

Hopping transport in granular metals

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I. Granular metallic systems.

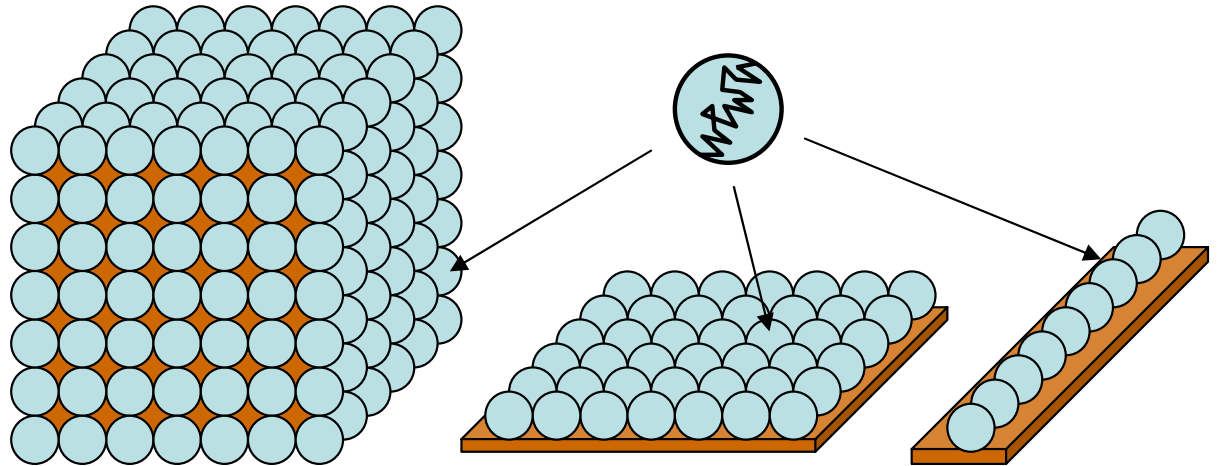
1. Effect of the Coulomb blockade in a single grain.
2. Regular periodic systems: Metallic and insulating phases at low temperature. Metal to insulator transition.
3. Granular arrays with electrostatic disorder. Hopping conductivity.

II. Hopping conductivity in granular superconductors.

Granular metallic systems

Array of metallic nanoparticels in an insulating substrate:

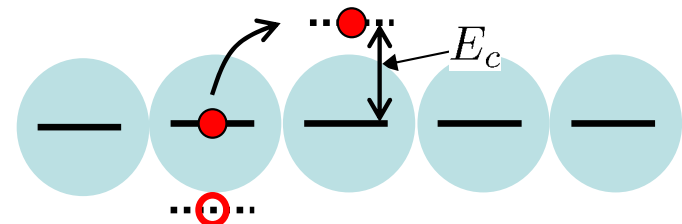
Grains are assumed to be dirty.
Electron motion inside each grain is diffusive



An electron spends some time inside a grain before it tunnels to a neighboring grain

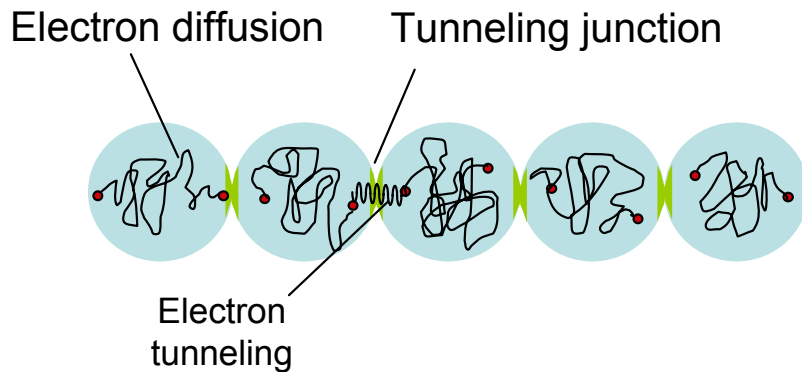
Coulomb interaction

Charging energy $E_c = e^2 n^2 / 2C$



Coulomb blockade.

Example: Motion of an electron in a 1D granular wire:



Important physical parameters of a granular sample:

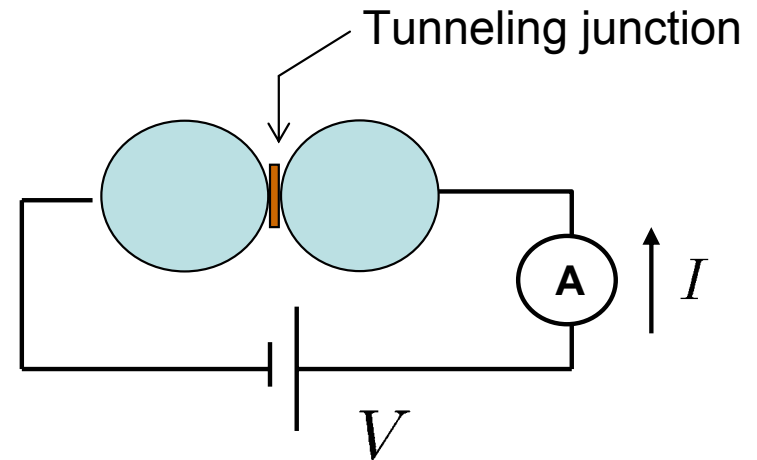
Tunneling conductance: $g = \frac{I}{V}$

Quantum conductance: $g_Q = \frac{e^2}{\hbar}$

Corresponding resistance: $R_Q = 4.05 \text{ k}\Omega$

Dimensionless tunneling conductance: $g \rightarrow \frac{g}{g_Q}$

Internal conductance of a single grain: g_0



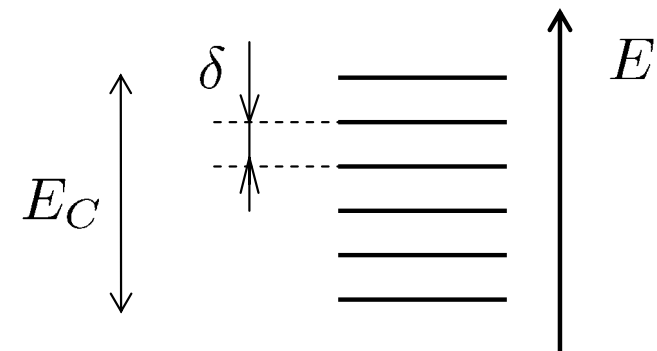
Granularity assumes :

