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Magnetoresistance in granular metallic systems

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Abstract. The magnetic and transport properties of ferromagnetic clusters embedded in a metallic host are studied. A simple model is presented to explain the observed behaviour of the magnetoresistance in such systems. The role played by both the dipolar interaction between the clusters and the ratio of the electronic mean free path in the non-magnetic material to the average inter-cluster distance is clarified. Finally, a suggestion is made of systems in which the best magnetoresistance ratios are expected to be found.

In the past few years, great interest has been focused on the study of granular metallic systems consisting of single-domain ferromagnetic clusters embedded in a non-magnetic metallic matrix. The discovery of the giant magnetoresistance (GMR) effect in these materials [1, 2] has raised the possibility of their use in the construction of reading heads, sensors, and other magnetic devices. Among those systems we have Cu–Co [1–3], Cu–Fe [2], Co–Ag [4, 5], and Fe–Co–Ag [6] alloys. From the point of view of practical applications, granular systems are very convenient. They are relatively easy to produce, thermally stable, and exhibit magnetoresistance amplitudes that may be comparable to or even larger than those of multilayers in the usual CIP geometry [7, 8]. All of these facts have motivated the search for the granular materials with the best GMR ratios.

In the present paper, we investigate the magnetoresistance of granular materials. We show that the magnitude of the effect is strongly dependent on two factors, namely, the presence of short-range ferromagnetic correlation between this and the clusters, and the relation between this and electronic mean free path. We use a simple model for the resistivity [9], which considers only the relative orientation of the magnetic moments of the clusters, ignoring scattering within the grains, all of which are of the same size.

We have found that, within this simple model, the magnitude of the effect is essentially controlled by two factors, namely, the presence of a short-range ferromagnetic correlation between the magnetic moments of the clusters, and the relation between the electronic mean free path in the non-magnetic material and the average inter-cluster distance. Taking these points into account, we have succeeded in explaining the observed behaviour of the GMR in granular metals, as well as in predicting the conditions under which the effect is expected to be maximized.

The GMR effect is thought to be related to the spin-dependent scattering of conduction electrons and the reorientation of the magnetic moments by an external magnetic field $\boldsymbol{H}$ [9, 10]. For $H = 0$, the total magnetic moments $\boldsymbol{\mu}_i$ of the clusters are oriented in such a way that the net magnetization of the system is zero and the resistance in both the up-spin

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and down-spin channels is high. However, the alignment of the moments by the magnetic field leads to a short-circuit effect in one of the spin channels, with a significant reduction in the total resistance R of the system. The field dependence of the alloy resistivity ρ is well described by a simple model proposed by Gittleman *et al* [9], according to which

$$\rho = \rho_0 - \kappa \langle \cos\theta_{ij} \rangle_\Lambda \tag{1}$$

where ρ_0 and κ are constants, and θ_{ij} is the angle between the magnetic moments $\boldsymbol{\mu}_i$ and $\boldsymbol{\mu}_j$ of clusters i and j. The average of $\cos\theta_{ij}$ in equation (1) is taken over pairs of clusters separated by distances not much larger than the electronic mean free path Λ in the non-magnetic material [11, 12]. The constant κ depends on spin-dependent scattering processes both inside and at the surface of the grains. Here, for simplicity, we have assumed that all of the total magnetic moments $\boldsymbol{\mu}_i$ of the clusters have the same magnitude μ. Given H, it follows from equation (1) and the usual definition of the magnetoresistance $\Delta R/R$ that

$$\Delta R/R = \kappa \frac{\xi_\Lambda(H) - \xi_\Lambda(0)}{\rho_0 - \kappa\xi_\Lambda(0)} \tag{2}$$

where $\xi_\Lambda(H) = \langle \cos\theta_{ij} \rangle_\Lambda$ is a field-dependent dipole–dipole correlation function. This quantity plays a central role in our present discussion of the GMR effect.

