Journal of Magnetism and Magnetic Materials 86 (1990) 197–206
North-Holland

A THEORETICAL STUDY OF INTERACTION EFFECTS ON THE REMANENCE CURVES OF PARTICULATE DISPERSIONS

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Received 14 October 1989

The remanence curves of strongly interacting fine-particle systems are investigated theoretically. It is shown that the Henkel plot of the dc demagnetisation remanence vs. the isothermal remanence is a useful representation of interactions. The form of the plot is found to be a reflection of the magnetic and physical microstructure of the material, which is consistent with experimental data. The relationship between the Henkel plot and the noise of a particulate recording medium, another property dependent on the microstructure, is also considered. The Interaction Field Factor (IFF), a single parameter characterising the non-linearity of the Henkel plot, is also investigated. The results are consistent with a previous experimental study. Finally, the effect of interactions on the Switching Field Distribution are investigated.

1. Introduction

There is presently a need in particulate magnetic recording media for an increased understanding of the magnetisation processes that take place and of the effect of interparticle interactions. It is the purpose of this paper to examine theoretically the effects of interactions on the principal remanence curves and to relate those to the magnetic and physical microstructure.

The two remanence curves we will consider are those arising from the attainment of isothermal remanent magnetisation $I_r(H)$ and dc demagnetisation $I_d(H)$, which are representative of non-equilibrium magnetic states at constant field but in different magnetic configurations. Wohlfarth [1] derived the remanence relation

$$I_d(H) = 1 - 2I_r(H) \tag{1}$$

between the two curves for a non-interacting assembly of uniaxial single-domain particles. This relationship is also valid [2] for multi-domain ferromagnets if the walls interact with the same density and distribution of pinning sites on both the initial and demagnetising branches of the magnetisation curve. It was first noted by Henkel [3] that the experimental variation of $I_d(H)$ with $I_r(H)$ gave characteristic plots which show both positive and negative curvature. The non-interacting case corresponds to a linear plot with a gradient of -2. It is reasonable to expect that many-body effects will exist in all magnetic systems, and these seem a reasonable explanation for the non-linear "Henkel plots". Gaunt et al. [2] have produced Henkel plots for PtCo which corre-

[1] Paper in preparation at the time of Professor Wohlfarth's death.

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sponds to the non-interacting case. They attribute agreement with eq. (1) to domain walls interacting with a similar density and distribution of pinning sites during initial and reverse magnetisation. Measurement on MnAlC, however, gave a non-linear relation in a "positive" sense with the curve concave downwards. The explanation given in ref. [2] assumes that at the maximum forward field, the walls became pinned at localised higher density pinning sites. Generally, the pinning sites activated during magnetisation are inactive during demagnetisation.

Pinkerton [4] and Pinkerton and Van Wingerden [5] have also studied Henkel plots in melt-spun NdFeB systems. These show generally a "positive" deviation from linearity which is ascribed to the magnetising effect of interactions between neighbouring grains. In this work a weakly interacting system with a small grain size was produced by a very rapid quench. This gives a closely linear Henkel plot. Spratt et al. [6] have also produced Henkel plots for powders of recording media particles. These show negative curvature which is believed to result from closed-loop configurations at the microscopic level (we will call these negative interactions).

The difference between the curves I_r(H) and I_d(H) has also been examined experimentally by Corradi and Wohlfarth [7]. Here, the interaction field factor (IFF) was used to characterise the type of interactions present. This allows a similar insight into the nature of interactions that Henkel plots provide although, since in this case the interactions are characterised by a single parameter, the IFF gives rather less detailed information.

In this paper we describe the results of theoretical studies by using our previously developed Monte Carlo model [8]. The Henkel plots are studied in relation to microscopic configurations which, in the case of recording media, allows a direct correlation with the medium noise. The major aim is to use the model calculations to give a reasonable basis for the interpretation of experimental Henkel plots. We also consider the relation between the coercivity and interaction field factor investigated by Corradi and Wohlfarth [7]. The model is also used to study the effects of interactions on the switching field distribution of recording media, which also compares favourably with experimental data.

2. The model

The model used in this study is that developed by Lyberatos et al. [8] to look at the anhysteretic properties of interacting magnetic tape particles. Here it is modified to allow the calculation of static remanence curves. The cell under consideration consists of 1000 particles on both a cubic and tetragonal lattice. Each spherical particle is assigned a volume according to a Gaussian distribution which essentially determines its switching field, and is then randomly assigned to a lattice point.

