

Variable range cotunneling and conductivity of a granular metal

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The Efros-Shklovskii (E-S) law for the conductivity of granular metals is interpreted as a result of a variable range cotunneling process. The cotunneling between distant resonant grains is predominantly elastic at low $T \leq T_c$, while it is inelastic (i.e., accompanied by creation of electron-hole pairs on a string of intermediate non-resonant grains) at $T \geq T_c$. The corresponding E-S temperature T_{ES} in the latter case is slightly (logarithmically) T -dependent. The magnetoresistance in the two cases is different: it may be relatively strong and negative at $T \ll T_c$, while at $T > T_c$ it is suppressed due to inelastic processes which destroy the interference.

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Introduction. The low-temperature conductivity of most granular metals (both three-dimensional samples and thin films) exhibits a typical insulating behavior, characterized by the Efros-Shklovskii (E-S) law

$$\sigma \sim e^{-\sqrt{T_{ES}/T}}. \quad (1)$$

In the samples with low room-temperature conductivity this law is observed in the whole range of T (from room temperature down to liquid helium temperatures) [1–3]. Two ingredients are known to be necessary [4, 5] for the existence of the behavior (1) in usual doped semiconductors with localized impurity centers: (i) soft “Coulomb gap” in the electron density of states, and (ii) long-range electron tunnelling between distant centers of their localization. The original idea on the building of the Coulomb gap as presented in Ref.[4] was recently adapted to granular arrays in Ref. [6]. It was argued that the principal source for this gap is random background charges and the physical mechanisms behind these charges were discussed in detail. The feature (ii) is quite natural for doped semiconductors as it follows from exponential decay of wave-functions of localized electrons. It is much less trivial for granular media where each grain is typically connected by tunnel junctions to its nearest neighbors only, therefore the very origin of long-range tunnelling needs some special explanation.

In this Letter we demonstrate that the variable range hopping in a granular metal involves the so called cotunneling process [7] (either elastic or inelastic). The elastic cotunneling is effective, if the temperature is low enough (namely, for $T < T_c \sim \mathcal{L}^{-1}(\delta \cdot E_C)^{1/2}$, where δ is the characteristic level spacing in the grains, E_C is

the characteristic charging energy, and $\mathcal{L} \sim 10$ is a large logarithmic factor, see below). At $T > T_c$ the conductivity is dominated by inelastic cotunneling processes. We directly show this for the case of the granular metal with poor room-temperature conductivity (small inter-grain conductances $g \equiv (h/e^2 R) \ll 1$). We expect that the same is also true for samples with moderately good conductivity (cf. e.g. [8, 9]) with properly renormalized T_{ES} , but this more delicate issue will be discussed in a separate publication. Experimentally, relative role of elastic vs inelastic cotunnelling processes could be detected by the presence of noticeable low-field magnetoresistance in the hopping regime: while elastic cotunnelling is expected to lead to negative magnetoresistance like it was predicted for doped semiconductors [10–13], inelastic cotunnelling is intrinsically incoherent and the whole effect of magnetic field upon conductivity is localized within individual grains and can, therefore, be only observed in very high fields $H \gtrsim 10$ T.

Co-tunnelling as a key mechanism of low-temperature charge transfer was proposed [7] and extensively discussed (cf. e.g. [14] for the review) with regard to transport via quantum dots. Quantum dot situated between two bulk metal reservoirs is characterized by its charging energy E_C and dimensionless conductances $g_{R,L}$. Semiclassical “orthodox theory” of Coulomb blockade [15] predicts exponential suppression of conductance through quantum dot at temperatures $T \ll E_C$, i.e. $G_{\text{orth}} \propto \min(g_L, g_R) \exp(-E_C/T)$, due to low probability of creation of real state of the dot with extra electron charge. Co-tunnelling process, on the other hand, occurs in the next order of perturbation theory in (small) tunnelling amplitudes $t_{R,L}$, but does not contain exponential suppression factor $\exp(-E_C/T)$ since the dot state with extra charge

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occurs as *virtual state* only. One should distinguish two kinds of co-tunnelling processes, elastic and inelastic ones. Elastic process occurs when tunnelling *in* and *out* of the dot deals with the same intra-dot electron eigenstate α , thus it leaves the dot in exactly the same quantum state as it was before it. On the contrary, inelastic co-tunnelling leaves behind it an excited electron-hole pair (since one electron tunnels *in* the dot and populates some eigenstate h , whereas another electron tunnels *out* of the dot from another, p -th eigenstate). Elastic co-tunnelling contribution to the conductance scales as $G_{el} \propto g_L g_R \delta / E_C$, whereas inelastic co-tunnelling contribution is $G_{inel} \propto g_L g_R (T/E_C)^2$, cf. [7, 14]. Thus, upon temperature decrease first the inelastic co-tunnelling prevails over classical conduction given by G_{orth} , and then at $T_* \sim \sqrt{E_C \delta}$ it gives way to the elastic co-tunnelling. Below we generalize the above ideas to the situation of variable-range hopping in granular arrays.

