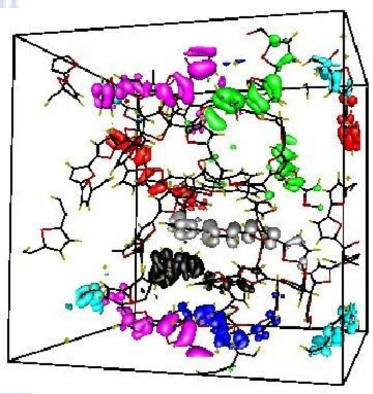
Charge Transport in Organic Electronic Materials



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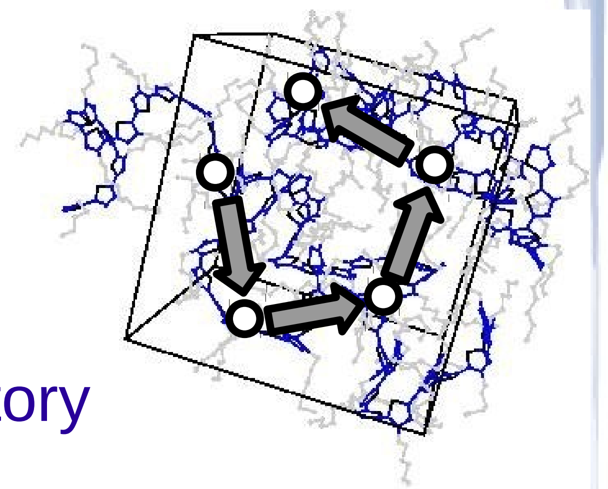

18th Symposium on Condensed Matter Physics,
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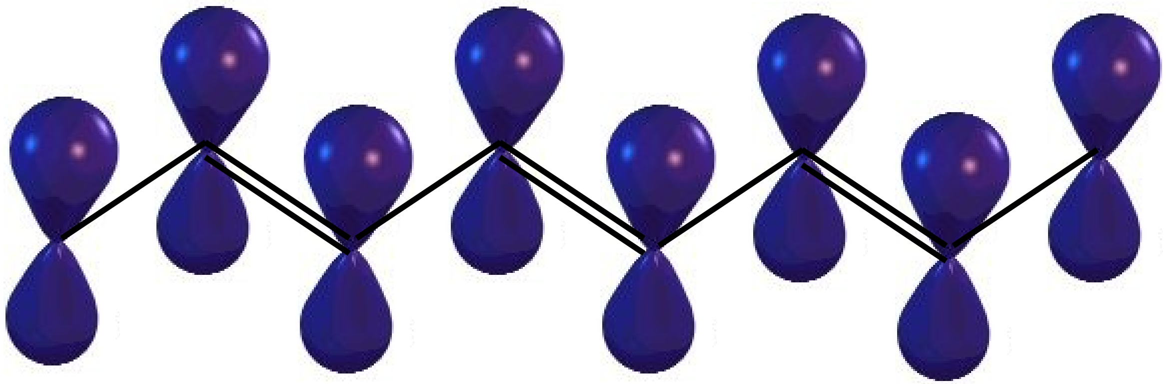
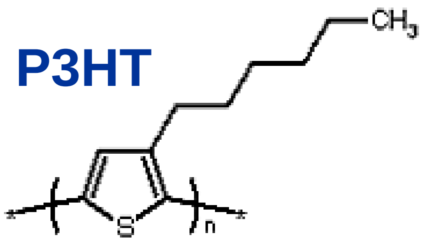
Conjugated polymers

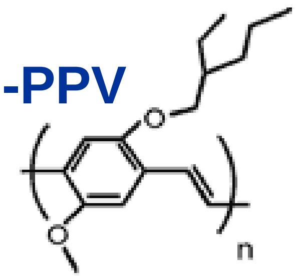
- Single polymer chains:



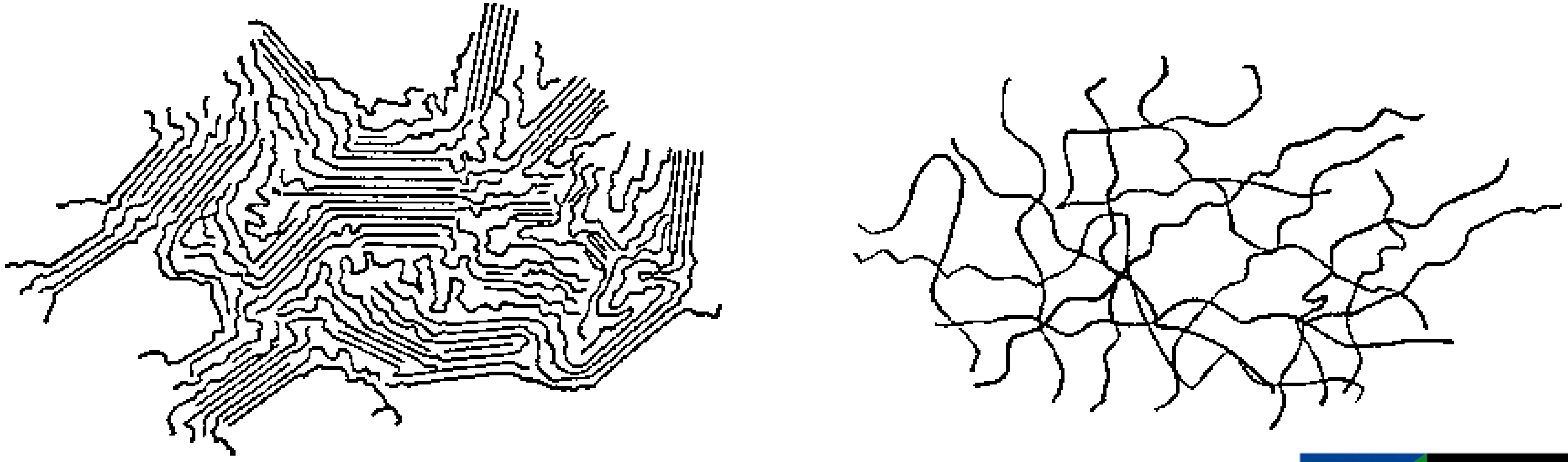
P3HT



MEH-PPV



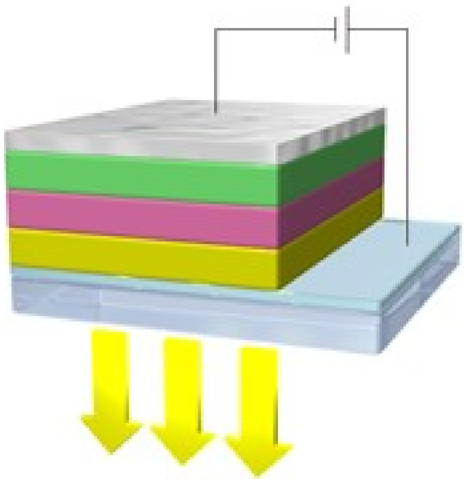
- Polymers forming a real material:



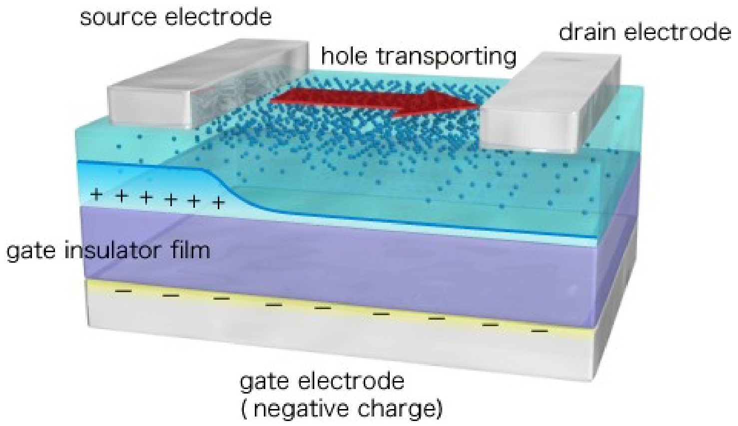
Advantages and applications

- Advantages
  - light and flexible
  - easy and cheap processing
  - tailored synthesis
- Drawbacks
  - low mobility
  - sensitive to UV
  - degradation with time
- Applications

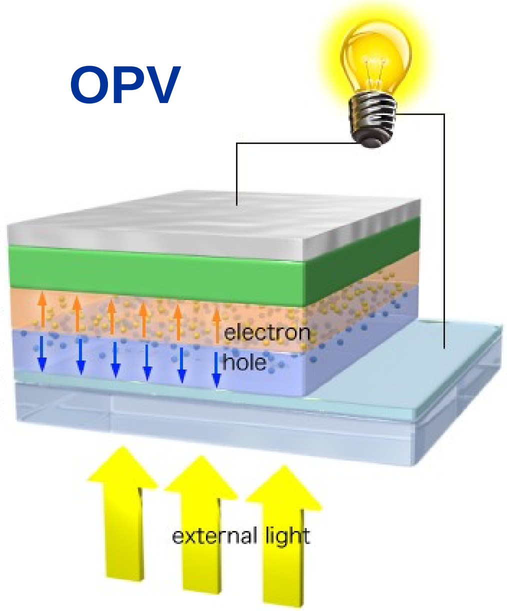
OLED



OFET



OPV



http://www.cstf.kyushu-u.ac.jp/~adachilab/research_b_e.html

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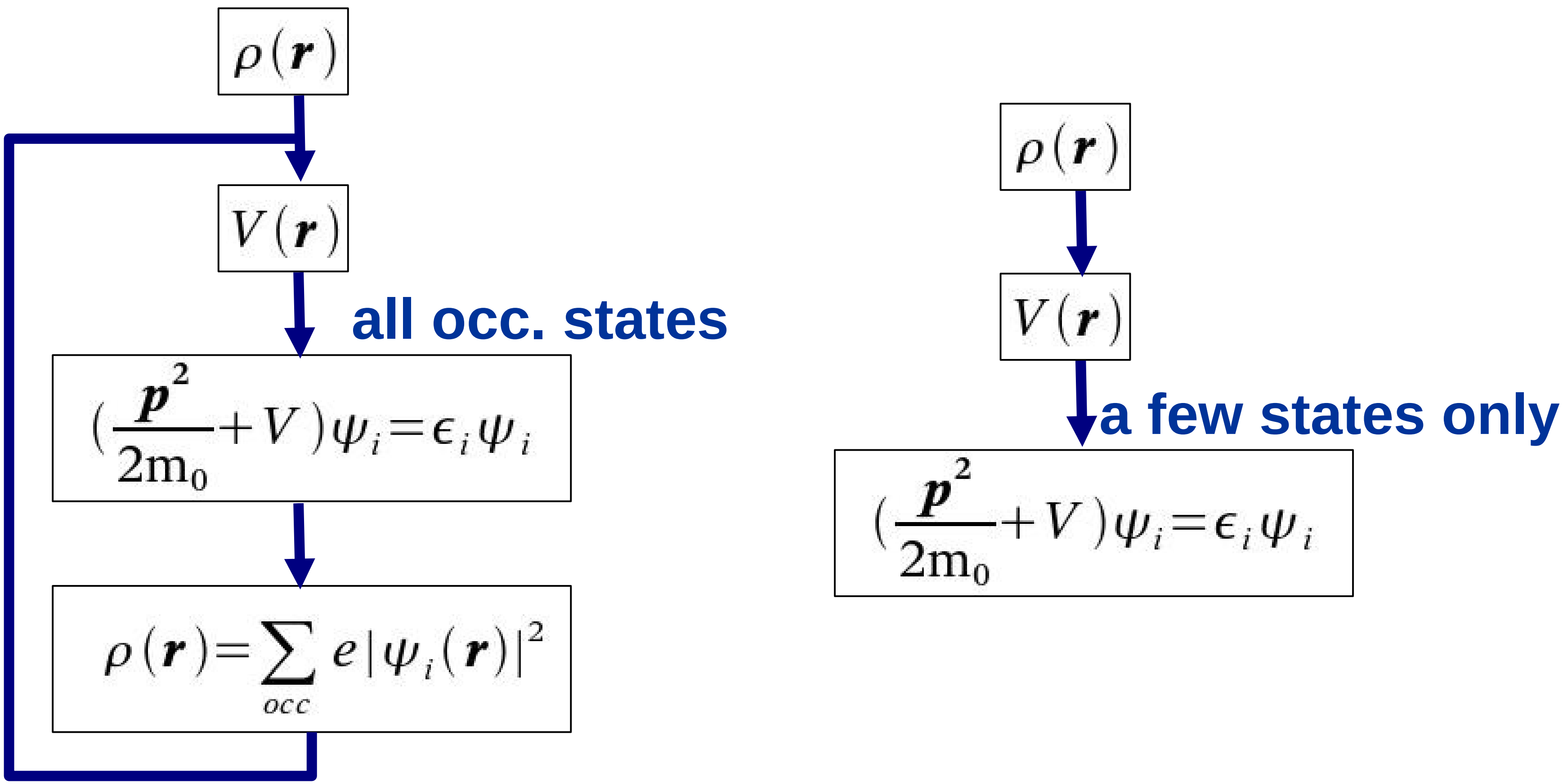
DFT vs. Charge patching



