

Transport in Granular Metals

I. Beloborodov, A. Lopatin, and V.V. Argonne National Laboratory







A U.S. Department of Energy laboratory managed by The University of Chicago



Outline

- Introduction: general features of granular conductors
- Metallic conductivity
- Insulating region hopping conductance
- Hopping conductivity in superconductors



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 .

Granular metals: experiment

Samples with larger coupling showed the behavior as

$$R \propto T^{-\alpha} \simeq 1 - \alpha \ln T$$
 , $\alpha \approx 0.117$

The samples are 3D, and the experiment was carried out in high magnetic field,

excluding thus the explanation of log as the weak localization behavior

Granular metals: experiment. R. W. Simon *at al*, PRL 36 (1987)



NbN in the insulating substrate

A. Gerber et al, PRL 78, 4277 (1997)

Samples (Al-Ge) with the high room temperature resistivity (weak tunneling) showed an exponential growth of resistivity as function of temperature.



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 .

$$\sigma \propto \exp\left[-\left(T_0 / T\right)^{1/2}\right] \qquad ???$$

The problem: description of transport in granular metals

Let us recall first the approach to general properties of homogeneously disordered metals

Conductivity of disordered metals







 $N = eVv_I(\varepsilon_F) \implies I = eN/\tau_I = e^2v_I(\varepsilon_F)V/\tau_I$

$$G = \frac{e^2 v_l(\varepsilon_F)}{\tau_L} = \frac{2\pi e^2}{\hbar} \left\langle t^2 \right\rangle v_l(\varepsilon_F)$$



$$G = \frac{e^2 v_l(\varepsilon_F)}{\tau}$$

 τ_L Divide the sample into blocks of size *L* and Introduce the energy associated With the lifetime within the block:

$$\Gamma_L = \frac{\pi\hbar}{\tau_L}$$

The electron diffusion over the step *L*:

$$D \sim L^2 / \tau_L$$

Now remembering that

Now let us make our separation of the blocks not fictitious, but real



The key characteristic: tunneling conductance



The tunneling conductance is measured in the units of the quantum conductance $e^2/2\hbar$

 $g_T \gg 1$ metallic transport properties

 $g_T \ll 1$ insulating behavior





Granular sample









bulk material

nanocrystal





nanocrystal



Coulomb blockade



 $g_T \ll 1$ - Coulomb blockade regime - charge quantization $g_T \gg 1$ - Charge quantization effects are exponentially small In a granular system: metal-insulator transition at $g_T \approx 1$

Granular metal



New energy scale:

$$\Gamma = g_T \delta$$

intragranule conductance $g_0 = \frac{G}{e^2 / \pi \hbar}$ tunneling conductance $g_T \ll g_0$

 δ : mean level spacing

 $E_C = e^2 / \kappa a$: charging Coulomb energy of a single granule

 $g_T \ll 1$ - Coulomb blockade regime - charge quantization $g_T \gg 1$ - Charge quantization effects are exponentially small In a granular system: metal-insulator transition at $g_T \approx 1$ Granular conductors: a new class of artificial materials with tunable electronic properties controlled at the nanoscale and composed of close-packed granules varying in size from a few to hundred nanometers



The granules are large enough to possess a distinct electronic structure, but sufficiently small to be mesoscopic in nature and exhibit effects of quantized electronic levels of confined electrons.

Conductivity of a granular sample

1. Metallic regime (strong coupling between the grains $g_{T} >> 1$)

$$D_{eff} \sim a^2 g_T \delta / \hbar = a^2 \Gamma / \hbar = a^2 / \tau_L$$

Interaction time: $\tau_{\rm T} \sim \hbar / T$



incoherent electron tunneling:

Temperature dependence of conductivity is controlled by electron tunneling between the neighboring grains

High temperature conductivity: $T > \Gamma$

Conductivity: $\sigma = \sigma_0 + \delta \sigma$ $\delta \sigma$ - interaction correction

$$\delta \sigma \approx -(e^2 / \pi \hbar) \ln(\tau_{\varphi} / \tau_C), \quad \tau_C = \hbar / E_C$$

 $\tau_{\varphi} = g_T \hbar / T:$ the dephasing time

Perturbation theory in $1/g_T$ results in the correction to conductivity

$$\frac{\delta\sigma}{\sigma_0} = -\frac{1}{2\pi dg_T} \ln\left(\frac{g_T E_C}{T}\right) \qquad \text{Efetov, Tschersich (2003)}$$

Can be understood as the renormalization of the tunneling conductance between the neighboring grains

$$\tilde{g}_T = g_T - \frac{1}{2\pi d} \ln\left(\frac{g_T E_C}{T}\right), \text{ valid as long as } \tilde{g}_T \gg 1$$

Conductivity depends logarithmically on temperature for in all dimensions !

