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Mesoscopic effects in macroscopic granular systems

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Abstract

Granular metals (systems of discontinuous metallic thin films) characterized by large distributions of grain sizes and inter-grain coupling were prepared by quench condensation. These samples, which are 2 mm $\times$2 mm in size, exhibit a number of experimental findings characteristic of low dimensional samples including magnetoresistance oscillations, sample-to-sample fluctuations and resistance switches. Such 'mesoscopic' effects are observed in a variety of metals such as Ni, Pb and Au. The results indicate that the transport in these systems is governed by a very small number of grains even though the sample contains about 10^9 grains. We interpret the findings as indications that the large distribution of resistances in the percolation network of the conduction trajectories facilitates the situation in which a small number of grains dominate the transport of the macroscopic system.

1. Introduction

Electric transport in a strongly localized system is an inherently inhomogeneous process. It has long been recognized that a percolation treatment is a proper way to treat such a system since it provides much insight into the physics of the conductivity in these systems [1–3]. In this approach each pair of localized states, i and j, is represented by a resistor with conductance proportional to the tunneling probability between the grains [4]:

$$\sigma \propto \exp -\left[\frac{2r_{ij}}{\xi} + \frac{E_{ij}}{KT}\right] \tag{1}$$

where r_{ij} is the spatial distance between the localized states, ξ is the localization length and E_{ij} is the difference in energy $E_i - E_j$. The percolation calculation leads to the understanding that the transport is governed by critical resistors, which act as 'red bonds' of the percolation network. The critical resistors determine the conductivity of the entire system. Hence, the scale of inhomogeneity is L_c, the percolation radius, which can be viewed as the average distance between critical resistors. This percolation treatment reproduces the well known variable range hopping transport behavior, which for weak electron interactions is [5]

$$R \propto \exp\left[\frac{T_0}{T}\right]^{d+1} \tag{2}$$

where d is the system dimension, and for strong interactions is [6]

$$R \propto \exp\left[\frac{\widetilde{T}}{T}\right]^{0.5}. \tag{3}$$

Transport in granular metals, i.e. systems of metallic islands imbedded in an insulating matrix, is similar to a continuous strongly localized system in many ways. If the grains are isolated from each other, the transport is governed by hopping between the grains. Hence, a percolating treatment is also natural in a granular system [7]. Each pair of grains is associated with a resistor and the system is governed by a relatively small number of critical resistors which carry the entire current flow. The main difference is that in a granular system many electrons can exist on a single localized dot and E_{ij} is determined by the difference in the charging energies between dots i and j. This depends mainly on the respective grain sizes. Experimentally, many granular systems exhibit transport curves similar to that of equation (3). Though this behavior has been observed for several decades its origin is still controversial and various theoretical models have been proposed to explain it [7–10].

The percolation nature of the transport in granular metals emphasizes the role of disorder. The distribution of geometrical parameters such as grain size and inter-grain spacing determines the distribution of resistances in the percolation network. L_c can be expected to grow as a function of resistance distribution. Recently Strelniker *et al* [11] showed that, as the disorder of a granular system is increased, the

