# ENGINEERING RESEARCH INSTITUTE UNIVERSITY OF MICHIGAN ANN ARBOR

#### REVERSIBLE PROPERTIES OF FERROMAGNETS

- I. THEORY OF THE EXPECTED VARIATION OF THE REVERSIBLE SUSCEPTIBILITY WITH MAGNETIZATION
- II. COMPARISON OF THEORETICAL AND EXPERIMENTAL SUSCEPTIBILITY CURVES\*
- III. SUMMARY

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#### FOREWARD

Parts I and III under the above title are bound in this jacket to enable a comparison of the theory of the first part and the data of the third. This is to be used during the time interim necessary to write and publish part II, the detailed experimental procedure, results and comparison with theory. The first part has been submitted to the <u>Journal of Physics and Chemistry of Solids</u>, the third part was presented at the Boston conference of Magnetism, October 18, 1956. It is expected that part II will be forthecoming.

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#### REVERSIBLE PROPERTIES OF FERROMAGNETS

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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.

#### REVERSIBLE PROPERTIES OF FERROMAGNETS

THEORY OF THE EXPECTED VARIATION OF THE REVERSIBLE SUSCEPTIBILITY WITH MAGNETIZATION

#### 1. INTRODUCTION

A major problem in the field of technical ferromagnetism is the precise description of the magnetization as a function of the applied magnetic field. Indeed the magnetization can be given only as an infinite-valued function of the applied magnetic field Hap dependent upon the magnetic history of the material. The reversible properties of a ferromagnet, which are discussed in this paper, result from the ferromagnetic character of the material when a vanishingly small disturbance of some kind is applied. From the criterion of reversibility it is implied that any oscillations in the material are carried out about fixed equilibrium positions and that the oscillations are vanishingly small. Thus reversibility indicates repeatability and, in the special case of a zero-frequency applied disturbance, it implies reversibility in the thermodynamic sense of zero energy dissipation.

The reversible quantities considered are the reversible susceptibility and the differential magnetostriction. They are discussed from the standpoint of their variation with the internal magnetization and the angle between the internal magnetization and the applied disturbance. The angular dependence is described by considering the components both parallel and normal to the internal magnetization. These phenomena are examined by first using as general a model as possible, then restricting the model so more detailed results can be obtained.

On the basis of the general model it is shown that previous equations for the variation of the parallel and transverse (normal) susceptibility

with magnetization 1,2,3 are applicable when the susceptibility is due to domain-wall motion. Equations are then developed, for the first time, for the corresponding variation of the reversible susceptibilities with magnetization when that susceptibility has its origin in the rotation of moments of domains as a whole against the anisotropy forces. Indeed, the entire rotational susceptibility matrix is derived as a function of the magnetization. The off-diagonal terms are in agreement with previous results of Rado.4

The variation of the differential magnetostriction with magnetization has previously been carried out only for parallel fields and domain rotation. In this paper, the results for the other three cases (domain rotation, transverse fields; domain-wall motion, parallel fields; domainwall motion, transverse fields) are given and errors as well as differences in points of view with the original work are noted.

Additional applicable references, including reviews, are listed in references 6 through 12.

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#### 2. THE GENERAL CASE

#### 2.1 The Magnetization

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

$$M = M_{s} \frac{\int_{0}^{\pi} f(\theta) \sin\theta \cos\theta d\theta}{\int_{0}^{\pi} f(\theta) \sin\theta d\theta} = M_{s} [\cos\theta]. \tag{1}$$

where the integration is over the unit sphere.  $\lceil \cos\theta \rceil$  is the weighted-average value of the cosine of the angle between the local atomic moments and the applied field. 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 the function  $f(\theta)$  will be left until later. The reversible susceptibility measured parallel to the applied magnetic field is given by

$$\chi_{\rm rp} = \frac{M_{\rm S} \, \delta \left[\cos\theta\right]}{\partial H} ,$$
 (2)

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 sign in Eq. 2 indicates that the external stress and the "history" of the specimen remains 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.

#### 2.2 The Domain Rotation Mechanism

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, 13 can be written as:

$$\frac{\partial \vec{M}}{\partial t} = \gamma(\vec{M} \times \vec{H}) - \frac{\lambda L}{M_s^2} \left[ \vec{M} \times (\vec{M} \times \vec{H}) \right]. \tag{3}$$

It is the usual convention to assume that the spontaneous moment is oriented along the z-axis of the material, that the effective magnetic field in this direction is large compared with the applied reversible field, and that the magnetization in this direction is effectively equal the saturation moment of the material. If all time-varying quantities vary as  $e^{j\omega t}$  the resulting relationships between the magnetization and the applied alternating field can be written as

$$(M) = (X) (H) \tag{4}$$

using matrix notation. To obtain a diagonalized matrix for the susceptibility it is necessary to use the coordinate system  $(x + iy, x - iy, z)^{1/4}$  where i is the imaginary operator representing a spatial rotation from the x to the y axis, not to be confused with the imaginary operator j representing the time phase dependence. The diagonal terms of the susceptibility matrix are then given by:

$$X_{+} = \begin{pmatrix} \frac{M_{x} + iM_{y}}{H_{x} + iH_{y}} \end{pmatrix} = \frac{M_{+}}{H_{+}} = \frac{\omega_{1}}{\omega_{0} + \frac{ij\omega}{1 + i\varepsilon}}$$

$$X_{-} = \begin{pmatrix} \frac{M_{x} - iM_{y}}{H_{x} - iH_{y}} \end{pmatrix} = \frac{M_{-}}{H_{-}} = \frac{\omega_{1}}{\omega_{0} - \frac{ij\omega}{1 - i\varepsilon}}$$

$$(5)$$

$$X_7 = 0$$

4

<sup>13</sup> L. Landau and E. Lifshitz, Physik. Zeits. Sowjetunion 8, 153 (1935)
14 D. Park, Phys. Rev. 98, 438-441 (1955)

where: 
$$\omega_1 = \gamma M_S$$

$$\omega_0 = \gamma H_t$$

 $H_{+}$  is the sum of all static fields.

$$\varepsilon = \frac{\lambda_{L}}{\gamma M_{S}}$$

ω is the applied radial frequency.

To consider the resulting susceptibility of the entire material, let us first average the resultant susceptibility matrix over a polycrystalline sample and later correct for interactions between adjacent domains. The procedure for averaging over a polycrystal involves first transforming from a set of axes based upon individual crystallites to a set based upon the applied biasing field.

Let the subscript d indicate that the matrix in question is described in a coordinate system with the z axis in the direction of the spontaneous moment, and let g indicate a system with the z axis in the direction of the biasing magnetic field. Then:

$$M_d = X_d H_d$$

$$M_g = QX_dH_d = QX_dQ^{-1}H_g = X_gH_g.$$
 (6)

C is the usual Euler matrix expressed in terms of the coordinates (x + iy, x - iy, z).  $^{1/4}$ 

If all angles around the applied field direction are equally likely-sphysically this means that if crystalline orientation exists, it has rotational symmetry about the field direction—then all nondiagonal terms of the matrix  $X_{\sigma}$  average to zero and the susceptibility is given by:

$$X_{g} = \frac{1}{4}$$

$$0 \qquad [(1-\cos\theta)^{2}] X_{+} + [(1-\cos\theta)^{2}] X_{-} 0 \qquad 0$$

$$0 \qquad 0 \qquad 2(1-[\cos^{2}\theta]) (X_{+} + X_{-}) \qquad 0$$

$$0 \qquad 0 \qquad 0 \qquad 0 \qquad 0 \qquad 0$$

The averages are once again the weighted-average values of the functions of  $\cos\theta$  over the macroscopic ferromagnetic sample.

In an attempt to account for the interaction of neighboring domains whose moments are rotating, consider first the effective field at a crystal-lite due to its neighbors. Following Park, <sup>15</sup> let us assume that the neighbors of any domain have the same average value of magnetization as the macroscopic material. For this case, Park showed that the interaction can be described by an equation of the form:

$$h = H = PM \tag{8}$$

where h is the effective magnetic field felt by a domain, H is the applied alternating field, M is the macroscopic alternating magnetization and  $\rho$  is a constant. The external demagnetizing factor of the material for general ellipsoids is also describable by a term proportional to  $\rho$ M. It is here assumed that both the demagnetizing and domain interaction effects are described by the term (-  $\rho$ M) of Eq. 8.

Putting Eq. 8 into Eq. 6,

$$M_g = \chi_{gH_g} = \frac{\chi_g}{1 + \rho \chi_g} H_g . \tag{9}$$

The susceptibilities (X) from Eq. 9 are formally identical with

<sup>15</sup> D. Park, Phys. Rev. <u>97</u>, 60-66 (1955)

the susceptibilities (X) of Eq. 6 when  $\omega_0$  is replaced by  $(\omega_0 + \rho \omega_1)$ . Thus the effect of external demagnetizing factors and nearest-neighbor domain interactions are describable as an increased anisotropy field. This was pointed out by Park<sup>15</sup> to be also true when the effective fields varied throughout the sample.

In terms of the coordinate system (x, y, z) and the  $x_i$ s, Eq. 7 becomes:

$$x_{g} = \frac{1}{4} \begin{bmatrix} (1 + [\cos^{2}\theta])(x_{+} + x_{+}) & 2 [\cos\theta](x_{-} + x_{+}) & 0 \\ -2 [\cos\theta](x_{-} + x_{+}) & (1 + [\cos^{2}\theta])(x_{-} + x_{+}) & 0 \\ 0 & 0 & 2(1 - [\cos^{2}\theta])(x_{-} + x_{+}) \end{bmatrix}$$
(10)

For the special case of a nonoriented polycrystal in the virgin state, it follows that  $[\cos^2\theta] = 1/3$  and that  $[\cos\theta] = 0$ . For this case the matrix of Eq. 10 is proportional to the unit matrix. For saturated material,  $[\cos\theta] = [\cos^2\theta] = 1$ , and the matrix is equal to the susceptibility matrix for a single crystal. As previously noted, the off-diagonal terms are proportional to the level of macroscopic magnetization. Since  $f(\theta)$  is not known, the diagonal terms cannot be calculated unambiguously from a knowledge of the normalized magnetization only.

