PHYSICAL REVIEW B VOLUME 26, NUMBER 1 1 JULY 1982

Theory of ferromagnetic resonance and static magnetization in ultrathin crystals

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(Received 14 December 1981)

A theory of the static magnetization orientations θ_0 and of the ferromagnetic resonance fields $\vec{H}_{\text{res}}$ in ultrathin ($\lesssim 50$ Å) monocrystalline films is presented. Both θ_0 and $\vec{H}_{\text{res}}$ are shown to depend crucially on the magnetocrystalline surface anisotropy, and a generalization of this anisotropy is proposed. The equation of motion of the magnetization is combined with the general exchange boundary condition, and realistic approximations are introduced in order to obtain closed algebraic expressions for θ_0 and $\vec{H}_{\text{res}}$. The spatial dependence of the oscillations in the presently calculated resonance modes do not represent spin waves but nondissipative exponential decays perpendicular to the film surfaces. The film thicknesses $2L$ are so small that the films are "beyond spin-wave cutoff." It is shown that the angular dependence of H_{res} differs strongly from that predicted by the uniform mode theory and that H_{res} is a linear function of $1/L$ rather than independent of L. An extension of the theory to somewhat thicker films is also presented.

I. INTRODUCTION

In this paper we predict the existence and properties of a certain magnetic resonance mode which can be induced in an ultrathin ferromagnetic crystal by its surface anisotropy. This mode will henceforth be referred to as "the" surface-induced mode even though it is related to surface wave modes discussed previously. We find that while the surface-induced mode does involve exchange interactions, it differs significantly from the spin-wave resonance modes usually observed in thin films as well as from all ferromagnetic resonance modes. An important property of the surface-induced mode is that in this mode the spatial dependence of the oscillations represents neither standing nor propagating waves but solely a nondissipative exponential decay. Specifically, we find the *entire* spatial dependence of the oscillations to be proportional to $\exp(k\eta)$, where k is a real number and η is a distance measured perpendicular to the crystal surfaces. The basic reason for this type of spatial dependence is the nature of the surface anisotropy. As shown by Eqs. (4.30) and (4.31) derived in Sec. IV, we find that for appropriate signs of the surface-anisotropy constants and appropriate orientations of the static magnetization, the ferromagnetic resonance field is larger than that obtained if the surface-anisotropy constants are zero. This means, according to Eqs. (4.31) and (4.9), that k^2 is positive so that k is indeed real rather than imaginary. It should also be noted that if the crystal thickness is small compared to $1/k$, we may regard the crystal as being "beyond cutoff" for spin-wave resonance. This is somewhat analogous to calling an electromagnetic waveguide beyond cutoff if its transverse dimensions are small compared to the wavelength of interest.

The particular problem posed and solved in this paper concerns the effects of surface anisotropy on the static magnetization orientations and on the ferromagnetic resonance fields in an ultrathin crystal. By ultrathin we mean a thickness which is typically less than about 50 Å. In Sec. II we specify an appropriate form of the equation of motion of the magnetization and of the equation expressing the exchange boundary condition. Also included in Sec. II is a generalization of the phenomenological representation of surface anisotropy.

Section III contains a static solution of the two above-mentioned equations. This solution describes the static orientations of the magnetization and predicts that these orientations are similar to those existing in a ferromagnetic domain wall. In Sec. IV A we present a dynamic solution of the two equations which describes small-amplitude oscillations of the magnetization. This solution predicts the ferromagnetic resonance field arising from the surface-induced mode and shows that along certain principal crystallographic directions the resonance field is not constant but a linear function of the reciprocal of the crystal thickness. Another qualitatively new prediction contained in this solution is that the angular dependence of the resonance field associated with the surface-induced mode differs strikingly from that associated with the uniform mode. Specifically, the crystallographic directions which appear to be magnetically easy and hard when investigated by means of the uniform mode may become magnetically hard and easy, respectively, when investigated by means of the surface-induced mode. It should also be noted that by introducing realistic approximations we are able to express both the dynamic and the static solution in closed algebraic form.

Section IV B is devoted to generalizations and discussions of our theoretical results on the resonance field. In particular, we extend our calculations to situations in which the volume anisotropy consists of a growth-induced, as well as an intrinsic contribution, to crystals which are not ultrathin but still sufficiently thin to prevent spin-wave resonance, and to cases in which the orientation of the static magnetization is arbitrary rather than parallel to a principal crystallographic direction.

The present work was motivated by ongoing ferromagnetic resonance experiments carried out at the Naval Research Laboratory[1] on ultrathin iron crystals grown by molecular-beam epitaxy on gallium arsenide substrates. A comparison between experiment and theory is included in Ref. 1 and augmented in Sec. IV B of the present paper.

II. BASIC EQUATIONS AND GENERALIZED SURFACE ANISOTROPY

Figure 1 shows the coordinate systems used in our calculations of the static magnetization orientations and of the ferromagnetic resonance fields. The axes of the x,y,z system are parallel, respectively, to the [100], [010], and [001] axes of the cubic crystal, and the ξ and η axes of the ξ, η, z system are parallel, respectively, to the cubic [110] and [$1\bar{1}0$] axes. Also shown are the planes $\eta=\pm L$ which are assumed to bound the crystal along the η axis. Along the ξ and z axes the crystal is assumed to be unbounded. It is further assumed that the applied static magnetic field $\vec{H}$ is always parallel to the ($1\bar{1}0$) crystal faces and oriented at an angle ψ with respect to the [001] axis. These crystal faces are assumed to be ideally flat and devoid of "islands." No time-dependent magnetic field is used because we are concerned with free oscillations only. The instantaneous orientation of the (position-dependent) magnetization $\vec{M}$ is described by the polar coordinates θ and ϕ.

