@ Nordita

2019/8/27

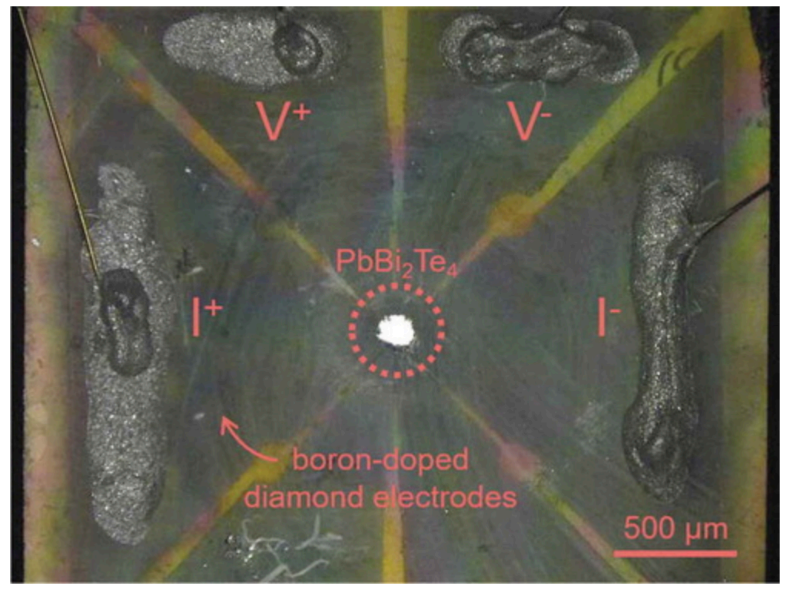
Effective model estimation for magnetic materials by machine learning

NIMS / U. Tokyo Ryo Tamura

Data-driven materials science in Japan

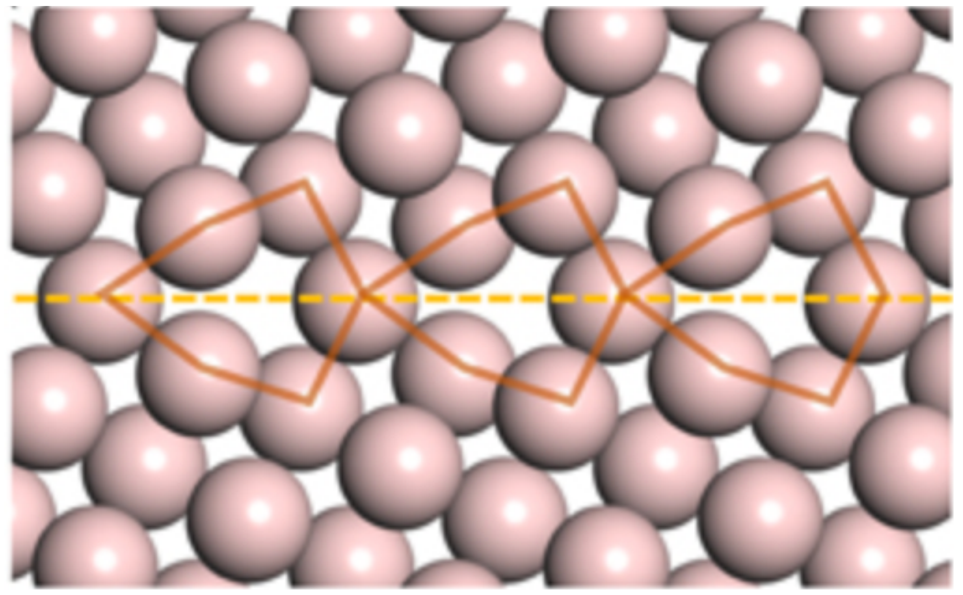
Fast screening, property prediction, improvement of analysis by data-driven techniques

