}+s_{12}^s\tau_{yy},\quad \varepsilon_{yy}=s_{12}^s\tau_{xx}+s_{11}^s\tau_{yy}\ ,$$
$$\varepsilon_{xy}=\tfrac{1}{2}s_{44}^s\tau_{xy}\ . \tag{8}$$

When perfectly isotropic in-plane stresses arise from differential thermal expansion or occur during the preparation of the film, $\tau_{xx}=\tau_{yy}$ and $\tau_{xy}=0$ and the in-plane anisotropy disappears. On the other hand, when an uniaxial stress, say, $\tau_{xx}$, is applied to the substrate, one may observe both in-plane and out-of-plane anisotropies, thus providing more information concerning the magnetoelastic coupling. Nevertheless, it is impossible to independently determine the two magnetoelastic coupling coefficients of the same degree ($l=0$ or 2) belonging to the $\alpha$ representation. The $\alpha$ magnetoelastic coupling coefficients appear always coupled with elastic coefficients belonging to the same irreducible representation, thus defining the effective coefficients

$$b^{\alpha,l}=\frac{b_1^{\alpha,l}(c_{12}^{\alpha}+\sqrt{2}c_{22}^{\alpha})-b_2^{\alpha,l}(c_{11}^{\alpha}+\sqrt{2}c_{12}^{\alpha})}{c_{11}^{\alpha}+2\sqrt{2}c_{12}^{\alpha}+2c_{22}^{\alpha}} \text{ with } l=0,2 . \tag{9}$$

The effective coefficient $b^{\alpha,2}$ will depend on the thickness in a way which may be derived from Eqs. (4) and (5):

$$b^{\alpha,2}=\frac{-B^{\gamma,2}C^{\alpha}+(2/t_f)[C^{\gamma}\sqrt{2}b_1^{\alpha,2}-C^{\alpha}b_2^{\alpha,2}]+(2/t_f)^2(\cdots)}{C^{\alpha}+2C^{\gamma}+(2/t_f)[c_{11}^{\alpha,s}+2\sqrt{2}c_{12}^{\alpha,s}+2c_{22}^{\alpha,s}]} , \tag{10}$$

and one sees that the thickness dependence of the elastic coefficients also plays an important role in the thickness dependence of this effective coefficient.

## IV. EXPERIMENTAL METHODS FOR STUDYING MAGNETOELASTIC EFFECTS IN THIN FILMS

We shall now discuss four experimental methods which have been employed up to now for determining SMC coefficients in thin films. In these four methods, phenomenological magnetoelastic coupling coefficients, namely, $b^{\gamma,2}$ and the effective coefficient $b^{\alpha,2}$, may be experimentally determined from in-plane and out-of-plane measurements, and the SMC coefficients may be derived from their thickness dependence. In any case, the substrate will be assumed to be thicker than the film.

In the first method, a film is strained by pulling on its substrate, generally a polyimide ribbon or a glass platelet:[4] The magnetoelastic properties are deduced from the induced magnetic anisotropy given by Eqs. (7) and (8).

A similar and promising method has been recently proposed by Bochi, Song, and O'Handley:[7] Instead of artificially created strains, the stress-induced anisotropy is due to the spontaneous strains observed in as-prepared films. These may be derived from the curvature of the substrate after film growth.

We shall now present, in more details, the last two methods, namely, the strain-modulated ferromagnetic resonance (SMFMR) and the magnetoelastic cantilever, since errors seem to have occurred in the recent past when analyzing both these techniques.

The strain-modulated ferromagnetic resonance allows the magnetoelastic coupling coefficients to be derived from the shift of the resonance frequency induced by applying a stress to any sample.[14] This method has been often applied to thin films.[6]

