Machine learning for ultracold quantum dynamics

Roman Krems

University of British Columbia

Quantum scattering theory is critically important for ultracold physics

Predictions of scattering lengths

Identification of resonances

Feasibility of sympathetic/evaporative cooling

Understanding collision losses

Ultracold chemistry

Understanding collisional decoherence

However, scattering calculations – particularly at low temperatures – are challenging

Atomic/molecular collisions are extremely sensitive to interaction potentials

External fields are non-perturbative

Converged calculations of molecular dynamics require large basis sets

Time-dependent quantum dynamics difficult to apply

J. Huang, S. Liu, D. H. Zhang, and R. V. Krems, Time-dependent wave packet dynamics calculations of cross sections for ultracold scattering of molecules, *Phys. Rev. Lett* ***120***, 143401 (2018)

Often, one needs to solve the equivalent of inverse problems

Measurements of resonances

$\longrightarrow$ scattering lengths

$\longrightarrow$ bound state energies

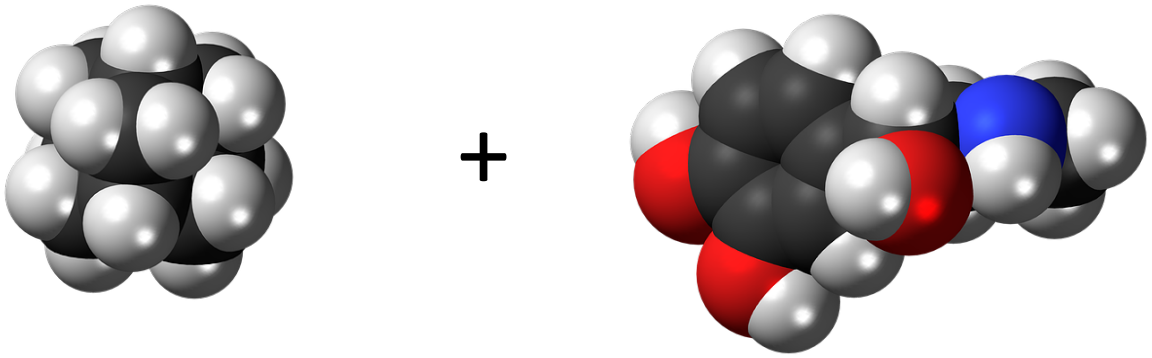
$\longrightarrow$ fine and hyperfine structure couplings

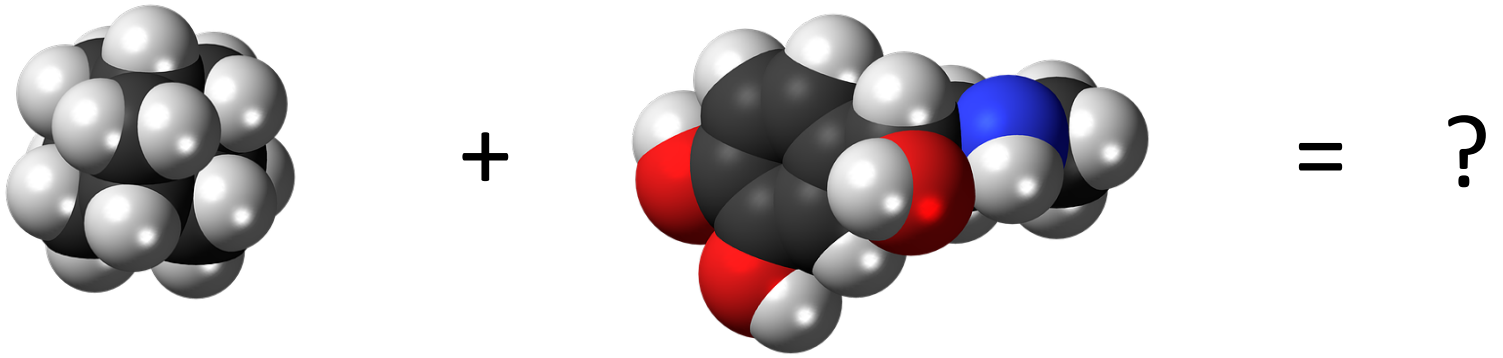
Trap loss / collision cross sections

$\longrightarrow$ reactive potential surfaces

$\longrightarrow$ long-range interactions

Inverse problems are difficult … especially for complex molecules



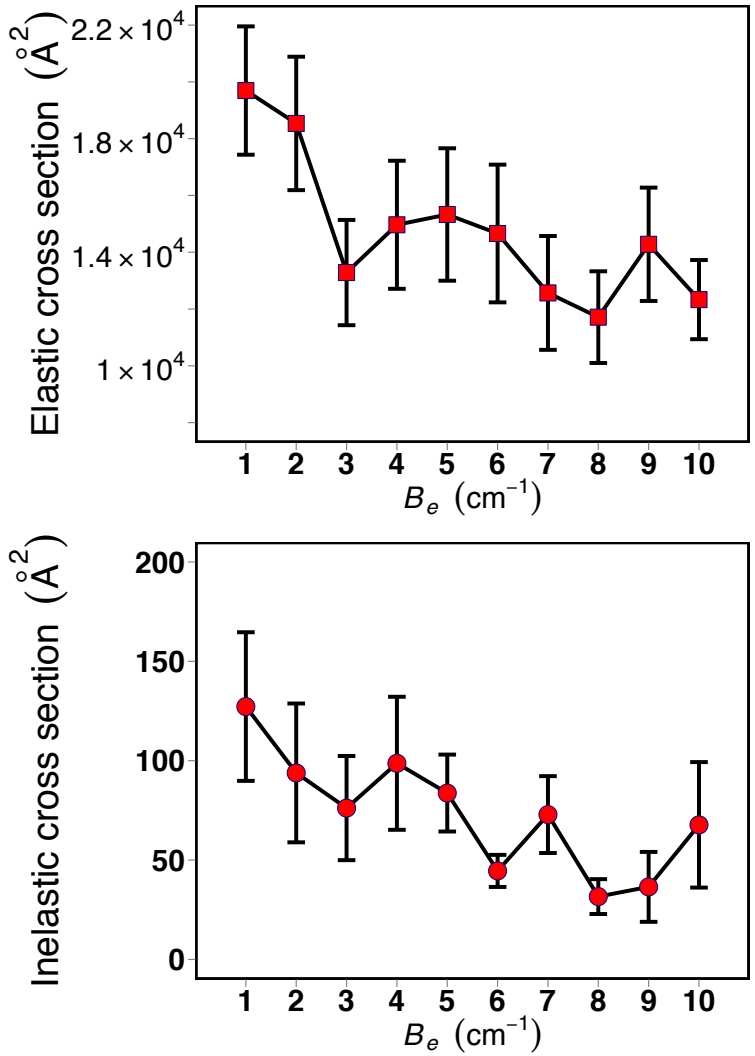


Direct problems:

PES always come with errors

How to determine the effect of errors in PES on predictions of dynamical observables?

Average over variations of the PES



Error bars are obtained by simple scaling of PES

But this does not account for errors in anisotropy

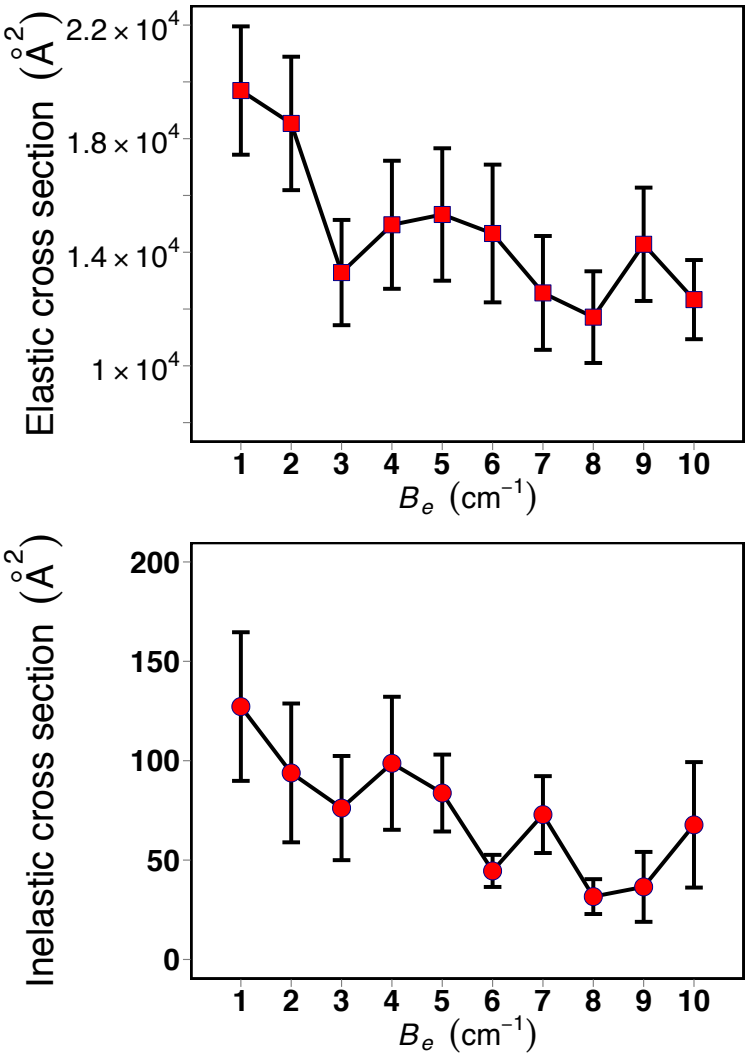
Required:

analysis of scattering observables as functions of strength and topology of PES

Particularly important for polyatomic molecules!

Jie Cui and RK, *Phys. Rev. A* **88**, 042705 (2013).

Average over variations of the PES



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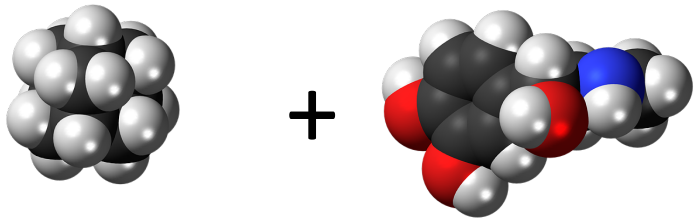
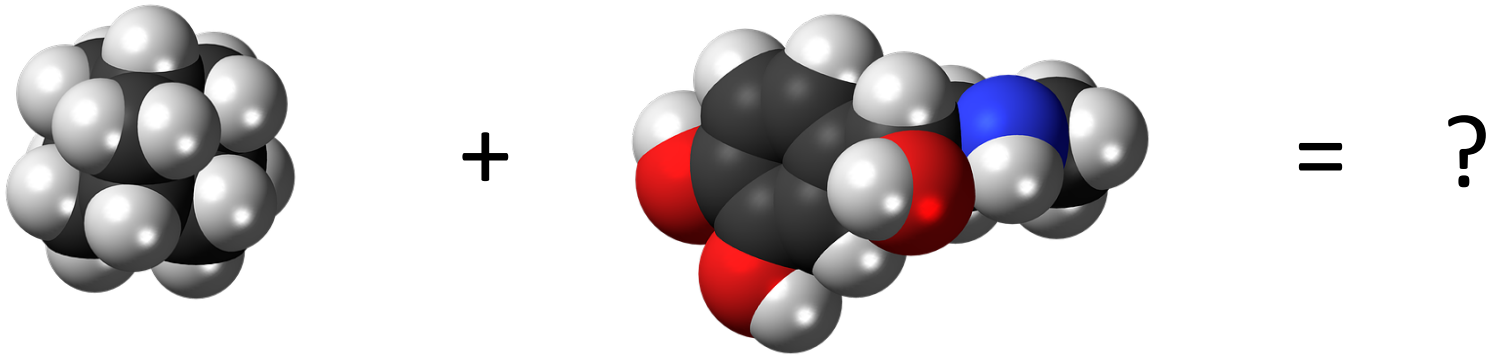


Jie Cui and RK, *Phys. Rev. A* **88**, 042705 (2013).



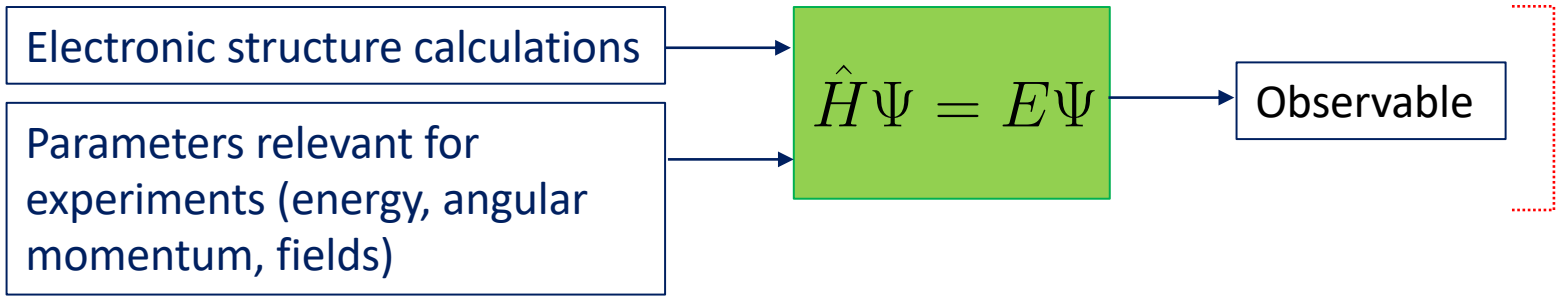
Inverse problems:

High-dimensional problems for complex species

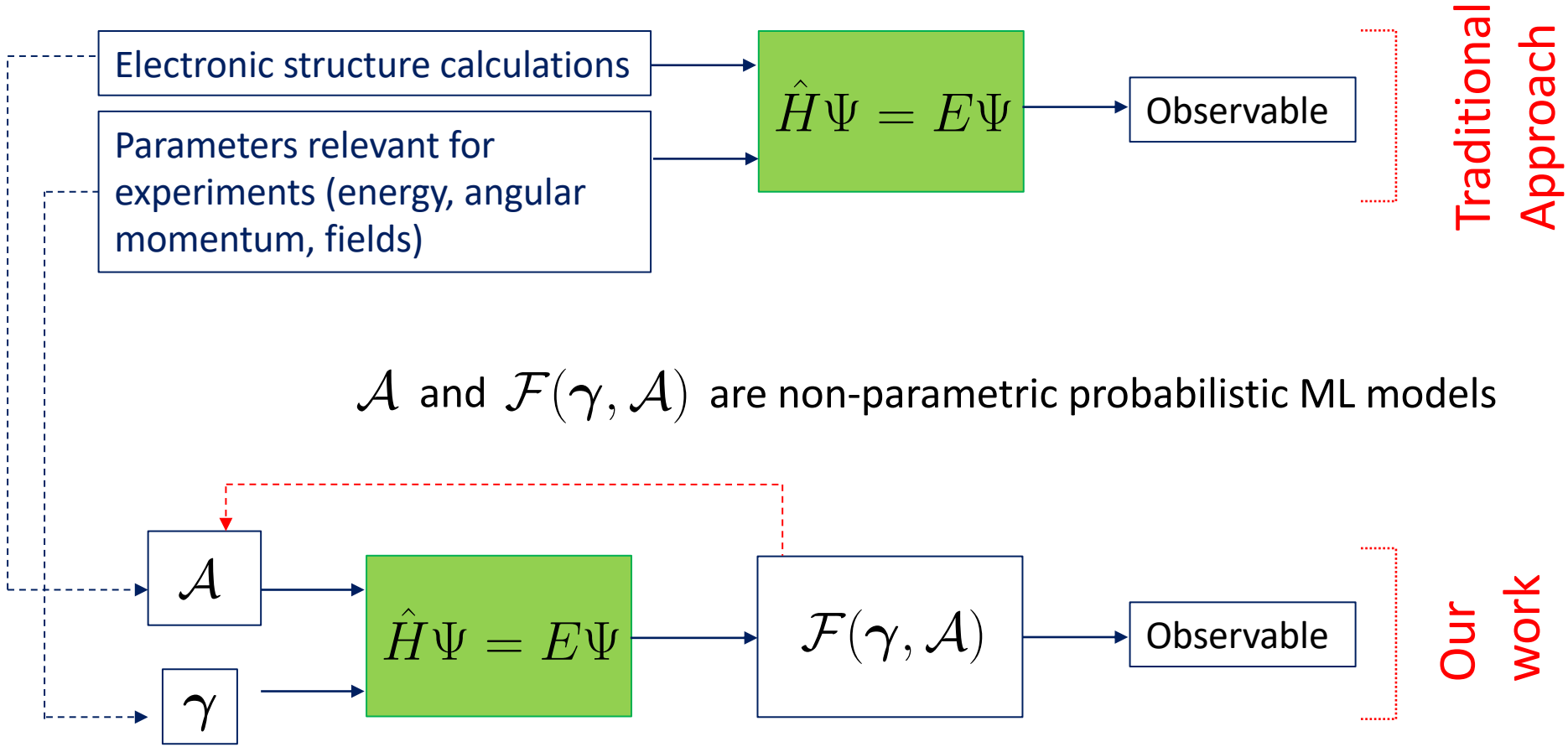
How to obtain PES from measured observables?

I will show that and how these challenges can be addressed with machine learning!

Quantum dynamics



Quantum dynamics

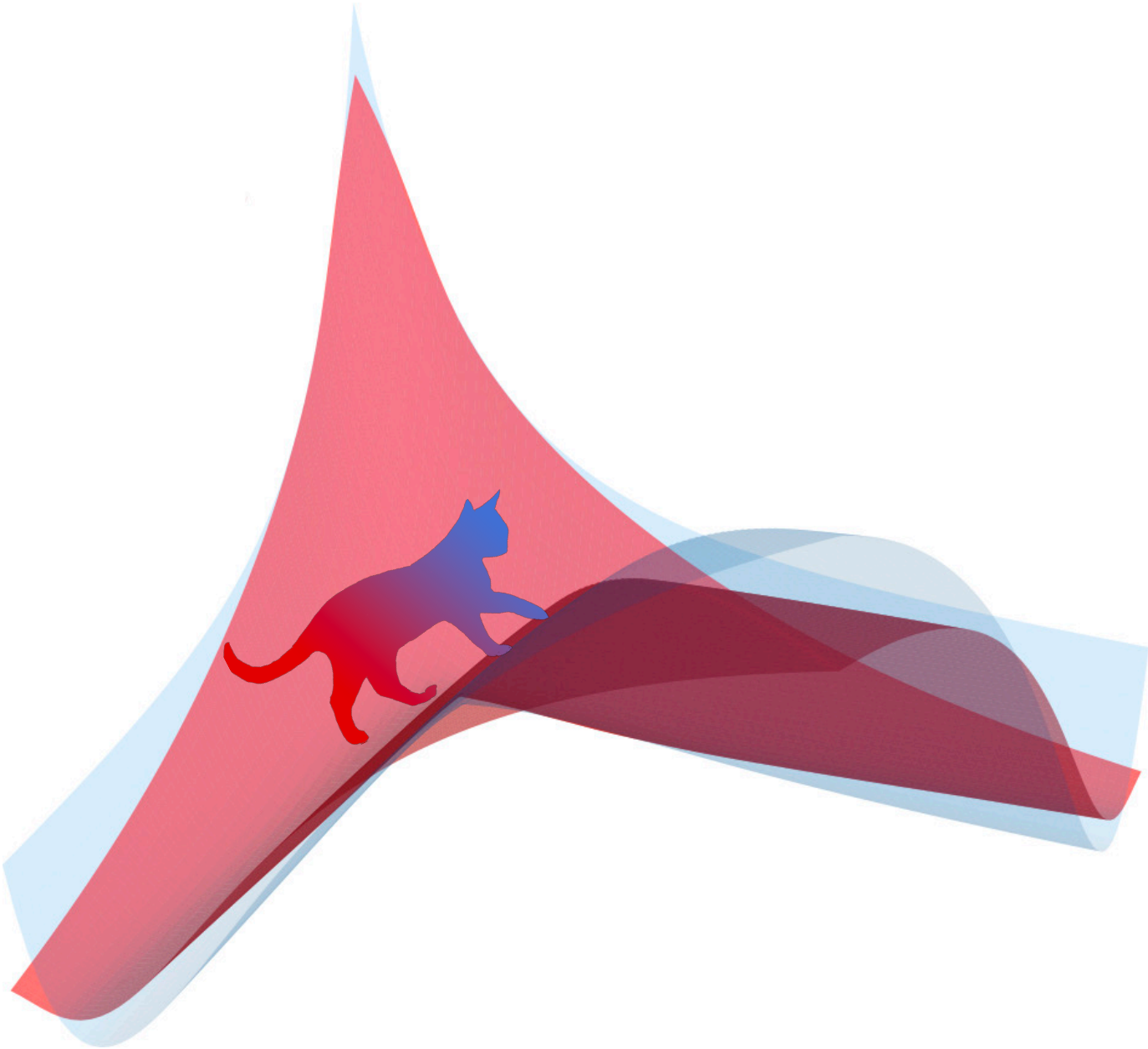


Traditional approach:

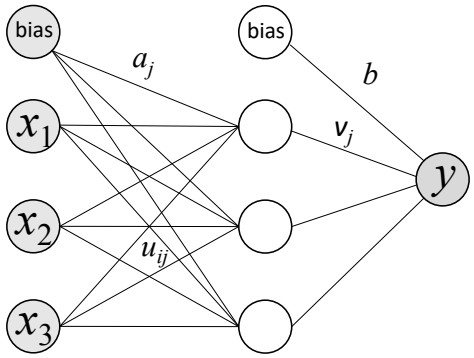
Treat atomic/molecular dynamics as evolving on a well-defined precomputed PES

Our approach:

Treat atomic/molecular dynamics as evolving on a non-parametric series of PES that is conditioned by observables



Neural networks



Require Big data
Often challenging to train

"…suffering is a perfectly natural part of getting a neural network to work well…" – Andrej Karpathy, https://karpathy.github.io/2019/04/25/recipe/

In physics

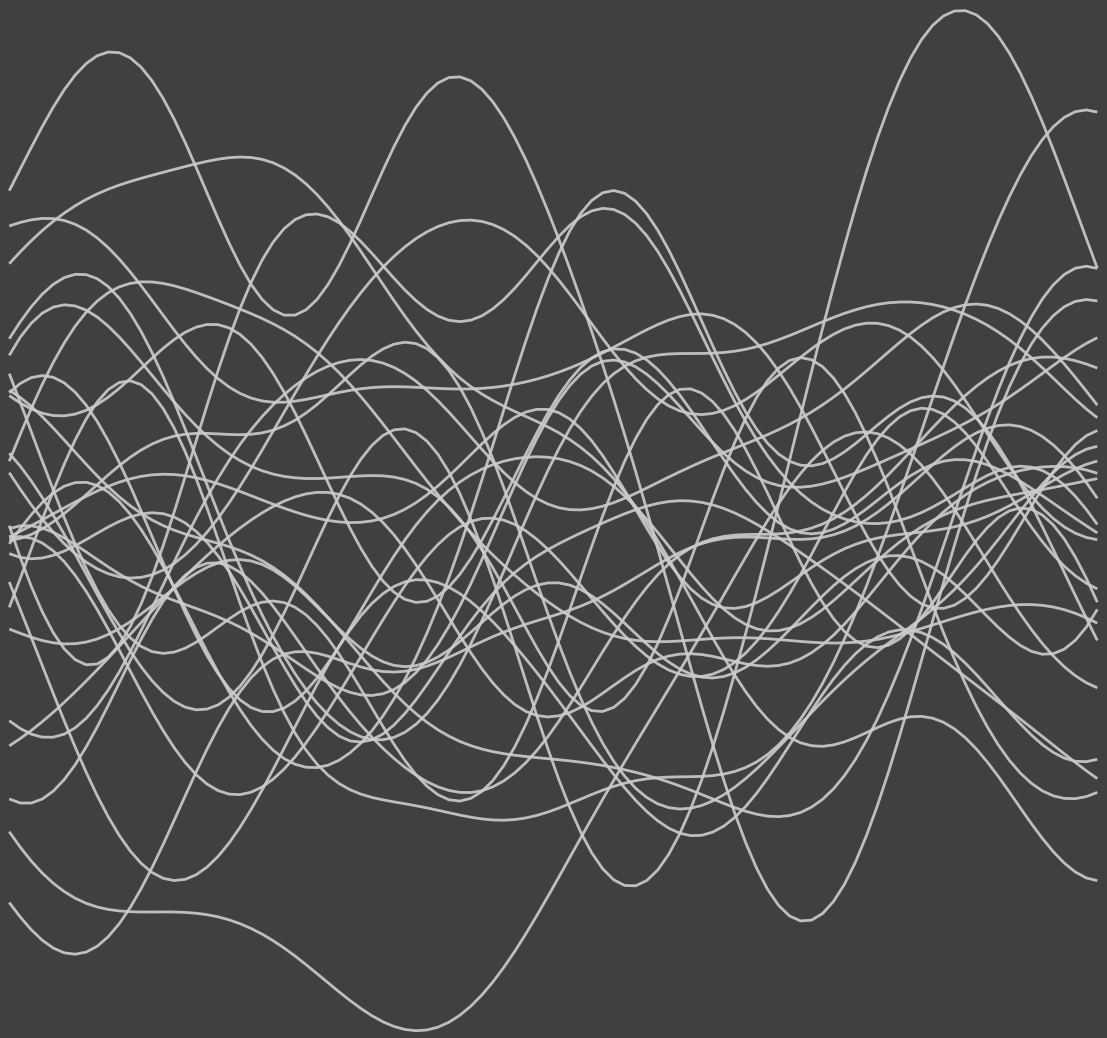
Data come from the solutions of complex equations
or from experiments
Data are expensive
The focus should be on making physical predictions based on small data

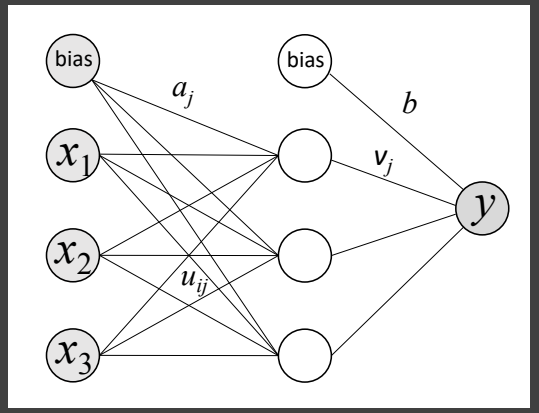
Deterministic neural networks cannot be used for problems with small data

