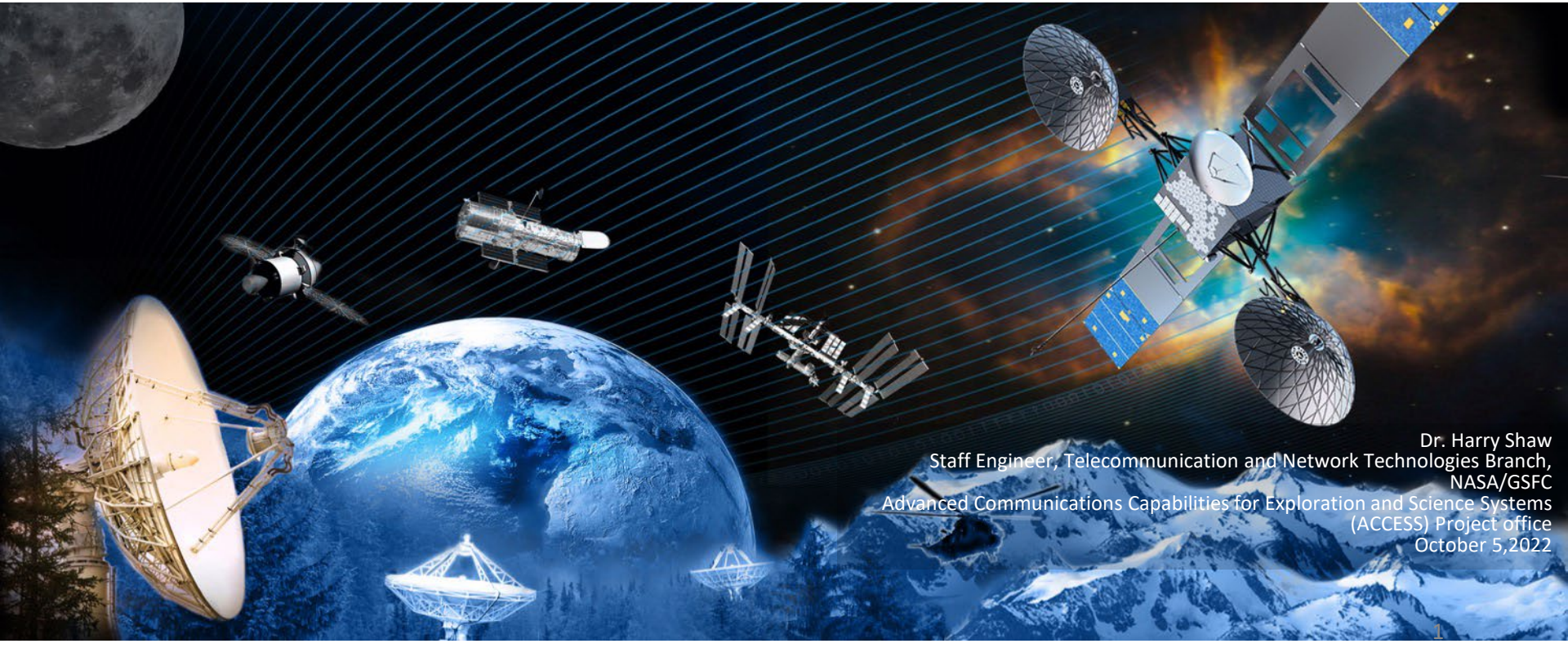
National Aeronautics and Space Administration

NASA

Exploration & SPACE Communications

More than you ever imagined...

Presentation to Michigan State University Electrical and Computer Engineering Department



Dr. Harry Shaw
Staff Engineer, Telecommunication and Network Technologies Branch, NASA/GSFC
Advanced Communications Capabilities for Exploration and Science Systems (ACCESS) Project office
October 5,2022

Introduction

- I lead the technical work in quantum research in the Exploration and Space Communications division, Advanced Communications Capabilities for Exploration and Science Systems (ACCESS) Project office at NASA/GSFC along with Dr. Haleh Safavi Our primary focus areas are:
  - Basic and applied research with our partners to support the development of doctorates in quantum-related fields.
    - Some of them will end up within NASA
  - Planning for eventual operations of quantum enabled satellites
  - Improving “quantum literacy” within the American student population with an emphasis on bringing women and underrepresented minorities into the quantum world
    - We have a very robust intern program.
  - We are not full-time researchers. We have “day jobs” operating communications satellites and ground stations

A little bit about how GSFC is organized

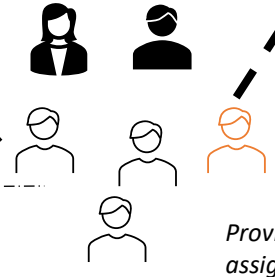
Engineering Directorate

A set of specific engineering disciplines called Branches. My team is in the Telecommunications and Technologies Branch

Signs timecards, does performance evaluations.
Serves as our "straight line" boss that we directly report to

Flight Project Directorate

Instead of Branches there is a set of Projects that executes missions and generally gets the funding from HQ for the matrixed Engineering Branch employees. We are matrixed to the ACCESS Project which operates the Tracking and Data Relay satellites and ground stations around the world. Our HQ boss is the Space Communications and Navigation Directorate (SCaN)



Science Directorate

A set of Branches that usually define what science is to be accomplished via spaceflight missions which are proposed and run by GSFC If awarded by HQ. We talk to them to understand what services they require.

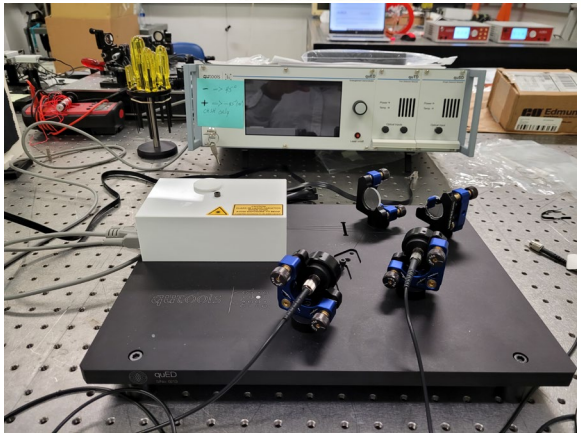
Our main customers

Provides funding and work assignments
Provides inputs to our Performance Evaluations
Serves as our "dotted line boss"

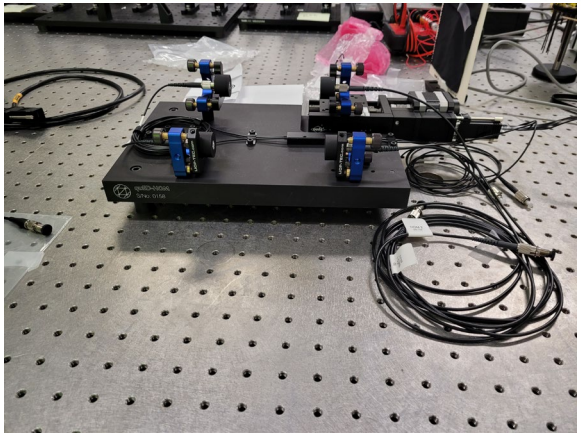
Some of the things we are doing

- We have a new quantum communications lab facility under development.
- In November 2021, we moved into a temporary lab to host some of our equipment and experiments
- We have a combination of Commercial-off-the-shelf systems and discrete hardware for a variety of quantum investigations.
  - There is an emphasis on having facilities for our graduate students (including our university partners) to conduct their research
  - Optical Angular Momentum (OAM) for quantum communications
  - QKD using Chaotic Communications
  - Polarization dispersion experiments

