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A Unified Description of the Electrical Properties of Low-Density Polyethylene via the Dispersion Parameter

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A Unified Description of the Electrical Properties of Low-Density Polyethylene via the Dispersion Parameter

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October 11th, 2019

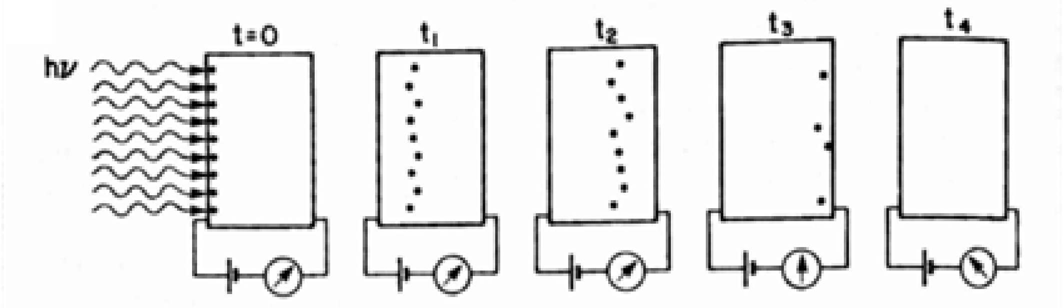
Outline

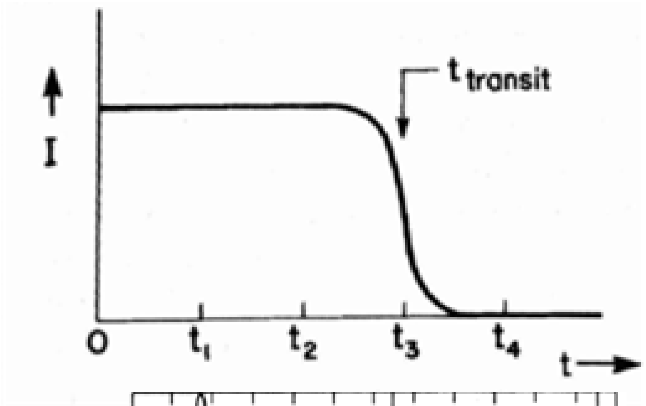
- Motivation
- Conduction
    - Crystalline Solids
    - Disordered Solids
- Measurements
    - Photoconductivity, DC conductivity, permittivity, radiation induced conductivity, electrostatic breakdown
- Critical Transitions
- Summary
- Conclusions

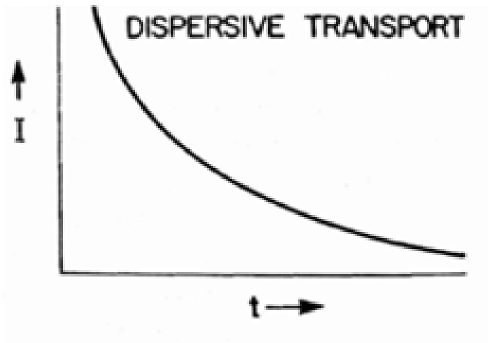
Why?

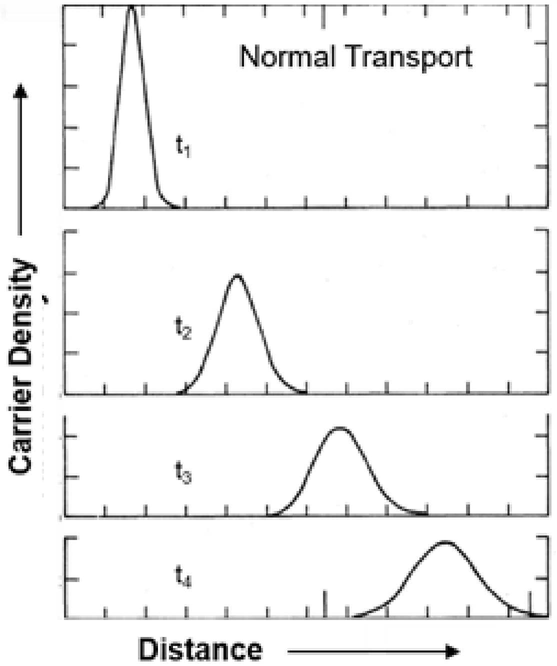
- Connect microscopic processes to macroscopic behavior
- Explain anomalous/dispersive behavior
- Theory has applications from spacecraft charging to HVDC cable insulation
- Defines many different material properties and measurements characteristics

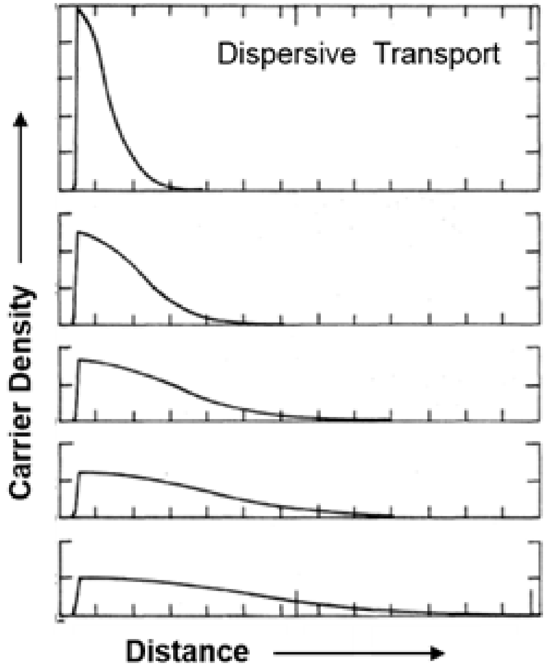
$$\alpha(T) = \frac{kT}{E_c} = \frac{T}{T_c} \qquad \alpha(E) = \frac{qaE}{2kT_c}$$









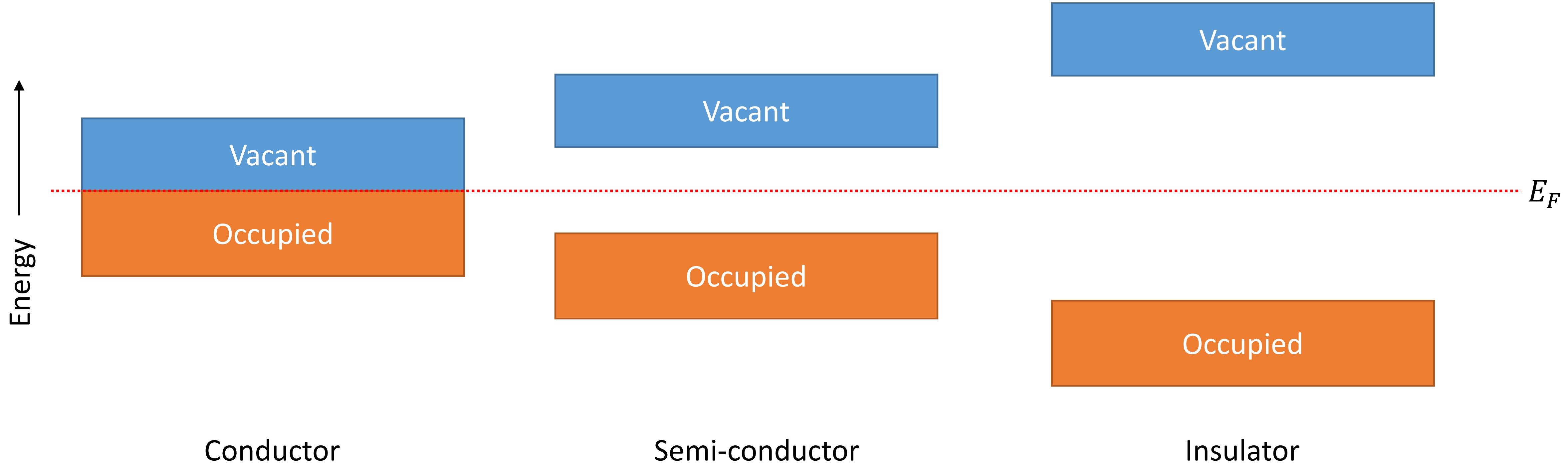


(Zallen, 1983)

