PHYSICAL REVIEW E **69**, 065105(R) (2004)

Percolation transition in a two-dimensional system of Ni granular ferromagnets

Yakov M. Strelniker, Richard Berkovits, Aviad Frydman, and Shlomo Havlin
Minerva Center, Jack and Pearl Resnick Institute of Advanced Technology, and Department of Physics, Bar-Ilan University, 52900 Ramat-Gan, Israel

(Received 9 February 2004; published 24 June 2004)

We model magnetotransport features of the quenched condensed granular Ni thin films by a random two-dimensional resistor network in order to test the condition where a single bond dominates the system. The hopping conductivity is assumed to depend on the distance between neighboring ferromagnetic grains and the mutual orientation of the magnetic moments of these grains. We find that the quantity characterizing the transition from *weak disorder* (not sensitive to a change of a single bond resistivity) to *strong disorder* (very sensitive to such changes) scales as $\kappa/L^{1/1.3}$, where L is the size of the system and κ is a measure of disorder.

DOI: 10.1103/PhysRevE.69.065105 PACS number(s): 64.60.-i, 05.50.+q

Transport in granular metals, i.e., systems of metallic islands embedded in an insulating matrix, has been an active area of research for a number of decades but many experimental findings are still not fully understood [1–4]. Recent magnetoresistance measurements performed on quench condensed granular Ni thin films which are on the verge of electric continuity seem to indicate that in these systems the electric conductivity is governed by the resistance of a local configuration of few grains although there are 10^9 grains in the system [5]. Sharp resistance jumps as a function of the applied magnetic field observed in this experiment were interpreted as the result of magnetomechanical distortions at a bottleneck grain. In order to verify this hypothesis, we model the granular system by network of random resistors and study transport properties using numerical simulations. Our main goal is to try and develop a comprehensive picture of this phenomena. The main points which we wish to clarify are how and under which conditions an order of a single grain can determine the transport properties of the granular mesoscopic system. In the picture of critical percolation, each red bond should affect the conductivity, but their number (for a system of 10^9 grain) is about 10^2 and not a few grains as observed.

An established method to fabricate systems of granular metals in general [6–10] and particularly in granular ferromagnets, [11] in a very controlled way is quench condensation. In this method, thin films are grown by sequential evaporation on a cryogenically cold substrate under UHV conditions while monitoring the film thickness and resistance. If the samples are quench condensed on a nonpassivated substrate, such as SiO_2, they grow in a granular manner so that the film is composed of separated islands. As more material is quench condensed, the average distance between the islands decreases and the resistance drops.

The hopping conductivity between two neighboring ferromagnetic grains depends on many factors but, for simplicity, we consider here the main two: Dependence on the grain-to-grain distance and dependence on the mutual orientation of the magnetic moments, **M**, of these grains. The effect of magnetic disorder is based on the assumption that the electron scattering, which takes place at the interfaces between the magnetic grains, depends on the electron spin direction. 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 $R_{\uparrow\downarrow}$. The resistance of the spin-up and spin-down electrons can be expressed as a function of the relative orientation angle Θ_{ij} between the magnetic moments $\mathbf{M}_i$ and $\mathbf{M}_j$ of the neighboring ferromagnetic grains denoted by $i=\{i_x,i_y\}$ and $j=\{j_x,j_y\}$, respectively. The spin-up and the spin-down electrons constitute two separate currents ("two-current" model [12,13]), and the total resistance R_{ij} can be found from expression

$$R_{ij}=R_0[1-(\delta R/2R_0)^2(1+\cos\Theta_{ij})^2], \tag{1}$$

where $R_0=(R_{\uparrow\uparrow}+R_{\uparrow\downarrow})/2$, $\delta R=(R_{\uparrow\uparrow}-R_{\uparrow\downarrow})/2$, $\cos\Theta_{ij}=\cos\theta_i\cos\theta_j+\sin\theta_i\sin\theta_j\cos(\phi_i-\phi_j)$, and θ,ϕ are the orientation angles of the magnetic moment of two neighboring ferromagnetic grains (labeled by "i" and "j").