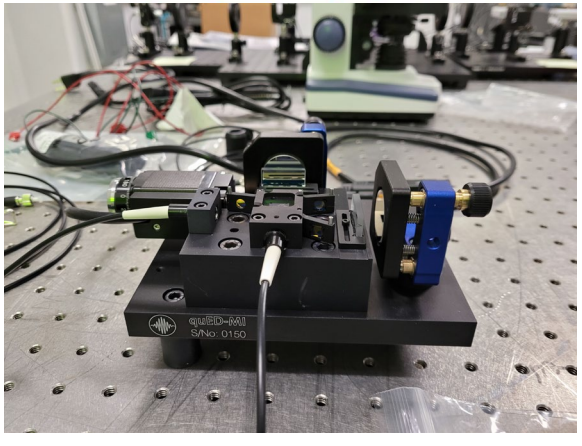
Qutools: This equipment will be used together to perform a variety of experiments



810nm Polarization Entanglement System



Hong-Au-Mandel fixture



Michelson Interferometer

Single Photon Experiments without Interference

Particle Nature of Photons
Quantum Cryptography/QKD: BB84 Protocol
Tomographic Single Photon State Reconstruction
Quantum Random Number Generation

Single Photon Experiments with Interference

Wave Nature of Photons: Single Photon Michelson Interferometer
Quantum Eraser
Wave-Particle Dualism: Michelson + HBT

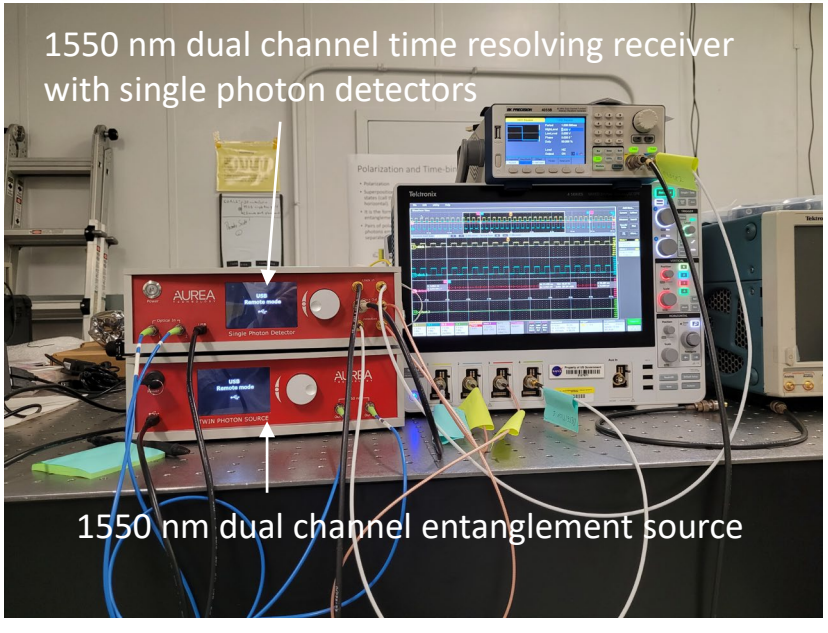
Photon Pair Experiments with Polarisation Entanglement

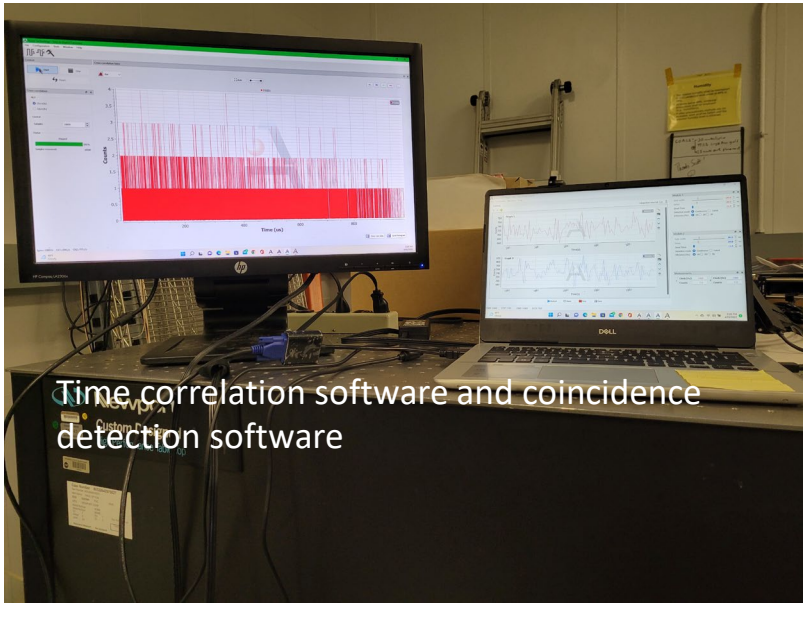
Violation of Bell's Inequality (CHSH)
"Non-Classical" Polarisation Correlations
Tomographic State Reconstruction of an Entangled Photon State
Quantum Cryptography/QKD: BBM Protocol
Quantum Cryptography: Ekert Protocol

Photon Pair Experiments without Polarisation Entanglement

Hong-Ou-Mandel 2-Photon Interference
Hong-Ou-Mandel Interference + Hanbury Brown & Twiss
Franson Interference

Aurea: This equipment will be used as building blocks for experiments with other equipment

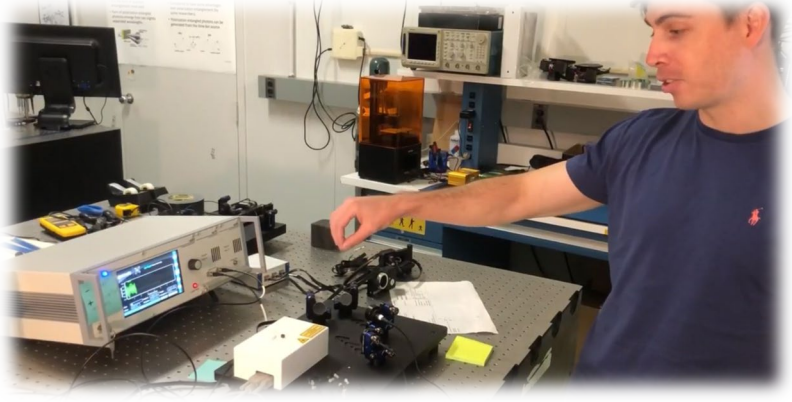




Planned work with Dr. Ayres

- Michigan State (PI Virginia Ayres) - Quantum Demonstration Project
  - Students and faculty will receive training on the ACCESS lab quantum equipment, develop understanding of key scientific principles and pursue investigations.
- Dr. Ayres's graduate student Nick Sebasco is working with NASA
  - Developed our Franson Interferometer setup and performed multiple entanglement verification experiments

