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THE MAGNETIZATION OF A RANDOM ASSEMBLY OF INTERACTING MOMENTS

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The magnetization in a field of 1 Oe, and subsequent demagnetization in zero field of a random assembly of single domain magnetite particles as a function of time has been measured. The effect of interparticle interactions is clearly evident. The results are in good agreement with a mean random field theory.

A DESCRIPTION of the internal fields of a random assembly of interacting particles is important for an understanding of the properties of spin glasses, dilute solid solutions of polar molecules, and rocks. We present results of experiments on a sample consisting of artificial single domain magnetite particles which clearly show the effect of a fluctuating random internal field. Such fields are central to many theories of spin glasses [1, 2]. However direct experimental verification is difficult due to a lack of a complete description of the spin glass phase.

The magnetic viscosity of assemblies of single-domain grains is a subject which is almost fifty years old. The central result of all the work is that

$$M = S \ln t,$$

where M is the magnetization, t the time and S the "viscosity coefficient". Here we wish to report on a study in which the relationship between M and $\ln t$ is clearly non-linear.

A study of the temperature dependence of the magnetic viscosity of assemblies of single and multi-domain grains has recently been published [3]. It was possible to account [4] for the data on single domain grains using Néel's theory [5] with the exception of a curvature in the plot of M against $\ln t$. Neel's theory predicts a straight line i.e. M proportional to $\ln t$. An extension of the theory [6] predicts that M should in fact be proportional to $(\ln ct)^n$ where c is a rate constant $\sim 10^8$ Hz [5] and n is determined by the grain size distribution. This will lead to curvature, but because c is so large in comparison to t the curvature is very small. However in the case of the samples of single domain grains the size distribution is known and could not account for the curvature. The possibility existed that this might be due to inter-particle interactions. The samples used in the study of [3] were produced by dispersing the magnetite powder in a non-magnetic matrix. It is very difficult to avoid some "clumps" of magnetite in such a sample. If this is the case, then a sample of the pure magnetite powder should show increased curvature.

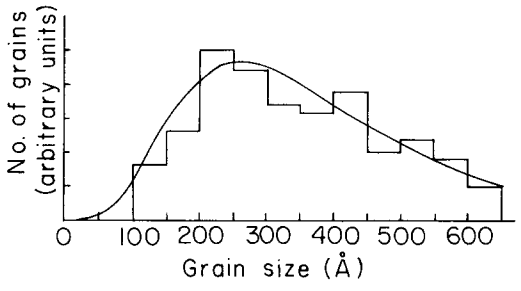

Fig. 1. Particle size distribution as determined by electron microscopy. The solid line is a fit to a log-normal distribution.

Our sample consisted of artificial magnetite grains whose size distribution, as determined by electron microscopy is shown as a histogram in Fig. 1 [7]. The solid line is the best-fitting log-normal distribution function. The sample consisted of approximately 0.5 gms. of the material packed in a non-magnetic container.

The magnetization as a function of time was measured at room temperature using a S.Q.U.I.D. 2d. order gradiometer. Unfortunately it was not possible to change the temperature without the appearance of irreversible changes in M due to mineral alteration of the sample. Therefore only data at room temperature were obtained. Results are only shown for a single field; the field dependence of the magnetization is identical to that which is commonly observed in low (< 10 Oe) fields, namely that $\partial M/\partial \ln t$ is proportional to H, where H is the applied field.

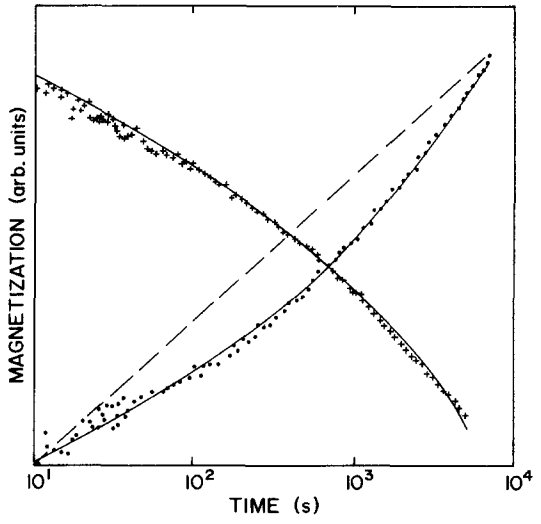

Fig. 2. Experimental results for the magnetization (dots) in a field of 1 Oe, and subsequent demagnetization (+) in zero field. The dashed line is the expected behaviour of a non-interacting system. The solid lines are calculated using the mean field theory of Klein [1].

