Modification of the saturated magnetization state

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The previous proof that a highly symmetric ferromagnetic particle cannot be saturated is extended to a case in which the symmetry is broken. The result is practically the same as that for the symmetric case, which must mean that a ferromagnetic particle can never be saturated, so that the magnetization reversal does not have to nucleate via the high energy barrier of a completely saturated sample.

I. INTRODUCTION

It has been known for a long time that in a sufficiently large ferromagnetic crystal, the energy of a domain structure is much smaller than that of the state of a uniform magnetization. The question, which was raised by Brown[1] in 1945, and which became known in the literature as the "Brown paradox," was how these domains can nucleate in a previously saturated sample, where a very high energy barrier must be overcome before reaching the lower-energy state. Many possibilities have been suggested during the years[2] to resolve this paradox, but none of them was quite satisfactory. It has even reached the point of suggesting[3] that micromagnetics should be discarded altogether, without offering any real alternative.

One of the suggested[4,5] possibilities for resolving this paradox was that ferromagnetic samples are never completely saturated, so that the theoretical high energy barrier is not encountered: Some small angles remain between the magnetization and the field, even when a very high field is applied, and these angles grow when the field is reduced. By the time a zero field is reached, the angles have already grown sufficiently to transform into the lower-energy domain structure, and there is no barrier. This idea, however, was only supported by some inconclusive experimental evidence, and by some unconvincing theoretical arguments.[2] It was, therefore, taken to be at most true for very special cases, and was never seriously considered as a general explanation. Nevertheless, it now turns out that it may be just that.

In a recent publication[6] it was shown that when certain approximations are removed from the theory, a small ferromagnetic particle is indeed never *completely* saturated. These approximations are inevitable in any practical calculation, and could be relaxed only for the study of very few atoms. Even for that case, the deviation from the saturated state is so small that it needed special tricks to be seen above the inherent inaccuracies of the computations. This means that all *other* micromagnetic calculations in the literature are still valid to a sufficiently high accuracy, and only the high energy barrier, which makes the Brown paradox, is affected.

However, that study[6] used the highly symmetric case of nine spins in a bcc configuration, and it cannot be clear *a priori* that this surprising result is not restricted to such a high symmetry. It thus seems quite necessary to see the effect of breaking such a symmetry, which is the purpose of the present paper. It will be shown here that the previous[6] result holds for this broken symmetry, and it may be safely concluded that real ferromagnetic particles cannot be saturated, with all the implications of this conclusion.

II. THEORY

The previous calculation[6] considered nine ferromagnetic ions (or spins) arranged in a bcc unit cell with a cube edge a. Spin No. 1 was the one at (0,0,0), and No. i, for $2 \leq i \leq 9$, was the spin at (k,l,m), where k, l, and m take the values ± 1, and where

$$i = 2 + 2k(k-1) + l(l-1) + \tfrac{1}{2}m(m-1). \qquad (1)$$

In the present study, spin No. 9 was moved from the position $-[1,1,1]$ to the position $-\lambda[1,1,1]$.

Since the exchange force has a very short range, the exchange constant C between the displaced spin $i = 9$ and its neighbors was taken as $10^{4(1-\lambda)}$ times the value of C between other spins. This means an energy term for the displaced spin which is 100 times larger than for other positions, in the case of an interstitial atom with $\lambda = \frac{1}{2}$, and 100 times smaller than in other positions for a spin drawn outside, to $\lambda = \frac{3}{2}$. In the magnetostatic energy term, λ just multiplies the distance vector for the displaced spin in an obvious way, and the anisotropy term is not affected by the displacement. On the whole, it does not seem necessary to repeat energy terms here, which can easily be modified from the expressions in Ref. 6.

