Computational model of the magnetic and transport properties of interacting fine particles

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(Received 7 June 2001; revised manuscript received 26 November 2001; published 29 April 2002)

A computational model is applied to the study of the hysteresis properties of a system of interacting single domain particles. The model is based on Monte Carlo techniques and takes into account both magnetostatic and exchange interactions. The results presented concentrate on a detailed study of the behavior of Co particles, with the interaction strength varied by variations in the packing density. It is found that the magnetic properties are strongly dependent on the parameter $\beta = KV/kT$, with K the anisotropy constant and V the mean particle volume. For small β—i.e., close to superparamagnetic systems—the microstructure is dominated by a tendency to flux closure. However, the interactions lead to an increase in the local energy barriers, resulting in an increase in H_c with packing density ϵ. For large β the anisotropy and magnetostatic interaction fields become comparable and the competition leads to a decrease in the coercivity H_c with ϵ. For intermediate values of β a maximum in the variation of H_c with ϵ is predicted. The irreversible susceptibility is shown to have a complex dependence on interactions, especially in small fields where frustration effects arising from the competition between exchange and magnetostatic interactions are apparent. Exchange and magnetostatic interactions give rise to local magnetic order which is strongly dependent on the relative strength of the exchange interactions. The magnetic order has a strong bearing on the magnetic properties. A link is also made to the transport properties of the system, which are dependent on a spin-spin correlation function. It is shown that exchange interactions give rise to a significant deviation from the quadratic dependence of the giant magnetoresistance on M^2.

DOI: 10.1103/PhysRevB.65.174417 PACS number(s): 75.40.Mg, 75.70.Pa, 75.60.Ej, 75.50.Tt

I. INTRODUCTION

The magnetic properties of nanoscale magnetic particles are unique in that, in this size range, energy considerations restrict the magnetic state to a single domain in each particle. The magnetization process must then involve rotation of the magnetic moment, a process which in energy terms is costly relative to domain wall motion, resulting in high coercivities. This phenomenon was first investigated theoretically by Stoner and Wohlfarth (SW).[1] The SW model provides the essential formalism for the understanding of the origin of hysteresis and for the effects of, for example, the orientational texture of the system requested by the degree of orientation of the easy axes. However, it is a static, athermal theory, the idealizations of which provide a severe limitation. In reality magnetization reversal takes place by thermal activation over finite energy barriers, which leads to a temperature and time dependence of magnetic behavior.

This effect was investigated by Néel[2] who characterized the reversal probability in terms of a relaxation time given by the Arrhenius-Néel law

$$\tau = f_0 \exp(-\Delta E/kT), \tag{1}$$

where ΔE is the energy barrier to rotation. The frequency factor f_0 is normally taken to be $\sim 10^9\ \mathrm{s}^{-1}$. By setting τ equal to the characteristic measurement time t_m, Bean and Livingston[3] derived a criterion separating thermally stable (TS) from thermal equilibrium or superparamagnetic (SPM) behavior as

$$\Delta E_c = kT \ln(t_m f_0). \tag{2}$$

Taking $t_m = 100$ s gives the usual criterion for SPM behavior as $\Delta E_c = 25kT$.

The combination of the SW and Néel models provides a strong framework for the interpretation of the time and temperature dependence of the magnetic properties in terms of a gradual evolution from TS to SPM behavior governed by the Bean-Livingston criterion. This gives rise to a semianalytical formalism in terms of the energy barrier distribution, which provides an important framework for the understanding and interpretation of experimental data. However, a major contribution to the energy barriers—namely, the effect of interparticle interactions—cannot easily be implicitly included, except as a mean-field approximation. Interparticle interactions have an important, occasionally dominant, effect on the magnetic properties of a fine-particle system. By the nature of the systems a magnetostatic (dipolar) interaction between particles is inevitable. Because this is a long-range interaction, its effects cannot be neglected except at the lowest densities. In addition, there exists the possibility of exchange interactions between the particles. Dormann *et al.*[4] have reviewed the effects of interparticle magnetostatic interactions on the general magnetic properties, but most especially the low-field susceptibility. The results are interpreted in terms of an analytical theory which predicts an increase in the local energy barrier due to interaction effects.[5] However, there is considerable evidence that interactions can lead to collective magnetic behavior, with consequences which cannot be ex-

plained in terms of simple increase of the energy barrier. For example, Morup[6] finds evidence for the existence of a ''superferromagnetic'' state arising from magnetostatic interactions. The ordered state depends on the physical microstructure of the system, which introduces the necessity to distinguish between two material types. The first of these is a system prepared by the solidification of a colloidal dispersion, either by freezing or polymerization. Here the local order has a form which arises from magnetostatic interactions and varies from flux closure structures for systems solidified in zero field[7] to large-scale chain formation in field-solidified samples.[8] Tronc *et al.*[9,10] have shown experimentally, for a system of frozen ferrotypes, that the existence of the collective state depends on the number of nearest neighbors and, hence, on the preparation conditions. The second type of system is a granular magnetic solid, a heterogeneous alloy which can be prepared for example by sputtering[11] and which consists of isolated grains of the magnetic phase dispersed in a nonmagnetic background. Such systems exhibit interesting magnetic and transport properties including a giant magnetoresistance (GMR).[12,13] Recent computational studies[14] have concluded that the randomness in the physical microstructure of a heterogeneous alloy results in a transition to a state with short-ranged order, whose effect on the energy barrier distribution is quite complex.

Magnetostatic intergranular interactions can be expected to be of importance in granular magnetic solids (GMS's). In the case of systems with grains dispersed within a metallic background there also exists the possibility of exchange coupling. Because of the oscillatory nature of the exchange coupling, this is a complex problem. It is known that the nature of the coupling in multilayer systems can change as the thickness of the interlayer is increased. In contrast, the coupling between spherical grains has not been extensively studied. However, it is known from experiment that the coupling tends to be ferromagnetic in nature. In two-dimensional (2D) sputtered films this is demonstrated from magnetic measurements and simulations.[15,16] In GMS's the recent work of Franco *et al.*[17] has demonstrated the presence of ferromagnetic exchange interactions and highlighted the importance of the interplay between magnetostatic interactions and exchange. The existence of a stripe domain structure in granular alloys with a perpendicular anisotropy[18] also indicates the existence of a ferromagnetic intergranular exchange coupling. Consequently, we have developed a model taking into account the intergranular exchange as a ferromagnetic term.

