MENG 35510/25510/CHEM26800/36800 (Spring 2021)

Quantum Molecular and Materials Modeling

Spring Quarter 2021

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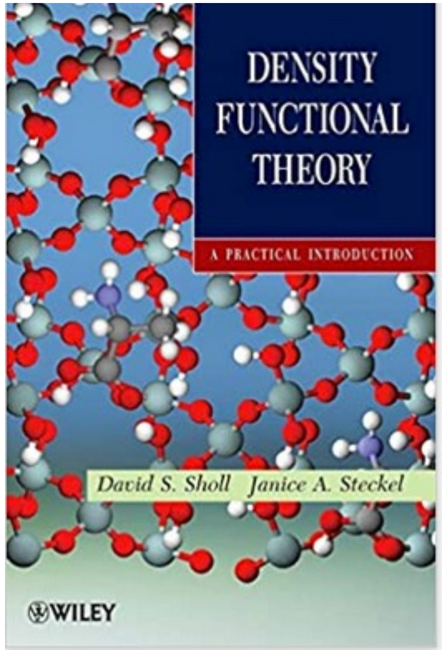
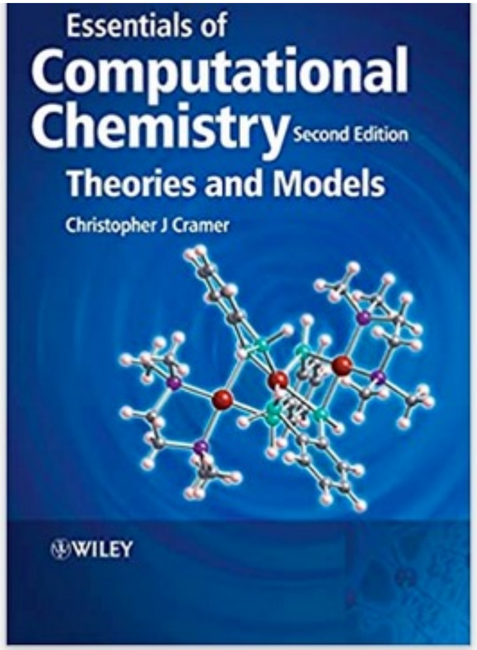
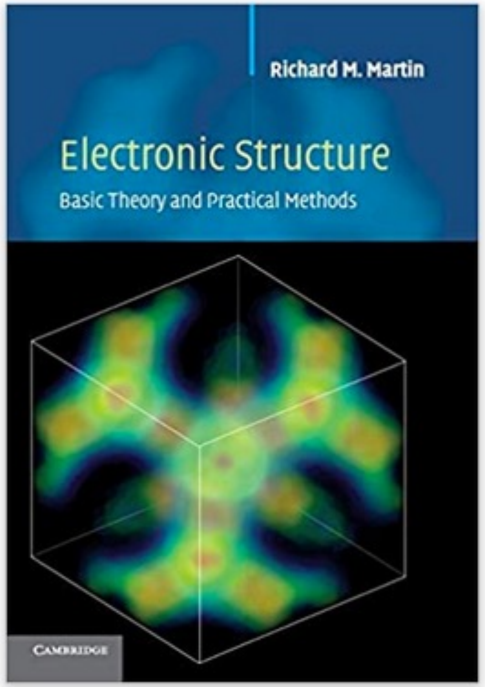
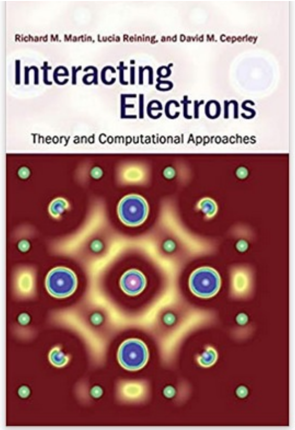
Applied quantum mechanics: materials and molecules through in-silico lenses

Books


DENSITY
FUNCTIONAL
THEORY
A PRACTICAL INTRODUCTION
David S. Sholl Janice A. Steckel
WILEY


Essentials of
Computational
Chemistry Second Edition
Theories and Models
Christopher J Cramer
WILEY


Richard M. Martin
Electronic Structure
Basic Theory and Practical Methods
CAMBRIDGE


Richard M. Martin, Lucia Reining, and David M. Ceperley
Interacting
Electrons
Theory and Computational Approaches

Theory, computation & software

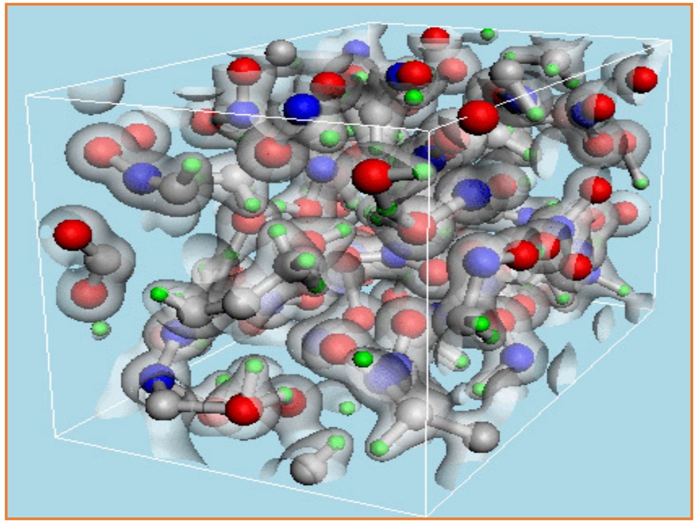
Theory → theoretical approximations → accuracy

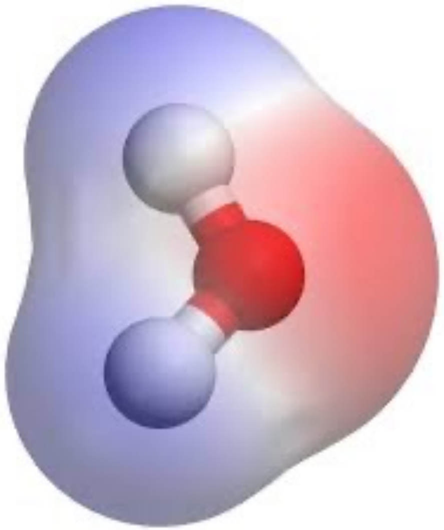
Algorithms used to solve the theoretical equations → numerical methods & approximations → accuracy & efficiency

Codes implementing the algorithms → efficiency

Verified, validated and optimized codes are key to successful predictions & open codes are critical to innovation

Materials (solids, liquids) and molecules





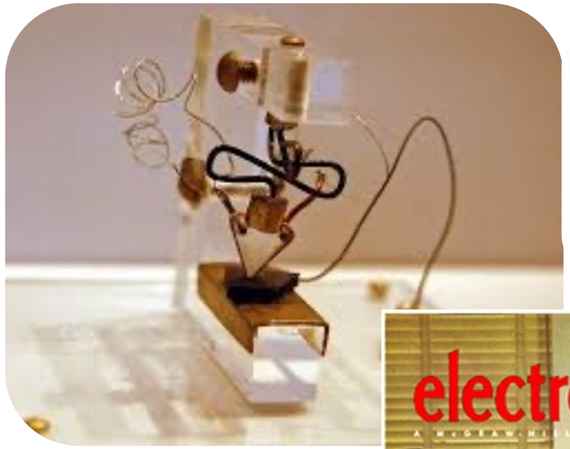
Material or molecule: Electrons (e-) and nuclei (p+)

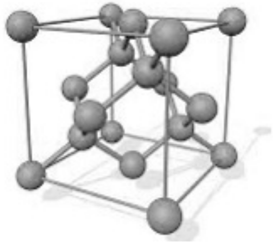
Atomic level stories of materials that changed or are about to change society

- Some well known and some emerging game changers
- Theory and computation as indispensable parts of the discovery process
- Understanding, predictions and design leading to discovery
  - Materials for sustainable energy sources
  - Materials leading us to the quantum information age

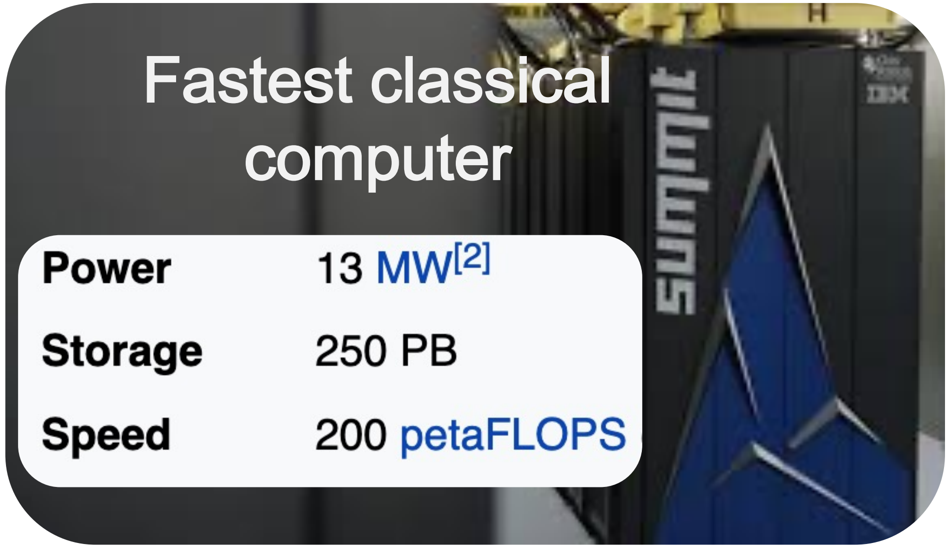
Materials as enablers of novel technologies and scientific discoveries

Silicon





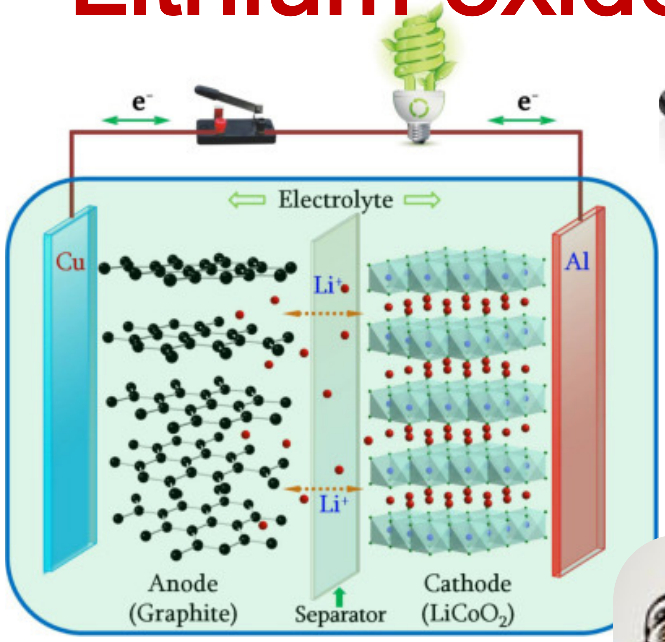
Brattain,
Bardeen,
Shockley
Nobel Prize in
Physics 1956

