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10TH CONFERENCE ON HOPPING AND RELATED PHENOMENA

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"Relaxation of Nonequilibrium Charge Carriers at Low Temperatures"

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Relaxation of nonequilibrium charge carriers at low temperatures

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Content

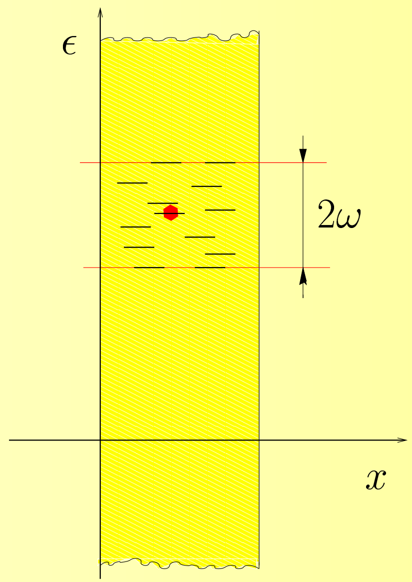
- The model and the kinetic constraint
- Relaxation at high temperatures
- Relaxation at low temperatures
- Conditions for the observability of the transition
- Relationship to some known results
- Conclusions

The model

- noninteracting electrons far from the Fermi energy

$$\frac{d\rho_m}{dt} = \sum_n [\rho_n W_{nm} - \rho_m W_{mn}]$$

$$W_{nm} = \nu\theta(\omega - |\epsilon_{nm}|)\exp(-2\alpha|R_{nm}| + \frac{\beta}{2}(\epsilon_{nm} - |\epsilon_{nm}|))$$



ρ_m- probability to find site m occupied, α^{-1}-localization length, $\beta = 1/kT$-inverse temperature, ν-attempt-to-escape frequency, R_{nm}-hopping length, ϵ_{nm}-difference between the site energies of the initial and the final site

The kinetic constraint

Idea:

- only long wave-length phonons are effective
- electron-phonon coupling constant restricts energy transfer to small values

$$W_{nm} \propto \sum_{\mathbf{q}} |\gamma(\mathbf{q})|^2 (1 - \cos(\mathbf{q}, \mathbf{R}_{nm})) \delta(\hbar\omega_q - |\epsilon_{nm}|)$$

$$\gamma(q) \propto F(q/2\alpha) \ * \ V(q)$$

overlap integral $\rightarrow 0$ for $q > 2\alpha$

Fourier transf. potential $\rightarrow 0$ for $q > q_c$

ω_q-phonon frequency, $\gamma(q)$-electron phonon coupling constant, q_c characteristic momentum of $V(q)$

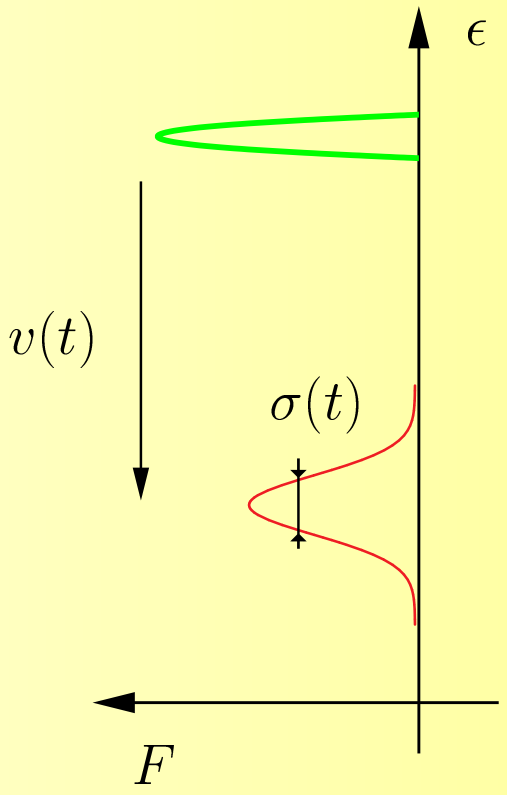
Interesting quantities

1. Energy distribution function $F(\epsilon_0, \epsilon|t)$
2. Energy relaxation rate

$$v(t) = \frac{d}{dt}\epsilon(t) = \frac{d}{dt}\langle \sum_n \epsilon_n \rho_n(t) \rangle$$

