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Transition from insulating to non-insulating temperature dependence of the conductivity in granular metals

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Abstract. – We consider interaction effects in a granular normal metal at not very low temperatures. Assuming that all weak localization effects are suppressed by the temperature we replace the initial Hamiltonian by a proper functional of phases and study the possibility for a phase transition depending on the tunneling conductance g. It is demonstrated for any dimension that, while at small g the conductivity decays with temperature exponentially, its temperature dependence is logarithmic at large g. The formulae obtained are compared with an existing experiment and a good agreement is found.

In an experiment [1] on granular Al-Ge thick films several interesting effects have been discovered. Destroying superconductivity by a magnetic field up to 17 T the authors could study, in particular, properties of the normal state. Some features of the normal state related to a negative magnetoresistance due to superconducting fluctuations have been discussed recently [2] but an unusual observation remained completely unexplained.

What we have in mind is a peculiar temperature dependence of the conductivity found in some samples. Samples that had a high room temperature resistivity showed an exponential decay of the conductivity as a function of temperature. This behavior is typical for insulators and has been interpreted in ref. [1] in this way. Samples with larger intergranular couplings did not show any exponential decay but the resistivity did not saturate at low temperatures and the authors described its temperature dependence by a power law

$$R = AT^{-\alpha}, \tag{1}$$

with $\alpha = 0.117$. Apparently, with such a small value of α a logarithmic temperature dependence

$$R = A\,(1 - \alpha \ln T) \tag{2}$$

(obtained by expansion of eq. (1) in α) could describe the experimental data as well. It is relevant to emphasize that the array of the grains was three-dimensional and one could not attribute such a behavior to weak localization effects.

In this paper, we consider a model for granular metals at not very low temperature and demonstrate that by changing the dimensionless tunneling conductance g one can have either exponential temperature dependence of the resistivity at small $g < 1$ or the logarithmic behavior, eq. (2), at large $g > 1$. It will be shown that the result is applicable for any dimensionality of the array of grains, which contrasts usual logarithmic corrections due to interference effects [3] typical for 2D. The Hamiltonian describing the model is chosen as

$$\hat{H} = \hat{H}_0 + \hat{H}_\mathrm{t} + \hat{H}_\mathrm{c}, \tag{3}$$

where $\hat{H}_0$ is the one-electron Hamiltonian of isolated grains including disorder within the grains. The tunneling of the electrons between the grains is given by

$$\hat{H}_\mathrm{t} = \sum_{i,j,\alpha,\alpha'} t_{ij} \hat{\psi}^\dagger_{\alpha i} \hat{\psi}_{\alpha' j}, \tag{4}$$

where the summation is performed over the states α, α' of each grain (spin is conserved) and over neighboring grains i and j. The possibility to tunnel from the state α to an arbitrary state α' of other grains introduces an additional disorder resulting in a finite tunnel conductance.

The term $\hat{H}_\mathrm{c}$ in eq. (3) describes the charging energy,

$$\hat{H}_\mathrm{c} = \frac{e^2}{2} \sum_{ij} \hat{N}_i C^{-1}_{ij} \hat{N}_j. \tag{5}$$

In eq. (5), $\hat{N}_i = \sum_\alpha \int \hat{\psi}^\dagger_\alpha (\boldsymbol{r}_i) \hat{\psi}_\alpha (\boldsymbol{r}_i) \, \mathrm{d}\boldsymbol{r}_i - \bar{N}$ is the excess number of electrons in the i-th grain ($\bar{N}$ is the dimensionless local potential) and C_{ij} is the capacitance matrix. Equation (5) describes the long-range part of the Coulomb interaction in the limit of weak disorder inside the grains and has been used in many works. Calculations with the Hamiltonian $\hat{H}$, eqs. (3),(5), can be replaced by computation of a functional integral over anticommuting fields $\psi_\alpha (\tau)$.

Although the model described by eqs. (3), (5) contains only the long-range part of the Coulomb interaction, it is still very complicated, because at very low temperatures interference becomes very important and one has to consider an interplay of localization and interaction effects. One could do this either using diagrammatic expansions [3] or writing a non-linear σ-model [4]. Both methods allow to consider the limit of large tunneling conductances g and the results are strongly dependent on the dimensionality. However, the behavior, eq. (1) or eq. (2) was not predicted for 3D in any of these works.

