

Quantum Percolation in Granular Metals

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Theory of quantum corrections to conductivity of granular metal films is developed for the realistic case of large randomly distributed tunnel conductances. Quantum fluctuations of intergrain voltages (at energies E much below the bare charging energy scale E_C) suppress the mean conductance $\bar{g}(E)$ much more strongly than its standard deviation $\sigma(E)$. At sufficiently low energies E_* any distribution becomes broad, with $\sigma(E_*) \sim \bar{g}(E_*)$, leading to strong local fluctuations of the tunneling density of states. The percolative nature of the metal-insulator transition is established by a combination of analytic and numerical analysis of the matrix renormalization group equations.

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Introduction— Low-temperature electron transport in granular metals has been intensively studied in recent years [1–4]. It was shown that in the temperature range $T \geq g\delta$ (where $g \gg 1$ is the characteristic value of dimensionless intergrain conductance in units of $e^2/2\pi\hbar$, and δ is the intragrain level spacing), quantum corrections to conductivity originate mainly from local fluctuations of voltages between neighboring grains. This effect can be treated within the Ambegaokar-Eckern-Schoen model [5] and leads to logarithmic temperature dependence of the effective conductance [1]:

$$g(T) = g_0 - \frac{4}{z} \ln \frac{g_0 E_C}{T}, \quad (1)$$

where $E_C \gg \delta$ is the charging energy of an individual grain, g_0 is the bare tunneling conductance of intergrain junctions (identical for all junctions), and z is the coordination number of the lattice. The result (1) is valid as long as the renormalized conductance $g(T)$ is large, i.e., down to temperatures $T_1 = g_0 E_C e^{-zg_0/4}$. It was argued [2,3] that transition from metal to insulator behavior (MIT) occurs at $T \sim T_1$ as long as $T_1 \geq g\delta$. The same conclusion for the two-dimensional (2D) case was reached [4] via instanton analysis.

Although the above results may well be applied to artificial 2D arrays of well-defined tunnel junctions, tunnel conductances g_{ij} are random in granular metals. In this Letter we investigate the role of g_{ij} randomness for energy (temperature) dependent properties of thin granular films, such as macroscopic conductance $g_{\text{eff}}(T)$ and the local tunneling density of states (LTDOS) $\nu_i(E)$. Quantum fluctuations lead to suppression of g_{ij} described by the one-loop renormalization group (RG) equation:

$$\frac{dg_{ij}}{dt} = -2g_{ij}R_{ij}, \quad (2)$$

where $t(E) = \ln(\bar{g}_0 E_C/E)$ is the auxiliary RG “time”, \bar{g}_0 being some mean bare conductance, and R_{ij} is the resist-

ance of the network between the points i and j . Physically, renormalization of g_{ij} is due to fluctuations of voltage between the grains i and j , which are governed by the corresponding resistance R_{ij} . Equation (2) is a straightforward generalization of the RG equation for a regular array [1] with $g_{ij} \equiv g$ and $R_{ij} = 2/gz$, whose solution is given by Eq. (1). The system of RG Eqs. (2) is nontrivial since R_{ij} is a complicated nonlocal function of all individual conductances g_{kl} .

In a regular system, Eq. (2) drives all conductances to zero simultaneously at $t = t_c = zg_0/4$, marking the point of the MIT. We will show that in a random system renormalized conductances of the junctions collapse to zero neither all at once, nor one by one, but in groups. These groups enclose clusters, consisting of one or several sites, which become disconnected from the rest of the network after the collapse (see Fig. 1). As a result, the MIT in a natural granular system is a percolative transition: it takes place when enough clusters have become disconnected so that the percolation via still conducting links is destroyed.