3.Mean squared deviation

$$\sigma^2(t) = \langle \sum_n \epsilon_n^2 \rho_n(t) \rangle - \epsilon^2(t)$$



$\langle \cdots \rangle$-configuration average

Calculation strategy

1. Solve problem with Green function

$$\rho_m(s) = \sum_n \rho_n(t=0) P_{nm}(s)$$

2. Use continuous coordinates

$$P(\rho, \rho'|s) = \sum_{nm} \delta(\rho - \rho_m) P_{mn}(s) \delta(\rho_n - \rho')$$

3. Calculate diffusion propagator

$$F(|\mathbf{R}' - \mathbf{R}|; \epsilon, \epsilon'|s) = \frac{1}{N(\epsilon)} \langle P(\rho, \rho'|s) \rangle$$

4. Obtain energy distribution function

$$F(\epsilon, \epsilon'|s) = \int d\mathbf{R} F(R; \epsilon', \epsilon|s)$$

5. Comparison of results with Monte-Carlo simulations

$N(\epsilon)$-density of states, $\rho_m = (\mathbf{R}_m, \epsilon_m)$, $\rho = (\mathbf{R}, \epsilon)$, s-Laplace frequency

Relaxation at $kT \gg \omega$

- Calculation of $F(\epsilon', \epsilon|s)$ in effective medium approximation[a]
- Quasielastic expansion of the equation for F
  assumption: ω is the smallest energy scale in the problem

$$sF(\epsilon', \epsilon|s) = \delta(\epsilon' - \epsilon) + kT\frac{d}{d\epsilon}[N(\epsilon, s)v(\epsilon, s)(\underbrace{\frac{d}{d\epsilon}\frac{F(\epsilon', \epsilon|s)}{N(\epsilon)}}_{\text{energy diffusion}} + \underbrace{\frac{F(\epsilon', \epsilon|s)}{kTN(\epsilon)}}_{\text{“current”}})]$$

$$v(\epsilon, 0) = \frac{\omega^2\nu}{3kT}\exp(-\rho_c(\epsilon)) \qquad \rho_c(\epsilon) = \#\frac{2\alpha}{(2\omega N(\epsilon))^{1/d}}$$

$$\frac{v(\epsilon, s)}{v(\epsilon, 0)}\ln\frac{v(\epsilon, s)}{v(\epsilon, 0)} = \frac{s}{\omega_0(\epsilon)} \qquad \omega_0(\epsilon) = \frac{d\nu}{\rho_c(\epsilon)}\exp(-\rho_c(\epsilon))$$

$v(\epsilon, s)$-spectral energy relaxation rate, d-spatial dimension, $\# = (d/S_d)^{1/d}$, S_d-solid angle in d dimensions

[a] method:O.Bleibaum, H. Böttger, V. V. Bryksin, Phys. Rev. B**62**, 13440 (2000)

Results for $N(\epsilon) = N$

1. characteristic time scale:

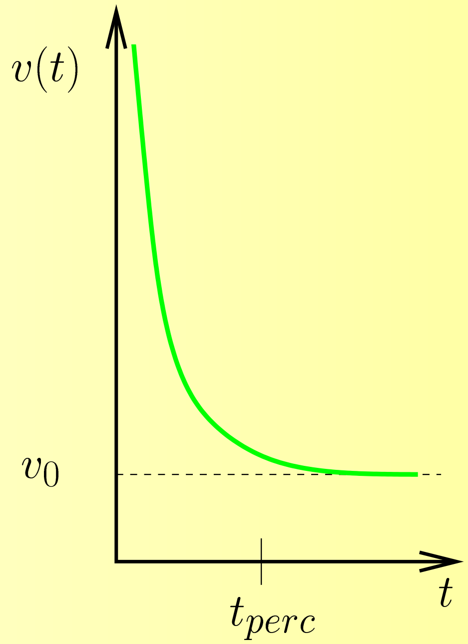
$$t_{perc} \propto \frac{1}{\nu}\rho_c \exp(\rho_c)$$

