Transport in Granular Normal Metals Materials

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Coauthors in several works: I.S. Beloborodov, A. Tschersich, A.V. Lopatin. Recent review: I.S. Beloborodov, A.V. Lopatin, V.M. Vinokur, K.B. Efetov, Rev. Mod. Phys. (2007) Hall resistivity: M.Yu. Kharitonov, K.E., PRL 2007, PRB 2008

Typical structure of a granular metal: d=50-200A



Coupling between the grains can vary: possibility of both macroscopically metallic and insulating states.

Nanoscience with numerous applications!

Experimental puzzles:

1. Strong coupling between the grains

$\sigma = \sigma_0 + \alpha \ln T$

2. Weak coupling between the grains

$$\sigma = a \exp(-b/\sqrt{T})$$

The dimensionality of the array does not seem to play any important role for both 1) and 2)!

Metal-Insulator transition?

If so, what is the reason for such a behavior?

Some experimental curves (after A. Gerber et al, PRL (1997))





FIG. 1. Resistance of sample 1 measured at zero (triangles) and 100 kOe field (circles) as a function of the inverse square root of the temperature. Sample 1 room temperature resistance is $2 \times 10^3 \Omega$.

FIG. 2. Resistance of sample 2 measured at zero (crosses) and 100 kOe field (open circles) as a function of the inverse square root of the temperature. Open circles indicate resistance measured with a constant dc current $I = 10^{-6}$ A. Solid squares are zero bias resistances approximated from I-V measurements. Sample 2 room temperature resistance is 800 Ω .

The weak coupling limit (insulator)



FIG. 3. Resistance of sample 3 as a function of temperature on a log-log scale, as measured at (zero) (×) and 100 kOe field (open circles). Open circles indicate resistance measured with a constant dc current $I = 10^{-5}$ A. Solid squares are zero bias resistances approximated from *I-V* measurements. Sample 3 room temperature resistance is 500 Ω .



FIG. 5. The resistance normalized to the room-temperature value vs $\ln T$ for two different superconducting samples. The graph on the left is for a sample with $R_{\Box}(300 \text{ K}) = 2000 \Omega/\Box$, while that on the right is for $R_{\Box}(300 \text{ K}) = 100 \Omega/\Box$. The lines are guides to the eye.

$$R = AT^{-\alpha}$$

or

$$R = A\left(1 - \alpha \ln T\right)$$

 $\alpha = 0.117$

R.W. Simon *et al*, Phys. Rev. B (1987)

Strong coupling limit. Metal?



High resistivity sample again.

Both 1) and 2) are typical for many experiments!

Why?

It is always so!

How to describe the granular metals?

Coulomb interaction plays a crucial role!



$$-e + e \implies E_c = e^2 / 2C$$

 E_c -charging energy

Other energies in the system:

 $\delta = (\nu V)^{-1}$ -mean level spacing in a grain

 $g\mathcal{S}$ -tunneling energy between the grains

g-tunneling conductance

The Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_t + \hat{H}_c$$

$$\hat{H}_{0} = \int \psi^{+}\left(\mathbf{r}\right) \left(-\frac{\nabla^{2}}{2m} + U\left(\mathbf{r}\right)\right) \psi\left(\mathbf{r}\right) d\mathbf{r}$$

$$H_t = \sum_{\mathbf{i},\mathbf{j},\alpha,\alpha'} t_{\mathbf{i}\mathbf{j}}\psi_{\alpha\mathbf{i}}^+\psi_{\alpha'\mathbf{j}} \qquad \hat{H}_c = \frac{e}{2}\sum_{\mathbf{i}\mathbf{j}} \hat{N}_{\mathbf{i}}C_{\mathbf{i}\mathbf{j}}^{-1}\hat{N}_{\mathbf{j}}$$

$$\hat{N}_{i} = \int \hat{\psi}^{+}(\mathbf{r}_{i}) \, \hat{\psi}(\mathbf{r}_{i}) \, d\mathbf{r}_{i} - \bar{N}$$

 t_{ij} tunneling amplitude from grain to grain, C_{ij} -capacitance matrix Methods of calculation:

1. Bosonization

2. Perturbation theory in the limit $g \ge 1$

(strong coupling between the grains).

Bosonization

Study of Coulomb interaction via bosonization is well known in superconductors where $\Delta_i(\tau) = \exp(2\phi_i(\tau))$ $\phi(\tau)$ is the phase. One can reduce the electron Hamiltonian to an effective phase Hamiltonian H_{eff} (Efetov (1980)) $H_{eff} = \sum \left[B \rho \rho - J \cos(2(\phi - \phi)) \right]$

$$H_{eff} = \sum_{ij} \left[B_{ij} \rho_i \rho_j - J_{ij} \cos(2(\phi_i - \phi_j))) \right]$$

Where $\rho_i = -i\partial/\partial\phi_i$ (eigenvalues are integers)

How can one "bosonize" a normal metal?

