Group A Wrap Up

- Charles Cheung (U of Delaware)
  - Theme: relativistic quantum chemistry calculations.
  - Parallelization: hundreds of nodes.

- Mursaleem Ansari
  - Theme: quantum chemistry calculations for catalyst design.
  - Parallelization: tens of nodes.

- Denis Mashkovtsev
  - Theme: O(N) chemistry calculations with the ONIOM method.
  - Parallelization: single node.

- Haruki Satoh
  - Theme: mesoscale calculations using classical models.
  - Parallelization: K computer experience in his lab.

- Akane Yamashita
  - Theme: high level modeling of biological systems.
  - Parallelization: single node (matlab)

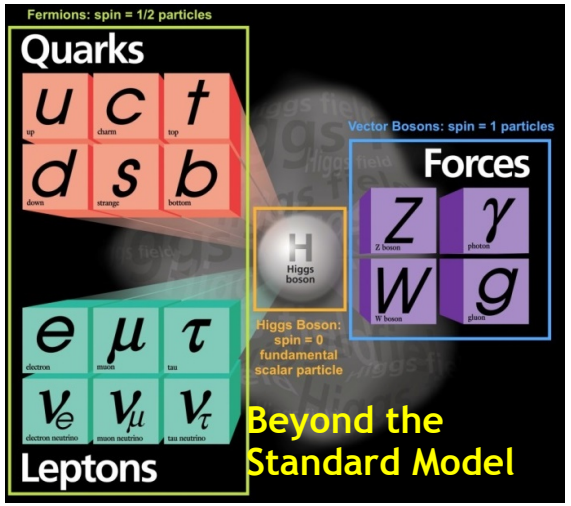
- Zhengyang Bai
  - Theme: development of new parallelization techniques for Hierarchical Matrices.
  - Parallelization: single node (extensive shared memory parallelization)

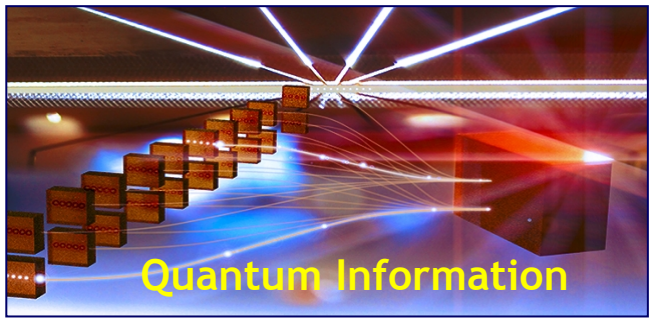
How do we bridge the gap to 150,000 nodes?

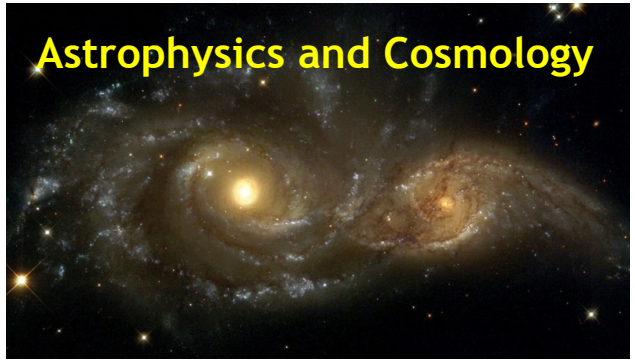
Development of next-generation relativistic atomic code for high precision calculations of properties of atoms and ions

C. Cheung, M. S. Safronova, S. G. Porsev, M. G. Kozlov, I. I. Tupitsyn

Applications



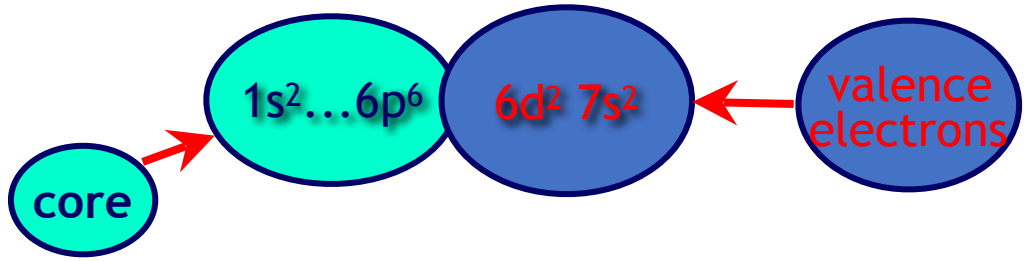




Current project: development of precision calculation methods for open-shell atom and ion properties

- Use theory to propose and design new experiments
- Which atom or ion should be used for said experiment?
- Need to know properties of atoms and ions to high precision for many applications!
- Need to know uncertainty of the theory values.