Clearly, ξ_Λ depends on the interaction between the clusters. In the simplest situation in which the clusters can be regarded as non-interacting, one can easily show [2, 11] that $\xi_\Lambda = m^2$, where $m = \left|\sum_i \boldsymbol{\mu}_i\right|/(N\mu)$ is the reduced magnetization and N is the number of clusters in the system. In such a case, it follows from equation (2) that $\Delta R/R$ is a quadratic function of m. Deviations from such a parabolic relation have been observed in several systems [11, 13], and have been interpreted as evidence of the interaction between the magnetic clusters [11, 13, 14]. However, as we demonstrate in the present communication, the magnitude of the deviation is also strongly dependent on the ratio of Λ to the average inter-cluster distance.

We remark that, even in the presence of interaction between the clusters, for values of H close to the saturation field H_S, one still finds that $\xi_\Lambda(H) \simeq m^2$. Hence, the magnetoresistance in that regime behaves as $\Delta R/R \simeq a - bm^2$, where a and b are appropriate constants. In view of this, it is convenient to introduce the so-called reduced magnetoresistance [11], defined as

$$\left(\frac{\Delta R}{R}\right)_{red} = \frac{(\Delta R/R) - a + b}{b}$$

which reduces immediately to $(\Delta R/R)_{red} = 1 - \xi_\Lambda(H)$. For real systems, curves for $(\Delta R/R)_{red}$ versus m correspond usually to flattened parabolas [11], instead of the simple $1 - m^2$ curve characteristic of non-interacting clusters.

Recently, we have investigated the coupling between magnetic clusters in granular materials [15]. We have shown that for clusters whose sizes and spatial distributions are consistent with the experimental data, the coupling between them is entirely dominated by the classical dipolar interaction. In addition, we have demonstrated that the total energy of the system is well described by replacing each cluster by an effective magnetic moment located at its 'centre of mass'.

Here, we are concerned with the magnetic field dependence of the resistance of granular materials. Our approach is based on the theory discussed above and Monte Carlo simulations, which enable us to determine ξ_Λ as a function of H and Λ. We obtain curves for $(\Delta R/R)_{red}$ versus m for different systems, and find that the observed flattening of these curves can be understood in terms of short-range ferromagnetic correlations and the ratio of Λ to the average inter-cluster distance. Finally, we discuss experimental situations in which the best GMR ratios can be found.

We consider systems with N single-domain magnetic clusters that are represented by effective magnetic dipoles $\boldsymbol{\mu}_i$ [15], and typically with 10^3 atoms each. We first show that in the limiting case in which Λ is very large ($\Lambda = \infty$), i.e. when the average of $\cos(\theta_{ij})$ is taken over all dipole pairs, a simple and general relation between $(\Delta R/R)_{red}$ and m is obtained. In fact, from the definition of the reduced magnetization we find that

$$Nm^2 = \frac{1}{N\mu^2}\sum_i \mu_i^2 + \frac{2}{N\mu^2}\sum_{i>j}\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j.$$

The first term on the r.h.s. of this equation is equal to 1, whereas the second is just $(N-1)\langle\cos\theta_{ij}\rangle_\infty$. Thus, we have $\xi_\infty = (Nm^2-1)/(N-1)$, and consequently

$$\left(\frac{\Delta R}{R}\right)_{red} = \frac{N}{N-1}(1-m^2). \tag{3}$$

We emphasize that this quadratic relationship does not depend on either the positions of the clusters or the interaction between them. It holds in general, provided that Λ is sufficiently large. In addition, for $N \gg 1$, it reduces to the result for non-interacting clusters. Therefore, it is clear that the observed deviations from the simple parabolic behaviour must be related to the actual magnitude of Λ and the dependence of $\langle\cos\theta_{ij}\rangle$ on that parameter. In order to make this point clearer, we investigate in detail some specific cases.