The total energy of this system was minimized numerically with respect to the 18 directions of the nine spins, for various values of the cube edge a. As in the previous study,[6] two sets of numbers were used for the saturation magnetization M_s and the anisotropy constant K_1. One set was made of the physical parameters of iron,

$$M_s = 1700 \text{ emu/cm}^3, \quad K_1 = 4.7 \times 10^5 \text{ erg/cm}^3, \qquad (2)$$

and the other set was made of the physical parameters of nickel (used even though real nickel is fcc and not bcc),

$$M_s = 484 \text{ emu/cm}^3, \quad K_1 = -4.5 \times 10^4 \text{ erg/cm}^3. \tag{3}$$

In all the numerical computations, $C = 1.73 \times 10^{-6}$ erg/cm was used for the exchange constant.

III. RESULTS

In all the cases studied here, the resulting configuration was the kind of curling structure reported in the previous study.[6] However, here the whole configuration was tilted somewhat with respect to the z axis, in the case of Fe, Eq. (2), or the [111] direction (which we label the z' axis) in the case of Ni, Eq. (3). The tilted configuration is essentially the same as postulated[7] in the context of superparamagnetic spheres, and the tilt angle was almost independent of the lattice constant a. The obvious reason for the tilt is that the displaced spin structure creates an effective anisotropy whose easy axis is at some angle to that of the crystalline anisotropy. This tilt was relatively large, of the order of half a degree, and concealed the details of the much finer deviations from saturation of the curling structure itself.

Since the problem addressed here is the amount of deviations from a homogeneously magnetized state, the average direction of that magnetization makes no difference, and the tilt may be removed by rotating the whole structure by any constant angle. Therefore, the computed values of the direction cosines α_i, β_i, and γ_i of the magnetization vector were transformed according to

$$\begin{aligned} \alpha_i^* &= \alpha_i \cos\theta_\alpha - \beta_i \sin\theta_\alpha \sin\theta_\beta + \gamma_i \sin\theta_\alpha \cos\theta_\beta, \\ \beta_i^* &= \beta_i \cos\theta_\beta + \gamma_i \sin\theta_\beta, \\ \gamma_i^* &= -\alpha_i \sin\theta_\alpha - \beta_i \cos\theta_\alpha \sin\theta_\beta + \gamma_i \cos\theta_\alpha \cos\theta_\beta. \end{aligned} \tag{4}$$

with

$$\theta_\alpha = -\left\langle \arctan \frac{\alpha}{\sqrt{\beta^2 + \gamma^2}} \right\rangle, \tag{5}$$

$$\theta_\beta = -\langle \arctan(\beta/\gamma) \rangle, \tag{6}$$

where $\langle\ \rangle$ denotes the average over the nine spins. These parameters, θ_α and θ_β, are thus independent of the index i of Eq. (4), but are computed separately for each value of a, although their dependence on a turned out to be quite weak.

Figure 1 shows the largest (in absolute value) of the magnetization components α^* and β^* as a function of a, for several values of the parameter λ. This figure, as well as the ones that follow, ends at $a = 0.3$ nm, because smaller values of a can have no physical meaning. The plotted values are those obtained *after* rotating the whole structure by the transformation of Eqs. (4)–(6). This transformation was also used for the case $\lambda = 1$, where the angles of rotation θ_α and θ_β were of the order of 10^{-3} of a degree or smaller. Yet, even with these small rotations, there is considerably less wiggling in the present figure than in the previous[6] $\lambda = 1$ results. It can thus be concluded that not all that wiggling was just due to the inaccuracy of minimizing the very small deviations from saturation. As mentioned there,[6] some of the wiggling represented the high degeneracy of the problem: Since the structure has the same energy after being rotated in several ways, it is difficult to prevent the computer from shifting between different solutions. Effectively it means that the small tilt is in different directions for different values of a.

