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Series expansions for the magnetisation of a solid superparamagnetic system of non-interacting particles with anisotropy

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Abstract

The calculation of the magnetisation curve of an assembly of non-interacting fine superparamagnetic particles, with uniaxial anisotropy and easy axes fixed in a solid non-magnetic matrix is considered. The presence of anisotropy complicates the calculation which otherwise would result in the Langevin function. The calculation for particles with anisotropy and easy axes fixed at arbitrary angles to the external field, requires the calculation of the partition function, which has previously been expressed exactly as a double integral or as a sum of single integrals. We have recently shown how the partition function can be reduced to a single integral and here we show how this can be expressed as a double infinite series containing known functions. Special cases are considered, some existing analytic formulae are reobtained, and some new analytic formulae are presented. For identical particles the deviation from the Langevin function is known to be considerable. The formulae presented should facilitate the incorporation of the effects of anisotropy. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Anisotropy; Superparamagnetism; Fine particles; Magnetisation curves

1. Introduction

In this paper we consider a superparamagnetic system, made up of fine non-interacting single domain magnetic particles possessing uniaxial anisotropy. We adopt the usual benchmark for superparamagnetism, being that the relaxation time of every particle is less than 100 s. The system considered is made up of an assembly of fine particles with easy axes fixed relative to a solid non-magnetic matrix. This we refer to as a solid superparamagnetic system. The particles are sufficiently dilute to ignore interactions. If the solid were cooled in the presence of an external magnetic field we would expect a preferential alignment of the particles easy axes in the direction of the field, resulting in a textured system. Cooling in the absence of an external field

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would result in a random texture. We have recently [1] shown how the magnetisation of a solid superparamagnetic system, can be calculated from a single integral form of the partition function, which had been expressed previously as a double integral [2] or as a sum of integrations [3]. Here we show how this single integral in turn can be written as a double infinite series of known functions. The series can be expanded until tolerance limits are obtained. This should make the calculation of the magnetisation with anisotropy more tractable, allowing a move away from the Langevin function of paramagnetism. As was originally stated by Bean and Livingston [4] the presence of anisotropy causes a deviation from the behaviour predicted by the Langevin function. For superparamagnetism the Langevin function is only valid for special cases. For identical particles, these cases are a magnetic fluid, where the viscosity is low enough for the particles to behave like a paramagnetic gas, a solid with zero anisotropy (physically unlikely) or a solid with anisotropy, random texture, and low external field. For the last case, specifically where the field energy to thermal energy ratio is less than one, the Langevin function is a valid approximation. When larger fields are involved, the anisotropy is an important factor in the description, and the predicted behaviour deviates significantly from that of the Langevin function. Specifically, saturation requires higher fields to achieve than the Langevin function would suggest. This is not surprising, as the anisotropy holds the magnetic vector of each particle closely to its easy axis, inhibiting saturation. However, as illustrated by Mamiya and Nakatani, recently [5] a size distribution can mask the effect of anisotropy on the magnetisation.

2. The magnetisation and the partition function with texture and size distributions

In general, the magnetisation can be calculated from the expected (equilibrium) value of $\cos\omega$, written $\langle\cos\omega\rangle$, where from Fig. 1, ω is the angle between the moment of each particle μ ($\mu = VM_s$) and the field $\boldsymbol{H}$. The magnetisation can be written

$$M(\beta,\alpha,\psi) = M_s\langle\cos\omega\rangle = M_s\frac{\partial Z/\partial\beta}{Z}, \tag{1}$$

where $\beta = HM_sV/kT$, with V the particle volume, H the applied field, M_s the saturation magnetisation, $\alpha = KV/kT$ with K being the anisotropy constant and kT the thermal energy. Z is the partition function given in Section 3 and ψ is the angle between the easy axis of the particle and the field as shown in Fig. 1. The

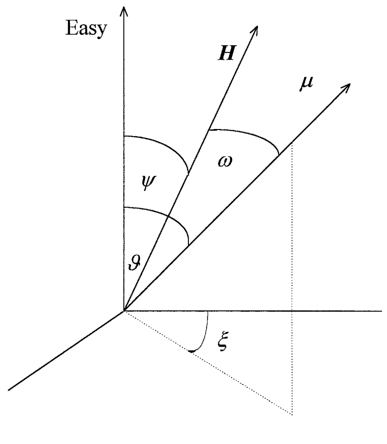

Fig. 1. The coordinate system used, with particle easy axis, field $\boldsymbol{H}$ and moment μ.