Method



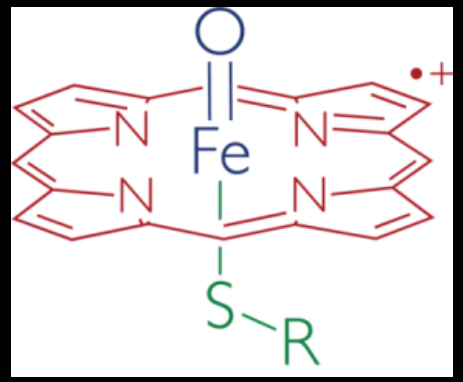
Problem 1: core-core and core-valence correlations (MBP
Problem 2: valence-valence correlations (CI+PT)

Mursaleem Ansari

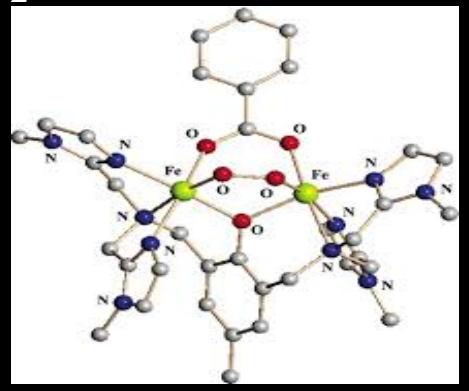
Design a new catalyst for C-H bond activation of CH_4

In biological system, methane is converted to methanol by the action of P450 enzyme

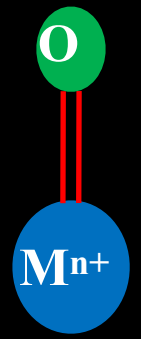
$$RH + O_2 + NADPH + H^+ \rightarrow ROH + H_2O + NADP^+$$



110.8 kJ/mol

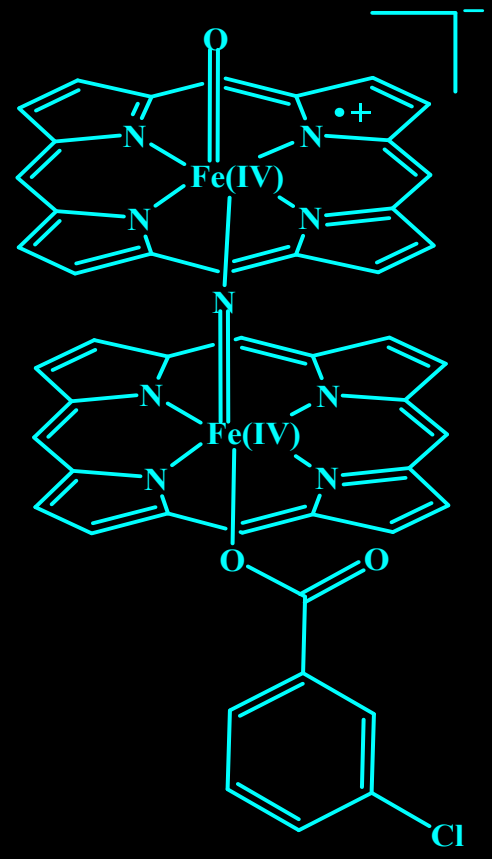


81.6 kJ/mol



Fe=O = 1.62–1.66 Å

> 1.62–1.66 Å



[(TPP)(*m*-CBA)Fe(IV)(μ-N)Fe(IV)(O)(TPP$^{\cdot+}$)]$^-$

C-H activation (CH_4)

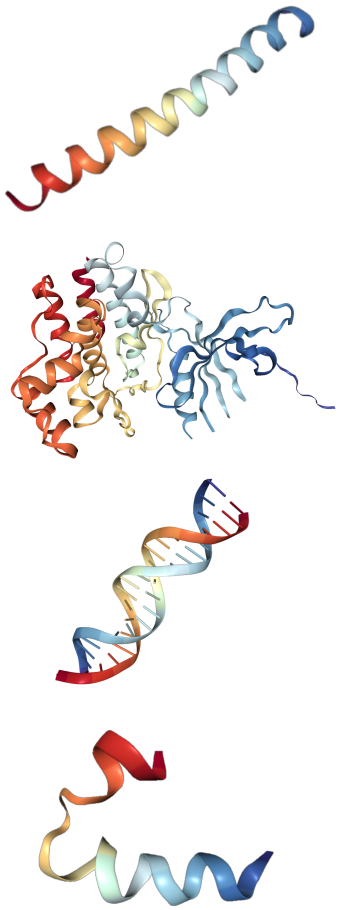
How to enhance catalytic activity ?

(1) Axial ligand effect (2) Changing donor atoms of ligand
(3) Changing ligand coordination sphere
(4) Changing geometries
(5) Effect of Lewis acid (6) Stabilized high spin state (d^4 configuration)
(7) Changing oxidation state (8) Exchange enhance reactivity (EER)

Nam, W. *et al. J. Am. Chem. Soc.* 2012, **134**, 11112.; Montellano, P. R. *et al. Chem. Rev.* 2010, **110**, 932.

