Journal of Magnetism and Magnetic Materials 215–216 (2000) 18–23

Journal of magnetism and magnetic materials

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Invited paper

An ideally soft ferromagnet: is it feasible?

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Abstract

The infinite initial susceptibility encountered in the micromagnetics computations of sufficiently small, amorphous ferromagnetic spheres is argued to be a real physical phenomenon, which may actually be realized in practice as a very large enhancement of the initial susceptibility of thin, amorphous ferromagnetic films, when they are subdivided into separate discs. The only difficulty is to make the films highly homogeneous, and to avoid mechanical strains as much as possible. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Ideally soft ferromagnets; Enhanced initial susceptibility; Micromagnetics; Improved read-heads

1. Introduction

My first encounter with the name 'ideally soft ferromagnet' was when I visited Tony Arrott in the summer of 1978. He coined this term to describe an isotropic ferromagnet without magnetostriction, such as iron whiskers just below the Curie temperature. On approaching T_c, the anisotropy of iron decreases [1] much faster than the magnetization. Therefore, there is a (small) temperature range in which the magnetization is still appreciable, while the anisotropy is already negligibly small. For this range, Arrott et al. developed a special theory [2] to explain their special experimental data, advancing from a crude approach [3] to one of the best [4] theoretical micromagnetics models ever done. It is remarkable even on today's standards, after all the numerical computations done in the meantime.

This model, however, did not interest many magneticians, because it was limited to a very special case at a very limited temperature range. This attitude should have changed when amorphous materials were discovered, but it did not. Somehow many workers refused to consider amorphous ferromagnets for what they basically are at room temperatures, namely samples which are essentially isotropic, at least on average. Instead, many were obsessed with looking for some unusual properties near T_c, as predicted by a theory of some fictitious, very large domains [5], that for a typical case turned out [6] to be bigger than the sample! This theory [5] did not mention amorphicity, nor was it meant to apply to this case. It only happened to be published at about the time these materials were invented, but it was somehow understood to describe them. A large number of experiments were tried to fit to this theory, proved [7] unfit for this purpose, in spite of a clear demonstration of an excellent fit of a conventional theory [8] to many experimental data.

Arrott's model leads to hysteresis loops with qualitatively similar properties to those of other soft materials, because both the theoretical and the experimental studies use highly elongated particles. There is thus no magnetocrystalline anisotropy, but there is a large *shape* anisotropy. In this respect, it is interesting to note that the model is also ignored by those criticized [9] for trying to account for experimental data on elongated particles by numerical computations in which the magnetocrystalline anisotropy is *assumed* to be zero. This assumption is a bad approximation [9] to the physical reality of the studied cases, but once made, its results must be compared with those of other cases for which the same assumption is made.

The possibility of a *complete* isotropy emerged when amorphous spheres [10], that have neither shape nor magnetocrystalline anisotropy or anisotropic mechanical stresses, were made. These spheres seemed to lead to

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PII: S0304-8853(00)00056-1

a new kind of extremely soft ferromagnets, but this goal was not achieved, for the reasons discussed in the next section. The same idea is extended in Section 3 to the case of thin circular discs, that theoreticallly can deliver an outstandingly soft ferromagnet, especially in applications for which only the initial susceptibility counts, and it is not necessary to use the soft magnetic properties all the way to saturation. The possibility of using the theory for real-life applications, such as transformers, sensors, or recording read-heads, is discussed in Section 4.

2. Amorphous spheres

Small particles of $Fe_{75}Si_{15}B_{10}$ made by spark erosion were [10] 'roughly spherical', so that they were 'almost free' from shape anisotropy, and the same conclusion can be drawn from the more detailed electron micrographs of particles made by the same [11,12] or other [13] techniques. Such particles were [14] even 'more amorphous' than ribbons of the same composition, in the sense that the short-range order in the particles was smaller than in the ribbons. This result may mean a complete isotropy in each particle, in as much as amorphicity implies isotropy, a point that is further discussed in Section 4. Magnetostriction should also have a much smaller effect than in ribbons, because the production of ribbons introduces high and anisotropic mechanical strains, while the particles cooled in a liquid, with no substrate, should [10] have no anisotropic stress. It thus seemed at first sight that these particles have all it takes to be ideally soft.

