Criterion for Uniform Micromagnetization

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Current theories of "single-domain" ferromagnetic particles compare the free energy in a state of uniform magnetization with that in an arbitrarily chosen state of nonuniform magnetization. In this paper, the comparison is made between an initial uniform state and all neighboring states, uniform or nonuniform, as an initially large applied field decreases. The initial state becomes unstable when, for some choice of the varied magnetization, the second variation of the free energy changes from positive to negative. This instability criterion leads to a boundary-value problem; the relative magnitudes of certain eigenvalues determine whether the deviation from the initial state occurs by uniform rotation or by development of nonuniform magnetization. Formulas for the critical radius are found in simple cases; they agree, except for a numerical factor, with formulas of Kondorskii.

1. INTRODUCTION

THE present theory of "single-domain" ferromagnetic particles is not rigorous. The usual procedure[1–4] is to compare the free energy of the uniformly magnetized state with that of some state of nonuniform magnetization; the latter is of arbitrarily selected form but contains one or more parameters, which are adjusted to minimize the energy for the chosen form. The state of lower energy is taken to be the actual one. Whichever way the calculation comes out, the conclusion is uncertain. If the nonuniform state has higher energy, there is no assurance that greater ingenuity might not have devised one of lower energy. If it has lower energy, there is no assurance that it is accessible from an initial uniform state; for there has been no investigation of intermediate states and of whether an activation energy is necessary for the transition.

A sounder method is that of Kondorskii.[5] The class of nonuniform states considered by him includes the uniform state as a limiting case. This eliminates the second uncertainty but not the first; for the minimization is carried out with respect to only two parameters.

The problem will be attacked here by a rigorous method. The energy of a specified initial uniform state will be compared with that of all neighboring states, both uniform and nonuniform. The conditions will be determined under which the initial state becomes unstable; and when instability occurs, the mode of deviation (whether uniform or nonuniform) will be determined.

2. BASIC THEORY

Consider a specimen with initially uniform magnetization in a specified direction, and take the z axis along this direction. Then for a rigid specimen the free energy is[6–9]

$$W=\tfrac{1}{2}C\int[(\nabla\alpha)^2+(\nabla\beta)^2+(\nabla\gamma)^2]d\tau + \int w(x,y,z,\alpha,\beta)d\tau - J_s\int(H_{0x}\alpha+H_{0y}\beta+H_{0z}\gamma)d\tau+W_M. \quad (2.1)$$

Here $d\tau$ is an element of volume, C is an exchange-energy constant, J_s is the magnitude of the spontaneous magnetization (assumed constant), (α,β,γ) are its direction cosines, $\mathbf{H}_0=H_{0x}\mathbf{i}+H_{0y}\mathbf{j}+H_{0z}\mathbf{k}$ is the applied field intensity, $w(x,y,z,\alpha,\beta)$ is the anisotropy energy density expressed as a function of position and of the two independent direction cosines (α,β), and W_M is the mutual energy of the poles that result from nonsolenoidal magnetization[10]:

$$W_M=-\tfrac{1}{2}J_s\int(\alpha H_x'+\beta H_y'+\gamma H_z')d\tau = (8\pi)^{-1}\int_{\text{space}}\mathbf{H}'^2d\tau. \quad (2.2)$$

The field intensity $\mathbf{H}'=-\nabla V$ of the poles can be calculated from a volume pole density $-\nabla\cdot\mathbf{J}$ and a surface pole density $\mathbf{n}\cdot\mathbf{J}$ by formulas (25) to (26) of reference 8; $\mathbf{J}=J_s(\alpha\mathbf{i}+\beta\mathbf{j}+\gamma\mathbf{k})$ is the vector magnetization, and $\mathbf{n}$ is a unit outward normal to the bounding surface S of the specimen.