Conduction - Crystalline

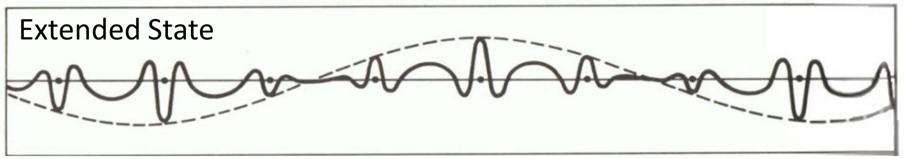
- Perfect periodic structure (long-range order)

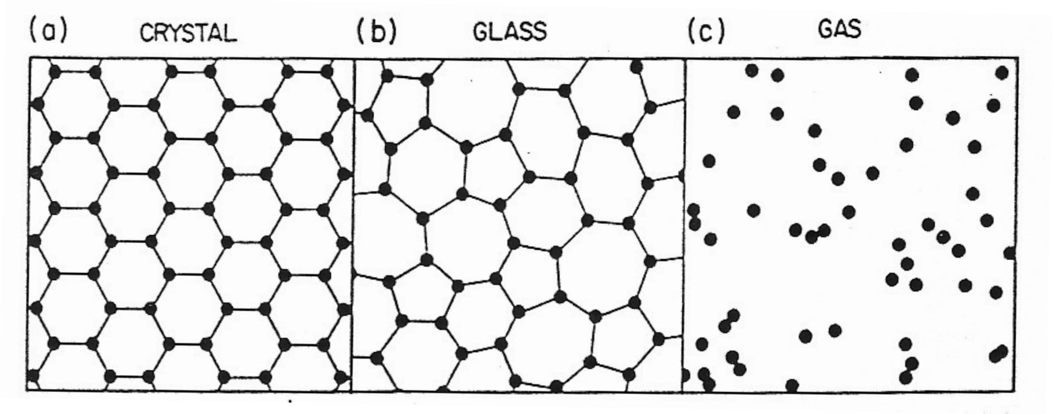
Schrödinger's Equation $\frac{-\hbar^2}{2m}\nabla^2\psi(\boldsymbol{r}) + V(\boldsymbol{r})\psi(\boldsymbol{r}) = E\psi(\boldsymbol{r})$ $\longrightarrow$ Bloch Functions $\psi_k(r) = u_k(\boldsymbol{r})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$

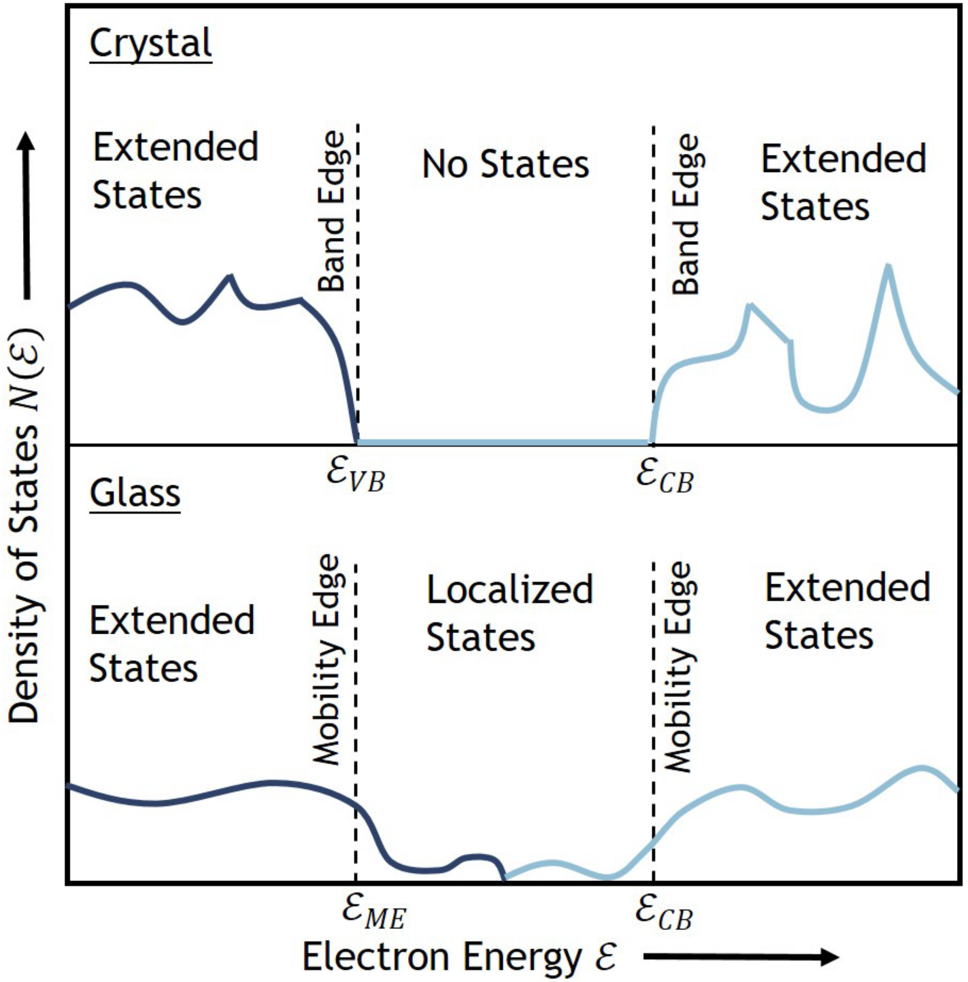


Conduction - Amorphous

$$\psi_k(t) = u_k(\boldsymbol{r})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$$

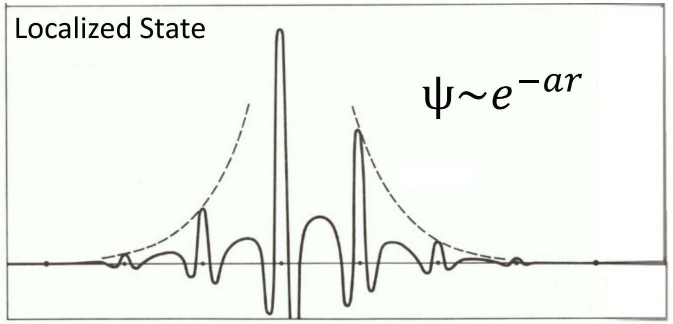






Amorphous solids exhibit

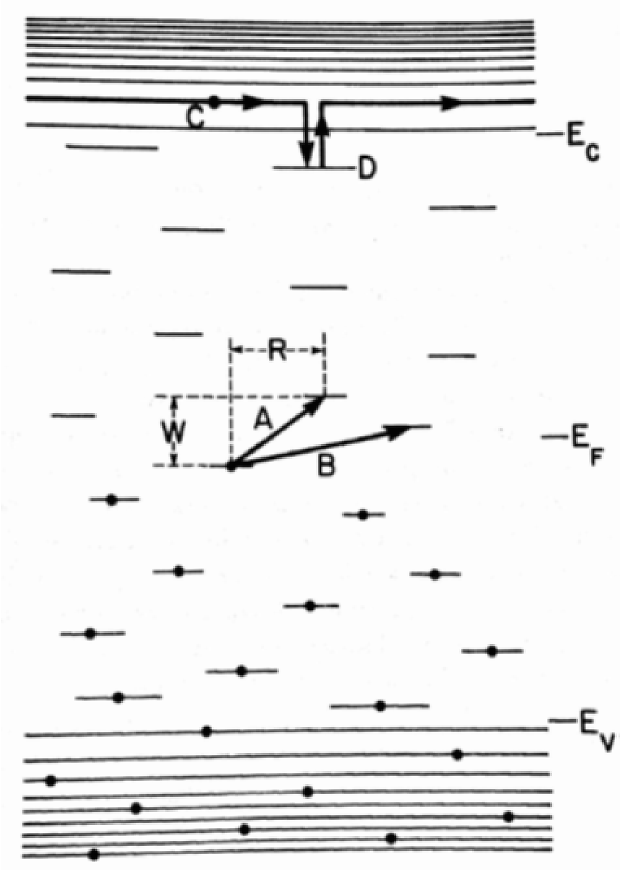
- No long-range order
- Short-range order
- Atoms have equilibrium positions



