# At and Across the Border:

# Understanding Chemistry with Computers

Zhenfei Liu

Department of Chemistry


November 15, 2023 @ Academy of Scholars Monthly Meeting

# How I Came to Wayne State


Peking University, Beijing, China
2003 - 2007

B.S. in **Chemistry**


University of California, Irvine
2007 - 2012

Ph.D. in Theoretical **Chemistry**

Working on Condensed Matter **Physics**



University of California, Berkeley
Lawrence Berkeley National Laboratory
2012 - 2018

Postdoc in **Physics**/Materials Sciences

Working on Computational Materials **Chemistry**


Assistant Professor
Since 2018

Department of **Chemistry**

# My Original Interest to Be a Chemist…

"If you wish to understand the fragrance of the rose or the tenacity of the oak; if you are not satisfied until you know the secret paths by which the sunshine and the air achieve these wonders; if you wish to see the pattern which underlies one large field of human experience and human measurement, then **take up Chemistry**."

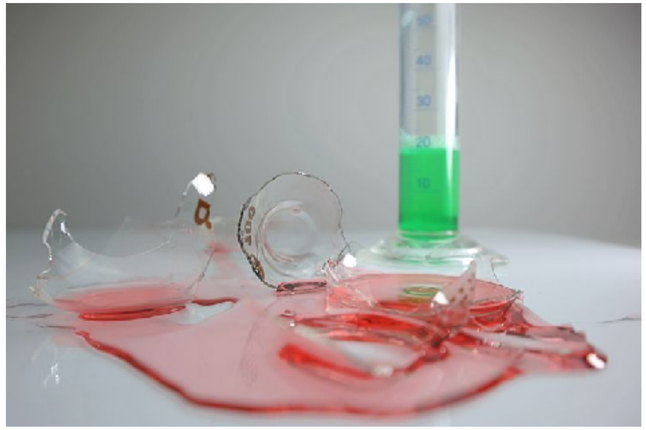
— Charles A. Coulson, 1973

Professor of Chemistry, Oxford University

Then I chose Chemistry as my major in college,

and did lots of

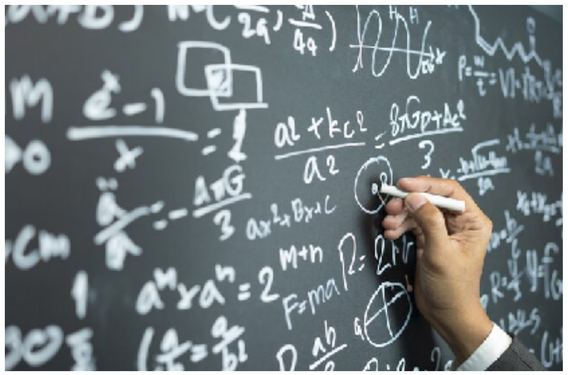
and

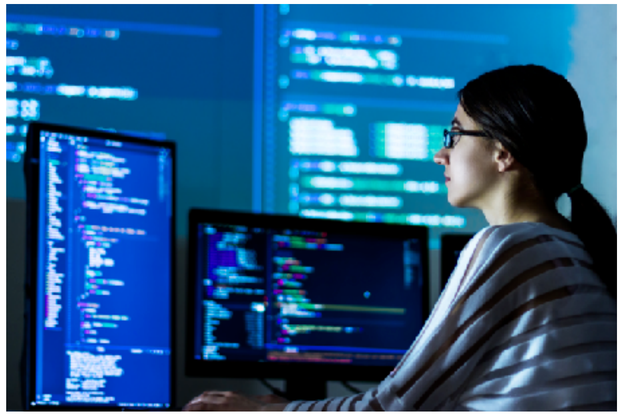


# Exposure and Engagement with Computational Chemistry

Conventional
"**Chemistry**"

Mathematics (and **Physics**)

