Transport Properties of Granular Metals at Low Temperatures

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We investigate transport in a granular metallic system at large tunneling conductance between the grains, $g_T \gg 1$. We show that at low temperatures, $T \leq g_T\delta$, where δ is the mean energy level spacing in a single grain, the coherent electron motion at large distances dominates the physics, contrary to the high-temperature ($T > g_T\delta$) behavior where conductivity is controlled by the scales of the order of the grain size. In three dimensions we predict the metal-insulator transition at the bare tunneling conductance $g_T^C = (1/6\pi)\ln(E_C/\delta)$, where E_C is the charging energy of a single grain. Corrections to the density of states of granular metals due to the electron-electron interaction are calculated. Our results compare favorably with the logarithmic dependence of resistivity in the high-T_c cuprate superconductors indicating that these materials may have a granular structure.

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A great deal of research in the current mesoscopic physics focuses on understanding properties of granular metals (see [1–3]). The interest is motivated by the fact that while their properties are generic for a wealth of strongly correlated systems with disorder, granular metals offer a unique experimentally accessible tunable system where both the interaction strength and degree of disorder can be controlled.

The key phenomenon revealing the most underlying physics is transport, where the effects of interactions play a crucial role. The processes of electron tunneling from grain to grain that govern electron transfer are accompanied by charging the grains involved after each electron hop to another grain. This may lead to a Coulomb blockade, and one justly expects this effect to be of prime importance at least in the limit of weak coupling. It thus makes it clear, on a qualitative level, that it is the interplay between the grain-to-grain coupling and the electron-electron Coulomb interaction that controls transport properties of granular metals; yet, despite the significant efforts expended, a quantitative theory of transport in metallic granular systems is still lacking.

A step towards formulating such a theory was made recently in Ref. [3]. It was shown that depending on the dimensionless tunneling conductance g_T one observes either exponential, at $g_T \ll 1$, or logarithmic, at $g_T \gg 1$, temperature dependence of conductivity. The consideration in [3] was based on the approach developed by Ambegaokar, Eckern, and Schön (AES) [4] for tunnel junctions. This technique, however, as shown in [5], applies only at temperatures $T > g_T\delta$, where δ is the mean energy level spacing in a single grain; in this regime the electron coherence does not extend beyond the grain size. At the low-temperature region, $T \leq g_T\delta$, the effects of the electron coherent motion at distances greatly exceeding the single grain size a must be included; thus, this important regime is not described by the AES approach [5].

Although experimentally the low-temperature regime is well within the experimental reach [1,2], it has never been addressed theoretically so far. The important question whether the system is a metal or becomes an insulator—in other words, whether the conductivity of the granular metals at large conductances remain finite in the limit of $T \to 0$—is still open.

In this Letter we investigate the low-temperature conductivity of granular samples focusing on the case of large tunneling conductance between the grains, $g_T \gg 1$. To this end we develop a technique that goes beyond the AES approach and includes effects of coherent electron motion at distances larger than the size of the grain. Without the Coulomb interaction, the granular system would be a good metal in the limit $g_T \gg 1$, and our task is to include the charging effects in the theory. We find that at temperatures $T \leq g_T\delta$ properties of the granular metal depend on the dimensionality of the array, and corrections to the conductivity and density of states due to the effects of Coulomb interaction are similar to those obtained in Ref. [6] for a homogeneous metal. Thus, at low temperatures the system behaves essentially as a homogeneous metal contrasting the case of large temperatures, $T \gg g_T\delta$, considered in Ref. [3].

This means that at large conductances the 3D system is a good metal. On the other hand, at $g_T \ll 1$ a granular sample is in the insulating state. Therefore, a 3D system should exhibit a metal-insulator transition at the critical value of the conductance g_T, such that samples with conductances $g_T > g_T^C$ are metals and their conductivity remains finite at $T \to 0$, while samples with $g_T < g_T^C$ are insulators and their conductivity vanishes at $T \to 0$.