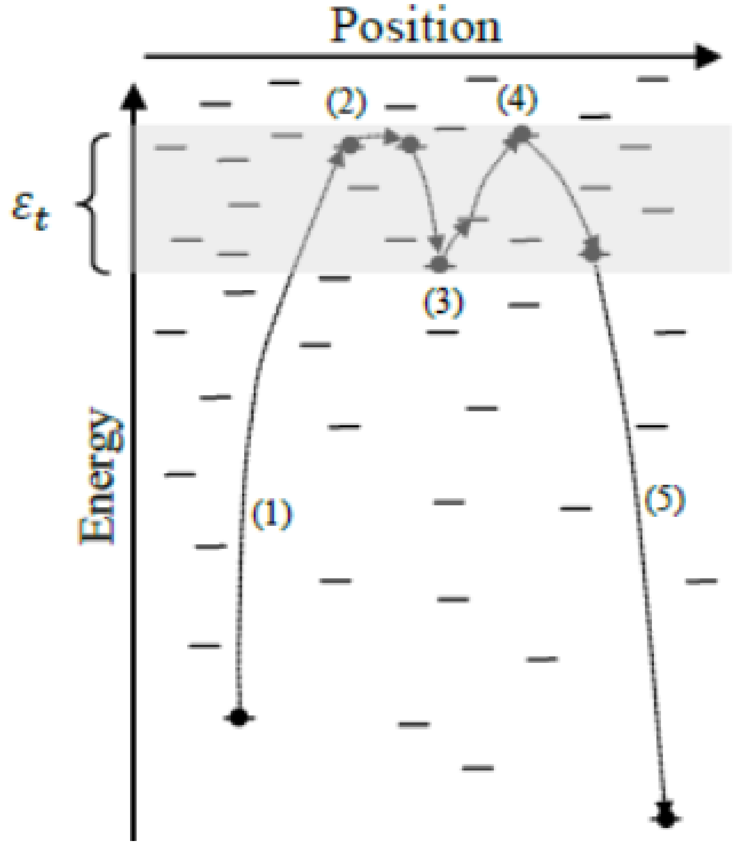
(Zallen, 1983)

Conduction - Amorphous

Conduction mechanisms in amorphous insulators:



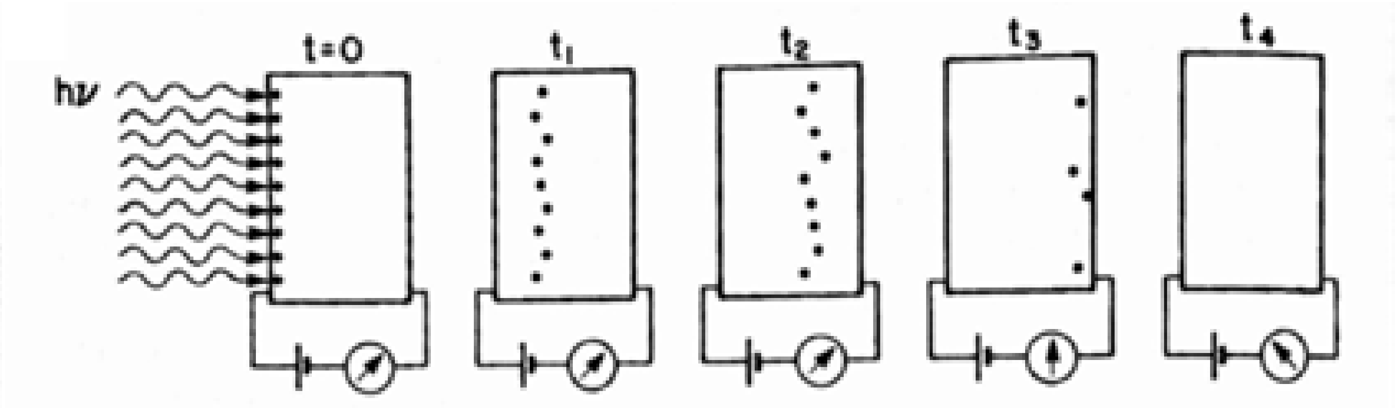
Multiple Trapping and Thermally Assisted Hopping



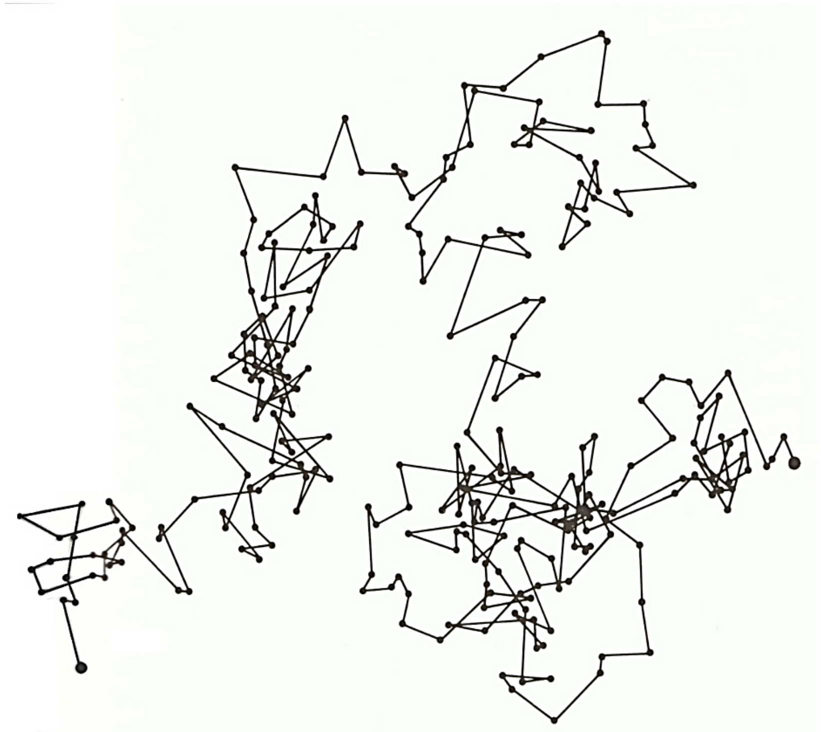
Variable Range Hopping and Radiation Induced Conductivity

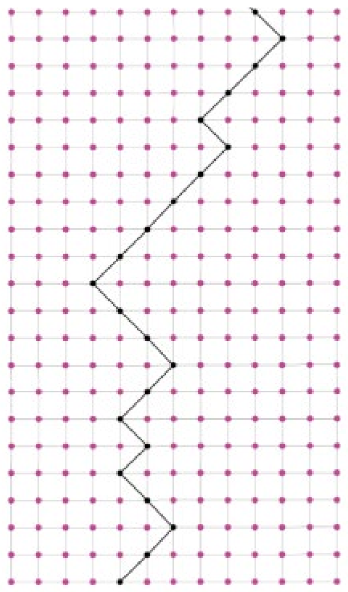
(Zallen, 1983; Sim 2013)

Photoconductivity



- Random Walks
  - Spatially disordered lattice
  - Discrete hopping times
  - Requires ensemble averages of all possible spatial disorder
- Continuous Time Random Walks
  - Characterized by hopping-time distribution function
  - Walker moves on periodic ordered lattice but probability of hopping is given as a function of time
  - Disorder is contained in distribution function