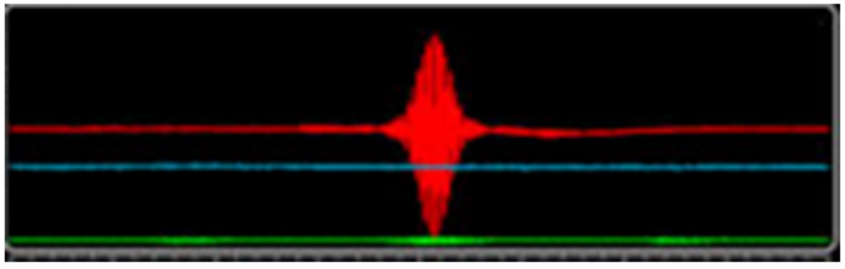




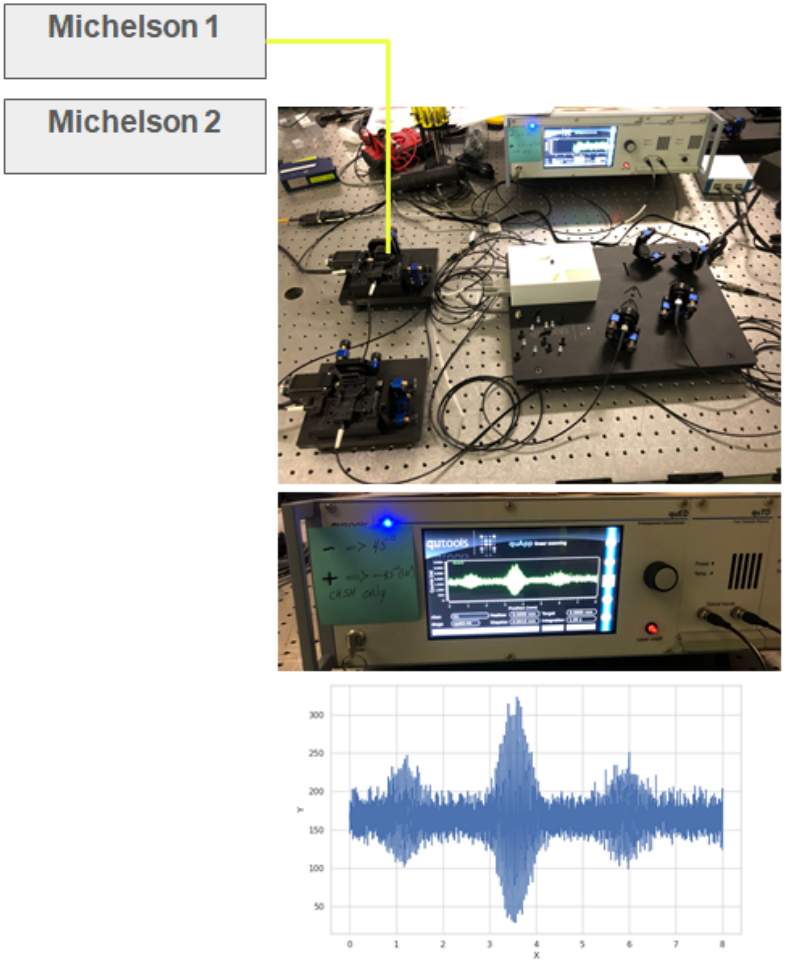
Franson Interferometer

- The Franson experiment demonstrates the impossibility of describing quantum phenomena within the framework of any local model of hidden variables.

Each arm of the Quantum Entanglement Demonstrator is connected to a Michelson interferometer.



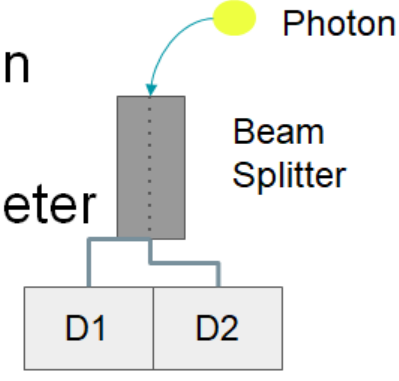
High visibility 2-photon interference fringes depict a violation of Bell's inequality.



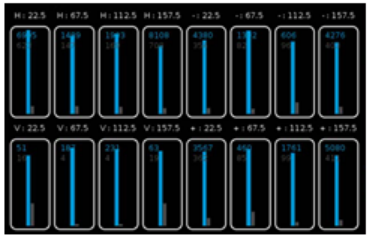
Entanglement verification

Nick Sebasco contributions: Setup, Calibrate, Test, Document

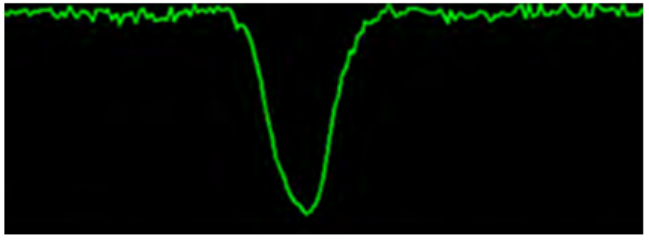
- Clauser-Horne-Shimony-Holt (CHSH) inequality violation
- Hong-Ou-Mandel (HOM) dip
- Single Photon Interference with a Michelson interferometer
- Hanbury-Brown Twiss (HBT) experiment
- (New Summer 22) Franson interference experiment



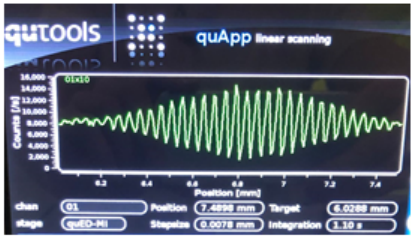
HBT experiment



CHSH Inequality



HOM dip



Single Photon Interference

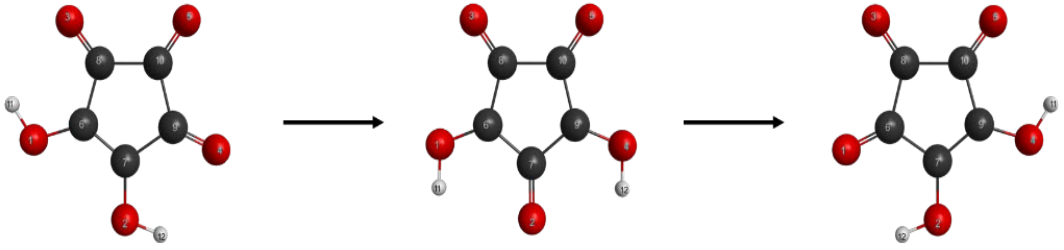
Unpublished Work in Progress

Low cost, above LN_2 temperature quantum circuits

- We have a research project to develop quantum circuits relying on stable tautomeric isomers
- Harry Shaw (NASA/GSFC), Kyle Sherbert(NASA/GSFC intern), Virginia Ayres (Michigan State), Deborah Preston (retired)
- Tautomers are conformational isomers that can take two states
- General questions we are looking at include:
  - Does the transition in states involve a superposition of states?
  - Does it involve quantum entanglement?
  - What is uniquely “quantum” about the circuit implementation?
  - How do we implement these materials in real-world quantum circuits?
- We have two tautomers we are looking at in great detail in simulation
  - Croconic acid
  - Azobenzene

Proton tautomerism in croconic acid

- Croconic acid is an important ferroelectric organic with favorable properties for classical circuits.
- The proton shift certainly provides for a possibility of a quantum oscillator
- There are a number of interesting thermodynamic issues to be explored