The main results of our work are as follows: (i) We estimate the critical value g_T^C of the bare tunnel

conductance at which the metal-insulator transition in 3D occurs as

$$g_T^C = (1/6\pi)\ln(E_C/\delta), \tag{1}$$

where E_C is the charging energy of an isolated grain. (ii) We find the expression for the conductivity of a granular metal that includes corrections due to Coulomb interaction and holds for all temperatures as long as these corrections are small. The corresponding answer can be conveniently written separating the correction due to the contribution from the large energy scales $\varepsilon > g_T\delta$ from that coming from the low energy scales $\varepsilon < g_T\delta$. Denoting corrections as $\delta\sigma_1$ and $\delta\sigma_2$, respectively, we have

$$\sigma = \sigma_0 + \delta\sigma_1 + \delta\sigma_2, \tag{2a}$$

where $\sigma_0 = 2e^2 g_T a^{2-d}$, with a being the size of the single grain, is the classical Drude conductivity for a granular metal (spin included). Correction $\delta\sigma_1$ in Eq. (2a) contains the dimensionality of the array d only as a coefficient and is given by the following expression [3]:

$$\frac{\delta\sigma_1}{\sigma_0} = -\frac{1}{2\pi d g_T}\ln\left[\frac{g_T E_C}{\max(T, g_T\delta)}\right]. \tag{2b}$$

On the contrary, the correction $\delta\sigma_2$ in Eq. (2a) that is important only at temperatures $T < \delta g_T$ strongly depends on the dimensionality of the array

$$\frac{\delta\sigma_2}{\sigma_0} = \begin{cases} \frac{\alpha}{12\pi^2 g_T}\sqrt{\frac{T}{g_T\delta}}, & d = 3, \\ -\frac{1}{4\pi^2 g_T}\ln\frac{g_T\delta}{T}, & d = 2, \\ -\frac{\beta}{4\pi}\sqrt{\frac{\delta}{T g_T}}, & d = 1. \end{cases} \tag{2c}$$

Here $\alpha = \int_0^\infty dx x^{-1/2}[1 - \coth(x) + x/\sinh^2(x)] \approx 1.83$ and $\beta = \int_0^\infty dx x^{-3/2}[\coth(x) - x/\sinh^2(x)] \approx 3.13$ are the numerical constants. For a 3D granular system, a temperature independent term of the order $1/g_T$ has been subtracted in the first line in Eq. (2c).

Corrections $\delta\sigma_1$ and $\delta\sigma_2$ are of a different origin: the correction $\delta\sigma_1$ comes from the large energy scales $\varepsilon > g_T\delta$, where the granular structure of the array dominates the physics. The fact that this correction is essentially independent of the dimensionality d means that the tunneling of electrons with energies $\varepsilon > g_T\delta$ can be considered as incoherent. On the other hand, correction $\delta\sigma_2$ in Eq. (2c) is similar to that obtained for homogeneous metals long ago [6] and comes from the low energy scales, $\varepsilon \leq g_T\delta$, where the coherent electron motion on the scales larger than the grain size a dominates the physics.

It is important to note that in the low-temperature regime all temperature dependence of conductivity comes from the correction $\delta\sigma_2$. At the same time, in this regime the correction $\delta\sigma_1$, though being temperature independent, still exists and can be even larger than $\delta\sigma_2$.

When deriving Eqs. (2) we neglected possible weak localization corrections that may originate from quantum interference of electron waves. This approximation is legitimate if a magnetic field is applied as in Ref. [1] or dephasing is strong due to inelastic processes.

Now we turn to the description of our model and the derivation of Eqs. (2): We consider a d-dimensional array of metallic grains with the Coulomb interaction between electrons. The motion of electrons inside the grains is diffusive, and they can tunnel from grain to grain. In principle, the grains can be clean such that electrons scatter mainly on grain surfaces. We assume that the sample in the absence of the Coulomb interaction would be a good metal. Although we assume that g_T is large, it should be still smaller than the grain conductance g_0, meaning that the granular structure is important and the resistivity is controlled by tunneling between the grains.

The system of weakly coupled metallic grains can be described by the Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_c + \sum_{ij} t_{ij}[\hat{\psi}^\dagger(r_i)\hat{\psi}(r_j) + \hat{\psi}^\dagger(r_j)\hat{\psi}(r_i)], \tag{3a}$$

where t_{ij} is the tunneling matrix element corresponding to the points of contact r_i and r_j of ith and jth grains. The Hamiltonian $\hat{H}_0$ in Eq. (3a) describes noninteracting isolated disordered grains and the term

$$\hat{H}_c = \frac{e^2}{2}\sum_{ij}\hat{n}_i C_{ij}^{-1}\hat{n}_j \tag{3b}$$

describes the Coulomb interaction with C_{ij} being the capacitance matrix and $\hat{n}_i$ being the operator of the electron number in the ith grain. In the regime under consideration one can neglect the coordinate dependence of a single grain diffusion propagator. The electron hopping between the grains can be included using the diagrammatic technique developed in Refs. [5,7].