According to the old, simple-minded approach [15], small ferromagnetic particles cannot support a multidomain structure, and become 'single domains'. For such particles, the coercive force is proportional to the anisotropy constant, if magnetocrystalline anisotropy dominates, or to the shape anisotropy constant when it dominates, or to the mechanical stress. [15] Therefore, if all these three factors vanish, the coercivity should be zero, and the reversal of magnetization should take place as one discontinuous rotation of all the spins from one direction to another, with an infinite susceptibility. Later studies changed [16] two major factors in this simple picture, namely:

(i) The 'critical size' for a particle becoming a single domain, as estimated in Ref. [15], changed considerably in later estimates, mainly because the magnetization was found to assume complicated structures just above that size, instead of the simple configurations assumed before.
(ii) It was found that very small particles become superparamagnetic, and lose their ferromagnetic properties altogether.

The second of these points, once realized, is less important for practical applications. Very small particles are indeed produced [12], but they are easy to separate from the others, because they remain suspended in the liquid [12,17] when the larger ones settle out.

The first point is crucial for any practical application of such materials. Since these particles were found at the time when the GE Co. was planning a high-speed magnetic printer [18] for computer output, it was only natural to ask if the predicted extremely soft properties could be used in the printing head. To answer this question I developed a Ritz model [19] for the magnetization configuration in small isotropic spheres, and used it to study the particular combination of the exchange constant, C, and the saturation magnetization, M_s, that should apply to $Fe_{75}Si_{15}B_{10}$,

$$R_0^2 = 9C/(8\pi M_s^2) = 60.4, \tag{1}$$

where R_0 is measured in nm. For this case, the average of the reduced magnetization component,

$$m_z = M_z/M_s, \tag{2}$$

was calculated as a function of the reduced field,

$$h = 3H/(2\pi M_s), \tag{3}$$

where H is the applied field. Some results are reproduced in Fig. 1, including the previously known magnetization

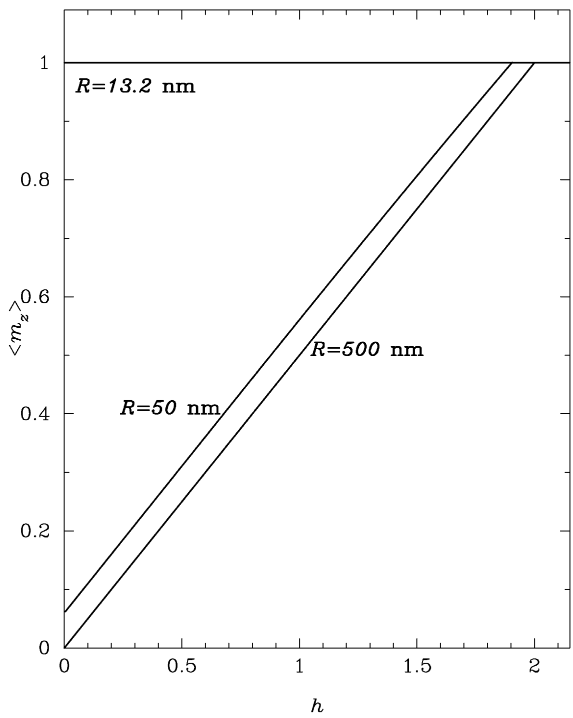

Fig. 1. The theoretical reduced average magnetization for isotropic spheres radius R as marked on the figure, with R_0 defined by Eq. (1), as a function of the reduced applied field h, defined in Eq. (3).

curve of a *large* sphere, which is a straight line extending from zero to saturation. Its slope is determined by the demagnetization of a sphere,

$$H_D = 4\pi M_s/3. \quad (4)$$

The susceptibility is thus $3/(4\pi)$ which will not do for a soft material, even though there is no coercivity.

