REVERSIBLE PROPERTIES OF POLYCRYSTALLINE FERROMAGNETS*†—I

THEORY OF THE EXPECTED VARIATION OF THE REVERSIBLE PROPERTIES WITH MAGNETIZATION

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Abstract—Using a statistical model, equations are developed for the variation of the reversible susceptibility both parallel with and normal to the biasing magnetization as a function of the magnetization assuming that the susceptibility arises by domain rotation. The results are contrasted with previous results based upon domain-wall motion. It is concluded that the theory points out a new technique for the separation of the origins of the susceptibility. Equations are also given for the expected variation of the differential magnetostriction with magnetization both parallel with and normal to the field and for both domain-wall motion and domain rotation.

Quantitative results depend upon the fraction of the moments oriented in each direction. A function describing this distribution is discussed.

An expression is given for the susceptibility matrix arising from domain rotation as a function of magnetization.

INTRODUCTION

THE reversible properties of polycrystalline ferro-magnets result from the ferromagnetic nature of the material when a differential disturbance of some kind is applied. To fit the reversible criterion, internal oscillations to the material must be carried out about fixed equilibrium positions and must be vanishingly small. Thus, reversibility indicates repeatability and, for the special case of a zero-frequency applied disturbance, it implies reversibility in the thermodynamic sense of zero energy dissipation. GANS[1] pointed out that the

reversible susceptibility of iron could be closely approximated analytically as a function of the magnetization only, independently of the magnetic history. BROWN[2,3,4] extended GANS' work to include the cases of the derived distribution function $f(\theta)\,d\theta$, the fraction of the presumed equal volume domains oriented between θ and $\theta + d\theta$, and with this knowledge was able to express the reversible susceptibilities and the magnetostriction in terms of the magnetization.

GRIMES and MARTIN[5] extended BROWN's work to include the reversible susceptibility measured both normal and parallel to the magnetization. They compared their results favorably with measurements on several ferrites. In the meantime, TEBBLE and CORNER[6] and STONER[7] pointed out that the reversible susceptibility is not a unique function of the magnetization, but does depend upon the magnetic history. It is the point of view of this paper that although the susceptibilities do indeed depend upon the magnetic history, as a first approximation they can be first considered as functions of the magnetization only. Variations

from this idealized behavior can then be discussed.

It is pointed out that under usual conditions the equations for the parallel reversible susceptibility χ_{rp} and the transverse (normal) reversible susceptibility χ_{rt} as given by BROWN[4] and by GRIMES and MARTIN[5] are applicable when the susceptibility has its origin in domain-wall motion. In this paper the susceptibility matrix is derived as a function of the magnetization when that matrix arises by domain rotation. The diagonal terms give the reversible susceptibilities χ_{rp} and χ_{rt} due to domain rotation.

The expected variation of the parallel differential magnetostriction with magnetization has been computed by BOZORTH and WILLIAMS.[8] They assumed that both the reversible susceptibility and the change in biasing moment had their origin in the rotation of the magnetic moments. In this paper equations are derived for each of the four possibilities when the biasing moment changes by domain-wall motion. The differential magnetostriction of several ferrites has been discussed by VAN DER BURGT.[9]

THE REVERSIBLE QUANTITIES, GENERAL CASE

The susceptibilities

The magnetic properties of a ferromagnet can be described in terms of the function $f(\theta)\,d\theta$, the fraction of the total atomic moments of the material which are oriented between θ and $\theta+d\theta$ with respect to the applied magnetic field. The magnetization is then given by:

$$M = M_s \frac{\int_0^\pi f(\theta)\sin\theta\cos\theta\,d\theta}{\int_0^\pi f(\theta)\sin\theta\,d\theta} = M_s \langle\cos\theta\rangle \quad (1)$$

where the symbol $\langle\ \rangle$ indicates the weighted average over the polycrystal, M is the gross magnetization, M_s is the spontaneous or saturation moment and θ is the angle between the gross magnetization and the moment vector in question. The validity of equation 1 is limited by the equality of the saturation and spontaneous moments but not by any particular magnetization mechanism. A discussion of $f(\theta)$ will be left until later. The reversible susceptibility measured parallel to the applied magnetic field is:

$$\chi_{rp} = M_s \frac{\partial\langle\cos\theta\rangle}{\partial H} \quad (2)$$

where H is the magnetic field, subject to the condition that the same mechanisms which allow the static value of M to be different from zero also give rise to the reversible susceptibility. The partial derivative indicates that the external stress and the magnetic history of the specimen remain constant. The latter implies that the susceptibility is initially measured opposite in direction to the change in H which brought the material to the position (M, H) at which the measurement was taken. This is assumed to be the susceptibility measured by a vanishingly small alternating field.

The reversible susceptibility for dilute material can also be calculated by applying the usual torque equation to all magnetic dipoles present in the system. This torque equation with the added damping term as given by LANDAU and LIFSHITZ[10] can be written as:

$$\frac{\partial M}{dt} = \gamma(M\times H) - \frac{\gamma\epsilon}{M_s}[M\times(M\times H)] \quad (3)$$

where t is the time, γ the magnetomechanical coupling coefficient, and ϵ a dimensionless parameter proportional to the energy loss. It is the usual convention in solving equation 3 to assume that the spontaneous moment is oriented along the z axis and that the effective magnetic field in the z direction is large compared with the applied reversible field. The magnetization from equation 3 can be written as a function of H using matrix notation, as:

$$(M) = (X)(H). \quad (4)$$

Let us now assume that all time varying quantities vary as $e^{\epsilon\omega t}$, and, to obtain a diagonalized matrix for (X), we use the co-ordinate system $(x+iy, x-iy, z)$.[11] Using this notation the operator j represents a time delay of $(\pi/2\omega)$ sec and the operator i represents a $\pi/2$ spatial rotation. The susceptibility matrix then becomes:

$$X_{11} = \frac{M_x+iM_y}{H_x+iH_y} = \frac{M_+}{H_+} = X_+ = \frac{\omega_0}{\omega_1+\dfrac{ij\omega}{1+i\epsilon}}$$

$$X_{22} = \frac{M_x-iM_y}{H_x-iH_y} = \frac{M_-}{H_-} = X_- = \frac{\omega_0}{\omega_1-\dfrac{ij\omega}{1-i\epsilon}}$$

$$(5)$$

$$X_{33} = X_{12} = X_{13} = X_{23} = X_{21} = X_{31} = X_{32} = 0,$$

where $\omega_0 = \gamma M_s$, $\omega_1 = \gamma H_z$ and ω is the applied radial frequency. The real parts of equation 5 with respect to i represent diagonal terms in the co-ordinate system (x, y, z) and imaginary parts represent off-diagonal terms. Real parts with respect to j represent energy stored and imaginary parts represent energy dissipated.

To extend equation 4 to a polycrystalline medium, let us first average the susceptibility matrix over a polycrystalline sample, and then correct for interdomain effects. The procedure for averaging is to transform the susceptibility matrix from one based upon the z axis to a general matrix in terms of the Euler angles (θ, φ, ψ), then to average the result over a polycrystal. If the subscript d indicates the z axis based system and g the arbitrarily based system, then $M_d = X_d H_d$ and $M_g = aX_d H_d = aX_d a^{-1} H_g = X_g H_g$ where a is the Euler transformation matrix expressed in the system $(x+iy, x-iy, z)$. Both a and a^{-1} have been computed by PARK.[11] In averaging over a polycrystal, if all angles around the applied field are equally likely (physically this means that if crystalline orientation exists, it has rotational symmetry about the field direction) then all off-diagonal terms of the matrix X_g average to zero and X_g is given by:

$$X_g = \tfrac{1}{4}\begin{bmatrix} \langle(1+\cos\theta)^2\rangle X_+ + \langle(1-\cos\theta)^2\rangle X_-, & 0, & 0 \\ 0, & \langle(1-\cos\theta)^2\rangle X_+ + \langle(1+\cos\theta)^2\rangle X_-, & 0 \\ 0, & 0, & 2(1-\langle\cos^2\theta\rangle)(X_+ + X_-) \end{bmatrix} \quad (6)$$

In an attempt to account for the interactions of neighboring domains whose moments are rotating consider the effective field at a particular domain because of its neighbors. Following PARK[12] let us assume that the neighboring domains have an averaged moment equal to that of the gross material. PARK showed that the interaction could be described as $h = H - \rho M$ where h is the effective magnetic field felt by the domain, H is the applied alternating field, 'M' the macroscopic alternating magnetization and ρ a constant. External demagnetizing factors are describable by a similar equation. Thus it is to be assumed that the term $(-\rho M)$ describes effects due to the external geometry and the internal domain interactions. This can be understood by assuming that the full demagnetizing factor of the domain by itself is only partially cancelled by its contact with neighboring domains. Putting the h's into the susceptibilities:

$$M_g = \frac{X_g}{1+\rho X_g} H_g = \chi_g H_g. \quad (7)$$

The susceptibilities (χ) from equation 7 are formally identical with the susceptibilities (X) of equation 6 when ω_1 is replaced by $(\omega_1 + \rho\omega_0)$. Thus the effect of external demagnetizing factors and nearest-neighbor domain interactions as here considered are describable as an increased effective anisotropy field. This is also true when the effective fields vary throughout the sample.[12]

In terms of the system (x, y, z) and the χ's, the averaged susceptibility matrix is:

$$\chi_g = \tfrac{1}{4}\begin{bmatrix} (1+\langle\cos^2\theta\rangle)(\chi_- + \chi_+), & 2\langle\cos\theta\rangle(\chi_- - \chi_+), & 0 \\ -2\langle\cos\theta\rangle(\chi_- - \chi_+), & (1+\langle\cos^2\theta\rangle)(\chi_- + \chi_+), & 0 \\ 0, & 0, & 2(1-\langle\cos^2\theta\rangle)(\chi_- + \chi_+) \end{bmatrix} \quad (8)$$

For a nonoriented polycrystal in the virgin state it follows that $\langle\cos^2\theta\rangle = 1/3$ and $\langle\cos\theta\rangle = 0$ so equation 8 is proportional to the unit matrix. For saturated material $\langle\cos\theta\rangle = \langle\cos^2\theta\rangle = 1$ and the matrix is the susceptibility matrix of a single crystal.[13] The off-diagonal terms are proportional to the macroscopic magnetization, a fact in agreement with previous calculations of RADO.[14] Since $f(\theta)$ is not known, the diagonal terms cannot be calculated unambiguously from a knowledge of the normalized magnetization only.

Remembering that equation 8 is based upon the biasing magnetization being in the z direction, the transverse and parallel reversible susceptibilities for domain rotation are seen to be

$$\chi_{rt}{}^r = \frac{\chi_- + \chi_+}{4}(1+\langle\cos^2\theta\rangle);$$
$$\chi_{rp}{}^r = \frac{\chi_- + \chi_+}{2}(1-\langle\cos^2\theta\rangle) \quad (9)$$

independently of the particular averaging function $f(\theta)$. The susceptibilities are then related by:

$$(\chi_+ + \chi_-) = 2\chi_{rt}{}^r + \chi_{rp}{}^r = 3\chi_0{}^r. \quad (10)$$

where the superscript r's indicate domain rotation, the subscript r's indicate a reversible property and

the subscripts p and t represent parallel and transverse respectively.

