

Effect of grain interactions on the frequency dependency of magnetic susceptibility

A.R. Muxworthy

*Institut für Allgemeine und Angewandte Geophysik, Universität München, Theresienstrasse 41,
80333 München, Germany. E-mail: adrian@geophysik.uni-muenchen.de*

Received 9th February 2000

SUMMARY

Environmental systems often contain superparamagnetic (SP) grains that cause a frequency dependency of low-field magnetic susceptibility (κ_{fd}). Previous models for κ_{fd} have been for non-interacting regimes, whereas environmental systems often display characteristics of magnetic interactions. In this paper, the magnetic susceptibility (κ) and κ_{fd} have been modelled for weakly-interacting assemblages of single domain (SD) grains of magnetite, near the SP and stable SD threshold known as the blocking volume v_b . Weak-interactions between SP grains effectively increase the anisotropy, which reduces v_b . The relationship between the grain distribution and the reduced v_b , causes a decrease in the peak values of κ_{fd} , and can reduce κ_{fd} by over 50 % for certain grain distributions. This helps to explain why κ_{fd} values for natural samples are very rarely seen above ≈ 15 %, as the effect of interactions is seen to reduce maximum $\kappa_{fd} > 20$ % in non-interacting models to values < 20 % for the same grain distribution. However, it is also found that the reduction of v_b as a result of interactions can also increase κ_{fd} for certain grain distributions. The model only accommodates weakly interacting systems, as the behaviour of strongly inter-

acting SP grains is not well understood, and no analytical formulation has yet been made.

Key words: Magnetic susceptibility, magnetic interactions, SP-SSD transition, environmental magnetism

1 INTRODUCTION

Low-field magnetic AC susceptibility (κ) measurements are routinely made in the field of environmental magnetism to help determine the magnetic mineralogy, concentration and grain size of a sample (Dearing et al. 1996a). In particular the measurement of the frequency dependency of low-field AC susceptibility (κ_{fd}) has become a standard tool to identify superparamagnetic grains (SP) near the SP and stable single domain (SSD) boundary (*e.g.*, Dearing et al. 1996b), where $\kappa_{fd} = (\kappa_{lf} - \kappa_{hf}) / \kappa_{lf}$, and κ_{lf} and κ_{hf} are the AC susceptibilities measured at a low and high frequency respectively. Being able to identify grains near the SP/SSD transition is of importance, because SP/SSD grain assemblages are very common in environmental systems, *e.g.*, Dearing et al. (1996b) found that approximately 50 % of Welsh and 25 % of English topsoils displayed significant κ_{fd} .

The measurement of κ_{fd} exploits the fact that there is a grain size range which behaves effectively as SP particles (high κ) in the low-frequency field, but SSD (low κ) in the presence of the higher frequency. According to the theory of Néel (1949), it is possible to have a κ_{fd} of > 90 % for a particular SD assemblage. However, in practice measurements on a large number of samples from various environments has indicated a general observational limit of ≈ 15 %, although there are a few reports of higher values for the standard decade increase in frequency for some volcanic tuffs, *e.g.*, ≈ 30 % (Worm & Jackson 1999). Note that multidomain (MD) grains display only a very small κ_{fd} (≤ 0.3 %) (Bhathal & Stacey 1969), whilst smaller SP and larger SSD display no κ_{fd} .

That values of κ_{fd} are very rarely seen above 15 %, has led to the development

of several theories which attempt to elucidate this apparent discrepancy. The theories for κ_{fd} fall in to two groups. The first group of theories considers the behaviour of population distributions of non-interacting Néel-type SD particles (Néel 1949) near the SP/SSD boundary in response to different AC field frequencies (*e.g.*, Stephenson 1971; Dabas et al. 1992; Eyre 1997; Worm 1998). These theories state, that as the blocking volume (v_b), *i.e.*, the boundary between SP and SSD grains, is a function of measuring frequency, then there is a range of grains which are blocked to the high frequency but not the lower one. The blocking volume for an independent SD grain in a small field is given by (Néel 1949)