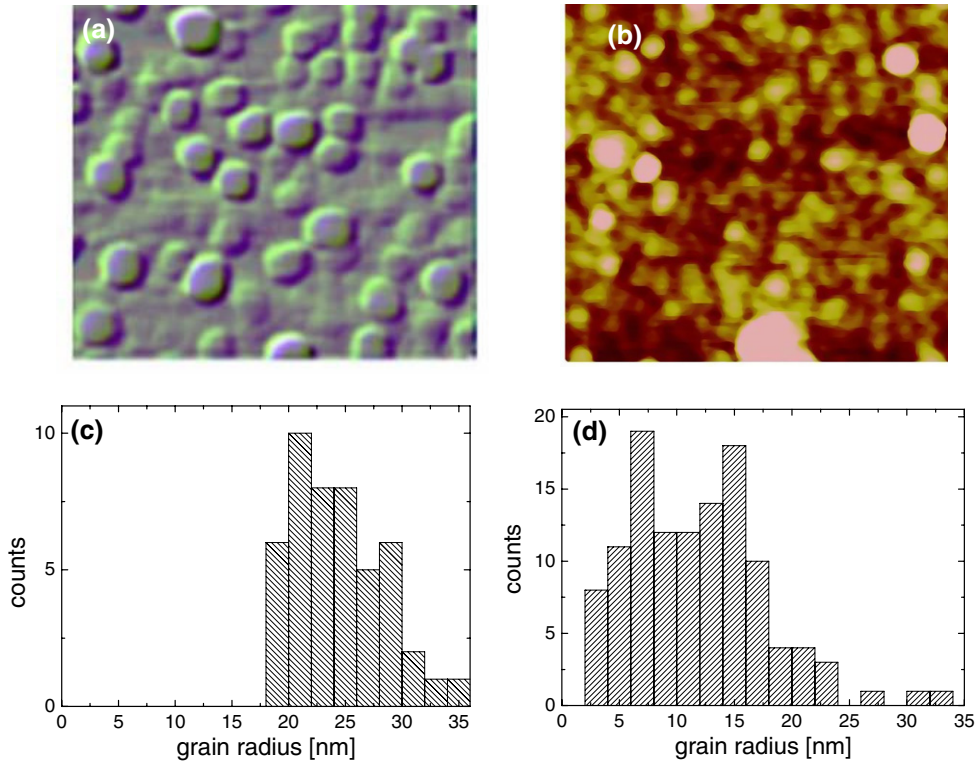

Figure 1. AFM images of 500 nm ∗ 500 nm granular Ni samples evaporated at 300 K in which the evaporation was terminated for a resistance of a few MΩ. The left image is for a sample that was evaporated at 10^{-8} Torr while the right is for a sample evaporated at 10^{-4} Torr. A histogram of the grain sizes is shown respectively below each AFM image.
(This figure is in colour only in the electronic version)

number of 'red bonds' (critical resistors) of the percolation network decreases and may reach an order of unity for extremely strong disorder. Under such conditions the transport can be expected to be governed by very few pairs of grains, hence the sample may exhibit properties of mesoscopic nature despite its macroscopic dimensions. Indeed, low temperature (50 mK) conductance fluctuations have been observed in a macroscopic continuous InO_x hopping system [12] and their origin was shown to be due to orbital effects similar to those observed in mesoscopic diffusive samples.

In this paper we compare two types of granular metals, one with a low distribution of resistors and the other with high resistance distribution. While the former show features typical of conventional granular systems, the later exhibit a number of experimental findings that indicate that the transport is governed by a very small number of grains even in systems in which the total number of grains is very large (10^9). In the granular case (unlike that of [12]) these phenomena are observed in all samples with high enough disorder and persist up to temperatures of the order of 10 K.

2. Experimental details

An established method to fabricate granular films in a very controlled way is quench condensation [13–19]. In this method an insulating substrate with metallic leads is placed on a sample holder within a vacuum chamber. After the chamber is pumped out, the substrate is cooled to cryogenic temperatures and thin films are grown on the substrate by sequential evaporations while monitoring the film thickness and resistance. If the samples are quench condensed on a non-passivated substrate such as SiO, the initial morphology is that of a discontinuous film constructed of an array of small metallic grains separated by vacuum. As more material is quench condensed, the average distance between the islands decreases and the resistance drops. Using this technique one can terminate the deposition at any desired thickness, thus achieving very fine control on the sample resistance.

The geometry of a quench condensed granular film turns out to be highly dependent on the pressure in the deposition chamber. Figure 1 compares AFM scans of two granular Ni samples, both having the same sheet resistance of ∼3 MΩ, in which the base pressure was of the order of 10^{-8} Torr and 10^{-4} Torr, respectively. In these cases the evaporation was performed at room temperature in order to enable AFM scans without distorting the as-deposited geometry. In general, the quench condensed samples are expected to have smaller grains than those of the room temperature evaporated samples. Hence, the AFM scans can not be used for exact qualitative analysis of the samples under study. Nevertheless, these micro-scans can provide qualitative information on sample morphology trends. Figure 1 shows that the low vacuum sample is much more disordered than the high vacuum sample. It shows a larger distribution of both the grain diameter and the inter-grain distance. Perhaps more important is the fact that the high pressure sample contains a large fraction of

smaller grains. These will be very unfavorable for hopping through because of their large charging energy (equation (1)). Therefore, the fact that the resistance at $T = 4$ K is similar for both samples indicates that the current percolation path is very different for both samples.

Though we can not be certain about the origin of the morphology difference between the samples, we postulate that evaporating at relatively high base pressure may have two effects. The first is an enhanced scattering of the deposited adatoms leading to a higher disorder of the film, thus giving rise to higher distribution of grain sizes and inter-grain distances. The second is the fact that the grains undergo a larger degree of oxidation in the chamber. This means that the effective grains for transport are smaller. This will have a particularly large effect on grains which are small to begin with, for which reducing the effective volume causes hopping through them to become very unfavorable (see equation (1)). In extreme cases some of the grains may become inaccessible for hopping, thus diluting the transport percolation network.