We consider compact arrays of magnetic clusters, represented by magnetic moments $\boldsymbol{\mu}_i$, both in two (2D) and three (3D) dimensions. The positions of the moments in all of the cases under consideration in this work have been determined according to the following prescription. The magnetic moments were initially located at the sites of either a square (2D case) or cubic (3D case) lattice, with the lattice parameter equal to 70 Å, which corresponds to the average distance between ferromagnetic Co clusters in Co_xCu_{1-x} systems ($x \sim 10$ at.%) [11]. However, because the spatial arrangements of the clusters in ordinary granular materials are disordered, we have randomly displaced the positions of the moments with respect to the lattice sites according to a gaussian probability distribution, with zero average and 10 Å standard deviation, in each space direction.

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

$$E\{\boldsymbol{\mu}_1\cdots\boldsymbol{\mu}_N, \boldsymbol{H}\} = \frac{1}{2}\sum_{i\neq j}E_{ij} - \sum_{j=1}^{N}\boldsymbol{\mu}_j \cdot \boldsymbol{H}$$

where E_{ij} is the classical dipolar pair interaction between clusters $\boldsymbol{\mu}_i$ and $\boldsymbol{\mu}_j$. Thus, using the standard Monte Carlo procedure [16] (10^6 thermalization steps and 10^3 steps for ensemble averages), we are able to find the equilibrium configurations of the various systems, and calculate their reduced magnetization m and the dipole–dipole correlation ξ_Λ for different values of Λ and H. Our calculations have been performed for temperatures T such that k_BT is small as compared to the interaction energy of the system.

Let us first discuss the 2D case. Figure 1 shows a snapshot of a system with 441 magnetic clusters in the absence of externally applied magnetic fields. It represents one of the possible microscopic states of the system, which has been reached after the thermalization procedure. The reduced magnetization m of this configuration is found to be negligible. Thus, if Λ were comparable to the linear dimensions of the system, $\xi_{\Lambda=\infty}(0)$ would also be negligible and $(\Delta R/R)_{red}$ very close to 1. However, the presence of disorder in the atomic arrangement in the non-magnetic metal, as well as temperature effects, should lead to much smaller values of Λ. The actual value of Λ depends on both the growth conditions and any heat

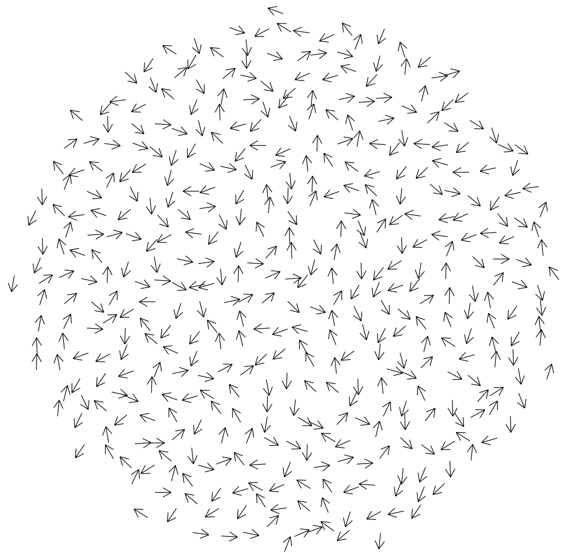

Figure 1. A snapshot of one of the equilibrium configurations of a 2D system with 441 dipoles (see the text).