Consider a hypothetical state of magnetization $(\alpha_0,\beta_0,\gamma_0)$, where α_0, β_0, and $\gamma_0=[1-\alpha_0^2-\beta_0^2]^{\frac{1}{2}}$ are specified functions of position. To find whether W is a

[1] William Fuller Brown, Jr., J. Appl. Phys. **11**, 160–172 (1940), Eqs. (10) and (11). There the method was applied not to fine particles but to long thin specimens.
[2] Charles Kittel, Phys. Rev. **70**, 965–971 (1946).
[3] Louis Néel, Compt. rend. **224**, 1488–1490 and 1550–1551 (1947).
[4] E. C. Stoner and E. P. Wohlfarth, Trans. Roy. Soc. (London) **A240**, 599–644 (1948).
[5] E. Kondorskii, Doklady Akad. Nauk S.S.S.R. **82**, 365–368 (1952); Izvest. Akad. Nauk S.S.S.R., Ser. Fiz. **16**, 398–411 (1952).
[6] L. Landau and E. Lifshitz, Physik. Z. Sowjetunion **8**, 153–169 (1935).
[7] W. C. Elmore, Phys. Rev. **53**, 757–764 (1938).
[8] William Fuller Brown, Jr., Phys. Rev. **58**, 736–742 (1940).
[9] Charles Kittel, Revs. Modern Phys. **21**, 541–583 (1949), Sec. 2.
[10] William Fuller Brown, Jr., Revs. Modern Phys. **25**, 131–135 (1953), Eq. (14).

minimum in this state, we must consider neighboring states $\alpha=\alpha_0+\epsilon u$, $\beta=\beta_0+\epsilon v$, $\gamma=[1-\alpha^2-\beta^2]^{\frac{1}{2}}$, where u and v are arbitrary functions of position and ϵ is an infinitesimal parameter. For given (u,v), let $\Phi(\epsilon)$ be the value of W in the state (α,β). Then sufficient (or with $\geqslant$ instead of $>$, necessary) conditions that W be a minimum in the state (α_0,β_0) are that

$$\Phi'(0)=0, \quad \Phi''(0)>0 \tag{2.3}$$

for arbitrary (u,v).[11]

We shall need $\Phi'(0)$ for arbitrary (α_0,β_0) and $\Phi''(0)$ for $\alpha_0=\beta_0=0$. The differentiation with respect to ϵ is most easily carried out by expanding the integrands to the second order in the variations $(\epsilon u,\epsilon v)$. In the expansion of W_M, the following magnetostatic theorem is useful: if $\mathbf{H}_i$ is the field intensity of poles due to magnetization $\mathbf{J}_i$, then

$$-\int \mathbf{J}_1\cdot\mathbf{H}_2 d\tau=-\int \mathbf{J}_2\cdot\mathbf{H}_1 d\tau=(4\pi)^{-1}\int_{\text{space}} \mathbf{H}_1\cdot\mathbf{H}_2 d\tau. \tag{2.4}$$

For arbitrary (α_0,β_0),

$$\begin{aligned}\Phi'(0)=C\int\left[\left(\frac{\partial\alpha_0}{\partial n}-\frac{\alpha_0}{\gamma_0}\frac{\partial\gamma_0}{\partial n}\right)u+\left(\frac{\partial\beta_0}{\partial n}-\frac{\beta_0}{\gamma_0}\frac{\partial\gamma_0}{\partial n}\right)v\right]dS\\ -C\int\left[\left(\nabla^2\alpha_0-\frac{\alpha_0}{\gamma_0}\nabla^2\gamma_0\right)u\right.\\ \left.+\left(\nabla^2\beta_0-\frac{\beta_0}{\gamma_0}\nabla^2\gamma_0\right)v\right]d\tau\\ +\int[w_\alpha(x,y,z,\alpha_0,\beta_0)u+w_\beta(x,y,z,\alpha_0,\beta_0)v]d\tau\\ -J_s\int\left[\left(H_x-\frac{\alpha_0}{\gamma_0}H_z\right)u\right.\\ \left.+\left(H_y-\frac{\beta_0}{\gamma_0}H_z\right)v\right]d\tau,\end{aligned} \tag{2.5}$$

where $\mathbf{H}=\mathbf{H}_0+\mathbf{H}'$ is the magnetizing force, evaluated with $\alpha=\alpha_0$, $\beta=\beta_0$. The exchange-energy term has been transformed by use of the divergence theorem.