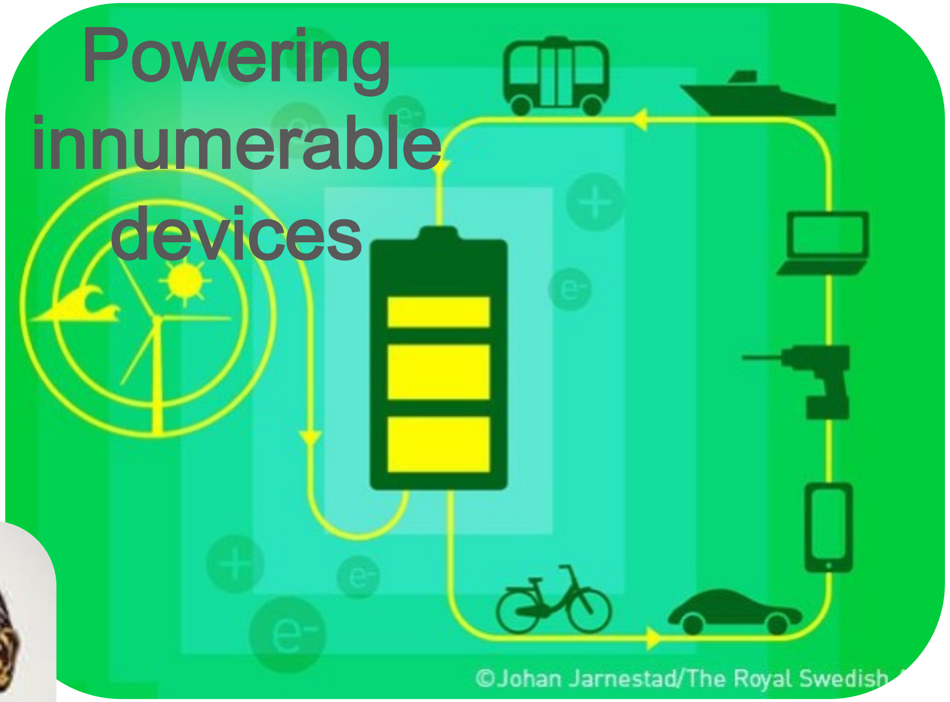


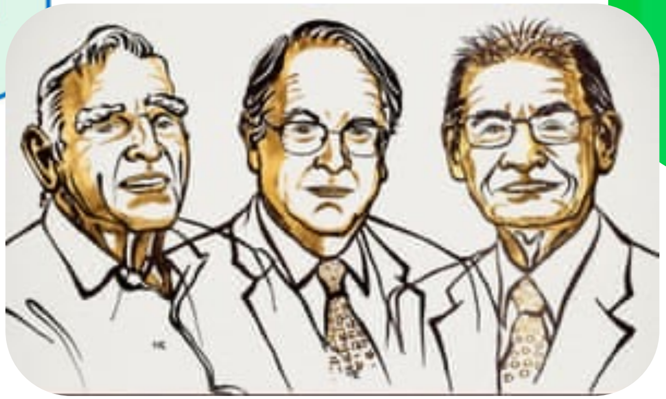
A total of 13 sextillion (1.3×10^{22}) MOSFETs have been manufactured **worldwide** between 1960 and 2018

Materials as enablers of novel technologies and scientific discoveries

Lithium oxides







Goodenough, Whittingham, Yoshino
Nobel Prize in Chemistry 2019

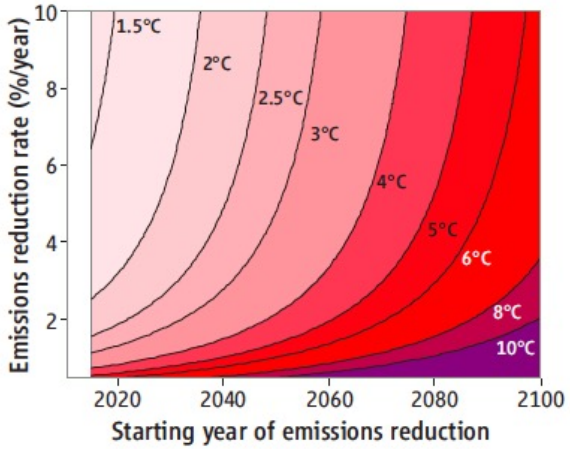
Materials for sustainability and the quantum information age

Sustainability:

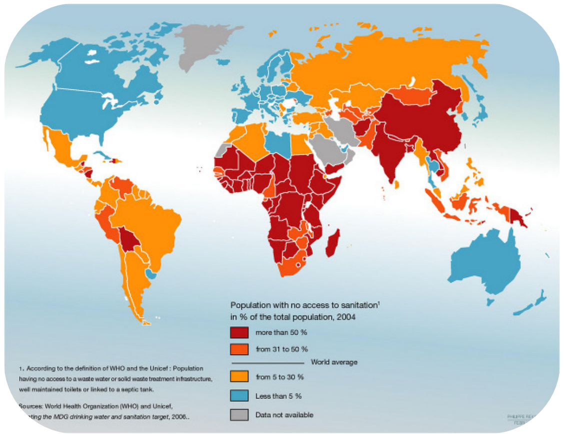
- Sustainable energy sources that do not destroy the ability of the human race to live on the planet
- Broadly available clean water for energy & food

The closing door of climate targets

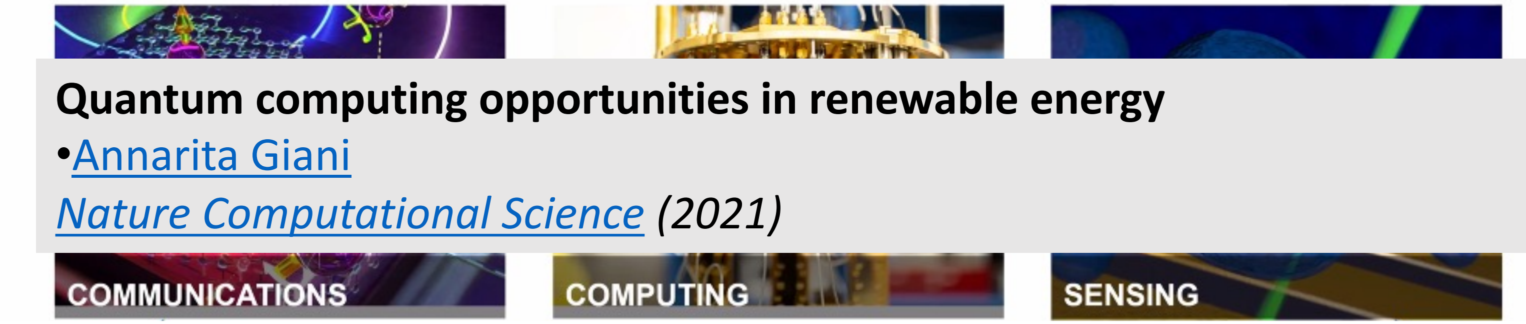
T.F.Stocker, Science 339, 280 (2013)



Water stress



Harnessing the power of quantum technologies:



Quantum computing opportunities in renewable energy

•Annarita Giani

Nature Computational Science (2021)

Theory and computation as integral and indispensable parts of scientific discovery

- Harnessing the laws of quantum mechanics to understand materials and molecules and predict and design their properties

- Using computer simulations to obtain accurate solutions to equations describing natural phenomena

Quantum mechanics: basic equations

$$H\Psi = E\Psi$$

$$i\hbar \frac{\partial \psi}{\partial t} = \mathrm{H}\psi$$

Dirac

The fundamental laws necessary for the mathematical treatment of a large part of physics and **the whole of chemistry** are thus completely known …

the difficulty lies only in the fact that application of these laws leads to equations that are too complex to be solved.

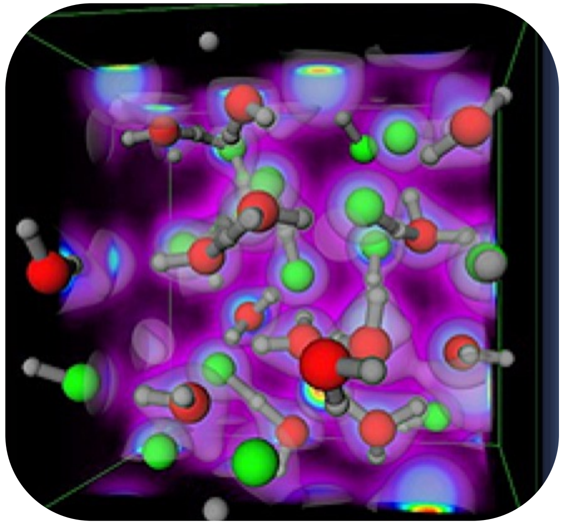
THE

PHYSICAL REVIEW

AN UNDULATORY THEORY OF THE MECHANICS OF ATOMS AND MOLECULES

BY E. SCHRÖDINGER

ZÜRICH, SWITZERLAND,
September 3, 1926.



Quantum mechanics: approximations

The approximations:

- **Mean-field theories:**
  - **Density Functional Theory (DFT)**
    - ***Local density approximations***
- **Hartree-Fock and Quantum Chemistry**
- **Stochastic approaches:**
  - **Quantum Monte Carlo (QMC)**
    - ***The fixed node approximation***

~ 1965

~ 1980-90

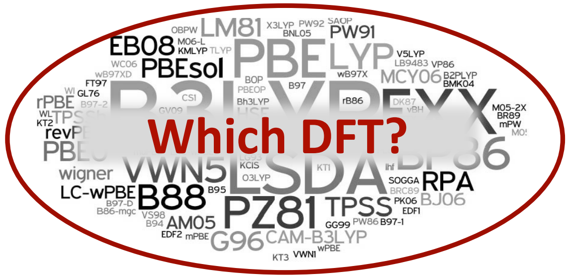
W.Kohn & J.Pople, Chemistry Nobel Prize,1998

The ability to compute:

algorithmic and computational developments

- **Solutions of DFT equations for solids & molecules**
- ***Ab-initio* Molecular Dynamics (Car-Parrinello method)**
- **Software development for HPC architectures**

