Model for Lowering the Nucleation Field of Ferromagnetic Materials*

C. ABRAHAM
Department of Electronics, The Weizmann Institute of Science, Rehovoth, Israel
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A model is considered which can reduce the theoretical nucleation field. It assumes nonuniformity in the ferromagnetic crystals due to the finiteness of the crystal, namely, the existence of a surface layer with different physical properties from those in the bulk. Since atoms near the surface of the specimen are in a lower crystal symmetry than the inner atoms which are far from the surface, different free energies are assumed for the bulk and the surface layer. These are taken as changes in the magnetocrystalline anisotropy constant, in the exchange energy constant, and in the magnitude of the magnetization vector. The nucleation field is calculated for an infinite slab of finite width and for an infinite circular cylinder. For a relatively small width of the surface layer, it was found that the nucleation field is reduced by a factor 2 with respect to a crystal in which no different property of the surface layer is assumed.

I. INTRODUCTION

THE micromagnetics theory predicts a very large energy barrier for domain nucleation in a previously saturated ferromagnetic crystal. The experimental observations show that the domains nucleate before the applied magnetic field reaches the predicted value[1] of the nucleation field $H_n=-(2K/J_s-NJ_s)$, where K is the magnetocrystalline anisotropy constant, J_s is the magnitude of the magnetization vector, and N is the demagnetization constant along the z axis. This discrepancy between the predicted value for the applied magnetic field and the observed value at nucleation is known as the Brown's paradox.[1] In this work an attempt is made to reduce the theoretical nucleation field H_n by assuming a nonuniformity in the physical properties of the ferromagnetic specimen. We shall consider a specimen formed from two regions: the inner part, which will be called the bulk, and a shell near the surface of the specimen, which will be called the surface layer. The physical meaning of this assumption lies in the fact that the atoms near the surface of the crystal are in a lower symmetry[2] than the atoms far away from the surface. The technique of spin-wave excitation provides information[3] about the existence of a thin layer near the surface of the specimen in which the properties of the matter are different than the properties of the bulk material.

The aim of the present model of reduction of the nucleation field is to show how much the finiteness of the specimen can influence the existence of Brown's paradox.

II. GENERAL THEORY

A ferromagnetic material infinite in the z direction, which has a uniaxial magnetocrystalline anisotropy, is considered. The applied magnetic field H_0 and the direction of easy magnetization are in the z direction.

The exchange energy constant $\bar{A}$ is assumed to be

$$\bar{A}=\begin{cases}A \text{ in the bulk}\\ \rho_1 A \text{ in the surface layer,}\end{cases} \tag{1a}$$

where ρ_1 is a positive parameter. Since a nonuniformity is assumed in the exchange energy, then there must be a nonuniformity in the magnitude of the magnetization vector. The magnitude of the magnetization vector $\bar{J}_s$ is assumed to be

$$\bar{J}_s=\begin{cases}J_s \text{ in the bulk}\\ \rho_2 J_s \text{ in the surface layer,}\end{cases} \tag{1b}$$

where $\rho_1<\rho_2<1$ is assumed.

For the surface layer a lower value of the magnetocrystalline anisotropy coefficient[4,5] $\bar{K}$ is assumed.

$$\bar{K}=\begin{cases}K \text{ in the bulk}\\ 0 \text{ in the surface layer.}\end{cases} \tag{1c}$$

A more general model will be obtained with the assumption that the easy direction of magnetization in the surface layer would make an angle[6] different from zero with the easy direction of the bulk. For an easy direction in the surface layer which is normal to the easy direction of the bulk, the nucleation field will be more positive than the present model,[7] but it complicates very much the numerical computation.