The magnetic moment $\mathbf{M}_i$ of the ferromagnetic grain is assumed to be always parallel or antiparallel to its randomly distributed easy axis $\hat{e}_i$, i.e., $\mathbf{M}_i=\pm M_i\hat{e}_i$. At zero-magnetic field, the direction of the magnetic moment of each grain is set to be parallel to its easy axis. Once a magnetic field **H** is turned on, it can switch the direction of the magnetic moment if $\mathbf{M}_i\cdot\mathbf{H}<0$. This switching from the parallel to antiparallel direction will occur once the strength of the magnetic field is such that the average magnetization of the sample (according to the Langevin relation) is larger than the grains magnetic moment $\mathbf{M}_i$. Thus, the grain magnetic moment will flip once,

$$\mathbf{M}_i\cdot\mathbf{H}<\mathcal{L}(h)\equiv\coth(h)-1/h, \tag{2}$$

where $\mathcal{L}(h)$ is Langevin function, $h\equiv M_iH/k_BT$, and T is the temperature.

The spacial disorder can be taken into account by construction of a Miller-Abrahams resistor network [14]. Each contact between a pair of grains i and j is represented by a resistor with conductance σ_{ij} proportional to the tunneling probability: $\sigma_{ij}=\sigma_0\exp(-2\alpha r_{ij})$, where σ_0 is a constant of conductivity dimensionality, r_{ij} is the distance between the ith and jth grains, and α is the coefficient of the exponential

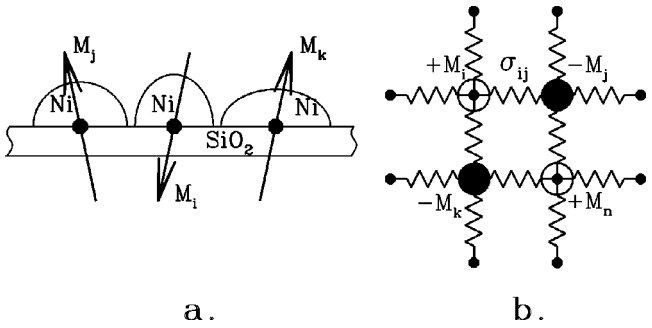

FIG. 1. (a) A schematic drawing of the Ni grains deposited on the insulating SiO_2 plate (site view). Magnetic moments of the Ni grains are directed randomly. (b) Square bond percolation net of resistors with random resistivity given by Eq. (3) mimicking the tunneling (hopping) conductance (see also Ref. [15]). The black circles on the sites of the network represent the magnetic moment of the grain directed downward (i.e., $-\mathbf{M}$), while the open circles represent the magnetic moment directed upward (i.e., $+\mathbf{M}$). The resistivity of each resistor depends on both the grain-to-grain distance [see Eq. (3)] and the mutual orientation of the magnetic moments $\mathbf{M}_i$ and $\mathbf{M}_j$ of the neighboring grains.