The part of Fig. 1 which has not been known before is that the behavior seen for the large radius of 500 nm, also applies approximately to a radius of 50 nm, and of course to all radii in between. Only at a radius of 13.2 nm (or smaller) the expected jump, with an infinite susceptibility, is attained, and this radius is already dangerously close to the superparamagnetic region, where this theory, and ferromagnetism as such, break down. Since amorphous $Fe_{75}Si_{15}B_{10}$ particles could only be made [10,11] with a diameter of 0.5 μm or larger (besides the very small, superparamagnetic particles, separated out), it is clear from Fig. 1 that they are useless for applications such as the printing head which the people in GE had in mind.

There is, however, another possibility. As mentioned in the foregoing, the magnetization configuration in small spheres (between the 'single-domain' size of 13.2 nm in Fig. 1 and the size at which a sphere is subdivided into domains) is much more complex than that assumed [15] in the old theories. Such structures were approximated in the earlier [19] Ritz model, and were later computed by a numerical minimization [20] of the energy. Magnetization curves obtained by averaging over the volume of the sphere the computed [20] m_z for each value of h, are plotted in Fig. 2 for the two radii, $R = 15$ and 20 nm. There is no hysteresis in these curves, but the discontinuous jump does not lead to a complete reversal of the magnetization, as is the case for the smaller radii in Fig. 1. The jump (with an infinite susceptibility in zero applied field) takes the average magnetization to a finite value, which increases with decreasing R. It then varies almost linearly in the field, till it reaches saturation at a field value which coincides with the analytic nucleation field [21] for a sphere of that radius. There is some hint to this behavior already in the curve for $R = 50$ nm in Fig. 1, but it only becomes clear and obvious for smaller radii, such as the ones in Fig. 2.

This strange and unusual behavior was found in the Ritz model [19] for isotropic spheres, but there was a place for some doubts then, because the point of saturation in the original model was too far from the analytic nucleation field. Better computations [20] made this feature more reliable, and it was also encountered (but ignored) in a three-dimensional [22] computation of an isotropic sphere. It has never been seen in any other case, before or after, theoretically or experimentally, except for a study [23] of amorphous Fe particles in a silica matrix, the magnetization curve of which is described as being

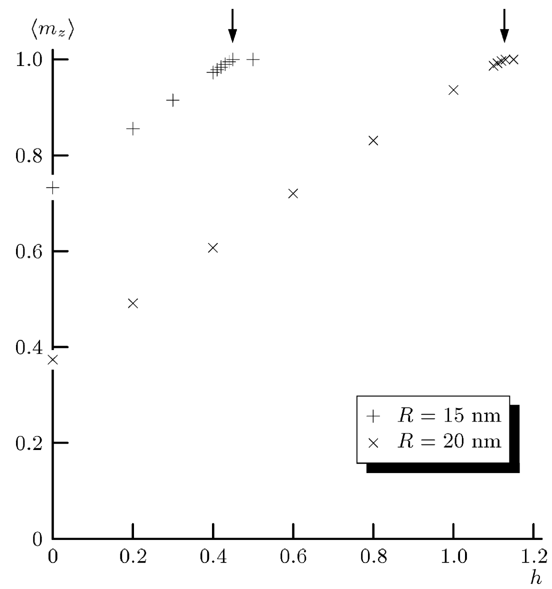

Fig. 2. The theoretical reduced average magnetization for isotropic spheres of radii 15 nm and 20 nm, for a material whose R_0 is given in Eq. (1), as a function of the reduced applied field h, defined in Eq. (3). The arrows mark the points where saturation is reached.

'quite similar' to the theoretical curves in Ref. [19]. There are, however, some unknown factors in this experiment, especially since some of the particles are [23] superparamagnetic. Therefore, this experiment can only be taken as a weak support, which is not sufficient for verifying the theory.