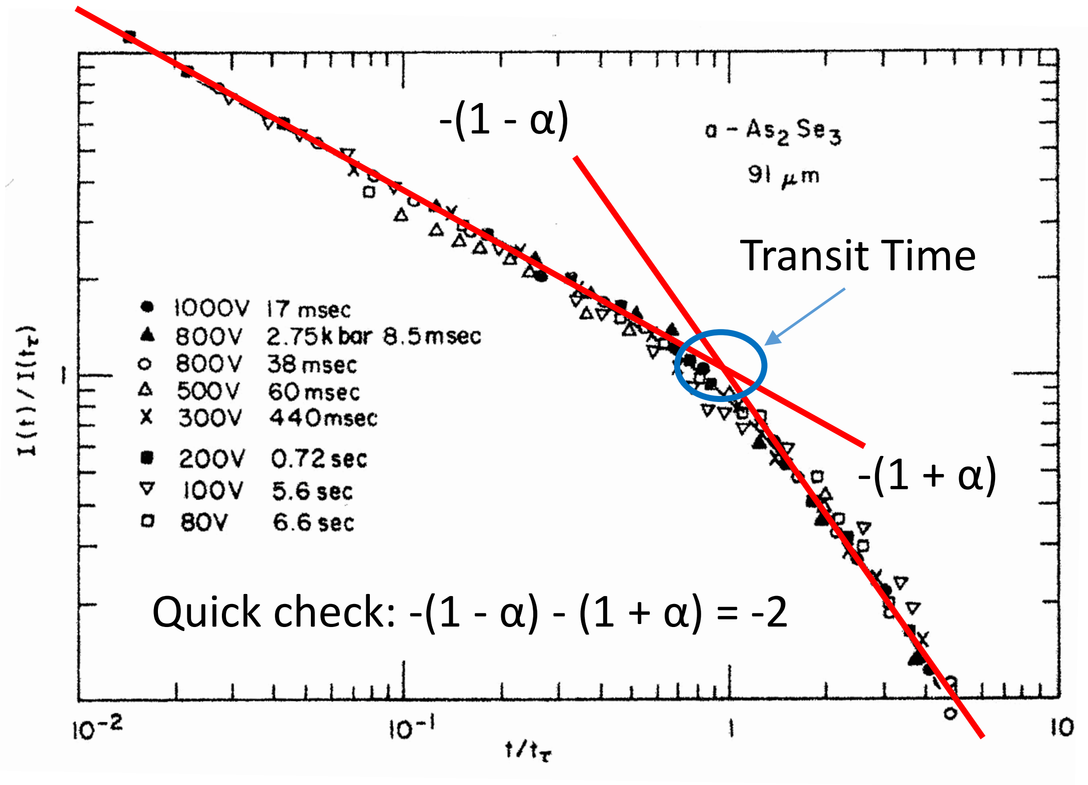




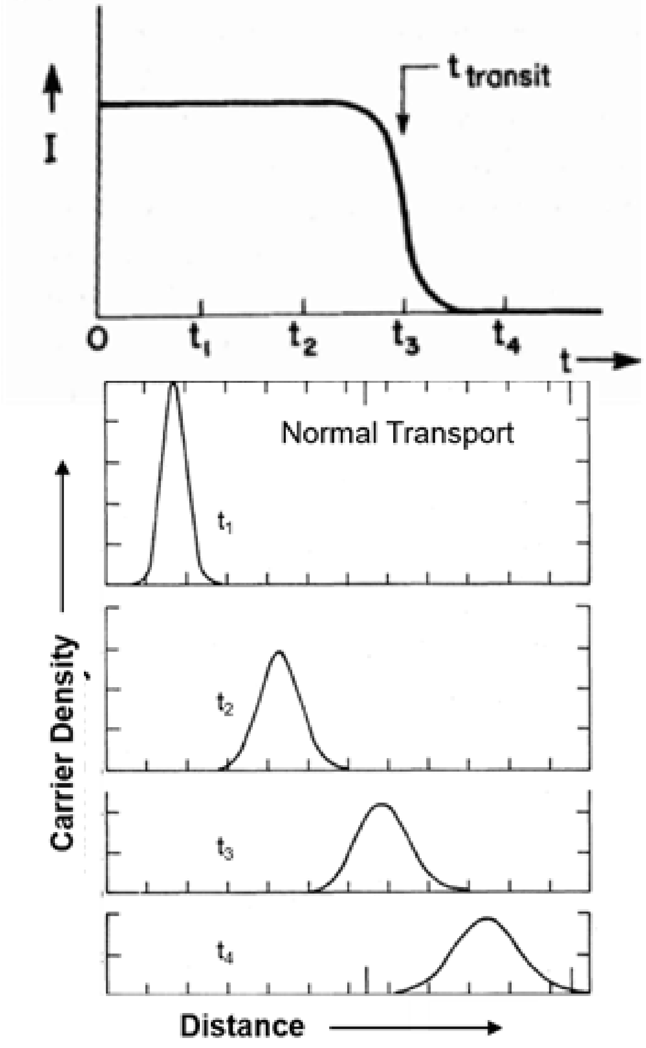
(Gillespie, 2017; Zallen, 1983)

Photoconductivity

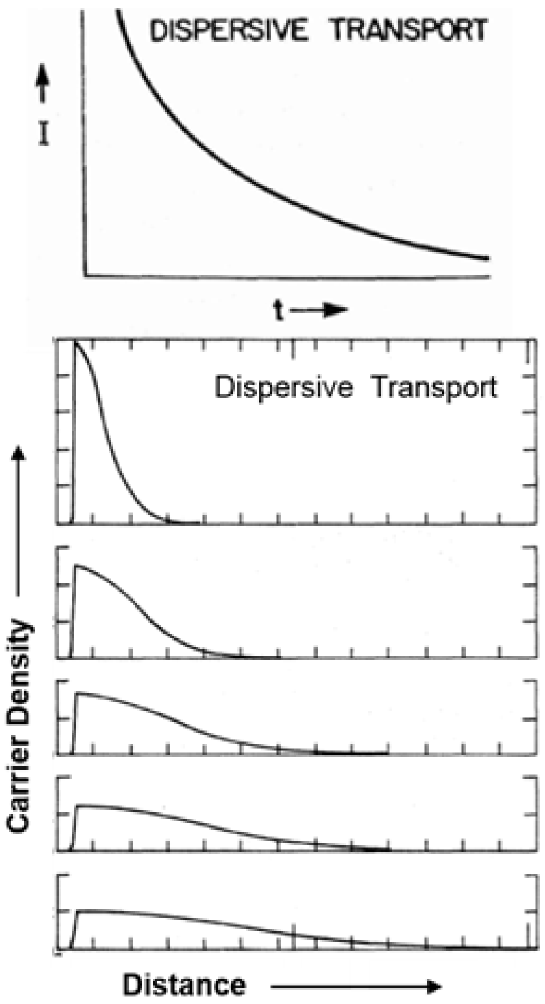
$$I(t) \sim \begin{cases} \dfrac{1}{\Gamma(\alpha) t^{1-\alpha}} & t \ll t_{transit} \\ \dfrac{1}{-\Gamma(-\alpha) t^{1+\alpha}} & t \gg t_{transit} \end{cases}$$