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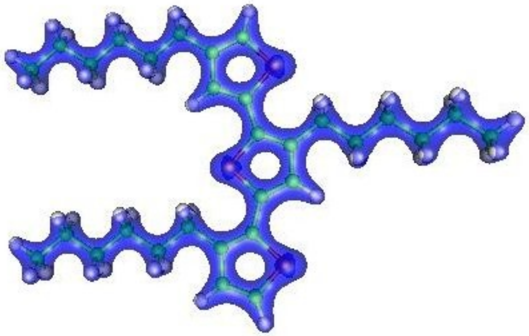
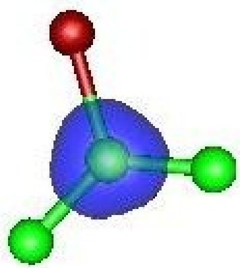
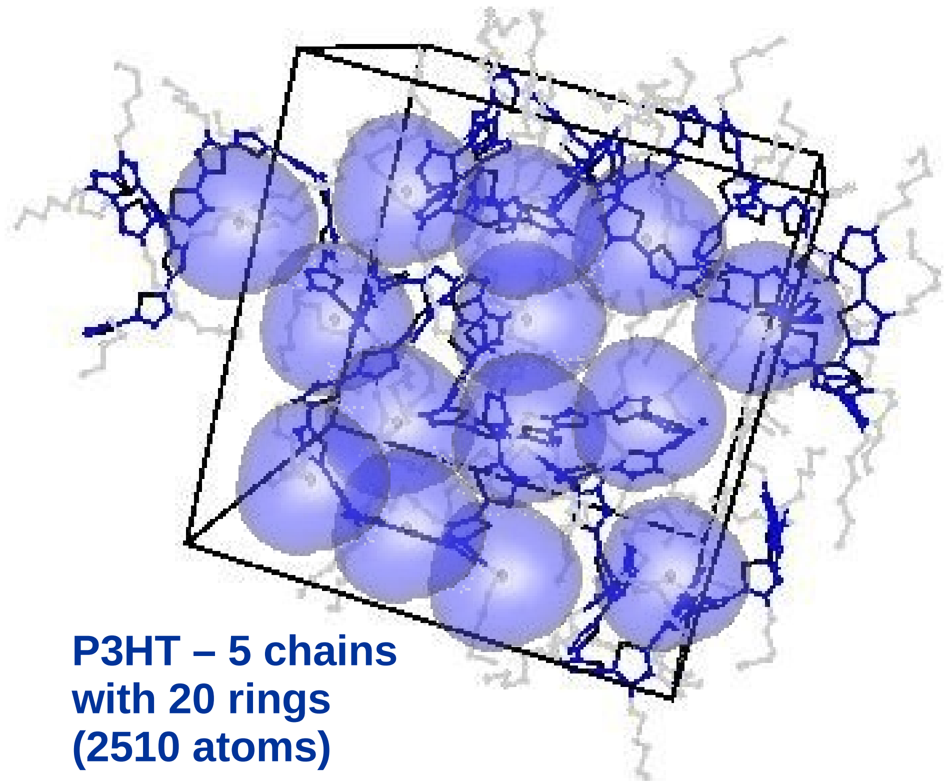
Charge patching method







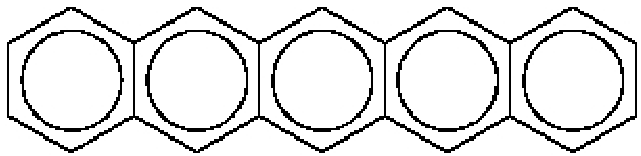
P3HT – 5 chains with 20 rings (2510 atoms)

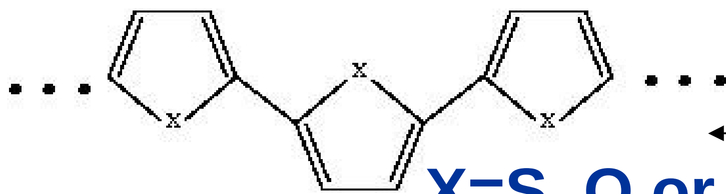
$$m_A(\boldsymbol{r}-\boldsymbol{R}_A)=\frac{w_A(\boldsymbol{r}-\boldsymbol{R}_A)}{\sum_B w_B(\boldsymbol{r}-\boldsymbol{R}_B)}\rho(\boldsymbol{r})$$

$$\rho_{patch}(\boldsymbol{r})=\sum_A m_A(\boldsymbol{r}-\boldsymbol{R}_A)$$

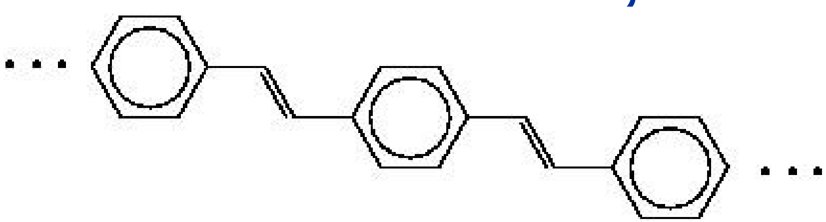
N. Vukmirović and L.-W. Wang, J. Chem. Phys. 128, 121102 (2008)

