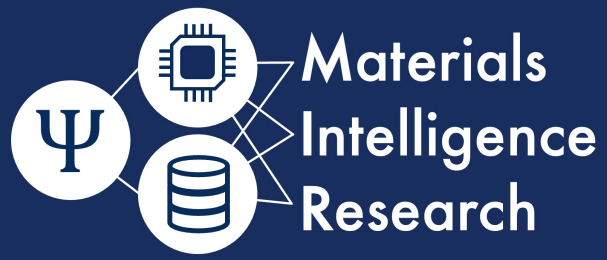
DOE CSGF

Materials Informatics for Catalyst Stability & Functionality

Steven B. Torrisi

Featuring work done at...



Harvard John A. Paulson School of Engineering and Applied Sciences

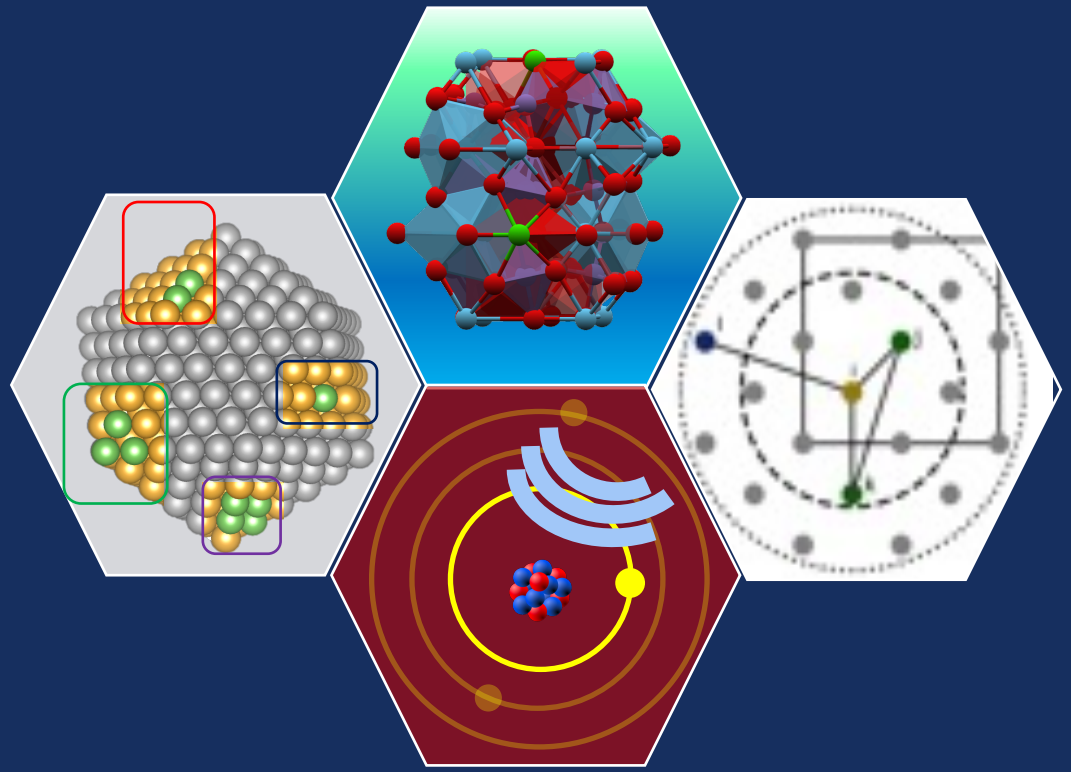
TOYOTA RESEARCH INSTITUTE

BERKELEY LAB

About Me

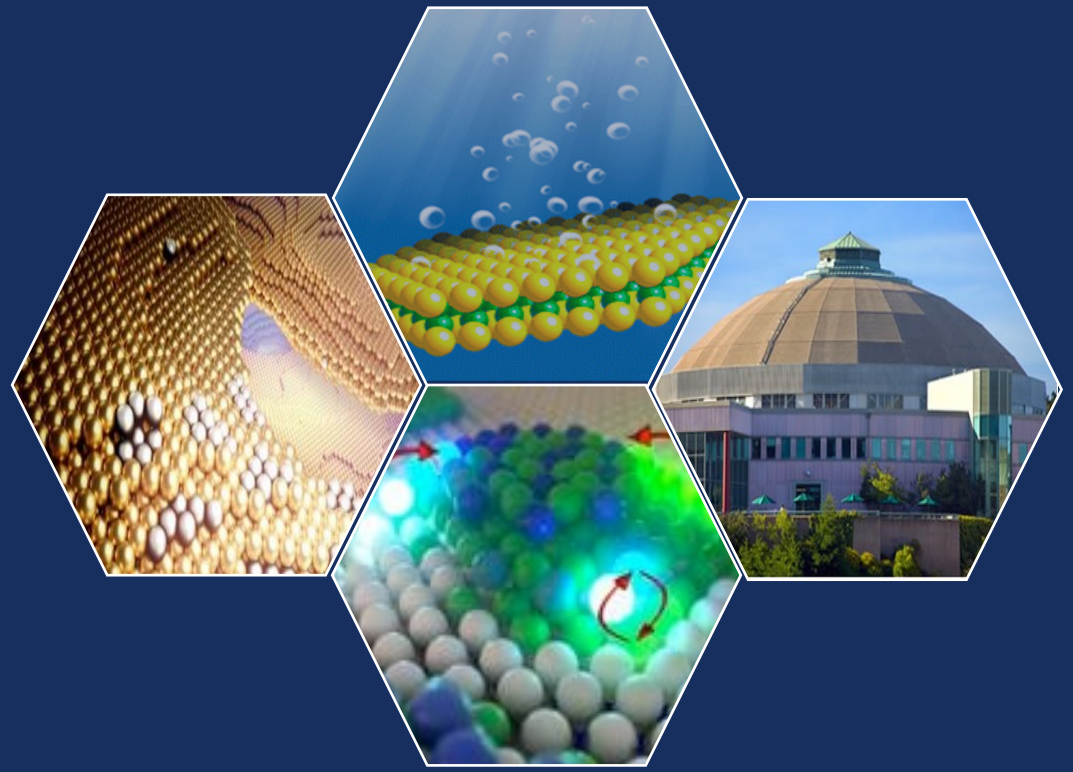
Sustainable Energy Development

Computational Materials Science



supporting

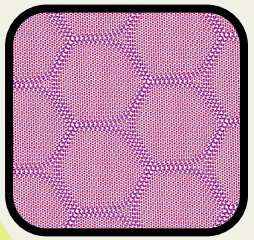
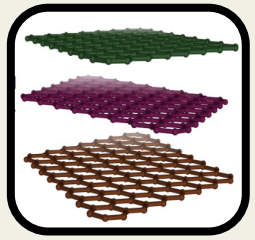
Catalyst Discovery, Optimization, and Understanding



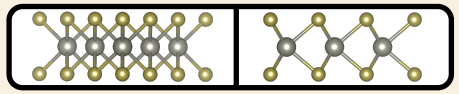
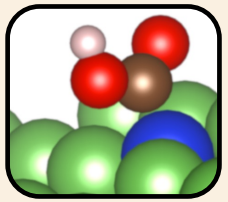
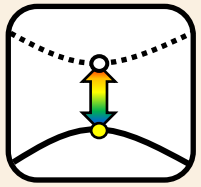
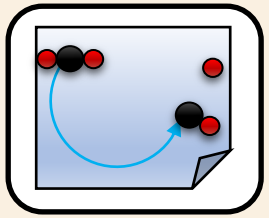
Credit: Top, A. Singh, Left, IMASC, Right, LBNL, Bottom, J.S. Lim

Scientific Directions During my Ph.D.

Stability & Mechanical Properties of Functional 2D Materials

Photocatalysis

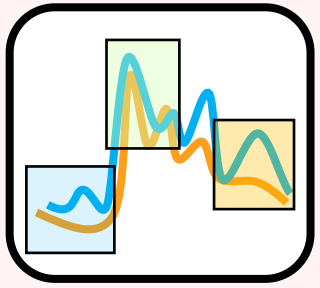
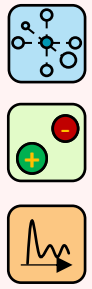
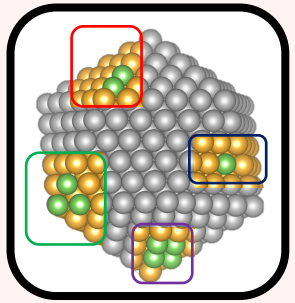

Scientific Software Development

TRIXS

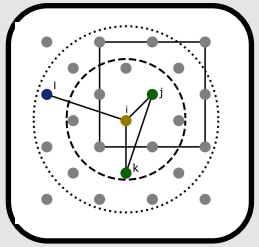
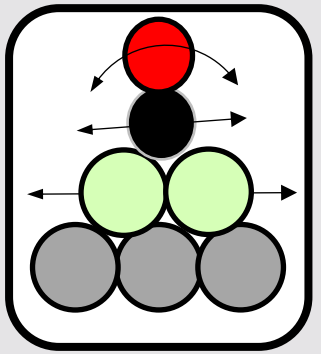
YGRW

InterMatch

Machine Learning for X-Ray Characterization

Machine-Learned Interatomic Potentials

1st author

"et al."

In prep.

Scientific Directions During my Ph.D.

2D Materials

Carr, Massat, **Torrisi** et al. *PRB (2018)*

Larson, Chen, **Torrisi** *et al.* PRB (2020)

Tritsaris, Carr, Zhu, Xie, **Torrisi** *et al.* 2D Mat. (2020)

Rhone, Chen, Desai, **Torrisi** *et al.* Sci. Rep. (2020)

Photoelectrocatalyst Discovery

Torrisi, Singh, Montoya, Biswas, Persson, *NPJ 2D Materials & Applications* 2020

Scientific Software Development

Sumner, **Torrisi**, Brickner, Brickner (Under Review, *eLife*)

Gerber, **Torrisi**, *et al.* (In prep.)

ML-Based X-Ray Characterization

Torrisi, *et al.*, NPJ Computational Materials (2020)

Marcella, Lim, Plonka… ***et al.*** (In prep.)

Machine-Learned Interatomic Potential Fitting

Vandermause, **Torrisi**, *et al.* NPJ Computational Materials (2020)

Torrisi, *et al.* (In prep)

(In prep)

1st author

"et al."

In prep.

My Practicum: Discovery of Two-Dimensional CO_2 Reduction Photocatalysts

In collaboration with…

Dr. Tathagata Biswas
Arizona State University

Prof. Arunima Singh
Arizona State University

Dr. Joseph Montoya
Toyota Research Institute

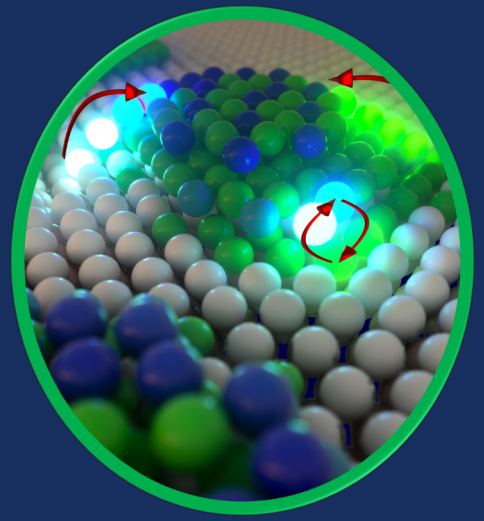
Prof. Kristin Persson
UC Berkeley

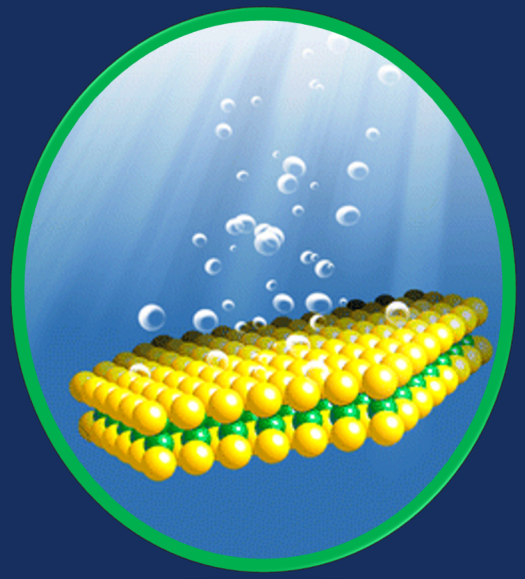
BERKELEY LAB

Energy-Relevant Materials

Catalysts

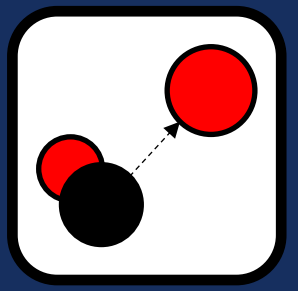
Materials which facilitate a useful chemical reaction

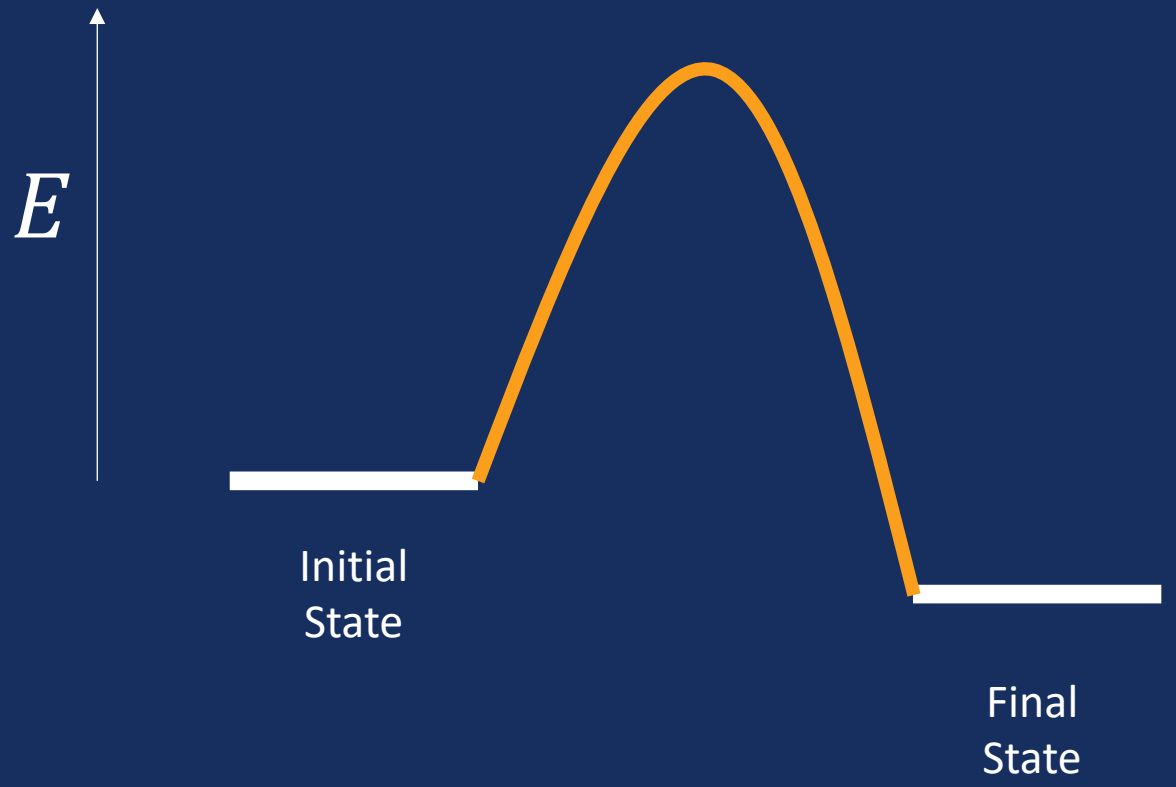




Left: Jin Soo Lim, Right: Arunima Singh

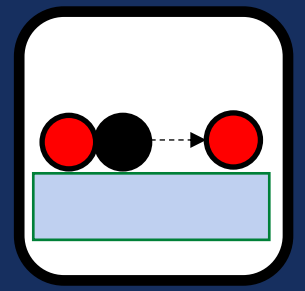
What are catalysts?

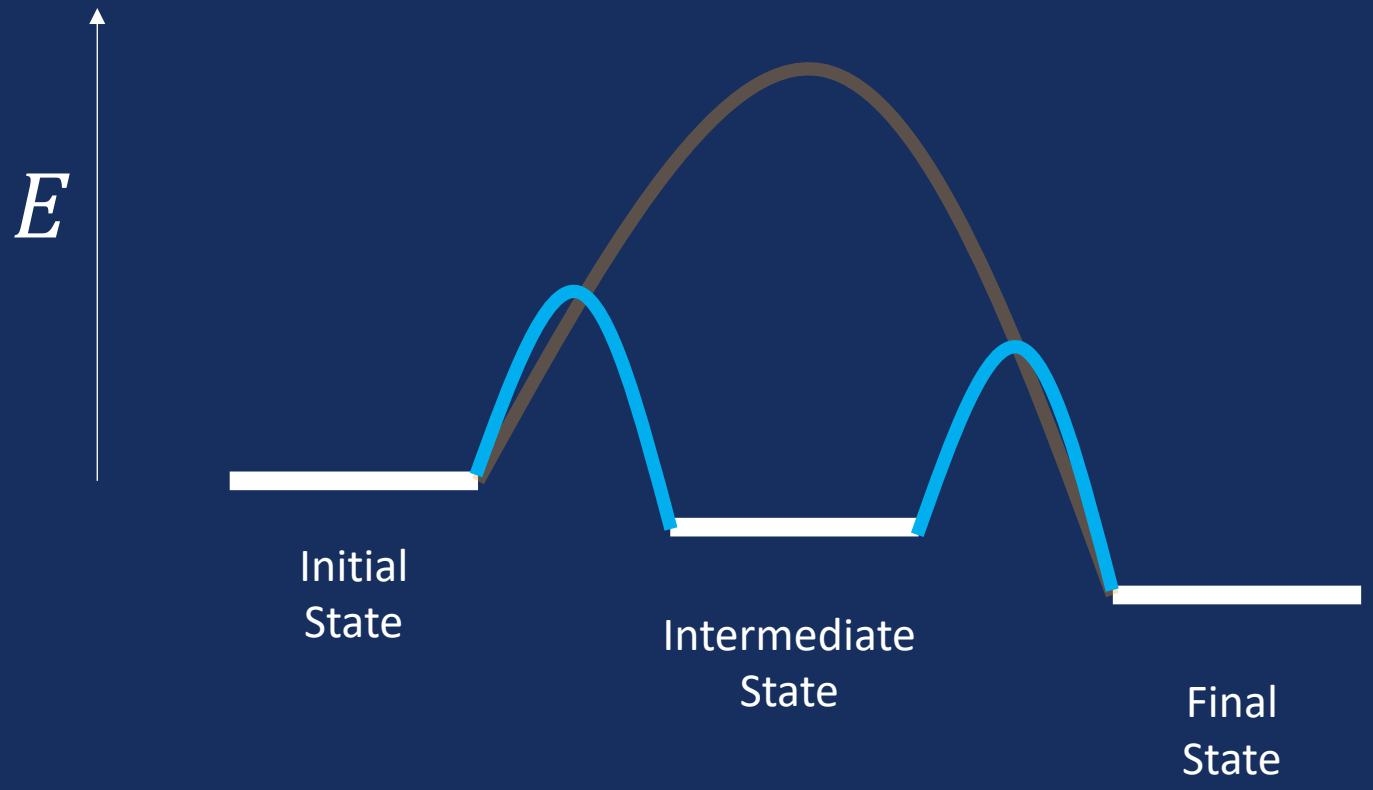




$$A \rightarrow B$$

What are catalysts?