In this paper we present a study of the magnetic and transport properties of a GMS with a random microstructure. This problem has recently been studied by Kechrakos and Trohidou[19] using a Monte Carlo (MC) model. These authors demonstrate that the magnetostatic interactions can introduce hysteresis effects. In this paper we study the relationship between the hysteresis and induced magnetic correlations. This provides a link to the magnetotransport properties, which are themselves sensitive to local correlations of the magnetization. We also study the effects of exchange coupling between grains, which is a feature of a GMS with a conducting nonmagnetic background. We study the effects of interactions on the energy-barrier dispersions via the switching field distribution (SFD). The competition between the flux closure structures favored by the magnetostatic interactions and the ''pseudodomains'' favored by the exchange coupling leads to frustration effects which are evident in the SFD.

II. DESCRIPTION OF THE MODEL

The model used takes into account the behavior of both the thermally stable and superparamagnetic particles and is described in Ref. 20. Interactions between the particles are included via the calculation of the local field. Importantly, these interactions are responsible for coupling the SPM and thermally stable fractions. The model consists of a cubic cell of interacting single-domain particles. The microstructure is created by randomly positioning the particles within the cell, rejecting any assignments leading to particle overlap. The particle sizes and the anisotropy fields are generated according to a log-normal distribution function. The anisotropy easy axes have a random three-dimensional distribution to make the system comparable to granular systems.

The magnetic field that acts on each particle is the vector sum of the applied field $\vec{H}_{\rm app}$ and the interaction field $\vec{H}_{\rm int}$, the latter having two components $\vec{H}_{\rm dip}$ and $\vec{H}_{\rm ex}$, the dipolar field and the exchange coupling field produced by the neighboring particles. The interaction field produced by the jth particle on the ith particle is

$$\vec{H}_{ij}=\frac{3(\vec{\mu}_j\cdot\vec{r}_{ij})}{r_{ij}^5}-\frac{\vec{\mu}_j}{r_{ij}^3}+C_{ij}H_K\vec{\mu}_j, \tag{3}$$

where C_{ij} represents the exchange coupling between the grains. Since the exchange coupling is not exactly known, C_{ij} is an empirical parameter. In order to model the short-ranged nature of the exchange coupling we introduce a cutoff distance $r_{\rm exch}$ such that for $d>r_{\rm exch}$, $C_{ij}=0$; otherwise, $C_{ij}=C^*$, where C^* is an empirical exchange field scaled with respect to H_k. We note that the model used here has randomly positioned grains, and as a result, even at low densities there exists the possibility of statistical clustering, which gives rise to some degree of exchange coupling. In all the computations given here, the cutoff radius was taken as 10% of the median grain diameter.

The total local field acting on an individual particle is

$$\vec{H}_i=\sum_{i\neq j}\vec{H}_{ij}+\vec{H}_{\rm app}, \tag{4}$$

where the summation is carried out over all particles within a range $r<r_{\rm max}$. The contribution from particles outside this range is calculated using a mean-field approximation.

For every thermally stable particle [i.e., with $KV>\ln(tf_0)$], the equilibrium position of the moment in the local field is calculated using the Stoner-Wohlfarth[1] model. However, if the SW model gives two equilibrium positions, the moment is allowed to jump between the two positions with a probability that depends on the temperature T, volume V, and anisotropy constant K of the particle. The probability is calculated using the Arrhenius-Néel law,[2] in which the en-

ergy barrier is calculated using a numerical approximation due to Pfeiffer.[21] If the transition is allowed, the moment is assigned to either minimum with a probability

$$p = e^{-E_i}/(e^{-E_1} + e^{-E_2}), \tag{5}$$

with $i = 1, 2$ labeling the minima. Physically, this process allows thermal equilibrium to be established.

This process applies when there is a well-defined two-state system, i.e., when the energy barrier is large enough that the moment is constrained to lie close to one or other of the energy minima. For smaller energy barriers ($KV < 3kT$) we use a standard Metropolis[22] algorithm: the angles of the magnetic moment, θ and ϕ, are modified randomly and the difference ΔE between the new energy state and the previous one is calculated and the moment is allowed to remain in this new position with the probability $p = \min(1, e^{-\Delta E/k_B T})$.

It should also be noted that standard MC moves are applied to the thermally stable particles (after determining the relevant minimum) in order to model the thermal equilibrium distribution about the energy minimum.

It is the intention of this paper to consider in detail the bulk magnetic and transport properties of a model granular solid. The essential link between the phenomena is a magnetic correlation length which is shown to be central to both the magnetic and transport properties. It has been shown[11,23] that the phenomena of GMR is related to the correlation $\langle \vec{\mu}_1 \cdot \vec{\mu}_2 \rangle$ between the magnetic moments of the grains. The phenomenon results from the spin-dependent scattering probability of electrons passing from a nonmagnetic to a ferromagnetic region (interface scattering) and also within the ferromagnetic regions. The model considers that this probability exponentially decreases with the distance covered by the electrons, $p = \exp(-r/r_0)$, where r_0, the *spin diffusion length*, is the distance after which the probability of the electron maintaining its initial polarization decreases by a factor of $1/e$. Considering this expression for probability, the correlation function related to the GMR becomes

$$\langle \vec{\mu}_1 \cdot \vec{\mu}_2 \rangle = \frac{\sum_{i=1,n} \sum_{j=1,n_i} \vec{\mu}_i \cdot \vec{\mu}_{n_i} e^{-r/r_0}}{\sum_{i=1,n} \sum_{j=1,n_i} \mu_i \cdot \mu_{n_i}}, \tag{6}$$

where n is the total number of particles in the system and n_i is the number of neighbors considered for the ith particle.

The paper presents a series of calculations of the bulk magnetic properties, with the aim of investigating the effects of both dipolar and exchange interactions.