Low temperature conductivity: $T < \Gamma$

$$\sigma = \sigma_0 + \delta \sigma$$
 where $\delta \sigma = \delta \sigma_1 + \delta \sigma_2$

 $\delta\sigma_1$ comes from the large energies, $\varepsilon > g_T \delta$, where the granular structure of the array dominates the physics. The fact that this correction is independent of the dimensionality *d* means that the tunneling of electrons is incoherent

 $\delta \sigma_2$ is similar to AA correction for homogeneously disordered metals. This is the contribution from the low energies, $\mathcal{E} < g_T \delta$, the behavior is dominated by coherent electron motion on large scales.

Large energy (small scales) correction



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

$$\gamma(\omega) = \frac{d}{d\omega} \omega \coth \frac{\omega}{2T}, \, \varepsilon_q = 2g_T \sum_{\mathbf{a}} (1 - \cos \mathbf{q} \mathbf{a})$$
$$\tilde{V}(\omega, \mathbf{q}) = \frac{2E_C(\mathbf{q})}{(\varepsilon_{\mathbf{q}} \delta - i\omega)[4\varepsilon_{\mathbf{q}} E_C(\mathbf{q}) - i\omega]}$$

$$\frac{\delta \sigma_1}{\sigma_0} = -\frac{1}{2\pi dg_T} \ln \left[\frac{g_T E_C}{\max(T, g_T \delta)} \right]$$
Effective & Techerology and the second se

21

Large scales correction



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

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

$$\frac{\delta\sigma}{\sigma_0} = -\frac{1}{2\pi dg_T} \ln\left[\frac{E_C}{\delta}\right] + \begin{cases} \frac{\alpha}{12\pi^2 g_T} \sqrt{\frac{T}{g_T \delta}}, & D=3\\ -\frac{1}{4\pi^2 g_T} \ln\frac{g_T \delta}{T}, & D=2\\ -\frac{\beta}{4\pi} \sqrt{\frac{\delta}{Tg_T}}, & D=1 \end{cases}$$

Phase Diagram of Granular Metallic Systems





2. Insulating phase

Typical experimental dependence: $\sigma \sim e^{-A/T^p}, \quad p \approx 1/2$

B. Abeles, P. Sheng, M. D. Coutts, and Y. Arie, Adv. Phys. 24, 407 (1975).

Earlier attempts to explain the conductivity temperature dependence were based on

ASCA phenomenological model:

Thickness of the insulating layer between two grains is $t \sim R$ proportional to grain sizes



Coulomb energy $E_c \sim e^2/R$

Tunneling probability $P \sim e^{-2t/L}$ L – localization length of the insulating layer

Optimization of $e^{-e^2/RT-2t/L}$ under constraint $R \sim t$

results in **p=1/2** dependence

ASCA model ??????? M. Pollak, C. Adkins (1992), R. Zhang, B.I. Shklovskii PRB (2004)

1. Capacitance disorder cannot remove the Coulomb gap completely





10 0.30

0.35

0.40

0.45

 $T^{-1/2}$ (K^{-1/2})

0.50

2. Recent experiments showed the 10 p=1/2 law for periodic arrays. 2d array of gold particles of 0.006 0.008 0.010 ⁶° [1Ω.] size ~ 5.5 nm. Particle sizes are controlled 10 within 5% accuracy. 10 Bilaye Trilave Parthasarathy, X.-M. Lin, K. Elteto, T. F. Rosenbaum, H. M. Jaeger PRL 2004 Tetralave Thick film 101 0.12 0.16 0.20 D. Yu, C. Wang, B. L. Wehrenberg, P. Guyot-Sionnest PRL 2004 T^{-1/2} [K^{-1/2}] T.B. Tran, et al, PRL 2005 FS law was also observed in the nanocrystal arrays of semiconducting onductance quantum dots I- Si ភ = Si : B

Yakimov, et al, JETP Lett. 2003

Hopping conductivity.

