University of Kentucky

Developments towards Machine-driven Design & Discovery of Organic Semiconductors

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Department of Chemistry &
Center for Applied Energy Research (CAER)

7th Annual Commonwealth Computational Summit
on AI in Education, Medicine, and Research

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Sashen Ruhunage

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Sean Ryno (Postdoc)
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Corrine Elliott (Undergraduate)
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Asare Nkansah (Undergraduate)
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Andrew Smith (Undergraduate)
William Allen Smith (Undergraduate)
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Brandyn Thompson (Undergraduate)
Nicholas Telesz (Undergraduate)
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Franklin Marrs (High School)
Ally Watrous (High School)

Arnold and Mabel Beckman Foundation
ACCESS Advancing Innovation
Award No. 1627428
Cooperative Agreement No. 2019574
NSF

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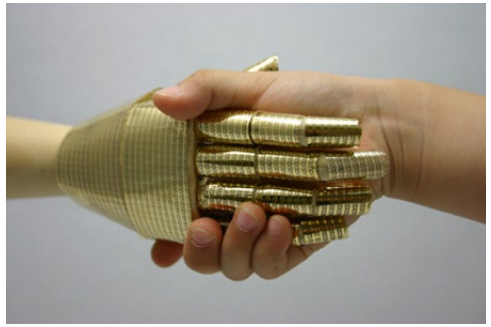
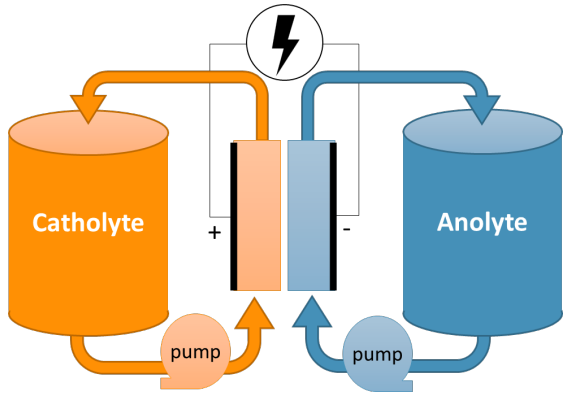
Organic materials for electronics, power generation, and storage

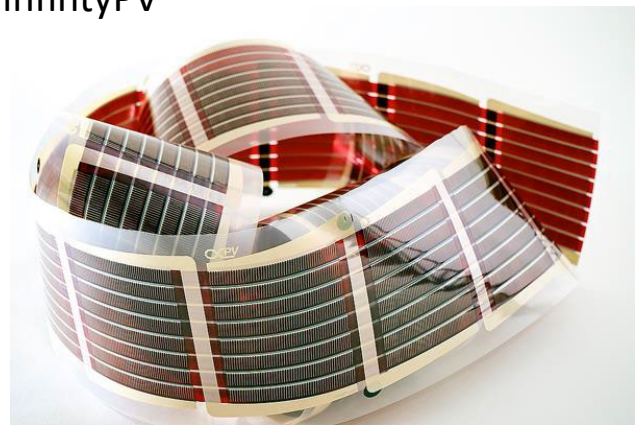
Samsung



University of Tokyo

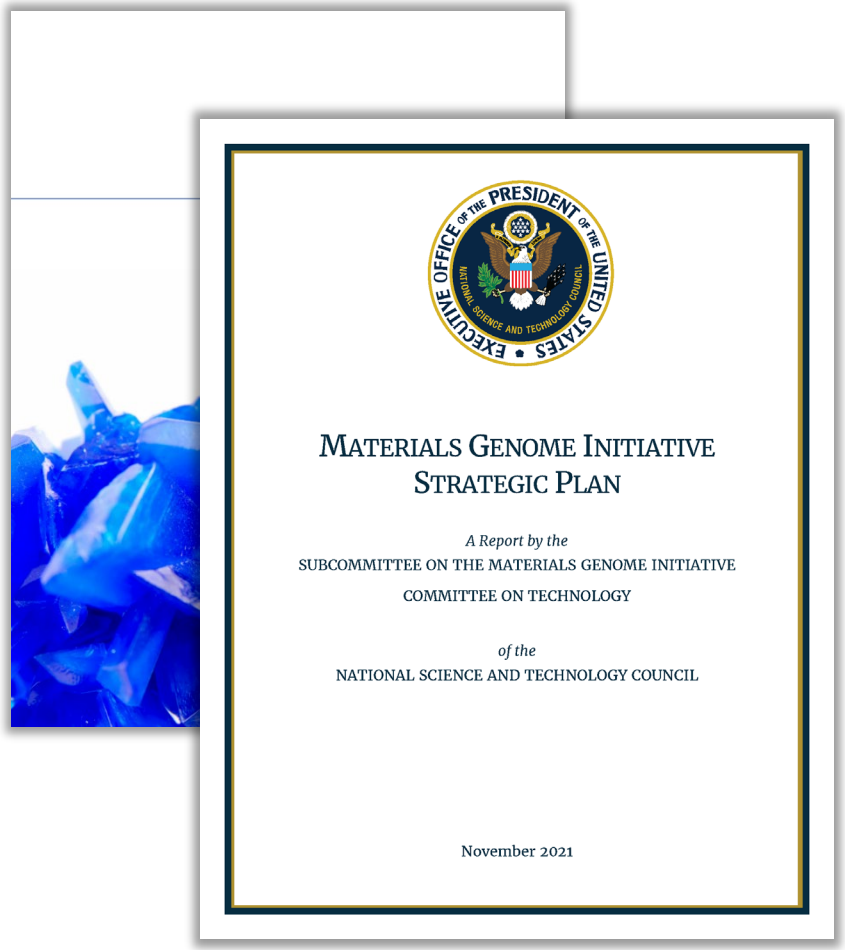


infintyPV



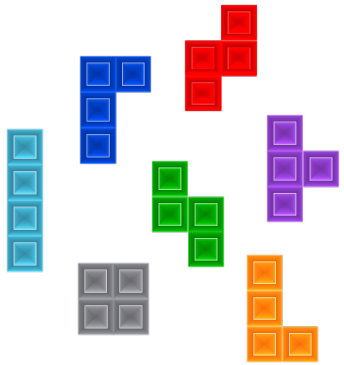
LG

Convergence of materials science, data science, and robotics



Launched in 2011 to accelerate the discovery, design, development, and deployment of new materials, at a fraction of the cost, by harnessing the power of data and computational tools in concert with experiment.

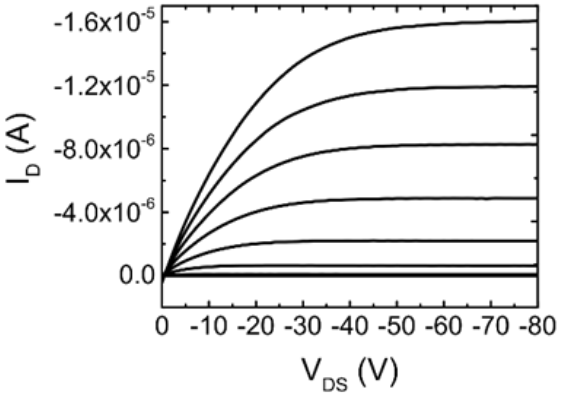
in silico design



A vast chemical space of organic chromophores

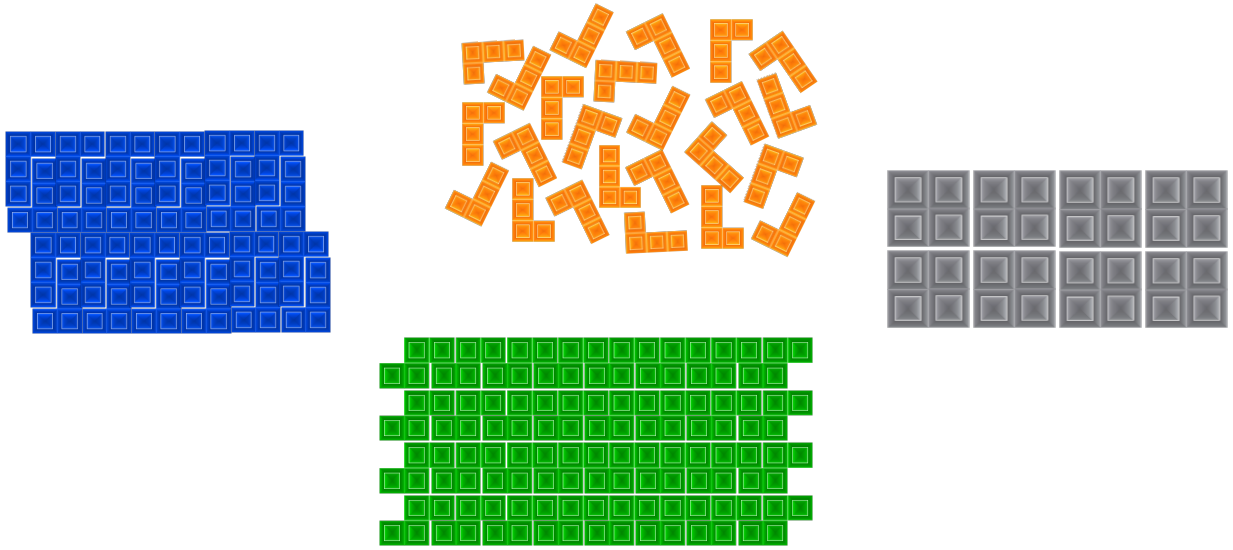
BUILDING BLOCK

?



