Scaling behavior of the giant magnetoresistance of magnetic aggregates

D. Kechrakos and K. N. Trohidou
Institute of Materials Science, NCSR ''Demokritos,'' 15310 Aghia Paraskevi Attiki, Greece

J. A. Blackman
Department of Physics, University of Reading, Whiteknights RG6 6AF, United Kingdom

(Received 5 October 2000; revised manuscript received 20 December 2000; published 12 March 2001)

We study numerically the giant magnetoresistance (GMR) of magnetic nanoparticle aggregates formed on a planar substrate by a diffusion process that is driven by the magnetostatic dipolar interactions between the particles. The growth of aggregates is modeled by a diffusion limited cluster aggregation model and the GMR of the assembly is expressed in terms of the short range spin correlation function. The latter is obtained by a Monte Carlo simulation. We show that the spin correlation function of the aggregates can be described by a scaling law that involves the fractal dimensionality of the structure. The formation of aggregates causes a large reduction of the low-field GMR because the role of interparticle dipolar interactions is enhanced due to the chainlike morphology of the aggregates.

DOI: 10.1103/PhysRevB.63.134422 PACS number(s): 75.70.Pa, 75.90.+w, 75.50.Mm, 75.50.Tt

Giant magnetoresistance (GMR) was first observed in granular films consisting of Co grains dispersed in a Cu matrix.[1,2] It was also demonstrated that in all samples the GMR scales as the square of the global magnetization M.[2] Deviations from the parabolic dependence at low fields ($M \sim 0$) were subsequently observed by various groups and were attributed to the grain size distribution[3] and to dipolar interactions among the grains.[4] Numerical studies gave support to the argument that dipolar interactions are mainly responsible for the flattening of the GMR vs M parabola at low fields[4–7] and it has recently been pointed out that dipolar interactions cause a more dramatic flattening when the system exhibits a wide distribution of grain sizes.[8] These results underline the essential role that interparticle interactions play in the modification of the GMR effect.

However, dipolar interactions among the magnetic particles are also of great importance during the growth process of the films. When magnetic particles diffuse on a substrate they move and relax their magnetic moments in a way that minimizes their mutual magnetostatic energy. The diffusion process is not a Brownian motion, but it is instead driven by the energetics of the interactions between the particles. When they unite, the morphology of the resulting aggregates is sensitive to the substrate temperature, the particle density and the strength of the interparticle interactions. At low particle concentrations (2–5 %), it has been shown that magnetostatic forces between the particles lead to formation of aggregates with a self-similar morphology characterized by a fractal dimensionality[9] (D_f). The magnetization of these aggregates obeys a scaling law with respect to the size of the aggregate (N) where the exponent is the fractal dimensionality.[10] This scaling law reflects the impact of the correlations in particle positions on the magnetic properties of the aggregate.

Recent numerical studies pointed out the importance of spatial correlations of the magnetic regions on the magnetotransport properties of a composite system. In particular, it was suggested that a linear array of superparamagnetic particles, coupled via dipolar forces, produces large GMR values when a magnetic field is applied perpendicular to the array axis.[5] In addition, the field dependent GMR of a magnetic multilayer with layer thickness following a Fibonacci sequence was shown to exhibit a self-similar sequence of plateaus as a consequence of the quasiperiodic structural order.[11] These results indicate that in systems such as granular films and magnetic multilayers where the GMR effect occurs as a result of spin dependent scattering, the spatial arrangement of the magnetic regions (whether granules or layers) has a strong impact on the GMR values, as it affects the micromagnetic configuration of the system.

The influence of the spatial morphology of a granular film on the GMR values has not been systematically addressed so far. An approximation that is most commonly adopted with respect to the morphology of the granular films is that the magnetic particles are dispersed randomly in the nonmagnetic matrix, or, in other words, that their locations are uncorrelated. This approximation describes satisfactorily the morphology of granular samples grown by an annealing process.[1–4] However, in some cases, granular systems with correlated structures occur. This is, for example, the case of ferrofluids where the aggregation process is dominated by magnetostatic dipolar interactions and leads to various fractal morphologies.[9] To the best of our knowledge, no GMR measurements on frozen ferrofluids have yet been reported. During the growth of granular metals or insulators by the recently developed technique of low-energy cluster beam deposition,[12,13] codeposition of nonmagnetic atoms, and magnetic particles of a transition metal takes place. In this case, we would expect that energy-driven diffusion and aggregation occur up to some extent, especially in dilute samples.