Alternate Reaction Pathway Provided;
Catalyst Is Not 'Used Up'

Killer App: "Artificial Leaves"

Fuel Generation
From CO_2 *e.g.*

$CO_2 + 2\,H_2 \rightarrow CH_3OH$

$CO_2 + 4\,H_2 \rightarrow CH_4 + 2H_2O$

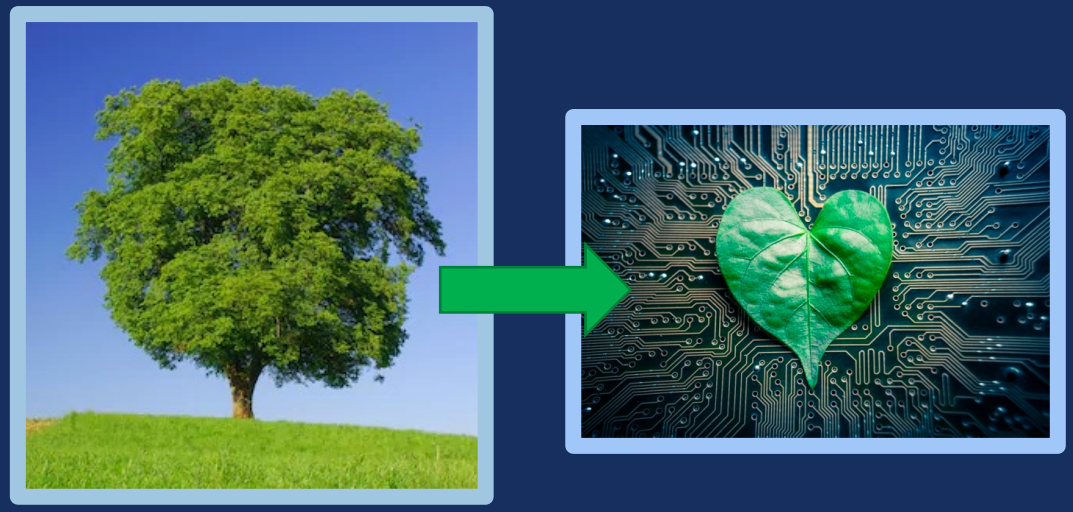
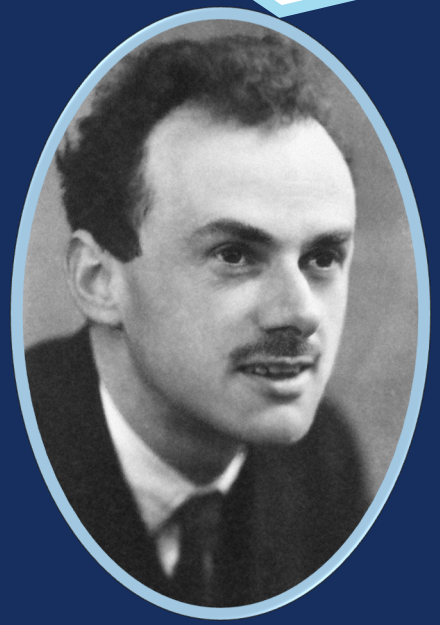


Photo credit: MIT News Office

Density Functional Theory, Briefly

The physical laws for a large part of physics and the whole of chemistry are completely known … "approximate practical methods of applying quantum mechanics should be developed"



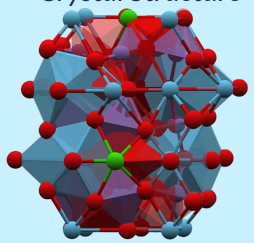
"Quantum Mechanics of Many-Electron Systems," P.AM. Dirac 1929

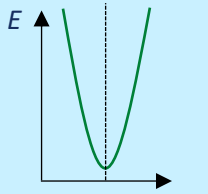
50+ years later:
Solve with functional of electron density ρ instead of all electrons Ψ

DFT-Accessible Properties

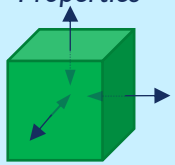
Mechanical & Structural Properties

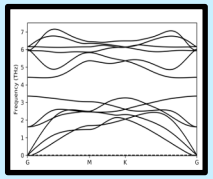
Crystal Structure





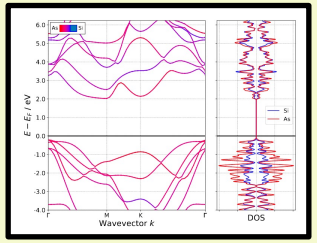
Elastic Properties





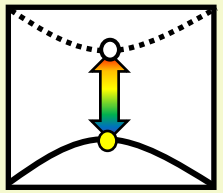
Vibrational Spectra

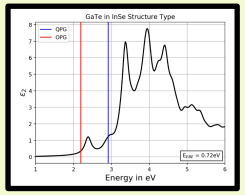
Electronic Properties

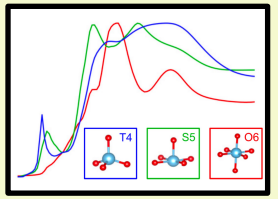


Bands & States

Band Gap



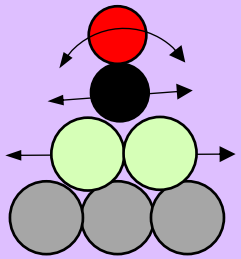




EM (Visible Light, X-Ray[1]…) Absorption

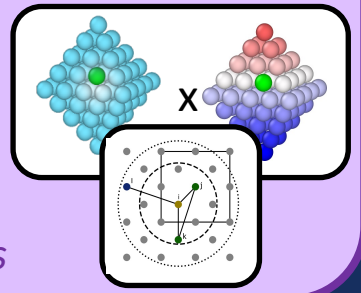
Dynamics

Accurate Forces



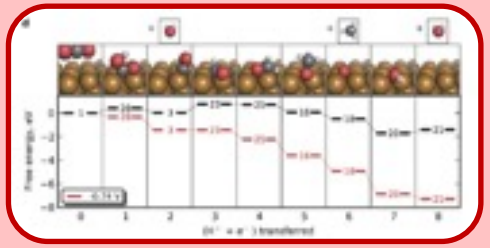
Ab-Initio Molecular Dynamics

Force Field Fitting



Reactivity

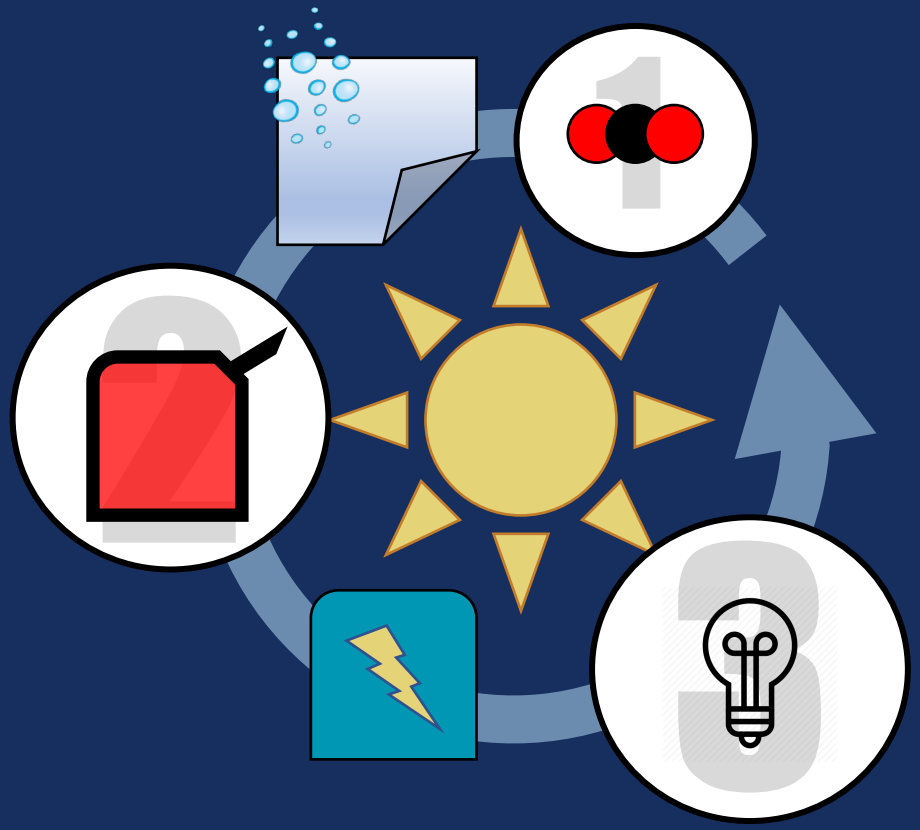
Binding Energies



Reaction Pathways[2]

1. Carbone *et al.* Phys. Rev. Mat. 2019
2. Peterson *et al.* Env. Sci,. & Technol. 2010

CO_2 Reduction & Artificial Photosynthesis

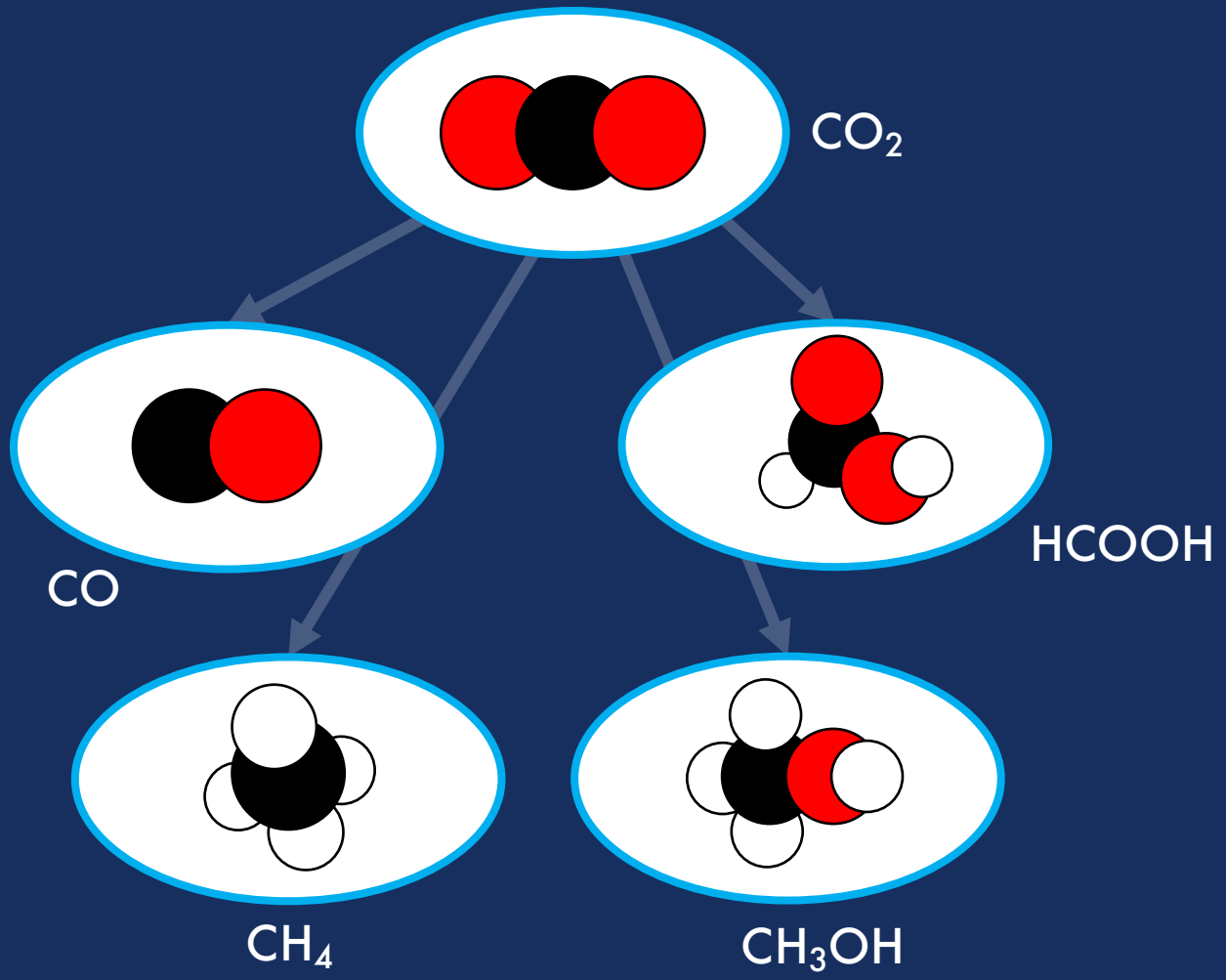


Central Challenges:

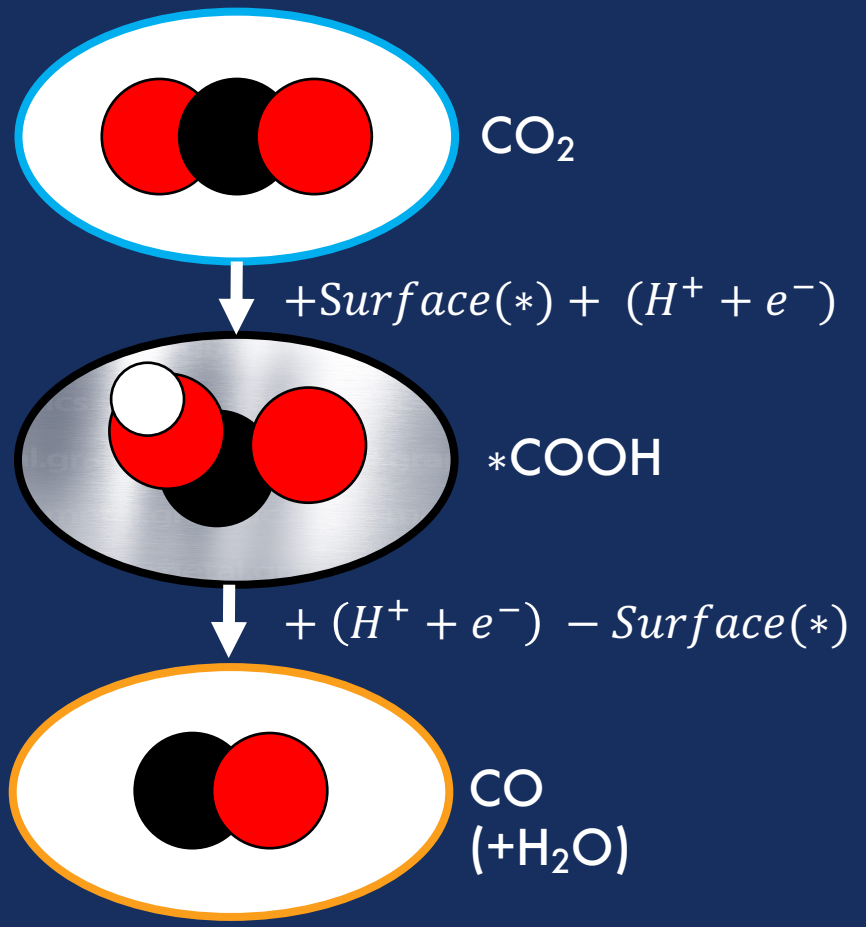
- CO_2 very stable
- Complex reaction pathway
- Hard to find reactive and stable materials