The model, eqs. (3), (5), becomes simpler if the temperature T is not very low so that low-energy diffusion modes are damped. As was discussed in a recent publication [5], the granular metal can be described at temperatures $T \gg g\delta$, where δ is the mean level spacing in a single grain, by the Ambegaokar, Eckern and Schön (AES) [6] functional of the free energy. If $g \lesssim 1$, this condition should be replaced by $T \gg \delta$. The limit of not very low temperatures not only simplifies the consideration but is interesting on its own because it leads to an unusual behavior of physical quantities and is easily accessible experimentally. In particular, we will see that by changing the tunneling conductance g one may have a transition from the exponential temperature dependence of the resistivity to the logarithmic behavior, eq. (2).

We calculate the conductivity $\sigma (\omega)$ using the Kubo formula and making an analytical continuation from Matsubara frequencies $i\omega_n$ to real frequencies ω [7]. In order to reduce the calculation of physical quantities to a computation of correlation functions with the AES action we decouple the interaction term, eq. (5), by integration over an additional $V_i (\tau)$ and

then, following refs. [5,8], remove this field from $\hat{H}_0$ by the gauge transformation

$$\psi_i(\tau) \to e^{-i\varphi(\tau)}\psi_i(\tau), \quad \dot{\varphi}_i(\tau) = V_i(\tau). \tag{6}$$

This is not a trivial procedure, because the new fields $\psi_\alpha(\tau)$ should obey, as before, the boundary condition $\psi_\alpha(\tau) = -\psi_\alpha(\tau+\beta)$, $\beta = 1/T$. So, the field $V_i(\tau)$ cannot be completely removed from $\hat{H}_0$ at arbitrarily low temperatures. Instead of $\varphi_i(\tau)$ let us consider $\tilde{\phi}_i(\tau)$:

$$\tilde{\phi}_i(\tau) = \phi_i(\tau) + 2\pi T k_i \tau, \tag{7}$$

where $-\infty < \phi_i(\tau) < \infty$, $\phi_i(0) = \phi_i(\beta)$, and $k_i = 0, \pm1, \pm2, ...$ are the so-called winding numbers. Performing the gauge transformation with $\tilde{\phi}_i$ instead of $\varphi_i(\tau)$, the antiperiodicity of the ψ_α is preserved, but the action still contains a term linear in $i(V_i - \dot{\tilde{\phi}}_i) \in (-i\pi T, i\pi T)$. Only in the limit $T \gg \delta$ this term can be neglected. The integration over $\tilde{\phi}_i(\tau)$ implies integration over $\phi_i(\tau)$ and summation over k_i. At large $g \gg 1$, one can put all $k_i = 0$. However, at $g \lesssim 1$ one should sum over all k_i and neglecting the contribution of the non-zero winding numbers, as done in ref. [8], leads to incorrect results.

Using the phase representation one can write the conductivity $\sigma(\omega)$ in the form

$$\sigma(\omega) = \frac{ia^{d-2}}{\omega}\left[\int_0^\beta \mathrm{d}\tau e^{i\Omega_n\tau} K(\tau)\right]_{\Omega_n\to -i\omega+\delta},$$

$$K(\tau) = \langle X_2^{\boldsymbol{a}}(\tau)\rangle - \sum_{\boldsymbol{i}}\langle X_{10}^{\boldsymbol{a}}(\tau)\, X_{1\boldsymbol{i}}^{\boldsymbol{a}}(0)\rangle, \tag{8}$$

$$X_2^{\boldsymbol{a}}(\tau) = e^2\pi g\int_0^\beta \mathrm{d}\tau'\left(\delta(\tau) - \delta(\tau'-\tau)\right)\alpha(\tau')\cos\left(\tilde{\phi}_{\boldsymbol{i},\boldsymbol{i}+\boldsymbol{a}}(\tau') - \tilde{\phi}_{\boldsymbol{i},\boldsymbol{i}+\boldsymbol{a}}(0)\right),$$

