Dipolar-glass behaviour of an insulating film containing nanogranular Fe particles
Norberto Majlis* and Martin J. Zuckermann**

* Physics Department, McGill University, 3600 University St., Montreal, QC H3A 2T8, Canada.
** Department of Physics, Simon Fraser University, 8888 University Boulevard, Burnaby BC V5A 1S6 Canada.
E-mail: martinz@sfu.ca
majlisn@physics.mcgill.ca

Abstract
We perform Metropolis Monte Carlo simulations of the behaviour of a film of insulating material containing a distribution of magnetic nanoparticles. We assume that these particles only interact through dipolar forces and we find that their behaviour at low $T$ shows characteristics of a spin-glass with a freezing $T_f$ at which the linear susceptibility and the specific heat show a maximum. We obtain the spin glass order parameter as a function of temperature and we also calculate the time auto-correlation of the spin at the center of the system. We find that these results are consistent with the temperature dependence of the variance and the mean of the local field at the central spin.

1 Introduction
At present there is growing interest in low dimensional magnetic systems. In particular, many studies are being conducted on granular systems, consisting of nanometric clusters of a magnetic metal dispersed in a non-magnetic solid matrix. Such systems have received much attention because of their potential application to ultra-high magnetic storage capacity. When the matrix is electrically conducting (metal or semiconductor) the composite system can exhibit collective behaviour due to RKKY interactions. In some cases these systems are ferromagnetic, with a critical $T_c$ in the room temperature range. Some of these films also display giant magnetoresistance.

When the host matrix is an insulator, or in general when the charge carrier concentration in the host is very low, the indirect RKKY exchange interactions between the magnetic particles can be neglected. If the average inter-particle separation is appreciably larger than the average particle size we can also neglect the effect of super-exchange, since the latter requires that the magnetic particles be in direct contact. In such a case, which is exemplified by some of the composite films mentioned above, the collective behaviour of the system is controlled by the magnetostatic (dipolar) interactions at low enough temperatures.

Detailed numerical calculations of the RKKY and the dipolar interactions between clusters of varying sizes show that for realistic cases the latter can be as important or even more...
important than the RKKY indirect exchange interactions for realistic situations and, furthermore, that the magnetostatic interactions can be well approximated by substituting each cluster by a point magnetic dipole located at its center.

Experimental work on the dynamics of several different systems in which Fe particles are dispersed shows that dipolar interactions can control the behaviour of the dynamic susceptibility for an adequate range of particle diameter and concentration. When the strength of the inter-particle interactions is increased, either by increasing the concentration of the particles or their radius, and hence their magnetic moment, the dynamic behaviour transitions from an interaction-modified super-paramagnetism to a glassy-type collective dynamics\[7, 8, 9\].

We have therefore chosen to study an assembly of point dipoles located at fixed random positions inside a non-magnetic matrix. The only restriction on the space configurations is that the distance between the centres of each pair of particles is greater than their diameter. Their magnetic moments can orientate freely because we neglect the effects of magnetic anisotropy. A Monte Carlo (MC) simulation of a planar triangular lattice of nanoparticles with random anisotropy and interacting through dipolar forces has shown the interplay of both energies.\[10\]

In this article we present the results of Metropolis Monte Carlo (MMC) simulations for a thin film containing a completely random but fixed spatial distribution of point particles, representing bcc Fe clusters, which only interact through magnetic dipolar forces. The MMC algorithm itself is described in the next section and the simulation data is exhibited and discussed in the following sections. Emphasis is placed on the spin glass nature of the results.

2 Model and Method

We incorporate the periodic boundary conditions in the film by introducing a square lattice of cells and we then randomly distribute \( N = n_x^2 n_z \) particles inside each cell. Here \( n_y = n_z \). This basic cell is then repeated indefinitely along the \((x,y)\) plane. The total dipolar energy of the film is given by:

\[
W = \frac{1}{2} \sum_{r_n} \sum_{i,j=1}^N \mu_i \cdot D(R_{ij} + r_n) \cdot \mu_j
\]  

where \( r_n \) is a site in the square \((x,y)\) lattice and the \( \alpha, \beta \) components of the dipolar tensor \( D \) are

\[
D^{\alpha,\beta}(R) = -\nabla_{R_\alpha} \nabla_{R_\beta} \frac{1}{|R|}
\]

Here frustration, which is a necessary ingredient for glassy behaviour, is a consequence of the disorder in the positions and orientations of the dipoles, resulting in random sign and amplitude fluctuations of the tensorial dipolar interaction. We have chosen a fixed value of \( n_z = 3 \), which corresponds to a width of the film equal to \( 3 \times f \times d \), where \( d = 10\text{Å} \) is taken to be the particle diameter and \( f > 1 \) is a factor which determines the average inter-particle
distance. We choose \( f = 2 \).