The results are shown in Fig. 2, plotted against the logarithm of the time. The dots are measured values of magnetization in an applied field of 1 Oe The crosses are the decaying moment in zero field. The expected behaviour of a dispersed assembly is indicated by the dashed line. This line shows a small amount of curvature which is predicted by theory [6]. This is due to the particle size distribution. For a detailed analysis the reader is referred to [6]. Our sample behaves quite differently, showing a pronounced curvature. The difference must be due to interparticle interactions. The solid lines in Fig. 2 are the result of the following calculation:

Our sample consists of a large number of particles of varying size with random orientations. The magnetic moment of a particle of volume V is JV where J is the saturation magnetization. It is assumed that there are two possible antiparallel orientations for the moment. The interparticle interaction is due to the magnetic dipole field produced by the neighbours of a particle. This problem is exactly the one considered by Klein in a number of publications [8]. Klein has solved both the Heisenberg and the Ising model in the random field approximation. Since our particles are fixed in space, and only one component of the field is of interest, it is the Ising solution which is appropriate [9]. We closely follow Klein's treatment.

The probability distribution of the internal field h is given by

$$P(h) = \exp(-h^2/D^2)2/\sqrt{\pi}D. \quad (1)$$

h, in other words, is treated as a Gaussian random variable. Klein [1] finds that $P(h)$ is a Lorentzian for small h, but for computational convenience we will preserve the Gaussian form for all h.

$$D = (2\pi/3)^{1/2} g\langle m\rangle,$$

where g is the field at a particle due to its neighbour multiplied by the number of nearest neighbours. In the present case the particle size is variable, as is the nearest neighbour distance, so these quantities are to be replaced by suitable averages. $\langle m\rangle$ is the average of the absolute value of the magnetization per particle divided by the particle moment. The magnetization of the sample, M is zero in zero field, but m is not:

$$\langle m\rangle = \int_0^\infty N(V)\mathrm{d}V \int_{-\infty}^{\infty} P(h)|n(V,h+B)|\,\mathrm{d}h, \quad (2)$$

where B is the external field, $N(V)\mathrm{d}V$ is the number of grains whose volume lies between V and $V + \mathrm{d}V$ and n is the distribution function.

Because we are dealing with nearly superparamagnetic systems the evaluation of 2 is complicated by the fact that n is time dependent. This is also the case for the magnetization

$$\langle M\rangle = \int JVN(V)\mathrm{d}V \int P(h)\, n(V, h+B)\mathrm{d}h. \quad (3)$$

n is obtained from

$$\mathrm{d}n/\mathrm{d}t = \tanh(x+b) - n)/\tau \quad (4)$$

where $x = JVh/kT$, and $b = JVB/kT$

$\tanh(x + b)$ is the equilibrium distribution function towards which n is relaxing. Actually this is only an approximation to the true distribution which lies somewhere between a Langevin function and the tanh function.

The solution to 4 is

$$n = n(0)\exp(-t/\tau) + \tanh(x+b)[1 - \exp-(t/\tau)] \quad (5)$$

and τ^{-1} is $\tau^{-1} = c\exp(-KV/kT)$ where K is the anisotropy constant; in this case it is due to the shape of the particles. KV is the energy barrier which must be overcome when the particle moment flips from one orientation to the other. c is an attempt frequency on the order of 10^8 Hz. $n(0)$ is the initial distribution, in this case $n(0) = 0$.

As shown in Fig, 1 the size distribution $N(V)$ is well fit by a log normal distribution,

$$N(V) = (A/V)\exp[-(3\ln(V/V_0))^2] \quad (6)$$

where V_0 is 4.3×10^{-17} cm^3, and A a constant proportional to the concentration.

