

# Theory of Disordered Condensed-Matter Systems

Walter Schirmacher

University of Mainz, Germany

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# Outline

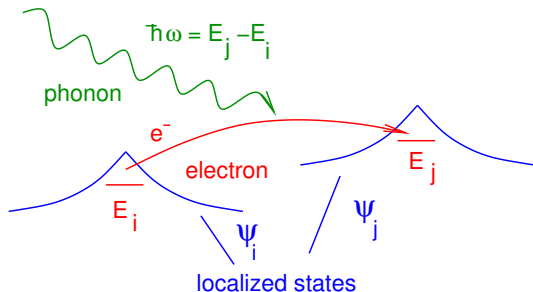
## 1 Disorder and the Glass transition

## 2 Diffusion in quenched-disordered systems

- Electronic hopping transport in disordered semiconductors
  - Phonon-assisted tunneling
  - Mott optimization
  - Master equation and Percolation construction
- Random walk on a lattice and regular diffusion
- diffusion in a disordered system: spatially fluctuating diffusivity
- Perturbation theory
- Analogy with a mass-spring vibrational problem
- disordered diffusivity and AC conductivity
- Coherent-potential approximation (CPA)
- Comparison of CPA calculations with measured AC conductivity data

## Electrons in doped/disordered semiconductors

- Phonon-assisted hopping transport between localized states
  - ▶ Lightly doped crystalline semiconductors
  - ▶ Structurally disordered semiconductors (OLED)



- Transition probability/time for an electron from  $\psi_i$  to  $\psi_j$

$$W_{ij} = \nu_0 e^{-\beta(E_j - E_i)/k_B T} e^{-2\alpha|r_i - r_j|}$$

$$\beta = 1/k_B T, \alpha = \text{inverse localization length}$$

## Mott optimization and $T^{1/4}$ law

- Mott's argumentation:

- ▶ The electron will try to use a hopping path, in which neither the first nor the second part of the exponential becomes too small. The optimal hop will maximize  $W_{ij}$ , i.e. minimize the exponent.
- ▶ The hopping distance will obey on the average

$$(\Delta r)^3 = [N_F \Delta E]^{-1} \quad N_F = \text{DOS at the Fermi level}$$

so that the exponent becomes

$$\xi_{ij} = \beta \Delta E + 2\alpha \Delta r = \frac{\beta}{N_F (\Delta r)^3} + 2\alpha \Delta r$$

which is minimized for

$$\Delta r_{\text{opt}} = \left( \frac{3\beta}{2\alpha N_F} \right)^{1/4}$$

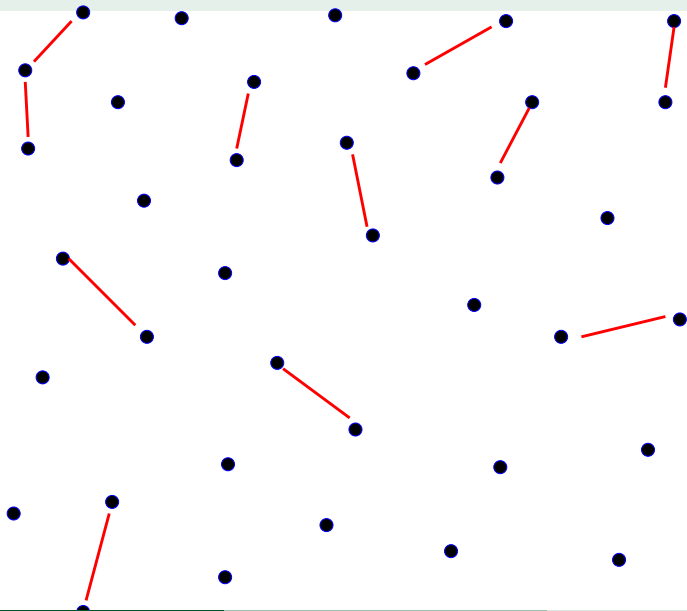
$$\Rightarrow W_{\text{opt}} \propto e^{-(T_0/T)^{1/4}} \quad T_0 \propto \alpha^3 / N_F$$

