Theoretical Search for Domain Nucleation

AMIKAM AHARONI

Department of Electronics, The Weizmann Institute of Science, Rehovoth, Israel

I. INTRODUCTION

THE energy of a sufficiently large ferromagnetic body can be enormously reduced by its being subdivided into domains that are magnetized in different directions. This division is actually observed experimentally by various techniques.[1] However, it is not understood how these domains nucleate in a previously saturated material, since the theory predicts a very large barrier, which should have practically prevented the observation of these domains. This discrepancy between theory and common observation, known as the Brown paradox, can be stated quantatitively as follows.

Consider a single crystal which has the shape of an ellipsoid of revolution and let the axis of symmetry, which is assumed to coincide with a direction of easy magnetization, be chosen as the z axis. Apply a large enough field in the $+z$ direction so that the sample is magnetized to saturation, then start reducing the field very slowly (so that eddy currents and other dynamic effects can be neglected), reversing it if necessary. In the course of this process, a certain value of the field will eventually be reached at which the state of magnetization to saturation is no longer stable. At this so-called "nucleation field," H_n, an infinitesimal additional change of the field will start some changes across a certain mode of magnetization reversal. What is most important, however, for this discussion, is that no change can occur, in particular no domain can nucleate, *before* the field H_n is reached. In his original paper,[2] Brown did not calculate the nucleation field, but he could give a lower limit; that is, he could prove that

$$-H_n \geqslant 2K/I_s - NI_s. \tag{1}$$

Here, I_s is the saturation magnetization, K is the constant of anisotropy (both considered constants of the material), and N is the demagnetization constant along the z axis. Here and in the following, the symmetry of the crystal does not enter, and K may be referred to cubic as well as to undirectional anisotropy, since at nucleation the material is assumed magnetized in an easy direction with only small deviation from this state.

Later, both Brown,[3] and Frei, Shtrikman, and Treves[4] calculated the nucleation field for a certain mode, called the "magnetization curling," for a sphere and for an infinite cylinder. Since a certain mode is assumed, one does not know if the field thus calculated is numerically the smallest possible, so that their results can be regarded only as an upper limit, yielding

$$-H_n \leqslant 2K/I_s - NI_s + 2\pi I_s k(R/R_0)^{-2}. \tag{2}$$

Here, R is the radius of the ellipsoid in a direction perpendicular to z, $R_0 = A^{\frac{1}{2}} I_s^{-1}$, A is the exchange constant,[1] and k is a numerical factor which equals 1.38 for a sphere and 1.08 for an infinite cylinder. They have also assumed[4] that (2) holds for any prolate spheroid, with $1.08 \leqslant k \leqslant 1.38$, a result which was later established by Aharoni,[5] who calculated k as a function of the elongation of a prolate spheroid. It was also shown[3,4] that for $R < 1.1R_0$ in the case of cylinder, and for $R < 1.4R_0$ in the case of a sphere, (2) can be written with $<$ instead of $\leqslant$, since other modes of magnetization reversal give numerically smaller values for H_n than are encountered by (2). This region, however, is of very small interest since R_0 is rather small (60 A for iron) and for such radii, at least the spherical particles become superparamagnetic[6] and the whole argument behind Eqs. (1) and (2) breaks down. In this review, therefore, we are interested only in the region where R is larger than, say, $1.5R_0$. In this region, it was proved that for an infinite cylinder[7] and for a sphere,[5] the curling is the lowest mode, so that (2) can be written with the equality sign. Being so at both limits, the equality sign in (2) probably holds for the general prolate spheroid, but this has not been proved.

For fine particles, i.e., in the region of sizes where the last term of (2) is still important, nucleation field was not measured for spherical or slightly elongated particles, so that (2) cannot be compared to experiment. For an infinite cylinder, however, both Brown[8] and Aharoni and Shtrikman[7] have shown that the hysteresis curve is rectangular, with the nucleation field identical with the coercive force. One can, therefore, compare (2) to the measured coercive force of *elongated*, fine particles. Figure 1 is a plot of (2) taking for iron[9] $2K/I_s = 560$ oe at room temperature. The experimental data for the coercive force of elongated, iron particles at room temperature in this figure are reproduced from Fig. 4 of Luborsky.[10]

[1] C. Kittel, Revs. Modern Phys. **21**, 541 (1949).
[2] W. F. Brown, Jr., Revs. Modern Phys. **17**, 15 (1945).
[3] W. F. Brown, Jr., Phys. Rev. **105**, 1479 (1957).
[4] E. H. Frei, S. Shtrikman, and D. Treves, Phys. Rev. **106**, 446 (1957).
[5] A. Aharoni, J. Appl. Phys. **30**, 70S (1959).
[6] C. P. Bean and J. D. Livingstone, J. Appl. Phys. **30**, 120S (1959).
[7] A. Aharoni and S. Shtrikman, Phys. Rev. **109**, 1522 (1958).
[8] W. F. Brown, Jr., J. Appl. Phys. **29**, 470 (1958).
[9] R. W. De Blois and C. P. Bean, J. Appl. Phys. **30**, 225S (1959).
[10] F. E. Luborsky, J. Appl. Phys. **32**, 171S (1961).

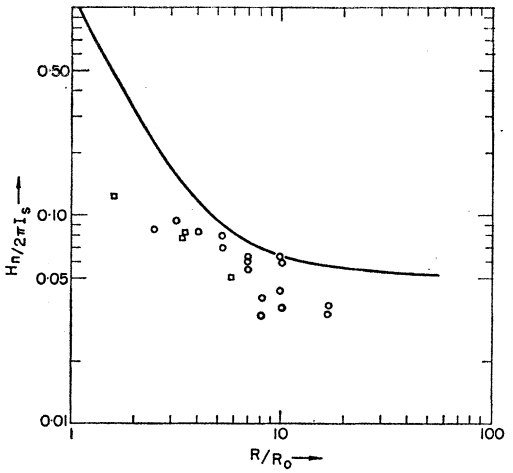

FIG. 1. Theoretical nucleation field, H_n, for an infinite cylinder of iron at room temperature as a function of its reduced radius compared to experimental results for the coercive force of fine elongated iron particles at room temperature and zero packing: the circles are wire drawing, the boxes are reduction of acicular oxides (after Luborsky[10]).

The disagreement between theory and experiment in Fig. 1 does not seem much worse than between one experiment and another. The agreement is remarkable if one remembers that the particles under study were not single crystals, but a rather poor substitution for *circular*-shaped infinite cylinders, and that for a large proportion of them the field was probably not applied along an easy axis. What is perhaps most striking is that all the experimental points lie *below* the theoretical curve and there are none above it. This can be regarded as a confirmation of the theory which predicts that, at a certain field, even a perfectly cylindrical, single crystal will reverse its magnetization. It should be noted, however, that the samples studied showed a $1-p$ dependence of the coercive force on packing,[11] whereas for the curling mode in a circular infinite cylinder there is no interaction, and, therefore, no dependence on packing. This is evidently due to the lack of cylindrical symmetry in the particles (see Fig. 13 of Luborsky[10]), which is essential for the ideal curling mode. When this symmetry is absent, it is reasonable to assume one has to deal with different modes than the curling, for which there is a packing dependence, yet the nucleation field does not differ much from that of the curling. It would be very interesting in this respect to study the substitute for curling in an elliptical infinite cylinder and the packing dependence it implies, but this has not yet been done. Additional data seem also desirable for the comparison, both from experimentalists (e.g., measurements on elongated particles of *hard* materials) and theoreticians (in particular the calculation of *coercive force* of spheres, for which there are some experimental data).

