

The THEORY of APERIODIC SOLIDS

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Lecture II

COHERENT & DISSIPATIVE TRANSPORT

Main References

J. BELLISSARD, H. SCHULZ-BALDES, *A Kinetic Theory for Quantum Transport in Aperiodic Media*,
J. Stat. Phys., **91**, 991-1026, (1998).

D. SPEHNER, J. BELLISSARD, *A Kinetic Model for Quantum Jumps*,
J. Stat. Phys., **104**, 525-572, (2001).

J. BELLISSARD, R. REBOLLEDO, D. SPEHNER, W. VON WALDENFELS,
The Quantum Flow of Electronic transport I: The finite volume case,
in preparation.

J. BELLISSARD,,
Coherent and dissipative transport in aperiodic solids,
Lecture Notes in Physics, **597**, Springer (2003), pp. 413-486.

Content

1. Why Revisiting Transport ?
2. Coherent Transport
3. Dissipative Transport
4. Conclusion

I - Why Revisiting Transport ?

- It is a very old problem
BOLTZMANN (1872-80) for classical systems;
DRUDE (1900) for electrons.
- It is treated in textbooks: phenomenology, perturbation theory, numerical calculations.

I.1)- Motivations

Conceptual Difficulties

1. No mathematically rigorous proof of the Kubo formulæ for transport coefficients.
(However substantial progress for classical systems (LEBOWITZ's school) and for quantum ones (PILLET-JAKSIC, FRÖHLICH *et. al.*)).
2. Low temperature effects are difficult to describe
ex. : Mott's variable range hopping
(see e.g. EFROS & SCHKLOVSKY)
3. Aperiodic materials escape Bloch theory : need for a more systematic treatment
(**ex.** : quasicrystals).
4. Aperiodic media exhibit anomalous quantum diffusion

Transport is complex

- Thermodynamic quantities are much easier to measure: experiments are cleaner, easier to control.
Ex. : heat capacity, magnetic susceptibility, structure factors... .
But they do not separate various mechanisms.
- Transport measurements are mostly indirect: harder to interpret (especially at low temperature). Too many mechanisms occur at once.

Few mechanisms

1. For metals, $\sigma(\mathbf{T})$ increases as temperature decreases

$$\sigma(\mathbf{T}) \stackrel{T \downarrow 0}{\sim} \mathbf{T}^{-2}, \quad (\text{Fermi liquid theory}).$$

2. For a thermally activated process

$$\sigma(\mathbf{T}) \stackrel{T \downarrow 0}{\sim} e^{-\Delta/\mathbf{T}} \quad (\text{If a gap holds at Fermi level}).$$

3. For weakly disordered systems

$$\sigma(\mathbf{T}) \xrightarrow{T \downarrow 0} \sigma(\mathbf{0}) > \mathbf{0} \quad (\text{residual conductivity}).$$

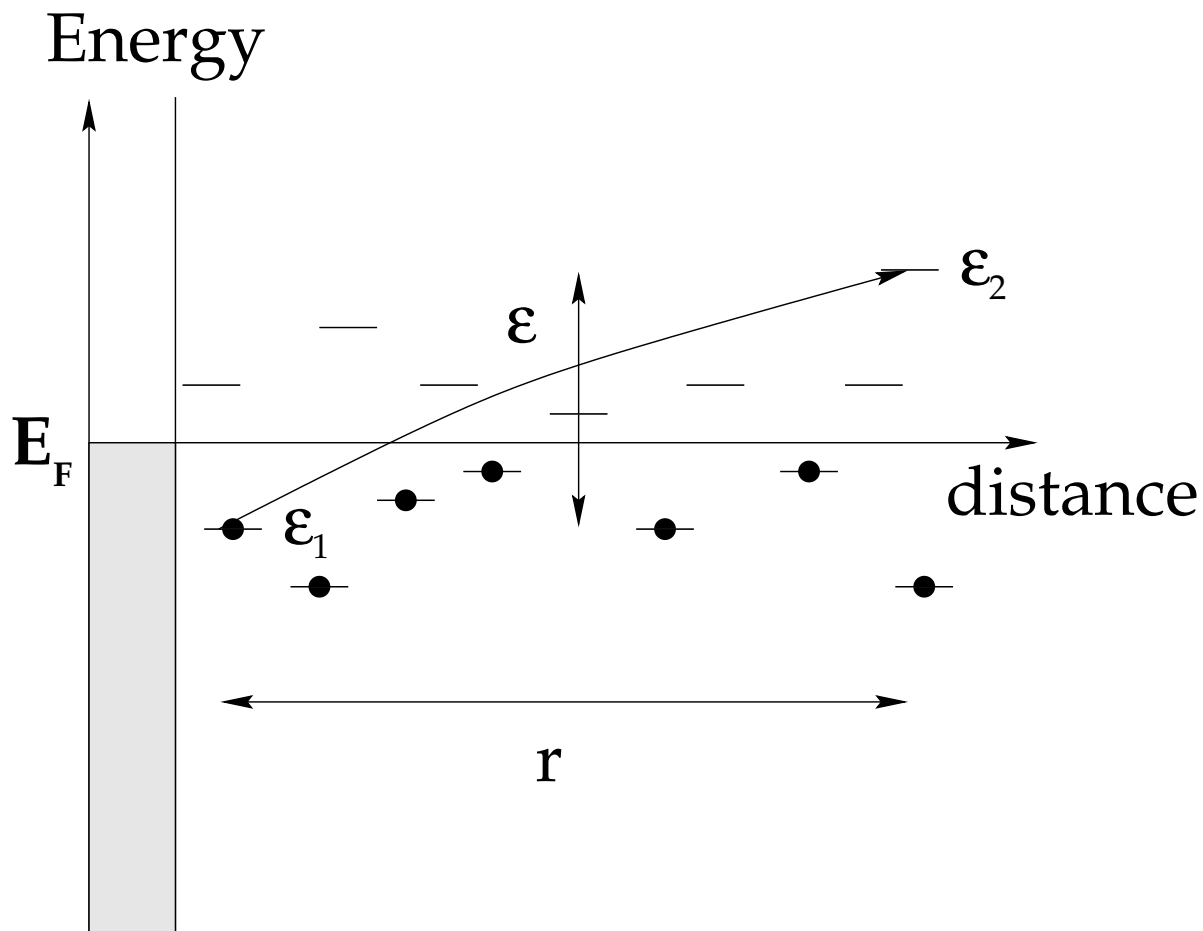
4. For strongly disordered systems in $3D$

