PURDUE
UNIVERSITY

Current Flow in Silicon-Molecule Heterostructure: First-Principles Solution

Titash Rakshit
G-C. Liang, A.W. Ghosh, S. Datta

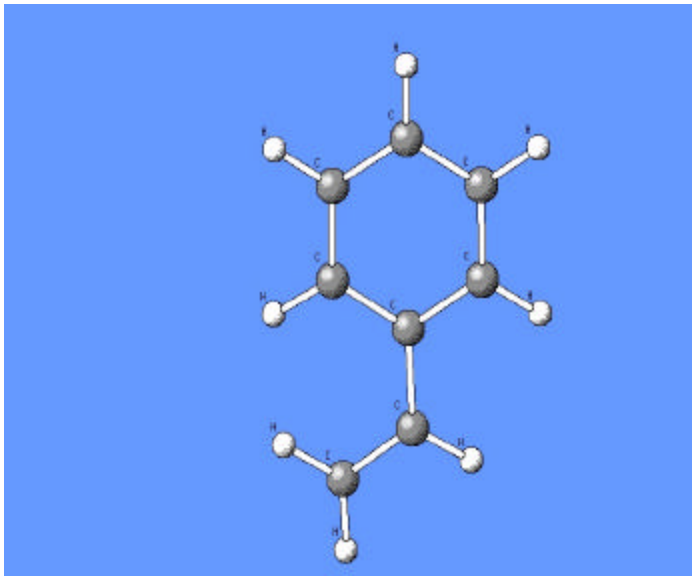
School of Electrical and Computer Engg,
Purdue University, IN

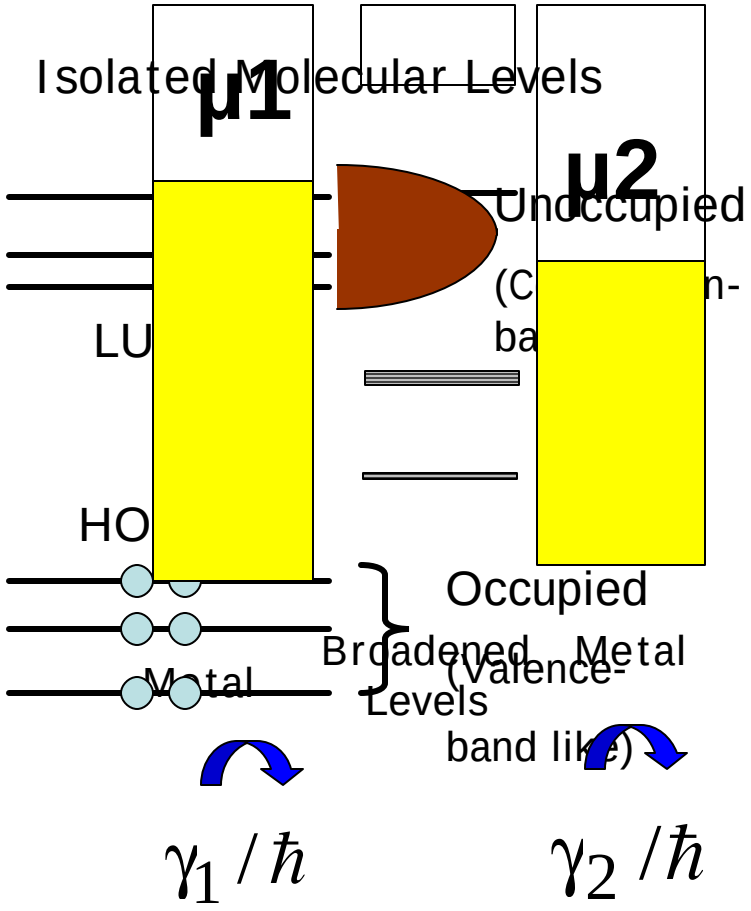
Outline

- **Qualitative picture: How current flows in a molecule**
- **Example: Molecules on Silicon Substrate: NDR predicted from Band-diagram**
- **Quantitative model: Coupled DFT-NEGF Fully Self-Consistent Solution**
- **Experiments: Agrees with broad features of the theoretical prediction**

