

Dipolar interaction and magnetic ordering in granular metallic materials

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The coupling between magnetic clusters in granular metals is investigated assuming a classical dipolar interaction to couple the clusters. It is shown that in general the dipolar interaction energy is well described by replacing the clusters by an effective magnetic moment located at the center of mass of the cluster. The magnetization and reduced magnetoresistance of these systems are described as a function of an external magnetic field for a variety of cluster distributions. An experimental design that yields a system with high reduced magnetoresistance is suggested. [S0163-1829(98)00521-9]

I. INTRODUCTION

Magnetic coupling between low-dimensional systems has been a subject of interest in recent years, due to the interesting basic properties they exhibit. In particular, the coupling of ferromagnetic clusters embedded in a metallic matrix has generated considerable interest because these clusters are easy to obtain in ribbon form and are characterized by an isotropic giant magnetoresistance (GMR) effect,¹ even at room temperature.

For these systems, the field dependence of the resistivity is interpreted by means of a spin-dependent scattering of conduction electrons.²⁻⁴ In such a model, the resistance is higher when the magnetic moments of neighbor clusters are not aligned, and the effect of the magnetic field, which aligns the magnetic moments, increases the magnetic ordering, hence decreasing the resistance of the system.² Another explanation for the GMR effect in granular systems was proposed by Kim *et al.*,⁵ who considered the interplay between the interfacial potential barrier and the dipole-dipole scattering, without requiring a spin-dependent scattering.

All existing theories, which do not consider any magnetic interaction between the clusters, assume a dependence of the fractional magnetoresistance $\Delta R/R$ as a function of the square of the reduced magnetization M/M_s ,⁶ where M_s is the saturated magnetization. However, experimental results show clear deviations from this predicted behavior at any field strengths, providing evidence for magnetic correlations among the clusters.^{1,7,8} These magnetic correlations arise from a long-range interaction among moments and two mechanisms have been proposed to account for this coupling: Ruderman-Kittel-Kasuya-Yosida (RKKY) and dipolar interactions. However, we have recently demonstrated that for clusters whose sizes and spatial distributions are consis-

tent with experimental data, the dipolar interaction is the dominant one.⁹

In this work we use a simple model to study the coupling between magnetic clusters. We demonstrate that, to a very good approximation, we can replace each cluster by an effective magnetic moment, when dipolar interactions are considered to couple the clusters. Within this approximation, we investigate the magnetic field dependence of the magnetization and magnetoresistance of granular materials. Our approach is based on a Monte Carlo simulation, which enables us to determine the dipole-dipole correlation as a function of an externally applied magnetic field. Finally, we study the reduced magnetoresistance and magnetization of a possible experimental setup, where a large GMR effect can be obtained, providing a good candidate for the design of electronic devices.

II. MODEL AND CALCULATIONS

We consider a single domain cluster of N magnetic atoms embedded in a nonmagnetic metallic matrix. The magnetic atoms are supposed to carry the same localized magnetic moment $\vec{m}_i = \vec{m}$ and are coupled, via a dipolar interaction, with a single magnetic moment \vec{m}_l , located outside the cluster but also within the nonmagnetic metallic matrix. This interaction is simply given by

$$E_{\text{dip}} = \sum_{i=1}^N \left[\frac{\vec{m}_l \cdot \vec{m}_i - 3(\vec{m}_l \cdot \hat{n}_i)(\vec{m}_i \cdot \hat{n}_i)}{r_i^3} \right], \quad (1)$$

where r_i is the distance between the magnetic moment \vec{m}_l and the magnetic moment i of the cluster, \vec{m}_i , and \hat{n}_i denotes

a unit vector along the direction that connects the magnetic moments \vec{m}_I and \vec{m}_i . The sum is carried out over all magnetic atoms in the cluster.