$$v_b = \frac{2kT \ln(t_m/\tau_o)}{\mu_0 M_s H_k} \quad (1)$$

where M_s is the spontaneous magnetisation, T the temperature, k is Boltzmann's constant, μ_0 is the permeability of free space, t_m the measurement time or for κ the reciprocal of twice the measurement frequency, H_k is the (micro-)coercive force associated with a grain, and h is the external field. τ_o is the atomic reorganisation time, which for magnetite is best taken as $\approx 10^{-9}$ s as argued by Worm (1998).

In the most recent paper of this type, Worm (1998) made calculations using log-normal grain distributions and demonstrated that for “realistic” narrow distributions of magnetite, low values of κ_{fd} are expected, *e.g.*, for a lognormal variation of 0.5, the maximum κ_{fd} is 22 % (Worm 1998).

Dearing et al. (1996a) proposed a different type of phenomenologically-based model for κ in SD grains, which estimates a maximum “theoretical” κ_{fd} of 16.3 %. However, as noted by both Eyre (1997) and Worm (1998), this model fails to incorporate the relationship of blocking volume and measuring time, and hence deviates from the well-established SD theory of Néel (1949).

All previous models for κ_{fd} have been for non-interacting systems, whereas it is known that magnetic interactions significantly effect the magnetic properties of assem-

blages of magnetic grains, both experimentally (*e.g.*, Dormann et al. 1999a) and theoretically (*e.g.*, Virdee 1999).

In this paper an analytical model based on the theory of Dormann et al. (1988) for distributions of interacting SP grains, is incorporated into the model of Worm (1998), and the effect of these interactions on κ_{fd} for assemblages of SP/SSD magnetite is presented. This is of great importance, firstly because it is exceptionally difficult to produce non-interacting synthetic SP/SD magnetic samples, making comparison between non-interacting theories and well-characterised synthetic samples futile, and secondly some environmental systems, *e.g.*, soils, usually display magnetic characteristics indicative of magnetic interactions (Maher 1988).

2 THEORY

The total κ for an assemblage of SD grains has a contribution from both the SP grains (κ_{sp}) and the SSD grains (κ_{ssd}). For non-interacting grains κ_{sp} and κ_{ssd} in an AC field, are given by (Néel 1949; Worm 1998)

$$\begin{aligned} \kappa_{sp} &= \frac{M_s \tanh(\mu_0 v M_s h / 3kT)}{h(1 + \omega^2 \tau^2)} \\ &\approx \frac{\mu_0 v M_s^2}{3kT(1 + \omega^2 \tau^2)} \quad \text{for small } h \end{aligned} \quad (2)$$

$$\kappa_{ssd} = \frac{2M_s}{3H_k} \quad (3)$$

where ω is the wave number, and τ is the relaxation time given by (Néel 1949),

$$\tau = \tau_0 \exp(-E_B/kT) \quad (4)$$

where E_B is the energy barrier to be overcome for the magnetic moment of a grain to switch direction. For a non-interacting grain E_B is equal to the anisotropy energy $E_a = \mu_0 M_s v H_k$.

2.1 Static and dynamic interactions

When an external field is applied to an assemblage of grains each particle experiences not only the external field, but also the dipole fields generated by neighbouring particles (Dunlop & West 1969). When calculating the effect of interactions it is necessary to consider the response of both SP and SSD grains to interactions, and the interaction fields they in turn generate.

The dipole field generated from a SSD grain is relatively constant compared to the time it takes for either an SP or SSD grain to rotate in the field. This makes it possible to treat such interactions as static (Spinu & Stancu 1998), and in a first approximation a mean field approach suffices for the dipole field generated by SSD grains (Dunlop & West 1969). This simple approximation is justified, because it is found that the interaction with SSD grains is relatively small compared to that between SP grains (EL-Hilo et al. 1992).