$\psi(t) \sim e^{-\tau}$



$\psi(t) \sim t^{-(1+\alpha)}$

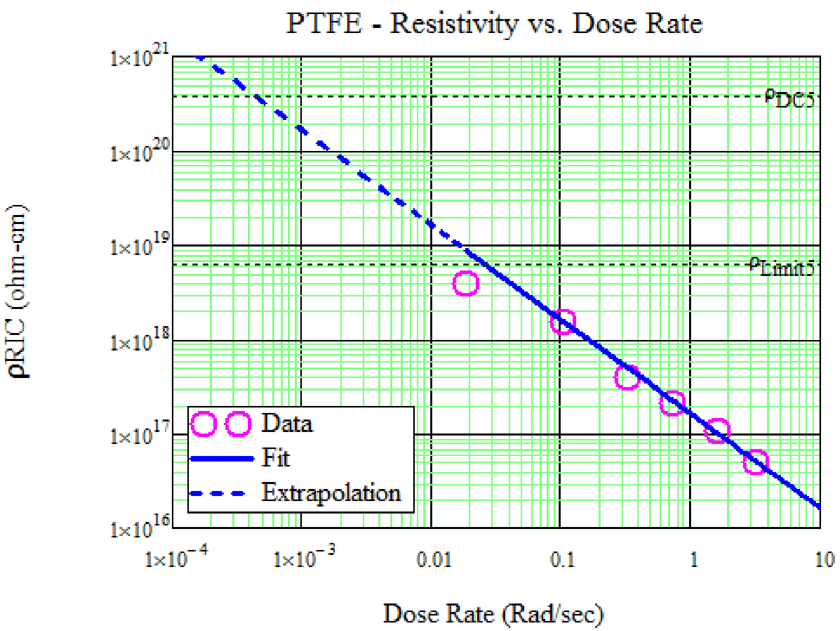


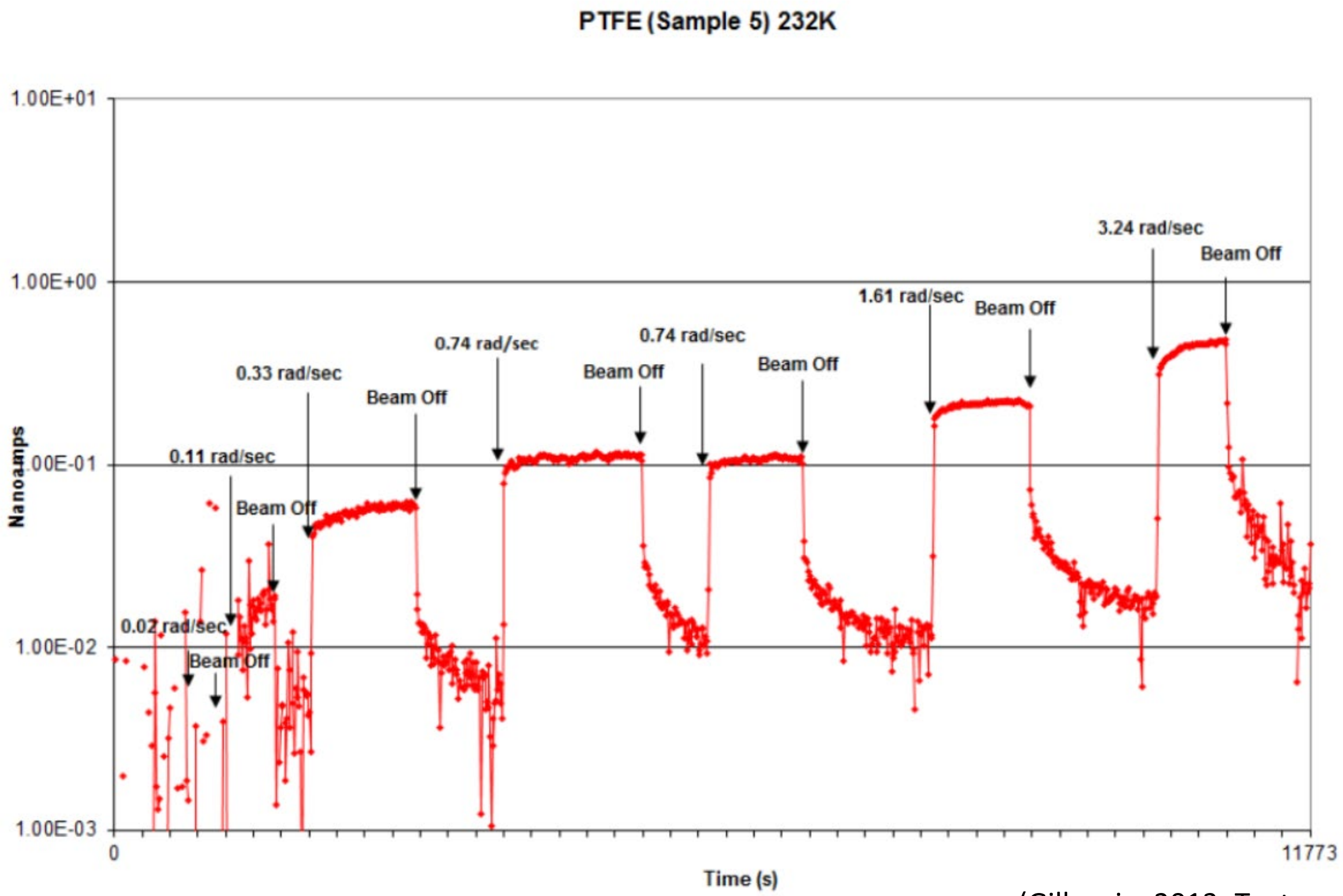
(Zallen, 1983; Scher, 1975)

Radiation Induced Conductivity

- Radiation induced conductivity is also defined by the dispersion parameter

$$\sigma_{RIC} = k_{RIC}(T)\dot{D}^{\Delta} \qquad \Delta = \frac{T_c}{T_c + T} = \frac{1}{1 + \frac{T}{T_c}} = \frac{1}{1+\alpha}$$

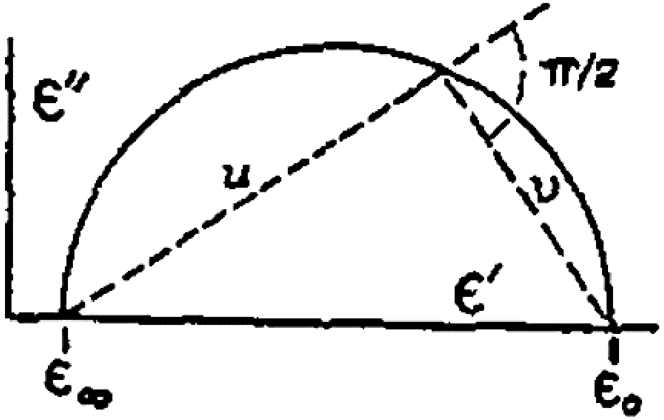




(Gillespie, 2013; Tyutnev, 2006)

Permittivity and DC Conductivity

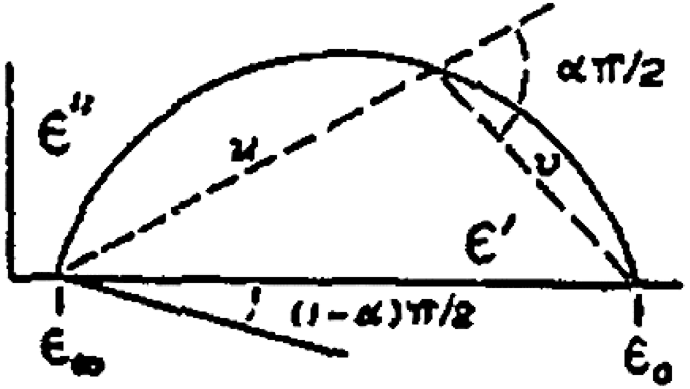
$$\varepsilon^* - \varepsilon_\infty = \frac{(\varepsilon_0 - \varepsilon_\infty)}{(1 + i\omega\tau_0)}$$



- Cole-Cole diagrams depict semi-circles or circular arcs
- Introduces the dispersion parameter through a geometrical argument
- Under DC conditions this gives a current of

$$\varepsilon^* - \varepsilon_\infty = \frac{(\varepsilon_0 - \varepsilon_\infty)}{[1 + (i\omega\tau_0)^\alpha]}$$