$$\sigma(\mathbf{T}) \stackrel{T \downarrow 0}{\sim} e^{-(\mathbf{T}_0/\mathbf{T})^{1/4}} \quad (\text{variable range hopping}).$$

I.2)- Mott's variable range hopping

N. MOTT, (1968).

B. SHKLOVSKII, A. L. EFFROS, *Electronic Properties of Doped Semiconductors*, Springer-Verlag, Berlin, (1984).



- Strongly localized regime, dimension d
- Low electronic DOS, Low temperature

- Absorption-emission of a phonon of energy ε

$$\text{Prob} \propto e^{-\varepsilon/k_B T}$$

- Tunnelling probability at distance \mathbf{r}

$$\text{Prob} \propto e^{-r/\xi}$$

- Density of state at Fermi level n_F ,

$$\varepsilon n_F r^d \approx 1$$

- Optimizing, the conductivity satisfies

$$\sigma \propto e^{-(T_0/T)^{1/d+1}} \quad \textit{Mott's law}$$

- Optimal energy $\varepsilon_{\text{opt}} \sim T^{d/(d+1)} \gg T$

- Optimal distance $\mathbf{r}_{\text{opt}} \sim 1/T^{1/(d+1)} \gg \xi$

I.3)- Transport in Quasicrystals

Lectures on Quasicrystals,

F. Hippert & D. Gratias Eds., Editions de Physique, Les Ulis, (1994),

S. ROCHE, D. MAYOU AND G. TRAMBLY DE LAISSARDIÈRE,

Electronic transport properties of quasicrystals, J. Math. Phys., **38**, 1794-1822 (1997).

Quasicrystalline alloys :

Metastable QC's: **AlMn**

(Shechtman D., Blech I., Gratias D. & Cahn J., PRL **53**, 1951 (1984))

AlMnSi

AlMgT ($T = Ag, Cu, Zn$)

Defective stable QC's: **AlLiCu** (*Sainfort-Dubost, (1986)*)

GaMgZn (*Holzen et al., (1989)*)

High quality QC's: **AlCuT** ($T = Fe, Ru, Os$)

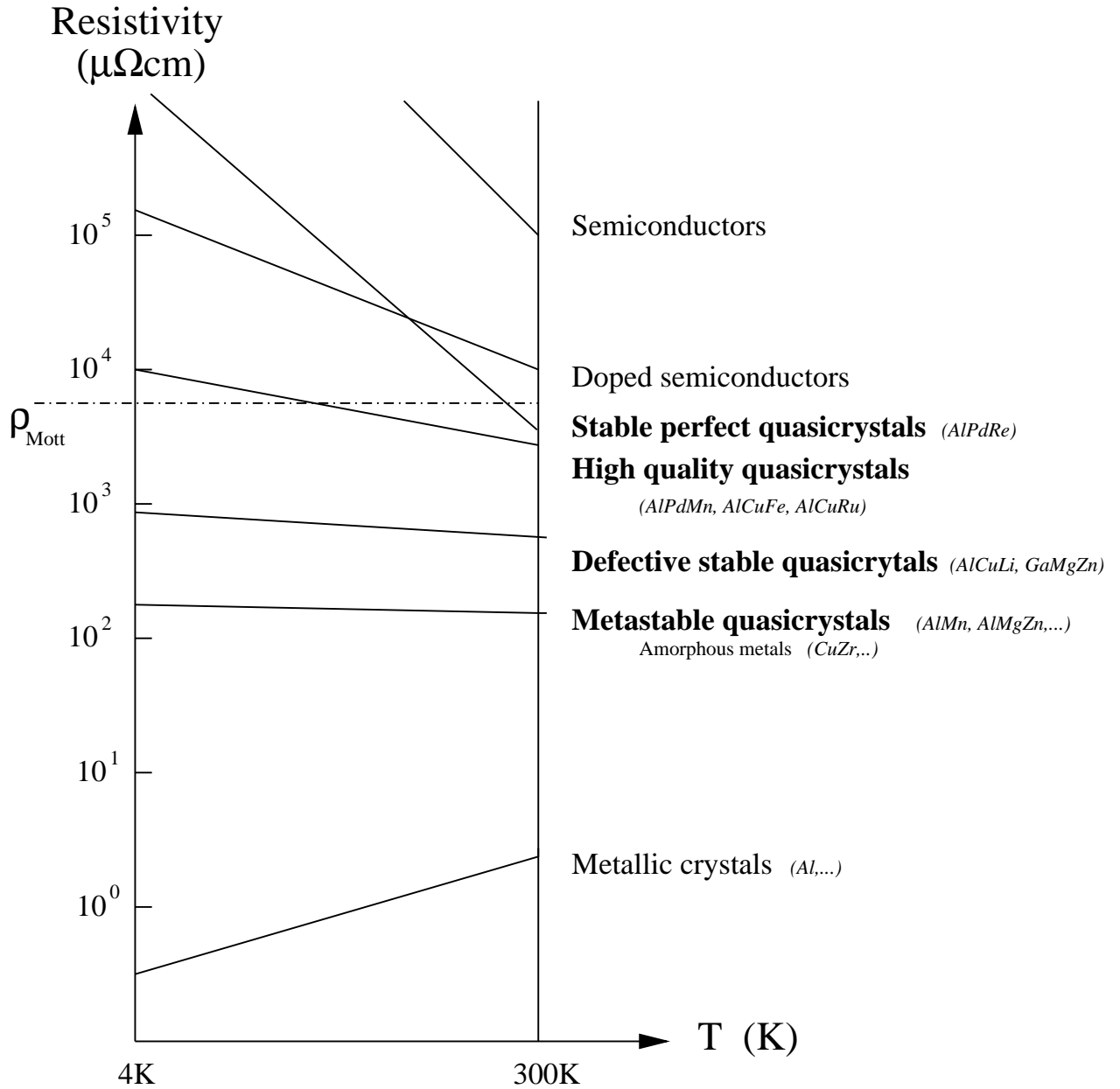
(*Hiraga, Zhang, Hirakoyashi, Inoue, (1988); Gurnan et al., Inoue et al., (1989);*