In the remainder of this paper we will compare results obtained on samples of these two types. We name the samples evaporated under UHV conditions 'conventional samples' and the samples prepared under lower vacuum conditions 'highly disordered' samples. It should be noted, though, that in both cases the samples are in the strongly localized regime, exhibiting hopping curves similar to equation (3). In the following section we present a number of experimental results indicating that the transport in highly disordered granular metals is governed by a very small section of the sample.

3. Results

3.1. *I–V characteristics*

Granular metals in the hopping regime are characterized by non-linear I–V characteristics. In many cases these curves follow a power law of the form

$$I = \sum_k a_k V^k \tag{4}$$

for which the dominant power, k, has been interpreted as corresponding to the number of grains involved in a single hopping event [20]. All of our quench condensed samples show I–V curves which, for low enough voltages, have the form of equation (4). Figure 2 presents I–V curves for a conventional sample and a highly disordered sample. In both cases the curve starts linear and crosses over to a quadratic dependence of I on V. However, the voltage scales are very different for both samples. This is demonstrated in figure 2(b) in which the R–V curves are plotted while scaling the voltage axis by an appropriate V_0. It is seen that, though the functional form is similar for both samples, the voltage scale, V_0, for the highly disordered sample is 260 times smaller than that of the conventional sample.

It is instructive to consider these results with the understanding that in a hopping system the nonlinearity is due to the energy which is supplied to the hopping electron by the electric field, $\tilde{F}$. This field plays an analogous role to the

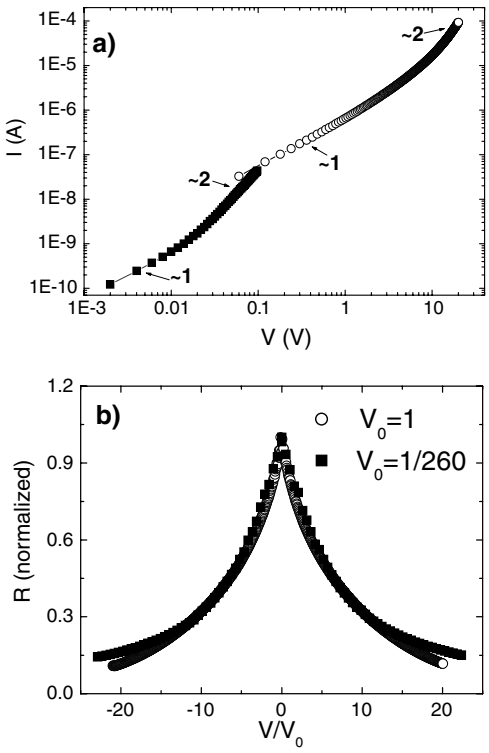

Figure 2. I–V measurements (a) and R–V plots (b) of conventional (empty circles) and disordered (full squares) samples. The numbers in the top panel correspond to the power in equation (4). In the R–V curves the V axis was scaled by a factor of V_0.

sample temperature. Thus, for finite electric field, equation (1) may be substituted by

$$\sigma \propto \exp -\left[\frac{2r_{ij}}{\xi} + \frac{E_{ij}}{k_B T + e\tilde{F} r_h}\right] \tag{5}$$

where r_h is the relevant hopping length. This defines a typical voltage $\tilde{V} = \tilde{F} r_h$ that drops across a single hop such that $V = \tilde{V} * n$, where V is the voltage applied across the sample and n is the number of significant hops in series. A smaller characteristic voltage across the sample corresponds to a smaller number of hops, implying that a small section of the sample is participating in the hopping process. The results presented in figure 2 indicate that the relevant length for transport in the disordered system is two orders of magnitude smaller than that in the ordered sample.

3.2. *Magnetoresistance fluctuations*

A distinct fingerprint of mesoscopic samples is magnetoresistance (MR) fluctuations due to Aharonov–Bohm effects on the electronic current trajectories. We observe reproducible perpendicular field MR fluctuations in many of our disordered granular samples. The effect is much more pronounced in the case where the granular material is a superconductor. In general, granular superconductors are characterized by a large positive magnetoresistance due to the suppression of superconductivity with magnetic field (MR) [21]. While 2D samples exhibit a smooth MR increase, in 1D samples there is a