For $\alpha_0=\beta_0=0$,

$$\begin{aligned}\Phi''(0)=C\int[(\nabla u)^2+(\nabla v)^2]d\tau\\ +\int(g_{11}u^2+2g_{12}uv+g_{22}v^2)d\tau\\ +J_s\int H_z(u^2+v^2)d\tau+(4\pi)^{-1}\int_{\text{space}}\mathbf{h}^2 d\tau,\end{aligned} \tag{2.6}$$

where $\mathbf{h}$ is the field intensity of poles due to magnetization $J_s(\mathbf{i}u+\mathbf{j}v+\mathbf{k}0)$; the g's may be defined as the coefficients in the expansion of w for small (α,β),

$$w=w_0+g_1\alpha+g_2\beta+\tfrac{1}{2}(g_{11}\alpha^2+2g_{12}\alpha\beta+g_{22}\beta^2)+\cdots. \tag{2.7}$$

In order that $\Phi'(0)$ may vanish for arbitrary (u,v), the coefficients of u and of v in the volume and surface integrals in Eq. (2.5) must vanish separately. This gives two partial differential equations and two boundary conditions [Eqs. (29) and (30) of reference 8]; they, together with the equations that determine V, must be solved in order to find functions (α_0,β_0) that make the first variation of W, $\epsilon\Phi'(0)$, vanish. The functions $\alpha_0=\beta_0=0$ satisfy the boundary conditions; they will also satisfy the partial differential equations provided

$$g_1-J_sH_x=g_2-J_sH_y=0. \tag{2.8}$$

If the specimen is homogeneous, so that w is independent of position, g_1 and g_2 are constants; if it is an ellipsoid in a uniform applied field (in arbitrary orientation), H_x and H_y are constants; if both these conditions are satisfied, then Eqs. (2.8) can be satisfied by adjustment of the applied field. We assume that the stated conditions are satisfied and the adjustment made; the applied field may still be varied in such a way as to change only H_z. For all values of H_z, the first of the two conditions (2.3) (the equilibrium condition) is now satisfied by $\alpha_0=\beta_0=0$. We turn our attention to the second (the stability condition), which requires that $\Phi''(0)$ as given by Eq. (2.6) shall be positive.

A *sufficient* condition for positiveness of $\Phi''(0)$ is that the quadratic form $T(u,v)=(g_{11}+J_sH_z)u^2+2g_{12}uv+(g_{22}+J_sH_z)v^2$ be positive definite. In a homogeneous crystal the g_{ij}'s are constants, and the x and y axes can be chosen so that $g_{12}=0$; then the state is stable if $-J_sH_z$ is less than g_{11} and g_{22}. Earlier,[12] this theorem was used to show that the usual postulates of domain theory were not capable of explaining the observed low coercive forces of crystals. In discussing that result, Stoner[13] suggested that imperfections of structure or of shape might promote instability at smaller values of $-J_sH_z$. Stoner's suggestion can be made quantitative by introducing perturbations of homogeneity or shape. For the present, however, we are concerned with an ideal ellipsoidal crystal; we must therefore investigate values of H_z for which $T(u,v)$, even without such perturbations, is no longer positive definite.

Consider a definite (u,v), and begin with H_z large enough to insure stability. If H_z is now decreased, it must eventually reach a value H_c at which, for the given (u,v), $\Phi''(0)$ vanishes; from Eq. (2.6), this value

[11] F. Schlögl, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 1, pp. 262–281.

[12] William F. Brown, Jr., Revs. Modern Phys. **17**, 15–19 (1945).