Another way to make predictions is Probabilistic Modelling

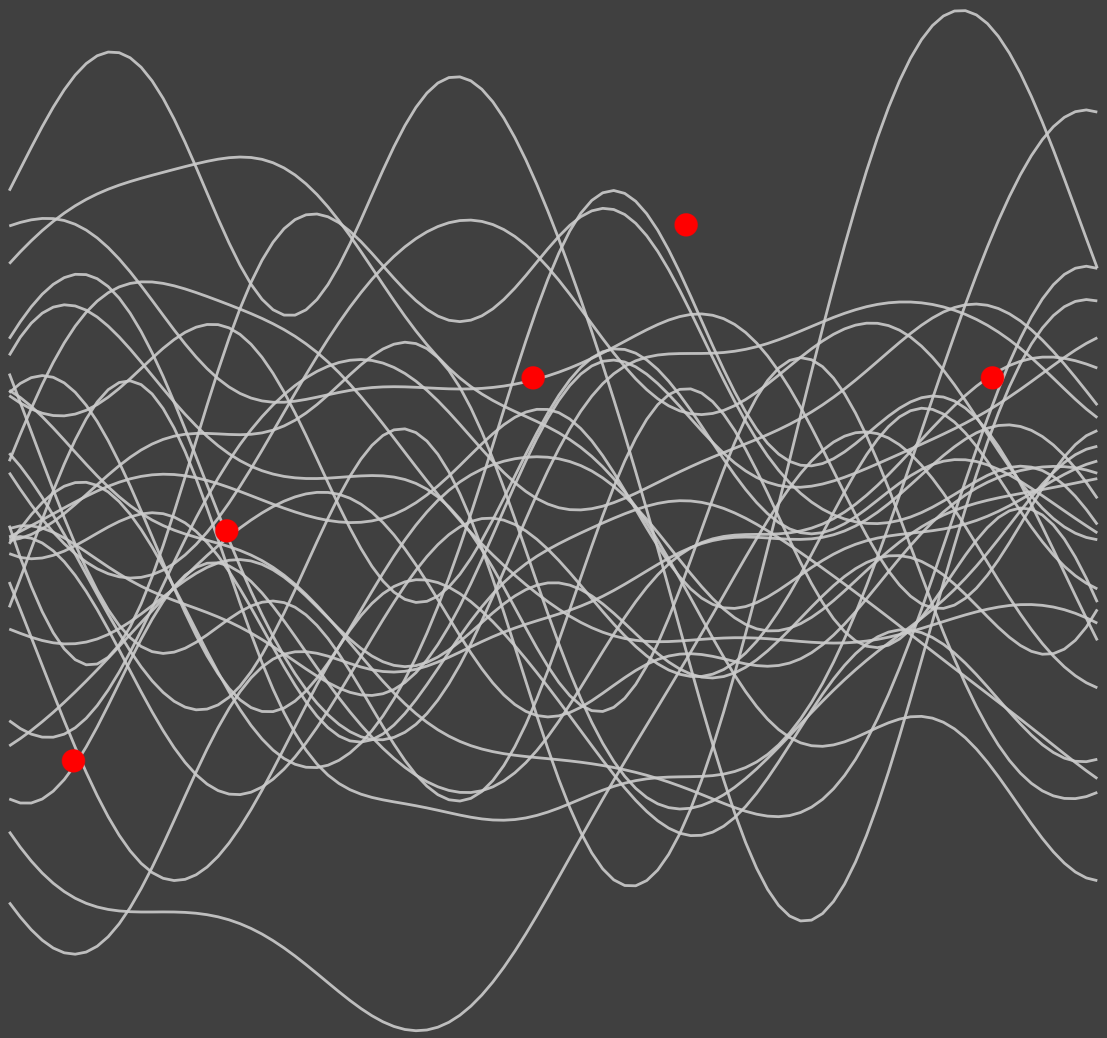
It is at the core of Bayesian machine learning

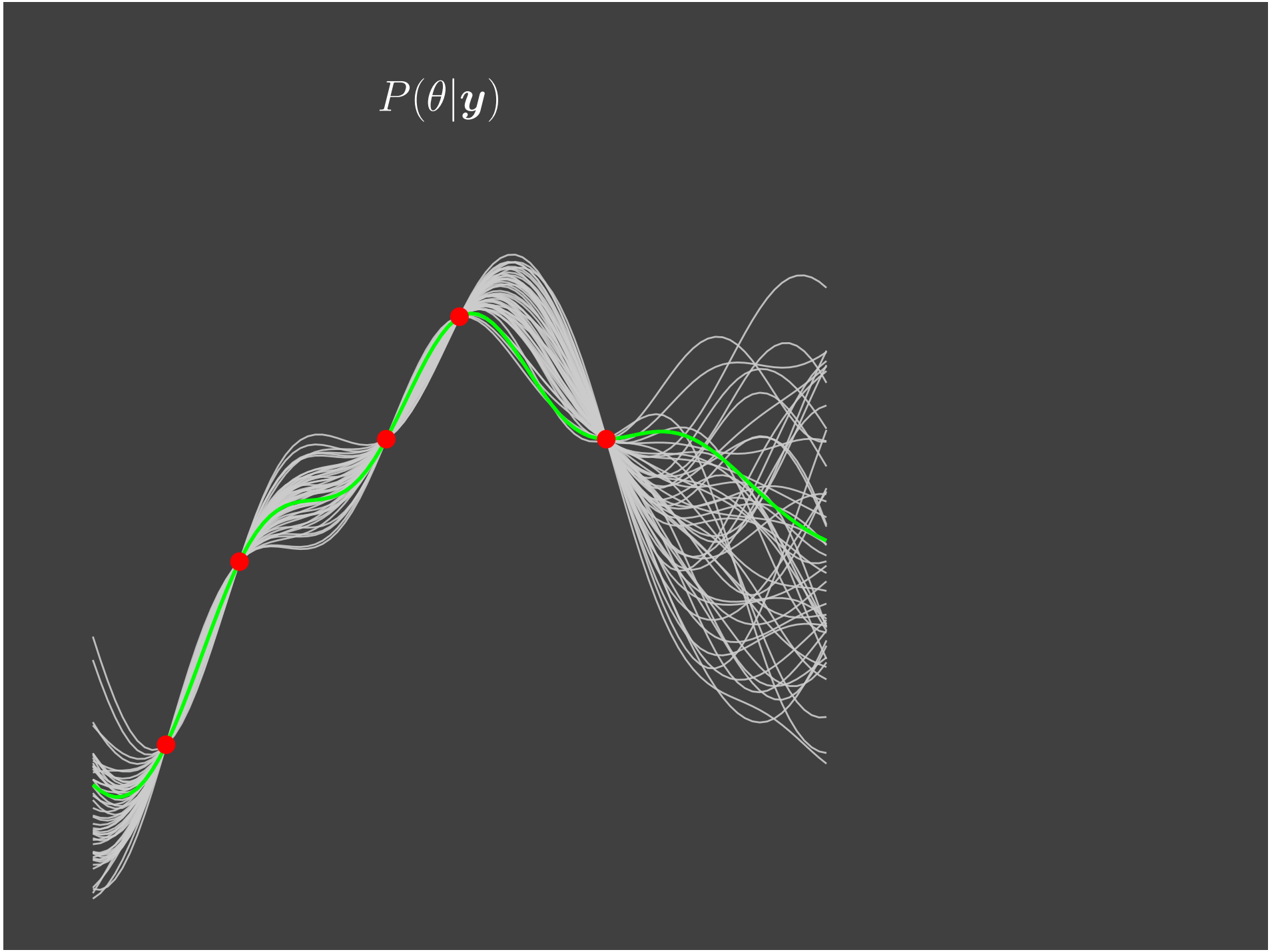
$P(\theta)$

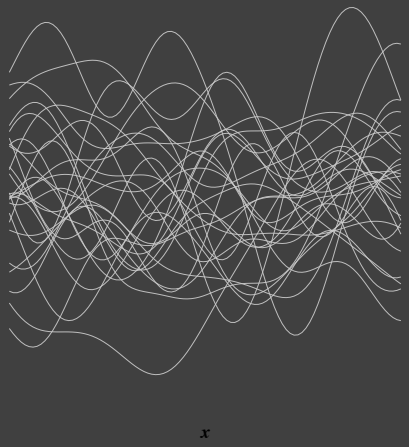




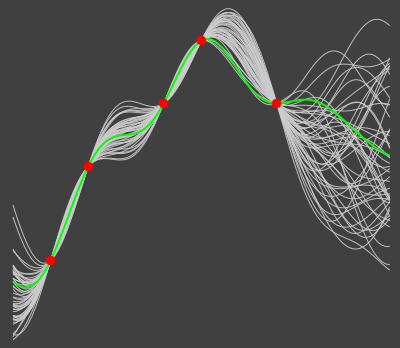
Goal: $P(\theta|\boldsymbol{y})$




$P(\theta|\boldsymbol{y})$



$P(\theta)$



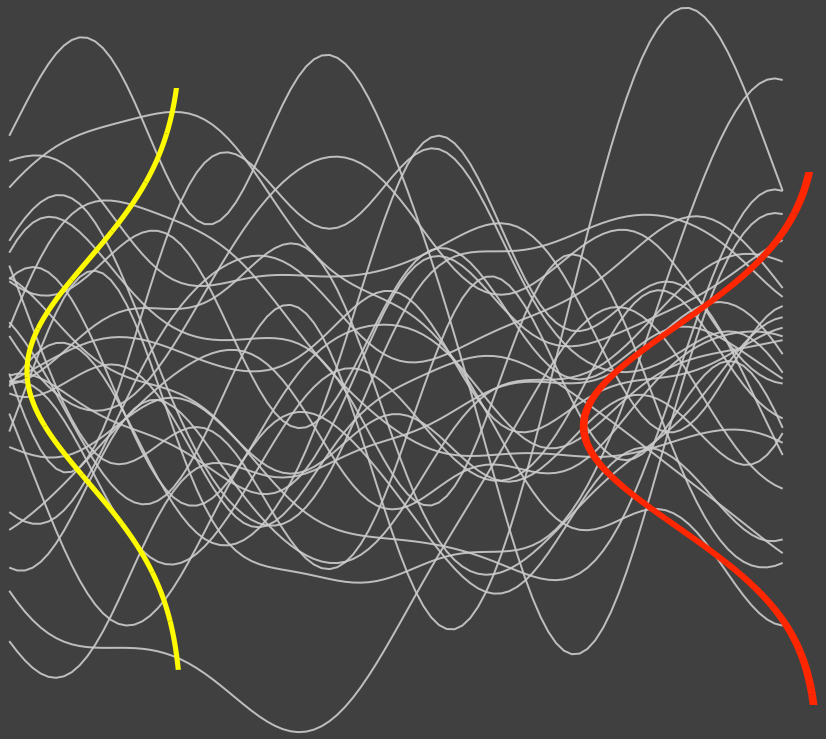
$P(\theta|\boldsymbol{y})$

Bayes' theorem:

$$P(\theta|\boldsymbol{y}) = \frac{P(\boldsymbol{y}|\theta)P(\theta)}{P(\boldsymbol{y})}$$

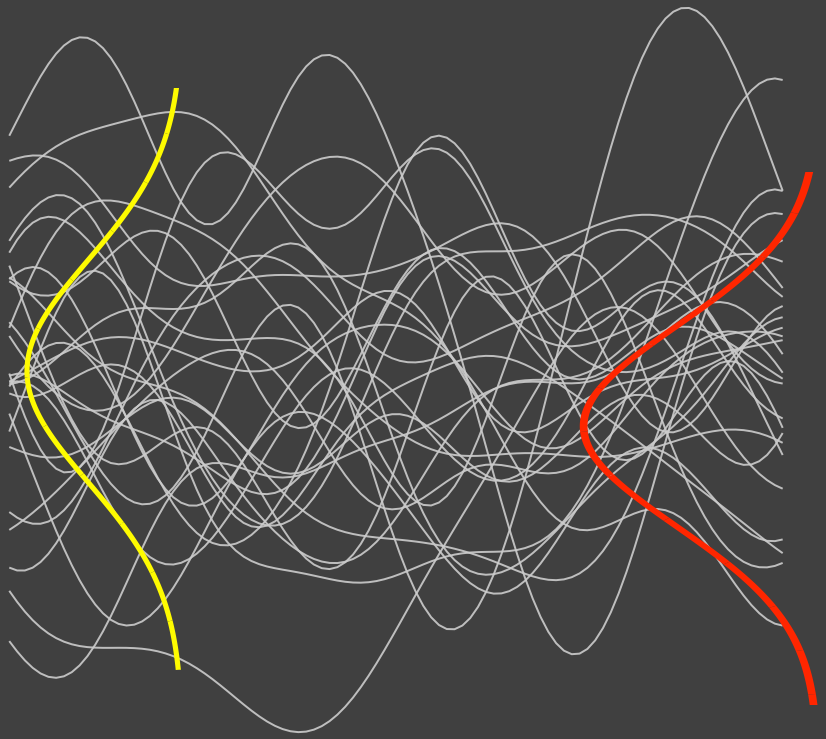
$$P(y^*|\boldsymbol{y}) = \int_\theta P(y^*|\theta)P(\theta|\boldsymbol{y})\mathrm{d}\theta$$

If the number of hidden neurons is taken to infinity, a Bayesian neural network becomes a Gaussian process



Gaussian processes are determined by a mean function and a covariance function (aka kernel function)

Choosing the proper kernel function is key to the performance of the models!



Gaussian processes are determined by a mean function and a covariance function (aka kernel function)

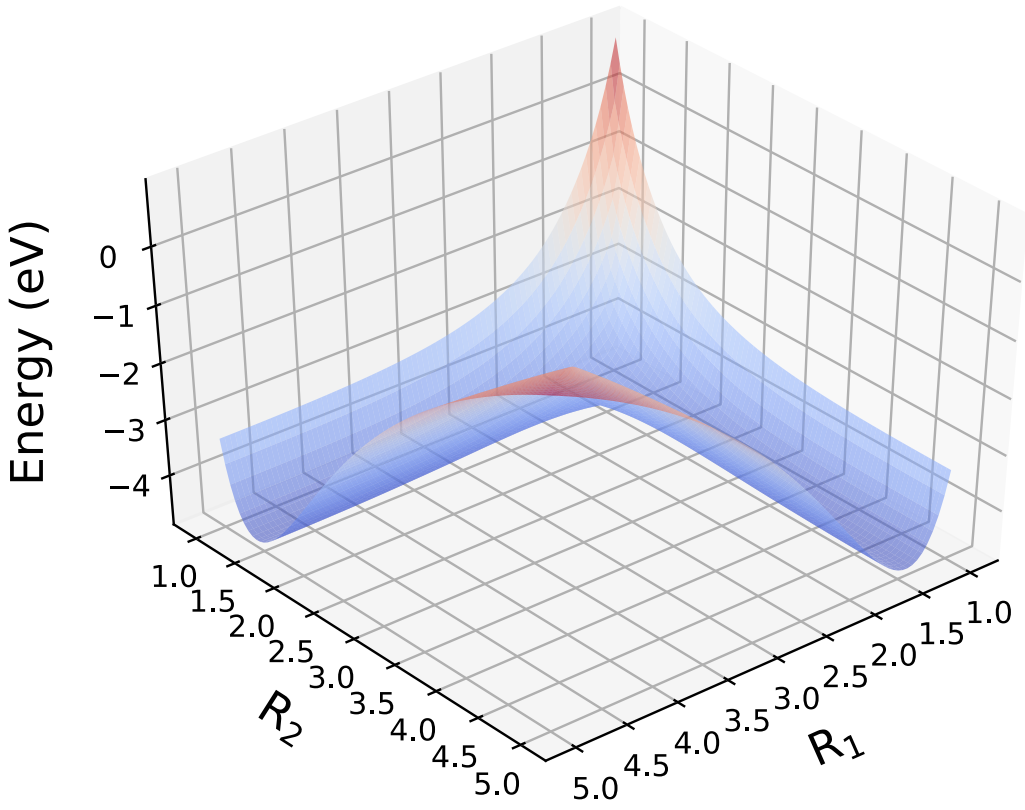
Choosing the proper kernel function is key to the performance of the models!

