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Effect of grain interactions on the frequency dependence of magnetic susceptibility

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SUMMARY

Environmental systems often contain superparamagnetic (SP) grains that cause a frequency dependence 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 per cent for certain grain distributions. This helps to explain why κ_{fd} values for natural samples are very rarely seen above $\approx$15 per cent, as the effect of interactions is seen to reduce maximum κ_{fd} $>$20 per cent in non-interacting models to values $<$20 per cent 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 interacting SP grains is not well understood, and no analytical formulation has yet been made.

Key words: environmental magnetism, magnetic interactions, magnetic susceptibility, SP–SSD transition.

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 dependence 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 low and high frequencies 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; for example, Dearing *et al.* (1996b) found that approximately 50 per cent of Welsh and 25 per cent of English topsoils displayed significant κ_{fd}.

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

That values of κ_{fd} are very rarely seen above 15 per cent has led to the development of several theories that 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), that is, the boundary between SP and SSD grains, is a function of measuring frequency, then there is a range of grains that 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

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

(Néel 1949), where M_s is the spontaneous magnetization, T is the temperature, k is Boltzmann's constant, μ_0 is the permeability of free space, t_m is the measurement time or for κ the reciprocal of twice the measurement frequency and H_k is the (micro)coercive force associated with a grain. τ_o is the atomic reorganization 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 lognormal grain distributions and demonstrated that for 'realistic' narrow distributions of magnetite, low values of κ_{fd} are expected; for example, for a lognormal variation of 0.5, the maximum κ_{fd} is 22 per cent (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 per cent. However, as noted by both Eyre (1997) and Worm (1998), this model fails to incorporate the relationship between 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 assemblages 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, first because it is exceptionally difficult to produce non-interacting synthetic SP/SD magnetic samples, making comparison between non-interacting theories and well-characterized synthetic samples futile, and second because 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, h, are given by (Néel 1949; Worm 1998)

$$\kappa_{sp} = \frac{M_s \tanh(\mu_0 v M_s h/3kT)}{h(1+\omega^2\tau^2)}$$

$$\approx \frac{\mu_0 v M_s^2}{3kT(1+\omega^2\tau^2)} \qquad \text{for small } h\,, \quad (2)$$

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

where ω is the wavenumber 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 an SSD grain is relatively constant compared to the time it takes for either an SP or an 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 that have a volume distribution, disordered arrangement and easy axes randomly distributed fall into one of three regimes depending on the interparticle 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 models only 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 fluctuates 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 cannot 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 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 (eq. 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-neighbour grains, ψ and d_{cc} represent the location of the particle's first neighbour, $\langle d_{cc}\rangle$ is the mean centre-to-centre interparticle 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, giving $d_{cc} = dd_o$, 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 ellipsoidal, making the DBF model applicable.

2.2 The influence of dipolar interactions on v_b

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

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

It is readily seen from eq. (6) that the effect of dynamic interactions is to reduce v_b, whilst the static interactions increase v_b. Eq. (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

$$m = \int\int M_s(v) H(H_k) n(v, H_k) dv dH_k . \tag{7}$$

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 demagnetized. This assumption is not critical as it is the dynamic interaction between SP grains that most strongly affects the behaviour of the assemblage.