$$X_{1\boldsymbol{i}}^{\boldsymbol{a}}(\tau) = e\pi g\alpha(\tau-\tau')\sin\left(\tilde{\phi}_{\boldsymbol{i},\boldsymbol{i}+\boldsymbol{a}}(\tau') - \tilde{\phi}_{\boldsymbol{i},\boldsymbol{i}+\boldsymbol{a}}(\tau)\right), \quad \alpha(\tau) = T^2\left(\mathrm{Re}\left(\sin(\pi T\tau + i\delta)\right)^{-1}\right)^2,$$

where $\boldsymbol{a}$ is a vector connecting the centers of neighboring grains $\boldsymbol{i}$ and $\boldsymbol{i}+\boldsymbol{a}$, $a = |\boldsymbol{a}|$, and d is the dimensionality of the array. In eqs. (8), $\tilde{\phi}_{\boldsymbol{ij}}(\tau) = \tilde{\phi}_{\boldsymbol{i}}(\tau) - \tilde{\phi}_{\boldsymbol{j}}(\tau)$ for $\boldsymbol{i}$ and $\boldsymbol{j}$ standing for neighboring grains and

$$\langle ... \rangle = \int (...)\exp[-S]\, D\tilde{\phi}\left(\int \exp[-S]\, D\tilde{\phi}\right)^{-1}, \tag{9}$$

where $D\tilde{\phi}$ stands for both the functional integration over $\phi(\tau)$ and summation over the winding numbers k_i. The AES action S can be written as

$$S = S_\mathrm{c} + S_\mathrm{t}, \tag{10}$$

where S_c describes the charging energy,

$$S_\mathrm{c} = \frac{1}{2e^2}\sum_{\boldsymbol{ij}}\int_0^\beta \mathrm{d}\tau C_{ij}\frac{\mathrm{d}\tilde{\phi}_i(\tau)}{\mathrm{d}\tau}\frac{\mathrm{d}\tilde{\phi}_j(\tau)}{\mathrm{d}\tau}, \tag{11}$$

and S_t stands for tunneling between the grains,

$$S_\mathrm{t} = 2\pi g\sum_{|\boldsymbol{i}-\boldsymbol{j}|=a}\int_0^\beta \mathrm{d}\tau d\tau'\alpha(\tau-\tau')\sin^2\left(\frac{\tilde{\phi}_{ij}(\tau) - \tilde{\phi}_{ij}(\tau')}{2}\right). \tag{12}$$

The dimensionless conductance g is given by $g = 2\pi\nu^2 t_{ij}^2$, where t_{ij} is the tunneling amplitude from grain i to grain j (spin is included).

Although the model described by eqs. (8)-(12) is simpler than the initial model, eqs. (3)-(5), explicit formulae can be written only in limiting cases. The same action S, eqs. (10)-(12), was used in ref. [9], and a metal-insulator transition has been predicted in a 2D array of tunnel junctions. However, the authors of ref. [9] did not calculate the conductivity but discussed properties of the partition function. For large g they did not account for phase fluctuations properly which, as we show here, are responsible for the behavior, eq. (2). Moreover, we find a transition in any dimensionality.

If the temperature T is very high, $T \gg E_c \sim e^2 C_{ij}^{-1}$, where E_c is the electrostatic energy of adding one electron to a grain, fluctuations of the phases $\tilde{\phi}$ are negligible and one can set $\tilde{\phi} = 0$ in the expressions for X_1 and X_2 in eqs. (8). Then, we obtain easily the conductivity

$$\sigma_0 = e^2 g a^{d-2}, \tag{13}$$

which shows that at such temperatures charging interactions are not important.

In the opposite limit $T \ll E_c$, transport in the granulated system has much more interesting characteristics. This inequality can be compatible with the inequality $T \gg \max\{g\delta, \delta\}$, used for the derivation of eqs. (8)-(12), because $E_c \gg \delta$ for 2D and 3D grains.

We calculate the conductivity at temperatures $T \ll E_c$ in the limits $g \gg 1$ and $g \ll 1$.