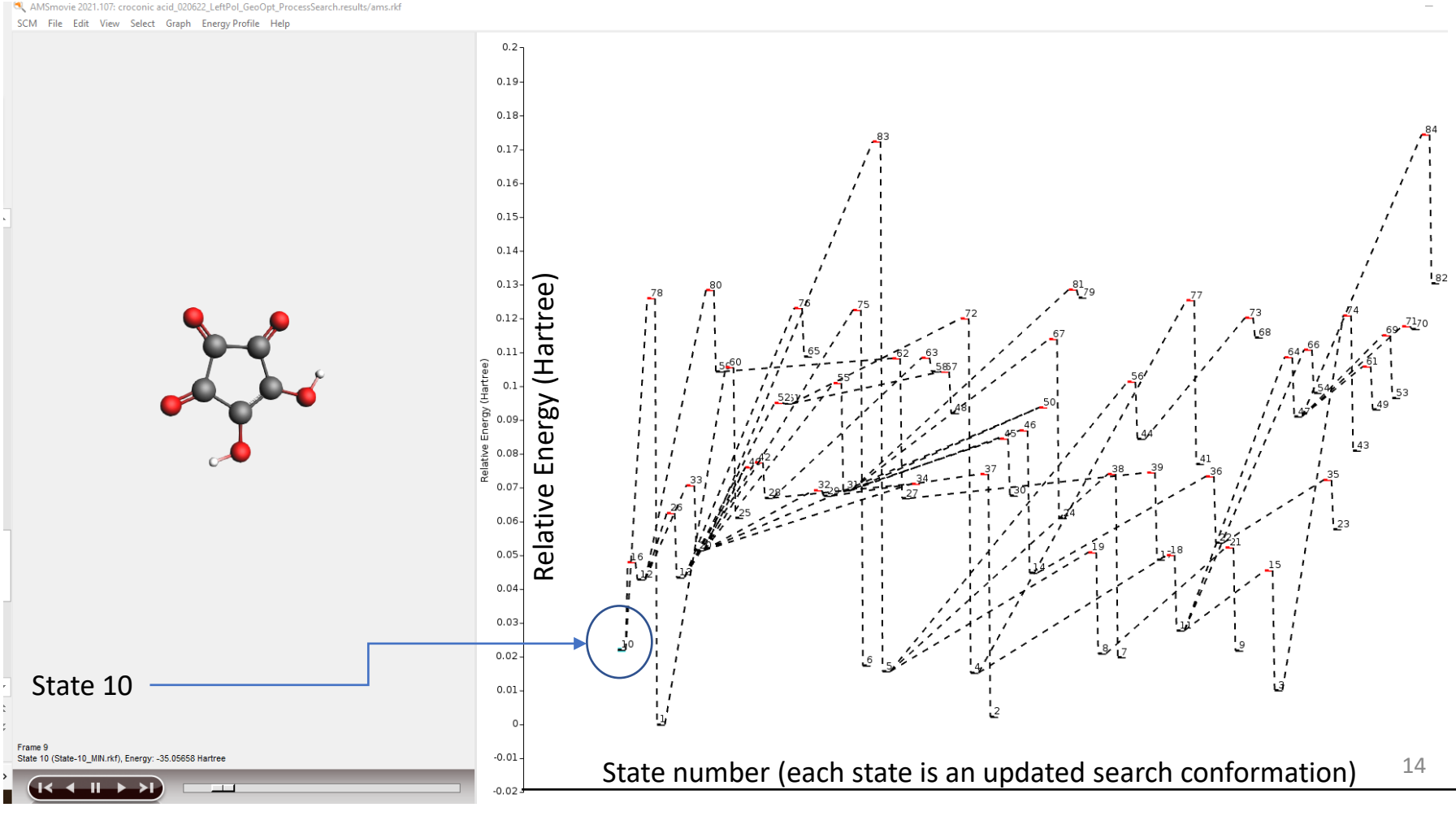
Kyle Sherbert's GAMESS simulation

Transition path and the potential energy surface

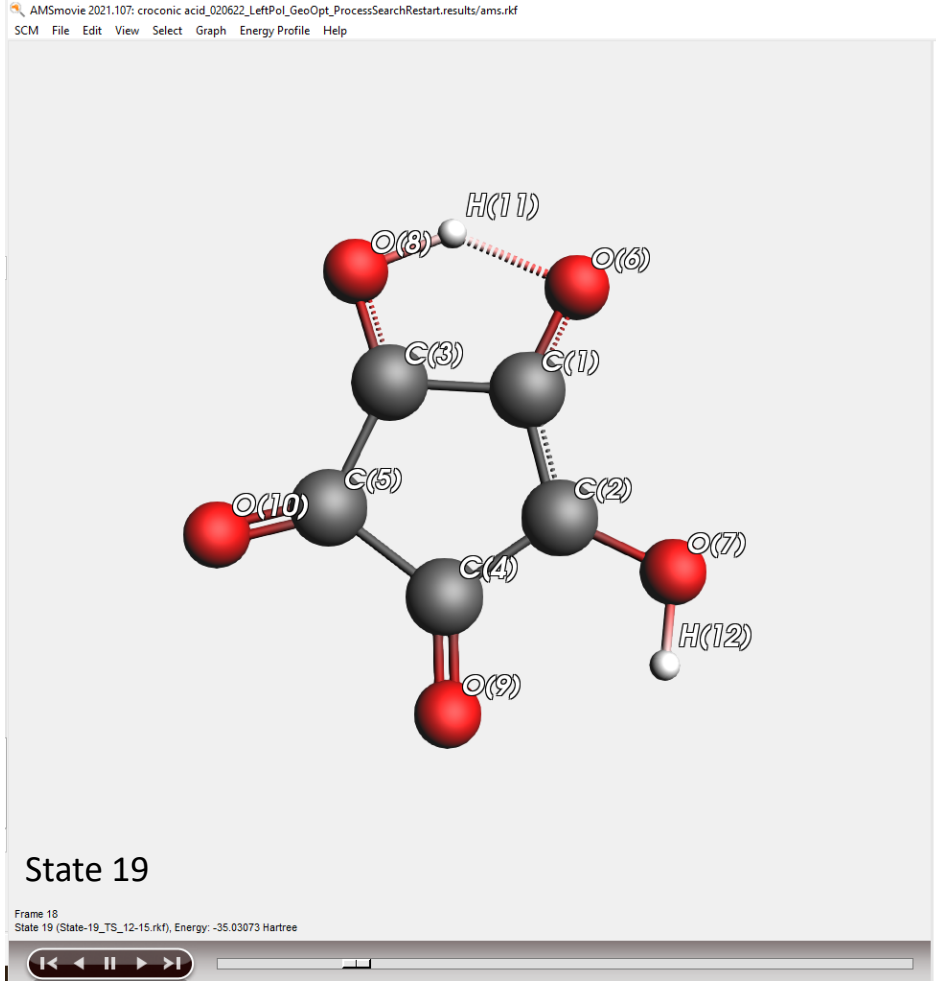
- We are performing simulations of the transition involving the proton transfer in croconic acid.
- We are using the Amsterdam Modeling Suite (AMS) from SCM and GAMESS
- AMS has a Potential Energy Surface explorer tool
- You define Expeditions and Explorers. They seek out potential energy scans with minimum energy.
- Required multiple iterations
  - 1st iteration: Use many expeditions and a few explorers
  - 2nd iteration. Narrow down candidates from the 1st iteration
  - Speed is tied to the number of cores in the machine. I have 4 cores, so I used only 12 explorers and 50 expeditions the first run and 12 explorers and 8 expeditions for second run