As we are interested only in the value of the nucleation field, it is sufficient to solve the linearized Brown's equations.[8]

$$2\bar{A}\nabla^2\alpha_i=\bar{J}_s(\partial U_i/\partial x)+\bar{J}_s\{(2\bar{K}/\bar{J}_s)+H_0-\partial U_i/\partial z\}\alpha_i, \tag{2a}$$

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[4] A. Aharoni, Phys. Rev. 119, 127 (1960).
[5] C. Abraham and A. Aharoni, Phys. Rev. 128, 2496 (1962).
[6] W. F. Brown, Jr., *Micromagnetics* (Interscience Publishers, John Wiley & Sons, Inc., New York, 1963), p. 96.
[7] W. F. Brown, Jr., Phys. Rev. 124, 1348 (1961).
[8] W. F. Brown, Jr., Phys. Rev. 58, 736 (1940).

$$2\bar{A}\nabla^2\beta_i=\bar{J}_s(\partial U_i/\partial y)+\bar{J}_s\{(2\bar{K}/\bar{J}_s)+H_0-\partial U_i/\partial z\}\beta_i, \quad (2b)$$

where α and β are the components of the magnetization vector in the x and y direction, respectively, $i=1$ refers to the bulk, and $i=2$ refers to the surface layer. The boundary conditions on the surface of the specimen are

$$\partial\alpha_2/\partial n=0, \quad \partial\beta_2/\partial n=0. \quad (2c)$$

Here n is an unit vector normal to the surface of the specimen. U_i is the magnetostatic potential and is related to α_i and β_i by Poisson's equation $\nabla^2U_i=4\pi\bar{J}_s(\partial\alpha_i/\partial x+\partial\beta_i/\partial y)$. U_2 is also related to the magnetostatic potential for the external region of the specimen by the usual boundary conditions[8] of potential theory on the surface of the specimen. The boundary conditions (2c) and the usual boundary conditions for the magnetostatic potential[8] involve only the functions α_2, β_2, and U_2, which belong to the surface layer. In order to determine a unique solution, one needs extra conditions on the boundary between the bulk and surface layer. These are obtained from the assumption of the micromagnetics theory that the magnetization direction and its first derivatives with respect to (x,y,z) changes continuously in the ferromagnetic specimen. The additional conditions imposed on the functions α_i, β_i, and U_i at the boundary between the two regions are

$$\alpha_1=\alpha_2, \quad \beta_1=\beta_2, \quad U_1=U_2, \quad (2d)$$

$$\partial\alpha_1/\partial m=\partial\alpha_2/\partial m, \quad \partial\beta_1/\partial m=\partial\beta_2/\partial m, \quad \partial U_1/\partial m=\partial U_2/\partial m. \quad (2e)$$

Here m is a unit vector normal on the boundary between the bulk and surface layer.

III. INFINITE SLAB

The form of the specimen is an infinite slab in the (y,z) directions of width $2(p+1)d$, where p is a parameter and d is the width of the assumed surface layer. Then the two different regions are (a) the surface layer $pd\leq|x|\leq(p+1)d$, and (b) the bulk $|x|\leq pd$. Now we will assume that $\alpha_i=U_i\equiv0$ and $\beta_i=\beta_i(x)$; then, from (2b) we will get the two equations for the two regions.

$$d^2\beta_1/dt^2=T^2(1-h)\beta_1; \quad |t|\leq p, \quad (3a)$$

$$d^2\beta_2/dt^2=-\rho^2T^2h\beta_2; \quad p\leq|t|\leq p+1, \quad (3b)$$

where

$$t=x/d, \quad T^2=d^2K/A, \quad h=-H_0J_s/2K, \quad \rho^2=\rho_2/\rho_1. \quad (3c)$$

In order to simplify the calculation of the eigenvalue h, we will deal separately with the odd and even solutions of (3). Because the numerical calculations show that the even solution gives a more positive nucleation field, we will deal only with this solution for $0\leq t\leq p$.