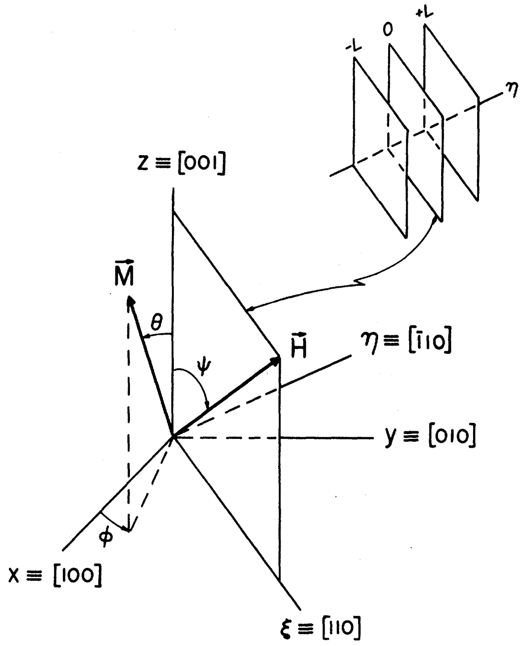

FIG. 1. Orientation of the applied static magnetic field $\vec{H}$, the instantaneous magnetization $\vec{M}$, and the monocrystal of thickness $2L$ with respect to the coordinate systems used in the calculations.

Throughout this paper the crystal is assumed to be bcc iron. For a different crystal or for a configuration differing from that of Fig. 1 the calculations would differ only in detail. Our assumed surface-induced mode may still exist, of course, because such a mode merely requires a nonzero surface anisotropy whose coefficients have appropriate signs and a crystal thickness $2L$ which is sufficiently small.

For the equation of motion of $\vec{M}$ we use the Landau-Lifshitz[2] equation and write it in the form

$$\left[\frac{1}{\gamma}\right]\frac{\partial\vec{u}}{\partial t}=\vec{u}\times\left[\vec{H}+\vec{H}_{\mathrm{dem}}-\left[\frac{1}{M}\right]\vec{\nabla}_{u}E_{\mathrm{ani}}+\left[\frac{2A}{M}\right]\nabla^{2}\vec{u}\right], \tag{2.1}$$

where

$$\vec{u}=\frac{\vec{M}}{M}=\vec{i}_{x}u_{x}+\vec{i}_{y}u_{y}+\vec{i}_{z}u_{z} \tag{2.2}$$

is a unit vector along $\vec{M}$ and $\vec{i}_x, \vec{i}_y, \vec{i}_z$ are unit vectors along the x, y, z directions, respectively. Here $\gamma = \gamma_e g/2$ is the magnetomechanical ratio, $\gamma_e = 2\pi$ (2.80) MHz/Oe is the value of γ for a free electron, and g is the spectroscopic splitting factor. The first term in the brackets of Eq. (2.1) is the field $\vec{H}$ defined above, the second term is the demagnetizing field $\vec{H}_{\rm dem}$, the third term is an effective field arising from the volume anisotropy energy density $E_{\rm ani}$, and the fourth term is an effective field due to the exchange interactions. Omitted from Eq. (2.1) is a relaxation term because the effect of such a term on the resonance field is generally negligible. The parameter A is the exchange stiffness constant and $\vec{\nabla}_u$ is an abbreviation for the operator

$$\vec{i}_x \frac{\partial}{\partial u_x} + \vec{i}_y \frac{\partial}{\partial u_y} + \vec{i}_z \frac{\partial}{\partial u_z} \ .$$

Since the dimensions of the crystal in the ξz plane are assumed to be infinite, we may write

$$\vec{H}_{\rm dem} = -4\pi(\vec{M}\cdot\vec{i}_\eta)\vec{i}_\eta \ , \qquad (2.3)$$

where $\vec{i}_\eta$ is a unit vector along the η direction. We further assume that the orientation of $\vec{M}$ varies solely with η so that ∇^2 can be replaced by $\partial^2/\partial\eta^2$. For $E_{\rm ani}$ we use only the term

$$E_{\rm ani} = K_1(u_x^2u_y^2 + u_y^2u_z^2 + u_z^2u_x^2) \ , \qquad (2.4)$$

where K_1 is the first-order cubic anisotropy constant. This restriction is made partly for the sake of simplicity and partly because in bulk iron, at least, all uniaxial volume-anisotropy terms are absent and the magnitude of the second-order cubic term $K_2u_x^2u_y^2u_z^2$ is presumably small (albeit nonnegligible) compared to that of the K_1 term. In epitaxially grown monocrystalline films, however, part of the volume anisotropy may consist of a magnetoelastic term originating in a substrate-film lattice mismatch or be caused by some other growth-induced mechanism. These possibilities and a method for dealing with them are discussed in Sec. IV B. Also contained in Sec. IV B is a justification of the fact that throughout this paper we omit not only the exchange effects induced by the skin depth but even spin-wave excitation by surface conditions.

For the exchange boundary condition we use the equation

$$\left[\frac{2A}{M^2}\right]\vec{M}\times\left[\frac{\partial\vec{M}}{\partial n}\right] + \vec{T}_{\rm surf} = 0 \qquad (2.5)$$

which was proposed along with some special forms by Rado and Weertman.[3] Here $\partial/\partial n$ denotes the partial derivative in the direction of a unit vector, $\vec{n}$, which is normal to, and points outward from, the thin "pill box" used in the derivation[3] of Eq. (2.5) by means of Green's theorem. It is important to realize that, as a consequence, the direction of $\vec{n}$ at each surface of a ferromagnetic film is always toward the film's interior. Thus we have

$$\frac{\partial}{\partial n} = \begin{cases} -\dfrac{\partial}{\partial\eta}, & \eta = +L \qquad (2.6a) \\[2ex] +\dfrac{\partial}{\partial\eta}, & \eta = -L \ . \qquad (2.6b) \end{cases}$$