We shall consider two geometries where the static magnetic field $H_0$ will lie in the $xOy$ plane and the rf magnetic field $h_z$, along the $Oz$ axis (see Fig. 1): In the first case, the film is perpendicular to $h_z$, and in the second case, it lies in the $xOz$ plane (see Fig. 1). In both cases, *uniaxial stress $\sigma$ is applied along the Ox direction.* The substrate is strained, with the following in-plane components: $\varepsilon_{xx}=s_{11}^{s}\sigma$, $\varepsilon_{yy}=s_{12}^{s}\sigma$, and $\varepsilon_{xy}=0$. We start from Eq. (7), using these strain components, dropping all the nonmagnetic terms and the magnetoelastic contribution to the magnetic anisotropy ($\approx b^2/c$). Let us assume the direction cosines of the static magnetic field $H_0$ to be $\{\cos\phi_H, \sin\phi_H, 0\}$ and those of the magnetization $\{\sin\vartheta\cos\phi, \sin\vartheta\sin\phi, \cos\vartheta\}$. The free energy density is

$$F=-\mu_0 MH\sin\vartheta\cos(\phi-\phi_H)+A\cos2\vartheta +B(1-\cos2\vartheta)\cos2\phi+C^t , \tag{11}$$

with

$$A_\perp=\tfrac{1}{4}\mu_0 M_s^2+\tfrac{1}{2}k_2+\tfrac{3}{4}b^{\alpha,2}(s_{11}^{s}+s_{12}^{s})\sigma ,$$
$$B_\perp=\tfrac{1}{4}b^{\gamma,2}(s_{11}^{s}-s_{12}^{s})\sigma , \tag{12}$$

for the first geometry (film perpendicular to $h_z$), and

$$A_\parallel=-\tfrac{1}{2}(A_\perp+3B_\perp),\quad B_\parallel=-\tfrac{1}{2}(A_\perp-B_\perp) , \tag{13}$$

for the second geometry ($h_z$ in film plane).

The resonance frequency has been given by Smit and Beljers:[15]

$$\left[\frac{\omega}{\gamma}\right]^2=\frac{1}{\mu_0^2 M_s^2\sin^2\vartheta}\left\{\left[\frac{\partial^2 F}{\partial\vartheta^2}\right]\left[\frac{\partial^2 F}{\partial\phi^2}\right]-\left[\frac{\partial^2 F}{\partial\vartheta\partial\phi}\right]^2\right\} . \tag{14}$$

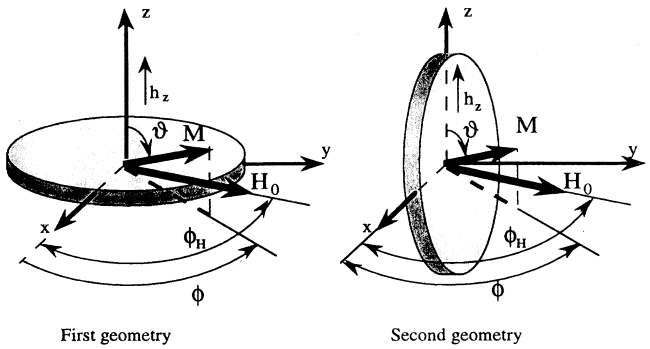


FIG. 1. Two different possible orientations of the magnetization (**M**), the static magnetic field ($\mathbf{H}_0$), and the hyperfrequence magnetic field ($\mathbf{h}_z$) in strain-modulated ferromagnetic resonance experiments.

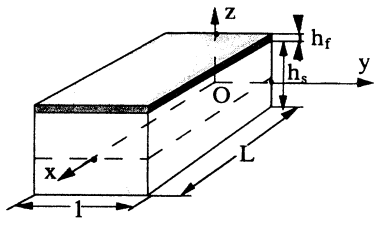


FIG. 2. Definition of the axes in an experiment for observing the deflection of a magnetostrictive bimorph.

We may derive the analytic solutions in the following specific cases.

(i) For $H_0 \| Ox$, we find the same resonance frequency in both geometries, namely,

$$\left[\frac{\omega}{\gamma}\right]^2 = \left\{H_0 + M_s + \frac{2k_2}{\mu_0 M_s} + \frac{3b^{\alpha,2}(s^s_{11}+s^s_{12}) - b^{\gamma,2}(s^s_{11}-s^s_{12})}{\mu_0 M_s}\sigma\right\}\left\{H_0 - \frac{2b^{\gamma,2}(s^s_{11}-s^s_{12})}{\mu_0 M_s}\sigma\right\} . \tag{15}$$

(ii) For $H_0 \| Oy$, in the first geometry, one gets

$$\left[\frac{\omega}{\gamma}\right]^2 = \left\{H_0 + M_s + \frac{2k_2}{\mu_0 M_s} + \frac{3b^{\alpha,2}(s^s_{11}+s^s_{12}) + b^{\gamma,2}(s^s_{11}-s^s_{12})}{\mu_0 M_s}\sigma\right\}\left\{H_0 + \frac{2b^{\gamma,2}(s^s_{11}-s^s_{12})}{\mu_0 M_s}\sigma\right\} . \tag{16}$$

Only the sign before $b^{\gamma,2}$ is changed with respect to (i).

