Electron Transport in Granular Metals

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We consider thermodynamic and transport properties of a long granular array with strongly connected grains (intergrain conductance $g \gg 1$). We find that the system's conductance and differential capacitance exhibits activated behavior, $\sim \exp\{-T^*/T\}$. The gap T^* represents the energy needed to create a long single-electron charge soliton propagating through the array. This scale is parametrically larger than the energy at which conventional perturbation theory breaks down.

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The low-temperature conductivity of granular materials continues to attract the attention of experimentalists [1] and theorists [2,3]. From a conceptual point of view, an attractive feature of these systems is the possibility to separately control the effects of electron interaction and quantum interference. A particularly interesting situation is realized in arrays with large intergranular conductance, $g \gg 1$ (in units of e^2/h), and large grain size (small electron mean level spacing, δ, in the grains). Under conditions where all characteristic energy scales related to δ are smaller than the temperature, the electron transport in such systems is purely inelastic and long range quantum coherence is inhibited [3]. As we show below, under these conditions interaction effects *alone* lead to an exponential suppression of conductivity, which is fully amenable to analytical treatment.

At high temperatures, the conductivity of a granular array is Ohmic, $\sigma = g$ (hereafter the length of the system is measured in the number of grains). At lower temperatures Altshuler-Aronov interaction corrections [4] impede the conduction behavior. For "inelastic" arrays this correction was found [2] to be $\delta\sigma = -\ln E_c/T$, where E_c is the charging energy of an individual grain. Comparison with the Ohmic contribution shows that this *perturbative* correction is small as long as $T > \tilde{E}_c \equiv E_c e^{-g}$. At the same energy scale, $\tilde{E}_c$, a *single* grain connected to external leads would cross over to the strong Coulomb blockade regime [5–7].

In this Letter we show that the conductivity of a $1d$ *array* of grains crosses over to a manifestly insulating (activated) behavior at a parametrically *larger* temperature, $T^* \gg \tilde{E}_c$. Below the crossover, the conductivity is exponentially small:

$$\sigma = g \exp\left(-\frac{T^*}{T}\right), \qquad T \lesssim T^*, \tag{1}$$

as characteristic for insulators. The gap, T^*, depends on the background charge, q, sitting on each grain. For an array with globally vanishing $q = 0$, we find $T^* \sim E_c e^{-g/4}$. The differential capacity is also suppressed in a way similar to the conductivity. Note that Eq. (1) is *not* a result of phonon-mediated hopping, but is a consequence of interactions between electrons only. In the intermediate temperature range, $T^* < T < E_c$, perturbation theory is applicable and the conductivity exhibits the logarithmic behavior [2].

The reason why the scale T^* and Eq. (1) were overlooked previously is that they are not visible in standard perturbative expansions in $1/g \ll 1$. In the conventional formulation of the theory in terms of *voltage* fluctuations [8], Eq. (1) is a consequence of large, topologically nontrivial fluctuations (instantons). Proliferation of instantons leads to insulating behavior at temperatures $T^* \gg \tilde{E}_c$, where Gaussian fluctuations are still small. Notice that for a *single* grain instantons affect the conductance only at much lower temperatures $T \approx \tilde{E}_c$ [9]. However, contrary to a single dot, an extended array provides a large "entropic volume" for the formation of instantons, which substantially increases the characteristic temperature. We shall return to a quantitative discussion of this picture below.

It turns out, however, that the effect is more naturally explained using a language of *charge* fluctuations. It is known that even a highly conducting barrier retains some ability to pin the charge on a single grain [5–7]. This mechanism is drastically enhanced in the array geometry, where it bears similarity to the pinning of charge density waves by a periodic potential. The elementary mobile excitations in this system are finite size solitons of unit charge. Their activation energy, T^*, is given by the geometric mean of the pinning strength and inverse charge compressibility (grain capacitance). Our main result, Eq. (1), simply reflects the thermal density of such single-charge solitons.