Nevertheless, within the limitations of Section 4 there is no reason to suspect any serious fault in the theory. Therefore, if amorphous spheres can be made in the appropriate size range, their initial susceptibility should be very large, making them useful for transformers or read-heads. Even if the amorphous particles of $Fe_{75}Si_{15}B_{10}$ made by spark erosion are too large to show this effect, there may be other methods or other amorphous materials that can do it. Thus, metallic alloys have been made in nearly spherical shape, in the micrometer [24,25] and sub-micrometer [26] size range, and with a very narrow size distribution. Such particles, or even smaller ones, may sometime be made of amorphous alloys. It should be noted that the calculations need not be repeated if a different material is involved, because all the radii mentioned in the foregoing are normalized to R_0 defined in Eq. (1). The theoretical results are the same for the same value of R/R_0. If R_0 is different from the numerical value in Eq. (1), the curves of Fig. 2 still apply, but for a different R that preserves the same value of R/R_0.

3. Thin discs

The advantage of the spherical geometry used in the foregoing is the lack of any shape anisotropy. This advantage is shared by a circular cylinder, as long as the magnetization is restricted to the plane perpendicular to the cylinder axis. Therefore, the main results of Figs. 1 and 2 should be similar to those that apply to circular discs made of thin films, for which at least most of the magnetization lies in the film plane.

On the one hand, amorphous spheres are better than discs because the former are free from anisotropic mechanical stresses, while even ribbons are known to be highly stressed, so that their magnetic properties change considerably [27] when they are annealed, and amorphous films must be worse in this respect. Annealing temperatures are often dangerously close to the crystallization point, which may sometimes [28] be an advantage, but not in the present case. It can only be hoped that a proper annealing is found that will relieve the stresses but let the film stay amorphous.

On the other hand, the thin film geometry has several advantages over the spherical geometry:

(i) Making amorphous ferromagnetic films, by sputtering or by vapor deposition, is already an old and well-established technique. [29]

(ii) Subdividing films into circular discs, from a few nm [30] to a few hundred nm [31] has also been done already, although not for amorphous films. Since the theory deals with only one particle, neglecting interactions among them, the separation among the discs will have to be determined by trial and error, and the manufacturing of discs is in a better control of this parameter, *and* of the disc size, than the manufacturing of spheres.

(iii) The size distribution of discs can be very narrow, and 'arrays of 10^8 identical particles' [31] have been reported. Such discs are also a better approximation to circles than particles are to spheres.

(iv) Recording heads are made of thin films anyway, usually multilayers [32], that must be easy to subdivide into circular discs. MR read heads have [33] shields, for which a high susceptibility is as important as for inductive heads. It may also be worth etching the MR part into separate discs, with thin lines for the electrical connection.

Even without a detailed theory for thin discs, a rough idea of their behavior can be obtained from the theoretical results for spheres. The nucleation field by curling has recently been claimed [34] to depend mostly on the particle volume, and hardly on its shape, except for the shape-dependent part of the additive demagnetizing factor. This conjecture is based only on experience with three-dimensional particles, and it is not clear if it may be extended to thin films. Nevertheless, it is assumed that the nucleation field of a sphere of radius R_s is approximately that of a disc of radius R_d if they have the same volume, namely if

$$4\pi R_s^3/3 = \pi R_d^2 t, \tag{5}$$

where t is the film thickness, or the total thickness of all the magnetic films in the case of a multilayer. Substituting for R_s in the curling nucleation field [16], normalizing dimensions to R_0 defined in Eq. (1), and using the normalized magnetic field in Eq. (3),

$$h_n = 6D_x - \left(\frac{4}{3}\right)^{5/3} q^2 \left(\frac{R_0}{R_d}\right)^{4/3} \left(\frac{R_0}{t}\right)^{2/3}, \tag{6}$$

where $q = 2.0816$ is the curling eigenvalue for a sphere, and D_x is the demagnetizing factor of the disc in the plane of the film. The latter factor must fulfill

$$D_x = (1 - D_z)/2, \tag{7}$$

where D_z is the (magnetometric) demagnetizing factor perpendicular to the plane of the film, which has been tabulated [35] under the name N_m.