Check these papers on how to build the best kernels for physics applications:

R. Vargas, J. Sous, M. Berciu and R. V. Krems, *Phys. Rev. Lett.* **121**, 255702 (2018)

Jun Dai and R. V. Krems, *J. Chem. Theory Comp.* 16, 1386 (2020).

How many potential energy points does one need to know to describe the reaction?



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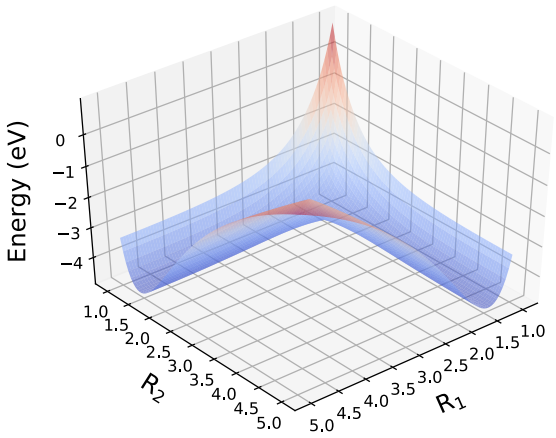
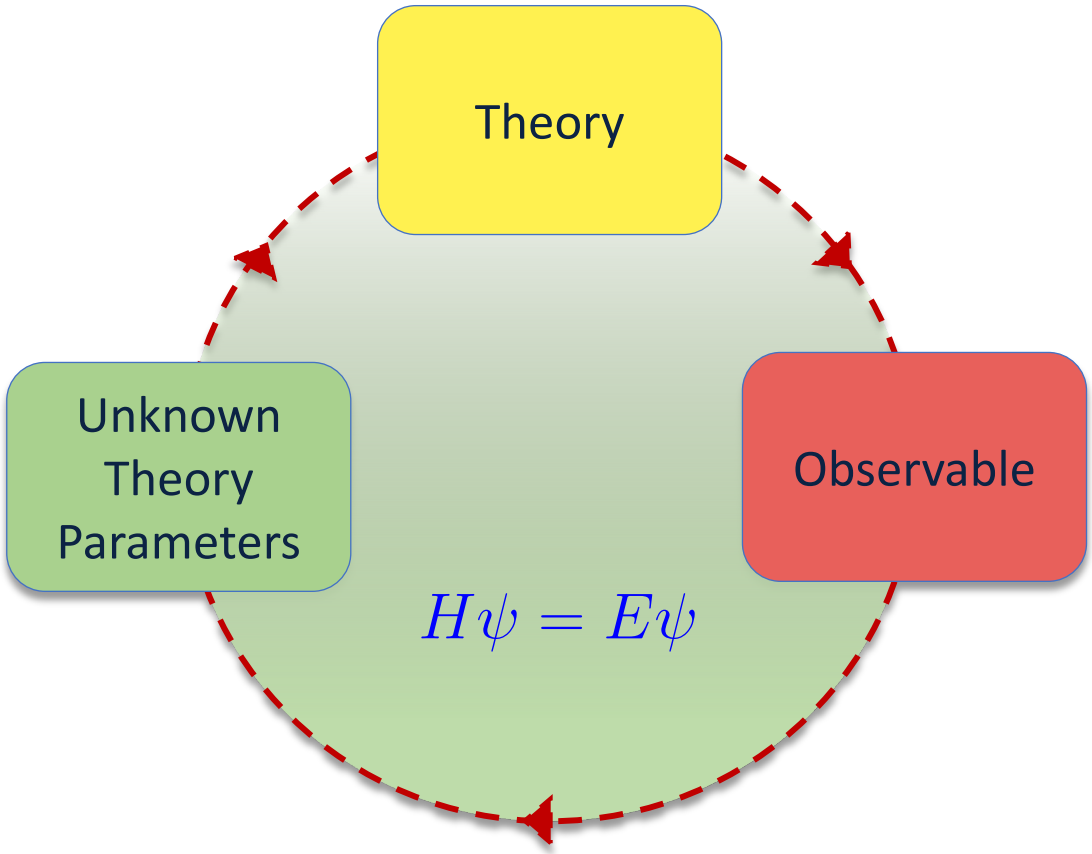
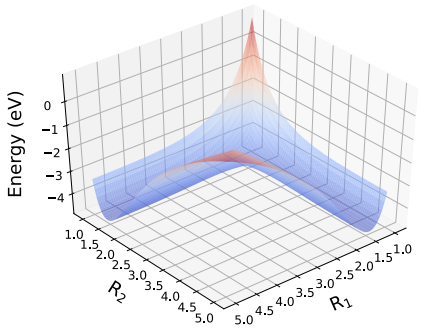
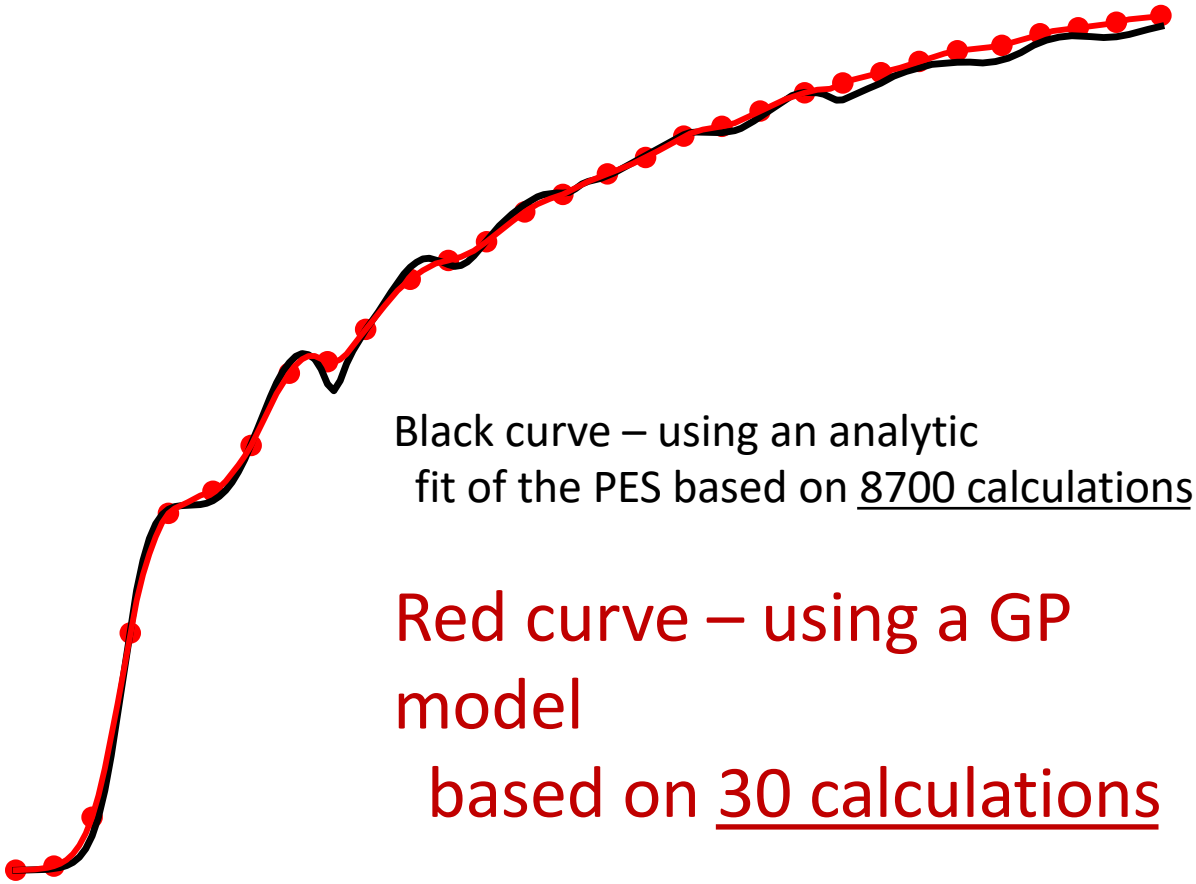






Quantum scattering calculation

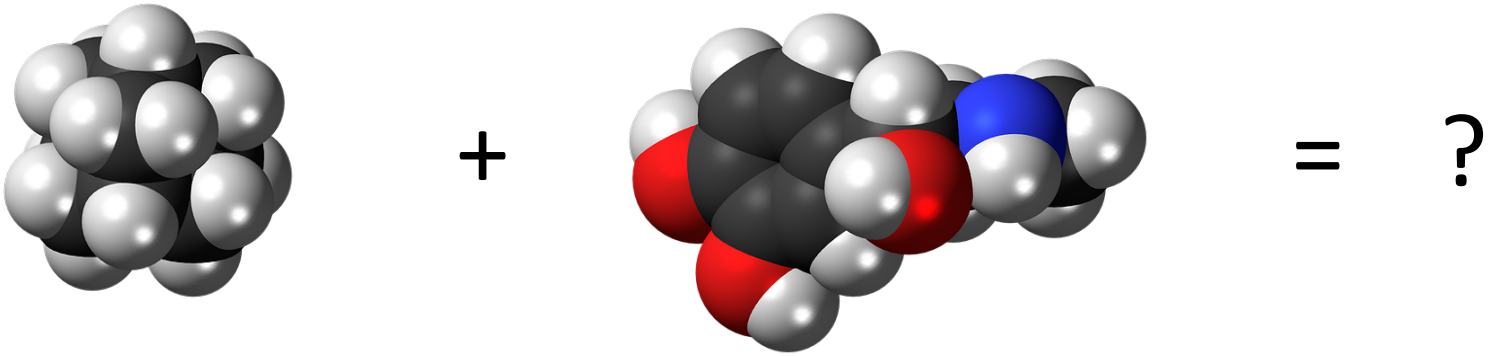


Black curve – using an analytic fit of the PES based on 8700 calculations

Red curve – using a GP model based on 30 calculations

Rodrigo Vargas, Yafu Guan, Dong Hui Zhang and RK, Bayesian optimization for the inverse scattering problem in quantum dynamics, **New J. Phys. 21, 022001 (2019)**

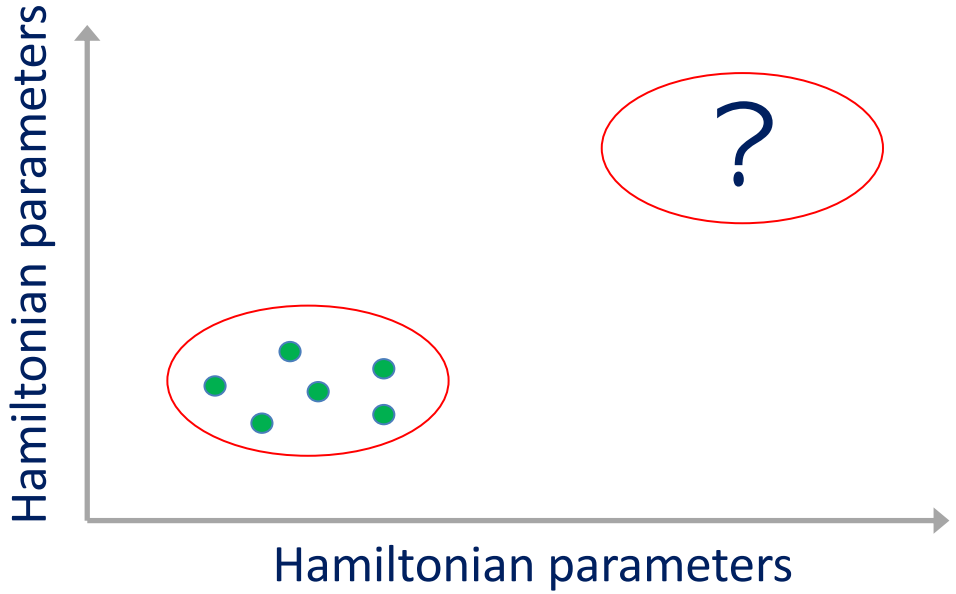
R. V. Krems, Bayesian Machine Learning for Quantum Molecular Dynamics, **PCCP 21, 13992 (2019)**

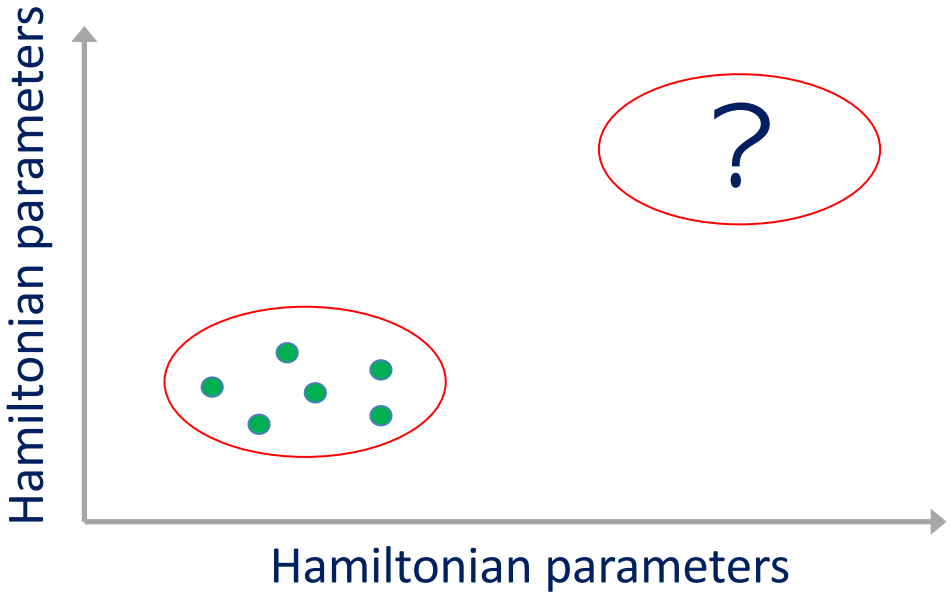


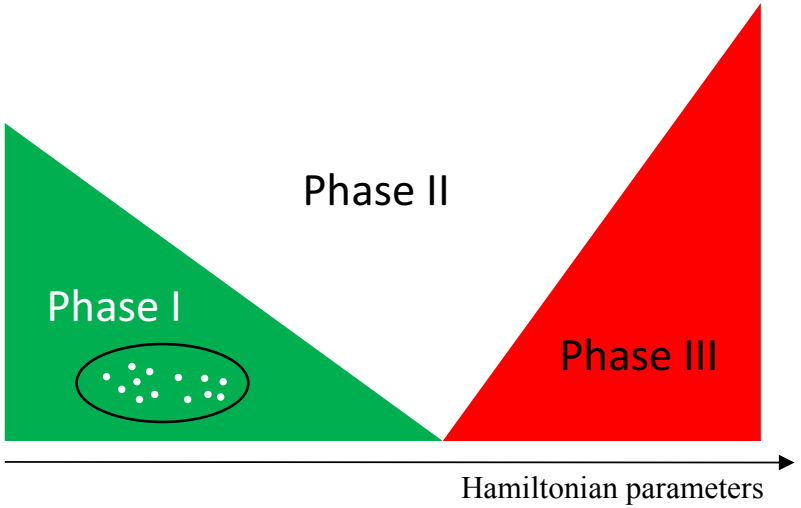
This is a problem with a high-dimensional Hamiltonian!