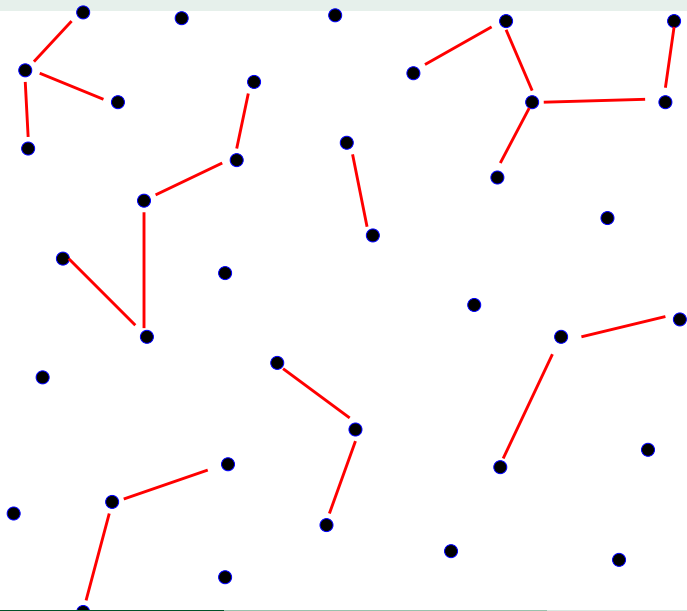
## Master equation and equivalent circuit

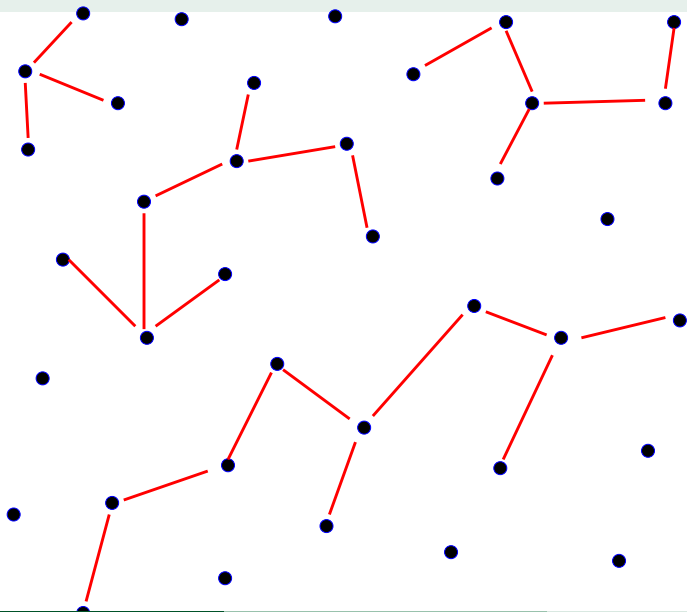
- The random walk of a hopping electron is described by the following Markovian Master equation

$$\frac{d}{dt}P_i(t) = - \sum_j W_{ij} \left( P_i(t) - P_j(t) \right) \quad P_i = \text{Probability to be at } i$$

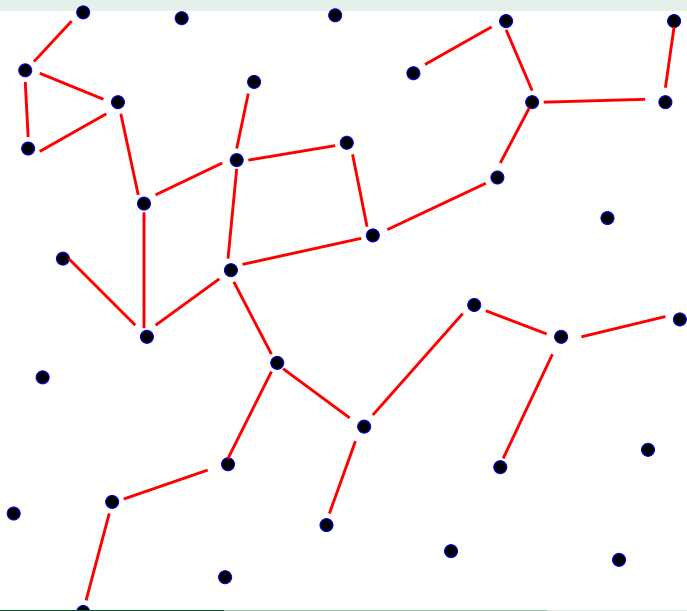
- In the steady state  $\frac{d}{dt}P_i(t) = 0$  this becomes equivalent to Kirchhoff's node equation with  $P_i$  being local voltages and  $W_{ij}$  conductances between nodes  $i$  and  $j$ .
- If the conductances vary exponentially one can use the following **percolation construction** to estimate the conductance of the network
  - ▶ A network of sites, connected with bonds  $i - j$  is considered
  - ▶ Each bond is assigned a conductance  $W_{ij} = W_0 e^{-\xi_{ij}}$
  - ▶ Sites with  $\xi_{ij} < \xi_{\text{threshold}}$  are connected, where  $E_{\text{threshold}}$  is a variable threshold, which is increased from a low value

