Abstract
It is shown that the Ngai model of relaxation accounts for the main experimental results of the magnetic relaxation in an assembly of interacting fine particles.
1 Introduction

The Ngai model for relaxation in complex systems was first introduced in 1979 [1, 2]. Since then, it has been found to offer an accurate description of a wide variety of systems [3]. However, its use in magnetism is more recent. See reference [4] and other references therein.

The fundamental difference between the Ngai model and previous models of relaxation is that these earlier models treat the effect of complexity on the relaxation as being essentially static, in the sense that the effect of complexity is to change the relaxation time or to produce the presence of a distribution of relaxation times. In contrast to this, in the Ngai model, the relaxation in complex systems, is dynamical in nature.

From this viewpoint, according to Ngai [5], a relaxing complex system consists of three parts:

i) Individual primary species (PS) of which are of interest for the relaxation.

ii) A heat-bath whose interaction with the PS provides a primary mechanism of relaxation.

iii) Other relaxing species, X, whose interaction with PS, the PS-X coupling, slows down the relaxation process. This is the main manifestation of complexity.

In this paper we propose another application of the Ngai model to a magnetic system: namely that of an interacting fine-particle assembly. In this system, the PS are the individual particles whose coupling with the other particles in the assembly slows down the relaxation process. In preceding attempts to tackle this problem [7, 8], the only effect of the presence of interparticle interactions is a change in the relaxation times. A discussion of these results in the light of the present model, together an evaluation of experimental results using this model, are also presented.

In this paper we show that many of the experimental results for an interacting system of small particles come out naturally within the Ngai model. In particular the model can explain, in a natural way, not only the observed differences for a system of interacting and non-interacting particles via the variation of the inverse of the blocking temperature with the logarithm of the measuring time, but also the values encountered for the relaxation times (table 1). The organization of the paper is as follows: in section 2 we review the Ngai model for the relaxation in a complex system. In section 3 we apply
this model to a system of interacting particles and show that the behaviour discussed previously can be accounted for by this model. Section 4 is devoted to the calculus of the magnetic viscosity in two different extentions of the model, the case of a distribution of particle-sizes and also the case of a distribution of switching fields. In the former, we encounter the ubiquitous logarithmic relaxation law, whereas in the latter our results agree with those of Charap [10] and therefore the model is compatible with the experimental results of Oseroff et al [11].

2 The Ngai model

After the justification of the necessity of the study of non simple-exponential relaxations we summarize briefly the more important results of the Ngai model. For more information about the theoretical derivations and the experimental successes of the model we refer to the bibliography [3].

It was shown by Ngai et al [6] that all the permissible relaxation functions have to verify the Paley-Wiener theorem of the Fourier transform theory. It is easy to show that the simple exponential relaxation function violates this theorem. Since a single, linear exponential form is unphysical, as demonstrated, the idea of a superposition of exponentially decaying functions may be also ruled out as a description of relaxation phenomena. There is therefore the necessity to move on non simple-exponential relaxations.

The Ngai model [3] proposes the existence of a temperature-insensitive crossover time, $t_c$, separating two regimes in which the dynamics of the relaxation are different. The relaxation function is a linear exponential $\exp\left(-\frac{t}{\tau_0}\right)$ for $t < t_c$ and cross-over at $t \approx t_c$ to a stretched exponential form $\exp\left(-\left(\frac{t}{\tau_\star}\right)^{(1-n)}\right)$ for $t > t_c$. The coupling parameter $n$ lies in the range $0 < n < 1$. Notice the appearance of two different characteristic time-scales, $\tau_0$ and $\tau_\star$, for the two regimes. These two time-scales are not independent, but continuity of the relaxation function at $t = t_c$ implies that:

$$\tau_\star = \left(t_c^{(1-n)} \tau_0\right)^{\frac{1}{1-n}}$$  \hspace{1cm} (1)

Notice an important consequence of equation 1 which will be important in the following, if $\tau_0$ describes the physics of some motion over an energy barrier
with activation energy $E_A$:

$$\tau_0 = \tau_\infty exp\left(\frac{E_A}{kT}\right)$$

(2)

Then for equation (1), $\tau_*$ describes a macroscopic relaxation time given by:

$$\tau_* = \tau^*_\infty exp\left(\frac{E^*_A}{kT}\right)$$

(3)

where:

$$E^*_A = \frac{E_A}{1 - n}$$

(4)

and:

$$\tau^*_\infty = \frac{1}{\tau^*_\infty} \frac{n}{t_c}$$

(5)

Where $E^*_A > E_A$.