For SP grains the situation is more complicated. The behaviour of magnetic assemblies of SP particles which have a volume distribution, disordered arrangement and easy axes randomly distributed, fall into one of three regimes depending on the inter-particle interaction (Dormann et al. 1999a,b): pure superparamagnetic (non-interacting case as modelled by Worm (1998)), superparamagnetic modified by interactions (weak-interaction regime), and a collective state. The properties of the last state, called the glass collective state (Dormann et al. 1999a), are close to those of spin glasses showing a phase transition. However, this state is presently not fully understood (Dormann et al. 1999a,b), and there is no analytical model for the collective state, there being only models for the non-interacting and weak-interaction regimes.

For the weak-interaction regime, near the blocking volume or temperature where relaxation is important in the system, the statistical interaction field is fluctuating at a high rate. These interactions are qualitatively different from static ones, and they are termed dynamic interactions (Dormann et al. 1988; Spinu & Stancu 1998). Such dynamically interacting systems are not in thermodynamic equilibrium and hence can not be directly

modelled using Boltzmann statistics, however, there are several approaches that have been developed to circumnavigate this problem (Dormann et al. 1988; Mørup & Tronc 1994). In this paper, the model developed by Dormann et al. (1988) (DBF) is incorporated to calculate the effect of interactions on κ_{fd} . This model was chosen in preference to a rival model (Mørup & Tronc 1994; Hansen & Mørup 1998), because of the extensive theoretical arguments and experimental evidence given in detail in Dormann et al. (1999b).

The DBF model estimates the energy interaction potential by averaging over all possible particle arrangements, and it is shown that the effect of dynamic interactions is equivalent to an increase in particle anisotropy for Néel-type SD particles (Dormann et al. 1988). To a first approximation, where only nearest neighbour interactions are considered the energy E_B (equation 4) can be rewritten as (O’Grady et al. 1993)

$$E_B = E_a + E_{int} \quad (5)$$

where $E_{int} = n\mu_0 M_s^2 v_m a_1 L\left(\frac{\mu_0 M_s^2 v_m a_1}{kT}\right)$

where E_a is the anisotropy energy of the non-interacting case, E_{int} is the interaction energy, v_m is the mean volume of the SP particles, where $a_1 = v_m \langle 3 \cos^2 \psi - 1 \rangle / \langle d_{cc}^3 \rangle$, n is the average number of nearest neighbours, ψ and d_{cc} represent the location of a particle which is the first neighbour, $\langle d_{cc} \rangle$ is the mean centre-to-centre inter-particle separation and L is the Langevin function. It is convenient to express d_{cc} in terms of the mean average diameter of the distribution, d_o , to give $d_{cc} = dd_o$, where d is the relative separation distance in terms of d_o .

The DBF model is only applicable to spherical or near-spherical grains. In nature most SD grains have an average aspect ratio of 1:1.5 (Dunlop & Özdemir 1997), *i.e.*, they are only slightly ellipsoid, making the DBF model applicable.

2.2 The influence of dipolar interactions on v_b

In a system where there are both blocked and superparamagnetic particles, the blocked particles create a static interaction field (h_{sd}) and the SP particles a dynamic interaction field. In determining v_b for an interacting system (equation 5), the dynamic interaction field is represented by the term E_{int} , whilst the static interaction field reduces the coercive force by the h_{sd} (Dunlop & West 1969). The equation for v_b in the presence of small external fields is then

$$v_b = \frac{-E_{int} + 2kT \ln(t_m/\tau_o)}{\mu_0 M_s (H_k - h_{sd})} \quad (6)$$

It is readily seen from equation 6 that the effect of dynamic interactions is to reduce v_b , whilst the static interactions increase v_b . Equation 6 converges, and it is possible to determine v_b for an assemblage of SSD and SP grains.