The aim of the present work is to establish a relation between the spatial morphology of a granular film containing aggregates of magnetic particles and the GMR value. The aggregates are formed by a diffusion process that is driven by magnetostatic dipolar interactions. We show that the GMR of an assembly of self-similar magnetic aggregates obeys a scaling law with the exponent being the fractal dimension of the aggregates and, furthermore, the formation of aggregates degrades substantially the GMR effect at low fields.

Our model assumptions correspond to the growth of samples by a cluster beam deposition technique,[12,13] that produces nearly monodisperse samples. For simplicity, we neglect the thickness of the film, because the diffusion of particles and the formation of aggregates is expected to take place during the growth of a single layer, i.e., before the particles are completely buried by the matrix material that obstructs the diffusion process. We model the aggregation of spherical magnetic nanoparticles on a planar substrate by a diffusion limited cluster aggregation process on a triangular lattice.[9]

The nanoparticles are initially located at random on the sites of the triangular lattice and they are allowed to diffuse. The interaction energy between a pair of particles with magnetic moments $\mathbf{m}_i$ and $\mathbf{m}_j$ separated by distance r_{ij} is

$$E_{ij}=g[\mathbf{m}_i\cdot\mathbf{m}_j-3(\mathbf{m}_i\cdot\mathbf{u}_{ij})(\mathbf{m}_j\cdot\mathbf{u}_{ij})]/r_{ij}^3, \tag{1}$$

where $\mathbf{u}_{ij}$ is the unit vector along $\mathbf{r}_{ij}$, $g=m^2/d^3$ is the dipolar energy strength, m is the magnitude of the dipole moment, and d the diameter of the particles. The other relative energy parameters used are the Zeeman energy $h=mH$ and the thermal energy $t=k_BT$. The energy unit in our simulations is the dipolar energy ($g=1$) and the unit of length is the particle diameter ($d=1$). Simulations[9] yielded a fractal dimensionality $D_f=1.23\pm0.04$ for the resulting aggregates. This value shows the tendency of the particles to form chainlike aggregates. The particle moments are aligned nose-to-tail in the plane.

The GMR in granular films is attributed to spin dependent scattering of the free electrons. Provided that the electron mean free path is longer than the size of the individual particles, each magnetic particle can be considered as a point scatterer described by a local potential which is proportional to the relative orientation between the local magnetization vector and the electron spin quantization axis.[14] Within the Born approximation for scattering, the resistivity of the sample is then proportional to moment-moment correlation function between nearest neighbor moments[15]

$$R(H)=R_0-R_1f(H), \tag{2}$$

where R_0 and R_1 are material dependent parameters, $f(H)=\langle\mathbf{m}_i\cdot\mathbf{m}_j\rangle_\lambda$ is the moment-moment correlation function and λ is the electronic mean free path. Here, we assume[14] that the electron mean free path is the relevant length scale for averaging the magnetic correlations. For the dilute granular metal $Cu_{1-x}Co_x$ at room temperature, the electronic mean free path is of the same order as the average distance between adjacent particles ($\lambda\geqslant d$).[4] Therefore, the moment correlation function between nearest neighbor particles is considered in Eq. (2). In the opposite limit, namely, in the case that the condition $\lambda\ll d$ was fulfilled, an appropriate description of transport in the granular system would require a resistor network approach,[16] rather than Eq. (2). The magnetoresistance at field H is defined as $\mathrm{MR}(H)=[R(H)-R(0)]/R(0)$, where H is the applied field. The material dependent parameters R_0, R_1 enter the definition of MR and, in principle, they can be obtained by fitting Eq. (2) to the experimental results for a particular system. Provided that the material parameters R_0, R_1 are field independent, the effect of interparticle interactions enters through the values the correlation function $f(H)$ only. If the high field parabolic asymptote of the MR versus magnetization is subtracted from the MR values, one obtains the reduced magnetoresistance,[4] given as

$$\mathrm{MR}_r(H)=1-f(H) \tag{3}$$

which is obviously independent of the material parameters R_0, R_1 and serves as a universal curve whose deviations from a parabola indicate deviations from the superparamagnetic behavior.