A. Zeidell *et al*. Chem. Mater. (2019) 31, 6962.

RESPONSE



STRUCTURE & SYNTHESIS PREDICTION

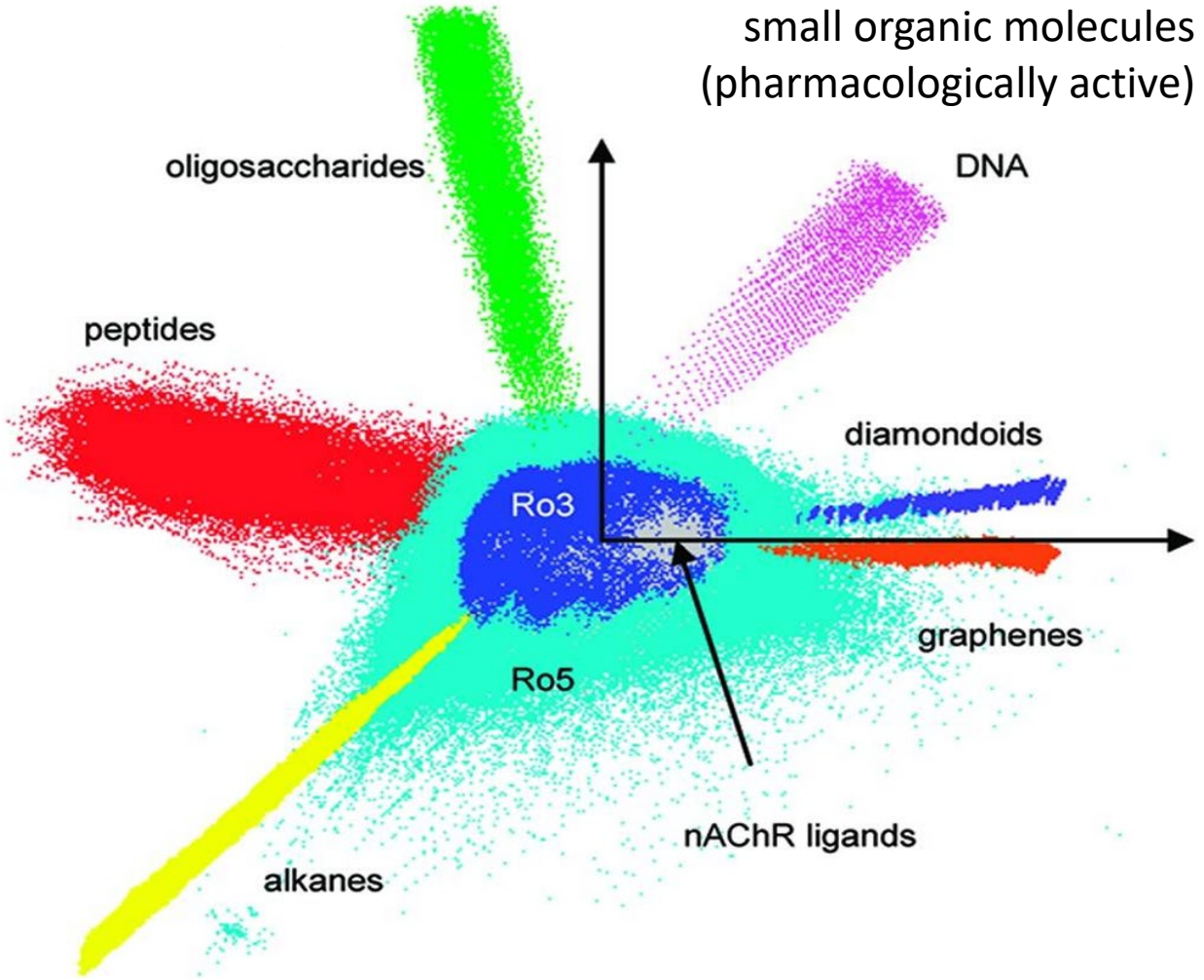
PROPERTY PREDICTION

Adapted from Q. Ai

The chemical space for organic materials is large

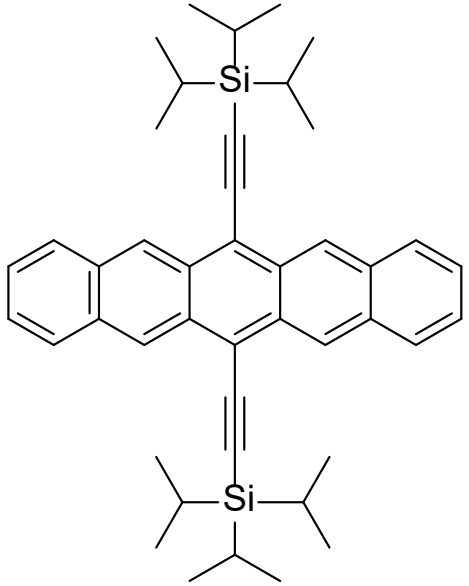
All things considered…chemical space is massive!

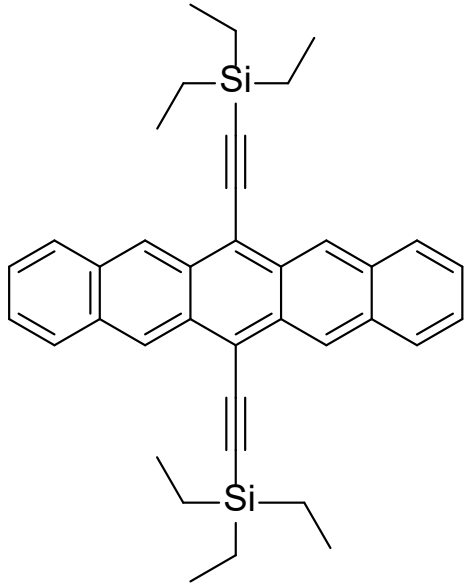
- 10^{18} grains of sand on Earth
- 10^{23} stars in the visible sky
- 10^{60} small organic molecules (pharmacologically active)

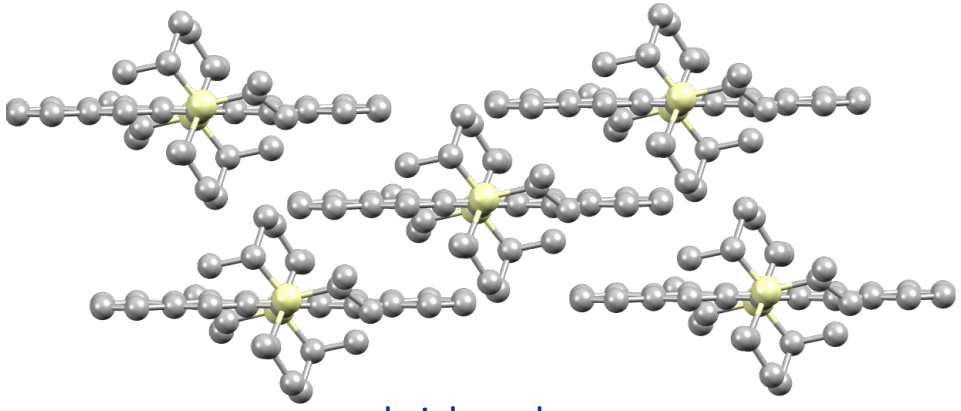


P. Kirkpatrick & C. Ellis, Nature 2004, 432, 823;
C. Lipinski & Andrew Hopkins, Nature 2004, 432, 855;
J.L. Reymond & M. Awale, ACS Chem Neurosci. 2012, 3, 649.

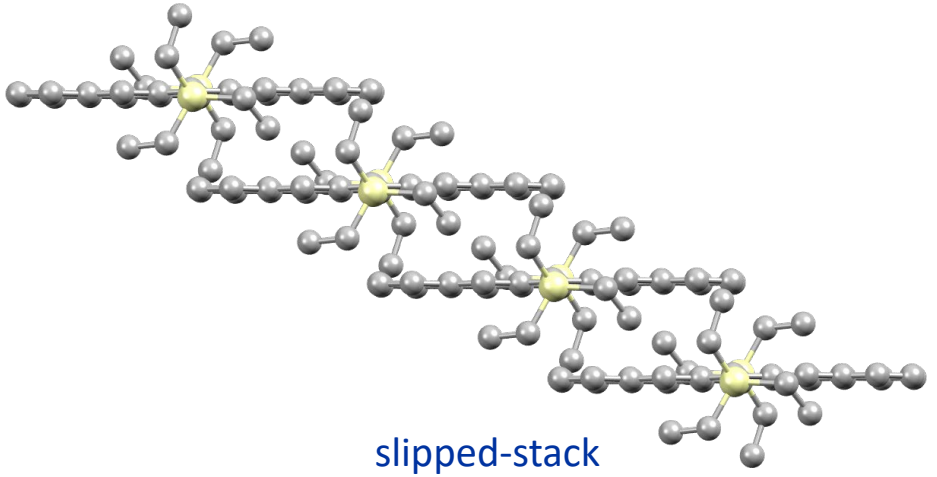
TAS-Pentacene: TIPS v. TES Substituents







brickwork

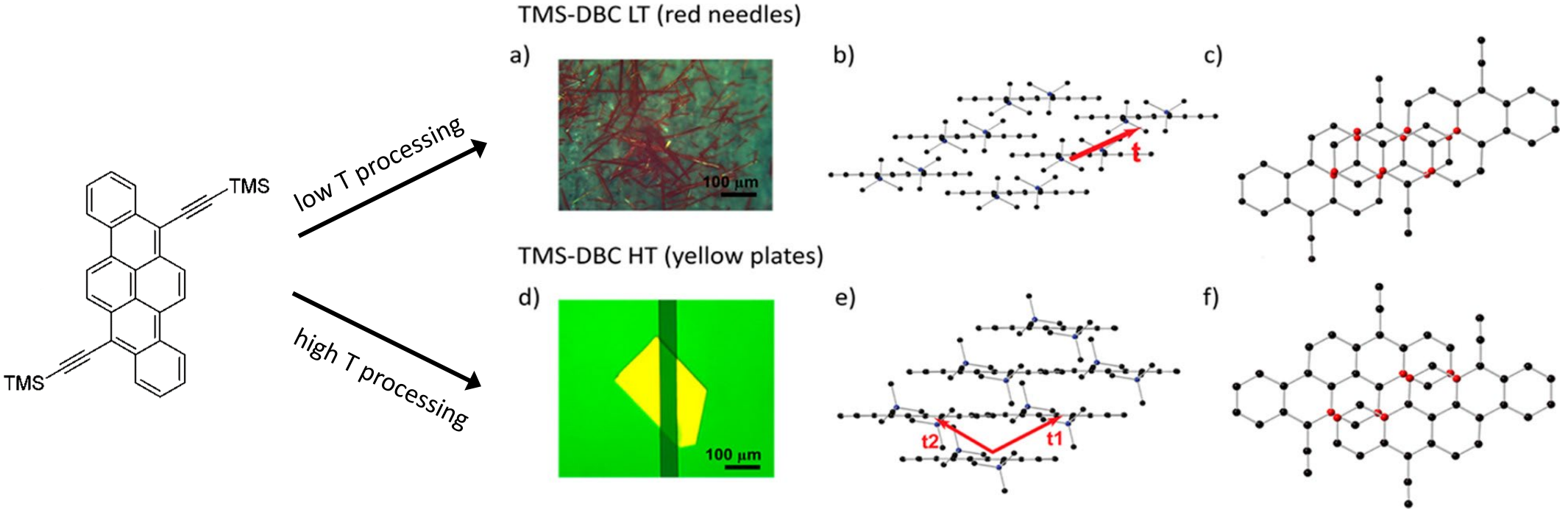


slipped-stack

K.J. Thorley, T.W. Finn, K. Jarolimek, J.E. Anthony, C. Risko, Chem. Mater. 2017, 29, 2502.

Polymorphs and processing

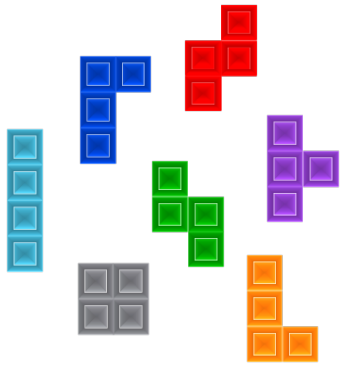
$$\mu_h = 0.028\ cm^2V^{-1}\ s^{-1}$$



$$\mu_h = 2.1\ cm^2V^{-1}\ s^{-1}$$

L. Stevens, V. Coropceanu, O.D. Jurchescu, G.E. Collis, et al. Chem. Mater. 2015, 27, 112.

in silico design



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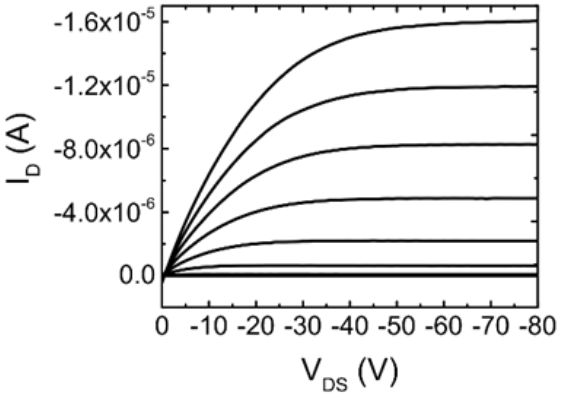