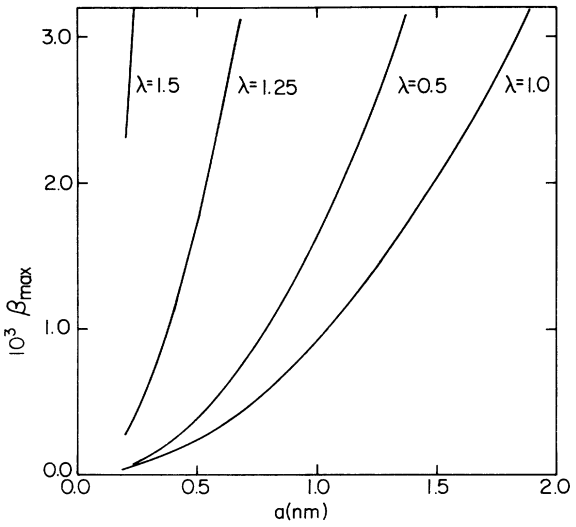

FIG. 1. The largest magnetization component perpendicular to the curling symmetry axis at the minimum-energy configuration of nine dipoles in a bcc lattice, as a function of the cube edge a for an exchange constant $C = 1.73 \times 10^{-6}$ erg/cm, and the parameters of Eq. (2). The parameter λ is a measure of the displacement of spin No. 9 along [111] away from its bcc position at $\lambda = 1$.

The *same* data of Fig. 1 are plotted in Fig. 2 in the

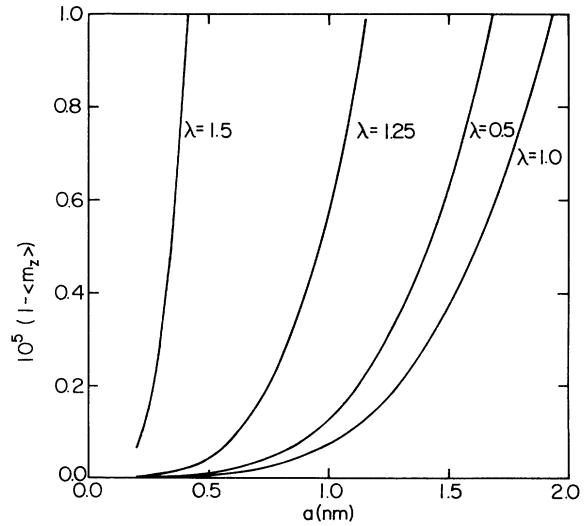

FIG. 2. Average over the 9 dipoles of the deviation of the z component of the magnetization from its saturation for the same data as in Fig. 1.

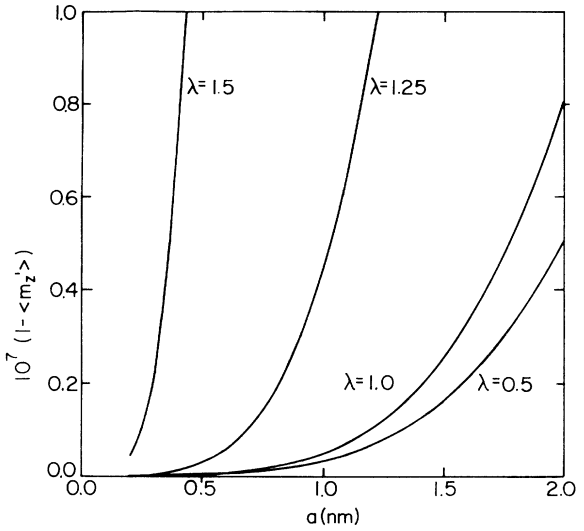

FIG. 3. Same as Fig. 2, for a fictitious material with the parameters defined in Eq. (3). The direction z' is along the body diagonal of the cube, after rotating according to Eqs. (4)–(6).

form of the deviation of the *average* of the nine spins from the z direction. It is clear from either figure that these deviations become very small when a approaches the lattice constant of Fe, but there is no a for which they actually become zero, and that this behavior is very much the same for the different values of λ in these figures. Hence, there is no "single-domain" particle in the atomic size limit, even when the high symmetry of the bcc structure is broken.

For the physical parameters of Ni, Eq. (3), the results of the minimization are very similar to those of Fe. The only difference is that the magnetostatic and anisotropy energies for Ni are much smaller than for Fe, so that the deviations from saturation, plotted in Fig. 3, are two orders of magnitude smaller than those in Fig. 2. Therefore the noise in Fig. 4, which is a similar plot to Fig. 1, is very large, although it is interesting to note that the noise for the other values of λ is much smaller than that for $\lambda = 1$. However, the results are qualitatively the same as for Fe, and do not depend on the value of λ. Thus the conclusion for Fe applies equally well to Ni.