For noninteracting particles, the magnetic moments are uncorrelated and therefore $f(H)=M^2$, where $M=\langle\mathbf{m}_i\rangle$ is the normalized sample magnetization, and consequently the observed parabolic dependence of MR_r on the sample magnetization is obtained. Deviations from the MR_r vs M parabola are observed at low fields ($M\sim0$) and were attributed to dipolar interactions between the magnetic moments.[4–8] Equation (3) indicates that the dependence of the MR on the micromagnetic configuration of the particle assembly enters via the short range moment correlation function. To calculate the correlation function we adopt the following model. We consider a granular film containing well separated aggregates of magnetic particles. Within each aggregate the particles are in contact and they interact strongly with each other, mainly through dipolar forces. The effective exchange interaction is proportional to the contact area of the spheres and it is therefore expected to be weak. On the other hand, dipolar interactions between particles belonging to different aggregates are also very weak due to the spatial separation of the aggregates. By comparing intercluster and intracluster dipolar interactions, one can show that this is justified if $d^3nN_{\mathrm{cl}}^{3/2}\ll1$, where n is the number of particles in a cluster and N_{cl} is the number of clusters per unit area. This condition holds in the case of a dilute system similar to the one we examine. Therefore, it is realistic to approximate the granular film by a collection of uncoupled aggregates with dipolar interactions between their constituent particles.

The moment correlation function of the kth aggregate containing N particles, is obtained as the thermal average of the inner product of moments averaged over all pairs of neighboring particles within the same aggregate:

$$f_N^k=\langle\mathbf{m}_i^k\cdot\mathbf{m}_j^k\rangle. \tag{4}$$

Next, the configuration average over the random shape of the aggregates is performed

$$f_N=\sum_k f_N^k \Big/ \sum_k 1 \tag{5}$$

under the assumption that all shapes are equally probable. Aggregates of various sizes and shapes are obtained from the aggregation model described previously. Finally, the moment correlation function of a film containing a distribution of aggregates is given as

$$f=\sum_N f_N \Big/ \sum_N 1 \tag{6}$$

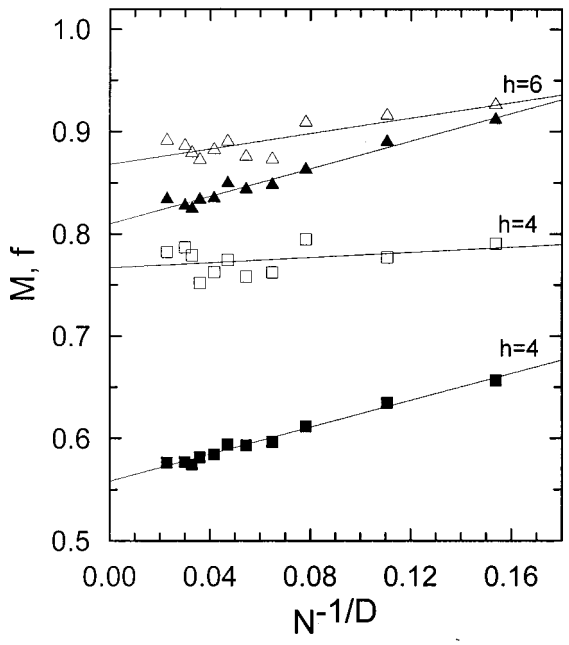

FIG. 1. Dependence of magnetization (full symbols) and short range correlation function (open symbols) on the linear size of the aggregate. Applied field (h) is normal to the substrate. Squares: $h=4$ and triangles: $h=6$. Parameters: temperature $t=10^{-4}$, dipolar strength $g=1$. Solid lines are the linear fit to the data.

provided that a uniform distribution of aggregate sizes exists in the sample.