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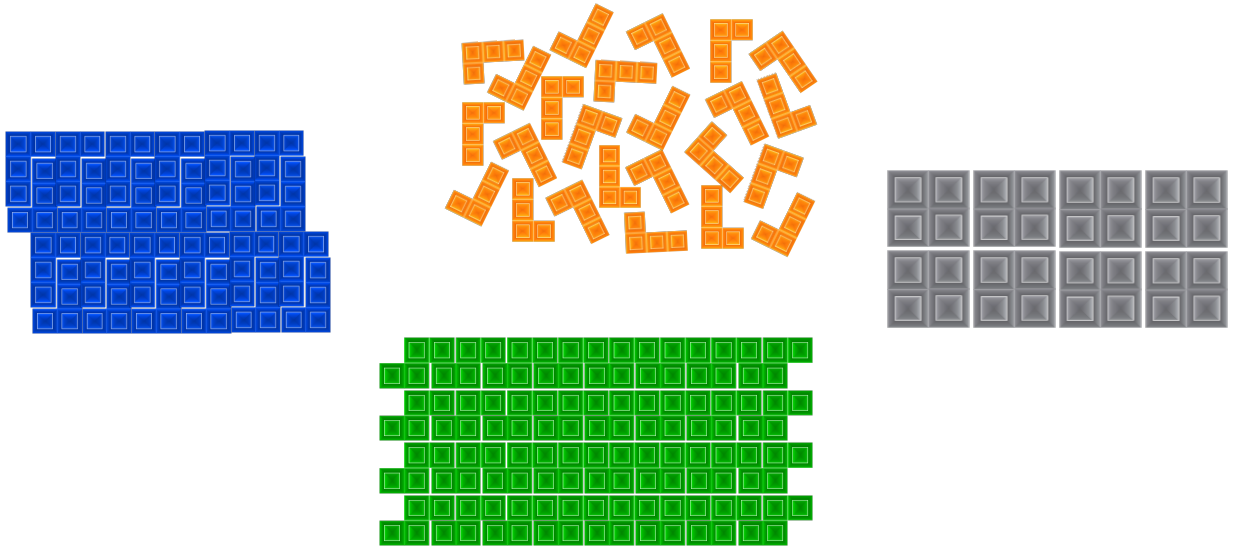


STRUCTURE & SYNTHESIS PREDICTION

PROPERTY PREDICTION

Adapted from Q. Ai

Organic Crystals in Electronic and Light Oriented Technologies (OCELOT)

Open Access
infrastructure

oscar.as.uky.edu

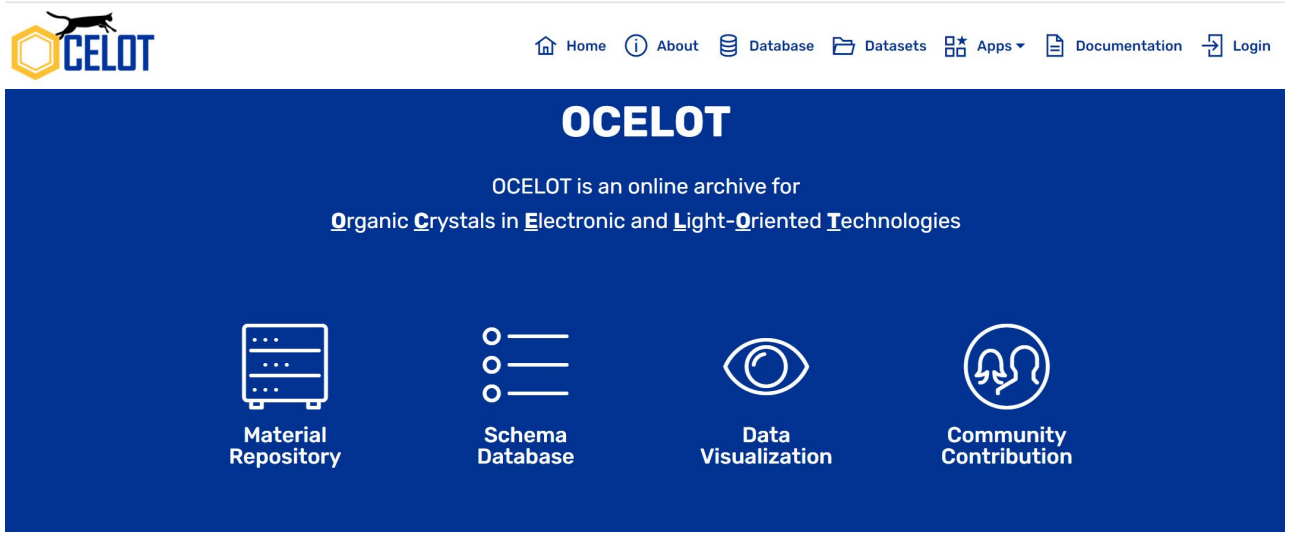
56k crystals
47k molecules
38k π-conjugated chromophores

OCELOT

- A Python tool kit for data analysis
- High-throughput computational workflows
- User-friendly web user interface
- Access to machine learning predictions

Q. Ai, V. Bhat, S.M. Ryno, L.Y. Huang, A. Smith, P. Sornberger, R. Duke, S. Goodlett, C. Risko with J.E. Anthony & M.M. Haley;
Q. Ai, V. Bhat, S.M. Ryno, K. Jarolimek, P. Sornberger, A. Smith, M.M. Haley, J.E. Anthony & C. Risko, J. Chem. Phys. 2021, 154, 174705.

Organic Crystals in Electronic and Light Oriented Technologies (OCELOT)



Database Statistics

as of March 26, 2023, 8:32 p.m. (US/Eastern)

56296
Crystals
with structure determined experimentally by XRD/ND.

2000
Descriptors
with optoelectronic properties computed via state-of-art DFT.

10e6
Core hours
have been used to fill this database.

1853
Researchers
are using this for their research since Feb 14, 2021.

Since 2021 launch →

- 2700+ users
  - 170 registered users that can access tools
- 48 countries

Tools →

- XtalTransform

generate crystal structures based on templates

- FilmDiffract

generate X-ray thin film diffraction patterns based on bulk crystal structures

- RepCheck

reproducibility checker for experimental procedures

- OCELOT-ML

open-access portal to ML models

Q. Ai, V. Bhat, S.M. Ryno, L.Y. Huang, A. Smith, P. Sornberger, R. Duke, S. Goodlett, C. Risko with J.E. Anthony & M.M. Haley;
Q. Ai, V. Bhat, S.M. Ryno, K. Jarolimek, P. Sornberger, A. Smith, M.M. Haley, J.E. Anthony & C. Risko, J. Chem. Phys. 2021, 154, 174705.

Organic Crystals in Electronic and Light Oriented Technologies (OCELOT)

Chromophore

[IP-tuned LC-ωHPBE/Def2SVP]

Electronic structure - Ground State

Vertical IP	6.365 eV
Adiabatic IP	6.321 eV
Vertical EA	-1.932 eV
Adiabatic EA	-1.959 eV
Hole reorganization energy	0.092 eV
Electron reorganization energy	0.130 eV
HOMO-LUMO gap	4.466 eV
Tuned w	0.178

Electronic structure - Excited State

Singlet transition (S0->S1)	1.865 eV
Triplet transition(S0->T1)	0.163 eV
Adiabatic (S0->T1)	0.616 eV

OCELOT Application Programming Interface (API)
https://github.io/ocelot_api

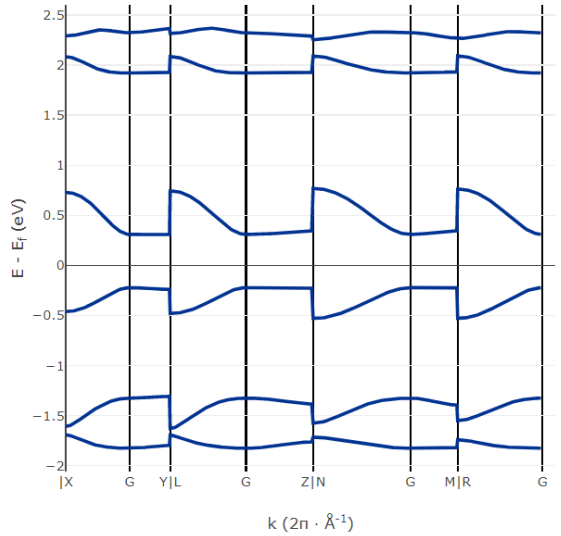
Crystal

[Periodic DFT | PBE with projector-augmented wave method]

Electronic Structure

Fermi energy	0.847 eV
Bandgap	0.532 eV
Bandgap is direct	False
Fitted bandgap	1.637 eV
Valence band maximum	0.623 eV
Hole line effective mass	0.889
Hole effective mass tensor	[1.189, 6.763, 314.853]
Valence band dispersion	0.239 ev
Conduction band minimum	1.155 eV
Electron line effective mass	0.480
Electron effective mass tensor	[83.278, 3.901, 0.624]
Conduction band dispersion	0.420 ev
Line segment	X-G:[0. -0.5 0.]-[0. 0. 0.]
K-point	[0.0, -0.067, 0.0]

Bandstructure

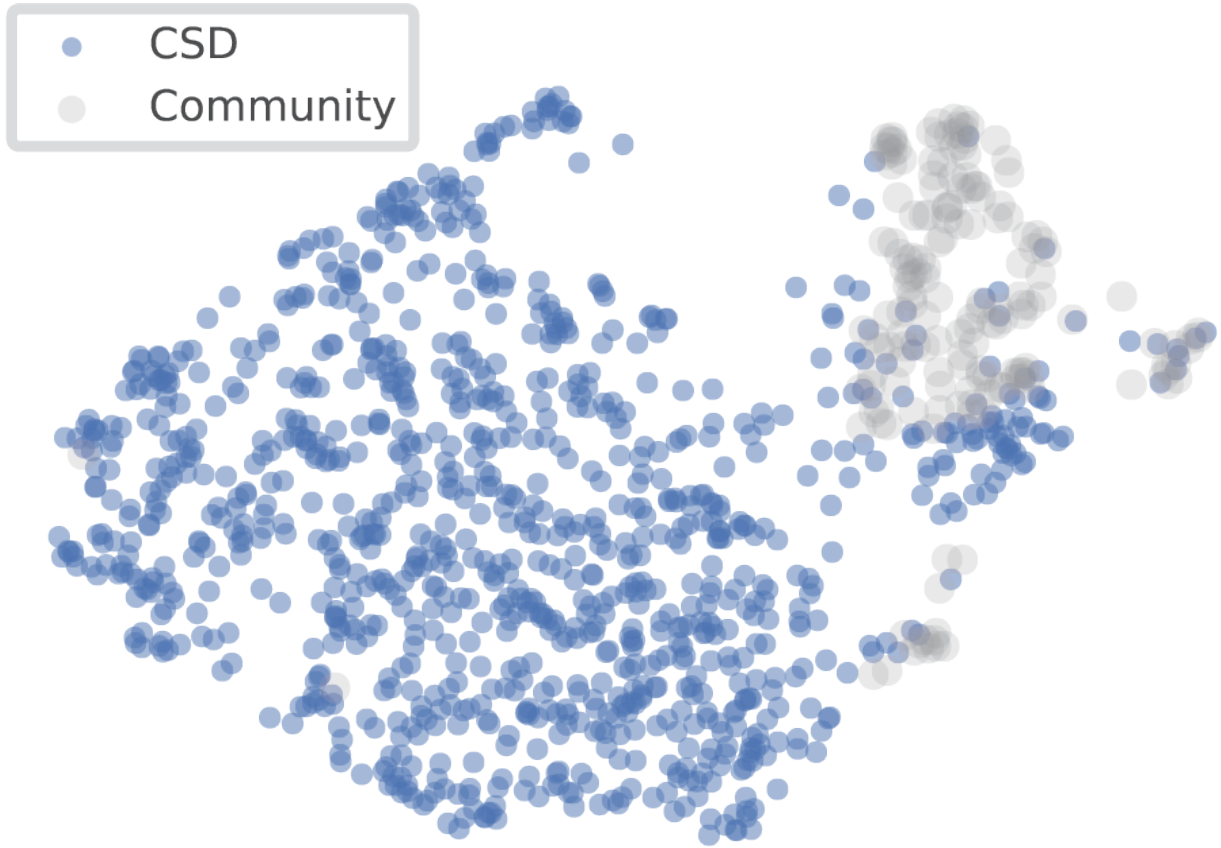


Percolation (DFT)

H-H coupling	0.059 eV
H-H dimer	**Long axis slip:** 6.29 Å **Short axis slip:** 0.93 Å **Vertical distance:** 3.39 Å
L-L coupling	0.113 eV
L-L dimer	**Long axis slip:** 6.29 Å **Short axis slip:** 0.93 Å **Vertical distance:** 3.39 Å