$$g \ll g_0$$

Coulomb energy: $E_c \approx e^2/a$

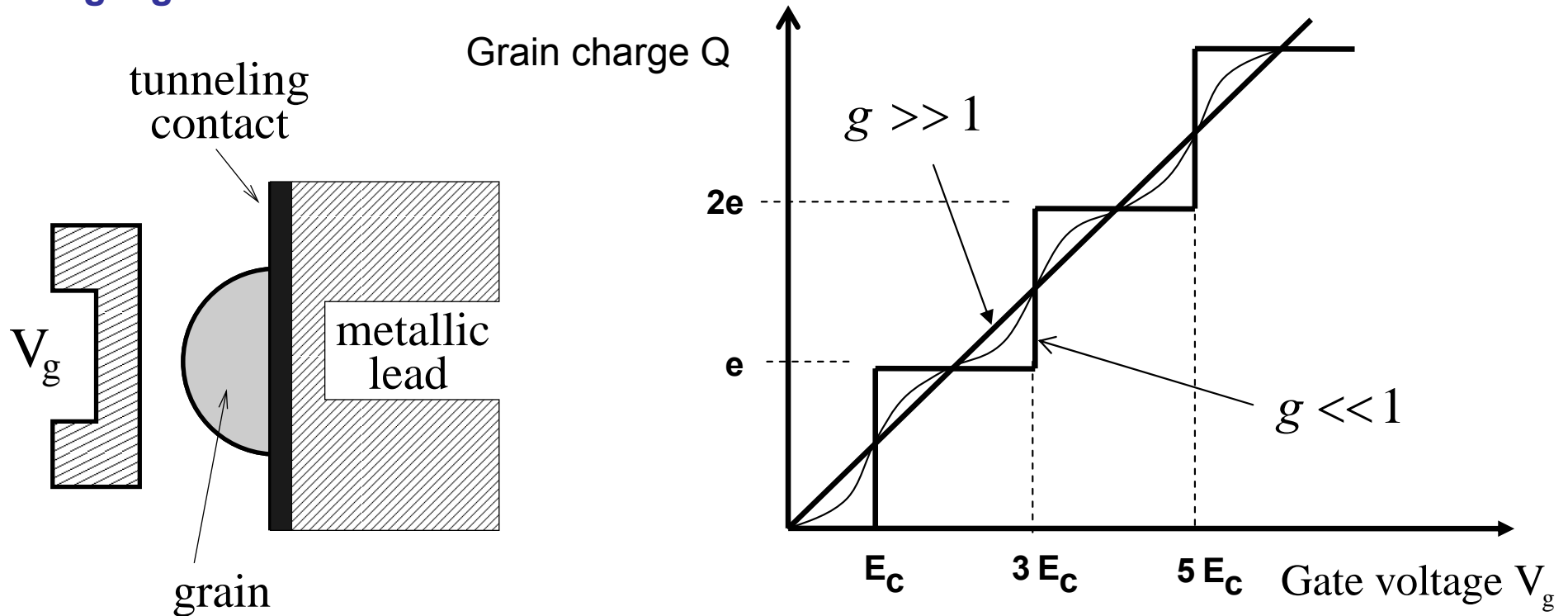
Mean distance between the energy levels δ :

For realistic grains: $E_c \gg \delta$



Coulomb blockade in a single grain.

Single grain + metallic lead



$g \ll 1$ - Coulomb blockade regime - charge quantization.

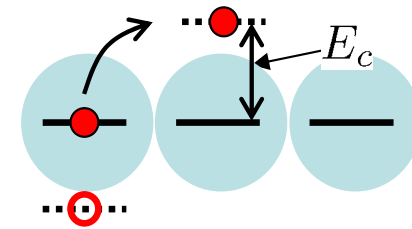
$g \gg 1$ - Charge quantization effects are exponentially small.

For a granular system one expects a metal insulator transition at $g \sim 1$.

Conductivity of a periodic granular sample in the insulating regime

Weak coupling between the grains: $g \ll 1$

Periodic granular sample :
activation conductivity with the Coulomb gap - E_c



K.B. Efetov, A. Tschersich (2003)

$$\sigma \sim e^{-\Delta_M/T}, \quad \Delta_M = E_c, \quad g \ll 1$$

Mott gap Δ_M is reduced due to intergranular electron tunneling

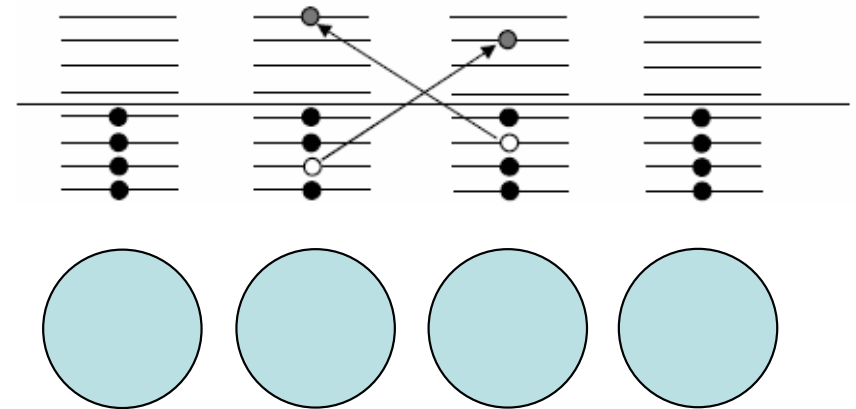
Weak coupling: perturbation theory

$$\Delta_M = E_c - \frac{2gz}{\pi} E_{eh} \ln 2, \quad gz \ll 1$$

E_{eh} - energy to create an electron-hole excitation

z - coordination number

$$\Delta E = - \sum_k \frac{|V_{k,0}|^2}{E_k - E_0}$$

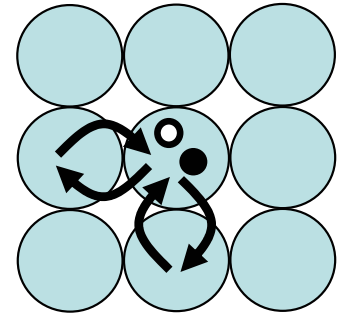


Reduction of the Mott gap at stronger coupling. Mott transition

1. Self consistent nearest neighbor hopping approximation

Mott gap is suppressed exponentially

$$\Delta_M = c g E_c e^{-\pi g z}, \quad g z \gg 1 \quad c - \text{const}$$



2. The above result neglects electron motion on scales of many grains

Diffusive time $\tau \sim D^{-1} r^2$ r – distance, D – diffusive coefficient

Diffusive processes are suppressed as long as $\tau > \Delta_M^{-1}$.