formula can be taken to apply to an ensemble of identical particles each of which has its easy axis fixed at the same angle ψ to the field. This would be the case if all the particles' easy axes were aligned, corresponding to a delta-like distribution of easy axes. In general, any distribution of easy axes might exist. A solid cooled in zero field should have a uniform distribution leading to a random texture or random anisotropy [5]. A solid which was fixed in the presence of a field would have a preferential alignment leading to a non-random texture. The delta-like case of all easy axes aligned is an extreme example of texture, resulting from anisotropy and high cooling field. In general, texture can be incorporated into the analysis by integrating over the distributions. For a system with texture given by a distribution of angles $D(\psi)$ this integration is

$$M(\beta, \alpha) = \frac{\int_0^\pi M(\beta, \alpha, \psi) D(\psi) \sin\psi \, d\psi}{\int_0^\pi D(\psi) \sin\psi \, d\psi}. \tag{2}$$

Distributions for non-random textures can be obtained following Raĭkher [6]. For a random texture resulting from a uniform distribution we have [3]

$$M(\beta, \alpha) = \int_0^{\pi/2} M(\beta, \alpha, \psi) \sin\psi \, d\psi, \tag{3}$$

where the symmetry allows the halving of the interval. This applies to an ensemble of identical particles. For a range of particle sizes a distribution such as that of Chantrell et al. [2] can be used. Eq. (1) requires the calculation of the two integrals Z and $\partial Z/\partial\beta$ and it is the aim of this paper to present formulae which will facilitate these calculations.

3. Reduction of the partition function to single integral form

The partition function Z as a double integral is given by

$$Z = \frac{1}{4\pi}\int_0^{2\pi}\int_0^{\pi} \exp(-\alpha \sin^2\vartheta + \beta\cos\omega) \sin\vartheta \, d\vartheta \, d\xi, \tag{4}$$

using the coordinate system of Chantrell et al. [2] given in Fig. 1. The angle ω is determined by the relation

$$\cos\omega = \cos\vartheta\cos\psi + \sin\vartheta\sin\psi\cos\xi. \tag{5}$$

Therefore, Z is given by

$$Z = \frac{1}{4\pi}\int_0^{2\pi}\int_0^{\pi} \exp(-\alpha\sin^2\vartheta + \beta\cos\vartheta\cos\psi + \beta\sin\vartheta\sin\psi\cos\xi) \sin\vartheta \, d\vartheta \, d\xi. \tag{6}$$

which we have shown in Ref. [1] can be reduced to the single integral form

$$Z = \int_0^{\pi/2} \exp(-\alpha\sin^2\vartheta) \cosh(\beta\cos\vartheta\cos\psi) I_0(\beta\sin\vartheta\sin\psi) \sin\vartheta \, d\vartheta. \tag{7}$$

This reduction is achieved by considering the symmetry of the functions to be integrated and using an integral definition of the modified Bessel function of zero order $I_0(z)$, that is Eq. (9.6.16) of Abramowitz and Stegun [7]. Furthermore, noting the differential properties of $I_0(z)$, given by Eq. (9.6.27) of Ref. [7]

$$\frac{d}{dz} I_0(z) = I_1(z),$$

we can calculate $\partial Z/\partial\beta$ from

$$\frac{\partial Z}{\partial \beta} = \int_0^{\pi/2} \exp(-\alpha\sin^2\vartheta)[\cosh(\beta\cos\vartheta\cos\psi)I_1(\beta\sin\vartheta\sin\psi)\sin\vartheta\sin\psi + \sinh(\beta\cos\vartheta\cos\psi)I_0(\beta\sin\vartheta\sin\psi)\cos\vartheta\cos\psi]\sin\vartheta\,\mathrm{d}\vartheta. \quad (8)$$