- More candidate catalysts
  → targets for future experiment and application

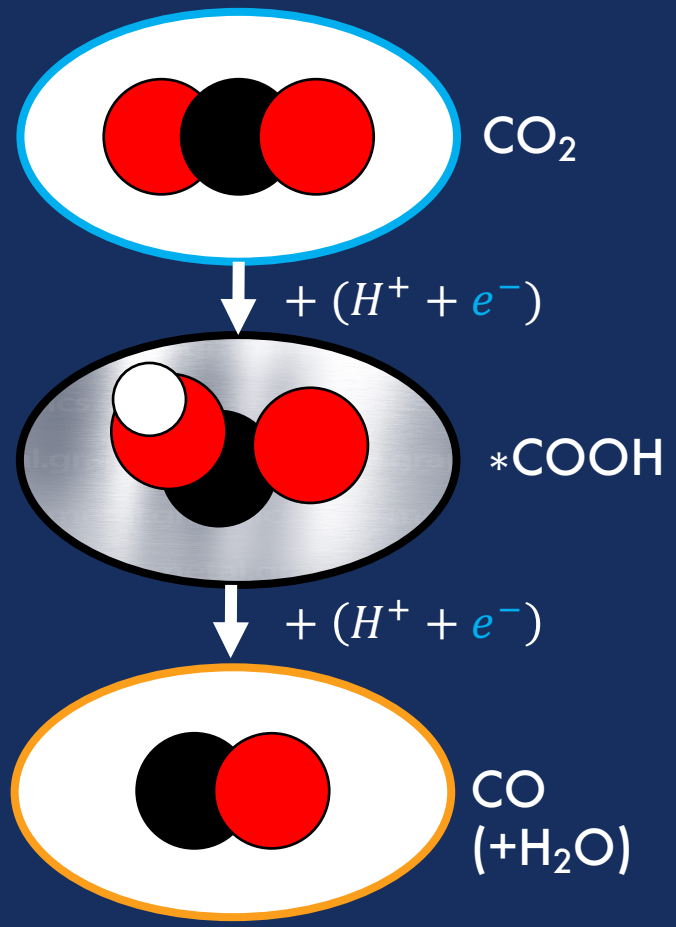
Many Reaction Products of Interest

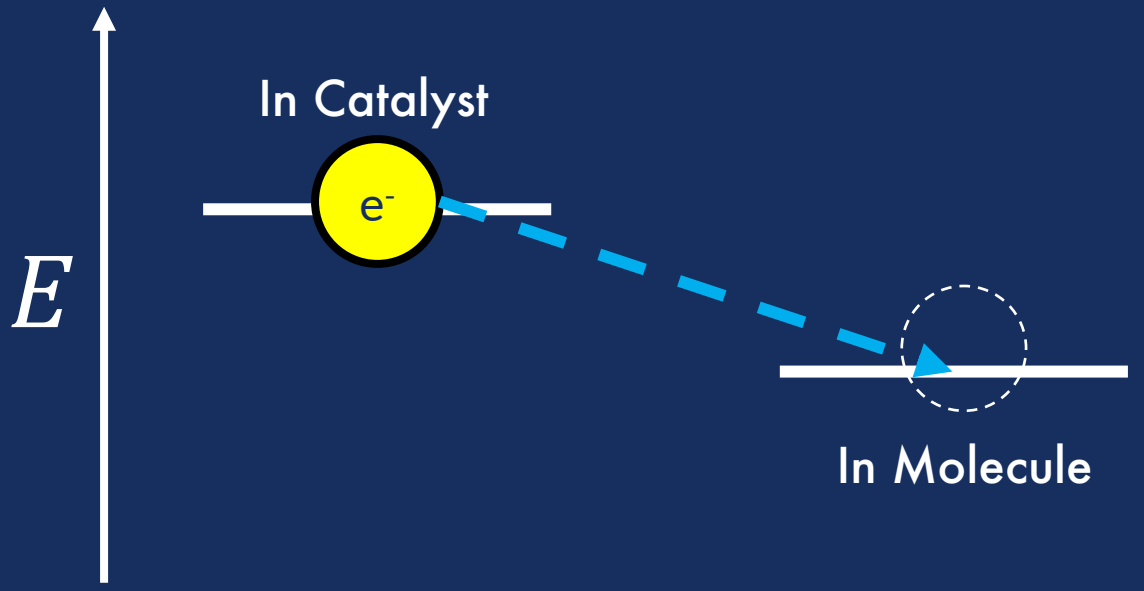


Reduction to CO

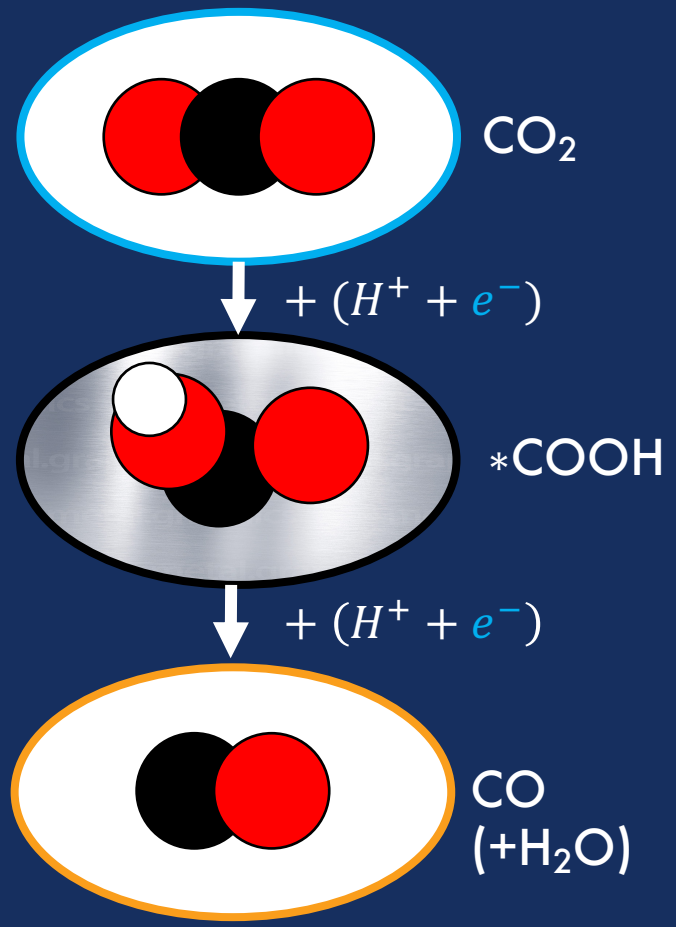


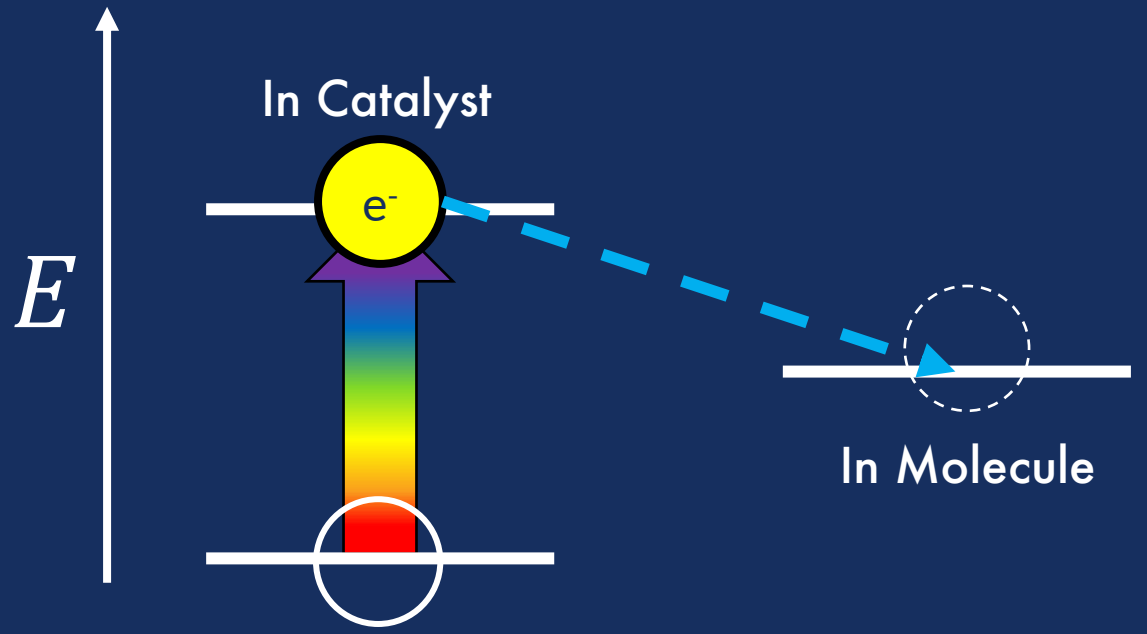
Reduction to CO





Reduction to CO



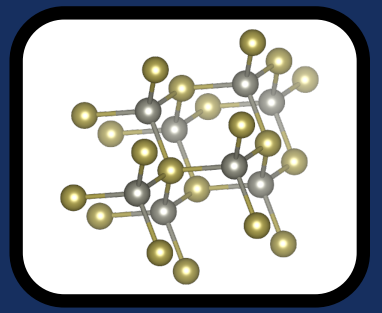


Previous Work: Discovery of Photocatalysts

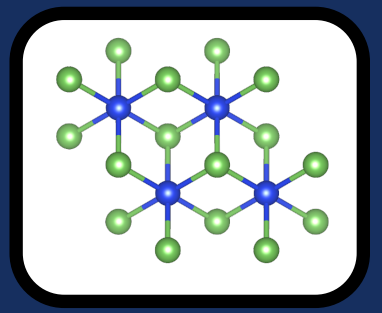
Computational Discovery of Photocatalyst Compounds

40+
New
Photocatalysts
(all 'bulk')

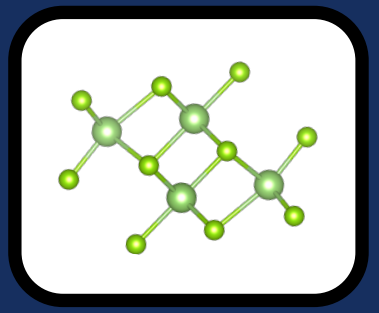
ZnTe



SiAs



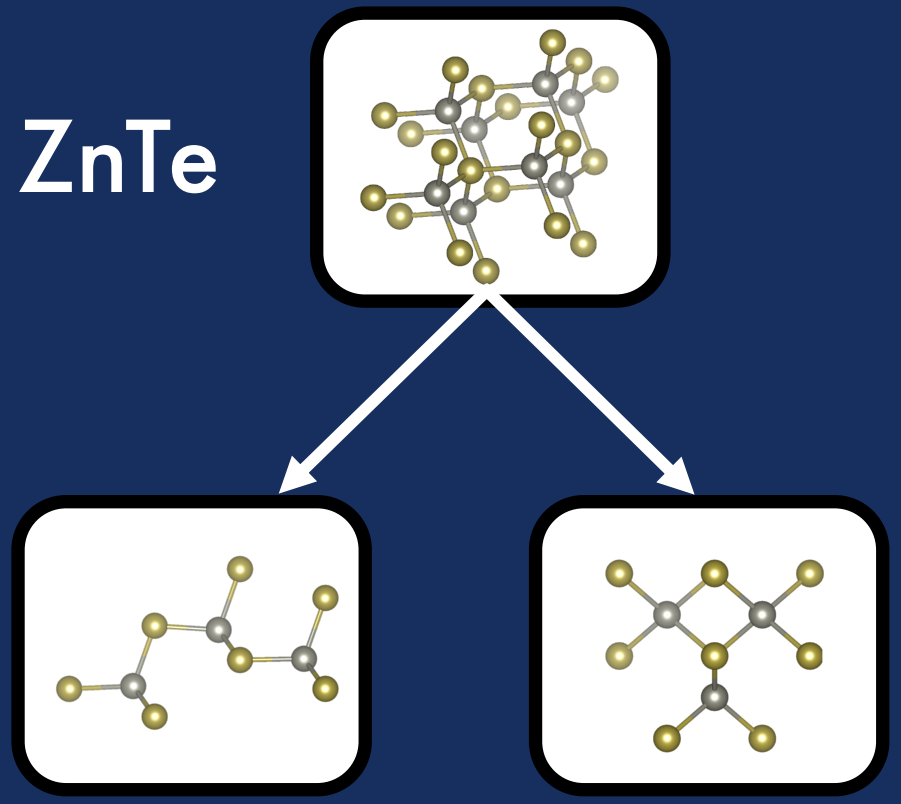
GaSe



A. Singh, J. Montoya, J. Gregoire, K. Persson, *Nat. Comm. 2019*

Could alternate structures do better?

ZnTe

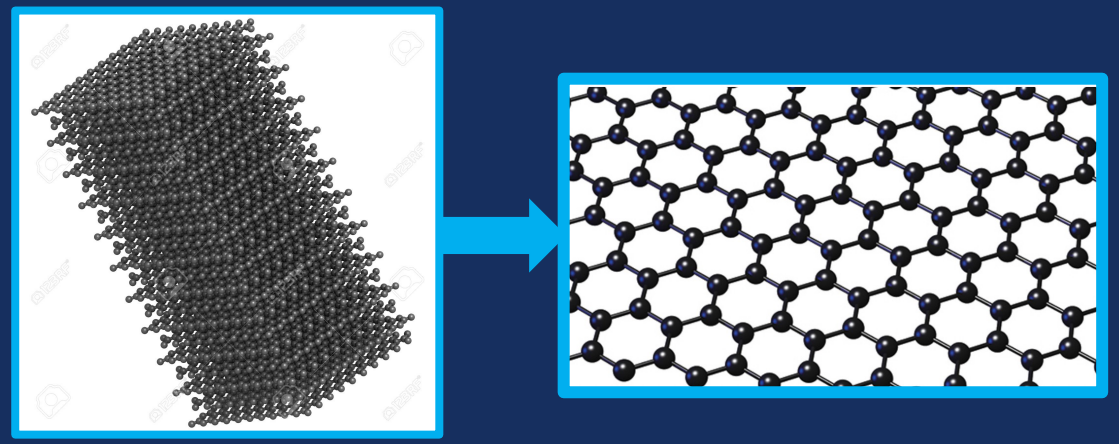


Better Light Absorption

Larger Surface Area

Better Excited e^- Properties

"Two-Dimensional" Structures

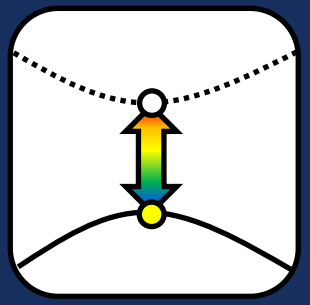


Materials in a sheet of atoms one atom, or layer thick

3D = "Bulk"

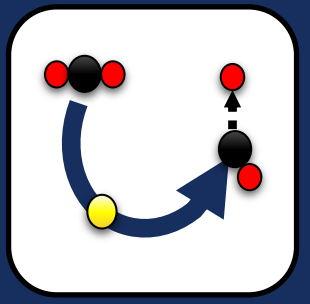
2D= "Monolayer"

Better Light Absorption



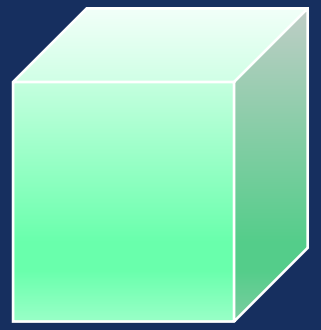
Larger Surface Area

Better Excited e^- Properties



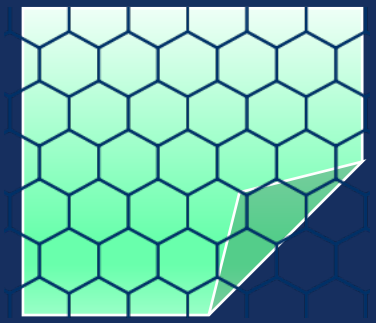
THE GOAL:

1. **Feasibility: For bulk structures, determine if two-dimensional phases can exist.**



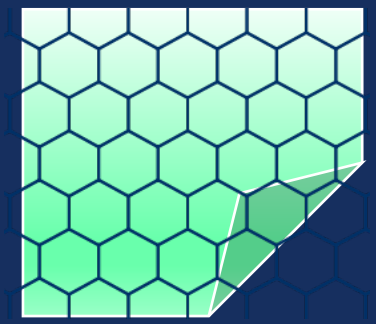
THE GOAL:

1. **Feasibility: For bulk structures, determine if two-dimensional phases can exist.**



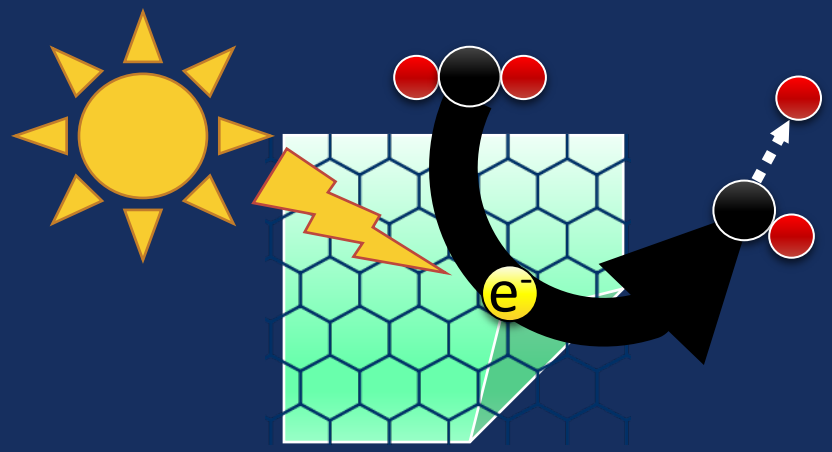
THE GOAL:

1. **Feasibility: For bulk structures, determine if two-dimensional phases can exist.**
2. **Suitability: For the resultant 2D phases, predict their catalytic properties.**

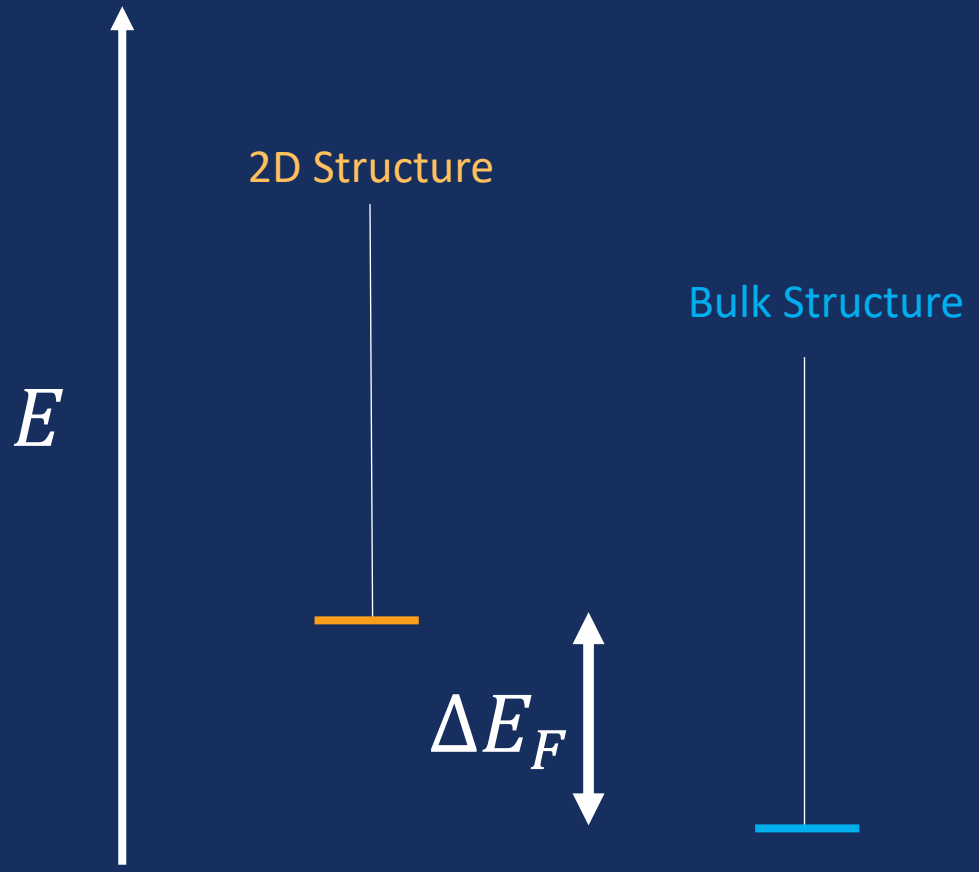


THE GOAL:

1. **Feasibility**: For bulk structures, determine if two-dimensional phases can exist.
2. **Suitability**: For the resultant 2D phases, predict their catalytic properties.