It is inferred from these data that correlated areas of magnetization develop, especially under the influence of exchange interactions. Transport properties are described in Sec. III D using the simple model outlined above. It is shown that the deviation from the parabolic dependence of GMR on M is predominantly due to interparticle exchange coupling. This leads to local ferromagnetically ordered areas larger than the spin diffusion length.

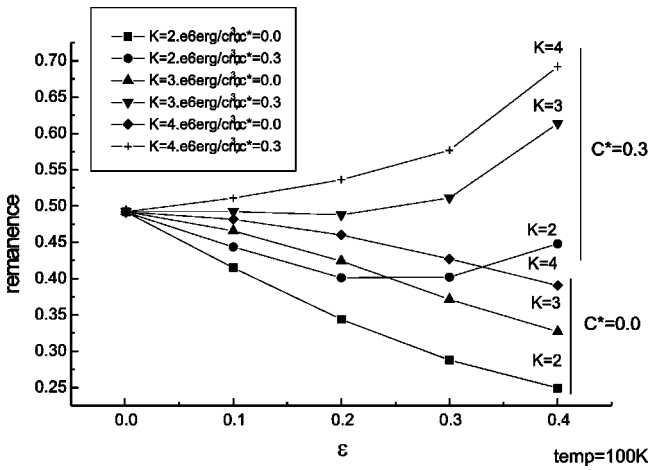

FIG. 1. Normalized remanence for different concentrations as a function of anisotropy constant K and exchange parameter C^*.

III. RESULTS AND DISCUSSION

All the calculations were made using a primary cell containing 5000 particles. The easy axes of the particles are randomly distributed. The particle diameters and size distribution (assumed lognormal) were usually kept constant with a median diameter of $D_m = 150$ Å, a standard deviation of $\ln(D) = 0.1$, and $M_s = 1400$ emu/cm^3, corresponding to bulk Co. We examined the effects of both dipolar and exchange interactions on the magnetization process as well as the effect of the anisotropy constant. Dipolar interactions were included in all the calculations, and when required, exchange coupling between grains was introduced by means of an exchange constant $C^* = 0.3$, which corresponds to a strongly exchange coupled system. In order to vary the interaction strength the packing density of the system was varied. The packing density of the system ϵ is the volume fraction of magnetic material in the cell.

A. Magnetization and remanence curves

Figure 1 introduces the effect of different anisotropy. In the presence of exchange interactions and for a higher anisotropy constant $K = 4 \times 10^6$ emu/cm^3 the remanence increases with concentration due to the increase in the energy barrier. For lower values of the anisotropy constant these calculations show an initial decrease of the remanence for low concentrations ($\epsilon = 0.001, 0.1, 0.2$), followed by an increase in the remanence value for high packing fractions when exchange interactions are effective. For low Co concentrations, the particles are not close enough to experience exchange interactions, allowing the longer-range magnetostatic interactions to dominate; at higher concentrations, the exchange interaction acts to keep the magnetic moments of the particles aligned to reduce the exchange energy.

For the case of zero-exchange coupling, the remanence decreases monotonically with ϵ for all values of K. This is in contrast to 2D simulations, as Ref. 24, and indicates the presence of strong demagnetizing fields. It might be expected that the presence of competing dipolar and exchange interactions would have some effects of frustration on the mag-

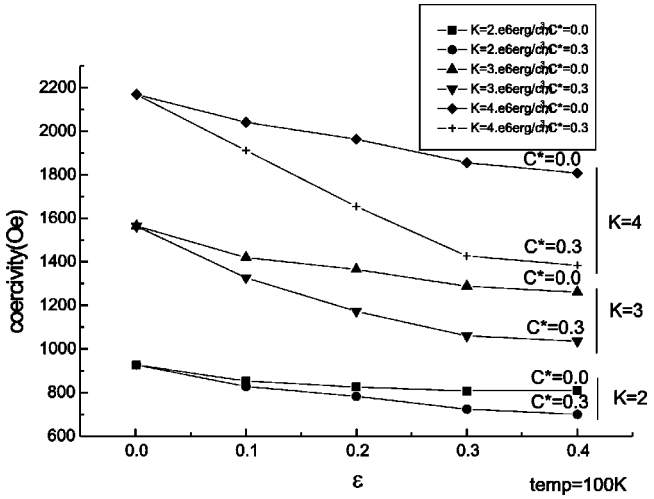

FIG. 2. Coercivity for different concentrations as a function of anisotropy constant K and exchange parameter C^*.

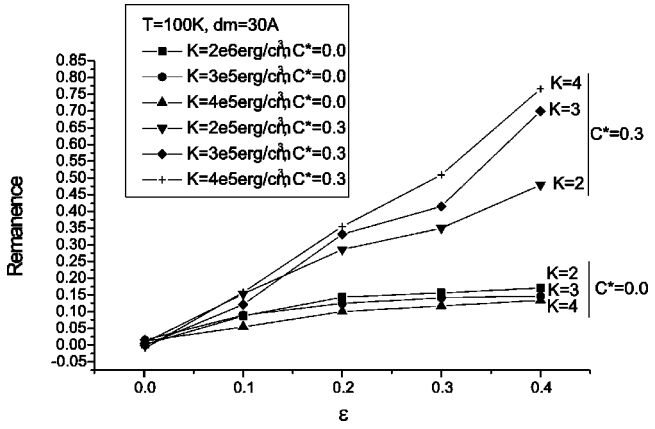

FIG. 3. Remanence for smaller values of KV/kT showing an increase in irreversible behavior with concentration.

netic properties. This is apparent in the minimum in the remanence predicted in Fig. 1 for $K=2\times10^6$ emu/cm^3: at low density the average particle separation is too large in relation to the cutoff of the exchange field to have an appreciable effect: hence, the low-density behavior is dominated by the dipolar interactions. At higher densities the exchange begins to dominate, leading to a predicted increase in remanence. At larger values of K the minimum vanishes and a monotonic increase of M_r with ϵ is seen. This is because, first, the large anisotropy mitigates against the formation of closed loop structures, leading to a relatively slow decrease in remanence. Second, it should be noted that in the calculations here the magnitude of the exchange field scales with H_k; thus, the magnitude of the exchange field relative to the dipolar interactions is larger for large K.