[13] Edmund C. Stoner, Repts. Progr. Phys. **13**, 83–183 (1950). See especially p. 116.

is given by

$$-J_sH_c=\Big\{C\int[(\nabla u)^2+(\nabla v)^2]d\tau + \int(g_{11}u^2+2g_{12}uv+g_{22}v^2)d\tau + (4\pi)^{-1}\int_{\text{space}}\mathbf{h}^2d\tau\Big\}\Big/\int(u^2+v^2)d\tau\equiv\frac{P}{Q}. \quad (2.9)$$

If H_z is decreased still further, Eq. (2.6) shows that $\Phi''(0)$ will become negative. The state $\alpha_0=\beta_0=0$ will then be unstable with respect to small deviations of the form $(\epsilon u,\epsilon v)$.

We wish to find the smallest value of $-J_sH_z$ at which instability sets in for *any* (u,v). We therefore seek the (u,v) for which P/Q is a minimum. The requirement that the first variation of P/Q vanish leads, by a method analogous to that used in deriving Eq. (2.5), to the partial differential equations

$$\begin{aligned}-C\nabla^2u+(g_{11}-\lambda)u+g_{12}v-J_sh_x=0,\\ -C\nabla^2v+g_{12}u+(g_{22}-\lambda)v-J_sh_y=0,\end{aligned} \quad (2.10)$$

and the boundary conditions

$$\partial u/\partial n=\partial v/\partial n=0; \quad (2.11)$$

here $\lambda=P/Q=-J_sH_c$.

The boundary value problem (2.10)–(2.11) can be solved only if λ has one of a set of eigenvalues λ_n. The corresponding eigenfunctions (u_n,v_n) are indeterminate by a constant factor. Eigenfunctions corresponding to different eigenvalues are orthogonal in the sense that

$$\int(u_mu_n+v_mv_n)d\tau=\delta_{mn} \quad (2.12)$$

for $m\neq n$, and the functions are conveniently normalized by imposing condition (2.12) also for $m=n$. To find the smallest $-J_sH_c$, we must find the smallest eigenvalue of λ; the corresponding $(\epsilon u_n,\epsilon v_n)$ describe the mode of deviation with respect to which the state $\alpha_0=\beta_0=0$ becomes unstable at this value of $-J_sH_z$.

An arbitrary (u,v) can be expanded as a series in the functions (u_n,v_n) with coefficients c_n. By substitution of the series in Eq. (2.9) and use of (2.10) and (2.12), we find

$$-J_sH_c=\textstyle\sum_n c_n^2\lambda_n/\sum_n c_n^2. \quad (2.13)$$

Equation (2.13) shows that $-J_sH_c$ can be as small as the smallest λ_n but no smaller. We have therefore achieved the desired minimum, and investigation of the second variation of P/Q is unnecessary. This assumes the completeness of the set of functions (u_n,v_n).

The partial differential equations and boundary conditions obtained from Eq. (2.5) reduce to Eqs. (2.10) to (2.11), to the first order in ϵ, if $\alpha_0=\epsilon u$ and $\beta_0=\epsilon v$, and if for $\epsilon=0$ Eqs. (2.8) are satisfied. Thus, formally, we can find the λ_n's and the functions (u_n,v_n) by seeking the (not necessarily stable) equilibrium first-order deviations from $\alpha_0=\beta_0=0$.

3. UNIFORM AND NONUNIFORM DEVIATIONS

We can always find a solution $u=\text{const}=u_0$, $v=\text{const}=v_0$. For this solution the boundary conditions (2.11) are satisfied, h_x and h_y are homogeneous linear functions of u_0 and v_0 (involving the demagnetizing factors of the ellipsoid and the direction cosines of its principal axes), and Eqs. (2.10) reduce to a pair of homogeneous linear simultaneous equations in u_0 and v_0. The requirement that the determinant vanish leads, in general, to two eigenvalues of λ. Let λ_0 be the smaller. If there are no other eigenvalues as small as λ_0, then when $-J_sH_z$ reaches the value λ_0, the system becomes unstable with respect to a uniform rotation of the magnetization vector. The behavior, for this particular orientation of the initial uniform magnetization with respect to the ellipsoid axes, is that of a Stoner-Wohlfarth particle.[4]