We want to solve it with very few quantum calculations

To do that, we need to be able to solve extrapolation problems

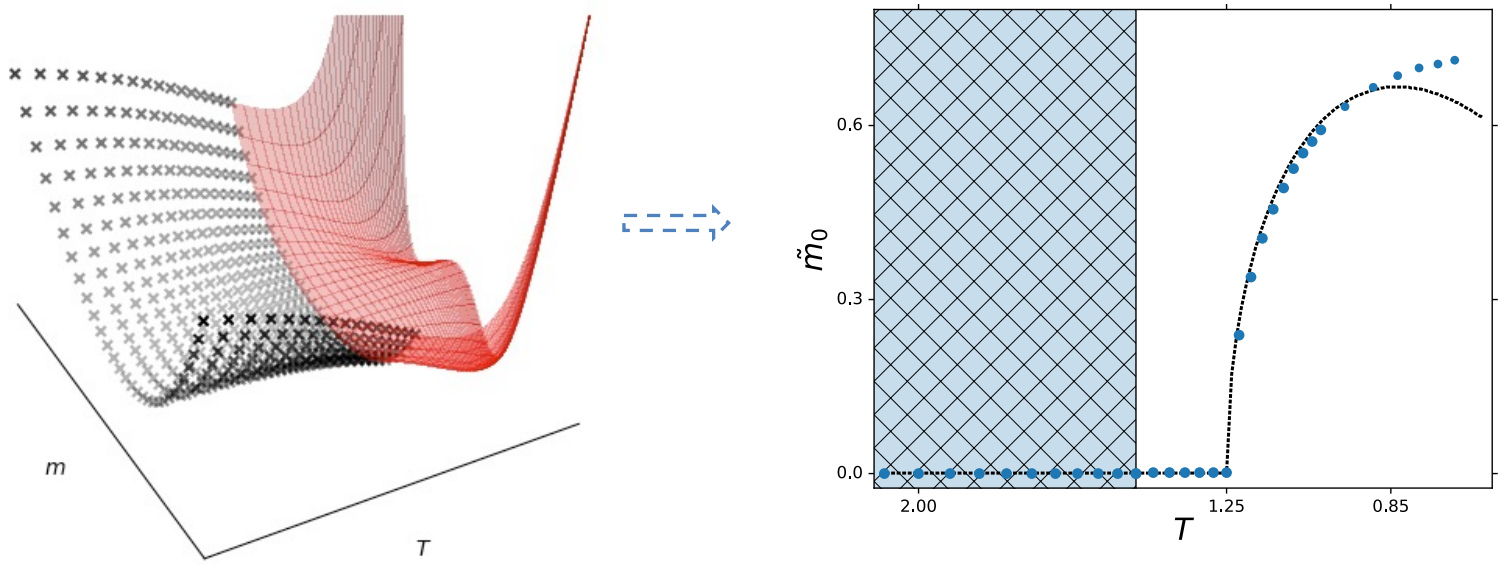






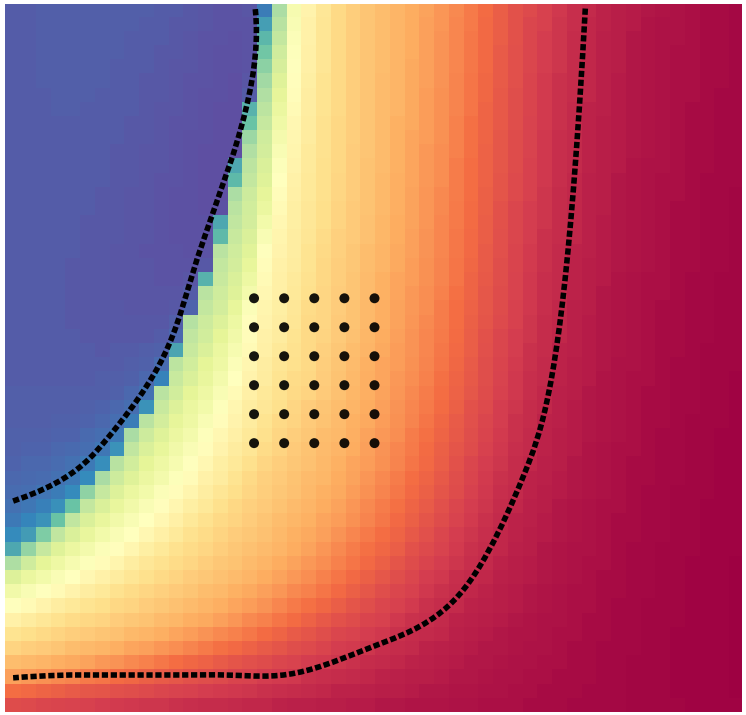
Rodrigo Vargas, John Sous, Mona Berciu and R. V. Krems, **Phys. Rev. Lett. 121, 255702 (2018)**
Jun Dai and R. V. Krems**, J. Chem. Theory Comp. 16, 1386 (2020)**

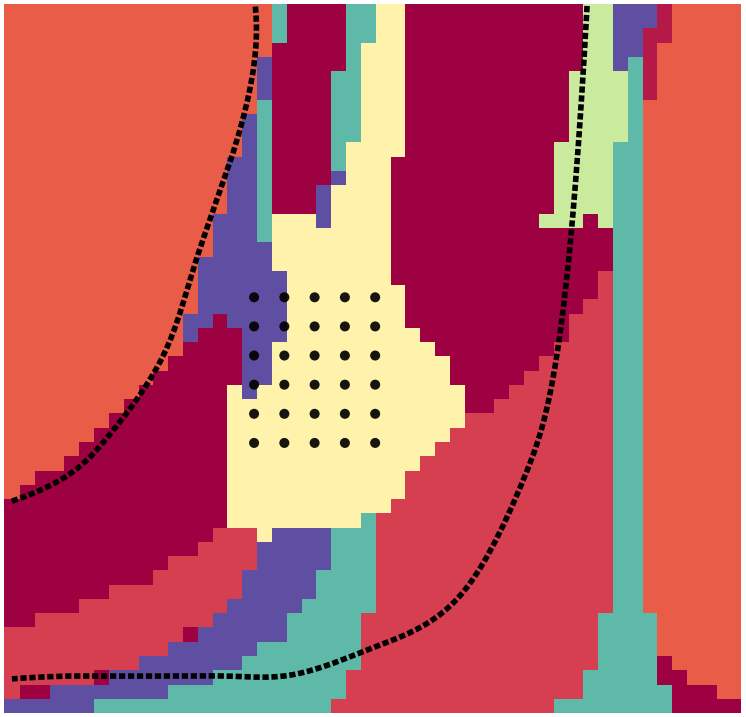
Heisenberg spin model



Rodrigo Vargas, John Sous, Mona Berciu and R. V. Krems, **Phys. Rev. Lett. 121, 255702 (2018)**
Jun Dai and R. V. Krems**, J. Chem. Theory Comp. 16, 1386 (2020)**

Generalized polaron model





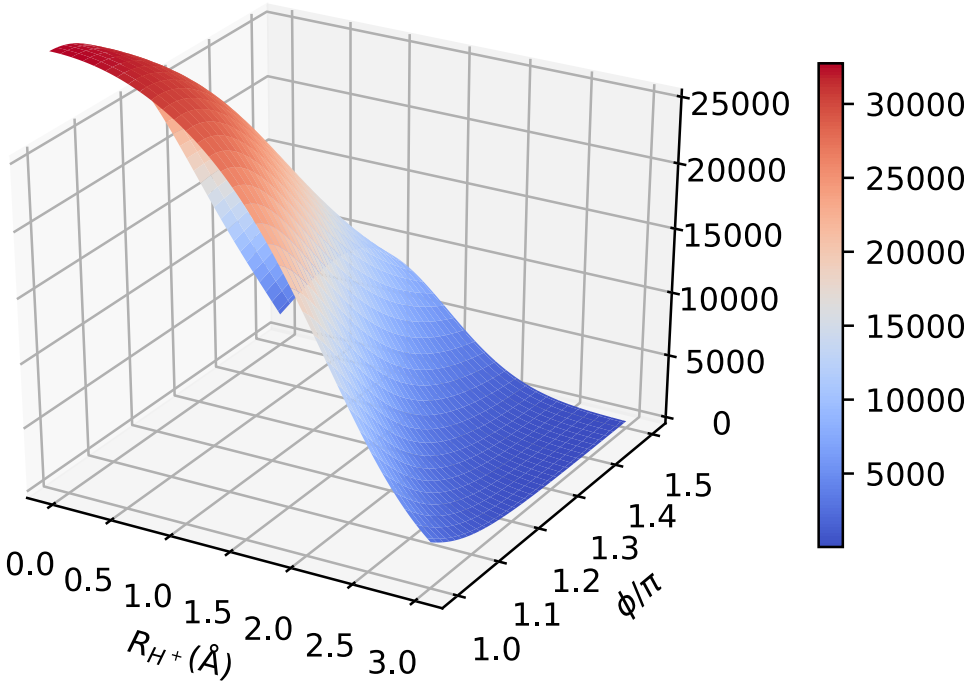
Choosing better kernels not only makes extrapolation possible, but also allows models to extrapolate farther

Choosing better kernels is like replacing spectacles with binoculars when it comes to quantum phase diagrams

Rodrigo Vargas, John Sous, Mona Berciu and R. V. Krems, *Phys. Rev. Lett. 121, 255702 (2018)*

Extrapolation of potential energy surfaces

Six-dimensional surface for H_3O^+



Jun Dai and RK, *J. Chem Theory Comp.* 16, 1386 (2020)

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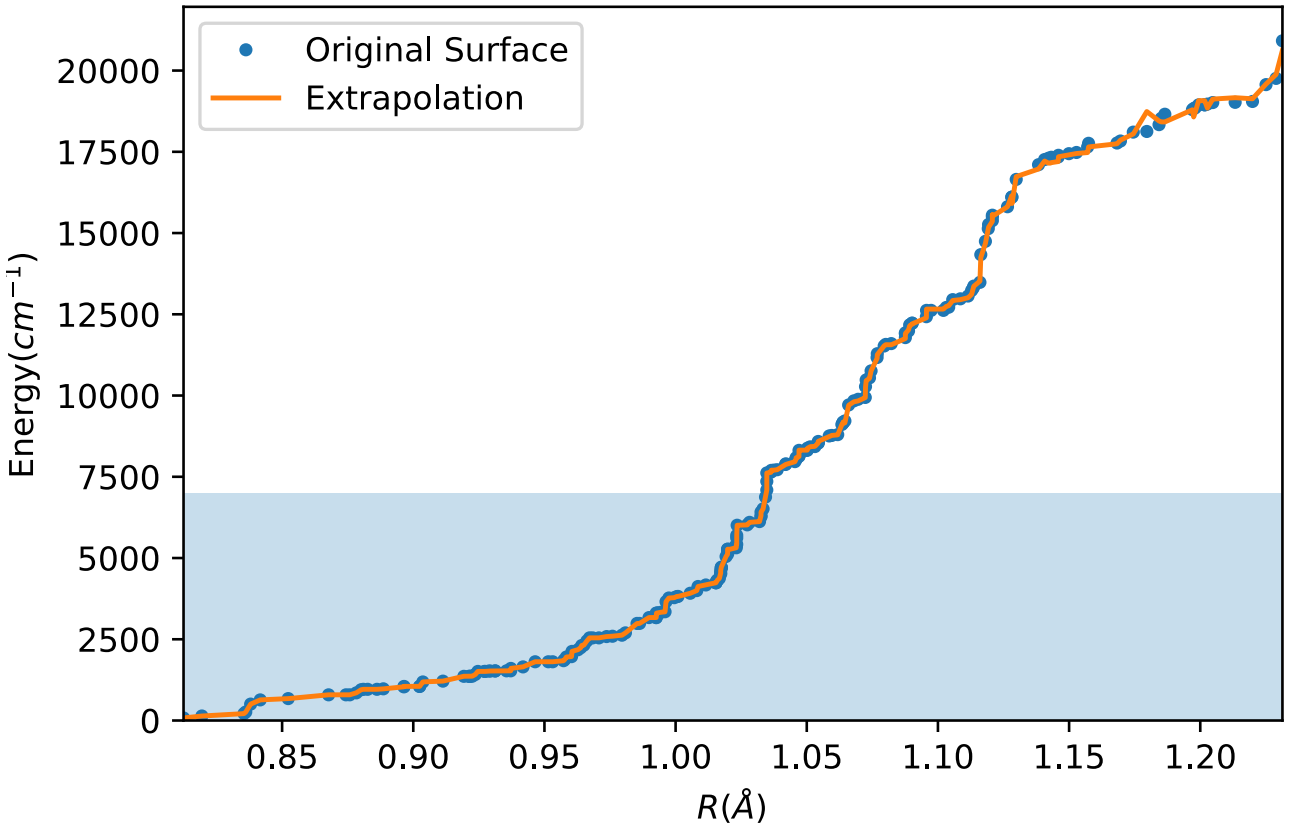
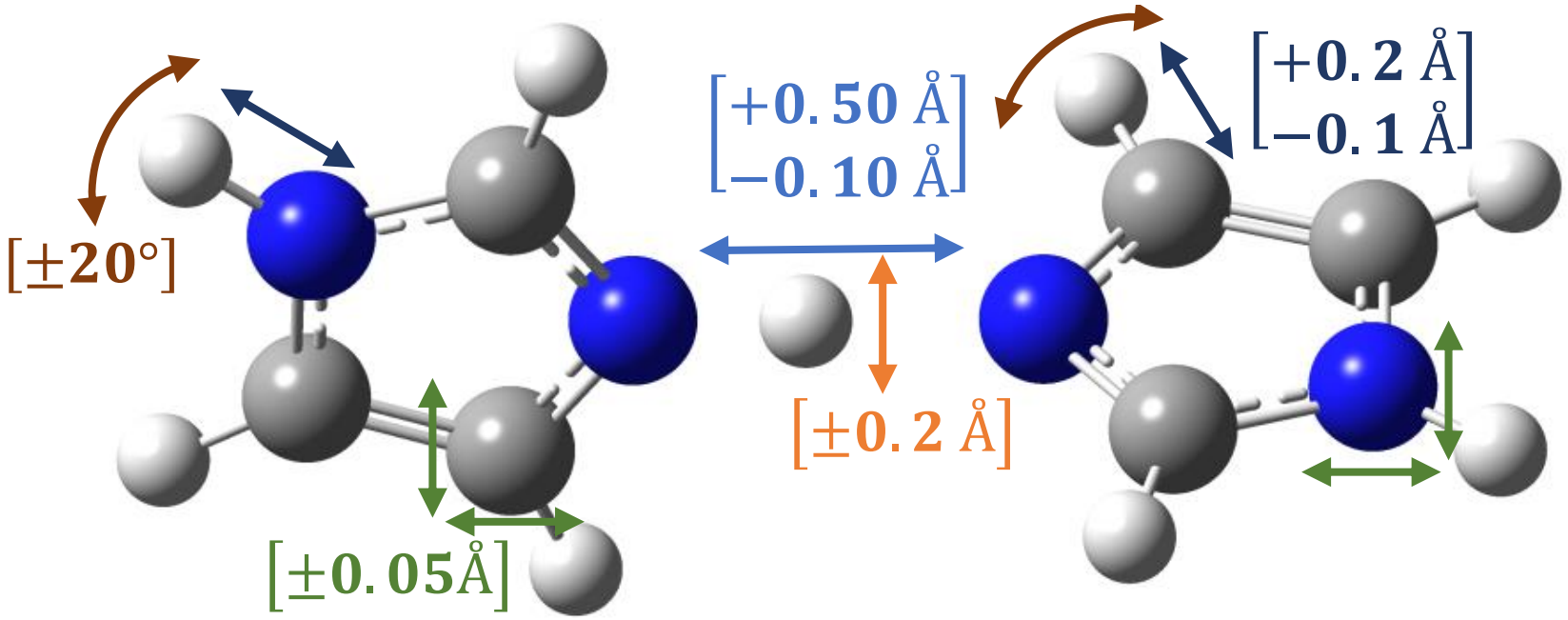