In the limit of large conductances $g \gg 1$, fluctuations of ϕ are small and all non-zero winding numbers k_i can be neglected. Non-zero k_i (as well as variations of $\bar{N}_i$) would lead to contributions of order $\exp[-g]$, and can be neglected in any expansion in $1/g$.

Keeping only terms quadratic in ϕ in eqs. (10)-(12) we reduce the action S to the form

$$S = T \sum_{\boldsymbol{q},n} \phi_{\boldsymbol{q},n} G^{-1}_{\boldsymbol{q},n} \phi_{-\boldsymbol{q},-n}, \tag{14}$$

$$G^{-1}_{\boldsymbol{q},n} = \omega_n^2 / (4E(\boldsymbol{q})) + 2g|\omega_n| \sum_{\boldsymbol{a}} (1 - \cos \boldsymbol{q}\bar{\boldsymbol{a}}),$$

where $E(\boldsymbol{q}) = e^2/(2C(\boldsymbol{q}))$ and $C(\boldsymbol{q})$ is the Fourier-transform of the capacitance matrix C_{ij} ($\boldsymbol{q}$ are quasi-momenta for the array of the grains). One should sum in eq. (14) over d unit lattice vectors $\bar{\boldsymbol{a}}$, where d is the dimensionality of the array.

Keeping only terms quadratic in ϕ in the action but not expanding the function X_2, eqs. (8), one reduces the correlator $\langle X_{2\boldsymbol{a}}(\Omega_n)\rangle$ to the form

$$\langle X_{2\boldsymbol{a}}(\Omega_n)\rangle = \pi e^2 g \int_0^\beta \alpha(\tau)\left(1 - e^{i\Omega_n \tau}\right) e^{-\tilde{G}_{\boldsymbol{a}}(\tau)} \mathrm{d}\tau,$$

$$\tilde{G}_{\boldsymbol{a}}(\tau) = 4T \int \frac{\mathrm{d}\boldsymbol{q}}{(2\pi)^d} G_{\boldsymbol{q}n} \sin^2 \frac{\boldsymbol{q}\bar{\boldsymbol{a}}}{2} \sin^2 \frac{\omega_n \tau}{2}. \tag{15}$$

One can check that the contribution coming from the correlator of the functions X_1 in eqs. (8) contains additional powers of $1/g$ and can be neglected in the main approximation. It is very important that the correlator $\langle X_1 X_1\rangle$ in eqs. (8) contains a summation over j, which corresponds to the zero quasi-momentum of the function K. If we carried out the computation for a single grain the contribution from $\langle X_1 X_1\rangle$ would not be smaller than the one from $\langle X_2\rangle$.

What remains to be done in order to calculate the conductivity for $g \gg 1$ is to compute the integral in eq. (15) for the Matsubara frequencies Ω_n and make the analytical continuation $\Omega_n \to -i\omega + \delta$. In the lowest order in α the result for the conductivity σ in the limit $\omega \to 0$ is

$$\sigma = \sigma_0 (1 - \alpha \ln(gE_c/T)), \quad \alpha = (2\pi g d)^{-1}. \tag{16}$$

Thus, at large $g \gg 1$ the conductivity decays with temperature logarithmically. Of course, one may not use this formula at very low temperature because the present consideration is valid at not very low temperatures $T \gg g\delta$ when the AES action may be used. At lower temperatures, one should take into account interference effects and, depending on the dimensionality d of the array, both metal and insulating states are possible. In contrast, eq. (16) is valid in *any* dimensionality.

If we used eq. (15) exactly we would obtain the power law, eq. (1), with the exponent α from eq. (16). A similar dependence was written for the voltage dependence of the conductance of a single junction in a model with an electromagnetic environment [10,11]. However, taking into account non-quadratic terms in the expansion of the action in phases ϕ changes this result and one comes again to eq. (16). For a single junction this result is known since the works of ref. [12] where a proper renormalization group (RG) equation was written. One can check that, in the first order of the RG, the equation is the same for a granular metal and does not depend on the arrangement of the grains in the array. It can be written as

$$\frac{\mathrm{d}g\left(\xi\right)}{\mathrm{d}\xi} = -\frac{1}{2\pi d}, \tag{17}$$

where $\xi = -\ln\tau$ and $g\left(\xi\right)$ is the effective conductance.