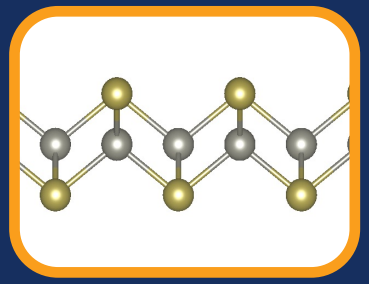


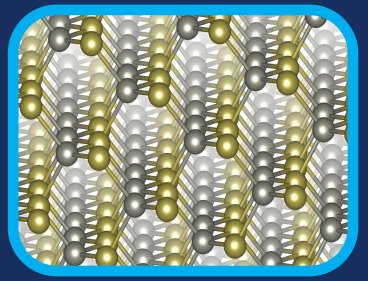
Feasibility *via* Stability



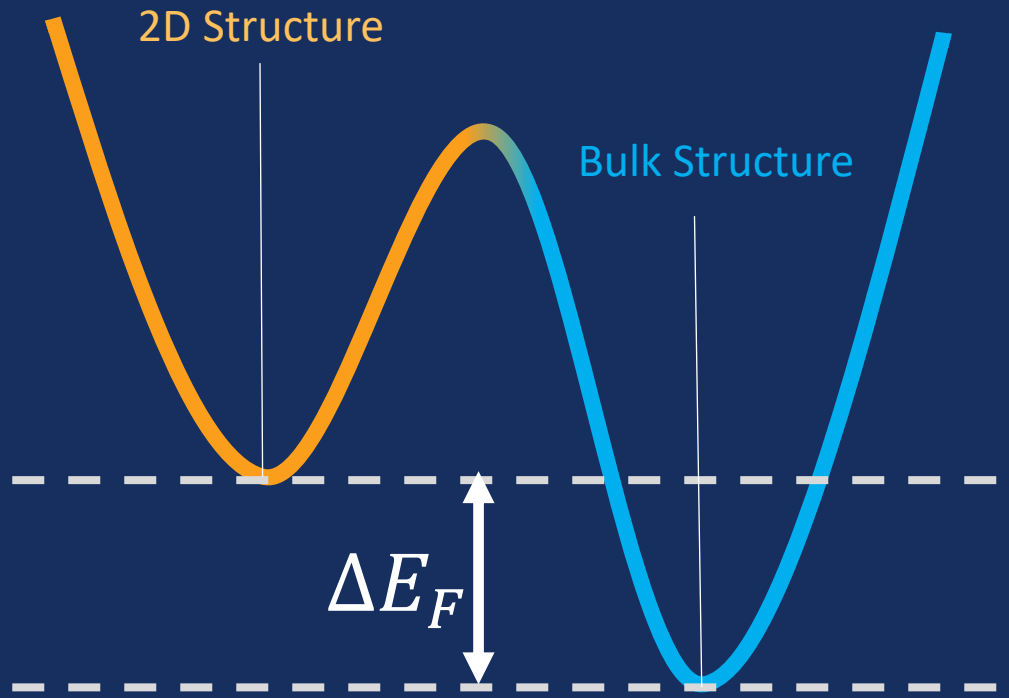
Thermodynamics

Rank candidate structures by energy
Compare ΔE_F vs. bulk



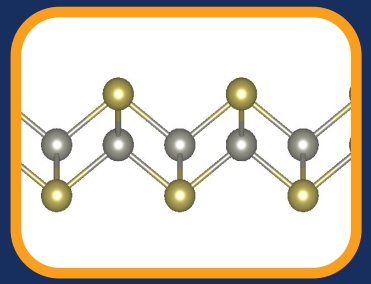


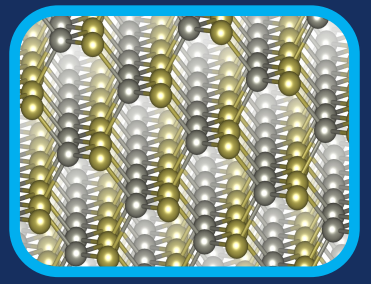
Feasibility *via* Stability



Thermodynamics

Rank candidate structures by energy
Compare ΔE_F vs. bulk



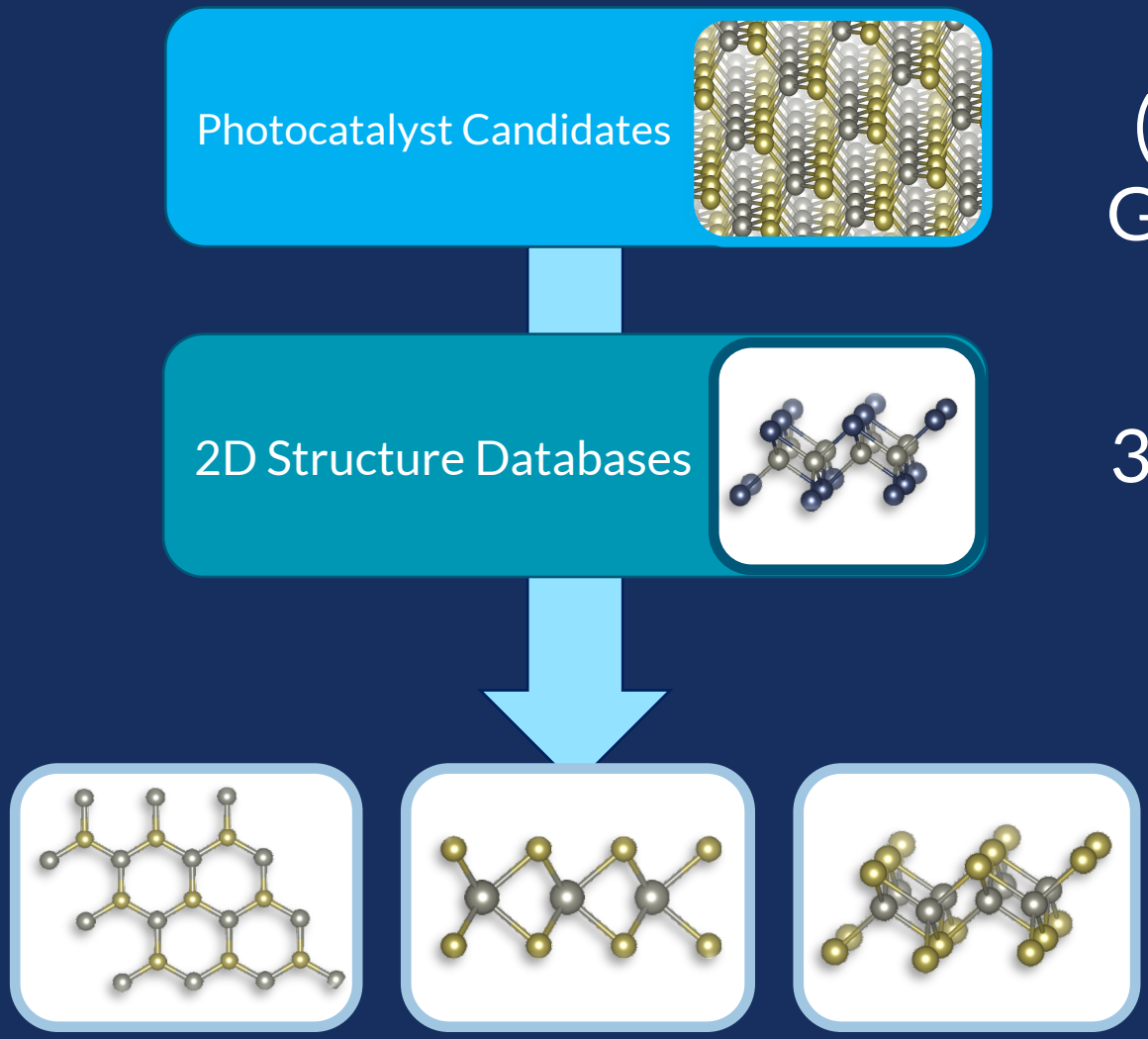


Heuristic Cutoff Energy (From literature[1]):

$$\Delta E_{f/atom} \leq 200 \text{ meV}$$

[1] B.C. Revard, W.W. Tipton, A. Yesypenko, R.G. Hennig, PRB 93 (2016)