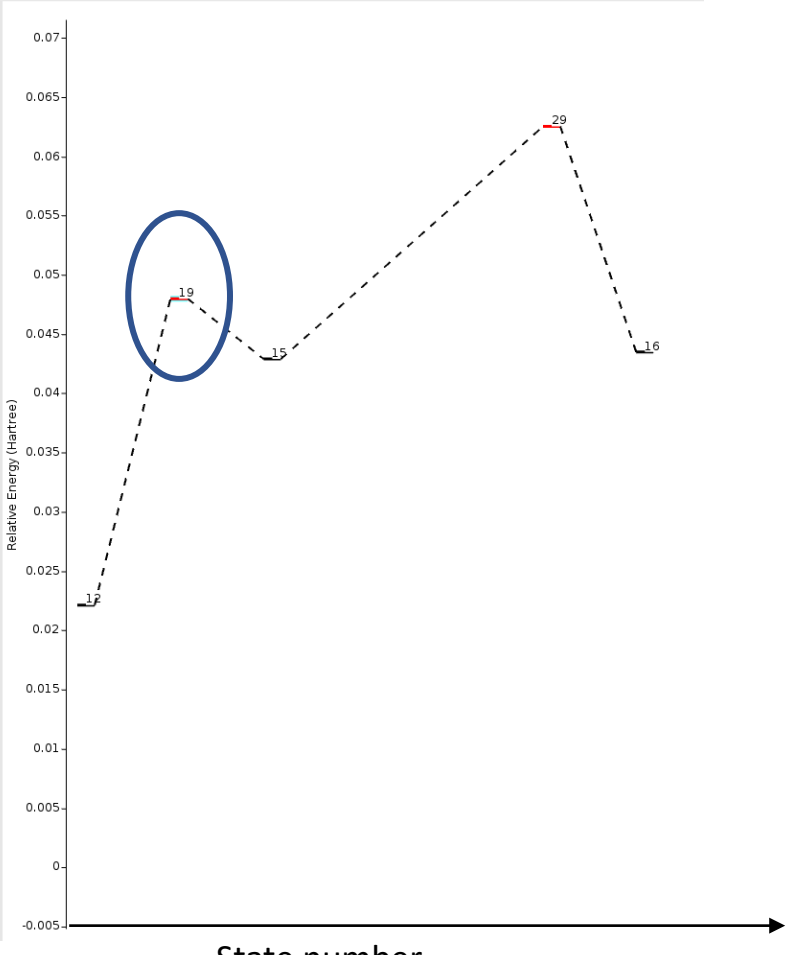
PES explorer 1st run

- Many states, most of which do not correspond to any physical reality



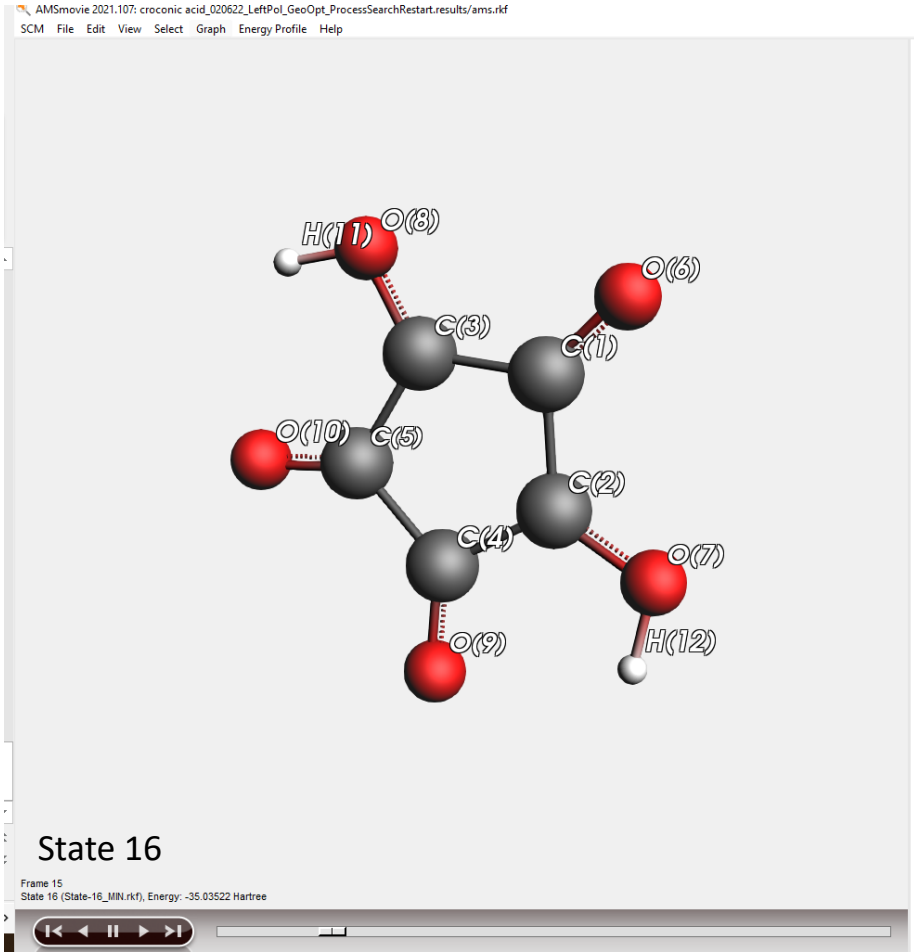
Transition State

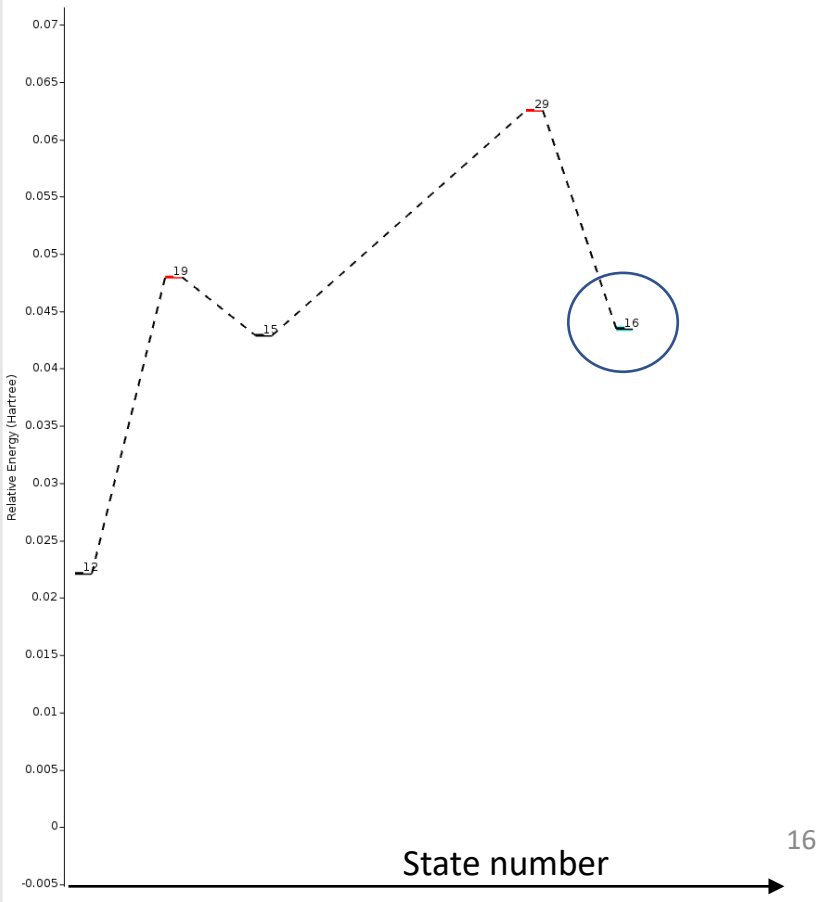




PES final run

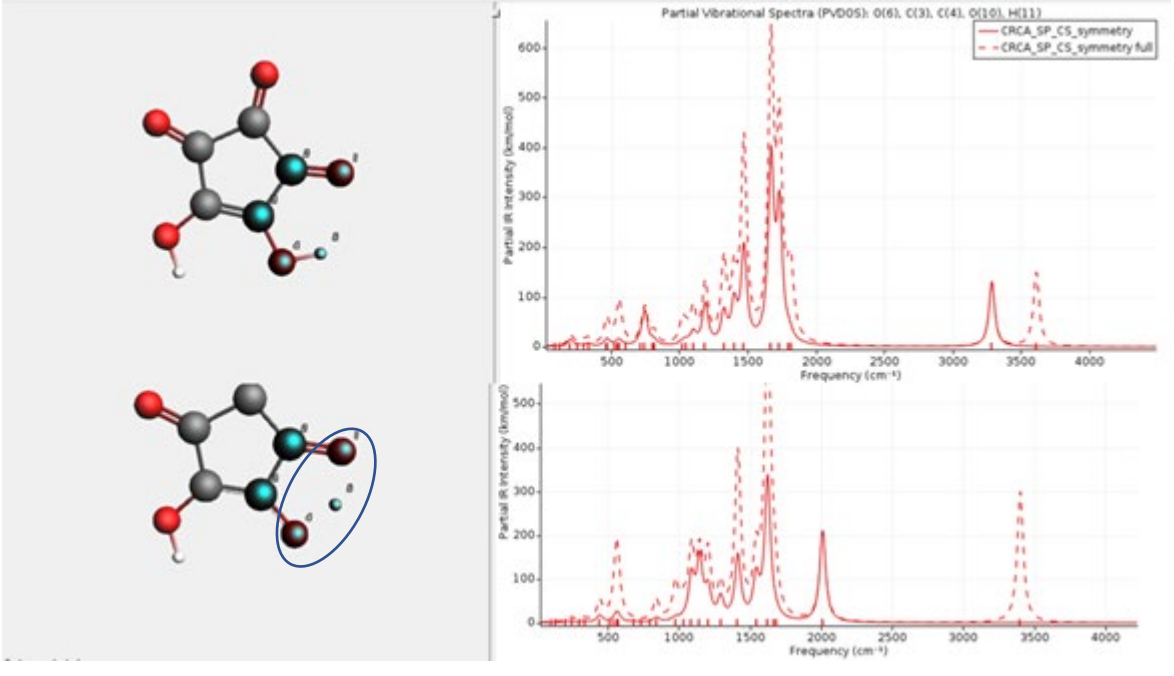
- It has been narrowed down to five states for which GAMESS and AMS seem to be in agreement (mostly)





Vibrational spectra and density of states

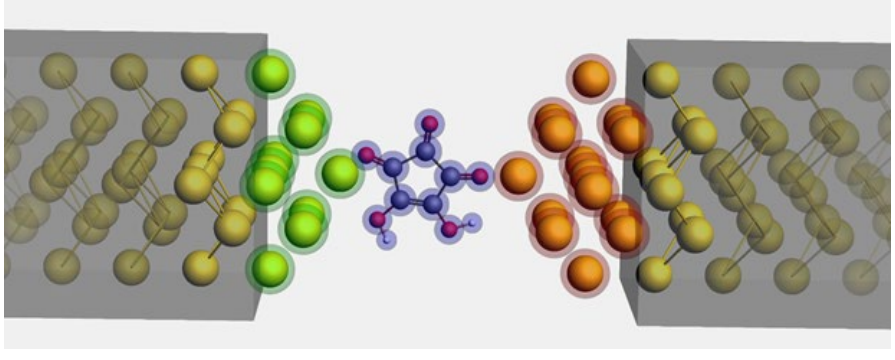
- Both are important in understanding the mechanisms at work and how we might implement a functioning circuit



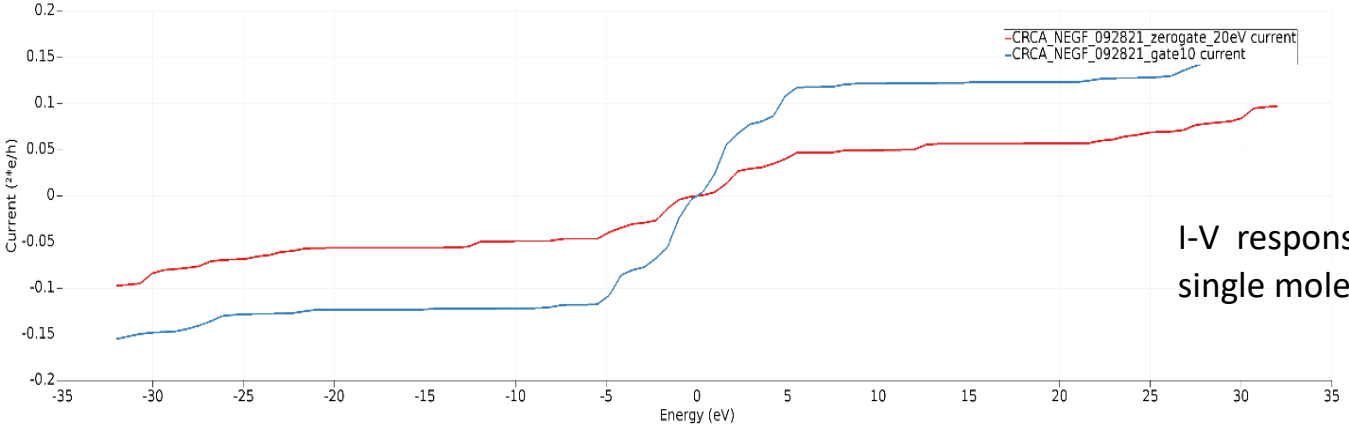
Vibrational spectra of the ground state (A) vs the transition state (B). The stretching mode of H11-O6 (circled) is the single imaginary vibrational mode at -886 cm^{-1} in the tautomeric transition state Note the appearance of the peak at 2005 cm^{-1} in the transition state and the split carbonyl peaks at 3282 cm^{-1} and 3607 cm^{-1} in the ground state appear as a single peak at 3395 cm^{-1} in the transition state.

Electronic transport through croconic acid. Single molecule circuit simulation

- AMS uses a non-equilibrium Green's function approach to simulate electron transport