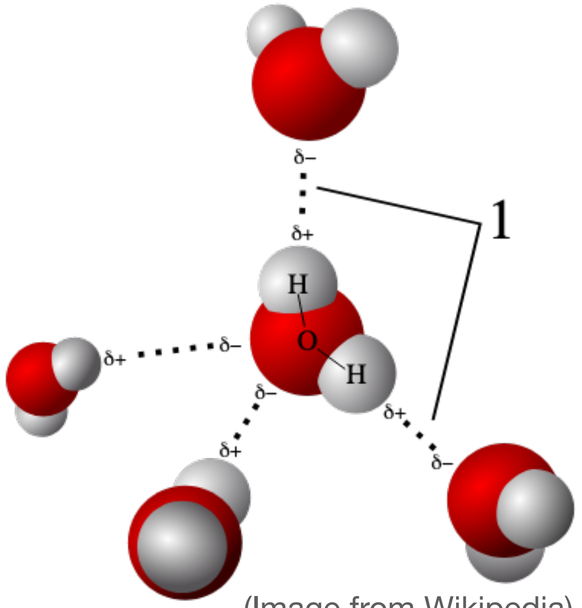




**Computer Science**

Circa 2005 (my junior year in college),
I did some undergraduate research:

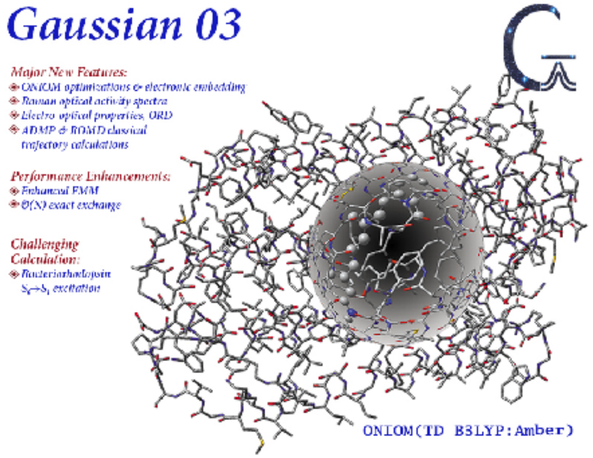
2003-2007




(Image from Wikipedia)

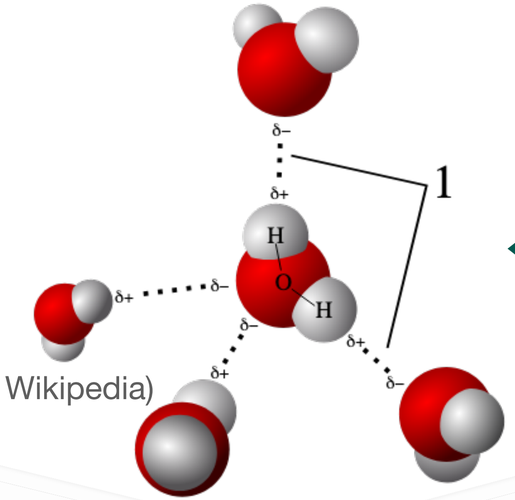
Used existing **computer** programs to solve **quantum mechanical** equations, to understand **chemistry**.

Berny Schlegel
(WSU Chemistry)



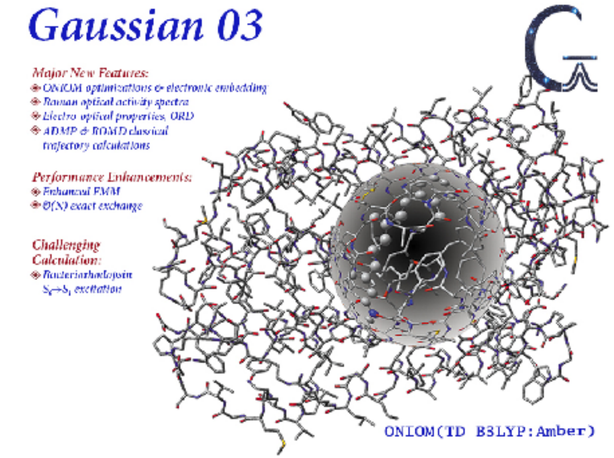
# Theoretical Chemistry, and Condensed Matter Physics