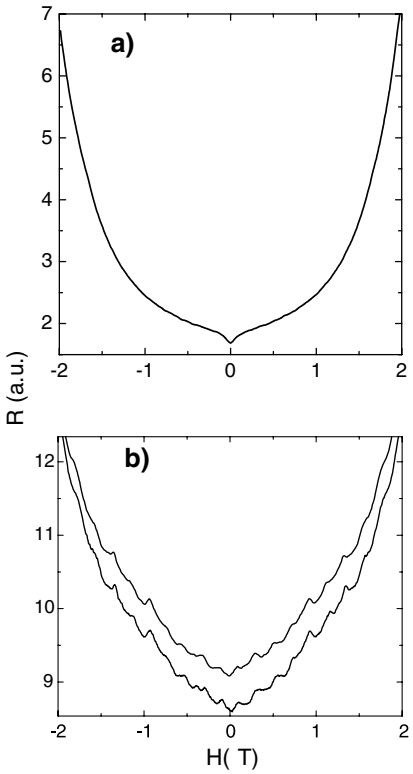

Figure 3. Magneto-resistance of 2 mm ×2 mm quench condensed granular Pb taken at 4.5 K for a conventional 2D sample (a) and a highly disordered sample (b). The different traces refer to different swipes shifted for clarity.

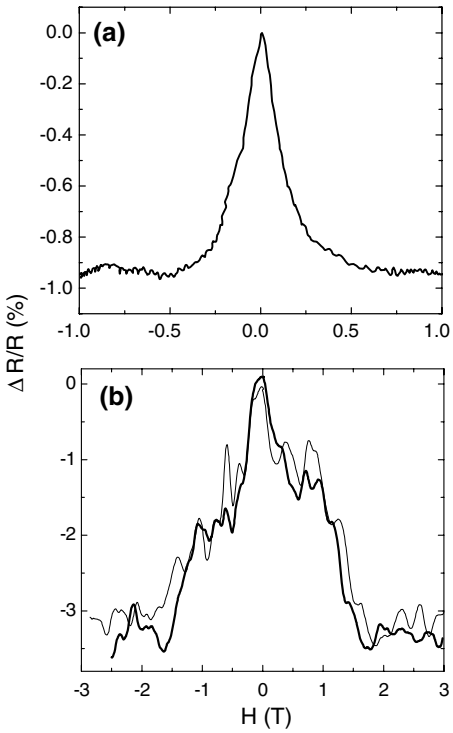

Figure 4. MR of a conventional Ni sample (top) and a highly disordered granular Ni sample exhibiting conductance fluctuations (bottom). The two traces are two different field sweeps, showing that most of the fluctuations are reproducible. The sample size was 2 mm ×2 mm for both cases.

MR quasi-oscillation superimposed on the positive MR background [22]. This behavior has been ascribed to Little–Parks oscillations that take place in current loops consisting of superconducting grains. In the 2D samples there are many loops enclosing different areas and characterized by different field oscillation periods. Hence, the resistance oscillations average out. In the 1D wires there is a characteristic loop area, which is determined by the width of the wire. Thus, there is no averaging out and the observed oscillation corresponds to this typical loop size. Figure 3 depicts the MR curves of two 2D granular films, one conventional and one highly disordered, in which the magnetic field, H, is applied perpendicular to the films. It is seen that the disordered sample exhibits MR oscillations which resemble those observed in 1D wires [22]. These oscillations have a quasi-period $\Delta H \sim 0.2T$, which corresponds to an area of 1000 Å^2. This is consistent with a loop consisting of about five to seven grains which dominates the conductivity in the sample. We reemphasize that the samples have macroscopic dimensions. The fact that the oscillations are not averaged out in these samples implies that very small regions dominate the electric properties of the entire sample.

Reproducible MR features are also observed in highly disordered granular normal metals in the hopping regime, though they are much smaller in amplitude and much less regular. Figure 4 depicts the MR curves of an ordered and a disordered granular Ni sample. The overall curve shows a negative MR at small fields which saturates at higher H. This behavior is due to alignment of the grain magnetic moments [17–19]. However, while the conventional sample exhibits a smooth curve, the disordered sample's MR curve is characterized by reproducible fluctuations superimposed on the negative background.

In a hopping system the phase breaking length, L_φ, relevant for mesoscopic fluctuations, was shown to be the hopping length [23, 24]. In granular hopping systems this is about 1000 Å [18]. Indeed, a typical fluctuation seen in our samples, ∼0.33 T, is consistent with a loop diameter of 780 Å. The fact that these fluctuations are not washed out in a sample with a spatial dimension which is four orders of magnitude larger than L_φ implies again that a very small section of the sample dominates the transport.