- There is much refinement to be done on these models. We need to include the polarization reversal that occurs under tautomerization. And deep dives into the practical interpretation of the simulation results are to be conducted.
- Additional work is planned to investigate electron and hole mobilities



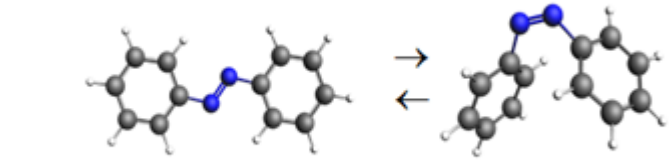
Single molecule circuit of croconic acid between gold electrodes



I-V response at zero bias and 10V bias for single molecule croconic acid circuit

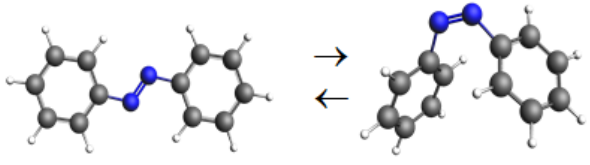
cis- & *trans-* tautomerism in azobenzene

- Azobenzene can be optically stimulated at UV between the ground state (*trans*-) and an excited state (*cis*-) with reasonable thermal stability and reverse switched with visible light
- Azobenzene, and a number of derivatives are also important candidates for classical organic circuit elements such as photoswitches. In our case, we are interested in quantum memories
- We are using AMS modeling suite, GAMESS and ChemOffice which has a good MM2 and MM94 modeling interface



trans-azobenzene *cis*-azobenzene

Azophenol-based Quantum Memories using excitable tautomeric state transitions



trans-azobenzene (ground state)