(iii) For $H_0 \| Oy$, in the second geometry, one gets

$$\left[\frac{\omega}{\gamma}\right]^2 = \left\{H_0 - M_s - \frac{2k_2}{\mu_0 M_s} - \frac{3b^{\alpha,2}(s^s_{11}+s^s_{12}) - b^{\gamma,2}(s^s_{11}-s^s_{12})}{\mu_0 M_s}\sigma\right\} \times \left\{H_0 - M_s - \frac{2k_2}{\mu_0 M_s} - \frac{3b^{\alpha,2}(s^s_{11}+s^s_{12}) + b^{\gamma,2}(s^s_{11}-s^s_{12})}{\mu_0 M_s}\sigma\right\} . \tag{17}$$

The elastic coefficients of the substrate ($s^s_{11}$ and $s^s_{12}$) enter these formulas, contrary to several papers where the resonance conditions have been expressed in terms of the *magnetostriction coefficients* of the film (Ref. 6 and references cited therein), which is incorrect: Here the film is stressed through the substrate and cannot expand freely.

We note that such resonance experiments do not provide all the magnetoelastic coupling coefficients, but only the $b^{\gamma,2}$ one and the effective $b^{\alpha,2}$ coefficient given by Eq. (9).

### A. Deflection of a bimorph

A bimorph made up of a magnetic film and its substrate is known to bend under the influence of an applied magnetic field. Klokholm has proposed that the deflection be used to derive the magnetostriction coefficient of the film;[16] recently, several authors have corrected his analysis of this cantilever method.[17] Again, we must point out that it is incorrect to speak of *magnetostriction* since the active element is the film and the passive one is the substrate: only *magnetoelastic coupling coefficients* may be determined from such experiments.

Extending a calculation developed in our previous paper for isotropic films where surface effects were not considered,[17] we give the general expression of the deflection for an isotropic thin film, starting from Eq. (7):

$$U_z = 3\frac{h_f}{h_s^2}\frac{1-\nu_s}{E_s}\frac{b^{\alpha,0}}{\sqrt{2}}\left[x^2+y^2+\frac{\nu_s}{1-\nu_s}z^2\right] + \frac{9}{2}\frac{h_f}{h_s^2}\frac{1-\nu_s}{E_s}b^{\alpha,2}\left[x^2+y^2+\frac{\nu_s}{1-\nu_s}z^2\right]\left[\alpha_3^2-\frac{1}{3}\right] + \frac{3}{2}\frac{h_f}{h_s^2}\frac{1+\nu_s}{E_s}b^{\gamma,2}\{(x^2-y^2)(\alpha_1^2-\alpha_2^2)+4xy\alpha_1\alpha_2\} , \tag{18}$$

where $Oz$ is the axis perpendicular to the film plane, $h_f$ and $h_s$ the thickness of the film and the substrate, respectively, and $\nu_s$ Poisson's ratio of the substrate (see Fig. 2). The deflection $U_z$ represents the vertical displacement of any material point $(x,y,z)$ in the film, which is assumed to be horizontal and fixed at its end $(0,0,0)$. The deflection $D$ is maximum at the other end, say, $x=L$. For $y=z=0$, one finds

$$D = \frac{3}{2}\frac{h_f}{h_s^2}\frac{L^2}{E_s}\left\{(1-\nu_s)\left[\frac{2b^{\alpha,0}}{\sqrt{2}}+3b^{\alpha,2}\left[\alpha_3^2-\frac{1}{3}\right]\right] + (1+\nu_s)b^{\gamma,2}(\alpha_1^2-\alpha_2^2)\right\} . \tag{19}$$

Again, this experiment gives only the $b^{\gamma,2}$ magnetoelastic coupling coefficient from the in-plane angular dependence of the deflection and the effective $b^{\alpha,2}$ coefficient from its out-of-plane angular dependence.