Turning now to $\vec{T}_{\rm surf}$, we note that it was defined[3] as the sum of all the surface-torque densities arising from forces other than ferromagnetic exchange. In the present paper, however, we consider solely that contribution to $\vec{T}_{\rm surf}$ which is due to surface anisotropy. Accordingly, Eq. (2.5) may be written as

$$\vec{u}\times\left[\vec{\nabla}_u E_{\rm surf} - 2A\frac{\partial\vec{u}}{\partial n}\right] = 0 \ , \qquad (2.7)$$

where $E_{\rm surf}$ denotes the surface-anisotropy energy density. To obtain the lowest-order expression for $E_{\rm surf}$ in terms of powers of the components of $\vec{u}$, we introduce a method which is based solely on symmetry and is, therefore, more general than the method of Néel.[4] We note that at the surface of a crystal the usual point symmetry is broken so that some of the terms bilinear in the components of $\vec{u}$ may not be forbidden by symmetry at the surface even if all such terms are forbidden by symmetry in the crystal's interior. Specifically, we find that at a {110} surface of bcc iron the $m3m$ point symmetry of the interior is replaced by a point symmetry characterized solely by three elements, namely, the twofold axis 2_η and the mirror planes $\bar{2}_\xi$ and $\bar{2}_z$. Thus the lowest-order expression for $E_{\rm surf}$ must have the form

$$E_{\rm surf} = K_s u_x u_y + K_{ss} u_z^2 \ , \qquad (2.8)$$

where the phenomenological coefficients K_s and K_{ss} are surface-anisotropy constants. Prior to comparing Eq. (2.8) with the result of Néel,[4] who originated the concept of surface anisotropy, we recall that Néel assumed phenomenological interactions between nearest-neighbor atoms and made explicit use of the detailed crystallographic structure. For a {110} surface of bcc iron, Néel found that $E_{\rm surf}$ is proportional to $u_x u_y$, just like the first term of our Eq. (2.8). But if we use Néel's method and extend it to include next-nearest-neighbor interactions, which are particularly important in bcc iron, then we obtain Eq. (2.8) in its entirety. The fact that this equation can be obtained in two different ways is important because we find (see Sec. IV B) that if K_{ss} is assumed to be zero then our final result for the ferromagnetic resonance field H_{res} may not be sufficiently general. It should also be noted that for the sake of simplicity we assume the values of the coefficients K_s and K_{ss} to be the same at $\eta=+L$ as at $\eta=-L$ even though at some future occasion this simplification may have to be dropped.

Having obtained the equation of motion (2.1) and the boundary condition (2.7), we replace each of these vector equations by three scalar equations and then express u_x, u_y, u_z in terms of θ and ϕ by means of the relations

$$u_x=\sin\theta\cos\phi,\quad u_y=\sin\theta\sin\phi\ ,\quad u_z=\cos\theta\ . \tag{2.9}$$

The two equations arising from Eq. (2.1) will henceforth be called the "θ,ϕ form" of the equation of motion and the two equations arising from Eq. (2.7) will be called the θ,ϕ form of the boundary condition. Prior to presenting and solving these four equations in Secs. III and IV we introduce the decompositions

$$\theta=\theta_0+\theta_1\ , \tag{2.10a}$$

$$\phi=\phi_0+\phi_1\ , \tag{2.10b}$$

where the subscripts zero and one denote the static and time-dependent components, respectively. Also to be noted is that the operator $\partial/\partial\eta$ will henceforth be abbreviated by a prime.

III. STATIC MAGNETIZATION ORIENTATIONS

By retaining only those terms in the θ,ϕ form of the equation of motion which do not contain either θ_1 or ϕ_1, we obtain the static magnetization equations

$$\phi_0=\pi/4\ , \tag{3.1}$$

$$2A\theta_0''+MH\sin(\psi-\theta_0)$$
$$-K_1\sin\theta_0\cos\theta_0(3\cos^2\theta_0-1)=0\ , \tag{3.2}$$

which determine the static orientations of the magnetization. Equation (3.1) is a consequence of symmetry and the fact (see Sec. II) that $\vec{H}$ is parallel to the $(1\bar{1}0)$ plane. Equation (3.2) has a solution in which θ_0 is independent of η but this solution must be rejected because for most values of θ_0 it violates the boundary condition [see Eq. (3.11) below] unless the surface anisotropy happens to be zero. The possibility that the surface anisotropy may give rise to spatially nonuniform magnetization orientations was first suggested by Rado and Weertman.[3] Although they noted that the resulting structure may resemble a domain wall, their suggestion does not seem to have been followed up by a detailed calculation until the present paper.

Rather than attempting to solve the nonlinear Eq. (3.2), we linearize it by putting

$$\theta_0=\theta_{00}+\delta\theta_0 \tag{3.3}$$

and introducing the approximation

$$2\,|\,\delta\theta_0\,|\ll 1\ , \tag{3.4}$$

where θ_{00} is independent of η and $\delta\theta_0$ is dependent on η. As shown below, moreover, θ_{00} is the value of θ_0 in the absence of surface anisotropy whereas $\delta\theta_0$ is that contribution to θ_0 which arises from surface anisotropy. With the use of Eq. (3.3) and the approximation (3.4), we now replace Eq. (3.2) by the equations

$$MH\sin(\psi-\theta_{00})-K_1\sin\theta_{00}\cos\theta_{00}(3\cos^2\theta_{00}-1)=0\ , \tag{3.5}$$

$$2A(\delta\theta_0)''-[MH\cos(\psi-\theta_{00})+\alpha_{00}K_1]\delta\theta_0=0\ , \tag{3.6}$$