It can, therefore, be concluded that, for the theoretical and experimental data available now, the discrepancy is not large and easily understood as far as fine particles are concerned. This does not give any information about *domain* nucleation, since fine particles are known[1,12] not to be divided into domains, from mere comparison of the energies involved, or more simply because they are smaller than the domain-wall thickness (which is usually taken[1] as 10^3 A for iron). The situation is quite different when large particles are considered.

If R is large enough with respect to R_0, the last term in (2) can be neglected, and (1) and (2) can be combined to read

$$-H_n=2K/I_s-NI_s. \tag{3}$$

For soft materials (i.e., those for which $K\ll I_s^2$) of almost spherical shape, there is again no discrepancy simply because there are no data to compare. The coercive force or other details of the magnetization curve were not calculated, and the nucleation field was not measured, except for one recent pulse-technique experiment of De Blois,[13] who measured (with a doubtful accuracy, see Sec. II) a nucleation field of +6.4 koe in a single-crystal iron cube at 77°K. The theoretical value for an iron *sphere* is, according to (3), about +6.6 koe, which shows a remarkable similarity to the De Blois figure and is at least in no possible contradiction to any other measurement of hysteresis curves.

However, if the soft materials are in a form of elongated particles, N becomes small, and (3) predicts an extremely negative, nucleation field. For iron, for example, $-H_n$ should be[9] 560 oe, whereas domains are observed in positive fields, and even the coercive force of iron whiskers (which are very good single crystals and with N practically zero) is of the order of 0.1 oe. (except in the De Blois and Bean experiment[9] which is described in the following).

For hard materials, in which $K\gg I_s^2$, the second term in (3) does not contribute much in any way. Since the maximum value of N is 4π, this can be taken as the best possible value for comparison. For manganese bismuth, K is[14] 8.9×10^6 erg cm^{-3} and πI_s is[15] 2200 gauss at room temperature, giving for the right-hand side of (3) about 29 koe, whereas the experimental coercive force, which should certainly exceed the nucleation field, ranges[1] from 12 koe for 5-μ particles to 0.6 koe for 100-μ particles. Not only is there a large discrepancy, but it increases with particle size (R_0 for Mn Bi is about 140 A so that at 5μ the contribution of the last term of (2) is very small indeed). Another example of a hard material is $BaFe_{12}O_{19}$, for which the theoretical value (with $N=4\pi$) is[16] −12 koe, which exceeds the

[11] F. P. Levi, J. Appl. Phys. **31**, 1469 (1960).

[12] L. Néel, Compt. rend. **224**, 1488 (1947).

[13] R. W. De Blois, Rev. Sci. Instr. **32**, 816 (1961).

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

[15] W. C. Ellis, H. J. Williams, and R. C. Sherwood, J. Appl. Phys. **28**, 1215 (1957).

[16] C. Kooy and U. Enz, Philips Research Repts. **15**, 7 (1960).

experimental, coercive force[17] of 3 koe at room temperature. For this material, there is even an observation of nucleation fields[16] of a single crystal, ranging from +500 to +2000 oe.

In contradistinction to these order-of-magnitude discrepancies which constitute the Brown paradox, De Blois and Bean[9] have measured nucleation fields approaching the theoretical value in certain *parts* of most-perfect iron whiskers. This and the fair agreement between theory and experiment for fine particles suggest that the theory might be essentially correct, but it is based on too idealistic a model to be realized in practical experiments. Three fundamental assumptions of the theory are immediate sources of doubt as to whether they actually hold for the usual experiment: that one starts from a complete saturation, that the material is a single crystal, and that its shape is an ellipsoid. In the following three sections, the present knowledge of the possible importance of these three parameters is reviewed, some preliminary results of new theoretical developments are given, and an attempt is made to outline how to determine finally whether they are actually the cause of the Brown paradox. There are also other possible flaws in the theory that might be important for its failure to account for the experimental results. Two of these are briefly discussed in Sec. V, namely, the possibility of using a wrong expression for the exchange energy, or wrong boundary conditions. Finally, temperature effects are neglected in the theory. A justification for this is to be published elsewhere.

II. INCOMPLETE SATURATION

Starting from a completely saturated sample is an essential part of the theory. Only for a highly idealized, one-dimensional case have some calculations been recently drawn without using this assumption.[18] Until techniques are developed to deal with the nonlinear equations involved, which are very complicated mathematical problems, one should be careful to compare with the experimental results only in cases where saturation was initially reached. It has already been mentioned as a possibility in Brown's original paper[2] that the measured crystals "were never really saturated, but always contained vestigial domains." and that the samples should be initially subject to a field of 10^5 gauss.

That large fields, above technical saturation, are necessary, has been demonstrated by Shur, Shtoltz, and Margolina.[19] They observed a polydomain structure on MnBi particles of 10- to 15-μ size. The domains disappear completely at a field of 5 koe, so that saturation can be assumed to take place at this field, and the coercive force is a few hundred oe. However, once such a particle is subjected to a field of 20 koe, the domains disappear and *do not appear again* at any field, even though the hysteresis cycle is transversed many times, the coercive force becoming an order of magnitude larger than before. The polydomain structure can be obtained again by applying a decaying ac field or by cooling the crystal to liquid-air temperature and then heating back to room temperature (near liquid-air temperature, K of MnBi happens to pass through zero).

Fowler, Fryer, and Treves[20] found domains still persisting at fields in which the crystal is considered normally completely saturated. By improving the resolving power of the Kerr-effect technique, they could see reversed domains at the tip of iron whiskers in a field as high as 6 koe (which was evidently the largest field at their disposal).

The fact that domains remain at the tip of the whiskers is definitely connected with the problem of surface roughness. Shtrikman and Treves[21] noted that the demagnetizing field at a sharp corner is infinite, so that in principle no saturation can be achieved in any finite field. Nevertheless, this infinity, being logarithmic, is not very serious in itself, since the corner can be smoothed out by a radius of curvature of the order of atomic distance. After all, on an atomic scale there can never be an absolutely sharp corner, just as there cannot be an absolutely smooth surface. Consider, for example, a circular finite cylinder, with its bottom plate on $z=0$ and the z axis along its center of symmetry, magnetized to saturation in the $+z$ direction. The surface charges of the circle at $z=0$ give a potential V, which implies a demagnetizing field with $\partial V/\partial\varphi=0$, $\partial V/\partial z$ finite everywhere, and $\partial V/\partial r$ infinite at $z=0$, $r=R$ (where R is the radius of the cylinder). Now, if one allows z at $r=R$ to reach only a value a, the field at this point is approximately

$$\partial V/\partial r=2I_s\{K(k)-2\}, \tag{4}$$

where K is the complete elliptic integral of the first kind,[22] and[23]

$$k^2\approx 1-(a/2R)^2. \tag{5}$$

One obtains an infinite field from (4) if $a\to 0$, but, if a/R is as small as 10^{-3}, the field is only about $4\pi I_s$, the maximum NI_s value for an ellipsoid, and reaches $18.5I_s$ when $a/R=10^{-4}$. Even if a is taken to be of the order of interatomic distances, 10^{-4} is the smallest value

[17] G. W. Rathenau, J. Smit, and A. L. Stuyts, Z. Phys. **133**, 250 (1952).

[18] A. Aharoni, Phys. Rev. **123**, 732 (1961). After the publication of this article, I have found out that the solution reported there is unstable. Actually, Brown has recently shown that *all* one-dimensional solutions are unstable (private communication). However, the technique of starting from values other than saturation is worth extending.

[19] I. S. Shur, E. W. Shtolz, and W. I. Margolina, J. Exper. Theor. Phys. (U.S.S.R.) **38**, 46 (1960).