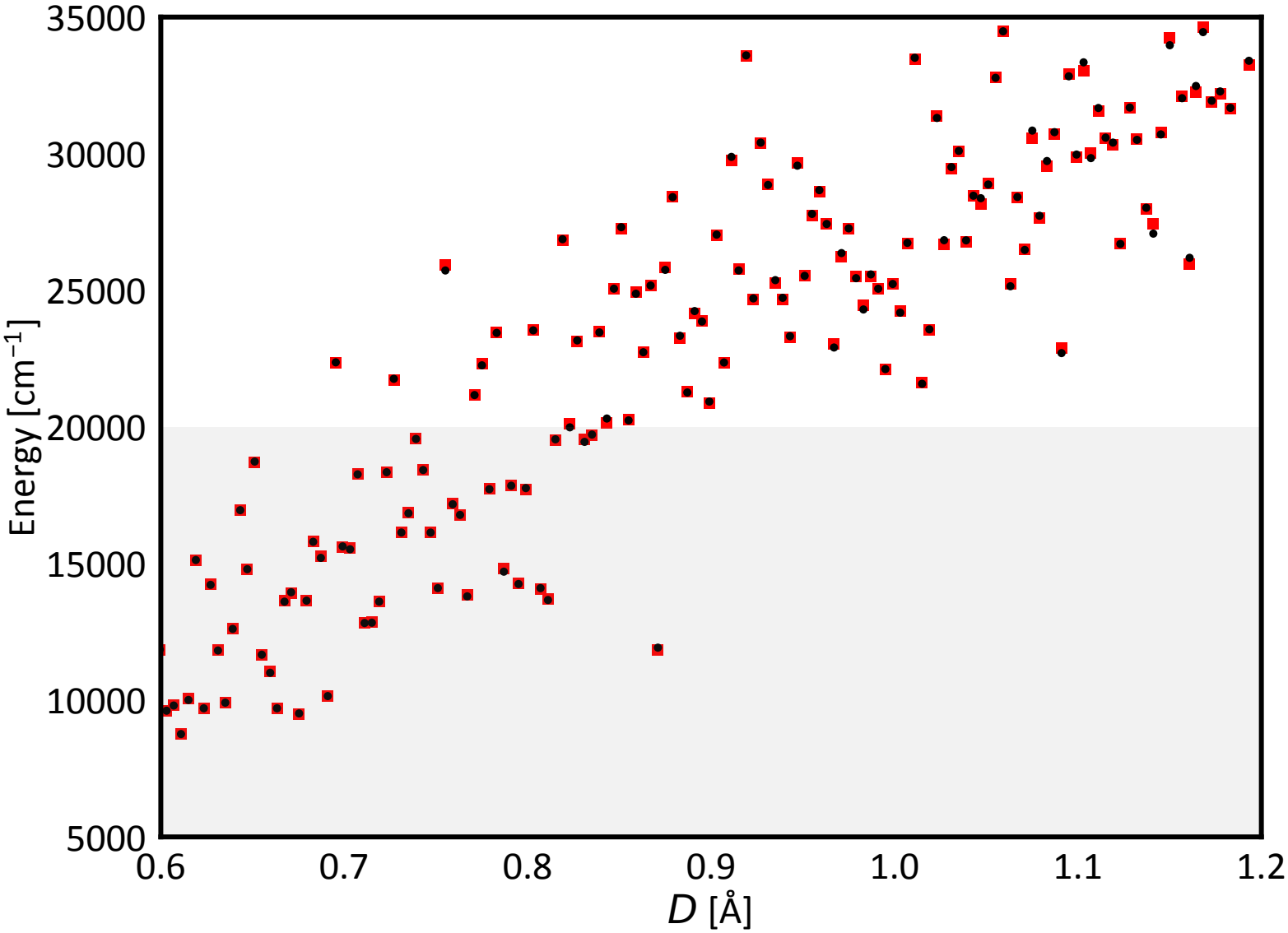
Jun Dai and RK, *J. Chem Theory Comp.* 16, 1386 (2020)

What about higher-dimensional problems?

51 dimensions

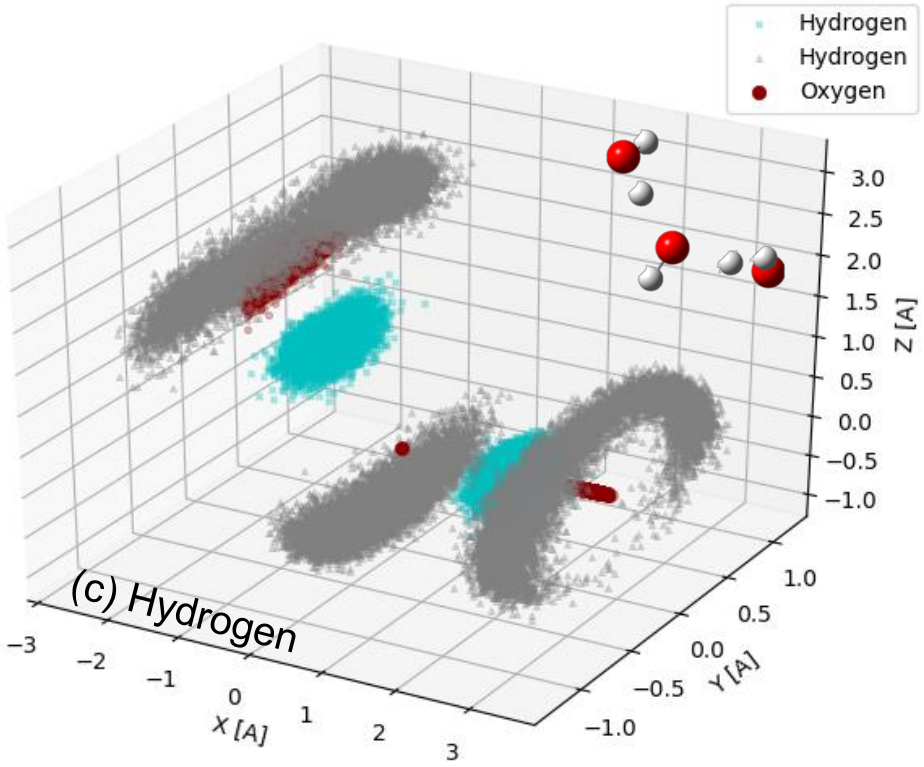


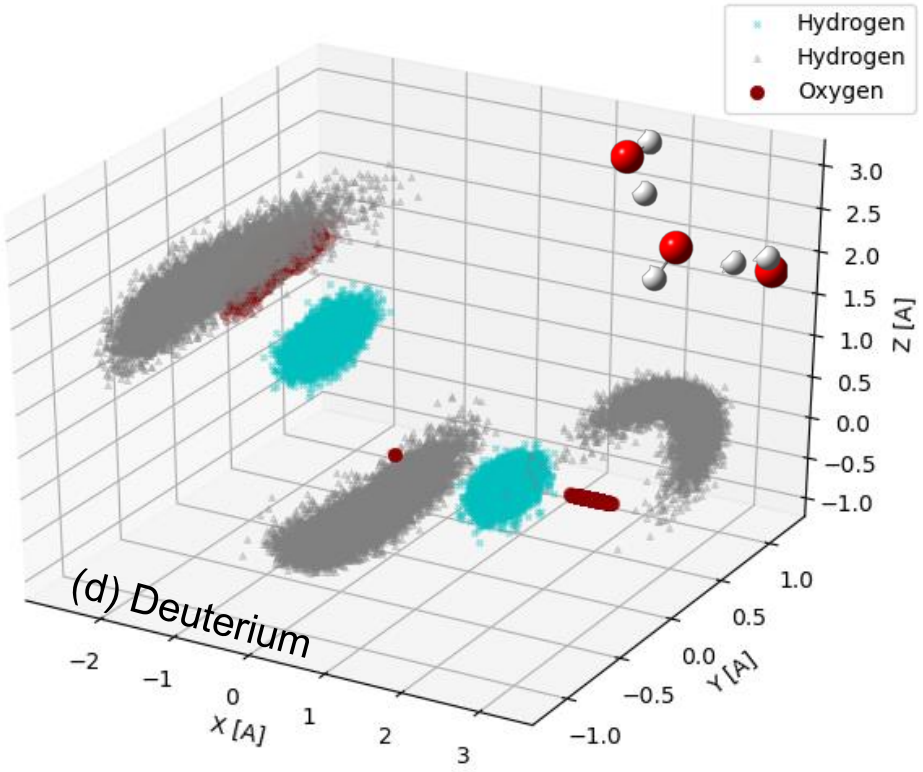
Hiroki Sugisawa, I. Sato and RVK, *J. Chem. Phys.* 153, 114101 (2020)



Hiroki Sugisawa, I. Sato and RVK, *J. Chem. Phys.* 153, 114101 (2020)

This allows us to compute things we couldn't imagine just a few years ago… Work of Hiroki Sugisawa





Wave function of $H_5O_3^-$ at the MP2 quantum chemistry level

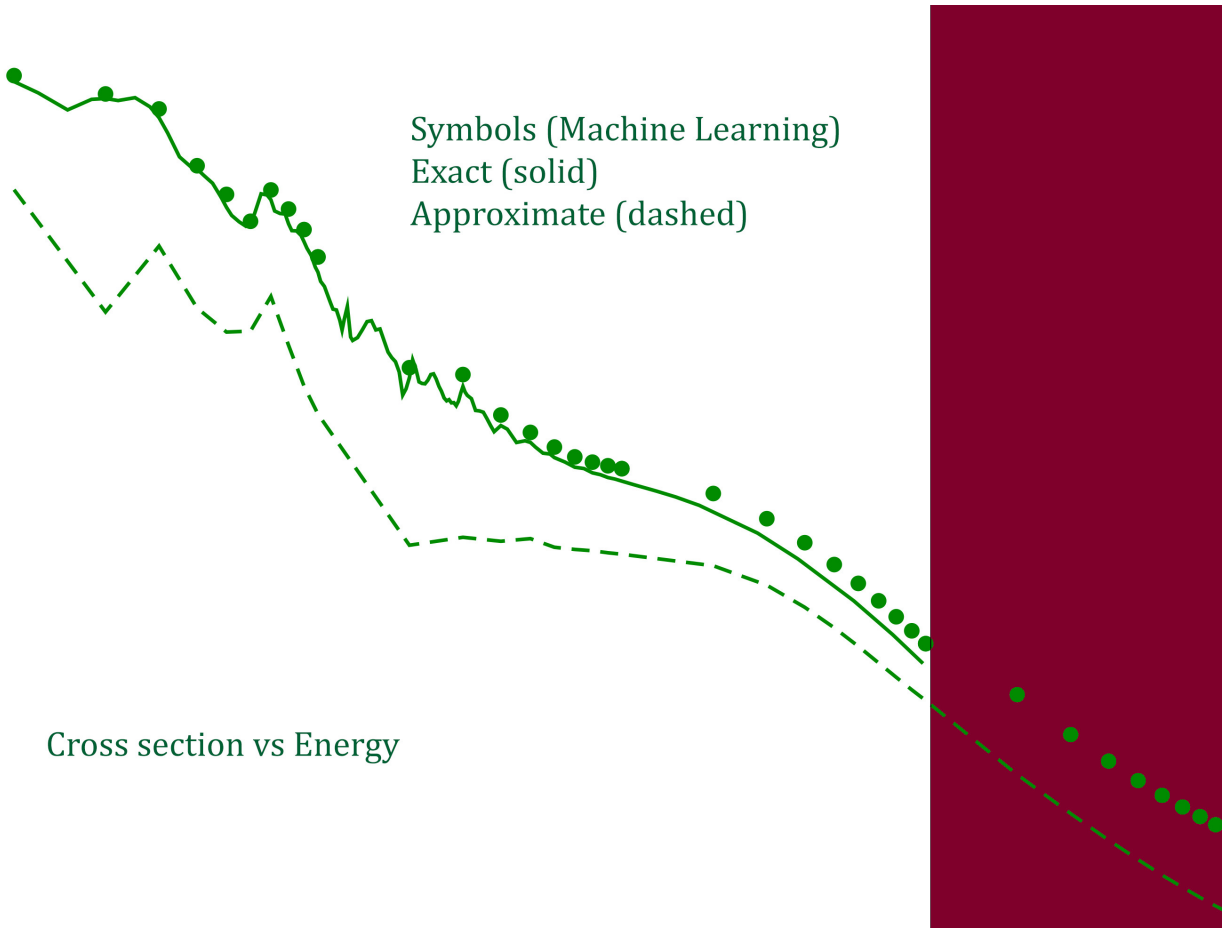
Direct approach: 220 years on a single core of Intel i7-9700K

Our approach: 8 days

Same models can be used for transfer learning

- Quantum dynamics calculations for complex systems are difficult
- Such calculations must rely on approximations
- Can the results of approximate quantum calculations be corrected by machine learning?