Superconductor



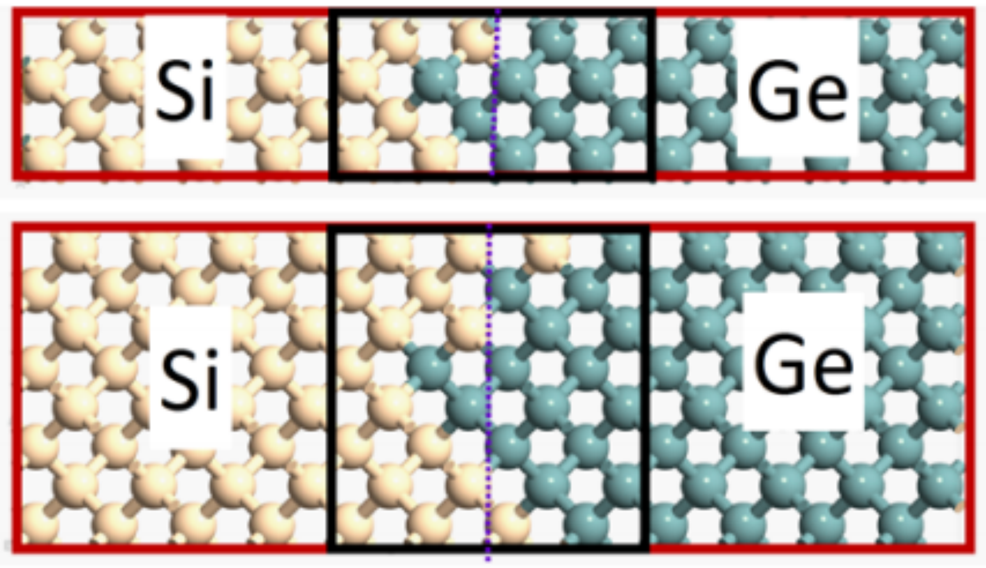
STAM 19, 909 (2018)

Interface



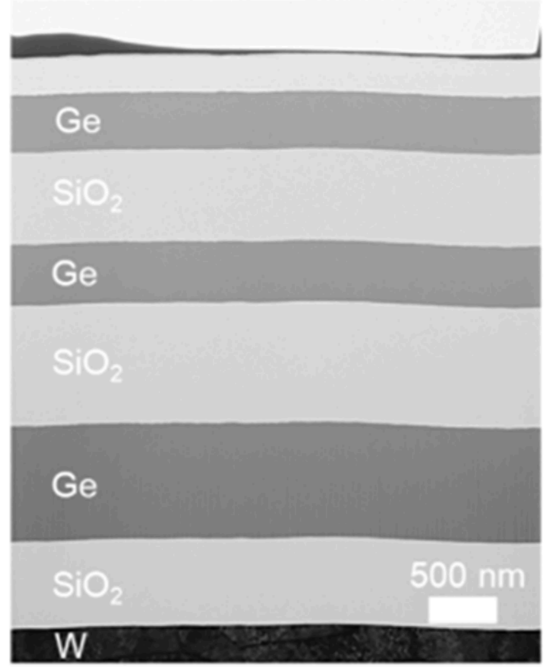
JJAP, 55, 045502 (2016)

Phonon conductivity



PRX 7, 021024 (2017)

Radiator

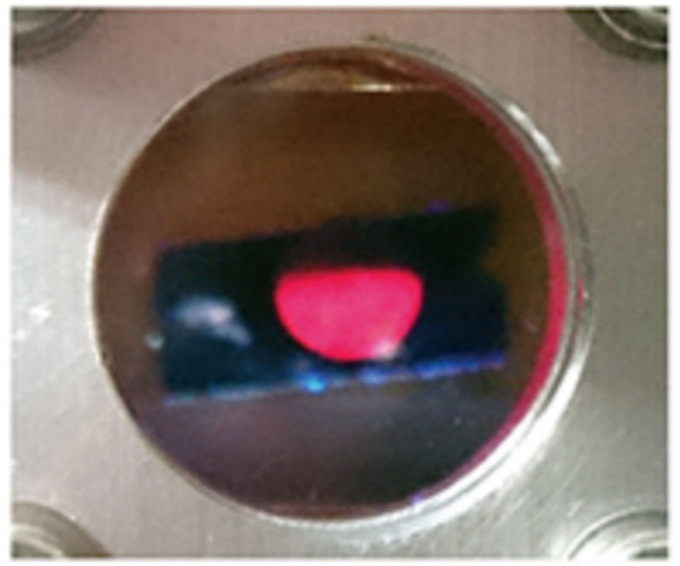


Cent. Sci. 5, 319 (2019)