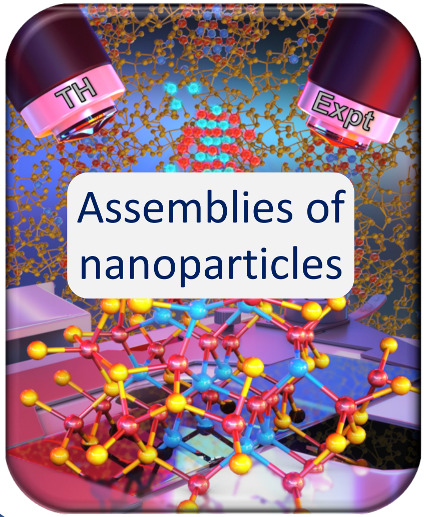
A general, predictive approach

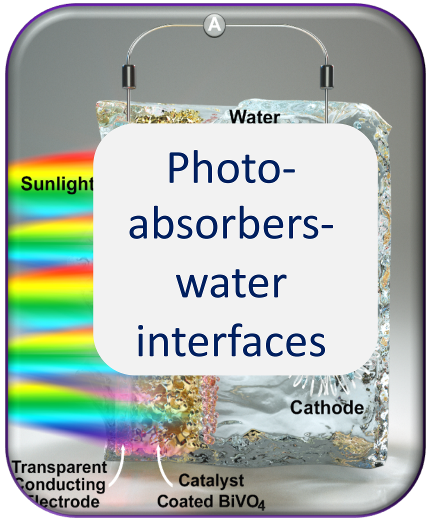


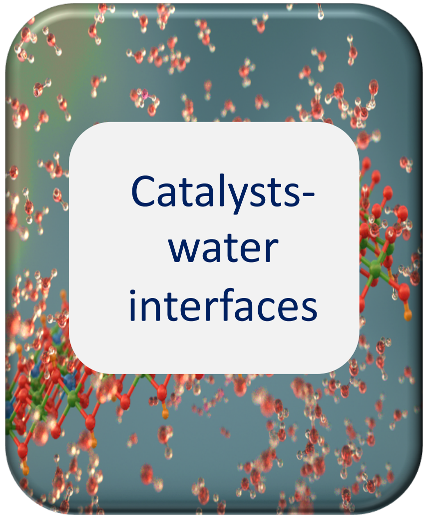
$$M\ddot{\mathbf{R}}_I = -\frac{\partial E}{\partial \mathbf{R}_I} = F_I[\{\mathbf{R}_J\}]$$

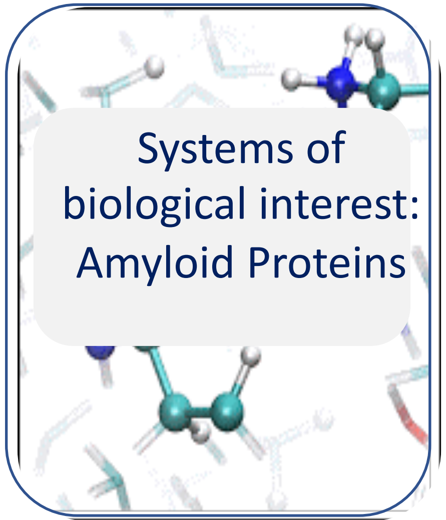
E from Density Functional Theory (DFT)

How do we solve these equations?









Ali Hassanali et al. 2020: http://users.ictp.it/~ahassana/research.html

A general, predictive approach

$$\boxed{M\ddot{\mathbf{R}}_I = -\frac{\partial E}{\partial \mathbf{R}_I} = F_I[\{\mathbf{R}_J\}]}$$

E from Density Functional Theory (DFT)

How do we solve these equations?

"Really efficient high-speed computing devices may, in the field of nonlinear partial differential equations [...] provide us with those heuristic hints which are needed in all parts of mathematics for genuine progress", J.v.Neumann and H.H.Goldstine, On the principles of large scale computing machines, Collected Works, Vol. V, 1963

A general, predictive approach

$$M\ddot{\mathbf{R}}_I = -\frac{\partial E}{\partial \mathbf{R}_I} = F_I[\{\mathbf{R}_J\}]$$

E from Density Functional Theory

2020 ACM Gordon Bell Prize Awarded to Team for Machine Learning Method that Achieves Record Molecular Dynamics Simulation

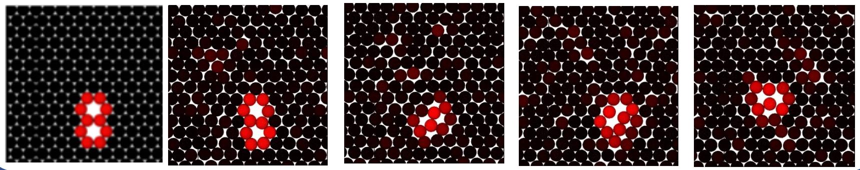
Nov 19th, 2020

ACM, the Association for Computing Machinery, named a nine-member team, drawn from Chinese and American institutions, recipients of the 2020 ACM Gordon Bell Prize for their project, "Pushing the limit of molecular dynamics with *ab initio* accuracy to 100 million atoms with machine learning."

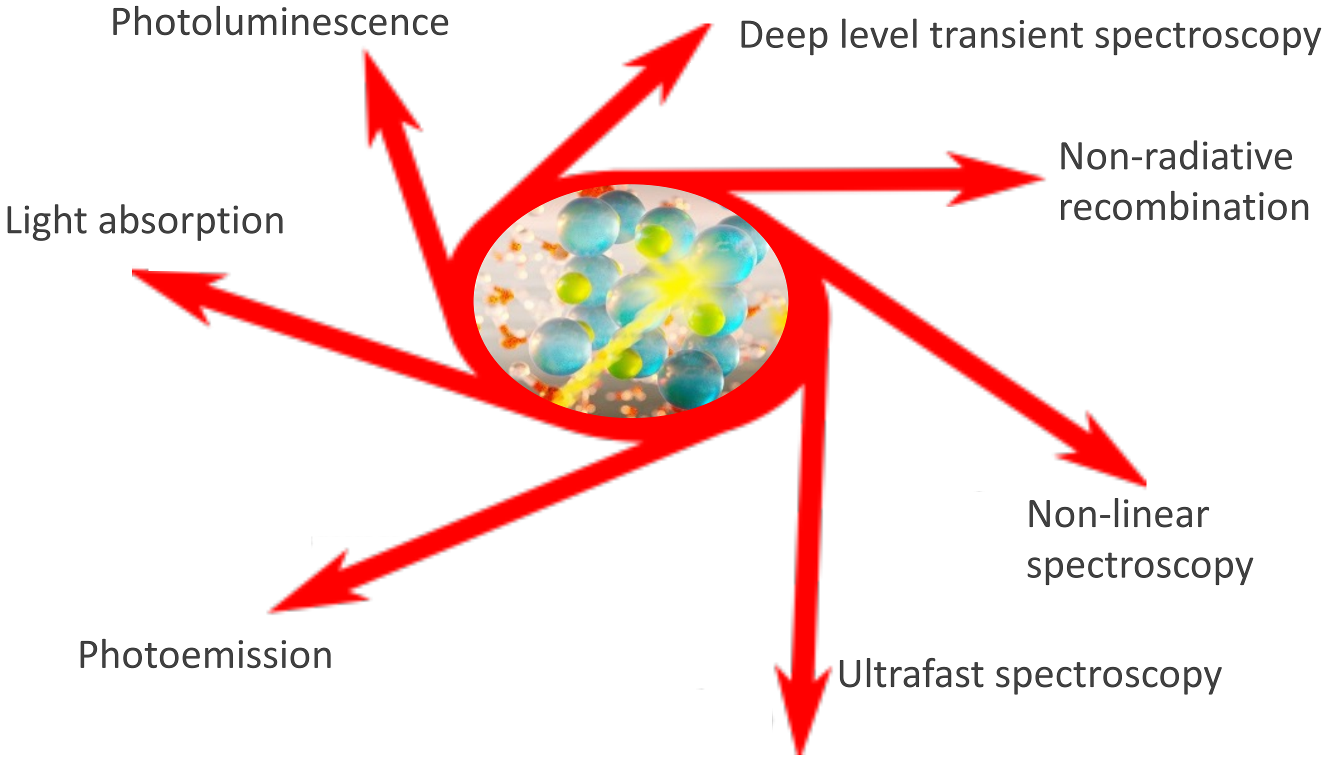
Winning team members include Weile Jia, University of California, Berkeley; Han Wang, Institute of Applied Physics and Computational Mathematics (Beijing, China); Mohan Chen, Peking University; Denghui Lu, Peking University; Jiduan Liu, Peking University; Lin Lin, University of California, Berkeley and Lawrence Berkeley National Laboratory; Roberto Car, Princeton University; Weinan E, Princeton University; and Linfeng Zhang, Princeton University.

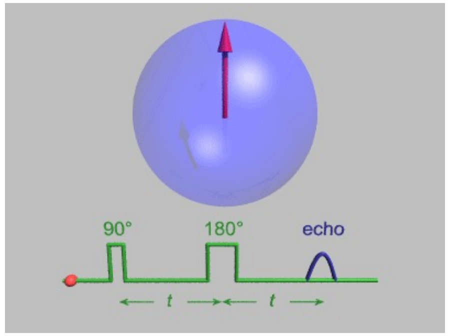
Theoretical and computational strategy

Structural properties of materials at finite T from atomistic & first principles simulations



Interrogate materials with light: Spectroscopy





Probe spins and how they interact

Theory, computation & software

Theory → theoretical approximations → accuracy

Algorithms used to solve the theoretical equations → numerical methods & approximations → accuracy & efficiency

Codes implementing the algorithms → efficiency

Verified, validated and optimized codes are key to successful predictions & open codes are critical to innovation

Theory, computation & software

Thursday, April 1	Lecture 2	Schoedinger Equation (SE): basic approaches to solve the time independent SE for modeling molecular and condensed systems
2. Tuesday, April 6	Lecture 3	Schoedinger Equation (SE): basic approximations in quantum chemistry → Hartree Fock (HF)
Thursday, April 8	Lecture 4 & Homework 1 assigned	Schoedinger Equation (SE): beyond HF & numerical solutions
3. Tuesday, April 13	Lecture 5	Quantum Chemistry: basis set expansions and error analysis
Thursday, April 15	Lecture 6	Introduction to Density Functional Theory (DFT): the electron gas, variational theorem, functional minimizations
4. Tuesday, April 20	Lecture 7 Homework 1 due & Homework 2 assigned	The Kohn-Sham (KS) equations: local and non-local functionals; examples of solutions for molecules and comparison with quantum chemistry methods
Thursday, April 22	Lecture 8	The Kohn-Sham (KS) equations: solutions for periodic systems (solids), including Bloch theorem and basic solid-state physics concepts (I)
5. Tuesday, April 27	Lecture 9	The Kohn-Sham (KS) equations: solutions for periodic systems (solids), including Bloch theorem and basic solid-state physics concepts (II)
Thursday, April 29	MIDTERM	Open book
6. Tuesday, May 4	Lecture 10 Homework 2 due & Homework 3 assigned	Geometry optimizations for molecules and solids; vibrational properties and phonons
Thursday, May 6	Lecture 11	Molecular dynamics
7. Tuesday, May 11	Lecture 12	First principles molecular dynamics (FPMD)
Thursday, May 13	Lecture 13 Homework 3 due & Homework 4 assigned	Application of FPMD, including computational vibrational spectroscopy of solids and molecules
8. Tuesday, May 18	Lecture 14	Computational spectroscopy: electronic and optical spectra of molecules and solids
Thursday, May 20	Lecture 15	Coupling FPMD and electronic structure theories beyond DFT
9. Tuesday, May 25	Lecture 16	Quantum simulations on quantum computers
Thursday, May 27	Lecture 17 (recap) Homework 4 due	Recap and general questions

Atomic level stories of materials that changed or are about to change society

- Some well known and some emerging game changers
- Theory and computation as indispensable parts of the discovery process
- Understanding, predictions and design leading to discovery
  - Materials for sustainable energy sources
  - Materials leading us to the quantum information age