### B. Case of a thicker film

We consider here a film where surface effects become negligible, but which remains, however, always much thinner than its substrate. The film becomes isotropic, thus giving $b^{\gamma,2}=B^{\gamma,2}$,

$$\frac{b^{\alpha,0}}{\sqrt{2}} = B^{\alpha,0}\frac{C^\gamma}{C^\alpha+2C^\gamma} = B^{\alpha,0}\frac{1-2\nu_f}{3(1-\nu_f)} \quad \text{and} \quad b^{\alpha,2} = -B^{\gamma,2}\frac{C^\alpha}{C^\alpha+2C^\gamma} = -B^{\gamma,2}\frac{1+\nu_f}{3(1-\nu_f)} , \tag{20}$$

where $\nu_f$ is Poisson's ratio for the film. Equation (19) may then be rewritten

$$D=\frac{h_f}{h_s^2}\frac{L^2}{E_s}\left\{B^{\alpha,0}\frac{(1-\nu_s)(1-2\nu_f)}{(1-\nu_f)}+\frac{3}{2}B^{\gamma,2}\left[(1+\nu_s)(\alpha_1^2-\alpha_2^2)-(1-\nu_s)\frac{(1+\nu_f)}{(1-\nu_f)}\left[\alpha_3^2-\frac{1}{3}\right]\right]\right\}. \tag{21}$$

The static deflection associated with the isotropic exchange magnetoelastic coupling coefficient $B^{\alpha,0}$ will be neglected hereafter. The deflection of the bimorph, which depends on the magnetization direction, is zero in an isotropic demagnetized state ($\langle\alpha_i^2\rangle=\frac{1}{3}$ and $\langle\alpha_i\alpha_j\rangle=0$). Otherwise, it is

$$D_{\parallel}=2\frac{B^{\gamma,2}}{E_s}\frac{h_fL^2}{h_s^2}\frac{(1-\nu_f\nu_s)-\frac{1}{2}(\nu_f-\nu_s)}{1-\nu_f}\quad\text{for }\alpha_1^2=1\ , \tag{22}$$

$$D_{\perp}=-\frac{B^{\gamma,2}}{E_s}\frac{h_fL^2}{h_s^2}\frac{(1-\nu_f\nu_s)-2(\nu_f-\nu_s)}{1-\nu_f}\quad\text{for }\alpha_2^2=1\ , \tag{23}$$

$$D_0=-\frac{B^{\gamma,2}}{E_s}\frac{h_fL^2}{h_s^2}\frac{(1-\nu_f\nu_s)+(\nu_f-\nu_s)}{1-\nu_f}$$

$$\text{for }\alpha_3^2=1\ \text{(out of plane)}\ . \tag{24}$$

One may verify that formulas (22) and (23) are just the two given in our previous paper[17] and that $D_{\parallel}+D_{\perp}+D_0=0$ and $D_{\perp}=D_0$, when $\nu_f=\nu_s$ (elastic isotropy).

On the other hand, if large spontaneous stresses induce a perpendicular magnetic anisotropy, the demagnetized state is defined by either $\alpha_3^2=1$ or $\alpha_3=0$ depending on the sign of the stresses and of the $B^{\gamma,2}$ coefficient. In the former case, the initial magnetostrictive deflection is just the one which would be observed if the film were magnetized perpendicular to its plane, i.e., $D_0$. Then the deflection of the bimorph observed when a magnetic field is applied in the plane becomes

$$\begin{aligned}D_{\parallel}-D_0&=3\frac{B^{\gamma,2}}{E_s}\frac{h_fL^2}{h_s^2}\frac{(1-\nu_f\nu_s)}{1-\nu_f}\ ,\\ D_{\perp}-D_0&=3\frac{B^{\gamma,2}}{E_s}\frac{h_fL^2}{h_s^2}\frac{(\nu_f-\nu_s)}{1-\nu_f}\ .\end{aligned} \tag{25}$$