Y. Calvayrac et al., (1990))

“Perfect” QC's:

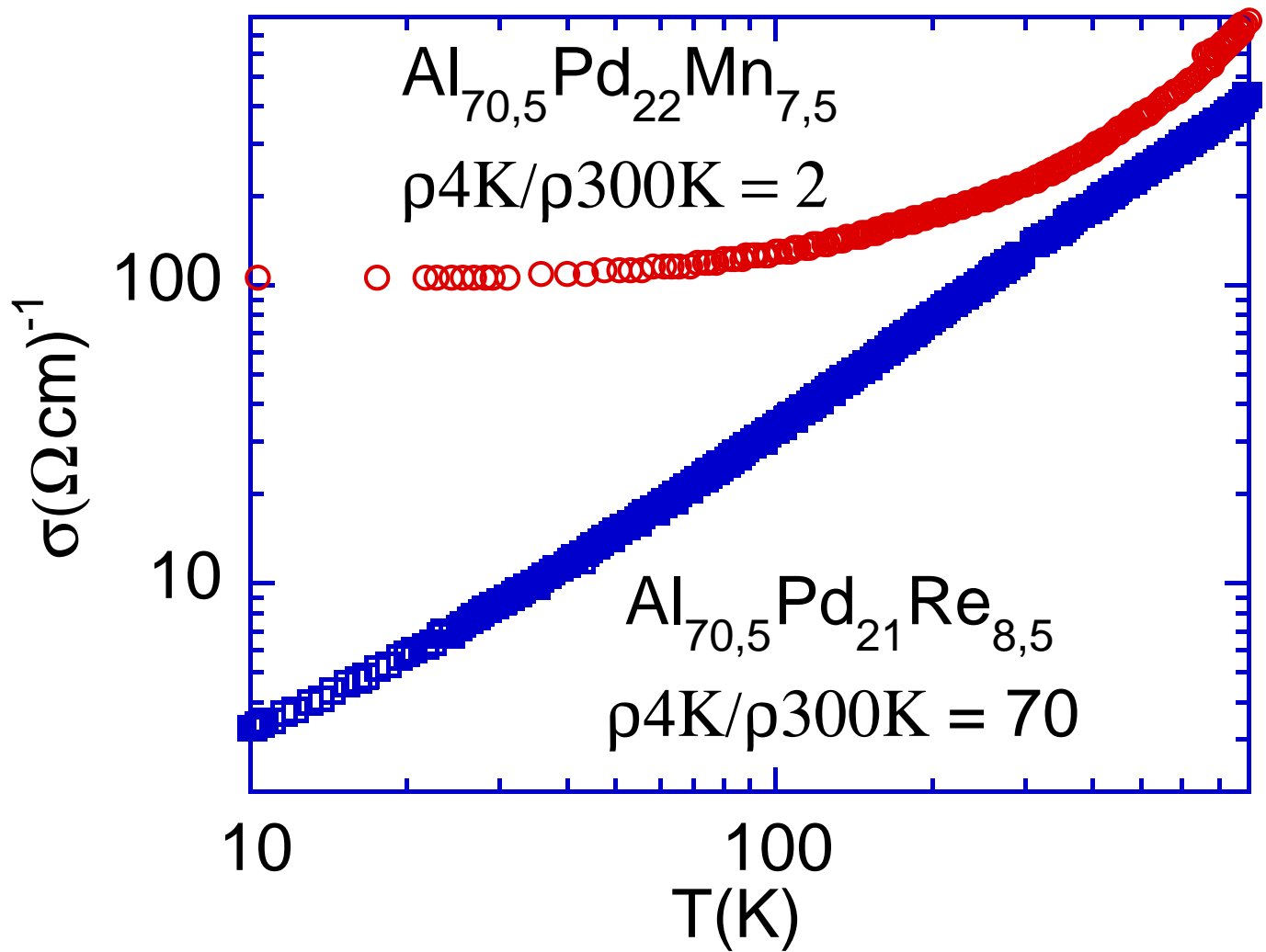
AlPdMn

AlPdRe



Typical values of the resistivity

(Taken from *C. Berger* in ref. *Lectures on Quasicrystals*)



Conductivity of Quasicrystals vs Temperature

$$\sigma \approx \sigma_0 + aT^\gamma \text{ with } 1 < \gamma < 1.5$$

$$\text{for } .01 \text{ K} \leq T \leq 1000 \text{ K}$$

For Quasicrystals

1. *Al, Fe, Cu, Pd* are very good metals : why is the conductivity of quasicrystalline alloys so low ?
Why is it decreasing ?
2. At high enough temperature

$$\sigma \propto T^\gamma \quad 1 < \gamma < 1.5$$

There is a new mechanism here!