$D = g \delta a^2$, δ - mean energy level spacing in a grain

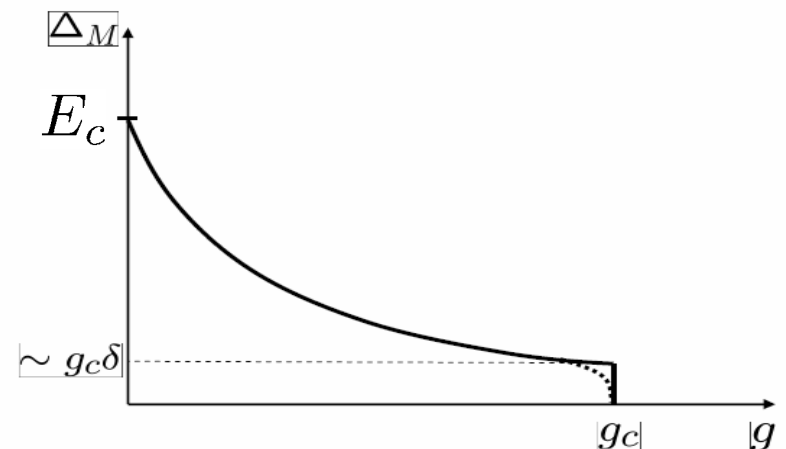
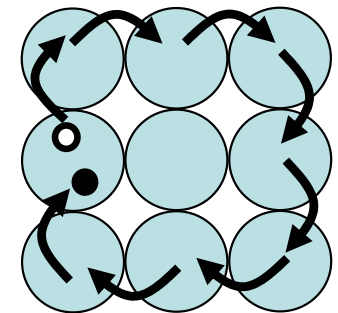
Taking: $r \sim$ grain size a

$$\Delta_M > g \delta \implies g < g_c = \frac{1}{\pi z} \ln(E_c / g \delta)$$

Mott transition at $T=0$

$g > g_c$ – metal

$g < g_c$ – insulator



Experimental observations, earlier explanation attempts

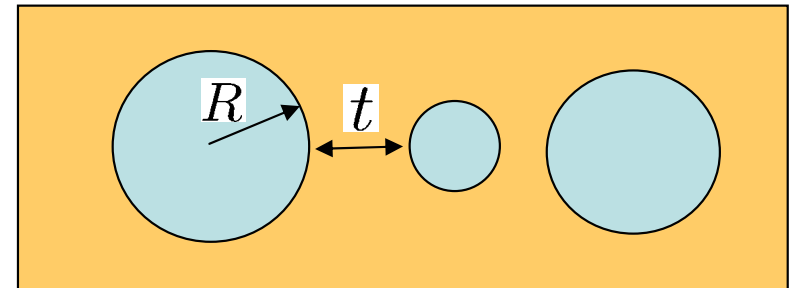
Typical experimental dependence: $\sigma \sim e^{-A/T^p}$, $p \approx 1/2$

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
proportional to grain sizes $t \sim R$



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 $R \sim t$

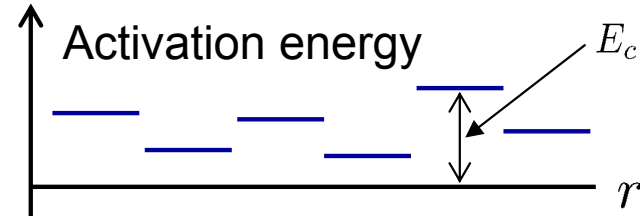
results in **$p=1/2$** dependence

Irregular arrays, role of the electrostatic disorder.

Critique of ASCA model: **M. Pollak, C. Adkins (1992), R. Zhang, B.I. Shklovskii PRB (2004)**

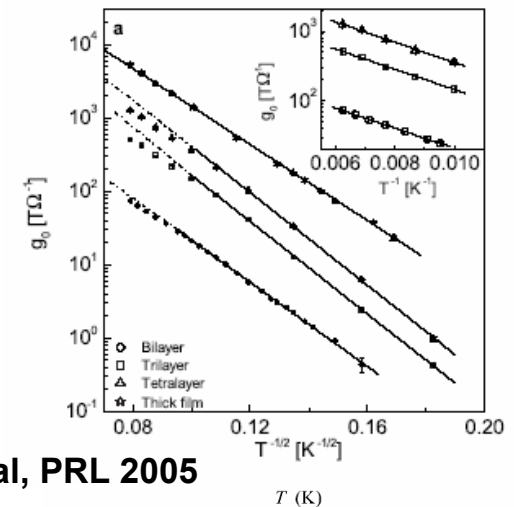
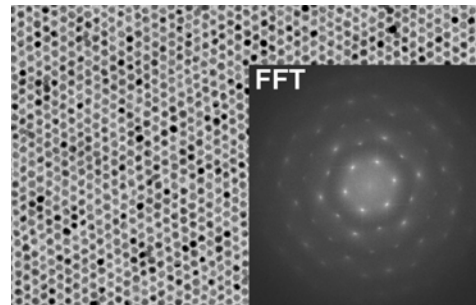
1. Capacitance disorder cannot remove the Coulomb gap completely

→ ASCA model cannot explain the observed behavior at low temperature



2. Recent experiments showed the $p=1/2$ law for periodic arrays.

2d array of gold particles of size ~ 5.5 nm.
Particle sizes are controlled within 5% accuracy.



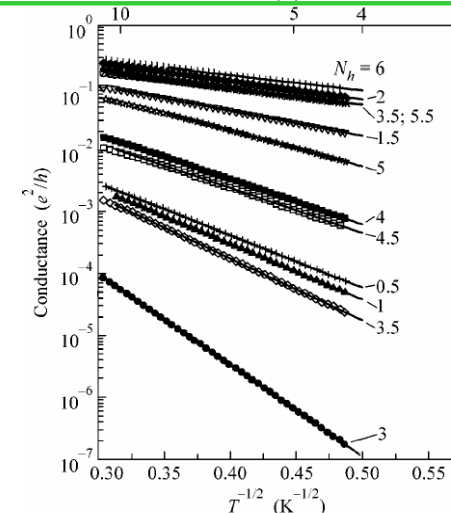
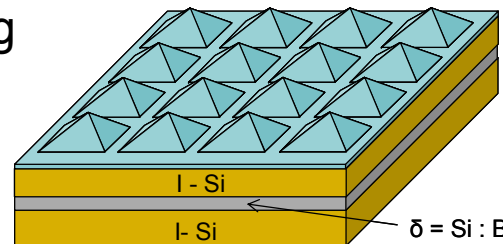
Parthasarathy, X.-M. Lin, K. Elteto, T. F. Rosenbaum, H. M. Jaeger PRL 2004

D. Yu, C. Wang, B. L. Wehrenberg, P. Guyot-Sionnest PRL 2004

T.B. Tran, et al, PRL 2005

ES law was also observed in the nanocrystal arrays of semiconducting quantum dots

Yakimov, et al, JETP Lett. 2003



Hopping conductivity: Random potential model

Two crucial ingredients of the hopping conductivity:

- a) Finite density of states in the vicinity of the Fermi level.
- b) Ability to hop on distances larger than a single grain size.