The conductivity of the granular metals is given by the analytical continuation of the Matsubara current-current correlator. In the absence of the electron-electron interaction the conductivity is represented by the diagram Fig. 1(a) that results in high-temperature (Drude) conductivity σ_0 which is defined below Eq. (2a). First order interaction corrections to the conductivity are given by the diagrams Figs. 1(b)–1(e). These diagrams are analogous to ones considered in Ref. [6] for the correction to the conductivity of homogeneous metals. We consider the contributions from diagrams 1(b) and 1(c) and 1(d) and 1(e) separately. The sum of diagrams 1(b) and 1(c) results in the following correction to the conductivity:

$$\frac{\delta\sigma_1}{\sigma_0} = -\frac{1}{2\pi d g_T}\mathrm{Im}\sum_{\mathbf{q}}\int d\omega\,\gamma(\omega)\varepsilon_{\mathbf{q}}\tilde{V}(\omega, \mathbf{q}), \tag{4}$$

where $\gamma(\omega) = \frac{d}{d\omega}\omega\coth\frac{\omega}{2T}$, $\varepsilon_q = 2g_T\sum_{\mathbf{a}}(1 - \cos\mathbf{q}\mathbf{a})$ with

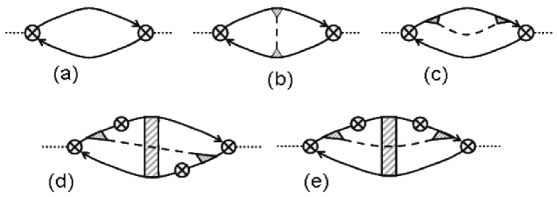

FIG. 1. Diagrams describing the conductivity of granular metals: diagram (a) corresponds to σ_0 in Eq. (2a), and it is the analog of Drude conductivity. Diagrams (b)–(e) describe the first order correction to the conductivity of granular metals due to electron-electron interaction. The solid lines denote the propagator of electrons, and the dashed lines describe effective screened electron-electron propagator. The tunneling vertices are described by the circles. The sum of diagrams (b) and (c) results in the conductivity correction $\delta\sigma_1$ in Eq. (2a). The other two diagrams, (d) and (e), result in the correction $\delta\sigma_2$.

a being the lattice vectors and

$$\tilde{V}(\omega, \mathbf{q}) = \frac{2E_C(\mathbf{q})}{(\varepsilon_\mathbf{q}\delta - i\omega)[4\varepsilon_\mathbf{q}E_C(\mathbf{q}) - i\omega]}. \tag{5}$$

Here the charging energy $E_C(\mathbf{q}) = e^2/2C(\mathbf{q})$ is expressed in terms to the Fourier transform of the capacitance matrix $C(\mathbf{q})$. Performing the integration over the frequency and summing over the quasimomentum $\mathbf{q}$ in Eq. (4) with the logarithmic accuracy, we obtain the correction (2b). One can see from Eq. (4) that the contribution $\delta\sigma_1$ in Eq. (2b) comes from the large energy scales, $\varepsilon > g_T\delta$ such that at low temperatures the logarithm is cut off on the energy scale $g_T\delta$.

To obtain the total correction to the conductivity of granular metal, the two other diagrams, Figs. 1(d) and 1(e), should be taken into account. These diagrams result in the following contribution to the conductivity:

$$\frac{\delta\sigma_2}{\sigma_0} = -\frac{2g_T\delta}{\pi d}\sum_\mathbf{q}\int d\omega\,\gamma(\omega)\mathrm{Im}\frac{\tilde{V}(\omega, \mathbf{q})\sum_\mathbf{a}\sin^2(\mathbf{qa})}{\varepsilon_\mathbf{q}\delta - i\omega}. \tag{6}$$