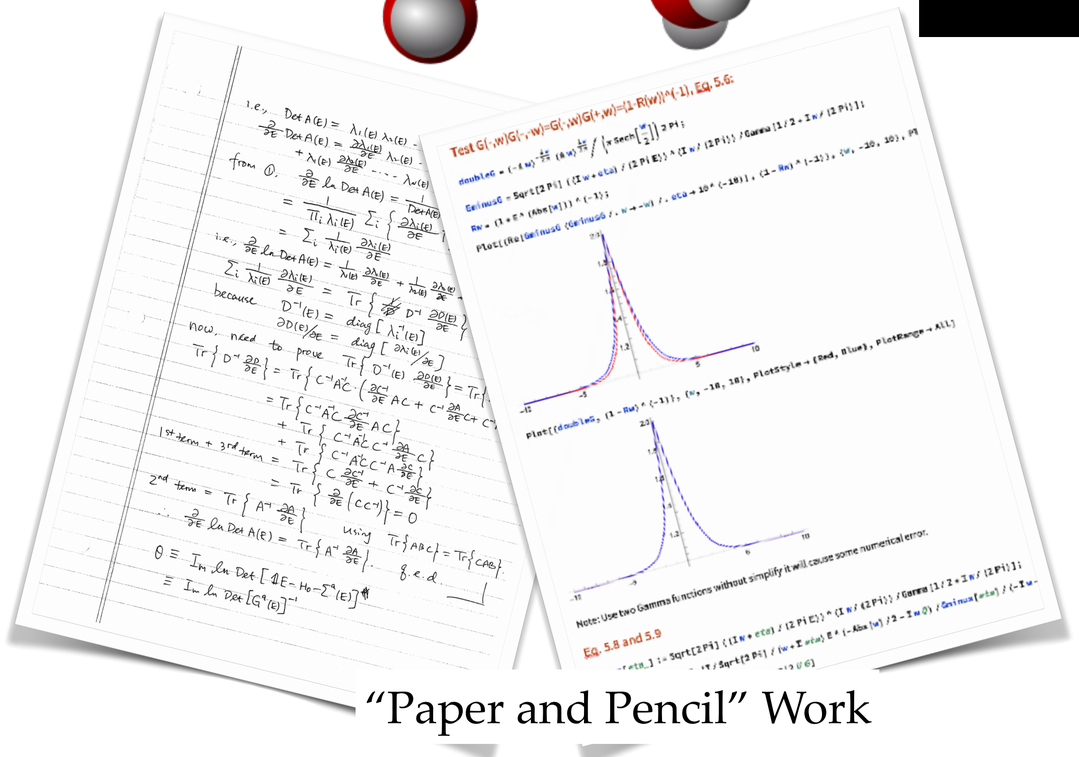
2007-2012



(Image from Wikipedia)

**What's in that black box?**



"Paper and Pencil" Work

All theoretical chemistry is really physics; and all theoretical chemists know it.

—Richard P. Feynman
(1918 - 1988)

(Image from nobelprize.org)

Nobel laureate, Physics 1965

# At the Border: Chemistry, Physics, and Computing

7/16/2013

$T(E) = \mathrm{tr}\{\Gamma_L(E)\, G(E)\, \Gamma_R(E)\, G^+(E)\}$

$\Sigma_L = \mathrm{Re}\Sigma_L(E) - \frac{i}{2}\Gamma_L(E)$ notice the

$\Sigma_L^+ = \mathrm{Re}\Sigma_L(E) + \frac{i}{2}\Gamma_L(E)$

$\Sigma_L - \Sigma_L^+ = -i\Gamma_L(E)$

$\Rightarrow \Gamma_L(E) = i\left(\Sigma_L(E) - \Sigma_L^+(E)\right)$

in transmission, F

$\begin{cases} U \leftarrow \Sigma_L - \Sigma_L^+ = -i\Gamma_L(E) \\ iU \leftarrow -i\Gamma_R(E) \end{cases}$

and used $U_{ij} = -U_{ji}^*$ (anti-Hermiti

later in the code, it does

$-[iU][GF]$

$\&\ -[U][GF]^+[iU][GF]$

which translates to $+\Gamma_L G^+ \Gamma_R G$ ✓

in my printing line. I print

GamR ← $-\mathrm{Im}(iU) = \Gamma_R(E)$ in eV

GamL ← $-\mathrm{Im}(U) = \Gamma_L(E)$ in eV

$G = \frac{1}{ES - H - \Sigma_L - \Sigma_R}$ in the code.