The above picture of conductances, eventually collapsing to zero, follows from the one-loop RG Eq. (2). The one-loop approximation breaks down at $g \sim 1$; at lower

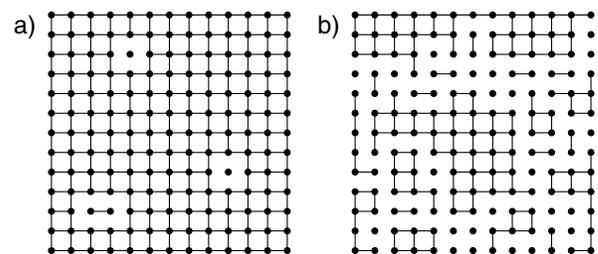


FIG. 1. Conducting bonds (with $g_{ij} > 0$) at different values of the RG time t . Results of numerical simulation of Eq. (2) on the lattice 20×20 with $P_0(g) = [2g_0\theta(g)/\pi]/(g^2 + g_0^2)$. (a) $t = 0.64g_0$ with the fraction of conducting bonds $N_{\text{cond}} = 0.95$; (b) $t = 1.08g_0$ with $N_{\text{cond}} = 0.55$.

energies the conductance decays exponentially with the RG time $t(E)$. Therefore, Eq. (2) can adequately describe only the initial stage of the MIT. Nevertheless, there exists a region near the transition where the percolative cluster contains good conductances with $g > 1$ so that Eq. (2) is still applicable.

We start from the case of relatively narrow original distribution $P_0(g)$, characterized by the mean value \bar{g}_0 and the standard deviation $\sigma_0 \ll \bar{g}_0$, and show that the renormalized distribution $P(g)$ broadens. In particular, for the square lattice ($z = 4$):

$$\frac{\sigma(E)}{\bar{g}(E)} = \frac{\sigma_0}{\bar{g}_0} \frac{\bar{g}_0/\bar{g}}{\sqrt{2 \ln(\bar{g}_0/\bar{g}) \ln \ln(\bar{g}_0/\bar{g})}}, \quad (3)$$

where $\bar{g} \equiv \bar{g}(E) = \bar{g}_0 - \ln(\bar{g}_0 E_C/E)$. Equation (3) is a large- $\ln(\bar{g}_0/\bar{g})$ asymptotics of a more general expression (see Eq. (7) below). It is valid as long as $\sigma(E) \ll \bar{g}(E)$, i.e., above $E_* = T_* = \bar{g}_0 E_C e^{-\bar{g}_0 + \sigma_0} \gg T_1$, where T_1 marks MIT in an ideal array with $\sigma_0 \ll 1$. Thus transition from metal into insulator in a granular array is intrinsically inhomogeneous. The vicinity of this transition at $\max(E, T) \leq T_*$ is difficult for exact analytical treatment as the width of distribution $P(g|E)$ becomes of the order of its mean value. In this region we employ the effective-medium approximation (EMA) and numerical solution of the RG Eqs. (2), and demonstrate that MIT is of a percolative nature.

Strong self-developed inhomogeneity of a granular array can be probed by scanning tunneling measurement of the LTDOS modified by the Coulomb zero-bias anomaly (ZBA) [6–8]. ZBA modification $Z(E) = \nu(E)/\nu_0$ of the average LTDOS in a regular array was considered in Refs. [1,3] and found to become very large before approaching MIT. Here we analyze spatial fluctuations of the ZBA suppression factor $Z_i(E)$. For an originally narrow distribution $P_0(g)$, the log-normal distribution of the ZBA factors is found, with $\text{std}[\ln Z_i(E)] \approx \sigma(E)/\bar{g}(E)$. Thus we predict order-of-unity local fluctuations of LTDOS at $\max(E, T) \leq T_*$. Spatial correlation length $\xi(E)$ of these fluctuations is found to grow moderately with decreasing E in the case of weak original disorder $\xi(E) \approx \sqrt{\ln[\bar{g}_0/\bar{g}(E)]}$, reaching $\sqrt{\ln(\bar{g}_0/\sigma_0)}$ at the border of strong inhomogeneity $E \sim T_*$. For the region in the vicinity of MIT, where relative fluctuations are large, we present numerical analysis of LTDOS fluctuations. Below we provide a brief derivation of our results.