The reversible susceptibility both parallel with and normal to the biasing magnetic field due to domain rotation can be seen from Eq. 10. Since the biasing field is in the z direction, the term on the diagonal giving the relationship between  $M_{\rm Z}$  and  $H_{\rm Z}$  will be the parallel susceptibility while the remaining diagonal terms are the transverse susceptibility. Thus,

<sup>16</sup> D. Polder, Phil. Mag. 40, 99 (1949)

$$\chi_{rp}^{r} = \frac{1}{2} (1 - [\cos^2 \theta]) (\chi_{+} + \chi_{-})$$
 and  $\chi_{rt}^{r} = \frac{1}{h} (1 + [\cos^2 \theta]) (\chi_{+} + \chi_{-}), (11)$ 

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

$$(x_{+}^{+} x_{-}) = 2 x_{rt}^{r} + x_{rp}^{r} = 3 x_{o}^{r}.$$
 (12)

As the magnetization is varied, if  $(x_{+}+x_{-})$  remains constant and if  $x_{rp}$  is a monotonic decreasing function of the magnetization, then  $x_{rt}$  must be a monotonic increasing function with the ratio of the slopes equal (-2/1).

The assumptions made in deriving Eqs. 11 and 12 are (1) that the susceptibility is describable by the small signal solution to the Landau-Lifshitz equation, (2) that either domain interactions do not exist or they are describable by Eq. 8, and (3) that the sum of biasing field and magnetic anisotropy effects are describable as an effective magnetic field. Assumptions 1 and 3 are related since the torque equation would presumably be valid so long as the two effects were describable as a magnetic field. The statement regarding the relative slopes is dependent upon the applied field being much less than the anisotropy field.

#### 2.3 The Magnetostriction

If a demagnetized sphere of polycrystalline ferromagnetic material is magnetized, the material ceases to be spherical and will become elongated or shortened in the direction of the applied field depending upon the sign of the magnetostrictive coefficient. The coefficient  $\lambda_s$  is defined to be

$$\left[\begin{array}{c} \ell_{m} - \ell_{\mathrm{Sp}} \\ \ell \end{array}\right] = \frac{\Delta \ell}{\ell}$$
 where  $\ell_{m}$  is the length of the material along the

direction of magnetization when the material is saturated and  $\ell_{\mathrm{sp}}$  is the original diameter of the sphere. The angular dependence of the magnetostriction must involve even powers of the cosine of the angle with respect to the applied biasing field. Under the restricting conditions that the magnetostriction is independent of the crystallographic direction of magnetization, i.e.  $\lambda_s = \lambda_{100} = \lambda_{111}$ , then for each crystallite: 17,18,19

$$\lambda_{d} = \frac{3}{2} \lambda_{s} (\cos^{2}\theta - \frac{1}{3}). \tag{13}$$

If the effects of microscopic strain interactions are completely described by the determination of  $f(\theta)$ , then:

$$\lambda_{g} = \frac{3}{2} \lambda_{s} (\left[\cos^{2}\theta\right] - \frac{1}{3}) \tag{14}$$

where  $\lambda_g$  represents the macroscopic magnetostriction.

As was the case with the susceptibilities, it is of interest to calculate the differential magnetostriction both parallel with and normal to the biasing magnetic field for both the wall-motion and domain-rotation mechanism of susceptibility. To calculate the differential magnetostriction it is convenient to set;

$$d_{p}^{W} = \frac{\partial \lambda_{g}}{\partial H} = \frac{3}{2} \lambda_{s} \frac{\partial \left[\cos^{2}\theta\right]}{\partial H}$$
(15)

where the partial indicates that the effective magnetic "history" and the external stress must remain constant.

D. Van Nostrand, New York, 1951, p. 634

M. Kersten, Z. Physik. 76, 505 (1932)
R. M. Bozorth, Ferromagnetism D. Van Nostrand, New York, 1951, p. 6
R. Becker and W. Doring, Ferromagnetismus Edwards Bros., Ann Arbor, 1939, p. 142

The differential magnetostriction can also be written in the form:

$$d = \left[\frac{d\lambda}{dH}\right] = \left[\left(\frac{d\lambda}{d\theta} / \frac{dM}{d\theta}\right) \frac{dM}{dH}\right]. \tag{16}$$

Thus combining Eq. 15 with the terms in parenthesis of Eq. 16:

$$d = \frac{3 \lambda_s}{M_s} \left[ \chi_r \cos \theta \right]$$
 (17)

for each domain. Although Eq. 17 is valid for any direction or susceptibility mechanism, for wall-motion it is more convenient to differentiate after integrating over (d0) as shown in Eq. 15. Nevertheless Eq. 17 shows that the macroscopic coefficient d is proportional to the weighted-average value of the product of cos 0 and the reversible susceptibility. Eq. 17 is in the proper form to be combined with Eqs. 11. The result is that the macroscopic differential magnetostrictions for domain-rotation are:

$$d_{p}^{r} = \frac{9 \lambda_{s} \chi_{o}^{r}}{2 M_{s}} ( [\cos \theta] - [\cos^{3}\theta] )$$
 (18)

$$d_{t}^{r} = \frac{9 \lambda_{s} \chi_{o}^{r}}{\mu_{s}} \left( \left[ \cos \theta \right] + \left[ \cos^{3} \theta \right] \right) . \tag{19}$$

Comparing Eqs. 18 and 19 it is obvious that for constant  $X_0$  the maximum value of d should occur for crossed magnetic fields with  $M = M_s$ . However, since  $H_{ap}$  is not negligible for  $M = M_s$ , this maximum value of  $\frac{9}{2} \frac{\lambda_s X_0}{M_s}$  can not be attained in nonoriented material.

<sup>\*</sup> Eq. 18 is similiar in form to the expression for  $\Lambda$  by Bozorth and Williams, box however 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  $[\cos \theta]^3 = [\cos^3 \theta]$ .

#### 3. RESULTS OF AN EFFECTIVE HISTORY FIELD

An expression has been given for the magnetization as a function of the weighted-average value of  $\cos\theta$ . If the magnetic moment vectors of each local moment are assumed to remain along "easy" crystallographic directions, then the parallel reversible susceptibility as calculated from Eq. 2 and the differential magnetostriction as calculated by Eq. 15 must also be calculated for material whose moments are oriented in the "easy" directions. This criterion is satisfied for domain-wall motion but not for domain rotation. Since for M not greater than about  $\rm M_s/2$  most materials are believed to have their magnetic moments oriented in easy directions, it follows that, at least for M < M<sub>S</sub>/2, Eqs. 2 and 15 will describe effects due to domain-wall motion.

It is now convenient to introduce the dimensionless parameter  $\eta_{\bullet}$  . By definition let

$$\eta = A (H + D). \tag{20}$$

where:

A is a constant with dimension of (H) -1

D is a history dependent term with the dimensions of H. It is necessary to make the restricting assumption that the effect of the magnetic history of the sample is completely described by an effective magnetic field D. Whatever the history effect is, it is possible to describe it in terms of an effective magnetic field; i.e. this implies that  $f(\theta)$  is a unique function of the magnetization M, and that the sum of the component of local moments in the field direction uniquely determines the component of the summed moments oriented in any other direction. A more detailed discussion of this point is given in the appendix.

Since  $f(\theta)$  is assumed to be a unique function of M, it follows that the weighted-average value of all powers of  $\cos\theta$  must be unique functions of M and thus of the parameter  $\eta$ . Let us therefore introduce the notation that:

$$F(\eta) = \left[\cos \theta\right]$$

$$G(\eta) = \left[\cos^2 \theta\right]$$

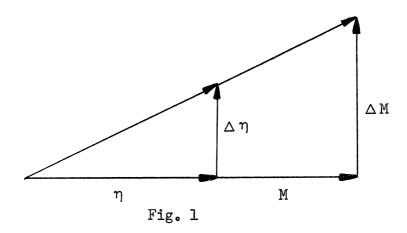
$$H(\eta) = \left[\cos^3 \theta\right]$$
(21)

From Eqs. 1, 2, 20 and 21 it follows that:

$$M = M_s F(\eta)$$
 (22)

$$\chi_{\text{rp}}^{\text{W}} = AM_{\text{S}} \frac{d F(\eta)}{d \eta}$$
 (23)

The partial with respect to H is taken at constant D. Since the effective history changes with each irreversible change, D must be assumed to change with each such change in domain configuration. To calculate the transverse susceptibility due to domain-wall motion refer to Fig. 1.



Relationship for determining the transverse reversible susceptibility for domain-wall motion.

So long as the changes in M are sufficiently slow so that the field and magnetization vectors remain always aligned, by similiar triangles:

$$\chi_{rt}^{W} = A \frac{\Delta M}{\Delta \eta} = A \frac{M}{\eta} = AM_{s} \frac{F(\eta)}{\eta}$$
 (24)

Upon comparing the results of Eqs. 23 and 24, it is seen that:

$$\chi_{rp}^{W} = \chi_{rt}^{W} + \eta \frac{d \chi_{rt}^{W}}{d \eta}$$
 (25)

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

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

$$2 \mu_{0} M_{S} H = \alpha x + \beta \dot{x} + m \dot{x}$$
 (26)

where x represents the spatial coordinate of the domain wall measured from its position in the absence of an applied field and  $\alpha$ ,  $\beta$  and m are constants. The assumption inherent in Eq. 24 is that  $\alpha$  x >>  $\beta$  x + m x. The magnetic Q, for a sinusoidal applied signal, is given by:

$$Q = \left(\frac{\alpha - \omega^2 m}{\omega \beta}\right) \quad \bullet \tag{27}$$

Thus so long as the material remains inductive and the Q is large, Eq. 24 will be valid.

<sup>\*</sup> The argument here is that if the incremental field is applied at right angles to the static field, the change in magnitude of the static field is second order in the increment. Thus Eq. 24 follows.