From equations to codes

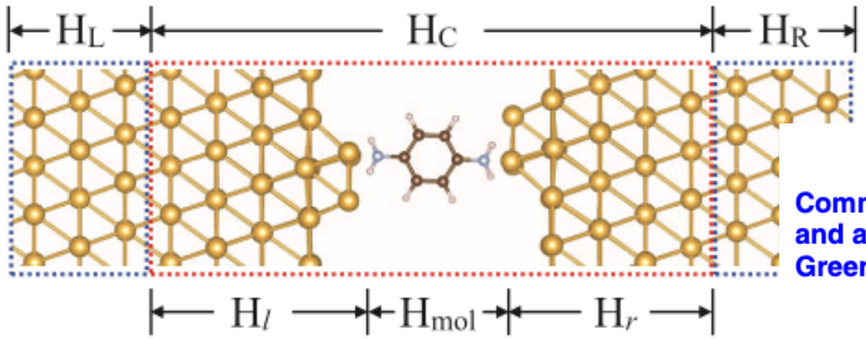
```
! debug Zhenfei Liu 08/02/2013

      IF (MOtrig) THEN
    do j = 0, nc-1
       jj=j+NGL
       do i = 0, nc-1
          ii=i+NGL
          Hc(i + j*nc) =
     .          Energy*Sbig(ii+jj*no) - Hdev(i + j*nc + 1)
     .        - SigmaR(i+nc*j) - SigmaL(i+nc*j)
! the +1 in Hdev is because Hdev's index goes from 1 to nc*nc
          GF(i+nc*j) = dcmplx(0d0,0d0)
       end do                        !i
       GF(j+nc*j) = dcmplx(1d0,0d0)
    end do                           !j

! debug done. (also the ELSE and END IF below)
```

siesta™

From codes to chemistry




My first exposure to **high-performance computing** (HPC)


NERSC

In service: 2010-2015

Ranked the world's #5 largest supercomputer, with 153,408 cores (Nov 2010)

THE JOURNAL OF CHEMICAL PHYSICS **141**, 131104 (2014)

**Communication: Energy-dependent resonance broadening in symmetric and asymmetric molecular junctions from an *ab initio* non-equilibrium Green's function approach**


Zhen-Fei Liu[1] and Jeffrey B. Neaton[1,2,3]

[1]*Molecular Foundry and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

[2]*Department of Physics, University of California, Berkeley, California 94720, USA*

[3]*Kavli Energy Nanosciences Institute at Berkeley, Berkeley, California 94720, USA*



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UNIVERSITY OF CALIFORNIA · BERKELEY · 1868

BERKELEY LAB

2012-2018

# Overview of My Research Program at WSU

$$\frac{\alpha + \beta \mathrm{erf}(\gamma|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|}$$

Density Functional Theory

$$\Sigma = iGW \qquad v_{\mathrm{xc}}(\mathbf{r}) = \frac{\delta E_{\mathrm{xc}}}{\delta \rho(\mathbf{r})}$$

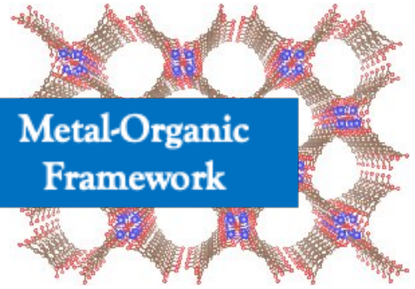
$$[\mathbf{H} + \mathbf{\Sigma}]\,|\psi\rangle = [\epsilon - i\Gamma/2]\,|\psi\rangle$$