Data set includes “dark” structures

- OCELOT contains structures not reported elsewhere – ‘dark’ or missing structures from the literature



Q. Ai, V. Bhat, S.M. Ryno, L.Y. Huang, A. Smith, P. Sornberger, R. Duke, S. Goodlett, C. Risko with J.E. Anthony & M.M. Haley;
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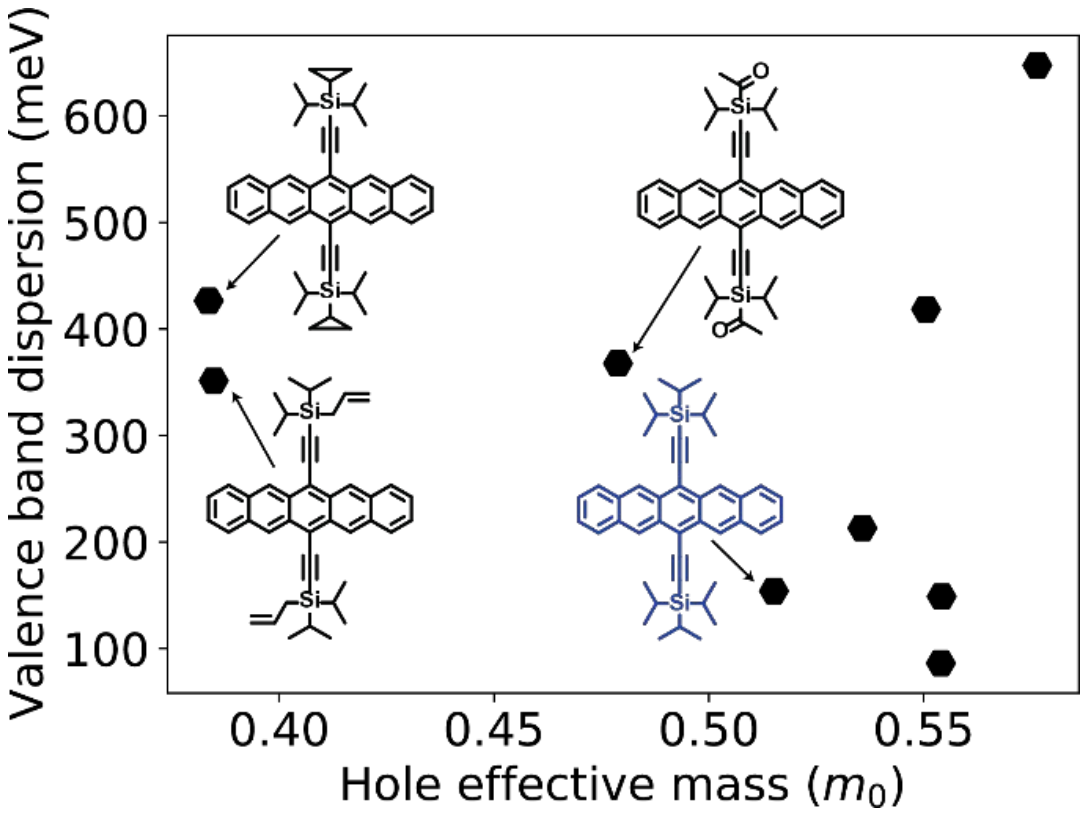
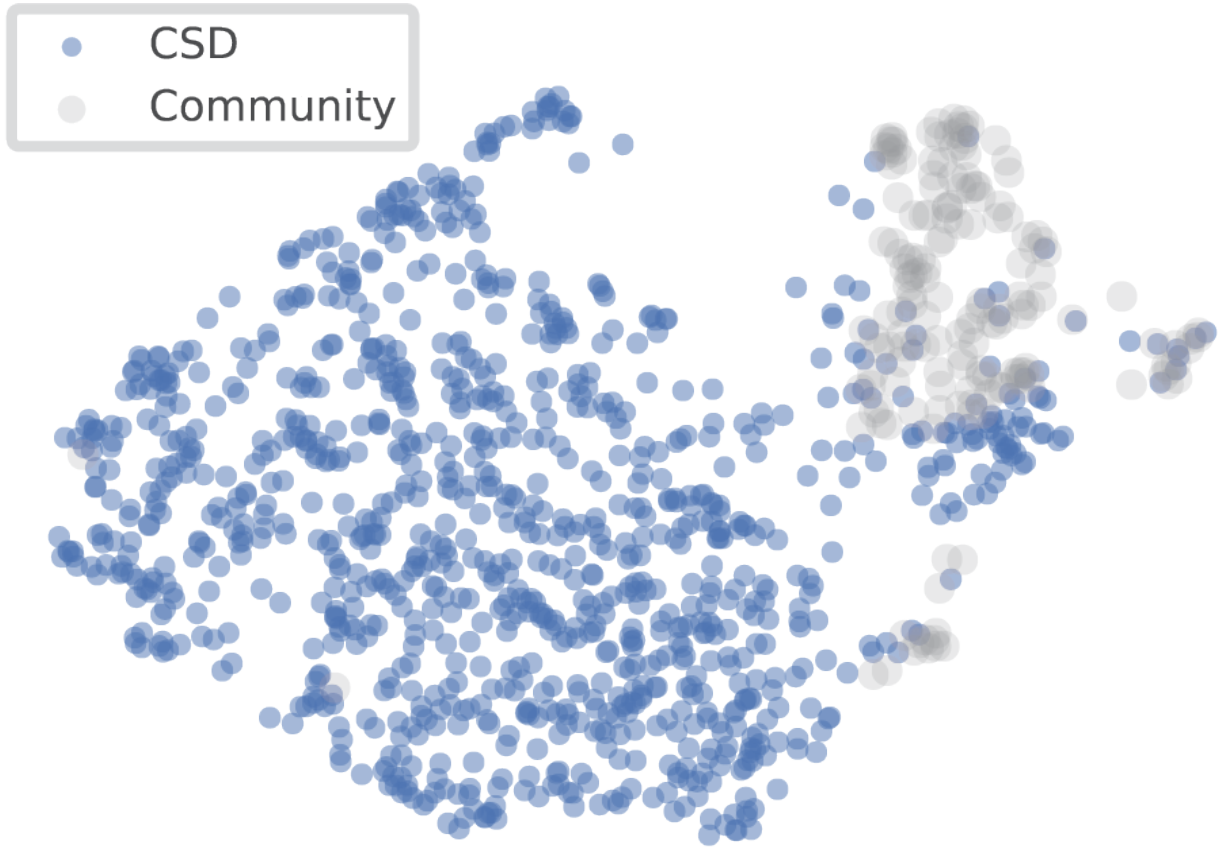
Data set includes “dark” structures

- Inclusion of dark structures aids in further exploration of chemical space of OSC





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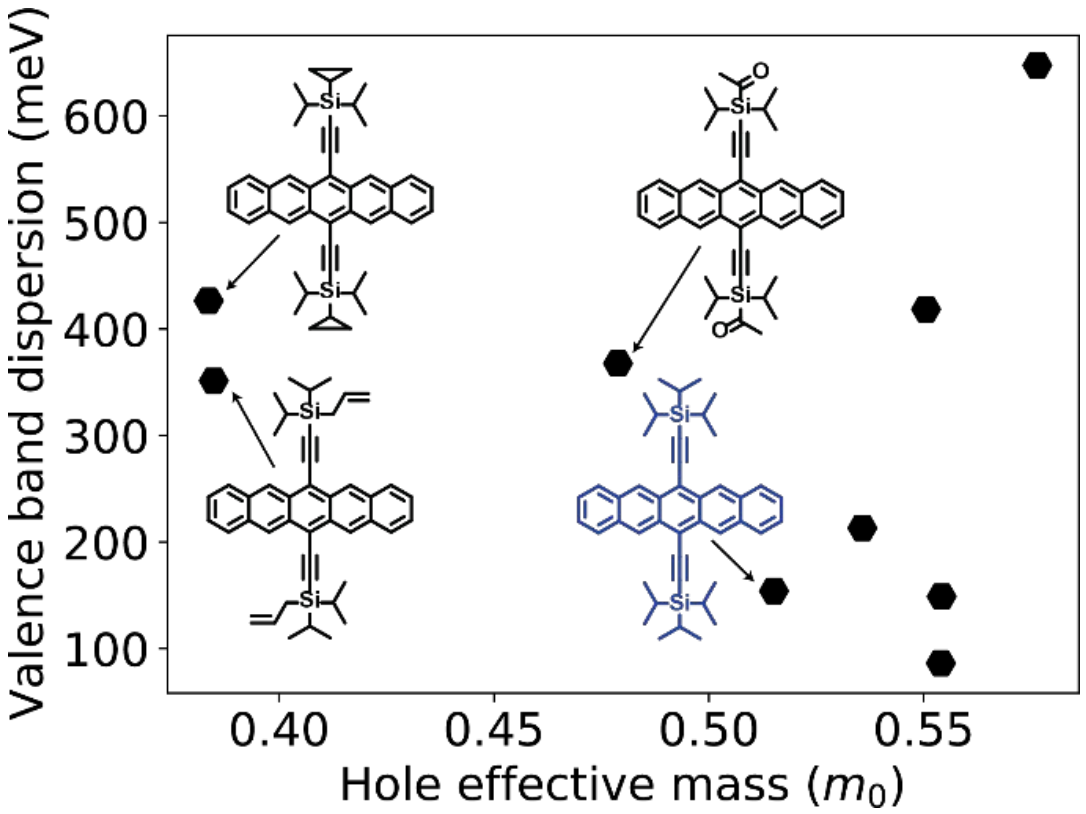
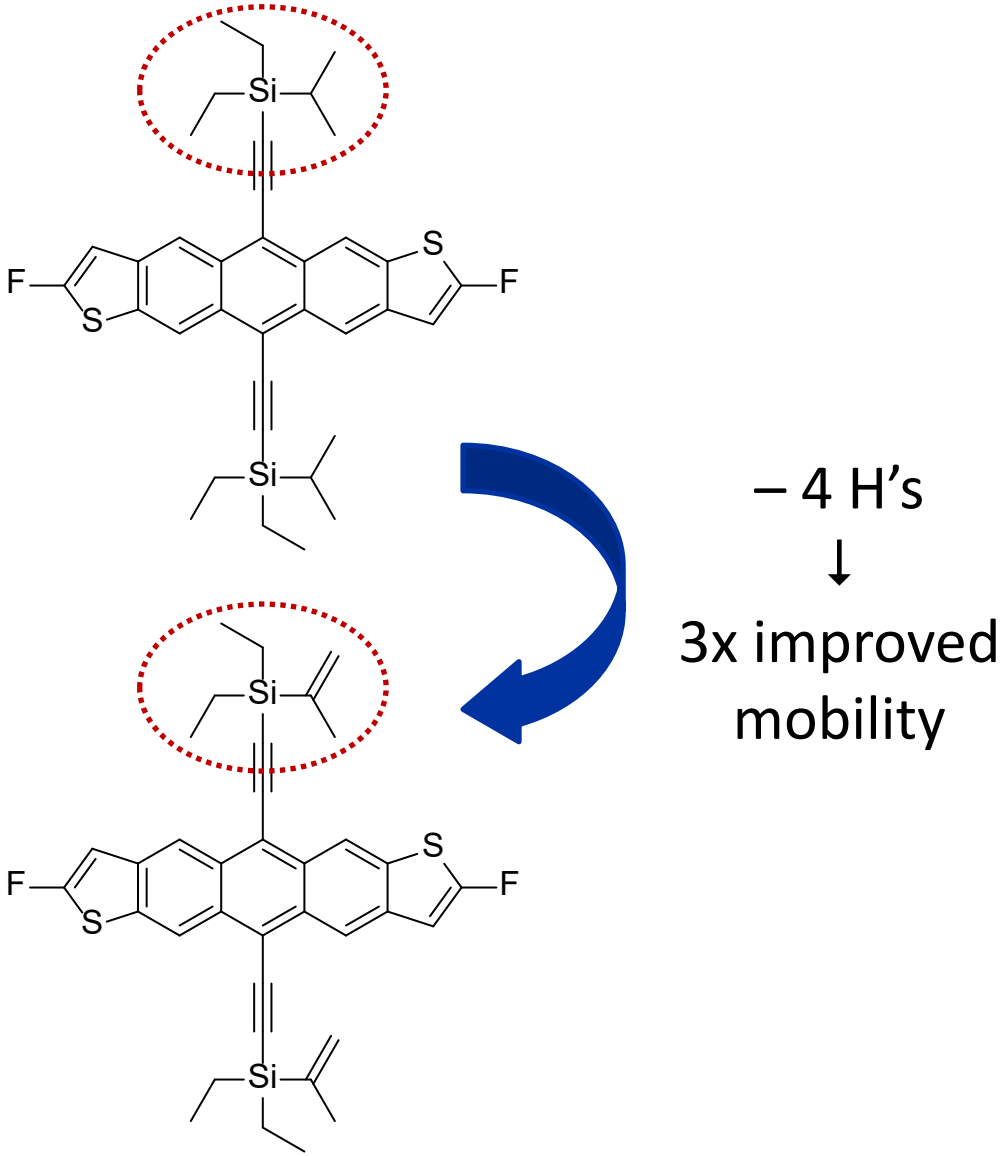