Considering that the cluster is far enough from the magnetic moment \vec{m}_I , we can make a second order expansion in $\delta\vec{r}_i$, where $\vec{r}_i = \vec{R} + \delta\vec{r}_i$, \vec{R} is a vector that connects the magnetic moment \vec{m}_I with the center of mass of the cluster, and $\delta\vec{r}_i$ connects the center of mass of the cluster with the magnetic moment $\vec{m}_i = \vec{m}$. The sums involving first order terms in $\delta\vec{r}_i/R$ are zero, because they involve precisely the definition of the center of mass of a system themselves. Then, considering only second order terms in $\delta\vec{r}_i/R$, we can approximate Eq. (1) by

$$\begin{aligned}
 E_{\text{dip}} \approx & N \frac{\vec{m}_I \cdot \vec{m}_i}{R^3} - 3N \frac{(\vec{m}_I \cdot \hat{n})(\vec{m}_i \cdot \hat{n})}{R^3} \\
 & - \frac{3}{R^3} \sum_{i=1}^N \left[(\vec{m}_I \cdot \vec{\epsilon}_i)(\vec{m}_i \cdot \vec{\epsilon}_i) + \frac{(\vec{m}_I \cdot \vec{m}_i) \vec{\epsilon}_i^2}{2} \right] \\
 & + \frac{15}{R^3} \sum_{i=1}^N \left[(\hat{n} \cdot \vec{\epsilon}_i)(\vec{m}_i \cdot \hat{n})(\vec{m}_I \cdot \vec{\epsilon}_i) + (\hat{n} \cdot \vec{\epsilon}_i)(\vec{m}_i \cdot \vec{\epsilon}_i) \right. \\
 & \left. \times (\vec{m}_I \cdot \hat{n}) + \frac{(\vec{m}_I \cdot \vec{m}_i)(\hat{n} \cdot \vec{\epsilon}_i)^2}{2} + \frac{(\vec{m}_I \cdot \hat{n})(\vec{m}_i \cdot \hat{n}) \vec{\epsilon}_i^2}{2} \right] \\
 & - \frac{105}{2R^5} \sum_{i=1}^N (\vec{m}_I \cdot \hat{n})(\vec{m}_i \cdot \hat{n})(\hat{n} \cdot \vec{\epsilon}_i)^2. \quad (2)
 \end{aligned}$$

Here we use the notation $\hat{n} = \vec{R}/R$ and $\vec{\epsilon}_i = \delta\vec{r}_i/R$. Inspecting Eq. (2), we notice that it includes factors that depend on the geometry of the cluster itself, in the form of the sum of the squared distances of the individual magnetic moments to the center of mass of the cluster. These second order contributions can be neglected for distances R greater than the size of the cluster. In this case, it is possible to replace the cluster by an effective magnetic moment located at its center of mass, and whose magnitude is equal to Nm_i , where N is equal to the number of atoms of the cluster.

It is straightforward to generalize the above situation if we consider two interacting clusters, where each one is replaced by an effective magnetic moment at its center of mass. Moreover, for conditions consistent with experiments concerning geometry of clusters and relative distances between them,¹ second order terms, as given by Eq. (2), can be neglected. Then, it is possible to approximate the dipolar interaction between two clusters by replacing each one by an effective magnetic moment \vec{M}_i , located at its center of mass, with magnitude equal to $N_i m_i$, where $i = 1, 2$, represents cluster 1 and 2, respectively, and N_i is the number of atoms in cluster i . This yields

$$E_{\text{dip}} = \left[\frac{\vec{M}_1 \cdot \vec{M}_2 - 3(\vec{M}_1 \cdot \hat{n})(\vec{M}_2 \cdot \hat{n})}{R^3} \right]. \quad (3)$$

Earlier we demonstrated for nearly spherical single magnetic domain clusters that if N is chosen approximately as a thousand atoms,⁹ the dominant interaction is the dipole-

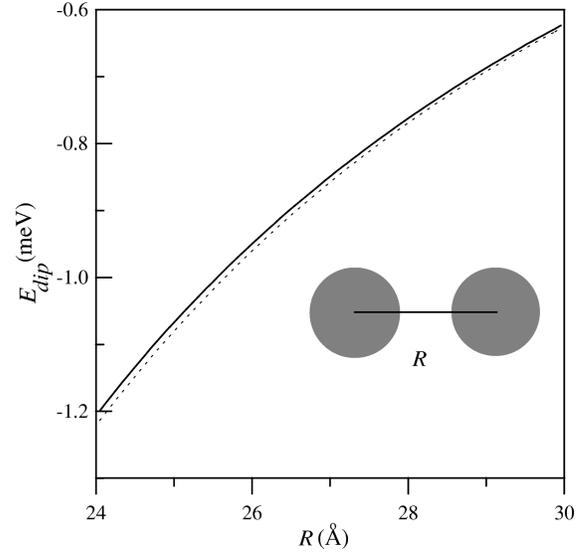


FIG. 1. Dipolar interaction energy as a function of distance between the centers of mass for two spherical clusters of 446 atoms each. The continuous line shows the exact results, whereas the dotted line shows the approximate result obtained by evaluating Eq. (3). The inset is a schematic view of the considered system.