[20] C. A. Fowler, Jr., E. M. Fryer, and D. Treves, J. Appl. Phys. **32**, 296S (1961).

[21] S. Shtrikman and D. Treves, J. Appl. Phys. **31**, 72S (1960).

[22] P. F. Byrd and M. D. Friedman, *Handbook of Elliptic Integrals for Engineers and Physicists* (Springer-Verlag, Berlin, 1954).

[23] For $z=0$ and $r=R-a$, the value of k is somewhat smaller than that of (5).

for a/R when whiskers are concerned, and for larger samples it is certainly not difficult to round off corners to this accuracy. The actual fields are thus seen to be not very large and to be restricted to a very short range from the corner. One might, therefore, still assume that complete saturation is possible in practice, although it takes fields of about 30 koe to saturate an iron whisker, preferably larger to allow for possible barriers. The theory can thus be in no contradiction to experiments in which sufficiently large fields are not used, since once the domain wall is there, its movement does not imply as high a barrier as its formation.

The main problem now is whether the large demagnetizing field at sharp corners would be sufficient for domains to be nucleated at these points, even after the whisker has been completely saturated once. Some theoretical aspects of this problem are discussed in Sec. IV, but because infinities, even though they are logarithmic, are very inconvenient for approximation, such as perturbation theory, it does not seem likely that a complete theoretical answer to this important problem can be given. On the other hand, its experimental solution does not seem particularly difficult, in view of the following discussion.

As mentioned in Sec. I, De Blois and Bean[9] obtained values more negative than 500 oe for the nucleation field at some parts of iron whiskers, thus approaching the theoretical value of -560. At other parts, the field was numerically much smaller. Later, De Blois[24] observed that electropolishing could increase $|H_n|$ in places where it was rather small before, and that corrosion due to water droplets on the surface reduced considerably a high peak of $|H_n|$. Of particular significance with respect to the above-mentioned work of Fowler, Fryer, and Treves[20] is that De Blois could extend the region of high $|H_n|$ even to the *tip* for electropolished smoothly tapered whiskers.[24]

Although he could not give quantitative results for the dependence of the nucleation field on surface roughness, De Blois thus proved that surface roughness is the major effect in the case of iron, reducing the theoretical H_n to its experimental values. However, the fields he used were rather small, so that one cannot tell if the theoretical value is obtained in previously magnetized regions and the low values for H_n in regions where the field was not sufficient to saturate, or if nucleation is actually that much easier because of the demagnetizing field. In the De Blois experiment, these two effects are evidently superimposed, but the problem could be settled by applying a large enough field in studying the "bad" regions and by measuring the nucleation field at these spots. It actually means combining the techniques of this experiment with the De Blois minature coil for high fields.[13] The other way is to try to calculate, as in Sec. IV, the effect of the demagnetizing field, assuming previous saturation and to see whether it checks quantitatively with the De Blois experiment.

There is at least some indication that, once a sample is actually completely saturated, the infinite demagnetizing field at the sharp corners is not as important for domain nucleation as assumed by Treves and his co-workers.[20,21] This might be concluded from the experiment of De Blois[13] (mentioned in Sec. I), who obtained for an iron *cube*, practically the theoretical nucleation field of an iron sphere, leaving a negligible contribution to the sharp edges of the cube. One should, however, be rather careful in drawing conclusions from this experiment, because of the stated,[13] possible zero-level error; because of the extraneous noise in the output coil, which comes much before the main signal but in principle could have obscured a signal of previously started magnetization reversal; because the technique measures the changes in the bulk magnetization and could hardly be expected to reveal domain nucleating or disappearing over a small portion of the volume; but mainly because at a highly positive fields many effects can superimpose, all yielding practically the same nucleation field. If this experiment were performed on a whisker, where even in relatively "bad" parts the nucleation field is negative, the conclusions would be more reliable. It is a pity that this has not been done yet. A most interesting experiment would, of course, be the comparison of a cube to a whisker tip.

In *hard* materials, where the demagnetization field is negligible, surface roughness cannot be expected to be very important. In these materials, on the other hand, crystalline imperfections (of which a whisker is practically free) can be very important factors. Dislocations, in particular, involve mechanical strains which change locally the magnetocrystalline anisotropy through magnetostriction effect. In Sec. III, a somewhat idealized model of this effect is discussed, but again with a previous complete saturation assumed. However, in this case too, one can devise relative directions for the dislocation axis and the ellipsoid axis, so that it takes an infinite field to saturate the material.[25] In practice, this means again the necessity of using rather large fields. The author tends to interpret the results of Kooy and Enz[16] on this basis of incomplete saturation. They observed the domains nucleate always at a well-defined spot in a single crystal of $BaFe_{12}O_{19}$. In each sample, there were two to four such nucleation centers, but only one of them was active in each single experiment. Each of these centers had a characteristic nucleation field ranging from $+500$ to $+2000$ oe. Usually, there was an increased or decreased local transparency to light at these spots,[26] thus indicating certain imperfections. It is possible, of course, that domains nucleated at these centers in a way discussed

[24] R. W. De Blois, J. Appl. Phys. **32**, 1561 (1961).

[25] W. F. Brown, Jr. (private communication).

[26] U. Enz (private communication).

in Sec. III. But, because the domains did not always nucleate at the centers having the largest nucleation field, in spite of the large difference in these fields, it seems to me that the field used was barely large enough to saturate the imperfect spots, and at different cycles somewhat different "saturating" fields were used. The 2000-oe field nucleation pertained thus to samples not previously saturated at the point of "nucleation," whereas only the 500-oe field nucleation probably pertained to a previously saturated sample, if this was reached at all. After all, they assumed the sample to be saturated only because they did not see any more domains, but we know from results of Shur *et al.*[19] that this is a rather poor indication. Repeating this experiment with the use of high fields can settle the problem of domain nucleation in *hard* materials. One could also, at the same time, determine the possible effect of surface roughness in hard materials by electropolishing, as De Blois did for whiskers, or by introducing a scratch, mechanically or by etching, and see whether this point can serve as a nucleation center. Theoretically, too, all the calculations were carried out for an easy axis (or hard axis, for a ferroxplama-type material,[27,28] which is essentially a similar *mathematical* problem) which coincided with the ellipsoid axis and the direction of the field, except for some rough estimation for an infinite cylinder inclined to the field[5,29] but not to an easy direction. It would be interesting to try to calculate the nucleation field for an easy axis inclined to a perfect-ellipsoid axis, even with the assumption of previous saturation (which is doubtful in this case) to see what difference it makes.

In spite of all the foregoing, it is still possible that, because of the angles between easy-direction axis and field, or because of crystalline imperfections, or because of surface roughness, saturation is unattainable in principle in a finite field, at least for large enough particles, for which subdivision into domains is favorable energetically. After all, Shur, Shtoltz, and Margolina[19] could remove the domains by a large field only when 10- to 15-μ particles were studied, i.e., close to the classical "critical size." Fifty-micron particles were always polydomain with a positive nucleation field. This might be explained, as in Secs. III or IV by imperfections, but it might also mean that one cannot saturate a large enough particle. Such a point of view implies leaving out all linearized-nucleation problems and dealing with Brown's nonlinear equations.[18,30] It means much more tedious computation, but might be unavoidable if the nonlinearity is proved to be an essential feature for domain nucleation, i.e., if the other possibilities to resolve the Brown paradox fail, or if additional experimental results are accumulated. In view of the present state of theory and experiment discussed in the foregoing, the author does not regard this as a serious possibility. Therefore, in the following two parts, the effects of crystalline and surface imperfections are studied by the linearized theory, i.e., by assuming previously saturated samples.