The boundary conditions (2) are in this case

$$(d\beta_1/dt)_{t=0}=0; \quad (d\beta_2/dt)_{t=p+1}=0 \quad (4a)$$

$$(\beta_1=\beta_2)_{t=p}; \quad [(d\beta_1/dt)-(d\beta_2/dt)]_{t=p}=0. \quad (4b)$$

The general even solution of (3) which satisfies the boundary condition (4a) is

$$\beta_1=a_1(\exp\{T(1-h)^{1/2}t\}+\exp\{-T(1-h)^{1/2}t\}), \quad t\leq p, \quad (5a)$$

$$\beta_2=a_2\cos\{\rho Th^{1/2}(p+1-t)\}, \quad p\leq t\leq p+1. \quad (5b)$$

From (4b) and (5) we get the transcendental equation (6a), which gives the eigenvalues h,

$$(1-h)^{1/2}\tanh\{pT(1-h)^{1/2}\}=\rho h^{1/2}\tan(\rho Th^{1/2}). \quad (6a)$$

For $p>100$, $h<0.99$, and $T>0.1$ one gets

$$\tanh\{pT(1-h)^{1/2}\}\approx1;$$

then for these indicated values of p,h and T, the transcendental equation (6a) reduces to

$$(1-h)^{1/2}=\rho h^{1/2}\tan(\rho Th^{1/2}). \quad (6b)$$

The least negative values of h which satisfy (6a) are plotted in Fig. 1 as functions of the parameter ρ for two values of T.

IV. INFINITE CYLINDER

A ferromagnetic material in the form of an infinite cylinder with the axis in the z direction of radius $R=(p+1)d$ is assumed. The bulk region is the inner cylinder of radius pd; the surface layer is the outer shell of width d. The problem has a cylindrical symmetry, and it is easier to deal with the equivalent differential equations (2a), (2b) and Poisson's equation written for the cylindrical components $(\alpha_\varphi,\alpha_r)$ of the magnetization vector in the cylindrical coordinates r, φ, and z. This transformation has already been done.[9] We shall solve the equations only for the curling[9] mode which is obtained by assuming: $\alpha_r\equiv0$, $U\equiv0$, $\alpha_\varphi=\alpha_\varphi(r)\not\equiv0$, since this mode of nucleation[1,9] is the easiest for cylindrical radii larger than $1.08\,A^{1/2}J_s^{-1}$ (which is about 60 Å for iron and about 300 Å for $BaFe_{12}O_{19}$).

The differential equations for the two regions are

$$\left\{\frac{d^2}{dt^2}+t^{-1}\frac{d}{dt}-t^{-2}-T^2(1-h)\right\}\alpha_{\varphi1}=0, \quad 0\leqslant t\leqslant p, \quad (7a)$$

$$\left\{\frac{d^2}{dt^2}+t^{-1}\frac{d}{dt}-t^{-2}+T^2\rho h\right\}\alpha_{\varphi2}=0, \quad p\leqslant t\leqslant p+1, \quad (7b)$$

where $t=r/d$ and (ρ,T,h) are defined as in (3c). The general solutions for (7) are

$$\alpha_{\varphi1}(t)=aJ_1(Qt), \quad 0\leq t\leq p; \quad (8a)$$

$$\alpha_{\varphi2}(t)=b\{BJ_1(qt)+Y_1(qt)\}, \quad p\leq t\leq p+1. \quad (8b)$$

Here a, b, and B are integration constants, $Q=T(1-h)^{1/2}$, $q=\rho Th^{1/2}$, J_1 and Y_1 are Bessel functions of first and second kind, respectively. The boundary condition

[9] A. Aharoni and S. Shtrikman, Phys. Rev. **109**, 1522 (1958).