dipole interaction, whereas the RKKY one is lower by a factor of 10. In this analysis we considered Co-Cu granular systems coupled by a dipolar interaction, neglecting the RKKY interaction. Because the spatial arrangement of the clusters in ordinary granular materials is distorted, we have randomly displaced the positions of the atoms with respect to the lattice sites according to a Gaussian probability distribution, with zero average and $0.1a_0$ of standard deviation, in each spatial direction, where a_0 is the lattice constant.

III. RESULTS AND DISCUSSION

Figures 1 and 2 illustrate results obtained for the dipolar energy interaction between two clusters (single magnetic do-

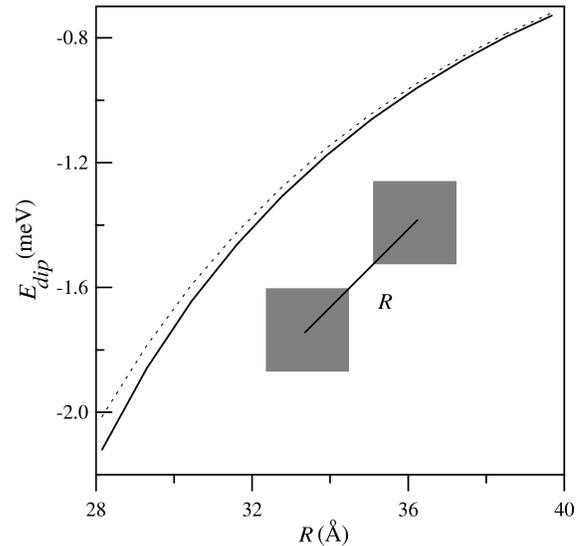


FIG. 2. Dipolar interaction energy as a function of the distance between the center of mass for two cubic clusters of 647 atoms each. The continuous line shows the exact results, whereas the dotted line shows the approximate result obtained by evaluating Eq. (3). The inset is a schematic view of the considered system.

mains) when we replace both by an effective moment located at the center of mass of each cluster, compared with the exact results obtained evaluating Eq. (1). In both cases we used $m_i = 1 \mu_B$, assuming that the magnetization of every atom of Co is $1.0 \mu_B$, as stated by Duc and Oanh,¹⁰ and we fixed the moments of the left cluster to lie in the $\langle 110 \rangle$ direction. The magnetic moment of the right cluster it found by minimizing the interaction energy. Figure 1 corresponds to results obtained for two spherical clusters, 446 atoms each, in a fcc structure with a lattice parameter of 3.6 Å. R is the distance between the centers of the spheres, and increases along the line that joins these centers, the $\langle 100 \rangle$ direction. Figure 2 illustrates results of the interaction of two cubical clusters, each one of 647 atoms. R is the distance between the centers of mass of both clusters and increases along the $\langle 110 \rangle$ direction.

It is worth pointing out that recent experimental results for melt spun granular $\text{Cu}_{1-x}\text{Co}_x$ alloys¹ indicate that Co clusters with $10^2 - 10^3$ atoms have approximately spherical shape, diameters ranging from 2.5 to 7.5 nm, and are 50 – 150 Å apart from each other. Therefore, we can conclude from these figures that replacing the clusters by these effective moments is a good approximation if we perform simulations for similar systems of different shapes. The approximation given by Eq. (3) is very useful and allows us to study many interacting single magnetic domains.