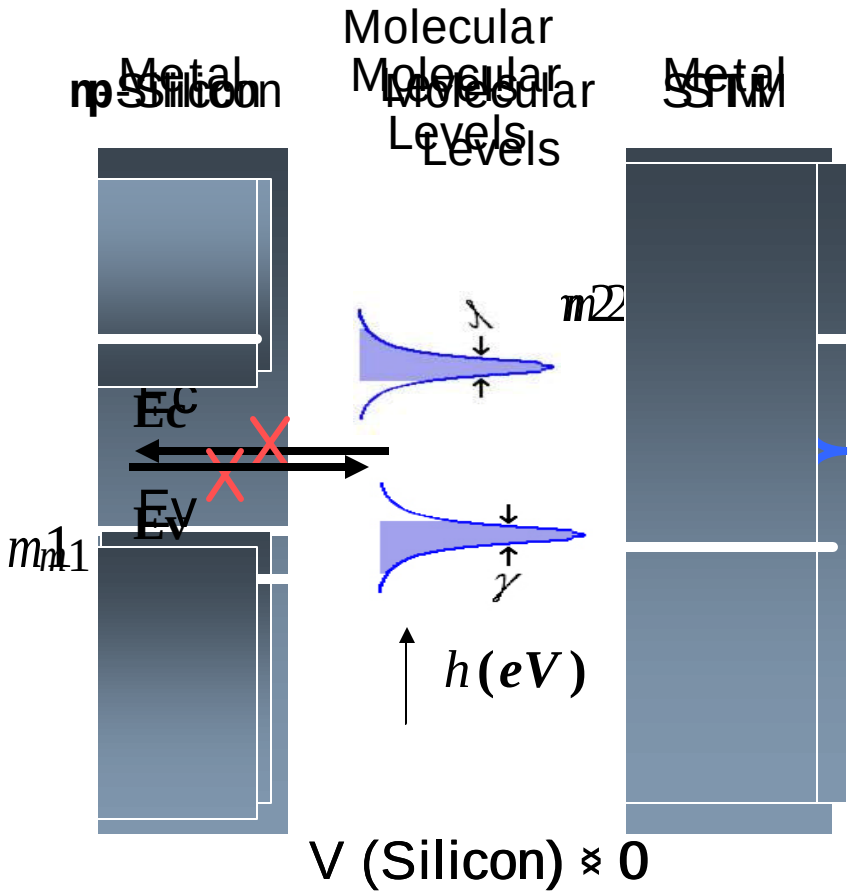
Molecules: Isolated (Closed) and Contacted (Open)

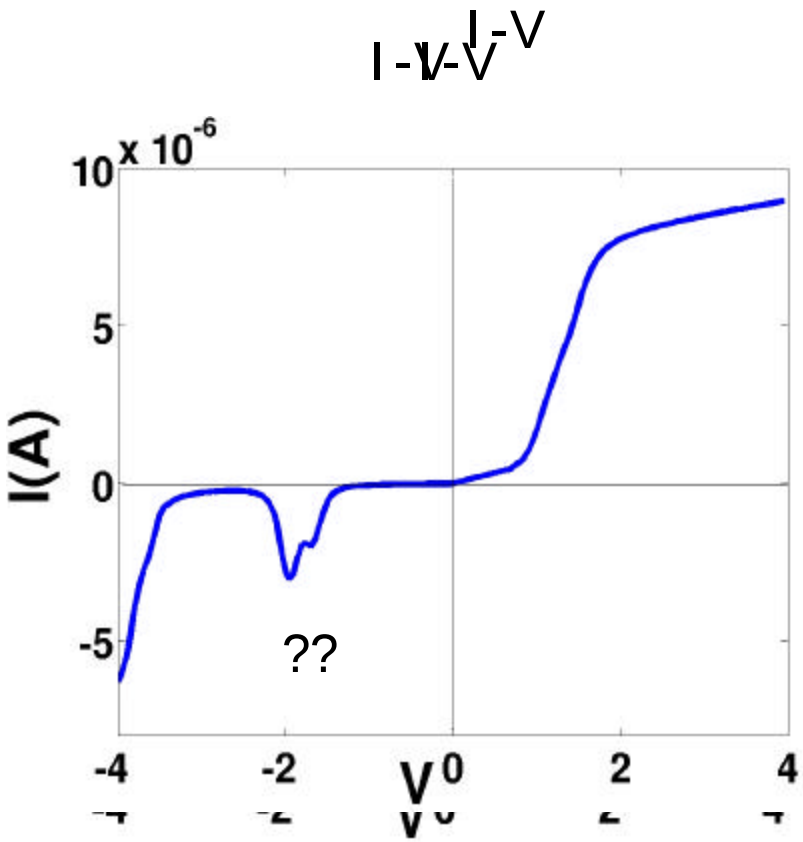
Styrene (C_8H_8)





Silicon-Molecule-STM: Negative Diff, Resistance





Detailed Solution Procedure : Open System, Out-of-equilibrium

Molecular Hamiltonian (H)

(Density Functional Theory)

Self-consistent solution

H

r

$H + \Sigma_1(E) + \Sigma_2(E)$

NEGF

r

Converged ??