For an aggregate containing of N magnetic particles we have shown in a previous work[10] that the magnetization follows the scaling law

$$M_N = a_m + b_m N^{-1/D_f}. \tag{7}$$

The first term (a_m) on the right hand side of Eq. (7) is interpreted as the bulk contribution to the magnetization and the second term ($b_m N^{-1/D_f}$) as the surface contribution. In Eq. (7), D_f is the noninteger (fractal) dimensionality of the aggregate. Since the bulk and the surface of a fractal object are not so clearly distinct, because the coordination number for a random fractal fluctuates Eq. (7) is a convenient way to describe finite-size effects. Similarly, finite-size effects are expected to be important for the short range correlation function of an aggregate. We therefore anticipate that the correlation function follows the scaling law

$$f_N = a_c + b_c N^{-1/D_f}, \tag{8}$$

where the first and the second term in Eq. (8) have the meaning of the bulk and surface contributions to the correlation function, respectively. The parameters a_c, b_c depend only on the temperature and the applied magnetic field.

In Fig. 1 we show the dependence of the magnetization (full symbols) and the correlation function (open symbols) on the inverse linear size ($R_g \sim N^{1/D_f}$) of the aggregate for certain values of the external field. The data in Fig. 1 are obtained at almost zero temperature ($t/g \sim 10^{-3}$) in order to maximize the ordering effects of the dipole moments and the magnetic field is applied normal to the substrate. Notice that the short range correlation shows stronger fluctuations than the long range correlation (magnetization), due to the large diversity of local environments of the sites in an aggregate. For the straight line fit, 20 aggregates, on average, for each of 11 different sizes N ($\pm 10\% N$) were used in a range of sizes from $N=10$ to 120 particles per aggregate. It is in this size range that the value of the fractal dimension $D=1.23$ describes the morphology of the aggregates.[9]

We discuss next the field dependence of the surface (parameters a) and bulk (parameters b) contributions to the magnetization and correlation function as they are defined by Eq. (7) and Eq. (8), respectively. In the zero field limit ($h \sim 0$) both b_m and b_c approach zero, indicating the disappearance of finite size effects. In the same limit, the bulk magnetization approaches zero while the bulk correlation function tends to a finite value $a_c(0) \sim 0.7$ (see Fig. 2). These results are interpreted as follows. In the zero external field limit, the dipole moments of the particles lie on the substrate plane and they are aligned in a nose-to-tail configuration. Deviations from the nose-to-tail arrangement only occur close to branching points of the aggregate. This is the minimum energy configuration as has been found previously[9] and indicated here by the positive value of the correlation function in the zero field limit. Owing to this micromagnetic configuration, the magnetization of the aggregates is zero, because it is measured along the field axis that is taken normal to the substrate. Also, the relatively high value ($a_c \sim 0.7$) of the ferromagnetic correlation function, shows that the alignment of the dipole moments is very efficient at low temperatures ($t/g=0.001$) and the demagnetizing field produced by the boundaries of the aggregate does not play an important role. This is expressed by the zero value of the surface correlation function (b_c). In the high field limit, the moments align themselves along the field axis and thus the bulk magnetization and the bulk correlation tend to unity, while surface effects become immaterial as the applied magnetic field becomes strong enough to overwhelm the local dipolar field at the surface of the aggregate.

The local dipolar field lies on the substrate plane while the externally applied field is normal to the substrate. As the external field increases it forces the magnetic moments to rotate and orient themselves off the substrate. The moments near the surface of the aggregate experience a weaker dipolar field, due to their reduced coordination, therefore, they are more susceptible to the external field. As a result of that, moments near the surface of the aggregate align easier along the field axis, thus making a positive contribution both to the magnetization ($b_m > 0$) and the correlation function ($b_c > 0$). The importance of finite size effects is maximized at intermediate field values, when the competition between the external and the local dipolar field is the strongest. This regime is identified by the presence of a peak in the field dependence of the surface parameters b_m and b_c at a certain field value (h_0). The relative positions of the peaks in b_m and b_c in Fig. 2 indicate that maximum finite size effects in the correlation function appear at higher fields ($h_0/g \sim 7$) than in the magnetization ($h_0/g \sim 5$). In other words, the