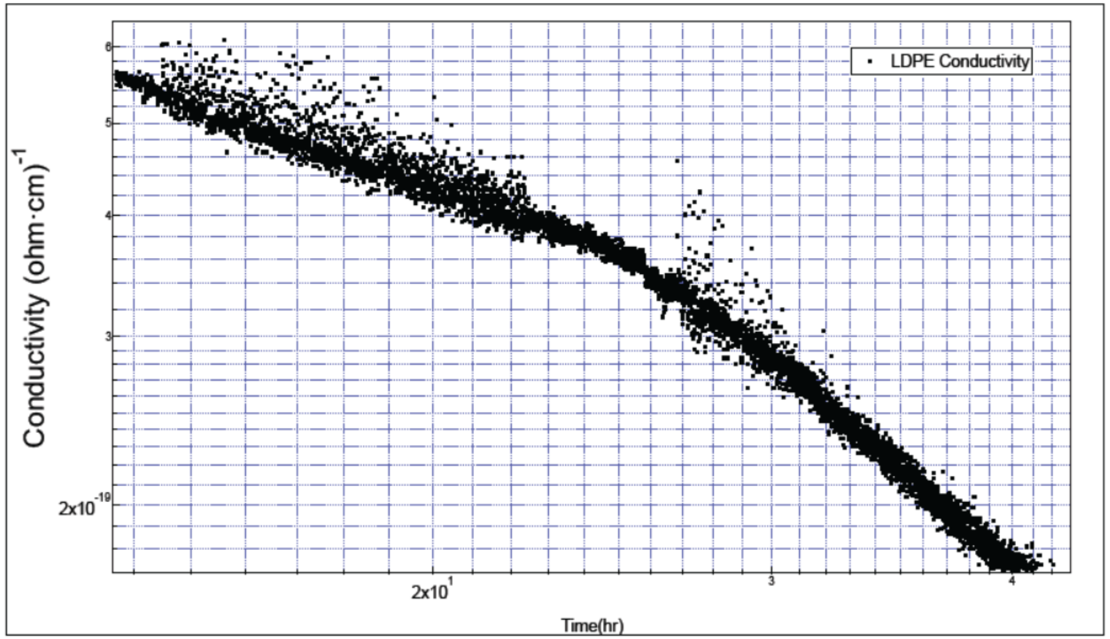
$$I(t) = \begin{cases} \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0}\frac{1}{\Gamma(\alpha)}\left(\frac{t}{\tau_0}\right)^{-(1-\alpha)} & t \ll t_{transit} \\ \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0}\frac{(-1)}{\Gamma(\alpha)}\left(\frac{t}{\tau_0}\right)^{-(1+\alpha)} & t \gg t_{transit} \end{cases}$$

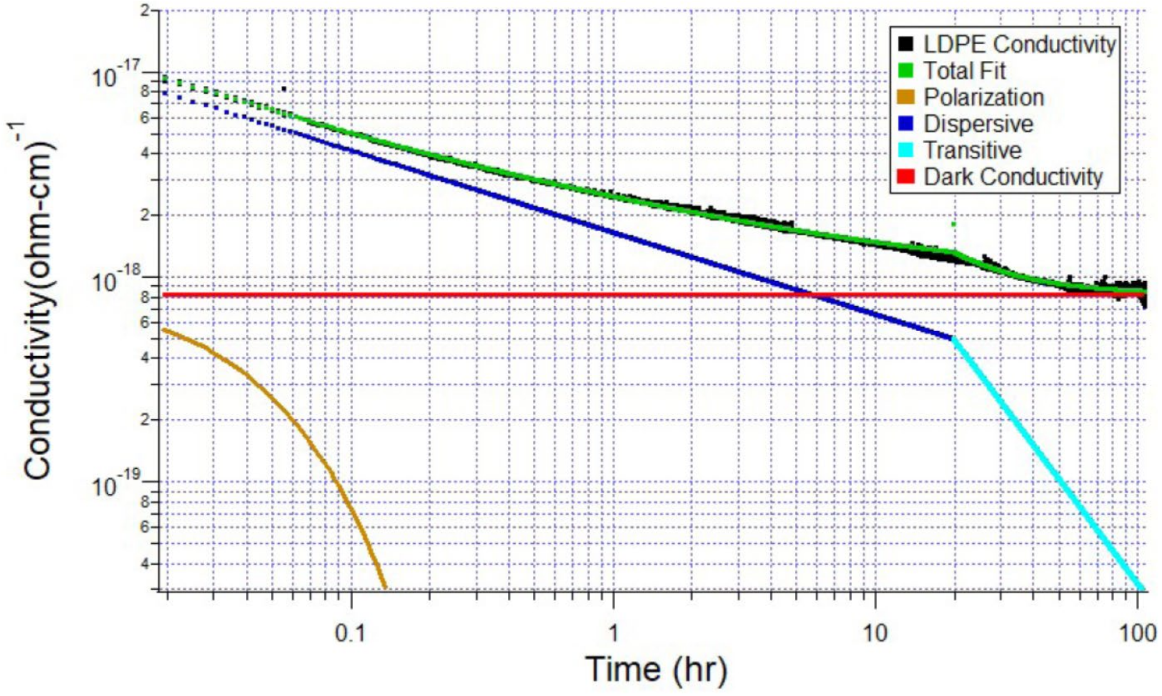


(Cole, 1941)

DC Conductivity

- Transient conductivity in constant voltage conductivity tests exhibit the same behavior as photoconductivity





$$\sigma(t) = \sigma_P^{\frac{-t}{\tau_P}} + \{\sigma_{disp} t^{-(1-\alpha)}\theta(\tau_{transit} - t) + \sigma_{trans} t^{-(1+\alpha)}\theta(t - \tau_{transit})\} + \sigma_{DC}$$

(Wood, 2018)