Aiming to elucidate if it is possible to enhance the GMR effect, we studied magnetic granular systems composed by a number N , greater than 100, of single domain clusters. Here, we are concerned with the magnetic field dependence of the resistance, within a simple model, where only the relative orientation between the magnetic moments of the clusters is considered, ignoring scattering within the grains. It is clear that a microscopic theory of transport would necessarily consider the spin-dependent scattering mechanism, and then, in addition, the particle size distribution and shape anisotropy of the grains would have to be included. It is well known that it is difficult to estimate the real distribution of grain sizes. However, recent reports on the manufacturing of Co-Ag granular films prepared by low energy cluster beam deposition¹¹ show that the grain size distributions can be very narrow (≈ 1 nm) around the mean grain diameter (≈ 3 nm). Also for Co-Cu systems obtained by planar flow casting on a CuZr wheel, the diameter of the grains is estimated to vary between 2.5 and 7.5 nm.¹ Hence, the size dispersion is in general no greater than 100% around the mean grain diameter. For such grain size distribution we have made calculations¹² that reveal that, within our model, the main effect of considering a broad distribution in the grain size of a disorder system is a small broadening of the reduced magnetoresistance as a function of the external magnetic field. The effect of the particle size distribution on the magnetoresistance, without considering dipolar interactions among the grains and based upon the theory of Zhang and Levy,² has been extensively studied by Ferrari *et al.*¹³ However, they included a very broad logarithmic size distribution to notice some effect. Also there is some evidence of spin disorder in the surface of the grains.¹⁴ This effect, and also spin anisotropy, would give rise to higher saturation fields than the ones obtained within our model, and would have to be included in further calculations in order to obtain

a better quantitative fitting to experimental results. However, in this work our attention was focused on the effect of the dipolar interaction between grains on the magnetic response of granular systems, and we will show that it is possible to obtain a very reasonable fitting considering only single magnetic domain clusters. Following our model, we will replace each cluster by an effective magnetic moment $\vec{\mu}$ located at its respective center of mass. For simplicity, though it's not necessary, we consider all the clusters with the same number of atoms.

The reduced magnetization m (average magnetization per site) of the system is defined as

$$m = \frac{|\sum_j \vec{m}_j|}{N\mu}.$$

This quantity varies from $m=0$, when all the effective moments are randomly oriented, to $m=1$, when all dipole moments are perfectly aligned. The latter occurs when a strong external magnetic field is applied to the system.

The resistance of the system, as a transport property, depends on the mean free path Λ of the electrons in the material, which defines the extension of the correlation between regions in our system.¹⁵ Considering the spin dependent tunneling probability and ignoring scattering within the grains, a simple expression for the reduced magnetoresistance^{6,1} can be obtained:

$$\left(\frac{\Delta R}{R} \right)_r = 1 - \langle \cos(\theta_{ij}) \rangle_\Lambda. \quad (4)$$

Here, $\theta_{i,j}$ is the angle between $\vec{\mu}_i$ and $\vec{\mu}_j$, and the average of $\cos \theta_{i,j}$ is taken over all clusters that are within a distance Λ of each other.

The dipole-dipole correlation and its behavior for different values of Λ were investigated by Vargas *et al.*,¹⁵ considering no other sources for blocking than the dipolar interaction itself. The reduced magnetoresistance was studied as a function of the electronic mean free path Λ in 2D and 3D systems. We demonstrated that the flattening of the magnetoresistance curve at low fields arises from the short range ferromagnetic correlation among the magnetic grains.¹⁵

In our present model, we considered 2D and 3D compact arrays of magnetic clusters, each composed of approximately 1000 atoms, represented by an effective magnetic moment $\vec{\mu}_i$. The positions of the moments in the following two systems under consideration have been determined according to the following prescription. The effective magnetic moments were initially located on the sites of either a square (2D case) or cubic (3D case) lattice, with a lattice parameter equal to 70 Å, which corresponds to the average distance between ferromagnetic Co clusters embedded in a Cu matrix.¹ However, in order to simulate real systems, we have randomly displaced the positions of the effective moments with respect to the lattice distribution, as described above. The results illustrated in Figs. 3 and 4 were obtained with $\Lambda = 80$ Å, according to experimental evidence.¹

Given the position of the clusters in space, the energy of the system can be written as

$$E\{\vec{m}_1 \cdots \vec{m}_N, \vec{B}\} = \sum_{i \neq j} E_{ij} - \sum_{j=1}^N \vec{m}_j \cdot \vec{B},$$