where the abbreviation α_{00} is defined by

$$\alpha_{00}=2-13\sin^2\theta_{00}+12\sin^4\theta_{00}\ . \tag{3.7}$$

Equation (3.5) expresses the static equilibrium condition in the absence of surface anisotropy and agrees exactly with Eq. (5) of Artman.[5] Equation (3.6), on the other hand, is new and remains valid in the presence of surface anisotropy. If the value of H is sufficiently large to justify the approximation

$$|\,\psi-\theta_{00}\,|\ll 1\ , \tag{3.8}$$

and to ensure that the bracketed expression in Eq. (3.6) is always positive, then Eq. (3.6) has the solution

$$\delta\theta_0=c_1\exp(\eta/l)+c_2\exp(-\eta/l)\ , \tag{3.9}$$

where l denotes the characteristic length

$$l=[2A/(MH+\alpha_{00}K_1)]^{1/2} . \quad (3.10)$$

We note that l resembles the wall-thickness parameter of ordinary domain theory and that the coefficients c_1 and c_2, which are unknown, must be determined by means of the static boundary condition. To derive the latter we retain only those terms in the θ,ϕ form of the boundary condition (2.7) which do not contain either θ_1 or ϕ_1. Thus we obtain

$$4A\frac{\partial\theta_0}{\partial n}+(K_s-2K_{ss})\sin2\theta_0=0 \quad (3.11)$$

as the boundary condition for θ_0. Equation (3.11) must be satisfied at the crystal boundaries ($\eta=\pm L$) by that value of θ_0 which results from combining Eq. (3.3) with Eq. (3.9). This requirement leads to an expression for c_1 and to an identical expression for c_2, as expected from symmetry. Substitution of this expression into Eq. (3.9) then yields

$$\delta\theta_0=-\frac{\frac{1}{2}r\sin2\theta_{00}[\cosh(L/l)]^{-1}\cosh(\eta/l)}{\tanh(L/l)+r\cos2\theta_{00}} , \quad (3.12)$$

where r is an abbreviation for the ratio

$$r=\tfrac{1}{2}(K_s-2K_{ss})l/A . \quad (3.13)$$

Although Eq. (3.12) was derived on the basis of the approximation (3.4), the former does not necessarily satisfy the latter if the parameters r and L/l are arbitrary and the value of the angle θ_{00} differs from zero or $\pi/2$. To assure that (3.12) is, in fact, consistent with (3.4) for all values of θ_{00}, we now require that the parameters satisfy the approximation

$$|r/\tanh(L/l)|\ll1 . \quad (3.14)$$

Combining (3.14) with Eqs. (3.12) and (3.13) gives

$$\delta\theta_0=-(K_s-2K_{ss})(l/4A)\sin2\theta_{00} \times[\sinh(L/l)]^{-1}\cosh(\eta/l) , \quad (3.15)$$

which is our final result for the general (i.e., η-dependent) form of $\delta\theta_0$. Among the predictions of Eq. (3.15) we note the following: (a) As L approaches infinity, $\delta\theta_0$ approaches zero, as expected for physical reasons; (b) as L approaches zero, the approximation (3.14) becomes invalid and hence Eq. (3.15) becomes inapplicable; (c) as H approaches infinity, both l and $\delta\theta_0$ approach zero, again as expected for physical reasons; (d) $\delta\theta_0$ is zero when θ_{00} equals zero or $\pi/2$, but not when θ_{00} equals $\cos^{-1}(1/\sqrt{3})$, as expected from the uniaxial character of the surface anisotropy.

To summarize, our general result for the static magnetization orientation is that ϕ_0 is given by Eq. (3.1) and that the dependence of θ_0 on ψ and η is contained partly in Eq. (3.5), which is supplemented by the approximation (3.8), and partly in Eqs. (3.3) and (3.15). For rough numerical estimates, however, it is useful to add the following special result. By introducing the further approximation

$$L/l\ll1 , \quad (3.16)$$

we can replace Eq. (3.15) by the expression

$$\delta\theta_0=-(K_s-2K_{ss})(l^2/4AL)\sin2\theta_{00} , \quad (3.17)$$

which is independent of η. We postpone the numerical estimates until the end of Sec. IV B because it is in connection with the dependence of H_{res} on θ_0 that the numerical value of $|\delta\theta_0|$ is of primary interest.

IV. FERROMAGNETIC RESONANCE FIELDS

A. Explicit calculation

By retaining only those terms in the θ,ϕ form of the equation of motion which are of first order in θ_1 and ϕ_1, we obtain three dynamical magnetization equations for θ_1 and ϕ_1. These equations will not be presented because we modify them in several respects. After expressing ϕ_0 by the static equilibrium condition (3.1), we assume temporarily (i.e., until the last paragraph of Sec. IV) that θ_0 is zero or $\pi/2$ so that θ_0 becomes identical with θ_{00} and each of the quantities $\delta\theta_0$, $(\delta\theta_0)'$, and $(\delta\theta_0)''$ vanishes. In other words, we temporarily restrict our calculation to situations in which the equilibrium direction of the magnetization is along [001] or [110] and thus spatially uniform. Later on, however, we show that our final result for the resonance field H_{res} [see Eq. (4.30) below] does remain valid, albeit less accurately, even if the value of θ_0 is arbitrary. Accordingly, we retain the symbol θ_0 and do not replace it by zero or $\pi/2$. After putting $\theta_0''=0$ into the static equilibrium condition (3.2) and introducing the approximation

$$|\psi-\theta_0|\ll1 , \quad (4.1)$$

we then express ψ in terms of θ_0. Next we assume that θ_1 and ϕ_1 are each proportional to $\exp(i\omega t+k\eta)$, where ω is the circular frequency and k denotes some (as yet unknown) *real* number. In this way the three above-mentioned dynamical equations lead to the two equations