To quantify this latter picture we consider a generalization of a model previously employed to study quantum dots [10]. Its simplest version treats the grains coupled by a single conducting channel and therefore has $g \lesssim 1$. (We shall show later that the predictions derived from it survive generalization to the complementary case $g \gg 1$.) The model is formulated in terms of a charge displacement field, $\theta_j(\tau)$, where $\theta_{j+1} - \theta_j = N_j$ is the charge on the jth grain. In the absence of backscattering at the contacts, the action reads

$$S_0 = \sum_{j=1}^{M-1} \frac{1}{T} \sum_m [E_c (\theta_{j+1} - \theta_j - q)^2 + \pi|\omega_m|\theta_j^2], \quad (2)$$

where the first term represents the charging energy of the grains, while the second originates from integrating out the continuum of the electronic degrees of freedom. Backscattering at the intergranular contacts is described [10] by a nonlinear action (2): $S_{\mathrm{bs}} = \frac{Dr}{\pi}\sum_j \int d\tau \times \cos(2\pi\theta_j(\tau))$. Here, r is the reflection amplitude and $D \gg E_c$ is conduction bandwidth.

A crucial observation that makes the problem solvable is that even for $r = 0$ the *quantum* fluctuations of $\theta_j(\tau)$ do *not* [11] diverge in the limit $T \to 0$:

$$\langle\theta_j(\tau)^2\rangle = \frac{T}{M} \sum_{k=0}^{M-1} \sum_{m\neq 0} \frac{e^{-|\omega_m|/D}}{E_k + \pi|\omega_m|} = \frac{1}{2\pi^2} \ln\frac{\pi D}{e^{\mathbf{C}} E_c}, \quad (3)$$

where $E_k = 4E_c \sin^2(\pi k/2M)$ is the excitation spectrum defined by Eq. (2), and $\mathbf{C} \approx 0.577$ is the Euler constant. One can thus safely integrate out these fluctuations, to arrive at a sine-Gordon type action that involves only the *classical* (zero Matsubara) component of the field:

$$S[\theta] = \frac{E_c}{T} \sum_{j=1}^{M-1} [(\theta_{j+1} - \theta_j - q)^2 - 2\gamma \cos(2\pi\theta_j)], \quad (4)$$

where $\gamma \equiv |r|e^{\mathbf{C}}/(2\pi^2)$. In the multichannel case, the coupling constant generalizes [7,12] to $\gamma \sim \prod |r_s|$, where r_s is the reflection coefficient of the sth channel.

Equation (4) is known as the action of the Frenkel-Kontorova model [13]. This model describes a harmonic elastic chain of "atoms" with stiffness E_c, placed on top of a periodic "substrate" potential with the amplitude $2\gamma E_c$. The "incommensurability parameter" q represents the periodicity mismatch between the chain and the substrate. For small values of q the system finds it favorable to retain a commensurate state [cf. Fig. 1(a)]; i.e., the chain stretches a little to still benefit from an optimal coupling to the substrate. Setting $\theta_j = 0$, one finds that the energy per "atom" in this configuration is given by $F_c = E_c(q^2 - 2\gamma)$. At $|q| > 2\gamma$ this energy becomes positive, and the state with $\theta_j = 0$ cannot persist as the lowest-energy state (i.e., it is obviously less favorable than the incommensurate state with $\theta_j \approx jq$ and $F_i \approx 0$). Indeed, in the limit of weak periodic potential, the transition between the commensurate [Fig. 1(a)] and incommensurate [Fig. 1(b)] phases occurs at $|q| = q^* \equiv \sqrt{2\gamma}$. For the average number of electrons per grain, $\bar{N}(q) \equiv q - \partial_q F/(2E_c)$, one thus expects $\bar{N}(q) = 0$ for $|q| \leq q^*$ (insulator) and $\bar{N} \to q$ for $|q| > q^*$ (metal).