Returning to the susceptibilities due to domain rotation, if Eq. 21 are combined with Eq. 10 the results are:

$$x_{g} = \frac{1}{4} \begin{bmatrix} 1 + G(\eta) \end{bmatrix} (x_{-} + x_{+}) & 2 F(\eta) (x_{-} - x_{+}) & 0 \\ -2 F(\eta) (x_{-} - x_{+}) & [1 + G(\eta)] (x_{-} + x_{+}) & 0 \\ 0 & 0 & 2[1 - G(\eta)] (x_{-} + x_{+}) & ,$$

and the diagonal components are:

$$X_{rp}^{r} = \frac{1}{2} \left[ 1 - G(\eta) \right] \left( X_{+} X_{+} \right)$$
 (29)

$$X_{rt}^{r} = \frac{1}{h} \left[ 1 + G(\eta) \right] \left( X_{r} + X_{r} \right) . \tag{30}$$

The magnetostriction, from Eqs. 14 and 21, is given by:

$$\lambda = \frac{3}{2} \lambda_{\varepsilon} \left[ \omega(\eta) - \frac{1}{3} \right]$$
 (31)

and the parallel-field differential magnetostriction is given by:

$$d_{p}^{W} = \frac{3}{2} \lambda_{s} \frac{Ad G(\eta)}{d \eta} . \tag{32}$$

By the same argument used to derive Eq. 24, the transverse-field differential magnetostriction is given by:

$$d_{t}^{W} = \frac{3}{2} \lambda_{s} \frac{A}{\eta} \left[ G(\eta) - \frac{1}{3} \right]. \tag{33}$$

For the case of domain rotation, from Eqs. 18 and 19 it follows that:

$$d_{p}^{r} = \frac{9 \lambda_{s} \chi_{o}^{r}}{2 M_{s}} \left[F(\eta) - H(\eta)\right]$$
(34)

$$d_{t}^{r} = \frac{9 \lambda_{s} \chi_{o}^{r}}{4 M_{s}} \left[ F(\eta) + H(\eta) \right]. \tag{35}$$

Upon examining Eqs. 1 and 21 through 35, it follows that the magnetization and the reversible quantities considered are related in a calculable fashion. Indeed each of the variables could be plotted against M and the parameter  $\eta$  eliminated subject to the condition that  $f(\theta)$  be known.

### $\downarrow$ THE DISTRIBUTION FUNCTION $f(\theta)$

#### 4.1 General Comments

The function  $f(\theta)$  is, by definition, proportional to the number of magnetic moments in the system of interest which make an angle between  $\theta$  and  $\theta + d\theta$  with respect to the biasing magnetic field. The magnetic moment of the macroscopic material gives only the net unbalanced moment in the direction of the field and by itself, except when saturated, does not uniquely determine the fraction of the moments in any specified direction. Any attempt to derive an expression for  $f(\theta)$  must begin by considering the mechanisms which cause some of the moments to be oriented nonparallel with the applied magnetic field.

It is assumed that all thermal effects are adequately described by the temperature dependence of such structure insensitive properties as the saturation magnetization, the magnetic anisotropy constants, and the magnetostrictive constants.

In the case of a nonmagnetized polycrystalline magnetic material, domains must be present with moments oriented in all possible directions. For the case of nonoriented polycrystalline material cooled through its Curie temperature, all directions must be equally probable. As the moment of a ferromagnet 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 anisotropy energy considerations. The exact result as the field is decreased further is open to considerable variation as well as conjecture. However, if the crystallites are large enough, domain walls will be nucleated.

In addition to magnetostatic energies, there will be local magnetostrictive energies arising because of the change in shape of the domains and the crystallites which undergo non-180° changes in direction of orientation of local moments.

As a result of these two energy types, as the total moment of a polycrystalline material is decreased from saturation the individual moments will tend to become "randomly" oriented in the sense of a most probable distribution. Deviations from the most probable configurations would depend upon magnetic history.

For increasing fields the picture is, in a sense, a model between that of a most probable distribution proposed for a single crystal by Heisenberg 20 and that of a parallel-antiparallel distribution as proposed by Akulov. 21 The behavior is expected to more closely parallel that proposed by Heisenberg due to the randomizing effect of the nonoriented polycrystalline system. This work has been extended in a series of three papers by Brown. 6,7,2

#### 4.2 The Idealized Case

The particular form of the distribution function  $f(\theta)$  that will be used in this paper is the Boltzman function.

$$f(\theta) = e^{\eta \cos \theta} . \tag{36}$$

Corrections will later be applied to modify Eq. 36 to more closely agree with

<sup>20</sup> W. Heisenberg, Z. Physik. 69, 287-297 (1931)

N. Akulov, Z. Physik. 69, 78-99 (1931)

\*An attempt is made in Appendix I to make it look reasonable that the Boltzman distribution would be applicable, in many cases, to nonoriented polycrystalline material.

the Akulov model, however Eq. 36 will still be used to give the fraction in each of the two possible directions.

The model assumed is a randomally oriented polycrystal which possesses effectively infinite anisotropy fields. Thus for a given crystallite only certain directions are possible, i.e. if only the first order anisotropy constant  $K_1$  is considered, only the [100] directions can be occupied for material with  $K_1 > 0$ . If the direction cosines of the applied magnetic field with respect to the crystalline axis of a crystallite are  $(\ell_1, \ell_2, \ell_3)$  then the cosine of the angle between field and moment can be given by  $(\pm \ell_1, \pm \ell_2, \text{ or } \pm \ell_3)$  for a total of six possible directions. The most probable value of  $\cos^m\theta$  for each crystallite is:

and the resultant average over the polycrystal is given by:

$$\begin{bmatrix} \cos^{m} \Theta \end{bmatrix} = \int \frac{d \Omega}{\frac{1}{2}} \frac{\sum_{i=1}^{3} \left( \ell_{i}^{m} e^{\eta \ell_{i}} + (-\ell_{i})^{m} e^{-\eta \ell_{i}} \right)}{3} . \tag{38}$$

Eqs. 37 and 38 were formulated by Brown. He also evaluated the integral of Eq. 38 for m = 1 and m = 2 in series form. The results, Eqs.

36-39 of reference 7, for low values of  $\eta$  and the value for m = 3 are:

$$F^{6}(\eta) = \left[\cos \theta\right] = \frac{\eta}{3} - \frac{\eta^{3}}{45} + \frac{2\eta^{5}}{945} - \frac{2\eta^{7}}{8505} + \dots$$

$$G^{6}(\eta) = \left[\cos^{2}\theta\right] = \frac{1}{3} + \frac{2\eta^{2}}{45} - \frac{14\eta^{14}}{945} + \frac{2\eta^{6}}{5670} - \dots$$

$$H^{6}(\eta) = \left[\cos^{3}\theta\right] = \frac{\eta}{5} - \frac{\eta^{3}}{105} + \frac{14\eta^{5}}{5670} - \dots$$
(39)

The superscript "6" indicates that there are six "easy" directions. The maximum values of the three functions, which occur for  $\eta = \infty$ , are  $\left[\cos\theta\right] = 0.8312$ ,  $\left[\cos^2\theta\right] = 0.7009$  and  $\left[\cos^3\theta\right] = 0.5991$ .

For material with  $K_1 < 0$ , the [lll] directions will be occupied by the atomic moments. If the applied field has the direction cosines  $\ell_1$ ,  $\ell_2$ ,  $\ell_3$  with respect to one of the crystallographic axes, then the cosine of the angle between field and moment will be given by  $(1/3)^{1/2}$   $(P_1\ell_1, P_2\ell_2, P_3\ell_3)$  where the P's are  $^{\frac{1}{2}}$ 1. There is a total of eight possible combinations of  $^{\frac{1}{2}}$ 1 and thus eight "easy" crystallographic directions. As before it is to be assumed that the probability that a particular domain in a given crystallite will have its moment oriented with a given cos  $\theta$  is given by Eq. 36. The macroscopic averages result from integrating the expected crystallite results over a unit sphere.  $\cos^m\theta$  is given by:

$$\begin{bmatrix} \cos^{m}\theta \end{bmatrix} = \int \frac{d}{d} \frac{\sum_{i} \left(\sum_{i} \frac{1}{\sqrt{3}} P_{i} \ell_{i}\right)^{m} \exp\left(\frac{\eta}{\sqrt{3}} \sum_{i} P_{i} \ell_{i}\right)}{P_{i} \exp\left(\frac{\eta}{\sqrt{3}} \sum_{i} P_{i} \ell_{i}\right)}$$
(40)

The integral of Eq. 40 has been evaluated as an infinite series by Brown for

m=1 and m=2, Eqs. 17, 18, 20 and 22 of reference 7. For small values of  $\eta$ , and the similar series for m=3, are:

$$F^{8}(\eta) = \left[\cos \theta\right] = \frac{\eta}{3} - \frac{\eta^{3}}{45} + \frac{2\eta^{5}}{945} - \frac{17\eta^{7}}{76545} + \cdots \right]$$

$$G^{8}(\eta) = \left[\cos^{2}\theta\right] = \frac{1}{3} + \frac{2\eta^{2}}{45} - \frac{4\eta^{4}}{945} + \frac{2\eta^{6}}{5103} - \cdots \right]$$

$$H^{8}(\eta) = \left[\cos^{3}\theta\right] = \frac{\eta}{5} - \frac{\eta^{3}}{105} + \frac{4\eta^{5}}{5103} - \cdots \right]$$

$$(41)$$

The superscript "8" indicates eight "easy" crystallographic directions. The maximum value of the three functions, which occurs for  $\eta = \infty$ , are  $\left[\cos\theta\right] = 0.8660$ ,  $\left[\cos^2\theta\right] = 0.7577$  and  $\left[\cos^3\theta\right] = 0.6693$ .