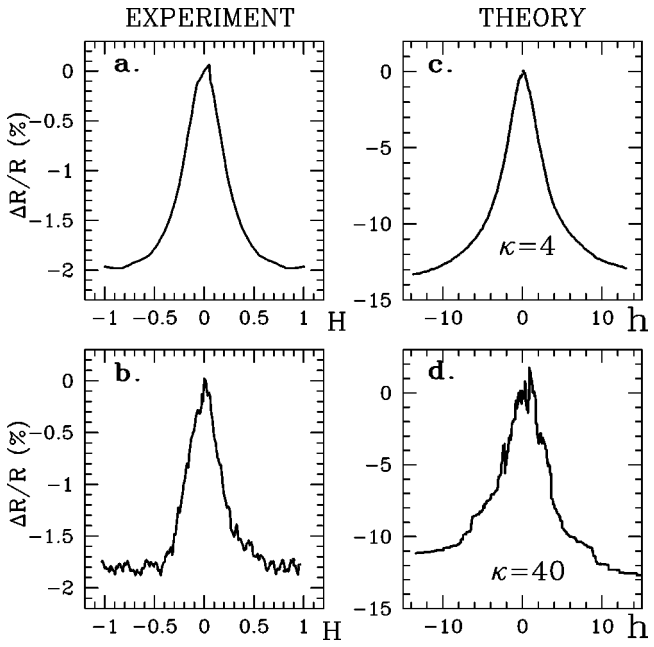

FIG. 2. (a) and (b) Experimental data of the relative magnetoresistance $\Delta R(H)/R(0)$ versus magnetic field $\mathbf{H}$ (in Tesla) of a dilute granular Ni sample for $T=4$ K (Ref. [5]). The sample in case (b) is more disordered than in case (a). The values of κ and system size, L, realized in these experiments [5] can be estimated as $\kappa \sim 10^2-10^3$, and $L \sim 10^5$. (c) and (d) Theoretical drawings of the similar quantity $\Delta R(h)/R(0)$ versus h [see Eq. (2)] obtained from numerical simulations on a random bond-percolating resistor network of the size $L=100$ with $\kappa=4$ [see (c)] and $\kappa=40$ [see (d)]. The ratio of the hopping resistivities when the magnetic moments of the grains are parallel compared to the antiparallel case [see Eq. (1)] is $R_{\uparrow\uparrow}/R_{\uparrow\downarrow}=4$.

decay (we do not consider here the thermal hopping over barriers of energy ΔE). In order to perform numerical simulations, we express the random distance between grains as $r_{ij}=\overline{l}\cdot \mathrm{rand}(ij)$, where $\mathrm{rand}(ij)$ is a random number taken from uniform distribution in the range (0,1), $\overline{l}=1/\sqrt{q}$ is the mean distance between metallic grains, and q is the two-dimensional (2D) density of these grains. Therefore, the bond conductance can then be rewritten as

$$\sigma_{ij} = \sigma_0(\Theta_{ij})\exp[-\kappa \cdot \mathrm{rand}(ij)], \tag{3}$$

where $\kappa \equiv 2\alpha\overline{l}=2\alpha/\sqrt{q}$ can be considered as the degree of disorder (a small density q of the deposited grains corresponds to strong disorder of the percolating system, i.e., large κ), $\sigma_0(\Theta_{ij})=1/R_{ij}$ where R_{ij} is defined by Eq. (1).

We model the film with Ni grains, reported in Ref. [5], deposited irregularly on a SiO_2 insulating plate [see Fig. 1(a)], as a square 2D bond-percolating resistor network [see Fig. 1(b)], where the conductivity of each resistor is assumed to have a random value. Between neighboring sites, we insert a resistor with random tunneling conductivity σ_{ij} [see Eq. (3) and Fig. 1(b)], whose conductivity is phenomenologically expressed as a function of the relative orientation angle Θ_{ij} of the effective magnetic moments of neighboring sites. Then, we solve the obtained system of linear Kirchhoff equations [16] and calculate the total effective resistance, R, of the 2D network as well as the values of the local current on each resistor.

In Figs. 2(a) and 2(b), we show 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. [5]). In Figs. 2(c) and 2(d), 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 [see Eq. (2)]. In the experimental data, pronounced noise due to sweeping the magnetic field back and forth can be clearly seen. 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, i.e., closer to percolation threshold) stronger jumps in the magnetoresistance curves are observed. 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 the resistor from the network 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 effect the system transport properties, e.g., the ohmic effective 2D resistivity. Therefore, the ratio R_{cut}/R (where R denotes the resistivity of the system prior to the removal of the resistor and R_{cut} is the resistivity after removing it) is an efficient characteristic of disorder. We expect that the ratio will be stronger for larger disorder.