The assumptions made in deriving equations 9 and 10 are:

1. that the susceptibility is describable by the small signal solution to the LANDAU-LIFSHITZ equation;

2. that the sum of the biasing field and magnetic anisotropy effects are describable as an effective magnetic field; and

3. that either domain interactions do not exist or that they are describable by the field $(-\rho M)$. Presumably, so long as assumption (2) is valid, assumption (1) will also be.

From equation 10 it is apparent that so long as $(\chi_- \div \chi_+)$ remains constant, if $\chi_{rp}{}^r$ is a monotonic decreasing function of M, then $\chi_{rt}{}^r$ will be a monotonic increasing function with ratio of slopes equal (-2).

The magnetostrictions

If a demagnetized sphere of polycrystalline material is magnetized the material ceases to be spherical and becomes elongated or shortened in the field direction. The coefficient λ_s is defined to be:

$$\lambda_s = \frac{\Delta l}{l} = \left(\frac{l_m - l_{sp}}{l} \right)$$

where l_m is the length of the saturated material in the field direction and l_{sp} is the diameter of the sphere. Because parallel-antiparallel orientation changes leave Δl unaltered the angular dependence of the magnetostriction must involve even powers of $\cos \theta$. Under the restricting conditions that the magnetostriction is independent of the crystallographic direction (i.e., $\lambda_s = \lambda_{100} = \lambda_{111}$) then for each crystallite:[15,16,17]

$$\lambda_d = \tfrac{3}{2} \lambda_s (\cos^2 \theta - \tfrac{1}{3}). \qquad (11)$$

where λ_d is the ratio $\Delta l / l$ for each crystallite as a function of θ. If the effects of microscopic strain interactions are completely described by the determination of $f(\theta)$, then each crystallite separately contributes according to equation 11 and the gross magnetostriction λ_g is given by:

$$\lambda_g = \tfrac{3}{2} \lambda_s (\langle \cos^2 \theta \rangle - \tfrac{1}{3}). \qquad (12)$$

Subject to the condition that the susceptibility has

its origin in the mechanism which produced a gross magnetization other than zero, the parallel differential magnetostriction is given by:

$$d_p = \frac{\partial \lambda_g}{\partial H} = \frac{3 \lambda_s}{2} \frac{\partial \langle \cos^2 \theta \rangle}{\partial H}, \qquad (13)$$

where the partial indicates that the magnetic history and the external stress are constant. Since λ and M are both functions of θ, the differential magnetostriction can also be expressed as:

$$d = \left(\frac{\partial \lambda_g}{\partial H} \right) = \left(\frac{d \lambda_g}{d \theta} \bigg/ \frac{dM}{d \theta} \right) \frac{dM}{dH}. \qquad (14)$$

For alternating differential fields dM/dH is taken to be the reversible susceptibility χ_r. Thus, combining equations 1 and 11 for each crystallite with the bracket in equation 14.

$$d = \frac{3 \lambda_s}{M_s} (\chi_r \cos \theta) \qquad (15)$$

for each domain. Equation 15 can be combined with the non-averaged values of the susceptibility for each domain corresponding to equation 9, then the product averaged over the polycrystal to obtain:

$$d_p{}^r = \frac{9 \lambda_s \chi_0{}^r}{2 M_s} [\langle \cos \theta \rangle - \langle \cos^3 \theta \rangle] \qquad (16a)$$

$$d_t{}^r = \frac{9 \lambda_s \chi_0{}^r}{4 M_s} [\langle \cos \theta \rangle + \langle \cos^3 \theta \rangle]. \qquad (16b)*$$

From equation 16 it is apparent that for constant χ_0 the maximum value of d should occur for crossed magnetic fields with $M = M_s$. Since H_{ap} is not negligible for $M = M_s$, this maximum value of $(9 \lambda_s \chi_0{}^r / 2 M_s)$ cannot be attained in nonoriented material.

Results of an effective history field

For many materials, the magnetic moment

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* The equation for $d_p{}^r$ is similar in form to the expression for Λ given by BOZORTH and WILLIAMS.[8] Using their approach (a) the biasing moment as well as the differential moment is assumed to vary by rotational processes, (b) it is not clear how the values of 1·37 to 1·60 were obtained for their f, and (c) they put $\langle \cos \theta \rangle^3 = \langle \cos^3 \theta \rangle$.

vectors remain oriented very nearly along crystallographically "easy" directions when $M \lesssim M_s/2$. Under these conditions the parallel reversible susceptibility and differential magnetostriction when calculated by equations 2 and 13 must also have magnetic moments in "easy" directions. This criterion is consistent with the movement of domain walls but not with rotation of domains. Thus so long as the static moment is oriented in "easy" directions equations 2 and 13 will be applicable to domain-wall motional effects.