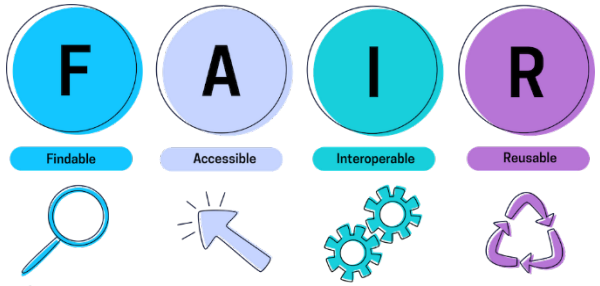
– 4 H’s

↓

3x improved mobility

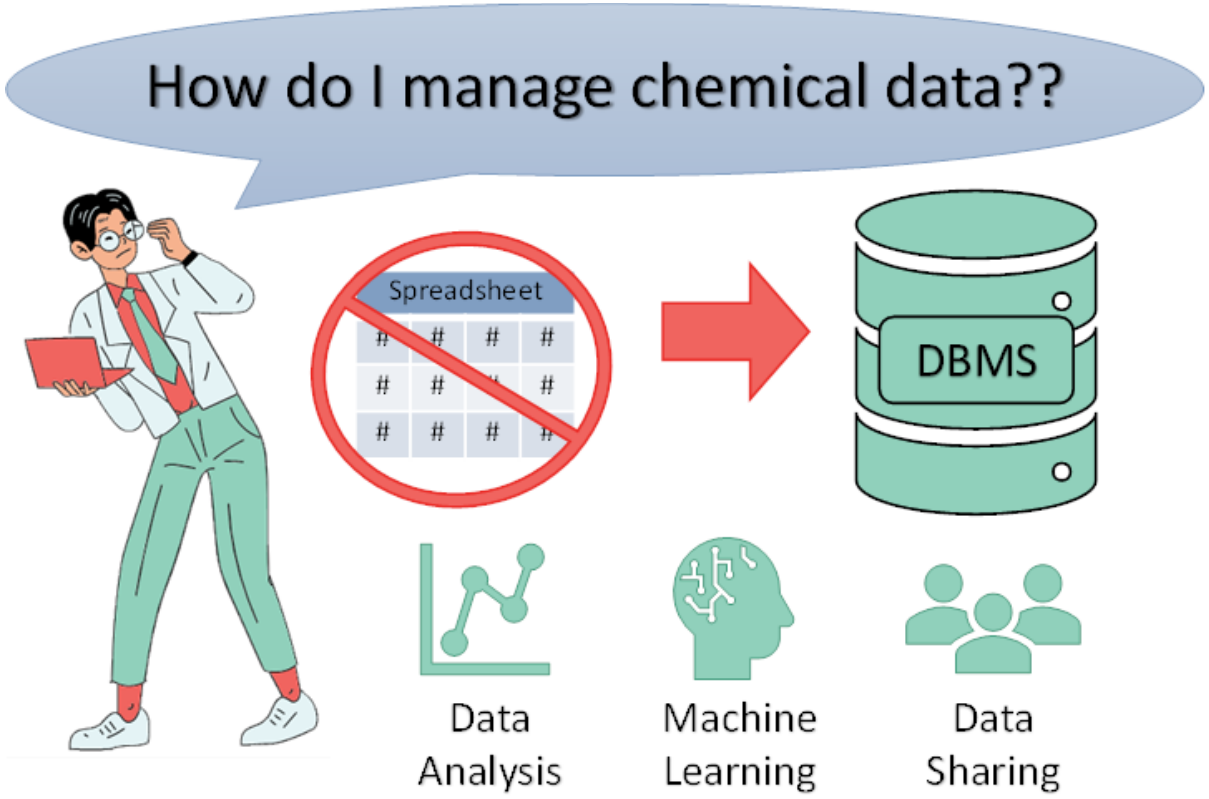
Q. Ai, V. Bhat, S.M. Ryno, L.Y. Huang, A. Smith, P. Sornberger, R. Duke, S. Goodlett, C. Risko with J.E. Anthony & M.M. Haley;
Q. Ai, V. Bhat, S.M. Ryno, K. Jarolimek, P. Sornberger, A. Smith, M.M. Haley, J.E. Anthony & C. Risko, J. Chem. Phys. 2021, 154, 174705.

FAIR scientific data principles



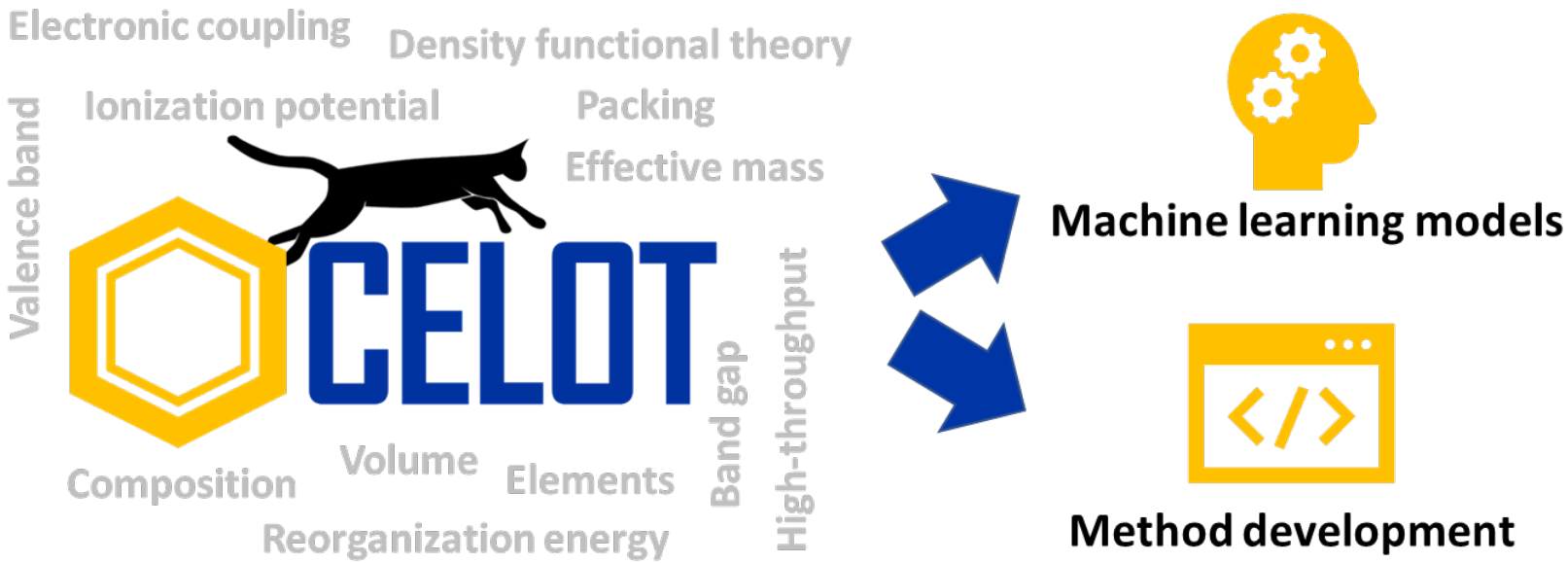
https://flur.ee/why-fluree/

- Findable
  - Could anyone find the data they are looking for within my data?
- Accessible
  - Could anyone access my data?
- Interoperable
  - Could anyone in my field understand the way my data is organized?
- Reusable
  - Could anyone use my data for validation or for their own analysis ?



M.D. Wilkinson, *et. al.* Scientific Data 2016, 3, 160018;
R. Duke, V. Bhat & C. Risko, Chem. Sci. (2022), 3, 13646.

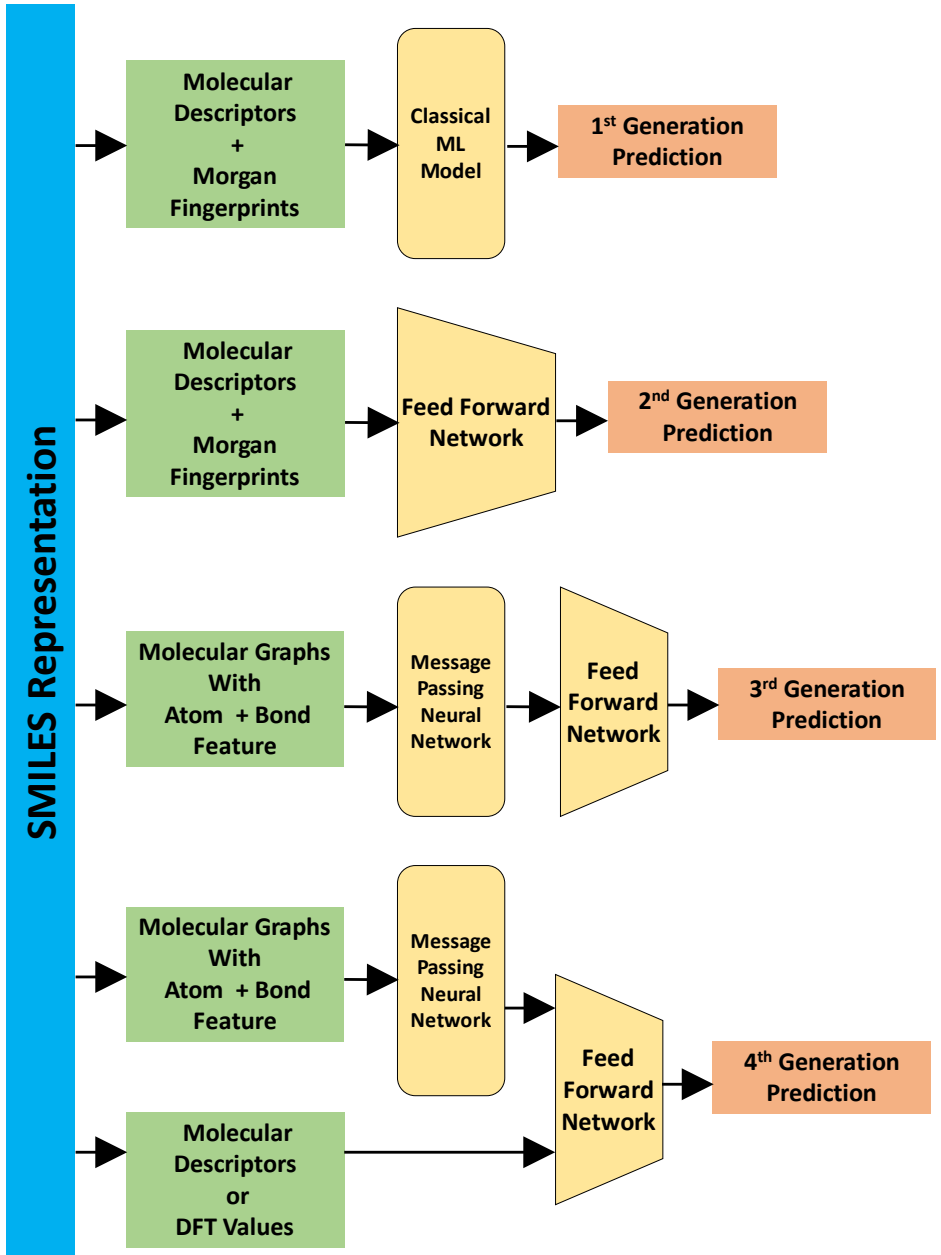
Let’s predict molecular & materials properties



How can we use this molecular & materials data to predict properties of new systems?