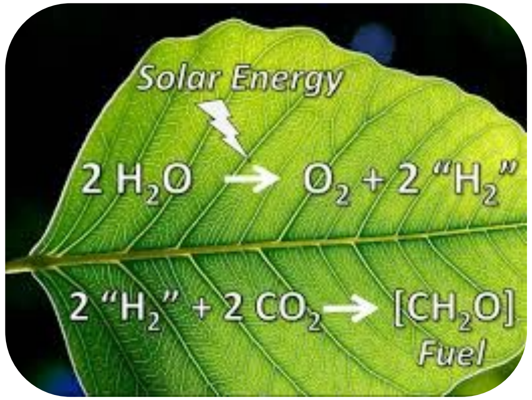
Materials for sustainability and the quantum information age

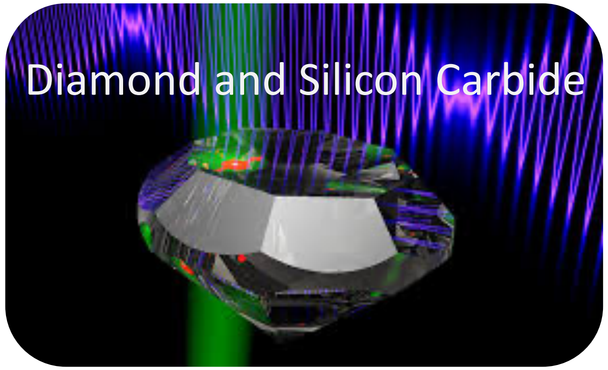
Sustainability:

- Materials to convert the energy we receive from the sun into **electricity**
- Materials to produce **clean fuels**: convert H_2O into hydrogen and oxygen and CO_2 to "benign" & useful species

Solar Cells

Photoelectrochemical cells

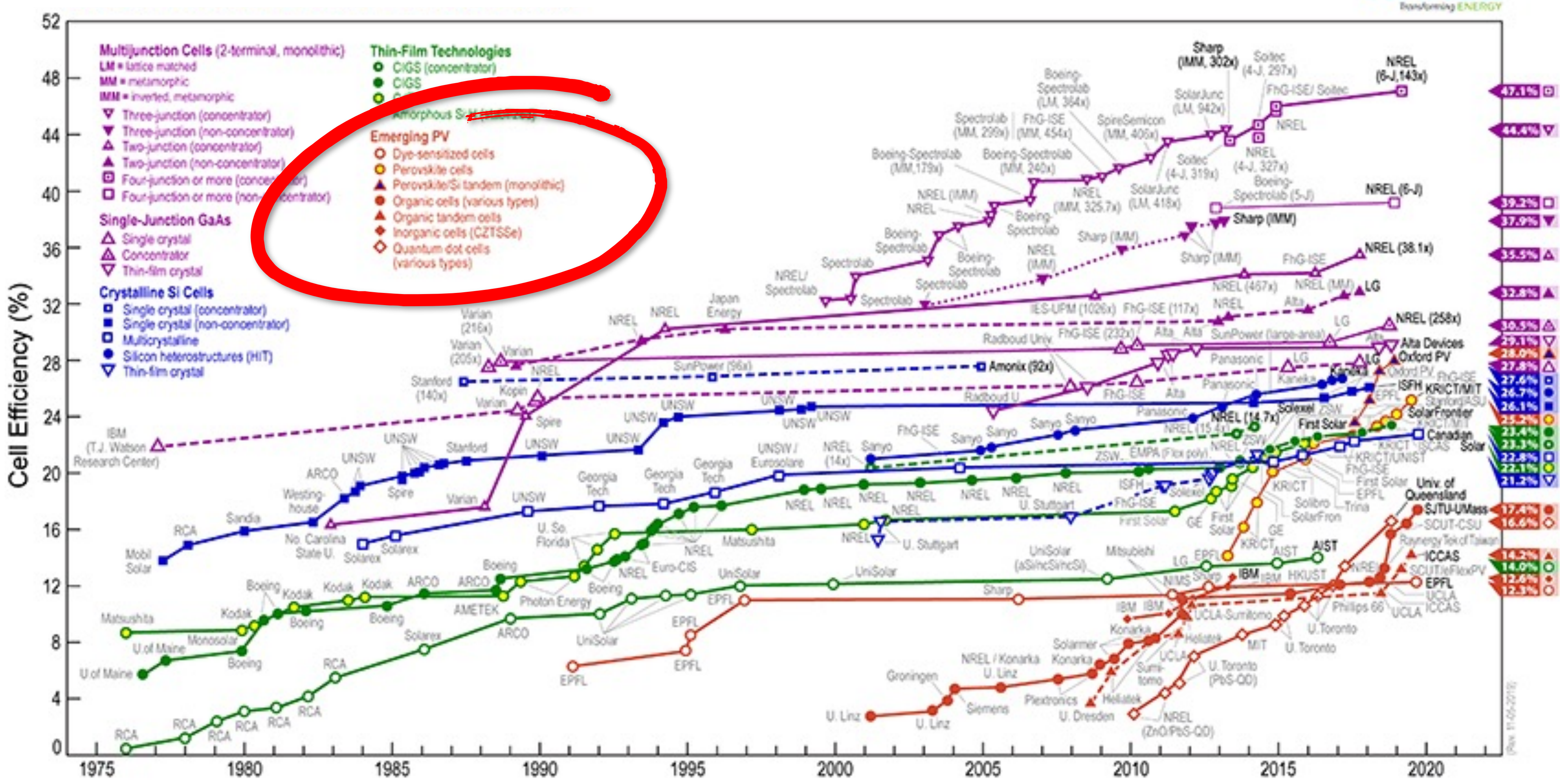




Harnessing the power of quantum technologies:

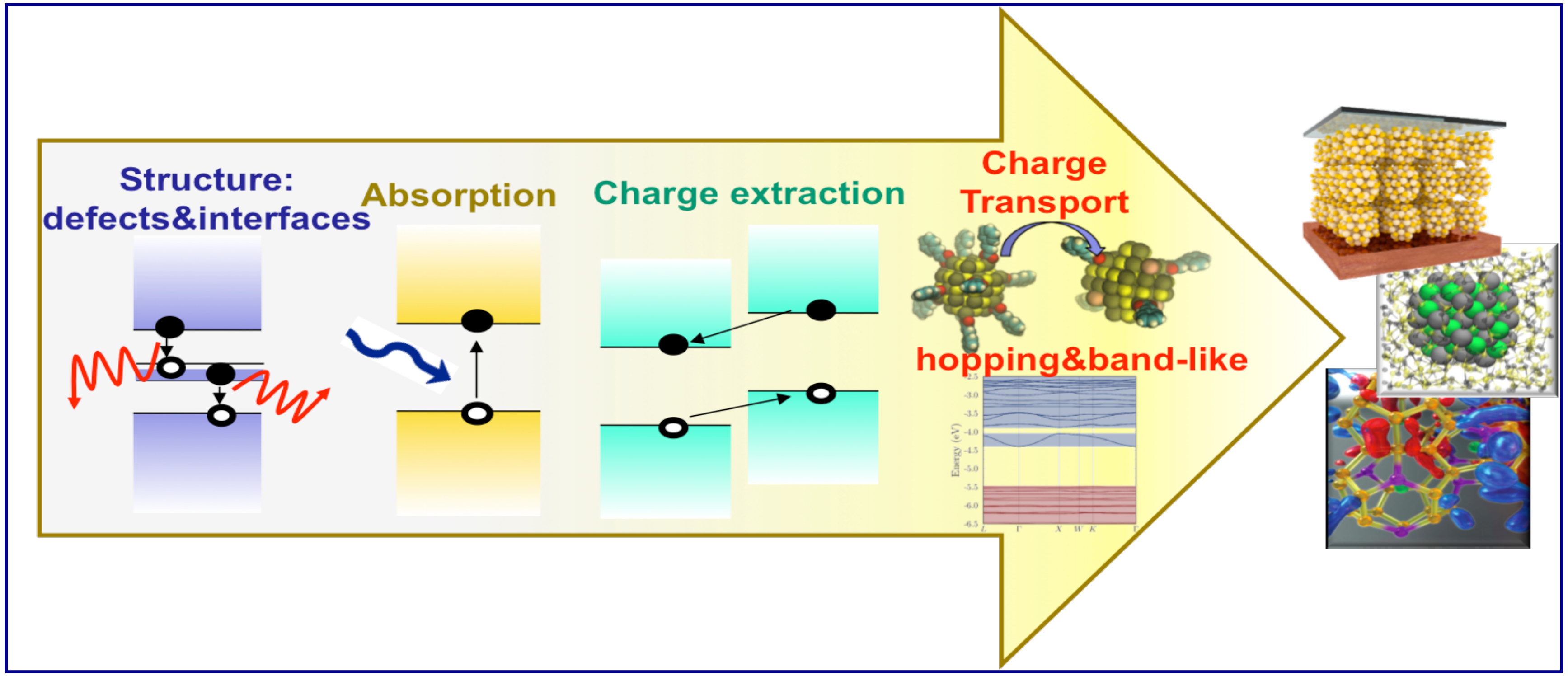
- Defective materials for **qubits** and **quantum sensors**

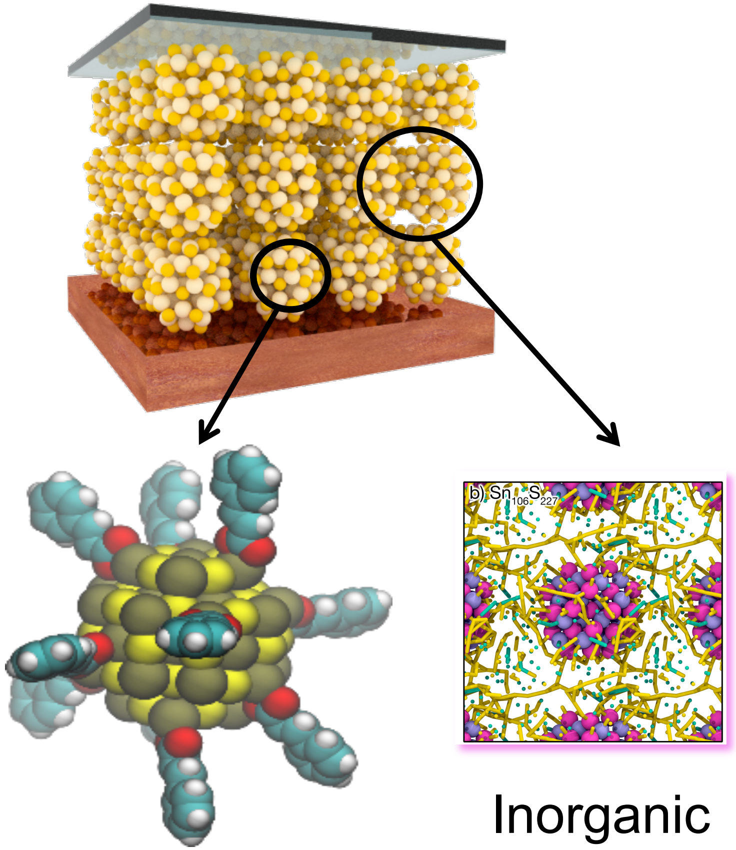
Solar cells: no size fits all & not only a matter of efficiency