The initial state when calculating $I_r(H)$ is produced either by randomly assigning the moment direction, or by ac erasure, so that the resulting reduced remanent magnetisation is zero. The initial state for the calculation of $I_d(H)$ is saturation remanence. Then, in both cases a positive or negative dc field is applied, which increases in magnitude until saturation is reached for $I_r(H)$ and negative saturation for $I_d(H)$. Each time the dc field is applied we evaluate the probability p_i that the ith particle will reverse the direction of its magnetic moment. A random number x is generated such that $0 < x < 1$, and if $p_i > x$ then we allow the particle to reverse the direction of its magnetic moment. The reversal probability for the ith particle is given by

$$p_i = 1 - \exp(t/\tau_i), \tag{2}$$

where t is the dc field application time, and τ_i is the relaxation time given by the Néel Arrhenius law,

$$\tau_i^{-1} = f_0 \exp\left[-\frac{VI_sH_k}{2kT}(1 - H_l/H_k)^2\right]. \tag{3}$$

Here f_0 is the precession frequency taken to be 10^9 s^{-1} and $H_k = 2K/I_{sb}$ where I_{sb} is the saturation magnetisation of the particles, T is the temperature and H_l is the component of the local field along the particle easy axis. The local field comprises the dc field and the interaction fields arising from all other particles in the assembly. A cell consists of a $10 \times 10 \times 10$ array of cobalt particles with median diameter $D_m = 0.02$ μm and standard deviation $\sigma = 0.15$. The easy anisotropy axes of the particles are directed along the z-axis and each particle centre is separated by nD_{max} in the z-direction where n (> 1) is the tetragonality factor. $D_{max} = D_m + 3\sigma$ is the largest particle diameter. The columns of particles are separated by mD_{max} (where $m > 1$) in the x- and y-directions. Variation of the parameters n and m allowed the production of lattices with distinct microstructures which forms a central part of the present study. In order to produce ac demagnetised states the system was subjected to a square wave field with a decrement of ΔH_{ac}/cycle. ΔH_{ac} was reduced in magnitude as in ref. [8] until a fully demagnetised state was acheived. The demagnetised state acheived in this manner is highly correlated as shown by previous noise studies [9].

The model assumes that the magnetic easy axes of the particles are aligned and hence there are no reversible magnetisation changes. Consequently, the dc demagnetisation curve is identical to the hysteresis loop in the second and third quadrants, and the coercivity (H_c) and remanence coercivity (H_r) are identical. Hence, we are easily able to calculate the variation of coercivity with packing density for each lattice type. We have also calculated the effective switching field distribution of the systems by differentiation of the remanence curves, which gives further information regarding effects of interactions on the behaviour of recording media.

3. Comparison of remanence curves

We have studied the behaviour of interacting fine particle systems with the specific aim of understanding the macroscopic magnetic behaviour in relation to the magnetic and physical microstructure. This was achieved by varying the lattice type and the initial magnetic configuration of the systems studied. The form of the remanence curves was found to depend strongly on the lattice type and (in the case of the IRM) the initial magnetic state. Fig. 1 shows typical data for the case in which the IRM curve was started from an ac erased initial configuration. Fig. 1 also serves to define the remanence coercivity H_r, at which I_d vanishes, and in addition an alternative estimate (H_r'), calculated using the expression $I_r(H_r') = 0.5 I_r(\infty)$, which essentially arises from eq. (1). The difference between these values (the Interaction Field Factor, IFF) has been used by Corradi and Wohlfarth [7] as a measure of many-body effects. Consideration of their experimental data in the light of the present calculations will be given later.

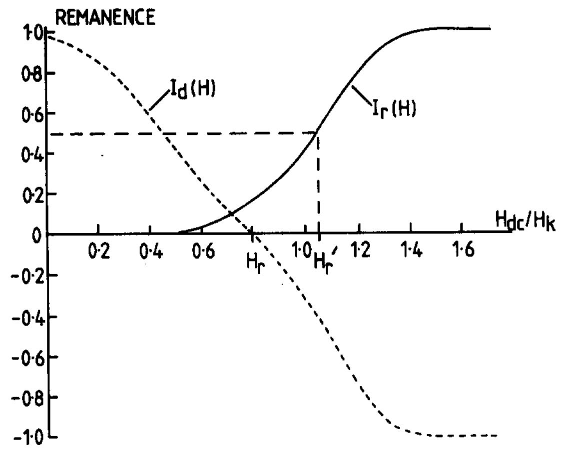


Fig. 1. IRM curve (broken line) and dc demagnetisation curve (solid line) calculated for a tetragonal lattice with $n = 3$. Definitions of the remanence coercivity (H_r) and an alternative estimate (H_r') are given.