In Fig. 3, we illustrate this by a color density plot of the

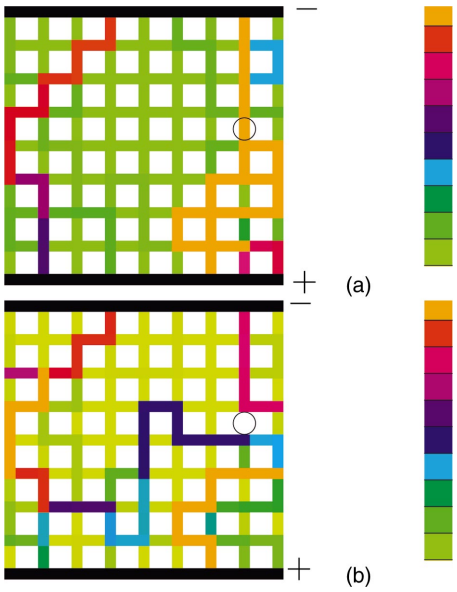

FIG. 3. (Color) A typical color density plot of the current distribution in a square bond-percolating lattice for which the voltage is applied in the vertical direction. This is a topological sketch in which the grains are located at the crossings of the grid lines and the spatial disorder is not presented. The current between the grains is shown by the different colors in the bonds composing the square grid. On the right-hand side, the current scales are shown: Orange corresponds to the highest value, green to the lowest. (a) Initial distribution of currents in the resistor network. The location of the resistor, on which the value of the local current is maximal, is shown by a circle. (b) The same for the modified network after cutting the spanning cluster by removing the resistor on which the local current is maximal. This removal results in a change of the current trajectories. Here, $L=8$ and $\kappa=22$.

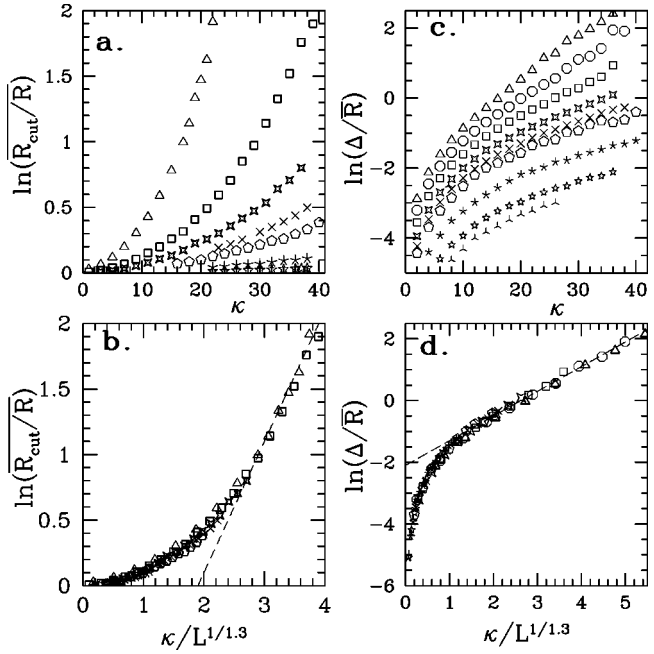

FIG. 4. (a) A semilog plot of the mean value of the ratio R_{cut}/R versus κ for various sizes of the system: $L=10$, 20, 30, 40, 50, 100, and 200 (from top to bottom). (b) A semilog scaling plot of the same quantity versus $\kappa/L^{1/1.3}$. (c) A semilog plot of the ratio of the variance of R normalized by the average resistivity $\Delta/\overline{R}$ versus κ for the various sizes of the system: $L=10$, 14, 20, 30, 40, 50, 100, 200, and 300 (from top to bottom). (d) A semilog scaling plot of the same quantity versus $\kappa/L^{1/1.3}$.

current distribution in the random resistor network before and after removing the bond with the maximal current. This cutting significantly changed the current trajectory.

To quantify this phenomena, we analyze the effect of κ on R_{cut}/R. In Fig. 4(a), we show a semilog plot of the statistically averaged value of the ratio R_{cut}/R [i.e., $\overline{R_{cut}/R} \equiv (1/N)\Sigma_{n=1}^{N} R_{n,cut}/R$] versus κ for various sizes of systems $L=10-200$. The average is taken over $N=10^4-10^6$ realizations. In Fig. 4(b), we present the scaling behavior of this ratio. It is found that R_{cut}/R scales well as a function of $\kappa/L^{1/1.3}$, while in the limit of strong disorder this ratio scales as $\ln(\overline{R_{cut}/R}) \propto \kappa/L^{1/1.3}$. This scaling is in agreement with the scaling found in Ref. [17] for the transition from weak disorder to strong disorder in the case of optimization. A similar scaling is also obtained for the ratio $\Delta/\overline{R}$ [where $\Delta \equiv \sqrt{\Sigma_{n=1}^{N}(R_n-\overline{R})^2/N}$ is the variance of R and $\overline{R} \equiv \Sigma_{n=1}^{N} R_n/N$ is the statistically averaged value of R]. In Fig. 4(c), we show the dependence of the ratio $\Delta/\overline{R}$ versus κ for different sizes of the system. In Fig. 4(d), we show its scaling behavior which is the same as in Fig. 4(b), i.e., $\ln(\Delta/\overline{R})$ scales as $\kappa/L^{1/1.3}$, again in agreement with Ref. [17]. It is plausible that the scaling parameter $\kappa/L^{1/1.3}$ should, in general, be $\kappa/L^{1/\nu}$, where in $d=2$ percolation $\nu=1.33$. Indeed, similar numerical studies [18] for $d=3$ yield good scaling with $\kappa/L^{1/0.9}$ consistent with $\nu=0.88$ in $d=3$ percolation.