Also connected with the problem of complete saturation is the question of whether I_s can be regarded at all as constant over the material. This constant is a function of temperature and is calculated as such[31] by averaging over an infinite body the contributions of spin waves excited by the temperature. To take this space average and consider it as independent of the coordinates for the other calculation can hardly be justified without a detailed study. It evidently is not true when the Curie temperature is approached, but even at room temperature it might be possible that, by assuming I_s a constant, the theory is affected and some justification seems necessary. A simple assumption, therefore, was tried, namely, that I_s is given by a standing spin wave of the form

$$I_s = I_0\{1+\epsilon\cos[\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}_0)]\}, \tag{6}$$

with ϵ assumed small (low temperature). If the magnetization vector $\mathbf{I}$ is assumed everywhere to have the magnitude I_s and if one follows Brown's derivation[3,32] of his equations with this I_s to a first order in ϵ, it turns out that this is equivalent[33] to substituting (6) in the final Brown equations. An important implication is that the state in which $\mathbf{I}$ is everywhere directed along the z axis is a solution of the equations for an ellipsoid. Although this is not saturation in the usual sense, one can regard it as such and work out the nucleation problem with the linearized equations. At least for an infinite slab, assuming one-dimensional dependence, this does not give any appreciable change in the nucleation field with respect to the case $\epsilon=0$.

III. CRYSTAL IMPERFECTIONS

Since the Brown paradox comes mostly from the large values of K, it seems reasonable to assume that local reduction in K will enable domains to nucleate. This has been suggested by Rathenau *et al.*[17] as a possible explanation to the paradox, but without any calculations. As a first approach to this problem, Aharoni[5,34] studied a one-dimensional model in which $K=0$ in a certain part of an infinite crystal. This did not make the nucleation as easy as one might expect from the fact that, at the region where $K=0$, there is no barrier at all. The barrier is provided by the exchange interaction on the boundary of this region, with the outside spins which are held by the magnetocrystalline anisotropy. The result was that the $2K/I_s$ term is reduced by one order of magnitude at most. He has

[27] M. W. Muller, Phys. Rev. **122**, 1485 (1961).
[28] M. W. Muller and A. Wehlau, J. Appl. Phys. **32**, 2448 (1961).
[29] S. Shtrikman and D. Treves, J. phys. radium **20**, 286 (1959).
[30] W. F. Brown, Jr., J. Appl. Phys. **30**, 62S (1959).
[31] F. J. Dyson, Phys. Rev. **102**, 1217 (1956).
[32] W. F. Brown, Jr., Am. J. Phys. **28**, 542 (1960).
[33] The exchange energy is taken as[1] proportional to $\mathbf{J}\cdot\nabla^2\mathbf{J}$ rather than the expression used by Brown.
[34] A. Aharoni, Phys. Rev. **119**, 127 (1960).

later shown that a linear decrease of K into the value zero affects considerably the coercive force but not the nucleation field,[35] and that the results are practically the same if more than one such "defect" is present.[36] According to Sec. I, one order of magnitude will not help at all in the case of elongated particles of *soft* materials, whereas for spherical particles of these materials there is no paradox. In the case of *hard* materials, if they are almost spherical, it is seen from (3) that one order-of-magnitude reduction in the first term is sufficient to make H_n positive. It is not so in elongated particles, but no data are available for measurements on elongated particles of hard materials. It seems, therefore, that for hard materials the resolution of the Brown paradox lies mainly in crystal imperfections, most probably dislocations at which K is reduced by the mechanical strains through magnetostriction. To support this assumption, it is desirable to measure nucleation fields of elongated particles and to extend the theoretical treatment to a more physical model of the dislocation, which can essentially be an adaptation of Brown's approach to saturation study.[37] Until this is done, it is interesting to see whether the one-dimensional calculations hold for three dimensions. The region with $K=0$ is, therefore, taken in the following to be an infinite cylinder, rather than a slab.

More specifically, a ferromagnetic material infinite in all directions is assumed which has a uniaxial, magnetocrystalline anisotropy $\bar{K}(r)$, where in cylindrical coordinates

$$\begin{aligned}\bar{K}(r)&=0, \quad \text{for} \quad r\leqslant R\\ &=K, \quad \text{for} \quad r\geqslant R.\end{aligned} \tag{7}$$

The external field is applied along the z axis, which is assumed to coincide with a direction of easy magnetization.

The differential equations for the nucleation problem are identical with those for an infinite cylinder[7] for $r\leqslant R$, whereas for $r\geqslant R$ the only change that should be made in them is to replace h by $h+g$, where

$$g=K/(\pi I_s^2). \tag{8}$$

One can, therefore, use the analytic solution[7,38] for these equations and write

$$A_r-A_\varphi=\sum_{n=1}^{3} a_n J_{m-1}(i\mu_n t), \tag{9a}$$

$$U_t=2iS\sum_{n=2}^{3}\frac{a_n\mu_n}{k^2-\mu_n^2}J_m(i\mu_n t), \tag{9b}$$

$$A_r+A_\varphi=\sum_{n=1}^{3}(-1)^{\frac{1}{2}n(n-1)}a_n J_{m+1}(i\mu_n t), \tag{9c}$$

for $t\leqslant 1$, and, similarly,

$$A_r-A_\varphi=\sum_{n=1}^{3} b_n H_{m-1}^{(1)}(i\alpha_n t), \tag{10a}$$

$$U_t=2iS\sum_{n=2}^{3}\frac{b_n\alpha_n}{k^2-\alpha_n^2}H_m^{(1)}(i\alpha_n t), \tag{10b}$$

$$A_r+A_\varphi=\sum_{n=1}^{3}(-1)^{\frac{1}{2}n(n-1)}b_n H_{m+1}^{(1)}(i\alpha_n t), \tag{10c}$$

for $t\geqslant 1$. Here, the notations are the same as in Aharoni and Shtrikman.[7] In particular,

$$t=r/R, \quad h=H/(2\pi I_s), \quad S=RI_sA^{-\frac{1}{2}}, \tag{11a}$$

$$\mu_1=(k^2+\pi S^2 h)^{\frac{1}{2}}, \tag{11b}$$

$$\mu_{2,3}=\left[k^2+\pi S^2(\tfrac{1}{2}h+1)\mp S\{2\pi k^2+\pi^2S^2(\tfrac{1}{2}h+1)^2\}^{\frac{1}{2}}\right]^{\frac{1}{2}}. \tag{11c}$$

A is the exchange constant and the α's are same as the μ's with $h+g$ substituted for h. The changeover from the Bessel function of the first kind J_m, in the region $t\leqslant 1$ to the Bessel functions of the third kind $H_m^{(1)}$, in the region $t\geqslant 1$ is necessary to insure zeros at infinity, since in this instance the material extends to infinity.

The boundary conditions in this case are that the three functions A_r, A_φ, and U_t will be continuous and have a continuous derivative at $t=1$. This implies six linear homogenous equations for the six coefficients, a_n, b_n of Eqs. (9) and (10). In order that a nonvanishing solution exist, the determinant of the coefficients of a_n and b_n must be zero. This condition implies a rather complicated relation between h, g, k, and S. For each value of the radius, the parameter k should be chosen so that the said relation implies the least-negative value for h, and this computation should, in principle, be carried out for every non-negative integral value[7] of m.