Optimization of tunneling probability: variable range hopping



$$\Gamma_{ij}^{0} = \gamma_{ij}^{(0)} \exp\left(-2r_{ij}/\xi\right) \exp\left(-\varepsilon_{ij}/T\right)$$



In the presence of Coulomb interaction the site energies are renormalized due to interactions with the surrounding sites.

Denoting the renormalized energies as $\tilde{\varepsilon}_i$, let us estimate the energy cost for an electron transfer from a filled, i, to an empty donor, j:



 e^2/r $\sigma \propto \exp(-r/\xi) \exp(-e^2/\kappa rT)$

Tunneling probability

Probability to overcome Coulomb barrier

Optimizing with respect to *r*, one finds:

$$\sigma \propto \exp[-(T_{ES} / T)^{1/2}]$$
$$T_{ES} = \frac{e^2}{\kappa \xi}$$



Coulomb interactions and hopping conductivity in granular arrays

$$H = \sum_{i} \mu_{i} n_{i} + \sum_{ij} n_{i} E_{ij}^{c} n_{j}$$
$$E_{i}^{\pm} = E_{i}^{c} \pm \mu_{i}$$

This looks exactly like the impurity levels in doped semiconductors:

VRH?

The puzzle of tunneling







tunneling via virtual states of intermediate grains



Elastic cotunneling mechanism

T <



Inelastic cotunneling mechanism

T

Cotunneling allows for charge transport through several junctions at a time by cooperative electron motion.

At low temperature the sequential tunneling is exponentially suppressed by the Coulomb blockade.

In this case, a higher-order tunneling process transferring electron charge coherently through two junctions can take place. The excess electron charge at the grain exists only virtually.

$$\Gamma_{i \to f} = \frac{2\pi}{\hbar} \left| \sum_{\psi} \frac{\langle i | \mathcal{H}_{\text{int}} | \psi \rangle \langle \psi | \mathcal{H}_{\text{int}} | i \rangle}{E_{\psi} - E_{i}} \right|^{2} \delta(E_{i} - E_{f})$$

- 1. There are 2 channels which add coherently
- The leads have macroscopic number of electrons. Therefore, with the overwhelming probability the outgoing electron will come from a different state than the one which the incoming electron occupies → After the process an electron-hole excitation is left in the grain.

D. V. Averin and A. A. Odintsov, Phys. Lett. A 140, 251 (1989)D. V. Averin and Y. V. Nazarov, Phys. Rev. Lett. 65, 2446 (1990).

Elastic co-tunneling mechanism



hopping probability $\propto \exp[-(2s/\xi_{el}) - (e^2/\tilde{\kappa}Tas)]$

Elastic co-tunneling mechanism

Tunneling through a chain of grains Model: Short range on-site interaction: Electron (hole) excitation energies 0 Ν N+1 $E_i^{\pm} = E_i^c \pm \mu_i$ NTunneling probability is a product $P_{el} = \delta(\xi_{N+1} - \xi_0) g_0$ k=1of elementary probabilities $P_k = \frac{g_k \delta_k}{\pi \tilde{E}_k}$ $\tilde{E}_k = 2 \left(\frac{1}{E_k^+} + \frac{1}{E_k^-} \right)^{-1}$ g_k - conductance between k-th and k+1 - st grains In terms of geometrical averages along the tunneling path the probability is $P_{el} = \bar{g}^{N+1} \left(\frac{\bar{\delta}}{\pi \bar{E}}\right)^N \delta(\xi_{N+1} - \xi_0) \quad \ln \bar{E} = \frac{1}{N} \sum_{k=1}^N \ln \tilde{E}_k$ Effective localization length: $P_{el} \sim e^{-2s/\xi_{el}}$ s – distance along the path $\xi_{el} = \frac{2a}{\ln(\bar{E}\pi/\bar{a}\,\bar{\delta})}$

Hopping conductivity in the regime of elastic cotunneling

Variable range hopping: Phonon assisted tunneling:

$$I \sim e^{-2r/\xi_{el} - \varepsilon/T}$$

(Granular metals: electrons also contribute to the energy relaxation)