V. Bhat, P. Sornberger, B. Pokuri, R. Duke, B. Ganpathysubramanium & C. Risko, Chem. Sci. (2023), 14, 203;
V. Bhat, B. Ganapathysubramanian & C. Risko, (2023) ChemRxiv, DOI: 10.26434/chemrxiv-2023-rvzmv.

Let's predict molecular & materials properties



- Challenges that need to be overcome:
  - Computing DFT-based electronic properties is time consuming
  - Recently published models are trained on datasets with ~25 atoms or rather niche chemical space

- Properties modeled:

  Vertical (VIE) and adiabatic (AIE) ionization energies
  Vertical (VEA) and adiabatic (AEA) electron affinities
  Cation (CR) and anion (AR) relaxation energies
  HOMO energies (HOMO), LUMO energies (LUMO)
  HOMO-LUMO energy gaps (H-L)
  Electron (ER) and hole (HR) reorganization energies
  Lowest-lying singlet (S0S1) and triplet (S0T1) excitation energies

OCELOT ML

oscar.as.uky.edu/ocelotml_2d

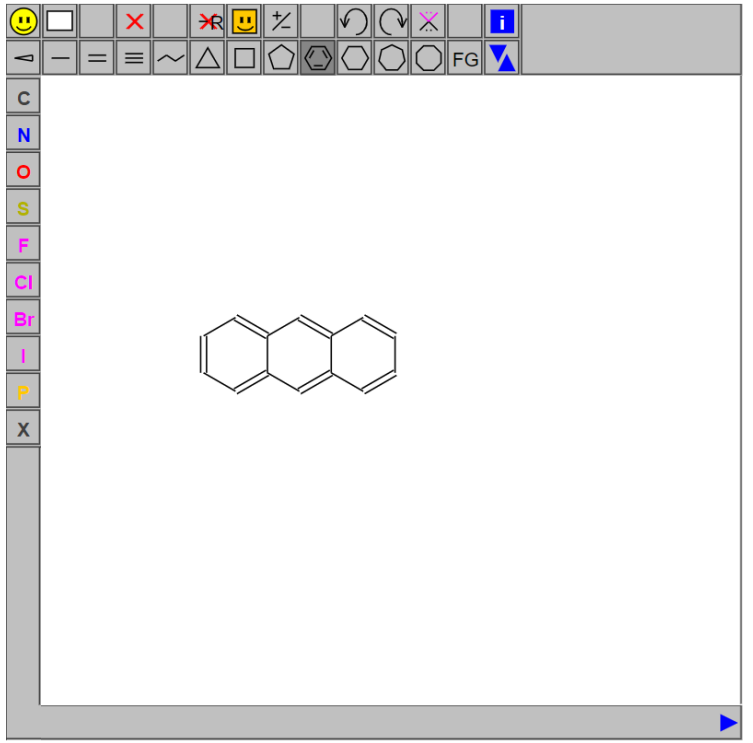
V. Bhat, P. Sornberger, B. Pokuri, R. Duke, B. Ganpathysubramanium & C. Risko, Chem. Sci. (2023), 14, 203;
V. Bhat, B. Ganapathysubramanian & C. Risko, (2023) ChemRxiv, DOI: 10.26434/chemrxiv-2023-rvzmv.

Let’s predict molecular & materials properties

- OCELOT ML provides the infrastructure to make the models publicly accessible
- Currently, molecular predictions with SMILES input is supported
- Best models from our training are available

OCELOT ML

oscar.as.uky.edu/ocelotml



Select propetry to predict

Adiabatic Ionization Energy (R2 = 0.87; MAE = 0.16)

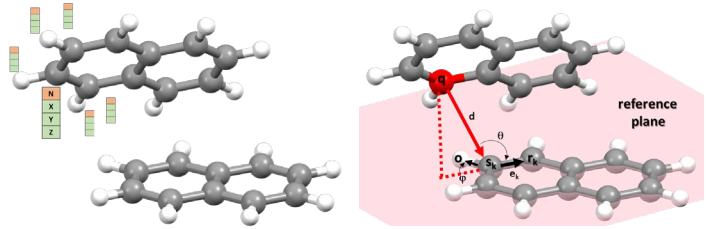
Predict

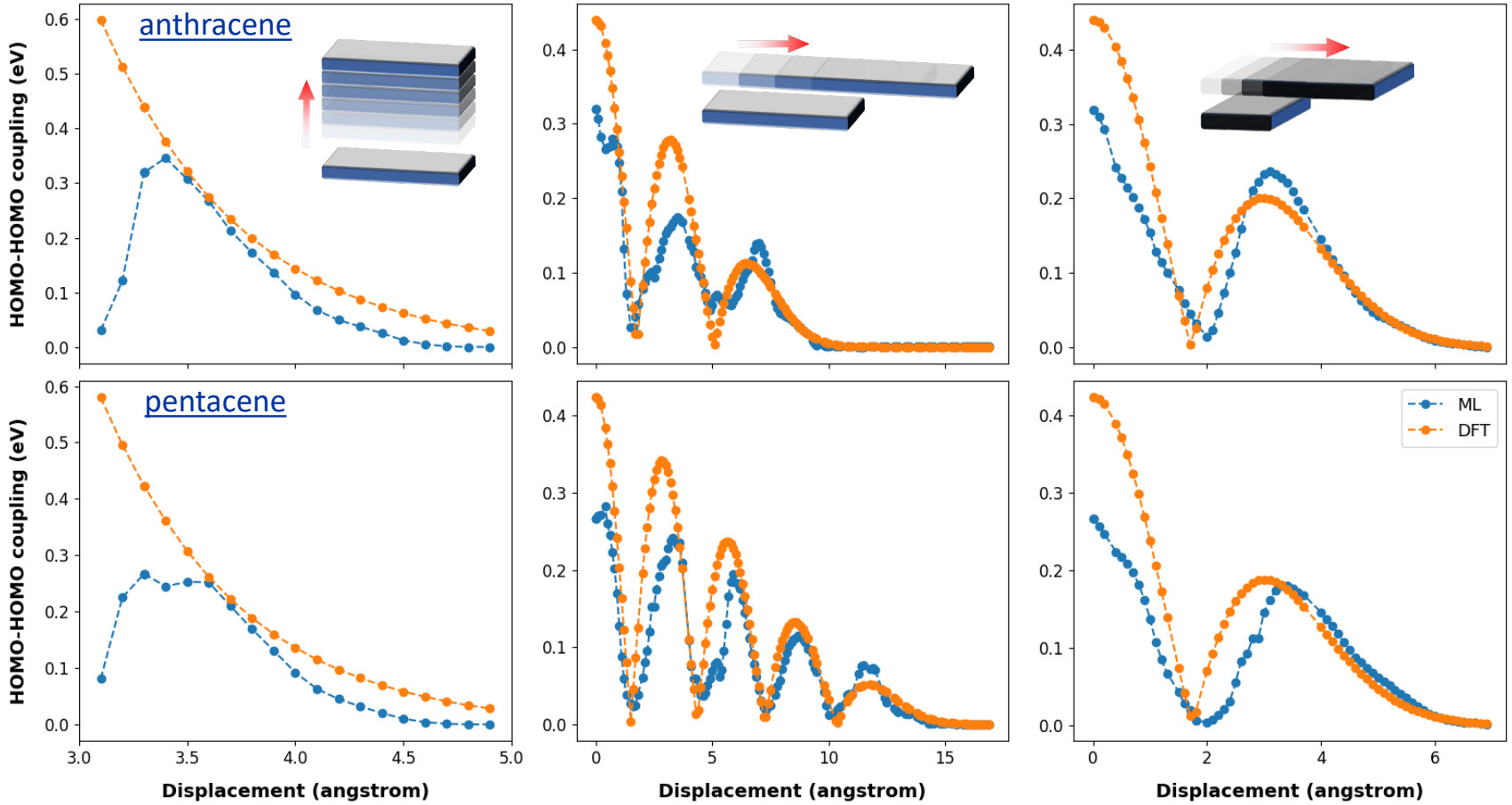
Predicted value is : 7.16 ± 0.151 eV

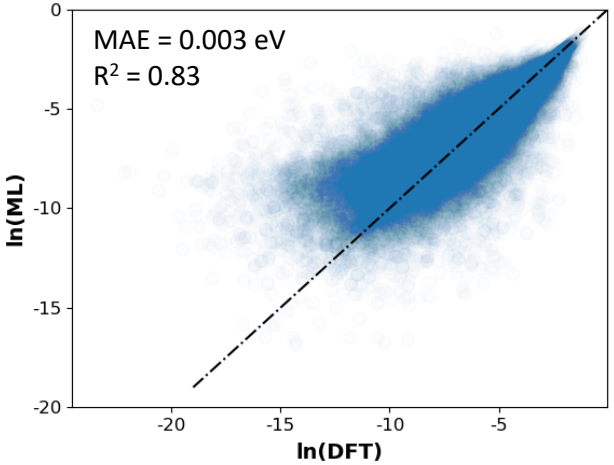
V. Bhat, P. Sornberger, B. Pokuri, R. Duke, B. Ganpathysubramanium & C. Risko, Chem. Sci. (2023), 14, 203.