Many-Body Perturbation Theory

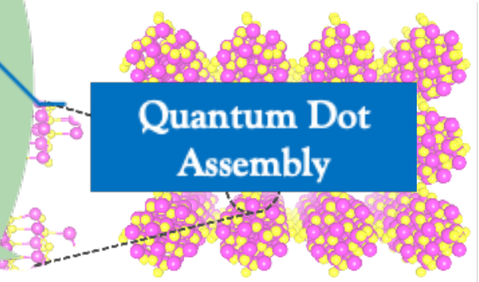
Electronic Structure Methods

Complex Materials and Interfaces

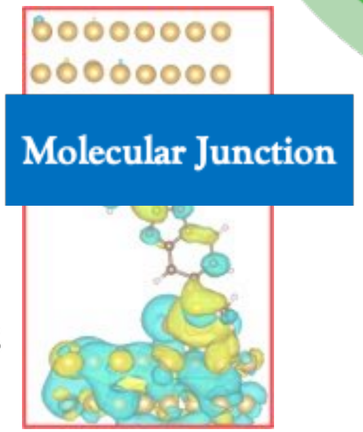
Charge Dynamics



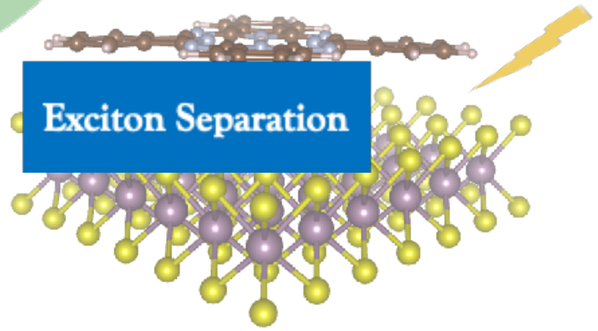
Metal-Organic Framework



Quantum Dot Assembly



Molecular Junction



Exciton Separation

Since August 2018

Collaboration with Experiments

Energy/Nano-Material Related Applications

New First-Principles Calculations

Implementations

New Theories and Methods

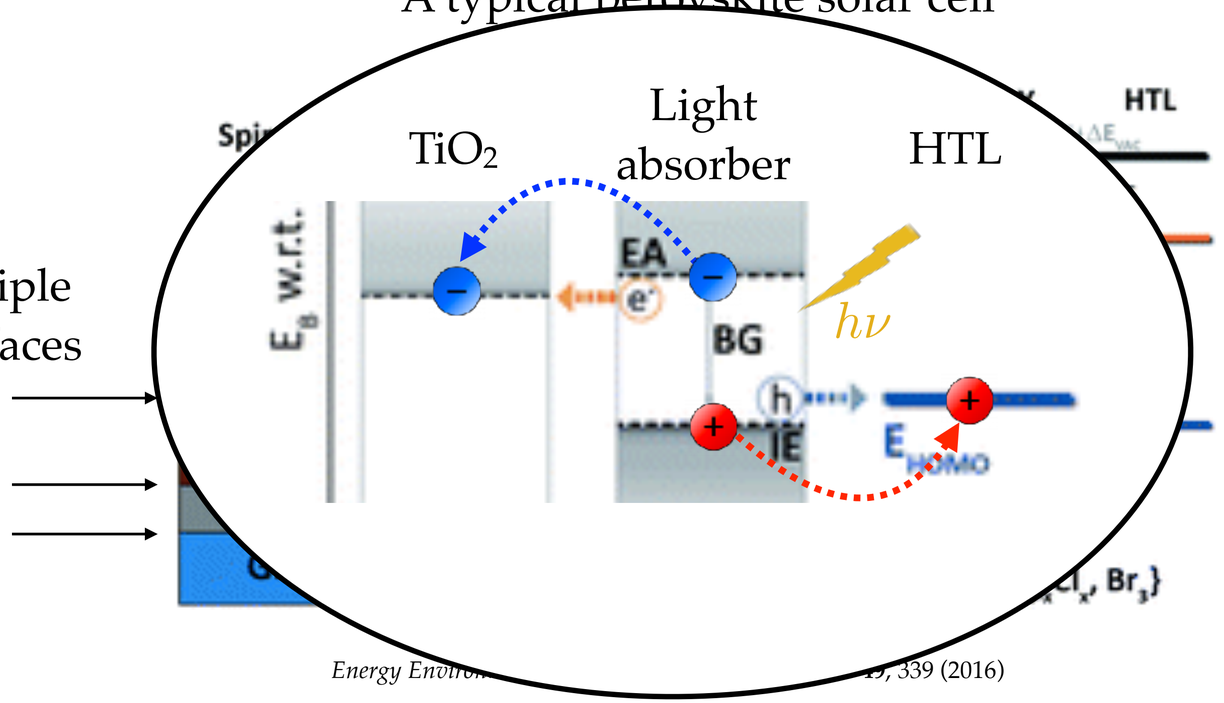
# Across the Border: Interfaces (Molecule/Substrate)

***"The interface is the device."***

— H. Kroemer, 2000 Nobel Lecture

A typical perovskite solar cell

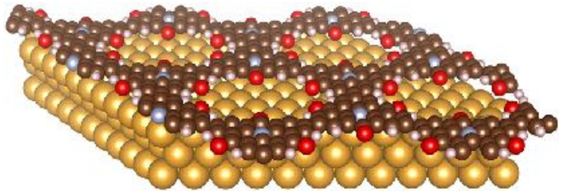
Multiple Interfaces




*Energy Environ.* 9, 339 (2016)

We develop new **accurate** and **efficient** computational methods, based on **quantum mechanics** + the **chemical structure**, using **high-performance computing** (HPC), to validate, understand, and predict experiments.