If the concept of the number of directions allowable in the single crystal is extended from six and eight to an infinite number of possible directions then the average for the polycrystal must equal the most probable value of the crystallite. Thus:

$$\begin{bmatrix}
\cos^{m}\theta
\end{bmatrix} = \frac{\int_{d} \Omega \cos^{m}\theta e^{\eta\cos\theta}}{\int_{d} \Omega e^{\eta\cos\theta}} \tag{42}$$

This result of Eq. 42 can be written in closed form. Introducing I,  $(\eta)$  = ctnh  $\eta$  -  $1/\eta$ , the Langevin function, then the functions together with the low field expansions are given by:

$$F^{\infty}(\eta) = \left[\cos \theta\right] = L(\eta) \cong \frac{\eta}{3} - \frac{\eta^{3}}{45} + \frac{2\eta^{5}}{945} - \frac{\eta^{7}}{4725} + \cdots$$

$$G^{\infty}(\eta) = \left[\cos^{2}\theta\right] = 1 - \frac{2L(\eta)}{\eta} \cong \frac{1}{3} + \frac{2\eta^{2}}{45} - \frac{4\eta^{14}}{945} - \frac{2\eta^{6}}{4725} - \cdots + \frac{2\eta^{6}}{4725} - \cdots + \frac{2\eta^{6}}{\eta^{6}} = L(\eta) \left(1 + \frac{6}{\eta^{2}}\right) - \frac{2}{\eta} \cong \frac{\eta}{5} - \frac{\eta^{3}}{105} + \frac{4\eta^{5}}{4725} - \cdots + \frac{2\eta^{6}}{\eta^{6}} = \frac{1}{\eta^{6}} + \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} + \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} + \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} + \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} + \frac{1}{\eta^{6}} = \frac{1}{\eta^{6}} + \frac{1}{\eta^{6}} = \frac{1}{\eta$$

The maximum value of each of the functions, which occurs for  $\eta = \infty$ , is unity.

For the case where all moments are oriented either parallel or antiparallel with the magnetic field, Eq. 36 yields directly:

$$F^{\parallel}$$
  $(\eta)$  = tanh  $\eta$   $G^{\parallel}$   $(\eta)$  = 1  $(44)$   $H^{\parallel}$   $(\eta)$  = tanh  $\eta$ 

Figure 2 shows plots of  $G(\eta)$  and  $H(\eta)$  as functions of  $F(\eta)$  for each of the anisotropy conditions.

The effect of a non-ideal distribution has been interpreted previously  $^3$  as resulting from portions of the material having their moments oriented in metastable positions. These conditions either increase or decrease the volume oriented parallel and antiparallel with the applied field for a specified magnetization. (See the Appendix.) The expected hysteresis when the susceptibility arises due to domain-wall motion was also discussed. Comparing Eqs. 11 and 44, it is apparent that when the susceptibility is due to domain rotation an increase in the parallel-antiparallel components would cause the transverse susceptibility to be larger than the parallel susceptibility. This effect would be especially noticeable when M = 0.

If the magnetization is decreased from saturation to some fixed value, consider that a fraction v of the moments are parallel with the direction of the moment where v is not consistent with Eqs. 20 and 36. If

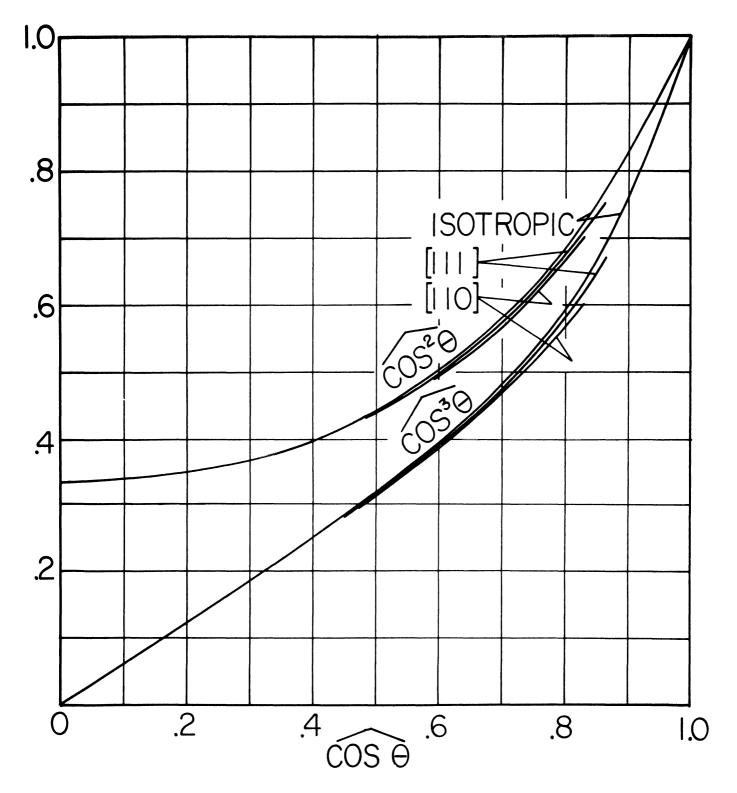


Fig. 2. The expected variation of the weighted-averaged value of the second and third powers of  $\cos\theta$  as a function of  $\cos\theta$  when the magnetic moment can be oriented in any direction in the single crystal (isotropic), in one of eight possible directions [111], and in one of six possible directions [100].

the temperature were altered in such a manner as to decrease  $K_1$  and  $\lambda_s$ , then v would presumably approach consistency with Eq. 36, placing the material in a more probable configuration. The result is an initial thermal hysteresis of susceptibility.

#### 5. THE REVERSIBLE FUNCTIONS

Upon comparing Eqs. 39, 41 and 43, the low field values of the functions are identical through the (7-m) power of  $\eta$ . Further, each function assumes infinite anisotropy fields. For large M the assumptions of six or eight possible directions cease to be valid and the equations become invalid. Thus Eqs. 43 will be used for plotting the functions.

Combining Eq. 43 with the equations for the reversible properties yields, from Eq. 1:

$$M = M_S L (\eta)$$
 (45)

From Eqs. 23 and 24, the susceptibilities due to domain-wall motion become:

$$\chi_{rp}^{W} = 3 \chi_{o} \frac{d L(\eta)}{d \eta}; \qquad \chi_{rt}^{W} = 3 \chi_{o} \frac{L(\eta)}{\eta}.$$
 (46)

where

$$x_{o} = \frac{AM_{s}}{3} \tag{47}$$

From Eq. 28, using 3  $X_0 = (X_+ + X_-)$  and 2  $K = (X_- - X_+)$ , the susceptibility matrix becomes:

$$\mathbf{x}_{g} = \begin{pmatrix} \frac{3}{2} \left[ 1 - \frac{\mathbf{L}(\eta)}{\eta} \right] \mathbf{x}_{o} & \mathbf{L}(\eta) \kappa & 0 \\ -\mathbf{L}(\eta) \kappa & \frac{3}{2} \left[ 1 - \frac{\mathbf{L}(\eta)}{\eta} \right] \mathbf{x}_{o} & 0 \\ 0 & 0 & 3 \frac{\mathbf{L}(\eta)}{\eta} & \mathbf{x}_{o} \end{pmatrix}. \tag{48}$$

For saturated material L  $(\eta)/\eta = 0$ , so Eq. 48 reduces to the more usual form. From Eq. 48 the susceptibilities due to domain rotation are:<sup>22</sup>

$$x_{rp}^{r} = 3 \times_{o} \frac{L(\eta)}{\eta} ; \qquad x_{rt}^{r} = \frac{3 \times_{o}}{2} \left[1 - \frac{L(\eta)}{\eta}\right] .$$
 (49)

As discussed in conjunction with Eqs. 11,  $\chi_0$  is inversely proportional to the total effective field at each crystallite. This field can be considered constant only if the applied magnetic field is much less than the anisotropy field.

The magnetostriction is considered to be given by Eq. 14. Thus, combining with Eq. 43:

$$\lambda_{g} = \lambda_{s} \left[ 1 - \frac{3L(\eta)}{\eta} \right] \tag{50}$$

The differential magnetostrictions, when the susceptibility is due to domain-wall motion, follow from Eqs. 32, 33 and 47:

$$d_{p}^{W} = d_{o} \cdot \frac{3}{\eta} \left[ \frac{L(\eta)}{\eta} - \frac{dL(\eta)}{d\eta} \right]$$
 (51)

$$d_{t}^{W} = d_{o} \cdot \frac{1}{\eta} \left[ 1 - \frac{3L(\eta)}{\eta} \right]$$
 (52)

where:

$$d_o = \frac{3\lambda_s \chi_o}{M_s} . \tag{53}$$

From Eqs. 34 and 35, the differential magnetostrictions, when the susceptibility is due to domain rotation, are given by:

<sup>22</sup> D. M. Grimes, Bull. Am. Phys. Soc. 1, 25 (1956)

$$d_p^r = d_o \cdot \frac{3}{\eta} \left[ 1 - \frac{3L(\eta)}{\eta} \right]$$
 (54)

$$d_p^t = d_o \cdot \frac{3}{2} \left[ L(\eta) - \frac{1}{\eta} \left( 1 - \frac{3L(\eta)}{\eta} \right) \right] . \tag{55}$$