Let's predict molecular & materials properties

- OCLEOT dimer v1 dataset has >439,000 DFT-determined electronic couplings





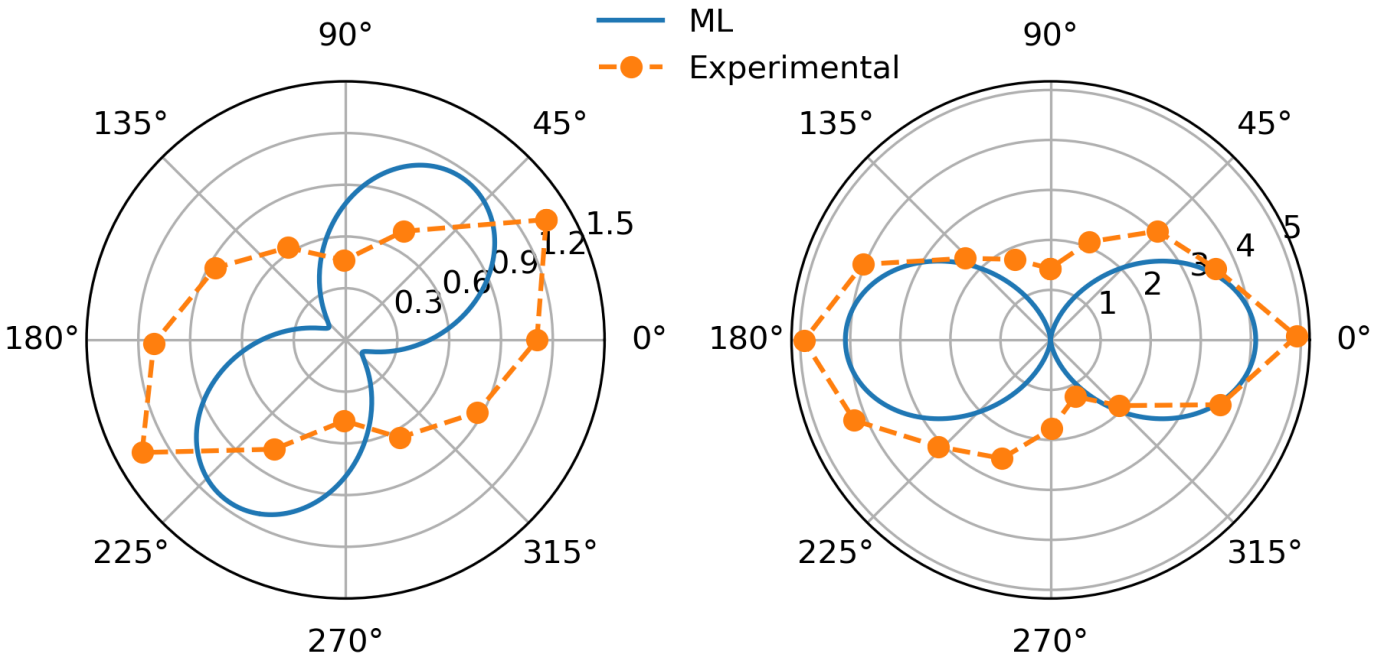


V. Bhat, B. Ganapathysubramanian & C. Risko, (2023) ChemRxiv, DOI: 10.26434/chemrxiv-2023-rvzmv;
V. Bhat, Q. Ai, & C. Risko, Provisional Patent Application 63/4888713

Let’s predict molecular & materials properties

via semi-empirical Marcus theory, prediction of charge-carrier mobility anisotropy

- ML intramolecular reorganization energy
- ML intermolecular electronic couplings



pentacene rubrene

OCELOT ML

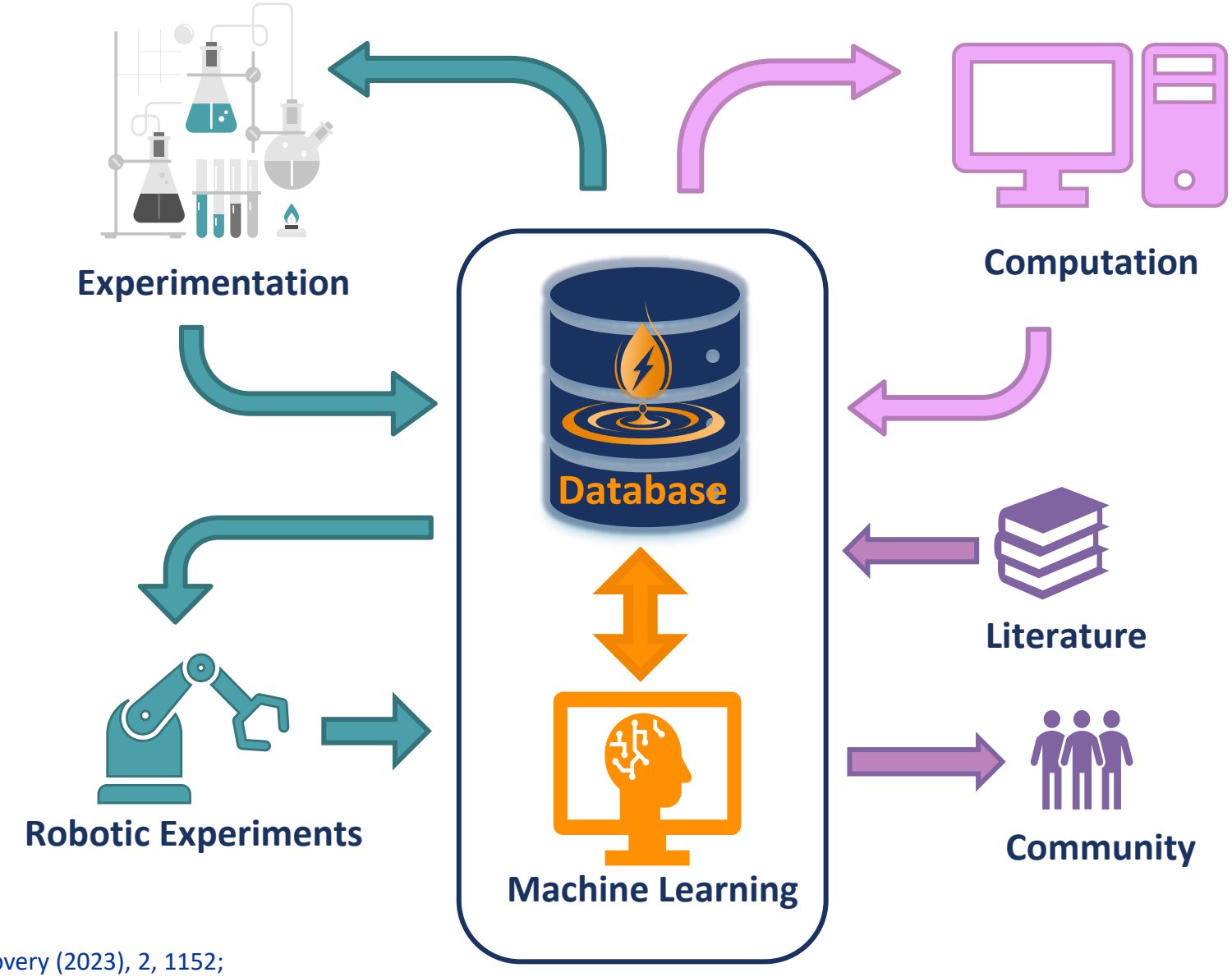
oscar.as.uky.edu/ocelotml

V. Bhat, B. Ganapathysubramanian & C. Risko, (2023) ChemRxiv, DOI: 10.26434/chemrxiv-2023-rvzmv;
V. Bhat, Q. Ai, & C. Risko, Provisional Patent Application 63/4888713

Now on to materials for batteries

Data-enabled **D**iscovery and **D**esign to **Tra**nsform **L**iquid-based **E**nergy **S**torage (D^3TaLES)

d3tales.as.uky.edu



R. Duke, V. Bhat, P. Sornberger, S.A. Odom & C. Risko, Digital Discovery (2023), 2, 1152;
R. Duke, S. Mahmoudi, A.P. Kaur, V. Bhat, I. Dingle, N.C. Stumme, S.K. Shaw, D. Eaton, A. Vego & C. Risko (2023), submitted.

Building more data pipelines

How can we incorporate (dynamic) experimental data?

How do we collect all the data associated with an experiment, i.e., the metadata?

A chemist performs an experiment, hand-writing measurements and observations in a laboratory notebook...

1 **Missing details**

Only selected details are transferred to the published paper. Other chemists have difficulty reproducing results.

2 **Machine readability**

It is difficult for computers to translate notebook text to experiment steps for analysis

3 **Manual analysis**

The chemist must manually transfer values from notebook to computer, then perform calculations.

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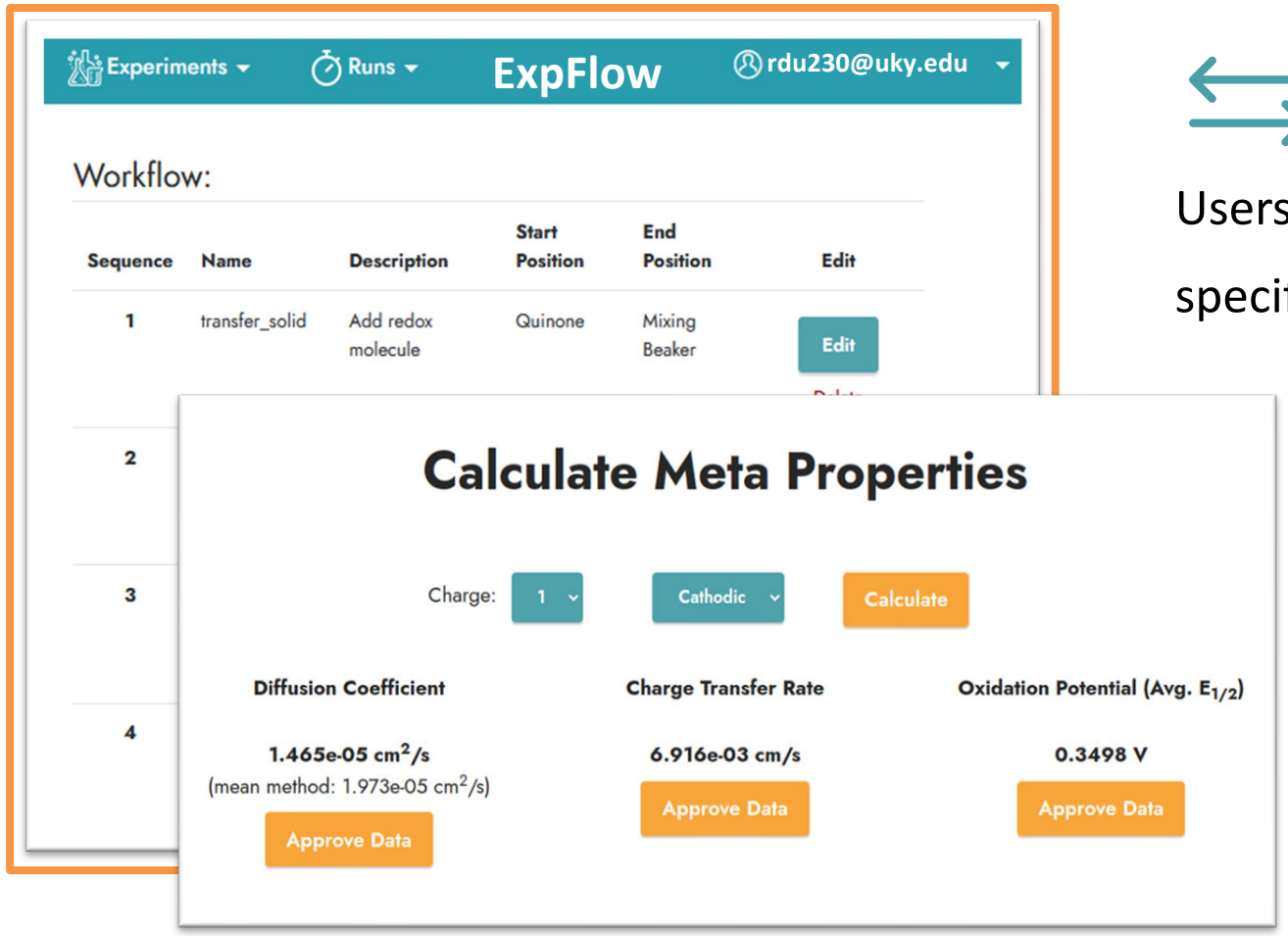
ExpFlow