*$D_{\perp}-D_0$ may exhibit the same sign as $D_{\parallel}-D_0$* provided Poisson's ratio of the film is larger than the one of the substrate: This is *different from the case of bulk samples.*

In the latter case, the magnetic moments lie in the film plane ($\alpha_3=0$) and if there is no in-plane anisotropy $\langle\alpha_1^2\rangle=\langle\alpha_2^2\rangle$ and the initial magnetostrictive deflection is now $D_0'=-D_0/2$. Then the deflections of the bimorph observed when a magnetic field is applied in the plane are very different:

$$\begin{aligned}D_{\parallel}-D_0'=-(D_{\perp}-D_0')&=\frac{3}{2}\frac{B^{\gamma,2}}{E_s}\frac{h_fL^2}{h_s^2}(1+\nu_s)\\ &=\frac{3}{4}\frac{B^{\gamma,2}}{G_s}\frac{h_fL^2}{h_s^2}\ .\end{aligned} \tag{26}$$

These are the two peculiar situations which may occur often as a result of the very large spontaneous stresses generally observed in thin films. The other extreme situation would be that of a film with anisotropic in-plane stresses, creating an easy magnetization direction parallel to $Ox$: Then one would observe $D_{\parallel}=0$.

In any case, $D_{\parallel}-D_{\perp}$ remains always independent of the initial state and provides $B^{\gamma,2}$, provided the shear modulus $G_s$ of the substrate is known:

$$D_{\parallel}-D_{\perp}=\frac{3}{2}\frac{B^{\gamma,2}}{G_s}\frac{h_fL^2}{h_s^2}\ . \tag{27}$$

### C. Conclusion

Only one of the four magnetoelastic coupling coefficients defined in Eq. (3), namely, $b^{\gamma,2}$ ($B^{\gamma,2}$ for thick films), may be determined by the four methods presented above. The measurement of $b^{\alpha,2}$ requires the magnetic film to be saturated under an applied magnetic field perpendicular to its plane. Although $b^{\alpha,2}$ is only an effective coefficient, *$b^{\alpha,2}$ and $b^{\gamma,2}$ will be the relevant parameters for predicting the performances of any actuator or sensor using magnetoelastic isotropic thin films.*

## V. LOWER SYMMETRIES

We shall consider now thin films of various symmetries: The lower the symmetry, the higher the number of coefficients necessary for describing the thermodynamic potential.

For instance, there are three magnetoelastic coefficients ($B^{\alpha,0}$, $B^{\gamma,2}$, and $B^{\varepsilon,2}$) for a crystal belonging to the $C_I$ cubic Laue group; the number of magnetoelastic coupling coefficients describing such crystalline thin films will be 7 ($b_1^{\alpha,0}$, $b_2^{\alpha,0}$, $b_1^{\alpha,2}$, $b_2^{\alpha,2}$, $b^{\gamma,2}$, $b^{\delta,2}$, and $b^{\varepsilon,2}$) for a (001) thin film (tetragonal symmetry), 6 ($b_1^{\alpha,0}$, $b_2^{\alpha,0}$, $b_1^{\alpha,2}$ $b_2^{\alpha,2}$, $b^{\varepsilon,2}$, and $b^{\zeta,2}$) for a (111) thin film (hexagonal symmetry), and 12 ($b_1^{\alpha,0}$, $b_2^{\alpha,0}$, $b_3^{\alpha,0}$, $b_1^{\alpha,2}$, $b_2^{\alpha,2}$, $b_3^{\alpha,2}$, $b_1^{\alpha,2'}$, $b_2^{\alpha,2'}$, $b_3^{\alpha,2'}$, $b^{\beta,2}$, $b^{\gamma,2}$, and $b^{\delta,2}$) for a ($1\bar{1}0$) thin film (orthorhombic symmetry). This latter case has been recently treated for magnetic multilayers within the microscopic pair model, but exchange contributions ($b_1^{\alpha,0}$, $b_2^{\alpha,0}$, $b_3^{\alpha,0}$) have not been considered because only pseudodipolar interactions have been introduced.[18]

In the same way, there are seven magnetoelastic coupling coefficients for a crystal belonging to the $T_I$ tetragonal Laue group: $B_1^{\alpha,0}$, $B_2^{\alpha,0}$, $B_1^{\alpha,2}$, $B_2^{\alpha,2}$, $B^{\gamma,2}$, $B^{\delta,2}$, and $B^{\varepsilon,2}$; the same is true for $T_I$ crystalline thin films cut in a (001) plane, since cutting this crystal in such a plane does not lower the symmetry. However, they will be 12 for (100) and ($1\bar{1}0$) planes (orthorhombic symmetry).