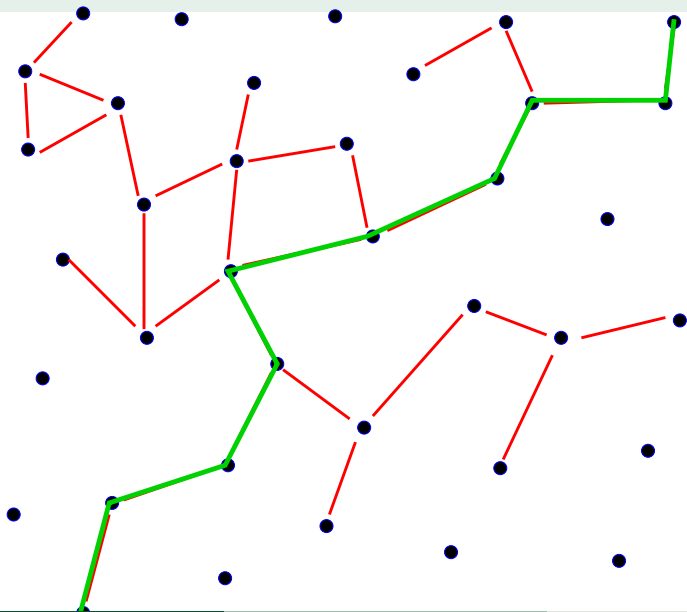
Increasing the threshold  $\xi$ 

Increasing the threshold  $\xi$ 

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## Percolation construction

- Mathematically this construction can be described in terms of the percolation threshold  $p_c$

$$\begin{aligned}
 p_c &= \left\langle \theta \left( \xi_c - \xi(\Delta r, \Delta E) \right) \right\rangle \\
 &= \underbrace{N(\langle \Delta E \rangle)}_{N_F} \int 4\pi(\Delta r)^2 d\Delta r \int d\Delta E \theta \left( \xi_c - \xi(\Delta r, \Delta E) \right) \\
 &= \frac{\pi N_F}{2\alpha^3 \beta} \int \int x^2 dx dy \theta \left( \xi_c - x - y \right) \\
 &\propto \xi_c^4 \frac{N_F T}{\alpha^3}
 \end{aligned}$$

which gives the same result as Mott's optimization.

## Random walk on a lattice

- Master equation for a random Walk on a 3-dimensional lattice with lattice constant  $a$  and nearest-neighbor hopping  $W_0$

$$\frac{d}{dt}P_i(t) = -W_0 \sum_j \left( P_i(t) - P_j(t) \right)$$

- ▶ Initial condition:  $P_i(0) = \delta_{i,i_0}$

- Laplace transform  $P_i(s) = \int_0^\infty e^{-st} P_i(t) dt$   $s = i\omega + \epsilon$

$$sP_i(s) - P_i(0) = -W_0 \sum_j \left( P_i(s) - P_j(s) \right)$$

- Bloch's theorem:

which leads to 
$$P_i(s) = e^{i\mathbf{k}r_i} u(\mathbf{k}, s)$$

$$su(\mathbf{k}, s) - 1 = 2W \left( \cos(k_x ax) + \cos(k_y ay) + \cos(k_z az) - 3 \right) u(\mathbf{k}, s)$$

- Solution

$$u(\mathbf{k}, s) = \frac{1}{s + 2W[3 - \cos(k_x ax) + \cos(k_y ay) + \cos(k_z az)]}$$

# Random Walk on a lattice

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- Solution

$$u(\mathbf{k}, s) = \frac{1}{s + 2W[3 - \cos(k_x ax) + \cos(k_y ay) + \cos(k_z az)]} \equiv G_0(\mathbf{k}, s)$$

- This is the **Green's function** for the regular random walk. For  $|\mathbf{k}| \rightarrow 0$  it becomes

$$G(\mathbf{k}, s) \rightarrow \frac{1}{s + Wa^2 k^2}$$

- This is the solution in  $\mathbf{k}, s$  space for the **diffusion equation**

$$\frac{d}{dt} P(\mathbf{r}, t) = D \nabla^2 P(\mathbf{r}, t) \quad D = Wa^2$$

## Ordinary diffusion equation

- Diffusion equation

$$\frac{d}{dt}P(\mathbf{r}, t) = D\nabla^2 P(\mathbf{r}, t)$$

- Spatial Fourier transform

$$\frac{d}{dt}P(\mathbf{k}, t) = -Dk^2 P(\mathbf{k}, t)$$

- Laplace transform

$$sP(\mathbf{k}, s) - P(\mathbf{k}, t = 0) = -Dk^2 P(\mathbf{k}, s)$$

$$P(\mathbf{k}, s) = \frac{P(\mathbf{k}, t = 0)}{s + Dk^2} = P(\mathbf{k}, t = 0)G(\mathbf{k}, s)$$

$$G(\mathbf{k}, t) = e^{-Dk^2 t}$$

$$G(\mathbf{r}, t) = \left[ \frac{1}{\sqrt{4\pi Dt}} \right]^3 e^{-r^2/4Dt}$$

## diffusion in a disordered system: spatially fluctuating diffusivity

- We want to explore (as before) the diffusive motion of particles in a disordered system, which move between sites  $i$  and  $j$

$$\frac{d}{dt}P_i(t) = - \sum_j W_{ij} \left( P_i(t) - P_j(t) \right)$$

- We first try to relate this equation to a kind of diffusion equation
- Then we solve this diffusion equation
  - ▶ By perturbation theory
  - ▶ By the coherent-potential approximation (CPA)