Narrow distribution.—If the standard deviation σ of the distribution is much smaller than the mean \bar{g} , the latter follows the homogeneous solution (1): $\bar{g}(t) = \bar{g}_0 - t$, while evolution of $\delta g_{ij} = g_{ij} - \bar{g}$ can be described perturbatively. Resistance can be written as $R_{ij} = G_{ii} + G_{jj} - 2G_{ij}$, where $G_{ij} = \hat{A}_{ij}^{-1}$ is the Green function of the diffusion operator on the network defined by the matrix elements $A_{ii} = \sum_j g_{ij}$ and $A_{ij} = -g_{ij}$ [9]. Using the per-

turbative series $G_{ij} = \bar{G}_{ij} - \bar{G}_{ik} \delta A_{kl} \bar{G}_{lj} + \dots$ we find

$$\delta G_{ij} = - \sum_{\langle kl \rangle} \delta g_{kl} (\bar{G}_{ik} - \bar{G}_{il}) (\bar{G}_{jk} - \bar{G}_{jl}). \quad (4)$$

In the momentum representation $\bar{G}(\mathbf{p}) = [2\bar{g}\varepsilon(\mathbf{p})]^{-1}$, for the square lattice, $\varepsilon(\mathbf{p}) = 2 - \cos p_x - \cos p_y$.

To proceed further we choose a vector representation for conductances g_i^α , when each edge is characterized by the lattice site i , which it goes from, and direction α , which can be either horizontal ($+x$) or vertical ($+y$). Using Eq. (4), introducing a new time variable $s = \ln[\bar{g}_0/\bar{g}(t)] = -\ln[1 - t/\bar{g}_0]$ and passing to Fourier representation, we get a linear evolution $d\delta g_\alpha(\mathbf{p})/ds = -\mathfrak{M}_{\alpha\beta}(\mathbf{p})\delta g_\beta(\mathbf{p})$, governed by the 2×2 time-independent matrix $\mathfrak{M}(\mathbf{p})$. One of its spectral branches, $\lambda_+(\mathbf{p})$, is gapped, whereas the other branch $\lambda_-(\mathbf{p})$, becomes gapless in the long wavelength limit $\lambda_-(\mathbf{p} \rightarrow 0) \approx (p^2/4\pi) \ln(1/p)$, see Ref. [9]. Individual conductances evolve as

$$\delta g_\alpha(\mathbf{r}, s) = \sum_{\mathbf{r}'} K_{\alpha\beta}(\mathbf{r} - \mathbf{r}', s) \delta g_\beta(\mathbf{r}', 0), \quad (5)$$

with the Fourier-transformed kernel given by

$$K(\mathbf{p}, s) = K_1(\mathbf{p}, s) + K_2(\mathbf{p}, s)(e^{i\alpha_p} \hat{\sigma}_+ + e^{-i\alpha_p} \hat{\sigma}_-), \quad (6)$$

where $K_{1,2}(\mathbf{p}, s) = (e^{-\lambda_-(\mathbf{p})s} \pm e^{-\lambda_+(\mathbf{p})s})/2$, $\alpha_{\mathbf{p}} = (p_x - p_y)/2$, $\hat{\sigma}_\pm = (\hat{\sigma}_1 \mp \hat{\sigma}_2)/2$ and $\hat{\sigma}_k$ are the Pauli matrices.

Assuming that different conductances are uncorrelated at $s = 0$, we find for the variance of the distribution:

$$\frac{\sigma^2(s)}{\sigma_0^2} = \frac{1}{2} \int (d\mathbf{p}) [e^{-2\lambda_-(\mathbf{p})s} + e^{-2\lambda_+(\mathbf{p})s}]. \quad (7)$$

For $s \ll 1$, $\sigma(s)/\sigma_0 = 1 - s/2 + \dots$, which is to be compared with the decay of the mean conductance $\bar{g}(s)/\bar{g}_0 = e^{-s} = 1 - s + \dots$. Thus, even at the initial stage of the evolution, the width of the distribution decays slower than its average. In the case $s \gg 1$, the integral (7) is dominated by the soft mode $\lambda_-(\mathbf{p})$ at $\mathbf{p} \rightarrow 0$ leading to $\sigma^2(s)/\sigma_0^2 \approx 1/(2s \ln s)$ and hence to Eq. (3).

