Hopping percolation transition in granular ferromagnets

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We present computer simulations of the magnetotransport properties of two-dimensional and three-dimensional granular ferromagnets using a random resistor network in which the conductivity, σ, between pairs of neighboring grains depends upon the intergrain distance [$\sigma \propto \exp(-\kappa r)$, where κ is a measure of disorder and r is a random number, $0 \leqslant r \leqslant 1$] and the relative magnetic orientations. We study the resistance (R) distribution function, $P(R)$, and find that in both the *weak disorder* regime $L/\kappa^{\nu} > 1$ (not sensitive to the removal of any single bond) and the *strong disorder* regime $L/\kappa^{\nu} < 1$ (very sensitive to such a removal) the distribution depends only on L/κ^{ν} and can be well approximated by a log-normal function with dispersion $b\kappa^{\nu}/L$, where b is a coefficient which depends on the type of the lattice and ν is the critical exponent of the percolation correlation length. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2176912]

Granular metals consist of composite mixtures of metal and nonmetal materials. When the metal concentration is small the metal forms small islands embedded in an insulating matrix. In this configuration the electric conductivity is governed by hopping between different grains. The electronic properties of such systems have been studied for many years;[1] nevertheless many issues related to the conduction mechanism are still poorly understood. The case in which the metallic grains are ferromagnetic is of special interest since these systems have been found to exhibit a giant magnetoresistance (GMR) effect due to spin dependent tunneling between grains.[2] A peculiar feature which is found in dilute discontinuous two-dimensional (2D) Ni films is the occurrence of sharp resistance jumps as a function of the applied magnetic field.[3] These were interpreted as the result of magnetomechanical distortions at a bottleneck grain which dominates the transport in systems which are on the percolation edge. This model is based on the notion that the electric conductivity is governed by the resistance of a local configuration of few grains, although there are 10^9 grains in the system. Recently we tested this hypothesis by modeling the granular system by a network of random resistors and studying their transport properties using numerical simulations.[4] We found a good agreement with the experimental results. In this paper we further develop the model and compare it to a three-dimensional (3D) sample.

Granular samples for which measurements have been performed[3] were prepared by quench condensation, i.e., sequential evaporation on a cryogenically cold nonpassivated substrate such as SiO_2 under UHV conditions while monitoring the film's thickness and resistance. The measurements which are presented in Fig. 1(a) were performed by evaporating Ni on a 2×2 mm^2 square between two large Ag pads so a two-probe electric contact configuration is obtained. The growth process was terminated at the point at which an electric conductivity could first be detected across the sample. At this stage, the resistance was larger than 100 MΩ and a typical Ni grain radius is about 100 Å (height ~20 Å). Thus, the sample contains about 10^{10} grains.

The hopping resistance R_{ij} between two neighboring ferromagnetic grains (labeled by i and j) depends on two main factors: the grain-to-grain distance [expressed in this work as $\kappa r(ij)$, where κ is as the degree of the spatial disorder and $r(ij)$ is a random number taken from uniform distribution in the range (0,1)] and the mutual orientation of the magnetic moments, **M**, of these grains (described by orientation angles θ and ϕ of the magnetic moments),

$$R_{ij} = R_0 \left[1 - \left(\frac{\delta R}{2R_0} \right)^2 (1 + \cos \Theta_{ij})^2 \right] e^{\kappa r(ij)}, \tag{1}$$