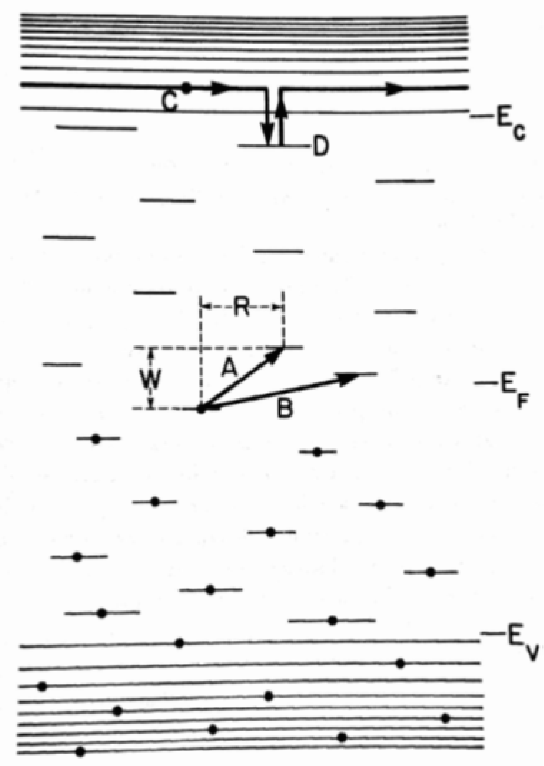
Critical Temperature Transition

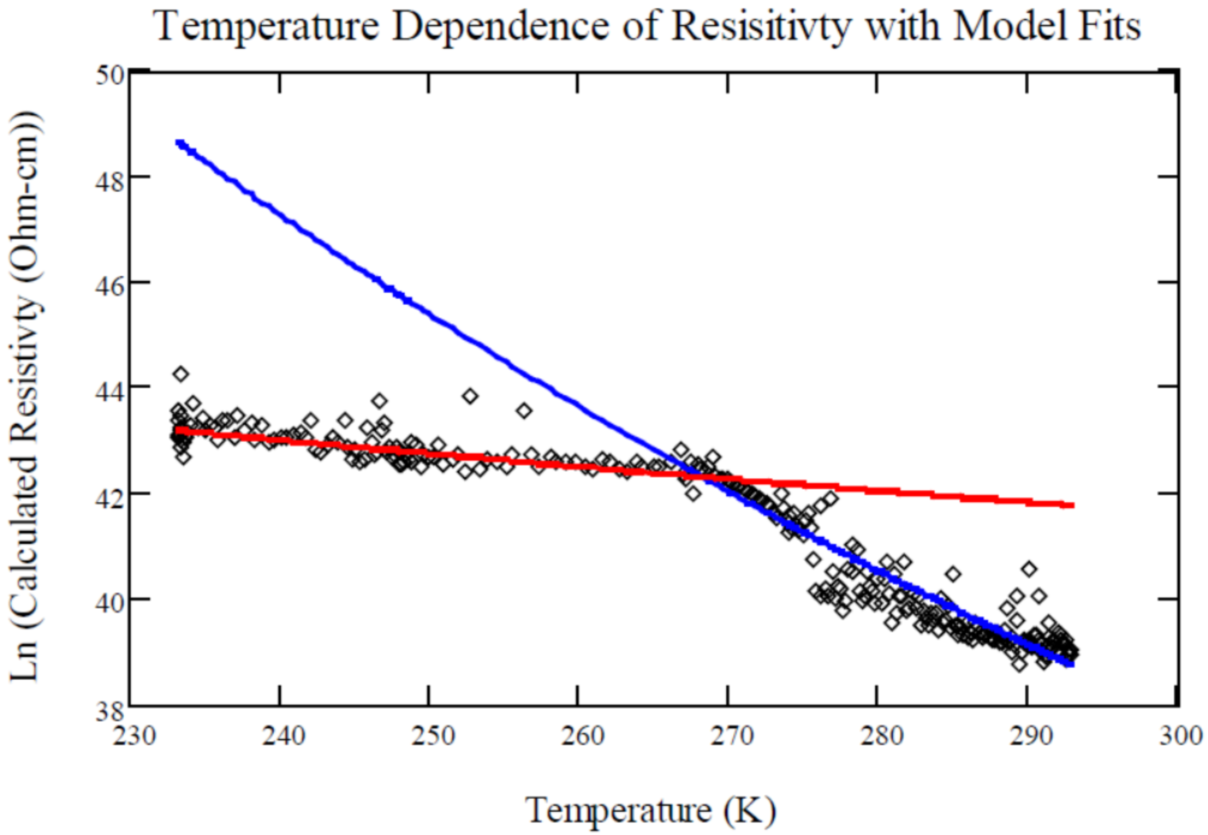
$$\alpha(T) = \frac{T}{T_c}$$

Two regimes:

- Assuming low applied field

1. $T \geq T_c$
   - Multiple trapping dominates
   - $\sigma \sim \exp(T^{-1})$
2. $T < T_c$
   - Variable range hopping dominates
   - $\sigma \sim \exp(T^{-1/4})$



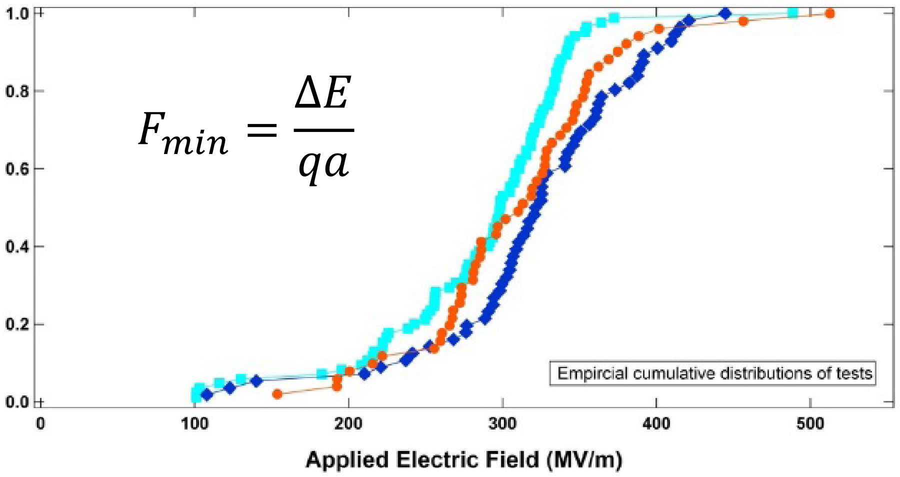


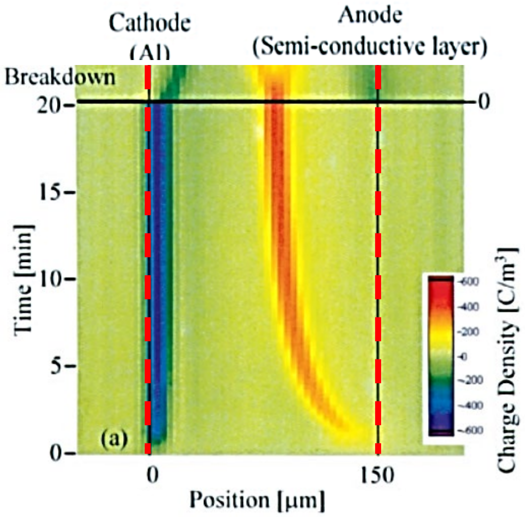
(Dennison, 2008; Brunson, 2007)

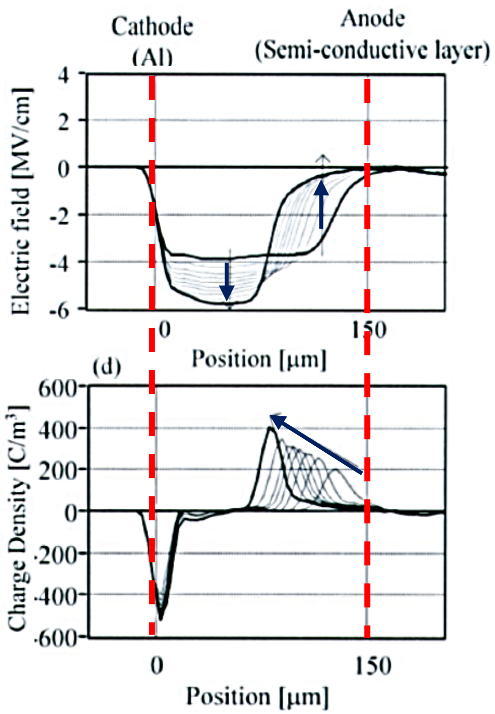
Critical Electric Field Transition

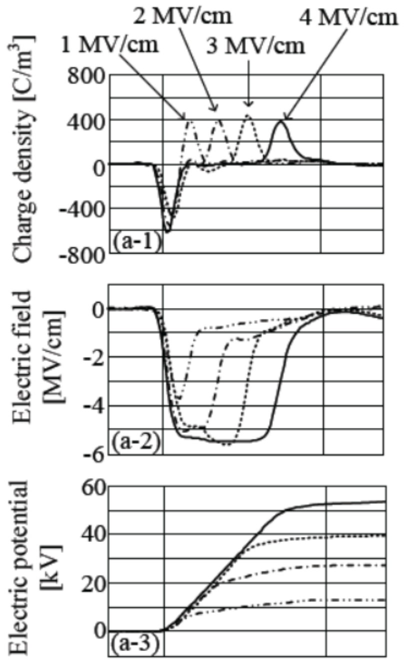
- Transition occurs at α = 1
- Dispersive to normal transport transition occurs at when E = $E_{Transition}$
- $E_{transition}$ denotes onset of electrostatic breakdown

$$\alpha(E) = \frac{qaE}{2kT_c} \quad \underset{\rightarrow}{\alpha = 1} \quad E_{Transition} = \frac{2kT_c}{qa}$$









(Andersen, 2017; Matsui, 2005)