The Henkel plot potentially reveals much more detail about the microscopic magnetic behaviour than the single IFF parameter. Fig. 2 shows Henkel plots calculated from the data of fig. 1. It can be seen that the curves have a characteristic (concave upwards) form with a deviation from the linear behaviour expected for a non-interacting system. This form of plot is characteristic of systems which are easier to demagnetise than to magnetise. The experimental data of Spratt et al. [6] on a compacted powder of CrO_2 particles are of this form. In contrast, the measurements of Pinkerton [4] on NdFe have a deviation in a "positive" sense. The difference probably originates in the different microstructures of the materials. The microstructure might be expected to have a significant effect on the interactions, perhaps the simplest example being closed chain configurations which, because of flux closure considerations, should intuitively have a demagnetising effect. In general, all microstructures exhibiting flux closure should have a similar effect. On the contrary, in systems containing for example isolated chains of particles this might be expected to have the effect of enhancing the magnetisation. This observation points to the importance of the texture of the material in determining the form of the Henkel plot. In this context the texture of the system is characteristic of the predominant local order, which in general will be dependent upon the particle size and the preparation condition, including the degree of dispersion and the magnitude and direction of any orienting field. Here we investigate these effects via the relation of the Henkel plot to the lattice geometry.

Calculated results for a microstructure with a net magnetising effect are given in fig. 3. This is essentially a 1-D lattice consisting of a series of chains oriented in the field direction. The IRM curve is calculated by starting from a demagnetised state obtained by orienting the magnetic moments at random subject to the constraint of zero magnetisation. At low packing densities, the chains are well separated and therefore the dipolar interactions between the chains are relatively weak. In this case the interparticle interactions might be expected to aid the magnetisation process, which is certainly reflected in the form of the Henkel plot of fig. 3. Again the deviation from linearity increases with increasing packing density.

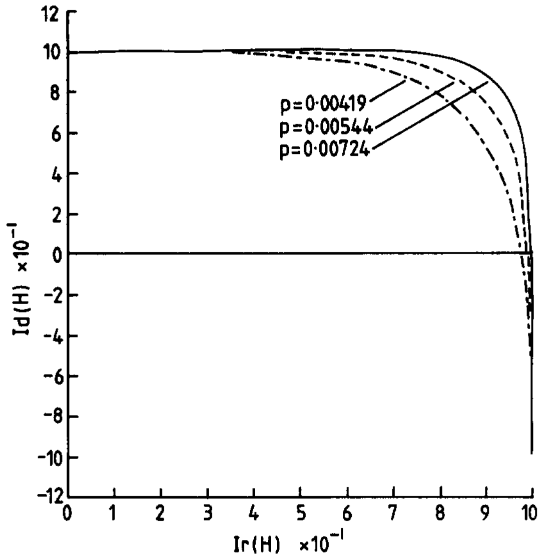


Fig. 3. Henkel plots for different packing fractions for a 1-D lattice.

The IRM curves were found to depend very strongly on the initial configuration of the system. This is very pronounced for a cubic lattice, as shown in fig. 4a. The starting configurations for these calculations were obtained in two distinct ways. The first was to assign positive and negative moments at random (within the constraint of achieving a demagnetised state). The second was obtained by subjecting the system to an ac erasure process which we have described previously [9]. In a strongly interacting system ac erasure allows the build up of long-ranged magnetic order as long as the process is carried out quasi-statically, so that equilibrium can be achieved. In the case of a cubic lattice this has a dramatic effect on the Henkel plot, completely changing the character of the deviation from linearity as can be seen in fig. 4b. The ac erased configuration contains correlated regions of closed-loop structures which are difficult to magnetise and which are responsible for