Stability Screening: Thermodynamics

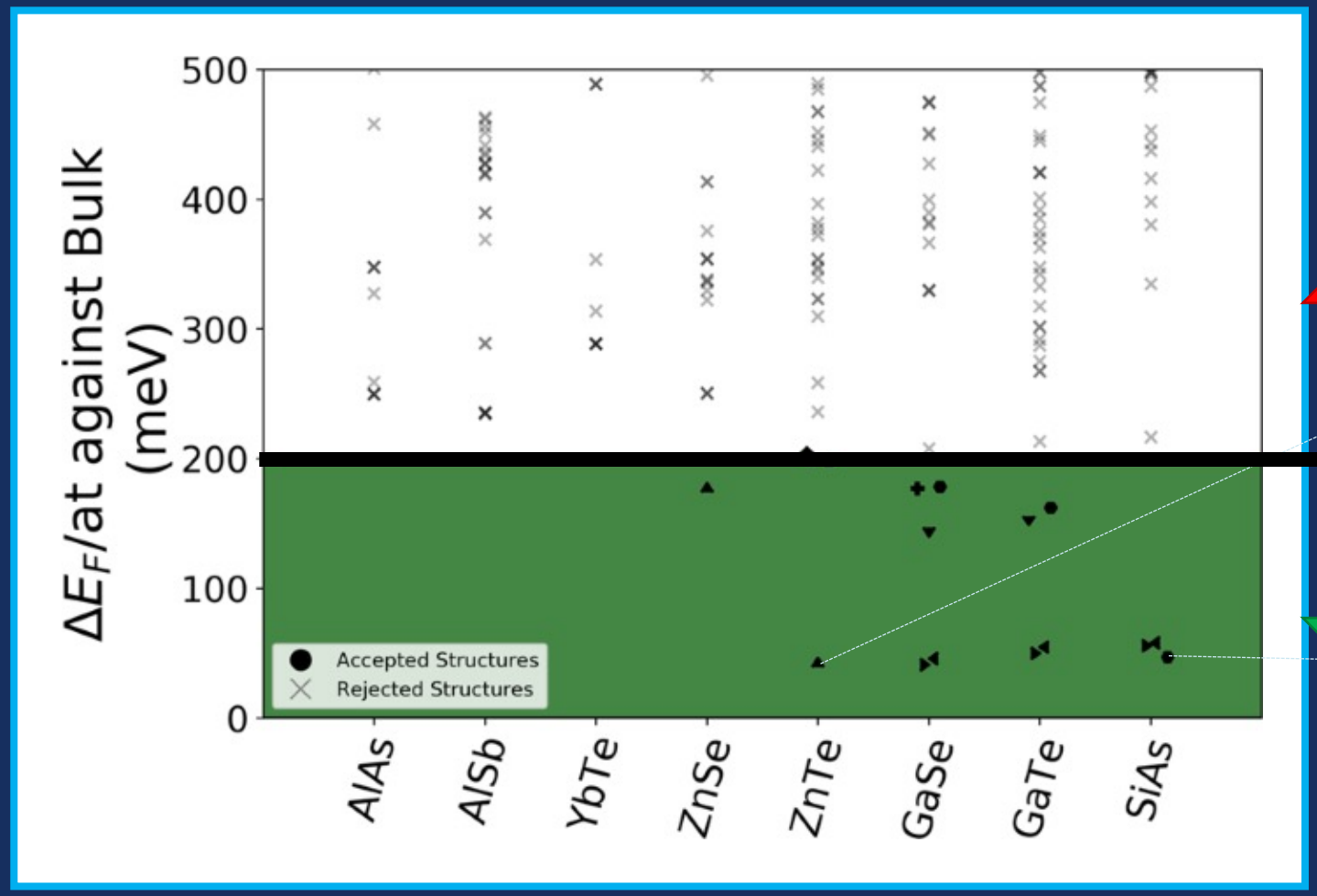


8 Binary Compounds
(SiAs, AlAs, AlSb, YbTe,
GaSe, GaTe, ZnTe, ZnSe)

36 Unique 2D Structures

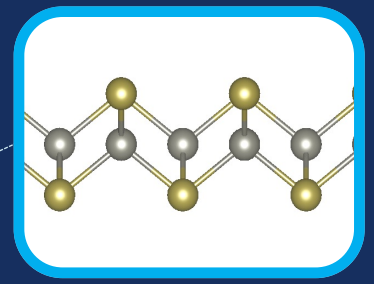
288 Candidate
2D Forms
Of Compounds

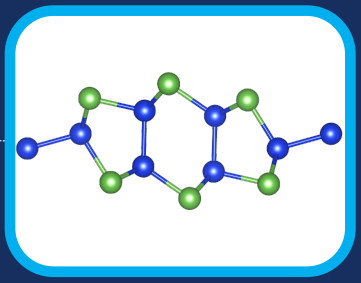
Thermodynamically Stable Forms



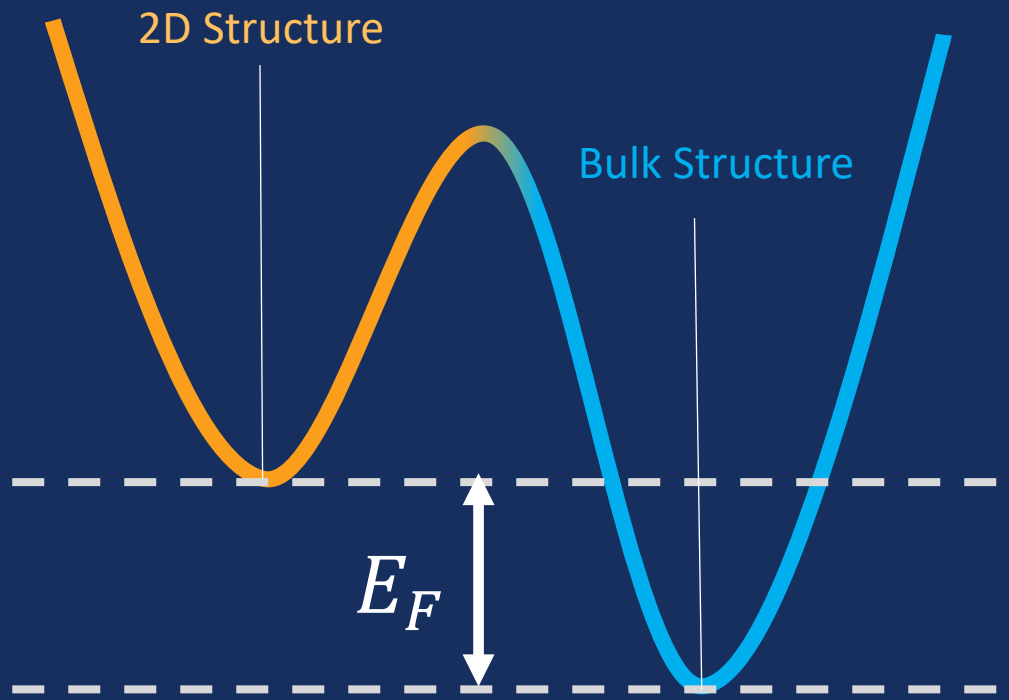
Exceeded Cutoff Energy

Below Stability Cutoff

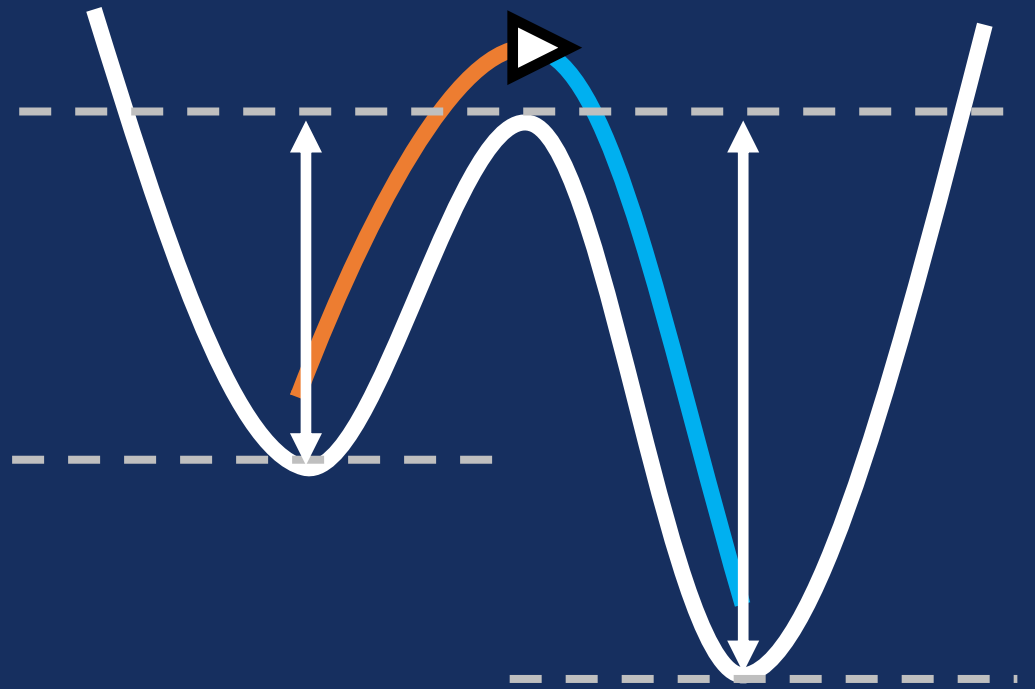




Stability Screening: Dynamics

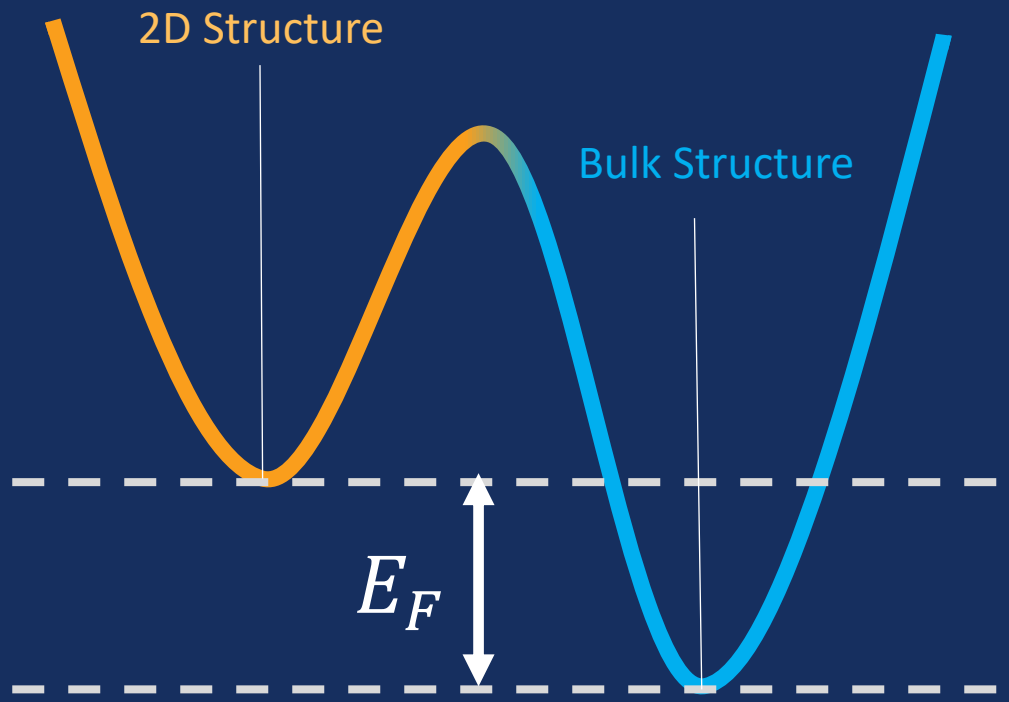


Thermodynamics

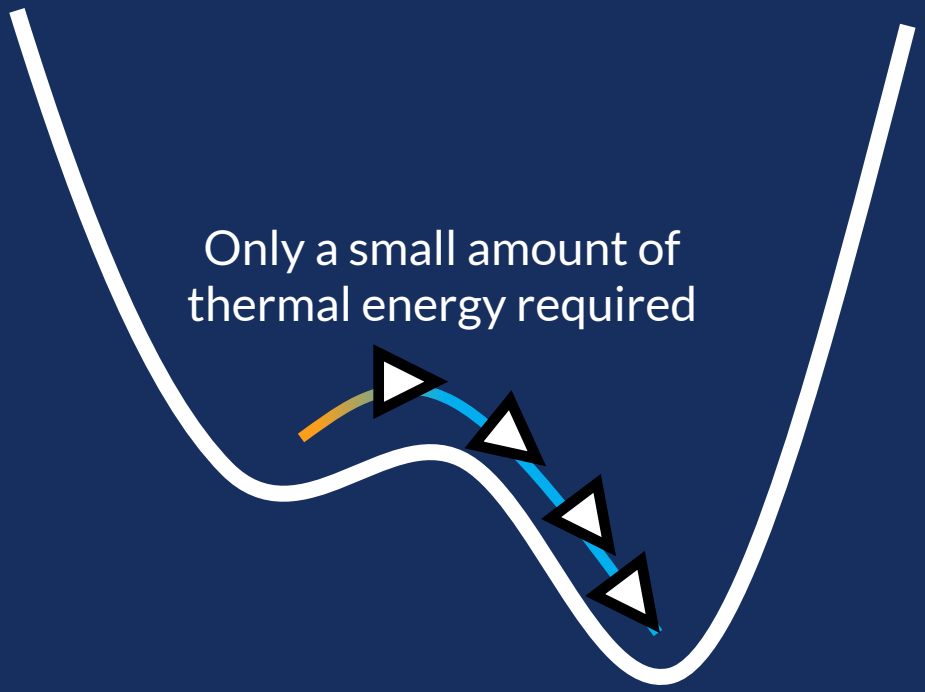


Kinetics

Stability Screening: Dynamics

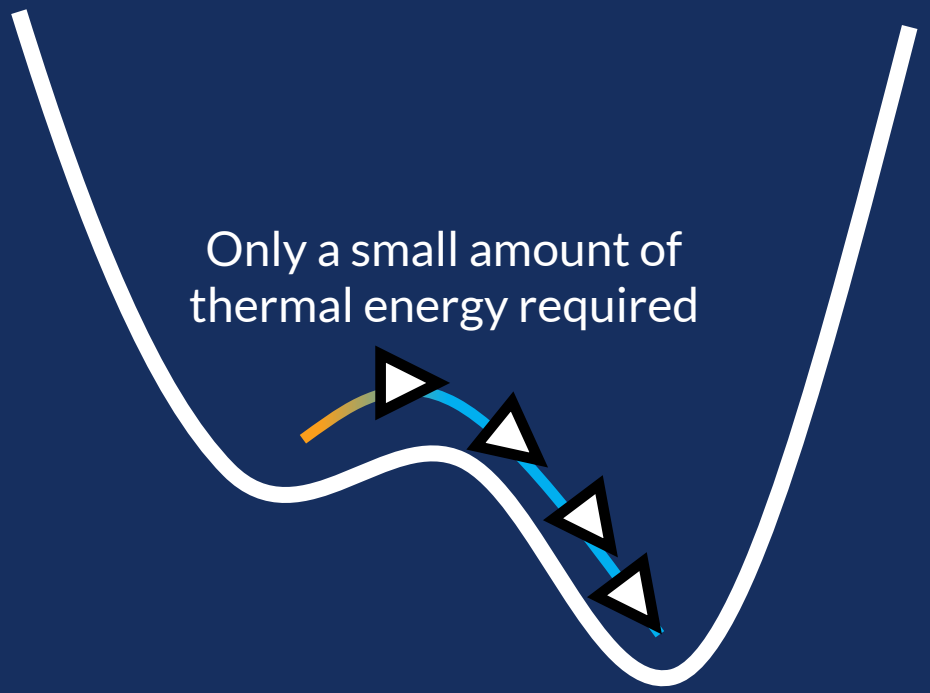


Thermodynamics



Dynamic Instability

Stability Screening: Dynamics



Dynamic Instability

Vibrations for Dynamic Instability

Phonon Spectra

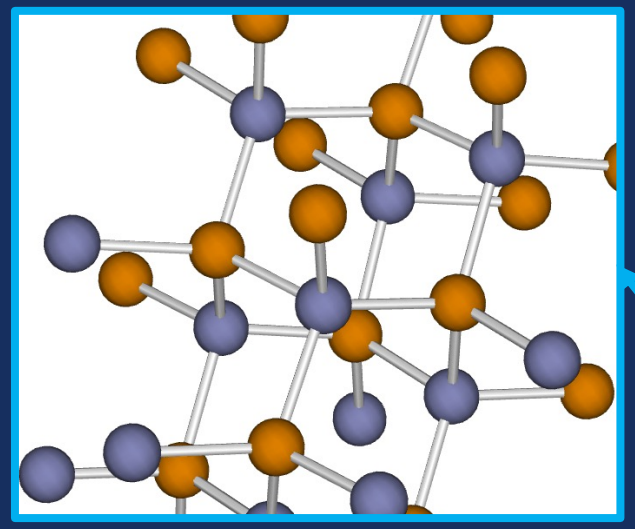
- ✓ Easy to check for dynamic instability

$$\boldsymbol{D}(\vec{q})\vec{u}_{n\vec{q}} = \omega_n^2(\vec{q})\vec{u}_{n\vec{q}} \quad (1)$$

