Ferromagnetic Resonance in Polycrystalline Magnetostrictive Films with Large Isotropic Planar Strain

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(Received 10 January 1973)

Generally, in a magnetic material the magnetostriction constants λ_{100} and λ_{111} are quite different. In the present paper we take this fact into account in a new calculation of the ferromagnetic resonance in polycrystalline magnetostrictive films under large isotropic strain. The results are quite different from those predicted by the MacDonald theory, where it was supposed that $\lambda_{100} = \lambda_{111}$. The calculation presented here is based on the independent-grain approach with randomly oriented grains and is expected to be valid when the strain-induced anisotropy field is of the same order of magnitude or is larger than the saturation magnetization. The calculation is developed for the case when the applied field is perpendicular to the plane of the film, and it is assumed that the dc field is large enough to keep the transverse components of the magnetization one order of magnitude lower than the longitudinal one. An analytical study shows that the absorption line presents singularities arising from those values of the resonance field that are stationary with respect to grain orientation. The singularities are located near H_{100} , H_{110} , and H_{111} , which are the resonance fields corresponding, respectively, to crystallites with [100], [110], and [111] direction along the applied field. Near H_{110} the absorption line shows a logarithmic singularity; the condition that a secondary peak should be resolved near H_{100} is determined. The results are illustrated by a numerical computation of the resonance line where the strain-induced anisotropy field, the elastic constants, and the crystalline anisotropy are varied independently. Finally, it is shown that the contribution of the crystalline anisotropy to the total hnewidth can be completely canceled out for a given value of the strain-induced anisotropy. The theory explains several results obtained in various magnetostrictive thin films, which are listed in the paper.

I. INTRODUCTION

It is well known that when a ferromagnetic or ferrimagnetic material is submitted to a strain (or stress), a strain-induced anisotropy field is created through the phenomenon of magnetostriction. In single-crystal samples submitted to an uniaxial stress or to isotropic planar strain (or stress), the effect of strain (or stress) on ferromagnetic-resonance (FMR) experiments has been frequently studied.¹⁻⁴ It has been clearly established that when the strain (or stress} is applied along a given symmetry axis, one observes a shift δH in the magnetic field required for resonance. In particularly, uniaxial stress was applied to bulk single crystals, and the λ_{100} and λ_{111} magnetostriction constants were deduced from δH with good accura $cy.$ ¹⁻³ Planar strain (or stress) are generally observed in a thin film deposited on a substrate of dissimilar material. The planar strain is assumed to be isotroyic if the film is perfectly adhered to the substrate. In single-crystal yttrium iron garnet (YIG) films the calculated and measured stressinduced field perpendicular to the film plane-deduced from δH -were in good agreement.⁴ In polycrystalline samples the situation has been much less clear, although isotropic planar strains induced by the thermal expansion mismatch between film and substrate were frequently observed by FMR in magnetostrictive thin films. 5 In previou experimental works, strain was only taken into account when the strain-induced field was particularly high, and in these cases the theory proposed by MacDonald was used. $6,7$

In this calculation the isotropic planar stress was obtained by averaging the stress contribution over all possible directions of the crystalline axes and by using the approximation of isotropic magnetostriction $\lambda_{100} = \lambda_{111}$. With this approximation, the effective demagnetizing field which is to be inserted in the well-known Kittel resonance formula is given by $4\pi M_{\text{eff}} = 4\pi M + 3\lambda_s \sigma/M$, where σ is the planar stress, λ_s is the magnetostriction constant of the polycrystalline sample, and M is the saturation magnetization. This calculation predicts for a polycrystalline sample a shift in the magnetic field required for resonance, but the resonance line shape and the corresponding linewidth should remain unaffected.

Recently new experimental results were obtained in polycrystalline $\mathrm{NiFe_{2}O_{4}}$ thin films with large isotropic strain.^{8,9} The experimental data could not be explained on the basis of MacDonald's calculation. Therefore a new calculation has been developed in which the fact that in most of magnetic materials λ_{100} is quite different from λ_{111} is taken into account. When $\lambda_{100} \neq \lambda_{111}$ MacDonald's theory is no longer valid and the shift of the resonance line and its shape is quite different than predicted, particularly for large strains.

For a correct formulation of the problem it is necessary to calculate first the anisotropy energy

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induced by isotropic planar strain when λ_{100} and λ_{111} are different. Using this expression the resonance line can be calculated-in the same way as was proposed for polycrystalline samples with crystalline anisotropy'0-by two basically different approaches. In the first method the dipolar interaction between the different parts of the sample is taken into account and the stress induced anisotropy field δH is considered as a small perturbation. This calculation is expected to be valid when δH is much smaller than $4\pi M_0$. On the other hand, the independent grain approximation can be used when the condition $|\delta H| \geq 4\pi M_0$ is satisfied. In this case-following Schlömann's assumptions¹⁰-dipolar interaction between grains can be neglected, each particular grain goes through resonance independently, and the resonance field corresponding to a given grain depends on its orientation with respect to the applied field.

The aim of the present paper is to present a new calculation of the resonance line using this last hypothesis. The calculation is developed for polycrystalline films of cubic structure, with the applied dc field parallel to the film normal. The spirit of the calculation is similar to that given by Schlömann^{11, 12} for polycrystalline materials with large anisotropy. First the magnetic field necessary for resonance is calculated for a particular grain. In a second step a distribution function $A(H)$ is calculated by integration and it is assumed that the absorption spectra $P(H)$ depends on the applied field in the same way as $A(H)$. Here $A(H)$ is defined as given by Schlömann¹¹: " $A(H)dH$ is proportional to the number of grains which have their resonance in the range of applied dc field H and $H+dH$." The results are presented in the following order: In Sec. II we develop the expression of the modified magnetoelastic energy in presence of isotropic planar strains. It will be shown, in particular, that the strain and stress hypothesis give the same results only when the material is elastically isotropic. In Sec. III, the calculation of the resonance equation and the corresponding absorption spectra is presented. The validity of the approximation used in the calculation is discussed. It is shown that the resonance line presents singularities, as in the case of ferrites with large crystalline anisotropy. The singularities are located at those values of the resonance field, that are stationary with respect to grain orientation. Their behavior is discussed based on an analytical study given in Appendix B. The magnetocrystalline anisotropy will be included also in the resonance formula. In Sec. IV, the results of some numerical calculations are presented to illustrate the results of Sec. III, for different values of the strain-induced anisotropy, of the crystalline anisotropy, and of the elastic constants. An important conse-