A (0001) thin film for a crystal belonging to the $H_I$ hexagonal Laue group will exhibit six coefficients since

the symmetry will remain the same as for the bulk material.

The complete description of the magnetoelastic coupling energies relevant to these symmetries as well as all the above coefficients can be found elsewhere.[11] For any symmetry, surface effects may split the coefficients relevant to the symmetry of the bulk material and/or add new coefficients, in a way quite similar to the one described above for isotropic thin films. For each specific case, it is necessary to introduce the condition of constant in-plane strains and then derive the equilibrium energy: Again, effective magnetoelastic coupling coefficients will appear for the modes belonging to the $\alpha(\Gamma_1)$ representation.

Contrary to the case of bulk crystals, the exchange magnetoelastic coupling coefficients ($b_i^{\alpha,0}$) are difficult to detect from anomalies in thermal expansion as a result of the huge stresses which appear when heating the samples, which are due to the differential thermal expansions of the film and the substrate.

## VI. MULTILAYERS

To illustrate the case of a crystalline magnetic film, we shall now deal with a multilayer consisting of a succession of magnetic and nonmagnetic (001) films of cubic symmetry. There are $2p$ interfaces, between $p$ magnetic films (thickness $t_m$) and $p$ nonmagnetic ones (thickness $t_{nm}$), taking into account the protective film (thickness $t_{nm}$ also) and the nonmagnetic much thicker substrate. The ratio $u$ of the magnetic volume $V_m$ to the total volume $V$ is $u=V_m/V=t_m/(t_m+t_{nm})$. The total thickness of the multilayer, without the substrate, is equal to $p(t_m+t_{nm})$ and remains small as compared with the length and width of the sample.

The symmetry of such a film is tetragonal. Choosing the [001] direction as the $Oz$ axis, we may write the free energy density for such a multilayer divided by $u$, i.e., the energy per unit volume *of the magnetic part* of the sandwich:

$$
\begin{aligned}
E/V_m = {} & b_1^{\alpha,0}\frac{1}{3}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})+b_2^{\alpha,0}\frac{\sqrt{2}}{3}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right] \\
& +\left[\frac{1}{2}\mu_0 M_s^2-k_2+b_1^{\alpha,2}\frac{\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}}{\sqrt{2}}+b_2^{\alpha,2}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]\right]\left[\alpha_3^2-\frac{1}{3}\right] \\
& +\frac{1}{2}b^{\gamma,2}(\varepsilon_{xx}-\varepsilon_{yy})(\alpha_1^2-\alpha_2^2)+2b^{\delta,2}\varepsilon_{xy}\alpha_1\alpha_2+2b^{\varepsilon,2}(\varepsilon_{yz}\alpha_2\alpha_3+\varepsilon_{zx}\alpha_3\alpha_1) \\
& +\frac{1}{6}c_{11}^{\alpha}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})^2+\frac{\sqrt{2}}{3}c_{12}^{\alpha}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right] \\
& +\frac{1}{3}c_{22}^{\alpha}\left[\varepsilon_{zz}-\frac{\varepsilon_{xx}+\varepsilon_{yy}}{2}\right]^2+\frac{1}{4}c^{\gamma}(\varepsilon_{xx}-\varepsilon_{yy})^2+c^{\delta}\varepsilon_{xy}^2+c^{\varepsilon}(\varepsilon_{yz}^2+\varepsilon_{zx}^2)\,. \qquad (28)
\end{aligned}
$$