Other eigenvalues correspond to functions (u,v) that vary with (x,y,z). Let λ_1 be the smallest such eigenvalue. If λ_1 is smaller than λ_0, then when $-J_sH_z$ reaches the value λ_1, the system becomes unstable with respect to a nonuniform deviation of the form $(\epsilon u_1,\epsilon v_1)$. The magnetization acquires a nonuniform character that may be described as an incipient domain structure.

Let the shape and orientation of the specimen be kept fixed and its size varied. In general λ_1 will vary when the size varies, but λ_0 will remain constant. Which of the two eigenvalues is smaller will depend on the specimen size, and at some critical size the two will become equal. This is, for the given shape and orientation, the critical size for existence of a "single-domain" particle.

4. SYMMETRIC CASE

We consider the special case in which the ellipsoid is one of revolution about $0z$, and $g_{11}=g_{22}=g$, $g_{12}=0$. This is the case treated in reference 12.

For uniform deviations (u_0,v_0), $h_x=-N_bJ_su_0$, $h_y=-N_bJ_sv_0$, where N_b is the transverse demagnetizing factor. Equations (2.10) become $(g-\lambda+N_bJ_s^2)u_0=(g-\lambda+N_bJ_s^2)v_0=0$ and lead to a coincident pair of eigenvalues $\lambda_0=g+N_bJ_s^2$. Instability with respect to uniform rotation develops at this value of $-J_sH_z=-J_s(H_{0z}-N_aJ_s)$, where N_a is the longitudinal demagnetizing factor; the corresponding value of H_{0z} is given by

$$-J_sH_{0z}=g+(N_b-N_a)J_s^2. \quad (4.1)$$

This is the Stoner-Wohlfarth formula.[4]

For nonuniform deviations, Eqs. (2.10) give

$$\begin{aligned}-C\nabla^2u+(g-\lambda)u-J_sh_x=0,\\ -C\nabla^2v+(g-\lambda)v-J_sh_y=0.\end{aligned} \quad (4.2)$$

To determine the whole spectrum of eigenvalues is difficult. We can find a solution with $\mathbf{h}=0$ by taking, in spherical coordinates (r,θ,ϕ) or cylindrical coordinates (z,ρ,ϕ),

$$u=-u_\phi \sin\phi, \quad v=+u_\phi \cos\phi, \quad \partial u_\phi/\partial\phi=0; \quad (4.3)$$

then the volume and surface pole densities vanish.

If the ellipsoid is a sphere of radius b,

$$u_\phi=A j_n(kr)P_n^1(\cos\theta) \quad (n\geq 1), \quad (4.4)$$

where

$$k^2=(\lambda-g)/C; \quad (4.5)$$

the notation is that of Stratton.[14] The eigenvalues of k are determined by $j_n'(kb)=0$ and are smallest for $n=1$. Let x_1 be the smallest root of $j_1'(x)=0$, approximately 2.08. Then the smallest eigenvalue of k is x_1/b, and the smallest eigenvalue of λ is $\lambda_1=g+Ck^2=g+Cx_1^2/b^2$, within the set of eigenvalues corresponding to solutions of the form (4.3). If solutions of other forms yield no smaller eigenvalue, then the deviation from $\alpha_0=\beta_0=0$ occurs by uniform rotation or by the nonuniform mode (4.3) according as λ_0 or λ_1 is smaller; for the sphere, $N_b=4\pi/3$, so that $\lambda_0=g+4\pi J_s^2/3$. The deviation is therefore uniform or nonuniform according as the radius b is smaller or larger than the critical value that makes $\lambda_1=\lambda_0$, namely