FIG. 1. The primed coordinates $x'y'z'$ correspond, respectively, to the $[100]$, $[010]$, and $[001]$ cubic crystal axes. The unprimed coordinates xyz correspond to the film axes, where the film is normal and the applied dc field are z directed.

quence of the results obtained is developed here: For a given value of the stress the total anisotropy, which is the sum of the magnetocrystalline and the stress induced anisotropy, cancels out and the linewidth can be reduced to that corresponding to a single crystal. Finally, in Appendix A a new formulation of Snit' s generalized resonance formula is given in a Cartesian coordinate system.

II. CALCULATION OF STRAIN-INDUCED ANISOTROPY ENERGY

We wish to calculate the magnetoelastic energy density E_{me} of a particular grain in a polycrystalline film subject to externally applied isotropic planar strain. The calculation applies for crystals of cubic symmetry. First we introduce two Cartesian coordinate systems x, y, z and x', y' , z' as shown in Fig. 1 with principal axes of the particular grain along the $x'y'z'$ direction. Let the x, y, z direction corresponding to the film coordinate with the film normal (axis normal to the plane where strain is applied) along the z axis, and the x, ν axes in the film plane. When the direction of the film normal is defined by the polar angle α measured from [001] and by the azimuthal angle β measured from $[100]$ the following rotation matrix can be determined:

$$
a_{ij} = \begin{pmatrix} \cos\alpha \cos\beta & -\sin\beta & \sin\alpha \cos\beta \\ \cos\alpha \sin\beta & \cos\beta & \sin\alpha \sin\beta \\ -\sin\alpha & 0 & \cos\alpha \end{pmatrix}.
$$
 (1)

In (1) the direction cosines of the normal to the film are given by the a_{i3} values. For the case of a cubic crystal the magnetoelastic energy in the x' $y'z'$ coordinate system is

$$
E'_{\mathbf{m}\mathbf{e}} = B_1 (\epsilon'_{11} \alpha'^2_1 + \epsilon'_{22} \alpha'^2_2 + \epsilon'_{33} \alpha'^2_3)
$$

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$$
+2B_2(\epsilon'_{12}\alpha'_1\alpha'_2+\epsilon'_{13}\alpha'_1\alpha'_3+\epsilon'_{23}\alpha'_2\alpha'_3), \qquad (2)
$$

where B_1 and B_2 are the magnetoelastic coupling constants related to λ_{100} and λ_{111} by the familiar equations $B_1 = -\frac{3}{2}(C_{11} - C_{12})\lambda_{100}$ and $B_2 = -3C_{44}\lambda_{111}$,
 $\epsilon_{11} = \epsilon_{x^2x^2}$ and $\epsilon_{12} = \epsilon_{x^2y^2}$ are the components of the strain tensor as defined by Landau,¹³ the α'_{i} are the direction cosines of the magnetization relative to the cubic crystal axes, and ϵ_{ij} and α_i are the values relative to the unprimed coordinates. The elastic energy is given by 13

$$
E'_{e} = \frac{1}{2}C_{11}(\epsilon_{11}^{\prime 2} + \epsilon_{22}^{\prime 2} + \epsilon_{33}^{\prime 2}) + C_{12}(\epsilon_{11}^{\prime} \epsilon_{22}^{\prime} + \epsilon_{11}^{\prime} \epsilon_{33}^{\prime} + \epsilon_{22}^{\prime} \epsilon_{33}^{\prime})
$$

$$
+ 2C_{44}(\epsilon_{12}^{\prime 2} + \epsilon_{13}^{\prime 2} + \epsilon_{23}^{\prime 2}) , \quad (3)
$$

where C_{11} , C_{12} , and C_{44} are the elastic stiffness constants.

The expression of the magnetoelastic energy in presence of the external strain was obtained with the following assumptions: (i) The stress on the film in a direction normal to the plane of the film is zero $(\sigma_{33} = 0)$. (ii) The shear strains in the plane of the film, and normal to the plane of the film are equal to zero: $\epsilon_{ij} = 0$ with $i \neq j$. (iii) The isotropic planar strain can be represented by two equal, orthogonal, uniaxial strains in the film plane: ϵ_{11} $=\epsilon_{22}=\epsilon$. A relation between the two remaining components of the strain tensor can be found easily as follows: First the equilibrium value of ϵ_{33} is determined from the total free energy $E_T = E'_{\text{me}} + E'_e$ by minimizing E_T with respect to ϵ_{33} :

$$
\sigma_{33} = \delta E_T / \delta \epsilon_{33} = 0 \quad . \tag{4} \quad \alpha_1^2 + \alpha_2^2 + \alpha_3^2 = 1
$$

The relation between the strain tensor and the direction cosines of the magnetization in the primed and unprimed system are, respectively, given by¹⁴

$$
\epsilon'_{ij} = \sum_{k,l} a_{ik} a_{jl} \epsilon_{kl} \quad , \tag{5}
$$

$$
\alpha_i' = \sum_j a_{ij} \alpha_j \quad . \tag{6}
$$

With hypothesis (2) and (3) the expression (5) becomes largely simplified:

$$
\epsilon'_{ij} = \epsilon + (\epsilon_{33} - \epsilon)a_{13}^2 ,
$$

\n
$$
\epsilon'_{ij} = a_{13}a_{jj}(\epsilon_{33} - \epsilon) , \qquad i \neq j .
$$
 (7)