Only for $m=0$ is the computation rather simple, since the determinant breaks down into a product of two determinants, one of which involves the coefficients of A_φ only and implies

$$\frac{i\mu_1 J_0(i\mu_1)}{J_1(i\mu_1)}=\frac{i\alpha_1 H_0^{(1)}(i\alpha_1)}{H_1^{(1)}(i\alpha_1)}. \tag{12}$$

The other one need not be calculated, since it reduces to (12) if the self-magnetostatic energy is neglected, which means that it will give more-negative eigenvalues h than (12). One can therefore, conclude that $A_r=U_t=0$ for $m=0$, and this is the curling mode.[7] The values of the nucleation field as a function of the cylinder radius R were calculated numerically from (12) and are plotted in Fig. 2, labeled "curling."

For $m\neq 0$, if the self-magnetostatic energy is neglected, one obtains a fourth-order determinant which can be written as a product of two second-order determinants, yielding relations somewhat similar to

[35] C. Abraham and A. Aharoni, Phys. Rev. **120**, 1576 (1960).
[36] A. Aharoni, J. Appl. Phys. **32**, 245S (1961).
[37] W. F. Brown, Jr., Phys. Rev. **58**, 736 (1940).
[38] A. Aharoni, E. H. Frei, and S. Shtrikman, J. Appl. Phys. **30**, 1956 (1959).

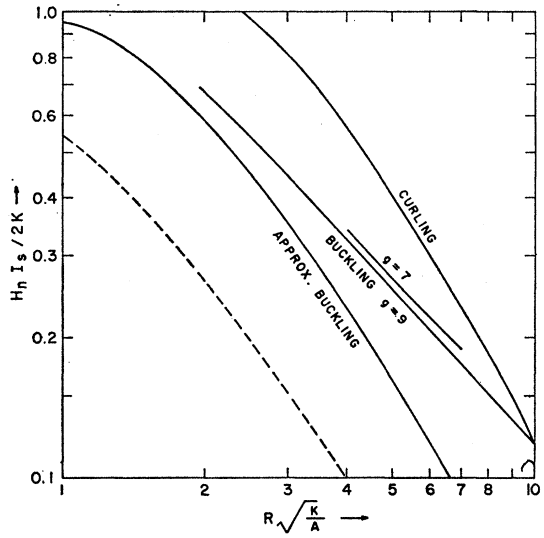

FIG. 2. Theoretical nucleation field H_n for an infinite, ferromagnetic material which has a defective region with $K=0$, in the form of an circular infinite cylinder of radius R (full curves) or of an infinite slab of width $2R$ (dashed curve). The "approximate buckling" is obtained for the cylinder by neglecting the self-magnetostatic energy in the buckling mode. Only the exact buckling mode depends on the additional parameter $g=K/(\pi I_s^2)$. Here, K is the magnetocrystalline anisotropy, A the exchange constant, I_s the saturation magnetization. The nucleation field is plotted in terms of $2K/I_s$ which is the nucleation field for perfect, infinite material.

(12). Computations showed that, for each of these branches, the values of H_n becomes more negative as m was increased and that they all were more negative, than for the curling, except for one of the branches of $m=1$. This branch is plotted in Fig. 2 labeled "approx. buckling." For the case $m=1$ only, the complete computation from the six-order determinant therefore, was carried out, with the values from the aforesaid branch used as starting values, carrying out the maximum with respect to k. The final results are plotted in Fig. 2 under the label of "buckling" for 2 values of the parameter g defined in (8), which correspond to barium ferrite and manganese bismuth, respectively.

For comparison, the results[34] for the nucleation in the case where the "defective" region is an infinite slab of width $2R$, are also plotted in Fig. 2. It can be seen that the slab one-dimensional results yield considerably easier nucleation than in the case of the infinite cylinder, which is certainly a better approximation to a dislocation. The form of the curves, however, is not much different, especially for hard materials, and the final conclusions with respect to the one-dimensional case can still be adopted. In particular, if one subtracts the demagnetizing field as in (3), the nucleation field for spherical particles of hard materials becomes positive when dislocations are present.

To justify the subtraction of the demagnetizing field consider an infinite plate with the region at which $K=0$ a circular *finite* cylinder in this slab. It is readily seen that the treatment of an infinite material in this section holds, if instead of the z dependence $\cos kp$ one writes a trigonometric function that will fulfill the additional boundary conditions $\partial\alpha_\varphi/\partial p=0$ at $p=\pm a$, say. That means $\cos(n\pi p/a)$ or $\sin\{(n+\frac{1}{2})\pi p/a\}$, with n an integer. One can, therefore, use the same transcendental equation for the eigenvalue h merely by substituting the internal field $H-4\pi I_s$ for H and by taking $k=N\pi/a$, where N takes integral or half-integral values.

For the curling mode of Eq. (12) and Fig. 2, $k=0$, i.e., $N=0$, and no change need be made except to subtract the demagnetizing field. For the buckling mode, on the other hand, k takes an optimum value, which might not be an allowed value for N. The buckling graph in this case is in principle higher than in Fig. 2. However, the minimum with respect to k is rather flat, and, if the width of the slab is large compared to R, the radius of the "defective" region (as is practically always the case), the allowed values for k are closely spaced and the correction in Fig. 2 is negligibly small. One can, therefore, say that Fig. 2 gives the correct result for a material in the form of a slab if $H_n+4\pi I_s$ is substituted for H_n.

Taking $BaFe_{12}O_{19}$ as an illustration, one obtains from (2) the nucleation field of $+500$ oe observed in certain cases for a thin-plate single crystal[16] if the $2K/I_s$ term is multiplied by about $\frac{1}{4}$. According to Fig. 2, this corresponds to a defective region having a radius of about 350 A, which is not unreasonable for a dislocation.

It can be concluded that for hard materials the solution to the Brown paradox lies mainly in dislocations, and that these are important only if the particles are almost spherical in shape. If this assumption is true, one can expect very elongated particles of reasonably good crystals to have a coercive force of the same order or of only one order of magnitude smaller than $2K/I_s$, even if the particles are rather large. This means that there is an enormous increase in the coercive force of MnBi for example, where the reported coercive force for 100-μ particles is only 600 oe and should be at least an order of magnitude larger were the particles elongated. It would consequently be very interesting to test this conclusion experimentally. Another possible experiment is to try to observe the domains nucleating on a cobalt whisker (provided a large enough field is used to saturate it). This would also settle the point of a possible surface-roughness effect in hard materials. One can also determine by conventional experimental techniques if the nucleation centers in the experiment of Kooy and Enz are dislocations.

IV. SURFACE ROUGHNESS

Nobody has ever tried a detailed calculation of nucleation modes in anything but a perfect ellipsoid (except for the rotation in unison, which is not actually an eigenfunction, for which mode it has been shown[39]

[39] W. F. Brown, Jr., and A. H. Morrish, Phys. Rev. **105**, 1198 (1957).

that any particle is equivalent to an ellipsoid). It is mainly because the internal field is not homogeneous once the form is anything but an ellipsoid, and mathematical difficulties arise. Nevertheless, after the experiment of De Blois[24] has proved that surface roughness is at least an important factor in determining the nucleation field, one cannot avoid studying the nucleation modes theoretically. In accordance with the discussion in Sec. II, previous saturation is assumed here, so that a model is chosen which does not imply an infinite demagnetizing field anywhere inside the material. Another assumption is made to make the mathematics less formidable, namely, to take a model which has a cylindrical symmetry, so that one has to deal with two dimensions instead of three. Moreover, in the case of cylindrical symmetry, there is a mode for which the equations for the two components of the magnetization vector are separated and can be solved individually.