Figure 2 shows the decrease of the coercivity with the packing fraction. For the case of dipolar interactions only, this decrease arises because the dipolar interactions favor the demagnetized state.[19] When we add exchange, we see a more rapid decline in the coercivity with concentration, due to the onset of collective magnetization reversal.[16] In 2D simulations such as those given in Ref. 16, exchange results in a large degree of correlation. In our case the correlation function lengths are relatively small, due to the low packing density and the spatial disorder. However, there remains a strong tendency for the formation of flux closure configurations under the influence of the magnetostatic interactions, leading to a short-ranged order. The exchange interactions give an enhancement of the remanence. However, the predicted hysteresis loops are significantly less square than the 2D case for similar parameters, suggesting that collective reversal is less important. We ascribe this to the spatial disorder which will naturally limit the correlation length, as will be discussed later.

The effects of interactions on superparamagnetic systems are somewhat complex in their dependence on ϵ and K. Figure 3 shows the remanence of a system of particles with a lower median diameter $D_m=30$ Å to illustrate the effects of the superparamagnetic particles in the system. It is important to note that, although the SPM fraction is treated separately, it is linked to the ferromagnetically stable fraction via the dipolar and exchange coupling. For these systems unlike those in Fig. 2, the remanence increases for both cases of dipolar only and dipolar and exchange interactions. For the case of dipolar interactions only, for small concentrations the particles do not experience any interactions and the remanence is equal to zero, as expected for behavior of SPM particles. When we increase the concentration, the particles start to experience the magnetic interactions of the surrounding particles. A study by Dormann *et al.*[4] shows the additional energy barrier due to dipolar interactions. Because of this, some of the SPM particles become blocked, leading to an increase in remanence, and consequently our predictions are in this sense supportive of the model of Dormann *et al.*

B. Switching field distributions

The effects of interactions on the energy barrier distribution are evident in the switching field distribution, which is a measure of irreversible magnetization changes. Here the SFD is calculated as the differential with respect to H of the dc demagnetization remanence curve. This is obtained by applying a negative field to a system initially in the saturated remanent state, following which the field is removed. The resulting remanence $I_d(H)$ is conventionally plotted as a function of H. An important related quantity is the remanence coercivity H_r, which is the field for which $I_d=0$.

Figure 4 shows the SFD, considering only dipolar interactions for a low-anisotropy constant of $K=2\times10^6$ emu/cm^3. For the noninteracting case, we can see that almost all the particles start to rotate at the same field with a small distribution due to the distribution of particle sizes. The smaller particles rotate earlier due to the lower-energy barrier, which is proportional to KV. As the Co concentration is increased, the dipolar interactions spread out the SFD and the shape of the curve becomes more asymmetric. From comparing Figs. 1 and 4 we observe that the dipolar interactions favor the demagnetized state, but make the rotation of the particles towards negative saturation more difficult. This is consistent with the formation of flux closure structures. The model of Dormann *et al.*[4] gives rise to a net increase in

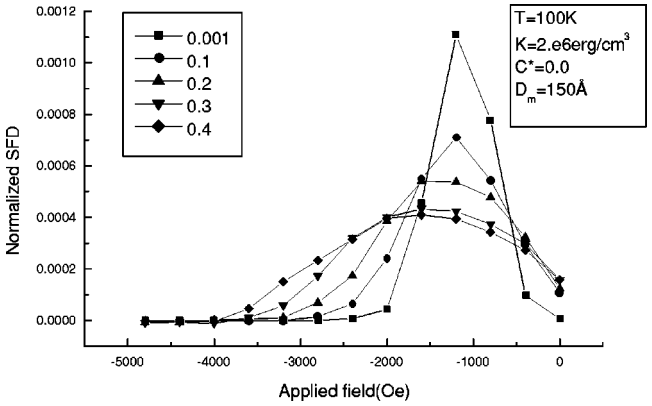

FIG. 4. Normalized switching field distribution as a function of concentration for a system with magnetostatic coupling only. Individual curves refer to different packing densities, as given in the legend.

the mean energy barrier evidenced by the shift in the peak to larger negative fields. Clearly, the change in width of the energy-barrier distribution implied by the increased width of the SFD is also an important factor and is not predicted by the model of Dormann *et al.* Essentially, the model of Dormann *et al.* uses an averaging technique to determine the change of mean energy barrier and ignores the fluctuations in the local environment of each particle. The increase in width of the SFD arises from the local fluctuations, which are clearly an important aspect of the physical description of the problem.

We are also interested in the study of the effect of the anisotropy on the switching field distribution. We have noticed significant variations in the SFD when we increase the anisotropy constant value. Figure 5 shows the SFD for the case with no exchange interactions for a value of $K=4\times10^6$ emu/cm^3. The high-anisotropy constant resists the formation of flux closure loops, preventing the rapid demagnetizing seen in Fig. 4 for the case without exchange interactions. This results in a more symmetric and broader SFD whose peak position is shifted to higher fields due to the

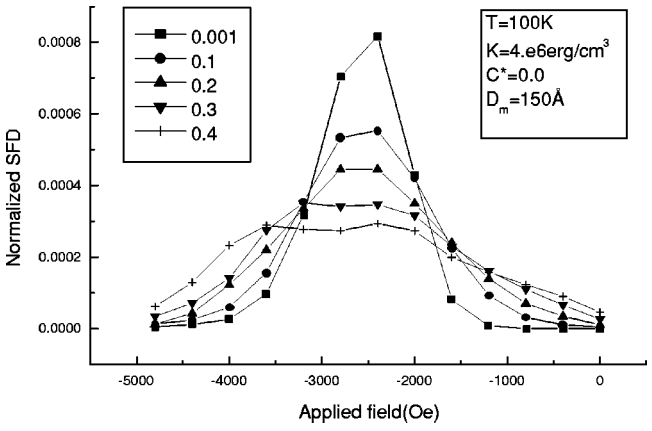

FIG. 5. Normalized switching field distribution as a function of concentration for a system with magnetostatic coupling only for systems with large anisotropy. Individual curves refer to different packing densities, as given in the legend.