where $R_0 = (R_{\uparrow\uparrow} + R_{\uparrow\downarrow})/2$, $\delta R = (R_{\uparrow\uparrow} - R_{\uparrow\downarrow})/2$, and $\cos \Theta_{ij} = \cos \theta_i \cos \theta_j + \sin \theta_i \sin \theta_j \cos(\phi_i - \phi_j)$. It is taken into account that the electron spin direction may be parallel or antiparallel to the direction of the magnetic moment of the initial grain and the moment of the final grain. If parallel, the electron experiences weak scattering and hence a low resistance $R_{\uparrow\uparrow}$; if antiparallel, the electron experiences strong scattering and hence a high resistance $R_{\uparrow\downarrow}$ (see Refs. 4–7). The magnetic moment $\mathbf{M}_i$ of the ferromagnetic grain is assumed to be always parallel or antiparallel to its randomly distributed easy axis. Once a magnetic field **H** is turned on, it can switch the direction of the magnetic moment if the strength of the magnetic field is such that the average magnetization of the sample is larger than the grain magnetic moment M_i. Thus the grain magnetic moment will flip once $\mathbf{M}_i \cdot \mathbf{H} < \mathcal{L}(h) \equiv \coth(h) - 1/h$, where $\mathcal{L}(h)$ is Langevin function, $h \equiv M_i H / k_B T$, and T is the temperature.[4]

We model the granular Ni film studied in Ref. 3 by a Miller-Abrahams[7] square 2D bond-percolating resistor network.[4] We insert a resistor with random tunneling resistance R_{ij} [see Eq. (1)] between neighboring sites, then we solve the obtained system of linear Kirchhoff equations and calculate the total effective resistance, R_e, of the 2D and 3D

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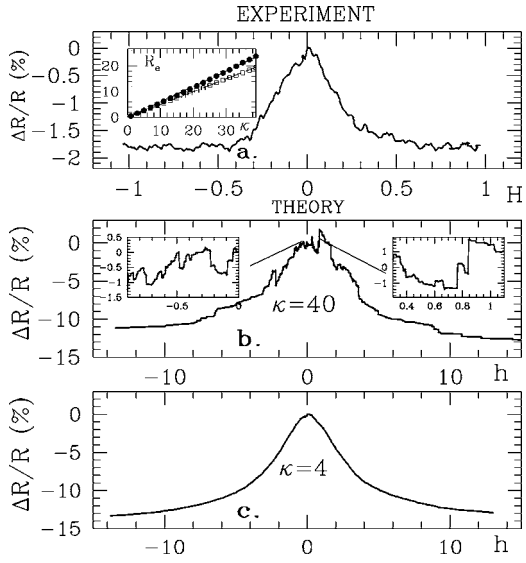

FIG. 1. (a) Experimental data of the relative magnetoresistance $\Delta R(H)/R(0)$ vs magnetic field H (in Tesla) of a dilute granular Ni sample for $T=4$ K (see Ref. 3). The values of κ and system size, L, realized in these experiments (Ref. 3) can be estimated as $\kappa \sim 10^2-10^3$ and $L \sim 10^5$. [(b) and (c)] Theoretical drawings of the similar quantity $\Delta R(h)/R(0)$ vs $h \equiv MH/k_BT$ obtained from numerical simulations on a random bond-percolating resistor network of the size $L=100$ with $\kappa=40$ [see (b)] and $\kappa=4$ [see (c)]. $R_{\uparrow\downarrow}/R_{\uparrow\downarrow}=4$. The inset in (a) is a semilog plot of the averaged resistance R_e vs κ. In the case of site percolation ($p_c=0.593$) the slope of the curve (shown by solid hexagons) is close to 0.6, while for the case of bond percolation ($p_c=0.5$) the slope of the curve (shown by empty squares) is equal to 0.5 [cf. with Eq. (3)]. The insets in (b) are the zoomed fragments of the megnetoresitance curve.