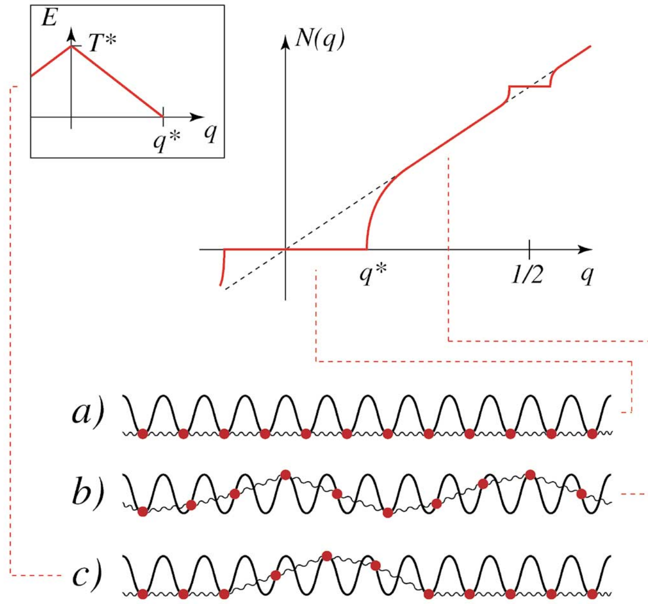

FIG. 1 (color online). Plot of the function (see text) $\bar{N}(q)$ of an atomic chain in proximity to a periodic substrate. (a) Commensurate configuration, (b) incommensurate configuration, and (c) solitary excitation with its excitation energy (inset).

The relevant *thermal excitations* in the commensurate state are so-called incommensurations (solitons in the language of the sine-Gordon model)—local defects, where the distortion θ_j "climbs" over a maximum of the substrate potential to relax back into a next minimum [cf. Fig. 1(c)]. Minimizing the action (4), subject to the boundary condition $\theta_{\pm\infty} = 0(1)$, and employing the condition $\gamma \ll 1$, one finds that the soliton action is given by $S_s = T^*(q)/T$, where

$$T^*(q) \overset{|q|\leq q^*}{=} 2\pi E_c(q^* - |q|); \qquad q^* = \sqrt{2\gamma}. \quad (5)$$

As a result, at $|q| < q^*$ the differential capacitance scales as $\partial_q \bar{N}(q) \sim \exp(-T^*(q)/T)$; i.e., $T^*(q)$ is the excitation gap of the system. Consequently, the conductivity exhibits the same activation behavior; cf. Eq. (1). (For an elaboration on the conductivity, see below.) Notice that for $|q| = q^*$ the gap vanishes. In agreement with our earlier estimate, this signals a proliferation of solitary excitations and the proximity of the incommensurate phase.

A more thorough discussion of the system (cf. Ref. [13]) shows that insulating "plateaus" along with superimposed solitary excitations form not only around $q = 0$, but also around other rational values of q. However, both the width of these plateaus and the corresponding activation energies decrease for higher rational fractions. Among the low lying rationals, $q = 1/2$ plays a particularly interesting role. Indeed, for a *single* grain, $q = 1/2$ represents charge degeneracy point, where the system is in a conducting state (Coulomb blockade peak). Unexpectedly, the *array* exhibits a very different behavior.

Using our current language, $q = \pm 1/2$ is special in that the atoms of the unperturbed chain alternatingly find themselves in minima/maxima of the substrate potential. Under these conditions, energy can be gained by building up a "Peierls distortion" of periodicity 2 and modulation amplitude $\delta\theta_j \sim \gamma$. This configuration is inert against small variations in q (insulating). The width of the insulating plateau estimates to only $\Delta q_{1/2} \sim \gamma$; i.e., it is much smaller than $\Delta q_0 \equiv 2q^* \sim \sqrt{\gamma}$.

The above discussion was based on the arguably artificial assumption that the background charges in every grain are the same. Under realistic conditions, though, one expects $q \to q_j$ to fluctuate. (The same applies to the tunneling conductances and charging energies; however, these latter fluctuations are of lesser relevance.) Let us briefly consider an extreme limit where $q_j \in [0, 1[$ on the different grains are uniformly distributed statistically independent random variables. For an undistorted chain, $\theta_{j+1} - \theta_j = q_j$, the potential terms $2\gamma \cos 2\pi\theta_j$ vary randomly, and the energy per atom is zero on average. The system can reduce its energy $\delta F \approx E_c \gamma^2$ per grain by slightly distorting the chain, so that $\delta\theta_{j+1} - 2\delta\theta_j + \delta\theta_{j-1} = \gamma \sin(2\pi j q_j)$. In analogy with the "clean" case, the *typical* excitation energy of the deformed state is $\sim\sqrt{E_c \delta F} \approx E_c \gamma$ [14]. The conductivity of the $1d$ random array is determined, however, by the *largest* pinning energy rather than the typical one. Because of the lack of space we defer this consideration to a subsequent publication [15].