3 NUMERICAL MODELS FOR κ AND κ_{FD}

Real samples have many grains of different sizes, shapes and internal stresses, and hence have a grain volume distribution $N(v)$ and a coercive force distribution $H(H_k)$. The total magnetic moment, m , for such an assemblage of SD grains is given by

$$m = \int \int M_s v N(v) H(H_k) n(v, H_k) dv dH_k \quad (7)$$

κ is found by dividing m by the total volume and the external field. For simplicity in this model it was assumed that the assemblage is initially demagnetised. This assumption is not critical, as it is the dynamic interaction between SP grains which most strongly affects the behaviour of the assemblage.

It is well documented that volume distribution usually take a lognormal form (*e.g.*, Krumbin & Graybill 1965). In this model a lognormal distribution of the form used in similar studies was utilised (Eyre 1997; Worm 1998)

$$N(v) \propto \exp(-\log(v/v_o)^2/2\sigma_l^2) \quad (8)$$

where v_o is the lognormal mean and σ_l is the lognormal variance. In the model κ was calculated as a function of v_o . Therefore, it must be realised that each volume calculated and depicted actually represents only the average volume for a distribution, but this is in accordance with experimental studies.

H_k can be associated to the bulk coercive force H_c , by the relationship $H_k \approx 2.09H_c$ (Stoner & Wohlfarth 1948). In the non-interacting model of Worm (1998), Worm considered an even distribution with $\mu_0H_c = 40 - 60$ mT. For comparison a similar approach is taken here, but instead of assuming an uniform distribution, a Gaussian distribution of the form $H(H_k) \propto \exp(-(H_k - \mu)^2/2\sigma_n^2)$ is used, where μ is the mean and σ_n the variation. The effect of varying μ and σ_n is considered. It was assumed that variations in H_k are due to variations in stress not shape. Initially this assumption may seem inappropriate for environmental systems where small grains usually originate by precipitation and are thought to have low internal stress, however, stress is often important for natural fine particles as they often possess an oxidised surface. Its importance increases with decreasing grain size. This assumption that the variation in coercivity is due to stress is not critical, as it is shown later, that κ_{fd} is more sensitive to variations in grain-size than in coercivity.

The inclusion of these two types of distribution is important, since it allows the direct calculation of κ for an assembly of magnetic grains, rather than considering the relaxation time of individual particles. However, such a statistical approach does not provide a simple analytical solution, but numerical calculations can be performed easily. For a given interaction regime it is first necessary to determine v_b (equation 6) of the system before calculating κ_{fd} , to determine the ratio of blocked and unblocked grains. E_{int} is varied by changing either or both of d or n (equation 5); E_{int} increases with n and decreases with d . The mean static field of the blocked grains was simply determined

by calculating the field associated with the mean SD grain size at the given separation distance d_{cc} . After determining v_b , equation 7 was integrated numerically using the mid-point method, allowing summation over grain volumes $v_o \rightarrow \infty$.

To determine κ_{fd} it is necessary to calculate both a high frequency (κ_{hf}) and low frequency (κ_{lf}). This is achieved by changing the measuring time, t_m , in equation 2. As κ_{fd} is usually measured using a Bartington dual-frequency susceptibility probe, which has a low-frequency of 470 Hz and high-frequency of 4700 Hz, these two frequencies were used in the calculations.

Following previous calculations in the literature, the model is for stoichiometric magnetite at room temperature, for which M_s was taken as $4.8 \times 10^5 \text{ Am}^{-1}$ (Dunlop & Özdemir 1997). The atomic arrangement time, τ_o , is weakly affected by the interaction field (Dormann et al. 1999b), however the effect is relatively small and in the following calculations it is assumed to be constant. The value for ψ in equation 5 is not significant compared to the other variables as $\langle 3 \cos^2 \psi - 1 \rangle$ can only vary between 1 and 2. For simplicity ψ was held constant at 30° .