It is convenient to introduce the dimensionless parameter η defined by $\eta = A(H_{ap}+D)$, where H_{ap} is the applied biasing field, A is a constant and D is a history dependent field. It is now necessary to make the restricting assumption that the magnetic history of the sample is completely described by the effective magnetic field D. The physical significance of the parameter η and the field D are discussed in the appendix. This assumption implies that the sum of the component of local moments in the field direction uniquely determines that in all other directions. With this assumption $f(\theta)$ and the resulting $\langle \cos^m \theta \rangle$ will be unique functions of η, and experimental plots of different functions of $\langle \cos^m \theta \rangle$ should eliminate hysteresis effects. Modification of this assumption will be discussed later. Since the $\langle \cos^m \theta \rangle$ are assumed to be unique functions of η, let us define:

$$F(\eta) \equiv \langle \cos \theta \rangle$$
$$G(\eta) \equiv \langle \cos^2 \theta \rangle \qquad (17)$$
$$H(\eta) \equiv \langle \cos^3 \theta \rangle.$$

The values of M and $\chi_{rp}{}^w$ as given in Table 1, row 1, follow from equations 1, 2, and 17. Note that the partial with respect to H is taken at constant D. D is presumed to change with each change in domain configuration. The values for $\chi_{rp}{}^r$, $\chi_{rt}{}^r$, $d_p{}^r$, $d_t{}^r$, $d_p{}^w$ and the susceptibility matrix in Table 1, row 1, follow from equation 17 and equations 9, 16, 13, and 8 respectively. To compute $\chi_{rt}{}^w$ note that when the biasing field is increased by a normal

FIG. 1. The relationship utilized to determine the transverse properties due to domain-wall motion.

K

differential field the increasing magnitude is second order in (dH/M) (i.e., $H = \sqrt{[H_{ap}{}^2 + (\Delta H)^2]} \cong H_{ap}\sqrt{[1 + \frac{1}{2}(\Delta H/H_{ap})^2]}$), but the change normal to H_{ap} is first order in $(\Delta H/H_{ap})$. Thus, for a differential field, the transverse susceptibility can be seen from the similar triangles of Fig. 1, if the field and moment remain parallel:

$$\chi_{rt}{}^w \equiv \lim_{\Delta\eta \to 0} A\frac{\Delta M}{\Delta \eta} = \frac{AM}{\eta} = AM_s\frac{F(\eta)}{\eta}. \quad (18)$$

The transverse-field differential magnetostriction due to domain-wall motion follows by a similar argument and is given by:

$$d_t{}^w = 3/2\lambda_s \frac{A}{\eta}[G(\eta) - \tfrac{1}{3}]. \quad (19)$$

Upon comparing the parallel and transverse susceptibilities it is apparent that:

$$\chi_{rp}{}^w = \chi_{rt}{}^w + \eta\frac{d\chi_{rt}{}^w}{d\eta}. \quad (20)$$

Thus, if $\chi_{rp}{}^w$ is a monotonic decreasing function of M, then $\chi_{rt}{}^w$ must also be decreasing in contrast with the expected results from domain rotation.

The differential equation of motion which describes the movement of 180° domain walls is

$$2\mu_0 M_s H = \alpha x + \beta\dot{x} + m\ddot{x} \quad (21)$$

where x represents the spatial co-ordinate of the wall measured from its equilibrium position in the absence of the field, and α, β and m are constants and μ_0 is the permeability of free space. Inherent in equations 18 and 19 is the assumption that $\alpha x \gg (\beta\dot{x} + m\ddot{x})$. The magnetic Q of the material for a sinusoidal applied signal is:

$$Q = \frac{\alpha - \omega^2 m}{\omega\beta}. \quad (22)$$

Thus, so long as the material remains inductive, the Q remains large, and wall-motion processes predominate, equations 18 and 19 will be valid.

The distribution function $f(\theta)$

The function $f(\theta) \, d\theta$ is, by definition, the fraction of the magnetic moments in the system of interest at an angle between θ and $\theta + d\theta$ with respect to the biasing field. $f(\theta)$ will be constant for nonoriented
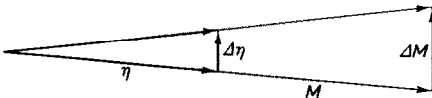
Table 1. The reversible quantities

	Magnetization	Reversible susceptibility		Differential magnetostriction	
		Wall motion	Domain rotation	Wall motion	Domain rotation
Parallel fields, Arbitrary $F(\theta)$	$M = M_s F(\eta)$	$\chi_{rp}{}^w = AM_s \dfrac{dF(\eta)}{d\eta}$	$\chi_{rp}{}^r = \dfrac{\chi_- + \chi_+}{2}(1 - G(\eta))$	$d_p{}^w = \tfrac{3}{2}\lambda_s A \dfrac{dG(\eta)}{d\eta}$	$d_p{}^r = \dfrac{9\lambda_s\chi_0}{2M_s}[F(\eta) - H(\eta)]$
Transverse fields		$\chi_{rt}{}^w = AM_s \dfrac{F(\eta)}{\eta}$	$\chi_{rt}{}^r = \dfrac{\chi_- + \chi_+}{4}(1 - G(\eta))$	$d_t{}^w = \tfrac{3}{2}\lambda_s A \dfrac{1}{\eta}\left(G(\eta) - \tfrac{1}{3}\right)$	$d_t{}^r = \dfrac{9\lambda_s\chi_0}{4M_s}[F(\eta) + H(\eta)]$
Parallel fields, Isotropic material $f(\theta) = e^{\eta\cos\theta}$	$M = M_s L(\eta)$	$\chi_{rp}{}^w = 3\chi_0 \dfrac{dL(\eta)}{d\eta}$	$\chi_{rp}{}^r = 3\chi_0 \dfrac{L(\eta)}{\eta}$ *	$d_p{}^w = \dfrac{3d_0}{\eta}\left[\dfrac{L(\eta)}{\eta} - \dfrac{dL(\eta)}{d\eta}\right]$	$d_p{}^r = \dfrac{3d_0}{\eta}\left[1 - \dfrac{3L(\eta)}{\eta}\right]$
Transverse fields		$\chi_{rt}{}^w = 3\chi_0 \dfrac{L(\eta)}{\eta}$	$\chi_{rt}{}^r = \tfrac{3}{2}\chi_0 \left(1 - \dfrac{L(\eta)}{\eta}\right)$	$d_t{}^w = \dfrac{d_0}{\eta}\left[1 - \dfrac{3L(\eta)}{\eta}\right]$	$d_t{}^r = \dfrac{3d_0}{2}\left[L(\eta) - \dfrac{1}{\eta}\left(1 - \dfrac{3L(\eta)}{\eta}\right)\right]$