Having discussed the charge pinning mechanism in the context of the few channel model, we next turn to a generalization to highly conducting arrays ($g \gg 1$). To this end we employ the so-called Ambegoakar-Eckern-Schön (AES) model [8]. This formalism describes the system in terms of the quantum phase, $\phi_j(\tau)$, conjugate to the charge $\theta_{j+1}(\tau) - \theta_j(\tau)$ of the jth grain. (Alternatively, one may think of ϕ_j as the time integral of the *voltage* on the grains, $i\dot{\phi}_j = V_j$.) The action of the model contains two terms, $S = S_c + S_t$, where $S_c[\phi] = \sum_j \int d\tau[\dot{\phi}_j^2/(4E_c) - iq\dot{\phi}_j]$ is the charging energy of a grain kept at voltage $V_j = i\dot{\phi}_j$, and

$$S_t[\phi] = \frac{gT^2}{2} \sum_{j=0}^{M-1} \int_0^\beta d\tau d\tau' \frac{\sin^2[\Delta\phi_j(\tau) - \Delta\phi_j(\tau')]}{\sin^2(\pi T(\tau - \tau'))} \quad (6)$$

describes the tunneling. Here $\Delta\phi_j \equiv (\phi_{j+1} - \phi_j)/2$ where $i\dot{\phi}_0$ and $i\dot{\phi}_M$ are the voltages on the leads connected to the array.

Before analyzing the array in terms of the above action, let us review a few general features of the AES approach: (i) ignoring effects of quantum interference, the applicability of the model is restricted [3] to temperatures $T > g\delta$; (ii) the quadratic approximation to the action, $S^{(2)}[\phi] = \frac{1}{T}\sum_{j,m}[\frac{\omega_m^2}{4E_c}|\phi_j|^2 + 2g|\omega_m||\Delta\phi_j|^2]$, provides a complete description of the *classical* RC-resistor network corresponding to the array; (iii) anharmonic fluctuations of the phase lead to the perturbative logarithmic correction to the dc conductivity [2] mentioned in the introduction; (iv) technically, the field ϕ_j represents a mapping $S^1 \to S^1$ from the unit circle (imaginary time augmented with periodic boundary conditions) into itself (ϕ_j is a phase). In addition to $\phi = 0$, the tunneling action $S_t[\phi]$ of a single grain (which, for low temperatures $T \ll E_c$, represents a good approximation to the *total* action of the grain) possesses a set of topologically nontrivial extremal phase configurations known as Korshunov instantons [5–7,16]:

$$\exp(i\phi^{(z)}(\tau)) \equiv \prod_{\alpha=1}^{|W|} \frac{e^{2\pi i\tau T} - z_\alpha}{1 - \bar{z}_\alpha e^{2\pi i\tau T}}. \quad (7)$$

Here, $W \in \mathbb{Z} \setminus 0$ is the winding number of the mapping $\phi^{(z)}$ and $z \equiv (z_1, \ldots, z_{|W|})$ is a set of $|W|$ complex parameters constrained by $|z_\alpha| < 1$. The instanton action, $S[\phi^{(z)}] \approx g|W| - 2\pi iqW$, is nearly z independent [17] which identifies the z_α's as zero modes. (Physically, $\arg z_\alpha$ determines the instance and $1 - |z_j|$ the duration of the voltage pulse, $i\dot{\phi}^{(z)}$.)

