Study of the Effect of Spin State on Conductivity in Thiophene Polymers Containing Iron-Carborane Cages

Benjamin Beach[1], Anjana Paudyal[1], Pedro Derosa[1,2]

[1]Louisiana Tech University

[2]Grambling University

Introduction

- The point: Spintronics
  - Potential in smaller and faster computers
  - Makes use of electron spins to store and process data
  - Memory Storage
    - Data writing: spin state
    - Data reading: conductivity

Introduction

- Our focus: thiophene metallacarborane polymers
  - The cages:
    - Interesting electromagnetic properties
    - Great chemical stability
  - The thiophenes:
    - Conducting polymers

Plan of Action

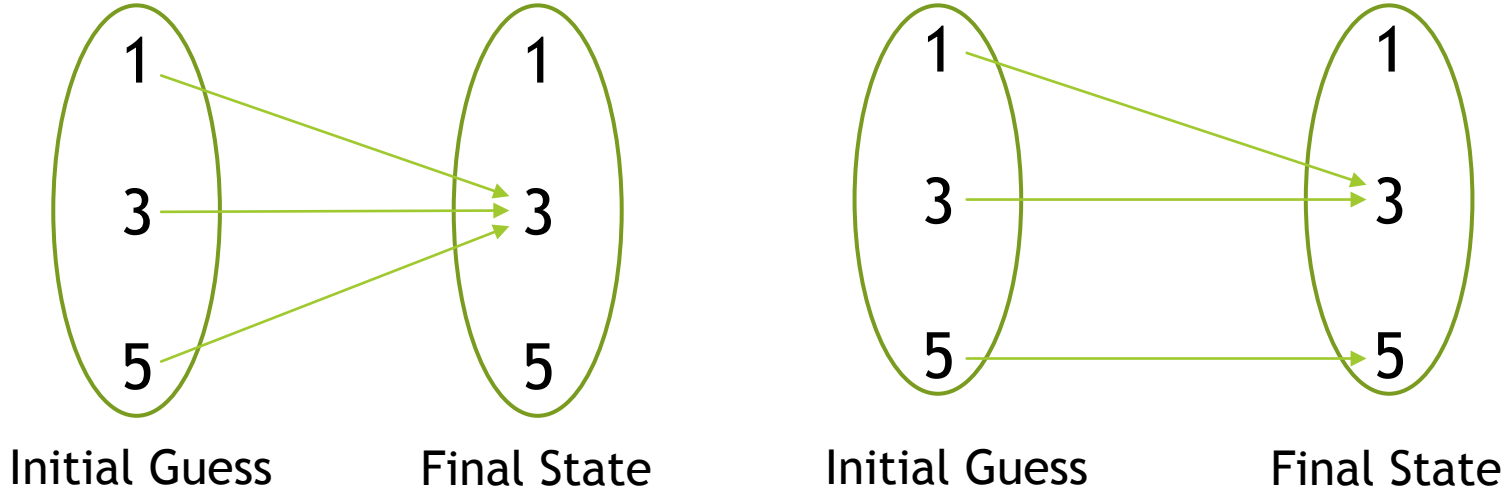
- Our Goal: Find the right central atoms
- Our plan:
  1. Learn how to determine the effect of spin on conductivity
  2. Study the effect for iron
  3. Expand scope to other magnetic metal atoms

Methodology

- Perform DFT geometry optimization via Gaussian 9
- Calculate I-V characteristics via Atomistix
  - DFT-Non-equilibrium Green's Function
- Submitted calculations on LONI

The Problem

- In Atomistix, we can only set the initial spin state
- The spin state is calculated by Atomistix
- Thus, we can end up with situations like these:

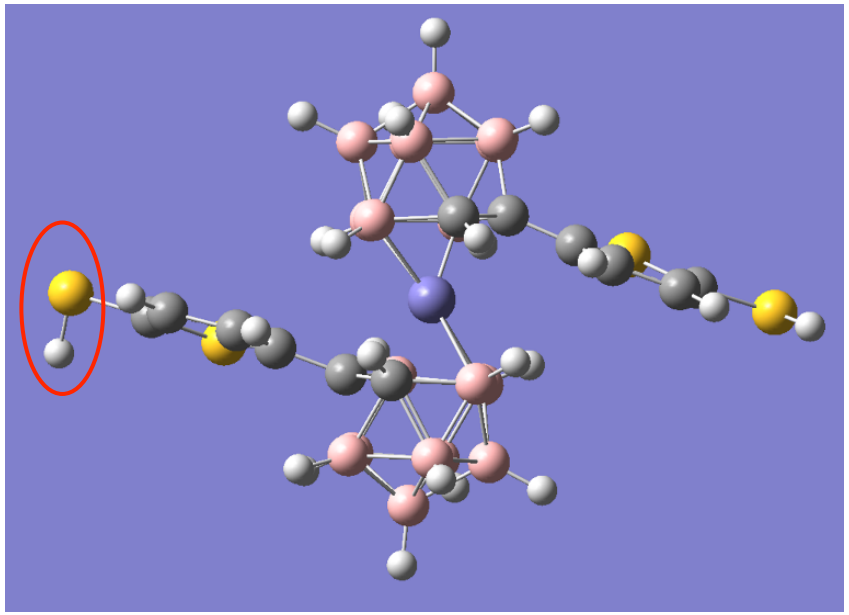


What we tried

- DFT Geometry Optimization
  - Change the charge
  - Change the spin
  - Change the atoms for electrode connection
- I-V calculations: Change the initial spin