3. At low temperature for $\text{Al}_{70.5}\text{Pd}_{22}\text{Mn}_{7.5}$,

$$\sigma \approx \sigma(0) > 0$$

4. At low temperature for $\text{Al}_{70.5}\text{Pd}_{21}\text{Re}_{8.5}$,

$$\sigma \propto e^{-(T_0/T)^{1/4}} \quad \text{C. BERGER } et al. (1998)$$

Is disorder playing any role at very low temperature ?

II - Coherent Transport

- Transport before collisions destroys *quantum coherence*.
- In aperiodic solids this transport can be *anomalous*.

II.1)- Mathematical Framework

1. Closing suitably the set of translated of the set of atomic positions leads to the *Hull* : it is a compact metrizable space Ω endowed with an \mathbb{R}^d -action.
2. An invariant ergodic probability measure \mathbb{P} is provided by the Gibbs state at zero temperature.
3. Observables are *random operators* $A = (A_\omega)_{\omega \in \Omega}$ acting on the Hilbert space \mathcal{H} of quantum states (such as $L^2(\mathbb{R}^d)$ for spinless electrons) with:
 - (a) Covariance : $T(a)A_\omega T(a)^{-1} = A_{\tau^{-1}a}$.
 - (b) $\omega \mapsto A_\omega$ is strongly continuous.
4. The trace per unit volume, defined by \mathbb{P} , exists:

$$\mathcal{T}_{\mathbb{P}}(A) = \lim_{\Lambda \uparrow \mathbb{R}^d} \frac{1}{|\Lambda|} \text{Tr}(A_\omega \upharpoonright_\Lambda) = \int_{\Omega} d\mathbb{P}(\omega) \langle x | A_\omega | x \rangle$$
5. Differential: $(\vec{\nabla} A)_\omega = -i[\vec{X}, A_\omega]$

II.2)- Local Exponents

Given a positive measure μ on \mathbb{R} :

$$\alpha_{\mu}^{\pm}(E) = \lim \left\{ \begin{array}{c} \sup \\ \inf \end{array} \right\}_{\varepsilon \downarrow 0} \frac{\ln \int_{E-\varepsilon}^{E+\varepsilon} d\mu}{\ln \varepsilon}$$

For Δ a Borel subset of \mathbb{R} :

$$\alpha_{\mu}^{\pm}(\Delta) = \mu\text{-ess} \left\{ \begin{array}{c} \sup \\ \inf \end{array} \right\}_{E \in \Delta} \alpha_{\mu}^{\pm}(E)$$

1. For all E , $\alpha_{\mu}^{\pm}(E) \geq 0$.
 $\alpha_{\mu}^{\pm}(E) \leq 1$ for μ -almost all E .
2. If μ is *ac* on Δ then $\alpha_{\mu}^{\pm}(\Delta) = 1$,
 if μ is *pp* on Δ then $\alpha_{\mu}^{\pm}(\Delta) = 0$.
3. If μ and ν are equivalent measures on Δ , then
 $\alpha_{\mu}^{\pm}(E) = \alpha_{\nu}^{\pm}(E)$ μ -almost surely.
4. α_{μ}^{+} coincides with the *packing dimension*.
 α_{μ}^{-} coincides with the *Hausdorff dimension*.

II.3)- Fractal Exponents

For $p \in \mathbb{R}$:

$$D_{\mu, \Delta}^{\pm}(q) = \lim_{q' \rightarrow q} \frac{1}{q' - 1} \lim_{\varepsilon \downarrow 0} \left\{ \begin{array}{c} \sup \\ \inf \end{array} \right\} \frac{\ln \left(\int_{\Delta} d\mu(E) \left\{ \int_{E-\varepsilon}^{E+\varepsilon} d\mu \right\}^{q'-1} \right)}{\ln \varepsilon}$$

1. $D_{\mu, \Delta}^{\pm}(q)$ is a non decreasing function of q .
2. $D_{\mu, \Delta}^{\pm}(q)$ is **not** an invariant of the measure class, in general.
- 3.(a) If μ is *ac* on Δ then $D_{\mu, \Delta}^{\pm}(q) = 1$.
 (b) If μ is *pp* on Δ then $D_{\mu, \Delta}^{\pm}(q) = 0$.