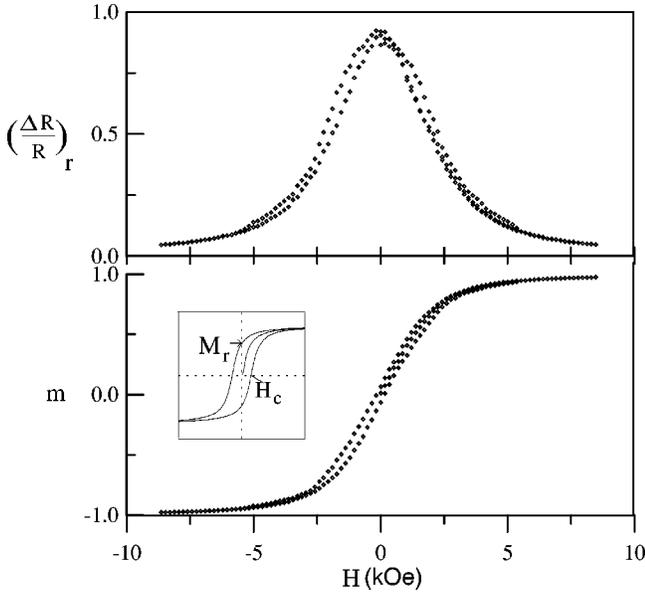


FIG. 3. Reduced magnetoresistance (top half) and reduced magnetization (bottom half) as a function of applied external field, for a 3D system of 437 single magnetic domains. The inset in the bottom half shows the definition of residual magnetization M_r and coercive field H_c .

where E_{ij} is the classical dipolar pair interaction between clusters i and j . Thus, using a standard Monte Carlo procedure¹⁶ (10^7 thermalization steps and 10^5 steps for ensemble averages), we were able to find the equilibrium configuration of the various systems, and calculate their reduced magnetoresistance and magnetization, as a function of an externally applied magnetic field H . The reduced magnetoresistance was calculated evaluating Eq. (4) for different values of the magnetic field. Our calculations have been performed for temperatures $T \sim 6$ K, where $k_B T$ is small as compared to the interaction energy of the system.

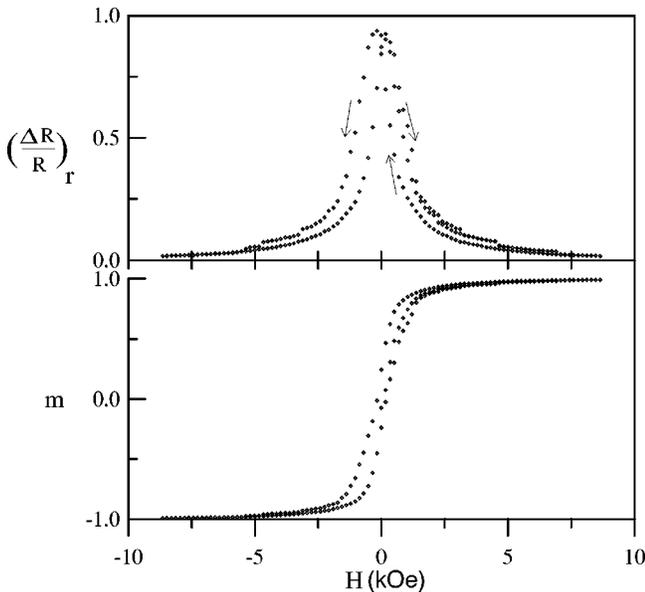


FIG. 4. Reduced magnetoresistance (top half) and reduced magnetization (bottom half) as a function of applied external field, for a 2D system of 441 single magnetic domains. The arrows indicate magnetization history.

Figure 3 shows results for a 3D system of 437 magnetic clusters when an external magnetic field is applied in the $\langle 100 \rangle$ direction. The upper panel illustrates results for the reduced magnetoresistance, and the lower one depicts the hysteresis loop. We can see a small coercive field ($H_c \sim 0.17$ KOe) and also very small residual magnetization ($M_r \sim 6\%$). The inset of the lower figure illustrates the usual definitions of the coercive field, H_c , and residual magnetization, M_r . The system described corresponds approximately to a $\text{Cu}_{97}\text{Co}_3$ alloy, with a saturated magnetization of $750\mu_B$ per cluster and then, equal to 2 emu/g, a value that is reached for an external field of approximately 7 KOe. These values are very close to the results illustrated by Xiao *et al.*⁷ in Fig. 1, for $\text{Cu}_{84}\text{Co}_{16}$ alloys, and also to the results illustrated by Allia *et al.*¹ in Fig. 2, for $\text{Cu}_{95}\text{Co}_5$ systems. This fact indicates that, in our model, the magnitude and distances among dipoles are adequate for studying real systems. According to Giteleman *et al.*,⁶ the alloy resistivity ρ is given by $\rho = \rho_0 - \kappa \langle \cos \theta_{i,j} \rangle$, where ρ_0 and κ are constants that depend on the material and temperature. For $\text{Cu}_{90}\text{Co}_{10}$ alloys at $T = 4$ K, the values commonly accepted are¹⁷ $\rho_0 = 11 \mu\Omega \text{ cm}$ and $\kappa = 3 \mu\Omega \text{ cm}$. Also, from the definition of the reduced magnetization $(\Delta R/R)_r$, we have that