In the spirit of the standard variable range hopping (VRH) theory, we consider a charge transfer between two distant grains i and j with anomalously small energies $\varepsilon_i, \varepsilon_j \ll E_C$ of the “charged ground state” (i.e. the states with an extra electron, or an extra hole; for explicit definition of the energy ε and detailed discussion of the corresponding “density of ground states” see [6]). Such a transfer between distant grains proceeds via a string of intermediate grains, where typically the energy of a state with an extra electron is high ($\varepsilon \sim E_C$). As in the case of a standard single-particle tunneling via resonant impurity states, the entire process is realized as a coherent sequence of local hops between adjacent grains in the string. There is an important difference, however. For the single-particle problem, where *the same* electron has to tunnel sequentially through all the impurities in the string, (starting from the first one and ending with the last), the order of these local hops is fixed. In our case there are many electrons in each grain, all these electrons being ready to tunnel to an adjacent grain at any time, so that the sequence of local hops in such a co-tunneling process can be *arbitrary*. As a result, the states with a number of excited electrons and holes on different grains appear as intermediate virtual states of the co-tunneling process, and the number of charged grains in these intermediate states can be larger than one. In the final state, however, all the charges of the intermediate grains should be compensated, and the only extra charge is transferred between the two terminal grains of the string. Long-range hopping process involves many intermediate grains; in general co-tunnelling through some of them will be of elastic type, whereas some other

will be inelastic. We will see below that elastic cotunneling dominates the variable-range hopping at rather low temperatures $T < T_c$, where T_c is significantly lower than in the case of single intermediate quantum dot.

Note also that in the case of not extremely low temperatures $T \gg \delta$ (when the spectrum of electrons in grains can be treated as a quasicontinuous one), one does not need to invoke phonons to ensure the energy conservation: the energy can be taken from the fermionic thermostat via inelastic cotunneling process.

General approach to variable-range cotunneling. The Hamiltonian of the system has the form $\hat{H} = \hat{H}_0 + \hat{H}_{tun} + \hat{H}_C$. Here the “single-grain” hamiltonian $\hat{H}_0 = \sum_i \hat{H}_0^{(i)}$, and the inter-grain tunneling hamiltonian $\hat{H}_{tun} = \sum_{\langle ij \rangle} \hat{H}_{tun}^{(ij)}$, in the latter the summation runs over the pairs $\langle ij \rangle$ of neighboring grains,

$$\hat{H}_0^{(i)} = \sum_{\alpha_i, \sigma} \epsilon_{\alpha_i} a_{\alpha_i, \sigma}^+ a_{\alpha_i, \sigma}, \quad \hat{H}_{tun}^{(ij)} = \sum_{\alpha_i, \alpha_j, \sigma} t_{\alpha_i, \alpha_j} a_{\alpha_i, \sigma}^+ a_{\alpha_j, \sigma},$$

where the operator $a_{\alpha_i, \sigma}^+$ creates an electron in a single-particle orbital eigenstate α_i with a spin projection σ on a grain i . The Coulomb interaction hamiltonian

$$\hat{H}_C = \frac{1}{2} \sum_{ij} E_c^{(ij)} (\hat{n}_i - q_i) (\hat{n}_j - q_j), \quad (2)$$

$\hat{n}_i = \sum_{\alpha_i, \sigma} a_{\alpha_i, \sigma}^+ a_{\alpha_i, \sigma} - n_i^{(0)}$ being the operator of excess number of electrons at the grain i , $n_i^{(0)}$ corresponds to the minimum of $\hat{H}_0 + \hat{H}_C$. Variables q_i (not necessarily integer!) are the so called background charges (in the units of e). We will treat them as independent continuous random variables $-1/2 < q_i < 1/2$ with symmetric distribution $P(q)$. Two different limits should be distinguished: (i) the case of strong charge disorder, when the background charges are large, so that q_i is distributed homogeneously in the interval $-1/2 < q_i < 1/2$; and (ii) the case of weak disorder, when the charges are small, so that $q_0 \equiv (\overline{q^2})^{1/2} \ll 1$ and the probability to have $q = 1/2$, related to the “bare density of ground states” at the Fermi-level (cf. [6]), is small: $P_{1/2} \equiv P(1/2) \ll 1$. While the former case seems to be most appropriate for a naturally disordered granular material, the latter one may be relevant for high quality artificial arrays of quantum dots.