Susceptibility matrix

$$\begin{pmatrix} \dfrac{\chi_- + \chi_+}{4}(1+G(\eta)), & \dfrac{\chi_- - \chi_+}{2}F(\eta), & 0 \\[2mm] \dfrac{\chi_- - \chi_+}{2}F(\eta), & \dfrac{\chi_- + \chi_+}{4}(1+G(\eta)), & 0 \\[2mm] 0, & 0, & \dfrac{\chi_- + \chi_+}{2}(1-G(\eta)). \end{pmatrix}$$

$$\begin{pmatrix} \dfrac{3\chi_0}{2}\left(1 - \dfrac{L(\eta)}{\eta}\right), & KL(\eta), & 0 \\[2mm] -KL(\eta), & \dfrac{3\chi_0}{2}\left(1 - \dfrac{L(\eta)}{\eta}\right), & 0 \\[2mm] 0, & 0, & 3\chi_0\dfrac{L(\eta)}{\eta} \end{pmatrix}$$

$$A = \frac{3\chi_0}{M_s}; \quad d_0 = \frac{3\lambda_s\chi_0}{M_s}; \quad L(\eta) = \operatorname{ctnh}\eta - \frac{1}{\eta}; \quad (\chi_- + \chi_+) = 3\chi_0; \quad (\chi_- - \chi_+) = 2K.$$

* GRIMES D. M. Bull. Amer. Phys. Soc. Series II, 1, 25 (1956).

material which has been cooled through its Curie temperature in the absence of any magnetic fields. For saturated material $f(\theta)$ will be unity in the field direction and zero in all other directions. If the magnetization is decreased from saturation, the first effect is the rotation of the magnetic moment vectors of the different crystallites towards "easy" crystallographic directions as determined by the anisotropy fields. The exact result as the field is further decreased is open to considerable variation as well as conjecture. However, demagnetizing fields will exist and, if the crystallites are sufficiently large, domain walls will be nucleated. These effects, as well as effective magnetostrictive fields arising from internal strains, will disorder the moments of the system. For single crystals, HEISENBERG[18] has proposed a model based upon the most probable domain behavior, while AKULOV[19] considered a model of either parallel or anti-parallel moments. Since the present system has the additional factor of random crystallite orientation, it is expected that the results will more nearly approximate the model proposed by HEISEN-BERG.

With the assumption that the domains are all of fixed and equal volume, BROWN[2] has shown that the HEISENBERG model leads to the distribution function

$$f(\theta) = e^{\eta \cos \theta} \qquad (23)$$

where η is the same as discussed in the previous section. In the appendix the same distribution function is derived without the restricting equal volume domain assumption, but with other restricting assumptions. For this section equation 23 will be assumed valid; then deviations from this equation are considered to give rise to the remaining hysteresis in plots of the susceptibilities versus the magnetization.

For calculation, we compute the weighted-average values by first averaging over each crystallite, then averaging the crystallite average over the polycrystal. We first assume effectively infinite first order anisotropy fields, then for rotational effects we assume these fields are still small enough to allow a finite moment rotation. The following cases will be considered: Cubic, $K_1 > 0$; cubic, $K_1 < 0$; cubic, an infinite number of possible directions; hexagonal, $K_1 > 0$; hexagonal, $K_1 < 0$; and all moments either parallel or anti-parallel

with the field direction. The results are shown in integral form in Table 2 along with the low field expansions and the numerical values for fields effectively infinite but still much less than the anisotropy fields.

The reversible functions

Upon comparing the low field expansion terms for the cubic and isotropic materials, they are identical to the $(7-m)$ power of η. Further each type assumes infinite anisotropy fields; the [100] oriented material has six possible directions, the [111] oriented material has eight possible directions, and the isotropic material an infinite number of directions. Since for large M the assumption of large anisotropy fields ceases to be valid, the
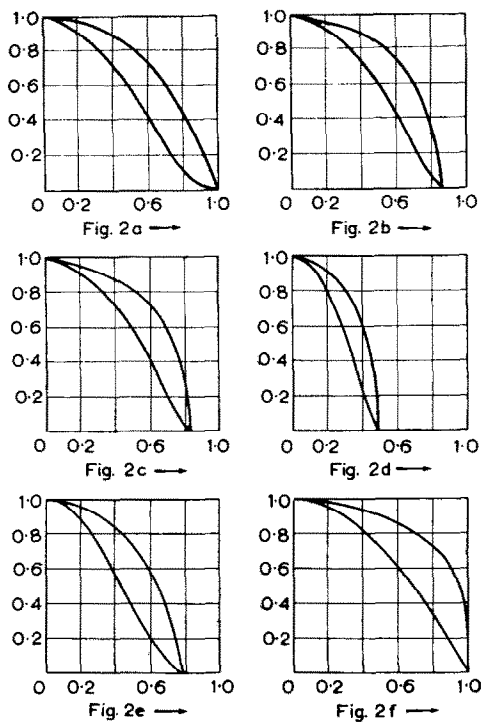

FIG. 2. The variation of the parallel (lower curves) and transverse (upper curves) normalized reversible susceptibilities with normalized magnetization when the susceptibility is due to domain-wall motion. The different figures represent different anisotropy classes. FIG. 2a, isotropic material; FIG. 2b, cubic, $K_1 < 0$; FIG. 2c, cubic $K_1 > 0$; FIG. 2d, hexagonal, $K_1 > 0$; FIG. 2e, hexagonal, $K_1 < 0$, zero basal plane anisotropy; FIG. 2f, all moments parallel or anti-parallel with H.