$$\Delta R/R = \frac{\kappa [(\Delta R/R)_r(H) - (\Delta R/R)_r(0)]}{\rho_0 + \kappa(\Delta R/R)_r(0) - 1},$$

where $\Delta R/R$ is the usual magnetoresistance defined by $\Delta R/R = R(H) - R(H=0)/R(H=0)$ and $(\Delta R/R)_r(H)$ is the value of the reduced magnetoresistance when an external magnetic field H is applied. Then, from our results illustrated in Fig. 3 and the mentioned values for ρ_0 and κ , we can obtain a curve for $\Delta R/R$. For these results, the reduction obtained for $\Delta R/R$ is close to 22%. This reduction is bigger than the result obtained by Xiao *et al.*⁷ for $\text{Cu}_{84}\text{Co}_{16}$ samples. However, we have to keep in mind that a grain size distribution will decrease the maximum of the reduced magnetoresistance, and hence will diminish the reduction of the magnetoresistance. Also other factors, such as spin anisotropy and surface spin disorder, could contribute to this difference.

Figure 4 depicts results obtained for a 2D system of 441 clusters. Here the external magnetic field was applied in the $\langle 10 \rangle$ direction. In contrast with Fig. 3, this 2D system has a bigger magnetic susceptibility, i.e., the coercive field is as small as before ($H_c \sim 0.17$ KOe) and the residual magnetization increases ($M_r \sim 25\%$).

As in normal ferromagnets, the systems studied above show typical hysteresis loops, indicating that dipole-dipole interactions among clusters produce a *ferromagnetic short range order*, as shown by Vargas *et al.*¹⁵ As we compared the hysteresis loops for 2D and 3D systems, we could observe that for the 2D system the magnetization reaches saturation for a weaker external applied field than in the 3D case, although the number of clusters in both situations is quite similar. This effect is a consequence of the greater number of neighboring dipoles in the 3D system, averaging six nearest neighbors in the 3D case and four nearest neighbors in the 2D case. This fact produces a larger internal field acting on every cluster in the 3D system, hence the external field needed to align them has to be stronger, as compared with the 2D case.

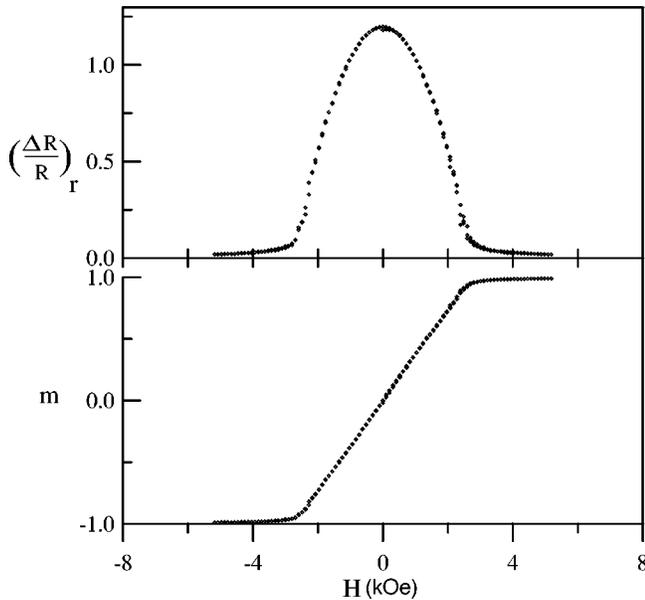


FIG. 5. Reduced magnetoresistance (top half) and reduced magnetization (bottom half) as a function of applied external field, for two parallel chains of 20 magnetic domains each. The magnetic field is applied perpendicular to the chains.