Apart from broadening the single-site distribution $P(g)$, the RG flow (2) produces correlations between δg at different links $C_{\alpha\beta}(\mathbf{r}; s_1, s_2) = \langle \delta g_\alpha(\mathbf{r}, s_1) \delta g_\beta(0, s_2) \rangle$. The Fourier transform of the correlation function reads:

$$C(\mathbf{p}; s_1, s_2) = \sigma_0^2 K(\mathbf{p}; s_1 + s_2). \quad (8)$$

At the initial stage of evolution, $s \equiv (s_1 + s_2)/2 \leq 1$, correlations are short ranged. At $s \geq 1$, correlations with large correlation length $\xi(s) = \sqrt{(4/\pi)s \ln s}$ develop: $C_{\alpha\beta}(\mathbf{r}, s_1, s_2) = \sigma^2(s) \exp[-r^2/\xi^2(s)]$.

Spatial fluctuations of g_{ij} lead to fluctuations of the LTDOS $\nu_i(E) = Z_i(E)\nu_0$. The ZBA suppression factor $Z_i(E)$ for granular media at $E \geq g\delta$ can be found according to the simple “environmental theory” [10]:

$$\ln Z_i(E) = -2 \int_0^t R_i(t') dt', \quad (9)$$

where $R_i(t)$ is the resistance between the site i and the far region of the array at the energy scale $E = \bar{g}_0 E_C e^{-t}$. The same result follows from the analysis provided in [1,3]. It is important to note that in a homogeneously disordered metal the short-length cutoff in the integral that determines the effective resistance $R(E)$ is given by the diffusion length $\sqrt{\hbar D/E}$, whereas in the present case it is just the grain size. The long-scale cutoff for the logarithmic divergency of $R_i(E)$ in 2D is $L(E) = e^2 g_{\text{eff}}/E$ (in the absence of external screening). Thus, one can write $R_i(E) = G_{ii}^{\text{reg}}$, where the otherwise divergent G_{ii} is regularized by the finite length $L(E)$. Local fluctuations of $\ln Z_i(E)$ are determined by a much smaller region of the size ξ around the site i so that the object $\delta R_i = \delta G_{ii}$ is already free of infrared divergency and is independent on the details of screening. Employing Eq. (4) and averaging $[\delta \ln Z_i(t)]^2$ with the help of Eq. (8) we get for the variance of the ZBA exponent in the limiting cases:

$$\langle [\delta \ln Z_i(t)]^2 \rangle \approx \begin{cases} \frac{0.07 \sigma_0^2}{\bar{g}_0^2} s^2, & s \lesssim 1; \\ \frac{\sigma_0^2}{8\pi^2 \bar{g}_0^2} \frac{e^{2s} \ln s}{s}, & s \gtrsim 1. \end{cases} \quad (10)$$

The low line of Eq. (10) can be rewritten as $\langle [\delta \ln Z_i(t)]^2 \rangle \approx (\ln s / 2\pi)^2 \sigma^2(s) / \bar{g}^2(s)$, indicating that fluctuations of the ZBA factors Z_i become of the order of unity simultaneously with the renormalized ratio σ/\bar{g} .

The results of numerical simulation for a model distribution $P_0(g) = \exp[-(\ln g/\bar{g}_0)^2 / 2\sigma_1^2] / (\sqrt{2\pi}\sigma_1 g)$ with the moderately small variance $\sigma_0^2 = \bar{g}_0^2 [e^{2\sigma_1^2} - e^{\sigma_1^2}] = (0.32\bar{g}_0)^2$ on the lattice 32×32 are shown in Fig. 2, where we present the distribution of the local values of $Z_i(E)$ at three values of the RG time t . Upon lowering the energy scale and approaching the MIT transition at $E \sim T_c = \bar{g}_0 E_C e^{-t_c}$ with $t_c = 0.99\bar{g}_0$, we observe a growing relative width of Z distribution with the zero- Z peak developing