No

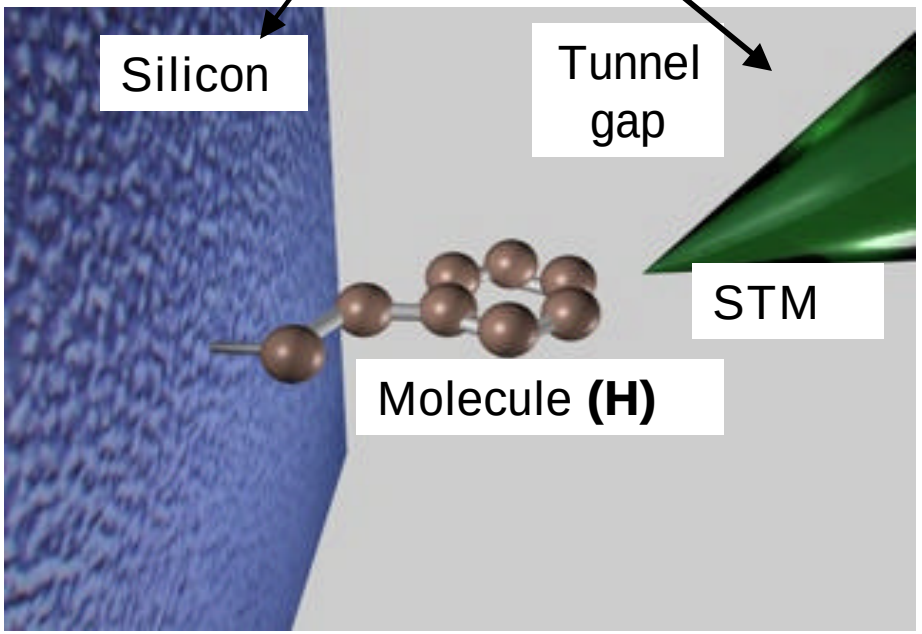
Yes

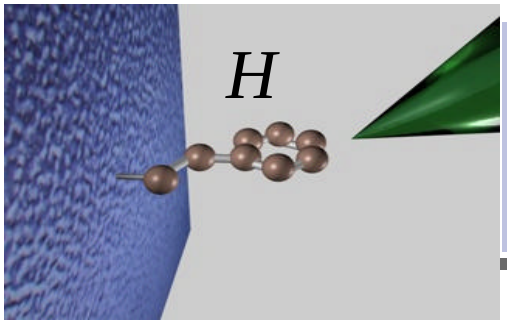
Calculate Current

Self-energy

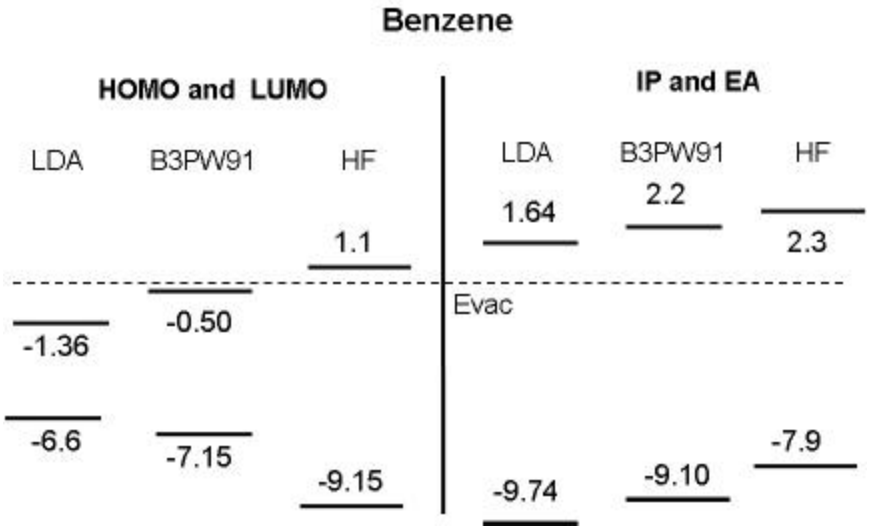
$\Sigma_1(E)$

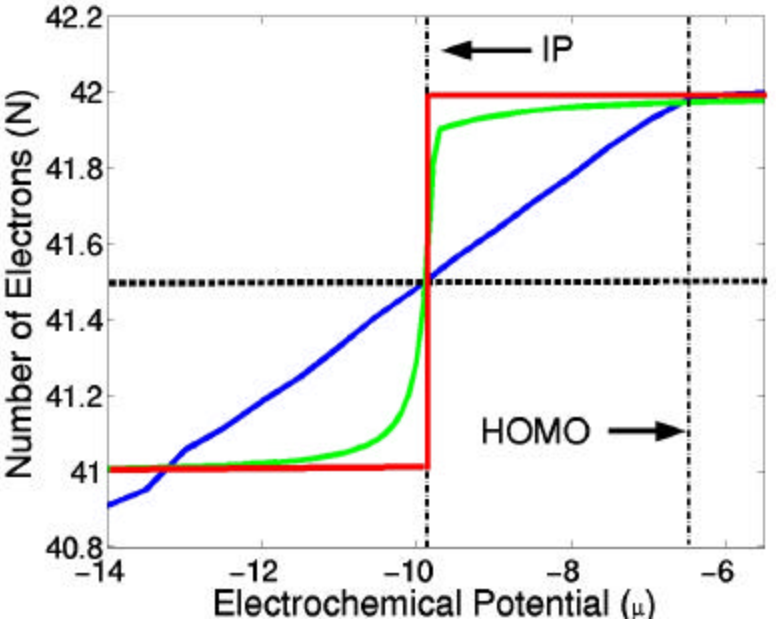
$\Sigma_2(E)$

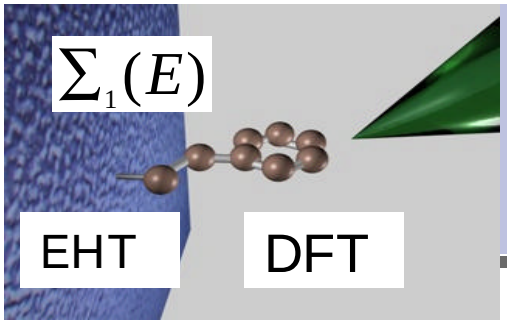




What Hamiltonian do we choose ??