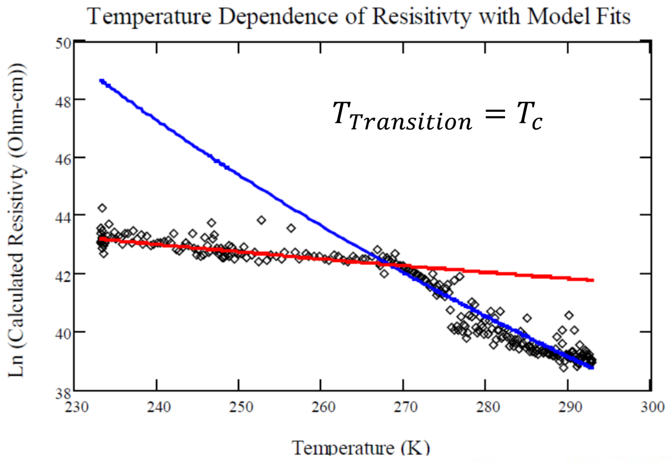
Summary

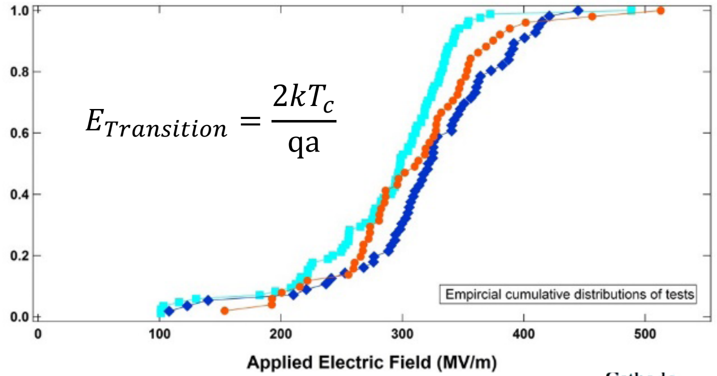
$$\alpha(T) = \frac{T}{T_c}$$

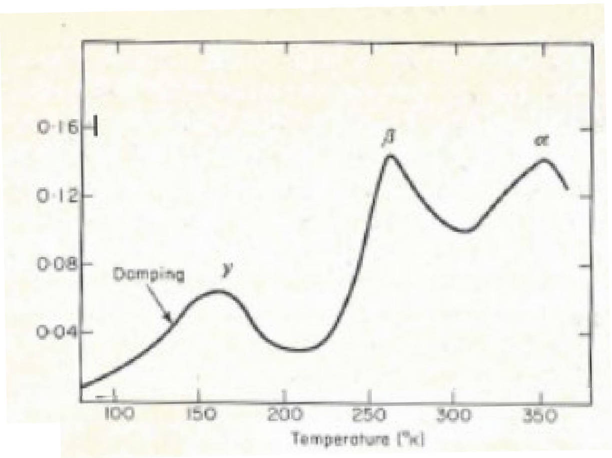
$$\alpha(E) = \frac{qaE}{2kT_c}$$

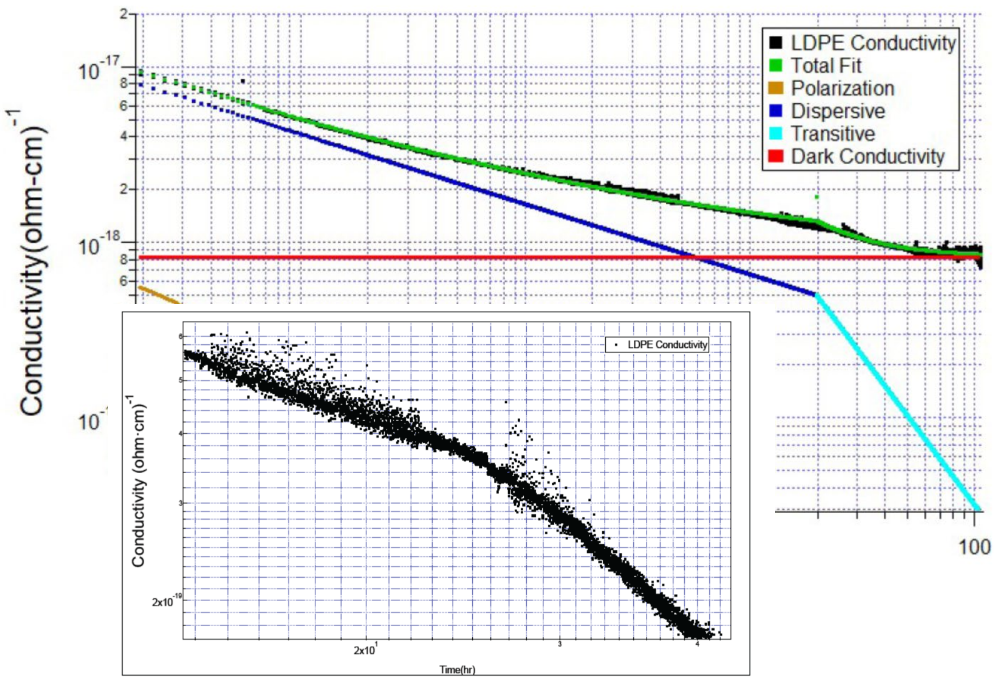
- T_c = 268 K from $\sigma(T)$
- β-phase transition at ~T_c
- ESD onset and dispersive to normal transport transition at E~100 MV/m
- RIC measurements predict T_c~255 K

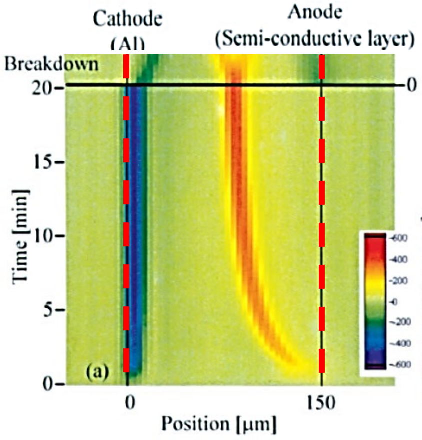
(Wood, 2018; Brunson, 2007; McCrum, 1967; Matsui, 2005; Andersen, 2017)

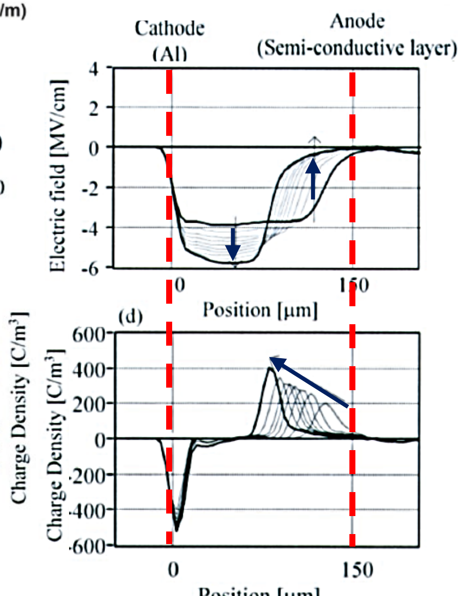


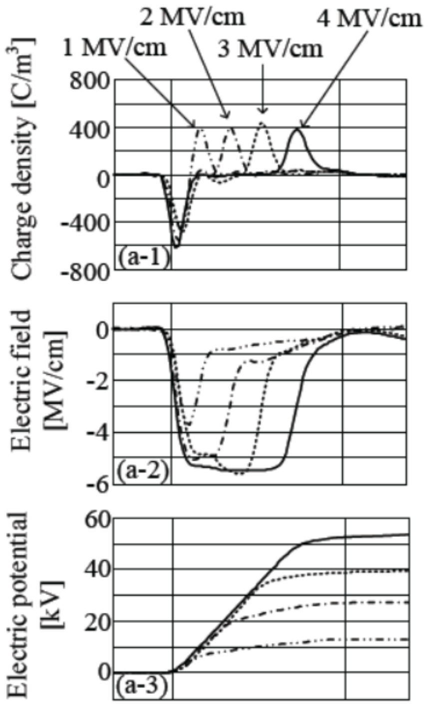












Conclusions

- Macroscopic electrical properties can be described via the dispersion parameter
- Experiments measuring various electrical properties have been connected via the dispersion parameter for low-density polyethylene
- For a deeper understanding of microscopic mechanisms, complimentary measurements are desired

Future Work

- Further corroborate measurements in literature for LDPE and other materials
- CVC and PEA measurements around critical transitions

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