Scheme of the bosonization:

1. Hubbard-Stratonovich transformation

$$\exp\left(-\frac{e^2}{2}\sum_{\mathbf{ij}}\hat{N}_{\mathbf{i}}C_{\mathbf{ij}}^{-1}\hat{N}_{\mathbf{j}}\right) = \int \exp\left(-i\sum_{i}\int\left(\psi^*\left(\mathbf{r}_{\mathbf{i}},\tau\right)\psi\left(\mathbf{r}_{\mathbf{i}},\tau\right)d\mathbf{r}_{\mathbf{i}}-\bar{N}\right)V_{\mathbf{i}}\left(\tau\right)d\tau\right) \\ \times \exp\left(-\frac{1}{2e^2}\sum_{\mathbf{ij}}\int d\tau V_{\mathbf{i}}\left(\tau\right)C_{\mathbf{ij}}V_{\mathbf{j}}\left(\tau\right)\right)DV$$
(2)

2. Gauge transformation

$$\psi\left(\mathbf{r_{i}},\tau\right)\rightarrow e^{-i\varphi_{\mathbf{i}}(\tau)}\psi_{\mathbf{i}}\left(\mathbf{r_{i}},\tau\right),\qquad\dot{\varphi}_{\mathbf{i}}\left(\tau\right)=V_{\mathbf{i}}\left(\tau\right)$$

However, one should satisfy the fermionic boundary condition:

$$\psi\left(\mathbf{r_{i}},\tau\right)=-\psi\left(\mathbf{r_{i}},\tau+\beta\right),\ \beta=1/T$$

This is possible in the limit $T \ge \delta$ \implies Integration over the phases $\tilde{\phi}_i(\tau)$

$$\tilde{\phi}_{\mathbf{i}}\left(\tau\right) = \phi_{\mathbf{i}}\left(\tau\right) + 2\pi T k_{\mathbf{i}}\tau,$$

where $-\infty < \phi_{\mathbf{i}}(\tau) < \infty, \ \phi_{\mathbf{i}}(0) = \phi_{\mathbf{i}}(\beta)$.

 k_i are integers (winding numbers)

Final action:
$$S = S_c + S_t$$
,
 S_c is the charging energy
 S_t describes the tunneling
between the grains
 $S_t = \pi g \sum_{|\mathbf{i}-\mathbf{j}|=a} \int_0^\beta d\tau d\tau' \alpha (\tau - \tau') \sin^2 \left(\frac{\tilde{\phi}_{\mathbf{ij}}(\tau) - \tilde{\phi}_{\mathbf{ij}}(\tau')}{2}\right)$ OD limit for a single grain!
where $\alpha (\tau) = T^2 \left(\operatorname{Re} \left(\sin \left(\pi T \tau + i \delta \right) \right)^{-1} \right)^2$ $g = 2\pi \nu_0^2 t_{\mathbf{ij}}^2$

This form is analogous to the Ambegaokar, Eckern and Schon (1982) action written to describe quantum dissipation.

However, it is applicable only for $T \ge \max(\delta, g\delta)$

No quantum dissipation and no dephasing at T=0!

The phase functional can be studied without difficulties in the limits $g \ge 1$ and $g \le 1$

 $g \ge 1$ One can use renormalization group integrating over fast variations of ϕ

RG Equation:
$$\frac{\partial g(\xi)}{\partial \xi} = -\frac{1}{2\pi d}$$
 $\sigma = e^2 g(T) a^{2-d}$
Result: $g(T) = g - \frac{1}{2\pi d} \ln \frac{gE_c}{T}$ Valid as long as $g \ge 1$

 $g \leq 1$ Expansion in g.

 $B \propto E_c$ -is the energy of the lowest excitation

Result: $\sigma = 2\sigma_0 \exp(-B/T)$

At $T \ge E_c$ one has $\sigma = \sigma_0$



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

Altshuler-Aronov corrections, the same as in a homogeneous metal.

Both $\delta \sigma_1$ and $\delta \sigma_2$ can be important (weak localization corrections are assumed to be killed by a magnetic field, which is of an experimental relevance).

3D

Density of states

$$\frac{\delta\nu_2}{\nu_0} = -\frac{1}{16g_T\pi^2} \begin{cases} 2\ln^2 \frac{g_T E_C}{T} & T \gg g_T \delta, \\ \ln \frac{g_T\delta}{T} \ln \frac{gE_0^4}{T\delta^3} + 2\ln^2 \frac{E_0}{\delta} & T \ll g_T\delta. \end{cases}$$
(10)

2D

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

Hall resistivity

<u>1. Classical picture of the Hall resistivity of the granular material.</u>

R-longitudinal resistivity: information about intergrain tunneling

$$R = \left(e^2 g(T) a^{2-d}\right)^{-1} \qquad g = 2\pi \nu_0^2 t_{ij}^2$$

Q. What kind of information can one extract from the Hall resistivity?

A. Carrier density inside the grains (no dependence on the tunneling!)

Most important formula:

$$R_{H} = \frac{\sigma_{xy}}{\sigma_{xx}^{2} + \sigma_{xy}^{2}}$$

Results of an explicit calculation:

$$\boldsymbol{\sigma}_{xx} \propto t_{ij}^2 \qquad \boldsymbol{\sigma}_{xy} \propto t_{ij}^4$$

 R_H does not depend on t_{ij} !