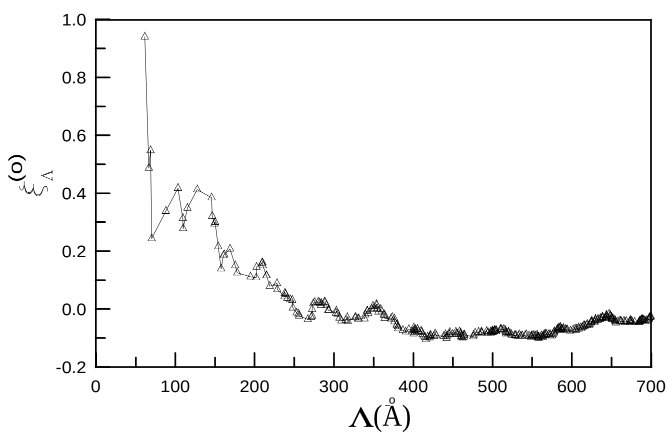

Figure 2. The correlation around the central dipole shown in figure 1.

treatment that the sample has been subjected to. If we assume that Λ is of the order of the average inter-cluster distance (in our case, 70 Å), we obtain for the configuration in figure 1 that $\xi_\Lambda(0) = 0.32$ and $(\Delta R/R)_{red} = 0.68$. This significant reduction in $(\Delta R/R)_{red}$ is in line with what has been found experimentally, and can be understood from a simple inspection of the orientation of magnetic moments in figure 1. It is clear from this figure that the dipolar interaction favours the formation of vortex structures [14], which leads to the appearance of a short-ranged ferromagnetic correlation between the moments. Thus, for small values of Λ, $\xi_\Lambda(0)$ turns out to be positive, which yields a smaller magnetoresistance effect. We can make this point more precise by introducing the quantity

$$\xi_\Lambda^{(i)} = \frac{1}{N_\Lambda^{(i)}} \sum_{j(\neq i)} \cos(\theta_{ij})\Theta(\Lambda - R_{ij})$$

where $N_\Lambda^{(i)}$ is the number of clusters whose distances R_{ij} to cluster i are smaller than or equal to Λ, and $\Theta(x)$ is the Heaviside step function. We present in figure 2 the curve for $\xi_\Lambda^{(i)}$ as a function of Λ for the central cluster in figure 1. It clearly shows a decreasing ferromagnetic correlation around the central cluster, extending up to a distance of 200 Å. Similar results are obtained for the other clusters in the system.

Having the above results in mind, we proceed to investigate the dependence on H of both $(\Delta R/R)_{red}$ and m, considering different values of Λ. For each field intensity, Monte Carlo calculations have been performed to determine the equilibrium configuration of the system. Figure 3 shows results for $(\Delta R/R)_{red}$ plotted against m for the 2D array of 441 clusters. For values of Λ close to the average inter-cluster distance, we find that the curves exhibit a pronounced flattening in the small-m region, in agreement with the existing experimental data [11]. However, as Λ increases the curves become less flattened and rapidly approach the $1 - m^2$ parabola, almost reaching this limiting behaviour when Λ is about twice the average inter-cluster separation. We have tested the accuracy of our results by repeating the calculations for systems with different numbers N of clusters, and found no significant deviations from the results presented here, provided that $N > 100$.

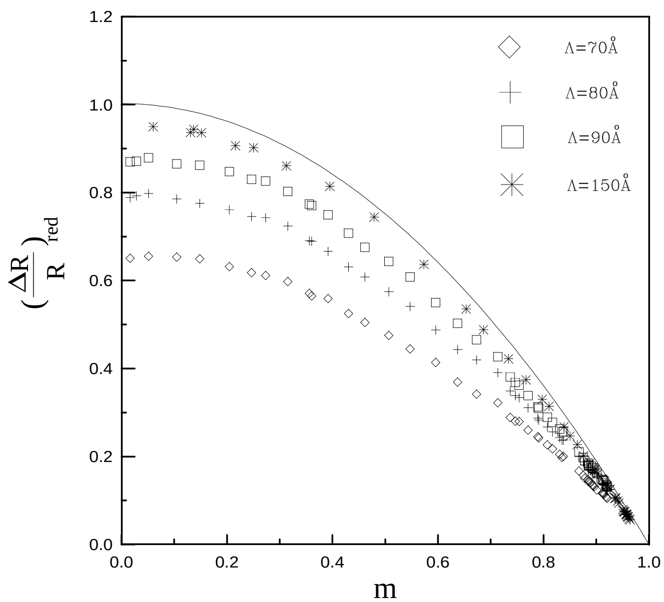

Figure 3. The reduced magnetoresistance as a function of the reduced magnetization of a 2D system with 441 dipoles, for different values of Λ. The full line corresponds to the behaviour predicted by equation (3) ($\Lambda = \infty$).