Dipolar sums are calculated by the adaptation of Ewald’s summation algorithm to the quasi-two-dimensional case and the MMC algorithm is used to calculate both thermodynamic and local properties of the system. All quantities are averaged for every temperature over 100 -200 different random space configurations of the particles. The MMC runs consisted of 80000 steps for the warming cycle, and 40000 steps for the actual calculation.

We place a particle at the center of the film cell in all cases and we calculate the average and the variance of the components of the local field \( B_0 \) and of the central spin \( S_0 \) for this site. In addition the following physical variables are calculated as functions of temperature for different numbers of particles in the basic cell: the total energy per particle, the susceptibility tensor and the specific heat. Their values are then extrapolated to obtain the limit of an infinite film of \( n_z = 3 \), namely the width corresponding to 3 average distances. We also obtain the averaged time auto-correlation function \( g(t) \) of the central particle spin \( S_0 \), defined as

\[
g(t) = \langle \langle S_0(t_0) \cdot S_0(t_0 + t) \rangle \rangle_{t_0}
\]

where an average over the initial time, \( t_0 \), is performed for each statistical average. Note that in our calculations time, \( t \), is defined as the number of MMC steps as the simulation progresses. This implies from the definition above that we calculate the scalar product of the value of central spin, \( S_0 \), after \( t_0 \) MC steps with the value of the central spin after a further \( t \) MMC steps. The time scale is not otherwise defined, being dependent on the spin-flip physical time, which in real systems is of the order of \( 10^{-13} \) sec. From the original Anderson definition of the order parameter \( q_{EA} \) we have:

\[
q_{EA} = \lim_{t \to \infty} g(t) \bigg|_{t \to \infty}
\]

An alternative order parameter has been defined as follows [11]:

\[
q_H = \frac{1}{N} \sum_{i=1}^{N} \left( \sum_{\alpha=x,y,z} \frac{1}{\tau} \sum_{t'=t_w} S_{0i}^\alpha(t') \right) \left( \sum_{t'=t_w} S_{0i}^\alpha(t') \right)^2 \right)^{1/2}
\]

We calculate both order parameters for every value of the temperature \( T \).

3 Thermodynamic properties

At \( T = 0 \) order parameters defined in the previous section should equal 1 while they are expected to vanish above the freezing temperature \( T_f \). However due to the system’s finite size, both \( q_{EA}(T) \) and \( q_H(T) \) have a long tail for high \( T \) as can be seen in figures 1 and 2.

One can fit the curves for \( q_{EA} \) and \( q_H \) vs. \( T \) with an algebraic function such that they intersect the temperature axis at an extrapolated temperature \( T^{(\alpha)}_{q}(N) \) ( \( \alpha = \{EA, H\} \))
Figure 1: The order parameter $q_{EA}$ as a function of $T$ in degrees Kelvin for three different sizes.

Figure 2: The order parameter $q_H$ as a function of $T$ for different sizes.

which is an estimate of the transition temperature from the spin-glass state to the disordered (super-paramagnetic) phase, for every value of $N$. We fit the low $T$ curves for
both order parameters with the function:

\[ f(T) = (1.0 - T/T_q)^\nu \]  

which for low \( T \) behaves as

\[ f(T) \approx 1.0 - \nu T/T_q \]  

The freezing temperatures thus obtained for both order parameters are in good agreement with each other, indicating that both definitions are consistent. However, \( q_{EA} \) is amenable to a better extrapolation, since it is less sensitive to the finiteness of the sample used in the simulation. At any rate, one has to cutoff the data at a reasonable value of \( T \approx 1.4 - 1.6 K \), since as mentioned above the order parameters for a finite system do not vanish at any finite \( T \). This arbitrary procedure introduces an uncertainty in \( T_q \) of the order of 0.1 – 0.2K. In Fig. 3 we show extrapolated values of \( T_q \) for three different values of \( N \). From these data we estimate:

\[ \lim_{N \to \infty} T_q \approx 1.79K \pm .07 \]

\[ \lim_{N \to \infty} \nu \approx 0.42 \pm .07 \]

so that at low \( T \) we get

\[ q_{EA} \approx 1.0 - 0.42T/T_q \]  

It is noteworthy that the mean field solution of the \( EA \) model of a spin glass, as mentioned by Mydosh [12] yields at low \( T \) :

\[ q_{EA}(T) = 1.0 - 0.4066T/T_f \]  

in close agreement with our result in Eq. 7. We also obtain an estimate of \( T_f \) by calculating, following Binder [13], the kurtosis of the distribution of the total magnetization as a function of \( T \), defined as

\[ cumul(T) = 1.0 - \langle |\vec{M}|^4 \rangle / 3 \langle |\vec{M}|^2 \rangle^2 \]  

The temperature at which the plots of this quantity vs. \( T \) intersect for different values of \( N \) should give an estimate of the freezing temperature for infinite size. It should be noted that the kurtosis is zero for a Gaussian distribution.