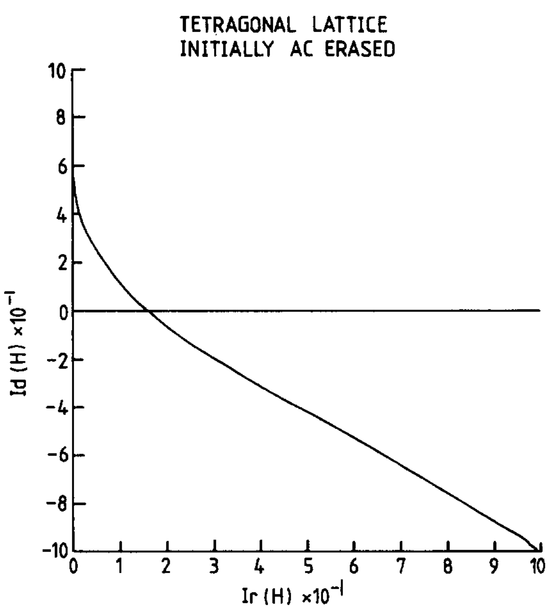


Fig. 2. Henkel plot of $I_d(H)$ vs. $I_r(H)$ for a tetragonal lattice calculated from the data of fig. 1.

the dramatic change in the Henkel plot. Interestingly the 1-D lattice gives close-to-linear Henkel plots when ac erased, as shown in fig. 4c. This is a result of columnar switching. The ac erased configuration gives rise to highly correlated states within a chain. Because of the relatively narrow intrinsic switching field distribution, whole columns tend to switch during the magnetisation process. This is similar to the case during the demagnetisation process where all the chains are fully magnetised in the same direction. Thus, in a sense the system consists of weakly interacting columns of particles. There is still, however, a slight deviation from linearity in the positive sense. This indicates that the positive intrachain interactions are more effective in overcoming the switching fields of the magnetically harder particles than are the negative interchain interactions which might be expected to have a demagnetising effect. The example of a 1-D lattice serves to illustrate