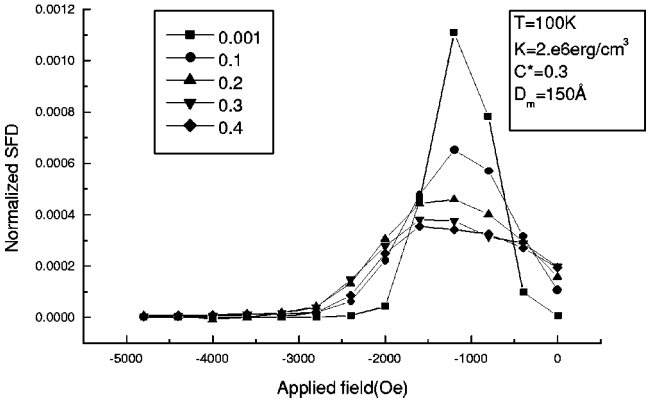

FIG. 6. Normalized switching field distribution as a function of concentration for a strongly exchange-coupled system. Individual curves refer to different packing densities, as given in the legend.

higher anisotropy. It is interesting to note that the SFD has become very broad and flat in this case. This is indicative of a very wide dispersion of energy barriers, perhaps resulting from the appearance of a large number of shallow local minima resulting from the competition between the anisotropy and the magnetostatic energy. The magnetization reversal then proceeds via a number of intermediate states defined by these minima. These results are consistent with previous work on the initial susceptibility of interacting fine particle systems[14] which showed that at high density the variation of susceptibility with temperature becomes very flat, again indicative of a very wide energy barrier distribution. We have also carried out some initial calculations of the time dependence of the magnetization (to be published separately) which are also consistent with the existence of an energy surface with many small local minima.

For the case of low anisotropy, in the presence of exchange as shown in Fig. 6 there is remanence enhancement due to exchange. However, the SFD in small negative fields is not significantly different from the case of magnetostatic coupling only. This is not consistent with the expected effect of exchange being to stabilize the magnetized state. The low anisotropy cannot stabilize the remanent state and the tendency to flux closure due to the magnetostatic interactions is the most significant factor as the field is made more negative. We note that the low-anisotropy case exhibits less cooperative reversal as evidenced by the long tail in the SFD in large fields. This suggests that coupling has a dramatic effect on the dynamic behavior of the system, the frustration arising from competition between the dipolar and exchange interactions. This same competition results in the dip observed in the remanence as a function of packing fraction for the same value of anisotropy constant shown in Fig. 1.

Figure 7 shows the same system with strong exchange coupling, but this time with the higher-anisotropy constant of $K=4\times10^6$ emu/cm^3. Once again, for small concentrations the dipolar interactions dominate, but when we increase the concentration the cooperative effects due to the exchange interaction become significant and the remanence coercivity decreases. In negative fields, particles of smaller size and anisotropy start to rotate first, with strong coupling to other particles producing cooperative demagnetization. The effect

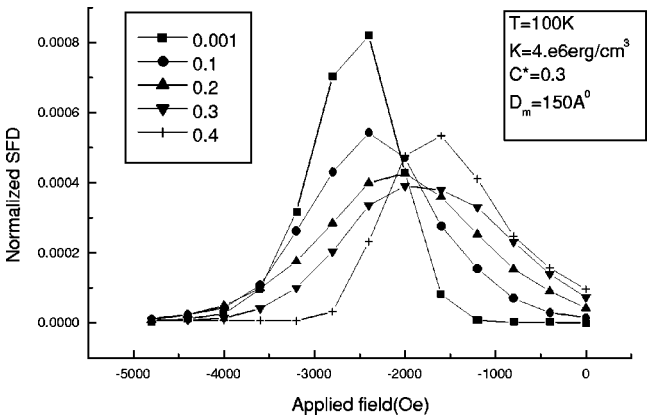

FIG. 7. Normalized switching field distribution as a function of concentration for a system with magnetostatic and exchange coupling for systems with large anisotropy. Individual curves refer to different packing densities, as given in the legend.

is more pronounced for the case of strong dipolar and exchange interactions. We can also observe that initially the SFD becomes wider, due to the dispersion in the local field when we increase the concentration. As the concentration is increased, the exchange becomes more important, and thereafter the increase of SFD in small fields is less pronounced. The exchange energy tends to maintain the alignment of the moments; this leads to a large gradient of the SFD in large fields, as once the field overcomes the energy barrier, cooperative rotation occurs.

C. Dependence of coercivity on KV/kT

To see the effects of the temperature in a system with dipolar interactions only, we have plotted the coercivity as a function of concentration for different temperatures in Fig. 8. For low temperatures the coercivity decreases with the concentration. Even when the particles are very small, if the temperature is low enough, almost all the particles are blocked. In this case we expect behavior similar to that of Fig. 2 in which the coercivity decreases with concentration. If we increase the temperature to 100 K, all the particles become SPM and the coercivity increases with packing fraction. Mørup demonstrates that in samples of fine particles, the dipolar energy becomes comparable to the thermal energy and therefore the particles start to block.[25] For small concentrations, the particles do not interact and the behavior is purely superparamagnetic with zero coercivity. As the particles become closer, the dipolar interactions start to be important and block the moments, generating a coercivity. At an intermediate temperature of 10 K the coercivity is constant with packing fraction.

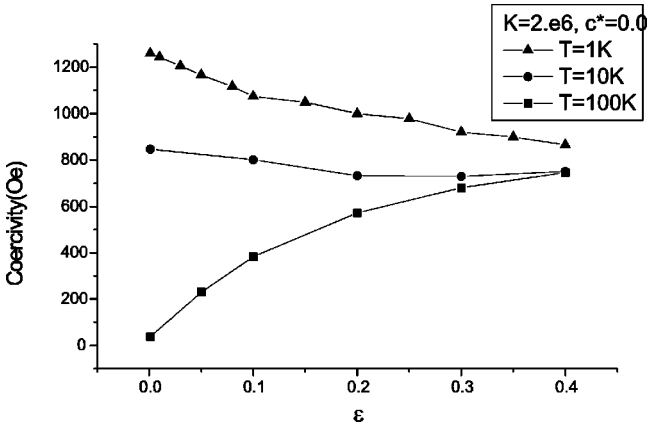

FIG. 8. Coercivity as a function of packing fraction showing three regimes of behavior dependent on KV/kT.