The specific heat is shown in Fig. 4 as the variance and in Fig. 5 as the numerical derivative, of the energy. Just as for the parallel static susceptibility curve in Fig. 6 they show a maximum at a temperature \( T_m(N) \) slightly lower than \( T_q(N) \). These results are compatible with spin-glass behaviour [14].

The graphs of the Binder cumulant combination, defined in Eq[9] as a function of \( T \) for three different sizes intersect approximately at a temperature \( T_c(N) \) slightly lower than \( T_q(N) \). These results are considered to be an estimate of \( T_f \). From Fig. 7 we obtain \( T_c \approx 1.5 K \).
We verify (Fig. 8) that the average modulus of the magnetization scales as $N^{-1/2}$ as corresponds to a completely random dipole distribution.

The energy per particle is shown in Fig. 9 for three different values of $N$. We verify that
Figure 5: The Specific Heat (Temperature derivative of the energy) as a function of $T$.

Figure 6: The Static Longitudinal Susceptibility as a function of $T$.

It converges as $N$ increases.
Figure 7: Binder cumulant expression vs. $T$ for different sizes.

Figure 8: Magnetization per particle vs. $T$ for different sizes.
4 Local properties

Besides the thermodynamic properties we have studied several local characteristics of the system. The time auto-correlation function $g(t)$ mentioned above was obtained for the central spin. A plot of $g(t)$ for $N = 108$, $T = 2.9K$ and 160 configurations is shown in Fig. 10. By definition $g(0) = 1$ at all temperatures.

The best fit to $g(t)$ was found to be

$$g(t) = b(T) + (1.0 - b(T)) \exp \left(-t/\tau(T)\right)$$

(10)

where, as indicated above, both parameters are functions of $T$. Our results for $g(t)$ at different temperatures allow us to obtain the functions $b(T)$ and $\tau(T)$, shown in Figs. 12 and 11 with the standard statistical error (SSE) bars of the fit.

From Eq. 3 we must have

$$b(T) = q_{EA}(T)$$

which we verify by comparing with Fig. 1. This is a consistency requirement on the fit to $g(t)$. The characteristic decay time $\tau$ of the transient term in Eq. 10 is shown as a function of $T$ in Fig. 12.

We see that as $T \to 0$ $g(t)$ tends to 1 while the decay time of the transient vanishes. As $T$ increases above $T_f$ the transient gets longer and the limiting amplitude decreases, and should eventually vanish. One again finds an abrupt change in the derivative of the curve for $\tau(T)$ which is the signal of the freezing temperature.
As a necessary step in the calculation of the energy and the specific heat we need to obtain the local field on every particle. The statistical distribution of the values of all three components of the field can be obtained as the MMC run proceeds, and we obtained their average and
variance from this distribution.

Fig. 13 shows that the modulus of the local field on the central spin *increases* as $T$ lowers, while its variance, shown in Fig. 14 *decreases* rapidly for $T$ below the freezing temperature $T_f \approx 1.5 - 1.6 \text{ K}$.

The central spin exhibits a similar behaviour: Figure 15 shows that the variance of the $x$ component of the central spin decreases abruptly below $T_f$.

We conclude that below $T_f$ each spin orientates in a given average direction, around which it fluctuates with a decreasing variance as $T$ decreases. This is shown by the change in the derivative shown in Fig 15. On the other hand, both the specific heat (Fig 4) and the static susceptibility (Fig. 6) show a maximum at the same $T_m$.

5 Conclusions

We present a simulation of the behaviour of a collection of nano-particles sustaining a magnetic dipole moment dispersed randomly in a non-magnetic film (the same conclusions are valid for electric dipoles, like for instance in a liquid crystal). We focused our study on the range of high concentrations, where collective behaviour can be expected.

We find that such a system exhibits at low temperatures a freezing transition similar to that of a spin-glass, as shown by the temperature dependence of both local and global statistical properties, namely:
Figure 13: Modulus of Local Field as a function of $T$

Figure 14: Variance of Local field vs. $T$

a) one can define an order parameter which is unity at very low $T$ and decreases as $T$ increases;

b) each magnetic dipole at low $T$ tends to orientate in a fixed direction, around which the
amplitude of its fluctuations decreases as $T$ lowers, which is shown by the correspondingly decreasing variance and increasing average amplitude of a given spin component. Besides, the directions along which the dipoles freeze at low $T$ are random, which is shown by the fact that the total magnetization scales with the number of particles as $N^{-1/2}$;

c) both the magnetic specific heat and the longitudinal static susceptibility show maxima at about the same temperature, which is close to the estimated freezing temperature obtained by the Binder criterium based on the $T$ dependence of the kurtosis of the statistical distribution of the magnetization;

d) the local magnetic field on a given spin starts increasing in amplitude and decreasing in variance as $T$ lowers below a given $T$, which coincides with the one at which the same properties occur for the spin components, and which we interpret as the temperature $T_f$ of the freezing transition.

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