$$b_c=\frac{x_1}{J_s}\left(\frac{3C}{4\pi}\right)^{\frac{1}{2}}. \quad (4.6)$$

If the ellipsoid is an infinitely long cylinder of radius b,

$$u_\phi=BJ_1((k^2-\mu^2)^{\frac{1}{2}}\rho)\cos(\mu z-\delta). \quad (4.7)$$

The eigenvalues are determined by $J_1'((k^2-\mu^2)^{\frac{1}{2}}b)=0$ and are smallest for $\mu=0$. Let x_1' be the smallest root of $J_1'(x)=0$, approximately 1.84. Then by an argument like that just given, the critical radius is

$$b_c=\frac{x_1'}{J_s}\left(\frac{C}{2\pi}\right)^{\frac{1}{2}}. \quad (4.8)$$

For a prolate spheroid in general, we may guess that the critical minor semiaxis will be given by a formula

$$b_c=\frac{x_1''}{J_s}\left(\frac{C}{N_b}\right)^{\frac{1}{2}}, \quad (4.9)$$

where x_1'' is a root of a transcendental equation and is intermediate between 1.84 and 2.08, varying with the ellipsoid eccentricity.

[14] Julius Adams Stratton, *Electromagnetic Theory* (McGraw-Hill Book Company, Inc., New York, 1941), pp. 404–406.

For a powder of identical and identically oriented spherical particles, far apart, the coercive force H_{0c} is not smaller than the value of $-H_{0z}$ at which the state $\alpha_0=\beta_0=0$ becomes unstable. Therefore

$$H_{0c}=g/J_s, \quad b<b_c;$$
$$\geq (g/J_s)[1-(\tfrac{4}{3}\pi J_s^2/g)(1-b_c^2/b^2)], \quad b>b_c. \quad (4.10)$$

The equality sign for $b<b_c$ is justified by a calculation by the Stoner-Wohlfarth method for finite (α,β), for hexagonal or cubic symmetry.

5. COMPARISON WITH EARLIER WORK

Kondorskii, for the case treated in Sec. 4, derived a formula equivalent to Eq. (4.9) with $x_1''=0.95\sqrt{5}=2.13$, larger than either of the values 1.84 and 2.08 derived above. His two-parameter method overestimates the energy of the nonuniform state and therefore predicts uniformity over a slightly too large range of radii. Stoner and Wohlfarth [their Eq. (6.10)] have $x_1''=\pi$, and N_a instead of N_b. The formulas of Kittel and of Néel are not of the form (4.9). Kittel's formulas contain the anisotropy constant, and Néel and Stoner and Wohlfarth believed that they were neglecting the crystalline anisotropy; but according to Kondorskii's calculation and the present one, the critical radius, in the symmetric case of Sec. 4, is independent of the anisotropy constant.

With Kondorskii's values [$10^6C=7.97$ (Fe), 3.52 (Ni); $J_s=1720$ (Fe), 500 (Ni); cgs emu], Eqs. (4.8) and (4.9) give for the critical radius in A: spheres, 167 (Fe), 382 (Ni); cylinders, 120 (Fe), 276 (Ni). Néel's sphere estimate for Fe was 160, Kondorskii's 170.

Kondorskii gives Eqs. (4.10) with equality signs, and with $4\pi J_s^2/3g$ written p and not explicitly evaluated.

6. CONCLUDING REMARKS

The following gaps in the theory should be filled. For the sphere and cylinder, the rest of the eigenvalue spectrum should be explored. For the prolate spheroid, the boundary-value problem should be solved with spheroidal wave functions.[15] More general cases, e.g., with $\mathbf{J}$ inclined to the ellipsoid axes, should be investigated. The completeness of the set of functions (u_n,v_n) should be established.

What may be more fruitful, however, is the extension of the theory to nonellipsoidal shapes and nonuniform g_{ij}'s. Such a study should improve understanding of domain nucleation and the coercive force.

[15] Stratton, Morse, Chu, Little, and Corbató, *Spheroidal Wave Functions* (The Technology Press of Massachusetts Institute of Technology and John Wiley and Sons, Inc., New York, 1956).