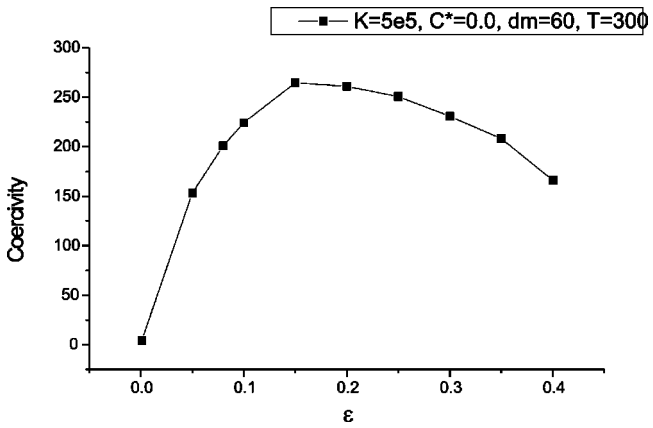

FIG. 9. Coercivity as a function of packing fraction for a system with small KV/kT. The peak reflects a slow transition to a system with local magnetic order.

The variation of the coercivity for different concentrations is plotted in Fig. 9 for small particles and a small value of the anisotropy constant. These data exhibit a maximum which we ascribe to the competition of two different processes. At low concentrations the particles are superparamagnetic, but if we increase the concentration and thereby the dipolar interaction, then the particle moments begin to block. If we keep increasing the concentration, the interactions become more important, and leading to longer-range correlations, specifically, the magnetostatic interaction produces flux closure loops which decrease the coercivity as can be seen for the case of blocked particles. Clearly, the effects of interactions on the properties of granular systems are highly complex. The model of Dormann *et al.*[4] predicts a monotonic increase of energy barrier with interaction strength. However, Figs. 8 and 9 indicate that the model of Dormann *et al.* cannot be applied under all circumstances, since it would be expected to lead to an increase of H_c with interaction strength. It can be seen that an increase in H_c with interaction strength is only reserved for weakly interacting systems having small values of KV/kT, in which the energy barriers arise principally from interactions. The maximum observed in Fig. 9 suggests that the transitions to an ordered state, leading generally to a reduced coercivity, cannot be neglected in practice. We also note that Kechrakos and Trohidou[19] have observed a similar peak in the coercivity as a function of concentration for a system with $K=0$. Here we have demonstrated that the important factor is KV/kT. In order to observe the peak experimentally it is necessary to study a system with KV/kT small enough to be predominantly superparamagnetic, but of sufficiently large moment so that the magnetostatic interactions are strong at high densities.

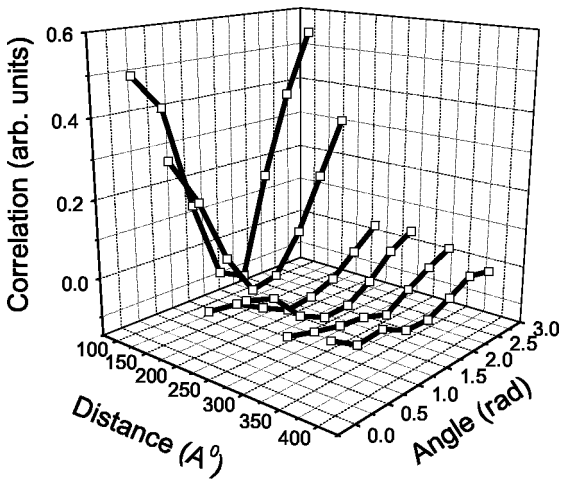

FIG. 10. Radial and angular dependence of the correlation function for a system with magnetostatic interactions only.

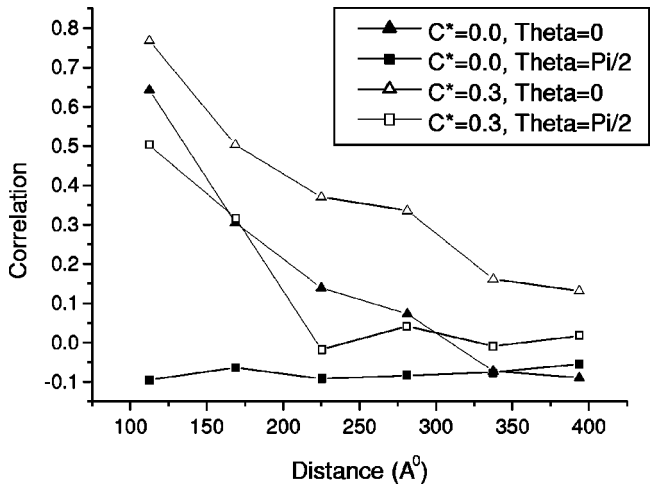

FIG. 11. Radial dependence of the correlation function for orientations parallel ($\theta=0$) and perpendicular ($\theta=\pi/2$) to the local magnetization. The data refer to systems with magnetostatic coupling only and with both magnetostatic coupling and exchange.

Evidence for the existence of correlations can be obtained by investigating a correlation function of the form

$$\xi=\langle \vec{\mu}_1 \cdot \vec{\mu}_2 \rangle. \tag{7}$$

ξ is clearly dependent on the position relative to the particle under consideration. Here we determine ξ in a coordinate system based on the magnetization direction of the particle under consideration. By this means ξ becomes sensitive to local anisotropies in the magnetic microstructure such as closed loop configurations which would be averaged out in the global coordinate system.