- a) Capacitance fluctuations – not enough. **R. Zhang, B.I. Shklovskii PRB (2004)**

Model: random potential is applied on each grain

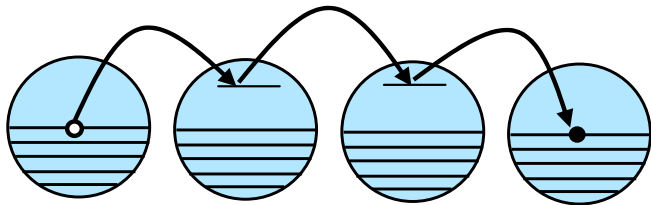
Coulomb part of the Hamiltonian:
$$H = \sum_i V_i \hat{n}_i + \sum_{ij} \hat{n}_i E_{ij}^c \hat{n}_j$$

\hat{n}_i - electron density on grain i V_i - random potential on grain i

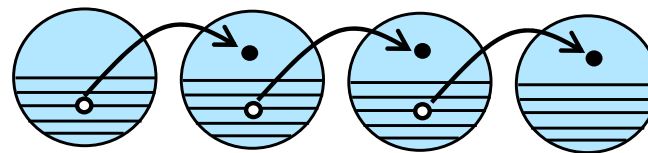
Random potential gives rise to the flat density of states.

Coulomb correlations → Efros-Shklovskii suppression of DOS.

- b) Tunneling via virtual states of intermediate grains **D. A. Averin and Yu. V. Nazarov, PRL (1990)**



Elastic cotunneling mechanism



Inelastic cotunneling mechanism $T > \sqrt{E_0^c \delta}$

Density of states

Weak intergrain tunneling coupling \rightarrow quantized charge \rightarrow classical description

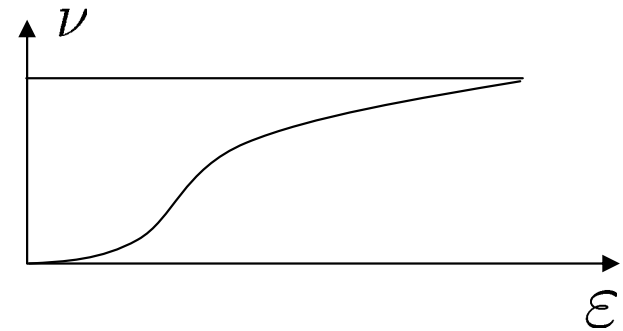
$$H = \sum_i V_i n_i + \sum_{ij} n_i E_{ij}^c n_j \quad n_i = 0, 1 - \text{classical electron charge}$$

The model is essentially equivalent to the one studied by Efros and Shklovskii.

Coulomb gap: Efros-Shklovskii result

$$\nu_g(\varepsilon) \sim (\tilde{\kappa}/e^2)^d \varepsilon^{d-1}$$

$\tilde{\kappa}$ – effective dielectric constant



For granular metals ES result gives the density of GROUND states.

Description in terms of the classical model is degenerate:
Many electron states within each grain correspond to the same charge.

DOS:

$$\nu(\varepsilon) \sim \nu_0 (\varepsilon \tilde{\kappa} / e^2)^d$$

In the Mott criterion for finding the hopping distance r within the energy shell \mathcal{M}

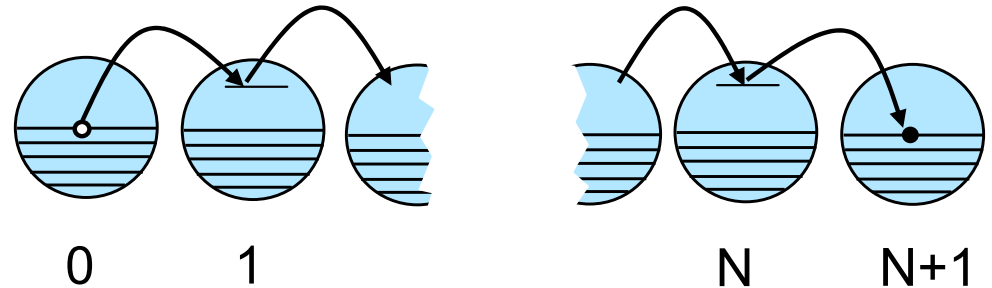
$$r^d \int_0^\varepsilon d\varepsilon' \nu_g(\varepsilon') \sim 1 \quad \text{one has to use the density of GROUND states (DOGS)}$$

Elastic cotunneling mechanism

Tunneling through a chain of grains

Model: Short range on-site interaction:
Electron (hole) excitation energies

$$E_i^\pm = E_i^c \pm \mu_i$$



Tunneling probability is a product of elementary probabilities

$$P_{el} = \delta(\xi_{N+1} - \xi_0) g_0 \prod_{k=1}^N P_k$$

$$P_k = \frac{g_k \delta_k}{\pi \tilde{E}_k} \quad \tilde{E}_k = 2 \left(1/E_k^+ + 1/E_k^- \right)^{-1}$$

g_k - conductance between k-th and k+1 - st grains

In terms of geometrical averages along the tunneling path the probability is

$$P_{el} = \bar{g}^{N+1} \left(\frac{\bar{\delta}}{\pi \bar{E}} \right)^N \delta(\xi_{N+1} - \xi_0) \quad \ln \bar{E} = \frac{1}{N} \sum_{k=1}^N \ln \tilde{E}_k$$

$$P_{el} \sim e^{-2s/\xi_{el}} \quad s - \text{distance along the path}$$

Effective localization length:

$$\xi_{el} = \frac{2a}{\ln(\bar{E} \pi / \bar{g} \delta)}$$

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)

Hopping distance r within the energy shell ε is given by $r^d \int_0^\varepsilon d\vare' \nu_g(\vare') \sim 1$

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

Minimization results in the E.S. law:

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

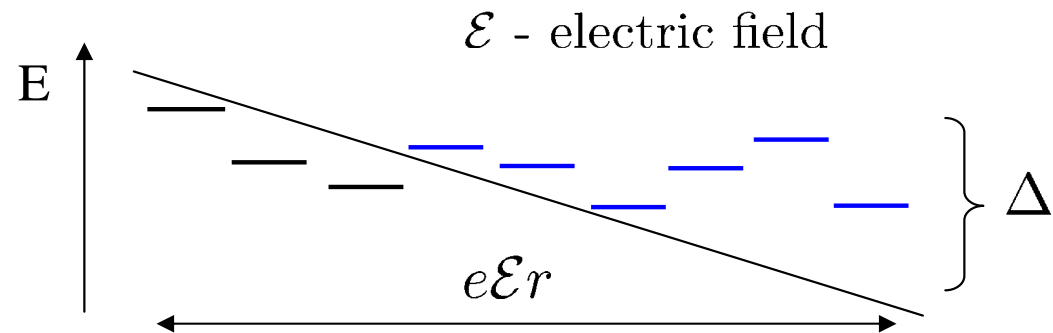
Nonlinear conductivity at strong electric fields

Hopping distance r within the energy shell Δ :