Turning to the array, the fact that the tunneling action depends only on the *differences* of neighboring phases, $\Delta\phi_j$, implies that a "plateau" formed by L instanton fields embedded into $M - L$ zeros, $(0, \ldots, 0, \phi^{(z)}, \ldots, \phi^{(z)}, 0, \ldots, 0)$, represents an extremal configuration. For $W = \pm 1$ its action is given by $S[\phi] = L(\pi^2 T/E_c \mp 2\pi iq) + g$, an expression that suggests an alternative interpretation of the instanton plateau: rather than monitoring a state of every grain, one may think of the plateau as a dipole of two charges located at the positions of the stepwise changes in the winding number, W_j: $0 \to 1$ and $1 \to 0$, respectively. Within this picture, $\exp(-g/2)$ represents the fugacity of the charges and $|L|\pi^2 T/E_c$ their interaction, and the q-dependent term describes the interaction of the dipole with a uniform electric field $2\pi iq$. More formally, a summation over all instanton configurations followed by integration over massive Gaussian fluctuations and zero modes [18,15] leads to the expression

$$\frac{Z}{Z_0} = \sum_{k=0}^{\infty} \frac{(\frac{\gamma E_c}{T})^{2k}}{(k!)^2} \sum_{j_1 \cdots j_{2k}}^{M-1} e^{-\frac{1}{2}\sum_{a,b}^{2k} V(j_a - j_b) - \sum_a^{2k} \Phi(j_a)}, \quad (8)$$

where $\gamma^2 \equiv g^3 e^{-g}$, and the interaction potentials are

$$V(j_a - j_b) = \frac{\pi^2 T}{E_c} e_a e_b |j_a - j_b|; \qquad \Phi(j_a) = 2\pi iqe_a j_a, \quad (9)$$

with $e_a \equiv (-1)^a$. These equations generalize from a single dipole to the statistical mechanics of a $1d$ Coulomb gas in a uniform external field. The fugacity of the gas, $\gamma E_c/T$, results from multiplication of the instanton action by the fluctuation factor [19].

To understand the properties of this system, we recall the standard mapping of a Coulomb gas onto the sine-Gordon model [20]. In the present context, the action of the latter is given by Eq. (4), which completes the proof of equivalence of the two approaches discussed in this Letter upon the proper identification of γ [21]. Therefore the activation temperature, $T^*(q)$, is given by Eq. (5) with $\gamma = g^{3/2}e^{-g/2}$. It is worthwhile to mention that a key element in establishing this equivalence is the factor $E_c/T \gg 1$ in the fugacity of the Coulomb gas, Eq. (8). This factor results from the large volume available to fluctuations in the array [19] geometry; i.e., no such factor exists for a single grain.

We finally turn to the discussion of the low-temperature ($T < T^*$) dc transport properties of the array. As mentioned above, in the insulating phase the fundamental excitations of our system are single-charge solitons. Referring for detailed discussion to Ref. [15], we here merely mention that in the presence of an external field, E, the dynamics of these objects is controlled by the Langevin equation

$$\frac{\partial\theta_j}{\partial t} - gE_c\left[\frac{\partial^2\theta_j}{\partial j^2} - \gamma\sin(\theta_j + jq)\right] = gE + \xi(t), \quad (10)$$

where $\xi(t)$ is a Gaussian correlated noise with $\langle\xi(t)\xi(t')\rangle_\xi = gT\delta(t-t')$ and $\partial^2\theta_j/\partial j^2 \equiv \theta_{j+1} - 2\theta_j + \theta_{j-1}$ is the discrete second derivative.

In the commensurate phase ($|q| < q^*$) the solutions of this equation are solitary configurations, $\theta_j(t) = \tilde{\theta}(j - vt)$, propagating with a constant velocity, v. Substituting this ansatz into Eq. (10), one finds $v = gE$, where $\gamma^{-1/2} \gg 1$ is the soliton length. As each of these objects carries unit charge, the current density is given by $J = env$, where $n = e^{-T^*/T}$ is the concentration of the thermally excited solitons [T^* is given by Eq. (5) with $\gamma = g^{3/2}e^{-g/2}$]. The linear dc conductivity of the array is thus given by Eq. (1). The linear I-V characteristics breaks down once the voltage drop per grain exceeds some critical value; even in the case of the largest energy gap ($q = 0$), this value is fairly low, $V_c \approx \gamma E_c \ll E_c$.