For the case of domain rotation, the initial susceptibility can be approximated by:

$$x_{o} \cong \frac{\mu_{o}^{M_{s}^{2}}}{3K_{1}} . \tag{56}$$

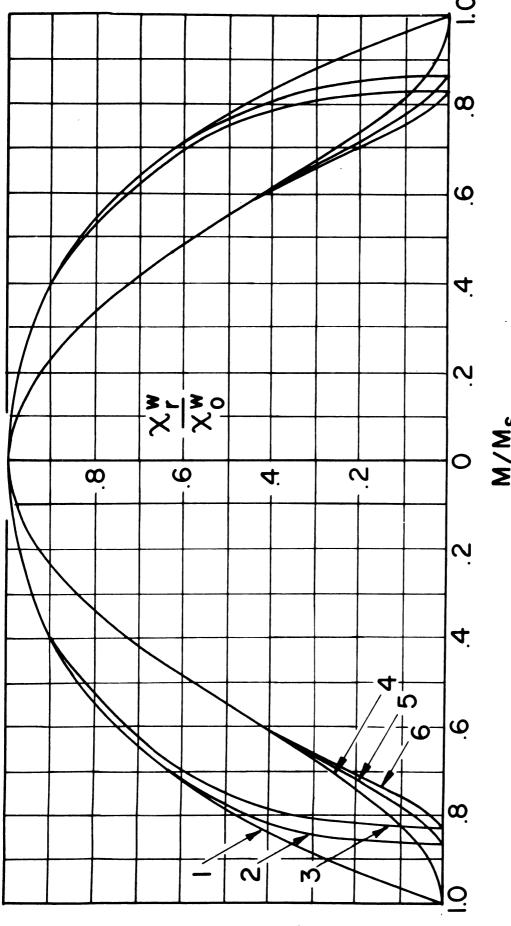
Putting Eq. 56 into Eq. 53 yields:

$$d_{o} = \frac{\mu_{o} \lambda_{s}^{M} s}{K_{7}} \quad \bullet \tag{57}$$

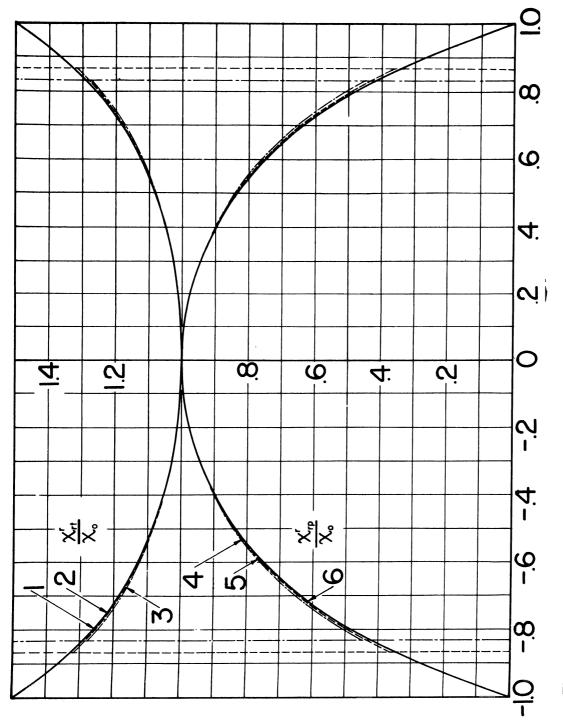
Eq. 54, with  $d_o$  defined according to Eq. 57, has a coefficient similiar to that obtained by Bozorth and Williams. 5

Figs. 3 and 4 show the expect variation of the susceptibilities with magnetization according to Eqs. 45, 46 and 49. Figs. 5 and 6 show the expected behavior of the differential magnetostriction with magnetization.

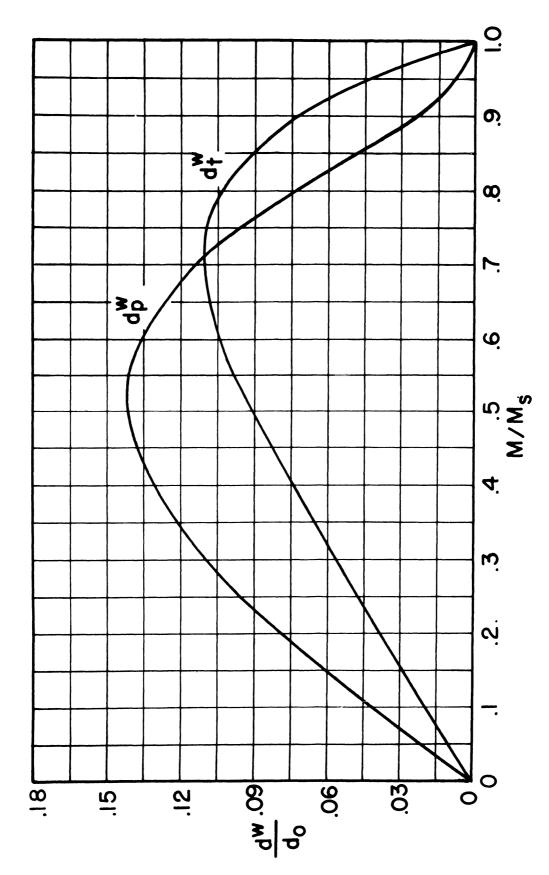
For the crossed-field domain-rotation effects the large values shown at M =  $M_S$  will not be realized because of the "H" term in the denominator of the expression for  $\chi_{0}$ . However, it is to be expected that the magneto-mechanical coupling will usually be larger for this case than for parallel fields.



with magnetization. Curves 1, 2, and 3 are for transverse fields, curves 4, 5, and 6 are for parallel fields. Curves 1 and 4 are for isotropic material, curves 2 and 5 are for [111] oriented material, and curves 2, 3, and 6 are for [100] oriented material. Fig. 5. Theoretical curves for the variation of the domain-wall motion reversible susceptibility



fields. Curves 1, 2, and 3 are for transverse fields, curves  $\mu$ , 5, and 6 are for parallel fields. Curves 1 and 6 are for isotropic material, curves 2 and 5 are for [lll] oriented material and curves 3 and  $\mu$  are for [100] oriented material. Fig.  $^{\!\! \mu}$ . Theoretical curves for the variation of the domain rotation reversible susceptibility with magnetization for values of biasing magnetic field small compared with the anisotropy



ization when the susceptibility has its origin in domain-wall motion.  $d_p^W$  is for parallel fields,  $d_t^W$  is for parallel fields, Fig 5. Theoretical curves for the variation of the differential magnetostriction with magnet-

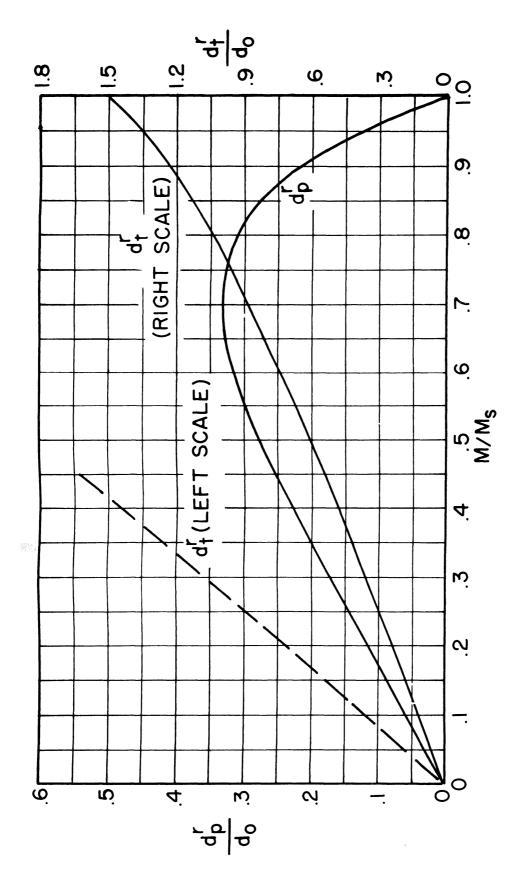


Fig. 6. Theoretical curves for the variation of the differential magnetostriction with magnetization when the susceptibility has its origin in domain rotation.  $\mathbf{d}_p^{r}$  is for parallel fields, i.  $\mathbf{r}$ . ization when the susceptibility has its origin in domain rotation.  ${\rm d}_{\tau}^{\rm r}$  is for transverse fields.

### 6. 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 both for parallel and for transverse moments. Because of the dependence of transverse field behavior upon susceptibility mechanism, it is concluded that the result gives rise to a new technique for evaluating the relative importance of these two mechanisms.

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 from each mechanism can be calculated.

Similiar results are derived for the differential magnetostrictions. Under ideal conditions, 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.

#### APPENDIX

Models for considering the normalized volume of material with magnetic moment between  $\theta$  and  $\theta$  +  $d\theta$ ,  $f(\theta)d\theta$ , have been proposed by Heisenberg<sup>20</sup> and by Akulov. Brown<sup>6</sup>,7,2 has attempted to utilize some of the techniques of statistical mechanics to derive, on a model much like that proposed by Hiesenberg, an expression for  $f(\theta)$ . Brown's model consisted of domains of 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 similiar to Brown and obtains the same  $f(\theta)$  as he. 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 randomally oriented crystallites. If  $\gamma$  represents a particular direction in the macroscopic system and the number of moments oriented in the  $\gamma$  direction is  $N_{\gamma\gamma}$ , then it follows that

$$N = \sum_{\Upsilon} N_{\Upsilon} . \tag{58}$$

The  $\gamma$ -direction denotes different directions in different crystallites.

The energy of the system due to exchange interactions can be expressed as

$$V_{e} = \sum_{\gamma} \sum_{n_{\bullet}n_{\bullet}} N_{\gamma} A_{e} \left[ 1 - (\Delta \theta)^{2} \right]$$
 (59)

where ( $\Delta$ 0) represents the angle between nearest-neighbor moments,  $A_e$  is the exchange interaction energy and  $\sum_{n \in \mathbb{N}} A_n$  represents a sum over all nearest-neighbor moments to the moment in question. If at this point domain theory is introduced by stating that inside a domain all moments will be aligned, then the first term of Eq. 59 represents a volume energy, while the second term is non-zero only in the region of a domain wall and therefore represents a surface energy.