Figure 10 gives the radial and angular dependence of the correlation function for a system with magnetostatic interactions only, for $K=2\times10^6$ erg cm^{-3}, $T=4$ K, and $\epsilon=0.4$. It can be seen that the system is strongly correlated with a correlation length of a few particle diameters. Equally important is the form of the correlation function, which is consistent with the existence of flux closure configurations. Specifically, we note the strong angular dependence of ξ for small separations. The strong correlations occur along the local magnetization direction, with smaller, negative, values at an orientation perpendicular to this direction, as would be expected for magnetostatically induced clusters. The correlation function for a system with the same parameters, but now including strong exchange coupling, does not show the anisotropy evident in Fig. 10. This is a result of the exchange coupling which leads to short-ranged ferromagnetic order. The difference in the form of local order between the two cases is demonstrated in Fig. 11, which shows the radial dependence of the correlation function for orientations parallel ($\theta=0$) and perpendicular ($\theta=\pi/2$) to the local magnetization. In the case of the magnetostatically coupled system the angular dependence is dramatic, with strong positive correlation parallel to the magnetization direction and a weaker, negative value perpendicular, which is to be expected given the form of the dipolar magnetostatic interaction. The behavior of the exchange-coupled system shows stronger correlations and the (isotropic) form of the correlation function is again consistent with the form of the exchange interaction. The magnetostatically coupled system shows a rapid decrease in ξ with distance for $\theta=0$, which indicates that the order in this case is either in the form of correlated particle pairs or that the flux closure structures have a small radius. The variation for $\theta=\pi/2$ is less pronounced and indicates negative correlations out to several particle diameters, which is consistent with the formation of clusters or, in the case of particle pair formation, is indicative that the pairs order antiparallel under the influence of the magnetostatic interaction. As mentioned previously, the correlation length, at around two particle diameters, is rather small, which presumably reflects the disordered physical microstructure. These results are consistent with the experimental data of Franco *et al.*[17] Here measurements of the parameter $\Delta M(H)$, derived from a comparison of the principal remanence curves, demonstrate a change from predominantly magnetostatic interactions to (ferromagnetic) exchange interactions with enhanced remanence as the exchange between grains is increased by annealing.

Clearly, there is strong evidence for short-ranged correlations in the system, whose form depends on the strength and form of the intergranular interactions. These correlations have a direct bearing on the magnetic properties. In addition, the transport properties of the system themselves are dependent on local correlations, and so we conclude this study with an investigation of the effects of correlations on the GMR.

D. Transport properties

The spin-dependent transport properties are studied via the spin-spin correlation function [Eq. (6)] according to the theory of Gittleman *et al.*[26] This has been evaluated by calculating the average spin-spin correlation function for the magnetization at a given field. We calculated the correlations within a cutoff distance corresponding to twice the spin diffusion length which in this case was $1.5D_m$. Clearly, the GMR is a macroscopic reflection of the micromagnetic cor-

relations considered at the end of the previous section. However, we stress the difference between the correlation functions described by Eqs. (6) and (7) in that the former is restricted in range by the spin diffusion length, which is a factor which must be considered in relating the GMR to micromagnetic correlations. The calculated MR curves at different concentrations for the dipolar-only and dipolar and exchange cases show a form similar to that observed experimentally. The experimental data given in Ref. 11 for Co-Ag films show dramatically different behavior for films with packing densities of $\epsilon = 0.3$ and 0.5. The lower packing density shows a GMR which is close to quadratic in M, whereas the higher packing density shows strong deviations from quadratic behavior. This change is accompanied by a change in character of the magnetic behavior from essentially superparamagnetic to ferromagnetic behavior at the higher density, dominated by the properties of quasidomains formed by the intergranular exchange coupling. This behavior (including a reduction in the absolute magnitude of the GMR) is perfectly consistent with our model calculations and the conclusion that the GMR is reduced in strongly ferromagnetically correlated systems.

Here we characterize the form of the GMR curves using the maximum height of the GMR curve and a ''linewidth'' given by the field width at half the maximum height of the GMR curve. The GMR can be used as a measure of the degree of local correlation in the system. For dipolar interactions only, the maximum value of the magnetoresistance is near the coercivity value in agreement with experiment. There is a small increase in the GMR when we increase the concentration. This is due to two reasons: first because, when we increase the concentration, the grains are closer and more scattering events can occur. Second, the dipolar interactions encourage the formation of flux closure loops, which increases the magnetic disorder of the system. On the other hand, we observe an increase in the linewidth, which means that we have more disorder at higher fields. This local disorder increases the spin-dependent scattering and increases the resistance. The effect of the exchange interactions is to give rise to a decrease in the magnetoresistance with concentration. This supports the theory of cooperative effects between neighboring grains which experience exchange coupling. The local alignment of the grains is detrimental to magnetoresistance because it reduces the magnetic disorder and the spin-dependent scattering of the electrons.

In order to quantify this behavior we consider first the variation of the magnetoresistance ''linewidth'' with concentration for different anisotropy constants and coupling as shown in Fig. 12. For the case of dipolar interactions only we observe an increase of the linewidth with concentration. Taking into account that the GMR is a measure of the misalignment of the grains in the cell this means that spin-dependent scattering starts at lower fields. For the case of exchange, the GMR linewidth decreases with concentration because the moments of the particles are aligned. The peak height of the GMR can be seen in Fig. 13 where the GMR can be seen to decrease when increasing the anisotropy. A system with a large value of anisotropy is less mobile, which means that there is less disorder on the local scale defined by the spin

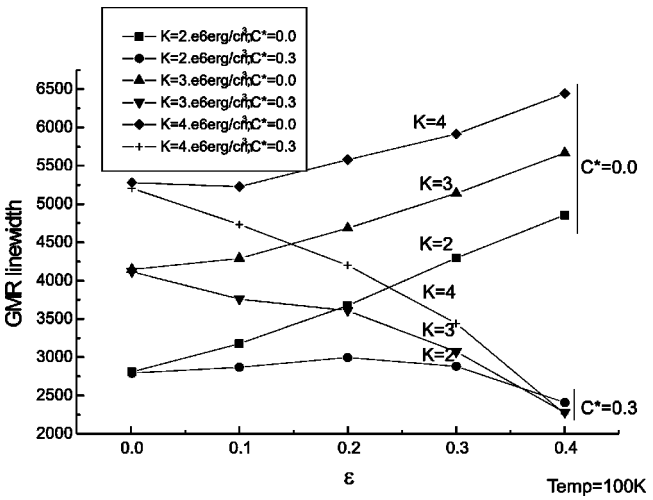

FIG. 12. GMR linewidth as a function of anisotropy constant.