## Conduction and diffusion in a disordered system

Let us consider a  $d$ -dimensional hypercube with side length  $L$  and volume  $V = L^d$ .

- Face-to-face conductance

$$g = \sigma L^{d-2} \quad \underbrace{\sigma = \frac{N}{V} \frac{e^2}{k_B T} D}_{\text{Einstein relation}}$$

- Let us now tile the volume  $V$  into a mesh of smaller hypercubes (voxels) with volume  $V_c$ . For each of the voxels we can calculate the conductance and apply Einstein's relation to arrive at diffusivities  $D_i = D(\mathbf{r}_i)$ , where  $\mathbf{r}_i$  is the center of a certain voxel.
- In a continuum description we arrive at a diffusion equation

$$\frac{d}{dt} P(\mathbf{r}, t) = \nabla D(\mathbf{r}) \nabla P(\mathbf{r}, t)$$



## Conduction and diffusion in a disordered system

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- In a continuum description we arrive at a diffusion equation

$$\frac{d}{dt}P(\mathbf{r}, t) = -\nabla D(\mathbf{r})\nabla P(\mathbf{r}, t)$$

and for the Green's function

$$\frac{d}{dt}G(\mathbf{r}-\mathbf{r}', t-t') + \nabla D(\mathbf{r})\nabla G(\mathbf{r}-\mathbf{r}', t-t') = -\delta(t-t', \mathbf{r}-\mathbf{r}')$$

# Perturbation theory

$$\frac{d}{dt}G(\mathbf{r}-\mathbf{r}', t-t') + \nabla D(\mathbf{r})\nabla G(\mathbf{r}-\mathbf{r}', t-t') = -\delta(t-t', \mathbf{r}-\mathbf{r}')$$

- We assume that the fluctuations of  $D(\mathbf{r})$  are **small**:

$$D(\mathbf{r}) = D_0 + \Delta D(\mathbf{r}) \quad D_0 = \langle D \rangle$$

- The unperturbed Green's function is  $G_0(\mathbf{k}, s) = \frac{1}{s + D_0 k^2}$
- Defining  $G(\mathbf{k}, \mathbf{k}', s) = \frac{1}{V} \int d\mathbf{r} \int d\mathbf{r}' e^{-i\mathbf{k}\mathbf{r}} e^{i\mathbf{k}'\mathbf{r}'} G(\mathbf{r}, \mathbf{r}', s)$   
we can re-write the diffusion equation for the full problem in  $\mathbf{k}, s$  space

$$G(\mathbf{k}, \mathbf{k}', s) = G_0(k, s)\delta_{\mathbf{k}, \mathbf{k}'} - \int \left( \frac{d\mathbf{q}}{2\pi} \right)^d \mathbf{k} \cdot \mathbf{q} \Delta D_{V_c}(\mathbf{k} - \mathbf{q}) G(\mathbf{q}, \mathbf{k}', s)$$

## Perturbation theory

$$G(\mathbf{k}, \mathbf{k}', s) = G_0(k, s) \delta_{\mathbf{k}, \mathbf{k}'} - \int \left( \frac{d\mathbf{q}}{2\pi} \right)^d \mathbf{k} \cdot \mathbf{q} \Delta D_{V_c}(\mathbf{k} - \mathbf{q}) G(\mathbf{q}, \mathbf{k}', s) \quad (1)$$

- We represent now the averaged Green's function in terms of a self-energy function  $\Sigma(\mathbf{k}, s)$  as

$$\langle G(\mathbf{k}, \mathbf{k}', s) \rangle = \delta_{\mathbf{k}, \mathbf{k}'} \frac{1}{G_0^{-1}(k, s) - \Sigma(\mathbf{k}, s)} \Leftrightarrow \Sigma(\mathbf{k}, s) = \frac{1}{G_0(k, s)} - \frac{1}{\langle G(\mathbf{k}, \mathbf{k}', s) \rangle_{\mathbf{k}=\mathbf{k}'}} \quad (2)$$