$$\theta_1[(i\omega/\gamma)\cos\theta_0 - H - H_\alpha + 2Ak^2/M] - \phi_1[(i\omega/\gamma)\sin\theta_0 + \tfrac{1}{2}(B + H_\beta - 2Ak^2/M)\sin 2\theta_0] = 0 \,, \tag{4.2}$$

$$\theta_1[(i\omega/\gamma)\cos\theta_0 + H + H_\alpha - 2Ak^2/M] + \phi_1[(i\omega/\gamma)\sin\theta_0 - \tfrac{1}{2}(B + H_\beta - 2Ak^2/M)\sin 2\theta_0] = 0 \,, \tag{4.3}$$

where B denotes $H + 4\pi M$ and the quantities

$$H_\alpha = \alpha K_1/M$$

and

$$H_\beta = \beta K_1/M \tag{4.4}$$

are anisotropy fields defined with the use of the abbreviations

$$\alpha = 2 - 13\sin^2\theta_0 + 12\sin^4\theta_0 \,, \tag{4.5}$$

$$\beta = 2 - 7\sin^2\theta_0 + 3\sin^4\theta_0 \,. \tag{4.6}$$

In order that Eqs. (4.2) and (4.3) possess a nonvanishing solution, the secular determinant of their coefficients must be zero. This requirement yields the dispersion relation

$$(\omega/\gamma)^2 = (H + H_\alpha - 2Ak^2/M) \times (B + H_\beta - 2Ak^2/M) \,, \tag{4.7}$$

which we now check in two special cases. By putting $k=0$ into Eq. (4.7) we obtain exactly Artman's[5] Eq. (15) for the resonance frequency of the uniform mode, i.e., Kittel[6] mode, in situations where the static magnetization and $\vec{H}$ are "quasi-aligned." If, on the other hand, we replace k by ik and assume $K_1 = 0$, then Eq. (4.7) reduces exactly to the simple spin-wave dispersion relation expressed by Eq. (10-10.6) of Morrish's[7] book. Also to be noted for later reference is that Eqs. (4.2) and (4.3) give the amplitude ratio

$$v \equiv \frac{\phi_1}{\theta_1} = \frac{i}{\sin\theta_0}\left[\frac{H + H_\alpha - 2Ak^2/M}{B + H_\beta - 2Ak^2/M}\right]^{1/2} . \tag{4.8}$$

Except in the case of the uniform mode, a calculation of $H = H_{\text{res}}$ from Eq. (4.7) requires that k be determined from the boundary conditions. In some cases, however, we find it more convenient to solve Eq. (4.7) for k and then use the boundary conditions for calculating H_{res}. Since Eq. (4.7) is quadratic in k^2, we can write its roots in the form

$$(2A/M)k_{1,2}^2 = H + \tfrac{1}{2}(H_\alpha + H_\beta) - 2\pi M\Delta \,, \tag{4.9}$$

$$(2A/M)k_{3,4}^2 = B + \tfrac{1}{2}(H_\alpha + H_\beta) + 2\pi M\Delta \,, \tag{4.10}$$

where Δ is given by

$$\Delta = \left[\left[1 + \frac{H_\beta - H_\alpha}{4\pi M}\right]^2 + \left[\frac{\omega}{2\pi M\gamma}\right]^2\right]^{1/2} - 1 \,. \tag{4.11}$$

In the case of experiments on iron at 16.3 GHz, the value of Δ satisfies $|\Delta| < 1$ but does not satisfy $|\Delta| \ll 1$. We assume $k_{1,2}^2 > 0$ and $k_{3,4}^2 > 0$ throughout so that all k values are indeed real and spin waves are absent. Using $k_2 = -k_1$ and $k_4 = -k_3$, we define k_1 and k_3 to be the positive roots of Eqs. (4.9) and (4.10), respectively. The solutions of Eqs. (4.2) and (4.3) can now be expressed as

$$\theta_1 = \sum_{n=1}^{4} C_n \exp(k_n \eta) \,, \tag{4.12}$$

$$\phi_1 = \sum_{n=1}^{4} C_n v_n \exp(k_n \eta) \,, \tag{4.13}$$

where v_n is the value of v for $k = k_n$ and the C_n are unknown coefficients.

To derive the dynamic boundary conditions we retain only those terms in the θ,ϕ form of the boundary condition (2.7) which are of first order in θ_1 and ϕ_1. Thus we obtain

$$2A\frac{\partial\theta_1}{\partial n} + \theta_1(K_s - 2K_{ss})\cos 2\theta_0 = 0 \,, \tag{4.14}$$

$$A\frac{\partial\phi_1}{\partial n} - \phi_1 K_s = 0 \,, \tag{4.15}$$

as the boundary conditions for θ_1 and ϕ_1. Substitution of Eq. (4.12) into Eq. (4.14) and of Eq. (4.13) into Eq. (4.15) then yields two equations which must be satisfied at each of the crystal boundaries ($\eta = \pm L$). This requirement leads to a set of four homogeneous linear equations for C_1, C_2, C_3, C_4 whose solution gives $H = H_{\text{res}}$. Before proceeding, however, we demonstrate that our formalism can indeed produce a solution for H_{res}. The dynamical problem involves not only the four equations that result from Eqs. (4.14) and (4.15) but also the equations of motion (4.2) and (4.3), or equivalently, Eqs. (4.7) and (4.8). Altogether, therefore, there are six independent equations. There are also six unknowns, namely, H, k, v, and the ratios C_4/C_1, C_3/C_1, C_2/C_1. Thus the six equations should indeed be soluble.