When (7) is substituted in (4) we obtain the following formula for ϵ_{33} as a function of ϵ :

$$
\epsilon_{33} = -2 \frac{C_{12} - C_{11} \Delta_C U}{C_{11} (1 + 2 \Delta_C U)} \epsilon - \frac{\delta E_{\text{me}}}{\delta \epsilon_{33}} \frac{1}{C_{11} (1 + 2 \Delta_C U)}, \qquad (8)
$$

with

when
\n
$$
\delta E_{\rm me}/\delta \epsilon_{33} = B_1 \sum_i \alpha_i^2 a_{13}^2 + 2 B_2 \sum_{i\nwhere
\n
$$
U = a_{13}^2 a_{23}^2 + a_{13}^2 a_{33}^2 + a_{23}^2 a_{33}^2
$$
$$

$$
= \sin^2 \alpha (\cos^2 \alpha + \frac{1}{4} \sin^2 \alpha \sin^2 2\beta)
$$
 (9)

and

$$
\Delta_C = (C_{12} + 2C_{44} - C_{11})/C_{11} \quad . \tag{10}
$$

For an elastically isotropic medium $\Delta C = 0$. The expression of the magnetoelastic energy in the primed system will be obtained if (7) and (8) are substituted in the total free energy E_T , which becomes the sum of two terms E_e and E_{me} , where

$$
E_e = \frac{(C_{11} + 2C_{12})(C_{11} - C_{12} + 3C_{11}U\Delta_C)}{C_{11}(1 + 2U\Delta_C)} \epsilon^2 + B_1\epsilon ,
$$

\n
$$
E_{\text{me}} = -\frac{(C_{11} + 2C_{12})\epsilon}{C_{11}(1 + 2U\Delta_C)} \left(B_1 \sum_i \alpha_i^2 a_{i3}^2 + 2B_2 \sum_i \alpha_i' \alpha_j' a_{i3} a_{j3}\right)
$$

\n
$$
-\frac{(B_1 \sum_i \alpha_i'^2 a_{i3}^2 + 2B_2 \sum_{i
$$

 E_e represents the elastic energy and it does not depend on the magnetization direction. In E_{me} , the first term depends on the strain and represents the magnetoelastic energy in a strained crystal. The last term in E_{me} does not depend on the strain and represents higher-order contributions to the magnetic anisotropy. This term is much smaller than the first one and generally also much smaller than the magnetic anisotropy itself. Consequently, it will be neglected in the following. To obtain the energy in the unprimed system, we substitute (6) into E_{me} , which leads to

$$
E_{\text{me}}/M = \frac{1}{2}H_1\alpha_3^2 + (H_1 - H_2)(a\alpha_1^2 + b\alpha_2^2 + c\alpha_3^2 + d\alpha_1\alpha_2 + e\alpha_1\alpha_3 + f\alpha_2\alpha_3),
$$
 (11)

$$
\alpha_1^2 + \alpha_2^2 + \alpha_3^2 = 1
$$

where

$$
a = \sin^{2} \alpha \cos^{2} \alpha (1 - \frac{1}{4} \sin^{2} 2\beta) ,
$$

\n
$$
b = \frac{1}{4} \sin^{2} \alpha \sin^{2} 2\beta ,
$$

\n
$$
c = -\sin^{2} \alpha (\cos^{2} \alpha + \frac{1}{4} \sin^{2} \alpha \sin^{2} 2\beta) ,
$$

\n
$$
d = -\frac{1}{4} \sin^{2} \alpha \cos \alpha \sin 4\beta ,
$$

\n
$$
e = -\sin \alpha \cos \alpha [\cos^{2} \alpha - \sin^{2} \alpha (1 - \frac{1}{2} \sin^{2} 2\beta)],
$$

\n(12)

$$
f = -\frac{1}{4}\sin^3 \alpha \sin 4\beta ,
$$

\n
$$
H_1 = \frac{H_1'}{1 + 2\Delta_C U} , \qquad H_1' = 3(C_{11} - C_{12}) \left(1 + \frac{2C_1}{C_1}\right)
$$

$$
H_2 = \frac{H'_2}{1 + 2\Delta_C U} , \quad H'_2 = 6C_{44} \left(1 + \frac{2C_{12}}{C_{11}} \right) \frac{\lambda_{111}}{M} \epsilon . \quad (13b)
$$

 $\frac{\lambda_{12}}{\lambda_{100}}$ ϵ

For some particular orientation of the crystallite axis Eqs. (11) reduce to a particular simple form: when the $[100]$ or $[111]$ crystallite axis is along the when the 100 or 111 crystallite axis is all film normal, E_{me} is given, respectively, by

$$
\frac{E_{\rm me}^{\rm 100}}{M} = \frac{H_1'}{2} \alpha_3^2 \quad , \qquad \frac{E_{\rm me}^{\rm 111}}{M} = \frac{H_2'}{2} \frac{\alpha_3^2}{1 + \frac{2}{3}\Delta c} + \text{const.} \quad (14)
$$

For these cases, E_{me} is an uniaxial anisotropy energy. The normal to the film plane is an easy axis when H_1 (or, in the case of the [111] direction, H_2) is negative and is a hard axis when H_1 (or H_2) is positive. In the literature the isotropic stress approximation $5,6$ is frequently used. In this approximation the components of the strain tensor are expressed as functions of the corresponding stress by minimizing the elastic energy and these values of ϵ_{i} , are substituted directly in the magnetoelastic energy. 15 When the condition of isotropic planar stress is written, $\sigma_{11} = \sigma_{22} = \sigma$ and all other stress components equal to zero, we obtain again Eqs. (11), with, however, H_1 and H_2 given by the following expression (15) and not by (13):

$$
H_1 = 3 \frac{\lambda_{100}}{M} \sigma , \quad H_2 = 3 \frac{\lambda_{111}}{M} \sigma . \tag{15}
$$

The stress σ is related to the strain by

$$
\sigma = \frac{Y}{1-\mu} \epsilon \quad , \tag{16}
$$

where Y is Young's modulus and μ is Poisson's ratio.