To summarize, we have considered a $1d$ array of metallic grains connected by highly conducting contacts. We have shown that the inelastic tunneling and weak charge quantization lead to the insulating behavior at the temperature below $T^* \sim E_c e^{-g/4}$. This scale is much larger than the energy $\tilde{E}_c \sim E_c e^{-g}$, where perturbative mechanisms inhibiting charge transport become sizable. The essence of this phenomenon is explained by the analogy between the array and an elastic "chain" pinned by a periodic potential. Most importantly, even an exponentially weak pinning potential leads to the formation of a "commensurate" phase where the differential capacitance and the linear conductivity exhibit activation behavior.

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[1] A. Gerber *et al.*, Phys. Rev. Lett. **78**, 4277 (1997).
[2] D. S. Golubev and A. D. Zaikin, Phys. Lett. A **169**, 475 (1992); G. Goeppert and H. Grabert, Eur. Phys. J. B **16**, 687 (2000); K. B. Efetov and A. Tschersich, Europhys. Lett. **59**, 114 (2002).
[3] I. S. Beloborodov, K. B. Efetov, A. Altland, and F. W. J. Hekking, Phys. Rev. B **63**, 115109 (2001).
[4] B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interaction in Disordered Systems*, edited by A. J. Efros, and M. Pollak (Elsevier Science Publishers, North-Holland, Amsterdam, 1985), pp. 1–153.
[5] S. V. Panyukov and A. D. Zaikin, Phys. Rev. Lett. **67**, 3168 (1991).
[6] X. Wang and H. Grabert, Phys. Rev. B **53**, 12621 (1996).
[7] Yu. V. Nazarov, Phys. Rev. Lett. **82**, 1245 (1999).
[8] V. Ambegaokar, U. Eckern, and G. Schön, Phys. Rev. Lett. **48**, 1745 (1982); G. Schön and A. D. Zaikin, Phys. Rep. **198**, 237 (1990).
[9] D. S. Golubev and A. D. Zaikin, JETP Lett. **63**, 1007 (1996).
[10] K. Flensberg, Phys. Rev. B **48**, 11156 (1993); K. A. Matveev, Phys. Rev. B **51**, 1743 (1995); A. Furusaki and K. A. Matveev, Phys. Rev. B **52**, 16676 (1995).
[11] This does not happen either for a single dot or for an array with an *elastic* tunneling. In both of these cases $\langle\theta_j(\tau)^2\rangle \sim \ln D/T$, and thus quantum fluctuations are increasingly important at small temperatures.
[12] I. L. Aleiner, P. W. Brouwer, and L. I. Glazman, Phys. Rep. **358**, 309 (2002).
[13] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, U.K., 1997).
[14] A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).
[15] A. Altland, A. Kamenev, and L. I. Glazman (unpublished).
[16] S. E. Korshunov, Pis'ma Zh. Eksp. Teor. Phys. **45**, 342 (1987) [JETP Lett. **45**, 434 (1987)].
[17] This is true for $1 - |z_\alpha|^2 \gg (T/E_c)$, which is always the case in our calculations.
[18] Doing so, we neglect temporal correlations (interactions) between instantons with $|W| > 1$ on a given grain. This approximation is justified for $T \gg \tilde{E}_c$.
[19] The massive Gaussian fluctuations bring the factor $(gE_c/T)^{2k(1-1/M)}$ [15], which for the single dot ($M = 2$, $k = 1$) reads as $gE_c/T\ln(E_c/T)$ [6]. An additional factor of g^k originates from the zero-mode integrations.
[20] See, e.g., N. Nagaosa, *Quantum Field Theory in Condensed Matter Physics* (Springer, Berlin, 1999).
[21] If transparency of all channels is small $|t_s|^2 = 1 - |r_s|^2 \ll 1$, one finds $\gamma \sim \exp\{\frac{1}{2}\sum_s \ln(1 - |t_s|^2)\} \approx \exp\{-g/2\}$, where $g \equiv \sum_s |t_s|^2$; i.e., the two definitions of γ agree with exponential accuracy.