Similar behaviour of the reduced magnetoresistance as a function of the reduced magnetization has been found in the case of 3D arrays of dipoles. In figure 4 we present results for a system of 437 dipoles and different values of Λ. Also in this case, the curves corresponding to Λ close to the average inter-cluster distance can be described as flattened parabolas. They, nevertheless, nearly coincide with the $1 - m^2$ parabola when Λ is about twice the average inter-cluster separation. It is important to mention that this particular behaviour of the GMR effect with changes in Λ, predicted by our present calculations, is in agreement with what has been observed experimentally by Allia *et al* [11]. In fact, Allia *et al* have found a clear reduction in the flattening of the curves for $(\Delta R/R)_{red}$ versus

m for Co_xCu_{1-x} systems after the samples had been subjected to heat treatment. Such a process allows the rearrangement of the atoms in the system, with a consequent increase in the electronic mean free path in the non-magnetic material.

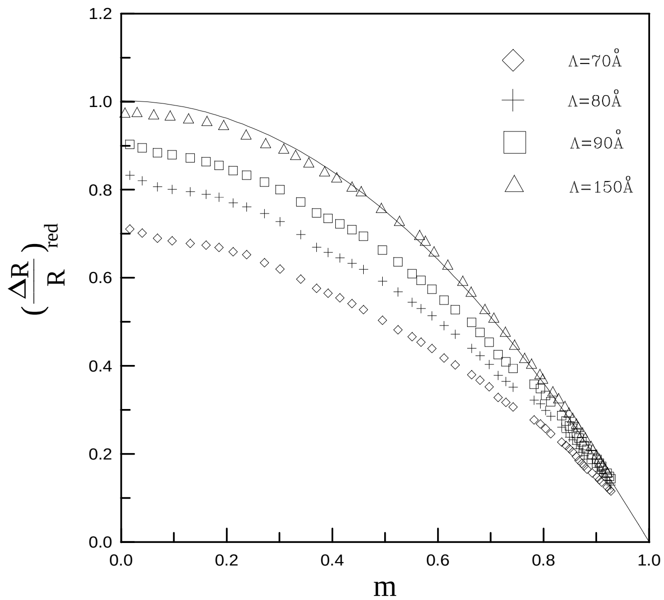

Figure 4. The reduced magnetoresistance as a function of the reduced magnetization of a 3D system with 437 dipoles, for different values of Λ. The full line corresponds to the behaviour predicted by equation (3) ($\Lambda = \infty$).

We emphasize that, according to our model, the flattening of the curves for $(\Delta R/R)_{red}$ versus m results from the combination of two factors, namely, the appearance of short-ranged ferromagnetic correlation between the moments induced by the dipolar interaction, and the magnitude of Λ relative to the average inter-cluster distance. These points can be used as guidelines for the design of systems with the best GMR ratios. The simplest case would be that of a 2D arrangement of magnetic clusters along parallel lines, such that the distance D between the lines is greater than the separation d between the clusters inside the lines, but smaller than Λ. To clarify our ideas, we have considered two chains of ten clusters each, in which the clusters are 70 Å apart, and such that the distance between the chains is 85 Å. As a consequence of the dipolar interaction, the orientation of the magnetic moments inside the lines is ferromagnetic, whereas the two lines have their magnetizations oriented antiferromagnetically. If we now take $\Lambda = 115$ Å, we find that $\xi_\Lambda(0) = -0.2$, which leads to an enhancement in the reduced magnetoresistance of about 20% with respect to the highest value so far observed for granular materials. Figure 5 shows curves for $(\Delta R/R)_{red}$ as a function of m for such a system, corresponding to different values of Λ. As we can see, the highest values of $(\Delta R/R)_{red}$ are achieved for a particular combination of geometrical factors D and d, and Λ. Finally, we point out that even higher values of the reduced magnetoresistance can be achieved by considering sequences of lines of clusters rather than a single pair of lines. Moreover, if we polarize the sample perpendicularly to the chains, it is mainly the correlation length of the electrons in the direction of the applied electric field that matters. In this particular case, we can easily verify that an enhancement of up to 100% in $(\Delta R/R)_{red}$ can be obtained, for appropriate values of D, d, and Λ.