Colloidal quantum dot solar cells: easy to make, with tunable properties

Assemblies of nanoparticles : Structure ←→ Function





Organic ligands

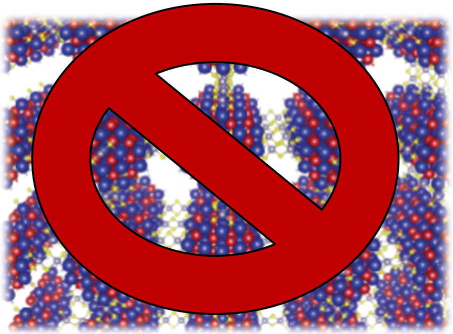
Inorganic ligands

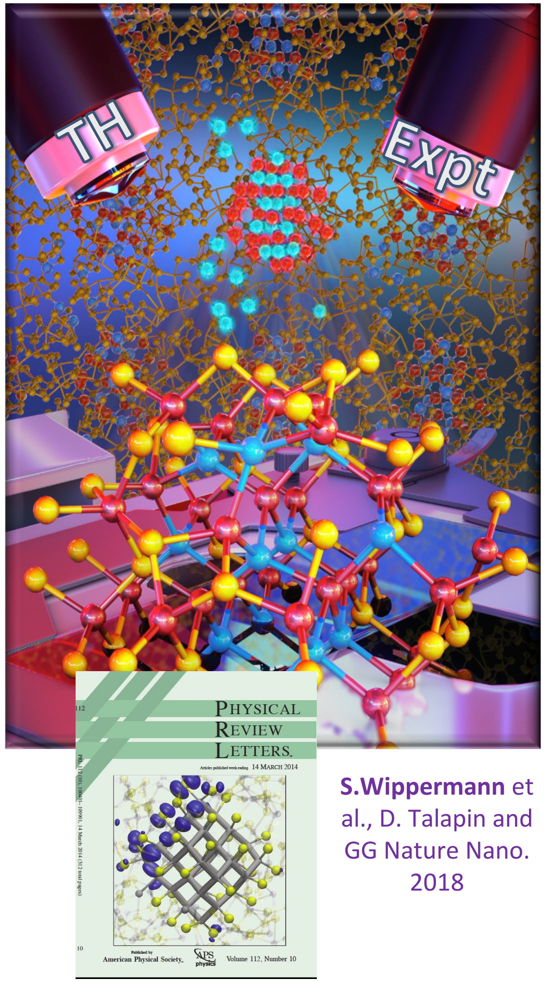
Colloidal quantum dot solar cells → design nanocomposites

- Nanoparticles "provide" the charges (and multi-exciton generation helps!) under illumination

- **Ligand & matrix engineering** is key to create favorable heterojunctions and to enable charge extraction: experiments explained & predictions verified

- Simplified structural models are insufficient to explain, even qualitatively, electronic and transport properties

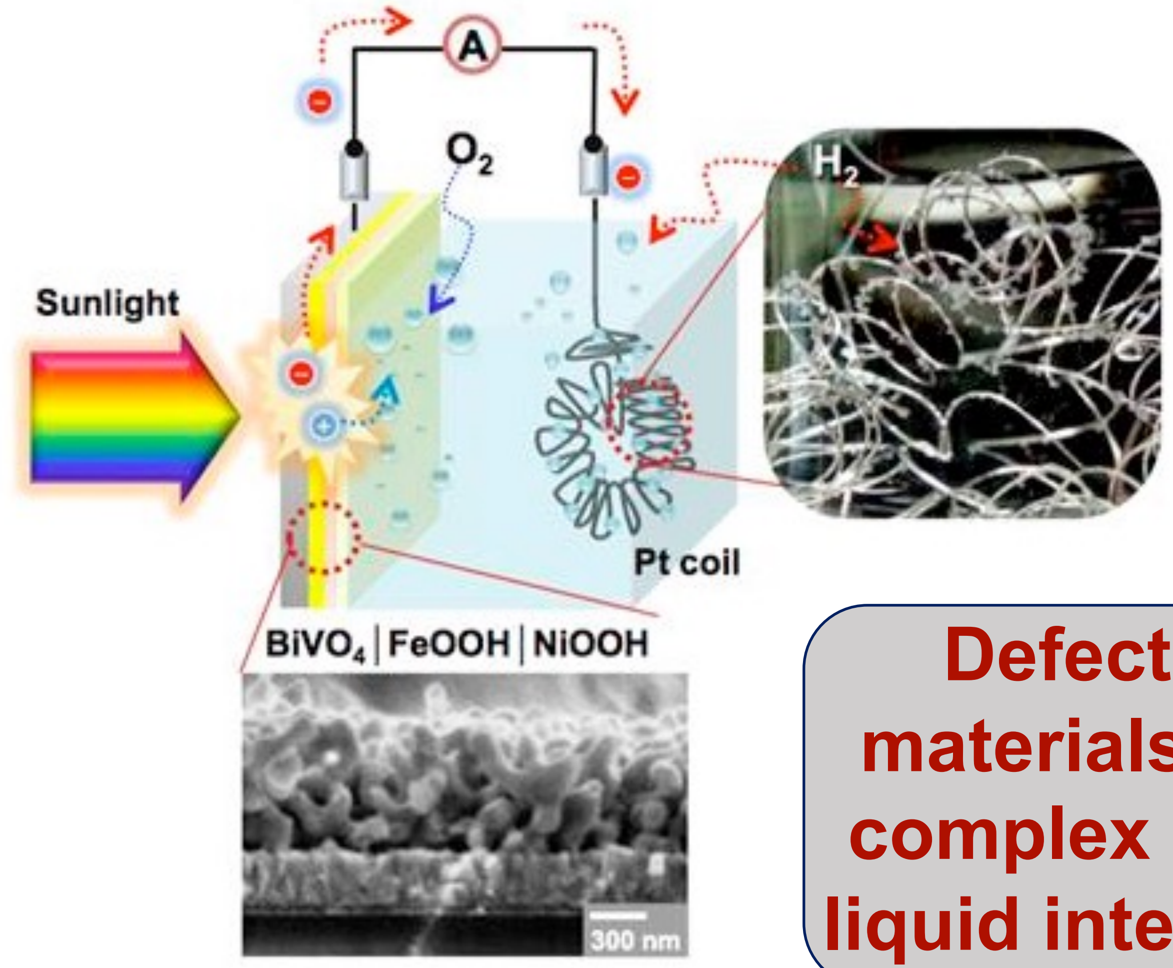




S.Wippermann et al., D. Talapin and GG Nature Nano. 2018

S.Wipperman, M.Voros, A.Gali, F.Gygi, G. Zimanyi and GG PRL 2014

Absorb light to trigger complex chemical reactions → photocatalytic water splitting



Defective materials with complex solid-liquid interfaces

Absorb light

$$2\,H_2O \xrightarrow{h\nu} 2H_2 + O_2$$

Transport (e^-, h^+) pairs from the solid absorber to the liquid

Harvest charges for chemical reactions

Navigating the puzzle pieces of water splitting

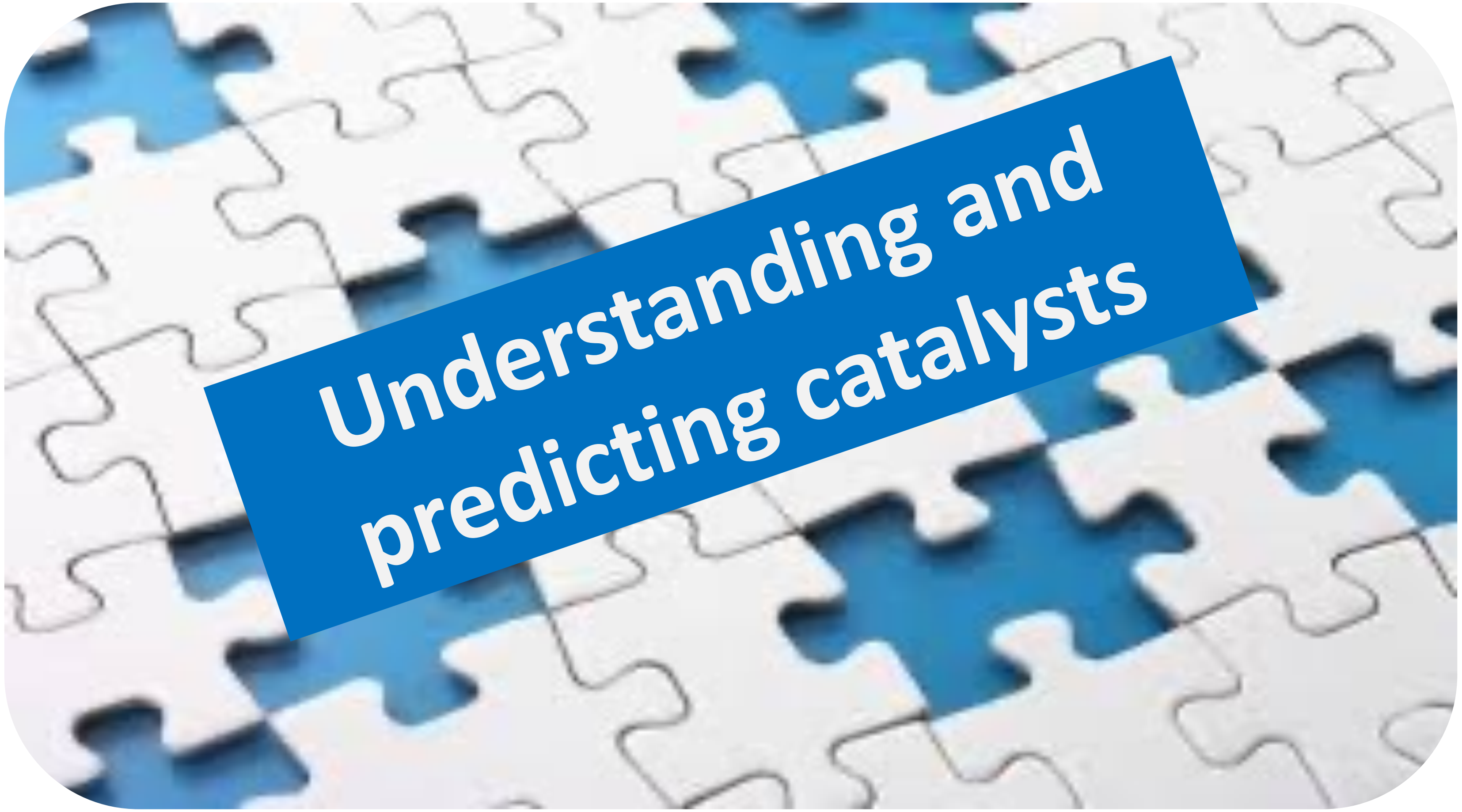
- Reasonable model of (salty) water

- Atomistic model of solid/liquid interfaces & their electronic properties (e.g. **band offsets** and **Schottky barriers**)