Shklovskii 1973

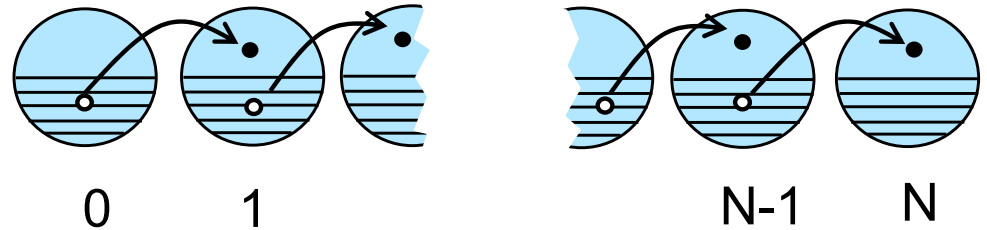
$$\left. \begin{array}{l} e\mathcal{E}r \sim \Delta \\ r \Delta \tilde{\kappa}/e^2 \sim 1 \end{array} \right\} r \sim \sqrt{e/\tilde{\kappa}\mathcal{E}}$$

$$j \sim j_0 e^{-(\varepsilon_0/\mathcal{E})^{1/2}}, \quad \varepsilon_0 \sim T_0/e\xi_{el}$$



Hopping conductivity in the regime of 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

1. Low electric field (linear regime)

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

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

with weakly temperature dependent effective localization length

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

Crossover temperature $\xi_{in} > \xi_{el} \implies T > \sqrt{\delta E_c}$ - inelastic mechanism dominates

Hopping conductivity via inelastic cotunneling

Regime of strong fields:

Temperature can be set to zero $T \rightarrow 0$

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

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

$$\left. \begin{array}{l} e\mathcal{E}r \sim \Delta \\ r \Delta \tilde{\kappa}/e^2 \sim 1 \end{array} \right\} \longrightarrow r \sim \sqrt{e/\tilde{\kappa}\mathcal{E}}$$

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

$$j \sim j_0 e^{-(\mathcal{E}_0/\mathcal{E})^{1/2}}, \quad \mathcal{E}_0(\mathcal{E}) \sim \frac{e}{\tilde{\kappa} a^2} \ln^2[\bar{E}^2/e^2 \mathcal{E}^2 a^2 \bar{g}]$$

Applicability:

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

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

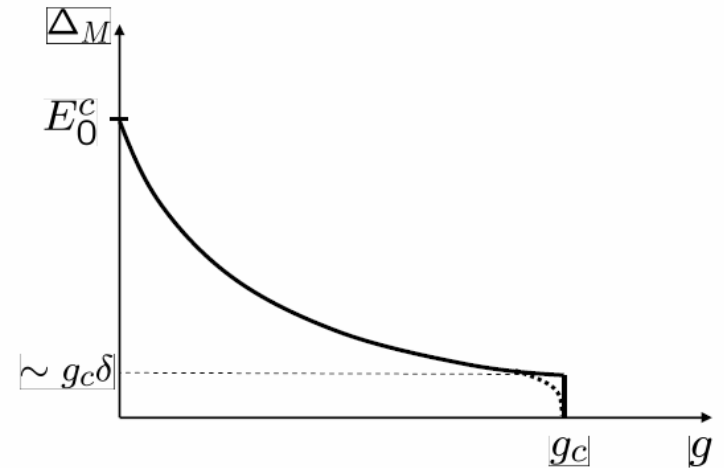
Results for metallic arrays

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:

$$\sigma \sim e^{-(T_0/T)^{1/2}}, \quad T_0 \sim e^2/\tilde{\kappa}\xi, \quad \xi \sim \begin{cases} \frac{2a}{\ln(\bar{E}\pi/\bar{g}\delta)}, & T < \sqrt{E_c\delta} \quad \text{elastic} \\ \frac{2a}{\ln[\bar{E}^2/16\pi T^2\bar{g}]}, & T > \sqrt{E_c\delta} \quad \text{inelastic} \end{cases}$$

Nonlinear regime:

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

Hopping conductivity in granular superconductors

Weak coupling regime $g \ll 1$

Each grain is superconducting but there is no global coherence.

Experimental data :

Granular aluminum samples.

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

Weak coupling insulating regime.

Grain size $\sim 120\text{\AA}$

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

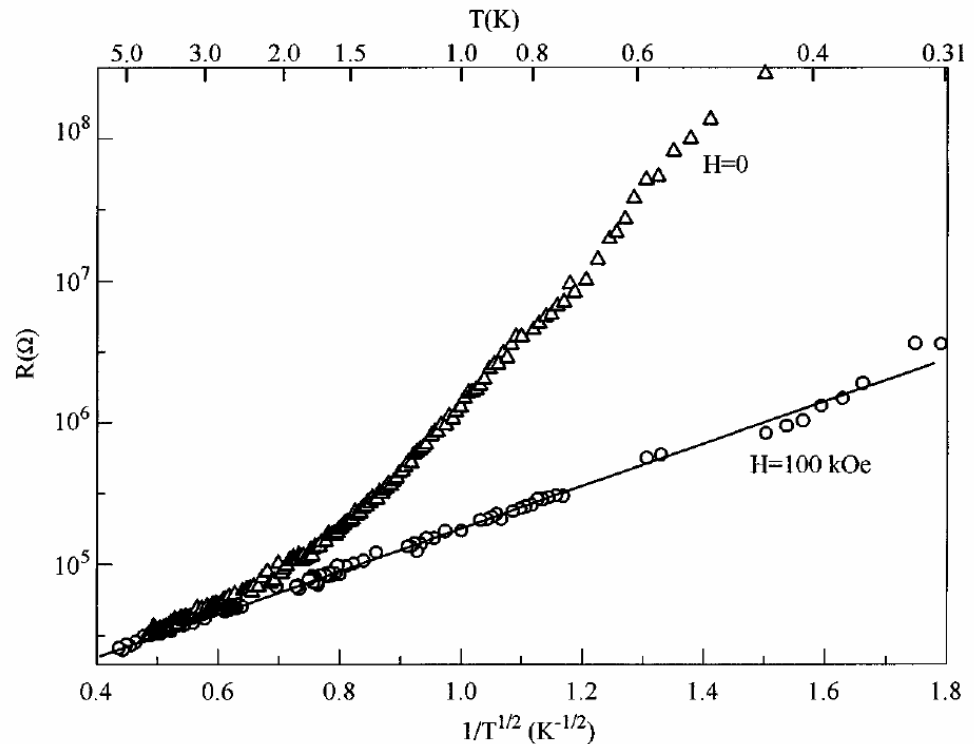
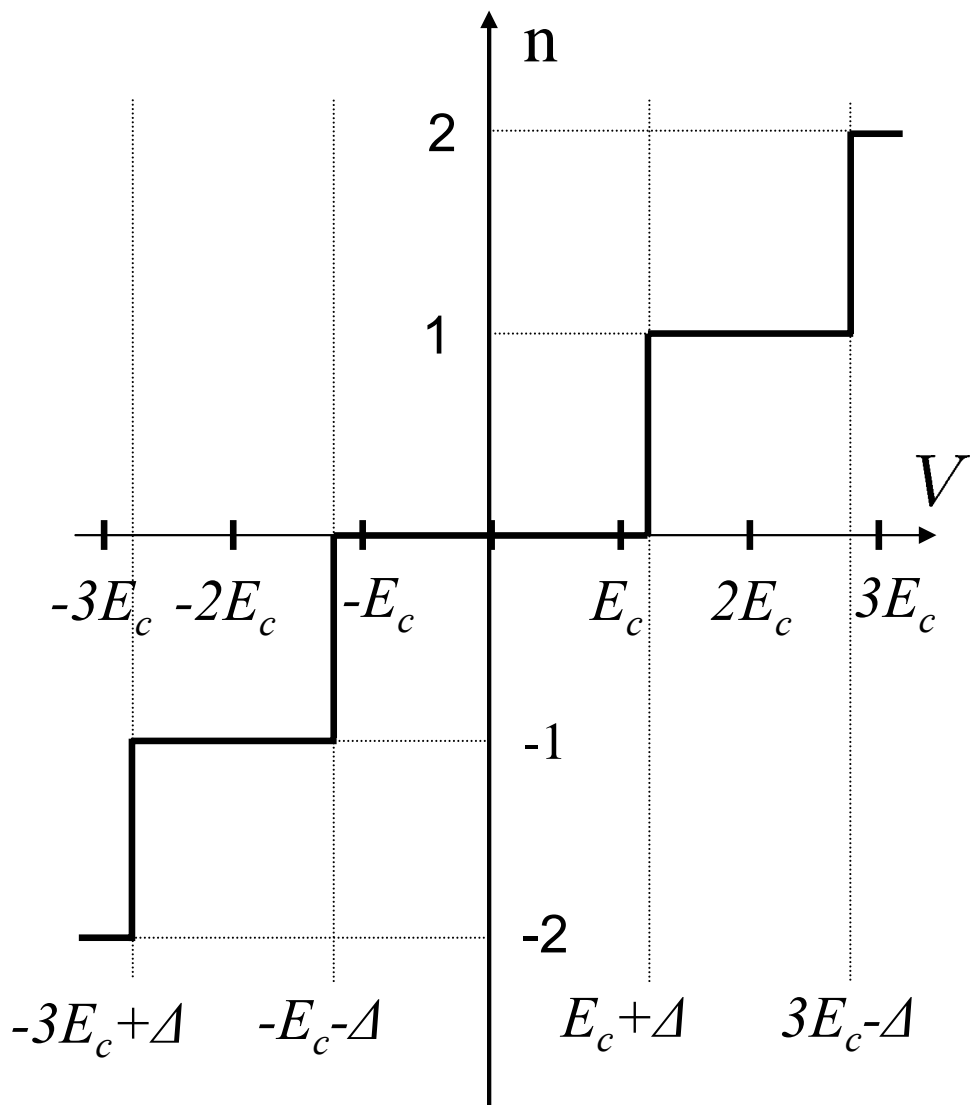