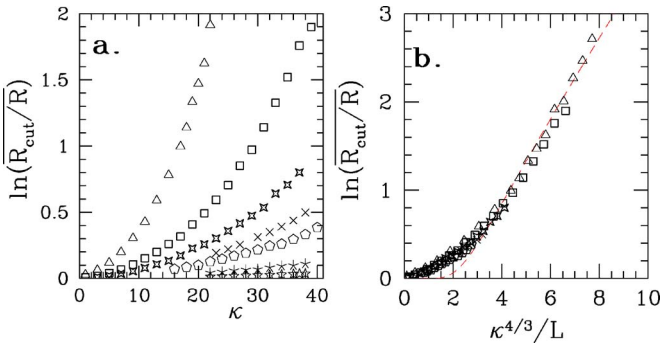

FIG. 2. (a) A semilog plot of the mean value of the ratio $R_{\rm cut}/R$ vs κ for various sizes of the system: $L=10,20,30,40,50,100,200$ (from top to bottom). (b) A semilog scaling plot of the same quantity vs κ^{ν}/L. The dashed line shows the analytical fit found using Eq. (4): $\overline{\ln R_{\rm cut}/R}=\int_{R^{(e)}_{\rm cut\ min}}^{R^{(e)}_{\rm cut\ max}} \ln(R'/R)P(R')dR'=(\mu/\sqrt{4\pi})\exp(-4.5^2/2\mu^2)$, since $R^{(e)}_{\rm cut\ max}\to\infty$ and the value $\ln[R^{(e)}_{\rm cut\ min}/R_e]=4.5$ is found empirically.

networks as well as the values of the local current on each resistor.[4,8,9]

In order to check our numerical scheme we present in the inset in Fig. 1(a) the dependence of the macroscopic effective resistance R_e vs κ and compare them with analytical formulas obtained by us using the symmetric self-consistency effective-medium approximation[10] (EMA) (where in the case of EMA $\langle R\rangle \equiv \langle R_{ij}\rangle$ and $\langle\cdots\rangle$ denotes the effective-medium averaging over the sample, while in the case of numerical simulations $\langle R\rangle \equiv \Sigma_{n=1}^{N} R_n/N$ means the statistically averaged value of R). If the local conductivities of R are distributed continuously in a range $R_{\min} \leqslant R \leqslant R_{\max}$ according to some distribution function $f(R)$, then the macroscopic effective R_e can be found by taking the integral

$$\int_{R_{\min}}^{R_{\max}} f(R)\left(\frac{R-R_e}{aR+R_e}\right)dR=0, \tag{2}$$

where $a=z/2-1$ and z is the number of bonds at each node of the network. If $r(ij)$ in Eq. (1) is uniformly distributed between 0 and 1, then $f(R)=1/\kappa R$ and R is varied in the range $R_0 \leqslant R \leqslant R_0 e^{\kappa}$. From the integral (2) we get $\ln[(aR+R_e)^{(1+a)/a}/R]_{R_0}^{R_0 e^{\kappa}}=0$, and, therefore,

$$R_e=R_0 e^{\kappa p_c}\left(\frac{1-p_c}{p_c}\right)\left[\frac{1-e^{-\kappa p_c}}{1-e^{-\kappa(1-p_c)}}\right], \tag{3}$$

where $p_c=1/(1+a)=2/z$. In the case of 2D square lattice ($z=4$, see Ref. 8), $R_e=R_0 e^{p_c\kappa}$, which could be found as an exact result directly from the Keller-Dykhne theorem[11] or as an approximate expression[12,13] [see also inset to Fig. 1(a)].

In Fig. 1(a) we show the experimental data of the relative magnetoresistance $\Delta R(H)/R(0) \equiv [R(H)-R(0)]/R(0)$ [where $R(H)$ is the total sample resistance at magnetic field H] obtained for a dilute Ni granular 2D sample versus the applied magnetic field (see Ref. 3) and in Figs. 1(b) and 1(c) we plot $\Delta R(h)/R(0) \equiv [R(h)-R(0)]/R(0)$ obtained from our numerical simulations versus h, which is proportional to the applied magnetic field. In the experimental data the pronounced noise, as the magnetic field is swept back and forth, can be seen clearly. Similar behavior is observed in the simulation curves when the disorder (κ) is large enough. For a higher value of κ (which corresponds to more dilute samples) stronger jumps in the magnetoresistance curves are observed [see insets in Fig. 1(b)]. Our results suggest that the jumps observed in both experimental and theoretical curves are a result of magnetic moment flip (due to applied magnetic field) at a bottleneck grain, leading to switches between different current trajectories.