3 Application of the Ngai model to the relaxation of a system of small interacting particles

There are several excellents treatment of the magnetic properties of an ensemble of particles. We shall, therefore, not enter into detail here [12, 13].

We consider, firstly, an ensemble of particles of equal volume $V$. For simplicity we assume that the particle anisotropy is uniaxial and that the particle’s easy axis of magnetization is parallel to the field direction. Then the energy of one of the particles in a field $H$ will be [14]:

$$E = KV \sin^2 \theta - M_s V H \cos \theta$$

(6)

where $K$ is the anisotropy constant, $M_s$ is the bulk saturation magnetization and $\theta$ measures the orientation of the magnetic moment of the particle with respect to the field. This energy gives rise to two energy minima separated by an energy barrier:

$$E_A = \frac{V}{2} M_s H_K (1 - \frac{H}{H_K})^2 = e_A V$$

(7)
At a finite temperature, we have a finite probability of a thermally activated motion over the energy barrier. The characteristic time-scale of the motion will be given by the expression:

\[ \tau_0 = \tau_\infty \exp\left(\frac{E_A}{kT}\right) \]  

where \( \tau_\infty \approx 10^{-9} - 10^{-12} \text{s} \). For such a system, the Ngai model predicts the existence of a cross-over time, \( t_c \), independent of temperature, to be determined from the experiment, separating two different regimes of the dynamics of the relaxation. The relaxation function is a linear exponential \( \exp(-t/\tau_0) \) for \( t < t_c \) and has a stretched exponential form \( \exp\left(-\left(t/\tau_\ast\right)^{1-n}\right) \) for \( t > t_c \), where \( 0 < n < 1 \). The relation between \( \tau_\ast \) and \( \tau_0 \) is given by equation 1.

An important consequence of the model can be now pointed out if, as a definition of the blocking temperature, we use the usual criterion that the characteristic time-scales of the relaxation will be of the order of the measuring time. We then expect that the logarithm of the measuring time plotted as a function of \( 1/T_B \) will be a “piece-wise” straight line with two different slopes, \( \frac{E_A}{k_B} \) for \( t < t_c \) and \( \frac{E_A}{k_B(1-n)} \) for \( t > t_c \). The slope corresponding to the small measuring times will be smaller than that corresponds to the longer measuring times. This fact was encountered experimentally in a detailed analysis of the dynamics of a system of small iron particles, magnetically interacting and dispersed in an amorphous alumina matrix by Dormann et al \[8\]. We reproduce here data from a figure from that paper, figure 2, which shows the predicted behaviour (figure 1). From this figure, we estimate that \( t_c \) is in the range \( 10^{-10} - 10^{-5} \text{s} \). However these results are not immediately comparable with the above developed theory because they correspond to samples with different sizes and interactions.

A better comparison with the theory can be made with the data of J. L. Dormann et al \[15\] in a system consisting of \( \gamma - Fe_2O_3 \) particles dispersed in polyvinilic alcohol (figure 2). In this case we have a series of samples with the same size and different interactions including the case of zero interaction (sample IF). The interaction increases from right to left. From the sample with zero interaction we can estimate \( \tau_\infty \approx 10^{-10} \text{s} \). By comparing the slopes of sample IF with the slopes of the other samples, we can calculate the value

\[ \tau_0 = \tau_\infty \exp\left(\frac{E_A}{kT}\right) \]
of \( n \). Using these values of \( n \), the afore-mentioned value of \( \tau_\infty \) and choosing \( t_c \approx 10^{-9} \), we can calculate theoretically the value of \( \tau^*_\infty \). These values are compared with the experimentally obtained values in table 1. We observe excellent agreement between theory and experiment.

Here we wish to stress the importance of this fact. The existence of the cross-over time, which separates two different time dependences with time-scales related by equation 5, is a stringent condition for the verification of the Ngai model. This is the first time that this cross-over has been encountered in a magnetic system.

A further important consequence is that the Ngai theory can explain the discrepancy between the size of the particles measured using two different time scales. This will be investigated in a further publication [16].

After showing that the Ngai can describe the relaxation of a system of small interacting particles we dedicate the remainder of this paper to the extension of the model to a calculation of the magnetic viscosity in the case of a distribution of particle sizes and a distribution of switching fields.