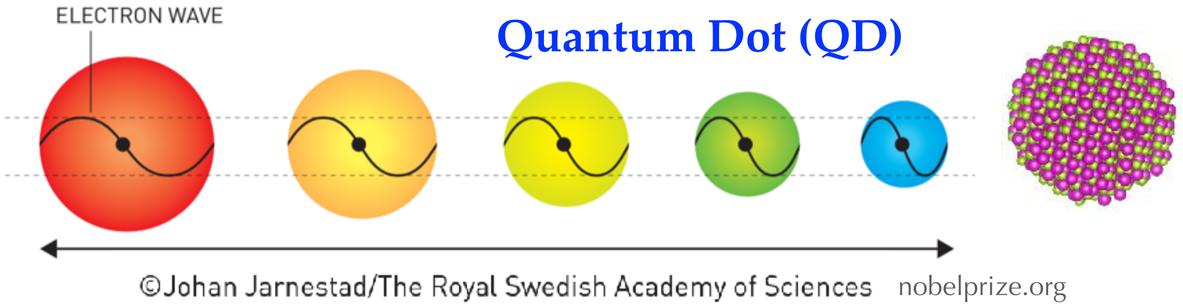
(Image from LLNL)

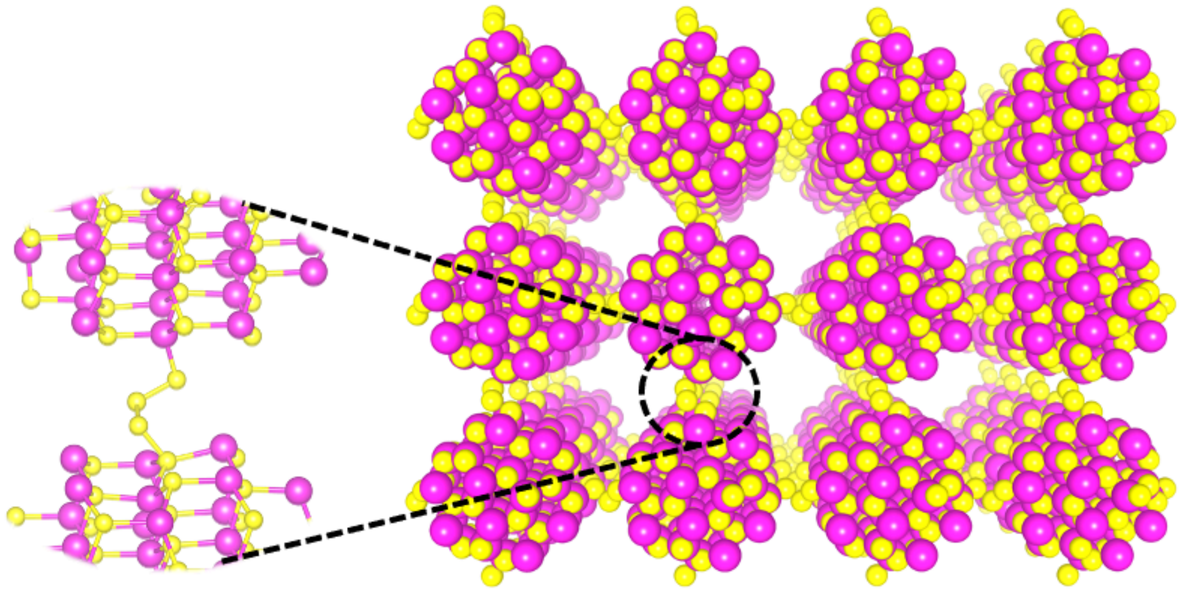


DMR-2044552
(CAREER)