Test of the CPM for various systems



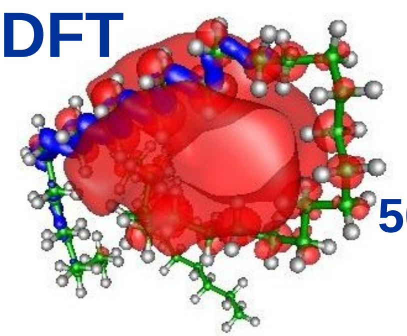


X=S, O or N-H

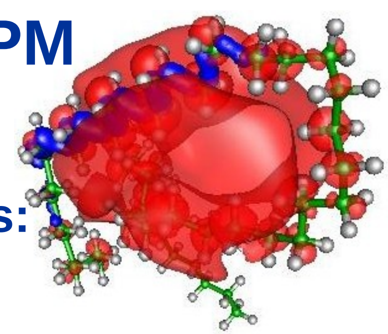


	av. err. (meV)
pentacene	10.0
polythiophene	1.6
6-ring thioph.	15.9
polyfurane	8.5
6-ring furane	27.9
polypyrrole	20.0
6-ring pyrrole	27.5
PPV	19.8

DFT



CPM



50% isosurfaces:
HOMO (blue)
LUMO (read)

Comparison in the case of 50 unit chain – av. error 7.6 meV

N. Vukmirović and L.-W. Wang, J. Chem. Phys. 128, 121102 (2008)

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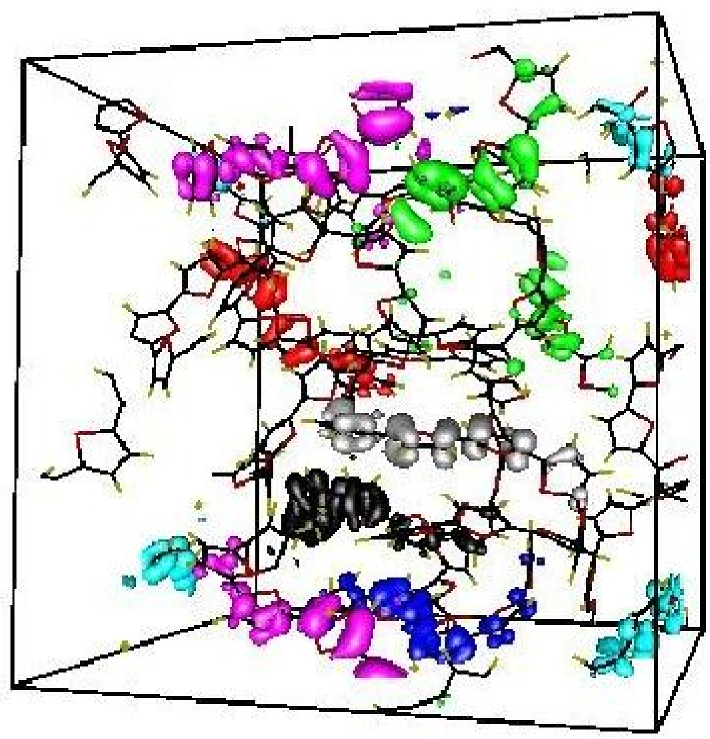
Wave functions

- Atomic structure – classical MD, simulated annealing
- Charge patching method for electronic structure
- Hole states in P3HT:
  - typically localised to 3-6 rings.

P3HT – 5 chains with 20 rings (2510 atoms)

blue: 18.910eV
green: 18.888eV
cyan: 18.755eV
red: 18.690eV
pink: 18.682eV
black: 18.675eV
white: 18.654eV



N. Vukmirović and L.-W. Wang, J. Phys. Chem. B 113, 409 (2009)

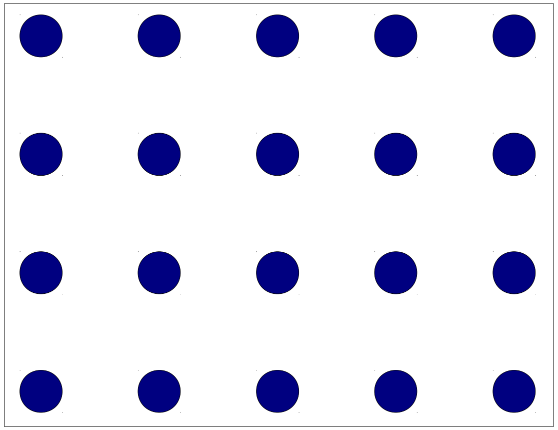
Previous approaches for transport

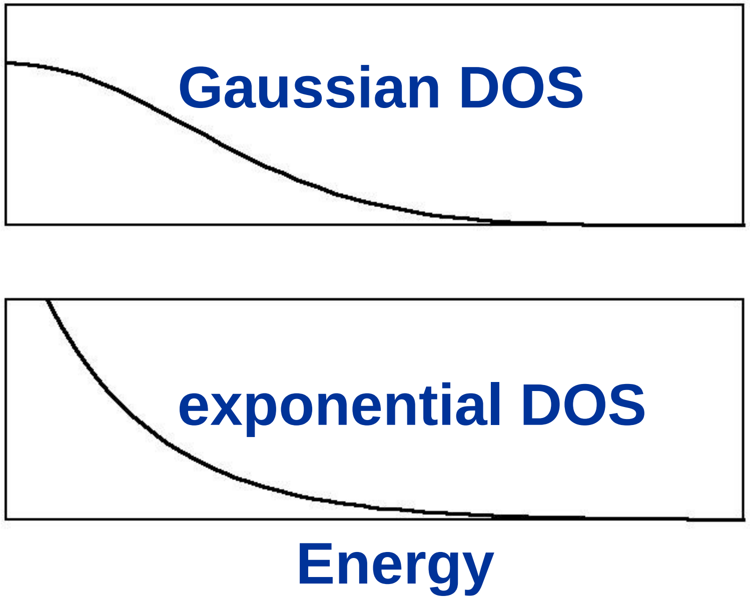
- Gaussian or exponential DOS
- Cubic lattice of sites
- Miller-Abrahams transition rates