All the above coefficients may be expressed in terms of surface coefficients, as for isotropic substances [see Eqs. (4) and (5)]. One gets

$$
\begin{aligned}
& k_2=\frac{2}{t_m}k^s,\quad b_1^{\alpha,0}=B^{\alpha,0}+\frac{2}{t_m}b_0^{\alpha,s},\quad b_2^{\alpha,0}=\frac{2}{t_m}b_0'^{\alpha,s}\,, \\
& b_1^{\alpha,2}=\frac{2}{t_m}b_1^{\alpha,s},\quad b_2^{\alpha,2}=B^{\gamma,2}+\frac{2}{t_m}b_2^{\alpha,s},\quad b^{\gamma,2}=B^{\gamma,2}+\frac{2}{t_m}b^{\gamma,s}\,, \\
& b^{\delta,2}=B^{\varepsilon,2}+\frac{2}{t_m}b^{\delta,s},\quad b^{\varepsilon,2}=B^{\varepsilon,2}+\frac{2}{t_m}b^{\varepsilon,s},\quad c_{11}^{\alpha}=C^{\alpha}+\frac{2}{t_m}c_{11}^{\alpha,s}\,, \qquad (29) \\
& c_{12}^{\alpha}=\frac{2}{t_m}c_{12}^{\alpha,s},\quad c_{22}^{\alpha}=C^{\gamma}+\frac{2}{t_m}c_{22}^{\alpha,s},\quad c^{\gamma}=C^{\gamma}+\frac{2}{t_m}c^{\gamma,s}\,, \\
& c^{\delta}=C^{\varepsilon}+\frac{2}{t_m}c^{\delta,s},\quad c^{\varepsilon}=C^{\varepsilon}+\frac{2}{t_m}c^{\varepsilon,s}\,.
\end{aligned}
$$

It is preferable to express the energy density with respect to the magnetic volume rather than to the total volume, because the $B^{\alpha,0}$, $B^{\gamma,2}$, $B^{\varepsilon,2}$, $C^{\alpha}$, $C^{\gamma}$ and $C^{\varepsilon}$ coefficients remain those of the bulk pure magnetic material.

As for the isotropic thin film, we shall consider that $\varepsilon_{xx}$, $\varepsilon_{yy}$, and $\varepsilon_{xy}$ are imposed by the substrate and derive the equilibrium values for the remaining strain components. We find $\varepsilon_{yz}=-(b^{\varepsilon,2}/c^{\varepsilon})\alpha_2\alpha_3$, $\varepsilon_{zx}=-(b^{\varepsilon,2}/c^{\varepsilon})\alpha_3\alpha_1$, and for $\varepsilon_{zz}$, the same expression as Eq. (6). Substituting these equilibrium values in Eq. (28), one gets

$$E/V_m = \left\{\frac{1}{2}\mu_0 M_s^2 - k_2\right\}\left[\alpha_3^2 - \frac{1}{3}\right] - \frac{1}{6}\frac{[b_1^{\alpha,0} + \sqrt{2}b_2^{\alpha,0} + (3/\sqrt{2})(b_1^{\alpha,2} + \sqrt{2}b_2^{\alpha,2})(\alpha_3^2 - \frac{1}{3})]^2}{c_{11}^{\alpha} + 2\sqrt{2}c_{12}^{\alpha} + 2c_{22}^{\alpha}} - \frac{[b^{\varepsilon,2}]^2}{c^{\varepsilon}}(\alpha_3^2 - \alpha_3^4)$$
$$+ \frac{\varepsilon_{xx} + \varepsilon_{yy}}{\sqrt{2}} b^{\alpha,0} + \frac{3}{2}(\varepsilon_{xx} + \varepsilon_{yy}) b^{\alpha,2}\left[\alpha_3^2 - \frac{1}{3}\right] + b^{\gamma,2}\frac{1}{2}(\varepsilon_{xx} - \varepsilon_{yy})(\alpha_1^2 - \alpha_2^2) + 2b^{\delta,2}\varepsilon_{xy}\alpha_1\alpha_2$$
$$+ \frac{3}{4}(\varepsilon_{xx} + \varepsilon_{yy})^2 \frac{c_{11}^{\alpha} c_{22}^{\alpha} - [c_{12}^{\alpha}]^2}{c_{11}^{\alpha} + 2\sqrt{2}c_{12}^{\alpha} + 2c_{22}^{\alpha}} + \frac{1}{4}c^{\gamma}(\varepsilon_{xx} - \varepsilon_{yy})^2 + c^{\delta}\varepsilon_{xy}^2 \ . \tag{30}$$

One finds again various contributions to the magnetic anisotropy, namely, the dipolar shape anisotropy, the magnetic anisotropy of the unstrained body ($k_2$), and two magnetoelastic contributions to the magnetocrystalline anisotropy (in $\alpha_3^2$ and $\alpha_3^4$).