E.S. DOGS $\nu_g(\varepsilon) \sim (\tilde{\kappa}/e^2)^d |\varepsilon|^{d-1} \longrightarrow r \varepsilon \tilde{\kappa}/e^2 \sim 1$

Minimization results in the E.S. law:

$$\sigma \sim e^{-(T_0/T)^{1/2}} \qquad T_0 \sim e^2 / \tilde{\kappa} \xi_{el} \qquad \xi_{el} = \frac{2a}{\ln(\bar{E}\pi/\bar{g}\,\bar{\delta})}$$

Hopping distance r within the energy shell ϵ is given by

Nonlinear conductivity at strong electric fields



Inelastic cotunneling: single granule



Hopping conductivity: inelastic cotunneling

Hopping through a chain of grains via inelastic cotunneling



$$P_{in} = \frac{1}{4\pi T} \frac{\bar{g}^{N+1}}{\pi^{N+1}} \left[\frac{4\pi T}{\bar{E}}\right]^{2N} \frac{|\Gamma(N+\frac{i\Delta}{2\pi T})|^2}{\Gamma(2N)} e^{-\frac{\Delta}{2T}}$$

 $\Delta = \xi_N - \xi_0$ - difference of the energies of initial and final states

Low electric field (linear regime)

Optimization under constraint $Na \tilde{\kappa} \Delta/e^2 \sim 1, \ (N \gg 1),$

results in the ES law:

$$\sigma \sim e^{-(T_0(T)/T)^{1/2}},$$
$$T_0(T) \sim e^2/\tilde{\kappa}\,\xi_{in}(T)$$

$$\xi_{in}(T) = \frac{2a}{\ln[\bar{E}^2/16\pi T^2\bar{g}]}$$

Crossover temperature between elastic and inelastic regimes

$$\xi_{in} = \xi_{el} \quad \longrightarrow \quad T = \sqrt{\delta E_c}$$

Hopping conductivity via inelastic cotunneling: strong fields

Low temperatures $T \rightarrow 0$:

$$P_{in}(T=0) = \frac{2^{2N}\pi}{(2N-1)!} \frac{|\Delta|^{2N-1}}{\bar{E}^{2N}} \left(\frac{\bar{g}}{\pi}\right)^{N+1}$$

Hopping distance can be found as in the case of elastic cotunneling

$$\left. \begin{array}{c} e\mathcal{E}r\sim\Delta\\ r\,\Delta\,\tilde{\kappa}/e^2\sim1 \end{array} \right\} \quad \Longrightarrow \quad r\sim\sqrt{e/\tilde{\kappa}\mathcal{E}}$$

Using that $N \sim r/a, \quad N \gg 1$

$$j \sim j_0 \ e^{-(\mathcal{E}_0/\mathcal{E})^{1/2}},$$

$$\mathcal{E}_0(\mathcal{E}) \sim \frac{e}{\tilde{\kappa} a^2} \ln^2[\bar{E}^2/e^2\mathcal{E}^2 a^2\bar{g}]$$

Applicability:

Nonlinear regime: ${\cal E}ea \gg T$

Inelastic cotunneling dominates elastic one: $\mathcal{E}ea \gg \sqrt{\delta E_c^0}$

Mapping onto the classical Coulomb gas

Phase action:
$$S_{AES} = -\frac{1}{2e^2} \sum_{ij} \int d\tau \, \dot{\phi}_i \, C_{ij} \, \dot{\phi}_j + S_t[\phi],$$

 $S_t[\phi] = \frac{1}{2\pi} \sum_{\langle ij \rangle} g_{ij} \int_{-\infty}^{+\infty} d\tau_1 \, d\tau_2 \frac{e^{i\phi_{ij}(\tau_1) - i\phi_{ij}(\tau_2)}}{(\tau_1 - \tau_2)^2}$

Mapping onto the Coulomb gas:

Expand partition $Z = Z_0 \sum_{N=1}^{\infty} \langle S_t^N[\phi] \rangle / N!$.