The effect of the anisotropy energy is to keep the moments of the system aligned along "easy" crystallographic directions. Let us assume 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:

$$V_{s} = \sum_{\gamma} N_{\gamma} AH_{t} cos\theta$$
 (60)

where:

A is a constant,

 $H_{t}$  is the total magnetic field,

 $\Theta$  is the angle between the magnetic field and the  $\gamma$ -direction. The effect of local magnetostatic and magnetostrictive energies throughout the lattice, as discussed in the text, 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  $N_{\gamma}$  have their moments in the  $\gamma$ -directions is:

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

By use of Stirling's approximation this becomes,

$$enW = NenN - \sum_{\gamma} N_{\gamma} enN_{\gamma}. \tag{61}$$

Since the energies just discussed are assumed to disorder the distribution of the moments, let us now assume that they act to put the moments in their most probable configuration, i.e. to require Eq. 61 to be stable with respect to variations in  $N_{\gamma}$ . Likewise, Eqs. 58, 60 and the volume energy term of Eq. 59 must be stable with respect to variations in  $N_{\gamma}$ . Using the techniques of Lagrange multipliers, and putting the sum of all such variations equal zero gives;

$$\delta N_{\gamma} \left( B + CAH_{t} \cos\theta + DA_{e} - \ell nN_{\gamma} \right) = 0$$
 (62)

from which it follows that,

$$N_{\star} = \exp (E + CAH_{t} \cos \theta)$$

and the fraction of the moments of the system of interest with their moments oriented in the  $\gamma$ -direction is given by

$$n_{\gamma} = \frac{\exp (AH_{t}\cos\theta)}{\sum \exp (AH_{t}\cos\theta)} = f(\theta).$$
 (63)

There are several points that should be emphasized regarding the foregoing discussion. First of all the disordering brought about by the forces considered is not so "random" in its effects as is thermal disorder. The reason is basically that in a thermal disorder problem the energy of a particular particle is independent of the energy of its momentary nearestneighbor. For the case of a ferromagnet the energy of one moment is very

much a function of the energy of its neighbor. Since the disorder is a static disorder, the configuration will have to move through a series of spatially interdependent configurations as the field is changed to arrive at a specified position. Under these circumstances the presence of potential maxima would weight a changing configuration towards the equilibrium position it just left.

An example might be the presence of single-domain size grains in an otherwise large grained media. These grains would not form domain walls, so would remain oriented towards the previous direction of saturation until a large field is produced in the opposite direction. While oriented in the previous direction they would contribute nothing to a wall-motional susceptibility but would result in a magnetization. For the case of domain rotation, this type of hysteresis acts through weighting the parallel and antiparallel components differently from the derived distribution.

In the foregoing discussion, it was assumed that the surface energy term of Eq. 59 remained constant. This term, however, is a function of the total wall area of the material. Around nucleating centers, the moments remain in metastable conditions until they possess sufficient energy to nucleate domain walls and thereby increase the surface exchange energy term. Thus the change in configurations with field lags the idealized model.

Consider material in a most probable configuration in the presence of a magnetic field  $H_0$  by virtue of being cooled through its Curie temperature in the presence of that field. When the field is changed by  $\Delta$  H where  $\Delta$ H is not small, the configuration will tend to the limiting value of a most probable configuration for  $(H_0 + \Delta H)$ . However, since potential maximum must first be surpassed, (i.e. domain walls must be nucleated and broken

free of localized potential barriers) the configuration will be between that of material in equilibrium in a field  $H_0$  and in a field  $(H_0 + \Delta H)$ . If this material represents some most probable configuration for a field value between  $H_0$  and  $H_0 + \Delta H$ , then an effective field will be considered as  $(H_0 + \alpha \Delta H)$  where  $0 \le \alpha \le 1$ . This is the meaning of the "effective history field" described by D of Eq. 20, where  $D = (\alpha-1) \Delta H$ .

In review, for a material which does not show hysteresis Eq. 36 must be very nearly correct. In the presence of potential maxima which give rise to hysteresis, that hysteresis can be partially described by requiring the configuration to be the most probable one for some hypothetical field. This field can then be eliminated between the reversible properties measured and the magnetization level.

The remaining error, after this type of correction, will depend upon that material and upon the magnitude of the  $\Delta$  H. Discussions are given in references 3 and 8.

# REVERSIBLE PROPERTIES OF FERROMAGNETS (SUMMARY)

The magnetic moment of a piece of ferromagnetic material is equal to the weighted-average value of the cosine of the angle between the direction of magnetization of the sample and that of each atomic magnetic moment. If it be assumed that the atomic moments, even though grouped into domains. remain oriented according to a Boltzmann distribution, 1,2,3 quantitative relationships between the magnetization and the reversible properties of ferromagnets can be given. Let  $\eta$  be a dimensionless parameter proportional to the applied magnetic field H plus a history dependent constant. Under these circumstances,

$$\frac{M}{M_{S}} = \left[\cos\theta\right] = \coth \eta - \frac{1}{\eta} = L(\eta) \tag{1}$$

The reversible susceptibility parallel to the biasing field is then given bу

$$\chi_{rp}^{W} = M_s \frac{\partial \left[\cos \theta\right]}{\partial H} = 3 \chi_o^{W} \frac{dL(\eta)}{d\eta}$$
 (2)

The parametric relationship involving the η was given as early as 1911 by Gans.4

The derivation of the Boltzmann distribution was put on a more

W. F. Brown, Jr. Phys. Rev., 52, 325 (1937)
D. M. Grimes, Thesis, University of Michigan, 1956; Bull. Am. Phys. 2 Soc. 1, 25 (1956)

D. M. Grimes, Preceeding Paper

R. Gans, Phys. Z., 12, 1053 (1911)

quantitative bases by Brown 1,5 in the late 30's. Although reasonable agreement with experiment was noted in many cases, as was pointed out by Tebble and Corner 6 in 1950, the susceptibility is not a single valued function of the magnetization particularly when going from one hysteresis loop to another. In 1954 Grimes and Martin extended the work of Brown to obtain the expression for low frequencies that:

$$\chi_{\text{rt}}^{W} = \frac{M_{\text{s}} \left[\cos \theta\right]}{(H + D)} = 3 \chi_{\theta}^{W} \frac{L(\eta)}{\eta}$$
 (3)

where D is a history dependent term with the dimensions of H. Early this year<sup>2,3</sup> it was shown that Equations 2 and 3 would be approximated for the case where the reversible susceptibility has its origin in domain-wall motion. It was also shown that for the case where the susceptibility is due to the rotation of all the moments of a domain that the susceptibilities were given by:

$$\chi_{rp}^{r} = \frac{3}{2} \chi_{o}^{r} \left(1 - \left[\cos^{2}\theta\right]\right) = 3 \chi_{o}^{r} \frac{L(\eta)}{\eta}$$
 (4)

and for low frequencies or infinite material:

$$\chi_{rt}^{r} = \frac{3}{4} \chi_{o}^{r} (1 + [\cos^{2}\theta]) = \frac{3}{2} \chi_{o}^{r} (1 - \frac{L(\eta)}{\eta})$$
 (5)

The exact form of the parametric equations depends upon the material remaining always oriented in "easy" crystallographic directions, thus they can be

W. F. Brown, Jr., Phys. Rev., <u>54</u>, 279 (1938)
R. S. Tebble and W. D. Corner Proc. Phys. Soc., <u>63B</u>, 1005 (1950)
D. M. Grimes and D. W. Martin Phys. Rev., <u>96</u>, 889 (1954)

considered to have quantitative significance only for M <  $\sim$  0.5 M<sub>s</sub>.

In a forthcoming paper<sup>3</sup> it is shown that the variation of the susceptibility matrix with magnetization when it results from domain rotation is given by:

$$X = \begin{bmatrix} \frac{3 \cdot X_{o}}{4} (1 + \left[\cos^{2}\theta\right]), & \kappa \left[\cos\theta\right], & 0 \\ -\kappa \left[\cos\theta\right], & \frac{3}{4} X_{o} (1 + \left[\cos^{2}\theta\right]), & 0 \\ 0, & 0, & \frac{3}{2} X_{o} (1 + \left[\cos^{2}\theta\right]) \end{bmatrix} = \begin{bmatrix} \frac{3 \cdot X_{o}}{2} (1 - \frac{L(\eta)}{\eta}), & \kappa L(\eta), & 0 \\ -\kappa L(\eta), & \frac{3 \cdot X_{o}}{2} (1 - \frac{L(\eta)}{\eta}), & 0 \end{bmatrix}$$

$$0, & 0, & \frac{3}{2} X_{o} (1 + \left[\cos^{2}\theta\right]) \end{bmatrix} = \begin{bmatrix} \frac{3 \cdot X_{o}}{2} (1 - \frac{L(\eta)}{\eta}), & \kappa L(\eta), & 0 \\ 0, & 0, & \frac{3}{2} X_{o} (1 + \left[\cos^{2}\theta\right]) \end{bmatrix}$$

In the usual form for which the matrix is written for microwave application the susceptibility term  $x_{11} = x_{22} = \frac{3}{2} x_0$ . The off-diagonal terms are proportional to M, a fact previously pointed out by Rado.<sup>8</sup>

In the same paper,<sup>3</sup> the differential magnetostriction "d" for each of the four cases where Eqs. 2, 3, 4 and 5 are applicable is described. The results are based upon the equation:

$$\lambda = \frac{3}{2} \lambda_{s} \left( \left[ \cos^{2} \theta \right] - \frac{1}{3} \right) \tag{7}$$

where  $\lambda_s = \lambda_{100} = \lambda_{111}$  is the saturation magnetostriction and  $\theta$  is the angle between the applied magnetic field and the magnetization. The results are that "d" for the case of domain-wall motion and parallel fields is given by:

$$d_{p}^{W} = \frac{3}{2} \lambda_{s} \frac{\partial \left[\cos^{2}\theta\right]}{\partial H} = \frac{3d_{o}^{W}}{\eta} \left(\frac{L(\eta)}{\eta} - \frac{dL(\eta)}{d\eta}\right)$$
(8)

8 G. T. Rado, Phys. Rev. 89, 529 (1953)

for domain-wall motion and transverse fields by:

$$d_{t}^{W} = \frac{3 \lambda_{s}}{2A(H+D)} \left( \left[ \cos^{2}\theta \right] - \frac{1}{3} \right) = \frac{d_{o}^{W}}{\eta} \left( 1 - \frac{3L(\eta)}{\eta} \right) \tag{9}$$

for domain rotation and parallel fields by:

$$d_{p}^{r} = \frac{9 \lambda_{s} \chi_{o}^{r}}{2M_{s}} \left( \cos \theta - \cos^{3}\theta \right) = \frac{3d_{o}^{r}}{\eta} \left( 1 - \frac{3L(\eta)}{\eta} \right)$$
 (10)

and for domain rotation and transverse fields by:

$$d_{t}^{r} = \frac{9 \lambda_{s} \chi_{o}^{r}}{4M_{s}} \left( \left[ \cos \theta \right] + \left[ \cos^{3}\theta \right] \right) = \frac{3d_{o}^{r}}{2} \left( L(\eta) \left( 1 + \frac{3}{\eta^{2}} \right) - \frac{1}{\eta} \right)$$
 (11)

where:

$$d_o = \frac{3 \lambda_s \chi_o}{M_s} . \tag{12}$$

Independent of the exact form of the distribution, so long as the effective magnetic field can be considered as the sum of the applied field and a history dependent field, the wall-motion susceptibilities are related by:

$$x_{rp}^{W} = x_{rt}^{W} + \frac{1}{\eta} \frac{dx_{rt}^{W}}{d\eta}$$
 (13)

Thus if  $X_{rp}^{W}$  is a monotonic decreasing function of M, then  $X_{rt}^{W}$  must also be monotonic decreasing if the two susceptibilities are equal when M = 0.