$$W_{ij} \sim \exp(-\alpha R_{ij}) \qquad E_i > E_j$$

$$W_{ij} \sim \exp(-\alpha R_{ij}) \exp(-\Delta E_{ji}/kT) \qquad E_i \leq E_j$$

- Several fitting parameters

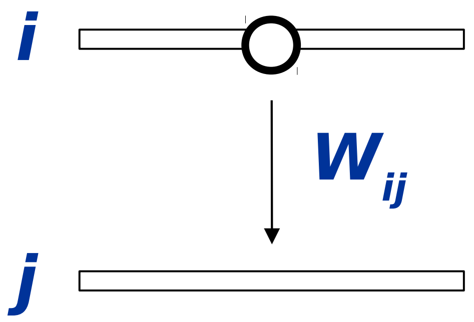




This approach

-Direct calculation of WFs and energies

-Transition rates calculated by considering interaction with all phonon modes



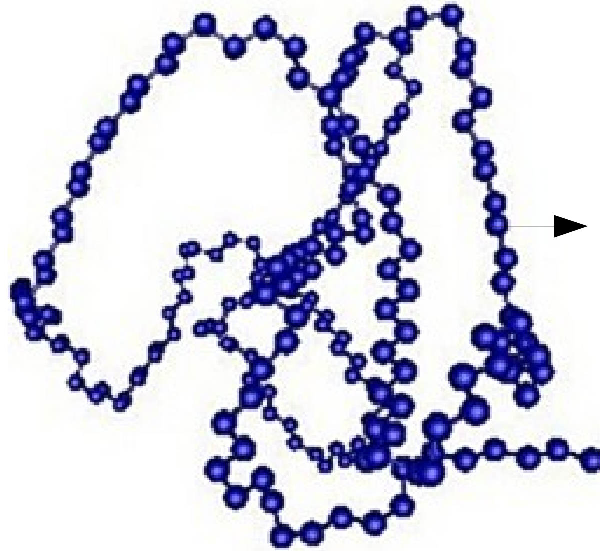
$$W_{ij} = \pi \sum_{\mu} \frac{|M_{ij,\mu}|^2}{\omega_\mu} [N(\hbar\omega_\mu)+1] \delta(E_i - E_j - \hbar\omega_\mu)$$

-Phonon modes from classical force field

-Electron-phonon coupling constants from charge patching

$$M_{ij,\mu} = \langle i | \frac{\partial H}{\partial v_\mu} | j \rangle$$

-No fitting parameters



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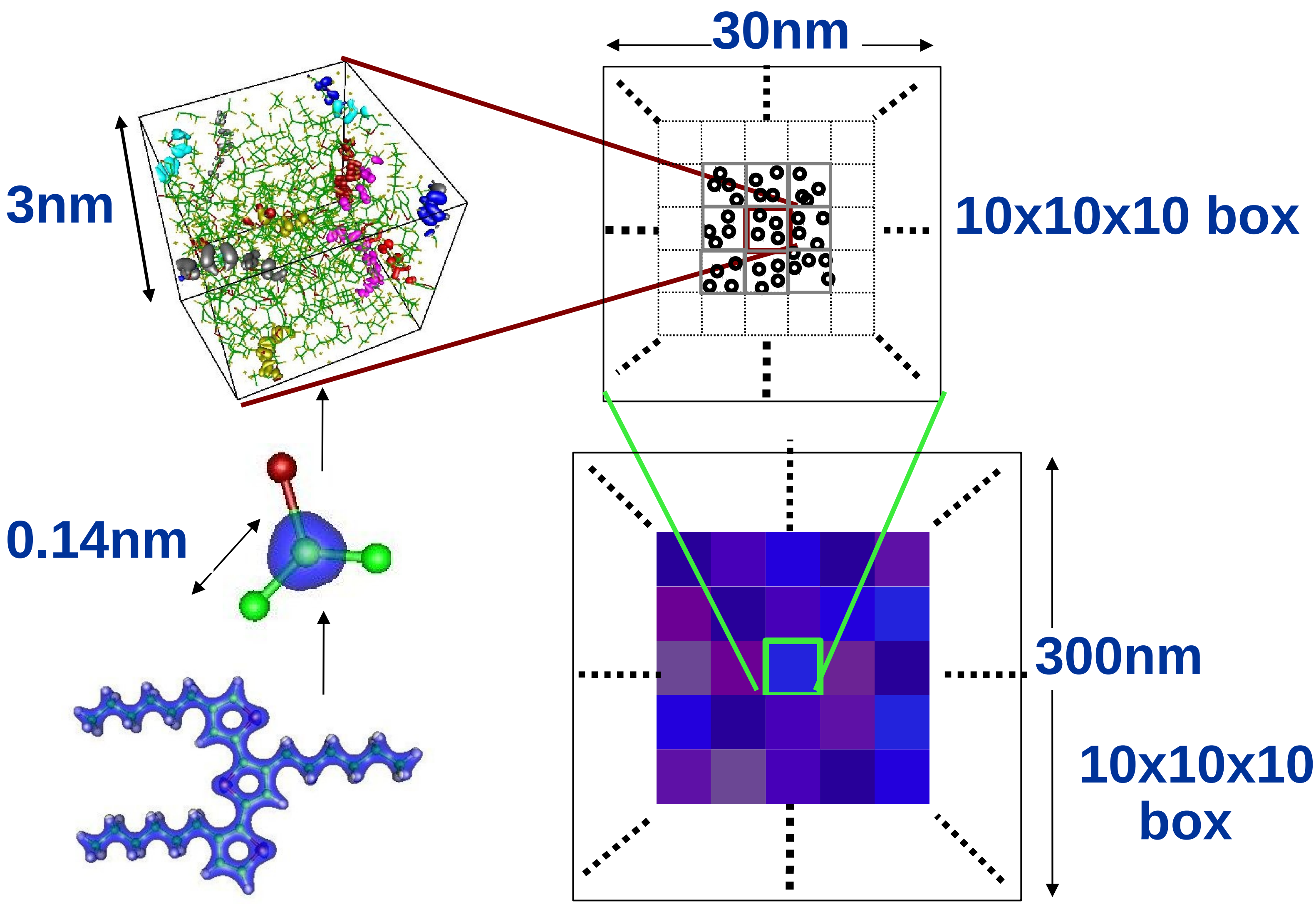
Multiscale method for carrier transport