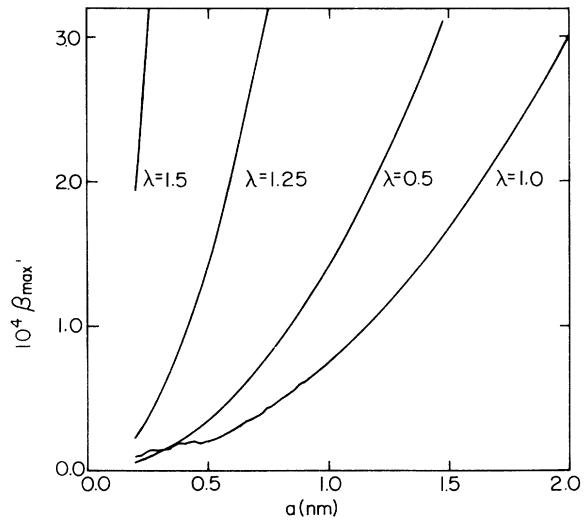

FIG. 4. Same as Fig. 1, for the material of Fig. 3.

IV. DISCUSSION

For the numerical values used in this study, the exchange energy is several orders of magnitude larger than the other energy terms, which can at most lead to very small deviations from a homogeneous magnetization at the energy minimum, even at quite large "lattice constant" a. For a realistic a the computer accuracy was not sufficient to determine the energy minimum, and it had to be approached from unphysically large values. But the extrapolation from a large to a realistic value of a is quite clear and conclusive.

Formally the present study deals with extremely small particles, which contain only a small number of atoms. However, as it is not quantum mechanical, it is not a true atomic theory; and particles that are so small are not ferromagnetic anyway. They become paramagnetic because thermal agitations can overcome the energy barrier for flipping the magnetization direction back and forth, as in Ref. 7 and the references cited therein. For somewhat larger, but rather small particles, the conclusion of the present study is that there is no "critical size" below which a particle is uniformly magnetized. However, the conventional micromagnetics calculations yield an energy which is lower for the curling configuration than it is for uniformly magnetized structures,[8] and this is not actually changed by the present results. The concept of a nucleation field as the field at which the magnetization just starts to deviate from saturation, cannot be defined as *rigorously* as assumed before, now that it has been shown that the saturated state cannot exist. However, the obtained deviations from uniformity are extremely small, and their contribution can be ignored, whenever common sense indicates that very small deviations can only have a small effect.

The real difference is for *large* particles, for which it has always been known that the energy of a domain structure is much lower than that of the saturated state, but it was not clear how these domains can enter the sample, *once it was completely saturated*.[2] Now that we have shown that even the smallest particle cannot be saturated by the exchange field (which is much larger than any applicable magnetic field) it is obvious that no particle can ever be *strictly* saturated by applying such a field, no matter how large. If the particle is not saturated, the *proof* of Brown[1] for the existence of a high barrier for *starting* the magnetization reversal does not hold any more. In the conventional, or historical, sense of the the word, the so-called "Brown paradox" has thus just disappeared.

It should be noted that the above argument does not prove that such a barrier does *not* exist; only that there is no proof for its existence. If such a barrier does exist, and the theory still does not agree with common observations,

it must be proved now on a different basis. The present treatment shows only the existence of very small angles between nearest-neighbor spins. It does not necessarily follow, but it is undeniably *possible*, that these angles may add up over the many spins in a sufficiently large crystal, even up to a fully developed vestigial domain configuration, as suggested[5] by some experiments.

Of course, there is no well-defined region for the applicability of the conventional calculations, or any clear-cut definition of what is a small particle and what is a large particle in this context. The saturated state is a very good approximation to the magnetization configuration in a large applied field, for *all* particles, and the real question is if one may still get away with treating the nucleation theory as a useful approximation. It is definitely not useful for bulk ferromagnets, but very useful for fine-particle magnets, for which the calculations often agree with experiments, especially when these particles are relatively free from defects.[8] The transition between these extreme cases remains ambiguous.

ACKNOWLEDGMENTS

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