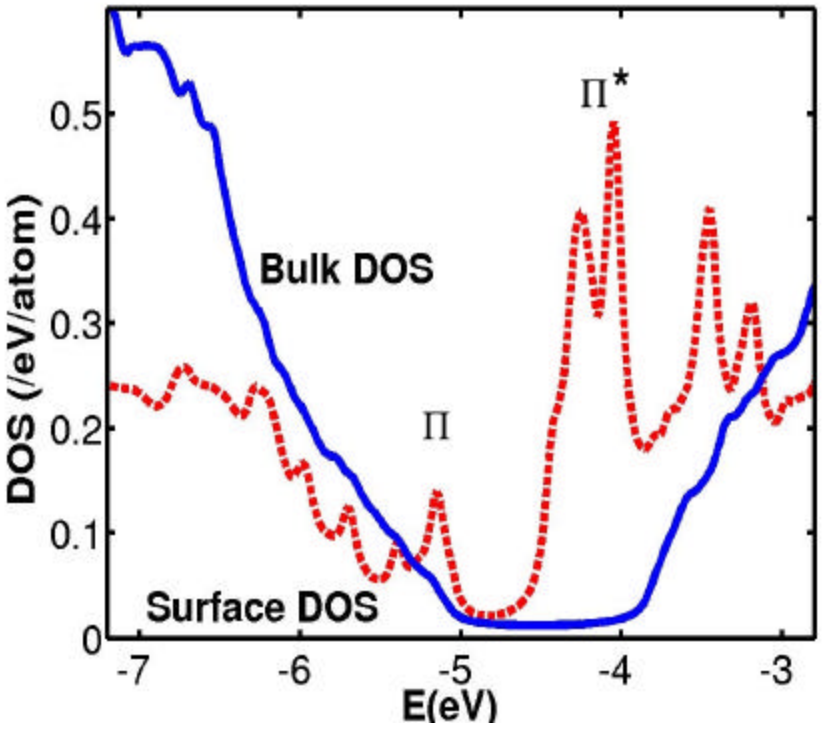




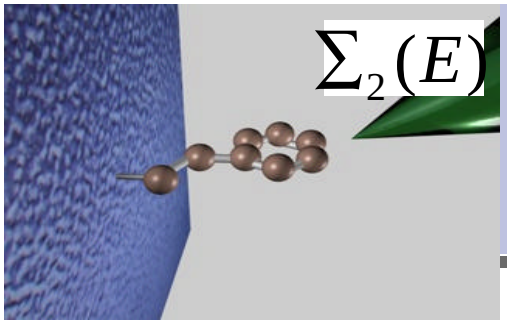


Self-energy $\Sigma_1(E)$, Silicon: Surface States

1. Ab-initio schemes not useful for Bulk or surface Si
2. Need for atomistic basis sets for Si -> can be coupled to atomistic molecule
3. Solution: EHT with Cerda's parameters



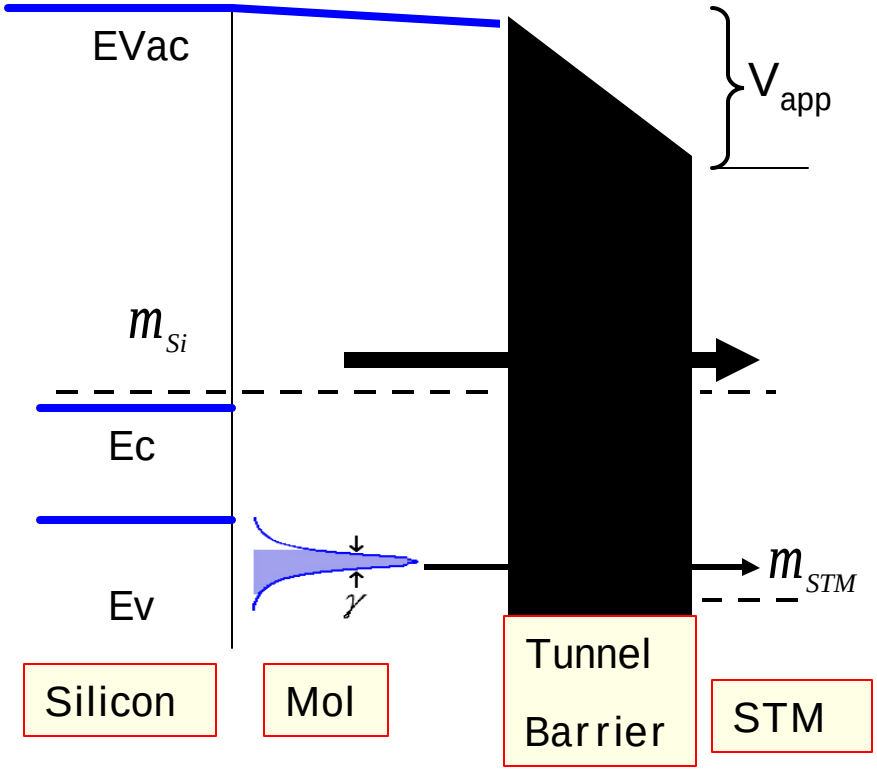
Calculated Si Bulk Density of States (DOS) and Si(100) Surface DOS