4 RESULTS

4.1 Effect of interactions on blocking volume

In Figure 1 the effect of interactions on the blocking diameter d_b is depicted for different grain distributions. In Figure 1a the effect of different AC frequencies on d_b is considered, whilst in Figure 1b different coercive forces are shown. It is seen that as both the interaction distance d and the number of nearest neighbours increase n (equation 5), then d_b is reduced depending on lognormal variance (σ_l) and d_o , to give a minimum value for d_b , e.g., for κ_{lf} ; for $\sigma_l = 0.2$, the minimum d_b is in the vicinity of $d_o = 0.012 \mu\text{m}$ for $\mu_o H_c = 40 \text{ mT}$, $n = 2$ and $d = 2$, and for $\sigma_l = 0.8$, $\mu_o H_c = 60 \text{ mT}$, $n = 1$, $d = 5$ the minimum is at $d_o = 0.0018 \mu\text{m}$. The position of the minimum decreases with increasing σ_l . The reduction of d_b indicates that the effect of the dynamic interactions is greater than that of static interactions (equation 6). For interaction parameters which give inter-

action energies greater than those shown in Figure 1, *i.e.*, $d \approx 2$ and $n \approx 2$ for $\sigma_l = 0.2$, the solution did not converge to give d_b for *all* values of d_o . It is suggested that this is the initial existence of the collective state, which is supported by experimental evidence, *e.g.*, Dormann et al. (1999a) found that for maghemite particles the boundary between the interactive and collective states was between $d \approx 1.44$ and 1.55. These values are slightly smaller than those suggested by the model for the glass collective state, *i.e.*, $d \approx 2$, however this may be due to differences in the grain distribution, mineralogy and temperature, and simplifications in the model.

4.2 Calculation of κ

Initially the results for the calculation of κ_{lf} for different grain sizes with different interaction parameters were considered (Figures 2). The coercivity distribution was kept constant with a mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT. κ_{hf} could just as easily be depicted, but it is standard practice to consider κ_{lf} . It can be seen that for increasing interactions, the effect is to reduce the intensity of κ_{lf} , for example, for $\sigma_l = 0.5$, the peak κ_{lf} is reduced from ≈ 16 for the non-interactive state to ≈ 11 for $n = 2$ and $d = 2.5$ (Figure 2b). The position of the peak κ_{lf} decreases with increasing interaction, *e.g.*, for $\sigma_l = 0.2$ the peak is shifted from $d_o = 0.0118 \mu\text{m}$ for the non-interacting case to $d_o = 0.0098 \mu\text{m}$ for $n = 2$, $d = 2$. The value at $d_o = 0.0118 \mu\text{m}$ decreases from 25 to 12.5, *i.e.*, 50 % (Figure 2a).

4.3 Calculation of κ_{fd}

The effect of weak interactions on κ_{fd} is shown in Figure 3. The κ_{fd} curves for the non-interacting cases are identical to those of Worm (1998). Interactions reduce the intensity of the peak values of κ_{fd} , and shift them to lower values of d_o changing the shape of the κ_{fd} versus d_o curves. For example, for $\sigma_l = 0.2$ the peak value is reduced from $\kappa_{fd} \approx 38$ % for the non-interacting case to 29 % for $n = 2$ and $d = 2$, with a shift in peak position from $d_o = 0.0154 \mu\text{m}$ to $0.0114 \mu\text{m}$. For $d_o = 0.0154 \mu\text{m}$, κ_{fd} is reduced by

38 %. An interaction regime with $n = 2$ and $d = 2.5$, reduces κ_{fd} at $d_o = 0.0078 \mu\text{m}$ from the non-interacting regime by 27 % for $\sigma_l = 0.5$, and for $\sigma_l = 0.8$ with interaction parameters $n = 2$ and $d = 4$ the peak κ_{fd} is reduced by 50 %.