Table 2. Distribution functions for nonoriented material if $f(\theta) = e^{\eta \cos \theta}$

Anisotropy condition	Isotropic	Cubic $K_1 < 0$	Cubic $K_1 > 0$
$\langle \cos^m \theta \rangle$	$\dfrac{\int d\Omega \cos^m \theta \, e^{\eta \cos \theta}}{\int d\Omega e^{\eta \cos \theta}}$	$\dfrac{\int \dfrac{d\Omega}{4\pi} \displaystyle\sum_p \left(\sum_i \dfrac{1}{\sqrt 3} p l_i \right)^m \exp\left(\dfrac{\eta}{\sqrt 3} \sum_i p l_i \right)}{\displaystyle\sum_p \exp\left(\dfrac{\eta}{\sqrt 3} \sum_i p l_i \right)}$ $p = \pm 1$ l_i = direction cosine of field to crystalline axis.	$\dfrac{\int \dfrac{d\Omega}{4\pi} \displaystyle\sum_i (e^{\eta l_i} + (-1)^m e^{-\eta l_i}) l_i^m}{\displaystyle\sum_i (e^{\eta l_i} + e^{-\eta l_i})}$
small η			
$F(\eta) = \langle \cos \theta \rangle$	$\dfrac{\eta}{3} - \dfrac{\eta^3}{45} + \dfrac{2\eta^5}{945} - \dfrac{\eta^7}{4725} + \cdots$	$\dfrac{\eta}{3} - \dfrac{\eta^3}{45} + \dfrac{2\eta^5}{945} - \dfrac{17\eta^7}{76{,}545} + \cdots$	$\dfrac{\eta}{3} - \dfrac{\eta^3}{45} + \dfrac{2\eta^5}{945} - \dfrac{2\eta^7}{8505} + \cdots$
$G(\eta) = \langle \cos^2 \theta \rangle$	$\dfrac{1}{3} + \dfrac{2\eta^2}{45} - \dfrac{4\eta^4}{945} + \dfrac{2\eta^6}{4725} - \cdots$	$\dfrac{1}{3} + \dfrac{2\eta^2}{45} - \dfrac{4\eta^4}{945} + \dfrac{2\eta^6}{5103} - \cdots$	$\dfrac{1}{3} + \dfrac{2\eta^2}{45} - \dfrac{4\eta^4}{945} + \dfrac{2\eta^6}{5670} + \cdots$
$H(\eta) = \langle \cos^3 \theta \rangle$	$\dfrac{\eta}{5} - \dfrac{\eta^3}{105} + \dfrac{4\eta^5}{4725} - \cdots$	$\dfrac{\eta}{5} - \dfrac{\eta^3}{105} + \dfrac{4\eta^5}{5103} + \cdots$	$\dfrac{\eta}{5} - \dfrac{\eta^3}{105} + \dfrac{4\eta^5}{5670} - \cdots$
$\eta \to \infty$			
$F(\infty)$	1·0000	0·8660	0·8312
$G(\infty)$	1·0000	0·7577	0·7009
$H(\infty)$	1·0000	0·6693	0·5991

Table 2. (continued)

Anisotropy condition	Hexagonal $K_1 > 0$	Hexagonal $K_1 < 0$ Zero basal anisotropy	All moments parallel or antiparallel to H_{ap}
$\langle\cos^m\theta\rangle$	$\displaystyle\int \frac{d\Omega}{4\pi}\left(\frac{(e^{\eta l}+(-1)^m e^{-\eta l})l^m}{e^{\eta l}+e^{-\eta l}}\right)$ l = cosine of angle between C-axis and applied field.	$\displaystyle\frac{\int d\Omega \int_0^{2\pi} d\phi\,(\sin\theta\cos\phi)^m e^{\eta\sin\theta\cos\phi}}{4\pi \int_0^{2\pi} d\phi\, e^{\eta\sin\theta\cos\phi}}$ The integrals over ϕ are expressible in terms of Bessel functions.	$\displaystyle\left(\frac{e^{\eta}+(-1)^m e^{-\eta}}{e^{\eta}+e^{-\eta}}\right)$
small η			
$F(\eta)=$ $\langle\cos\theta\rangle$	$\dfrac{\eta}{3} - \dfrac{\eta^3}{15} + \dfrac{2\eta^5}{105} - \dfrac{17\eta^7}{2835} + \cdots$	$\dfrac{\eta}{3} - \dfrac{\eta^3}{15} + \dfrac{\eta^5}{84} - \dfrac{29\eta^7}{15120} + \cdots$	$\eta - \dfrac{\eta^3}{3} + \dfrac{2\eta^5}{15} - \dfrac{17\eta^7}{315} + \cdots$
$G(\eta)=$ $\langle\cos^2\theta\rangle$	$0\cdot3333$	$1 - \dfrac{\eta^2}{3} + \dfrac{\eta^4}{30} - \dfrac{\eta^6}{210} + \dfrac{\eta^8}{1689} + \cdots$	$1\cdot0000$
$H(\eta)=$ $\langle\cos^3\theta\rangle$	$\dfrac{\eta}{5} - \dfrac{\eta^3}{21} + \dfrac{2\eta^5}{135} - \cdots$	$\dfrac{\eta}{5} - \dfrac{2\eta^3}{105} + \dfrac{\eta^5}{360} - \cdots$	$\eta - \dfrac{\eta^3}{3} + \dfrac{2\eta^5}{15} - \cdots$
$\eta \to \infty$			
$F(\infty)$	$0\cdot5000$	$0\cdot7854$	$1\cdot0000$
$G(\infty)$	$0\cdot3333$	$0\cdot6667$	$1\cdot0000$
$H(\infty)$	$0\cdot2500$	$0\cdot5890$	$1\cdot0000$