A. Jasinski, J. Montaner, R. C. Forrey, B. H. Yang, P. C. Stancil, N. Balakrishnan, R. Vargas-Hernandez, J. Dai and R. V. Krems, **PRR 2, 032051 (2020)**



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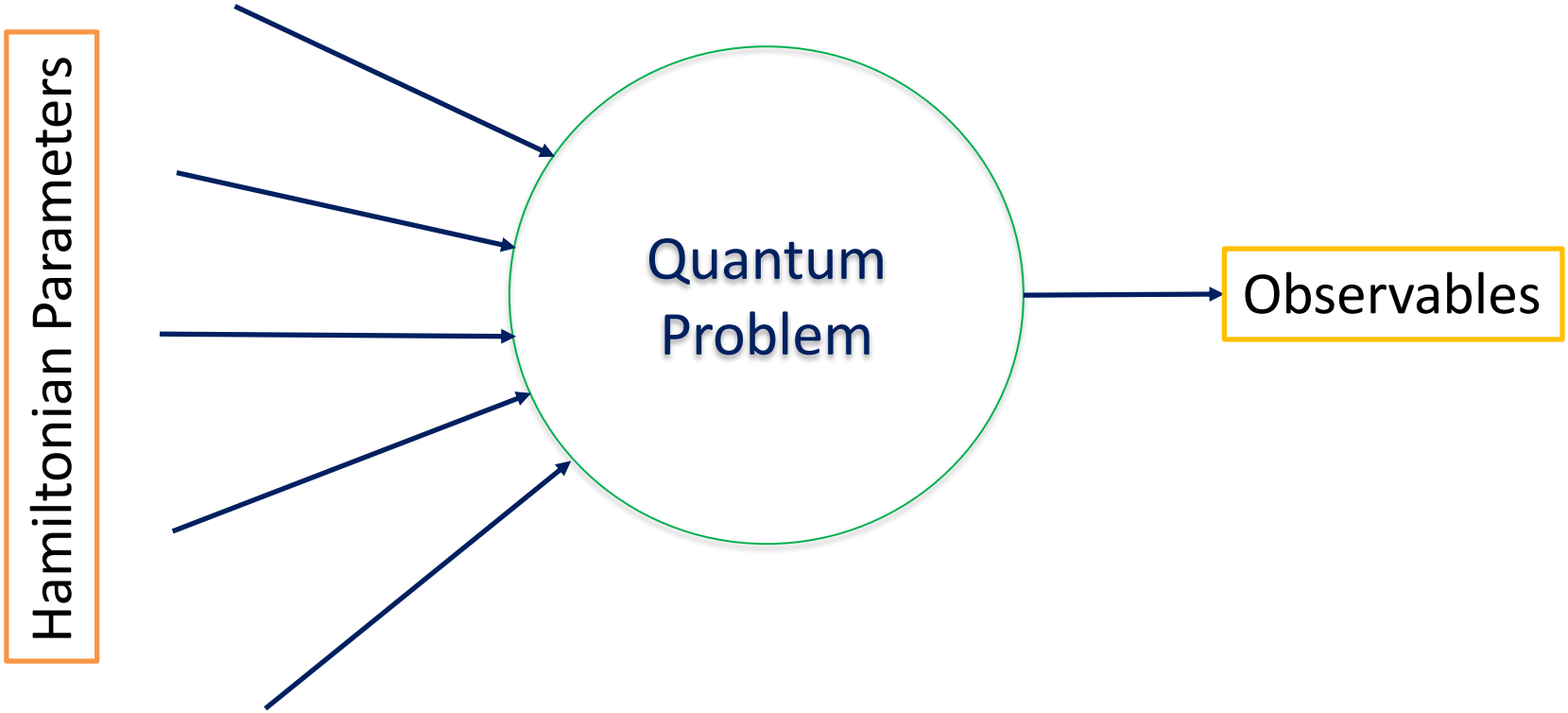
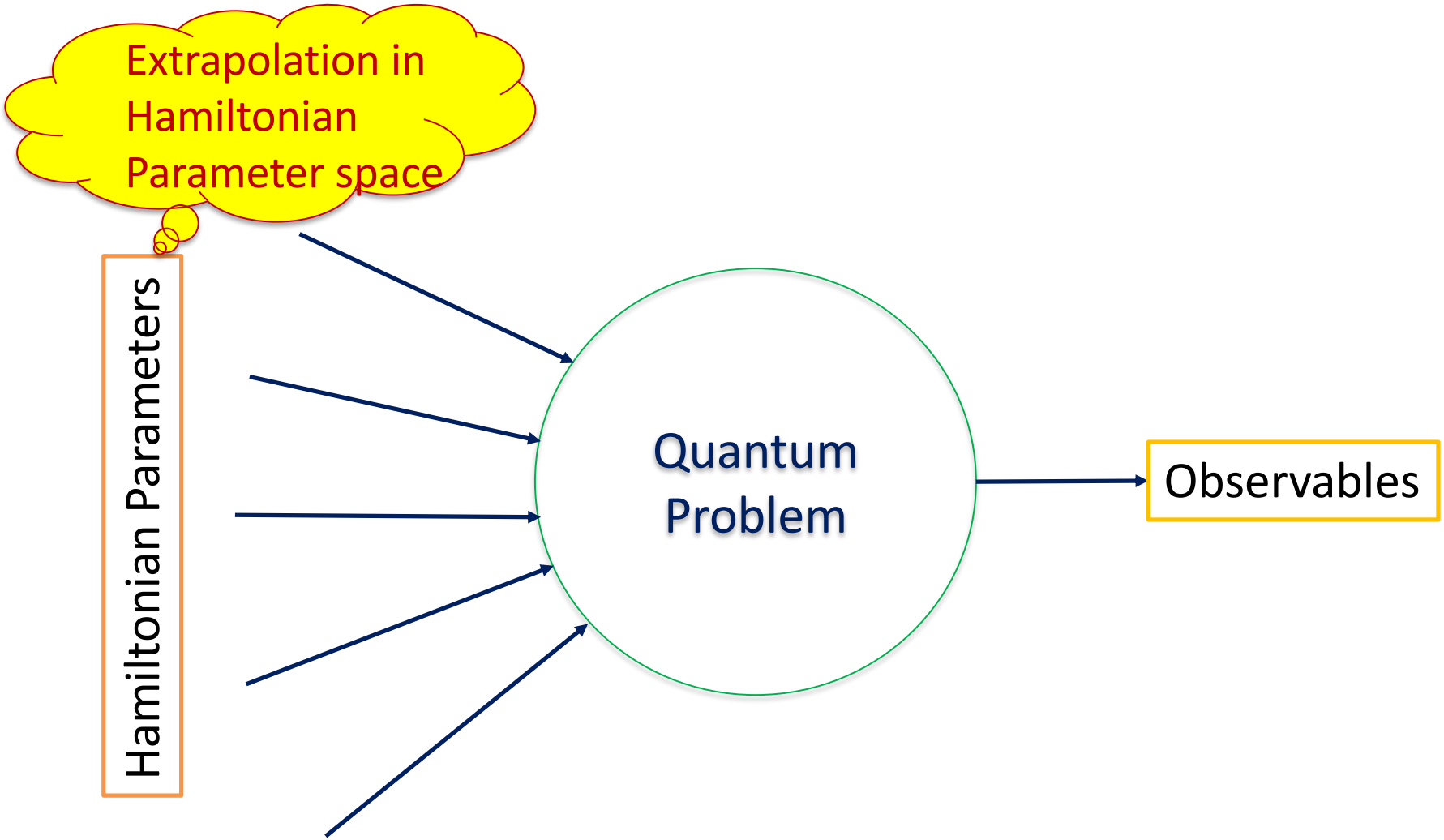
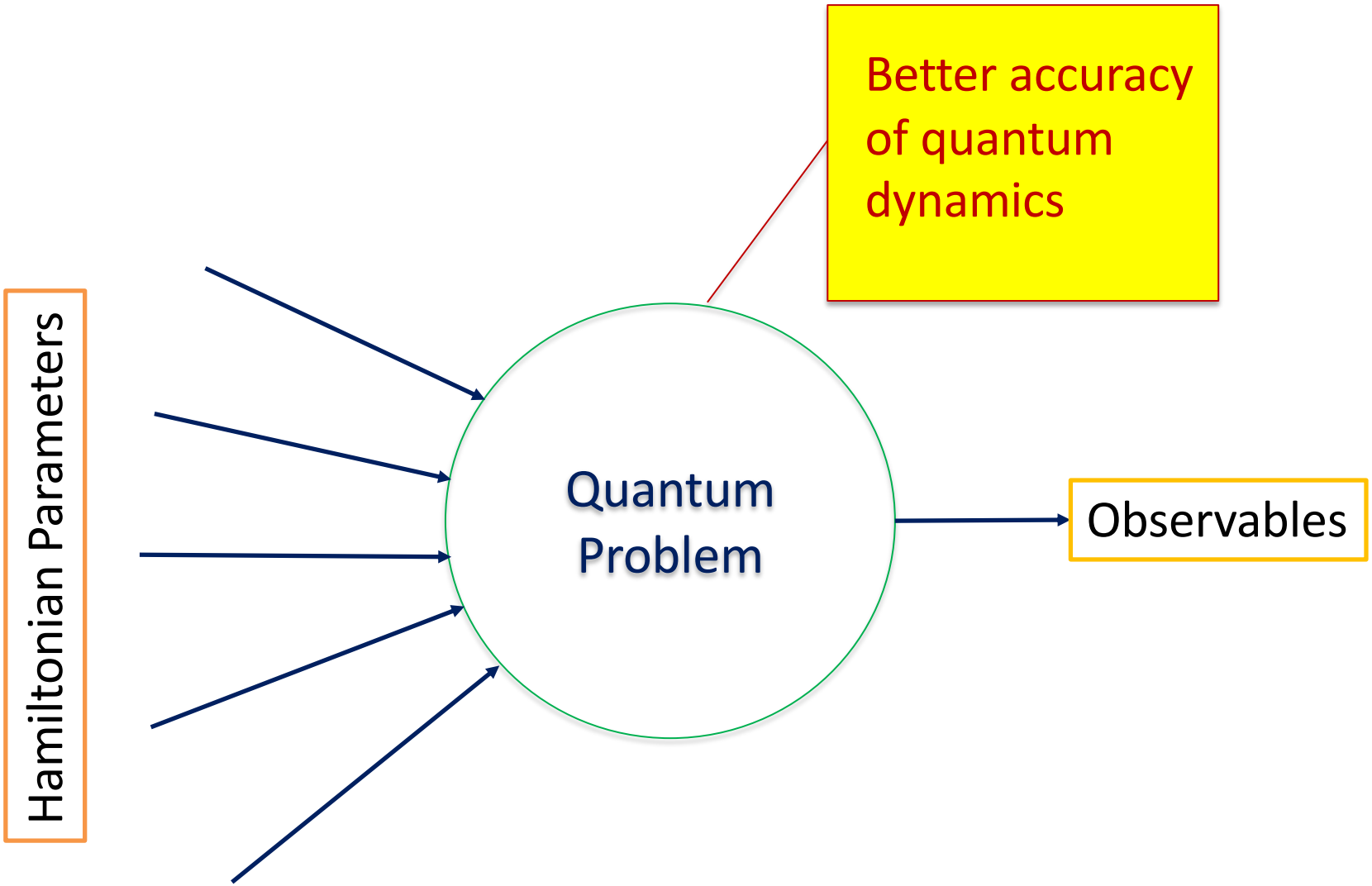
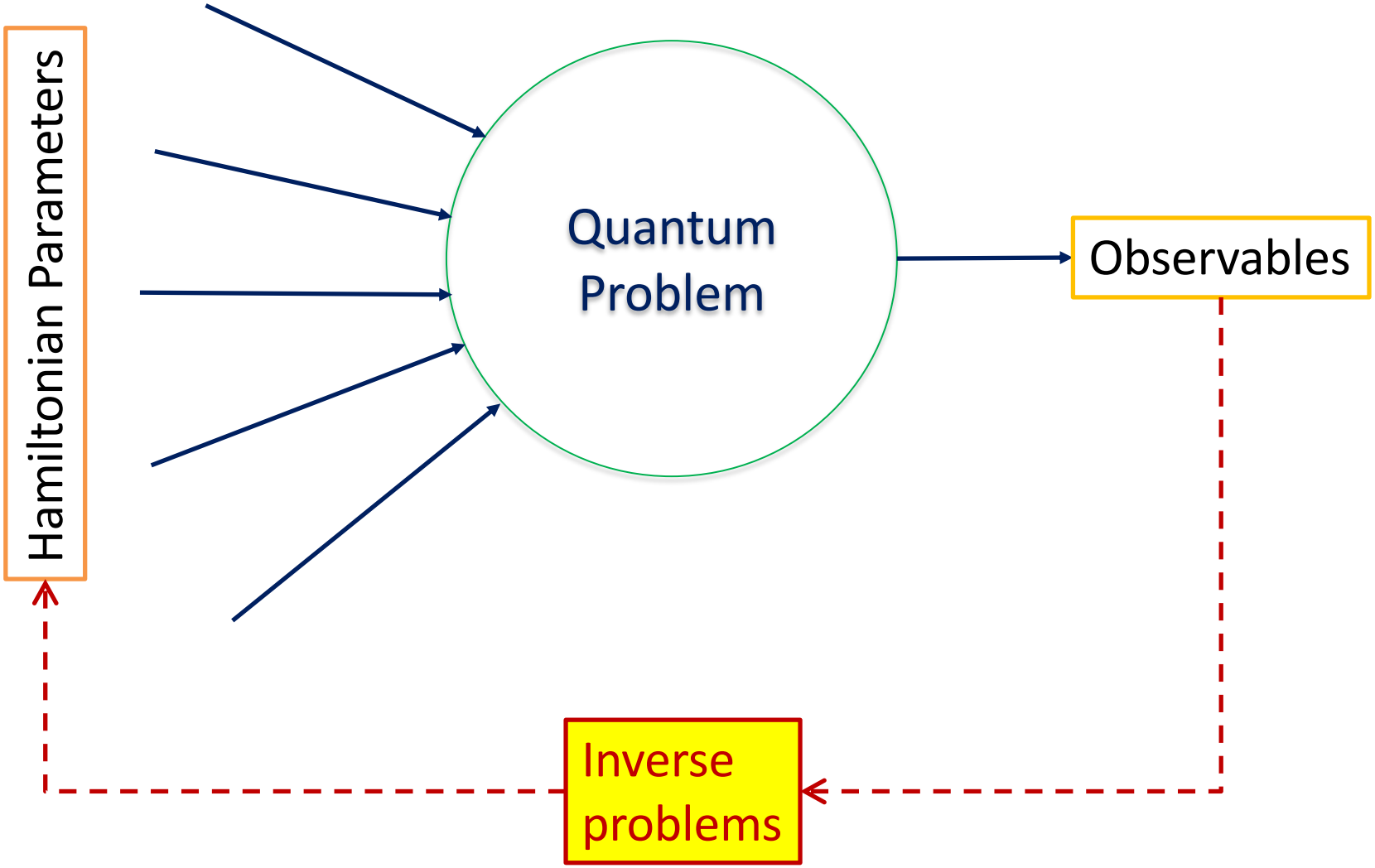
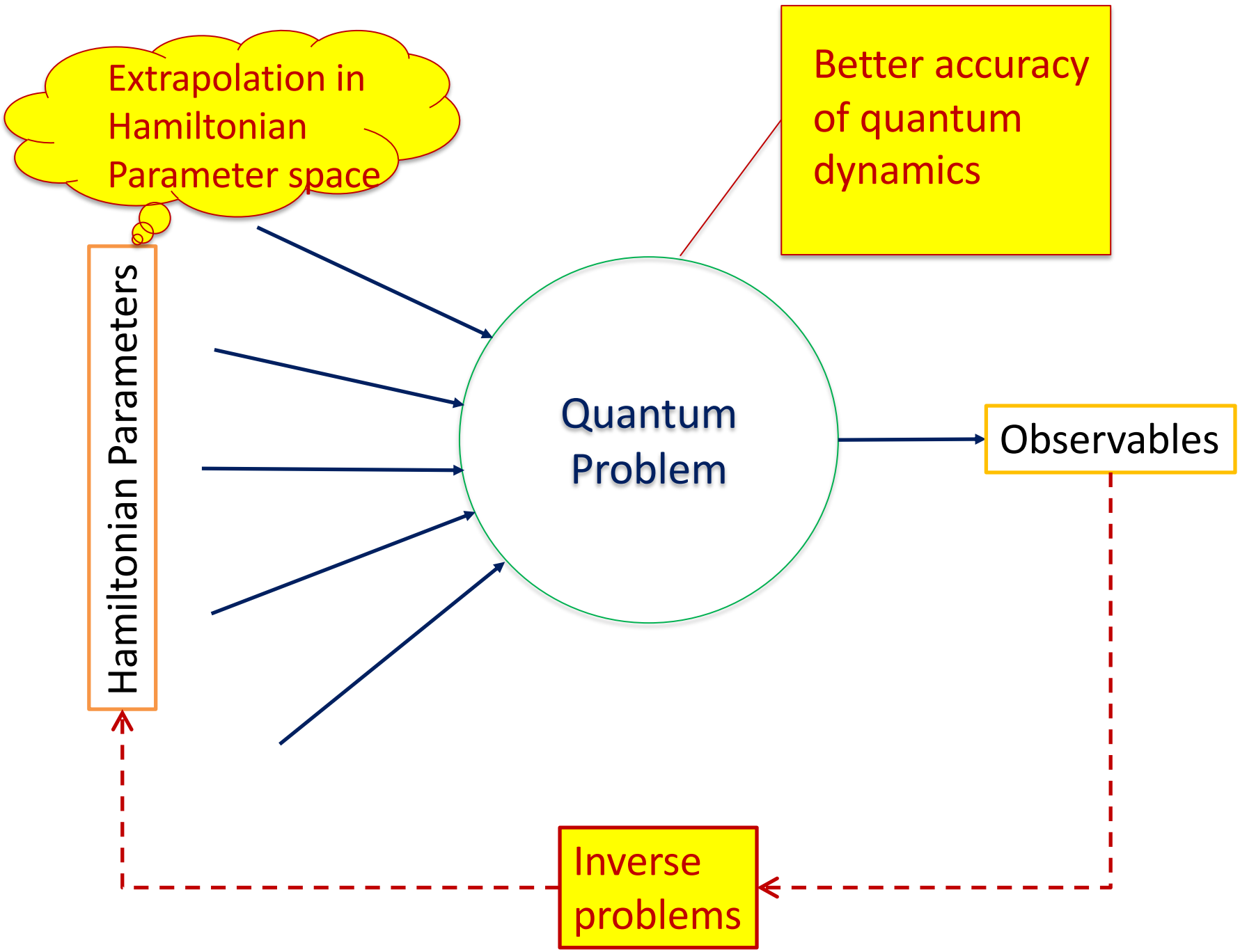