The effect of interactions is to decrease d_b , therefore there are a range of small grains which display κ_{fd} only in the interacting regime, *i.e.*, the effect is to increase κ_{fd} for small d_o . This is demonstrated by considering the change in κ_{fd} , *i.e.*, $(\kappa_{fd}(\text{inter}) - \kappa_{fd}(\text{non-inter}))$ versus d_o (Figure 4), where it is seen that for most d_o the effect of interactions is to reduce κ_{fd} , however κ_{fd} for smaller d_o is seen to increase with increasing interactions. For larger values of d_o , interactions can also give rise to a very small increase in κ_{fd} , although this is not readily seen in Figure 4.

If the coercive force distribution is changed (Figure 5), then for the non-interacting regime, the position of the peak κ_{fd} is seen to increase with decreasing mean coercive force ($\mu_o H_c$). The position of the peak κ_{fd} is less affected in the interacting regime by changes in mean $\mu_o H_c$, therefore the distance between the non-interacting and interacting κ_{fd} peak increases with decreasing mean $\mu_o H_c$. Differences in σ_n were found to be less significant than changes in mean $\mu_o H_c$.

5 DISCUSSION

In general the effect of interactions is to reduce both κ and κ_{fd} , but for certain values of d_o , κ_{fd} can also increase slightly. The increase in κ_{fd} for certain d_o is due to the reduction in v_b caused by interactions (Figure 4) which change the range of grain sizes which display significant κ_{fd} . Thus, not only do interactions reduce κ and generally κ_{fd} , they also change the size range which display significant κ_{fd} . The reduction in peak κ_{fd} is relatively small, but for non-peak values of d_o , κ_{fd} can be reduced by over 50 % (Figure 3). Hence, the addition of grain interactions to the model of Worm (1998), further explains why κ_{fd} values for natural samples are very rarely seen above ≈ 15 %, because the effect of interactions is seen to reduce maximum $\kappa_{fd} > 20$ % in the non-interacting model of Worm (1998) to values < 20 % (Figure 3). Unfortunately the system is highly

non-unique so it is not possible to evaluate grain distributions and interaction energies from values of κ_{fd} and κ_{lf} alone. However, knowledge of how interactions effect κ_{fd} contributes to a better understanding of a sample.

In fact, there is a general difficulty in identifying and quantifying interactions in natural magnetic systems, because unless both the grain distribution and dominating anisotropy is accurately known then there is no definitive test for identifying SD grain interactions (Dunlop & Özdemir 1997). At present, measurement of the Wohlfarth ratio (R) (Wohlfarth 1958) is the most common technique for identifying levels of magnetic interactions (*e.g.*, Maher 1988; Worm & Jackson 1999). For non-interacting, uniaxial SD grains $R = 0.5$, and the effect of interactions is to reduce R . However, there are problems with this simplified approach as the presence of MD grains also reduces R , and recent calculations found that $R > 0.5$ for non-interacting grains with cubic anisotropy (García-Otero et al. 2000), making the interpretation of R for natural systems rather ambiguous.

The effect of strong interactions which produce a collective glass state on κ_{fd} are unknown, and cannot be modelled analytically at present as there is no formulation for modelling SD grain assemblages of this type (Dormann et al. 1999b). Hence one can only speculate the behaviour of such clusters and the effect on κ and κ_{fd} . The glass collective state displays many of the characteristics of spin glasses, which are characterised by “frozen” long range order and a slowing down of relaxation time. Whether the glass collective state displays long range order is debatable (Hansen & Mørup 1998; Dormann et al. 1999b), however, recent experimental evidence suggests that the relaxation time increases (Dormann et al. 1999a). It is therefore speculated, that the effect of a collective state would be to further decrease both κ and κ_{fd} , as the superparamagnetic character of the grains would be removed and they would display a more SSD-like behaviour. understanding of a sample.

Worm (1998) stated correctly that bi-modal distributions would also significantly reduce κ_{fd} . This effect would be even more enhanced in interacting regimes, because

κ for the small SP grains is more strongly effected than larger SSD or MD grains. It should be noted that there are several other effects not considered in the model which are expected to reduce κ_{fd} , e.g., mixed mineral assemblages.