$(d\alpha_{\varphi_2}/dt)_{t=p+1}=0$, gives

$$B=-\frac{\rho T(p+1)h^{1/2}Y_0\{\rho Th^{1/2}(p+1)\}-Y_1\{\rho Th^{1/2}(p+1)\}}{\rho T(p+1)h^{1/2}J_0\{\rho Th^{1/2}(p+1)\}-J_1\{\rho Th^{1/2}(p+1)\}}.$$

The conditions (2e) give the transcendental equation

$$(1-h)^{1/2}\frac{I_0\{pT(1-h)^{1/2}\}}{I_1\{pT(1-h)^{1/2}\}}=\rho h^{1/2}\frac{Y_0(\rho pTh^{1/2})+BJ_0(\rho pTh^{1/2})}{Y_1(\rho pTh^{1/2})+BJ_1(\rho pTh^{1/2})}, \quad (9)$$

where I is the modified Bessel function.

The least values of $h(h_n)$ which satisfy the equation (9) are plotted in Fig. 1 under the name "Cylinder Curling Mode" as a function of the parameter ρ for $p=50$ and two values of T. For large values of p (this means large cylindrical radii) the transcendental equation (9), transforms to the equation (6b). The nucleation field is the same for a cylinder of a large radius or a thick slab.

V. DISCUSSION

It has been shown that for large values of p the nucleation field of the infinite cylinder equals that of the infinite slab. The nucleation field is reduced by increasing the width d of the assumed surface layer, and by increasing the value of the parameter ρ.

For hard ferromagnetic materials ($K\gg J_s^2$) like MnBi and $BaFe_{12}O_{19}$, $T=0.2$ corresponds to a width d of about 6 and 11 Å, respectively. For soft ferromagnetic materials ($K\ll J_s^2$) the same value of T corresponds to much larger values of d. This means that the present model gives a negligible reduction to the nucleation field for soft ferromagnetic crystals when reasonable width is assumed for the surface layer. That fact is not in contradiction with the observed values for the nucleation field of iron whiskers. De Blois and Bean[10] measured nucleation fields approaching the theoretical value in certain parts of most perfect iron whiskers. For hard ferromagnetic crystals, a big discrepancy still exists between the calculated and observed values of the nucleation field. But for hard ferromagnetic crystals, the present model gives a significant reduction in nucleation field by assuming only a relatively small width for the surface layer, and an arbitrary, but reasonable, value for the parameter ρ. The observed coercive force[1] of MnBi (which should certainly be more negative than the nucleation field) ranges from -12 kOe for 5-μ particles to -0.6 kOe for 100-μ particles. The theoretical nucleation field for an elongated particle of MnBi is about -26 kOe. For large particles the discrepancy might arise from the fact that the particles are not single crystals, but, for small particles, it is most probable that they are single crystals and the discrepancy arises from the finiteness of the specimen. The present model is able to fit the observed nucleation field for small particles of MnBi if one assumes a surface layer of 6-Å width (where for the bulk, K is[11] 8.9×10^6 erg/cm^3, πJ_s is[12] 2200 G, and the exchange constant is taken as 10^{-6} erg/cm^3). The micromagnetics theory is a continuum model and it breaks down at such a small scale (6 Å), but in this work the surface layer region has a meaning of a boundary condition and not of a real region.

A series of papers[4,5,13] used Aharoni's imperfection model (which assumes only that the value of the anisotropy constant is lower at the crystal imperfection region) in order to show the influence of assumed inner crystal imperfection on the nucleation field. It is shown[5] that Aharoni's model for inner crystal imperfection can resolve Brown's paradox for hard ferromagnetic material. Also, the present model can show that for hard ferromagnetic materials, Brown's paradox is influenced by the finiteness of the sample. No one of the two models