For elastically isotropic media

$$
Y = \frac{(C_{11} - C_{12})(C_{11} + 2C_{12})}{C_{11} + C_{12}}, \quad \mu = \frac{C_{12}}{C_{11} + C_{12}}, \quad (17)
$$

and it can be easily shown that for this case the values of H_1 and H_2 given by formulas (13) and (15) are identical. In the following sections the expression of the magnetoelastic energy obtained for isotropic strain will be used; the isotropic stress approximation can be considered to correspond to an elastically isotropic media.

III. CALCULATION OF MAGNETIC FIELD NECESSARY FOR RESONANCE AND STUDY OF RESONANCE LINE

We wish to calculate the resonance field of a particular grain for the case when the external homogeneous dc magnetic field is applied along the normal to the film. The resonance condition can be obtained from the total magnetic energy E . In presence of the applied dc field, and including the crystalline anisotropy energy E_k , the total energy E is given by

$$
E = E_Z + E_d + E_{\text{me}} + E_k \quad , \tag{18}
$$

where $E_z = -\mathbf{\bar{M}} \cdot \mathbf{\bar{H}}$ is the Zeemann energy, $E_d = \frac{1}{2}$ $(\underline{N}\overline{M})\cdot\overline{M}$ the demagnetizing energy, E_{me} the magnetoelastic energy as defined by Eqs. (11) where H is the applied external field, N the demagnetizing tensor, and \overline{M} the saturation magnetization. The anisotropy energy in the unprimed coordinate system can be obtained from the familiar formula E_k = $K_1(\alpha_1'^2 \alpha_2'^2 + \alpha_2'^2 \alpha_3'^2 + \alpha_2'^2 \alpha_3'^2)$ when the α'_i are expressed as functions of the α_i [Eq. (6)].

Using the expression of the energy as given by Eq. (18), the resonance condition can be obtained from the generalized resonance formula (A8) of

Appendix A, in principle, without any approximation. In (A8) the first and second derivatives of the magnetic energy must be taken at the equilibrium position of the magnetization. However, when the applied field is too low the magnetization vector deviates considerably from the field direction and the expression of the components of the magnetization at the equilibrium position becomes extremely complicated. The following hypothesis greatly simplifies the calculation: We use the large-field approximation, supposing that the applied dc field is sufficiently great that the component of the magnetization along it is an order of magnitude greater than those in the film plane. (This condition can be easily realized in experiments with a sufficiently high measuring frequency). When H_1 and H_2 in Eq. (11) are both negative (and much larger than the anisotropy field) the normal to the film is an easy axis and the magnetization vector tends to be aligned with it. Then the approximation should be valid even when the applied field is not too great. When the high-field approximation $M_z \simeq M \gg m_x$, m_y is introduced in Eq. (18), the Zeemann and demagnetizing energies become, respectively, $E_z = -HM_z$ \simeq - HM and $E_d = 2\pi M_Z^2 \simeq 2\pi M^2$. Now the equilibrium value of m_x^0 and m_y^0 of the static components of the magnetization can be easily computed from Eq. (A3). It can be shown that with the approximation presently used one can obtain an expression of the resonance condition valid up to the second order in H_1 , H_2 and in $H_a = 2K_1/M$. At this point of the calculation we suppose that H_1 , H_2 , and H_a are of the same order of magnitude. When $H_a \ll 4\pi M$, as is the case in most ferromagnetic and ferrimagnetic materials, H_a can be considered as small compared to δH , which we have assumed is of the order of $4\pi M$. Nevertheless in some specific cases-as will be shown in Sec. III-it is necessary to obtain a formula valid up to the second order in both H_1 , H_2 , and H_a . For this order, we need to determine the equilibrium value of $m_\texttt{x}^{\texttt{0}}$ and $m_\texttt{y}^{\texttt{0}}$ only up to the first order:

and
$$
H_a
$$
. For this order, we need to deter-
ne the equilibrium value of m_x^0 and m_y^0 only up to
first order:

$$
\frac{m_x^0}{M} \approx \frac{H_a - (H_1 - H_2)}{H} e
$$
,

$$
\frac{m_y^0}{M} \approx \frac{H_a - (H_1 - H_2)}{H} f
$$
.
(19)

The generalized resonance formula (A8) can be simplified when terms leading to third power in H_1 , H_2 , and H_a are neglected:

$$
(\omega/\gamma)^2 = (E_1^0)^2 + (E_2^0)^2 + (E_3^0)^2 + (x_3^0)^2 [E_{11}^0 E_{22}^0 - (E_{12}^0)^2]
$$

$$
- E_3^0 [x_3^0 (E_{11}^0 + E_{22}^0) - (x_1^0 E_{13}^0 + x_2^0 E_{23}^0)]
$$

$$
+ x_3^0 (E_1^0 E_{13}^0 + E_2^0 E_{23}^0), \qquad (20)
$$

where the subscript 3 corresponds to the z axis.

When the first and second derivatives of the energy in (20) are calculated at the equilibrium value given by (19), the resonance condition for a particular grain is given by

$$
H_0(\alpha, \beta) = 4\pi M + H_1 - H_a - [3(H_1 - H_2) - 5H_a]U
$$

+
$$
[(\omega/\gamma)^2 + a_1 U^2 - b_1 U - c_1 V]^{1/2},
$$
 (21)

$$
V = a_{13}^2 a_{23}^2 a_{33}^2 = \frac{1}{4} \sin^4 \alpha \cos^2 \alpha \sin^2 2\beta \tag{22}
$$

$$
a_1 = 21(H_1 - H_2)^2 - 102(H_1 - H_2)H_a + 85H_a^2,
$$

\n
$$
b_1 = 5(H_1 - H_2)^2 - 24(H_1 - H_2)H_a + 19H_a^2,
$$
\n(23)

$$
c_1 = 6[3(H_1 - H_2)^2 - 15(H_1 - H_2)H_a + 14H_a^2].
$$

If in Eq. (21) we take $H'_1 = H'_2$ and $H_a = \Delta_c = 0$, the resonance field reduces to $H = 4\pi M + H'_1 + \omega/\gamma$, which is the expression given by MacDonald. For $H_1 = H_2$ $= 0$ we find again the expression given in Ref. 11.