- **Charge transport** @interfaces

K-S Choi and GG collaboration, Nature Energy 2021

Navigating the puzzle pieces of water splitting

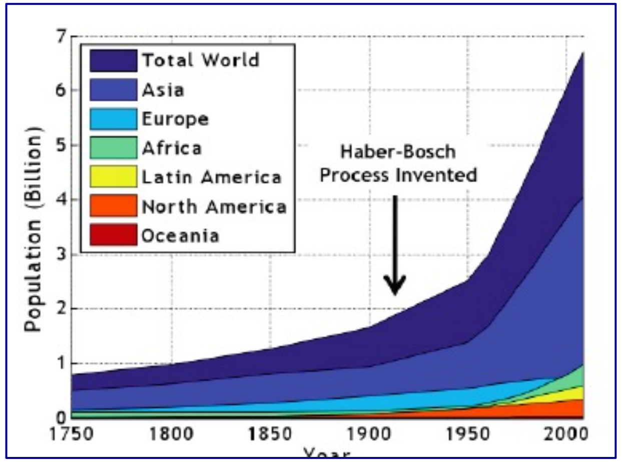


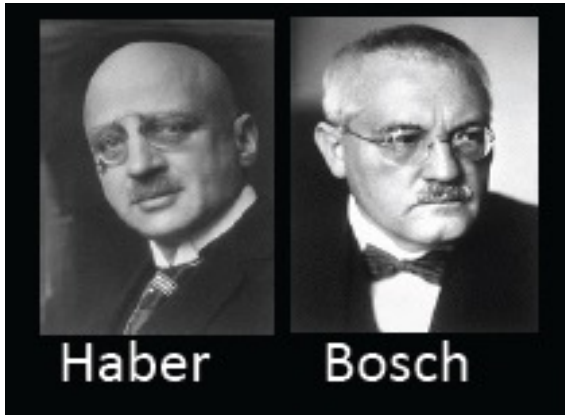
A new catalyst can be a game changer

- In 1898 Sir William Crookes, President of the British Assoc. for the Advancement of Science, was “calling upon science to save the world from impending starvation”

- In 1908 Fritz Haber discovered a catalyst that would combine atmospheric nitrogen with hydrogen to form ammonia (catalyst: uranium!)

- In 1913 Carl Bosch developed process to mass produce ammonia and made fertilizers





A new technology can be a game changer

'Great Horse Manure Crisis of 1894'.

- New York had a population of 100,000 horses producing around 2.5m pounds of manure a day.

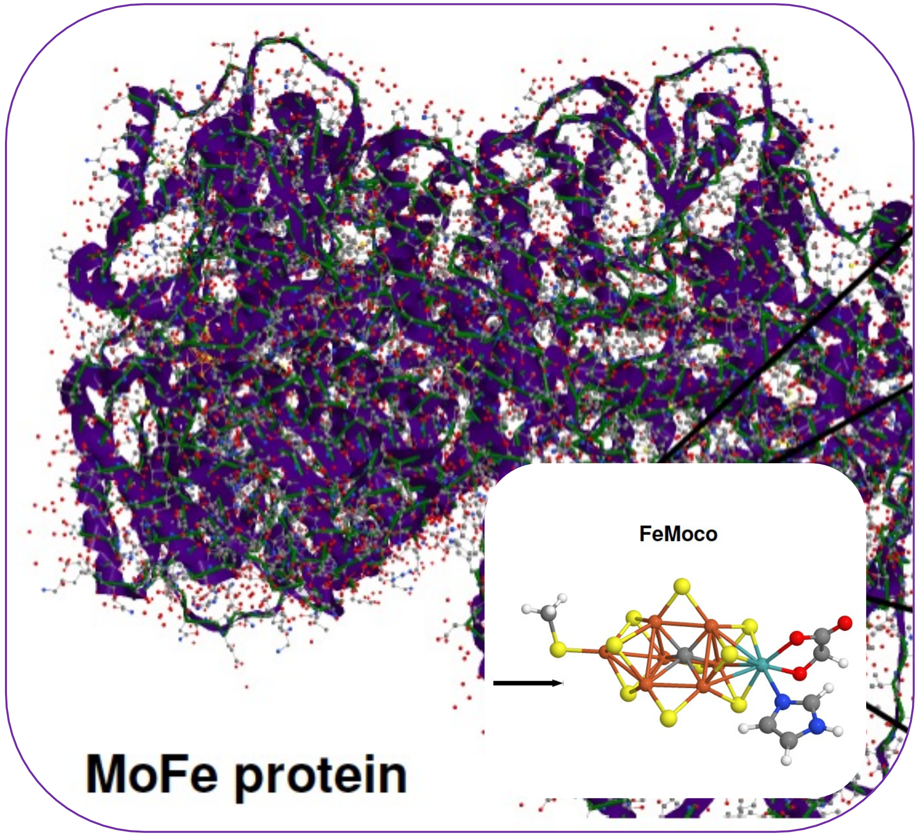
- London was in even worse conditions; in 1894 the Times newspaper predicted... "In 50 years, every street in London will be buried under nine feet of manure."

- **From horse power to horsepower**: in 1912 Henry Ford made the use of the motor a viable technology for transportation

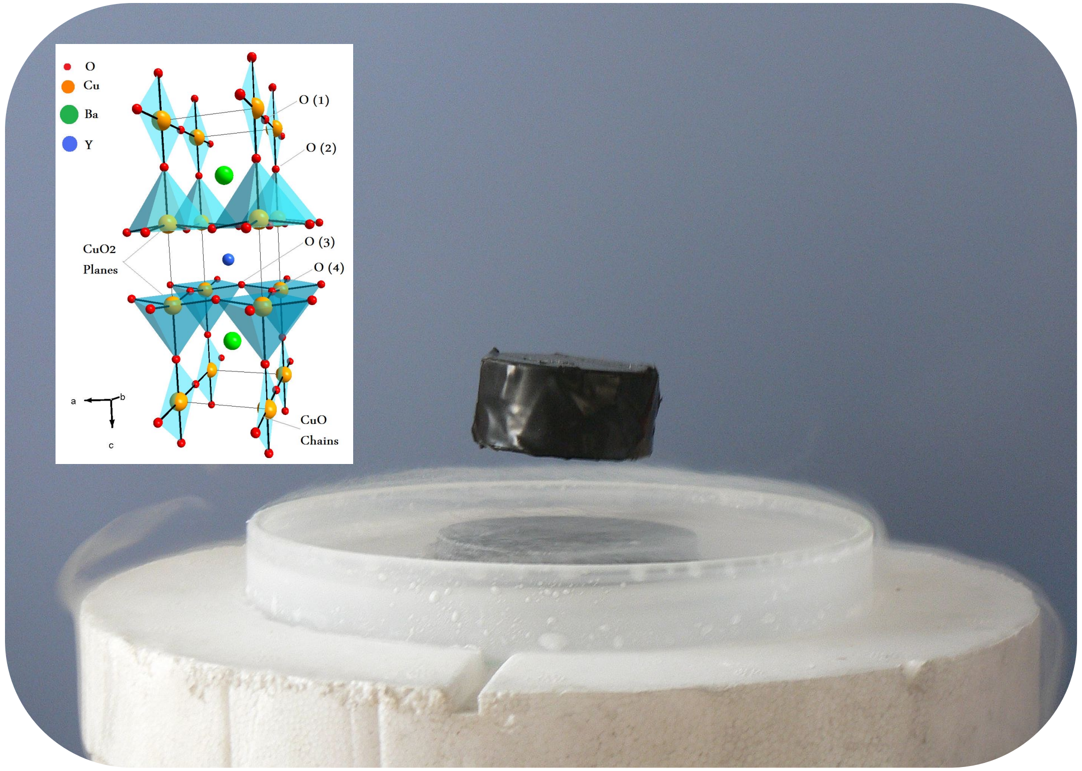
How are we solving more complex problems?

How are we solving more complex problems?

Nitrogenase MoFe protein



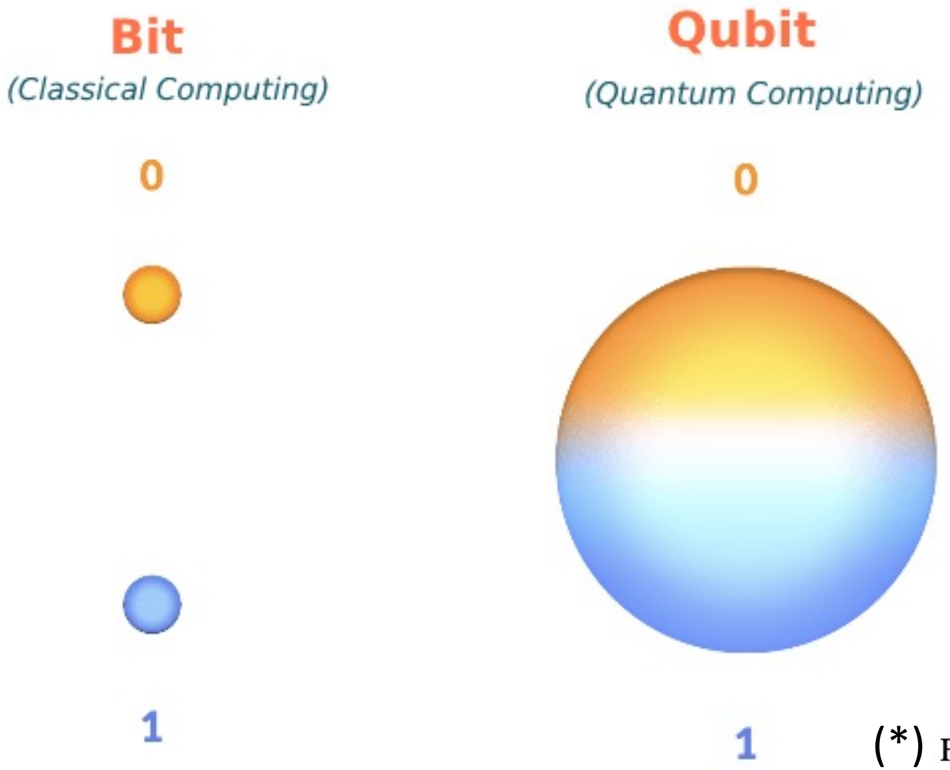
High Tc Superconductors



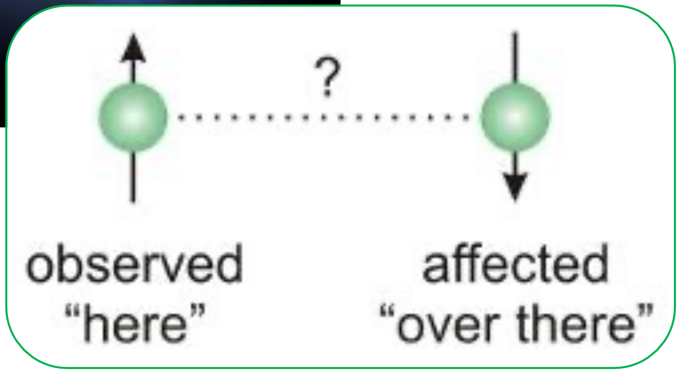
Change of Perspective

Richard Feynman (82') proposed the idea of creating machines based on the laws of **quantum mechanics** instead of the laws of classical physics (*).