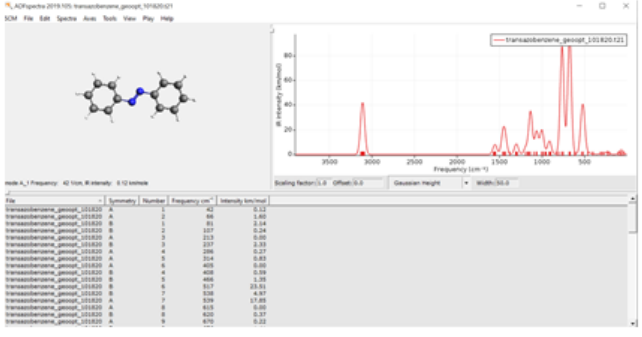
cis-azobenzene (excited state)

- The *trans-cis* transition in the class of azo-compounds has been recognized as a potential organic memory system for some time[3]. Both electron spin and nuclear spin are available as memory phenomena
- In this work we study azo-compounds to determine how to fabricate an ensemble of tailorable, tunable quantum memories[4]
- A variety of azobenzene and their derivatives are being studied using DFT-LDA, DFT-GGA:BLPY, Hartree-Fock and TDDFT methods to evaluate ground and excited state transitions suitable for developing quantum memories[5]

Calculate the frequencies and normal modes of the trans- and cis-configurations to obtain the Hessian eigenvalues and eigenvectors[1,2]

$$H_{ij} = \frac{\partial^2 E}{\partial R_i \partial R_j}$$

H_{ij} = 2nd derivative of energy over the set of nuclear coordinates R_i, Rj

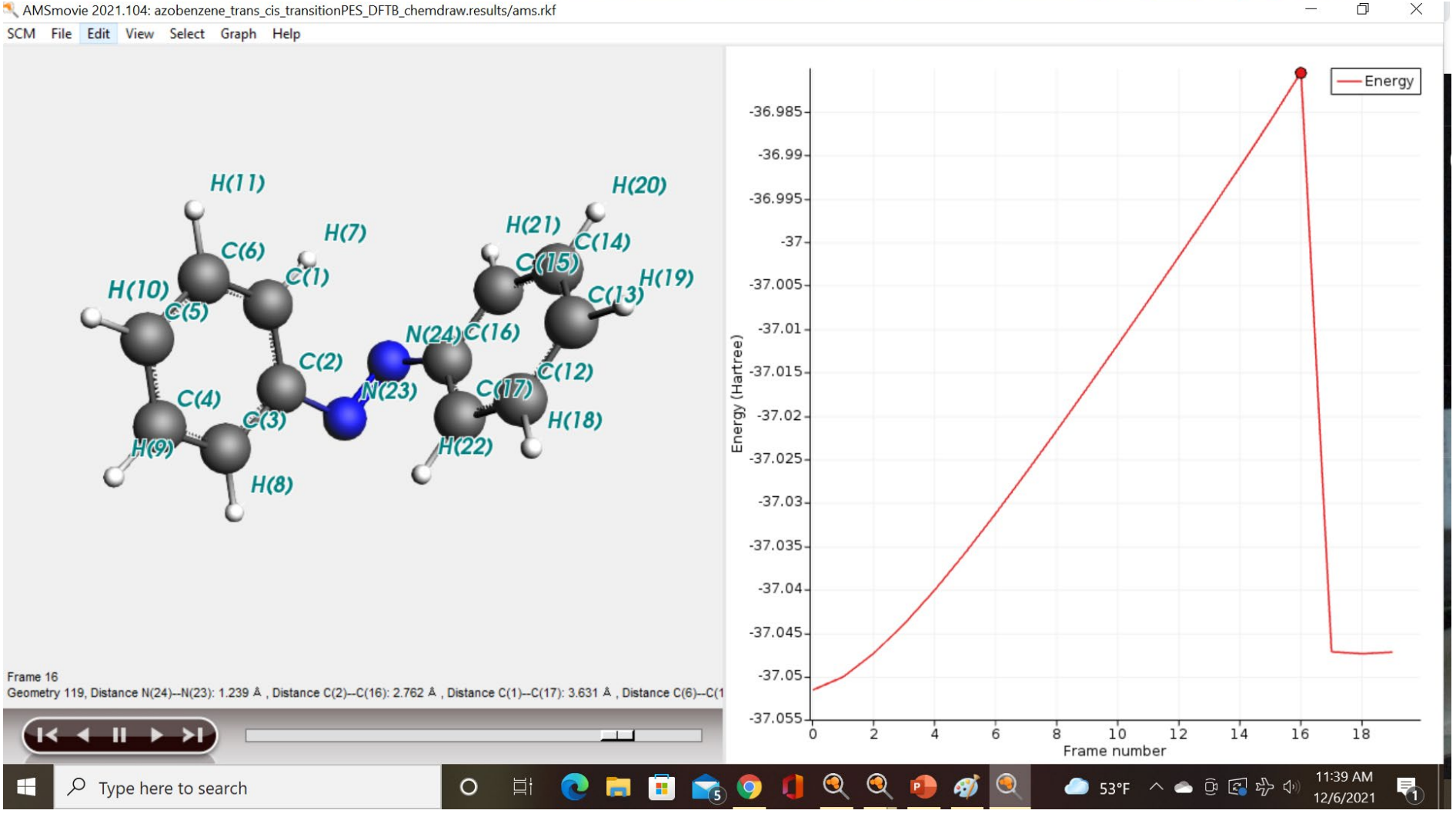


DRAFT

Evaluate options for quantum coding via vibrational modes with UV, IR or Raman and resulting spectra as representation of quantum states.

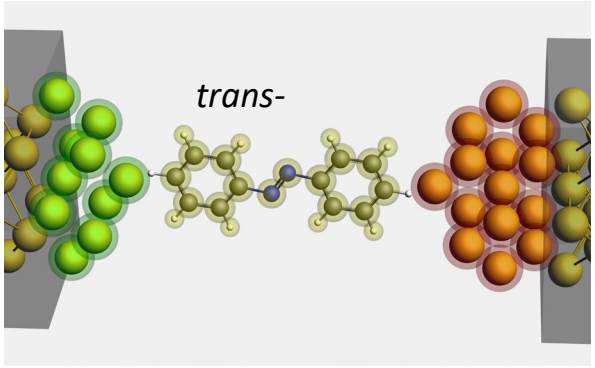
1.https://www.scm.com/doc/AMS/Vibrational_Spectroscopy.html#irfrequencies
2.Petrone, Alessio, et al. "Ab Initio Excited-State Transient Raman Analysis." *The Journal of Physical Chemistry A* 121.20 (2017): 3958-3965.
3.Reimer, Jeffrey A. "Nuclear hyperpolarization in solids and the prospects for nuclear spintronics." Solid state nuclear magnetic resonance 37.1-2 (2010): 3-12
4.Goswami, Sreetosh, et al. "Robust resistive memory devices using solution-processable metal-coordinated azo aromatics." Nature materials 16.12 (2017): 1216-1224.
5.Jacquemin, Denis, et al. "Extensive TD-DFT benchmark: singlet-excited states of organic molecules." Journal of Chemical Theory and Computation 5.9 (2009): 2420-2435.