II.4)- Spectral Exponents

Given a Hamiltonian $H = (H_\omega)_{\omega \in \Omega}$, namely a selfadjoint observable, we define:

1. The *local density of state* (LDOS) is the spectral measure of H_ω relative to a vector $\varphi \in \mathcal{H}$.
2. The corresponding local exponent is obtained after maximizing (+) or minimizing (-) over φ . It is denoted α_{LDOS}^\pm . It is $\mathbb{P} - a. s.$ independent of ω .
3. The *density of states* (DOS) as the measure defined by

$$\int d\mathcal{N}_{\mathbb{P}}(E) f(E) = \mathcal{T}_{\mathbb{P}}(f(H))$$

4. The local exponent associated with the DOS is denoted by α_{DOS}^\pm .
5. Inequality : $\alpha_{\text{LDOS}}^\pm(\Delta) \leq \alpha_{\text{DOS}}^\pm(\Delta)$.
6. The fractal exponents for the LDOS are defined in the same way, provided we consider the average over ω before taking the logarithm and the limit $\varepsilon \downarrow 0$.

II.5)- Transport Exponents

1. For $\Delta \subset \mathbb{R}$ Borel, let $P_{\Delta, \omega}$ be the corresponding spectral projection of H_{ω} . Set:

$$\vec{X}_{\omega}(t) = e^{itH_{\omega}} \vec{X} e^{-itH_{\omega}}$$

2. The averaged spread of a typical wave packet with energy in Δ is measured by:

$$L_{\Delta}^{(p)}(t) = \left(\int_0^t \frac{ds}{t} \int_{\Omega} d\mathbb{P} \langle x | P_{\Delta, \omega} | \vec{X}_{\omega}(t) - \vec{X} |^p P_{\Delta, \omega} | x \rangle \right)^{1/p}$$

3. Define $\beta = \beta_p^{\pm}(\Delta)$ similarly so that $L_{\Delta}^{(p)}(t) \sim t^{\beta}$.

4. $\beta_p^{-}(\Delta) \leq \beta_p^{+}(\Delta)$.

$\beta_p^{\pm}(\Delta)$ are non decreasing in p .

5. *Heuristic*

$\beta = 0 \rightarrow$ absence of diffusion (*ex: localization*),

$\beta = 1 \rightarrow$ ballistic motion (*ex: in crystals*),

$\beta = 1/2 \rightarrow$ quantum diffusion

(*ex: weak localization*).

$\beta < 1 \rightarrow$ subballistic regime,

$\beta < 1/2 \rightarrow$ subdiffusive regime

(*ex: in quasicrystals*).

II.6)- Inequalities

1. *Guarneri's inequality*: (Guarneri '89, Combes , Last '96)

$$\beta_p^\pm(\Delta) \geq \frac{\alpha_{\text{LDOS}}^\pm(\Delta)}{d}$$

2. *BGT inequalities*: (Barbaroux, Germinet, Tcheremchantsev '00)

$$\beta_p^\pm(\Delta) \geq \frac{1}{d} D_{\text{LDOS},\Delta}^\pm\left(\frac{d}{d+p}\right)$$

3. *Heuristics*:

- (a) *ac* spectrum implies $\beta \geq 1/d$.
- (b) *ac* spectrum implies ballistic motion in $d = 1$
- (c) *ac* spectrum is compatible with quantum diffusion in $d = 2$. This is expected in weak localization regime.
- (d) *ac* spectrum is compatible with subdiffusion for $d \geq 3$.

II.7)- Current-Current Correlation

1. The *coherent* current operator is the product of the electric charge of charge carriers by their velocity namely (It belongs to the algebra of the Brillouin zone.):

$$\vec{J}_\omega = i \frac{e}{\hbar} [H_\omega, \vec{X}]$$

2. There is a Borel measure m on \mathbb{R}^2 such that, for $f, g \in \mathcal{C}_0(\mathbb{R})$

$$\mathcal{T}_{\mathbb{P}} \left\{ f(H) \vec{J} g(H) \vec{J} \right\} = \int_{\mathbb{R}^2} dm(E, E') f(E) g(E')$$

m is called the *current-current correlation*.

3. If $L_H = i[H, \cdot]$ is the *Liouville operator* acting on $L^2(\mathcal{A}, \mathcal{T}_{\mathbb{P}})$, then its spectral measure relative to the cyclic vector is the marginal of m with respect to the variable $E - E'$.
4. The diffusion exponent is the spectral exponent of L_H at eigenvalue 0.

II.8)- Results in Models

1. For Jacobi matrices (1D chains), the position operator is defined by the spectral measure (orthogonal polynomials) \Rightarrow transport exponents should be defined through the spectral ones.
2. For Jacobi matrices of a Julia set, with μ the σ -balanced measure (*Barbaroux, Schulz-Baldes '99*)

$$\beta_p^+ \leq D_\mu(1-p) \quad \text{for all } 0 \leq p \leq 2$$

3. If H_1, \dots, H_d are Jacobi matrices, η_1, \dots, η_d are positive numbers and if

$$H^{(\eta)} = \sum_{j=1}^d \eta_j \mathbf{1} \otimes \dots \otimes \mathbf{H}_j \otimes \dots \otimes \mathbf{1}$$

then (*Schulz-Baldes, Bellissard '00*)

$$\beta_p^+(H^{(\eta)}) = \max_j \beta_p^+(H_j)$$

$$\alpha_{\text{LDOS}}(H^{(\eta)}) = \min\left\{1, \sum_j \alpha_{\text{LDOS}}(H_j)\right\}$$

for a.e. η . In addition if $\sum_j \alpha_{\text{LDOS}}(H_j) > 1$, $H^{(\eta)}$ has *a.c. spectrum*.

4. For any $\epsilon > 0$, there is a Jacobi matrix H_0 such that if $H_j = H_0$, $\forall j$, $H^{(\eta)}$ has *a.c. spectrum* for $d \geq 3$ and spectral exponent $\leq 1/d - \epsilon$ for a.e. η .