4. Expansion of the functions in the integrand

The integrand of Eq. (7) involves the exponential, modified Bessel function of zero order and the hyperbolic cosine functions. In this section we expand the first two of these as infinite sums to find that the remaining integral involving the hyperbolic cosine reduces to a known function. The series expansion of the exponential is Eq. (4.2.1) of Ref. [7]

$$\exp(-\alpha\sin^2\vartheta) = 1 + \sum_{n=1}^{\infty}\frac{(-\alpha)^n}{n!}\sin^{2n}\vartheta,$$

where for non-zero argument we can write

$$\exp(-\alpha\sin^2\vartheta) = \sum_{n=0}^{\infty}\frac{(-\alpha)^n}{n!}\sin^{2n}\vartheta, \quad (9)$$

and the modified Bessel function as given by Eq. (9.6.10) of Ref. [7] is (in the notation of that text)

$$I_v(z) = (z/2)^v\sum_{k=0}^{\infty}\frac{(z^2/4)^k}{k!\Gamma(v+k+1)},$$

which for $v=0$ gives

$$I_0(z) = \sum_{k=0}^{\infty}\frac{(z^2/4)^k}{k!\Gamma(k+1)} = \sum_{k=0}^{\infty}\frac{(z/2)^{2k}}{(k!)^2}, \quad (10)$$

where the integer value of the Gamma function

$$\Gamma(k+1) = k!$$

given in Eq. (6.1.6) of Ref. [7] is used. In this case, taking

$$z = \beta\sin\vartheta\sin\psi,$$

we arrive at

$$I_0(\beta\sin\vartheta\sin\psi) = \sum_{k=0}^{\infty}\frac{(\frac{1}{2}\beta\sin\psi)^{2k}\sin^{2k}\vartheta}{(k!)^2}. \quad (11)$$

We note that the zero argument should be avoided in Eq. (11) for $k=0$. Use of the sums given in Eqs. (9) and (11) leads to

$$Z(\beta,\alpha,\psi) = \sum_{n=0}^{\infty}\frac{(-\alpha)^n}{n!}\sum_{k=0}^{\infty}\frac{(\frac{1}{2}\beta\sin\psi)^{2k}}{(k!)^2}\int_0^{\pi/2}\sin^{2(n+k)}\vartheta\cosh(\beta\cos\vartheta\cos\psi)\sin\vartheta\,\mathrm{d}\vartheta. \quad (12)$$

The remaining integral can be dealt with by noting Eq. (9.6.18) of Ref. [7] which is

$$I_v(z) = \frac{(z/2)^v}{\sqrt{\pi}\Gamma(v+1/2)}\int_0^{\pi} \mathrm{e}^{\pm z\cos\theta}\sin^{2v}\theta\,\mathrm{d}\theta \quad (\Re v > -1/2),$$

here $I_v(z)$ is a modified Bessel function. By adding both $\pm$ cases, and dividing by 2, in place of the exponential, we obtain the hyperbolic cosine. In the notation of Ref. [7], we rearrange as

$$\int_0^{\pi} \cosh(z \cos \vartheta) \sin^{2v} \theta \, d\theta = \frac{I_v(z)\sqrt{\pi}\Gamma(v + 1/2)}{(z/2)^v}. \tag{13}$$

The limits of this can be halved based on the symmetry of the integrands around $\pi/2$. Comparing this with the remaining integral we see that the substitution of

$$z = \beta \cos \psi,$$

and

$$v = n + k + 1/2,$$

gives

$$\int_0^{\pi/2} \cosh(\beta \cos \vartheta \cos \psi) \sin^{2(n+k+1/2)} \vartheta \, d\vartheta = \frac{I_{n+k+1/2}(\beta \cos \psi)\sqrt{\pi}(n + k)!}{2(\frac{1}{2}\beta \cos \psi)^{n+k+1/2}}, \tag{14}$$

where the Gamma function is replaced by the factorial as in Eq. (10), and where the functions $I_{n+k+1/2}(z)$ are the modified spherical Bessel functions of the first kind. Replacing the remaining integral in the partition function gives

$$Z(\beta, \alpha, \psi) = \sum_{n=0}^{\infty} \frac{(-\alpha)^n}{n!} \sum_{k=0}^{\infty} \frac{(n + k)!}{(k!)^2} \frac{(\frac{1}{2}\beta \sin \psi)^{2k}}{(\frac{1}{2}\beta \cos \psi)^{n+k}} \sqrt{\frac{\pi}{2\beta \cos \psi}} I_{n+k+1/2}(\beta \cos \psi). \tag{15}$$