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

$$N(v) \propto \exp[-\log(v/v_o)^2/2\sigma_l^2] , \tag{8}$$

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 realized 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 is related to the bulk coercive force H_c by the relationship $H_k \approx 2.09 H_c$ (Stoner & Wohlfarth 1948). In the non-interacting model of Worm (1998), Worm considered an even distribution with $\mu_0 H_c = 40-60$ mT. For comparison a similar approach is taken here, but instead of assuming a 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 oxidized 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 to variations 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 (eq. 6) of the system before calculating κ_{fd} in order to determine the ratio of blocked to unblocked grains. E_{int} is varied by changing either d or n or both (eq. 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, eq. (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 (κ_{hf}) and a low-frequency κ (κ_{lf}). This is achieved by changing the measuring time, t_m, in eq. (2). As κ_{fd} is usually measured using a Bartington dual-frequency susceptibility probe, which has a low frequency of 470 Hz and a 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 4.8×10^5 A m^{-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 of ψ in eq. (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 Fig. 1 the effect of interactions on the blocking diameter, d_b, is depicted for different grain distributions. In Fig. 1(a) the effect of different AC frequencies on d_b is considered, whilst in Fig. 1(b) different coercive forces are shown. It is seen that as both the interaction distance d and the number of nearest neighbours n increase (eq. 5), d_b is reduced depending on the lognormal variance (σ_l) and d_o, to give a minimum value for d_b; for example, for κ_{lf} for $\sigma_l = 0.2$, the minimum d_b is in the vicinity of $d_o = 0.012$ µm for $\mu_o H_c = 40$ mT, $n = 2$ and $d = 2$, and for $\sigma_l = 0.8$, $\mu_o H_c = 60$ mT, $n = 1$, $d = 5$ the minimum is at $d_o = 0.0018$ µm. 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 the static interactions (eq. 6). For interaction parameters that give interaction energies greater than those shown in Fig. 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*

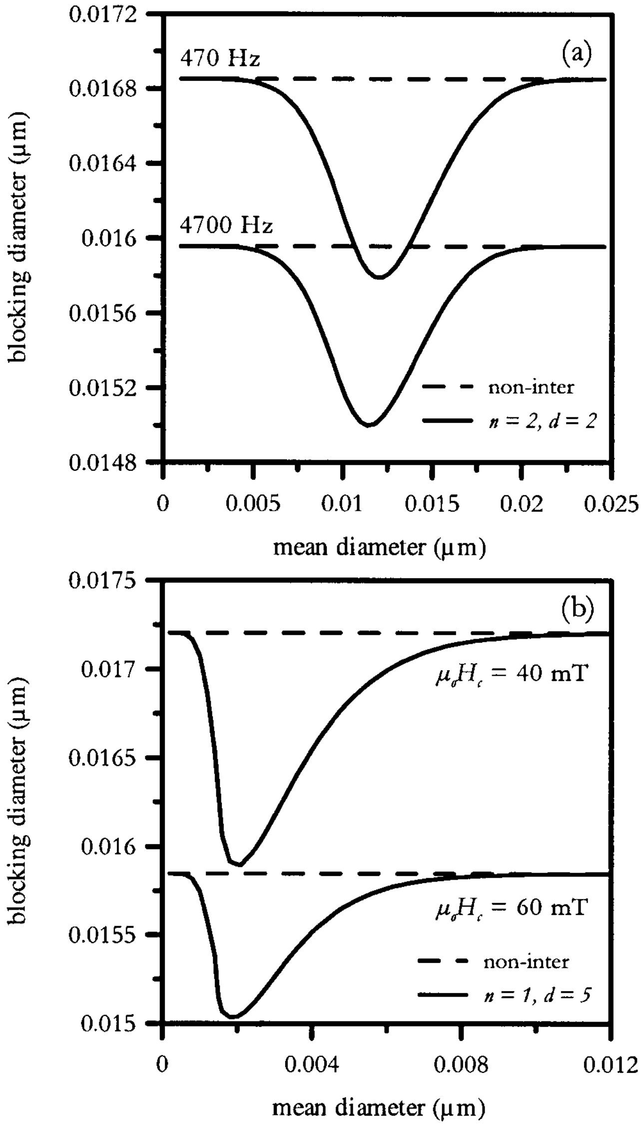

Figure 1. d_b as a function of d_o, i.e. the mean diameter of the grain distribution, for (a) two different measuring frequencies with $\sigma_l = 0.2$ and $\mu_0 H_c = 40$ mT and (b) two different coercive force values with $\sigma_l = 0.8$ and a measurement frequency of 4700 Hz.