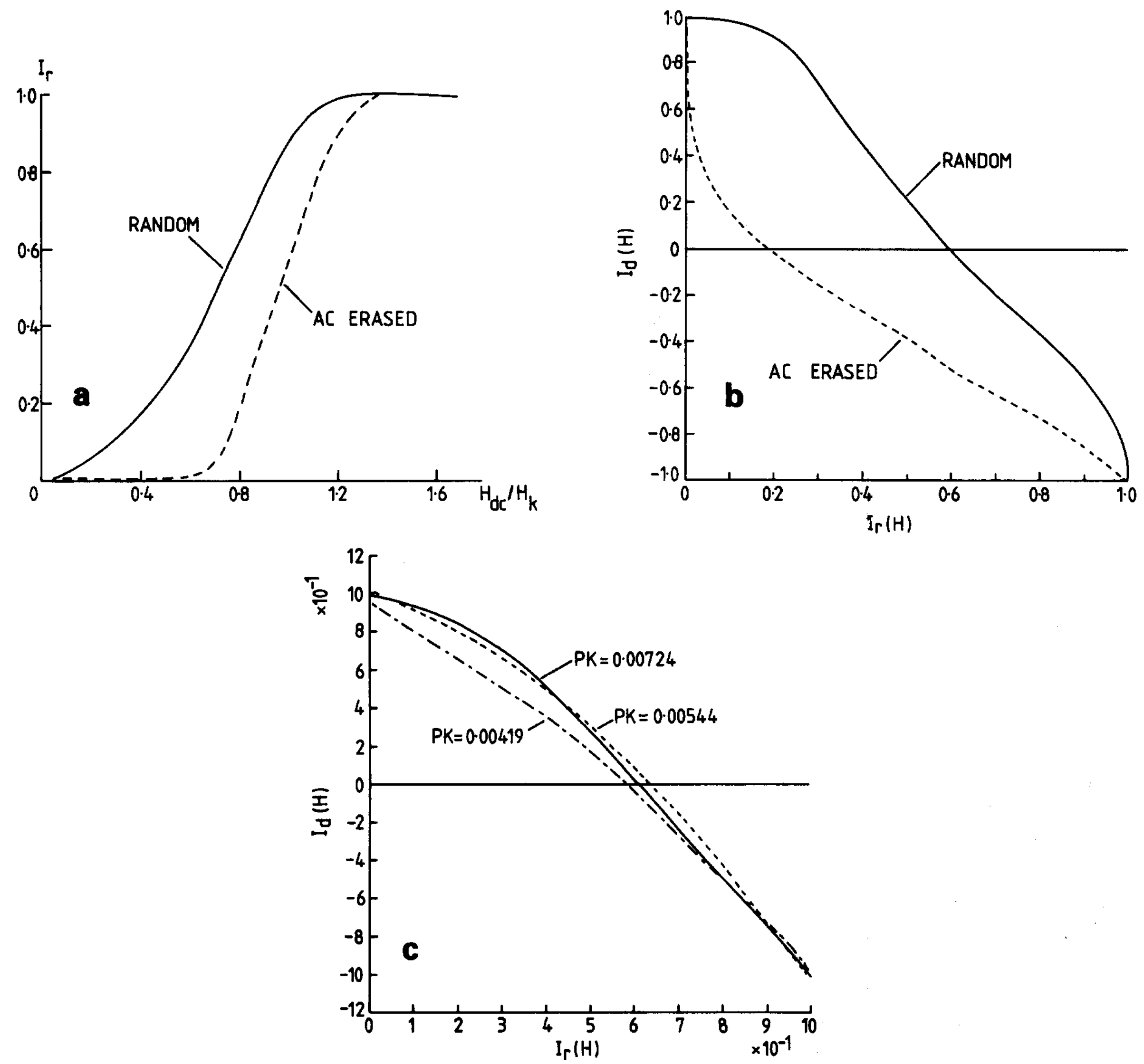


Fig. 4. (a) IRM curves for the cubic lattice (packing fraction = 0.182) starting from random and ac erased initial configurations; (b) corresponding Henkel plots for the IRM curves of fig. 4a; (c) Henkel plot for the 1-D lattice.

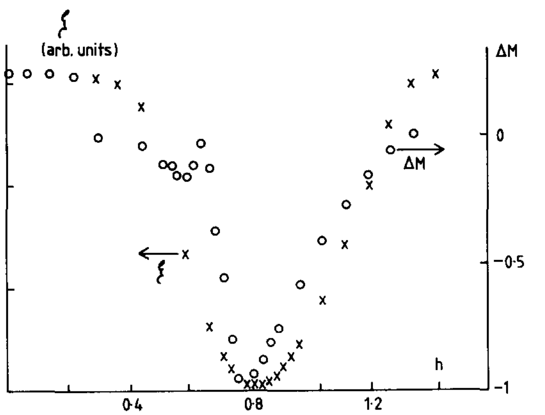


Fig. 5. ΔM (broken curve) and the long-wavelength noise power (solid curve) as a function of field during the demagnetisation process.

that the Henkel plot is dependent on interactions, but in a way which is strongly affected by the type of microstructure of the medium.

The relationship with the magnetic microstructure has been further investigated by calculating the variation during the demagnetisation process of a spin–spin correlation function

$$\xi = \langle \mu_i \cdot \mu_j \rangle, \tag{4}$$

where μ_i is the magnetic moment of particle i and the brackets represent an ensemble average. In order to effect a comparison with the macroscopic behaviour, we have characterised the non-linearity of the Henkel plot by using the parameter

$$\Delta M = I_d - (1 - 2I_r).$$

Fig. 5 shows the variation of ΔM (calculated using the IRM curve for an initially ac erased system) and ξ with field. ΔM has a maximum, indicating that the maximum deviation from linearity occurs at the remanence coercivity. The variation of ξ is very similar to that of ΔM. The value at H_r is close to zero. From the definition of ξ this corresponds to a highly ordered structure having an "antiferromagnet-like" order. Thus, the maximum in ΔM corresponds to a highly correlated microstructure, as seems reasonable. The correlation function defined by eq. (4) is used here because, for a recording medium, it is directly related to the noise produced as a result of the discrete amount of flux produced by each particle at the recording head. Hence, the calculations given here are in agreement with the observation that the noise of a particulate recording medium goes through a minimum during the dc demagnetisation process. The relation demonstrated by fig. 5 between the variation of ξ and ΔM with field represents an interesting connection between the microscopic properties and bulk magnetic behaviour of the medium, the essential link being the microscopic magnetic correlations. This is currently the subject of further theoretical and experimental investigations.