- Iterating (1) twice and expanding the fraction in (2) to second order in the fluctuations we obtain (using  $\langle \Delta D_{V_c} \rangle = 0$ )

$$\Sigma(\mathbf{k}, s) = \frac{1}{(2\pi)^d} \int d^d \mathbf{p} K(\mathbf{k} - \mathbf{p}) (\mathbf{k} \cdot \mathbf{p})^2 G_0(\mathbf{p}, s)$$

where  $K(\mathbf{k}) = \int_{V_c} d^d \mathbf{r} e^{i\mathbf{k}\mathbf{r}} \langle \delta D(\mathbf{r}_0 + \mathbf{r}) \delta D(\mathbf{r}_0) \rangle$  is the

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- For short-range correlated fluctuation  $K(\kappa)$  is constant and  $\propto \langle \Delta D^2 \rangle$  and we obtain

$$G_0^{-1}(\mathbf{k}, s) = s + D_0 k^2 - k^2 D_1 |s|^{d/2} \equiv s + D(s)$$

## Perturbation theory

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with  $D_1 \propto \langle \Delta D^2 \rangle$ .

- It can be shown that  $D(\omega)$  is the Laplace transform of the velocity autocorrelation function

$$D(\omega) = \int_0^\infty e^{-st} \underbrace{\langle v_x(t) v_x(0) \rangle}_{Z(t)}$$

- This implies

$$Z(t) \propto t^{-(d+2)/2}$$

non-exponential long-time tail

## Analogy with a mass-spring vibrational problem

- Consider a system of point masses connected with springs with equation of motion for **scalar** elongations

$$\rho_m \frac{d^2}{dt^2} u_i = - \sum_j W_{ij} (u_i - u_j)$$

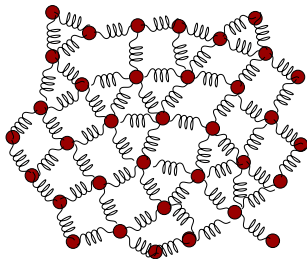
- By the replacement

$$\frac{d^2}{dt^2} \leftrightarrow \frac{d}{dt}$$

or

$$-\omega^2 \leftrightarrow i\omega$$

one can transfer conclusions from the disordered diffusion problem to the disordered vibrational problem.