The contribution to σ_{xy} comes from the diffuson D with non-zero space harmonics (in contrast to σ_{xx} !) Classical Hall resistivity: $R_{H}^{(0)} = \frac{H}{n^{*}ec}$, $n^{*} = An$

A-numerical coefficient, depending on the geometry of the grains (A=1 for cubic grains, $A = \pi/4$ for spherical grains)

2) Effect of the Coulomb interaction: Again, logarithmic in temperature corrections δR_H

$$\frac{\delta R_H}{R_H^{(0)}} = \frac{c_d}{4\pi g_T} \ln \left[\frac{g_T E_c}{T}\right]$$

in the region $g_T \delta \leq T \leq g_T E_c$

Insulating regime, $g_T \leq 1$

Activation law: clear explanation in terms of electron hopping from grain to grain.

How to explain $\exp(-a/\sqrt{T})$ law?

Beloborodov et al (2005): elastic and inelastic co-tunneling through many grains and random fluctuation of the Fermi energy in the grains due to charged imp

Are underdoped or optimally doped high- T_c cuprates granular?

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Logarithmic Divergence of both In-Plane and Out-of-Plane Normal-State Resistivities of Superconducting La_{2-x}Sr_xCuO₄ in the Zero-Temperature Limit

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The low-temperature normal-state resistivities of underdoped $La_{2-x}Sr_xCuO_4$ crystals with T_c of 20 and 35 K were studied by suppressing the superconductivity with pulsed magnetic fields of 61 T. Both in-plane resistivity ρ_{ab} and out-of-plane resistivity ρ_c are found to diverge logarithmically as $T/T_c \rightarrow 0$. Logarithmic divergence is accompanied by a nearly constant anisotropy ratio, ρ_c/ρ_{ab} , suggesting an unusual three-dimensional insulator.

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Metal-to-Insulator Crossover in the Low-Temperature Normal State of $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$

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We measure the normal-state in-plane resistivity of $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ single crystals at low temperatures by suppressing superconductivity with 60 T pulsed magnetic fields. With decreasing hole doping, we observe a crossover from a metallic to an insulating behavior in the low-temperature normal state. This crossover is estimated to occur near 1/8 doping, well inside the *underdoped* regime, and not at optimum doping as reported for other cuprates. The insulating regime is marked by a logarithmic temperature dependence of the resistivity over two decades of temperature, suggesting that a peculiar charge localization is common to the cuprates.



FIG. 3. Logarithmic plot of $\rho_{ab}(T)$ for BSLCO crystals in 0 T (solid lines) and in 60 T (filled circles), labeled by La concentration, x. The straight line emphasizes the log(1/T) behavior in the x = 0.84 sample and open circles are 30 T data from the long-pulse magnet. LSCO data in 0 T (dashed lines) and in 60 T (open squares), labeled by Sr concentration, y, are from Ref. [3]. All data are vertically scaled to directly compare the resistivity per CuO₂ layer in units of $(k_F l)^{-1}$.

$$\sigma = b + \frac{e^2}{\pi d\hbar} \ln T$$

Results of the fittingFor
$$La_{2-y}Sr_yCuO_4$$
:d=2.3 at y=0.08 and
d=1.08 at y=0.15

For
$$Bi_2Sr_{2-x}La_xCuO_{6+\delta}$$
:

d=4.5 at x=0.84 and d=3.57 at x=0.76

Imaging the granular structure of high-*T*_c superconductivity in underdoped Bi₂Sr₂CaCu₂O_{8+δ}

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Granular superconductivity occurs when microscopic superconducting grains are separated by non-superconducting regions; Josephson tunnelling between the grains establishes the macroscopic superconducting state1. Although crystals of the copper oxide high-transition-temperature (high-T_c) superconductors are not granular in a structural sense, theory suggests that at low levels of hole doping the holes can become concentrated at certain locations resulting in hole-rich superconducting domains2-8. Granular superconductivity arising from tunnelling between such domains would represent a new view of the underdoped copper oxide superconductors. Here we report scanning tunnelling microscope studies of underdoped Bi2Sr2CaCu2O8+à that reveal an apparent segregation of the electronic structure into superconducting domains that are ~3 nm in size (and local energy gap <50 meV), located in an electronically distinct background. We used scattering resonances at Ni impurity atoms6 as 'markers' for local superconductivity7-9; no Ni resonances were detected in any region where the local energy gap $\Delta > 50\pm$ 2.5 meV. These observations suggest that underdoped Bi2Sr2Ca-Cu.O., is a mixture of two different short-range electronic orders

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Conclusions:

1. Bosonization and diagrammatic schemes are developed to study granular metals.

2. A crossover from the logarithmic to the exponential dependence of the conductivity on temperatures is found at not very low temperatures.

3. Hall resistivity gives information about the carrier density inside the grains and has also logarithmic in temperature corrections.

4. Logarithmic dependence of the conductivity observed in many granular materials is explained.

5. High T_c superconducting cuprates may be granular, too.