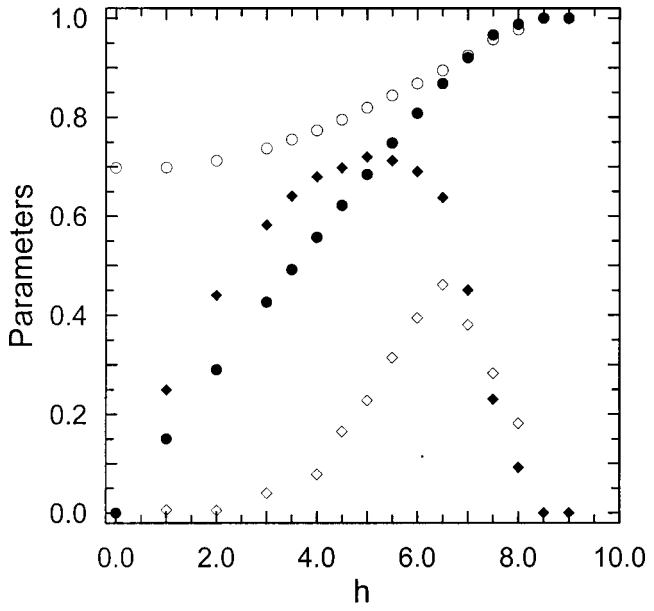

FIG. 2. Field dependence of scaling parameters. Full circles: bulk magnetization (a_m), full squares: surface magnetization (b_m), open circles: bulk correlation (a_c), and open squares: surface correlation (b_c). Parameters: temperature $t=10^{-4}$, dipolar strength $g=1$.

in-plane long range order of the magnetic moments (magnetization) is destroyed at weaker external fields than the short range order.

Knowing the field dependent scaling parameters of Eq. (7) and Eq. (8) one can compute the magnetization and reduced magnetoresistance for an aggregate of any size. This is done in Fig. 3 for two aggregates of size $N=10$ and $N=100$, in order to demonstrate the size effects on these macroscopic quantities. Obviously, higher field values are required for the larger cluster in order to reach saturation. The lack of surface effects in the zero field limit is also demonstrated in these curves. Notice also that for zero field one has $b_c(0)=0$ and $\mathrm{MR}_r(0)=1-a_c(0)$. Therefore the parameter $a_c(0)$ provides the maximum flattening of the MR_r vs M parabola (Fig. 4) which is caused by the dipolar interactions. Finite size effects are rather suppressed in the MR_r curves, because of the small values of the coefficient b_c. However, these effects can be observed more clearly if we define the normalized MR by $\mathrm{MR}_n=\mathrm{MR}/\mathrm{MR}_{\max}$, and use the scaling law, Eq. (8), to obtain

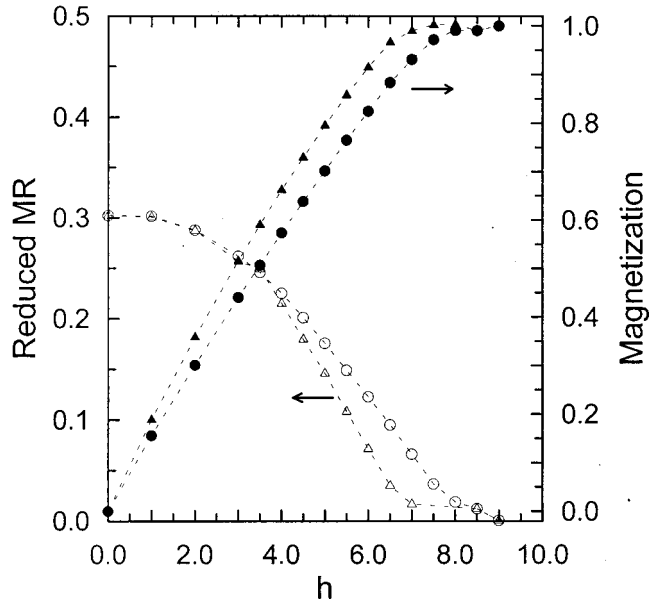

FIG. 3. Finite size effects in the field dependence of magnetization (full symbols) and reduced magnetoresistance (open symbols). Size of aggregates: $N=10$ (triangles), $N=100$ (circles).