Smell sensor

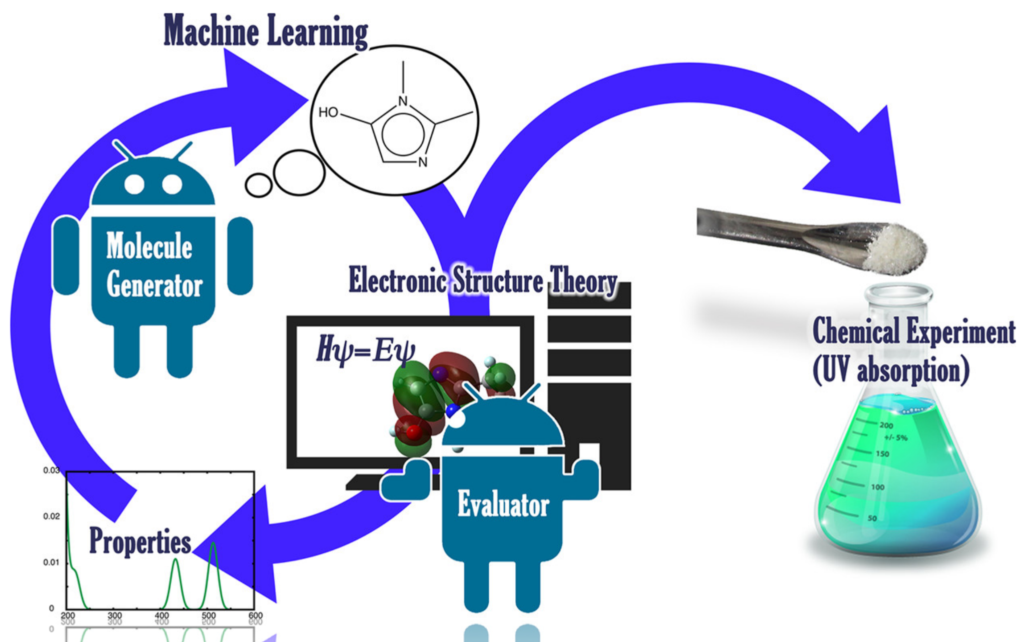
Sci. Rep. 7, 3661 (2017)

Semiconductor



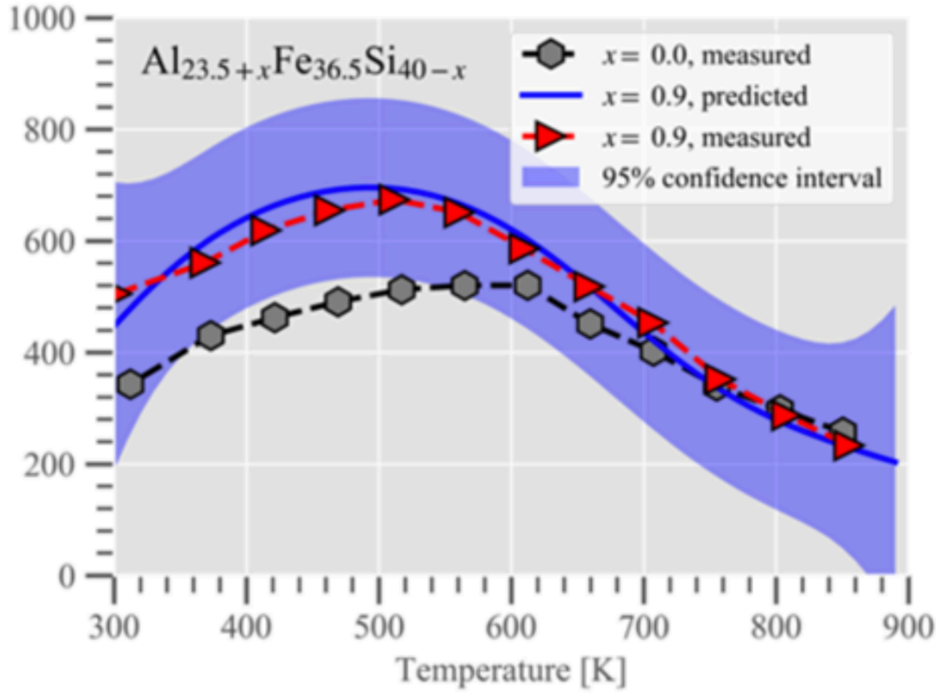
Nat. Com. 7, 11962 (2016)

Molecule



Cent. Sci. 4, 1126 (2018)

Thermoelectric

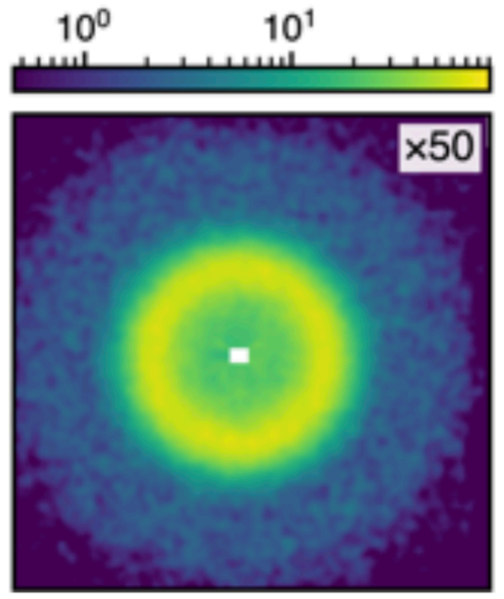


Appl. Mater. Inter. 11, 11545 (2019)

Li-ion conductivity

BCSJ 92, 1100 (2019)