Using the values in Table 1 of Ref. [35], Eq. (6) was solved for the case $h_n = 0$, namely the values of R_d for which *all* the magnetization switches in one jump with an infinite susceptibility at zero applied field. The results are plotted in Fig. 3 as a function of the film thickness, t. A partial switching as in Fig. 2 can be expected to occur

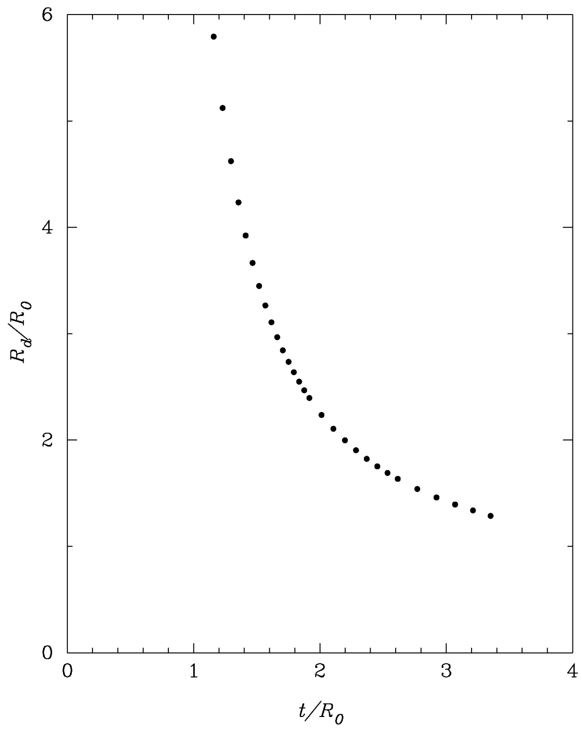

Fig. 3. Theoretical disc radius, R_d, for a one-switch magnetization reversal as a function of the film thickness, t. Both are normalized to R_0 defined by Eq. (1).

for considerably larger radii of the disc. Fig. 3, however, seems to indicate that even a total switching of the magnetization may be attainable in practice, so that the advantage of high susceptibility is not offset by the disadvantage of not using full saturation. If this indeed turns out to be the case, it may also be worth trying to subdivide write heads, which are also made [36] of thin films, into discs.

4. Practical considerations

There are no infinities in real life, and the foregoing infinite susceptibility should not be taken literally, being just the result of simplifications in the theory. It is like a resonance that is calculated as an infinite signal, but in practice is a finite peak. An important point in the present case is that a real particle cannot be a perfect sphere, or a perfect circular disc. The deviations from the perfect shape will reduce the infinity to a hopefully large, but finite, value. Also, the films must be as smooth as possible, because hysteresis loss in amorphous films is known to increase dramatically [37] with increased surface roughness.

For thin films in particular, there is an extra problem with strains during the deposition, and it can only be hoped that an appropriate annealing procedure is found that will reduce the effect of magnetostriction to a negligibly small value, without crystallizing the film. The theory presented here for films is also much less rigorous than that for spheres, but the qualitative picture of isotropy in the film plane must be valid. The quantitative estimation in Fig. 3 may thus be inaccurate, but it is only a hint for experimentalists of where to look for the appropriate size by trial and error.

A more serious problem is to justify the assumption, used here and in previous [20] studies, that amorphicity implies a complete isotropy throughout the particle. There is no clear evidence for or against this assumption, but it should be noted that others think differently, emphasizing [38–40] the strong, ‘single ion’ anisotropy. This anisotropy, with a randomly oriented easy direction, certainly exists [41] on an atomic scale, but then it must obviously be zero on average. Therefore, the working assumption of zero anisotropy seems justified for micromagnetics that always deals with averages, even for the exchange energy, and *not* with local fluctuations [16] on atomic scale. Also, those who take the local anisotropy very seriously will find it difficult to account for the experimental results of Ref. [23]. Randomly oriented crystallites are found [28] to be softer the smaller the grain size, presumably because it takes a sufficient number of grains to average over the ‘exchange length’. In the present case, the grain size is replaced by the limit of a single ion, so that a particle radius of at least several times R_0 seems to be adequate for averaging the anisotropy to zero, without worrying about the break-down of this assumption (and of micromagnetics in general) at very small particle size. Moreover, here the averaging over a particle is followed by averaging over a large number of such particles, which should also diminish the effect of slight irregularities in the shape of each particle. I thus believe that this experiment is well worth trying.

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