Superposition



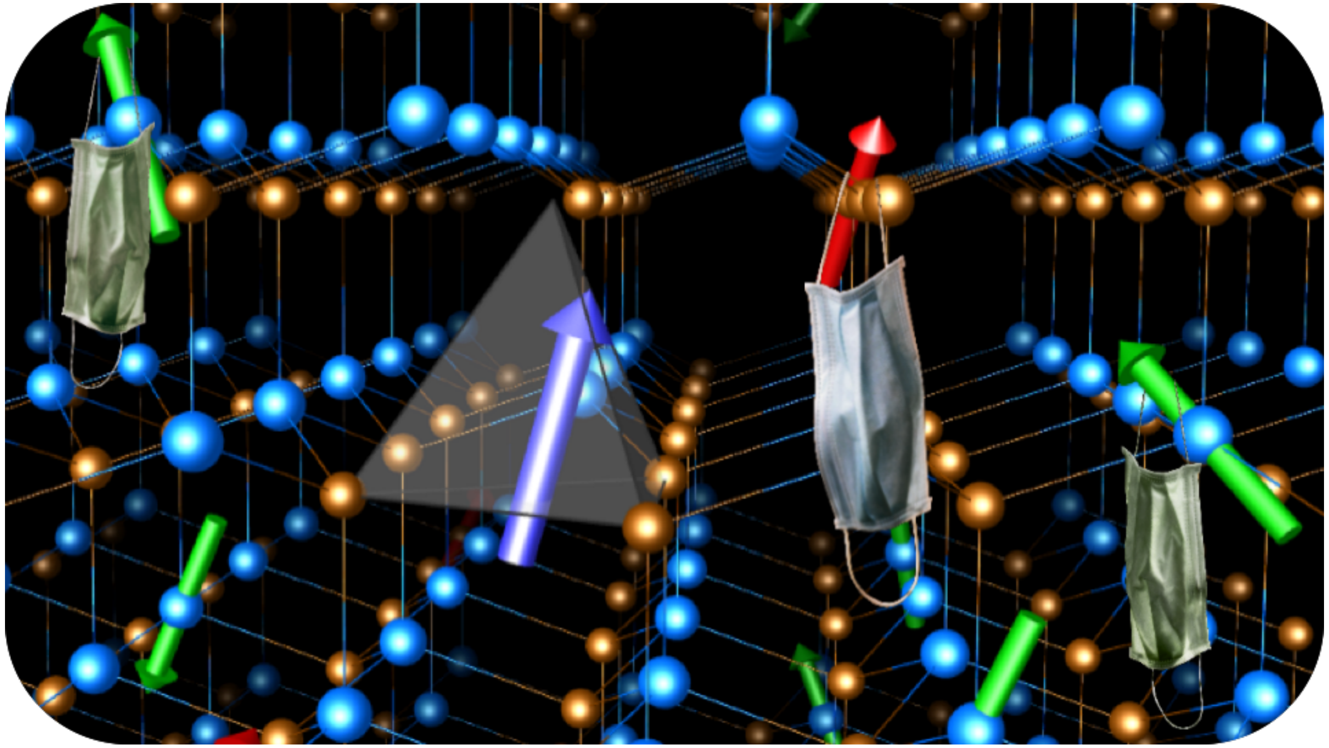
Entanglement



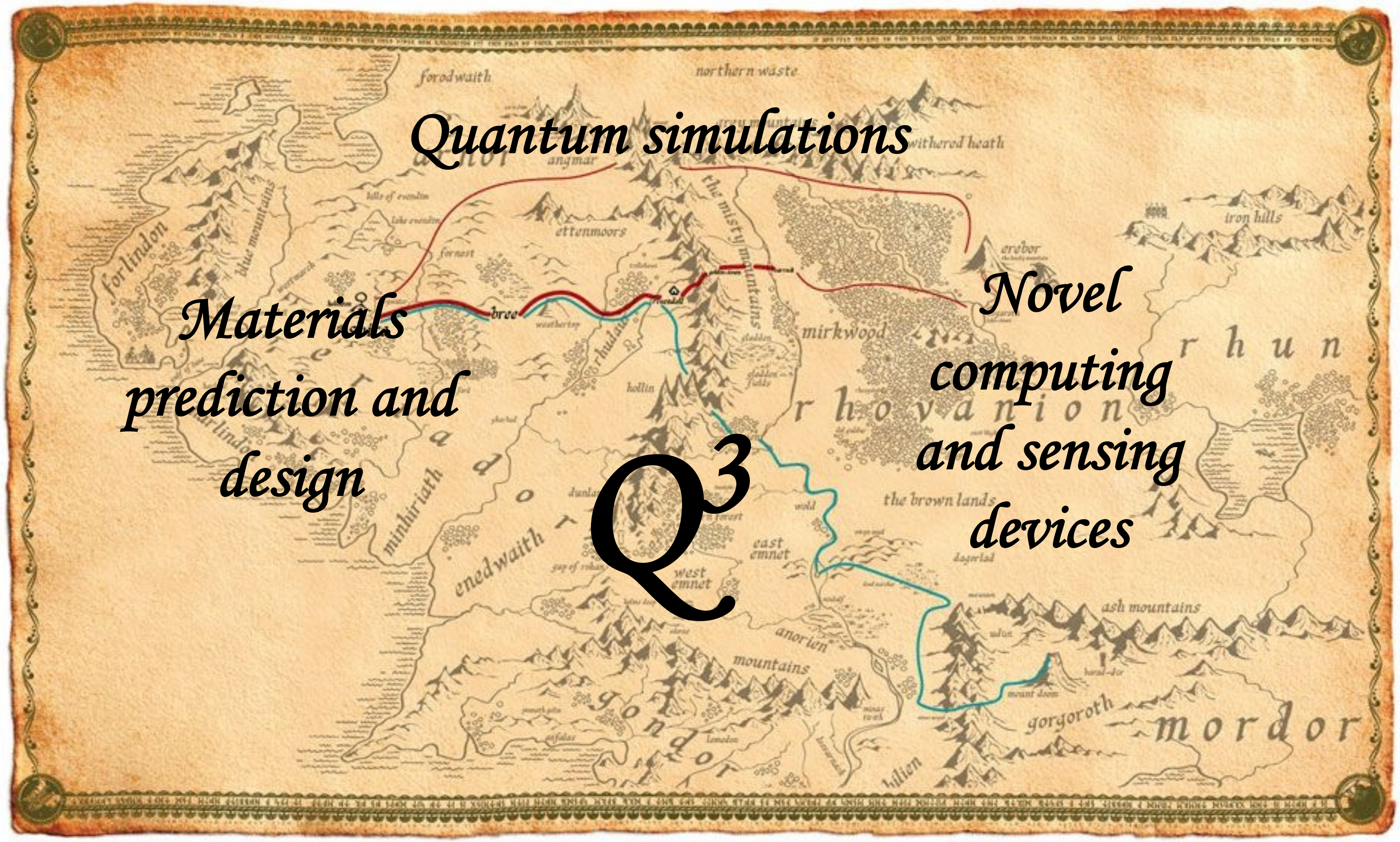
(*) Feynman, R. 1983. Tiny Computers Obeying Quantum Mechanical Laws. Talk delivered at Los Alamos National Laboratory. Published in *New Directions in Physics: The Los Alamos 40th Anniversary Volume*.

Spin defects

- Manipulating spin states of defects in solids with light, to design new computing technologies and new generation of nanoscale sensors:
  - —**Diamond and Silicon Carbide**



From materials to quantum devices and back again

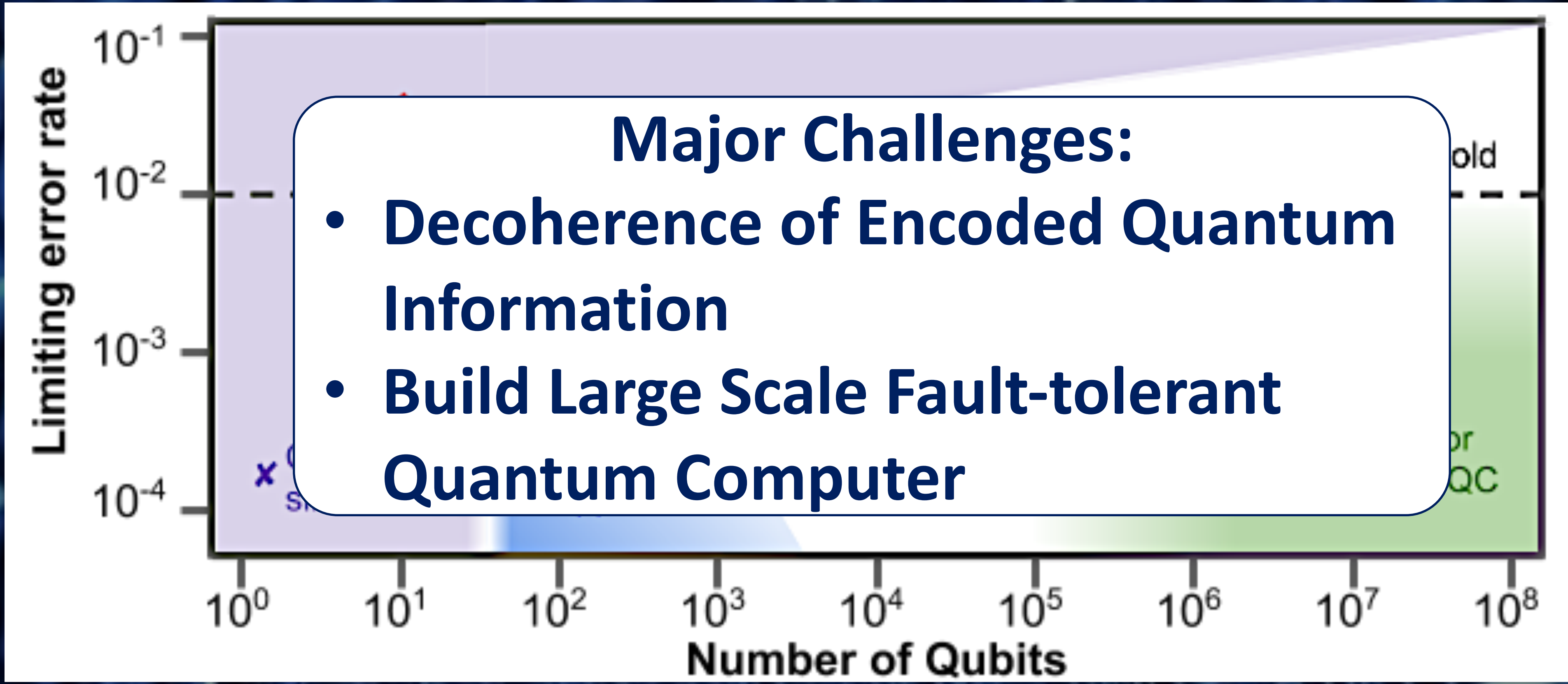


Quantum Computations

[…] With a [different] entanglement pattern […] the Summit supercomputer at ORNL could solve the problem in a matter of days.