# Complex Materials

## Quantum Dot (QD)

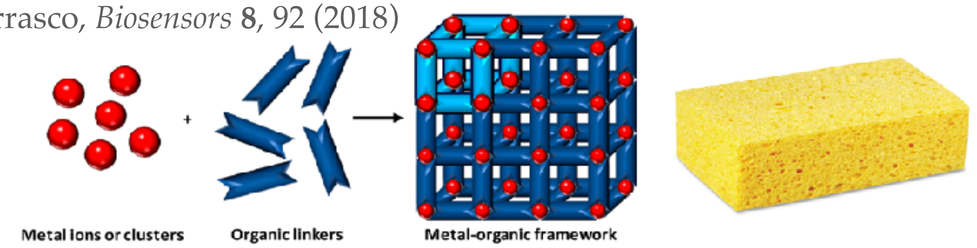


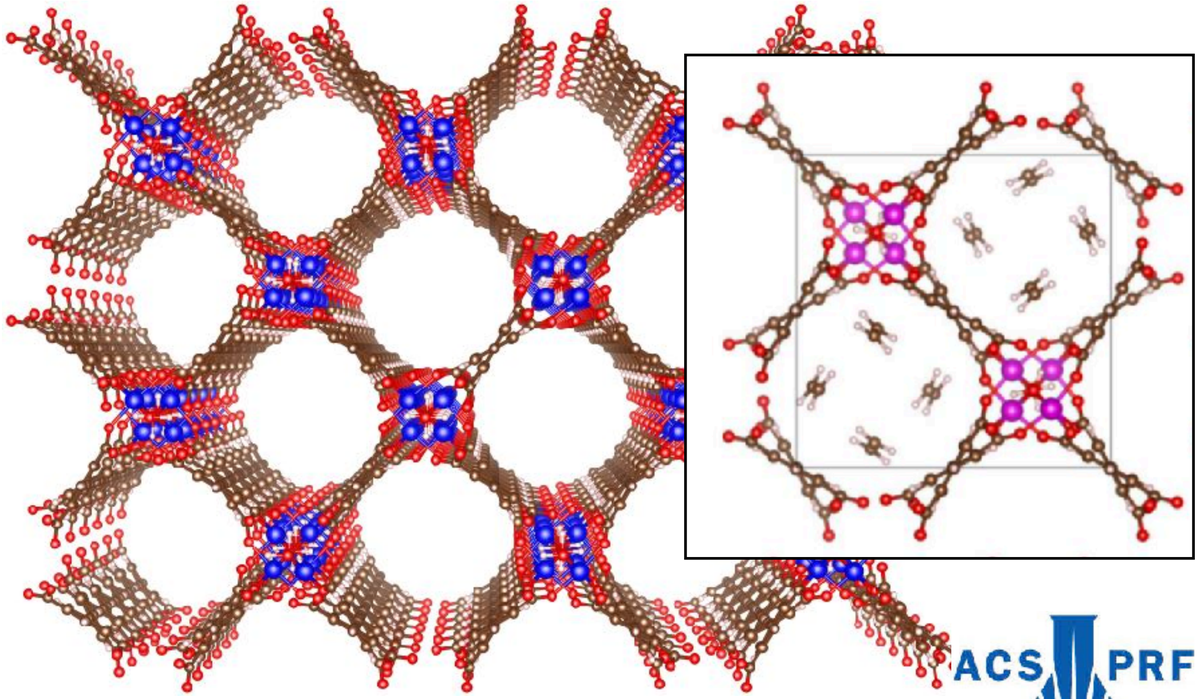



S. Aryal, J. Frimpong, and Z.-F. Liu,
*J. Phys. Chem. Lett.* **13**, 10153 (2022)

## Metal-Organic Framework (MOF)

S. Carrasco, *Biosensors* **8**, 92 (2018)



T. Quainoo, S. N. Lavan, and Z.-F. Liu,
*J. Mater. Res.* **37**, 334 (2022).

DE-SC0023324

Stephanie Brock (WSU Chemistry)

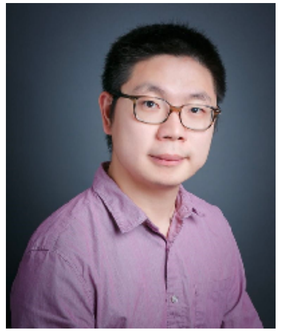
61117-DNI10

# Ongoing Collaborations

WSU Chemistry

Stephanie Brock (Academy Member)

Aaron Rury (Jr. Faculty Award 2023)



Long Luo (Jr. Faculty Award 2021)

Jeff Neaton
(Berkeley)

Michael Inkpen
(USC)

Pierre Darancet
(Argonne)

Li Yang
(WashU)

Jier Huang
(Boston College)

# And More



Sivan Refaely-Abramson
(Weizmann Institute)