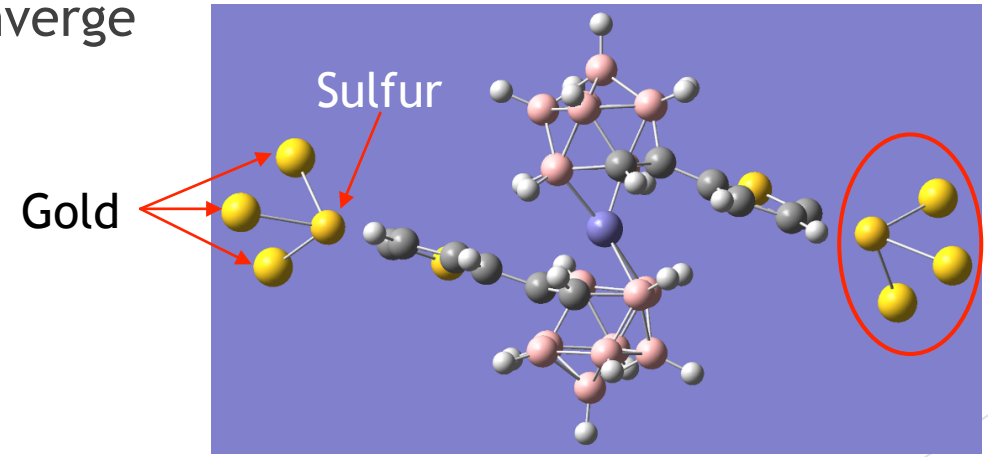
Geometry Optimization: S-H Connection

- Net charges: +1, 0, and -1
- 3 multiplicities per charge
- Not all converged



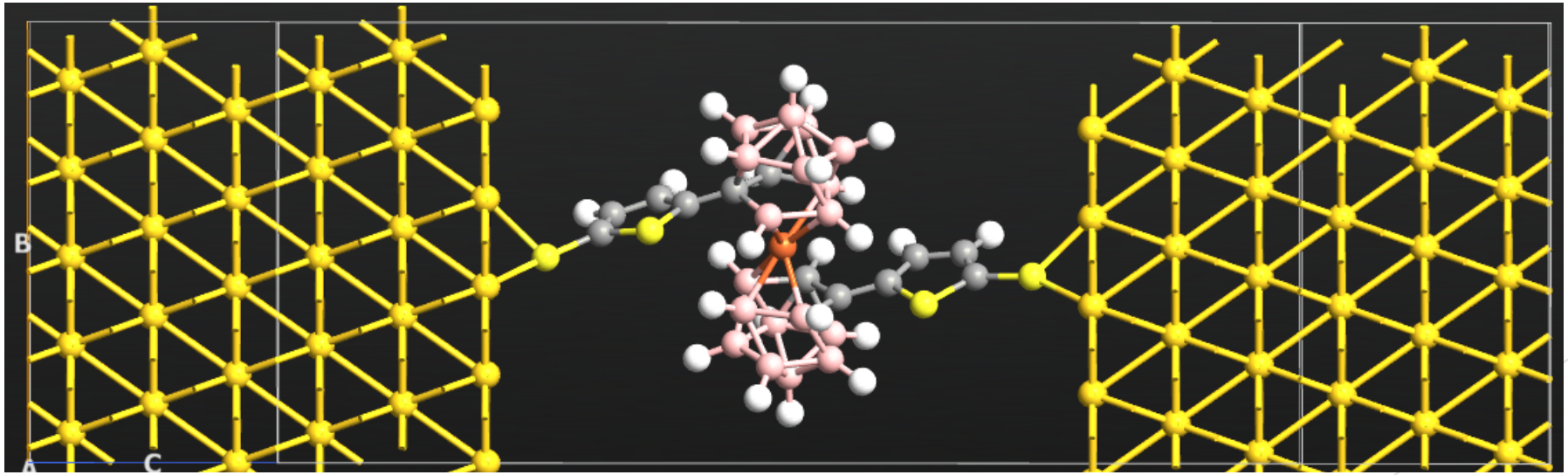
Geometry Optimization

- 3-atom Gold electrodes
  - Fixed electrode structure
  - Stiff, movable electrode structure
    - Attempted the same charges and multiplicities as before
    - Most didn't converge



Atomistix I-V Calculations

- Voltages: 0-0.8V with a step size of 0.1V
- Periodic boundary conditions
- For the mobile gold optimization, tried 3 different spin states on the iron



HPC Platform

- Ran DFT calculations on LONI
- Geometry optimizations: 1-3 days
- I-V curves: 7-10 days

Further Analysis

- Explaining the curve
  - Transmission Spectra
  - Energy Levels
  - Orbitals
- Final spin and net charge: Mulliken Population