Returning to the above-mentioned set of four homogeneous linear equations for C_1, C_2, C_3, C_4, we denote the secular determinant of their coefficients by D and call it the "boundary condition determinant." In order that these four equations possess a nonvanishing solution, we must have

$$D = 0 , \tag{4.16}$$

where D is given by

$$D = \begin{vmatrix} (K_s^* + Ak_1)\exp(k_1 L) & (K_s^* - Ak_1)\exp(-k_1 L) & \cdots & \cdots \\ (K_s^* - Ak_1)\exp(-k_1 L) & (K_s^* + Ak_1)\exp(k_1 L) & \cdots & \cdots \\ (K_s - Ak_1)v_1\exp(k_1 L) & (K_s + Ak_1)v_1\exp(-k_1 L) & \cdots & \cdots \\ (K_s + Ak_1)v_1\exp(-k_1 L) & (K_s - Ak_1)v_1\exp(k_1 L) & \cdots & \cdots \end{vmatrix} . \tag{4.17}$$

The third and fourth columns of D are each denoted by $\cdots$ for brevity. They are identical to the first and second columns, respectively, except that k_1 is replaced by k_3 and v_1 by v_3. The symbol K_s^* is an abbreviation for the quantity

$$K_s^* = \tfrac{1}{2}(K_s - 2K_{ss})\cos 2\theta_0 . \tag{4.18}$$

Next we multiply each row of D by L and form suitable linear combinations of rows and of columns. In this way we manipulate D into a form in which eight of its elements are zero. Noting that neither $\cosh k_1 L$ nor $\cosh k_3 L$ can vanish, we then divide two of the columns by $\cosh k_1 L$ and the other two columns by $\cosh k_3 L$. Upon suitable interchanges of rows and of columns the matrix of D can be brought into block form. Specifically, we find that Eq. (4.16) is replaced by the requirement

$$D_{\text{I}} D_{\text{II}} = 0 , \tag{4.19}$$

where D_{I} and D_{II} are given by

$$D_{\text{I}} = \begin{vmatrix} K_s^* L + Ak_1 L \tanh k_1 L & K_s^* L + Ak_3 L \tanh k_3 L \\ (K_s L - Ak_1 L \tanh k_1 L)v_1 & (K_s L - Ak_3 L \tanh k_3 L)v_3 \end{vmatrix} , \tag{4.20}$$

$$-D_{\text{II}} = \begin{vmatrix} K_s^* L \tanh k_1 L + Ak_1 L & K_s^* L \tanh k_3 L + k_3 L \\ (K_s L \tanh k_1 L - Ak_1 L)v_1 & (K_s L \tanh k_3 L - k_3 L)v_3 \end{vmatrix} . \tag{4.21}$$

The condition (4.19) can be satisfied only if at least one of the quantities D_{I} and D_{II} always vanishes. Let us first make the assumption $D_{\text{II}} = 0$. If we now consider the special case $| k_1 L | \ll 1$ and $| k_3 L | \ll 1$, then we easily find that $D_{\text{II}} = 0$ leads to $v_1/v_3 = 1$. But Eqs. (4.8)–(4.10) may be combined to give $v_1/v_3 = R$, where R is an abbreviation for the quantity

$$R = \frac{4\pi M\Delta - (H_\beta - H_\alpha)}{4\pi M(2+\Delta) + (H_\beta - H_\alpha)} . \tag{4.22}$$

Thus the assumption $D_{\text{II}} = 0$ leads to the prediction $R = 1$ in this special case. But Eq. (4.31) (see below) can easily be combined with Eq. (4.22) to show that (since $M \neq 0$) the prediction $R = 1$ is incorrect. Accordingly, D_{II} is not always zero so that Eq. (4.19) forces us to conclude that it is D_{I} which always vanishes. By combining $D_{\text{I}} = 0$ with Eq. (4.20) we obtain

$$\frac{(\Gamma_1 + H_s^*)(\Gamma_3 - H_s)}{(\Gamma_1 - H_s)(\Gamma_3 + H_s^*)} = R \tag{4.23}$$

which contains the abbreviations ($i=1,3$):

$$\Gamma_i=(2A/M)k_i^2(\tanh k_iL)/(k_iL)\ , \tag{4.24}$$

$$H_s=2K_s/(ML);\ \ H_s^*=2K_s^*/(ML)\ . \tag{4.25}$$

So far we have made only three principal assumptions in our treatment of ferromagnetic resonance, namely, linearization of the Landau-Lifshitz equation, quasialignment of the static magnetization with $\vec{\mathrm{H}}$, and predominance of the surface-induced mode. In addition, we assumed temporarily that θ_0 is zero or $\pi/2$. At this point we introduce three further assumptions. Firstly, we assume that the approximation

$$|\,k_1L\,| \ll 1 \tag{4.26}$$

is satisfied, i.e., that half the crystal thickness is small compared to the decay distance of the oscillations associated with k_1 and k_2. Secondly, we assume that the approximation

$$|\,H_s/\Gamma_3\,|^{1/2} \ll 1 \tag{4.27}$$

is also satisfied, i.e., that the "surface-anisotropy field" H_s is negligible compared to the product of the "exchange field" $(2A/M)k_3^2$ and the dimensionless ratio $(\tanh k_3L)/(k_3L)$. Thirdly, we assume that $|K_s^*/K_s\,|$ is at most unity so that [because of Eq. (4.18)] the values of K_s and K_{ss} satisfy the limitation

$$|\,1-2K_{ss}/K_s\,| \le 2\ . \tag{4.28}$$

On the basis of these three assumptions Eq. (4.23) leads to the simple relation

$$(2A/M)k_1^2=H_s(R+K_s^*/K_s)/(R-1) \tag{4.29}$$

which we combine with Eqs. (4.25) and (4.18) and then substitute into Eq. (4.9). Thus we obtain our central result for the resonance field

$$H_{\mathrm{res}}=(H_{\mathrm{res}})_{\mathrm{unif}}-\frac{K_s[(1-2K_{ss}/K_s)\cos2\theta_0+2R\,]}{LM(1-R)}\ , \tag{4.30}$$

where

$$(H_{\mathrm{res}})_{\mathrm{unif}}=2\pi M\Delta-\tfrac{1}{2}(H_\alpha+H_\beta) \tag{4.31}$$

is the resonance field of the uniform mode. The quantities Δ and R depend on θ_0 and are given by Eqs. (4.11) and (4.22), respectively.