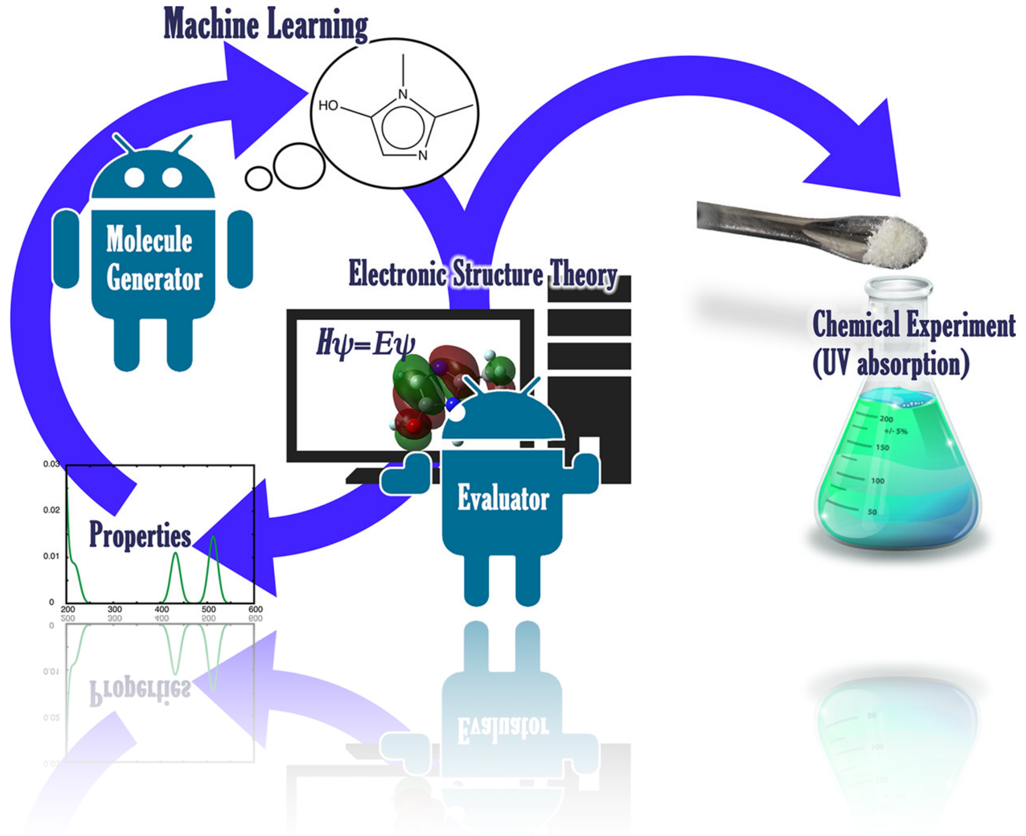
SAXS



Sci. Rep. 9, 1526 (2019)