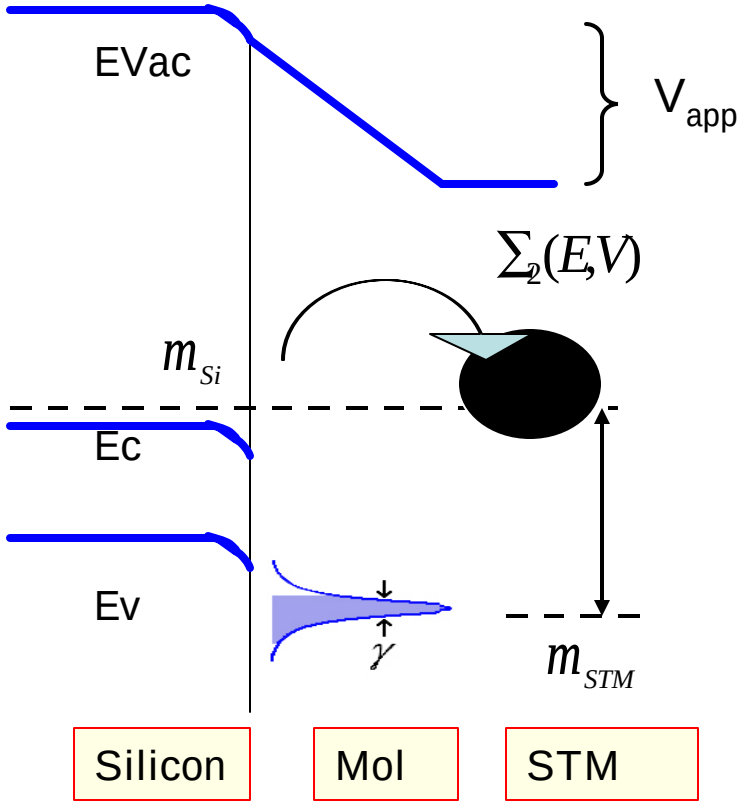


Self Energy $\Sigma_2(E)$ Modeling STM and Vacuum

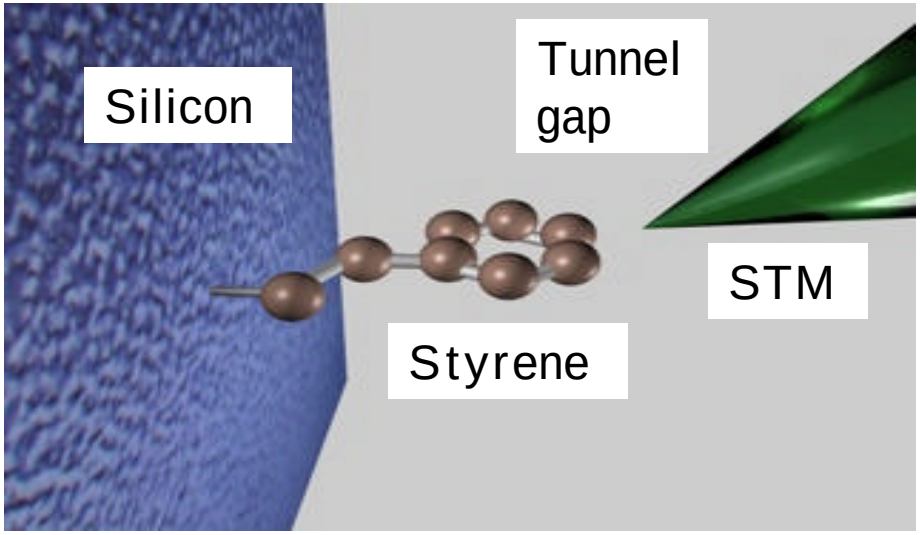
Case I: STM far away from molecule:
Effects (i) Electrostatic (ii) Quantum