Dependent only upon the applied magnetic field being much smaller than the anisotropy field, the following relationship holds:

$$3\chi_0^r = 2\chi_{rt}^r + \chi_{rp}^r$$
.

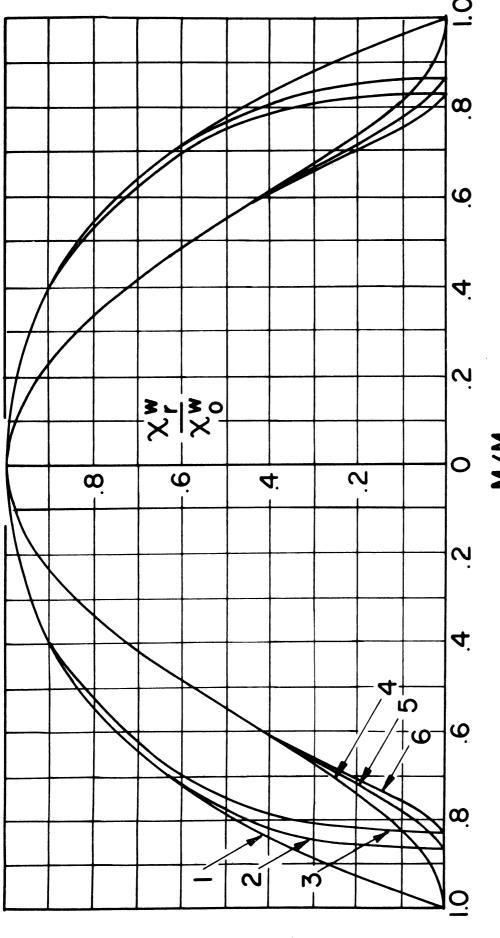
So if  $x \frac{r}{rp}$  is a monotonic decreasing function of M,  $x \frac{r}{rt}$  must be monotonic increasing function. These differences point out a new technique for the separation of the magnetization mechanisms: namely the measurement and comparison of the parallel and transverse reversible susceptibilities.

Fig. 1 shows the expected variation of the wall-motional susceptibilities according to Eqs. 1, 2, and 3; Fig. 2 shows the expected variation of the domain-rotational susceptibilities according to Eqs. 1, 4 and 5; Fig. 3 shows the expected wall-motional differential magnetostrictions according to Eqs. 8 and 9; Fig. 4 shows the expected domain-rotational differential magnetostrictions according to Eqs. 1, 10, and 11. Note that the largest value of d is expected from  $d_{\rm t}^{\rm r}$ , and remember no quantitative correlation is expected for M <  $\sim$  0.5 M<sub>s</sub>.

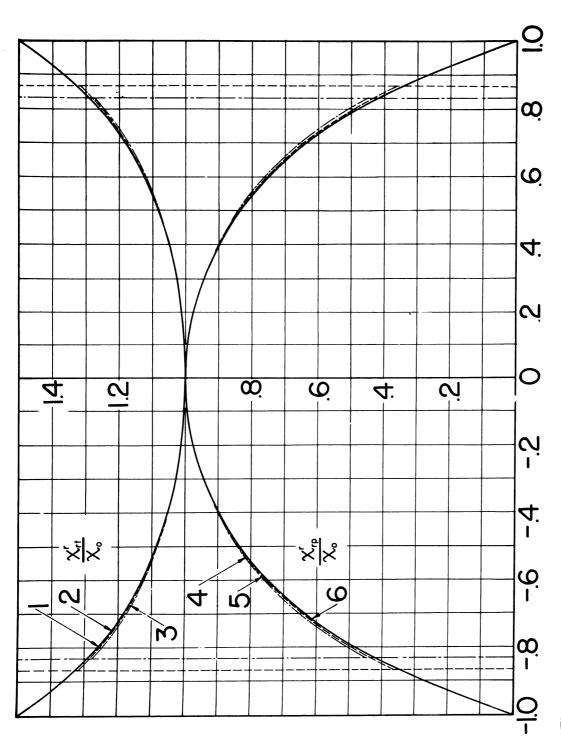
Experimental data have been taken on several ferrite samples to compare with the theoretical curves. Fig. 5 shows susceptibility data from specimen F-6-2, which was made in our laboratory by mixing to the composition Ni<sub>.1682</sub>Co<sub>.2992</sub>Zn<sub>.5326</sub>Fe<sub>2</sub>O<sub>l<sub>1</sub></sub>, firing at 1375°C. for 1/2 hour, 1200°C. for two hours and then slowly cooling---all in a N<sub>2</sub> atmosphere. The shaded area of Fig. 5 represents the area between the susceptibility curves calculated from Eqs. 2 and 3 and from assuming all moments to be either parallel or antiparallel with the applied magnetic field. From the data of Fig. 5 it is concluded that the susceptibility of F-6-2 is very predominantly due to domain rotation. To check this point the relaxation frequency was calculated from the equation:

$$f = \frac{\gamma}{2\pi} \cdot \frac{2M_s}{3\chi_0} \tag{15}$$

and found to be on the order of 140 mc/sec. Experimentally, the peak in the



for transverse fields, curves  $\mu$ , 5, and 6 are for parallel fields. Curves 3 and 6 are for six pos-Fig. 1. Theoretical reversible susceptibility due to domain-wall motion. Curves 1, 2, and 3 are sible directions of orientation of the magnetic moment in the single crystal. Curves 2 and 5 are for eight possible directions, and curves 1 and  $\mu$  are for an infinite number of such directions.



5 are for eight possible directions and curves 1 and 6 are for an infinite number of such directions. sible directions of static orientation of the magnetic moment in the single crystal. Curves 2 and Fig. 2. Theoretical reversible susceptibility due to domain rotation. Curves 1, 2, and 3 are for transverse fields, curves 4, 5, and 6 are for parallel fields. Curves 3 and 4 are for six pos-

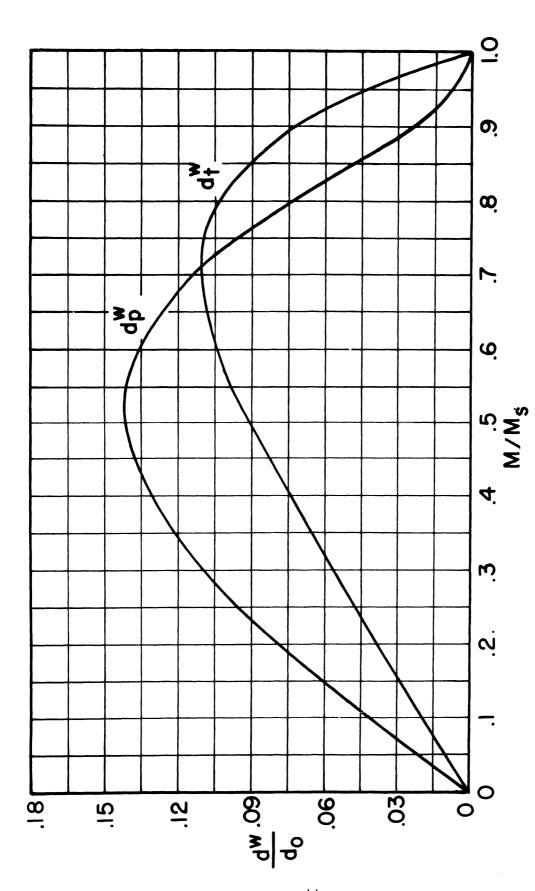


Fig. 3. Theoretical differential magnetostriction if the reversible susceptibility is due to domain-wall motion.  $d_{\mathrm{p}}^{\mathrm{W}}$  represents parallel fields and  $d_{\mathrm{t}}^{\mathrm{W}}$  represents transverse fields.