The stress-induced anisotropy exhibits in-plane and out-of-plane components, given by the above equation, where the strain components $\varepsilon_{xx}$, $\varepsilon_{yy}$, and $\varepsilon_{xy}$ are given by Eq. (8). As for isotropic thin films, the magnetoelastic coupling coefficients belonging to the $\alpha(\Gamma_1)$ representation may not be determined independently, but only the effective coefficient $b^{\alpha,2}$ given by Eqs. (9) and (10) is associated with out-of-plane anisotropy. On the other hand, for in-plane stress-induced anisotropy, two coefficients must be considered now, since this anisotropy will be different for a stress applied along a [100] direction ($b^{\gamma,2}$ coefficient) or a [110] direction ($b^{\delta,2}$ coefficient). Hence, for a (001) thin film of cubic symmetry rigidly fixed onto a substrate, three coefficients are relevant to the problem of stress-induced anisotropy, $b^{\gamma,2}$, $b^{\delta,2}$, and the effective $b^{\alpha,2}$ coefficient, and not only two as sometimes found in the literature.

## VII. CONCLUSIONS

In the framework of the theory of Callen and Callen of magnetostriction, the thermodynamical potential relevant to thin films rigidly fixed onto a thick substrate has been given for various crystalline symmetries and crystallographic planes. The number of SMC coefficients is larger than the number of magnetoelastic coupling coefficients of the bulk material, but only a few of them contribute to the magnetic anisotropy induced by in-plane stresses. The SMC coefficients belonging to the $\Gamma_1(\alpha)$ representation contribute to the energy density only through linear combinations (effective coefficients). A magnetoelastic contribution to the magnetocrystalline anisotropy arises from the minimization of the free energy under the boundary conditions at the interface with the substrate: It provides a contribution to the out-of-plane uniaxial anisotropy.

SMC coefficients may be derived from the $1/t_m$ dependence of the stress-induced anisotropy: This requires excellent samples to be prepared with densities as high as possible, because porosity would reduce magnetization, introduce other surfaces, and hence modify also the values of the magnetoelastic coupling coefficients. Evidently, the $1/t_m$ dependence of the magnetoelastic coupling coefficients must not be masked by any spurious thickness dependence of the magnetization.


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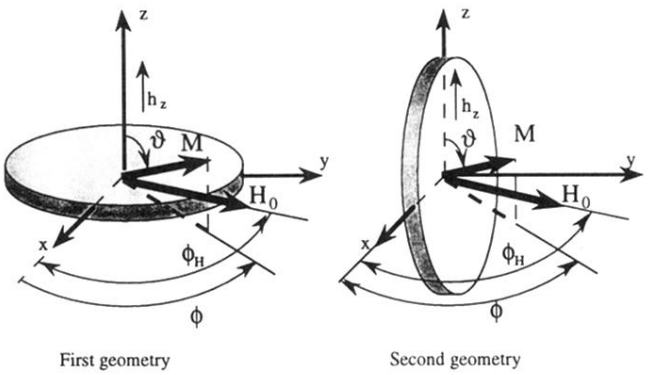


First geometry Second geometry

FIG. 1. Two different possible orientations of the magnetization ($\mathbf{M}$), the static magnetic field ($\mathbf{H}_0$), and the hyperfrequence magnetic field ($\mathbf{h}_z$) in strain-modulated ferromagnetic resonance experiments.

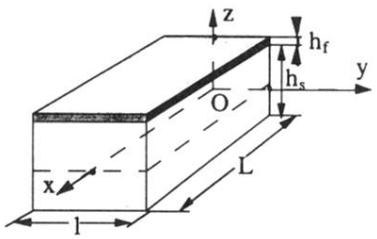


FIG. 2. Definition of the axes in an experiment for observing the deflection of a magnetostrictive bimorph.