Case II: STM touches the molecule:
Collapse of the tunnel barrier



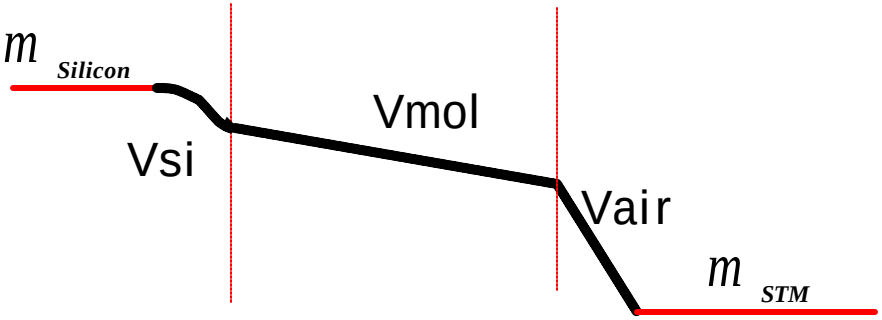


Electrostatics: Laplace

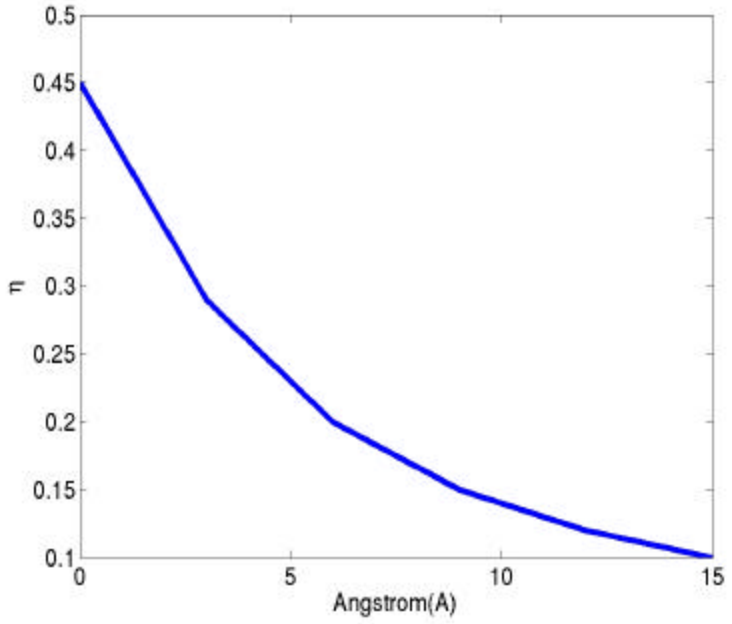


Avg. fraction of bias on molecule

$$h = \frac{V_{Si} + \frac{V_{mol}}{2}}{V_{total}}$$

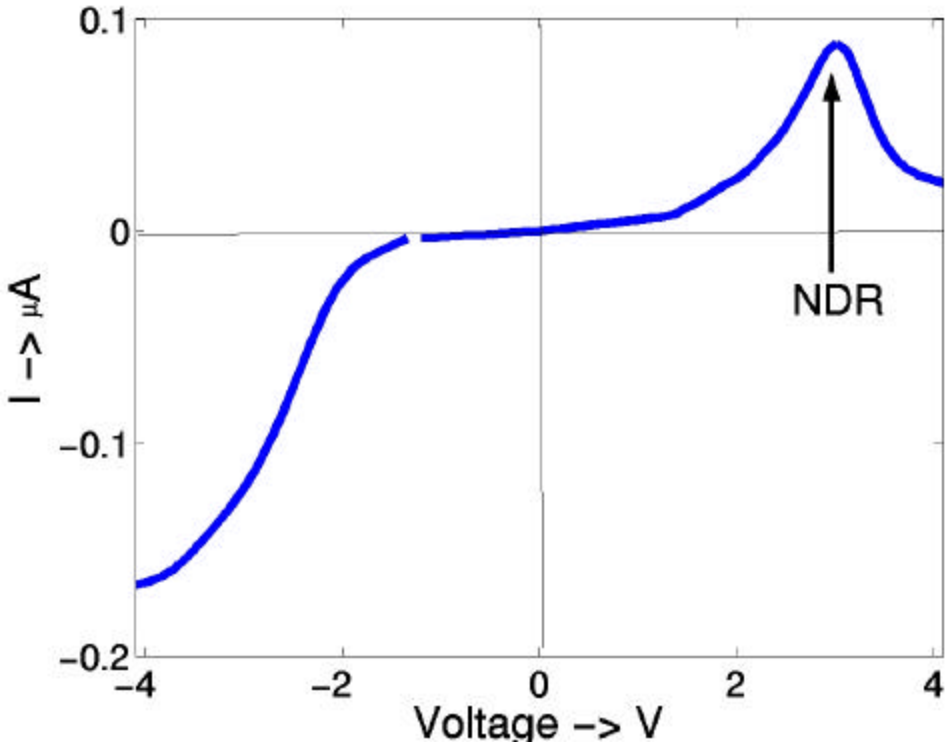