Current my works

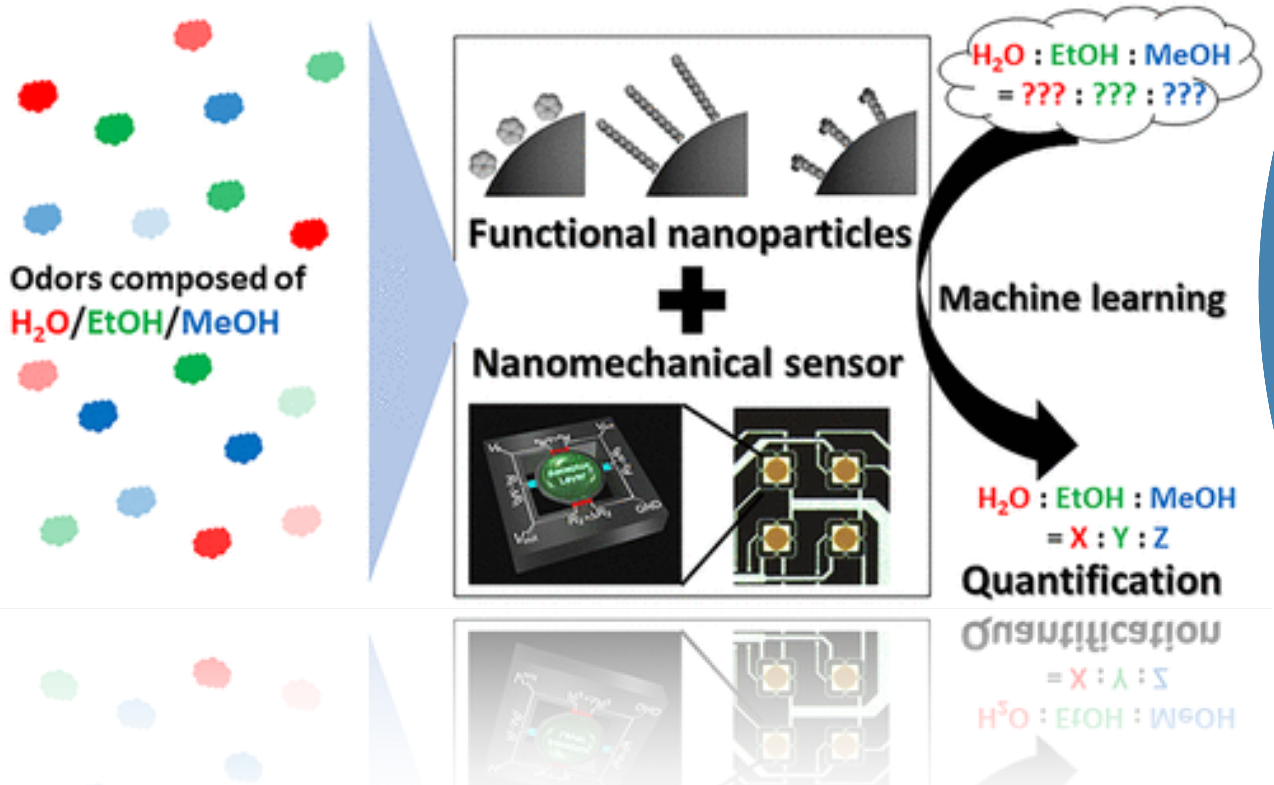
Organic



ACS Cent. Sci. 4, 1126 (2018)

Smells Sensor

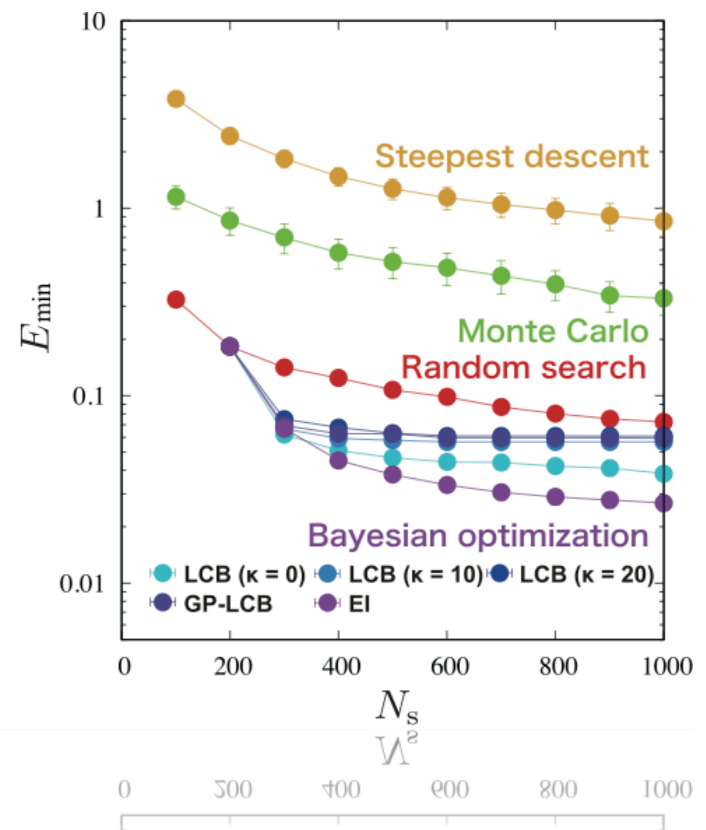
Sci. Rep. 7, 3661 (2017)



ACS Sensors 3, 1592 (2018)