It is helpful to introduce the notation

$$f_n(z) = \sqrt{\frac{\pi}{2z}} I_{n+1/2}(z), \tag{16}$$

whereby

$$Z(\beta, \alpha, \psi) = \sum_{n=0}^{\infty} \frac{(-\alpha)^n}{n!} \sum_{k=0}^{\infty} \frac{(n + k)!}{(k!)^2} \left(\frac{\beta}{2}\right)^{k-n} \frac{\sin^{2k} \psi}{\cos^{n+k} \psi} f_{n+k}(\beta \cos \psi). \tag{17}$$

The differential properties of $f_n(z)$, given by Eq. (10.2.20) of Ref. [7], are

$$\frac{d}{dz} f_n(z) = f_{n+1}(z) + \frac{n}{z} f_n(z), \tag{18}$$

which lead to

$$\frac{\partial Z(\beta, \alpha, \psi)}{\partial \beta} = \sum_{n=0}^{\infty} \frac{(-\alpha)^n}{n!} \sum_{k=0}^{\infty} \frac{(n + k)!}{(k!)^2} \left(\frac{\beta}{2}\right)^{k-n} \frac{\sin^{2k} \psi}{\cos^{n+k} \psi} \left[\cos \psi f_{n+k+1}(\beta \cos \psi) + \frac{2k}{\beta} f_{n+k}(\beta \cos \psi)\right]. \tag{19}$$

Considering the terms for $n = 0$ and $k = 0$ we can write

$$\begin{aligned} Z(\beta, \alpha, \psi) = {} & f_0(\beta) + \sum_{n=1}^{\infty} \left(\frac{-2\alpha}{\beta \cos \psi}\right)^n f_n(\beta \cos \psi) \\ & + \sum_{n=1}^{\infty} \frac{(-\alpha)^n}{n!} \sum_{k=1}^{\infty} \frac{(n + k)!}{(k!)^2} \left(\frac{\beta}{2}\right)^{k-n} \frac{\sin^{2k} \psi}{\cos^{n+k} \psi} f_{n+k}(\beta \cos \psi), \end{aligned} \tag{20}$$

where the leading term is seen to have no ψ dependence as is shown in Appendix A and also

$$\frac{\partial Z(\beta,\alpha,\psi)}{\partial\beta} = f_1(\beta) + \sum_{n=1}^{\infty}\left(\frac{-2\alpha}{\beta\cos\psi}\right)^n \cos\psi\, f_{n+1}(\beta\cos\psi)$$

$$+ \sum_{n=1}^{\infty}\frac{(-\alpha)^n}{n!}\sum_{k=1}^{\infty}\frac{(n+k)!}{(k!)^2}\left(\frac{\beta}{2}\right)^{k-n}\frac{\sin^{2k}\psi}{\cos^{n+k}\psi}\left[\cos\psi\, f_{n+k+1}(\beta\cos\psi) + \frac{2k}{\beta}f_{n+k}(\beta\cos\psi)\right]. \quad (21)$$

The functions $f_n(z)$ can be calculated directly from hyperbolic functions and using the recurrence relations (10.2.18) of Ref. [7],

$$f_{n-1}(z) - f_{n+1}(z) = \frac{2n+1}{z}f_n(z). \quad (22)$$

The first three are given in Appendix A.

5. Discussion of evaluation techniques

We now have a infinite series for the partition function Z. This involves the modified spherical Bessel functions of the first kind and is given in Eq. (20). These functions are available in most mathematical packages including *Maple* and *Mathematica*. This solution requires no numerical integration. The differential properties allow us to write down another series for $\partial Z/\partial\beta$, Eq. (21). In order to avoid the repetition of evaluations which might take place when evaluating the Bessel functions in packages the recurrence relations in Appendix A can be used. These require just a single evaluation of the hyperbolic sine and cosine functions and the generation of coefficients $g_n(\beta\cos\psi)$. Alternatively, f_0 and f_1 can be calculated and the recurrence relations of Eq. (22) can be used to generate the appropriate coefficients. Also numerical integration of the single integral form of Z, Eq. (7) and of $\partial Z/\partial\beta$ Eq. (8) can be reconsidered when it appears that convergence requires a large numbers of terms. Overall, this should offer an improvement on previous methods such as that outlined in Ref. [3], which required sums of integrations.