Gaussian Results: Hydrogen End Atom

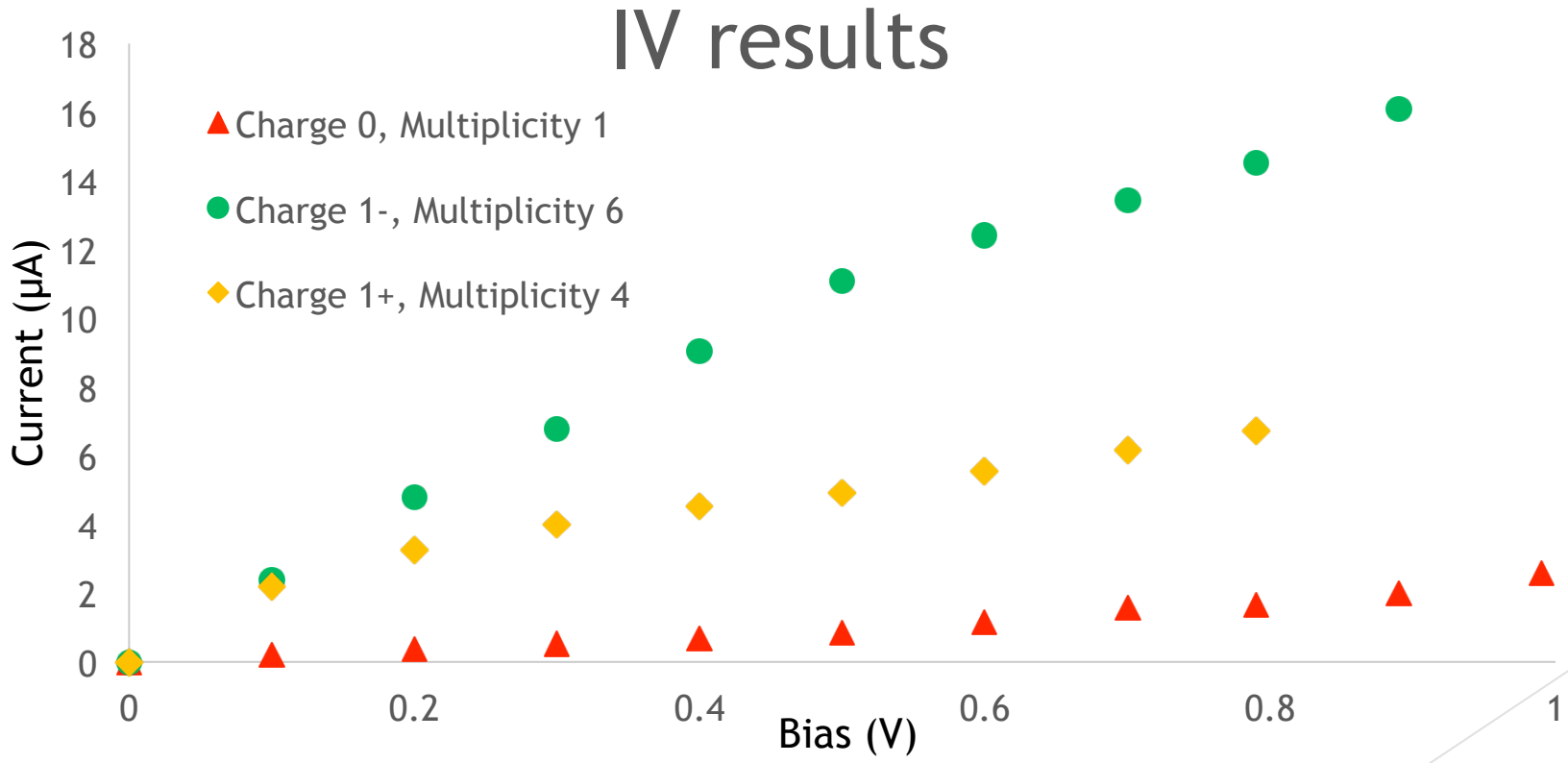
- Early on: more accurate DFT functional; one of each charge converged
- Later: less accurate functional->more accurate functional
  - Two of each charge converged
  - The geometries with the highest energies for each charge were the ones that converged originally

Binding energies (Hartrees):