The following two figures illustrate a 2D arrangement of magnetic clusters along two parallel lines. Each chain has 20 clusters, in which the clusters are 70 Å apart, and the distance between the chains is 85 Å. For these calculations we used $\Lambda = 135$ Å. In Fig. 5, the external magnetic field was applied perpendicular to the chains. We observe no hysteresis loop, as in the case of an ideal superparamagnetic sample. As compared with the previous 2D sample, this system has a smaller susceptibility. Consequently, the reduced magnetoresistance curve is broader as compared to that of Fig. 4.

Figure 6 illustrates results for the same system described in Fig. 5, but with the external field applied parallel to the dipole chains. Here we find a hysteresis loop, while the coercive field was higher (~ 0.41 KOe) as compared with the previous results, and corresponds to the field needed to flip completely one chain of 20 dipoles. Consequently, the residual magnetization reaches a maximum value. Near the coercive field, the reduced magnetoresistance shows a dramatic effect, because the magnetization jumps from zero to the saturation value. Magnetic recording materials normally have high remanence and coercivity to prevent unwanted demagnetization, therefore magnetic materials used in recording will commonly have hysteresis loops of square shape like the one shown in Fig. 6. The coercive field can be adjusted by varying the number of single magnetic domains in the chains.

Whether the external field is applied parallel or perpendicular to the chains, a completely different behavior of the magnetization is obtained for these dipole chains. When the external magnetic field is applied perpendicular to the chains, a smooth linear behavior of the magnetization is obtained, up to the saturation value. However, when the external field is applied parallel to the chains, nothing happens until the external field reaches a value (coercive field) that allows us to invert the magnetization of one complete chain. Near this

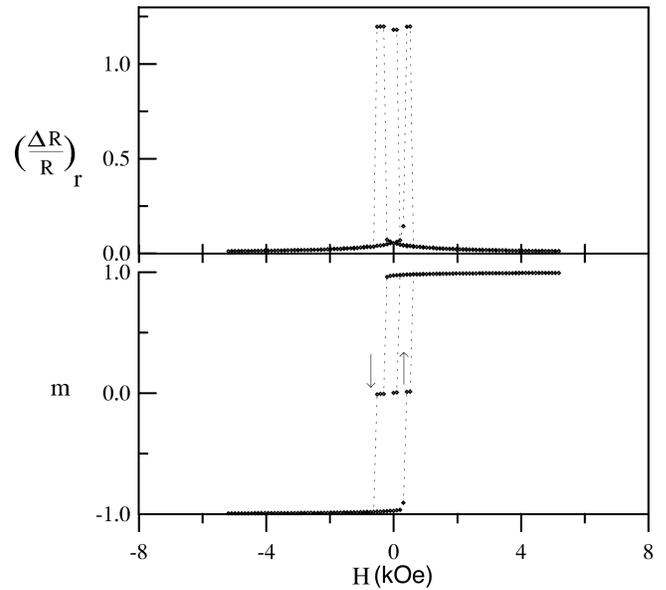


FIG. 6. Reduced magnetoresistance (top half) and reduced magnetization (bottom half) as a function of applied external field, for two parallel chains of 20 magnetic domains each. The magnetic field is applied parallel to the chains. Arrows indicate magnetization history.

coercive field, the magnetoresistance shows a dramatic effect because the sample magnetization jumps from zero (maximum reduced magnetoresistance) to its saturation value (minimum reduced magnetoresistance). These particular two-dimensional distributions of dipoles may be useful for technological applications, because of the enhancement obtained for the reduced magnetoresistance, particularly for the case illustrated in Fig. 6. With techniques such as x-ray photolithography, it will be possible, very soon, to pattern on a metallic substrate any particular array of single domain magnetic clusters of nanometer sizes. This will allow experimentalists to test designs similar to the one presented here.

IV. CONCLUSIONS

By means of a simple model, we have investigated the behavior of magnetic granular systems when an external magnetic field is applied. We have demonstrated that, for systems comparable to those that are experimentally studied, it is valid to replace a cluster by an effective magnetic moment located at its center of mass. Also, we have obtained magnetization and reduced magnetoresistance curves in close agreement with experimental results, strongly suggesting that the magnetic dipolar interaction is the dominant interaction in granular systems like the ones used in our analysis. We also suggest an experimental design by which the best GMR ratios could be obtained. Such experiments are expected to be useful for technological applications.

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