## Analogy with a mass-spring vibrational problem

- Unperturbed Green's function

$$G_0 = \frac{1}{i\omega + \epsilon + k^2 D_0}$$

- Green's function with disorder

$$G = \frac{1}{i\omega + \epsilon + k^2 D(\omega)}$$

- Frequency-dependent diffusivity  $D(\omega)$
- Nonanalytic property

$$D(\omega) \propto |\omega|^{d/2}$$

- Unperturbed Green's function

$$G_0 = \frac{1}{-\omega^2 + i\epsilon + k^2 c_0^2}$$

- Green's function with disorder

$$G = \frac{1}{-\omega^2 + i\epsilon + k^2 c^2(\omega)}$$

- Frequency-dependent sound velocity  $c^2(\omega)$
- Nonanalytic property

$$\text{Im}\{c(\omega)\} \propto \Gamma(\omega)/\omega \propto |\omega|^d$$

# Analogy with a mass-spring vibrational problem

- Green's function with disorder

$$G = \frac{1}{i\omega + \epsilon + k^2 D(\omega)}$$

- Frequency-dependent diffusivity  $D(\omega)$
- Nonanalytic property

$$D(\omega) \propto |\omega|^{d/2}$$

- Longtime tail

- Green's function with disorder

$$G = \frac{1}{-\omega^2 + i\epsilon + k^2 c^2(\omega)}$$

- Frequency-dependent sound velocity  $c^2(\omega)$
- Nonanalytic property

$$\text{Im}\{c(\omega)\} \propto \Gamma(\omega)/\omega \propto |\omega|^d$$

- Rayleigh scattering



# Diffusivity in disordered materials and AC conductivity

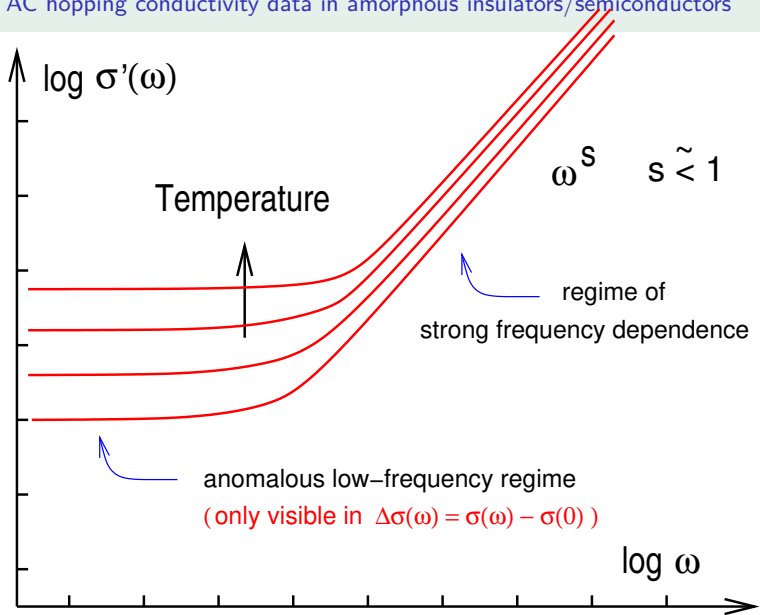
- Nernst-Einstein relation

$$\sigma(\omega) = \frac{N}{V} \frac{e^2}{k_B T} D(\omega)$$

⇒ Loss function

$$\epsilon''(\omega) \propto \frac{\sigma'(\omega) - \sigma(0)}{\omega} \propto \omega^{1/2}$$

## Typical AC hopping conductivity data in amorphous insulators/semiconductors



## Coherent-potential approximation (CPA) on a lattice

- We consider again the Master equation

$$\frac{d}{dt}P_i(t) = - \sum_j W_{ij} \left( P_i(t) - P_j(t) \right)$$

where we now assume that the sites are on a **simple-cubic lattice** and the transition rates  $W_{ij}$  are random numbers with some distribution density  $P(W_{ij})$ .

- The Green's function of this equation  $\mathcal{G}_{ij}(t, t')$  obeys the equation

$$\frac{d}{dt}\mathcal{G}_{ij}(t, t') + \sum_{\ell} W_{i\ell}(\mathcal{G}_{ij} - \mathcal{G}_{\ell j}) = \delta_{ij}\delta(t - t')$$

- Defining a "Hamiltonian Matrix"

$$\mathcal{H}_{ij} = \begin{cases} -\sum_j W_{ij} & i = j \\ W_{ij} & i \neq j \end{cases}$$

and Laplace transforming we obtain the following matrix equation for the Green matrix  $\langle i|\mathcal{G}(\omega)|j \rangle = \mathcal{G}_{ij}(\omega)$

$$(\tilde{\xi} - \mathcal{H})\mathcal{G} = (i\omega^2 + \epsilon - \mathcal{H})\mathcal{G} = 1$$

## Coherent-potential approximation (CPA) on a lattice

- The CPA is now constructed as follows

We invent an *effective medium*, which is not disordered (i. e. it has the cubic symmetry), but the force constants are frequency dependent:

$$W_{ij}^{eff}(s) = \Gamma(s)$$

Let  $Z = 6$  be the coordination number of the sites. Then effective-Medium Hamiltonian is

$$H_{ij} = \begin{cases} -\sum_j W_{ij}^{eff}(s) - Z\Gamma(s) & i = j \\ W_{ij}^{eff}(s) = \Gamma(s) & i \neq j \end{cases}$$

- The Green's function of the effective medium obeys the equation of motion

$$\tilde{s}G_{ij} - \delta_{ij} = Z\Gamma(s)(G_{\ell j} - G_{ij}) \quad \ell \text{ arbitrary neighboring site}$$

- We have solved this equation already with the result

$$G(\mathbf{k}, s) = \sum_{ij} e^{i\mathbf{k}\mathbf{r}_i - \mathbf{r}_j} G_{ij}(s) = \frac{1}{s + \Gamma(s)[6 - 2\cos(k_x a) + 2\cos(k_y a) + 2\cos(k_z a)]}$$

## Coherent-potential approximation (CPA) on a lattice

- We now "dig a hole" into the effective medium. As we deal with *pairs* of sites this hole must contain a pair  $(i_0, j_0)$
- Inside the hole we replace the effective-medium force constant  $\Gamma(\tilde{z})$  by the actual one  $W_{i_0 j_0}$  so that we obtain a "perturbation"

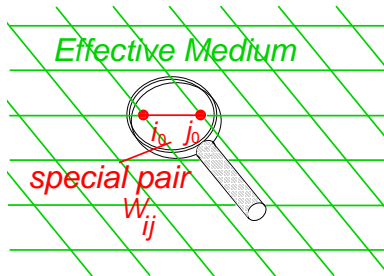
$$v_{i_0 j_0}(s) = W_{i_0 j_0} - \Gamma(s)$$

- The corresponding perturbing Hamiltonian matrix  $V$  has four non-zero entries, namely

$$v_{i_0 i_0}, v_{j_0 j_0}, v_{i_0 j_0}, \text{ and } v_{j_0 i_0}$$

- In the  $i_0 j_0$  subspace we have

$$V = \begin{pmatrix} -v_{i_0 j_0}(s) & v_{i_0 j_0}(s) \\ v_{i_0 j_0}(s) & -v_{i_0 j_0}(s) \end{pmatrix}$$



## Coherent-potential approximation (CPA) on a lattice

- We now demand that introducing this perturbation should have **on the average** no influence on the effective medium which is equivalent to demanding that the Green's function of the effective medium should be equal to the configuratonally averaged Green's function of the disordered system.