ONIOM approach in an optimization of structures of polymers via elongation

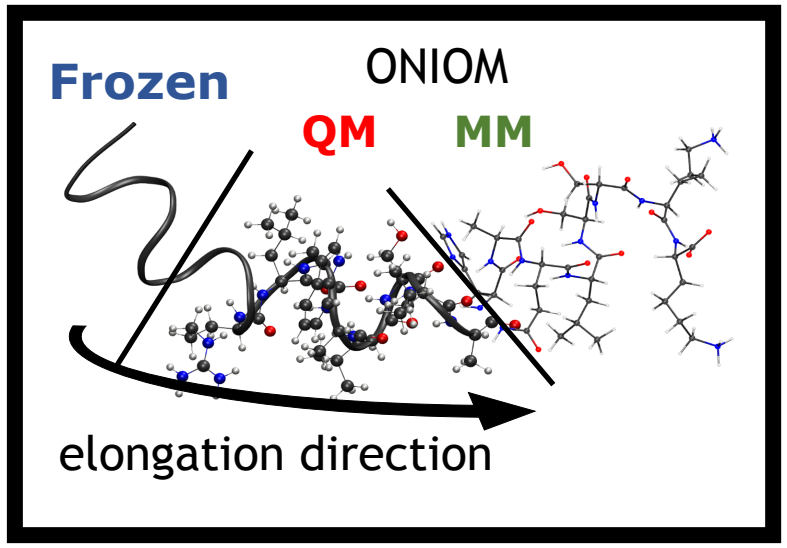
D. Mashkovtsev



polymers

Elongation: fragmentation quantum chemical method for polymers.

Simulation is polymerization like process.



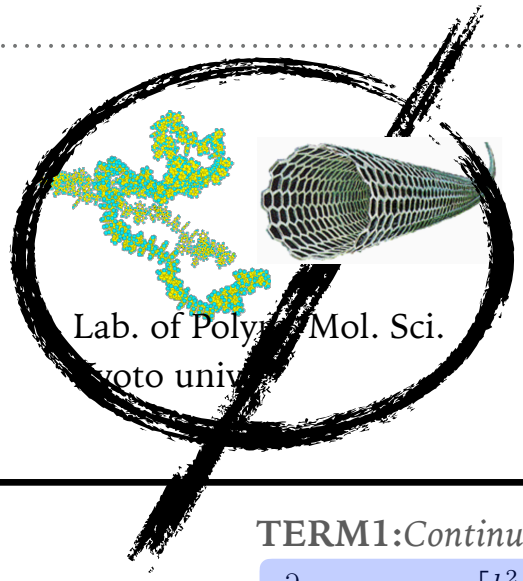
wavefunction, i.e. electronic structure

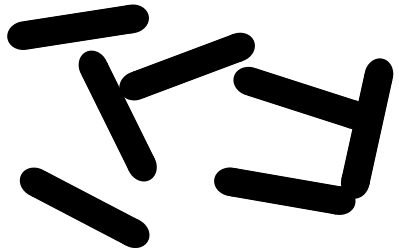
optimized geometry

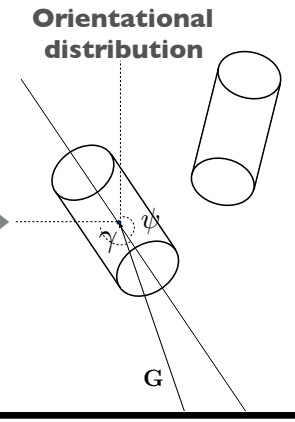
Computational complexity → O(*N*) with growing size of system

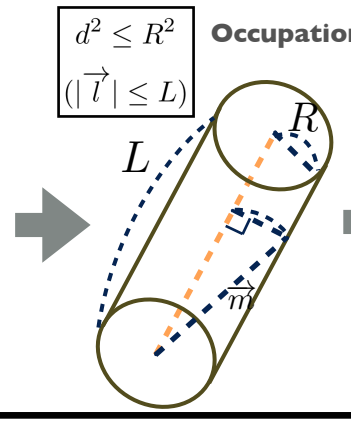
○The selection rules for mesoscale structure of block-copolymer/carbon nanotube complex system

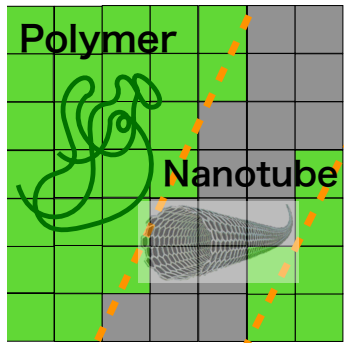
H.Y.S.Satoh Dept. of phys., TOHOKU univ.











TERM1:*Continuum approximation*

$$\frac{\partial}{\partial n} Q_K(n,\boldsymbol{r}) = \left[\frac{b^2}{6}\Delta - \beta \mathcal{V}_K(\boldsymbol{r})\right] Q_K(n,\boldsymbol{r})$$

TERM2:*Expectation from path integral*

$$\phi_K^*(\boldsymbol{r}) = \sum_m \frac{\int d\boldsymbol{r}_k \int d\boldsymbol{r}_l Q_K(k \mid m) Q_K(m \mid l)}{\int d\boldsymbol{r}_k \int d\boldsymbol{r}_l Q_K(k \mid l)}$$

$$\begin{cases} Q_K(\vec{r}) \\ Q_K(\vec{r}) = 0 \quad (\forall K) \end{cases}$$

partition function $Q_K(k, \boldsymbol{r}_k; l, \boldsymbol{r}_l)$

potential field $\mathcal{V}_K(\boldsymbol{r})$

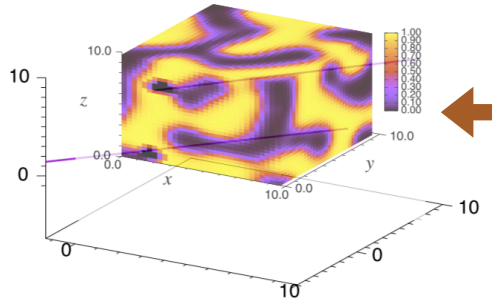
density field $\phi_K(\boldsymbol{r})$

$$\mathcal{V}_K(\boldsymbol{r}) = \frac{1}{2}\sum_{K'} k_B T \chi_{KK'} \phi_{K'}(\boldsymbol{r})$$