3.3. Magnetoresistance switches

An additional unique effect is observed in many of the highly disordered granular ferromagnets, i.e. reproducible sharp resistance jumps at a specific magnetic field [25, 26]. The origin of such a jump, seen in figure 5, has been interpreted as being due to magneto-mechanical distortions of the grains which act as bottlenecks in the percolation network of the conductance. Whether this interpretation is correct or not, it seems clear that the sharp resistance changes are associated with the behavior of single (or a small number of) grains. It is hard to think of a scenario for such abrupt transport changes if a large number of grains are involved, in particular due to the fact that these features are observed only in the highly

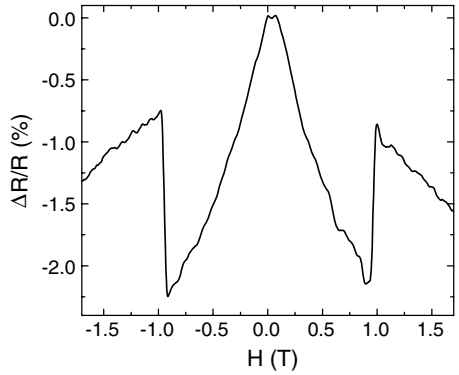

Figure 5. MR of a 2 mm ×2 mm highly disordered granular Ni sample exhibiting sharp resistance jumps.

disordered samples and were never seen in conventional 2D granular ferromagnets.

3.4. Sample to sample fluctuations

A clear difference between the conventional and the highly disordered samples is the sample-to-sample variations in the transport properties. It is impossible to compare the conductivity of two quench condensed samples because the conductivity is extremely sensitive to the film thickness. A nominal thickness change of a fraction of an Å can cause an order of magnitude resistance change. It is therefore more useful to consider the fluctuation with regards to other parameters that characterize the sample. We chose to focus on two such parameters, the MR amplitude, $\Delta R/R$, and the saturation field, H_S, in granular ferromagnets. Figure 6 shows these two parameters as a function of sample resistance for a number of conventional granular Ni films and a number of highly disordered films. Each data point corresponds to one sample, or to a single evaporation stage.

While in the conventional case the MR never exceeds the value of 2%, and decreases smoothly with the sheet resistance, the MR amplitude in the disordered samples exhibits strong sample-to-sample fluctuations, reaching values as high as 50%, and has no clear dependence on the sheet resistance. A similar behavior is seen with regards to H_S The conventional samples exhibit a constant value of 0.5 T, whereas in the highly disordered samples there is a wide spread of saturation fields, reaching values of a few tesla.

Both these effects, i.e. large MR amplitudes and large saturation fields, have been observed in small granular Ni films as well [27]. These findings were interpreted as arising from the fact that in systems smaller than L_c the transport is forced to flow through relatively small grains. This leads to large saturation fields [28], and presumably also to large MR magnitudes. The same effects appearing in our macroscopic samples demonstrate, once again, that these systems behave as 'small systems' with regards to electric transport.

4. Summary

We compare transport and magneto-transport results obtained on two types of granular samples, made of the same material, having similar resistances and fabricated using the same method. The only apparent difference was the chamber pressure, which presumably affected the distribution of resistances in the conduction percolation network. Our results demonstrate that the highly disordered samples exhibit behavior typical of low dimensional samples. This is consistent with the notion invoked in [11] that a very small fragment of the sample dominates the entire transport properties. This perception can be used to study a small granular system without the need for advanced lithography techniques.

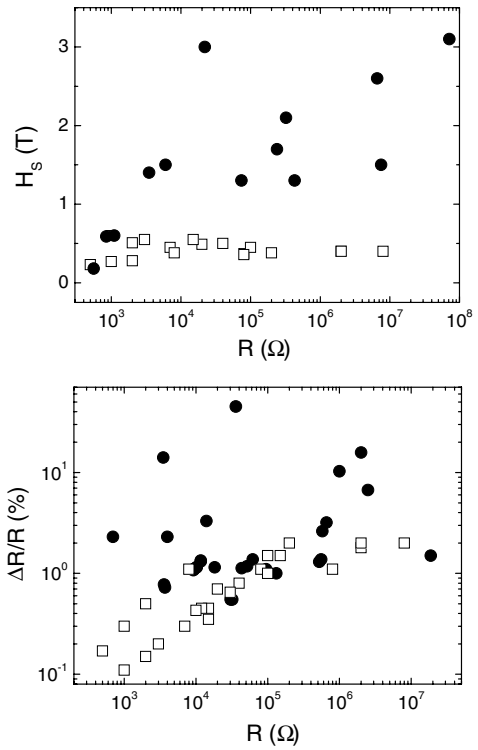

Figure 6. (a) Saturation field, H_s, and (b) $\Delta R/R$ as a function of sheet resistance for a conventional 2D granular Ni (empty squares) and a highly disordered sample (full circles).

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