N. Vukmirović and L.-W. Wang, Nano Lett. 9, 3996 (2009)

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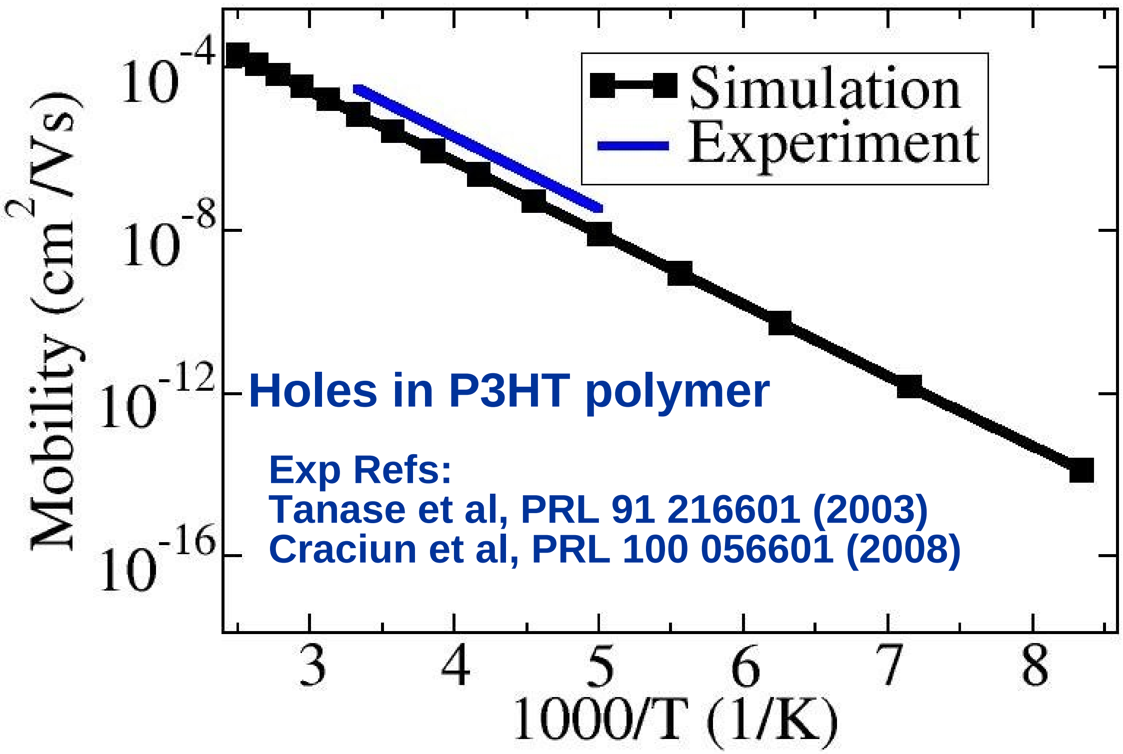
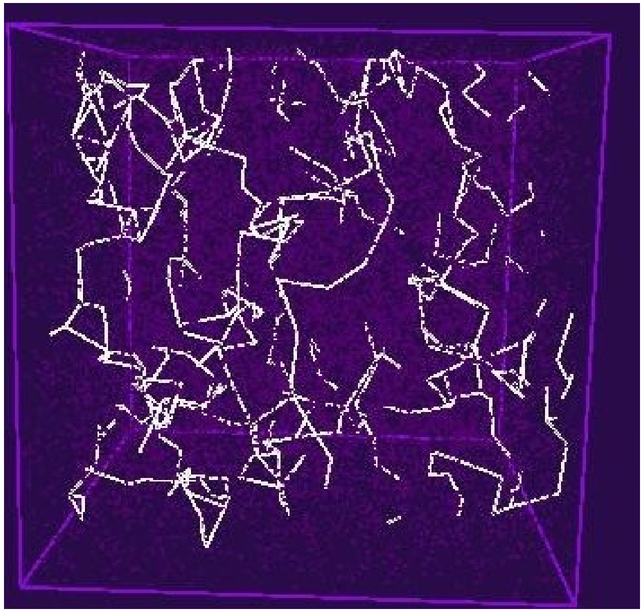
Mobility