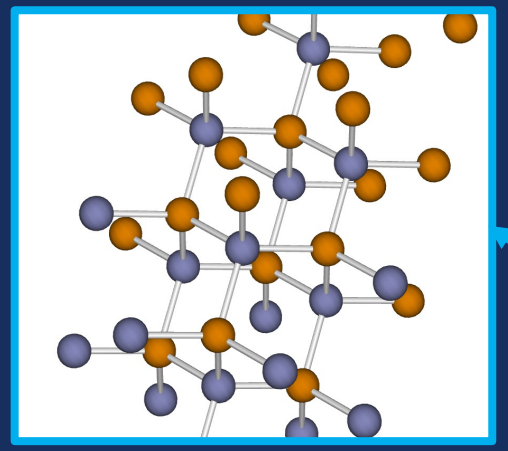
$$\omega_n^2 > 0: Stable$$

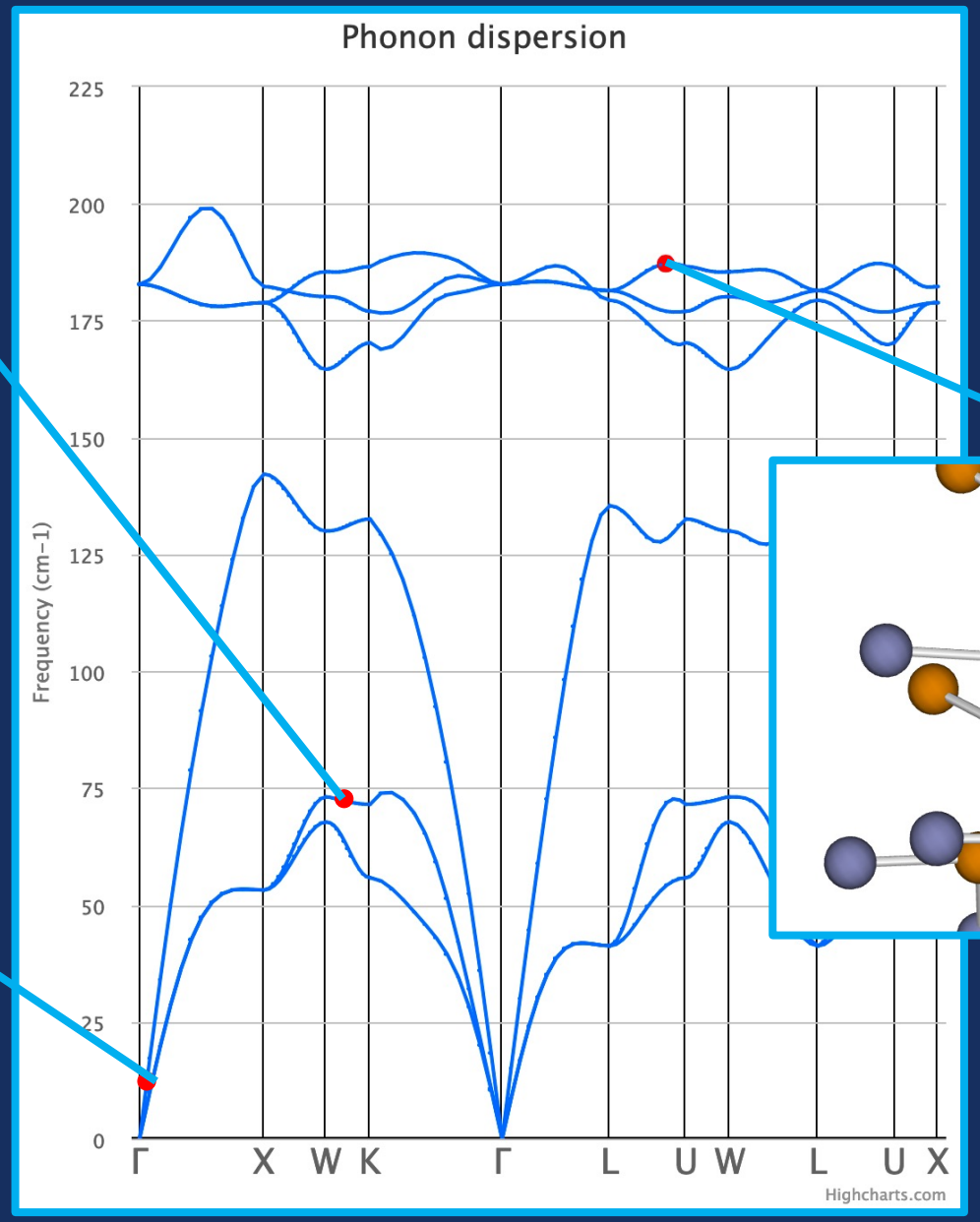
$$\omega_n^2 < 0: Unstable$$

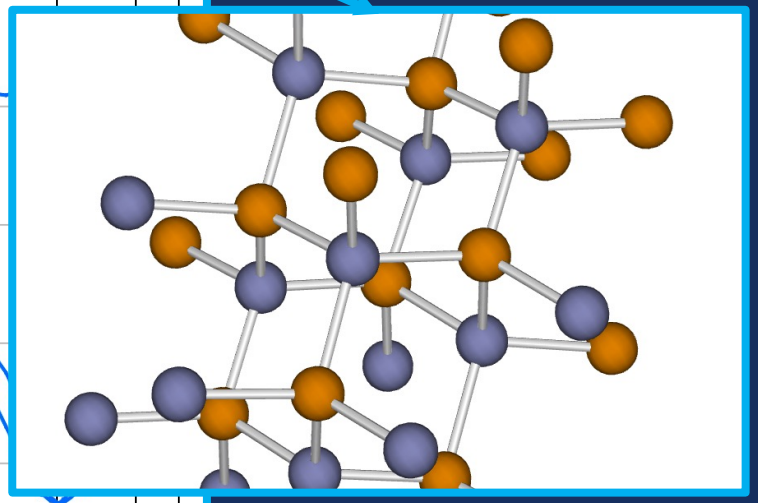
Bulk ZnTe



$\omega_n^2(\vec{q})\vec{u}_{n\vec{q}}$



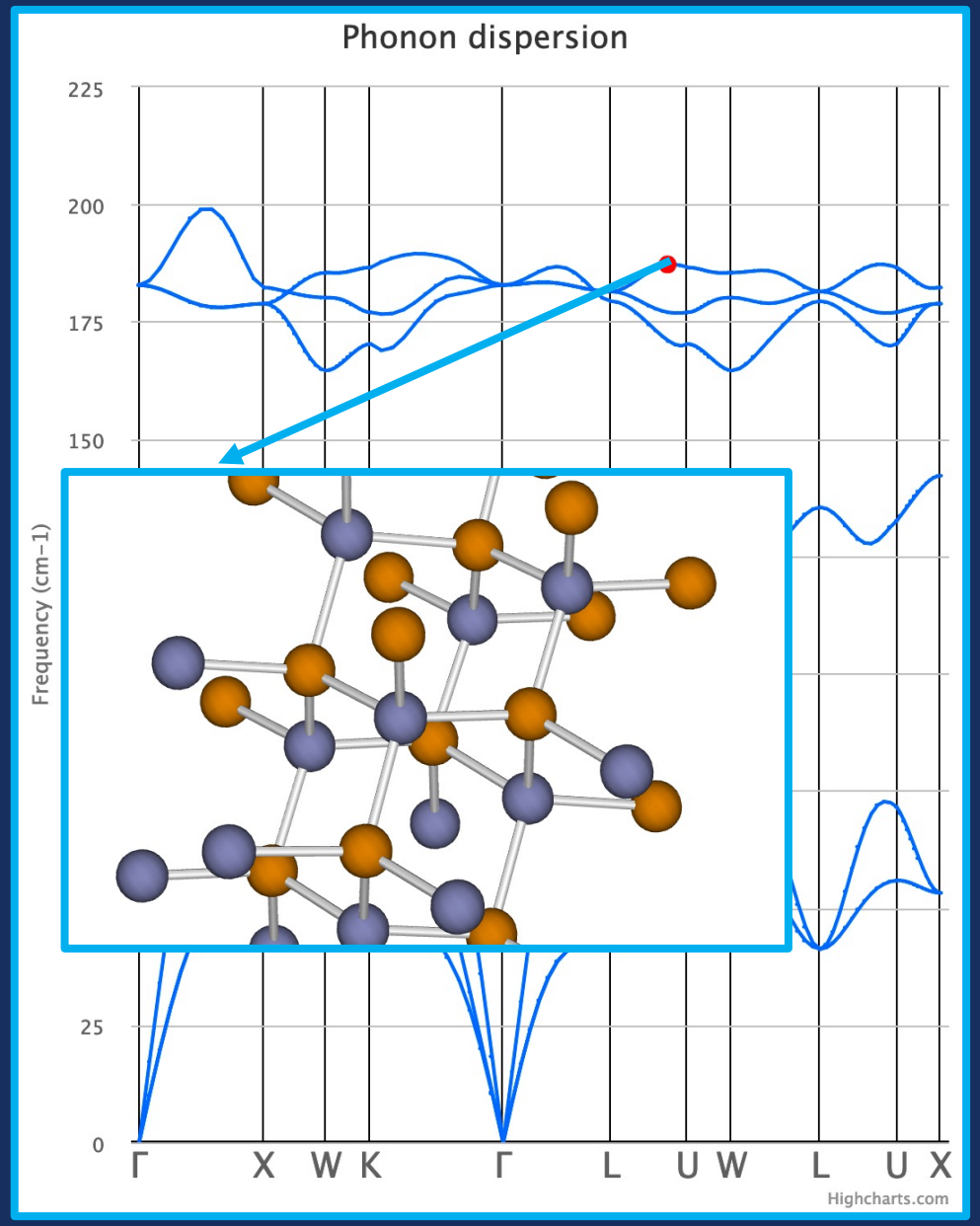




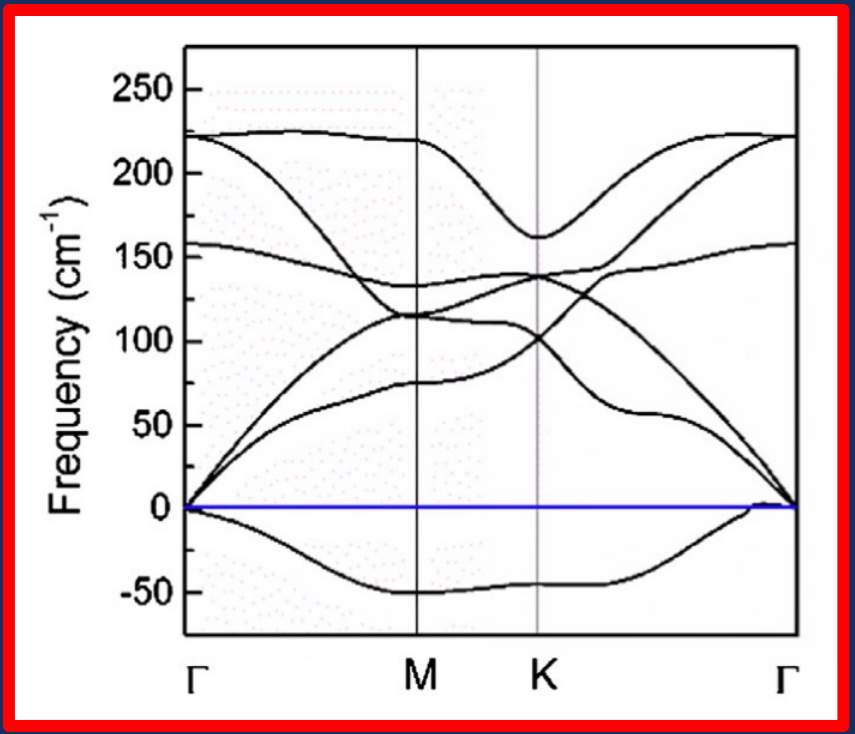
http://henriquemiranda.github.io, Materials Project, ZnTe (mp-2176)

Bulk ZnTe

$\omega_n^2(\vec{q})\vec{u}_{n\vec{q}}$



Example From Strained Silicene



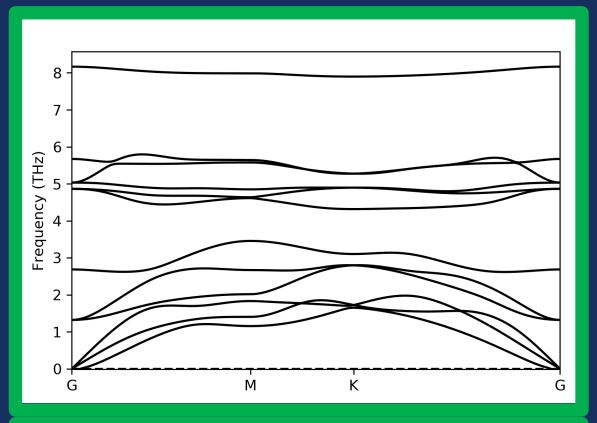
http://henriquemiranda.github.io, Materials Project, ZnTe (mp-2176)

Unstable Phonon Mode from Yang et al. Comp. Mat. Sci. 95 (2014)

Dynamical Stability Found!

GaTe

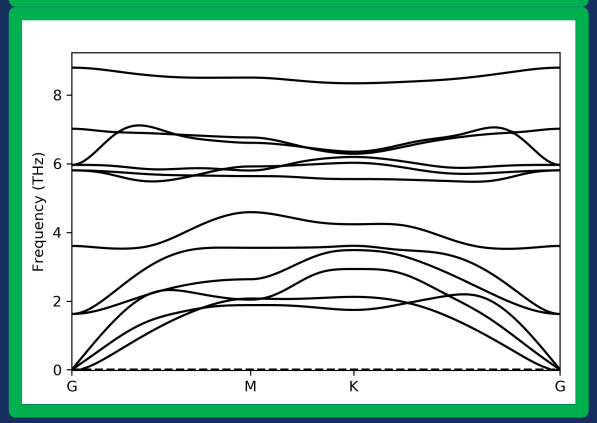
x 4 structure



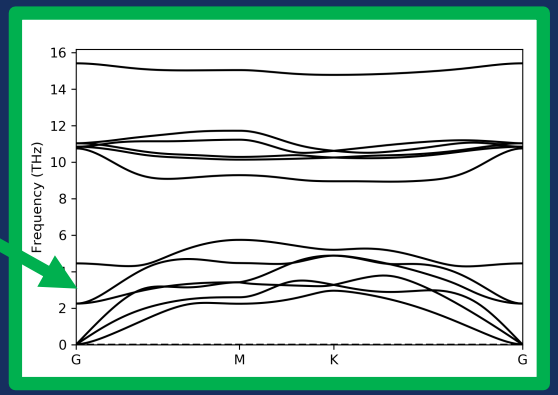
GaSe

x 4 structure

1 unstable

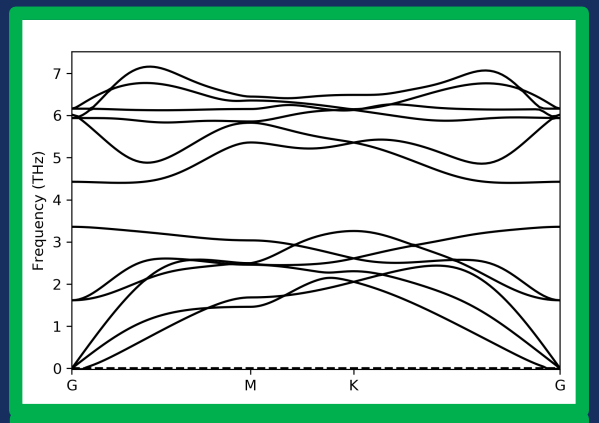


$\omega_n^2 > 0: Stable$



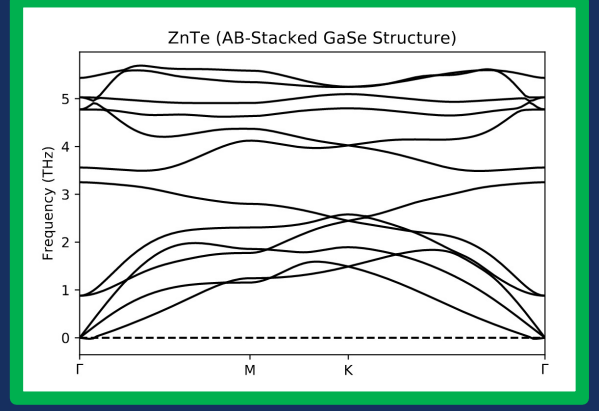
ZnSe

x 1 structure



ZnTe

x 2 structure



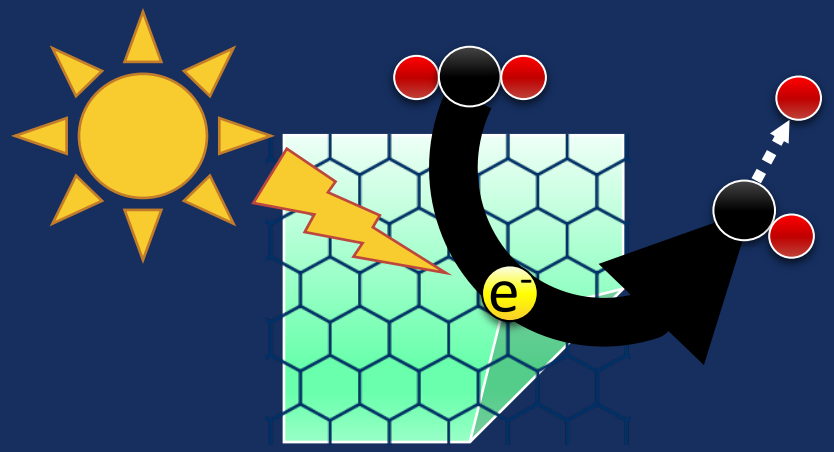
SiAs

x 3 structure

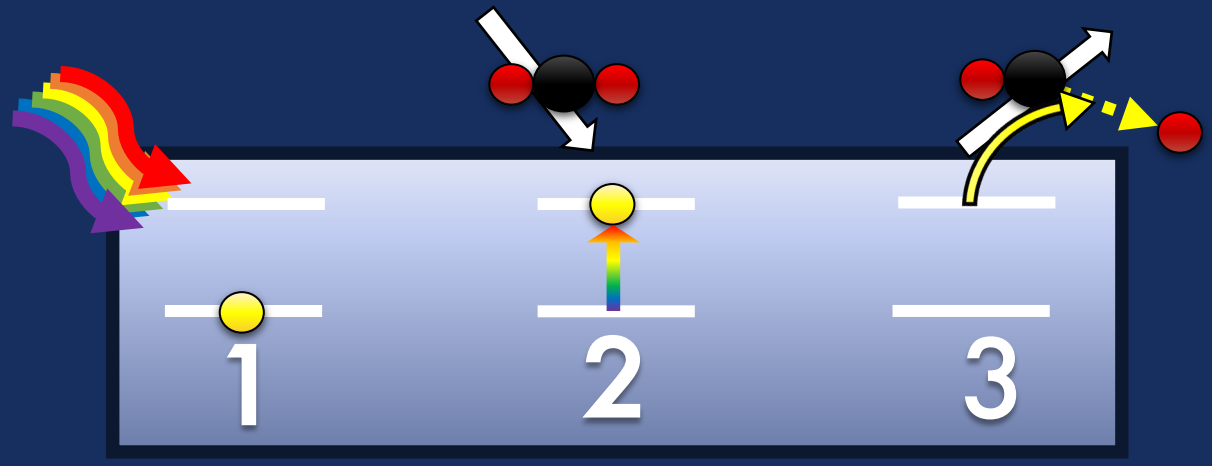
THE GOAL:

✓ **Feasibility: For bulk structures, determine if two-dimensional phases can exist.**

❑ **Suitability: For the resultant 2D phases, evaluate their catalytic properties.**



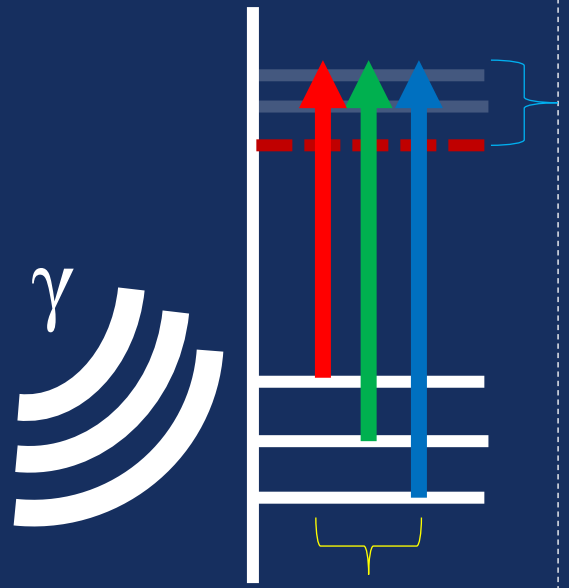
Catalytic Suitability in Three Steps



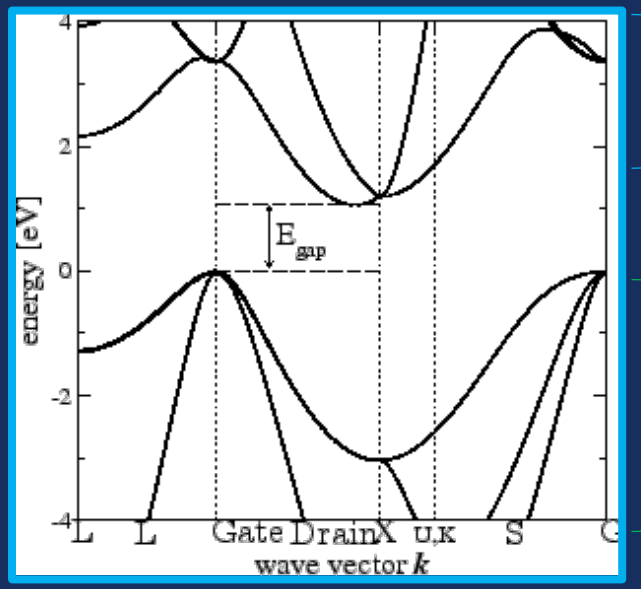
1. Light Absorption
2. Sufficiently Energetic e^-
3. Reactant Binding

Light Absorption in Materials

Atoms : Crystals
Orbitals : Bands



Varying E



Unoccupied

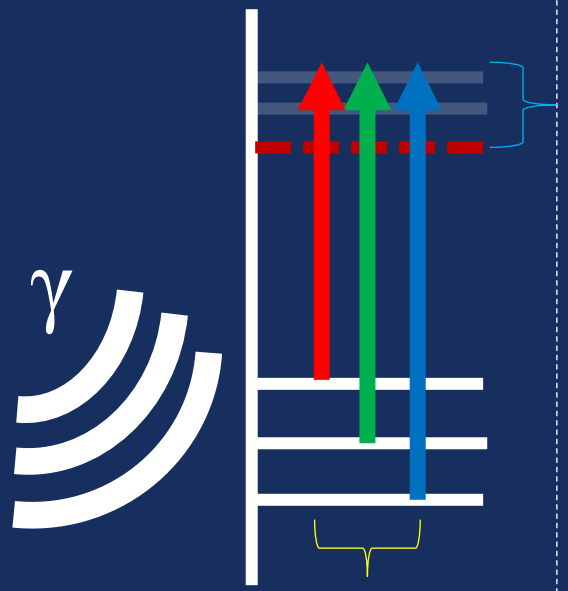
Occupied

Silicon Band Structure

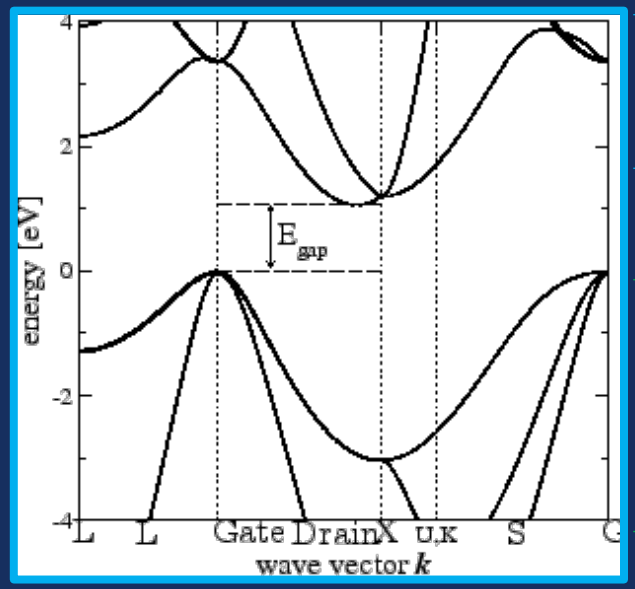
Light Absorption in Materials

Atoms : Crystals
Orbitals : Bands

For solar light harvesting…
Band gap size should correspond to visible light energy
(Direct gap preferrable)