In contrast to the contribution $\delta\sigma_1$ in Eq. (4), the main contribution to the sum over the quasimomentum $\mathbf{q}$ in Eq. (6) comes from the low momenta, $q \ll 1/a$. In this regime the capacitance matrix $C(\mathbf{q})$ in Eqs. (5) and (6) has the following asymptotic form:

$$C^{-1}(\mathbf{q}) = \frac{2}{a^d}\begin{cases}\ln(1/qa), & d = 1,\\ \pi/q, & d = 2,\\ 2\pi/q^2, & d = 3.\end{cases} \tag{7}$$

Substituting Eq. (7) into Eq. (5), integrating over the frequency, and summing over the quasimomentum $\mathbf{q}$ in Eq. (6), we obtain the result for the correction $\delta\sigma_2$ in Eq. (2c).

Comparing our results in Eqs. (2) with those obtained in Ref. [3] using the AES functional, we see that the correction to the conductivity obtained in Ref. [3] is equivalent to the correction $\delta\sigma_1$ in Eq. (2a), which corresponds in our approach to the sum of diagrams Figs. 1(b) and 1(c). The correction $\delta\sigma_2$ in Eq. (2a) becomes important only at low temperatures, $T < g_T\delta$, where an AES functional is not applicable. While in our approach both corrections to the conductivity must be small $\delta\sigma_1, \delta\sigma_2 \ll \sigma_0$, the method of Ref. [3] gives the possibility to show that for $T \gg g_T\delta$ the dependence of the conductivity is logarithmic as long as $\sigma/e^2a^{2-d} \gg 1$.

It follows from Eq. (2c) that at low temperatures, $T < g_T\delta$, for a 3D granular array, there are no essential corrections to the conductivity coming from the low energies since the correction $\delta\sigma_2$ is always small. This means that the result for the renormalized conductance, $\tilde{g}_T$ of Ref. [3] for 3D samples within the logarithmic accuracy can be written in the following form:

$$\tilde{g}_T(T) = g_T - \frac{1}{6\pi}\ln\left[\frac{g_TE_C}{\max(\tilde{g}_T\delta, T)}\right], \tag{8}$$

such that it is valid for *all* temperatures as long as the renormalized conductance $\tilde{g}_T \gg 1$. One can see from Eq. (8) that for bare conductance, $g_T \gg (1/6\pi) \times \ln(E_C/\delta)$, the renormalized conductance $\tilde{g}_T$ is always large and the system remains metallic down to zero temperature. In the opposite limit $g_T < (1/6\pi)\ln(E_C/\delta)$, the system flows when decreasing the temperature to the strong coupling regime $\tilde{g}_T \sim 1$ that indicates the onset of the insulating phase. We see that with the logarithmic accuracy the critical value of the conductance g_T^C is given by Eq. (1).

The result for the bare critical conductance in Eq. (1) agrees with the estimate for g_T^C that follows from the consideration of Coulomb blockade phenomena in a single grain [8]: the contribution of the Coulomb blockade to thermodynamic quantities in the regime of strong coupling is controlled by the factor $\sim\exp[-\pi g(T)]$, where $g(T) = g_T - (1/Z\pi)\ln(g_TE_C/T)$ with Z being the number of contacts. Coulomb blockade effects become strong at $g(T) \sim 1$. Taking $T \sim g_T\delta$ and $Z = 6$ we estimate the bare conductance as $g_T^C \sim (1/6\pi)\ln(g_TE_C/T)$ that coincides with Eq. (1).

In a similar way we can obtain interaction corrections to the density of states (DOS) of granular metal,

$$\frac{\delta\nu(\varepsilon)}{\nu_0} = -\frac{1}{4\pi}\sum_\mathbf{q}\mathrm{Im}\int d\omega\frac{\tanh[(\varepsilon - \omega)/2T]}{(\varepsilon_\mathbf{q}\delta - i\omega)[\varepsilon_\mathbf{q} - i\omega/4E_C(\mathbf{q})]}. \tag{9}$$

Here ν_0 is the DOS for noninteracting electrons, and $\varepsilon_\mathbf{q}$ and $E_C(\mathbf{q})$ were defined below Eqs. (4) and (5), respectively. Using Eq. (9) for a 3D granular array we obtain

$$\frac{\delta\nu_3}{\nu_0} = -\frac{A}{2\pi g_T}\ln\left[\frac{E_Cg_T}{\max(T, g_T\delta)}\right], \tag{10a}$$

where $A = g_T a^3 \int d^3q/(2\pi)^3 \varepsilon_{\mathbf{q}}^{-1}$. For temperatures $T \gg g_T\delta$ the correction to the DOS (10a) coincides with the one obtained in Ref. [3] using the AES approach. It follows from Eq. (10a) that for a 3D array of grains, as in the case with conductivity, the main contribution to the DOS comes from the large energy scales, $\varepsilon > g_T\delta$.