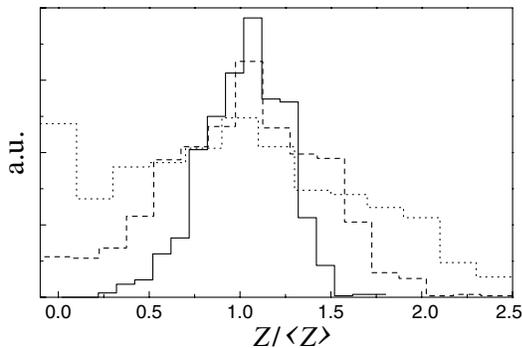


FIG. 2. Histograms for the distribution of the ZBA factors $Z(E)$ near the percolation threshold $t_c = 0.99\bar{g}_0$, obtained numerically for $\sigma_0/\bar{g}_0 = 0.32$: $t = 0.8t_c$ (solid line); $t = 0.9t_c$ (dashed line); $t = 0.95t_c$ (dotted line).

near the percolation threshold due to the considerable weight of disconnected clusters.

Effective-medium approximation (EMA).—In this approximation one takes into account only the simplest—local—correlations between g_{ij} and R_{ij} , while all distant conductances are replaced with a homogeneous medium with effective conductance g_{eff} (see, e.g., [11]). Spatial correlation is neglected within EMA, and the system at all “RG times” t is completely described by the single-conductance distribution function $P(g|t)$. While being an uncontrolled approximation, EMA provides an instrument to attack the final stage of evolution of any initial distribution—the stage with $\sigma \sim \bar{g}$. We will see that, as is typical for EMA, it works quite well except for the immediate vicinity of MIT, for determination of energy-dependent effective conductance $g_{\text{eff}}(t)$.

Within the EMA, $R_{ij} = [g_{ij} + (z/2 - 1)g_{\text{eff}}]^{-1}$ should be substituted into Eq. (2). The effective conductance is then found from the self-consistency condition [11] $\langle R_{ij}(g_{ij} - g_{\text{eff}}) \rangle_{g_{ij}} = 0$. This procedure leads to a nonlinear integral equation for the function $g_{\text{eff}}(t)$:

$$\int_0^\infty P_0(g_0) dg_0 \frac{g[g_0, \{g_{\text{eff}}\}|t] - g_{\text{eff}}(t)}{g[g_0, \{g_{\text{eff}}\}|t] + (\frac{z}{2} - 1)g_{\text{eff}}(t)} = 0. \quad (11)$$

For a general $P_0(g_0)$ this equation can be solved only numerically. If the distribution $P_0(g_0)$ is narrow, its standard deviation is given by $\sigma_{\text{EMA}}(s)/\sigma(0) = e^{-s(1-2/z)}$. At earlier stages ($s \lesssim 1$) it coincides (for the square lattice case $z = 4$) with the exact perturbation theory (7), but it deviates from it at large s , where \mathbf{p} dependence of the eigenvalue $\lambda_-(\mathbf{p})$ becomes important.

An important and physically relevant class of distributions, which allow for analytical EMA treatment, is defined by the condition that $\ln g$ is symmetrically distributed around some mean value. For all such distributions on the square lattice a simple solution for the effective conductance can be obtained: $g_{\text{eff}}(t) = \bar{g}_0 - t$ for $t < t_c = \bar{g}_0$ and $g_{\text{eff}}(t) = 0$ at $t \geq t_c$, where $\bar{g}_0 = \exp\langle \ln g_{ij}(0) \rangle$. Solutions $g_{ij}(t)$ for the individual conductances with $g_{ij}(0) > \bar{g}_0$ go above $g_{\text{eff}}(t)$ and eventually— one by one—become identical zeros at $t = t_c^{ij} > t_c$. Solutions with $g_{ij}(0) < \bar{g}_0$ go below $g_{\text{eff}}(t)$ and become zeros all at once at $t = t_c$, together with $g_{\text{eff}}(t)$. The fraction $N_{\text{cond}}(t)$ of *conducting* bonds, i.e., those having $g_{ij} > 0$ within the one-loop accuracy of Eq. (2), jumps from 0 to 1/2 at $t = t_c$ and then decreases monotonically, vanishing at $t \gg t_c$.