If the tunnel matrix elements t_{α_i, α_j} are small enough, the rate w_{ij} of cotunneling between distant grains i and j can be found in the high order perturbation theory in \hat{H}_{tun} :

$$w_{ij} = 2\pi f_F(\varepsilon_i) f_F(-\varepsilon_j) \sum_{\mathcal{P}} \sum_{\sigma_0, \{\sigma_l\}} \sum_{\{p_l, h_l\}} \sum_{\{\alpha_m\}} \prod_{k=1}^{N+1} |t_{h_k p_{k-1}}|^2 f_F(\varepsilon_{p_0}) f_F(-\varepsilon_{h_{N+1}}) \prod_l f_F(\varepsilon_{p_l}) f_F(-\varepsilon_{h_l}) \times \\ \times \delta \left[\varepsilon_{p_0} - \varepsilon_{h_{N+1}} + \sum_l (\varepsilon_{p_l} - \varepsilon_{h_l}) - \Delta_{ij} \right] \sum_{\mathcal{T}, \mathcal{T}'} (-1)^{K(\mathcal{T}) - K(\mathcal{T}')} \prod_m \delta_{\lambda_m(\mathcal{T}) \lambda_m(\mathcal{T}')} f_F[\lambda_m(\mathcal{T}) \varepsilon_{\alpha_m}] Q(\mathcal{T}) Q(\mathcal{T}'). \quad (3)$$

Here we have assumed that the $i \rightarrow j$ transition is dominated by tunneling along a unique “string” – a chain of neighboring grains, denoted by numbers $k = 0, 1, \dots, N, N+1$, so that $0 \equiv i$ is the initial grain; $N+1 \equiv j$ is the final grain; each pair $k, k+1$ are in contact. The possibility for several relevant strings to exist, and the effect of interference of their contributions will be discussed in the last section of this Letter. The summation runs over all possible partitions \mathcal{P} of the string into two subsets: $\{k\} = \{m\} \cup \{l\}$; on the grains $\{m\}$ the elastic cotunneling (via a state $h_m, p_m \equiv \alpha_m$) occurs, while at grains $\{l\}$ the inelastic cotunneling with a creation of an electron-hole pair with quantum numbers h_l, p_l takes place. The summation over all eigenstates α_m and pairs of eigenstates $h_l \neq p_l$ is assumed. The energies $\varepsilon_{p_l}, \varepsilon_{h_l}$ are measured with respect to the Fermi level. The spin-variable σ_0 corresponds to the state p_0 ; variables σ_l correspond to p_l -components of electron-hole pairs. All other spin-variables are not independent because of the spin-conservation by tunneling hamiltonian. The spin summation gives the factor $2^{L(\mathcal{P})}$, where $L(\mathcal{P})$ is a number of inelastic grains in the partition \mathcal{P} . The interference cross-terms between the processes with different α_m are neglected because of violent sign-fluctuations of $t_{h_k p_{k-1}}$.

The “time-orderings” \mathcal{T} are all the possible orderings $\{k_1, k_2, \dots, k_r, \dots, k_{N+1}\}$ of the set of grains $k = 0, \dots, N$ (note, that there is no final grain $N+1$ in this set!). The contribution to the composite transition amplitude $\hat{H}_{\text{eff}}^{(ij)}$ corresponding to particular \mathcal{T} has the structure

$$\hat{H}_{\text{eff}}^{(ij)}(\mathcal{T}) = \hat{H}_{\text{tun}}^{(k_{N+1} k_{N+1+1})} \hat{G} \hat{H}_{\text{tun}}^{(k_N k_{N+1})} \hat{G} \dots \hat{G} \hat{H}_{\text{tun}}^{(k_1 k_1+1)},$$

where the many-particle Green-function $\hat{G} = [\hat{H}_0 + \hat{H}_C]^{-1}$. The function $\lambda_k(\mathcal{T}) = 1$, if in the ordering \mathcal{T} the grain $k-1$ comes earlier, than the grain k , and $\lambda_k(\mathcal{T}) = -1$ otherwise. The sign-factor $(-1)^{K(\mathcal{T})}$ arises due to permutations of fermionic operators. The Fermi-functions $f_F(\varepsilon)$ take into account the Fermi filling factors. The δ -function ensures the energy conservation, and $\Delta_{ij} = \varepsilon_j - \varepsilon_i - E_c^{(ij)}$ is the difference of energies of the initial and final state. The factors

$$Q(\mathcal{T}) = \prod_{r=1}^N \left[H_C \{n(r)\} + \sum_{r'=1}^r (\varepsilon_{h_{k_{r'+1}}} - \varepsilon_{p_{k_{r'}}}) \right]^{-1} \quad (4)$$

are products of energy denominators, appearing in \hat{G} . Here n_k are numbers of excess electrons on k -th grain after p local hops; they can be found from the following recursion formula:

$$n_k(r) = \begin{cases} n_k(r-1) - 1, & \text{if } k = k_r, \\ n_k(r-1) + 1, & \text{if } k = k_r + 1, \\ n_k(r-1), & \text{otherwise,} \end{cases} \quad (5)$$

while $n_k(0) = n_k^{(0)}$ is the equilibrium distribution.