Customizable

Users can modify the tools for specific use

Reusable

Experimental templates can be used by others

Sharable

All raw data and experimental procedures can be shared

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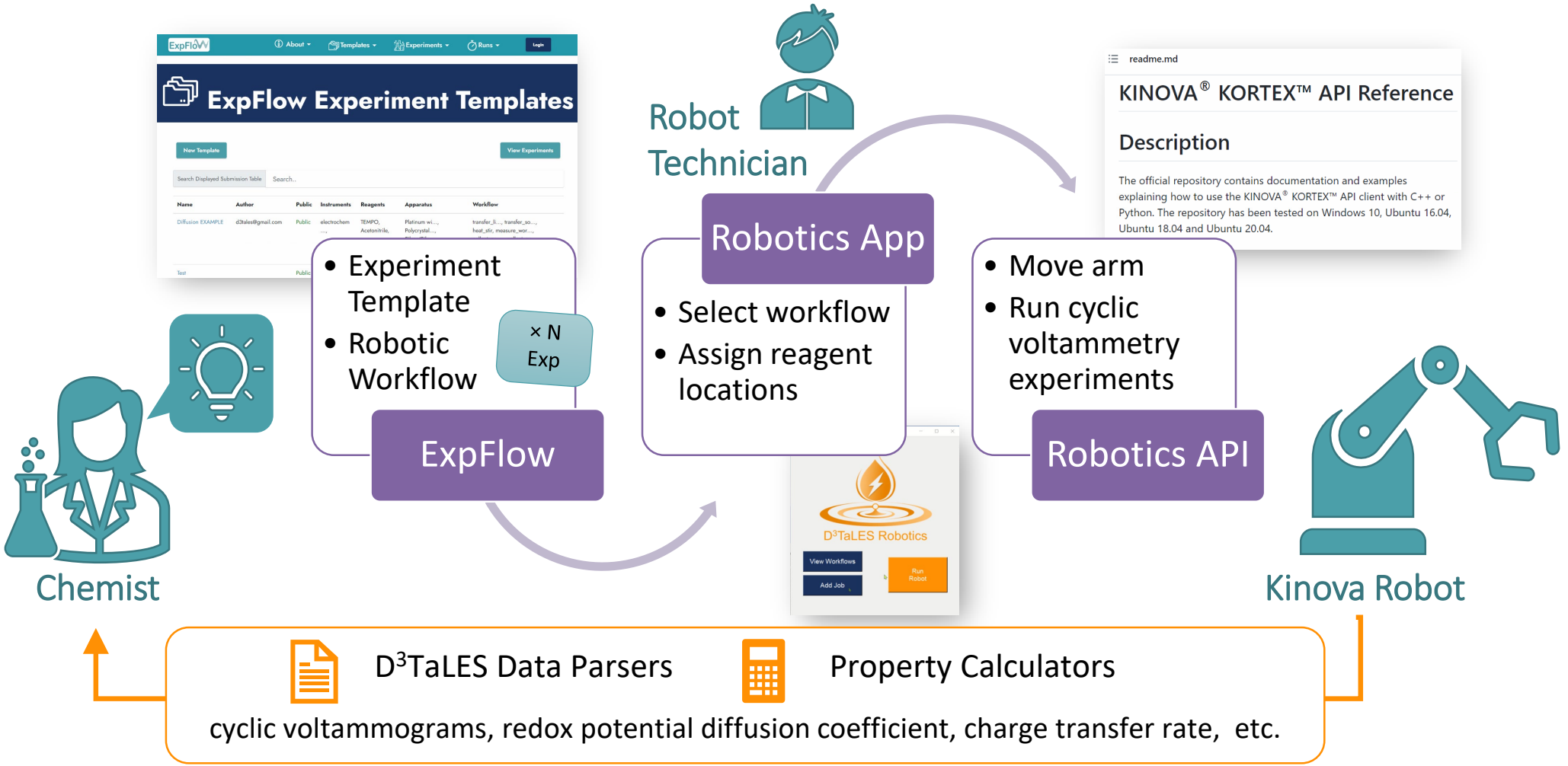
Automated electrochemistry



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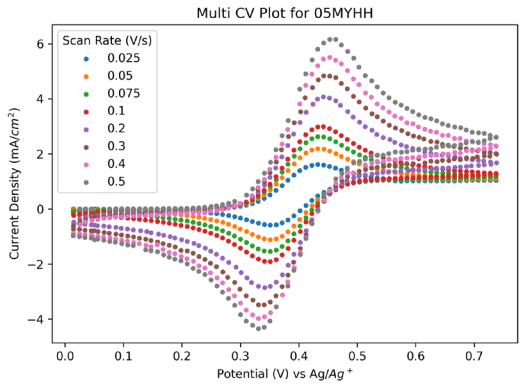
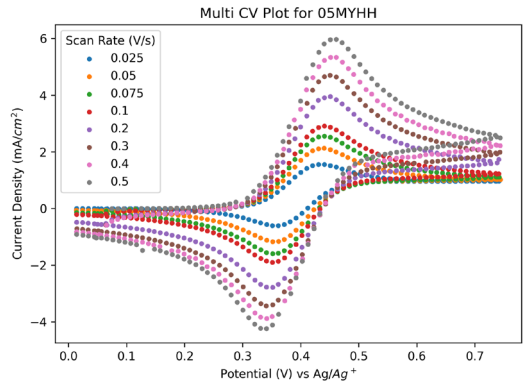
Automated electrochemistry

Exp 2: N-[2-(2-Methoxyethoxy)ethyl]-phenothiazine (**MEEPT**) CVs

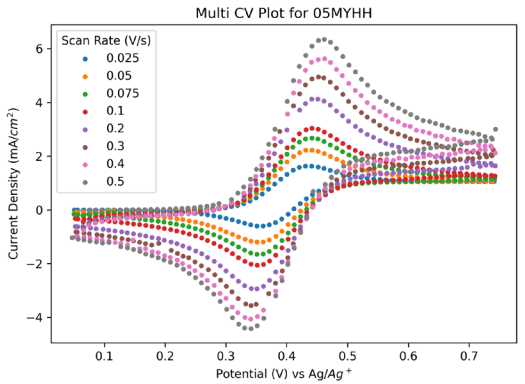
Trail 1



Trial 2



Trial 3



	Robotic/ExpFlow						Literature Reported vs. Ag/Ag+		Literature Reported vs. Fc/Fc+	
ROM	Trial 1	Trial 2	Trial 3	Avg. (vs. Ag/Ag^{+})	Std. Dev.	vs. Fc/Fc^{+} [α]	Value	Ref.	Value	Ref.
Fc	0.081	0.082	0.082	0.082	0.001	0.000	0.086	Ref.[6]		
MEEPT	0.396	0.396	0.396	0.396	0.000	0.314	0.410*	Ref.[14]	0.310	Ref.[8]
DMPZ	-0.156	-0.156	-0.156	-0.156	0.000	-0.238	-0.150	Ref.[9]		
4-MeO TEMPO	0.371	0.376	0.375	0.374	0.003	0.292	0.68[++]	Ref.[5]		
DBB	0.773	0.773	0.773	0.773	0.000	0.691	0.710	Ref.[10]		
DBBB	0.773	0.768	0.773	0.771	0.003	0.690			0.60[‡]	Ref.[13]
TH	0.910	0.910	0.910	0.910	0.000	0.828	0.900	Ref.[12]	0.840	Ref.[7]
ECZ**	0.678	0.678	0.672	0.676	0.003	0.594				

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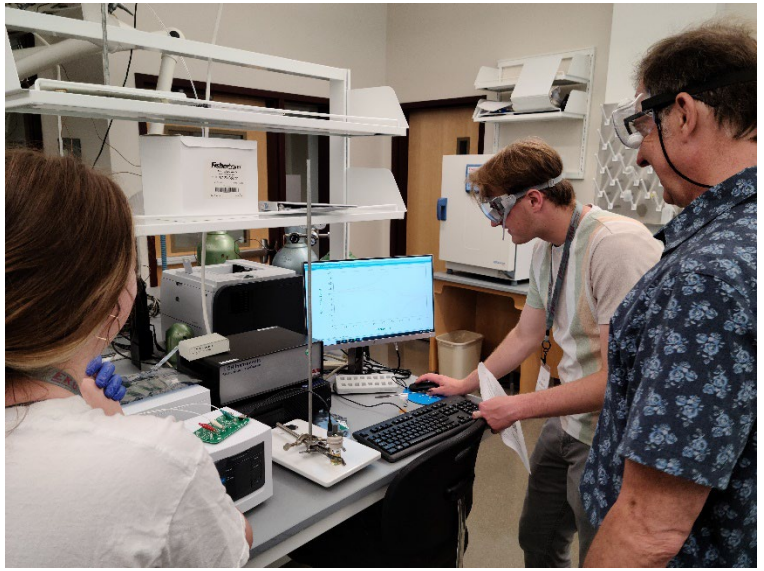
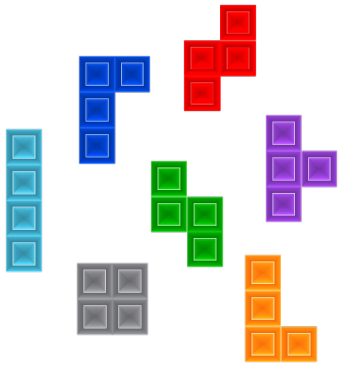
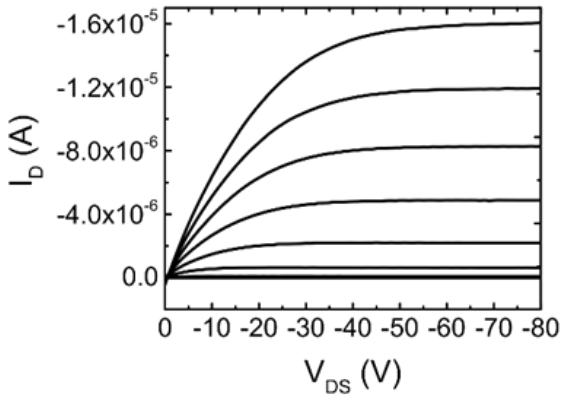
In the Mix: An immersive educational experience



in silico design



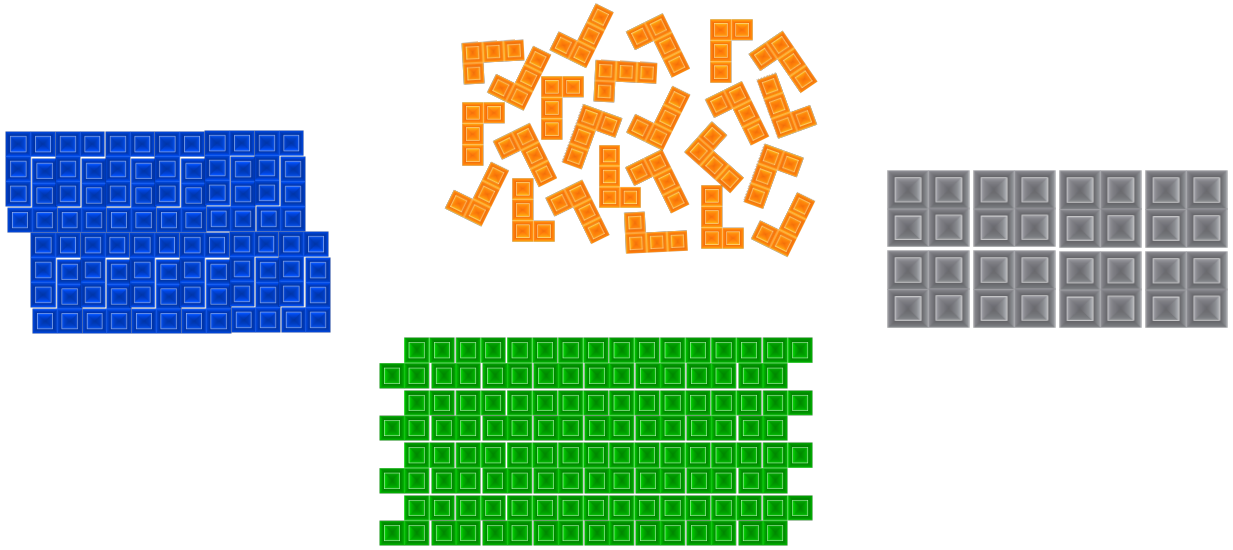
A library of organic chromophores



A. Zeidell *et al*. Chem. Mater. (2019) 31, 6962.

BUILDING BLOCK

RESPONSE



STRUCTURE & SYNTHESIS PREDICTION

PROPERTY PREDICTION

Adapted from Q. Ai

THANK YOU!