Hamiltonian Parameters
Quantum Problem
Observables


Extrapolation in Hamiltonian Parameter space
Hamiltonian Parameters
Quantum Problem
Observables


Better accuracy of quantum dynamics
Hamiltonian Parameters
Quantum Problem
Observables


Hamiltonian Parameters
Quantum Problem
Observables
Inverse problems


Extrapolation in Hamiltonian Parameter space
Better accuracy of quantum dynamics
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Physical Chemistry Chemical Physics
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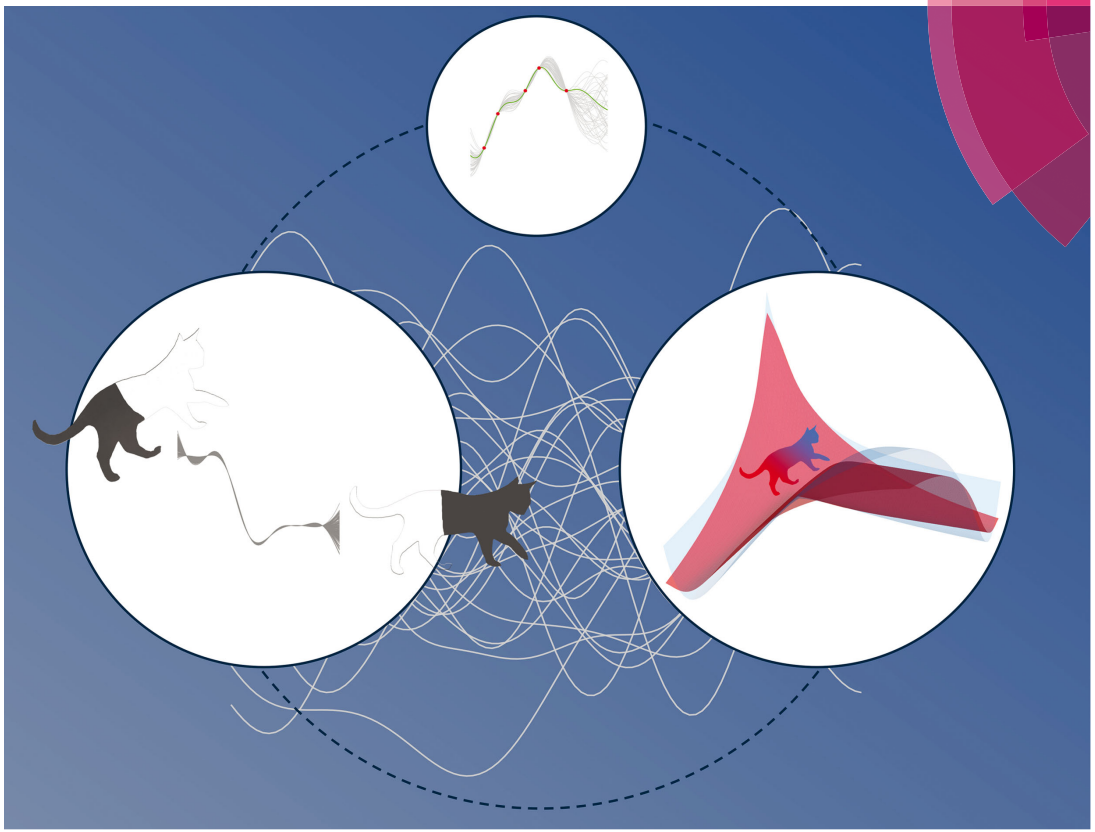

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PERSPECTIVE
R. V. Krems
Bayesian machine learning for quantum molecular dynamics

R. V. Krems,
PCCP 21, 13392 (2019)

Machine learning for quantum dynamics:

Sensitivity of observables to interaction PES, and their errors. [Jie Cui and RK, **PRL 115, 073202 (2015)**]

Inverse quantum problems

[R. Vargas-Hernandez, Y. Guan, D. H. Zhang, and RK, **NJP 21, 022001 (2019)**]

Extrapolation in Hamiltonian parameter spaces

[R. Vargas-Hernandez, J. Sous, M. Berciu and RK, **PRL 121, 255702 (2018)**]

Enhancing the accuracy of approximate dynamical calculations [A. Jasinski et al, **PRR 2, 032051 (2020)**]

More references at https://groups.chem.ubc.ca/krems

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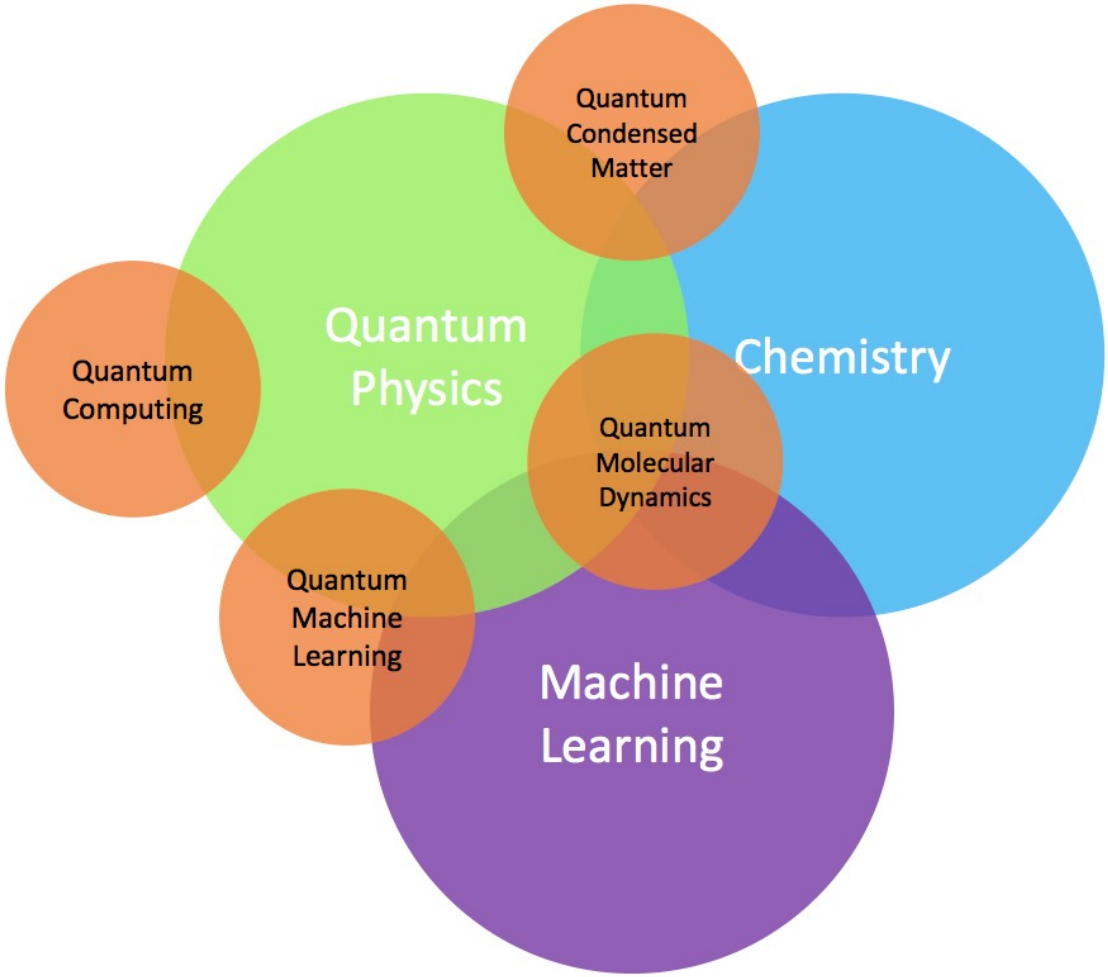

Roman Krems University of British Columbia