Today's main topic

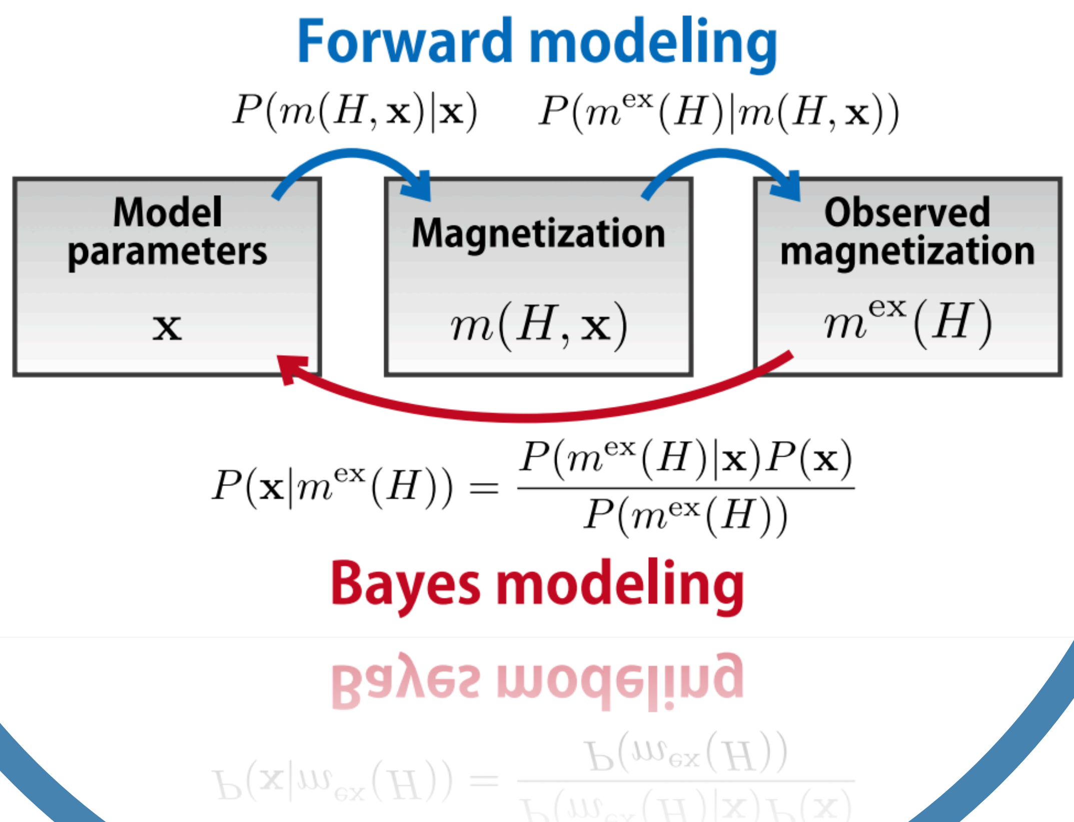
Bayesian Opt.



PLoS ONE 13, e0193785 (2018)