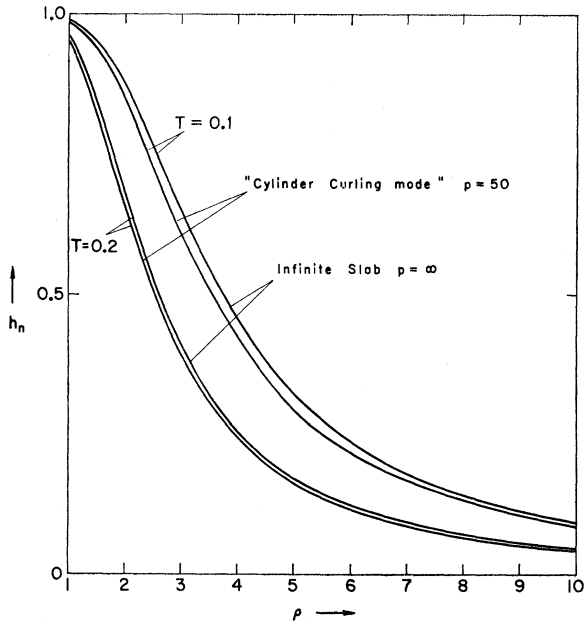

FIG. 1. The reduced nucleation field h_n for an infinite slab and an infinite cylinder as a function of the parameter ρ for two values of T.

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[11] R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1951), p. 575.

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excludes the other, and they may be taken together in order to reduce the calculated nucleation field.

In Sec. I, it was assumed that $\rho_i<1$ (for $i=1, 2$), but from the equations (6) and (9) we can see that, if it is assumed $\rho_i>1$, the calculated nucleation field remains the same as plotted in Fig. 1 for the same value of ρ. This means that the exchange energy and the magnitude of the magnetization vector can be assumed to be larger in the surface layer than in the bulk and still the calculated nucleation field is lower.

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High-Temperature Series Expansions for the Spin-$\frac{1}{2}$ Heisenberg Model by the Method of Irreducible Representations of the Symmetric Group*

GEORGE A. BAKER, JR.
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico

AND

G. S. RUSHBROOKE
Physics Department, King's College, Newcastle-upon-Tyne, England

AND

H. E. GILBERT
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico

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We show how the partition functions for finite clusters with spin-$\frac{1}{2}$ Heisenberg interactions may be computed efficiently and generally to any desired number of powers in reciprocal temperature. As an example, we have expanded the zero-magnetic-field free energy to the twenty-first power for the linear Heisenberg model and for nonzero magnetic field give an expression good through the tenth power. We introduce the concept of the two-point Padé approximant and use it to analyze the energy for the linear Heisenberg model.

1. INTRODUCTION AND GENERAL THEORY

RECENT advances in the ability of experimental physicists to measure the nature of the singularity in various thermodynamics functions near the critical point have raised anew the question of the adequacy of the Heisenberg model of magnetism to describe real substances in the critical region.[1] Studies by various authors[2] have shown that in the analogous Ising model, the most precise method now known of determining the predictions of models of this sort is the analysis of the exact power-series expansions (in reciprocal temperature, etc.) of the various thermodynamic functions. The major problem involved in extending the power series for the Heisenberg model has been the calculation of the traces of the spin operators involved. In this section of our paper we show how that step can be greatly simplified and easily adapted for a computer. In the last section of our paper we will apply our method, as an example, to the linear Heisenberg model, and analyze, by means of the Padé approximant method, the energy and magnetic susceptibility. We digress in the second section to introduce the concept of the 2-point Padé approximant, which turns out to be extremely useful in discussing the linear ferromagnetic Heisenberg model.

Domb[3] has pointed out that the partition function of an infinite lattice can be simply expressed in terms of the partition functions for finite clusters. That this procedure is possible follows from the fact that the logarithm of the partition function for a general lattice can be written in the form

$$\ln Z^{(j)}=\sum_\alpha p_\alpha{}^{(j)}\varphi_\alpha, \tag{1.1}$$

where α denotes a connected graph, $p_\alpha{}^{(j)}$ is the number of distinct ways it occurs on lattice (j), and φ_α is a unique function associated with graph α. By applying (1.1) successively to various finite clusters we may solve for the φ_α's, and then, knowing the lattice constants

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