2. energy relaxation rate

$$v(t) \propto v_0 \frac{t_{perc}}{t \ln^2(t_{perc}/t)} \qquad t \ll t_{perc}$$

$$v(t) = v_0(1+\sqrt{\frac{e}{2\pi}}\frac{\exp(-t/t_{perc})}{(t/t_{perc})^{3/2}}) \qquad t \gg t_{perc}$$

$$v_0 = \frac{\omega^2 \nu}{3kT}\exp(-\rho_c)$$



3. Diffusive contribution to the propagator irrelevant at low temperatures ($kT \ll \sqrt{6d\omega\rho_c}$)

$$sF(\epsilon', \epsilon|s) = \delta(\epsilon' - \epsilon) + kT\frac{d}{d\epsilon}(F(\epsilon', \epsilon|s)v(\epsilon, s))$$

4. Mean squared deviation

$$\sigma^2(t) = 2kT\,v_0 t \qquad\qquad t \gg t_{perc}$$

5. Distribution function at large times

$$F(\epsilon', \epsilon|t) = \frac{1}{\sqrt{2\pi\sigma^2(t)}}\exp(-\frac{(\epsilon' - \epsilon - v_0 t)^2}{2\sigma^2(t)})$$

Relaxation at $T = 0$ *for* $N(\epsilon) = N$

Results of effective medium approximation (EMA):

$$sF(\epsilon', \epsilon|s) = \delta(\epsilon' - \epsilon) + \omega \frac{d}{d\epsilon}(F(\epsilon', \epsilon|s)v(\epsilon, s))$$

$$v(\epsilon, 0) = \frac{\omega\nu}{2} \exp(-\rho_c(\epsilon))$$

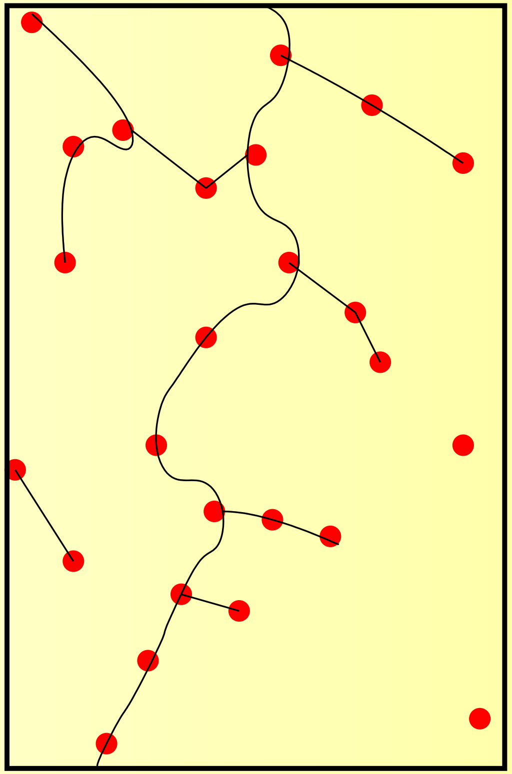
$$\rho_c(\epsilon) = \# \frac{2\alpha}{(\omega N(\epsilon))^{1/d}}$$

$$\omega_0(\epsilon) = \frac{d\nu}{\rho_c(\epsilon)} \exp(-\rho_c(\epsilon))$$

<u>Observation:</u> EMA predicts percolation like transport at zero temperature!

Observation:

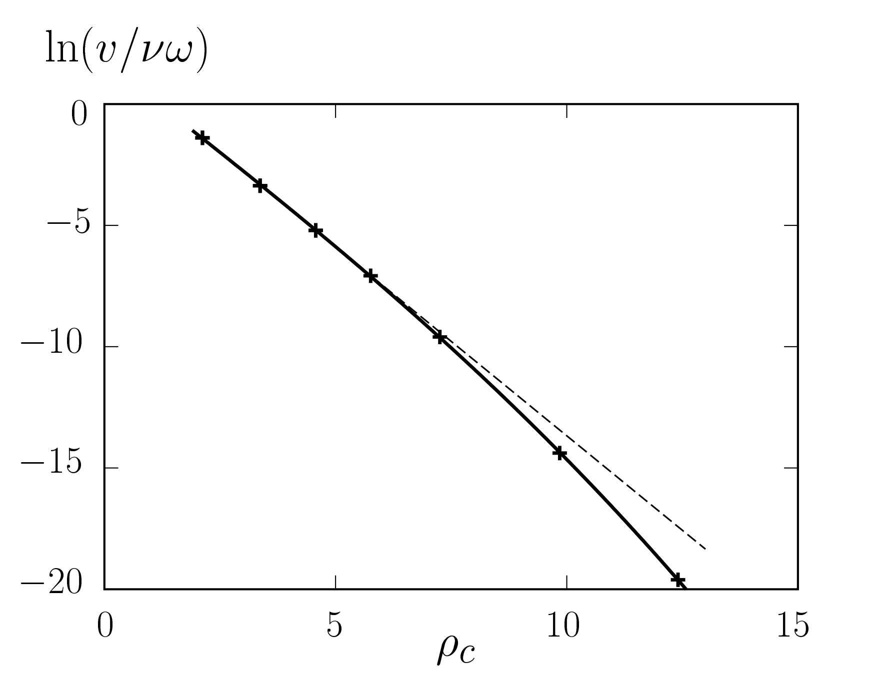
1. Percolation paths are the fastest possible relaxation paths → they determine the lower tail of the distribution function.
2. A lot of other relaxation paths are possible. Accordingly, we expect that the center of the particle packet is not determined by percolation like paths.



Monte-Carlo Simulations of v for d=3:

EMA versus MC-Simulation

1. Both the numerical preexponetial factor and the magnitude of the exponent of the EMA are too small to compare with the numerical simulations.
2. The concentration dependence of v as predicted by the EMA differs from the numerical result.
   (— numerical result,
   - - - guide for the eye)

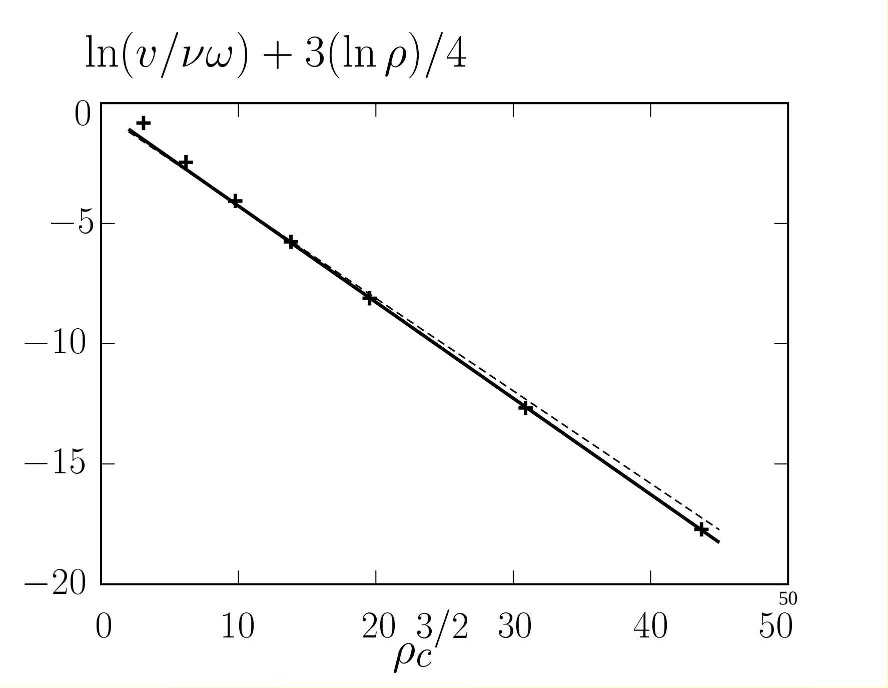


Concentration dependence of $v(s=0)$ for d=3

- Analogy: Energy relaxation at $T=0$ is like conduction in a strong electric field!
- Conductivity in a strong electric field:

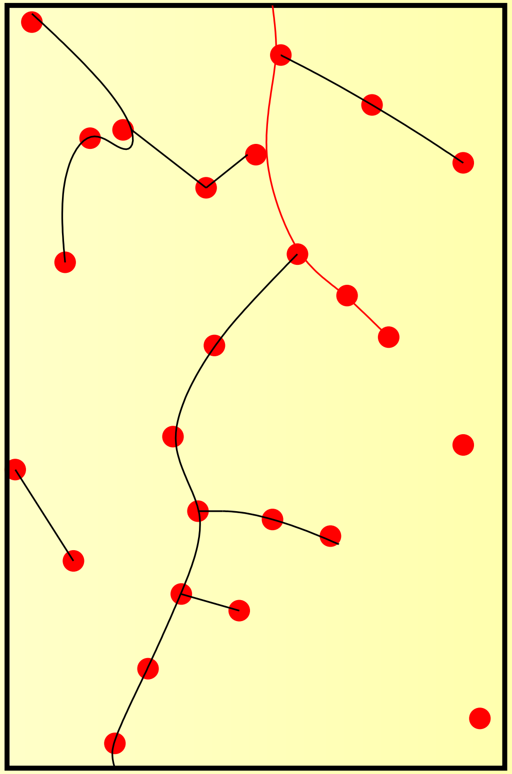
$$\sigma \propto \exp(-c\rho_c^{d/(d-1)})$$

("Miller-Abrahams transport") (I. P. Zvyagin (1979), H. Böttger and V. V. Bryksin (1980), N. Van Lien and B. I. Shklovskii (1981))



Why are relaxation paths different from low-field percolation paths?

1. The particle can not return.
2. There is no particel source.



Analytical description of the relaxation

- CTRW+Quasielasticity

$$sF(\epsilon',\epsilon|s) = \delta(\epsilon'-\epsilon) + kT\frac{d}{d\epsilon}(F(\epsilon',\epsilon|s)v(\epsilon,s))$$

$$v(\epsilon,0) = C_1\omega\nu\rho_c^{-d/(2(d-1))}\exp(-(d-1)(\rho_c(\epsilon)/d)^{d/(d-1)})$$

$v(\epsilon, s \neq 0)$: complicated integral of $s, d, \rho_c(\epsilon)$

C_1-number

(E. Haba, O. Bleibaum, H. Böttger and V.V. Bryksin, Phys. Rev. B **68**, 142203 (2003))

Some further results for $N(\epsilon) = N$

1. Characteristic time scale:

$$t_{MA} = \frac{1}{\nu} \exp((d-1)(\rho_c(\epsilon)/d)^{d/(d-1)})$$

2. Energy relaxation rate:

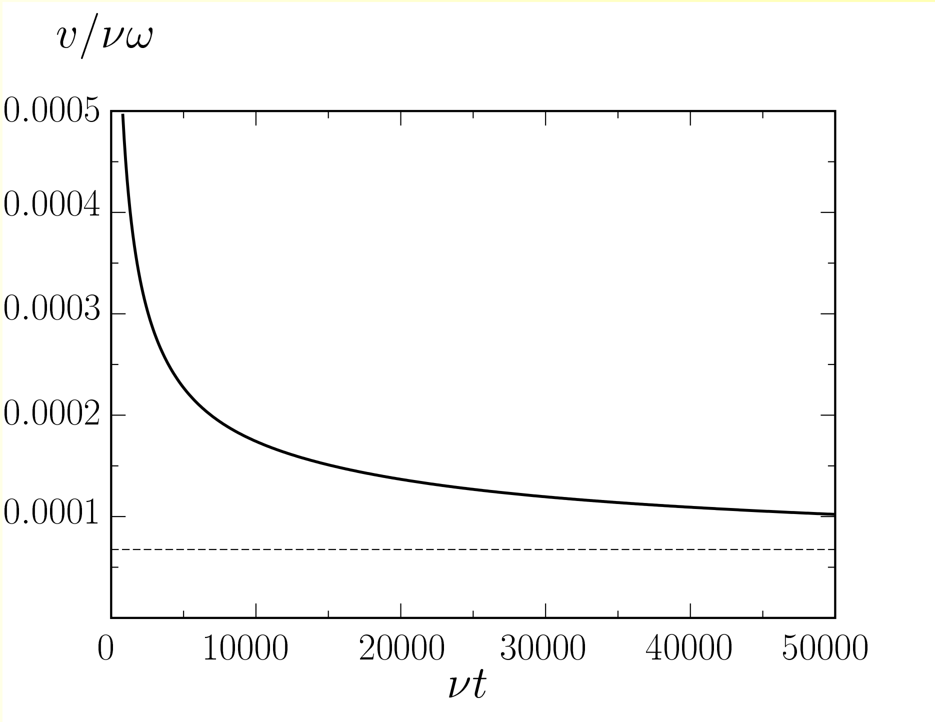
$$v(t) = v(0)(1 + \frac{2v(0)}{d\omega\nu} \frac{\rho_c^d}{\ln^{d-1}(\nu t)} \nu t \exp(-\frac{\ln^d(\nu t)}{\rho_c^d})) \qquad t \gg t_{MA}$$