TERM3:*External field of inhomogeneous mixture*

$$\begin{cases} \sum_K \phi_K(\vec{r}) = 1 \\ \phi_K(\vec{r}) = 0 \quad (\forall K) \end{cases}$$

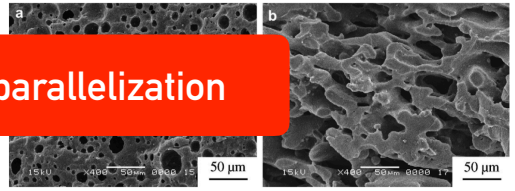
$$\begin{cases} W_K(\vec{r}) = W_K^i(\vec{r}) + W_K^s(\vec{r}) \\ \mu_K(\vec{r}) = \infty \quad (\forall K) \end{cases}$$



Global structure: No effect from fillers

$10\text{nm} \rightarrow 10\mu\text{m}$

By parallelization

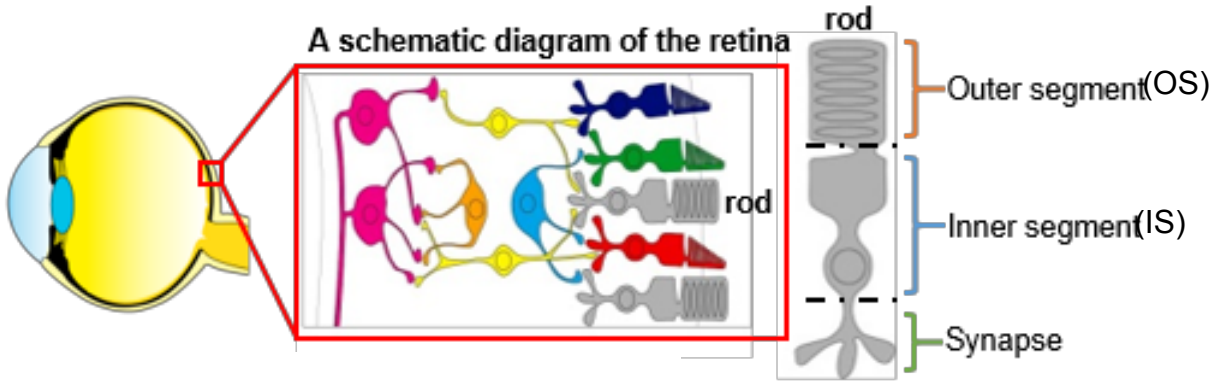


Y.Chen et al; *J. Nanomat.*,(2014)614797

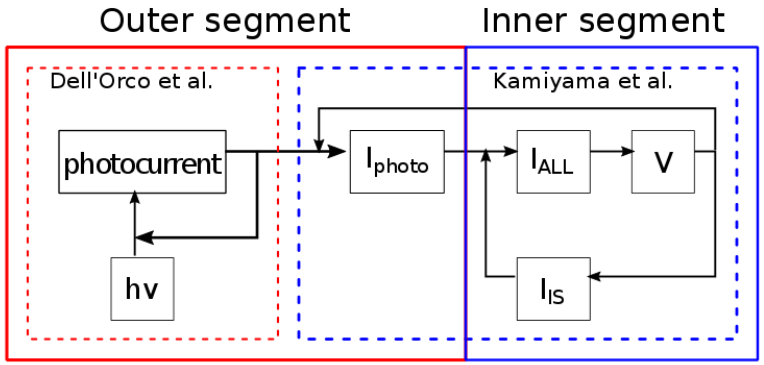
Computational modeling of rod

Akane Yamashita

①A schematic diagram of rod



② A schematic diagram of integrated model of rod

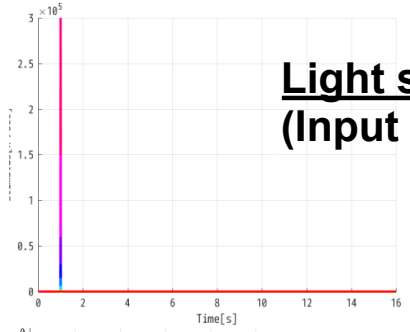


methods

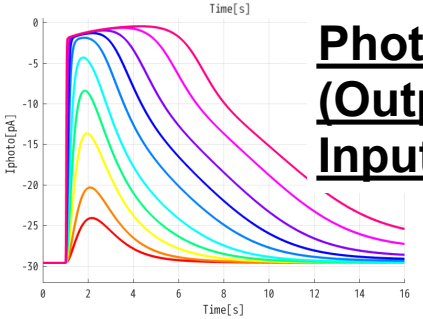
Tool : Matlab
ODE type : ODE15s
Step size : 0.01
Max step size : 0.001

We developed a computational model of rod photoreceptors with outer and inner segment.