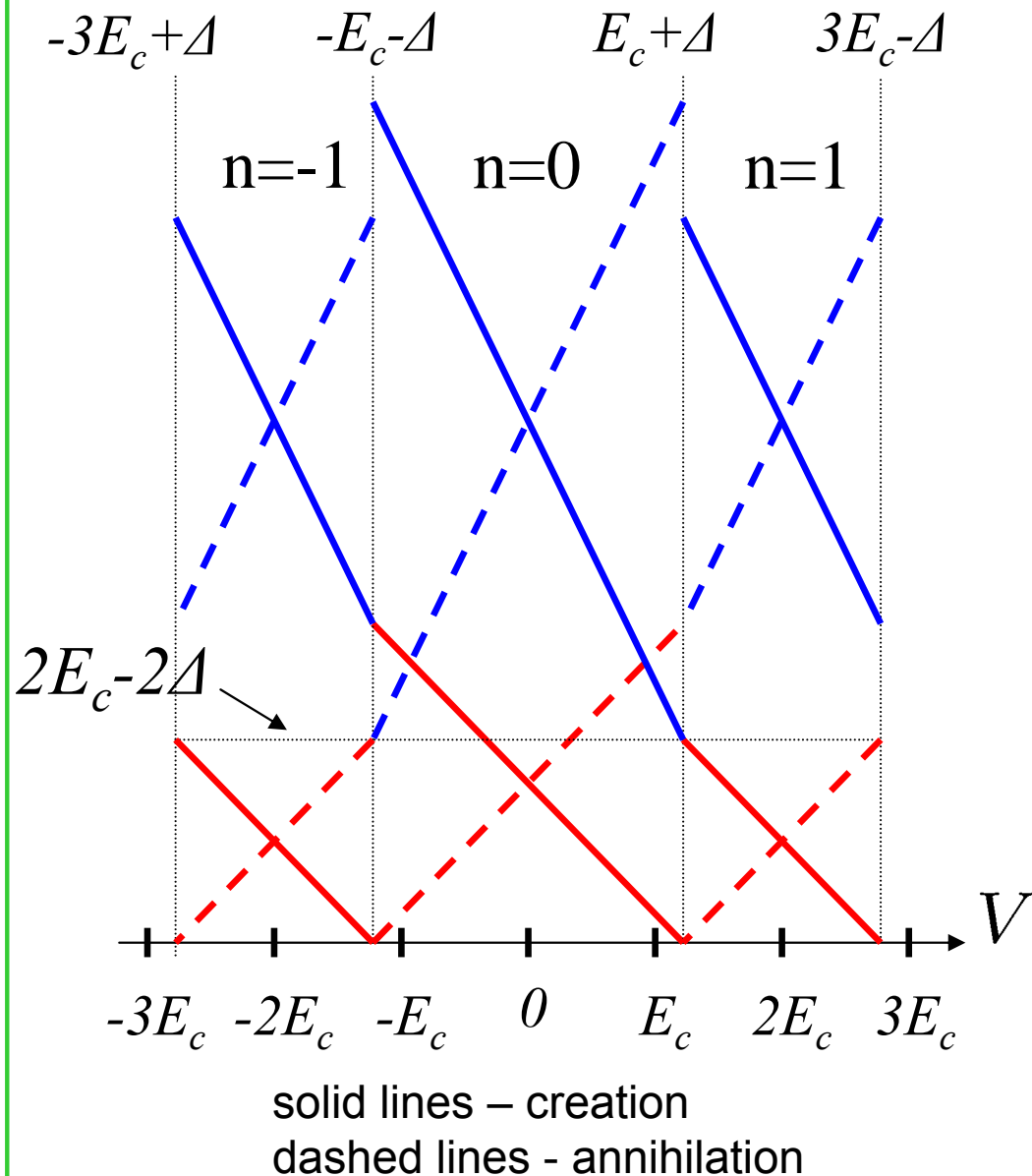
FIG. 1. Resistance of sample 1 measured at zero (triangles) and 100 kOe field (circles) as a function of the inverse square root of the temperature. Sample 1 room temperature resistance is $2 \times 10^3 \Omega$.