diffusion length. We can also observe that the GMR height increases for the dipolar interactions when we increase the concentration. For the case of exchange interactions we can observe a decrease in the height of the peak when we increase the concentration due to the correlation in the system between the magnetic moments of the particles. Finally, Fig. 14 shows the GMR peak height versus the reduced magnetization, which is given by M/M_s. The exchange interactions result in a reduction of the GMR and a deviation from the theoretically predicted parabola. If the moments within a radius drawn out by the spin diffusion length are correlated due to exchange, then there will be little or no spin-dependent scattering and hence no GMR. This is evidence for cooperative reversal.

IV. CONCLUSIONS

In this article we have studied the effect of both dipolar and exchange interactions on the remanence, coercivity, switching field distribution, and giant magnetoresistance for different concentrations and anisotropy constant. The model system consists of randomly situated fine particles of Co with uniaxial anisotropy.

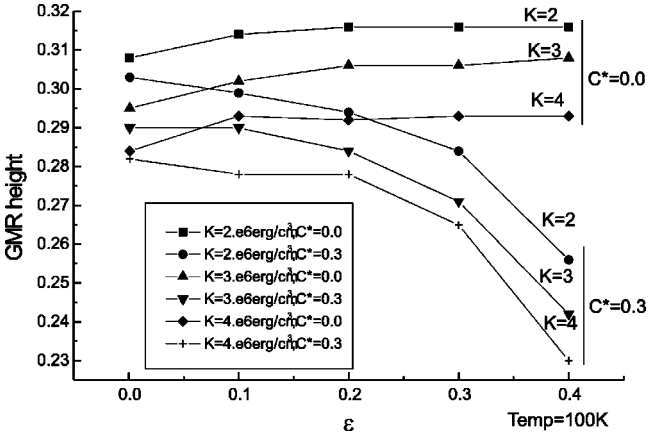

FIG. 13. Maximum GMR as a function of concentration for different anisotropy values K and exchange constant C^*.

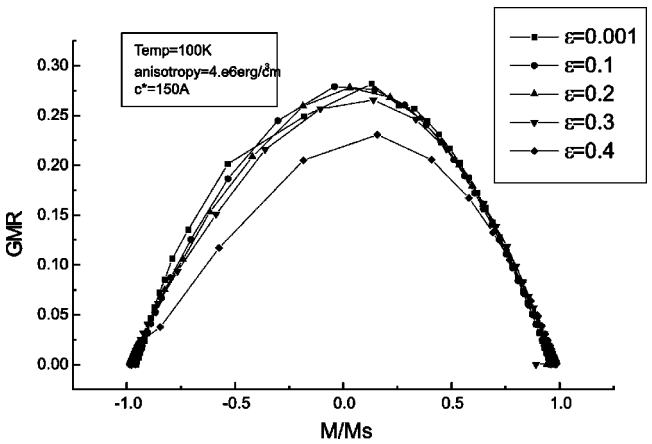

FIG. 14. GMR vs reduced magnetization as a function of concentration for an exchange coupled system with $C^*=0.3$.

The results suggest that in the absence of exchange interactions, the system is dominated by the formation of closure vortex structures due to magnetostatic interactions. The closure structures generally favor the demagnetized state and increase the magnetic disorder. As the packing fraction—and hence the magnetostatic interactions—is increased, the remanence and coercivity therefore decrease and the GMR increases. The SFD reveals a rapid demagnetizing process followed by a slower approach to negative saturation, producing an asymmetric curve. The presence of local interactions also has the effect of broadening the SFD and GMR curves.

A strong particle anisotropy mitigates against the formation of these closure loop structures, thus adding extra inertia to the reversal process. An increase in the anisotropy constant therefore results in a higher remanence and coercivity and lower GMR. As the rapid demagnetizing is mitigated, the SFD becomes broader with increased K and more symmetrical.

For superparamagnetic particles with zero remanence in the noninteracting case, the increase in packing fraction and the onset of interaction effects represent an increase in the energy barrier, which results in an increase in remanence. This interdependence of the effect of interactions with the value of KV/kT was demonstrated by investigating the variation of coercivity with packing fraction for three different temperatures representing three different values of KV/kT. For the superparamagnetic case the coercivity increases with dipolar interactions; for the blocked case the coercivity decreases, and an intermediate temperature can be found where the coercivity remains constant with increasing packing fraction. Similarly, a system with low anisotropy which is superparamagnetic at room temperature with zero coercivity will become increasingly blocked as the packing fraction is increased due to the increase in energy barrier caused by the increasing interactions. However, as the packing fraction is increased still further, flux closure structures are able to form which have a demagnetizing effect, eventually depressing the coercivity.

The introduction of short-range exchange interactions typically results in alignment of the particle moments and encourages cooperative reversal. This also tends to increase the size of the vortex structures. The tendency for local alignment results in an increased remanence and the cooperative reversal in a decrease in the coercivity. Both features result in a reduction in the GMR and its linewidth. As the exchange interaction is a much shorter range force than the magnetostatic interactions, there is some competition between these effects as the packing fraction is increased and a system that was dominated by dipolar interactions becomes increasingly influenced by the exchange field. This can be seen in the dip of the remanence as a function of packing fraction curve and in the SFD. In particular, the remanence decreases relatively rapidly in small fields, which is not consistent with the expected effect of exchange coupling in stabilizing the magnetization. This indicates the importance of the competition between magnetostatic and exchange coupling which leads to a decrease in local energy barriers via the effects of frustration. The computations indicate that even in the relatively disordered systems studied here there are significant correlations at the micromagnetic level which must be considered in interpreting the magnetic and transport properties of the systems.

ACKNOWLEDGMENTS

The financial support of the EU is gratefully acknowledged, Contract No. CT97-0124.

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