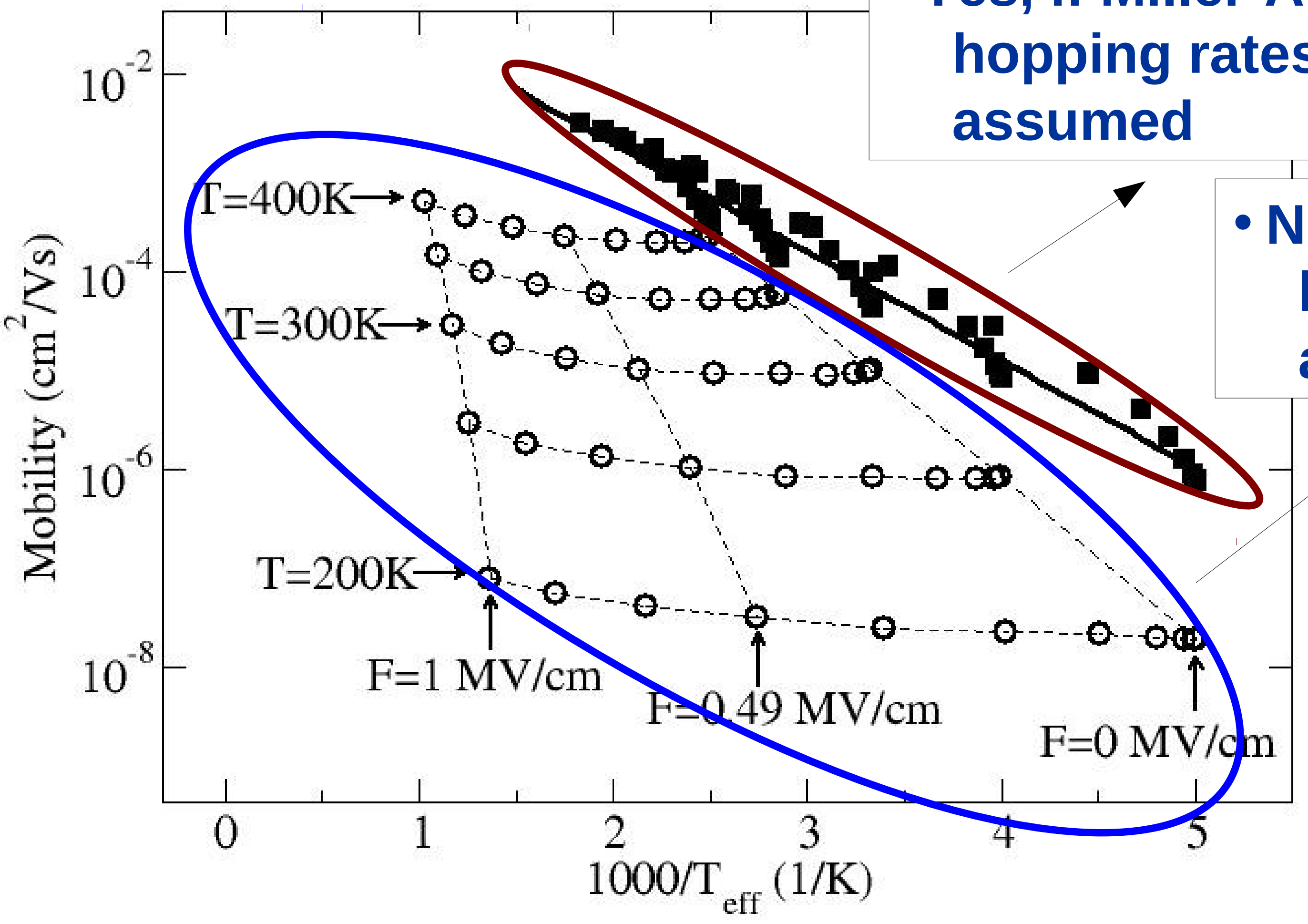


http://www.colourlovers.com/uploads/2008/02/sydney_lightning_bolts.jpg

- Microscopic insight into the current paths in the material.

N. Vukmirović and L.-W. Wang, Nano Lett. 9, 3996 (2009)

Is the concept of elec. temperature useful?



- Yes, if Miller-Abrahams hopping rates are assumed
- No, if realistic hopping rates are assumed
- Qualitatively different conclusions!