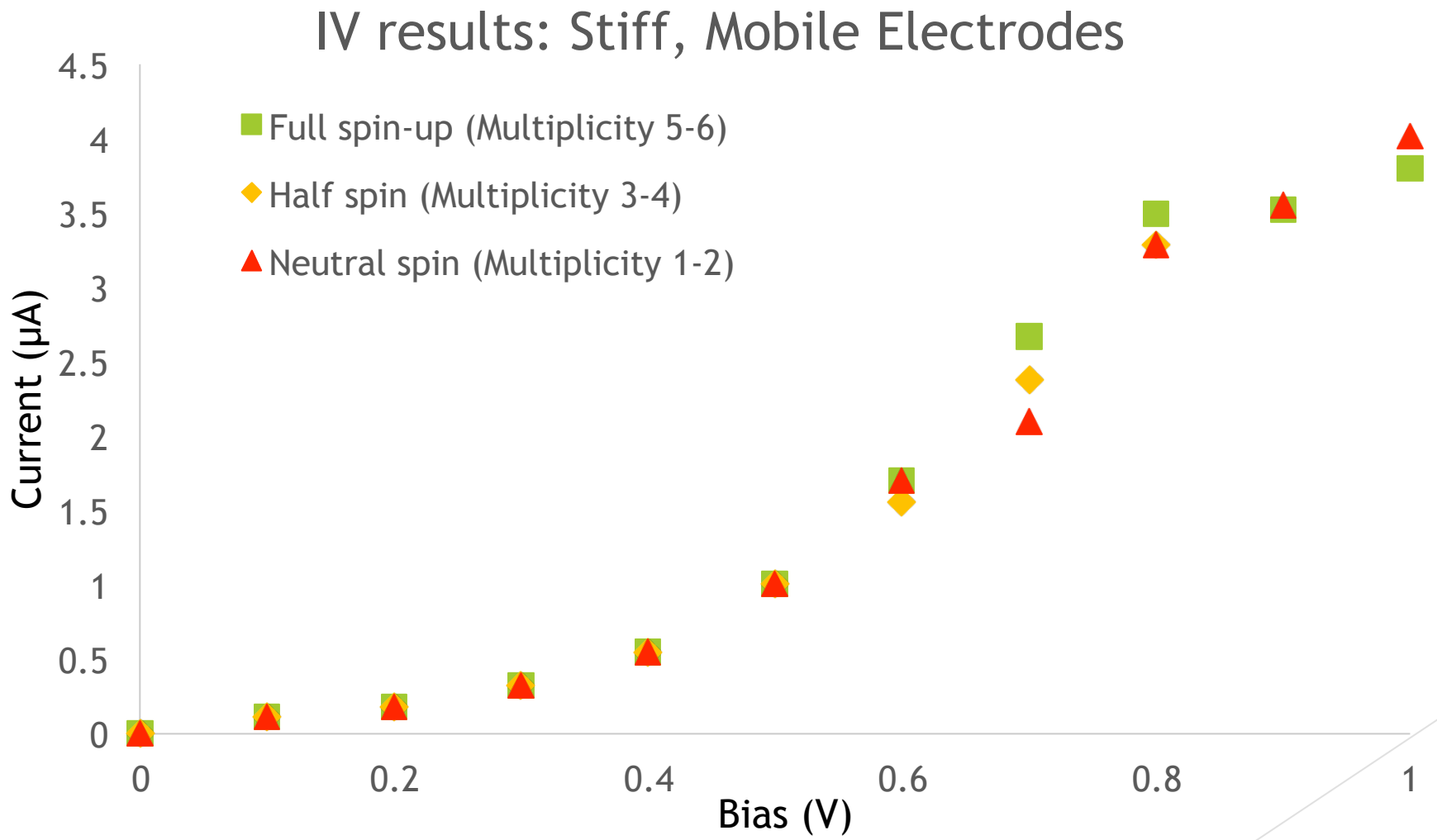
Charge 1-		Multiplicity 4	Multiplicity 6
		-3776.941	**-3776.529**
Charge 0	Multiplicity 1		Multiplicity 5
	-3776.334		-3776.787
Charge 1+	Multiplicity 2	Multiplicity 4	
	-3776.537	**-3776.111**	