- This means that the  **$T$  matrix** corresponding to  $V$  should vanish on the average

$$\langle T \rangle = \left\langle \frac{V}{1 - VG} \right\rangle$$

- This is a  $2 \times 2$  matrix equation, with 4 identical matrix elements

$$\left\langle \frac{W - \Gamma(\tilde{z})}{1 + (W - \Gamma(\tilde{z}))^2 [G_{ij}(s) - G_{ii}(s)]} \right\rangle = 0$$

and, taking the medium equation of motion into account

$$\left\langle \frac{W - \Gamma(\tilde{z})}{1 + (W - \Gamma(\tilde{z})) \frac{2}{Z\Gamma(\tilde{z})} (1 - \tilde{z}G_{ii}(\tilde{z}))} \right\rangle = 0$$

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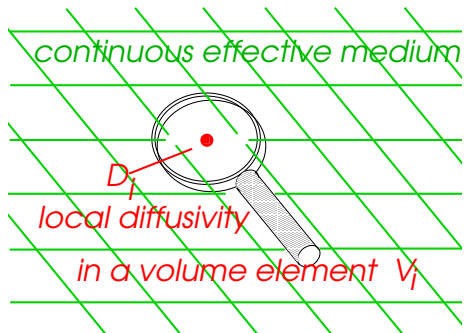
and, taking the medium equation of motion into account

$$\left\langle \frac{W - \Gamma(\tilde{z})}{1 + (W - \Gamma(\tilde{z})) \frac{2}{z\Gamma(\tilde{z})} (1 - \tilde{z}G_{ii}(\tilde{z}))} \right\rangle = 0$$

- This is the self-consistent CPA equation for the frequency-dependent rate  $\Gamma(s)$ , which is proportional to  $D(s)$  on the lattice with fluctuating  $W_{ij}$

# Off-lattice CPA for a spatially fluctuating diffusivity

S. Köhler, G. Ruocco, WS, PRB 2013



obeying the coarse-grained equation

$$(-i\omega + D(\omega)k^2)\rho(k, \omega) = 0$$

**Self-consistent CPA condition:**

$$\left\langle \frac{D(\omega) - D_i(\mathbf{r})}{1 - \frac{\nu}{3}[D(\omega) - D_i(\mathbf{r})]\Lambda(\omega)} \right\rangle_E = 0$$

$$\Lambda(\omega) = \frac{1}{N} \sum_{|\mathbf{k}| < k_g} \frac{k^2}{-i\omega + D(\omega)k^2} \rightarrow \frac{1}{D(0)} \text{ for } \omega \rightarrow 0$$

- derived as saddle-point of replica field theory
- can also be obtained as the continuum limit of the lattice CPA.



## The dc diffusivity in CPA and percolation

CPA equation in the dc ( $\omega \rightarrow 0$ ) limit:

$$\frac{\nu}{3}D(0) = D(0) \left\langle \frac{1}{1 + 2\frac{D(0)}{D_i}} \right\rangle \Rightarrow \frac{\nu}{3} = \left\langle \frac{1}{1 + 2\frac{D(0)}{D_i}} \right\rangle \text{ if } D(0) \neq 0$$

• Percolation model:  $P(D_i) = p\delta(D_i - \tilde{D}) + (1 - p)\delta(D_i)$

$$\Rightarrow \frac{\nu}{3} = p \frac{1}{1 + 2\frac{D(0)}{\tilde{D}}} \Rightarrow D(0) = \frac{3}{2\nu} \tilde{D}(p - p_c) \quad \text{with } p_c = \frac{\nu}{3}$$

$$D(0) = 0 \quad \text{for } p < p_c$$

• The parameter  $\frac{\nu}{3}$  is the percolation threshold  $p_c$ .