Varying E

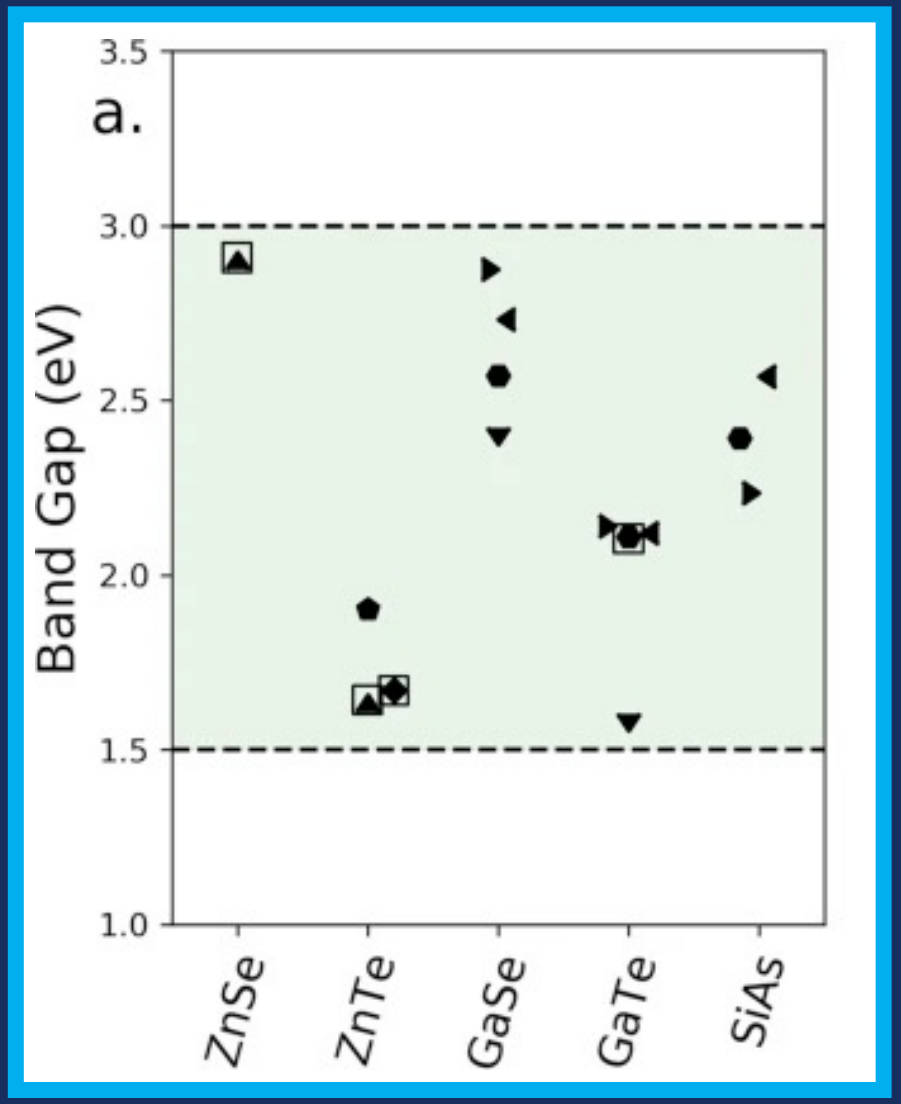


Unoccupied

Occupied

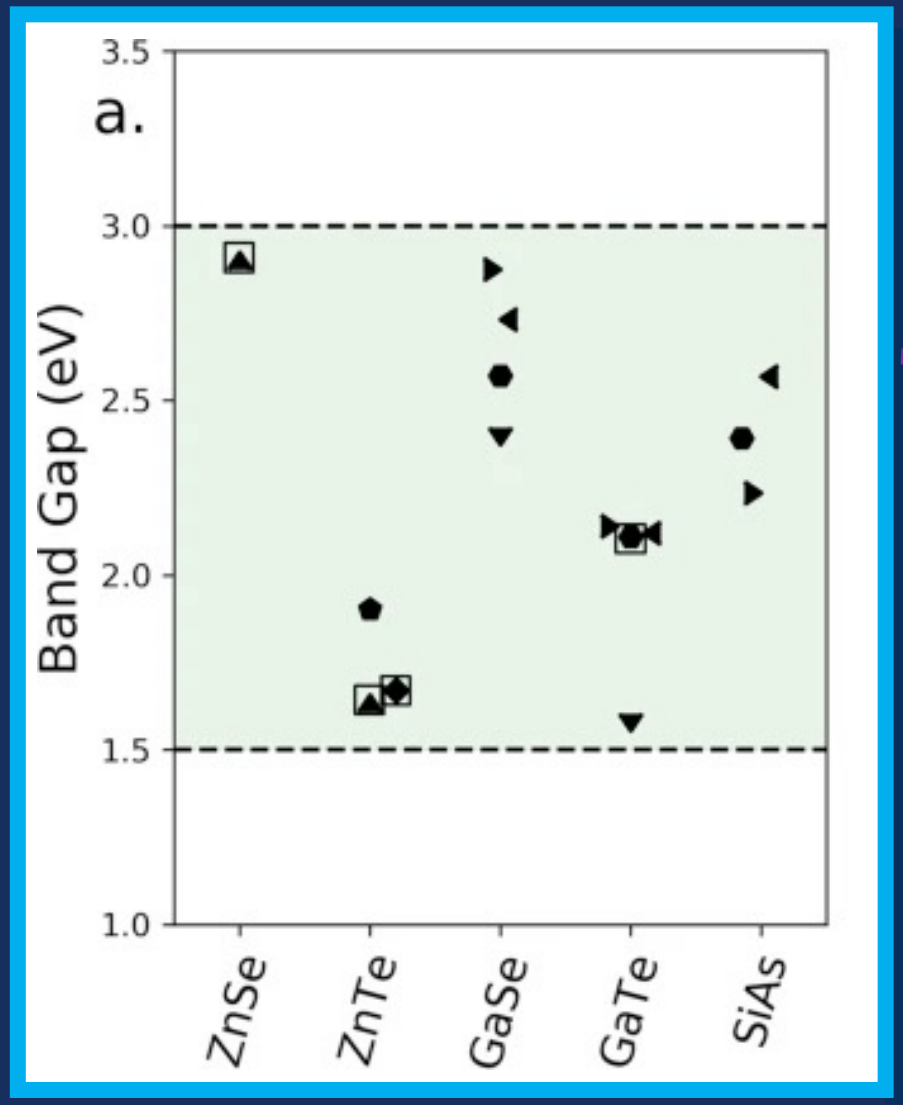
Silicon Band Structure

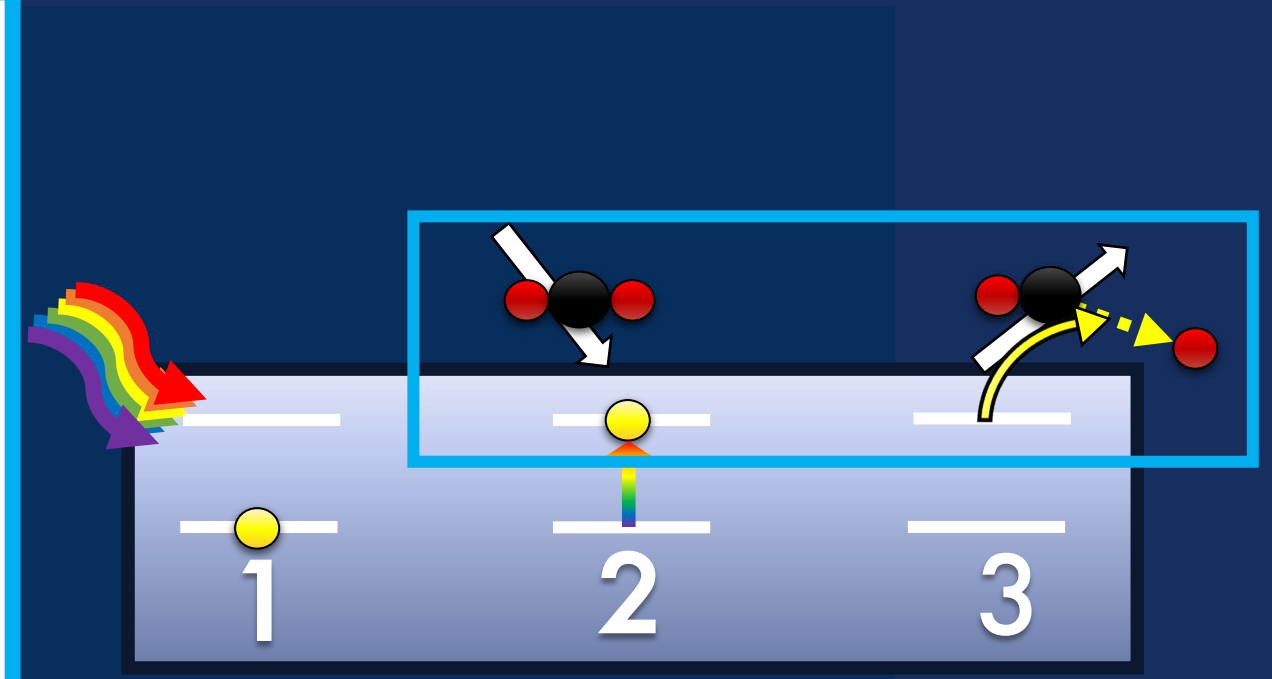
Band Gaps Lie In Visible Spectrum



Boxed: Direct Gap
Note: Computed using HSE06 functional

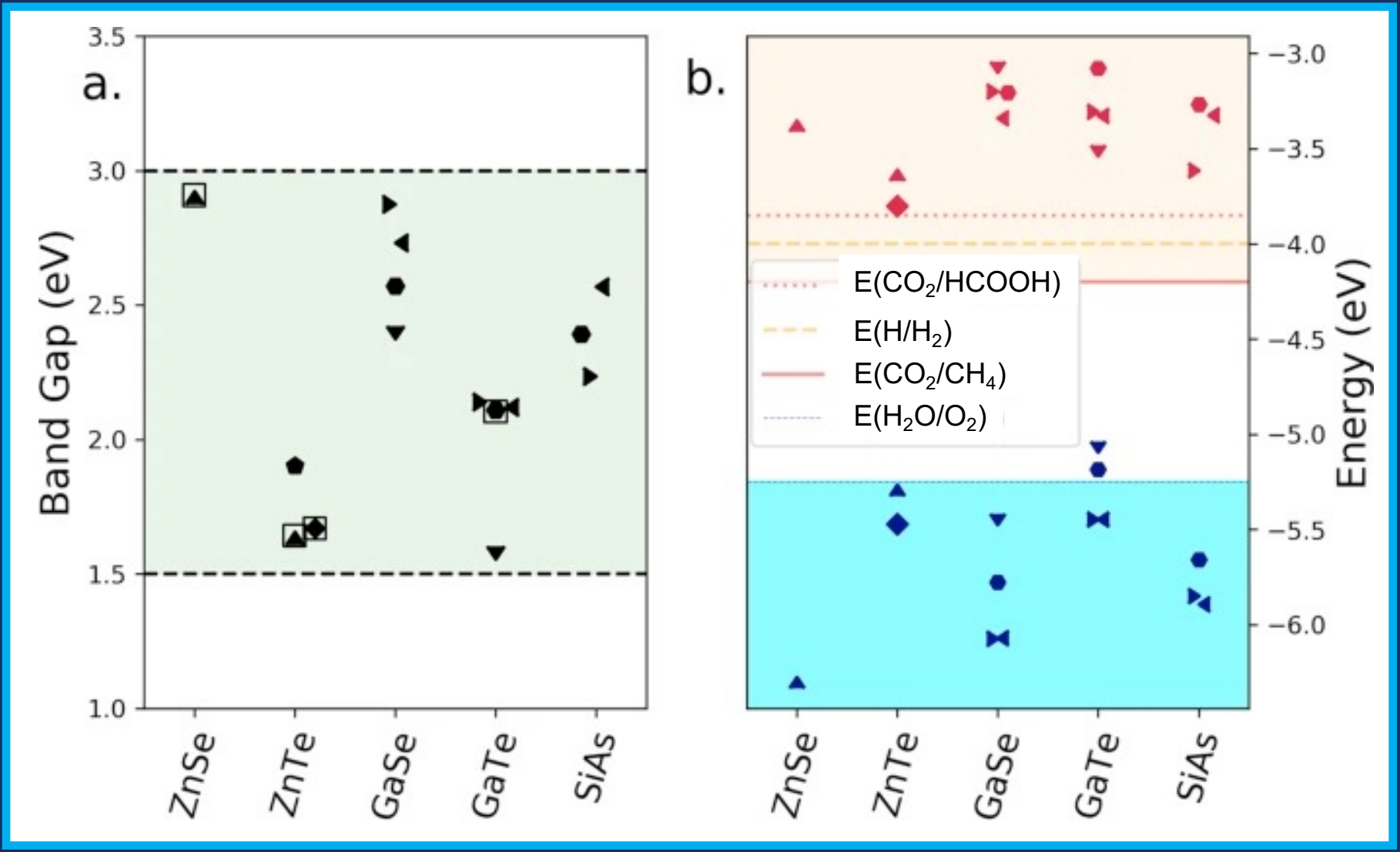
Excited Electrons must be high-energy!





Boxed: Direct Gap
Note: Computed using HSE06 functional

Band Edges Facilitate CO_2 Reduction

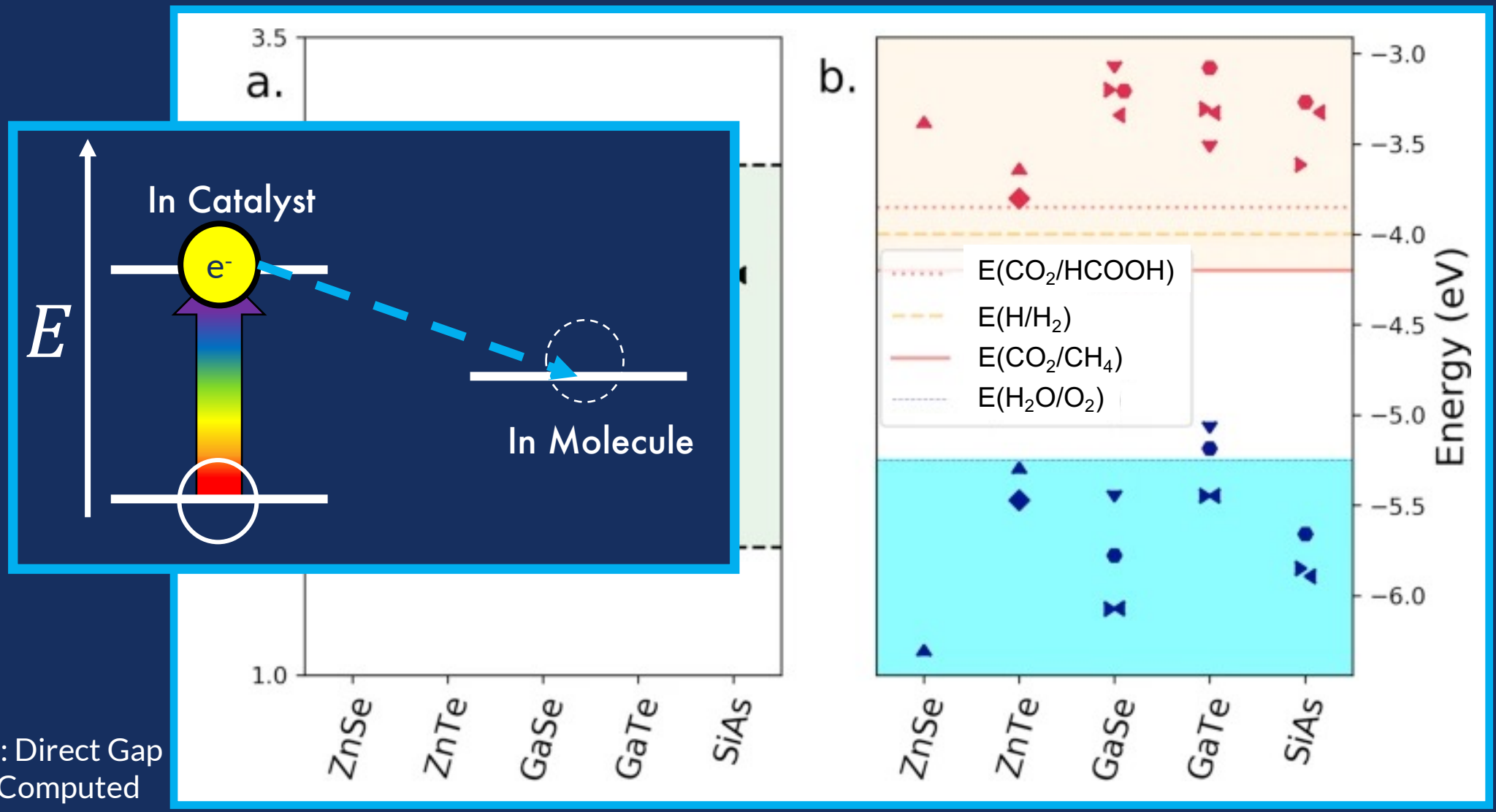


Excited states are *above* reaction energies

→ they may participate in reactions

Boxed: Direct Gap
Note: Computed using HSE06 functional

Band Edges Facilitate CO_2 Reduction



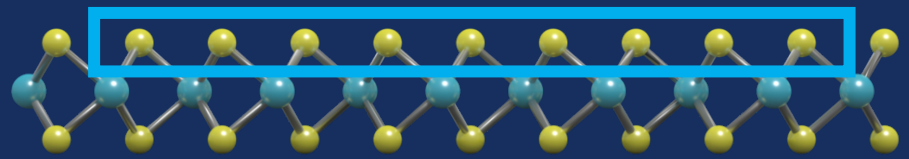
Excited states are *above* reaction energies

→ they may participate in reactions

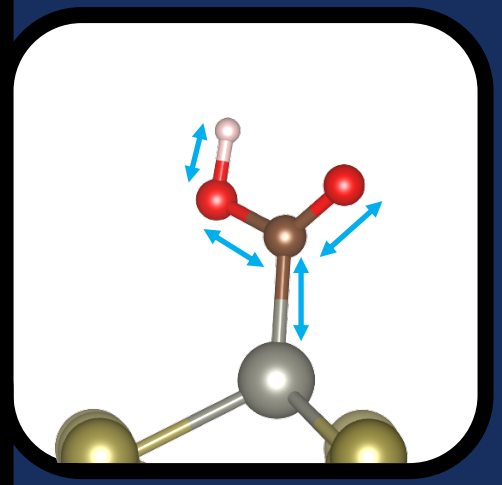
Boxed: Direct Gap
Note: Computed using HSE06 functional

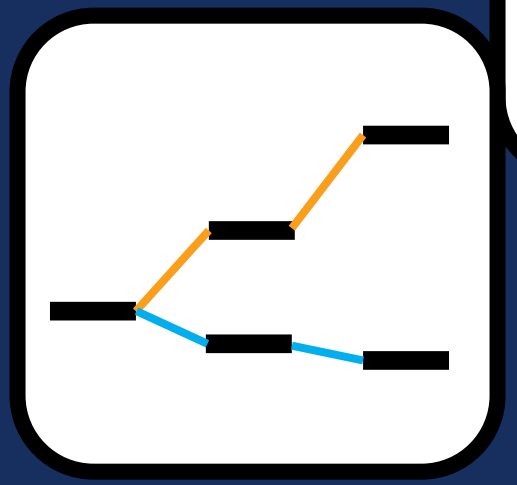
Calculation of "Theoretical Overpotential"

- Compute binding energies of reactants on 'basal plane' (top of monolayer)