I-V results: Hydrogen Optimizations

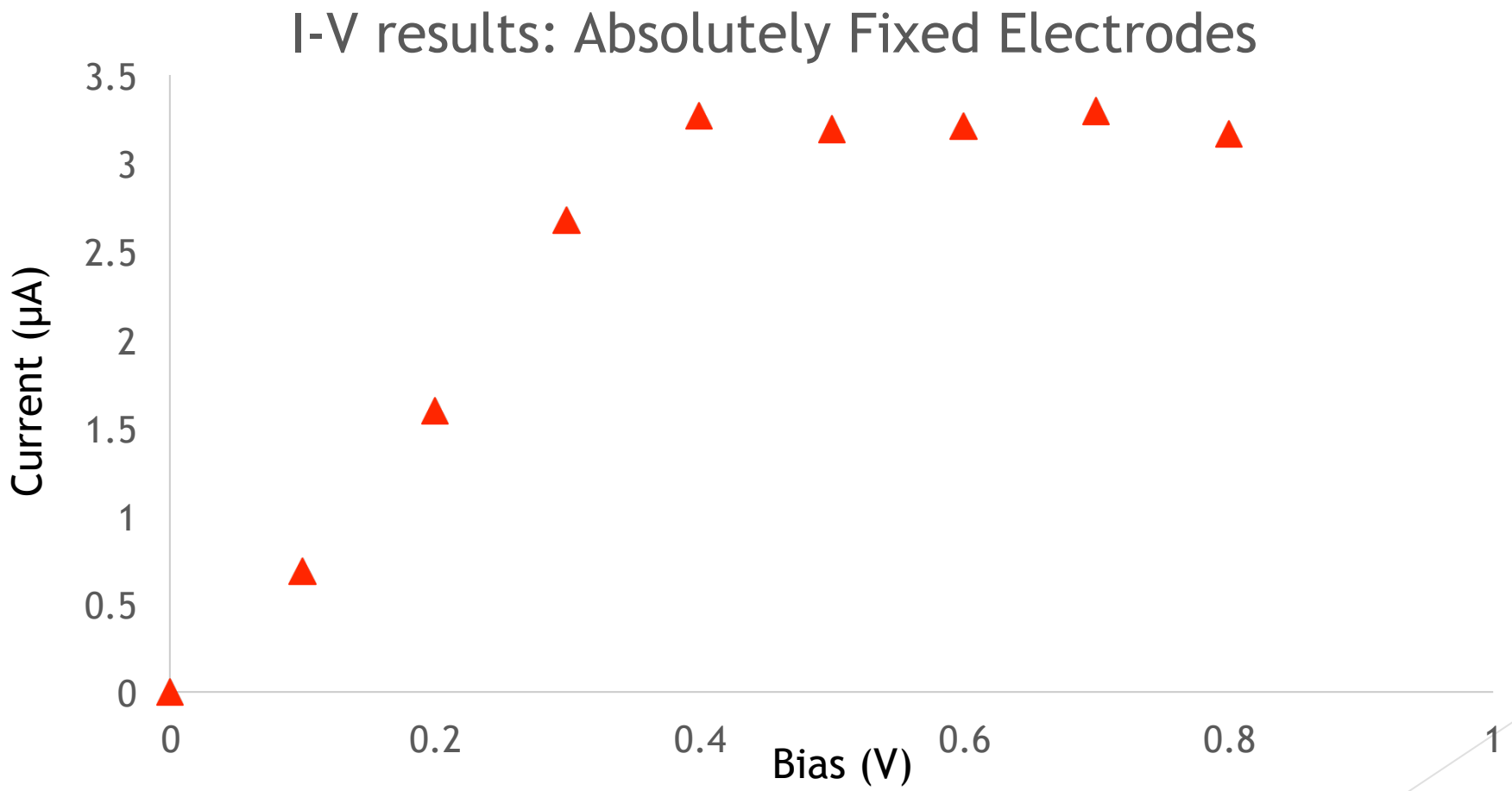
Optimized Charge in Gaussian:	-1	0	1
Final multiplicity:	5	3	3



I-V results: Gold Optimizations



I-V results: Gold Optimizations (cont.)



Conclusions

- Changing the initial spin in Atomistix didn’t seem to have much effect on conductivity
- Changing the charge in Gaussian had a significant effect on conductivity

Future Work

- Calculate I-V curves for the most recently optimized geometries
- Calculate all I-V curves with varied initial spin
- Expand the research to explore the properties of the molecule with other magnetic central atoms

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References

- **Polythiophenes Containing In-Chain Cobaltabisdicarbollide Centers**
  Bruno Fabre, Erhong Hao, Zorabel M. LeJeune, Edith K. Amuhaya, Frédéric Barrière, Jayne C. Garno, and M. Graça H. Vicente ACS Applied Materials & Interfaces 2010 2 (3), 691-702
- Gaussian software
- Atomistix software

Questions?