6 CONCLUSIONS

Both κ and κ_{fd} have been calculated from first principles, for weakly interacting assemblages of SD magnetite grains. Weak-interactions between SP grains effectively increase the anisotropy, which reduces v_b . The decrease in v_b is dependent on the grain-size distribution and the measuring frequency. The result is that interactions decrease the peak values of κ_{fd} , and can reduce κ_{fd} by over 50 % for certain grain distributions. However, the reduction of v_b as a result of interactions can also increase κ_{fd} for certain grain distributions (Figure 4). The effect of interactions is seen to reduce maximum $\kappa_{fd} > 20$ % in the non-interacting model of Worm (1998) to values < 20 %, whereas experimental κ_{fd} values are very rarely seen above ≈ 15 % (Dearing et al. 1996a).

The effect of interactions further supports the arguments given by both Eyre (1997) and Worm (1998), that there is no “theoretical” limit of 16.3 % for κ_{fd} as given in the model of Dearing et al. (1996a). Instead, the fact that values for κ_{fd} are rarely seen above ≈ 15 %, is because real assemblages of magnetic grains can be both magnetically interacting and have wide grain-size distributions.

ACKNOWLEDGMENTS

Financial support from the European Union funded European Network for Mineral Magnetic Studies of Environmental Problems (MAG-NET) (contract number ERBFM-RXCT98-0247) is gratefully acknowledged.

REFERENCES

- Bhathal, R. S. & Stacey, F. D., 1969, Frequency independence of low-field susceptibility of rocks, *J. Geophys. Res.*, **74**(2025-7).

- Dabas, M., Jolivet, A., & Tabbagh, A., 1992, Magnetic-susceptibility and viscosity of soils in a weak time-varying field, *Geophys. J. Int.*, **108**, 101–109.
- Dearing, J. A., Dann, R. J. L., Hay, K., Lees, J. A., Loveland, P. J., & O'Grady, K., 1996, Frequency-dependent susceptibility measurements of environmental materials, *Geophys. J. Inter.*, **124**, 228–240.
- Dearing, J. A., Hay, K. L., Baban, S. M. J., Huddleston, A. S., Wellington, E. M. H., & Loveland, P. J., 1996, Magnetic susceptibility of soil: an evaluation of conflicting theories using a national data set, *Geophys. J. Inter.*, **127**, 728–734.
- Dormann, J. L., Bessais, L., & Fiorani, D., 1988, A dynamic study of small interacting particles - superparamagnetic model and spin-glass laws, *J. Phys. C-Solid State Physics*, **21**, 2015–2034.
- Dormann, J. L., Fiorani, D., Cherkaoui, R., Tronc, E., Lucari, F., Dorazio, F., Spinu, L., Nogues, M., Kachkachi, H., & Jolivet, J. P., 1999, From pure superparamagnetism to glass collective state in γ -Fe₂O₃ nanoparticle assemblies, *J. Magn. Magn. Mater.*, **203**, 23–27.
- Dormann, J. L., Fiorani, D., & Tronc, E., 1999, On the models for interparticle interactions in nanoparticle assemblies: comparison with experimental results, *J. Magn. Magn. Mater.*, **202**, 251–267.
- Dunlop, D. J. & Özdemir, O., 1997, *Rock Magnetism: Fundamentals and Frontiers*, Cambridge University Press.
- Dunlop, D. J. & West, G. F., 1969, An experimental evaluation of single domain theories, *Rev. Geophys.*, **7**, 709–757.
- EL-Hilo, M., O'Grady, K., & Chantrell, R. W., 1992, The ordering temperature in fine particle systems, *J. Magn. Magn. Mater.*, **117**, 21–28.
- Eyre, J. K., 1997, Frequency dependence of magnetic susceptibility for populations of single domain grains, *Geophys. J. Int.*, **129**, 209–211.
- García-Otero, J., Porto, M., & Rivas, J., 2000, Henkel plots of single-domain ferromagnetic particles, *J. Appl. Phys.*, **87**(10), 7376–7381.
- Hanesch, M. & Petersen, N., 1999, Magnetic properties of a recent parabrown-earth from Southern Germany, *Earth Planet. Sci. Lett.*, **169**(85-97).
- Hansen, H. F. & Mørup, S., 1998, Models for the dynamics of interacting magnetic nanoparticles, *J. Magn. Magn. Mater.*, **262-274**, 184.
- Krumbin, W. C. & Graybill, F. A., 1965, *An introduction to statistical models in geology*, McGraw-Hill, New York.
- Maher, B. A., 1988, Magnetic properties of some synthetic sub-micron magnetites, *Geophys. J.*, **94**, 83–96.
- Mørup, S. & Tronc, E., 1994, Superparamagnetic relaxation of weakly interacting particles,