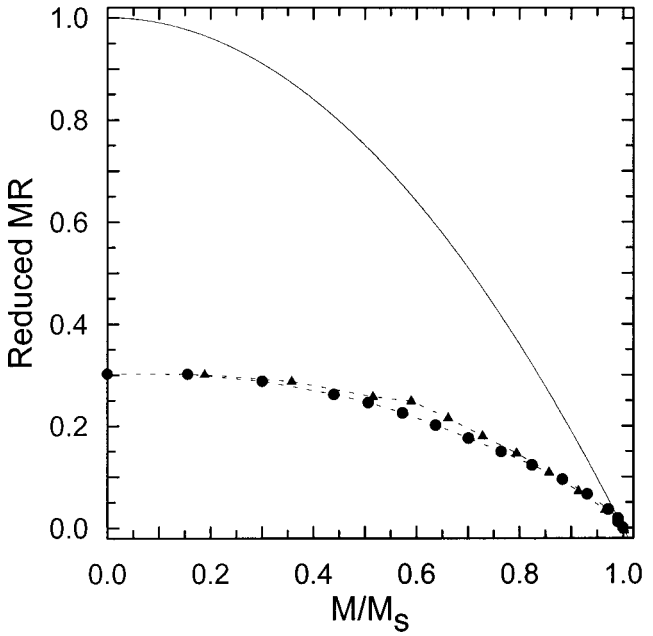

FIG. 4. Finite size effects in the reduced magnetoresistance. Size of aggregates: $N=10$ (triangles), $N=100$ (circles). Solid line: $1-(M/M_s)^2$ parabola.

$$\mathrm{MR}_n(h)=\frac{1-a_c(h)}{1-a_c(0)}-\frac{b_c(h)}{1-a_c(0)}N^{-1/D_f}. \tag{9}$$

In Eq. (9), the surface term is enhanced due to the denominator $1-a_c(0)$, being smaller than unity. Notice also, as the strength of the dipolar interactions increases the value of the zero field correlation function $a_c(0)$ increases and the second term in Eq. (9) that describes finite size effects increases. It is therefore anticipated that MR_n is the appropriate quantity to plot in order to observe finite size effects. This is demonstrated in Fig. 5, where MR_n is plotted as a function of the reduced magnetization M/M_s. Notice that, due to dipolar interactions, the MR_n values are always above those for the superparamagnetic particles (solid line in Fig. 5). This behavior is due to the positive zero-field correlation function for the dipolar particles which enters in the denominator $\mathrm{MR}_{\max}=1-a_c(0)$.

In conclusion, we have studied the finite-size scaling of the giant magnetoresistance in a system containing self-

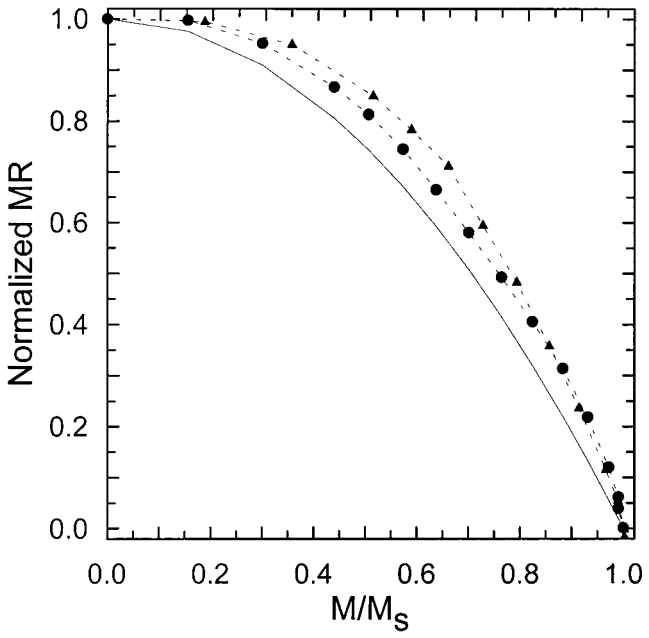

FIG. 5. Finite size effects in the normalized magnetoresistance. Size of aggregates: $N=10$ (triangles), $N=100$ (circles). Solid line: $1-(M/M_s)^2$ parabola.