4. The interaction field factor (IFF)

The IFF is defined in ref. [7] as $(H_r' - H_r)/H_c$, expressed as a percentage. Here, however we use $H_r' - H_r$ which is a parameter dependent only on interactions. It is essentially a measure of the non-linearity of the Henkel plot and was found [7] to increase with packing density for powders of Fe_2O_3 particles. Our calculations show the variation of IFF with packing densities to depend upon the type of lattice and the initial magnetic configuration. Fig. 6 shows the the data for a 1-D lattice which has a negative IFF which increases in mag-

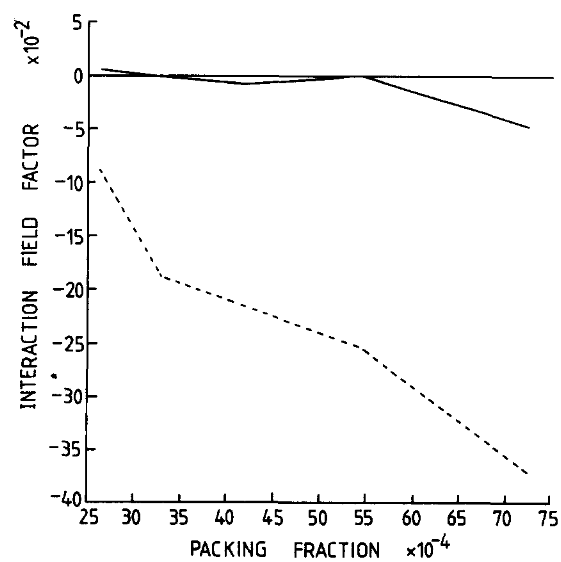


Fig. 6. IFF vs. packing fraction for a 1-D lattice with IRM curves calculated for different starting configurations. Solid line – ac erased; dotted line – randomly orientated moments.

nitude with the packing density, in agreement with the experimental data of ref. [7]. The strong dependence on the starting configuration arises from the sensitivity of I_r to the initial magnetic microstructure, as noted previously. Similar data for a cubic lattice where the dependence on the initial configuration is more pronounced are shown in fig. 7, with a sign change between the random and ac erased IRM state. In the random initial state the interactions tend to have a magnetising effect, whereas the more correlated state produced by ac demagnetisation actually hinders the magnetisation process.

An additional important parameter, strongly dependent on particle interactions, is the coercivity H_c, which is known [7,10,11] to depend strongly upon the packing density. In ref. [7] the coercivity was shown to be correlated with the IFF. Here, we have investigated this relation for all lattice types, using the ac erased IRM curves to calculate the IFF. Our data are not entirely conclusive. There is a definite increase of H_c with IFF for the cubic lattice as shown in fig. 8. This is contrary to the experimental data of ref. [7] since for the cubic lattice the interactions have a magnetising rather than a demagnetising effect. No significant trend can be detected for the tetragonal lattice, although the data do indicate a broad maximum. If this is

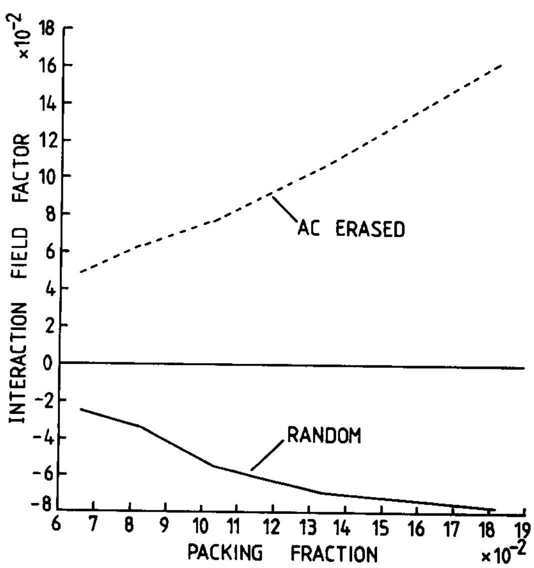


Fig. 7. IFF as a function of packing fraction for a cubic lattice.

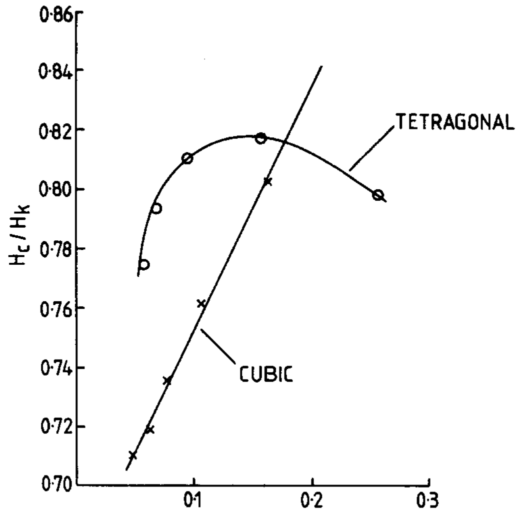


Fig. 8. Coercivity as a function of packing fraction for both the cubic (×) and tetragonal (○) lattices.

the case the sense of the interaction effects is dependent on the proximity of the particles rather than being solely dependent on the physical microstructure. In our model this effect could arise because of the discretisation of the interaction energy used in the calculation. Further work is necessary in order to clarify this.