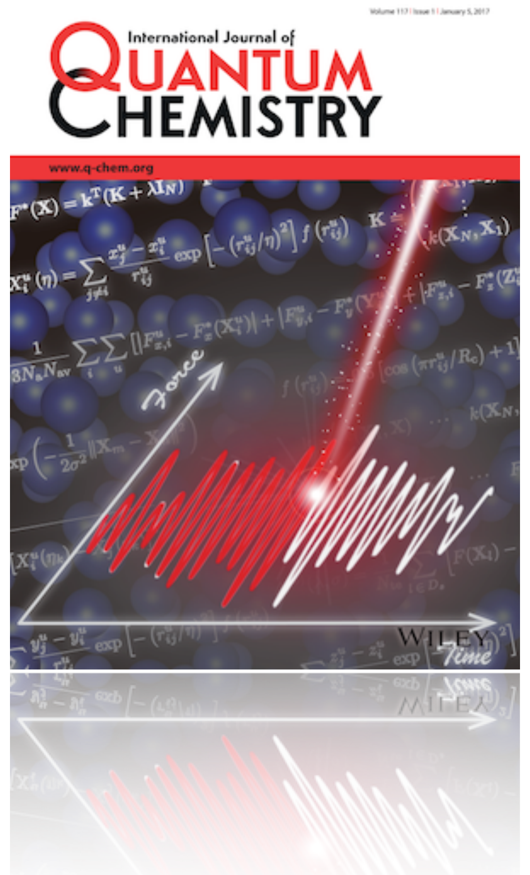
Today's main topic

Magnet



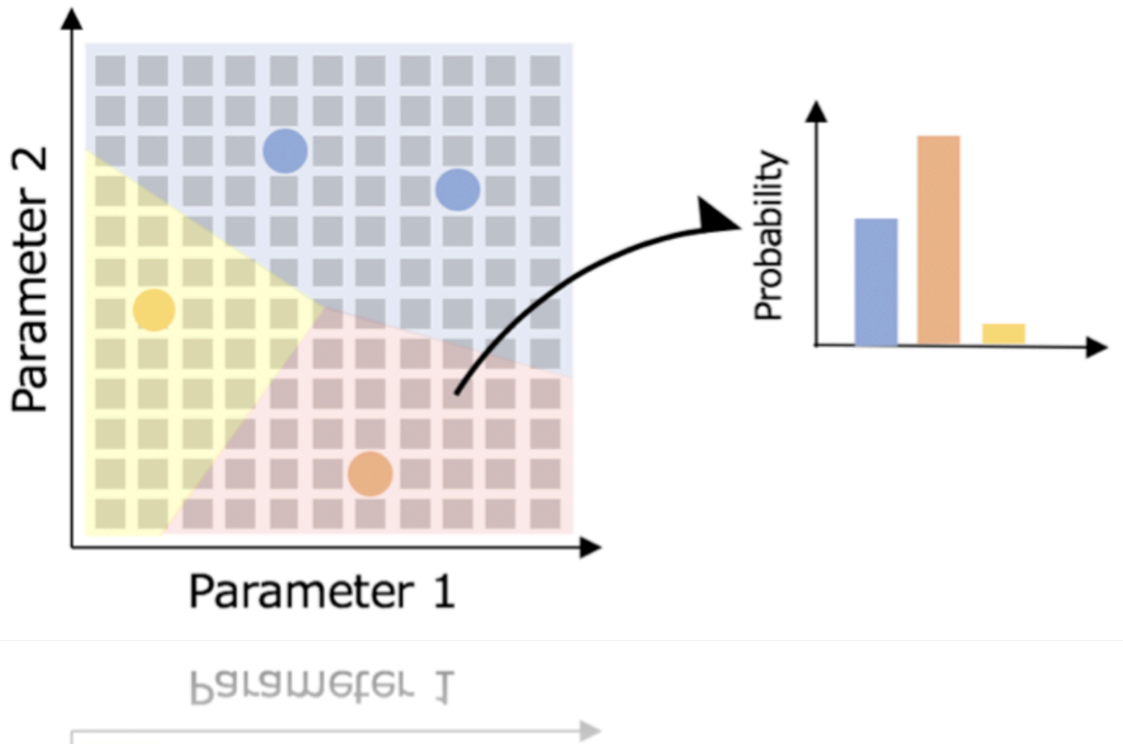
Phys. Rev. B 95, 064407 (2017)

Atomic Force



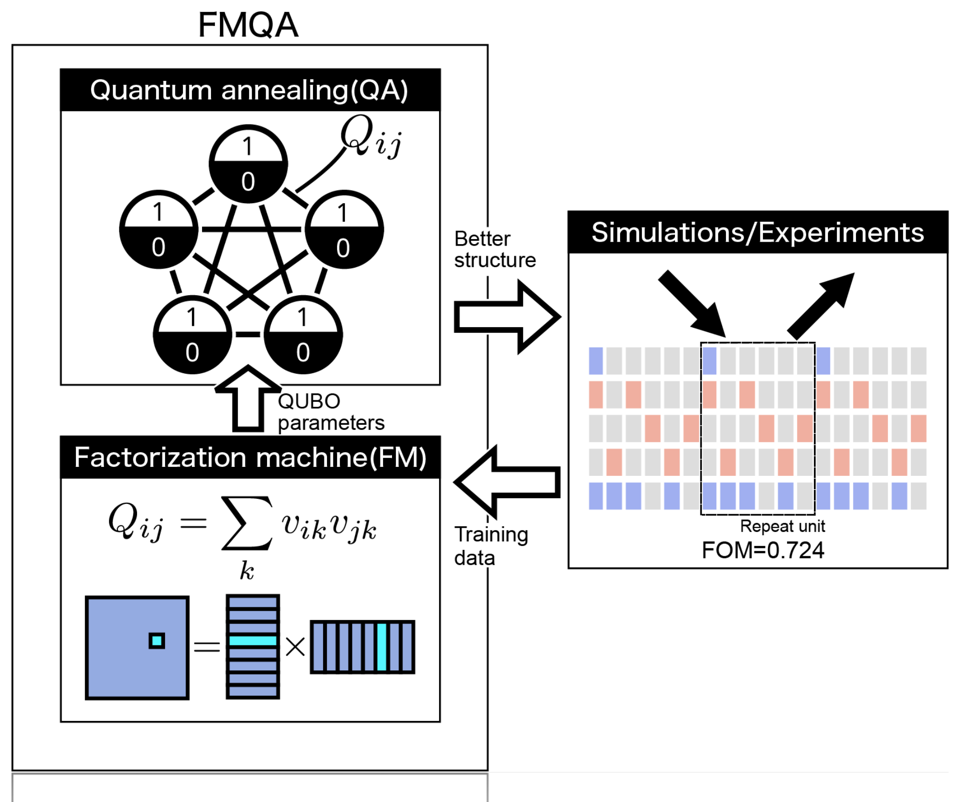
IJQC. 117, 33 (2017)
JPSJ. 88, 044601 (2019)

Phase Diagram



Phys. Rev. Mat. 3, 033802 (2019)