Using Eq. (9) for a 2D array we obtain the following result for the correction to the DOS:

$$\frac{\delta\nu_2}{\nu_0} = -\frac{1}{16 g_T \pi^2} \begin{cases} 2\ln^2 \frac{g_T E_C}{T}, & T \gg g_T\delta, \\ \ln\frac{g_T\delta}{T}\ln\frac{g E_C^4}{T\delta^3} + 2\ln^2\frac{E_C}{\delta}, & T \ll g_T\delta. \end{cases} \tag{10b}$$

Using the relation between the tunneling conductance, g_T, and the diffusion coefficient, $D = g_T a^2 \delta$, one can see that the temperature dependence of the DOS for $T \ll g_T\delta$ given by Eq. (10b) coincides up to the temperature independent constant with the result for the correction to the DOS of the homogeneous metal [6].

The logarithmic behavior (2b) of the conductivity is in good agreement with experimental findings [1,2]. It would be very interesting to extend the resistivity measurements to the low-temperature regime where we predict the temperature dependence (2c). A similar logarithmic dependence of resistivity on temperature was recently found in high-T_c compounds $La_{2-y}Sr_yCuO_4$ (LSCO) and $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ in a very strong magnetic field [9,10]. A possible granularity of these samples was suggested in Ref. [3]. Recently the microscopic granularity was directly experimentally observed in the superconducting state of $Bi_2Sr_2CaCu_2O_{8+\delta}$ by the scanning tunneling microscopy probe [11]. If we accept that samples studied in [9,10] are indeed microscopically granular, we can compare the results of the experiments with our predictions. When doing so, it is convenient to scale three-dimensional conductivity to the conductivity of CuO planes, σ_{plane}. According to our predictions,

$$d\sigma_{\text{plane}}/d\ln T = (e^2/\pi\hbar)k, \tag{11}$$

where the coefficient $k = 1/2\pi$ in the low-temperature and $k = 1/d$ in the high-temperature regimes. While in the low-temperature regime the application of Eq. (11) is legitimate only under the assumption that electrons in different CuO planes are incoherent, in the high-temperature regime the behavior of conductivity according to Eq. (2b) is logarithmic for any dimension. In this regime the real dimensionality d should be replaced by $d = Z/2$, where Z is the (average) number of the contacts of each grain with all the adjacent grains. Describing the data shown in Fig. 3 of Ref. [10] by our log dependencies at temperature $T \approx 5$ K we extract $k \simeq 0.4$, for Sr concentration of $y = 0.08$ for $La_{2-y}Sr_yCuO_4$ [12]; for the $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ compound we find $k \simeq 0.2$ for $x = 0.84$ La concentration, and $k \simeq 0.3$ for $x = 0.76$. For each particular curve the values k extracted from Fig. 3 of Ref. [10] increase with temperature (especially in the case of LSCO), and this is in a complete agreement with our results provided that the ''coherent-incoherent'' crossover occurs at about $T \sim 5$ K. At higher temperatures k noticeably exceeds $1/2\pi$, supporting the idea of a granularity of doped cuprates.

In conclusion, we have investigated transport properties of granular metals at large tunneling conductance and obtained corrections to the conductivity, Eqs. (2), and DOS, Eqs. (10), due to electron-electron interaction. We have shown that at temperatures $T > g_T\delta$, the granular structure of the array dominates the physics. On the contrary, at temperatures $T \leq g_T\delta$ the large-scale coherent electron motion is crucial. Comparing our results with experimental data supports the assumption of a granular structure of doped high-T_c cuprates.

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[12] This curve for $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$, $x = 0.15$ shows a very wide crossover at about 20 K extending to low temperatures. Thus, there are just a very few points in the narrow temperature interval 1–3 K where our approach is applicable. This invalidates the comparison of the data with our results for this particular curve.