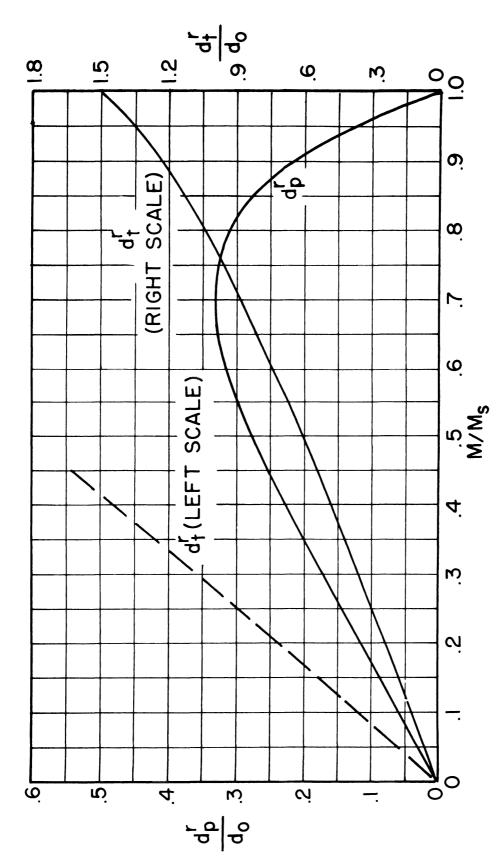
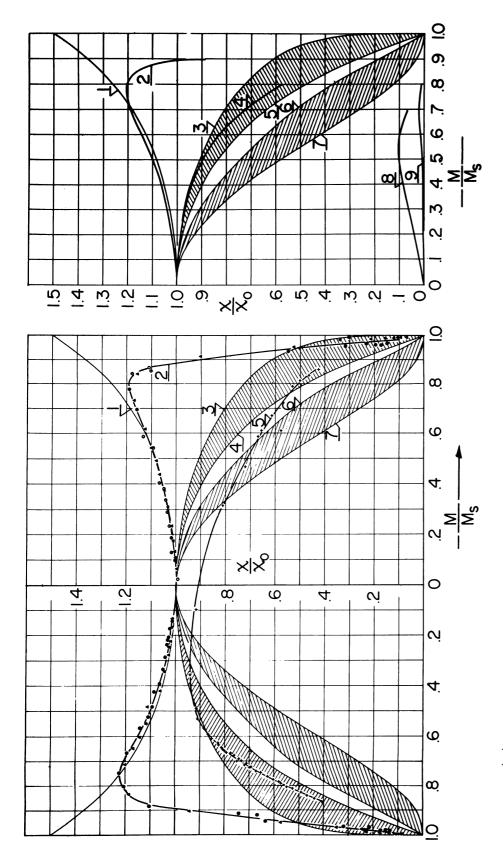


Fig. 4. Theoretical differential magnetostriction if the reversible susceptibility is due to domain rotation.  $d_{\mathrm{p}}^{\mathrm{r}}$  represents parallel fields and  $d_{\mathrm{t}}^{\mathrm{r}}$  represents transverse fields.



Curves verse fields for domain rotation and domain-wall motion respectively; curves 4 and 7 represent the 2 and 5 represent the experimental data taken on Core F-6-2 for transverse and parallel fields re-Curves 1 and 4 represent the theoretical curves for trans-Curves 3 and The arrow indicates the curves; curve 6 represent transverse and parallel fields for all moments either parallel or antiparallel with 2 represents the symmetrical part of the experimental transverse field data; curve 9 represents the antisymmetrical part; curve 4 represents the symmetrical part of the experimental parallel (a) Represents the variation of the reversible susceptibility with magnetization. variation for parallel fields, domain rotation and domain-wall motion respectively. (b) Shows the corresponding theoretical the applied field when the susceptibility is due to domain-wall motion. field data; curve 8 represents the antisymmetrical part. direction of the change in magnetization. spectively at both 10 and 320 kc/sec.

loss portion of the susceptibility was found to be 150 mc/sec. 9, and the frequency at which the susceptibility was one-half of the initial value was 160 mc/sec., verifying the rotational susceptibility mechanism.

Fig. 6 shows a similar curve for core I-15-1. It is from a batch of material prepared by Dr. D. Fresh at the U.S. Bureau of Mines, College Park, Md. The frequency dependence of this sample has been measured by Rado<sup>10</sup> et. al., and will be analyzed by him at the forthcoming London conference. The composition is assumed to be the same as that of their "Ferrite F": Mg.97Fe.03Fe204. The measurement of the reversible susceptibility of this particular material was initiated by request of Dr. Rado. The conclusion from Fig. 6 is that one-half or more of the initial susceptibility is due to domain wall motion.

Fig. 7 shows still another similar set of data on core F-10-2. F-10-2 was fired with F-6-2, but is believed to be of the composition Ni  $_{3800}$  Fe  $_{1282}$ Co  $_{0330}$ Zn  $_{14588}$ Fe  $_{20}$ l $_{1}$  $_{0}$  From the data of Fig. 7 it is inferred that over half of the susceptibility is due to domain-wall motion.

Fig. 8 shows a comparison of the experimental differential magnetostriction data of Bozorth and Williams with the results of Eqs. 1, 8 and 12. do was taken from their values for B<sub>s</sub> and  $\lambda_s$  while  $\lambda_s$  was taken from Bozorth. There have been no arbitrary scale corrections. Eq. 8 is deemed

<sup>9</sup> The frequency measurements were made by the High Frequency Impedance Standards Section, National Bureau of Standards, Boulder, Colorado.

<sup>10</sup> G. T. Rado, V. J. Folen, W. H. Emerson. To be presented at the Oct. 29 to Nov. 2 London Conference and published by the Institution of Electrical Engineers.

C. F. Jefferson, Tech. Report No. 66, Electronics Defense Group, Engineering Research Institute, University of Michigan. June 1956.

<sup>12</sup> R. M. Bozorth and H. J. Williams, Rev. Mod. Phys., 17, 72 (1945)

<sup>13</sup> R. M. Bozorth, Ferromagnetism D. Van Nostrand, 1951, p. 541

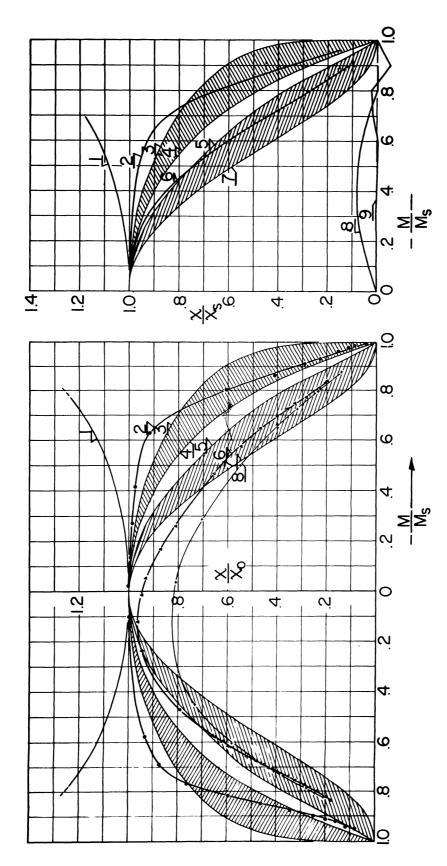


Fig. 6. This figure represents the same data for specimen I-15-1 as that illustrated by Fig. 5 for F-6-2.

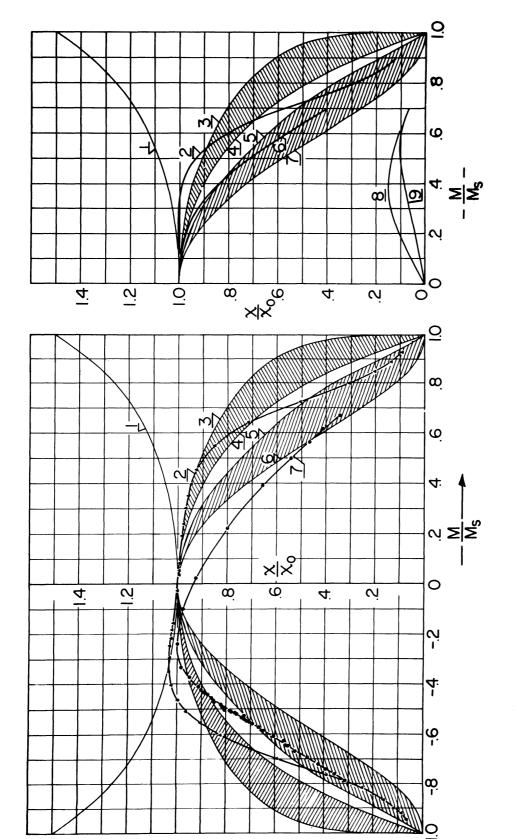
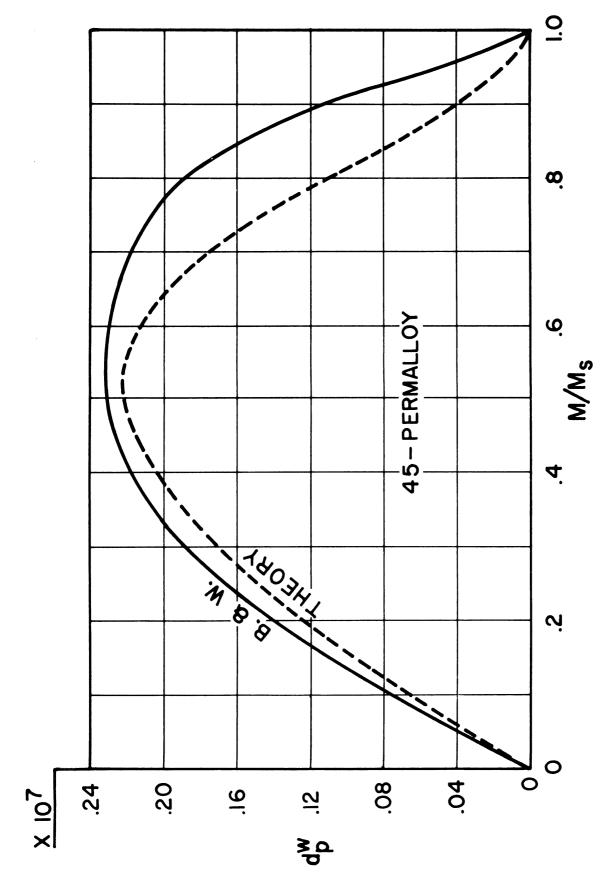


Fig. 7. This figure represents the same data for specimen F-10-2 as that illustrated by Fig. 5 for F-6-2 except that the data were taken only at  $320~{\rm kc/sec.}$ 



magnetostriction due to domain-wall motion with that reported by Bozorth and Williams for Per-Fig. 8. A comparison of the theoretical curve for the variation of the parallel differential malloy 45. There have been no arbitrary scale corrections.

superior to their corresponding equation for (a) as they pointed out, they used a domain-rotation model where the susceptibility was due to domain wall motion, and (b) they substituted  $[\cos\theta]^3$  for  $[\cos^3\theta]$ .

In conclusion, I wish to thank Dr. Rado for making their manuscript available to me before the conference.

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