6. Special cases

We can obtain some analytic approximations from the exact forms considered. We consider the limiting case of no anisotropy, which leads to the Langevin function. For all easy axes aligned, we expand for small anisotropy, where the Langevin function is the leading term. Also, for all easy axes aligned and very large anisotropy we reobtain the asymptotic limit of hyperbolic tangent.

6.1. The case of no anisotropy

For the case of zero anisotropy we have only the leading terms in Eqs. (20) and (21) so that

$$Z(\beta) = f_0(\beta) = \frac{\sinh\beta}{\beta}, \quad (23)$$

and

$$\frac{\partial Z}{\partial\beta} = f_1(\beta) = \frac{\cosh\beta}{\beta} - \frac{\sinh\beta}{\beta^2}, \quad (24)$$

where we have noted Eqs. (10.2.13) and (10.2.14) of Ref. [7] given in Appendix A of this paper. From Eq. (1) this leads to

$$\frac{M(\beta)}{M_s} = L(\beta) \tag{25}$$

where $L(\beta)$ is the Langevin function given by

$$L(\beta) = \frac{f_1(\beta)}{f_0(\beta)} = \coth\beta - \frac{1}{\beta}. \tag{26}$$

6.2. For all particles aligned with the parallel field and $2\alpha < \beta$

For the case of low anisotropy and all easy axes aligned with the field we have $\psi = 0$ giving $\sin\psi = 0$ and $\cos\psi = 1$ so that

$$Z_{\parallel}(\beta, \alpha) = f_0(\beta) + \sum_{n=1}^{\infty} \left(\frac{-2\alpha}{\beta}\right)^n f_n(\beta). \tag{27}$$

For $2\alpha < \beta$ we can expand as

$$Z_{\parallel}(\beta, \alpha) = f_0(\beta) - \frac{2\alpha}{\beta} f_1(\beta) + \frac{4\alpha^2}{\beta^2} f_2(\beta) + \cdots. \tag{28}$$

From Eq. (21) we can write $\partial Z/\partial\beta$ as

$$\frac{\partial Z_{\parallel}(\beta, \alpha)}{\partial\beta} = f_1(\beta) + \sum_{n=1}^{\infty} \left(\frac{-2\alpha}{\beta}\right)^n f_{n+1}(\beta) \tag{29}$$

and expand as

$$\frac{\partial Z_{\parallel}(\beta, \alpha)}{\partial\beta} = f_1(\beta) - \frac{2\alpha}{\beta} f_2(\beta) + \frac{4\alpha^2}{\beta^2} f_3(\beta) + \cdots. \tag{30}$$

From Eq. (1) the magnetisation is then

$$\frac{M_{\parallel}(\beta, \alpha)}{M_s} = \frac{f_1(\beta) - (2\alpha/\beta) f_2(\beta) + (4\alpha^2/\beta^2) f_3(\beta) + \cdots}{f_0(\beta) - (2\alpha/\beta) f_1(\beta) + (4\alpha^2/\beta^2) f_2(\beta) + \cdots} + \cdots. \tag{31}$$

In terms of the Langevin function this can be written as

$$\frac{M_{\parallel}(\beta, \alpha)}{M_s} = L(\beta) + \frac{2\alpha/\beta(L(\beta)/\beta - L'(\beta)) - (2\alpha/\beta)^2((3/\beta)(L'(\beta) - 3L(\beta)/\beta + 2/3))}{1 - (2\alpha/\beta)L(\beta) + (2\alpha/\beta)^2(1 - 3L(\beta)/\beta)}. \tag{32}$$

For easy axes parallel to the external field we expect the presence of anisotropy to result in saturation at lower fields. Noting that the approach of the Langevin function to saturation is given by [4]

$$L(\beta) \cong 1 - \frac{1}{\beta} \tag{33}$$

for large β, we find the approach to saturation for $\beta \gg 1$ and $1 < 2\alpha < \beta$ to be

$$\frac{M_{\parallel}(\beta, \alpha)}{M_s} \cong 1 - \frac{1}{\beta} + \frac{2\alpha}{\beta^2}. \tag{34}$$