Inspecting the expression (3) we see that the characteristic value $\bar{\varepsilon}_{\text{inel}}$ of the “inelastic energies” $\varepsilon_{p_l}, \varepsilon_{h_l}$ is controlled by the combination of the δ -function in (3) and the product of the corresponding Fermi-functions. As a result, $\bar{\varepsilon}_{\text{inel}} \sim \Delta_{ij}/\bar{L}$, where \bar{L} (which is T -dependent) is the number of inelastic cotunneling events in the total process. Actually $T \ll \bar{\varepsilon}_{\text{inel}} \ll E_C$, so that, in particular, the dependence of $Q(\mathcal{T})$ on the inelastic energies can be neglected. On the other hand, the characteristic value of $\bar{\varepsilon}_{\text{el}}$ of the “elastic energies” ε_{α_m} is limited only by the energy denominators $Q(\mathcal{T})$, so that $\bar{\varepsilon}_{\text{el}} \sim E_C$.

Thus, performing the integration over $\varepsilon_{p_l}, \varepsilon_{h_l}$ and ε_{α_m} , we obtain for the effective Miller-Abrahams dimensionless conductance g_{ij} between two distant grains i and j :

$$g_{ij} \propto e^{-\frac{\varepsilon_{ij}}{T}} \left(\frac{t}{E_C} \right)^{2N} \times \\ \times \sum_{L=0}^N \frac{\left(\frac{2|\Delta_{ij}|^2}{\delta^2} \right)^L \left(\frac{E_C}{\delta} \right)^{N-L}}{(2L+1)!} F_{NL}. \quad (6)$$

Here $\delta = E_0 e^{-\ln(E_0 \nu \nu)}$ is the mean level spacing (ν is the electronic density of states per one spin projection at the Fermi level in a particular grain, ν is the grains

volume, and E_0 is an arbitrary energy unit). The characteristic Coulomb energy $E_C = E_0 e^{\ln(E_{kk}/E_0)}$ (normally $E_C \sim e^2/\kappa_{\text{eff}}a$, where a is the average diameter of grains, and κ_{eff} is the effective dielectric permeability of the material, see [6]). Finally, the mean tunneling amplitude $t = E_0 e^{1/2 \ln(|t_{kk+1}|^2/E_0^2)}$, where $|t_{kk+1}|^2$ is the "coarse grained" (i.e., averaged over an interval of energies near the Fermi level, large compared to the level spacing, but small, compared to any other relevant scale) value of $|t_{\alpha_k, \alpha'_{k+1}}|^2$. The "averaging of the logarithm" rule appearing in the above definitions of mean values, arises as a result of self-averaging of large ($\sim N$) number of similar independent random factors with identical distributions. Note also the presence of the spin-factors 2 in the multipliers, corresponding to inelastic processes and absence of such factors for the elastic processes.

The local activation energy ε_{ij} for the ij hop is the combination of $\varepsilon_i, \varepsilon_j$ and $E_C^{(ij)}$, standard for the hopping conductivity theory (see [5] for the explicit definition).

The weight-function $F_{NL} = \sum_{\mathcal{P}} \delta_{L, L(\mathcal{P})} C(\mathcal{P})$, where $C(\mathcal{P})$ are numerical coefficients, depending only on the partition \mathcal{P} and on the explicit form of the charging energy matrix $E_C^{(kk')}$:

$$C(\mathcal{P}) = \sum_{\mathcal{T}, \mathcal{T}'} (-1)^{K(\mathcal{T}) - K(\mathcal{T}')} \times \int \prod_m d\bar{\varepsilon}_m \delta_{\lambda_m(\mathcal{T}) \lambda_m(\mathcal{T}')} \theta[-\lambda_m(\mathcal{T}) \bar{\varepsilon}_{\alpha_m}] \bar{Q}(\mathcal{T}) \bar{Q}(\mathcal{T}'), \quad (7)$$

$\bar{\varepsilon}_m \equiv \varepsilon_m/E_C$ and $\bar{Q}(\mathcal{T}) \equiv Q(\mathcal{T})E_C^N$ being the dimensionless variables.