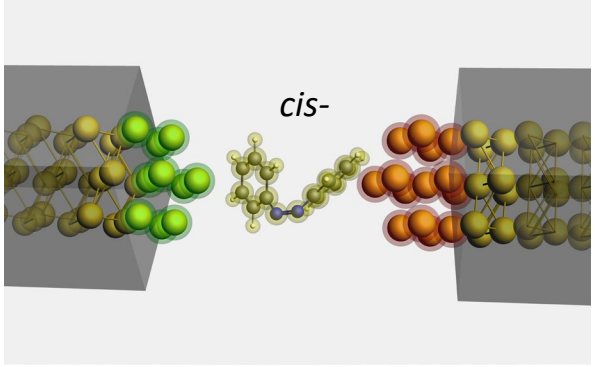
Used the Potential Energy Scan procedures to find the transition point

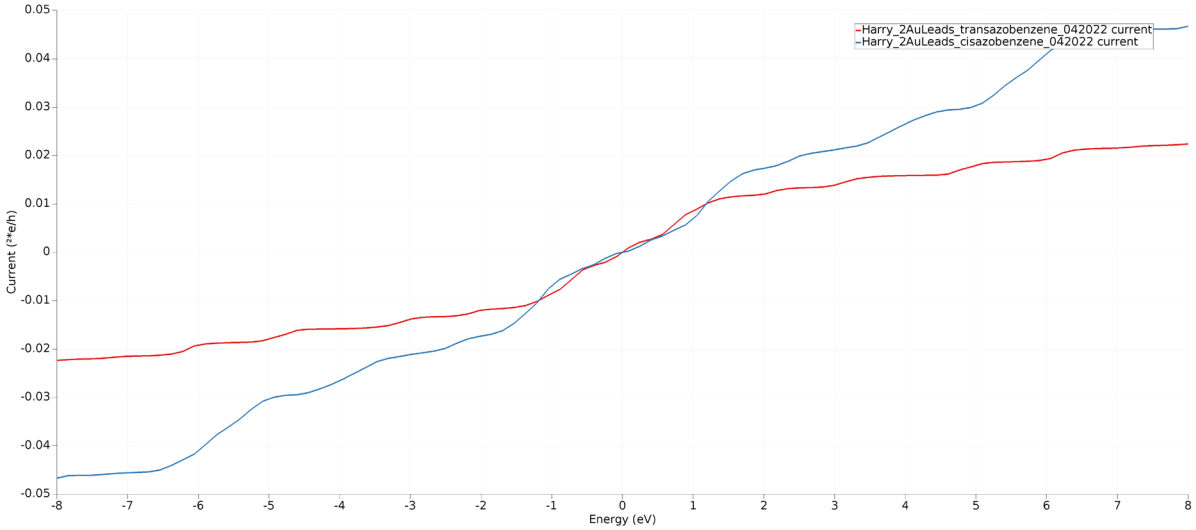


Electronic transport through azobenzene. Single molecule circuit simulation

- We are looking at transport properties through, *trans-*, *cis-*, and transition state
- There is much refinement to be done on these models. And deep dives into the practical interpretation of the simulation results are to be conducted
- Additional work is planned to investigate electron and hole mobilities
- We're investigating read out options of either an electronic or spectroscopic nature, probably Raman

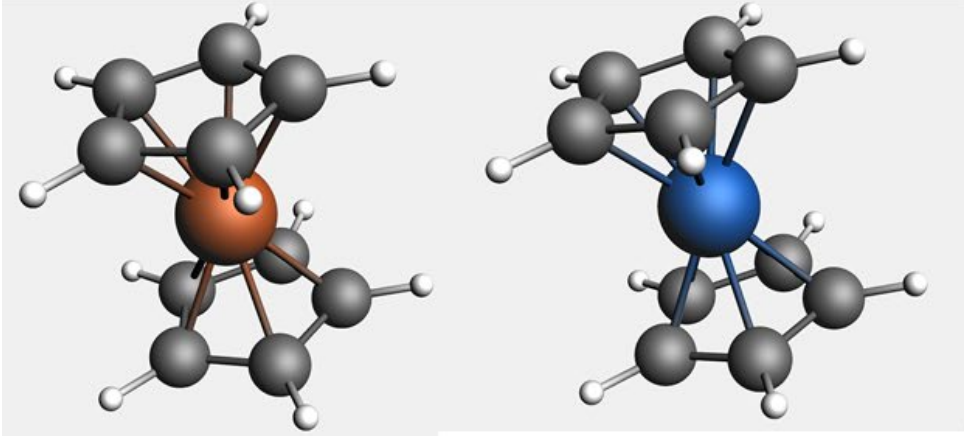




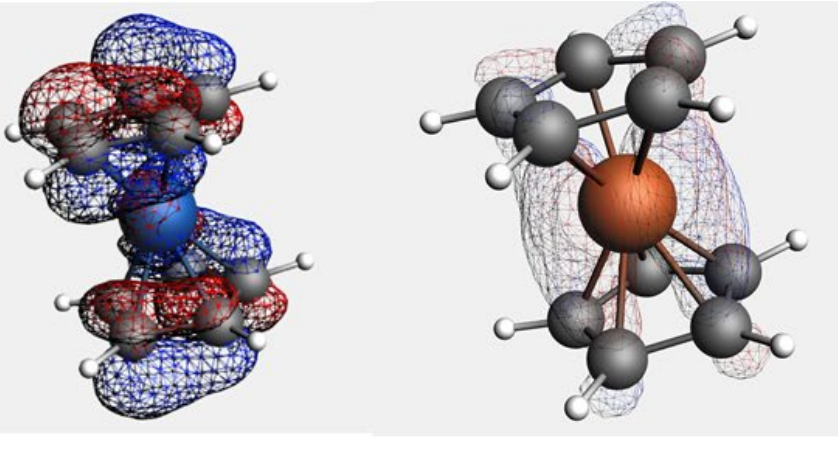


Spin transport through metal cyclopentadienyls

- We are investigating the use metal cyclopentadienyls such as ferrocene and cobaltocene for quantum spin transport
- These molecules are exceeding difficult to model correctly. The highly delocalized nature of the central transitional metal atom to the cyclopentadienyl rings must be accounted for in evaluating the simulation options and results.
- Ferrocene, Cobaltocene and their derivatives are under evaluation with considerations for Aufbau and non-Aufbau[6] solutions, staggered, eclipsed and skewed configurations, with each constituting a separate symmetry basis



Ground state, Ferrocene and Cobaltocene eclipsed configuration



Cobaltocene and Ferrocene HOMO occupations

6. Ghosh, Sayantani, et al. "Entangled quantum state of magnetic dipoles." Nature 425.6953 (2003): 48-51

Sol-gels

- We have our quantum materials, how are we going to implement them in a practical application?
- Dr. Preston and I are investigating doping silica sol-gel as a substrate.
- We specialize in low temperature synthesis of silica sol-gel and doping them for novel applications [8,9]
- Next applications are quantum dots



The sol-gel synthesis process involves acid catalyzed hydrolysis and condensation resulting in polymerization and shrinkage over time. Patience is required to synthesize monoliths.



Mid-cure blank silica sol-gel



Mid-cure azobenzene doped Sol-gel. Note the two phases



3 months later Shrinkage into a Single doped monolith

8. US 6,445,861 B1, 9/3/2002, Sol-Gel Processing To Form Doped, Sol-Gel Monoliths Inside Hollow Core Optical Fiber And Sol-Gel Core Fiber Devices Made Thereby. Harry Shaw, Melanie Ott, Michele Manuel (U of Florida)

9. Preston, Deborah, Jean Claude Pouxviel, Thomas Novinson, William C. Kaska, Bruce Dunn, and Jeffrey I. Zink. "Photochromism of spiropyrans in aluminosilicate gels." Journal of Physical Chemistry 94, no. 10 (1990): 4167-4172.

Conclusion

- Looking forward to continuing our successful relationship and investigating new problems.
- I would like to acknowledge the support of the NASA/GSFC ACCESS Project, Exploration and Space Communications Division and the NASA HQ Space Communications and Navigation (SCaN) Program Office.