assumption of isotropic material should closely approximate the results for cubic material. The reversible functions for isotropic material are listed in Table 1, row 2. The variations of the wall motional and rotational susceptibilities are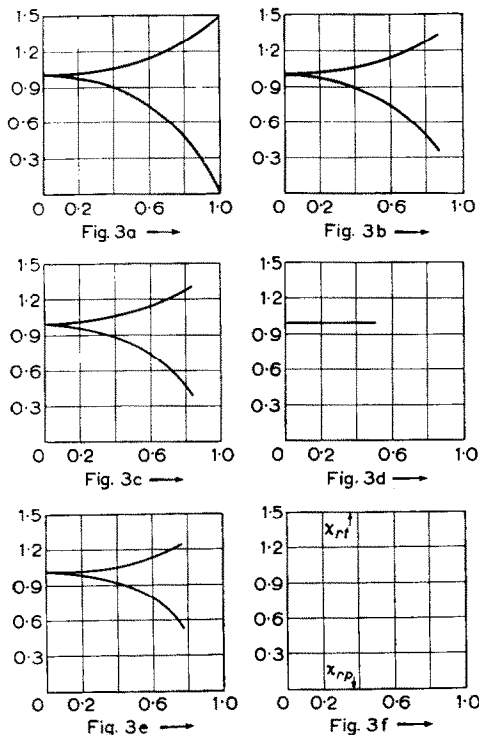 shown in Figs. 2 and 3, respectively, for each of the six cases. Fig. 4 illustrates the wall motional differential magnetostriction, and Fig. 5 shows the rotational differential magnetostrictions for isotropic material.
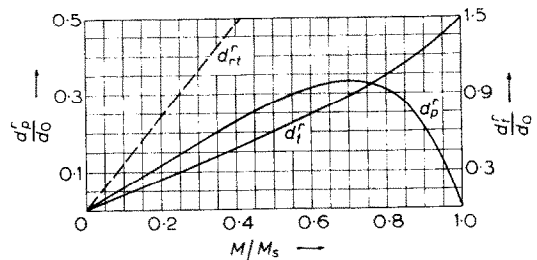
FIG. 5. The variation of the parallel and transverse differential magnetostrictions with magnetization in isotropic material when the susceptibility is due to domain rotation.

The rotational initial susceptibility can be approximated by:

$$\chi_0 = \frac{\mu_0 M_s^2}{3K_1}. \tag{24}$$

Putting equation 24 into the value of d_0 as given in Table 1 yields:

$$d_0 = \frac{\mu_0 \lambda_s M_s}{K_1}. \tag{25}$$

If this form of d_0 is substituted into equation 16a, the coefficient differs from that of the equation given for Λ by BOZORTH and WILLIAMS[8] only by their "small factor" f.

The large values of the rotational reversible properties for crossed fields will not be realized because of the H term in the denominator of the expression for χ_0.

FIG. 3a ⟶   FIG. 3b ⟶
FIG. 3c ⟶   FIG. 3d ⟶
FIG. 3e ⟶   FIG. 3f ⟶

FIG. 3. The variation of the parallel (lower curves) and transverse (upper curves) normalized reversible susceptibility with normalized magnetization when the susceptibility is due to domain rotation. The anisotropy conditions for the figures are the same as in FIG. 2.
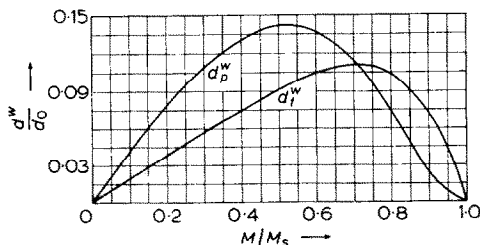
CONCLUSIONS

Theoretical equations are developed for the variation of the reversible susceptibility with magnetization if the susceptibility has its origin in domain rotation or in domain-wall motion. These equations are developed for both parallel and transverse moments. Because of the dependence of transverse field behavior upon susceptibility mechanism, it is concluded that the result provides a new technique for evaluating the relative importance of these two mechanisms (see equations 10 and 20).

FIG. 4. The variation of the parallel and transverse differential magnetostrictions with magnetization in isotropic material when the susceptibility is due to domain-wall motion.

Without a detailed knowledge of domain distribution, if the ratios of the slope of transverse and parallel reversible susceptibilities with magnetization is negative, and if only the wall motional and domain rotational mechanisms exist, then the negative ratio can only result from domain rotation. If a more restrictive model is assumed, then correspondingly more detailed results can be calculated. With a detailed model as well as knowledge of the anisotropy constants, the percentage of the susceptibility for each mechanism can be calculated.

Similar results are derived for the differential magnetostrictions under the condition that $\lambda = \frac{3}{2}\lambda_s[\langle\cos^2\theta\rangle - \frac{1}{3}]$. For an ideal moment distribution, the maximum value would arise for transverse fields and domain rotation. In most cases this result would carry over to useable samples. It is expected that a paper will follow comparing experimental susceptibility results on selected ferrite samples with the theory developed here.

An equation is given for the rotational susceptibility matrix applicable to microwave measurements on nonsaturated material on the condition that only nearest-neighbor domain interactions are present and that the total moments of all nearest-neighbor domains to any particular domain be equal to the macroscopic moments.