(*Schulz-Baldes, Bellissard '00*)

5. There is a class of models of Jacobi matrices on an infinite dimensional hypercube with *a.c. spectrum* and vanishing transport exponents.

(*Vidal, Mosseri, Bellissard '99*)

II.9)- Anderson's Model

The Anderson model represents electrons in a disordered systems and is given by the Hamiltonian H acting on $\ell^2(\mathbb{Z}^d)$ as

$$H = -\Delta + \lambda V, \quad V = (V_x)_{x \in \mathbb{Z}^d}, \lambda > 0$$

where the V_x 's are *i.i.d* with $\langle V_x \rangle = 0$, $\langle V_x^2 \rangle = 1$.
Then (*Bellissard, Hislop '05*)

Theorem 1 *If H is the Hamiltonian for the Anderson model, at large λ the current-current correlation is absolutely continuous with analytic density away from the diagonal*

III - Dissipative Transport

- Transport beyond the collision time.
- Several mechanisms of dissipation may be considered: *electron-electron* collisions, interactions with *acoustic* phonons or *optical* ones. *etc..*

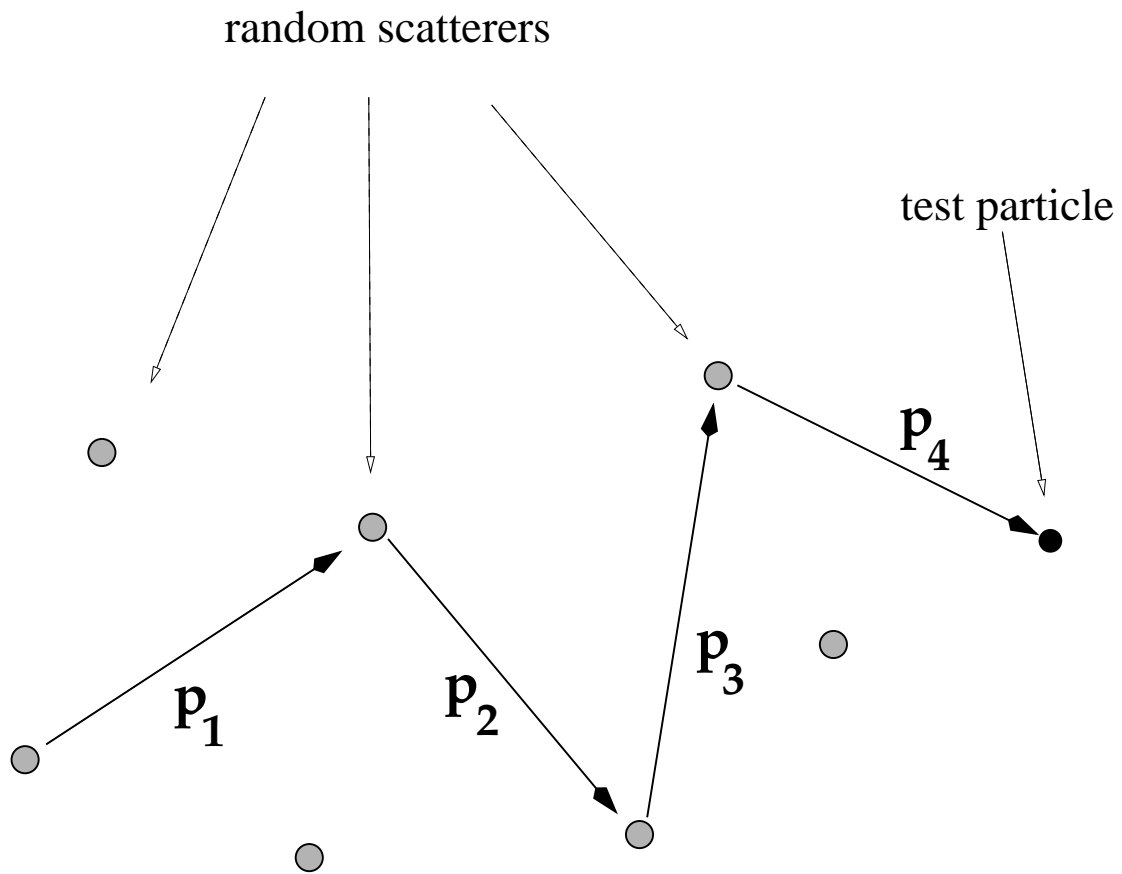
III.1)- The Drude Model

Assumptions :

1. Electrons in a metal are free classical particles of mass m_* and charge q .
2. They experience collisions at random Poissonian times $\dots < t_n < t_{n+1} < \dots$, with average relaxation time τ_{rel} .
3. If p_n is the electron momentum between times t_n and t_{n+1} , then the p_{n+1} 's is updated according to the Maxwell distribution at temperature T .

Then the conductivity follows the *Drude formula*

$$\sigma = \frac{q^2 n}{m_*} \tau_{rel}$$



The Drude Kinetic Model

III.2)- The Kubo Formula

(*Bellissard, Schulz-Baldes '95*)

1. Replace the classical dynamics by the quantum one electron dynamic in the aperiodic solid.
2. At each collision, force the density matrix to come back to equilibrium. (*Relaxation time Approximation* or RTA).
3. There is then one *relaxation time* τ_{rel} . The electric conductivity is then given by Kubo's formula:

$$\sigma_{i,j} = \frac{q^2}{\hbar} \mathcal{T}_{\mathbb{P}} \left(\partial_j \left(\frac{1}{1 + e^{\beta(H-\mu)}} \right) \frac{1}{1/\tau_{rel} - \mathcal{L}_H} \partial_i H \right)$$

Here q is the charge of the carriers, $\beta = 1/k_B T$, μ is the chemical potential and $\mathcal{L}_H = \iota/\hbar [H, \cdot]$.