6.3. For all particles aligned with large anisotropy and arbitrary parallel field β

For the case of $\alpha \to \infty$ and arbitrary β we consider the single integral form of the partition function of Eq. (7). For all particles aligned with the field we have $\psi = 0$ giving $\sin\psi = 0$ and $\cos\psi = 1$. Thus, since $I_0(0) = 1$, Z is

$$Z = \int_0^{\pi/2} \exp(-\alpha \sin^2 \vartheta) \cosh(\beta \cos \vartheta) \sin \vartheta \, d\vartheta. \tag{35}$$

Making the substitution $x = \cos\vartheta$ we get

$$Z = \int_0^1 \exp(\alpha(x^2 - 1)) \cosh(\beta x) \, dx. \tag{36}$$

Separating the integral limits into two parts

$$Z = \int_{1-x_1}^1 \exp(\alpha(x^2 - 1)) \cosh(\beta x) \, dx + \int_0^{1-x_1} \exp(\alpha(x^2 - 1)) \cosh(\beta x) \, dx. \tag{37}$$

We find that as α becomes large the first integral becomes very much larger than the second for smaller values of the arbitrary value x_1, so that only the value of $\cosh(\beta x)$ at $x = 1$ is significant, resulting in the limit

$$Z \to \cosh \beta e^{-\alpha} \int_{1-x_1}^1 \exp(\alpha x^2) \, dx \quad \text{as } \alpha \to \infty. \tag{38}$$

From Eq. (1) the parallel magnetisation can then be written as

$$\frac{M_{\|}(\beta, \alpha \to \infty)}{M_s} = \tanh \beta. \tag{39}$$

This is the formula for aligned grains suggested in Ref. [8]. Also it is worth noting that it corresponds to the Brillouin function for paramagnetism when the lowest spin number $S = \pm\frac{1}{2}$ is used in that function [9,10] with only the discrete parallel and anti-parallel alignments possible. This agreement is not surprising as in the limit $\alpha \to \infty$ the discrete constraint is consistent with the magnetic moments being fixed tightly to their easy axes. Furthermore, for a random texture we can write the asymptotic limit for Eq. (3) as

$$\frac{M(\beta, \alpha \to \infty)}{M_s} = \int_0^{\pi/2} \cos\psi \tanh(\beta \cos\psi) \sin\psi \, d\psi, \tag{40}$$

which is in agreement with that employed in Ref. [5]. We further note that Garanin [11] has presented correction terms for Eq. (39) given by

$$\frac{M_{\|}(\beta, \alpha)}{M_s} \cong \tanh \beta - \frac{1}{2\alpha}\left(\frac{\beta}{\cosh^2 \beta} + \tanh \beta\right) + \frac{\beta}{(2\alpha)^2}.$$

7. Conclusions

It is hoped that the options presented here will facilitate accurate calculations of magnetisation curves for superparamagnetic systems with anisotropy.

We offer two routes: (a) direct numerical integration of one single integral for Z and another for $\partial Z/\partial \beta$; (b) evaluation of truncated sums of known functions for both quantities. As improving preparation techniques should tend towards the possibility of a narrower distribution of particle sizes it is to be expected

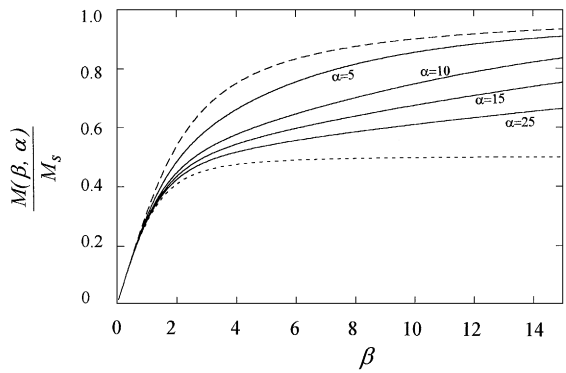

Fig. 2. The reduced magnetisation $M(\alpha, \beta)/M_s$ as a function of $\beta = HM_sV/kT$ for a system with random texture. The dashed and dotted curves are $L(\beta)$ and Eq. (40), respectively. The solid lines are calculated numerically using the single integral forms Eqs. (7) and (8) with Eqs. (1) and (3), for $\alpha = 5$, 10, 15 and 25.