We have also observed a small decrease of the IFF with increasing temperature, which is consistent with the recently observed increase in the anhysteretic susceptibility with temperature [12]. This was interpreted in terms of a decrease in interaction strength as characterised by $H_r' - H_r$. The present calculations confirm that a significant temperature variation of $H_r' - H_r$ does indeed occur because of a related decrease in interaction effects.

5. Interactions and the switching field distribution

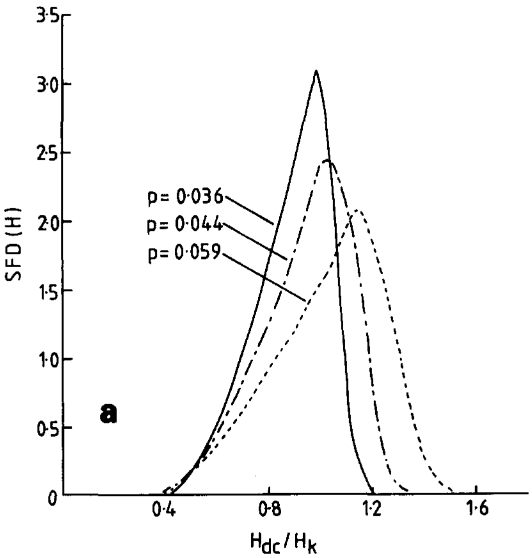
From the point of view of the characterisation of recording media, the switching field distribution (SFD) is of major importance. SFD(H) is a measure of the number of particles reversing their moments in an applied field H. With this definition it is intuitively clear that the SFD is given by the differential of the remanence curve. In principle, the forward and reverse remanence curves should give the same SFD for a non-interacting system, although the work presented here shows that, since the remanence curves differ significantly due to interactions, this will not be the case in practise. This is in agreement with previous experimental data [13].

In this case we concentrate on the use of the SFD determined from the forward remanence curve in order to investigate interaction effects. In general, the effect of interactions is to considerably broaden the SFD which seems reasonable since the dispersion of interaction fields must add considerably to the intrinsic energy barrier distribution. This observation is common to all lattice types studied, with the broadening dependent on the interaction strength.

Typically, the SFD's obtained were non-symmetric with a tail at the small field side. This reflects the asymmetry in the intrinsic SFD, which in our model system results from the volume distribution of the particles. The dependence of the remanence curve on the initial configuration gives rise to a similar dependence for the SFD. This is illustrated in fig. 9 which gives the SFD for a tetragonal lattice with various packing fractions for an initially random configuration and for an ac erased system. The ac erased case (fig. 9a) illustrates the very significant broadening, associated with the effects of interactions, which increases with increasing packing density.

Fig. 9b shows the result of the initially ramdom case in which a significant shoulder appears at the low-field side of the distribution. This results from the random magnetic microstructure which produces many particle pairs oriented in high-energy configurations, which because of its inherent instability is a configuration that is very easily magnetised. Thus, the IRM is initially enhanced, resulting in the observed increase of the SFD in the low-field region. This effect disappears at higher fields where the initial magnetic configuration decreases in importance. In this region the SFD's calculated from both initial states are essentially identical.

Our data for the cubic lattice are summarised in fig. 10 which gives the variation with packing density of the parameter ΔH characterising the width of the SFD at half height. This can be seen to be strongly dependent on the packing fraction for both cases. This is consistent with the fact that the magnitude of the interaction fields scale with this factor. The observed increase in ΔH with packing fraction is also in agreement with the



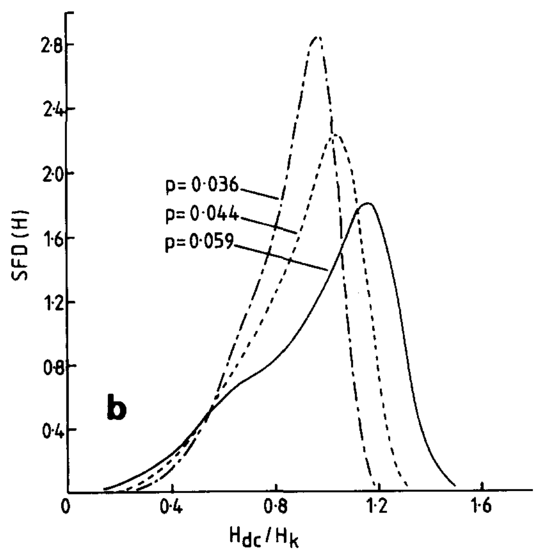


Fig. 9. Switching field distribution calculated for the following initial configuration: (a) tetragonal lattice, initially ac erased, (b) tetragonal lattice, randomly oriented moments.