## Activated diffusivity and percolation

### Activated diffusion:

$$D_i = D_0 e^{-E_i/k_B T} \quad \text{with a given } P(E_i)$$

### CPA equation for $\omega = 0$ :

$$\frac{\nu}{3} = p_c = \left\langle \frac{1}{1 + 2 \frac{D(0)}{D_i}} \right\rangle$$

Parametrizing  $D(0) = \frac{D_0}{2} e^{-E_D/k_B T}$  we obtain

$$p_c = \int dE P(E) \frac{1}{e^{(E-E_D)/k_B T} + 1}$$

For low enough temperatures this gives

$$p_c = \int_0^{E_D} dE P(E) \quad (*)$$

- $D(0)$  obeys an Arrhenius law irrespective of the shape of  $P(E)$ !
- Equation (\*) corresponds to the **percolation construction** of hopping conductivity (Ambegaokar, Halperin, Langer, 1971, Shklovski, Efros 1984)

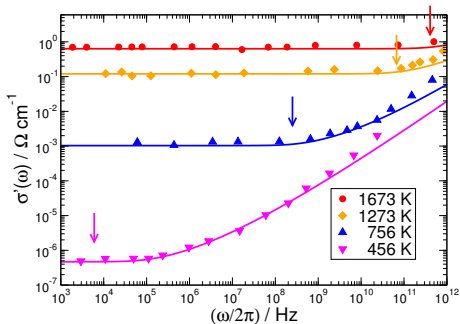
## Activated diffusivity and percolation

- The summed energies up to the percolation threshold  $E_D$  must be equal to the bond-percolation concentration:

$$p_c = \int_0^{E_D} dE P(E) \quad D(0) = \frac{1}{2} D_0 e^{-E_D/k_B T}$$

- This equation is identical to the low-temperature limit of the dc CPA equation.
- Therefore the CPA in this limit (which is equal to Bruggeman's (1935) EMA) is equivalent to the percolation construction.  
(This result is already known for some time  
See e.g. Dyre, Schrøder, Rev. Mod. Phys. **72**, 873, 2000)

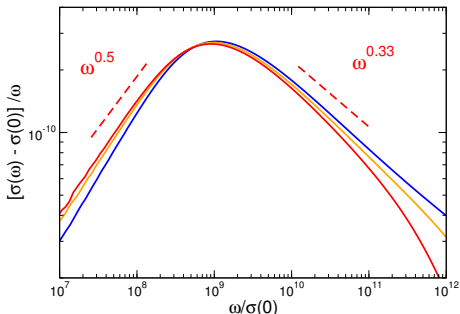
## CPA ac diffusivity calculation for a constant barrier distribution



 The data points are ac ionic conductivity data compiled by Wong and Angell in their book *Glass: Structure by spectroscopy*

## Loss function for the same data

$$\text{Loss function} = \left( \sigma'(\omega) - \sigma(0) \right) / \omega$$



• For small frequency  $D(\omega)$  exhibits a non-analyticity of the form

$$D(\omega) = D(0) + A\omega^{3/2}$$

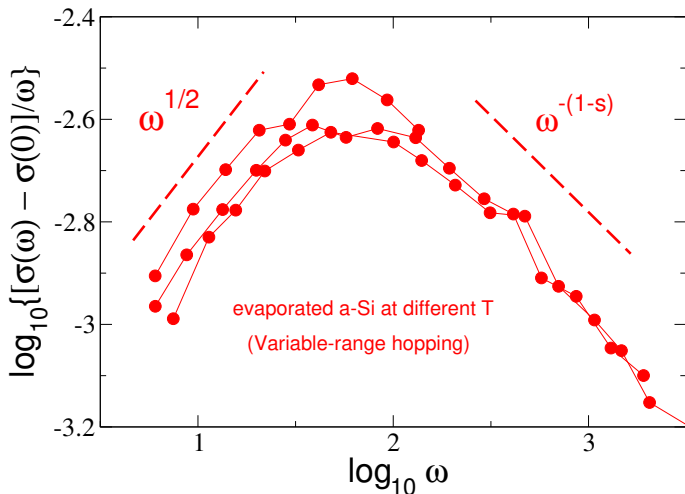
• Because  $D(\omega)$  is just the Laplace transform of the velocity autocorrelation function  $Z(t)$  this is equivalent to a long-time behaviour

$$Z(t) \propto t^{-5/2} \quad (\text{"long-time tail"})$$

## Measured ac conductivity data in amorphous silicon

$$\text{Loss function } [\sigma'(\omega) - \sigma'(0)] / \omega$$

(A. R. Long et al. Philos. Mag. B **58**, 153 (1988) )



## Take-home messages

- In electronic and ionic hopping transport with exponentially varying hopping rates there exists an underlying percolation aspect: The carriers choose the way of the least resistance.
- Diffusive transport can be well described with a spatially fluctuating diffusivity.
- Lowest-Order perturbation theory provides an explanation for the long-time tail of the velocity-autocorrelation function.
- In an analogy between the heterogeneous diffusion equation and heterogeneous sound propagation the diffusional anomaly corresponds to Rayleigh scattering.
- For disordered diffusion there exists an extensive frequency regime with a strong frequency dependence of the conductivity.
- The coherent-potential approximation (CPA) describes well the percolative aspect and the frequency-dependence.