Now the distribution function of the resonance field strength is calculated. For an analytical study of the absorption line it is convenient to take the linewidth Of the individual grains equal to zero. The distribution function is given by

$$
A(H) = \frac{1}{4\pi} \int_0^{\pi} \sin \alpha \, d\alpha \int_0^{2\pi} \delta(H - H_0(a, \beta)) d\beta , \quad (24)
$$

where $\delta(H - H_0(\alpha, \beta))$ is the Dirac δ function.

It can be easily shown that this integral presents singularities for the stationary values of $H_0(\alpha, \beta)$, i. e., $\partial H_0 / \partial \alpha = \partial H_0 / \partial \beta = 0$. The three stationary values deduced from the first derivative of the resonance equation (21) are given by H_{100} , H_{110} , and H_{111} which are the resonance field for grain with $[100]$, $[110]$, and $[111]$ axis parallel to the applied field.

In the discussion which follows, the anisotropy field will be considered as a small perturbation $(H_{\alpha} \ll 4\pi M)$ and the independent-grain approximation—as defined by Schl $\ddot{\text{om}}$ ann¹⁰—will be satisfied if $\delta H > 4\pi M$, where

$$
\delta H = H_1' - \frac{H_2'}{1 + \frac{2}{3}\,\Delta_C} \quad . \tag{25}
$$

This new anisotropy field depends on the anisotropy Of the magnetostriction and on the elastic anisotropy. It is also proportional to the magnitude of the strain ϵ . It can be shown (Appendix B) that the absorption line presents a logarithmic singularity for H_{110} :

$$
H_{110} = 4\pi M + \frac{1}{4} (H_1 + 3H_2 + H_a)
$$

+ {(ω/γ)² + [$\frac{1}{4}$ (H₁ - H₂ - 3H_a)]²}¹², (26)

with

$$
H_1 = \frac{H_1'}{1 + \frac{1}{2}\Delta_C} \quad \text{and} \quad H_2 = \frac{H_2'}{1 + \frac{1}{2}\Delta_C}
$$

The distribution function starts and ends with a finite value of the resonance field corresponding to

$$
H_{100} = \omega / \gamma + 4\pi M + H_1' - H_a \t{,}
$$
 (27)

$$
H_{111} = \omega / \gamma + 4\pi M + H_2 + \frac{2}{3} H_a , \qquad (28)
$$

with

$$
H_2 = \frac{H_2'}{1+\frac{2}{3}\Delta_C} \ ,
$$

which are the two other singularities of the resonance line. The total width of the spectra is given by

$$
H_{100} - H_{111} = \delta H - \frac{5}{3} H_a \tag{29}
$$

In the following sections, we shall consider only the cases $H_i < 0$, $H_2 < 0$, and $|H_1| > |H_2|$, ⁹ which correspond to data obtained in $NiFe₂O₄$ and Ni thin films. Then H_{100} and H_{111} are, respectively, the minimum and the maximum of the resonance field distribution. It can be easily shown that 'when $H_1 - H_2$ is higher than a critical value (Appendix B) a secondary resonance peak is resolved near H_{100} . For an elastically isotropic material (with $H_a = 0$) this condition is $|H'_1| - |H'_2| > 0.3\omega/\gamma$. It can be shown that the peak near H_{111} is generally not resolved.

IV. NUMERICAL COMPUTATION OF RESONANCE LINE

Now we consider the physically more realistic case in which the individual crystallites have a Lorentzian absorption curve with the same linewidth ΔH . (This last hypothesis in only approximately satisfied: In single crystals ΔH varies slowly with the direction of the applied field relative to the axes of the crystal.) Then the absorption line becomes

$$
P(H) = \frac{\Delta H}{8\pi^2} \int_0^{\pi} \sin \alpha \, d\alpha
$$

$$
\times \int_0^{2\pi} \frac{d\beta}{\left(\frac{1}{2}\Delta H\right)^2 + \left[H - H_0\left(\alpha, \beta\right)\right]^2} , \quad (30)
$$

where ΔH is the full linewidth and we use the normalization condition

$$
\int_{-\infty}^{+\infty} P(H) dH = 1 \tag{31}
$$

It follows from the static equilibrium condition that the magnetization vector deviates slightly from the applied field. Thus the theoretical resonance line should be compared with the sum of the absorption measured in two transverse directions and in the longitudinal direction relative to the normal Of the film: The absorption is proportional to the imaginary part of the trace of the susceptibility tensor when phenomenological loss is included. The susceptibility tensor can be calculated from Eq. (AS).

FIG. 2. Theoretical absorption curves of a polycrystalline magnetostrictive film for different values of the parameter $\delta H' = H_1' - H_2'$. We take here $H_a = 0$, $\Delta_c = 0$, $\Delta H = 0.0035\omega/\gamma$ for the computation and H_i is the internal field in the sample: $H_{ext} - 4\pi M - H_2'$. As $\delta H'$ increases, the main peak intensity decreases, the linewidth increases, and a second peak tends to be resolved on the lowfield side of the curve.

As $M_{\star} \gg m_{\star}$, m_{\star} , the longitudinal absorption is negligible.

The ellipticity of the precession can be now estimated from the two-dimensional susceptibility tensor for the case when the driving field is linearly polarized and with sufficiently low linewidth of a particular grgin. A simple calculation leads to $P(H) \approx \frac{1}{2}(\chi_{xx} + \chi_{yy})$ and a multiplicative correction factor of the form

$$
[1+(\gamma/\omega)^2(a_1U^2-b_1U-c_1V)]^{1/2}
$$
 (32)

should be incorporated in Eq. (30). This expression shows that for crystallites with $[100]$ || H and $[111] \parallel H$, the precession is circular (in fact in this case $m_x^0 = m_y^0 = 0$) and the ellipticity is the largest

FIG. 3. Influence of the elastic anisotropy on the absorption curve of a polycrystalline magnetostrictive film. We keep constant the quantity $\delta H = H_1 - H_2/(1 + \frac{2}{3}\Delta_p)$ = 0.45 ω/γ rather than $\delta H'$, so that H_{100} and H_{111} remain located at the same field. The elastic anisotropy can both increase or decrease the tendency of the second peak to be resolved. Approximately, the case of Δ_c =0.6 corresponds to that of nickel.