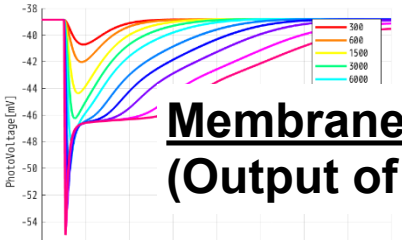
③Simulation results



Light stimuli (Input to OS)

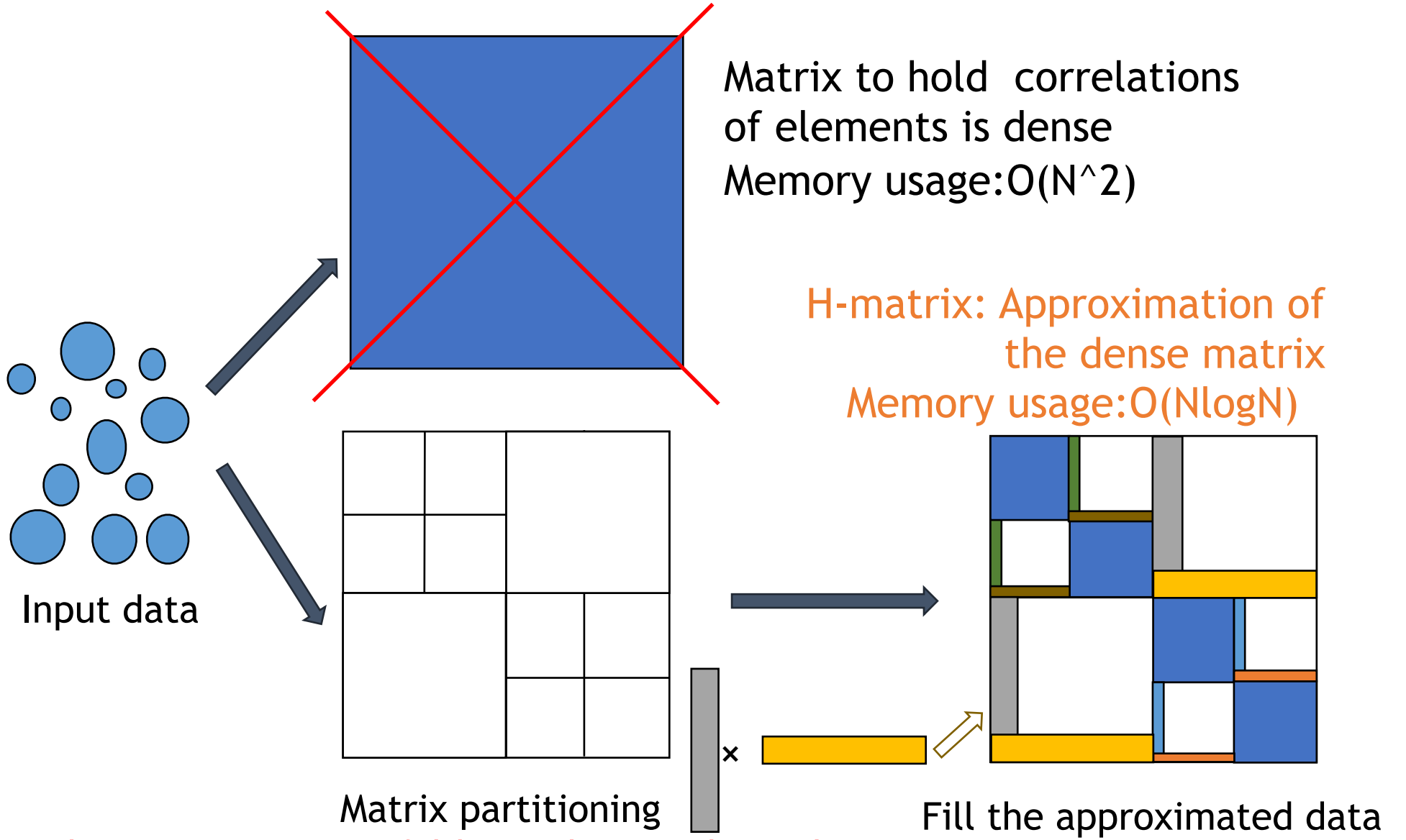


Photocurrent (Output of OS Input to IS)