Clustering and percolation.—The t dependence of $g_{\text{eff}}(t)$ and $N_{\text{cond}}(t)$ is shown in Fig. 3 together with the results of numerical simulations for the initial distribution $P_0(g) = [2\bar{g}_0\theta(g)/\pi]/(g^2 + \bar{g}_0^2)$ on the square lattice 32×32 . One can see that the simulated $g_{\text{eff}}(t)$ follows the EMA in the wide range of t , while in the vicinity of the transition it clearly deviates from the EMA and ap-

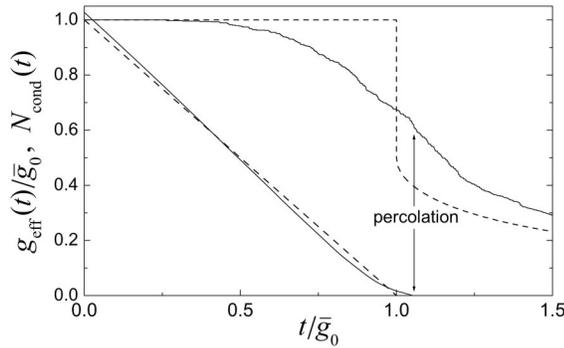


FIG. 3. Results of the numerical simulation (solid lines) and EMA (dashed lines) for the global conductivity $g_{\text{eff}}(t)$ and the fraction of conducting bonds $N_{\text{cond}}(t)$ (see text for details).

proaches zero with an exponent $\mu > 1$. The numerically found $N_{\text{cond}}(t)$ behaves, however, quite smoothly, showing no jump at $t = t_c$. The reason for this smoothness is, apparently, clusterization as demonstrated in Fig. 1 and discussed in detail in Ref. [9]. The position t_c of the percolative transition is a function of the initial distribution $P_0(g)$. In general, t_c is on the order of some mean initial conductance \bar{g}_0 , while the correct coefficient should be determined numerically.

The evolution of the network of conductances with the growth of the parameter $t(E)$ is rather similar to what would be expected at the classical percolation transition. However, our system can not be described by either purely “bond” or “site” percolation, due to development of local correlations (clustering) along with the RG flow. In particular, the numerically observed (cf. Figure 3) value of $N_{\text{cond}}(t_c)$ is clearly larger than 1/2, contrary to expectations for purely bond percolation on the square lattice. More detailed numerical work is needed to determine the nature of this new kind of percolative transition; in particular, “measurements” of the conductivity exponent μ (equal to 1.3 in the standard percolation problem [12]) would be very desirable.

Conclusions.—We have shown that at low temperatures strong intrinsic inhomogeneities are developing in granular metal arrays with moderately large random bare conductances $g_{ij} \gg 1$. As a result, the Coulomb-driven metal-insulator transition expected if $g_0 \leq \ln(E_C/\delta)$ [2,4] acquires features of percolation transition. Most directly, the predicted behavior can be detected by measuring the distribution of the local tunneling density of states at low temperatures. The best object for such a study would be a granular cermet of metal grains in the insulating matrix, like those studied in Refs. [13,14]. In these materials the ratio E_C/δ was about 1×10^3 , indicating the existence of a broad range for logarithmic corrections to conductivity. It is hardly possible that local tunnel conductances in such a granular cermet are all equal; at best, they can be distributed with the width on the order of the mean conductance. Our results, presented

in Fig. 3, show that a simple logarithmic dependence $g_{\text{eff}}(T) = \bar{g}_0 - \ln(\bar{g}_0 E_C/T)$ holds in a wide range of T for moderately random granular arrays as well, at least for the class of practically important symmetric distributions of $\log(g)$ in the 2D space.

If a granular metal has a tendency to become superconductive with $T_{\text{sc}} \sim T_c$, its local superconductive properties are expected to be strongly inhomogeneous due to position-dependent Coulomb effects. In other terms, superconductive properties of granular metal can be much more of a “granular nature” than its normal properties at elevated temperatures. In this regard we mention very interesting recent experimental results [15].

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