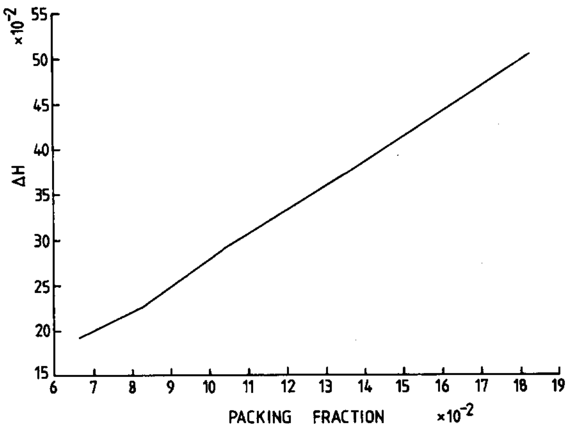


Fig. 10. ΔH, representing the width of the switching field distribution, as a function of the packing fraction.

experimental data of Corradi and Wohlfarth [7] on compacted powders. In this work the width of the SFD was characterised by the coercivity factor $CF = (H_r - H_c)100/H_c$ which was also found to increase with increasing packing density.

6. Conclusion

We have studied the effects of dipolar interactions on the remanence curves of a fine-particle system and on various derived parameters. The remanence curves are extremely sensitive to interactions. Non-linearities in the plot of I_d vs. I_r can be ascribed directly to the interaction effects. The data show a marked dependence of the Henkel plot on the lattice type. Thus, the different types of experimental curves in, for example, NdFe and CrO_2 powders probably reflects the different microstructure of the systems. We have also found that the IRM curve is very strongly dependent on the magnetic state.

Clearly, remanence curves and, in particular, the Henkel plot are a useful link between the magnetic and physical microstructure of the systems. As such, they represent a potentially useful technique for the investigation of interaction effects in strongly interacting particulate media. In addition, they can also act as a useful characterisation of recording media.

The dependence of the IRM on the starting state is of interest. The differences between the random and ac erased states are very pronounced, especially in the case of the 1-D lattice. Here, the Henkel plot became very closely linear after ac erasure. The reason for this is the formation of highly correlated chains of particles. Because of the narrow spread of particle sizes these tend to switch collectively; hence, the system behaves effectively as a set of weakly coupled particles which have a closely linear Henkel plot. In this respect the plot must be interpreted with care when analysing data with regard to interactions. In practice, this extreme case is unlikely to occur since, for example, an increased size distribution or imperfections in the chain structure will both reduce the collective behaviour. However, care should be taken in studying systems in which extreme collective behaviour is possible.

In practice, it is difficult to obtain a completely random initial state even by rapid cooling. Some correlations are inevitable, although these might be expected to differ greatly from those obtained by ac demagnetisation. Perhaps a more controlled experiment is to compare the IRM curves for an ac erased system with those of a system demagnetised firstly by applying a saturating field and then by applying a negative field equal to the remanence coercivity. The complication here is that the larger particles will be switched in the opposite direction to the smaller ones. Initial studies, however, show that there is a marked difference between the data which can be ascribed to differences in the initial state. Further work is presently in progress in this area. Previous work [14] has shown that there exists a correlation between the SFD and the magnitude of the time dependence coefficient $S = dM/d \ln t$ which characterises the time decay of the magnetisation. In general $S = S(H, T)$ and has a similar form to the SFD. Our studies of remenance curves would imply that the SFD is different when measured during the magnetisation and demagnetisation processes. As a consequence, it is reasonable to assume that there should be an equivalent difference between values of S measured during the magnetisation and demagnetisation processes. In fact this has been observed experimentally by

Flanders and Sharrock [15], and by Uren et al. [16] who also demonstrated the relation to the SFD measured under the appropriate conditions. This is an interesting approach which would provide useful study of interaction effects. Further theoretical work in this area is presently underway.

Acknowledgement

This work was supported by a grant from the Science and Engineering Research Council.

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