Averaging over the Coulomb action gives rise to the classical charges:

$$\left\langle e^{i\sum_{n}\phi_{i}(\tau_{n})e_{n}}\right\rangle = e^{-U^{c}}, \qquad U^{c} = \frac{1}{2}\sum_{n_{1},n_{2}}E^{c}_{i_{n_{1}}i_{n_{2}}}|\tau_{n_{1}} - \tau_{n_{2}}|e_{n_{1}}e_{n_{2}} \\ \text{Internal interaction:} \\ \frac{g}{2\pi}\frac{1}{(\tau_{n} - \tau_{n}')^{2}} = e^{-\mathcal{U}_{n}^{q}(\tau_{n} - \tau_{n}')}, \qquad \text{Id Coulomb interaction along the "time" axis} \\ \mathcal{U}_{n}^{q} = \ln[2\pi(\Delta\tau_{n})^{2}/g]$$

Total classical energy (internal +Coulomb parts) :

$$\sum_{n=1}^{N} \mathcal{U}_n^q + U_{4N}^c$$

 $U^q =$

Elastic cotunneling is beyond the AES approach



Feynman diagram

maps onto the Coulomb gas:

The electron world line representing the probability of elastic cotunneling from the 0th to the *N*+1st grain.



Inelastic cotunneling is described within the AES approximation



General process: tunneling through the granule chain





Experiment

H. Jaeger's group at the University of Chicago

Transmission electron micrographs showing the region between the in-plane electrodes for a) bilayers, b) trilayers, c)tetralayers and d) thick films. The darker regions on top and bottom of a-c are the electrodes. The insets on the right sides are diffraction patterns computed by fast fourier transform. The insets on the left sides of panels a&c are the zoomed-in images. The scale bars correspond to 200nm (a-c) and 40nm (d, all insets).







a) Zero-bias conductance g_0 versus inverse temperature $T^{-1/2}$ for multilayer and thick film data. Inset: For the high-temperature range, where the multilayer data in the main panel deviate form the dotted lines, g_0 has been replotted as a function of T^{-1} , indicating Arrhenius behavior from 100-160K(b-e) Evolution of the I – V characteristics with temperature for bilayers (b,c) and thick films (d,e). Panels (c) and (e) are log-log plots of the data shown in the plots above them. The straight solid lines are guide to the eye, indicating power law behavior. Insets to b&d: Temperature dependence of the hopping distance N obtained from $g_0(T)$ and the I-V power-law exponents obtained from panels c&e in the range 2V < V < 7V.



Determining T_{01} in the Efros-Shklovski hopping formula we find (at T = 10K), N = 4 for multilayers and N= 4-5 for the thick films.

$$I_{in} \sim V \left[\frac{g_T}{h/e^2}\right]^j \left[\frac{(eV)^2 + (k_B T)^2}{E_C^2}\right]^{j-1}$$

N=4 or j=3 implies I ~ V ⁵. This is what was experimentally observed

Unresolved questions (or what we do not know):

R~logT behavior

- Transport in the arrays of quantum dots (semiconductors)
- •Theory of Hall effect
- Phononless (hopping) transport

1. Periodic granular array:

Activation conductivity $~~\sigma~\sim~e^{-\Delta_M(g)/T}$

T=0: Insulator to metal transition occurs at $g_c = \frac{1}{\pi z}\,\ln(E_c/g\delta)$



2. Arrays with electrostatic disorder:

Linear regime:

$$\sigma \sim e^{-(T_0/T)^{1/2}}, \quad T_0 \sim e^2/\tilde{\kappa}\xi, \quad \xi \sim \begin{cases} \frac{2a}{\ln(\bar{E}\pi/\bar{g}\,\bar{\delta})}, & T < \sqrt{E_0^c\,\bar{\delta}} & \text{elastic} \\ \frac{2a}{\ln(\bar{E}\pi/\bar{g}\,\bar{\delta})}, & T < \sqrt{E_0^c\,\bar{\delta}} & \text{elastic} \end{cases}$$

Nonlinear regime:

$$j \sim j_0 \ e^{-(\mathcal{E}_0/\mathcal{E})^{1/2}}, \qquad \mathcal{E}_0 \sim \begin{cases} \frac{e}{\tilde{\kappa} a^2} \ \ln^2[\bar{E}\pi/\delta\bar{g}], & \mathcal{E}ea < \sqrt{\delta E_c^0} \\ \frac{e}{\tilde{\kappa} a^2} \ \ln^2[\bar{E}^2/e^2\mathcal{E}^2a^2\bar{g}], & \mathcal{E}ea > \sqrt{\delta E_c^0} \end{cases} \text{ inelastic}$$