APPENDIX

Models for considering the normalized volume of material with its magnetic moment between θ and $\theta + d\theta$ have been proposed by HEISENBERG[18] and by AKULOV.[19] BROWN[2,3,4] has attempted to derive, on a model much like that proposed by HEISENBERG, an expression for $f(\theta)$ in a polycrystal. BROWN's model was one of domains with fixed and equal volume. The reasons for using such a model are, primarily, that it can be handled mathematically and that the important features are the energy densities. These energies will vary but little from model to model, so the results derived on the basis of the fictitious model must be expected to carry over to a more realistic one. This appendix uses mathematics similar to BROWN's and obtains the same $f(\theta)$. The model is more realistic but the technique of handling the exchange energy is questionable.

Consider N atomic magnetic moments per unit volume distributed among an unknown but large number of randomly oriented crystallites. If γ represents a particular direction in the macroscopic system and the number of moments oriented in the γ direction is $N\gamma$, then it follows that

$$N = \sum_{\gamma} N_{\gamma}. \tag{26}$$

The γ direction denotes different directions in different crystallites.

The effect of the anisotropy energy is to keep the moments of the system aligned along "easy" crystallographic directions. It is to be assumed that all moments remain oriented along these easy directions, and that the effect of the anisotropy energy is entirely described by this assumption.

Another energy is the volume magnetostatic energy, i.e., the energy of a magnetic dipole in a magnetic field. This energy can be written:

$$V_s = \sum_{\gamma} N_{\gamma} A H_t \cos\theta \tag{27}$$

where:

A is a constant,
H_t is the total magnetic field,
θ is the angle between the magnetic field and the γ-direction.

The effect of local magnetostatic and magnetostrictive energies throughout the lattice will be to disorder the moments of the system. The number of ways in which the moments of the system can be distributed among N atoms such that N_{γ} have their moments in the γ-direction is:

$$W = \frac{N!}{\prod_{\gamma}(N_{\gamma}!)},$$

which by use of Stirling's approximation becomes

$$\ln W = N \ln N - \sum_{\gamma} N_{\gamma} \ln N_{\gamma}. \tag{28}$$

Equations 26, 27, and 28 are applicable to a paramagnet. For a ferromagnet the exchange energy must also be considered and is given by:

$$V_{\text{ex}} = \sum_{\gamma} \sum_{\text{n.n.}} N_{\gamma} A_{\text{ex}} [1 - (\Delta\theta)^2]$$

where $\Delta\theta$ represents the angle between nearest-neighbor moments, A_{ex} is the exchange energy and $\sum_{\text{n.n.}}$ represents a sum over all nearest-neighbor moments to the moment in question. If at this point domain theory is introduced and intradomain moments are considered to be aligned, then the exchange energy can be written as:

$$V_{\text{ex}} = \sum_{\gamma} s N_{\gamma} A_{\text{ex}} - \frac{\delta}{a}(\Delta\theta)^2 A_{\text{ex}} \cdot S\tau \tag{29}$$

where s is the number of nearest neighbors, δ is the wall

thickness, a is the distance between moments, τ is a constant of the order of unity and S is the domain wall area. The sum is exclusive of the walls. Note that the first term of equation 29 is a volume energy term and the second a surface energy term. It is now necessary to make the restricting assumption that the wall area surrounding material oriented in the γ-direction is proportional to $N_\gamma{}^*$. With this assumption:

$$V_{\text{ex}} = \sum_\gamma N_\gamma(sA_{\text{ex}} - D_1) \tag{30}$$

where D_1 is a constant.

Since the magnetoelastic energies and the internal demagnetizing factors act to "randomize" the distribution of the magnetic moments, it is now to be assumed that they act to put the material in a most probable orientation in the sense of an extremum of equation 28. Equations 26, 27, and 30 must also be stable with respect to variations in N_γ. Thus, using the technique of Lagrange multipliers, and putting the sum of all such variations equal zero gives:

$$\delta N_\gamma(B + CAH_t \cos \theta - \ln N_\gamma) = 0$$

where B and C are constants. From this it follows that

$$N_\gamma = \exp(B + CAH_t \cos \theta)$$

and that the fraction of the moments of the system in the γ-direction is:

$$f(\theta) = \frac{\exp(AH_t \cos \theta)}{\sum_\gamma \exp(AH_t \cos \theta)}. \tag{31}$$

In the foregoing discussion, the result of the magnetoelastic energy and the internal demagnetizing fields is not such a "random" orientation as the corresponding thermal problem where the energy of a particular particle is independent of the energy of its momentary nearest neighbor. In a ferromagnet the disorder is a static disorder and the configuration must move through a series of spatially interdependent configurations to arrive at a specified position. Under these circumstances the presence of potential maxima would weight a changing con-

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* Brown makes an attempt to make this assumption look reasonable in Brown W. F., Jr. *Phys. Rev.* **55**, 570 (1939).

figuration away from an extremum of equation 28 and towards the previous equilibrium position.

If a material is in a most probable configuration in the presence of a magnetic field (H_{ap}), by virtue of being cooled through its Curie temperature in the presence of that field, and the field is changed by ΔH (where ΔH is not small), then the configuration will tend to the limiting value of a most probable configuration for $(H_{ap} + \Delta H)$. However, since potential maxima must first be surpassed (i.e., domain walls must be broken free of localized potential barriers and in some cases nucleated), the configuration will be between that of material in equilibrium in a field H_{ap} and in a field $(H_{ap} + \Delta H)$. If this material represents some most probable configuration for some intermediate field then the effective field can be represented as $(H_{ap} + \alpha \Delta H)$ where $0 \leqslant \alpha \leqslant 1$. The meaning of the "effective history field" D, used to define the parameter η, is $D = (\alpha - 1) \Delta H$. The remaining error, after this type of correction, will depend upon the material and upon the magnitude of the ΔH. Discussions are given in references 4 and 5.

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