The explicit form of the weight-function $F_{N,L}$ for general $E_C^{(kk')}$ can not be found. However, as it is argued below, the asymptotics of F , relevant for the purely elastic and purely inelastic limits, are $F_{N,0} \approx \bar{A}_1^N$ and $F_{N,N} \approx \bar{A}_2^N$, correspondingly. The numerical constants \bar{A}_1 and \bar{A}_2 are not known. As a result,

$$g_{ij} \propto \exp\left\{-\frac{\varepsilon_{ij}}{T}\right\} \begin{cases} \left(\frac{\bar{A}_1 g \delta}{8\pi^2 E_C}\right)^{N_{ij}}, & \text{elastic,} \\ \left(\frac{e^2 \bar{A}_2 g |\Delta_{ij}|^2}{16\pi^2 N_{ij}^2 E_C^2}\right)^{N_{ij}}, & \text{inelastic,} \end{cases} \quad (8)$$

where $g \equiv Gh/e^2 = 8\pi^2(t/\delta)^2 \ll 1$ is the average dimensionless conductance of a contact between two adjacent grains. Note, that this definition of g differs from that in Refs.[8, 9]. Applying standard Mott-Efros-Shklovskii arguments to the random network with conductances (8), we obtain Eq. (1) with $T_{ES} = \mathcal{L}(T)E_C$

$$\mathcal{L}(T) = \begin{cases} c_1 \ln\left(\frac{8\pi^2 E_C}{\bar{A}_1 g \delta}\right), & T \ll T_c, \\ c_1 \ln\left(\frac{16\pi^2 E_C^2}{e^2 \bar{A}_2 g T^2 \mathcal{L}^2}\right), & T \gg T_c, \end{cases} \quad (9)$$

where the crossover temperature $T_c \sim \sqrt{E_C \delta}/\mathcal{L}$, and $c_1 \sim 1$ is an unknown constant, depending on the statistical geometry of the granular material. Since $E_C \propto a^{-1}$ and $\delta \propto a^{-3}$, we conclude that roughly $T_{ES} \propto a^{-1}$ and $T_c \propto a^{-2}$.

It should be noted that the above consideration is justified and the VRH regime is actual only if the characteristic length of the hop is large: $\bar{N} \sim (E_C/\mathcal{L}T)^{1/2} \gg 1$. For $\bar{N} < 1$ the Nearest Neighbor Hopping regime, characterized by the Arrhenius law $\sigma \propto \exp(-E_A/T)$ with the activation energy $E_A \sim E_C$ should be observed. The crossover temperature between the two regimes is controlled by the intergrain conductance g ; the NNH is likely to be found in the samples with very low g .

Model of local repulsion: Mott law for granular array. The expression (7) can be explicitly evaluated for the model case of the short range Coulomb interaction $E_C^{(kk')} = E_k \delta_{kk'}$. Then dimensionless local energies of the charged states are $\bar{E}_k^{(\pm)} = (E_k/E_C)[1/2 \mp q_k]$, and $\varepsilon_i = E_k(\frac{1}{2} - |q_i|) \text{sign}(q_i)$. In this case (7) can be written as a product of single particle Green-functions with energies, depending on the local charge. As a result

$$C(\mathcal{P}) = \prod_m \left(\frac{1}{\bar{E}_m^{(+)} + \frac{1}{\bar{E}_m^{(-)}}}\right) \prod_l \left(\frac{1}{\bar{E}_l^{(+)} - \frac{1}{\bar{E}_l^{(-)}}}\right)^2,$$

so that, for $N \gg 1$, when the number of similar factors in (10) is large and an effective self-averaging takes place,

$$F_{NL} = C_N^L A_1^{N-L} A_2^L, \quad (10)$$

$$A_1 \equiv e^{-\overline{\ln(1/4 - q^2)}}, \quad A_2 \equiv 4e^{\overline{\ln q^2 - 2\overline{\ln(1/4 - q^2)}}}, \quad (11)$$

C_N^L being binomial coefficients. The constant A_1 does not show any dramatic dependence on the strength of the random potentials: $A_1 = e^2 \approx 7.4$ for strong charge disorder, and $A_1 = 4$ for a weak one. The constant $A_2 = e^2$ for a strong disorder, while for a weak one $A_1 \sim q_0^2 \ll 1$. The reason is the destructive interference between two possible processes of the pair production: in the "e-h-process" the electron is created first and the hole is created the second, while in the "h-e-process" the order is inverted. As a result, for $q_0 \ll 1$ the crossover temperature strongly depends on q_0 : $T_c(q_0) \sim (q_0 \mathcal{L})^{-1} \sqrt{E_C \delta}$. The growth of T_c is saturated at $T_c^{\text{max}} \sim (\mathcal{L})^{-1} (E_C^3 \delta)^{1/4}$ for $q_0 \lesssim (\delta/E_C)^{1/4}$, when the energies $\varepsilon_{pl}, \varepsilon_{hl}$ of the pairs

come into play. Thus, we conclude that for the case of weak charge disorder the inelastic cotunneling is suppressed and the crossover between elastic and inelastic cotunneling is shifted to higher temperatures.