N. Vukmirović and L.-W. Wang, Phys. Rev. B 81, 035210 (2010)

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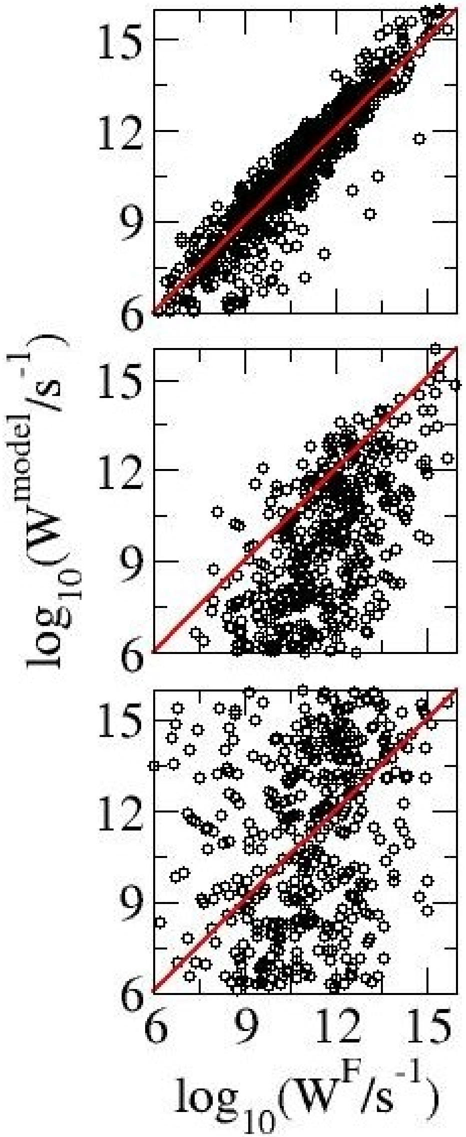
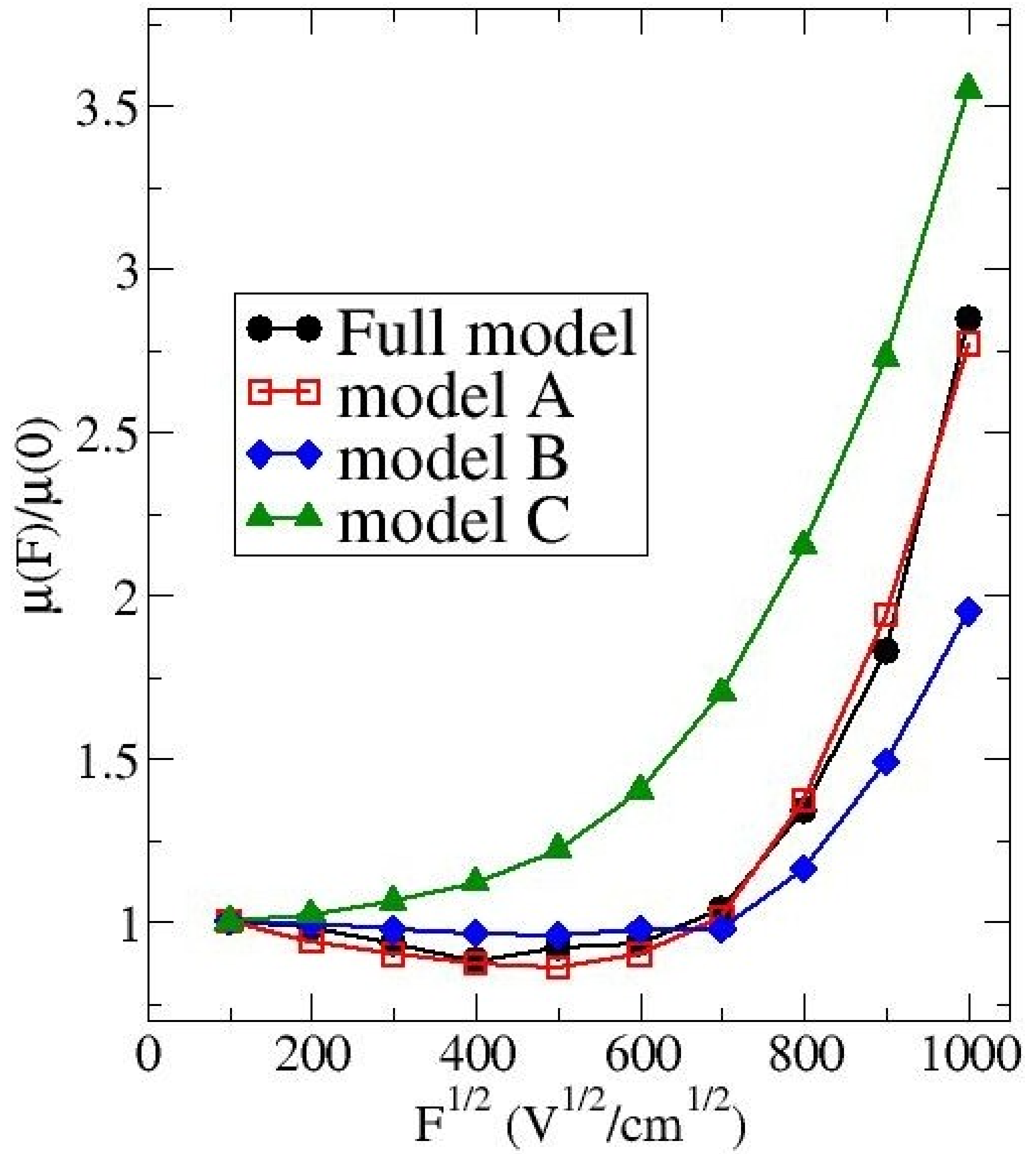
Test of different models



model A

model B

model C



N. Vukmirović and L.-W. Wang, Appl. Phys. Lett. 97, 043305 (2010)

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So, what determines the transport?

$$W_{ij} = \pi \sum_{\mu} \frac{|M_{ij,\mu}|^2}{\omega_\mu} [N(\hbar\omega_\mu)+1] \delta(E_i - E_j - \hbar\omega_\mu)$$

$$W_{ij} = \beta^2 S_{ij}^2 [N(E_{ij})+1] D_{ph}(E_{ij}) / E_{ij}$$

- $\beta^2 S_{ij}^2$: wavefunction moduli overlap
- $N(E_{ij})$: phonon occupation number
- $D_{ph}(E_{ij})$: phonon DOS
- E_{ij}: transition energy

• electronic DOS?	• Yes.
• phonon DOS?	• Yes.
• details of WF overlaps?	• Yes.
• details of phonon modes?	• No.

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Take home messages

- Simulations that link the atomic structure of the disordered polymer material to its electrical properties are now possible.
- Electronic transport
  - Electronic temperature in a finite electric field is not useful for the description of carrier transport.
  - Phonon DOS and details of WF overlaps are important (in addition to Electronic DOS).

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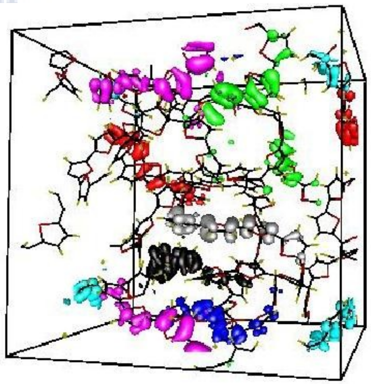
Charge Transport in Organic Electronic Materials



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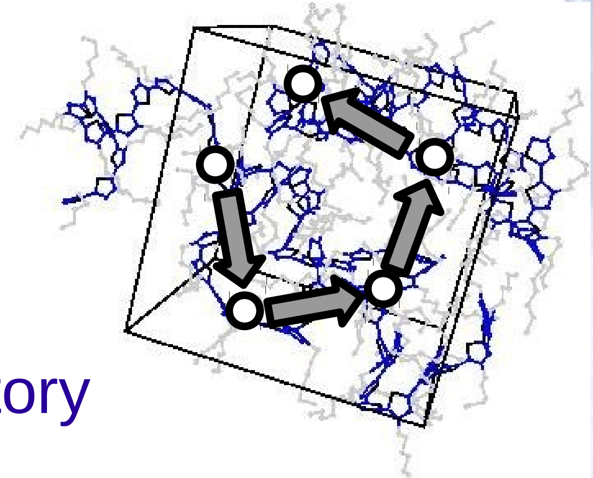

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