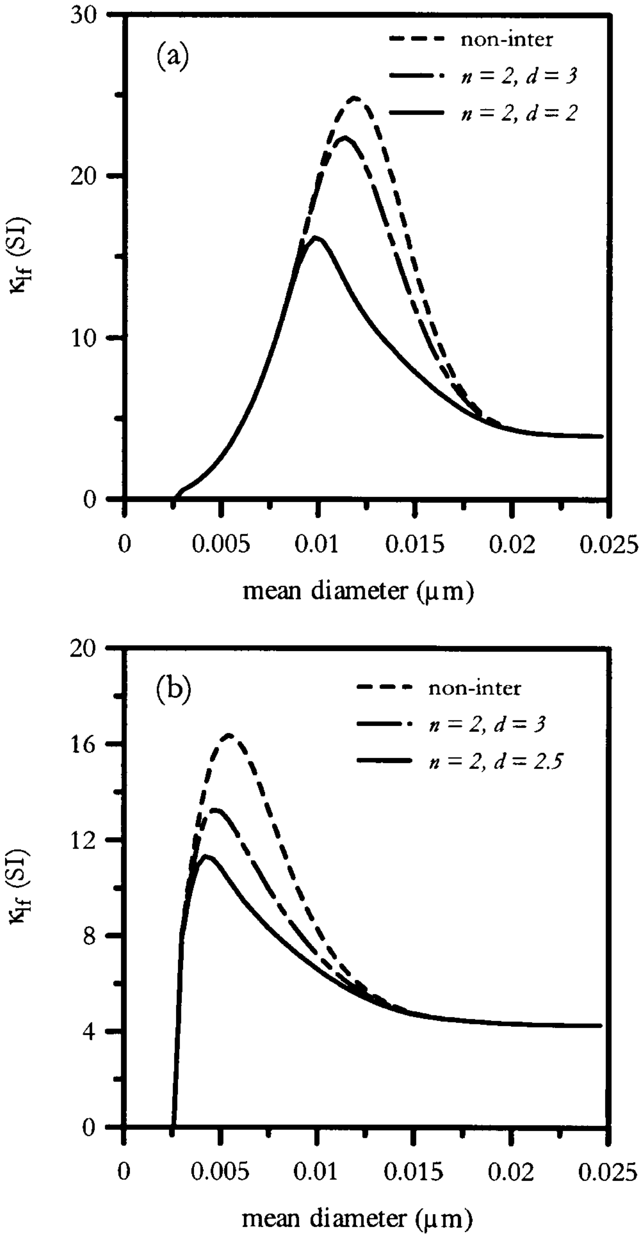

Figure 2. κ_{lf} versus d_o for (a) $\sigma_l = 0.2$ and (b) $\sigma_l = 0.5$ with three different interaction parameters and coercive force distribution between 40 and 60 mT, mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT. The wide coercive force distribution is similar to that used in (Worm 1998).

values of d_o. It is suggested that this is the initial existence of the collective state, which is supported by experimental evidence; for example, (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 (Fig. 2). The coercivity distribution was kept constant with a mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT. κ_{hf} can be depicted just as easily, 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 $d = 2.5$ (Fig. 2b). The position of the peak κ_{lf} decreases with increasing interaction; for example, for $\sigma_l = 0.2$ the peak is shifted from $d_o = 0.0118$ μm for the non-interacting case to $d_o = 0.0098$ μm for $n = 2$, $d = 2$. The value at $d_o = 0.0118$ μm decreases from 25 to 12.5, i.e. by 50 per cent (Fig. 2a).

4.3 Calculation of κ_{fd}

The effect of weak interactions on κ_{fd} is shown in Fig. 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