$$v(t) \propto \frac{\omega}{t} \frac{\ln^{d-1}(\nu t)}{\rho_c^d} \exp(-\frac{\ln^d(\nu t)}{\rho_c^d}) \qquad t \ll t_{MA}$$

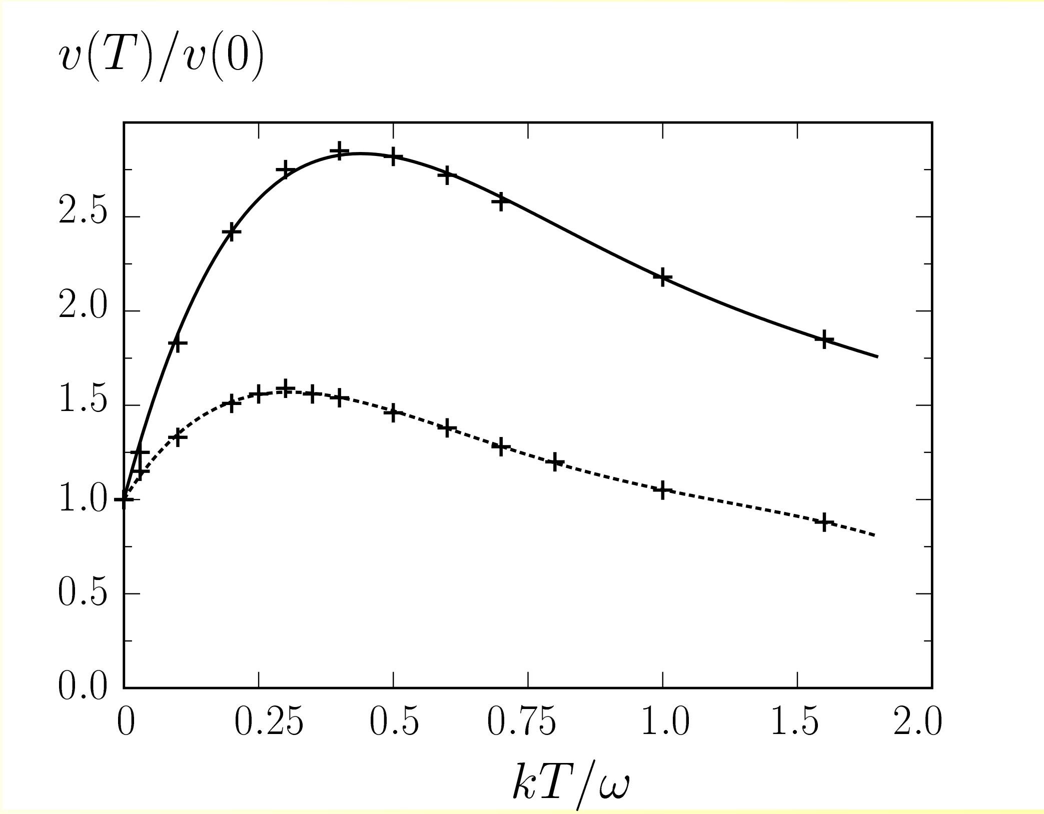
3. Structure of the energy diffusion function for $t \gg t_{MA}$: Gaussian

(E. Haba, O. Bleibaum, H. Böttger, V. V. Bryksin, Phys. Rev. B **68**, 14203 (2003))

Energy relaxation for $\rho_c = 7.25$ und $d = 3$.