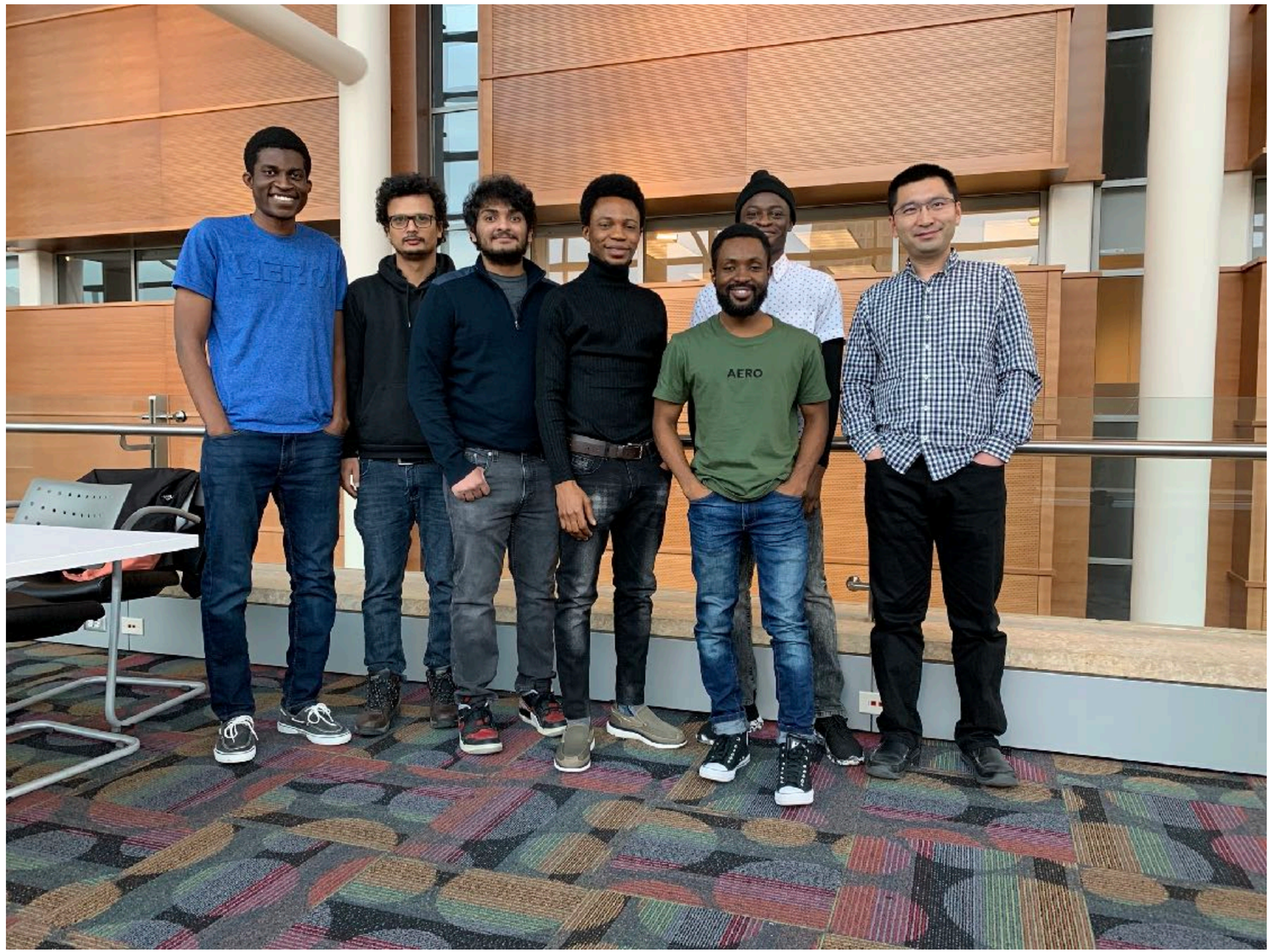


Haixing Li
(CityU of HK)



Zhongyue Zhang
(Kumamoto U)

# Acknowledgements




**Current:**

Joseph Frimpong
Timothy Quainoo
Amos Afugu
Moses Adeyemo
Gyanu Kafle (postdoc)
Rishat Dilmurat (postdoc)
Tejas Karun (undergrad)

**Alumni:**

Dr. Sandip Aryal (postdoc)
Dr. Naseem Ud Din (postdoc)
Dr. Sydney Lavan (PhD)
Dr. Olugbenga Adeniran (PhD)
Lyric Elliott (undergrad)

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