B. Generalizations and discussions

We note that Eq. (4.30) does not contain the exchange stiffness constant A even though it could not have been derived if A were zero. The absence of A from Eq. (4.30) turns out to be a consequence of the approximation (4.26). If we carry the calculation to second rather than first order in $|\,k_1L\,|$, we find that H_{res} does indeed contain A even if the approximation (4.27) remains satisfied.

Among the most interesting aspects of Eq. (4.30) we discuss the following: (a) Equation (4.30) predicts that within its range of applicability, i.e., for ultrathin crystals, H_{res} is a linear function of $1/L$. (b) Equation (4.30) shows that the slope of the predicted H_{res} vs $1/L$ line associated with the [001] axis may differ in sign and/or in magnitude from the slope associated with the [110] axis. One of our motivations for introducing K_{ss} in addition to K_s was, in fact, to endow Eq. (4.30) with sufficient generality for predicting two *different positive* values for the slopes of H_{res} vs $1/L$ along [001] and [110], i.e., for $\psi=0$ and $\psi=\pi/2$. (c) Equation (4.30) predicts that as a consequence of surface anisotropy the easy and hard axes of magnetization may be interchanged if the crystal thickness is decreased sufficiently. (d) Equation (4.30) becomes invalid for the case $L\rightarrow0$ because then the approximation (4.27) is no longer satisfied. In this case, moreover, Eq. (4.23) becomes $R=1$, a prediction which we showed to be incorrect, so that for the case $L\rightarrow0$ even Eq. (4.23) is invalid.

The *behavior* predicted in (a), (b), and (c) has been observed *experimentally*[1] at room temperature. As stated previously,[1] the values of the surface-anisotropy constants ($K_s=-3.0$ ergs/cm^2 and $K_{ss}=-1.4$ ergs/cm^2) deduced from the experimental data of Fig. 7 of Ref. 1 were highly tentative because with the use of these values the "small surface-anisotropy approximation" (4.27) of the present paper is only poorly satisfied. In order to clarify this situation, we now avoid the use of the approximation (4.27) by fitting the same experimental data[1] (for four thicknesses in the range 18 Å $\le 2L \le$ 55 Å) on the basis of Eq. (4.23) rather than Eq. (4.30). We find that the predicted curves of H_{res} vs $1/L$ associated with both the [001] and [110] axes are almost straight lines and that the deduced values of the surface-anisotropy constants are $K_s=-(3.2\pm2)$ ergs/cm^2 and $K_{ss}=-(1.5\pm1)$ ergs/cm^2. Since the predictions of Eq. (4.23) depend on the quantity K_s-2K_{ss} very sensitively, an adequate fitting of the experimental data requires that the values of K_s and K_{ss} be specified more precisely than indicated above. We note that the data can be fitted within the experimental error of $2L$ (which is $\pm 10\%$) by using, for example, $K_s - 2K_{ss} = -0.298$ erg/cm^2 for the [001] axis, $K_s - 2K_{ss} = 0.0545$ erg/cm^2 for the [110] axis, and $K_s = -3.156$ ergs/cm^2 for both axes. When combined with this value of K_s, both of these values of $K_s - 2K_{ss}$ lead to K_{ss} values which agree with the value $K_{ss} = -(1.5 \pm 1)$ ergs/cm^2 specified above. It should be emphasized that the values of K_s and K_{ss} deduced from the experiments are *effective* values because they do not refer to iron crystals surrounded by a vacuum but to iron crystals grown epitaxially on GaAs and covered by an epitaxial film of Al. This means, in particular, that K_s and K_{ss} represent some sort of average values and that a more accurate theoretical description of the data (which is not warranted at present) would require the introduction of *two* sets of surface-anisotropy constants. Furthermore, the values of K_s and K_{ss} may represent, in part, a simulation of the effects of *inhomogeneous* strains arising from imperfect epitaxy. Although homogeneous strains can, of course, give rise to a uniaxial volume anisotropy and thus shift the H_{res} vs $1/L$ curves for the [001] and [110] axes in opposite directions along the H_{res} coordinate axis, such strains cannot produce any dependence of H_{res} on $1/L$. Still another reason for labeling K_s and K_{ss} as "effective" surface anisotropies is that they may include effects caused by gradients of the spontaneous magnetization near the film surfaces.

Next we illustrate the predictions of Eq. (4.30) by means of a hypothetical example pertaining to bcc iron at room temperature. For this example, in contrast to the experimental example discussed above, both of the approximations underlying Eq. (4.30) are satisfied. By adopting the representative values $4\pi M = 2.154 \times 10^4$ emu, $K_1 = 4.5 \times 10^5$ ergs/cm^3, $A = 2.0 \times 10^{-6}$ erg/cm, $g = 2.09$, and choosing $\omega/(2\pi) = 16.3$ GHz, we compute Δ, R, and $(H_{\text{res}})_{\text{unif}}$ from Eqs. (4.11), (4.22), and (4.31), respectively. Thus we obtain $\Delta = 0.126$, $R = 0.0592$, $(H_{\text{res}})_{\text{unif}} = 830$ Oe for $\vec{H}$ along [001] and $\Delta = 0.0935$, $R = 0.0632$, $(H_{\text{res}})_{\text{unif}} = 1138$ Oe for $\vec{H}$ along [110]. To compute the $1/L$-dependent shift of H_{res}, we use the last term of Eq. (4.30) and arbitrarily choose $L = 2 \times 10^{-7}$ cm. Since neither K_s nor K_{ss} has been measured reliably so far, we use Néel's[4] theoretical estimate $K_s = -0.229$ erg/cm^2 and assume, as in Ref. 1, the value $2K_{ss}/K_s = 0.9$. The shift $H_{\text{res}} - (H_{\text{res}})_{\text{unif}}$ computed in this way is 155 Oe for $\vec{H}$ along [001] and 19 Oe for $\vec{H}$ along [110]. Using for $\vec{H}$ along [001] the resulting value $H_{\text{res}} = 985$ Oe, we obtain from Eqs. (4.9) and (4.10) the values $k_1 = 2.58 \times 10^5$ cm^{-1} and $k_3 = 3.23 \times 10^6$ cm^{-1}. For $\vec{H}$ along [110] we have $H_{\text{res}} = 1157$ Oe and obtain $k_1 = 8.96 \times 10^4$ cm^{-1} and $k_3 = 3.18 \times 10^6$ cm^{-1}. It should be noted that the values of the parameters used in this example do satisfy the approximations (4.26) and (4.27) so that the use of Eq. (4.30) is indeed justified. If we had assumed $K_{ss} = 0$ then we would have obtained $H_{\text{res}} - (H_{\text{res}})_{\text{unif}} < 0$ for $\vec{H}$ along [110]. Because of Eq. (4.9), this would have meant $k_{1,2}^2 < 0$ which is contrary to our assumption and leads to spin waves.