In summary, the recently observed features of the electrical transport in dilute granular Ni films [5], 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 are similar to the experimental measurements and indicate 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 $L^{1/\nu}$, i.e., of 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.

In addition, the unique geometry of the samples enables one to detect the properties of a single grain even if the sample has macroscopic dimensions. The considered granular samples also provide a unique opportunity to study magnetoconductive effects on nanosized structures. The results obtained on these extreme samples shed some light on the behavior of the ferromagnetic granular samples in general and they may also be relevant for the development of new types of magnetoresistive-based single-grain devices.

This research was supported in part by grants from the U.S.–Israel Binational Science Foundation (Grant Nos. 2002–402 and 98–370), the Israel Science Foundation (Grant Nos. 326/02, 276/01, and 274/01), and the KAMEA Fellowship program of the Ministry of Absorption of the State of Israel. We gratefully acknowledge useful conversations with S. V. Buldyrev.

[1] J. I. Gittleman, Y. Goldstein, and S. Bozowski, Phys. Rev. B **5**, 3609 (1972).
[2] A. Frydman, O. Naaman, and R. C. Dynes, Phys. Rev. B **66**, 052509 (2002).
[3] C. J. Adkins, J. Phys. C **15**, 7143 (1982).
[4] I. P. Zvyagin and R. Keiper, Philos. Mag. B **81**, 997 (2001).
[5] A. Cohen, A. Frydman, and R. Berkovits, Solid State Commun. **129**, 291 (2004).
[6] A. Ghazali and J.-C. Lévy, Phys. Rev. B **67**, 064409 (2003).
[7] M. Strongin, R. Thompson, O. Kammerrer, and J. Crow, Phys. Rev. B **1**, 1078 (1970).
[8] R. C. Dynes, J. P. Garno, and J. M. Rowell, Phys. Rev. Lett. **40**, 479 (1978).
[9] H. M. Jaeger, D. B. Haviland, B. G. Orr, and A. M. Goldman, Phys. Rev. B **40**, 182 (1989).
[10] R. P. Barber and R. E. Glover III, Phys. Rev. B **42**, 6754 (1990).
[11] A. Frydman and R. C. Dynes, Solid State Commun. **110**, 485 (1999).
[12] W.-G. Yin and R. Tao, Phys. Rev. B **62**, 550 (2000).
[13] D. M. Edwards, J. Mathon, and R. B. Muniz, IEEE Trans. Magn. **27**, 3548 (1991).
[14] A. Miller and E. Abrahams, Phys. Rev. **120**, 745 (1960).
[15] V. Ambegaokar, B. I. Halperin, and J. S. Langer, Phys. Rev. B **4**, 2612 (1971).
[16] Following S. Kirkpatrick, Rev. Mod. Phys. **45**, 574 (1973) and A. K. Sarychev, D. J. Bergman, and Y. M. Strelniker, Phys. Rev. B **48**, 3145 (1993), we write the Kirchhoff equations for each site, i.e., that the sum of the currents flowing into every site is zero: $\Sigma_i I_i = 0$, where $I_i = \Sigma_j (V_j - V_i)\sigma_{ij}$, V_i is the voltage at site i. We solve the system of coupled linear equations for the voltages at every site using standard SSPOTRF and SSPOTRS sparse solver subroutines accommodated for Cray supercomputers.
[17] M. Porto, N. Schwartz, S. Havlin, and A. Bunde, Phys. Rev. E **60**, R2448 (1999).
[18] Y. M. Strelniker and S. Havlin (unpublished).