To prove this, consider a ferromagnetic particle of any shape that has a cylindrical symmetry about the z axis, which is also assumed to be a direction of an easy magnetization, and the direction of the external field H_0. Suppose that it is possible to saturate it in a finite field, and let the potential of its surface charges in the saturated state be denoted by V_0. In cylindrical coordinates, this V_0 will always be a function of r and z only, since the surface and, therefore, the charges themselves do not depend on φ. Assuming now a small variation of the magnetization from the saturated state with direction cosines α_φ and α_r and writing Brown's equation[30] in the cylindrical coordinates, it is seen that the equation in α_φ contains only derivatives with respect to φ of α_r and the potential, whereas in the other equations α_φ is contained only as its derivative with respect to φ. Therefore, if one is interested in a mode that has a cylindrical symmetry, i.e., if α_r and α_φ are assumed to be independent of φ, the equation for α_φ turns out to be

$$2A(\nabla^2-r^{-2})\alpha_\varphi=I_s(H_0+2K/I_s-\partial V_0/\partial z)\alpha_\varphi, \quad (13)$$

with the boundary conditions

$$\partial\alpha_\varphi/\partial n=0. \quad (14)$$

Here A is the exchange constant[1] ($=\frac{1}{2}C$ of Brown[30]) and n is the normal to the boundary surface. The equation for α_r, which should be solved in connection with the Poisson equation for the potential of surface and volume charges of the *change* in magnetization, might also give another mode for nucleation but is disregarded here.

Equation (13) is thus the general equation for the curling in cylindrically symmetric particles. For the special case in which the particle is an ellipsoid of revolution, $\partial V_0/\partial z=N$, the demagnetizing factor and the coefficient of α_φ on the right-hand side is a constant. This yields then a solution in terms of tabulated functions.[5] In other cases, $\partial V_0/\partial z$ is a function of space, which is known in principle, since it is a solution of Laplace equation with the appropriate boundary conditions.

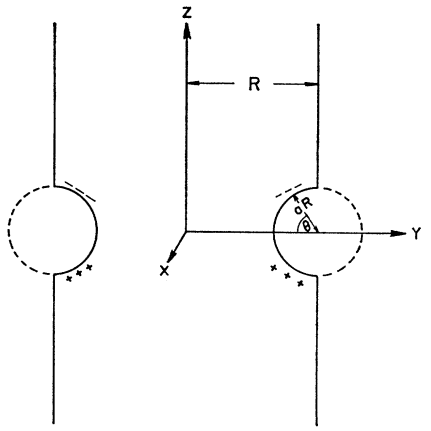

FIG. 3. Model used for a theoretical study of a scratch on the surface of an circular infinite cylinder.

The model of scratch on the surface chosen here for detailed calculations is shown in Fig. 3. It consists of an circular infinite cylinder with a semitoroid removed. When the particle is saturated in the z direction, V_0 caused by the surface charges on the semitoroid is given by

$$V_0(r,z,R,a)$$
$$=-R^2aI_s\int_0^{2\pi}\int_{-\pi/2}^{\pi/2}\sin\theta(1-a\cos\theta)\{r^2+z^2+R^2(1+a^2)$$
$$-2R^2a\cos\theta-2rR(1-a\cos\theta)\cos\varphi$$
$$-2zRa\sin\theta\}^{-\frac{1}{2}}d\theta d\varphi. \quad (15)$$

Since one is interested only in a $a\ll1$, this expression is expanded in a Taylor series in a. Now both V_0 and $\partial V_0/\partial a$ vanish for $a=0$, while $\partial^2V_0/\partial a^2$ gives a finite expression for $a=0$, which can be expressed in terms of complete elliptic integrals of the first and second kind. Multiplied by $\frac{1}{2}a^2$, this expression is, therefore, taken as an approximation for V_0. It turns out that both V_0 and its derivatives with respect to r and z are finite everywhere within the materials (there is a logarithmic infinity at $r=R$, $z=0$, but this is outside the material and, therefore, is disregarded), which justifies the choice of this particular model from the point of view of convenience. Whether the choice is justified for the physical case is quite a different question. It seems, nevertheless, a rather good first approach and will certainly justify itself if the calculated nucleation as a function of a (and R) will check with a De Blois-type measurement[24] of the nucleation field as a function of scratch dimensions on a whisker. It should be noted one is fully justified to assume just one scratch and that the cylinder is infinite, since, in the experimental

arrangement of point-to-point measurement, the rest of the material is held saturated so that it can well be assumed to extend to infinity and have no other surface roughness, which have no effect on the measurement.

To solve (13), α_φ is expanded in a Dini series[40]

$$\alpha_\varphi = \sum_{n=1}^{\infty} f_n(z) J_1(q_n r/R), \tag{16}$$

where q_n are the zeros of the derivative of J_1, which takes care of (14) for $|z| > aR$, and f_n are functions to be determined. Substituting (16) in (13) and expanding again in Dini series the following expression

$$\left(\frac{\partial V_0}{\partial z}\right) J_1(q_n r/R) = \sum_{m=1}^{\infty} F_m(z) J_1(q_m r/R), \tag{17}$$

the r dependence can be eliminated and one ends up with a set of linear equations for f_n, namely,

$$\frac{d^2 f_n}{dz^2} = \sum_{m=1}^{\infty} g_{nm}(z) f_m(z), \quad n = 1, 2, 3 \cdots. \tag{18}$$

Here, g_{mn} can be calculated from (17) by the usual way of calculating the coefficients of Dini series,[40] i.e., by certain integrations involving $\partial V_0/\partial z$. All the g_{mn} are, therefore, known in principle once $\partial V_0/\partial z$ is known, except that they also contain (linearly) the unknown eigenvalue H_0.

The general solution of the set of linear equations (18) can always be written as a linear combination

$$f_n(z) = A_n f_n^{(1)}(z) + B_n f_n^{(2)}(z), \tag{19}$$

where, for $z = 0$,

$$f_n^{(1)}(0) = 0, \quad (d f_n^{(1)}/dz)_{z=0} = 1, \tag{20a}$$

$$f_n^{(2)}(0) = 1, \quad (d f_n^{(2)}/dz)_{z=0} = 0. \tag{20b}$$

Let $2N$ equidistant points be chosen in the region $0 \leq z \leq aR$ and let the summation in (18) and (16) be stopped at N. For any value of H_0, the values of the $2N$ functions $f_n^{(1)}$, $f_n^{(2)}$ at the $2N$ discrete points can be evaluated from (18) and (20) by the Runge-Kutta method, yielding a *unique* solution. Substitution of (16) and (19) into (14) then implies $2N$ relations between the constant A_n and B_n. If the determinant of their coefficient is computed as a function of the previously selected value of H_0, the zero of this determinant will determine the eigenvalues H_0 of the problem. The least-negative eigenvalue H_0 is then the nucleation field, and the computation can be repeated for various values of a.

Although this scheme is straightforward in principle, it is rather cumbersome, especially as N becomes large. C. Abraham and I plan to put it on the electronic computer of this Institute in the near future in order to try to relate the results to De Blois-type measurements, as mentioned in the foregoing. The same numerical approach will also be used to study the effects of cavities, which is mathematically exactly the same problem.

Cavities or nonmagnetic precipitates are known[1] to be important factors in determining the *shape* of domains around them. They might also change the nucleation field by their demagnetizing field to the same extent that surface imperfections do it. This has actually been suggested, without detailed calculations, as a solution of the Brown paradox.[41] Theoretically, the effect of cavities is usually calculated by assuming the wall to be already there and a comparison of energies (for a recent publication see Nix and Huggins[42]), so that it yields no information about nucleation fields. For the rotation in unison mode, it was shown[39] that, for a prolate spheroidal particle containing a spheroidal cavity, the nucleation field is sometimes smaller and sometimes larger than when the cavity is absent, depending on relative directions and eccentricities. In particular, for a cavity of the shape of a sphere centrally located within a spherical particle (or actually, whenever the polar axes of the ellipsoids are parallel and the eccentricities are equal), the cavity implies no change in the nucleation field for the rotation in unison mode.[39] For this simple case, the nucleation by the curling mode can be easily studied analytically by the perturbation theory, and it is shown in the following that the nucleation becomes somewhat *more difficult* because of the presence of the cavity. Other cases which have cylindrical symmetry can be studied by means of a technique similar to the one described using spheroidal wave functions instead of Bessel functions for ellipsoidal particles.