Unfortunately, the result (10) can not be generalized for the case of nonlocal interaction $E_c^{(ij)}$. It can be shown, however, that $F_{N,0} \approx \bar{A}_1^N$, and $F_{N,N} \approx \bar{A}_2^N$, with certain renormalized constants \bar{A}_1 and \bar{A}_2 . Roughly, the reason is as follows (details will be presented elsewhere): simple exponential form of the F -function holds for any "effectively short-range" interaction (not necessarily strictly local one), while for effectively long-range one the functional form of F can be changed dramatically. The clue is that, despite the long-range character of the Coulomb potential, the interaction of effective degrees of freedom in our case is the short-range one. Indeed, actual charge configurations, relevant to our problem, are those, generated by local electronic hops between neighboring grains. These hops create *local dipoles*, and the dipole-dipole interaction decays with distance r as r^{-3} .

With the explicit formula (10) at hand one can perform the summation in (6) and find

$$\ln g_{ij} = N_{ij} \left\{ \ln \left(\frac{A_1 g \delta}{8\pi^2 E_C} \right) + \varphi \left(\frac{|\varepsilon_i - \varepsilon_j|}{2N_{ij}} \sqrt{\frac{2A_2}{A_1 E_C \delta}} \right) \right\} - \frac{\varepsilon_{ij}}{T},$$

where the function $\varphi(z)$ is implicitly defined by the relations $\varphi(z) = 2y - \ln(1-y)$, $y^3 = (1-y)z^2$. The function $y(z) \equiv \bar{L}/N$ (with asymptotics $y(z) \approx z^{2/3}$ at $z \ll 1$) has the meaning of a relative fraction of inelastic events.

The average number of grains N_{ij} in a string, connecting two distant grains, is proportional to the distance r_{ij} between them: $N_{ij} = c_2 n_g^{1/d} r_{ij}$, where n_g is the concentration of grains, and $c_2 \sim 1$ is a geometric constant, depending only on the statistics of grains packing. Estimates, made for c_2 for several two-dimensional models show that $c_2 \approx 1$. Thus, we have arrived at the $d+1$ -dimensional percolation problem in the \mathbf{r}_i and ε_i space. The density of sites $\nu_{d+1} = n_g P_{1/2}/E_C$ in this space is the density of marginal grains, whose ground states are almost degenerate. The connectivity criterion reads

$$\begin{aligned} & \xi(\mathbf{r}_i, \varepsilon_i | \mathbf{r}_j, \varepsilon_j) < \xi, \quad \text{where} \\ & \xi(\mathbf{r}_i, \varepsilon_i | \mathbf{r}_j, \varepsilon_j) = \frac{\varepsilon_{ij}}{T} + c_2 n_g^{1/d} \times \\ & \times r_{ij} \left\{ \ln \left(\frac{8\pi^2 E_C}{A_1 g \delta} \right) - \varphi \left(\frac{|\varepsilon_i - \varepsilon_j|}{2c_2 n_g^{1/d} r_{ij}} \sqrt{\frac{2A_2}{A_1 E_C \delta}} \right) \right\}. \end{aligned} \quad (12)$$

As usual (see [5]), one should find a value $\xi = \xi_c$ corresponding to the first appearance of an infinite cluster of grains, connected according to the criterion (12). Then, with the exponential accuracy, the global conductivity of the system $\sigma \propto \exp(-\xi_c)$.

The arising percolational model differs, however, from the standard VRH one (see [5, 16]) due to an additional dependence on r_{ij} and $\varepsilon_i, \varepsilon_j$ appearing in the argument of the function φ . However, since the variation of this function on the relevant scale of its argument is $\delta\varphi \sim 1 \ll \mathcal{L}$, the corresponding relative variation of $\xi(\mathbf{r}_i, \varepsilon_i | \mathbf{r}_j, \varepsilon_j)$ is small and can be treated by the standard perturbational method (see [5]). As a result, we obtain the Mott law

$$\sigma \sim \exp\{-(T_M/T)^{1/(d+1)}\}, \quad (13)$$

with T_M , which is slightly temperature-dependent:

$$T_M = \frac{\mathcal{L}^d (T/T_c) n_c}{2P_{1/2}} E_C, \quad T_c = \frac{2c_2}{\mathcal{L}} \sqrt{\frac{A_1 E_C \delta}{2A_2}}, \quad (14)$$

$$\mathcal{L}(T/T_c) = c_2 \left\{ \ln \left(\frac{8\pi^2 E_C}{A_1 g \delta} \right) - \chi(T/T_c) \right\}. \quad (15)$$