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

Electron occupation number: $n(V)$

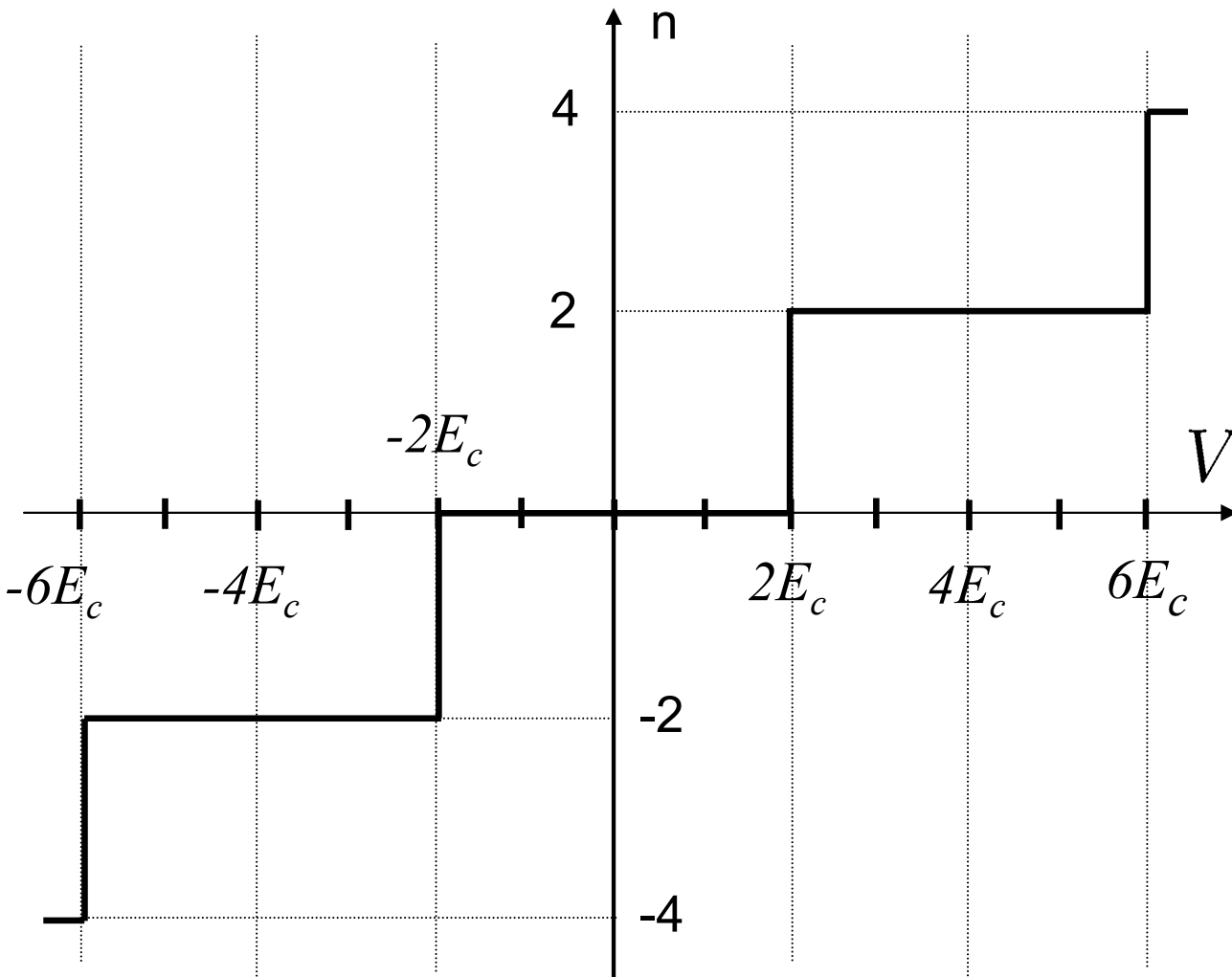


Single - and two particle excitation energies

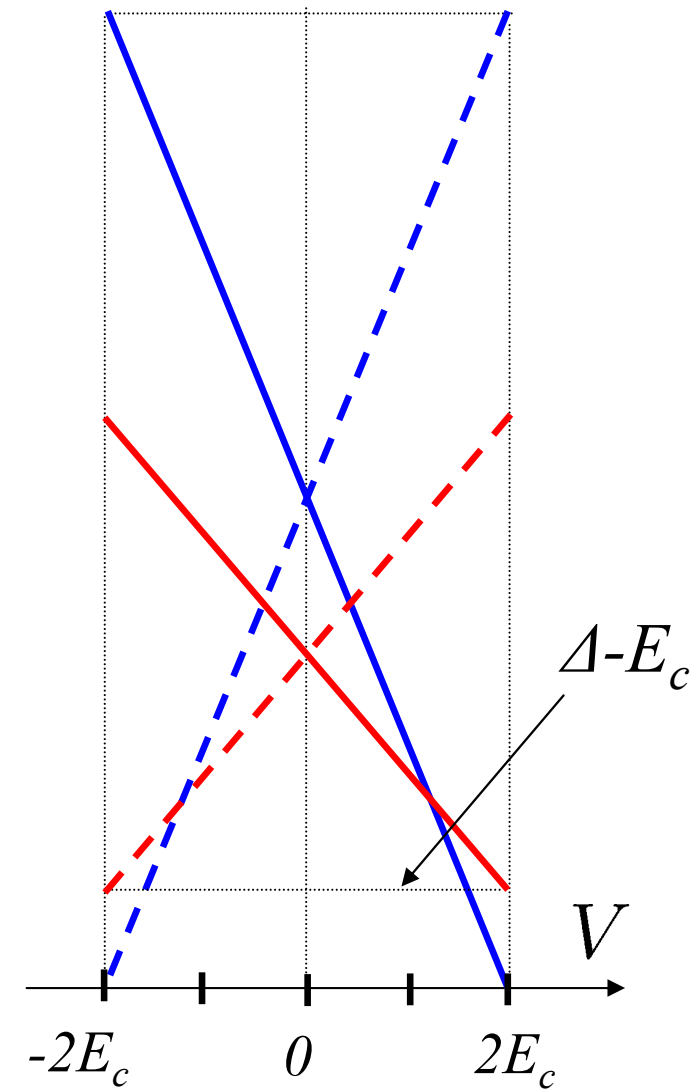


Electron number and excitation energies as functions of V , $\Delta > E_c$

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Single - and two particle excitation energies