Solving eq. (17) with the boundary condition $g\left(0\right) = g$ we come immediately to eq. (16).

However, next orders in the RG (expansion of the Gell-Mann-Low function in $g\left(\xi\right)$) are dependent on the type of the array and differ from the corresponding terms for a single junction. The applicability of the one-loop approximation, eq. (17), and of its solution, eq. (16), implies an additional inequality for the temperature: $T \geq T_{\mathrm{c}} = gE_{\mathrm{c}}\exp[-1/\alpha]$. In other words, eq. (16) remains valid until σ/e^2 becomes of the order unity. For not very large grains this inequality is less restrictive because T_{c} is exponentially small for large g. The dimensionality of the array d enters eqs. (16), (17) as a parameter only. Actually, the number of contacts with neighboring grains (coordination number) rather than the dimensionality itself enters eqs. (16), (17). This difference may be important in situations when the grains are close packed in a cubic lattice.

The logarithmic behavior, eq. (16), describes the granular system at sufficiently large $g \gtrsim 1$. At smaller g, the temperature dependence becomes exponential and we check this statement in the limit $g \ll 1$ expanding the functional integral in eq. (9) in the tunneling part S_{t}, eq. (12), of the action. The main contribution comes again from the function $\langle X_2\left(\tau\right)\rangle$ in eqs. (8). In the lowest order one can completely neglect S_{t}, which leads to computation of the correlator $\Pi\left(\tau\right)$:

$$\Pi\left(\tau\right) = \left\langle \exp\left[-i\left(\tilde{\phi}_i\left(\tau\right) - \tilde{\phi}_i\left(0\right)\right)\right]\right\rangle_{S_{\mathrm{c}}}, \tag{18}$$

where the phases $\tilde{\phi}_i\left(\tau\right)$ are introduced in eq. (7), S_{c} is given by eq. (11) and the averaging should be performed with this functional. (Strictly speaking, the function $\Pi\left(\tau\right)$ is sufficient for calculating $K\left(\tau\right)$ only for diagonal C_{ij}. However, a proper modification for an arbitrary C_{ij} is simple.) The computation of the average in eq. (18) can be performed using two different methods. A more straightforward way of calculating is to use the definition of $\tilde{\phi}_i\left(\tau\right)$, eq. (7), which allows to represent the action S_{c} as $S_{\mathrm{c}} = S_{\mathrm{c}}[\phi] + S_{\mathrm{c}}[k]$ and to carry out integration over the phase ϕ and summation over the winding numbers separately. Integrating over the phase $\phi_i\left(\tau\right)$ we obtain for $0 < \tau < \beta$ (see also [8,9])

$$\langle e^{-i(\phi_i(\tau)-\phi_i(0))}\rangle = \exp\left[-B_{ii}\left(\tau - T\tau^2\right)\right], \tag{19}$$

where $B_{ij} = \frac{e^2}{2}\left(C^{-1}\right)_{ij}$.

However, eq. (19) is not the final result and the summation of the winding numbers is essential. This can be performed using the Poisson summation formula. As a result, the function $\Pi(\tau)$ can be represented as

$$\Pi(\tau) = \frac{1}{Z} e^{-B_{ii}\tau} \sum_{\{n_k\}} e^{-\sum_k 2\tau n_k B_{ki} - \beta \sum_{k,l} B_{kl} n_k n_l}, \tag{20}$$

where all n_k are integers and Z is a normalization coefficient ($\Pi(0) = 1$). The necessary periodicity in τ of the function $\Pi(\tau)$ with the period β is evident from eq. (20).