FIG. 4. Influence of the magnetocrystalline anisotropy on the absorption curve of a polycrystalline magnetostrictive film. We take here for the different parameters the values of Ref. 9, which correspond approximately to the experimental nickel ferrite thin-film case: $\Delta = 0$, $\omega/\gamma = 5580$ Oe, $4\pi M = 3060$ Oe, $\Delta H = 20$ Oe, $\delta H' = -2500$ Oe. H is the external field.

for those for which $H \parallel [110]$. So the theoretical curve underestimates slightly the height of the spectra near H_{110} . However, this correction is only of the second order and in most cases is negligible.

The numerical computation of Eq. (30) was made by standard integration methods, with the use of an electronic computer (CII 10070). The computation was developed only for the cases $H_1 < 0$, $H_2 < 0$, and $|H_1| > |H_2|$. The analytical study in Sec. III and Appendix B shows that the width and the shape of the resonance line depend on the three following parameters: $\delta H' = H'_1 - H'_2$, Δ_c , and H_a . Figures 2-4 illustrate the evolution of the resonance line when these three parameters are varied indedependently. Figure 2 shows the resonance line for various values of $\delta H' = H_1' - H_2'$ for an elastically isotropic material ($\Delta_c = 0$) and for $H_a = 0$. One can note that with increasing $\delta H'$ the peak near H_{110} is conserved, the center of gravity of the line is displaced to lower fields, and a separate peak is resolved near H_{100} as predicted by the calculation described in Sec. III. These results are similar to that obtained by Schl j mann¹¹ with increasing crystalline anisotropy. Figure 3 shows the influence of the elasticity parameter: In this calculation δH has a fixed value and again we took $H_a = 0$. The curves with $\Delta_c \neq 0$ are to be compared with the line corresponding to elastic isotropy: for $\Delta_c > 0$, H_{110} is shifted to higher fields, the width of the spectra between the singularities corresponding to H_{100} and H_{110} increases, and the peak near H_{100} is no longer resolved. For $\Delta_c < 0$, the contrary is observed.¹ However in this case the peak near H_{100} is of the same amplitude as that near H_{110} . These results are explained when one considers the resonance equation (21). The resonance fields between H_{100}

FIG. 5. Influence of $\delta H'$ on the shape of the absorption curve in the vicinity of the magnetocrystalline anisotropy cancellation: $H_a = -600$ Oe and the cancellation occurs near $\frac{5}{3}H_a = -1000$ Oe = $\delta H'$. Here $\delta H'$ is expressed in k Oe and H is the external field with same values for other parameters than in Fig. 4.

and H_{110} are shifted to higher values for $\Delta_c > 0$ and to lower values for $\Delta_c < 0$, while the position of H_{100} is not affected as $U=0$ for this case. Finally in Fig. 4, the influence of the anisotropy field is presented in an elastically isotropic case $(\Delta_{\alpha} = 0)$, for a given value of $\delta H'$. For $K_1 < 0$, the width of the line decreases, H_{110} is shifted to lower fields and the peak at H_{100} is no longer resolved. These results are explained by the fact that for negative crystalline anisotroyy the normal to the film is, respectively, the easy and hard axis for crystallites with $[111]$ II \overline{H} and $[100]$ II \overline{H} , which is contrary to the case of the anisotropy induced by the strains. For K_1 > 0 the opposite results are obtained.

A singular situation occurs when the stress induced anisotropy field, and the crystalline anisotropy field are of the same order of magnitude and have the same sign. 17 First we consider the case of elastic isotropy $(\Delta_c=0)$: For $H'_1 - H'_2 = \frac{5}{3}H_a$, the first-order term in the resonance equation is canceled out and only the second order term contributes to the resonance linewidth. When ω/γ is sufficiently large, this contribution is negligible, and the width of the line is reduced to the same order of magnitude as that corresponding to a single crystal. Figures 5 and 6 present the resonance line and the corresponding linewidth, when H_a is fixed and $H'_1 - H'_2$ is varied. (The calculation was made with parameters close to that measured on bulk $NiFe₂O₄$.) We can remark that for the numerical values taken here, the independent grain model is not a good approximation because the dipolar interactions have to be taken into account. It can be shown that the dipolar interaction only affects the linewidth, which is narrowed by the dipolar interlinewidth, which is narrowed by the dipolar inter
action except for the case $H'_1 - H'_2 = \frac{5}{3} H_a$. ¹⁸ Finall it can be shown that the same effect occurs even for a material with large elastic anisotropy. The

value of δH giving the minimum linewidth is now obtained by numerical computation. The fact, that the effect of crystalline anisotropy oa the resonance linewidth can be suppressed in polycrystalline material with a given strain-induced anisotropy field, could be useful for practical devices.

V. CONCLUSION

We presented a new calculation of the resonance line in polycrystalline film under large isotropic planar strain, where the fact that generally λ_{100} $\neq \lambda_{111}$ was taken into account. The calculation was made with the independent-grain approach and we used the large-applied-field approximation. The validity of this approximation can be easily controlled in a given experimental case: H_1 and H_2 deduced from the value of the applied field corresponding to the singularities must be independent of the measuring frequencies.

The results are qualitatively the same as for ferrites with large anisotropy¹¹: The absorption line presents singularities, the logarithmic singularity occurs for the applied field equal to H_{110} , and the two other singularities are located at the extreme values of the distribution function. However, in the present case the shape of the spectra depends on a great number of parameters: λ_{100} , λ_{111} , C_{11} , C_{12} , C_{44} , and ϵ , while it is sufficient to know the single parameter Ha when only the crystalline anisotropy is considered.