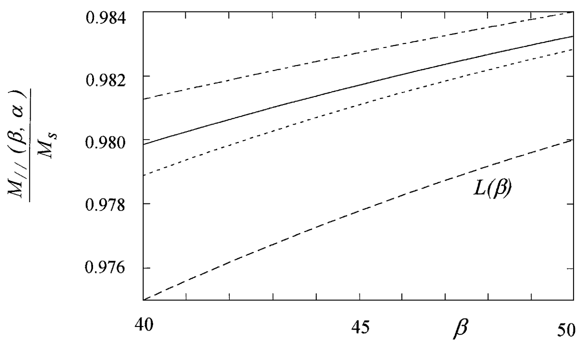

Fig. 3. The reduced parallel magnetisation $M_{\parallel}(\alpha, \beta)/M_s$ for aligned particles $\alpha = 5$. The solid is from the single integral forms. The dashed curve is $L(\beta)$. The dotted and the dot-dashed curves are Eqs. (32) and (34), respectively.

that the effects of anisotropy will become more manifest. Fig. 2 shows the effect of increasing anisotropy on a system with random texture. The overall effect is to leave the low-field region of the magnetisation curve and so the initial susceptibility unaffected, but to inhibit saturation. Fig. 3 shows the magnetisation curve for a delta-like distribution of easy axes where the field is applied parallel to the easy axes. The system saturates at lower fields than predicted by the Langevin function.

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Appendix A

In Eq. (17) when $n = 0$ we obtain a sum, which, by considering the original expansion in Eq. (7) is seen to equal the integral

$$\sum_{k=0}^{\infty} \frac{(\beta/2)^k}{(k!)} \frac{\sin^{2k}\psi}{\cos^k\psi} f_k(\beta\cos\psi) = \int_0^{\pi/2} \cosh(\beta\cos\vartheta\cos\psi) I_0(\beta\sin\vartheta\sin\psi)\sin\vartheta\,\mathrm{d}\vartheta. \tag{A.1}$$

With no anisotropy, ψ has no physical meaning. Therefore, any angle ψ could be chosen, thus Eq. (A.1) should be independent of ψ. From Watson [12] Eq. (12.14) we can write the following:

$$\int_0^{\pi/2} \cosh(\beta\cos\vartheta\cos\psi) I_0(\beta\sin\vartheta\sin\psi) P_n(\cos\vartheta)\sin\vartheta\,\mathrm{d}\vartheta = (-1)^{n/2} P_n(\cos\psi) f_n(\beta), \tag{A.2}$$

where $P_n(x)$ are the Legendre polynomials and where n must be even. This gives

$$\int_0^{\pi/2} \cosh(\beta\cos\vartheta\cos\psi) I_0(\beta\sin\vartheta\sin\psi)\sin\vartheta\,\mathrm{d}\vartheta = f_0(\beta), \tag{A.3}$$

which is the leading term in the partition function as required.

Also, in the specific case of $\psi = \pi/2$ we find

$$\int_0^{\pi/2} I_0(\beta\sin\vartheta)\sin\vartheta\,\mathrm{d}\vartheta = \frac{\sinh(\beta)}{\beta}, \tag{A.4}$$

which is in agreement with Eq. (11.4.10) of Ref. [7].

The modified spherical Bessel functions of the first kind can be calculated from Eq. (10.2.12) of Ref. [7]

$$f_n(z) = \sqrt{\frac{\pi}{2z}} I_{n+1/2}(z) = g_n(z)\sinh(z) + g_{-n-1}(z)\cosh(z),$$

where

$$g_0(z) = \frac{1}{z}, \quad g_1(z) = -\frac{1}{z^2} \qquad g_{n-1}(z) - g_{n+1}(z) = (2n+1)\frac{g_n(z)}{z}.$$

From Eq. (10.2.13) of Ref. [7] the first three functions are

$$f_0(z) = \sqrt{\frac{\pi}{2z}} I_{1/2}(z) = \frac{\sinh z}{z}, \qquad f_1(z) = \sqrt{\frac{\pi}{2z}} I_{3/2}(z) = -\frac{\sinh z}{z^2} + \frac{\cosh z}{z},$$

and

$$f_2(z) = \sqrt{\frac{\pi}{2z}} I_{5/2}(z) = \left(\frac{3}{z^3} + \frac{1}{z}\right)\sinh z - \frac{3}{z^2}\cosh z.$$

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