Temperature dependence of the energy relaxation rate (d=3)



(— $\rho_c = 7.25$, - - - $\rho_c = 5.75$)

Observation conditions

How rare are Miller-Abrahams pores?

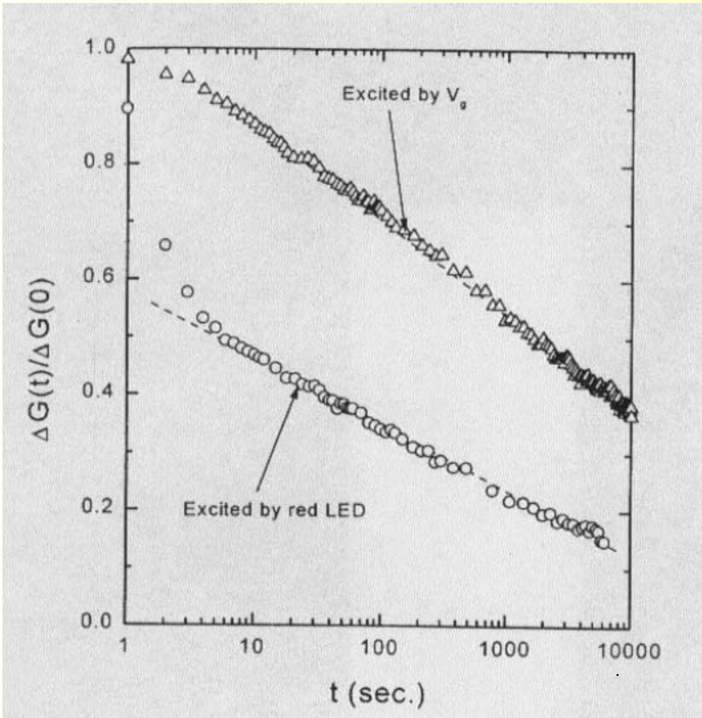
Poisson distribution: $p(R) = \exp(-\kappa_d) = \exp(-(\frac{2\alpha R}{\rho_c})^d)$

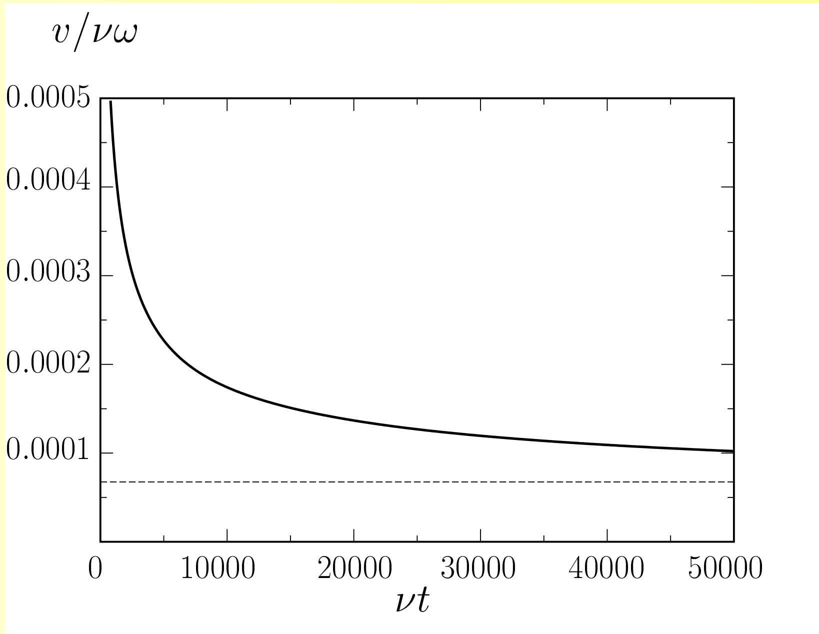
Hopping length in MA-regime for $s = 0$: $R_{MA} = \frac{(d-1)}{2\alpha}(\frac{\rho_c}{d})^{d/(d-1)}$

$$\kappa_3 = 3.65\rho_c^{3/2} \qquad \kappa_2 = \frac{1}{16}\rho_c^2$$

Miller-Abrahams processes are in particular important in d=2 !