The present theory was successfully used to explain the peculiarities of the resonance line observed in polycrystalline NiFe₂O₄ thin films.⁹ It can be also useful to explain some other published experiments where the results were not understood. Mather¹⁹ has recently reported experimental results on ferromagnetic resonance at oblique angles in stressed Ni films. He attributed the peaks of the absorption curve to standing spin-

FIG. 6. Influence of $\delta H'$ on the linewidth in the vicinity of the magnetocrystalline field cancellation. Same values as in Fig. 5 for the other parameters. When $\Delta_c \neq 0$, cancellation still occurs, but for different values of $\delta H'$, as is pointed out on the diagram.

wave modes. Location of the peaks at perpendicular resonance and their angular behavior are well explained on the basis of our theory,²⁰ while the results could not be interpreted using the wellknown angular dependence of spin-wave modes. In $Fe₃O₄$ thin films, Nosé et al.²¹ observed three resonance peaks, in perpendicular resonance. The positions of these peaks as functions of the applied field were generally independent of the film thickness. Therefore, the peaks probably correspond to singularities of the resonance line. We believe that the great number of experimental resultslisted in Ref. 5-obtained in various magnetostrictive films under large strain, have to be carefully reexamined on the basis of our calculation.

The present theory is only valid for a material with randomly distributed crystallites. When thermally induced strain exists during the film preparation, the crystallites are not oriented randomly because the grains having low strain energy are favored during growth. 22 This effect was observed in several thin films. 22 The present calculation then has to be modified in order to take into account the strain-induced texture.

APPENDIX A: GENERALIZED RESONANCE FORMULA IN A CARTESIAN COORDINATE SYSTEM

For the calculation we suppose the external field sufficiently high to saturate completely the magnetic media. The coordinate axes i , j , and k form a right-handed system. First we compute the static equilibrium position of the magnetization vector by requiring that the torque to which the magnetization is submitted is equal to zero:

$$
\vec{H}_{\text{eff}}^0 \times \vec{M}_0 = 0 , \qquad (A1)
$$

where the effective static internal field $\overline{H}_{\text{eff}}^{0}$ is derived from the total magnetic energy E . If E is independent of the partial derivatives of the magnetization with respect to the coordinate system, \vec{H}^0_{eff} is defined by

$$
\vec{H}_{\text{eff}}^0 = -\delta E / \delta \vec{M}_0 \tag{A2}
$$

and the components of the magnetization vector can be deduced from

$$
x_i^0 E_j^0 - x_j^0 E_i^0 = 0 , \quad i \neq j
$$

\n
$$
x_i^0 = m_i^0 / M_0 , \quad E_i^0 = \delta E / \delta x_i^0 , \quad \sum_i (x_i^0)^2 = 1 .
$$
 (A3)

The resonance condition can be obtained from the equation of motion of the magnetization vector:

$$
\frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{H}_{\text{eff}} \,, \tag{A4}
$$

where the components of magnetization vector are given by $m_i = (x_i^0 + x_i)M_0$, x_i being the rf component of the magnetization. Then the total energy can be expressed in a Taylor expansion up to the second order in the rf magnetization components as follows:

$$
E = E_0 + \sum E_i^0 x_i + \frac{1}{2} \sum E_{ii}^0 x_i^2 + \sum_{\substack{i \neq j \\ i \neq j}} E_{ij}^0 x_i x_j , \qquad (A5)
$$

where $E_{ij}^0 = \delta^2 E / \delta x_i^0 \delta x_j^0$ is the second derivative of the free energy taken at the equilibrium position. The effective field can be obtained from the total energy, assuming small motion of the magnetization vector around the equilibrium position. Then the components of the effective field are given by

$$
(H_{\text{eff}})_i = -\delta E / \delta x_i = -(E_i^0 + E_{ii}^0 x_i + E_{ij}^0 x_j + E_{ik}^0 x_k) \tag{A6}
$$

Finally, the three linearized equations in x_i of the following form are obtained when (A6) is substituted in $(A4)$:

$$
\frac{1}{\gamma} \frac{dx_i}{dt} = x_i (x_k^0 E_{ij}^0 - x_j^0 E_{ik}^0) + x_j (x_k^0 E_{jj}^0 - x_j^0 E_{jk}^0 - E_k^0)
$$

$$
+ x_k (x_k^0 E_{jk}^0 - x_j^0 E_{kk}^0 + E_j^0) . \quad (A7)
$$

The determinant of the coefficient of x_i yields a secular equation giving the resonance frequency:

$$
\left(\frac{\omega}{\gamma}\right)^2 = \sum_i (E_i^0)^2 - \sum_{\substack{i \neq j \neq k \\ j < k}} E_i^0 \left[x_i^0 (E_{jj}^0 + E_{kk}^0) - x_j^0 E_{ij}^0 - x_k^0 E_{ik}^0 \right] + 2 \sum_{\substack{i \neq j \neq k \\ i \neq j \neq k}} x_i^0 x_j^0 (E_{ik}^0 E_{jk}^0 - E_{ij}^0 E_{kk}^0) + \sum_{\substack{i \neq j \neq k \\ j < k}} (x_i^0)^2 \left[E_{jj}^0 E_{kk}^0 - E_{ijk}^0 \right]. \tag{A8}
$$

APPENDIX B: ANALYTICAL STUDY OF RESONANCE LNE NEAR ITS SINGULARITIES

The behavior of the resonance line near a singularity can be studied as follows:

Assuming that the stationary point corresponds to the $[uvw]$ direction we chose a coordinate system whose polar axis is $[uvw]$. The direction of the normal to the film is defined by the polar and azimuthal angles α_1 and β_1 in this special system. Then the resonance field is developed as a function of α_1 and β_1 , and the distribution function given by Eq. (19) is integrated for $H - H_{uvw} \simeq 0$, where H is the applied field and H_{uvw} the resonance field corresponding to the stationary direction $(\alpha_1 = 0)$.

A. Behavior near Logarithmic Singularity 100

 $Schlömann showed¹¹$ that when the resonance field can be put into the form

$$
H_0(\alpha_1 \beta) = H_{uvw} \left[1 + (a \sin^2 \beta - b \cos^2 \beta) \sin^2 \alpha \right] \quad (B1)
$$

and for $ab > 0$, the corresponding distribution function has a logarithmic singularity and the singular part of $A(H)$ for $|H-H_{uvw}| \approx 0$ is

$$
A(H) \approx \frac{1}{4\pi H_{uvw} (ab)^{1/2}} \ln \frac{|H - H_{uvw}|}{H_{uvw}} \ . \tag{B2}
$$

We show that in the present case the resonance field H_{110} corresponds to this logarithmic singularity. The calculation is made for $\Delta_c = 0$ and $H_a = 0$.