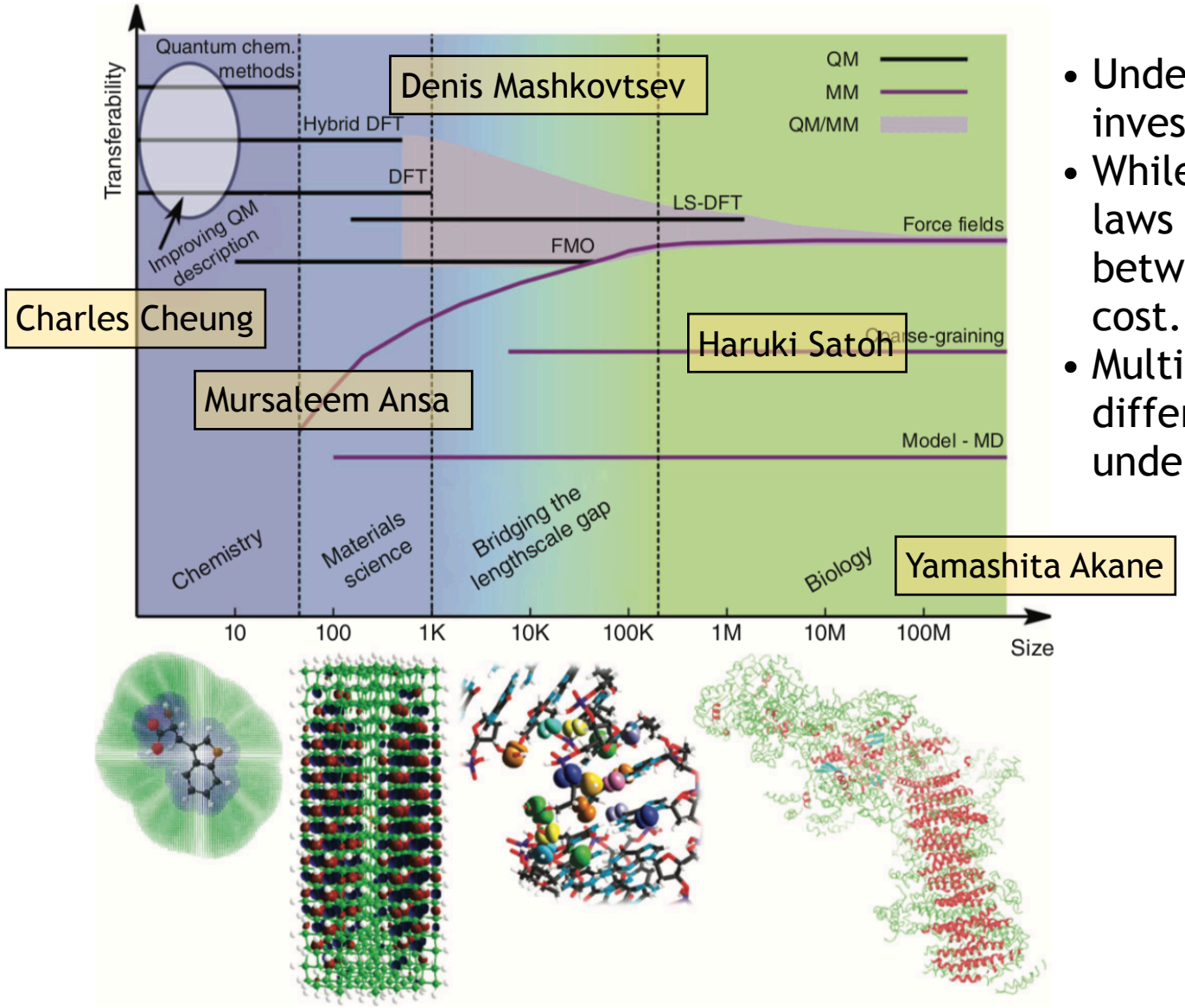
Membrane potential (Output of OS)

In simulation, the characteristics observed by experiments or previous study were reproduced.

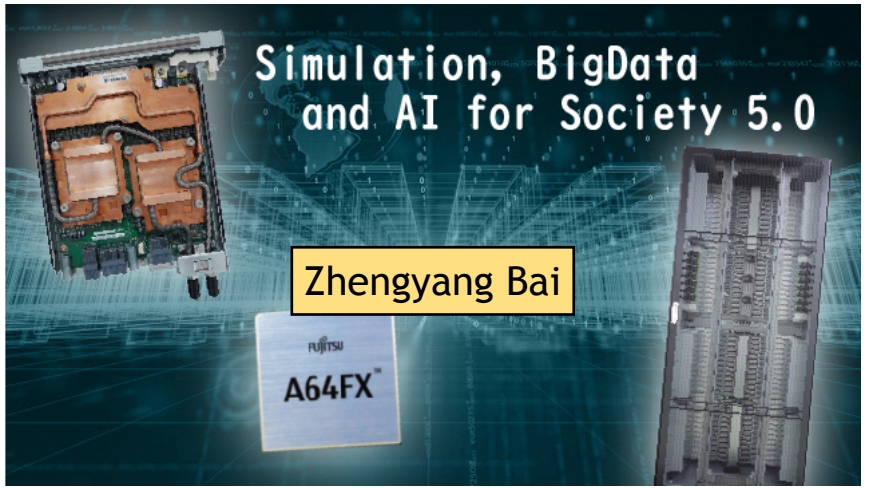


In my research, at most an 11.8-fold speedup is achieved using Task Parallel Languages

Multi-Scale Modeling of Chemical Processes



- Understanding fundamental chemical processes requires investigations at many different scales.
- While ultimately the universe is governed by the same laws of physics, in practice we need to make trade offs between the accuracy of our model, and the calculation cost.
- Multi-scale modeling works to bring together many different theories and length-scales in order to understand complex systems.