Hopping conductivity in granular superconductors

Weak coupling regime $g_{T} << 1$

Simplest Model: Coulomb energy + Josephson couplings	$H = 4\sum_{ij} \hat{n}_i E_{ij}$	$_{j}\hat{n}_{j} + \sum_{ij} J_{ij} \cos(\phi_{i} - \phi_{j})$
ϕ – Cooper pair phase	$\hat{n}=-i\partial/\partial\phi$	- Cooper pair number operator
Anderson-Abeles criterion for the global superconductivity development $J > E_c$ Applicable at g<<1 - as long as the charge renormalization effects may be neglected.		
We assume g<<1 g ∆ <<	Ec N	o global coherence

In the presence of electrostatic disorder the transport can be mediated by Cooper pair hopping

Questions:

- 1. Electron hopping transport in the presence of the superconducting gap -?
- 2. Multiple cotunneling in the presence of the gap ?
- 3. Conductivity temperature dependence -?

Parity term.

Energy of a single superconducting grain.

$$E = n^2 E_c - Vn + P(n+p)\Delta$$

Charging energy Random potential

n - number of excessive electrons, counted with the respect to N_0 – the total charge of the neutral state

Total number of electrons $N=N_0 + n$.

Parity effect: A state with odd number of electrons has an extra energy △.

Parity function:
$$P(n) = \begin{cases} 0 - \text{even n} \\ 1 - \text{odd n} \end{cases}$$

Matveev, Averin Nazarov 1992

p=1,2 – the total charge N of the neutral state can be even or odd.

The energy E is at minimum with respect to electron number n for a given potential V.



Single grain model: Electron occupation number

Electron excitation energy:

$$\mathcal{E}_{\pm} \equiv E(n+1) \pm E(n)$$

= $(\pm 2n+1)E_c \mp V + \Delta \cos \pi (n+p)$

Occupation number n jumps $n \to n \pm 1$ at $\mathcal{E}_{\pm} = 0$

Pair excitation energy:

$$\mathcal{E}_{2\pm} \equiv E(n+2) - E(n)$$
$$= 4(\pm n+1)E_c \mp 2V$$

Occupation number n jumps $n \to n \pm 2$ at $\mathcal{E}_{2\pm} = 0$

Depending on the mutual relation of E_C and \triangle one finds qualitatively different dependences n(V):

1. Charging energy dominates $E_{C} > \Delta$.

Modified Coulomb staircase:

Occupation number changes by one

n→n+1 at

 $V_n = (2n+1)E_c + \Delta \cos \pi (n+p)$

Pair excitations are gapped for all V! $\mathcal{E}_{2\pm} > 2(E_c - \Delta)$

2. Parity term dominates $\triangle > E_{C.}$

Usual staircase but for Cooper pairs:

Occupation number changes by two

n→n+2 at $V_n = E_c(4n+2)$

Electron excitations are gapped for all V!

$$\mathcal{E}_{\pm} > \Delta - E_c$$

Electron number and excitation energies as functions of V, $E_c > \Delta$



Electron number and excitation energies as functions of V, Δ



Density of states

Long range Coulomb interaction

$$H_c = \sum_{ij} n_i E_c^{ij} n_j - V_i n_i + \Delta P(n_i + p_i) \qquad E_c \sim e^2/2 \,\tilde{\kappa} \, r \quad r \to \infty$$

Main conclusions of the single grain model stay the same!

1. $E_C > \triangle$: Gapless electrons.

Pair gap: $2(E_{C}- \triangle)$

2. $E_C < \triangle$: Gapless pairs.