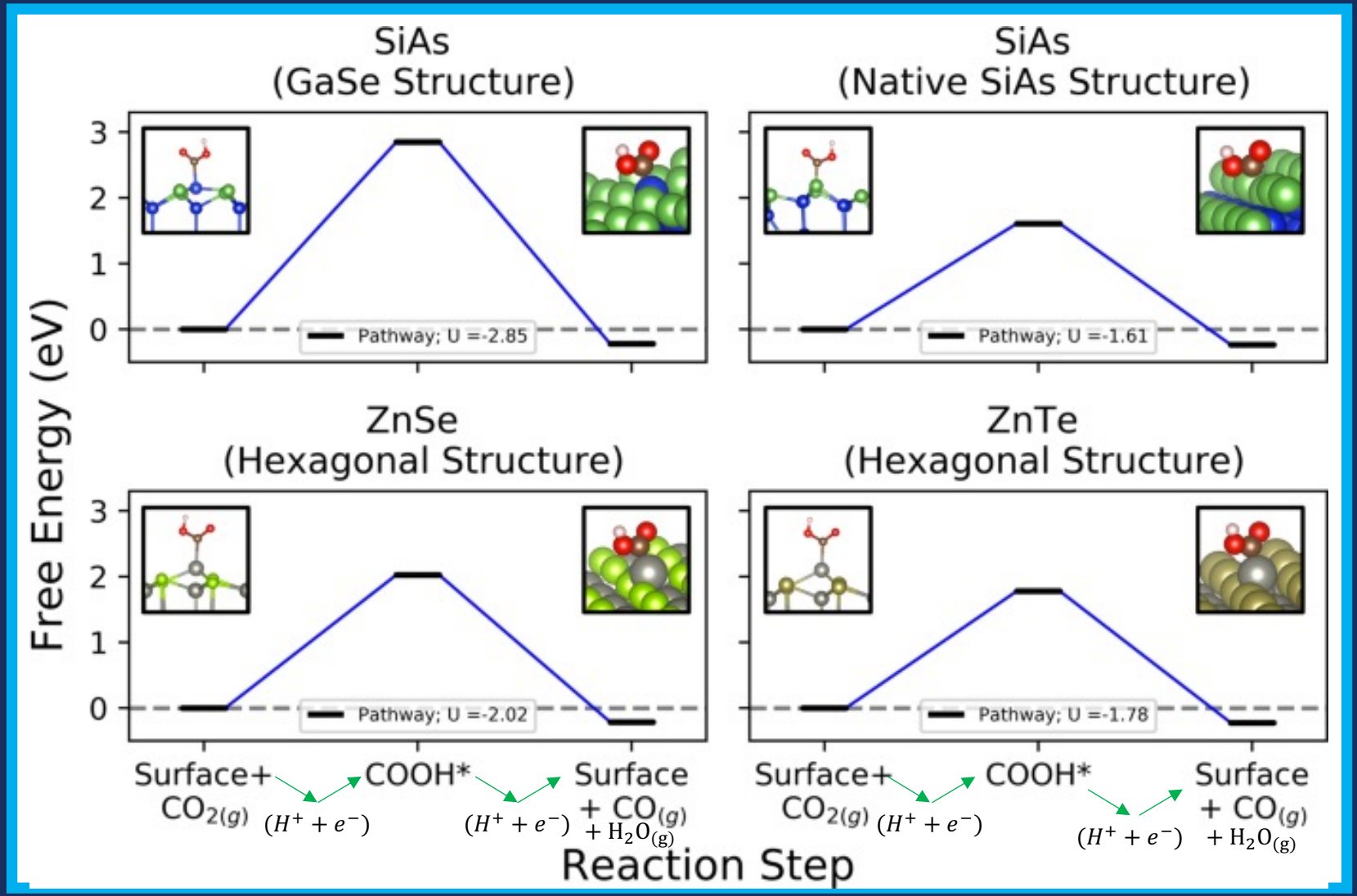
- Account for contributions to free energy
- Compare reaction pathway; determine 'in the dark' bias voltage to induce reaction





Basal Plane Binding Energies

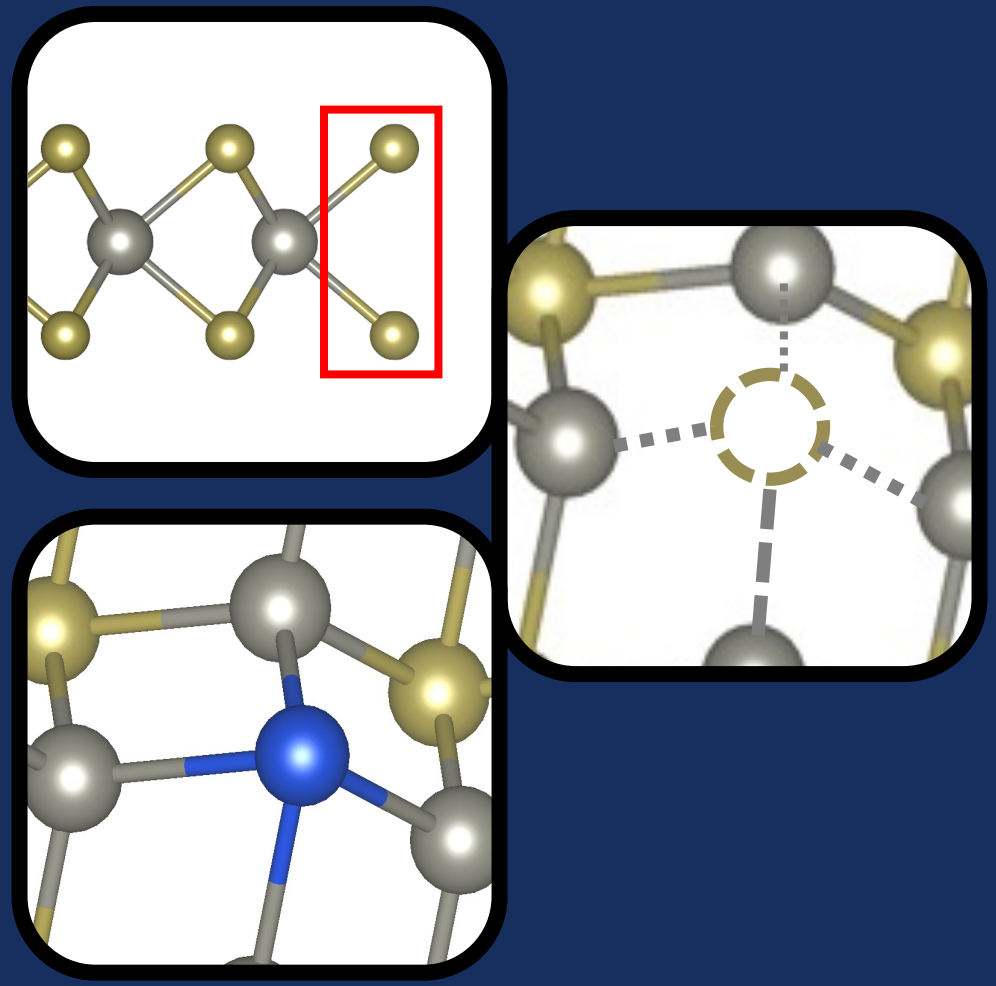
- This is “in the dark”; no illumination present
- Height of barrier: positive binding energy
- May change when excited electrons present
- Detailed evaluation of mechanism would be study in its own right



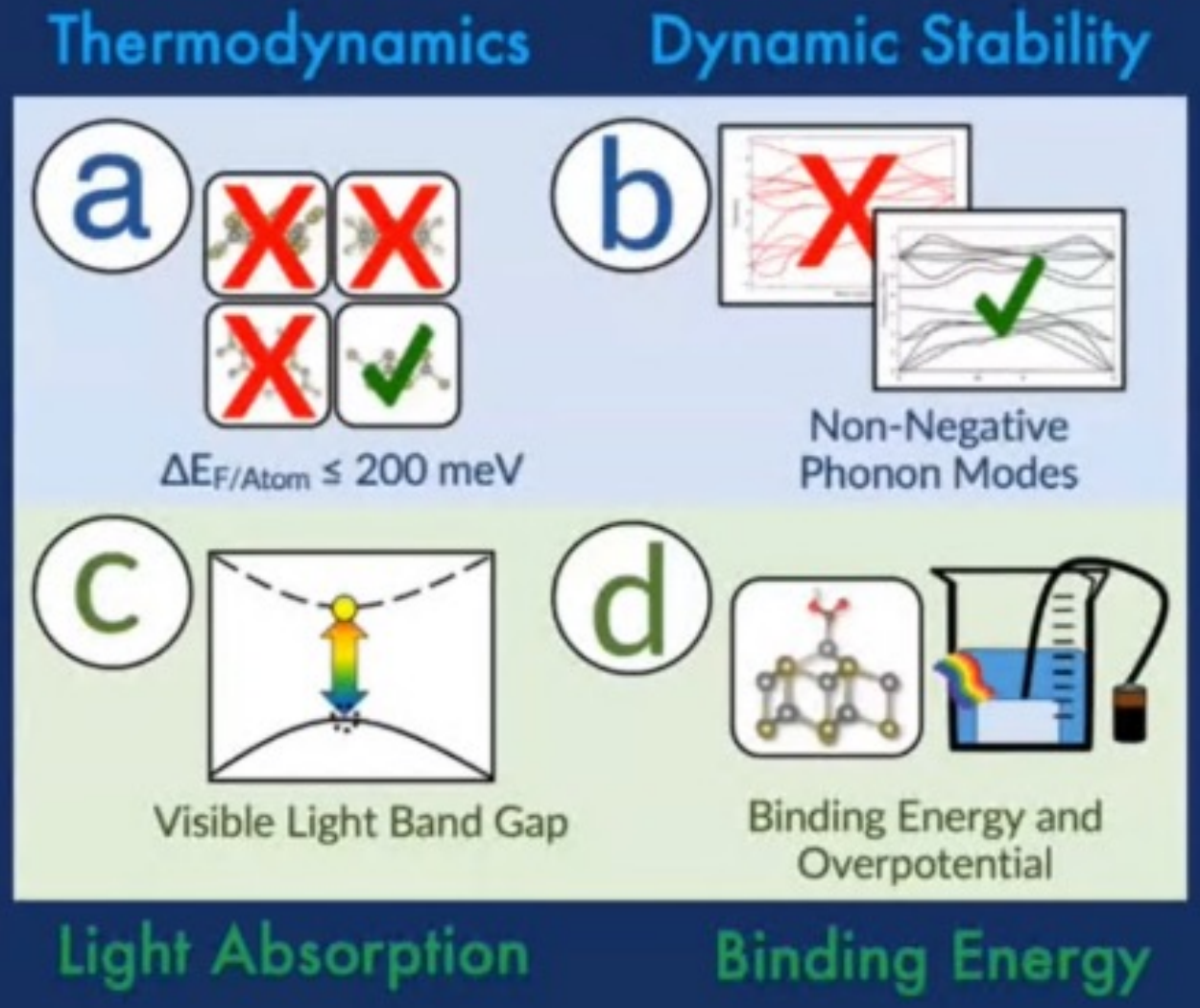
STP
(CO, CO_2, H_2)
3k Pa H_2O

Next Steps: Improving Reactivity

- Model reactivity of surface edges
- Preliminary results - vacancies on surface bind stronger
- Explore role of different dopants



Conclusions



S.B. Torrisi, A. Singh, J. Montoya, T. Biswas, K.A. Persson, NPJ 2D Mat. & Appl. 2020

Acknowledgements

Boris Kozinsky

Efthimios "Tim" Kaxiras

Jennifer Hoffman

Cynthia Friend

DOE CSGF

Materials Intelligence Research

Catalysis Subgroup

Boris Kozinsky

Jon Vandermause

Simon Batzner

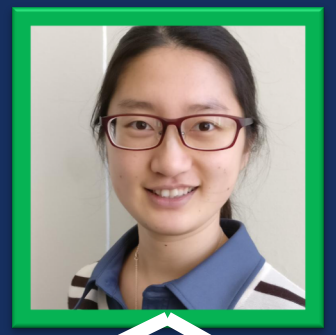
Anders Johansson

Jenny Coulter

Yu Xie

Urban Axes 2019

Lixin Sun



David Lim

Cameron Owen

Isabel Diersen

Summer 2018

The Materials Project

Summer 2019


Linda Hung


Santosh Suram


Abe Anapolsky

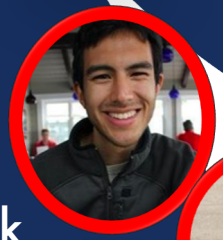

Jens Hummelshøj

Brian Storey


Daniel Schweigert

TOYOTA
RESEARCH INSTITUTE


Joey Montoya (again)


Patrick Herring


Chirru Gopal


Ha-Kyung Kwon


Murat Aykol

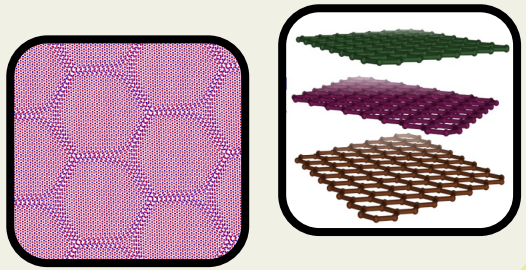

Junko Yano (LBNL)


Matt Carbone (Columbia Chemistry, Rochester 2016)

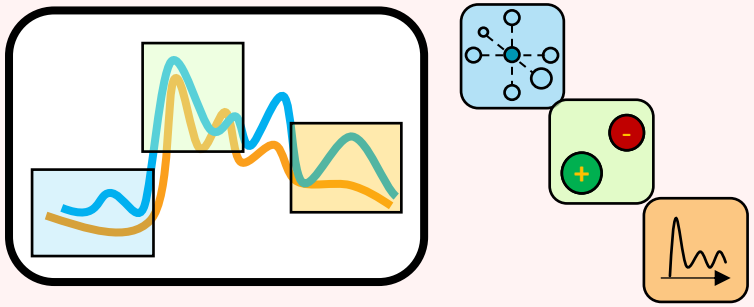

Aditi Krishnapriyan

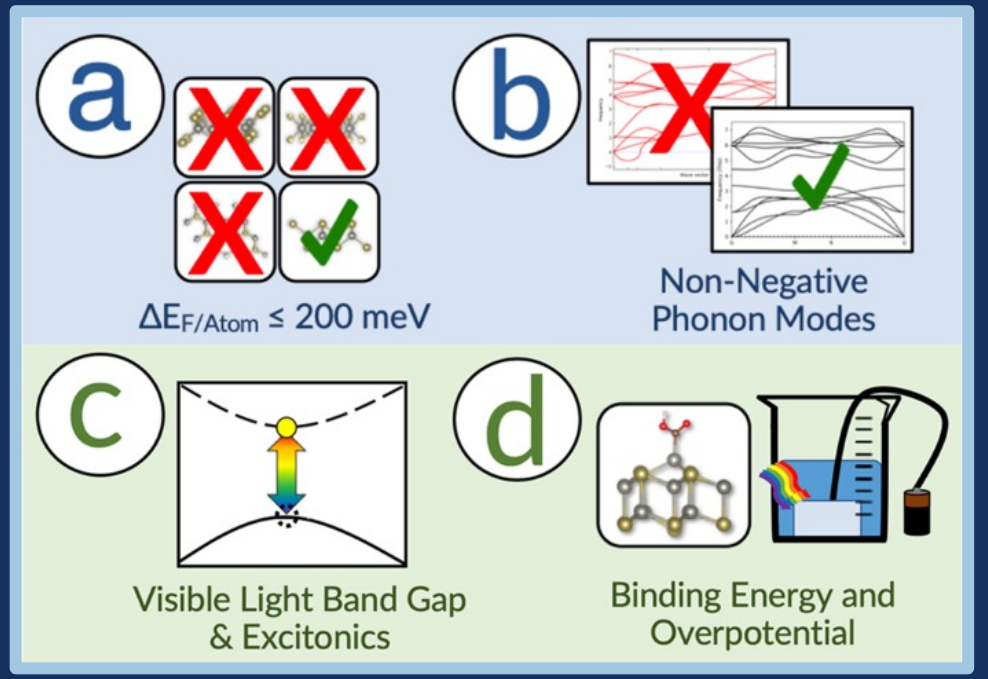
Questions?

Metals, 2D Materials

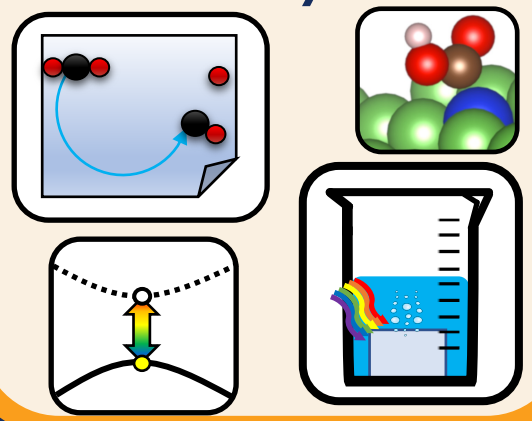


X-Ray Absorption Spectroscopy

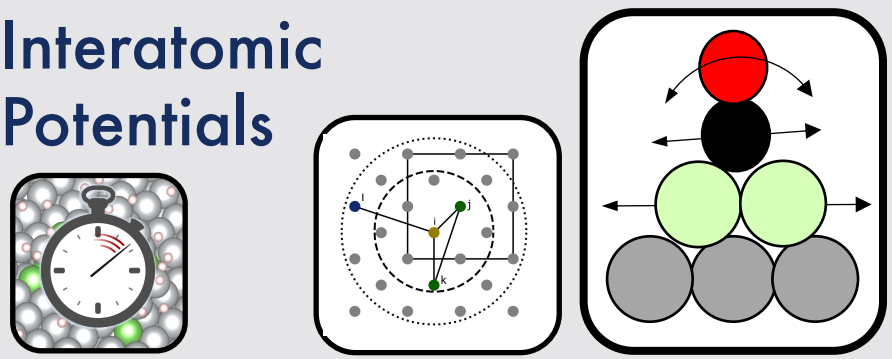




Catalysis



Machine-Learned Interatomic Potentials



Thank you