First, the magnetoelastic energy is expressed in the $[001]$, $[1\overline{1}0]$, $[110]$ coordinate system:

$$
E = \frac{1}{2} B_1 [(\epsilon'_{11} + \epsilon'_{22}) (\alpha'^2_1 + \alpha'^2_2) + 2\epsilon'_{22} \alpha'^2_2 + 4\epsilon'_{13} \alpha'_1 \alpha'_3]
$$

+
$$
\frac{1}{4} B_2 [(\epsilon'_{11} - \epsilon'_{33}) (\alpha'^2_1 - \alpha'^2_3)
$$

+
$$
4\epsilon'_{12} \alpha'_1 \alpha'_2 + 4\epsilon'_{23} \alpha'_2 \alpha'_3], \quad (B3)
$$

and then the magnetoelastic energy is calculated in the unprimed system, where α_1 and β_1 are the polar and azimuthal angles with respect to the [110] and [001] direction. Finally, when the resonance equation is developed as a function of α_1 and β_1 near the stationary point, we find

$$
H_0(\alpha_1 \beta_1) = H_{110}[1 + (a \sin^2 \beta_1 - b \cos^2 \beta_1) \sin^2 \alpha_1], \quad (B4)
$$

with

$$
a = \frac{(H_1 - H_2)(12\{(\omega/\gamma)^2 + \left[\frac{1}{4}(H_1' - H_2')\right]^2\}^{1/2} - (H_1' - H_2'))}{4H_{110}((\omega/\gamma)^2 + \left[\frac{1}{4}(H_1' - H_2')\right]^2\}^{1/2}} ,
$$

$$
b = \frac{(H_1' - H_2')(12\{(\omega/\gamma)^2 + \left[\frac{1}{4}(H_1' - H_2')\right]^2\}^{1/2} - (5H_1' - 3H_2'))}{8H_{110}((\omega/\gamma)^2 + \left[\frac{1}{4}(H_1' - H_2')\right]^2\}^{1/2}} .
$$
 (B5)

It can be easily shown that when Δ_c and H_a ($|H_a|$

FIG. 7. Variation of the critical value $\delta H' / (\omega / \gamma)$ [Eq. (16)] as a function of the elasticity parameter Δ_{c} . In the calculation $H_a=0$, $H_2=0$, and $\delta H=H_1$.

FIG. 8. Shape of the spectra near the H_{100} singularity as a function of the crystallite linewidth ΔH . The numerical values are the same as for Fig. 4: $H_a = \Delta_c = 0$.

 $\ll |\delta H|$) are taken into account the singularity at H_{110} is conserved.

B. Behavior of Resonance Line near Extrema of $H_0(\alpha, \beta)$

The two other singularities are located at the minima and maxima of the distribution function. We develop the calculation for the case corresponding to the numerical computation $H_1 < 0$, $H_2 < 0$, and $|H_1| > |H_2|$. Then the resonance field H_{100} corresponds to a minimum of $H_0(\alpha, \beta)$. We determine now the condition that a secondary peak should be resolved in its vicinity.

When Eq. (19) is developed as a function of α near H_{100} , it takes the form $H_0 (\alpha, \beta) = H_{100} (1 + az)$ + bz²). If a new variable $y = H_{100}$ (az + bz²) is introduced (where $z = \sin^2 \alpha$), by inversion of this equation we obtain

$$
z = \frac{1}{A} \left(\frac{y}{H_{100}} \right) - \frac{\overline{B}}{A^3} \left(\frac{y}{H_{100}} \right)^2 ,
$$

where

$$
\overline{B} = \frac{1}{2\pi} \int_0^{2\pi} B d\beta . \tag{B6}
$$

Finally it can be shown that the distribution function for $H - H_{100} \approx 0$ takes the form

$$
A(H) = \frac{1}{|A| H_{100}} \left(1 + \frac{A - 4\overline{B}}{A^2} \frac{H - H_{100}}{H_{100}} \right) \tag{B7}
$$

and the condition that a secondary peak should be resolved at H_{100} for $H - H_{100} > 0$ is

$$
\frac{\partial A(H)}{\partial H} = \frac{A - 4\overline{B}}{A^2} < 0
$$

This condition as a function of H_1, H_2, Δ_c , and H_a is finally given by

$$
\frac{\omega}{\gamma} < \frac{195[|H'_1| - |H'_2|]^2 + 942[|H'_1| - |H'_2|]H_a + 767H_a^2 + 160\Delta_c[|H'_1| - |H'_2|]^2}{54[|H'_1| - |H'_2|] + 90H_a + 4\Delta_c[|H'_1| [9 + 4\Delta_c] + 24[|H'_1| - |H'_2|]]}
$$
\n(B8)

Without magnetocrystalline anisotropy and for an elastically isotropic material $(H_a = \Delta_c = 0)$ the singularity near H_{100} is resolved if $H_1'-H_2'$ is larger than the following critical value: $|H'_1| - |H'_2| > 0$. $3 \omega / \gamma$.

When the material is not elastically isotropic, Fig. 7 shows the variation of the critical value of $|H_1'| - |H_2'|$ as a function of the elasticity parameter Δ_c . In the same way one can predict that this critical value increases for $H_a < 0$ and decreases

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for $H_a > 0$. The preceding condition-necessary to obtain a well-resolved peak at H_{100} -is no longer valid when we consider crystallites with nonzero linewidth. Figure 8 shows (for $\Delta_c = 0$, $H_a = 0$) how the peak gradually disappears as the linewidth is increased. A well-resolved peak occurs for ΔH = 20 Oe, but the peak is no longer resolved when $\Delta H = 100$ Oe, although the condition (B8) is well satisfied $(\gamma \delta H / \omega \simeq 0.45)$.

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