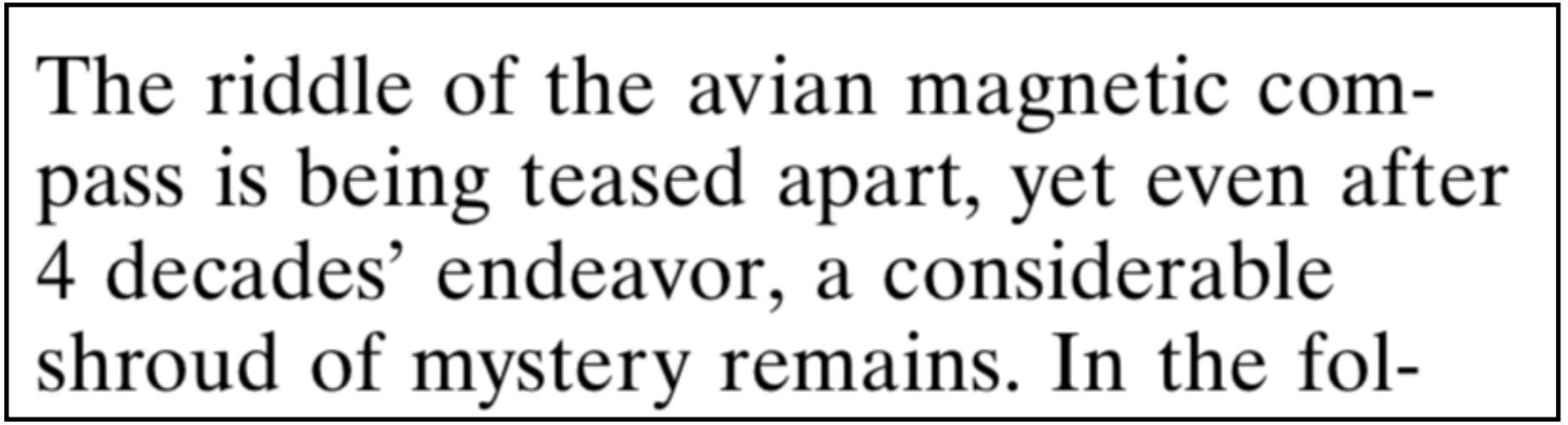
Ratcliff, Laura E., et al. "Challenges in large scale quantum mechanical calculations." *Wiley Interdisciplinary Reviews: Computational Molecular Science* 7.1 (2017): e1290.

Collaboration Opportunities - Topic

Research topic: how is it possible for birds to know how to fly north and south?

Birds somehow have the ability to see the earth's magnetosphere.

How can we understand this complex phenomenon?


The riddle of the avian magnetic compass is being teased apart, yet even after 4 decades' endeavor, a considerable shroud of mystery remains. In the fol-

Rodgers, Christopher T., and Peter J. Hore. "Chemical magnetoreception in birds: the radical pair mechanism." *Proceedings of the National Academy of Sciences* 106.2 (2009): 353-360.

Collaboration Opportunities - Topic

- Crytpochrome protein: it is theorized that this protein can let the birds see the direction as a color.
- The underlying mechanism is electron spin, and its interaction with the magnetosphere.
- Requires understanding the interaction between proteins and blue light, and thus calculations with high level quantum chemistry.
- But the system contains 10,000 atoms, which is out of reach.

The role of the electron spin in chemistry and biology has been receiving much attention because of a plausible relation to electromagnetic field effects on living organisms (1), and due to the seemingly importance of the earth's magnetic field on birds and fish navigation (2). Part of the difficulty in studying the subject arises from the lack of a physical model that can rationalize these phenomena. Recently, the chiral-induced spin selectivity (CISS) effect was observed in electron transmission and conduction through organic molecules (3). The spin selectivity was observed for photoelectron transmission through monolayers of double-stranded DNA adsorbed on gold (4). Another study discovered a spin dependence in the conduction through single molecules of double-stranded DNA. In this configuration, one end of the molecule was adsorbed on a Ni substrate, whereas the other was attached to a gold nanoparticle (5).

Mishra, Debabrata, et al. "Spin-dependent electron transmission through bacteriorhodopsin embedded in purple membrane." ***Proceedings of the National Academy of Sciences*** **110.37 (2013):**