similar aggregates grown by an energy-driven diffusion process on a substrate and we compare the scaling behavior of the magnetoresistance to that of the magnetization of the same system. We have shown that the fractal dimensionality that characterizes the spatial morphology of the aggregates is a suitable exponent to describe the finite size effects of both the magnetization and the giant magnetoresistance. We have found that strong ferromagnetic correlations exist in a system containing chainlike aggregates of magnetic particles causing a serious flattening in the zero field MR_r ($\sim$70%). With increasing particle density the resulting aggregates tend towards the compact limit ($D_f=2$) and the flattening of the MR_r parabola is expected to be weaker. Thus, in monodisperse nanoparticle systems, where particle aggregation takes place, the different flattening values are expected to reflect the degree of particle aggregation.

In experiments on granular $Cu_{1-x}Co_x$ samples formed by a fast annealing process, similarly large reductions of the zero field MR_r (Fig. 4) were measured by Allia *et al.* (Ref. 4) and attributed to solely to dipolar interaction effects.[4] Numerical simulations[8] taking into account the dipolar interactions and the size distribution have predicted a similar flattening of the MR_r curve. However, particle aggregation during the heat treatment and the precipitation of Co particles cannot be excluded as a factor that contributes to the MR degrade.

On the other hand, experiments with Co/Ag films grown by cluster deposition,[12] have demonstrated a reduction of the MR values from the theoretical predictions for separated and noninteracting granules. A puzzling finding in those experiments was that the degrade of the GMR effect became increasingly important as the concentration of magnetic particles approached the extreme dilution limit ($\sim$5 % at.). It has been suggested that the reason for this behavior could be the breakdown of the self-averaging hypothesis,[12] that is made in the transport theory of random alloys. However, low concentration of particles is a prerequisite for the formation of aggregates through a diffusion process driven by interparticle magnetostatic interactions. Formation of aggregates with an almost linear structure, as the ones described in this work, would seriously reduce the GMR effect and could thus provide a possible explanation for the observed behavior. Further experiments concerning MR measurements on systems containing magnetic aggregates (e.g., frozen ferrofluids) would be beneficial in order to compare directly with the calculations presented above.

[1] E. Berkowitz, J. R. Mitchell, M. J. Carey, A. P. Young, S. Zhang, F. E. Spada, F. T. Parker, A. Hutten, and G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992).
[2] J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).
[3] E. F. Ferrari, F. C. S. da Silva, and M. Knobel, Phys. Rev. B **56**, 6086 (1997).
[4] P. Allia, M. Knobel, P. Tiberto, and F. Vinai, Phys. Rev. B **52**, 15 398 (1995).
[5] D. Altbir, P. Vargas, J. d'Albuquerque e Castro, and U. Raff, Phys. Rev. B **57**, 13 604 (1998).
[6] M. El'Hilo, R. W. Chantrell, and K. O'Grady, J. Appl. Phys. **84**, 5114 (1998).
[7] D. Kechrakos and K. N. Trohidou, J. Appl. Phys. **87**, 5179 (2000).
[8] D. Kechrakos and K. N. Trohidou Phys. Rev. B **62**, 3941 (2000).
[9] K. N. Trohidou and J. A. Blackman, Phys. Rev. B **51**, 11 521 (1995).
[10] K. N. Trohidou and D. Kechrakos, J. Phys.: Condens. Matter **10**, L255 (1998).
[11] C. C. Bezerra, J. M. de Araújo, C. Chesman, and E. L. Albuquerque, Phys. Rev. B **60**, 9264 (1999).
[12] F. Parent, J. Tuaillon, L. B. Stern, V. Dupuis, B. Prevel, A. Perez, P. Melinon, G. Guiraud, R. Morel, A. Barthelemy, and A. Fert, Phys. Rev. B **55**, 3683 (1997).
[13] S. Rubin, M. Holdenried, and H. Micklitz, Eur. Phys. J. B **5**, 23 (1998).
[14] P. M. Levy, *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1994), Vol. 47, p. 367.
[15] J. I. Gittleman, Y. Goldstein, and S. Bozowski, Phys. Rev. B **5**, 3609 (1972).
[16] X. Portier, E. Y. Tsymbal, A. K. Petford-Long, T. C. Anthony, and J. A. Brug, Phys. Rev. B **58**, 591 (1998).