The universal percolation constant $n_c \approx 5.7$ for $d=3$ and $n_c \approx 3.5$ for $d=2$. The function $\chi(z)$ is related to $\varphi(z)$ by

$$\chi(z) = \langle s \rangle_{\text{perc}}^{-1} \left\langle s \varphi \left(\frac{|\zeta - \zeta'|}{s} z \right) \right\rangle_{\text{perc}}, \quad (16)$$

where the averaging over the "percolation hypersurface" in the space of dimensionless energy ζ and dimensionless coordinate \mathbf{s} has the following explicit meaning:

$$\begin{aligned} \langle F \rangle_{\text{perc}} & \equiv \\ & \equiv \frac{\int d\zeta d\zeta' \int d\mathbf{s} F(\zeta, \zeta', \mathbf{s}) \delta \left(1 - s - \frac{|\zeta| + |\zeta'| + |\zeta - \zeta'|}{2} \right)}{\int d\zeta d\zeta' \int d\mathbf{s} \delta \left(1 - s - \frac{|\zeta| + |\zeta'| + |\zeta - \zeta'|}{2} \right)}. \end{aligned}$$

The asymptotics of the function $\chi(z)$ are

$$\chi(z) \approx \begin{cases} b_1 z^{2/3} & \text{for } z \ll 1, \\ 2(\ln z + b_2 + \dots) & \text{for } z \gg 1, \end{cases} \quad (17)$$

where b_1, b_2 are universal constants, depending only on the space dimensionality d . In particular, $b_2 = -1/6$ for $d=2$ and $b_2 = -1/2$ for $d=3$.

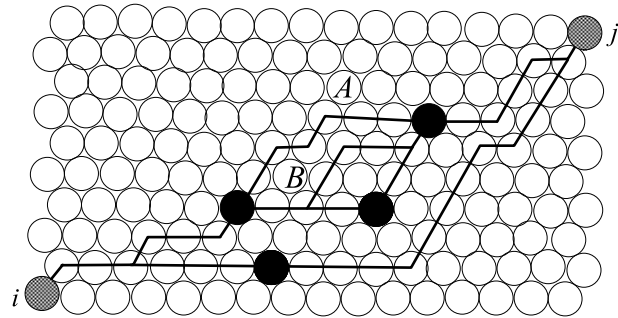
Experimentally the Mott law (13) is likely to be observed in materials with weak charge disorder, where the density of states at $\varepsilon=0$ is very small because of the factor $P_{1/2} \ll 1$, and the Coulomb gap is irrelevant,

except for extremely low T range. In such materials, however, T_M is very large (cf. (14)) and the crossover between the Mott and the Arrhenius laws should take place at relatively low T .

Magnetoresistance. In the entire above consideration we have neglected a possible interference between contributions of different strings (if any), connecting the same pair ij . The main reason for this approximation is the strong sign-fluctuations of the matrix elements $t_{h_k p_{k-1}}$: even in the coherent (purely elastic) limit the signs of contributions of different strings to the composite amplitude of $i \rightarrow j$ transition fluctuate independently. The interference effects, although irrelevant to the zero-field effects, are sensitive to magnetic field, so that they may be the source for a low temperature effect of a negative orbital magnetoresistance, similar to the one discussed in [10–13] for the case of VRH in conventional disordered semiconductors. The key component of this effect is the interference between the contributions of different spatial paths, leading from i to j . In our case this means the existence of several strings $i \rightarrow j$ giving comparable contributions to g_{ij} . For a fairly homogeneous material, where all g_{kk+1} are of the same order of magnitude one can expect such different strings to exist already for $N_{ij} \gtrsim 1$. For a strongly disordered material (with exponentially large fluctuations of g_{kk+1}) there is typically only one leading string for $N_{ij} < N_{\min}$, and only for $N_{ij} > N_{\min}$ several strings act in parallel. The crossover length N_{\min} is a function of the magnitude of fluctuations $D = (\ln g - \overline{\ln g})^2$, the explicit form of this function is model-dependent, and we will not discuss it in the present Letter. For us it is only important that $N_{\min}(D) \sim 1$ for $D \sim 1$, and $N_{\min}(D) \gg 1$ for $D \gg 1$.