$$V_{total} = V_{Si} + V_{mol} + V_{air}$$



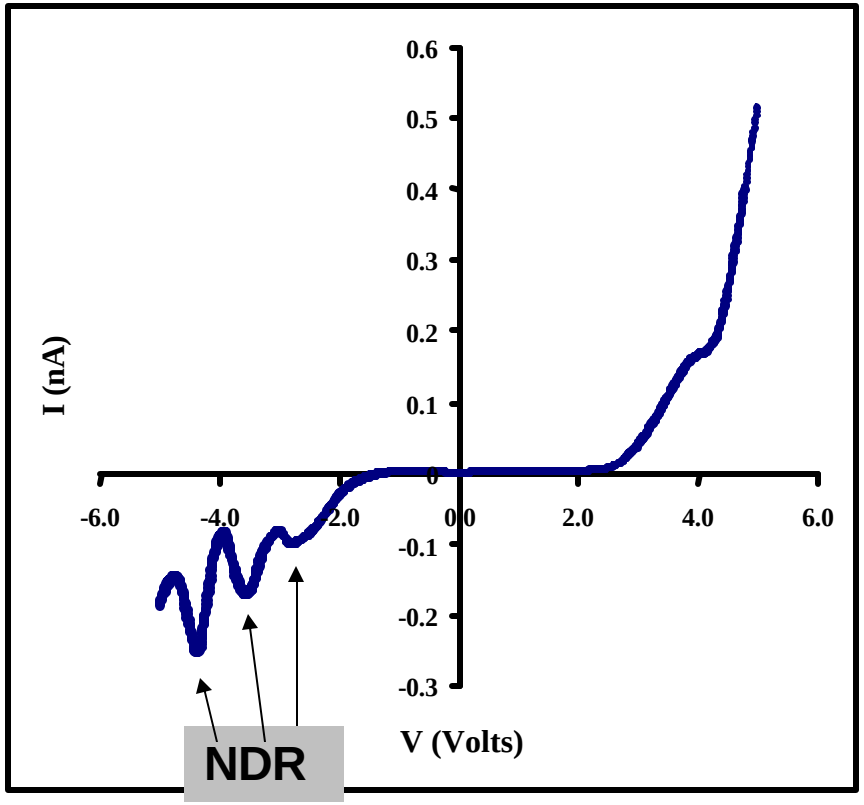
Fully Self-Consistent I-V

Ab-initio fully self-consistent I-V of Styrene on p-Si(100):
LDA/6-31g(d), 163x163

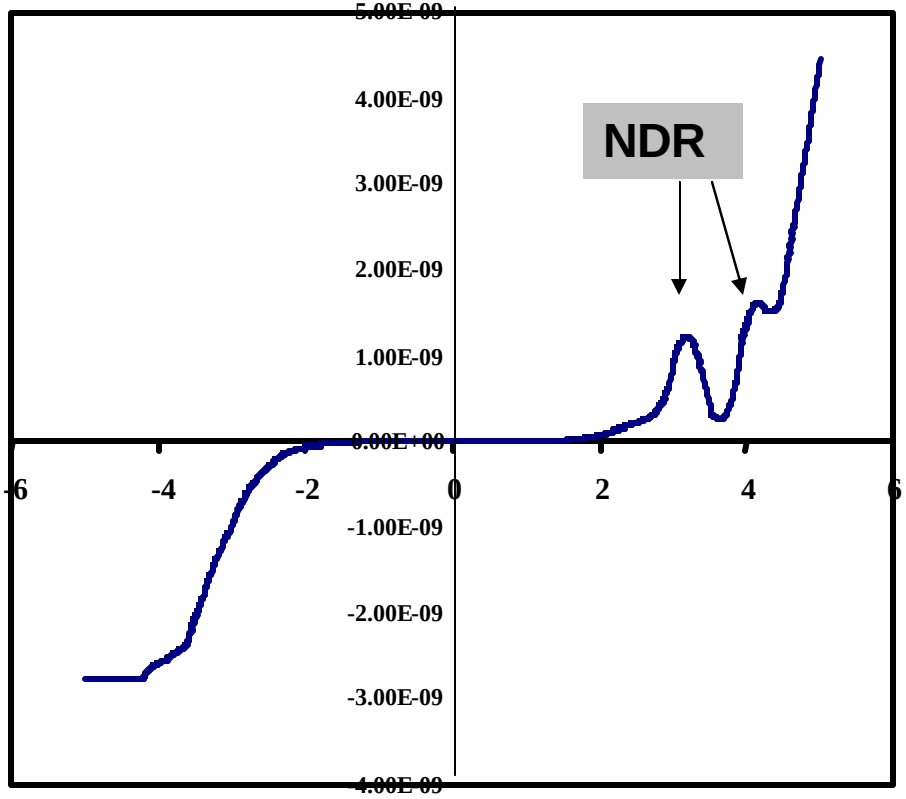


Experimental Verification

Molecule on n-type Si(100)



Molecule on p-type Si(100)