In the case of a spherical cavity centrally located in a spherical particle, V_0 consists of $(4\pi/3) I_s z$ due to the surface charges on the outer sphere and $-(4\pi/3) \times a^3 R^3 \rho^{-2} I_s \cos\theta$, due to surface charges on the inner sphere of radius aR. Here ρ and θ are the spherical coordinates and R is the radius of the particle. Using the notations

$$h = H_0/(2\pi I_s) + K/(\pi I_s^2) - \tfrac{2}{3}, \qquad S = R I_s A^{-\frac{1}{2}}, \quad T = \rho/R \tag{21}$$

and writing (13) in spherical coordinates, the differential equation for the curling mode becomes

$$\left[\frac{\partial^2}{\partial T^2} + \frac{2}{T}\frac{\partial}{\partial T} + \frac{1}{T^2}\frac{\partial^2}{\partial \theta^2} + \frac{\cos\theta}{T^2 \sin\theta}\frac{\partial}{\partial \theta} - \frac{1}{T^2 \sin\theta} - \pi S^2 h + \tfrac{2}{3}\pi S^2 a^3 \frac{2\cos^2\theta - \sin^2\theta}{T^3}\right] \alpha_\varphi = 0, \tag{22}$$

[40] G. N. Watson, *A Treatise on the Theory of Bessel Functions* (Cambridge University Press, New York, 1958), p. 577.

[41] L. F. Bates and D. H. Martin, Proc. Phys. Soc. (London), **69**, 145 (1956).

[42] W. D. Nix and R. A. Huggins, Phys. Rev. **121**, 1038 (1961).

with the boundary conditions

$$(\partial\alpha_\varphi/\partial T)_{T=a}=(\partial\alpha_\varphi/\partial T)_{T=1}=0. \tag{23}$$

Consider the last term (with a^3) as a perturbation. The unperturbed equation can be solved by separation of variables yielding

$$\alpha_\varphi{}^{(0)}=T^{-\frac{1}{2}}\{AJ_{n+\frac{1}{2}}(\mu T)+BJ_{n-\frac{1}{2}}(\mu T)\}P_n{}^1(\cos\theta). \tag{24a}$$

Here,

$$\mu^2=-\pi S^2h_0 \tag{24b}$$

is the unperturbed eigenvalue, and n is a positive integer, to insure regularity at $\theta=0$. By substituting the boundary conditions (23) into the solution (24), one obtains 2 linear equations in A and B, which determine A/B and yield a transcendental equation for μ. The smallest value of μ is presumably obtained for $n=1$, in which case the transcendental equation is

$$\mu(1-a)(2+\mu^2a)+\{\mu^2(1-a)^2-2-\tfrac{1}{2}\mu^4a^2\}\times\tan\{\mu(1-a)\}=0, \tag{25}$$

and

$$\frac{A}{B}=\frac{\mu^2\cos\mu-2\mu\sin\mu-2\cos\mu}{\mu^2\sin\mu+2\mu\cos\mu-2\sin\mu}. \tag{26}$$

Using now first-order perturbation theory in (22), one can write, after carrying out the integration with respect to the angles, for the change in the eigenvalue:

$$h_n-h_0=-\frac{4a^3\mu^3}{15}\cdot\frac{\int_{\mu a}^{\mu}\left\{\left(\frac{A}{B}\right)(x^{-\frac{5}{2}}\sin x-x^{-\frac{3}{2}}\cos x)-x^{-\frac{3}{2}}\sin x-x^{-\frac{5}{2}}\cos x\right\}^2dx}{\int_{\mu a}^{\mu}\left\{\left(\frac{A}{B}\right)(x^{-1}\sin x-\cos x)-\sin x-x^{-1}\cos x\right\}^2dx} \tag{27}$$

for $x=\mu T$. Carrying the integrations, using A/B from (26) and using (25) to shorten the expressions, one obtains finally for the nucleation field

$$h_n=-\frac{\mu^2}{\pi S^2}-\frac{\mu^2a^3}{15}(1+a)\frac{4\mu^2(1+a^2)-4+\mu^4a^2}{2(\mu^2-2)(1+a+a^2)+\mu^2a^3[\mu^2(\frac{1}{7}\mu^2a+1)+2(1+a)]}. \tag{28}$$

For $a=0$, the nucleation field is $-\mu_0{}^2/(\pi S^2)$, with μ_0 the root of

$$(\mu_0{}^2-2)\tan\mu_0+2\mu_0=0 \tag{29}$$

yielding $\mu_0=2.08$. For small values of a, Eq. (25) yields $\mu=\mu_0$. Actually, for $a\leq10^{-2}$, μ equals μ_0 to 4 significant digits. For small values of a, therefore, it is seen from (28) that

$$|h_n|>\mu^2/(\pi S^2)=\mu_0{}^2/(\pi S^2),$$

and the nucleation by the curling mode is more difficult for a hollow than for a rigid sphere in spite of the demagnetizing field. The difference, however, is extremely small, being of the order of a^3.

V. OTHER POSSIBILITIES

In this section, we discuss briefly some possible, more-fundamental flaws in the micromagnetics approach. Very difficult problems are involved and no attempt is made actually to solve them. It is not felt, however, that they can be important to the Brown paradox in its present state, for reasons discussed under each section.

Brown[30,32] has already mentioned the difficulty in writing the self-magnetostatic energy, since the internal field cannot be defined in places where the magnetization changes rapidly. When the change is gradual (it should actually be linear over small distances) and the crystal is cubic, it can be shown that the internal field is $\mathbf{H}'+(4\pi/3)\mathbf{I}$ in which the last term does not contribute to the energy since $\mathbf{I}\cdot\mathbf{I}$ is constant. If inside a domain wall the change is too rapid, the use of this Lorentz-field approximation might be doubtful.[30,32] (The second complication mentioned by Brown[30] is of quite different nature in principle. It is just the difficulty of visualization, but the problem is well defined mathematically with the Poisson equation.) A much more difficult situation arises when one considers under the same basis the term for the exchange energy. Therefore, this is considered here first.

A. The Exchange Energy

This energy is of quantum-mechanical nature. For obtaining the Brown equations, this energy is used[3] in a classical-type expression which is obtained[1] by taking the first term of an expansion in Taylor series, assuming neighboring spins to be almost parallel. This argument certainly fails if the spatial change in orientation of magnetization is rather rapid. In particular, it definitely excludes from the calculation functions which do not have a continuous first derivative (at least). This might be a rather severe limitation.

It has been argued[1] that the Taylor-series expansion is justified because the Bloch wall extends over many atoms such that the change in magnetization is quite gradual. It is not a very convincing argument, since even an over-all gradual change can be rather abrupt

in certain parts of it, especially as improved techniques reveal[43] a rather complicated structure for a domain wall. Also, this assumption, when used to estimate the wall thickness,[1] actually gives an *infinite* thickness for simple models, a difficulty overcome by a rather doubtful technique.[1] But even if it were true that smooth functions describe the stable state after walls have settled, it could still be argued that when domains *start* to nucleate they can do it through sectionally smooth functions which possibly reduce the barrier for nucleation. It is true that such an argument does not affect the original statement of the Brown paradox,[2] since all one needs there is the existence of an energy which opposes a change from the saturation magnetization, its magnitude being neglected anyway. For the treatment of crystal imperfections (Sec. III), this makes a lot of difference, since at the region where $K=0$ magnetization is held only by the exchange interaction with the outer part, mathematically expressed as the boundary condition that the magnetization should be smooth on the boundary.