Phys. Rev. Lett., **72**, 3278–3281.

Néel, L., 1949, Théorie du traînage magnétique des ferromagnétique en grains fins avec applications aux terres cuites, *Ann. Géophys.*, **5**, 99–136.

O’Grady, K., EL-Hilo, M., & Chantrell, R. W., 1993, The characterisation of interaction effects in fine particle systems, *IEEE Trans. Mag.*, **29**(6), 2608–2613.

Spinu, L. & Stancu, A., 1998, Modelling magnetic relaxation phenomena in fine particles systems with a Preisach-Néel model, *J. Magn. Magn. Mater.*, **189**, 106–114.

Stephenson, A., 1971, Single domain grain distributions: I. a method for the determination of single domain grain distribution, *Phys. Earth Planet. Inter.*, **4**, 353–360.

Stoner, E. C. & Wohlfarth, E. P., 1948, A mechanism of magnetic hysteresis in heterogeneous alloys, *Phil. Trans. R. Soc. Lond.*, **240**, 599–642.

Virdee, D., 1999, *The influence of magnetostatic interactions on the magnetic properties of magnetite*, Ph.D. thesis, Univeristy of Edinburgh.

Wohlfarth, E. P., 1958, Relations between different modes of acquisition of the remanent magnetization of ferromagnetic particles, *J. Appl. Phys.*, **29**, 595–596.

Worm, H. U., 1998, On the superparamagnetic-stable single domain transition for magnetite, and frequency dependence of susceptibility, *Geophys. J. Inter.*, **133**, 201–206.

Worm, H. U. & Jackson, M., 1999, The superparamagnetism of Yucca Mountain tuff, *J. Geophys. Res.*, **104**, 25415–25425.

Figure 1. d_b as a function of d_o , *i.e.*, the mean diameter for the grain distribution, for a) two different measuring frequencies with $\sigma_l = 0.2$ and $\mu_0 H_c = 40$ mT, and b) two different coercive force values with $\sigma_l = 0.8$ and a measurement frequency of 4700 Hz.

Figure 2. κ_{lf} versus d_o for a) $\sigma_l = 0.2$ and b) $\sigma_l = 0.5$ with three different interaction parameters, and coercive force distribution between 40-60 mT, mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT. The wide coercive force distribution is similar to that used in Worm (1998).

Figure 3. κ_{fd} versus d_o for σ_l equal to a) 0.2, b) 0.5 and c) 0.8 with three different interaction parameters, and coercive force distribution between 40-60 mT, mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT.

Figure 4. Change in κ_{fd} , *i.e.*, $(\kappa_{fd}(\text{inter}) - \kappa_{fd}(\text{non-inter}))$, versus d_o for $\sigma_l = 0.5$ with three different interaction parameters, and coercive force distribution between 40-60 mT, mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT.

Figure 5. κ_{fd} versus d_o for $\sigma_l = 0.5$ for two different mean coercive forces ($\mu_0 H_c = 30$ mT and 50 mT) for a non-interacting and an interacting regime. Both coercive force distributions were integrated over a range of 20 mT with the mean at the mid-point, and $\sigma_n = 45$ mT.