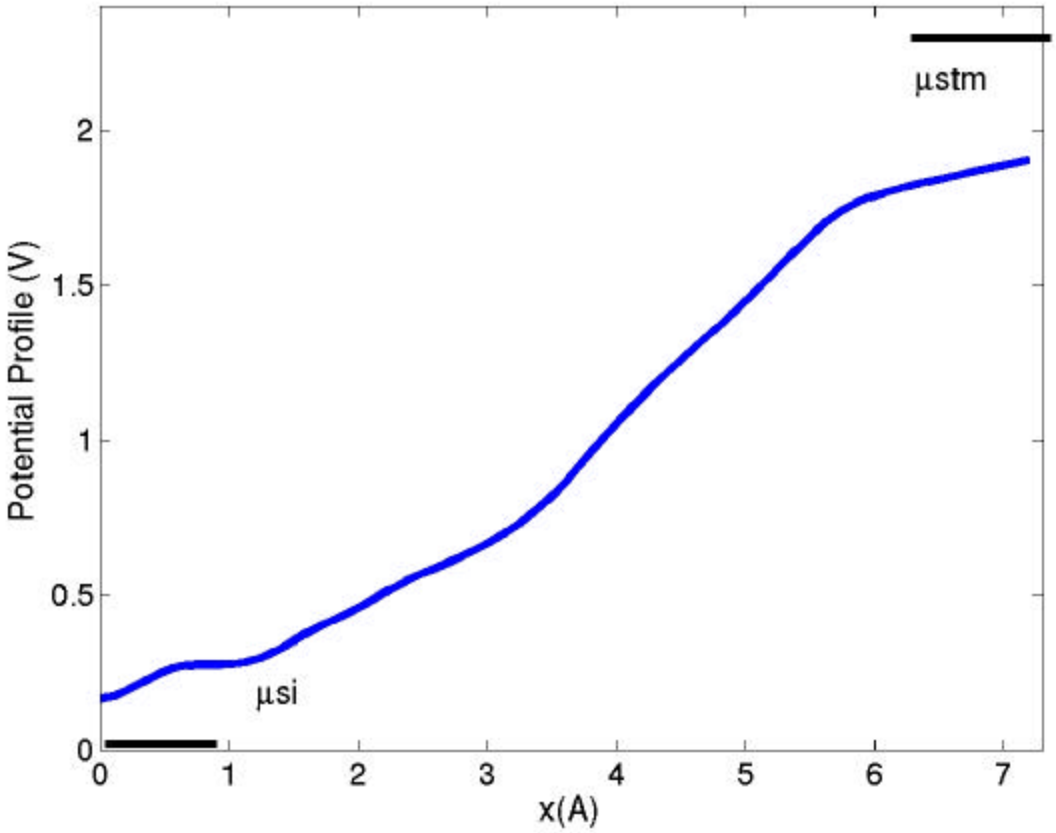


Mark Hersam Group (Northwestern University)

Nanoletters, 01/04 **Cover story**

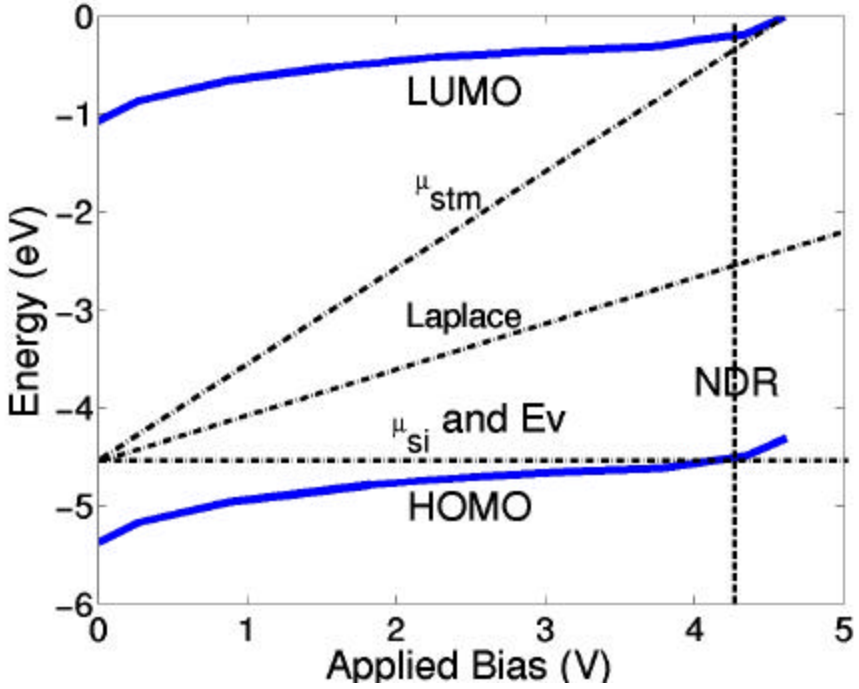
Summary and Conclusions

1. General Framework of coupled DFT-NEGF self-consistent solution
2. Novel RTD proposed for a Molecule-Silicon Heterostructure: polarity reversal predicted dependent on type of doping
3. Experiments agree with the broad feature of polarity reversal, however the exact location of NDR peaks on n-type substrates remain to be understood


µstm
µsi
Potential Profile (V)
x(A)
0
0.5
1
1.5
2
0
1
2
3
4
5
6
7

Fully Self-Consistent I-V

Ab-initio fully self-consistent I-V of Styrene on p-Si(100):
LDA/6-31g(d), 163x163, 4 days on 1 processor machine



Assumption: STM close to molecule, small drop across vacuum gap