4. For the Hilbert-Schmidt inner product defined by $\mathcal{T}_{\mathbb{P}}$, \mathcal{L}_H is anti-selfadjoint. Thus as $\tau_{rel} \uparrow \infty$, the resolvent of \mathcal{L}_H is evaluated closer to the spectrum near 0.

III.3)- The Anomalous Drude Formula

(*Mayou '92, Sire '93, Bellissard, Schulz-Baldes '95*)

$$\sigma \stackrel{\tau_{rel} \uparrow \infty}{\sim} \tau_{rel}^{2\beta_F - 1}$$

where β_F is the transport exponent $\beta_2(E_F)$ evaluated at Fermi level.

1. In practice, $\tau_{rel} \uparrow \infty$ as $T \downarrow 0$.
2. If $\beta_F = 1$ (*ballistic motion*), $\sigma \sim \tau_{rel}$ (*Drude*). The system behaves as a **conductor**.
3. For $1/2 < \beta_F \leq 1$, $\sigma \uparrow \infty$ as $T \downarrow 0$: the system behaves as a **conductor**.
4. If $\beta_F = 1/2$ (*quantum diffusion*), $\sigma \sim const.$: residual conductivity at low temperature.
5. For $0 \leq \beta_F < 1/2$, $\sigma \downarrow 0$ as $T \downarrow 0$: the system behaves as an **insulator**.

III.4)- Time Scales

1. The phonon mediated electron-electron interaction gives $\tau_{rel} \sim T^{-2}$ (*Fermi liquid* theory).
2. Electron interaction with acoustic phonons, gives $\tau_{rel} \sim T^{-5}$ (*Bloch* theory).
3. *Quantum chaos* in the one electron spectrum makes the Hamiltonian look like a *random matrix* like in *weak localization* regime (*Bellissard, Magnen, Rivasseau, '02*). This leads to a conductivity independent of the temperature.
4. Optical phonons are important in aperiodic system. They produce a band similar to the spectrum of a random matrix (*quantum chaos*). The influence of optical phonons on the relaxation time *is unknown*.

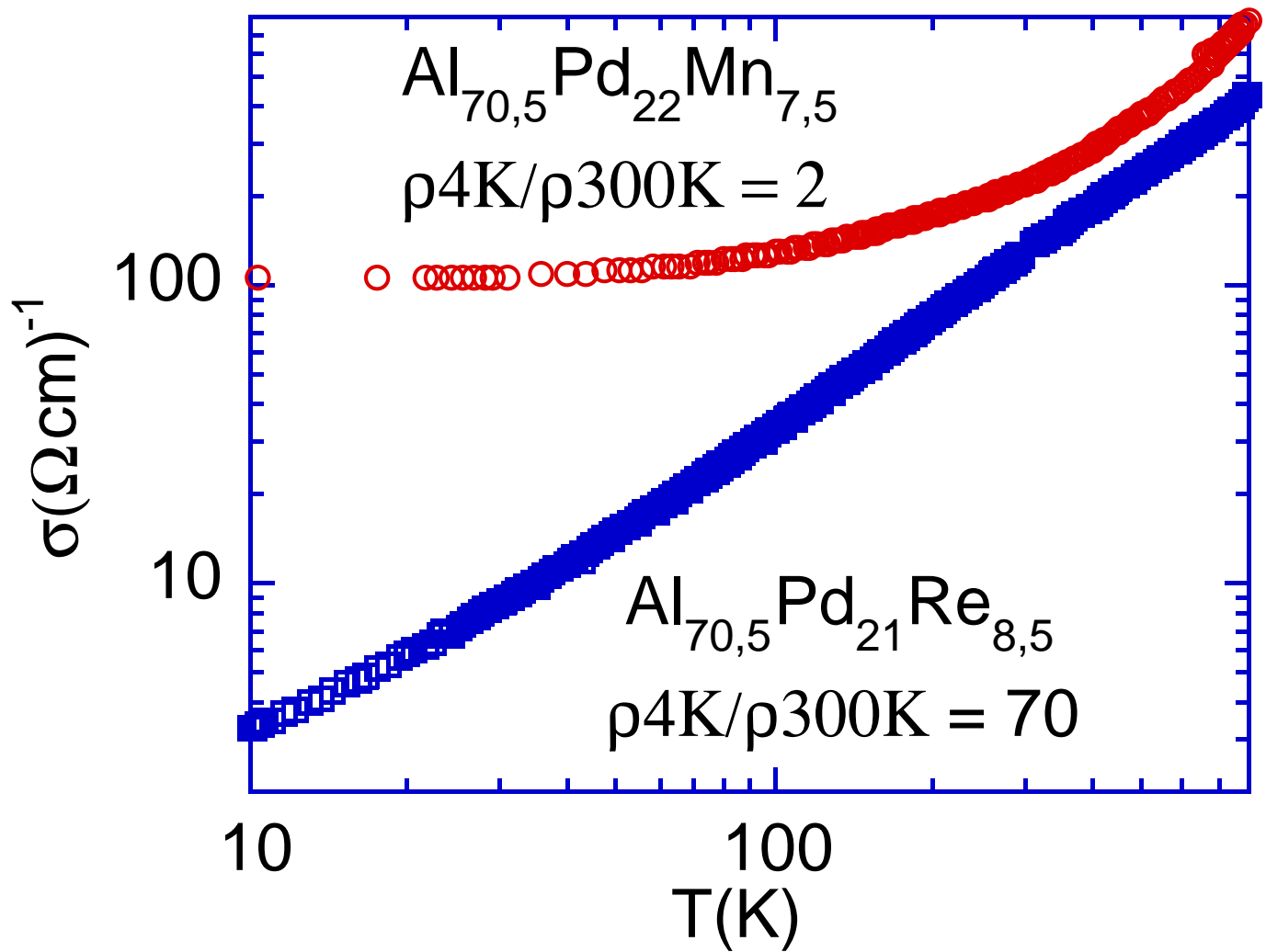
5. In the conducting regime, the *shortest* dissipative time scale dominates, favoring Fermi liquid theory:

$$\sigma \stackrel{T \downarrow 0}{\sim} T^{-2(2\beta_F - 1)} \quad \text{if } \beta_F > 1/2,$$

6. In the insulator regime, the *longest* dissipative time scale dominates. Thus Bloch theory is likely to dominate

$$\sigma \stackrel{T \downarrow 0}{\sim} T^{5(1 - 2\beta_F)} \quad \text{if } \beta_F < 1/2,$$

7. If there is an infinite number of time scales the low temperature behaviour is not a scaling law: for example in the Mott variable range hopping regime.



Conductivity of Quasicrystals vs Temperature

$$\sigma \approx \sigma_0 + aT^\gamma \text{ with } 1 < \gamma < 1.5$$

$$\text{for } .01 \text{ K} \leq T \leq 1000 \text{ K}$$

III.5)- Conductivity in Quasicrystals

S. Roche & Fujiwara, Phys. Rev., B58, 11338-11396, (1998).

1. LMTO *ab initio* computations for $i - AlCuCo$ give $\beta_F \approx 0,375 < 1/2$. Thus *Bloch's law* dominates giving

$$\sigma \underset{\sim}{\overset{T \downarrow 0}{\sim}} T^{5/4} \quad \text{if } \beta_F < 1/2,$$

compatible with experimental results !

2. If *disorder* dominates at low temperature
 - (a) for $AlPdMn$, the occurrence of a *residual conductivity* implies *weak localisation*. Thus there should be a *high density of defects or impurities*
 - (b) $AlPdRe$ behaves like an insulator with *Mott's variable range hopping conductivity*. This is a sign for *strong localization*, implying a *low density of defects or impurities*.

This seems with these materials being structurally similar and samples being high quality.

III.6)- Quantum Chaos in Quasicrystals

J.B. & D. Mayou, speculations '99

1. Numerical simulations performed for the octagonal lattice exhibit level repulsion and Wigner-Dyson's distribution (M. Schreiber, U. Grimm, R. A. Roemer, J. -X. Zhong, *Comp. Phys. Commun.*, **121-122**, 499-501 (1999)).
2. For a sample of size L in dimension d :
 Mean level spacing $\Delta \sim L^{-d}$.
 Thus Heisenberg time $\tau_H \sim L^d$.
3. Time necessary to reach the boundary (*Thouless*)
 $L \sim \tau_{Th}^{\beta_F}$. Thus $\tau_{Th}^{\beta_F} \sim L^{1/\beta}$.
4. Hence :
 - (a) if $\beta_F > 1/d$ level repulsion dominates implying
 - quantum diffusion $\langle x^2 \rangle \sim t$
 - *residual conductivity*
 - absolutely continuous spectrum at Fermi level;
 - (b) if $\beta_F < 1/d$ level repulsion can be ignored and
 - anomalous diffusion dominates $\langle x^2 \rangle \sim t^{2\beta_F}$
 - insulating behaviour with scaling law
 - *singular continuous* spectrum near Fermi level.

III.7)- Beyond the RTA

1. At low temperature, the RTA is invalid. There is a spectrum of relaxation times.
2. A kinetic model of *quantum jumps* has been proposed leading to the validity of linear response.
(*Spehner, Bellissard '00, Bellissard, Rebolledo, Spehner, von Waldenfels '00*).
3. The current admits two parts : the coherent one, induced by $\vec{J} = \imath[\vec{X}, H]$, and a dissipative one including other effects like *phonon drag*, etc.
4. The Kubo formula becomes more involved and can be decomposed into five contributions in general.
5. Applied to strongly localized electrons, this formalism gives rise to a justification of the *Abrahams and Miller random resistor network model*,
(*Spehner, Thesis '00, Spehner, Bellissard, '00*)
describing the *Mott variable range hopping*.

IV)- Conclusions :

1. The electron dynamics in an aperiodic solid can be described by using random operators and rules of Non Commutative Calculus.
2. The quantum evolution of a typical wave packet leads to anomalous diffusion, described through various spectral and transport exponents.
3. These exponents are related by inequalities that allow subdiffusion together with absolutely continuous spectrum for $d \geq 3$.
4. Dissipative mechanisms, such as electron-phonon interaction, may be described through kinetic models, generalizing the Drude model.
5. The interplay between coherent and dissipative transport is revealed at low temperature. Anomalous diffusion then leads to an anomalous Drude formula within the RTA.
6. The anomalous Drude formula may explain the behaviour of quasicrystals.
7. Beyond the RTA, the kinetic models are still valid but involve more conditions. One consequence is the justification of the Abraham-Miller random resistor network which usually leads to a better understanding of the Mott variable range hopping conductivity, in strongly disordered systems.