## 4 Calculation of the magnetic viscosity

A typical viscosity measurement in a small particle assembly runs as follows: After saturation in a strong field, a small measuring field is applied in the reverse direction. The magnetization is then measured as a function of time. Typical times between application of the saturation and the first measurement are of the order of seconds and the time-scale of the measurement is of the order of minutes. In the previous section, we estimate that \( t_c \approx 10^{-10} - 10^{-5} \) s (figure 1) and thus we expect that the relaxation function

| Sample | \( n \) | \( \tau^*_\infty (th) \) (s) | \( \tau^*_\infty (exp) \) (s) |
|--------|-----|----------------|----------------|
| CH     | 0.4 | \( 4 \times 10^{-12} \) | \( 10^{-12} \) |
| IN     | 0.73| \( 3.87 \times 10^{-16} \) | \( 10^{-16} \) |
| FLOC   | 0.76| \( 3 \times 10^{-17} \) | \( 10^{-17} \) |
Figure 1: Variation of the blocking temperature, $T_B$, with the logarithm of the measuring time for different samples of iron particles, according to [7]. The average particle size and the interparticle interaction strength increases on going from right to left.
Figure 2: Variation of the blocking temperature, $T_B$, with the logarithm of the measuring time for different samples of $\gamma - Fe_2O_3$ particles, according to [14]. The interparticle interaction strength increases from right to left. The average particle size is held constant.
for the present experiment will be of the stretched exponential type. Using the boundary conditions, \( M(t = 0) = -M_0 \) and \( M(t = \infty) = M_0 \) the time dependence of the magnetization can be written in the form:

\[
M(t) = M_0 - 2M_0 \exp\{-\left(\frac{t}{\tau_\star}\right)^{1-n}\}
\]  

(9)

We now extend the model to two cases: distribution of particle sizes and distribution of switching fields.

### 4.1 Distribution in the particle size

We now consider the fact that we have a distribution in the particle size, \( f(V) \) such that:
\[
\int_{0}^{\infty} f(V) dV = 1
\]  

(10)

In this case, our relaxation law transforms to:
\[
\frac{M(t)}{M_0} = \int_{0}^{\infty} (1 - 2 \exp\{-\left(\frac{t}{\tau_\star}\right)^{1-n}\}) f(V) dV
\]  

(11)

As a simple example we consider the distribution \[18\]:
\[
f(V) = \begin{cases} 
\frac{1}{V_2 - V_1} & V_1 < V < V_2 \\
0 & \text{otherwise}
\end{cases}
\]  

(12)

For this distribution function, the relaxation function can be evaluated in terms of the exponential integral:
\[
E_i(x) = \int_{x}^{\infty} \frac{e^{-y}}{y} dy
\]  

(13)

A simple calculation gives:
\[
M(t) = M_0 - 2M_0 \frac{kT}{e_A(V_2 - V_1)} (E_i(z_2) - E_i(z_1))
\]  

(14)

where:
\[
z_i = \left(\frac{t}{\tau_\star \exp\left(\frac{e_A V_i}{kT}\right)}\right)^{1-n} \quad i = 1, 2
\]  

(15)
We stress here that this opens up a new possibility for the analysis of the relaxation data.

For $V_1$, $V_2$, $n$ and $t$ such that $z_1 \gg 1$ and $z_2 \ll 1$ we have:

$$M(t) = M_0 - \frac{2M_0k_BT}{e_A(V_2 - V_1)}((1 - n)\log\tau_2 - 0.577 - (1 - n)\log t)$$  \hspace{1cm} (16)$$

where:

$$\tau_2 = \tau^{\star}_{\infty}e^{\frac{e_AV_2}{k_BT(1-n)}}$$  \hspace{1cm} (17)$$

We have therefore encountered in this limit the ubiquitous logarithmic relaxation. In the literature, the parameter $S$ is usually defined by:

$$S = \frac{dM(t)/M_0}{d\log t}$$  \hspace{1cm} (18)$$

which in this case is given by:

$$S = \frac{2k_BT(1-n)}{e_A(V_2 - V_1)}$$  \hspace{1cm} (19)$$

Two important comments can now be made. Firstly, the value of $S$ depends on $n$ and therefore on the particle interactions and secondly, there are several possibilities to obtain the logarithmic relaxation: (i) from equation (9) and, assuming that $(1 - n)\log(t/\tau_\star)$ is very small, we have:

$$M(t) = M_0 - 2M_0e + 2(1 - n)M_0elog(t/\tau_\star)$$  \hspace{1cm} (20)$$

and (ii) assuming a distribution of volume sizes.