Dreyfus remarked in the discussion to Brown's Grenoble paper[44] that the variational calculation might exclude the domain walls because it deals with functions having a continuous derivative. Brown answered[44] that a wall according to Landau and Lifshitz *is* such a function and that the discontinuities imply an infinite exchange energy. This really means going in circles, since at the discontinuities the expansion in a Taylor series is not allowed and one cannot use the expression which implies infinity for the energy. The quantum-mechanical original expression will *not* lead to infinities, or even its approximation as Eq. (2.1.5) of Kittel.[1]

Nevertheless, it seems that discontinuous functions can be excluded, because on a microscopic scale it means certain pairs of spins have an energy larger by orders of magnitude than other pairs, and this does not seem a reasonable distribution for an energy minimum. The continuum approximation is, therefore, most probably justified, except for a possible discontinuity in the derivative. In this case, neighboring spins at the discontinuity are parallel and there is no loss of energy. It seems to me, although I cannot prove it, that appropriate *smooth* functions can always lower the energy of sectionally smooth ones. Yet sectionally smooth functions should definitely be allowed modes, when one needs an estimation and not the lowest-eigenvalue mode.

The possibility of using sectionally smooth functions can find important implications in cases when the calculation of the exact modes is complicated and of no special interest. For example, an estimation can be readily given for the nucleation field in an infinite cylinder of *any* cross section. Let R be the radius of the largest circle that can be drawn inside this cross section. Let the magnetization be held in the z direction outside the circular cylinder, and be changed by the curling mode inside it. One can then use the solution for a unidirectional cylinder[38] (the boundary conditions thus insuring continuity of the *function* but not of the derivative), and get for the nucleation field in the inequality (2) with[38] $k=4.67$. Together with (1), this implies (3) for the large enough radius R without any detailed calculation of the actual-nucleation mode. It is thus proved than an elongated particle of any cross section, provided the latter is large enough, should have a nucleation field that equals $2K/I_s$ to a good approximation. Even for a small cross section, the estimation is not too bad and it can explain why Eq. (2) was found in a rather good agreement with experiment for fine, elongated particles, which yet showed dependence on packing. The actual mode evidently depends on packing, but the nucleation field is at least easier than given by (2). Preliminary study of the nucleation field in a *rectangular* cylinder shows that the approximation discussed here holds and that, at least in this case, the actual smooth modes are easier than this unsmooth one.

Another implication is in the interpretation of the experiment of De Blois and Bean.[9] They have subtracted from the measured nucleation field a demagnetizing field of a prolate ellipsoid whose major axis was $2z_0$, i.e., the length of the coil, and whose minor axis was the average diameter of the whisker. Thus, for a reversing field of 496 oe (after subtracting the bucking field), they took the nucleation field as 483 oe. To show that this correction is not necessary, consider a magnetizing coil to apply a field over a distance $2z_0$ of a circular, infinite cylinder, and let the rest of the cylinder be held magnetized to saturation. Again, if one looks for a continuous function, but allows a discontinuity in the derivative (with respect to z), the solution of Aharoni and Shtrikman[7] can be used with the additional boundary conditions

$$\alpha_r(z_0)=\alpha_r(-z_0)=\alpha_\varphi(z_0)=\alpha_\varphi(-z_0)=0. \qquad (30)$$

These conditions determine k and p_0 in (11) of Aharoni and Shtrikman,[7] which are not arbitrary any more. The smallest value for k is thus $\frac{1}{2}\pi R/z_0$, which yields for the curling mode

$$|H_n|=2K/I_s+\pi I_s\{2.16(R/R_0)^{-2}+\tfrac{1}{2}\pi(z_0/R_0)^{-2}\}. \qquad (31)$$

Here, the last 2 terms are definitely negligible, so that the "demagnetizing field" does not come in. The correction was, therefore, left out in De Blois' later experiments.[24]

Even when the Taylor expansion of the exchange energy is allowed, the expression used in the derivation of Brown's equations[3] is based[1] on the assumption of cubic symmetry. Herring and Kittel[45] remarked that for hexagonal crystals the exchange constant A should

[43] S. Methfessel, S. Middelhoek, and H. Thomas, IBM J. Research Develop. **4**, 96 (1960).

[44] W. F. Brown, Jr., J. phys. radium **20**, 101 (1959).

[45] C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).

be replaced by two constants. Again, this would not affect the derivation[2] of (1) but might make quite a difference in more-detailed calculations for hard materials which have usually very elongated unit cells. However, Brown[25] showed that one obtains just one constant for the ideal, close-packed, hexagonal lattice.

One can also question the basic, quantum-mechanical expression which is based on the assumption that the 3d electrons with unpaired spins are fixed to individual atoms, a model which can hardly account for the nonintegral, effective number of electrons per atom which contribute to the magnetization.[1] One of the arguments in favor of this model[1] is that it accounts for domain-wall energy, but, if the other possibilities discussed in this paper do not resolve the Brown paradox satisfactorily, this argument would fail since the model would not account for the domain nucleation. The model gives a temperature dependence of I_s which is in agreement with experiment in contradistinction to the Stoner's school model of collective electrons, but even this has been questioned in certain cases.[46] It seems, therefore, that the models should be combined, but the mathematics involved does not raise any hope of even an estimation on how a contribution from collective-electron ferromagnetism can affect the domain nucleations.

B. Boundary Conditions

In Brown's equations, the boundary conditions are[18] $\partial \mathbf{I}/\partial n=0$. They should probably be replaced by[25] $\partial \mathbf{I}/\partial n+\lambda \mathbf{I}=0$, where λ is a tensor of some kind, which originates from surface anisotropy. In the first place, in the Taylor expression of the exchange energy,[1] one is left with second derivatives only because the first derivatives cancel out due to symmetric contributions from the neighbors. This is true for an interior spin, but is no longer true on the surface.[47] However, Brown[25] derived the same expression by summing over a single line of spins, then over parallel lines. There is again some doubt about the end, but it does not look as bad in the Kittel[1] derivation. There are also other possible contributions to surface anisotropy[47] the most obvious of which is an antiferromagnetic-oxide layer on the surface,[38,48] and the question certainly needs looking into in detail. It should be remarked, though, that at least for infinite cylinders with large-enough radii the boundary conditions cannot change the state of the Brown paradox as discussed here. Since an analytic solution of the equations is known,[7] different boundary conditions can easily be substituted in the infinite-cylinder case, and it is readily seen that they affect only a term which is negligible for large radii, leaving the disagreement with experiment as it is.

ACKNOWLEDGMENTS

The help of E. Neeman and C. Abraham in the numerical computation of Fig. 2 is gratefully acknowledged.

[46] E. P. Wohlfarth and J. F. Cornwell, *Berichte der Arbeitsgemeinschaft Ferromagnetismus* 1959 (Verlag Stahleisen G. m. b. H., Düsseldorf, Germany, 1960), p. 9.

[47] P. Pincus, Phys. Rev. **118**, 658 (1960); G. T. Rado and J. R. Weertman, J. Phys. Chem. Solids **11**, 315 (1959).

[48] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956); J. Appl. Phys. **29**, 454 (1958). See also F. J. Darnell, *ibid.* **32**, 186S (1961), J. H. Greiner, I. M. Croll, and M. Sulich, *ibid.* **32**, 188S (1961).