Apparently, the magnetoresistance is controlled by the typical area $S(N_{\text{loop}})$ of the interference loop (N_{loop} being the "length" of this loop). Loops with $N_{\text{loop}} < N_{\min}$ are extremely rare, and can be ignored. For $N_{\text{loop}} > N_{\min}$ the scaling law $S(N_{\text{loop}}) \sim N_{\text{loop}}^u$, with an unknown exponent $u < 2$ holds. The problem of statistics of loops is closely related to the well-studied theory of directed polymers in a random field [17].

One of the essential ingredients of our cotunneling process is the presence of inelastic events, which certainly destroy the interference and suppress the magnetoresistance. Namely, the interference between two different strings A and B is possible only if the cotunneling at all grains of A and B , which are not common for them, is elastic (see Figure). Since the relative fraction of inelastic cotunnelings y depends on the temperature, so does the length $M_{\text{el}} \sim 1/y$ of a typical stretch on a string, containing only "elastic grains". It is just M_{el} , not the entire length N of the distant hop, that should



Four different strings contributing to the $i \rightarrow j$ transition in a particular realization of array. For a shown partition ("inelastic grains" $\{l\}$ are depicted as filled circles, elastic grains $\{m\}$ – as open ones) only two strings (A and B) contribute to interference effects

play the role of the effective length N_{loop} of the interference loop. Clearly, for $T \gtrsim T_c$ one has $M_{\text{el}} \sim 1$, while $M_{\text{el}} \sim (T_c/T)^{2/3} \gg 1$ for $T \ll T_c$.

Thus, we can conclude that for temperatures $T > T_{\text{mag}} \sim T_c N_{\min}^{-3/2}(D) < T_c$ the orbital magnetoresistance is strongly suppressed (since the typical elastic stretch is shorter than N_{\min}), while at $T \ll T_{\text{mag}}$ it can be relatively strong: the characteristic magnetic field H_c , at which the conductivity would saturate at $\sigma(H \gg H_c) \sim \sqrt{2}\sigma(H=0)$ (cf. Ref.[12]), is

$$H_c \sim \Phi_0/S(M_{\text{el}}) \sim H_c^{(0)}(T/T_c)^{2u/3}, \quad (18)$$

where $H_c^{(0)} = \Phi_0 n_g^{2/d}$ is the field, corresponding to a flux Φ_0 through an elementary triangle of neighboring grains. The dependence $\sigma(H)$ at $H < H_c$ can be different in different ranges of H : either $\Delta\sigma \propto H^2$ (at the smallest fields, see [11, 13]) or $\Delta\sigma \propto H$ (at the intermediate fields, see [10, 13]), or $\Delta\sigma \propto H^{1/2}$ (at relatively high fields, see [12]).

Conclusions. In conclusion, we have developed a theory of variable-range hopping in granular arrays with poor intergrain coupling. Long-range hopping of electrons is provided by the multiple co-tunnelling "strings" which contains both elastic and inelastic processes within individual grains. In the presence of long-range Coulomb interaction, Efros-Shklovskii law for the temperature dependence of conductivity is derived in the asymptotic limits of purely inelastic or purely elastic co-tunnelling. Upon temperature decrease, relative contribution of elastic co-tunnelling increases; For the model case of local (screened) Coulomb interaction, general situation of partially elastic co-tunnelling was studied and crossover temperature T_c was determined, cf. Eqs.(9), (14). For real granular metals this crossover temperature happens to be rather low. In particular, for Al grains of size $a \sim 20$ nm one estimates $E_C \sim 500$ K and

$\delta \sim 0.05$ K, which leads (at $g \sim 0.3$, so that $\mathcal{L} \sim 12$) to $T_c \sim 0.5$ K. For $a \sim 10$ nm the same estimates give $T_c \sim 2$ K. Therefore the major part of experimental temperature range (from room to liquid helium temperatures) is dominated by inelastic co-tunnelling. This is the reason for magnetoresistance to be very weak in granular metals, contrary to disordered semiconductors. Observation of noticeable negative magnetoresistance due to interference of different tunnelling “strings” might be possible with granular media made of small (≤ 10 nm) grains of non-superconductive metals like copper, silver or gold, at temperatures below 1 K. In the inelastic regime (at $T > T_c$) the “constant” T_{ES} , entering Eq. (1), is itself T -dependent: it logarithmically increases with the decrease of T . This dependence should lead to somewhat faster growth of the resistivity upon lowering T , than predicted by the standard Efros-Shklovskii law.

After the present study was completed, we became aware of the preprint [18] where purely elastic variable-range cotunnelling was proposed as the conduction mechanism for granular metals; their results seem to agree with our ones as long as inelastic processes are neglected.

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