### 4.2 Distribution of switching fields.

Following Charap\cite{Charap}, we consider an ensemble of interacting magnetic particles with uniaxial anisotropy, thus with a switching barrier given by equation 7 and a switching field distribution $p(H_K)$. We can transform the $p(H_K)$ in a density of energy barriers $G(E^*_A)$ through:

$$G(E^*_A) = \left(\frac{\partial E^*_A}{\partial H_K}\right)^{-1}p(H_K)$$  \hspace{1cm} (21)$$
In the calculation of $G(E^*_A)$ we use the Charap method. For a given $H_K$, only for fields $H \approx H_K$ we encounter appreciable switching. In this range of fields we can write approximately:

$$E^*_A \approx \frac{V}{2} M_s \frac{(H_K - H)^2}{H(1-n)} \quad (22)$$

which simplifies the evaluation of equation 21. To take a specific case, we assume a switching field distribution of Lorentzian form:

$$p(H_K) = \frac{1}{\pi \Delta} (1 + \left(\frac{H_K - H_c}{\Delta}\right)^2) \quad (23)$$

where $\Delta$ measures the half-width of the distribution at the half-maximum. Then $G(E^*_A)$ can be written as:

$$G(E^*_A) = \frac{1/\pi \Delta}{VM_s \sqrt{2E^*_A/V M_s H(1-n)(1 + (\frac{H_H + H(1-n)}{\Delta})^{2})}} (24)$$

The magnetization is now described by:

$$\frac{M(t)}{M_0} = 1 - 2 \int_0^\infty G(E^*_A) e^{-\left(\frac{t}{\tau^*}\right)^{1-n}} dE^*_A \quad (25)$$

Defining:

$$\chi = \frac{2E^*_A}{VM_s H(1-n)} \quad (26)$$

$$\beta = \frac{VM_s H}{k_B T} \quad (27)$$

we have:

$$\frac{M(t)}{M_0} = 1 - \frac{H(1-n)}{\pi \Delta} \int_0^\infty d\chi \frac{\exp\left(-\left(\frac{t}{\tau^*_\infty}\right)^{1-n}\right)}{\sqrt{\chi(1 + (\frac{H(H(1-n)\sqrt{\chi}) - H_c)^2}{\Delta})}} \quad (28)$$

and:

$$S(t) = \frac{H(1-n)^2}{\pi \Delta} \left(\frac{t}{\tau^*_\infty}\right)^{(1-n)} \int_0^\infty d\chi \frac{\exp(-\beta \chi(1-n)^2 - \left(\frac{t}{\tau^*_\infty}\right)^{1-n} e^{-\beta \chi(1-n)^2})}{\sqrt{\chi(1 + (\frac{H(H(1-n)\sqrt{\chi}) - H_c)^2}{\Delta})}} \quad (29)$$
We have calculated numerically the dependence of $S(t)$ for $t = 100s$ with the applied field at different temperatures for several values of $n$. A typical example, for $n = 0.5$, is shown in figure 3. The choice of parameters is the same as that of Charap: $\tau_0 = 10^{-9}s$, $V = 10^{-15}cm^3$, $H_c = 200Oe$, $\Delta = 10Oe$ and $t_c = 10^{-9}s$. Several comments can now be made. $S_{max}$, the maximum value of $S$, decreases with $n$ and the curve is broader than the corresponding curve for $n = 0$. We have fitted the temperature dependence of $S_{max}$ to a $T^\alpha$ law (figure 4) and have found $\alpha = 0.443$. This does not deviate significantly from the $n = 0$ estimate $\alpha = 0.478$. From a similar calculation given by Charap, we expect in the first approximation, a dependence of $S_{max}$ with $T$ of $T^{\frac{1}{2}}$. Finally, we mention that these results are in qualitative accord with the experimental results of Oseroff el al [11].

5 Conclusions

After showing that the Ngai model of relaxation can account for the main experimental results of the magnetic relaxation in an interacting fine particle assembly, we have extended the model to the case of a distribution of particles sizes and switching fields. We mention finally that we look forward to this model being used for an analysis of the magnetic relaxation in an ensemble of small magnetic particles.

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