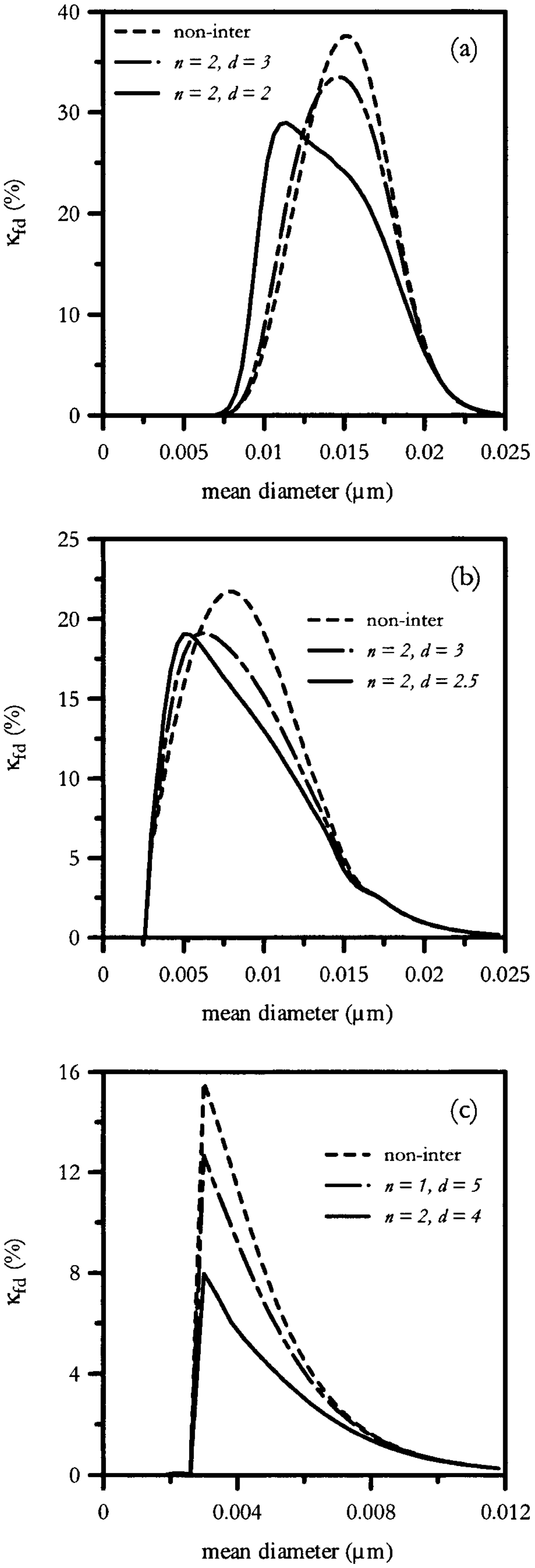

Figure 3. κ_{fd} versus d_o for σ_l equal to (a) 0.2, (b) 0.5 and (c) 0.8 with three different interaction parameters and coercive force distribution between 40 and 60 mT, mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT.

shape of the κ_{fd} versus d_o curves. For example, for $\sigma_l = 0.2$ the peak value is reduced from $\kappa_{fd} \approx 38$ per cent for the non-interacting case to 29 per cent for $n = 2$ and $d = 2$, with a shift in peak position from $d_o = 0.0154$ μm to 0.0114 μm. For $d_o = 0.0154$ μm, κ_{fd} is reduced by 38 per cent. An interaction regime with $n = 2$ and $d = 2.5$ reduces κ_{fd} at $d_o = 0.0078$ μm from the non-interacting regime by 27 per cent 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 per cent.

The effect of interactions is to decrease d_b, therefore there is a range of small grains that display κ_{fd} only in the interacting regime, that is, the effect is to increase κ_{fd} for small d_o. This is demonstrated by considering the change in κ_{fd}, i.e. [κ_{fd} (inter) − κ_{fd} (non-inter)] versus d_o (Fig. 4). It is seen in Fig. 4 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 Fig. 4.