The second method is to use the standard quantum-mechanical formalism instead of calculating the functional integrals in eq. (18), which has been suggested in an earlier work on granulated superconductors [13]. Within this approach one writes instead of the action S_c, eq. (11), an effective Hamiltonian $\hat{H}_{\text{eff}}$,

$$\hat{H}_{\text{eff}} = \sum_{ij} B_{ij} \hat{\rho}_i \hat{\rho}_j, \qquad \hat{\rho}_i = -i\partial/\partial\phi_i, \tag{21}$$

and calculates the thermodynamic average with $\hat{H}_{\text{eff}}$. For the function $\Pi(\tau)$ one should calculate the average

$$\langle e^{-i\left(\hat{\phi}_i(\tau) - \hat{\phi}_i(0)\right)} \rangle_{\hat{H}_{\text{eff}}}, \qquad \hat{\phi}_i(\tau) = e^{\hat{H}_{\text{eff}}\tau} \phi_i e^{-\hat{H}_{\text{eff}}\tau}.$$

Eigenvalues of the operators $\hat{\rho}_i$ are integers (eigenfunctions of H_{eff} must be periodic in ϕ with period 2π) and one comes easily to eq. (20). This consideration explicitly demonstrates that accounting for the winding numbers leads to the charge quantization.

We see from eq. (20) that, in order to get an explicit expression for the conductivity, one should sum over all configurations of charge. At high temperatures $T \gg E_c$ the sum over n_k in eq. (20) can be replaced by integrals and we get $\Pi(\tau) = 1$, which leads to eq. (13).

In the opposite limit, $T \ll E_c$, the main contribution comes from charge configurations with the lowest energy. The ground state with all $n_k = 0$ does not contribute to the conductivity. If the lowest excited state corresponds to one charged grain with charge ± 1 (depending on a particular C_{ij}), we come, using eqs. (8) in the limit $\omega \to 0$, to the rather simple formula

$$\sigma = 2\sigma_0 \exp\left[-B_{ii}/T\right]. \tag{22}$$

Since B_{ii} is the energy corresponding to the charge ± 1, eq. (22) corresponds to conduction of an activated electron and hole (the factor 2 means that both of them are taken into account).

Comparing eq. (22) with eq. (16) we come to the conclusion that there must be a critical value g_c separating in the limit $T \to 0$ the logarithmic behavior from the exponential one. Whether the activation energy (Coulomb gap) turns to zero or has a jump at $g = g_c$ is not clear from the present consideration. (Strictly speaking, there should not be any singularity of the conductivity at finite temperatures but the change of the behavior may be noticeable experimentally or numerically.) The model of the granular metal may be used to describe disordered electron systems at low electron density. In this case, potential wells would correspond to the grains.

The sample of the experiment [1] that showed the "power law behavior", eq. (1), had the room temperature resistivity $R_0 = 7.3 \times 10^{-3}\ \Omega\,\text{cm}$. The diameter of the grains was 120 ± 20 Å, which allows, using the value $\hbar/e^2 = 4.1 \times 10^3\ \Omega$, to estimate the dimensionless tunnel conductivity as $g = 0.7$. If we put $d = 2$ in eq. (16) we obtain $\alpha = 0.116$, which exactly corresponds to the experimental value from eq. (1). However, the arrays used in

ref. [1] were rather thick and, at first glance, one should use $d = 3$. Nevertheless, the value of d in eq. (16) corresponds rather to the half of the contacts of a single grain than to the real dimensionality. Then, the experimental value of α indicates that either the grains are not closely packed so that the typical number of contacts per grain is 4 or our calculation is too rough to provide a quantitative agreement with the experiment (the value of α, eq. (16), is based on the assumption $g \gg 1$ but the experimental value of g is of order 1). The resistivity of samples with a high room temperature resistivity behaved as $\exp\left[a/T^{1/2}\right]$ rather than obeying the activation law, eq. (22). But this can be attributed to a variation of the size of the grains or of the local potential [14].

In conclusion, we suggested a scheme of calculating the conductivity of a granular metal at not very low temperatures. On the basis of explicit results we demonstrated the existence in any dimensionality of a transition between states with exponential dependence of the conductivity on temperature and a logarithmic one. Relating the coefficient α to the room temperature conductivity we were able to compare our results with an existing experiment and got a good agreement. The model of the granular metal may also serve as a good description of disordered systems with a low electron density.

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