The double exponential in 5 restricts the values of V so the logarithm in 6 does not change very much over the region of interest. We approximate $N(V)$ by C/V where

$$C = A\exp-(3\ln\bar{V}/V_0)^2 \cong 0.2A.$$

Equation 5 is essentially a step function, and integration over V need only be taken to a maximum value

$$V_m = (kT/K) \ln ct \tag{7}$$

At room temperature JVD/kT turns out to be less than 1, $P(h)$ is small for $h > D$, and $\tanh(x + b)$ may be replaced by its argument. Equation 2 then becomes

$$\langle m \rangle = \int_0^{(kT/K)\ln ct} (C/V)\, dV (2JV/kT) \left[\int_B^{\infty} (h/D\sqrt{\pi}) \exp -(h^2/D^2)\, dh + \int_0^{B} (B/D\sqrt{\pi}) \exp -(h^2/D^2)\, dh \right]. \tag{8}$$

Approximating the resulting error functions by their leading terms, and letting $K = 1/2\, JH_c$

$$\langle m \rangle = 2C(B/H_c) \ln ct. \tag{9}$$

Klein [10] obtains an analogous result for high temperatures.

To calculate M we again expand $\tanh(x + b)$, this time retaining terms in Bh^2 and Bh^4; we neglect terms in powers of B greater than one because B is small:

$$M = \int_0^{(kT/K)\ln ct} JC\, dV \int_0^{\infty} 2/\sqrt{\pi} D\, e^{-h/D}\, 2B \times \{(JV/kT) + (JV/kT)^3 h^2 + (JV/kT)^5 h^4\}\, dh.$$

On integrating

$$M = 2kTC(\ln ct)^2\, B\{1 + (d \ln ct)^2 + 2(d \ln ct)^4\} \tag{10}$$

where $d = D/H_c$.

Since D is proportional to $\ln ct$ from equation 9, in this approximation the corrections are proportional to the fourth and eighth power of $\ln ct$ respectively.

After the sample was held in field for a time t_f the field was turned off. At the moment the field was turned off the distribution function was (from 5)

$$n_0 = \tanh(x + B)\{1 - \exp[-ct_f \exp(-KV/kT)]\} \tag{11}$$

The field is now zero so the equilibrium distribution is zero; therefore equation 5 becomes

$$n = n_0 \exp[-ct \exp(-KV/kT)]$$

with n_0 given by equation 11.

Following the same procedures used in deriving 10

$$M_{\text{demag}} = 2kTC(\ln t_f/t)^2 B \times \{1 + (d \ln t_f/t)^2 + 2(d \ln t_f/t)^4\}.$$

Without the terms in the square brackets the expressions are those for non interacting grains, which agrees well with experiment. The terms in d therefore, contain the effect of the random field.

In order to compare equation 11 with experiment values for H_c and g are required. For this material H_c is equal to 175 Oe [7]. It is difficult to obtain a precise value for g, but it should be approximately equal to the field at the average nearest neighbour distance. If this distance is set equal to twice the diameter of the average grain, g is about 50 Oe. The fit in Fig. 2 was obtained by allowing g to vary; the value in that case is 20 Oe. The discrepancy could possibly be due to oxidation of the magnetite grains [7].

In general spin-glasses, at least at low concentrations of the magnetic species, show the standard $M = S \ln t$ behaviour [11]. No clear evidence of curvature exists in the data of reference [11]. On the other hand it is interesting to note that Chamberlin, Mozurkewich and Orbach [12] have recently observed the time decay of thermal remanent magnetization in spin glasses that is similar to the demagnetization data shown in Fig. 2. Namely, both sets of data show an accelerating rate of decay with increase in $\ln t$. In their data the curvature increased as the temperature was lowered below the spin-glass transition. The following is a completely speculative application of the random field model used here to that case. Assume that the spin glass phase consists of a large number of spin clusters. The size of a cluster would be determined by the number of spins whose interaction energy exceeded kT. The size of the clusters would be expected to increase as the temperature decreased since this would allow the more weakly coupled spins to be bound. Thus close to the transition the clusters would be few and small, because only the strongly interacting spins could order. The behaviour would then be the same as that of a non-interacting system, and a plot of M against $\ln t$ would show only a slight curvature. Well below the transition temperature the clusters would be larger and more numerous, and these moments would be effective in providing significant local fields, leading to curvature in the plot of kT against $\ln t$.

We have observed the effect of interparticle interactions on the magnetization of a random assembly of single domain magnetite grains. The experimental results are in good agreement with a mean random field theory.

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