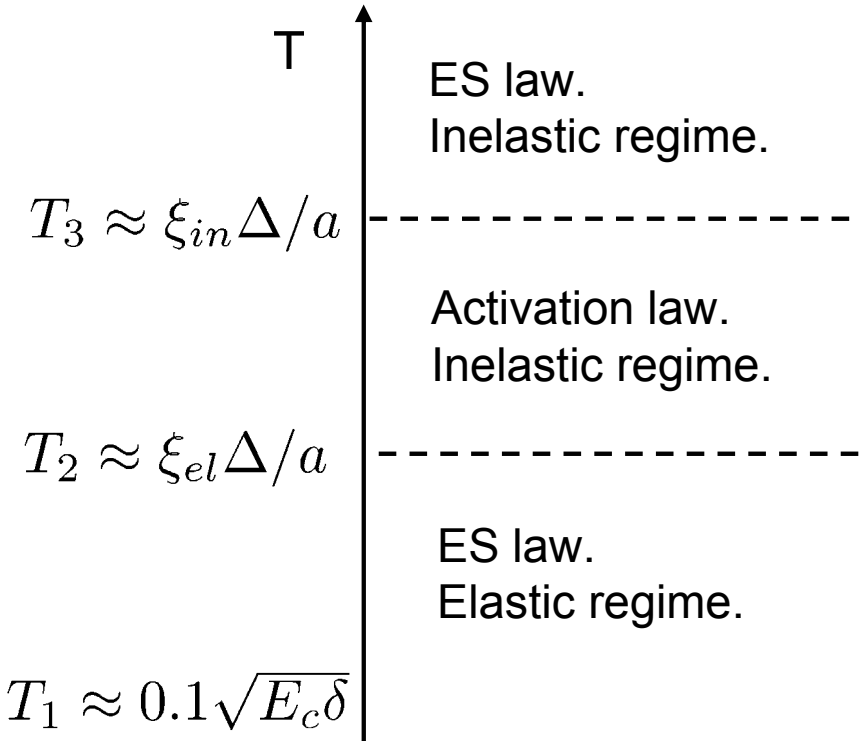
solid lines – creation
dashed lines – annihilation

Hopping conductivity in superconductors: Results.

Hopping law

$E_c > \Delta$. Electron hopping.

$E_c < \Delta$. Cooper pair hopping.

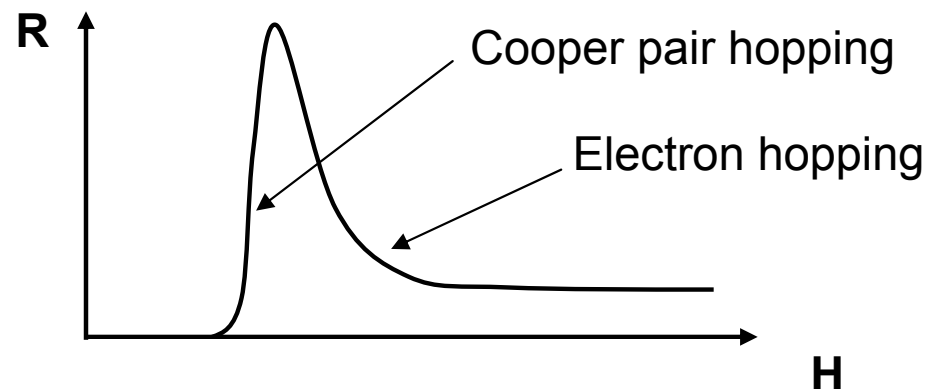


Negative magnetoresistance

1. ES law for Cooper pair transport.

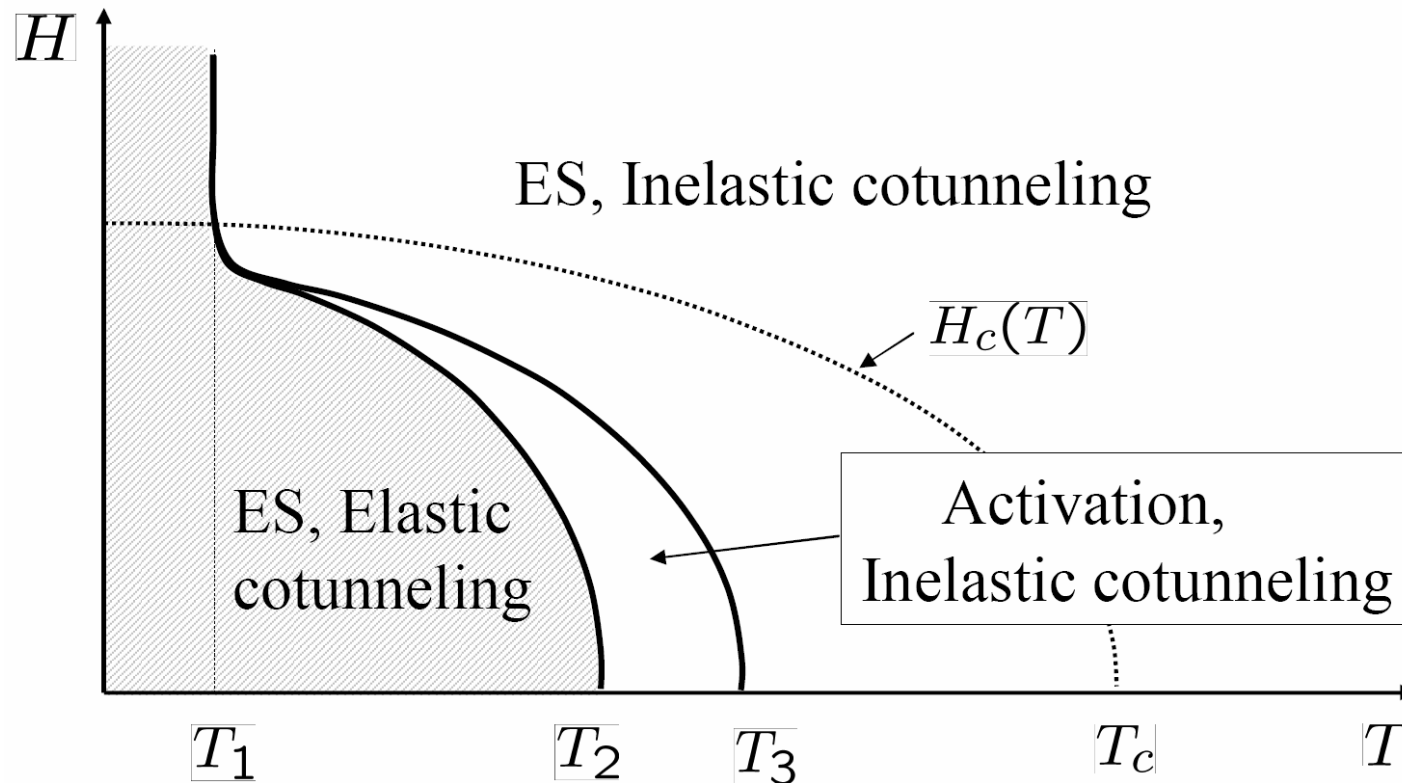
2. **Positive** magnetoresistance.

3. Possible scenario at $g \sim 1$:
Renormalization of the charging energy due to tunneling coupling.



Transport phase diagram in the magnetic field in the EH regime

The gap Δ can be tuned by the magnetic field



$T_1 \approx 0.1\sqrt{E_c\delta}$ - Crossover between elastic and inelastic regimes at $\Delta=0$

$T_2 \approx \xi_{el}\Delta/a$ - 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