Electron gap: \triangle -E_C

DOS: Efros-Shklovskii approach:

Energy to replace electron form i to j

$$\mathcal{E}_{-+}^{ij} = \mathcal{E}_{-}^i + \mathcal{E}_{+}^j - 2E_c^{ij} > 0$$

$$\nu_1(\varepsilon) = \alpha_{d1} \, (\tilde{\kappa}/e^2)^d \varepsilon^{d-1}$$

Energy to replace a pair from i to j

$$\mathcal{E}_{2-2+}^{i j} = \mathcal{E}_{2-}^{i} + \mathcal{E}_{2+}^{j} - 8E_c^{ij} > 0$$

$$\nu_2(\varepsilon) = \alpha_{d2} \, (\tilde{\kappa}/(2e)^2)^d \varepsilon^{d-1}$$

The difference is due to the Cooper pair doubled charge only, therefore $\alpha_{d1} \approx \alpha_{d2}$.



Multiple cotunneling in granular superconductors. Hopping Conduct

Electron hopping regime: Ec $>> \triangle$.



Hopping conductivity

$$\sigma \sim e^{-(T_0/T)^{1/2}} \quad T_0 \sim e^2 / \tilde{\kappa} \xi_{el}$$

The presence of the gap results in a small correction to the localization length:

$$\xi_{el} = \frac{2 a}{\ln(\bar{E}(\Delta) \pi / \bar{g} \,\bar{\delta})}$$
$$\bar{E}(\Delta) = \bar{E}(0) + c\Delta$$

Noticeable negative magneto-resistance.

Inelastic regime



 T>>∆: ES law with essentially unaffected localization length:

$$\xi_{in}(T) = \frac{2 a}{\ln[\bar{E}^2/16\pi T^2 \bar{g}]}$$

2. T<< ∆: Strong suppression of the inelastic cotunneling !

$$\sigma \sim \exp\left[-N\left(\ln(\bar{E}^2/4\bar{g}T\Delta) + 2\Delta/T\right)\right]$$

N is the typical tunneling order:

$$N = \sqrt{b e^2 / 16 a \tilde{\kappa} \Delta} \sim \sqrt{E_c / \Delta}$$

Giant negative magneto-resistance!

Transport phase diagram in the magnetic field in the EH regime



 $T_1 \approx 0.1 \sqrt{E_c \delta}$ - Crossover between elastic and inelastic regimes at Δ =0 $T_2 \approx \xi_{el} \Delta/a$ - Crossover between the elastic and inelastic activation behavior at H=0 $T_3 \approx \xi_{in} \Delta/a$ -Crossover between ES and activation inelastic regimes

Cooper pair hopping (CPH) regime

Hopping of Cooper pairs can be described in terms of the effective Hamiltonian

$$H = 4\sum_{ij} \hat{n}_i E_c^{ij} \hat{n}_j - 2\sum_i \hat{n}_i V_i + \frac{1}{2} \sum_{\langle ij \rangle} J_{ij} e^{i\varphi_i - i\varphi_j}$$

arphi - Cooper pair phase $\hat{n} = -i\partial/\partial \varphi$ - Cooper pair number operator $J_{ij} = g_{ij}\pi\Delta/2_{\sigma}$ - Josephson couplings

Tunneling amplitude via perturbation theory in J:

$$A \sim \prod_{i=1}^{N} J_{i,i+1} / \tilde{\mathcal{E}}_{2}^{i}, \qquad \tilde{\mathcal{E}}_{2}^{i} = 2 / [1 / \mathcal{E}_{2+}^{i} + 1 / \mathcal{E}_{2-}^{i}]$$

 $\mathcal{E}_{2+}^{i}, \quad \mathcal{E}_{2-}^{i}$ - Cooper pair creation and annihilation energies

Tunneling probability $P = A^* A \sim e^{-2r/\xi_{CPH}}$

Conductivity:

$$T_0 \sim e^2 / \tilde{\kappa} \, \xi_{CPH}$$

$$\xi_{CPH} = \frac{a}{\ln(8\bar{E}/\pi\bar{g}\Delta)}$$

Positive magnetoresistance !

Experimental data

Granular aluminum samples.

A. Gerber, A. Milner, G. Deutscher, M. Karpovsky, A. Gladkikh PRL 1997.

Weak coupling insulating regime.

Grain size ~ 120A

 $Ec>> \land \rightarrow Electron hopping !$

Theory predicts the giant negative magneto-resistance at T< △.

Explanation: suppression of the inelastic cotunneling by the superconducting gap.



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 $2 \times 10^3 \Omega$.

Hopping conductivity in superconductors: Results