October 21, 2019 | Written by: Edwin Pednault, John Gunnels & Dmitri Maslov, and Jay Gambetta

Quantum Computations



Major Challenges:

- **Decoherence of Encoded Quantum Information**
- **Build Large Scale Fault-tolerant Quantum Computer**

Where is the data?
How can one access the data?
What remains of the data described in scientific papers?

http://www.qresp.org/

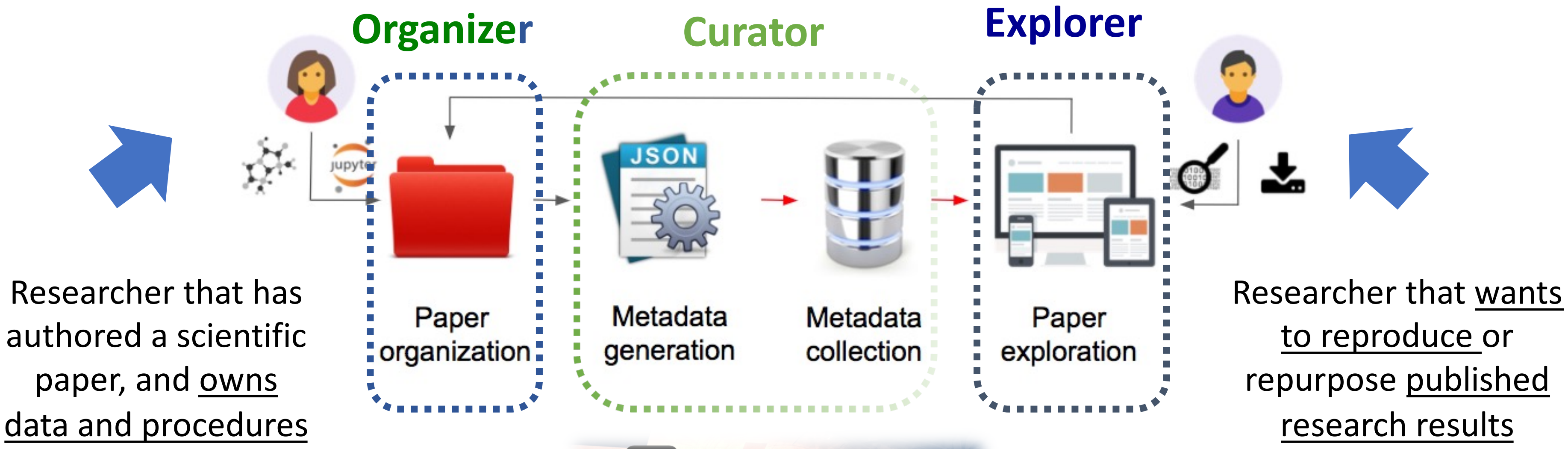
Qresp

Curation and Exploration of Reproducible Scientific Papers

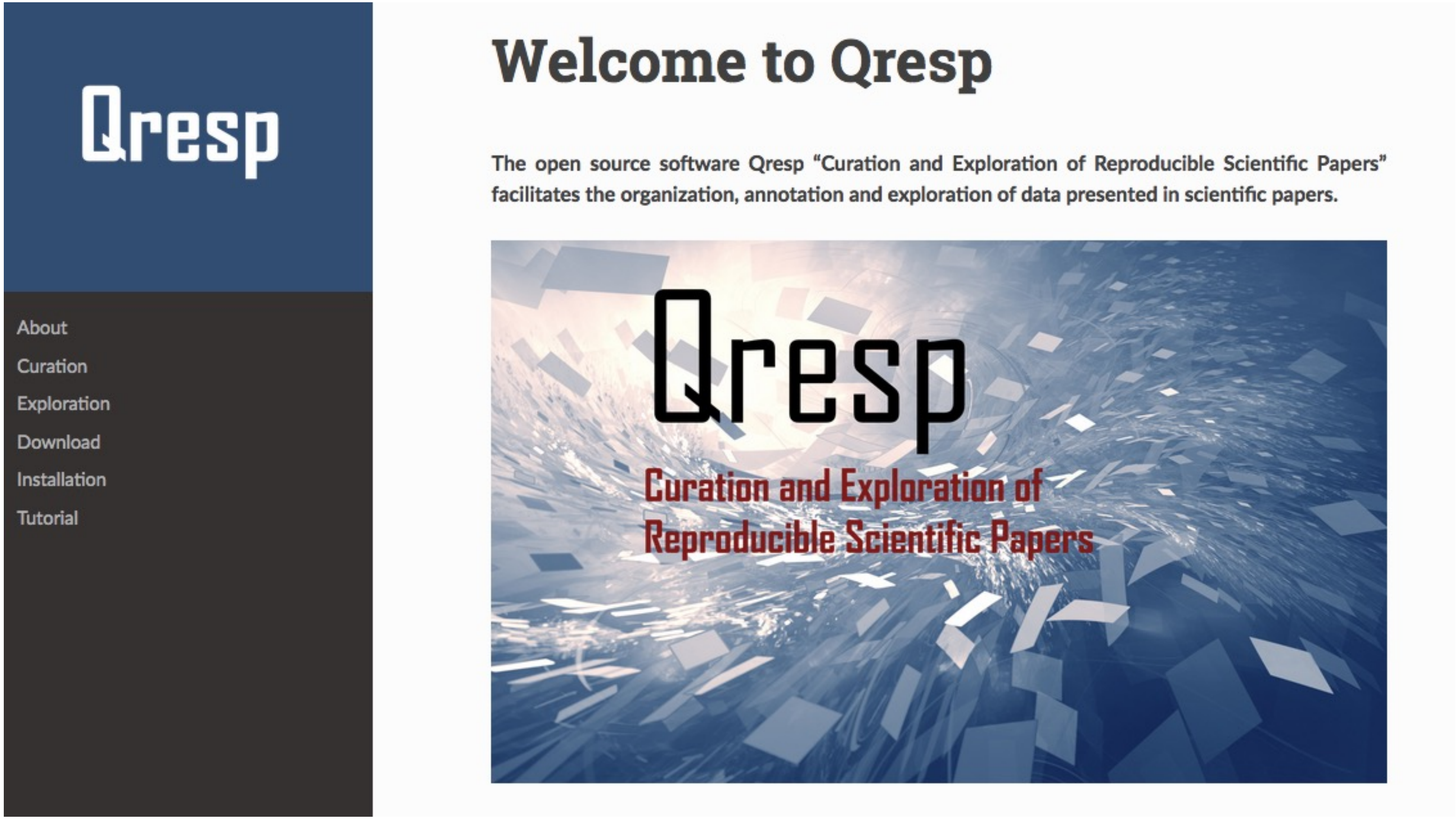
Govoni et al. Scientific Data, 2019

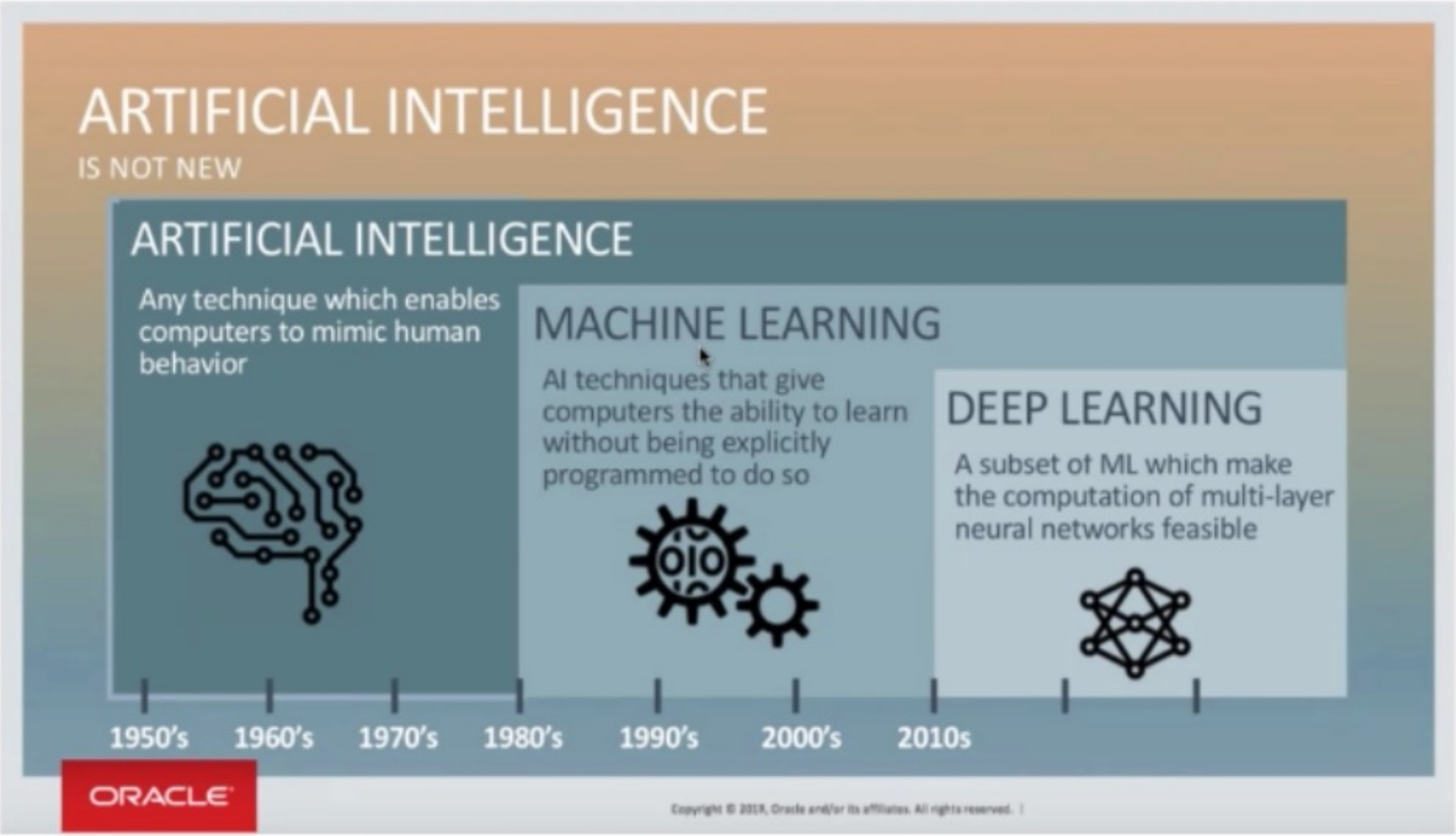
Reproducibility of scientific papers

- We developed a platform for the dissemination and reproducibility of data on a *per-publication basis*
- Qresp is an *open-source suite* of programs that facilitates organization, annotation and exploration of data presented in scientific papers



http://www.qresp.org/





https://blogs.oracle.com/bigdata/difference-ai-machine-learning-deep-learning

Artificial Neural Networks: A kind of machine learning approach that uses networks of neuron-like units to process information