In order to test this hypothesis, we perform the following numerical simulations: We remove from the network the resistor on which the local current is maximal. In this way we hope to determine the conditions for a single bond to dominate the conductivity of the system. Such a removal of a dominating single bond could change the trajectory of the current along the spanning cluster which should affect the system transport properties, e.g., the Ohmic effective 2D resistivity. Therefore, the ratio $R_{\rm cut}/R$ (where R denotes the resistivity of the system prior to the removal of the resistor and $R_{\rm cut}$ is the resistivity after removing it) is an efficient characteristic of disorder. We expect that the ratio will be stronger for a larger disorder. In Fig. 2(a) we show a *semilog* plot of the statistically averaged value of the ratio $R_{\rm cut}/R$ vs κ for various sizes of systems $L=10-200$. In Fig. 2(b) we present the scaling behavior of this ratio. It is found that $\overline{R_{\rm cut}/R}$ scales well as a function of κ^{ν}/L, where ν is the critical exponent of the percolation correlation length $\xi \propto (p-p_c)^{-\nu}$ [in 2D $\nu=4/3\simeq 1.33$, while in 3D $\nu\simeq 0.88$ (Ref. 12)].

We study also the probability distribution function $P(R)$ (i.e., the probability that the total resistance of the system is

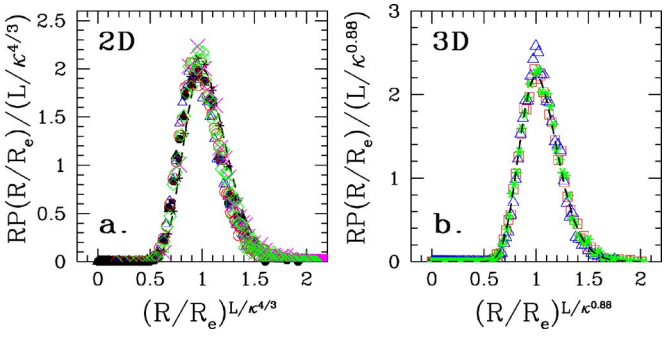

FIG. 3. (a) A scaling plot of $RP(R/R_e)$ vs $(R/R_e)^{L/\kappa^{\nu}}$. The dashed line represents the analytical result (4), with $b=0.2$ and $\nu=4/3$ for nine systems with $L=500$, $\kappa=22$ (▲); $L=300$, $\kappa=15$ (■); $L=200$, $\kappa=10$ (●); $L=100$, $\kappa=10$ (∗); $L=50$, $\kappa=30$ (△); $L=40$, $\kappa=40$ (□); $L=30$, $\kappa=20$ (○); $L=20$, $\kappa=40$ (×); and $L=20$, $\kappa=30$ (◇); (b) similar to (a), but for 3D, with $b=0.18$ and $\nu=0.88$ for three systems with $L=20$, $\kappa=15$ (□); $L=26$, $\kappa=15$ (△); and $L=10$, $\kappa=6.8$ (∗). ($h=1.85$). The dashed line is the analytical result (4).

R) for systems of different sizes L and different disorders κ. In Fig. 3(a) we present numerical results suggesting that $P(R)$ can be approximated by the log-normal form

$$P(R) \simeq (\sqrt{2\pi}\mu R)^{-1} \exp[-\ln^2(R/R_e)/(2\mu^2)], \tag{4}$$

where $\mu=\alpha(p_c\kappa)^{\nu}/L$ for both 2D and 3D cases (in 2D is found that $\alpha \simeq 0.5$).

In summary, the recently observed features of the electrical transport in dilute granular Ni films,[3] which are believed to be governed by a very small number of grains, are explained using Monte Carlo resistor network simulations. The dependence of the simulated magnetoresistance versus the applied magnetic field is similar to the experimental measurements and indicates that few resistors or even a single one can govern the total conductivity. This is not expected from a pure percolation picture where the number of red bonds on which the current is maximal scales as $L^{1/\nu}$ (see Ref. 14), i.e., of the order of a few hundreds in the macroscopic system considered here. On the other hand, the strong disorder limit of our model yields a single bond that dominates the conductivity. The obtained experimental and theoretical results can be used in the development of magnetoresistive devices based on single-grain effects.

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