Some estimates in d=2





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(M. Pollak and Z. Ovadyahu, J. Phys. 1 France, **7**, 1595 (1997))

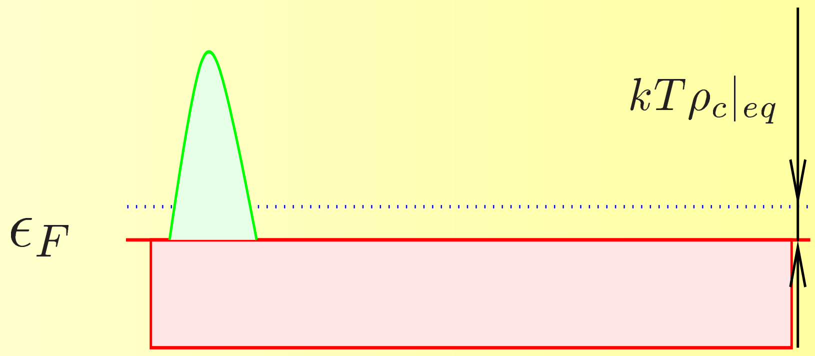
Question: When is $t_{\dots} = 1s$?

$$\nu = 10^{12}\text{Hz}$$

$$t_{perc} = 1s \quad \rightarrow \quad \rho_c = 25$$

$$t_{MA} = 1s \quad \rightarrow \quad \rho_c = 7.4$$

excess charge carriers



How many hops are necessary to find a MA-hole?

$$e^{-\kappa_2} = e^{-(7.4)^2/16} = \frac{1}{20.08}$$

Results for an exponential density of states

- A strongly energy dependent density of states affects the relaxation process.$\rightarrow$ Characteristic time scale is not set by the MA-time.
- $N(\epsilon) = N_0 \exp(\frac{d\epsilon}{\Delta})$

Question: Are there differences between the results for the quasi-elastic and the inelastic theory?

$$O(\nu t)_{\text{inelastic}} \quad \rightarrow \quad O(\# \frac{\omega}{\Delta} \nu t)_{\text{quasi-elastic}}$$

$$\epsilon(t) = \Delta \ln \ln(\nu t) \quad \rightarrow \quad \epsilon(t) = \Delta \frac{d-1}{d} \ln \ln(\# \frac{\omega}{\Delta} \nu t)$$

Conclusion: Systems with weakly energy dependent density of states should show cleanest MA-behavior.

O-observable, $\#$-number of order 1

Conclusions

- The simple model describes a transition between two different relaxation regimes.
- The relaxation in this model is percolation like at high temperatures, like in a low-field conduction problem. In this case the characteristic time scale is given by the percolation time.
- The relaxation is Miller-Abrahams like at very low temperatures. In this regime the characteristic relaxation time is given by the Miller-Abraham time, which is much larger than the percolation time.
- 2-dimensional systems with weakly energy dependent density of states seem to provide the most favorable conditions for the observation of the Miller-Abrahams relaxation.

Literature

The material presented in this talk is based on the papers

- O. Bleibaum, H. Böttger, V. V. Bryksin and A. N. Samukhin, *Dispersive energy transport and relaxation in the hopping regime*, Phys. Rev. B **62**, 13440 (2000).
- O. Bleibaum, H. Böttger, V. V. Bryksin and A. N. Samukhin, *Einstein relationship and relaxation current in a tail at vanishing temperature*, Phys. Rev. B **65**, 94203 (2002)
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- E. Haba, O. Bleibaum, H. Böttger, and V. V. Bryksin, *Energy relaxation in a tail at zero temperature in the hopping regime*, Phys. Rev. B **68**, 14203 (2003).
- O. Bleibaum, H. Böttger and V. V. Bryksin, *Impact of the density of states on the dynamical hopping conductivity*, Phys. Rev. B **66**, 104203 (2002).