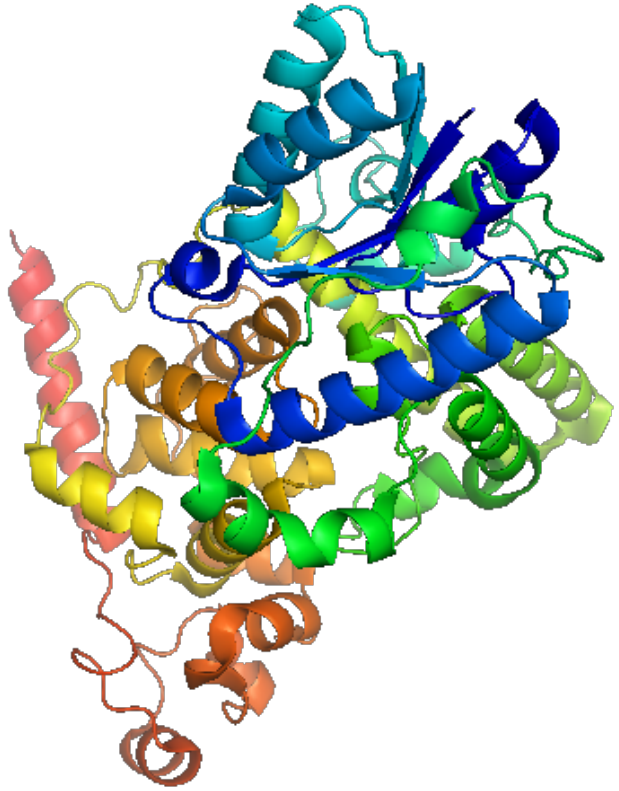
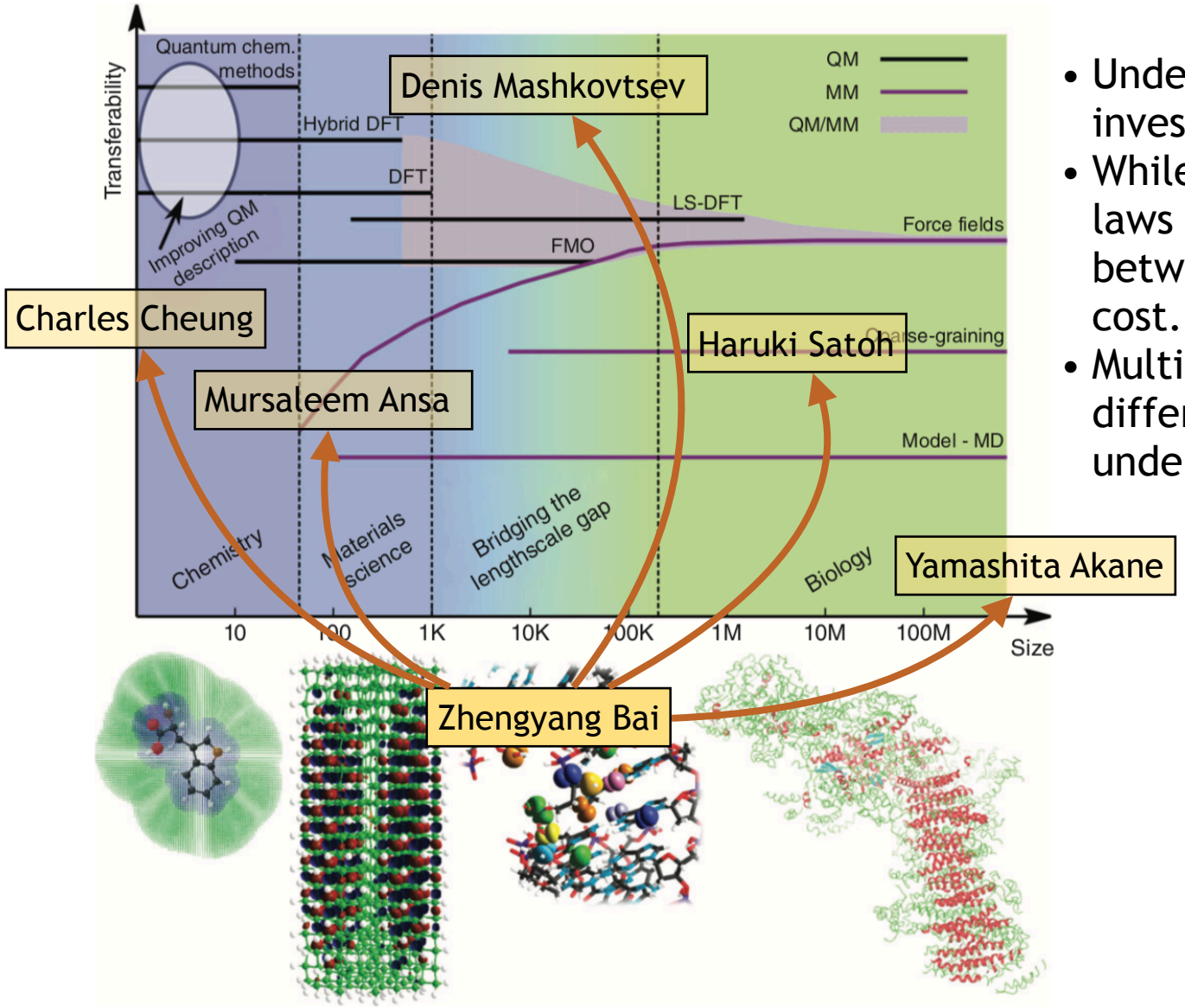


Image taken from: https://en.wikipedia.org/wiki/Cryptochrome

Multi-Scale Modeling of Chemical Processes



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Ratcliff, Laura E., et al. "Challenges in large scale quantum mechanical calculations." *Wiley Interdisciplinary Reviews: Computational Molecular Science* 7.1 (2017): e1290.

Collaboration Opportunities - Challenges

- How do you combine all these programs together?
  - CI+PT, SCF Solver, GAMESS, Gaussian, Matlab, Task Manager
- How can we load balance these operations, when each task takes different amounts of time.
- How to test that it is working when all results are involve complex couplings?
- How to include experimentalists into the pipeline?
- Who can manage this long sequence of calculations?
- How many computer hours do you need?

Collaboration Opportunities - How To Start?

- Determine what the final result should look like.
  - Movie showing how the molecules behave at various scales.
  - High level plot of the impulse response.
- Tune each part of the calculation, and find the system sizes that can be quickly computed.
- Describe the various steps of the calculation and dependencies.
  - For each part, what dominate interactions.
- Gather the starting experimental data.
  - Initial conditions.
  - Experimental data for each scale that we can reproduce.
- Find parts of the calculation that need new kinds of theory.
- Create model systems to test on for each step of the calculation.