In order to explore a possible generalization of Eq. (4.30), we now consider an experimental situation in which the extrapolation to $1/L = 0$ of a linear plot of H_{res} vs $1/L$ fails to yield the calculated value of $(H_{\text{res}})_{\text{unif}}$. This kind of situation may well indicate (provided the assumed value of the magnetization is correct) that the volume anisotropy contains some contribution in addition to K_1. To show by means of an example how such a contribution can be incorporated into Eq. (4.30), we assume that (as a result of imperfect epitaxy or other growth conditions) the iron crystal possesses a uniaxial volume anisotropy of the form $K_u \sin^2\theta_0$. This anisotropy can be included in Eq. (4.30) by simply adding $(2K_u/M)\cos 2\theta_0$ to H_α and adding $(2K_u/M)\cos^2\theta_0$ to H_β. It should be noted that this modification of Eq. (4.30) affects not only $(H_{\text{res}})_{\text{unif}}$ but also R. The method described can be used, of course, for incorporating volume anisotropies additional to K_1 into Eq. (4.23) as well as into Eq. (4.30).

Turning now to cases in which the approximations (4.26) and (4.27) are not both satisfied, we find that the calculated H_{res} are no longer given by an expression as simple as Eq. (4.30). The method we suggest for such cases is to substitute Eqs. (4.9) and (4.10) into Eq. (4.23) and then solve the resulting equation for H_{res} numerically. This method requires, of course, that K_s and K_{ss} be known or else determinable by fitting the curves of H_{res} vs $1/L$ measured along the [001] and [110] axes. It should be noted, however, that the usefulness of the H_{res} value calculated in this way may well be questionable if L is too large. The reason is that the derivation of Eq. (4.23) is based on the surface-induced mode and ignores spin waves. For sufficiently large values of L, furthermore, the exchange interactions existing in the skin depth should also be considered, as discussed in Ref. 3. Nevertheless, the suggested numerical calculation of H_{res} should still be useful because L values do exist which are too large to satisfy the approximation (4.26) but too small to allow skin-depth-induced exchange effects or even spin-wave resonance excitation by surface conditions.

Up to this point our calculations of H_{res} have assumed that θ_0 is zero or $\pi/2$ so that each of the quantities $\delta\theta_0$, $(\delta\theta_0)'$, and $(\delta\theta_0)''$ vanishes, as mentioned near the beginning of Sec. IV A. Now we seek to calculate H_{res} for values of θ_0 which are arbitrary and thus (see Sec. III) spatially nonuniform. We find that this task is quite complicated because if θ_0 is arbitrary then the equation of motion and the boundary condition are augmented by new terms, and because the coefficients of the resulting differential equations are no longer constant. By introducing two additional approximations, however, we are able to show that they cause the equation of motion, the boundary condition and the quasialignment correction to be unchanged. This means that Eq. (4.30), our explicit result for H_{res}, remains approximately valid even if θ_0 is not restricted to zero or $\pi/2$. A proof of these assertions is rather lengthy and will be omitted, but a formulation and discussion of the newly introduced approximations is clearly necessary. The less stringent one of these requires that $|(K_s-2K_{ss})/(ML)|$ be negligible compared to $4\pi M$. Using the values of M, L, and K_s adopted above, we find that this approximation is well satisfied for $2K_{ss}/K_s=0.9$. The more stringent one of the new approximations requires that $|\delta\theta_0|$ be negligible compared to unity. To determine whether this can be achieved we make a numerical estimate based on the value $\theta_{00}=\cos^{-1}(1/\sqrt{3})$ which was chosen because it is "far" from the values $\theta_{00}=0$ and $\theta_{00}=\pi/2$ where $\delta\theta_0$ vanishes. Using $2K_{ss}/K_s=0.9$ and the parameters adopted above, we obtain $H=H_{\text{res}}=1765$ Oe by assuming the (as yet unproven) applicability of Eq. (4.30) to arbitrary θ_0 values and find that Eq. (3.10) gives $l=1.28\times10^{-6}$ cm. If we take $L=2\times10^{-7}$ cm, as before, then the approximation (3.16) is well satisifed so that Eq. (3.17) is valid. Since the latter yields $\delta\theta_0=0.022$, which is negligible compared to unity, it is clear that Eq. (4.30) is a good approximation if the parameters have the values chosen above. It is seen, therefore, that K_{ss} as well as K_s must be known (or determined by measurements involving $\theta_0=0$ and $\theta_0=\pi/2$) before the applicability of Eq. (4.30) to arbitrary values of θ_0 can be ascertained reliably.

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