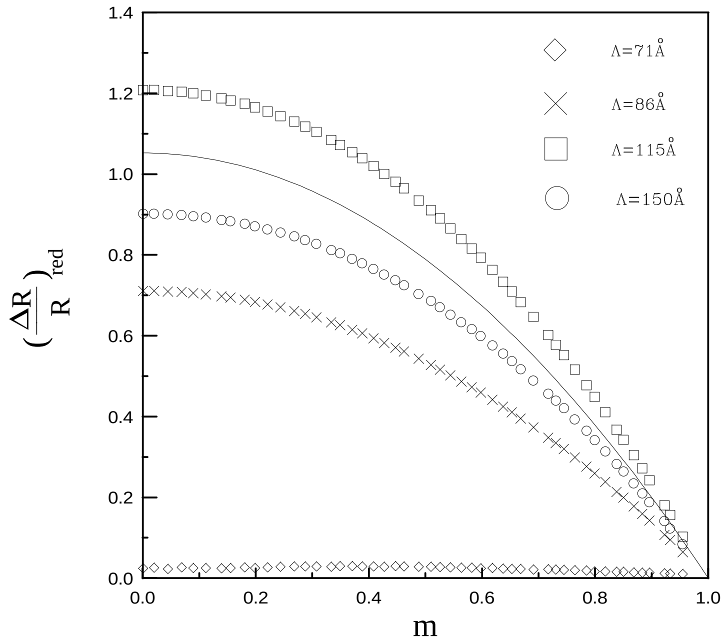

Figure 5. The reduced magnetoresistance versus the reduced magnetization of a 2D system consisting of two lines of ten dipoles each, for different values of Λ. The full line corresponds to the behaviour predicted by equation (3) ($\Lambda = \infty$).

At this point, we would like to clarify that a cluster of 1000 Co atoms in a perfect fcc structure (with a lattice constant of 3.6 Å) has a radius of 14.2 Å. Then, distances between grains of the order of 70 Å and Λ values of the order of 70 Å or greater are perfectly compatible within our model. We have also made simulations without the restriction that all grains have the same size. Within our model, size effects also contribute to the flattening of the magnetoresistance curve.

The role of the magnetic moment distribution as regards the magnetoresistance, without considering dipolar interactions among the grains, and based on the theory of Zhang and Levy [17] and the measurements of Rabedau *et al* [18], has recently been studied by Ferrari *et al* [19].

It is clear that a microscopic theory of transport would necessarily consider the spin-dependent scattering mechanisms, and then the particle size and shape anisotropy of the grains have to be considered. However, in this work our attention was centred on the dipolar interaction between grains and how this affects the correlation between magnetic moments, and, consequently, the scattering probability for an electron moving from one magnetic grain to another. Thus, the effect of the dipolar interaction and electronic mean free path on the magnetoresistance of the system has been investigated.

In conclusion, we have presented a simple model to explain the observed behaviour of the magnetoresistance of granular materials as a function of the reduced magnetization of the system. We have clarified the role played by both the dipolar interaction between the clusters and the magnitude of the electronic mean free path in the non-magnetic material in determining the magnitude of the GMR effect. In addition, on the basis of the results obtained here, we have proposed systems in which the best GMR ratios are expected to be found.

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