If the coercive force distribution is changed (Fig. 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 (Fig. 4) that change the range of grain sizes that display significant κ_{fd}. Thus, not only do interactions reduce κ and generally κ_{fd}, but they also change the size range that

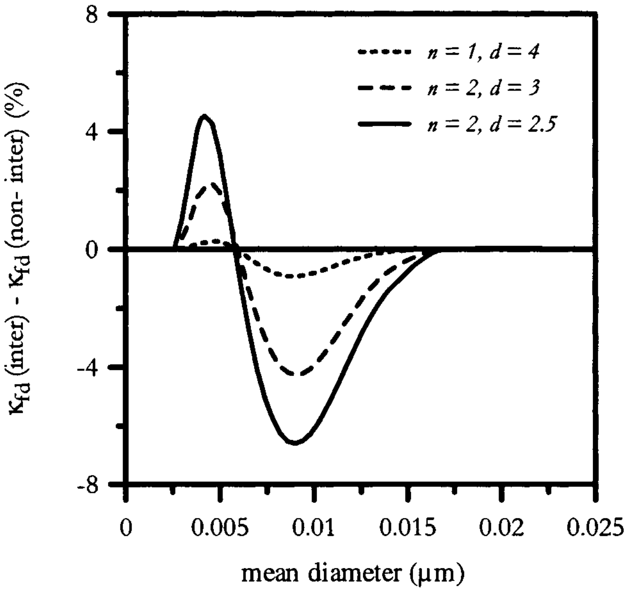

Figure 4. Change in κ_{fd}, i.e. [κ_{fd} (inter) − κ_{fd} (non-inter)], versus d_o for $\sigma_l = 0.5$ with three different interaction parameters and coercive force distribution between 40 and 60 mT, mean $\mu_0 H_c = 50$ mT and $\sigma_n = 45$ mT.

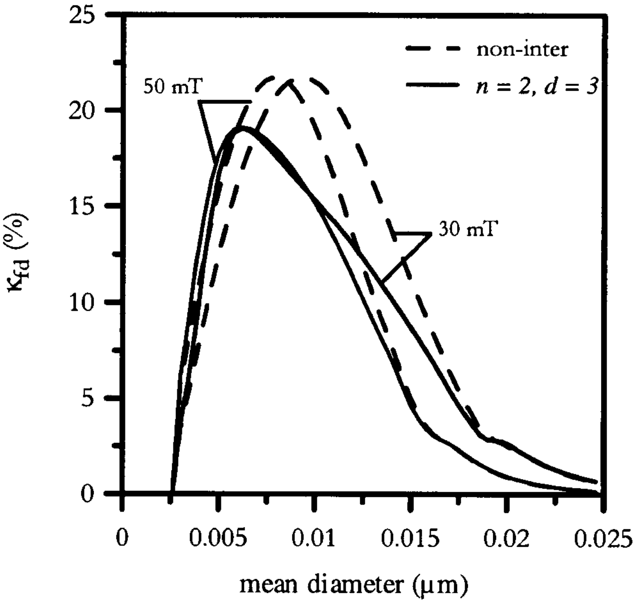

Figure 5. κ_{fd} versus d_o for $\sigma_l=0.5$ for two different mean coercive forces ($\mu_0 H_c=30$ and 50 mT) for a non-interacting and an interacting regime. Both coercive force distributions were integrated over a range of 20 mT with the mean at the mid-point and $\sigma_n=45$ mT.

displays 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 per cent (Fig. 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 $\approx$15 per cent, because the effect of interactions is seen to reduce maximum κ_{fd}>20 per cent in the non-interacting model of Worm (1998) to values 20 per cent (Fig. 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 affect 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 the dominating anisotropy are accurately known, 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 that 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 on 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 characterized 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 decrease further both κ and κ_{fd} because the SP character of the grains would be removed and they would display a more SSD-like behaviour.

Worm (1998) stated correctly that bimodal 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 affected than for larger SSD or MD grains. It should be noted that there are several other effects not considered in the model that 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 per cent for certain grain distributions. However, the reduction of v_b as a result of interactions can also increase κ_{fd} for certain grain distributions (Fig. 4). The effect of interactions is seen to reduce maximum $\kappa_{fd}>20$ per cent in the non-interacting model of Worm (1998) to values <20 per cent; experimental κ_{fd} values are very rarely seen above $\approx$15 per cent (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 per cent for κ_{fd} as given in the model of Dearing *et al.* (1996a). Instead, the fact that values of κ_{fd} are rarely found above $\approx$15 per cent arises because real assemblages of magnetic grains can both be magnetically interacting and have wide grain-size distributions.

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