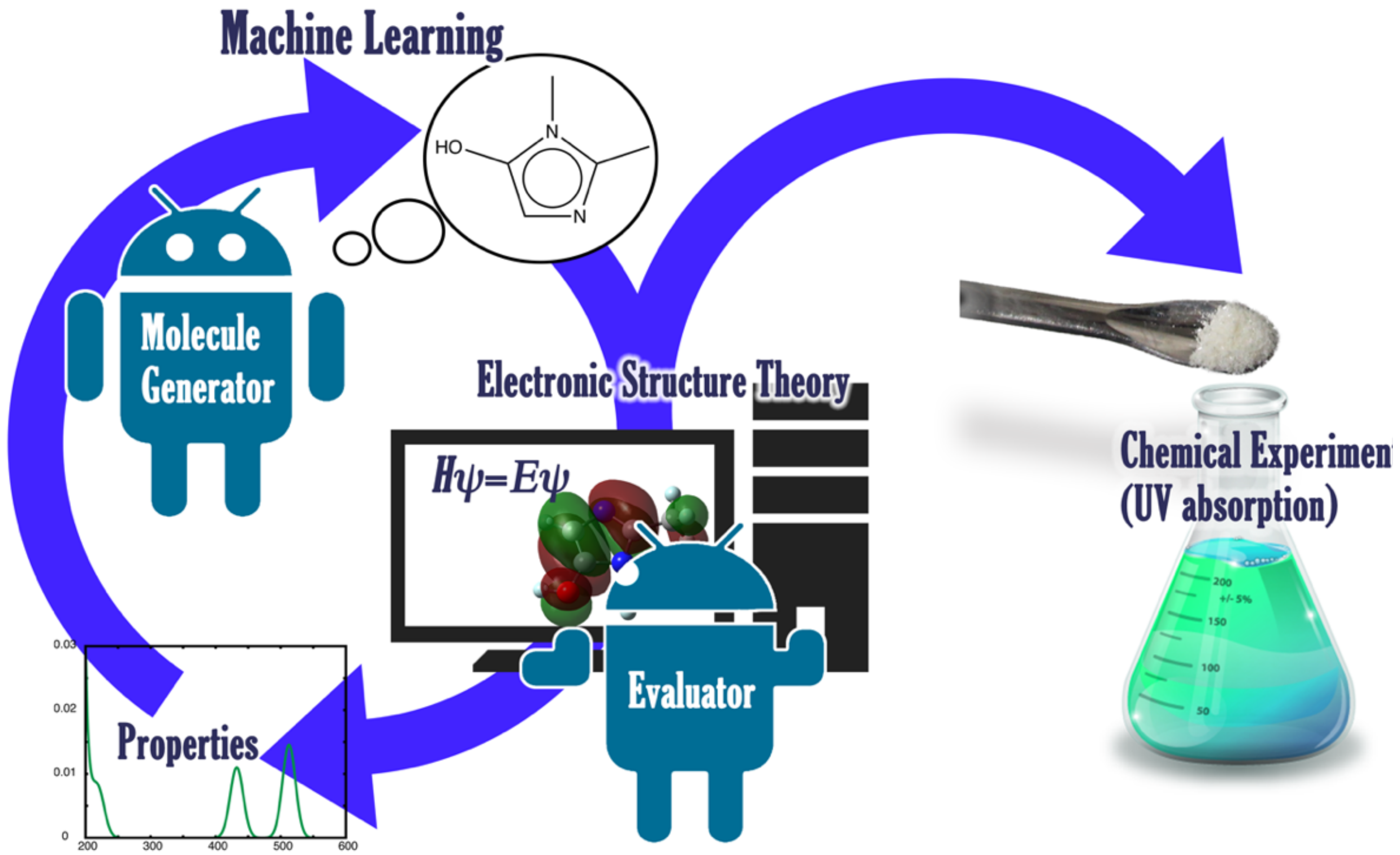
Metamaterial



arXiv: 1902.06573 (2019)

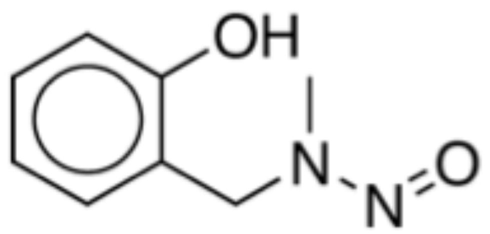
Design of molecule by AI

By combination of ChemTS and Gaussian, we can design synthesizable, novel functional molecules.



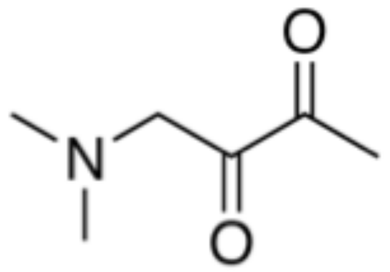
400nm

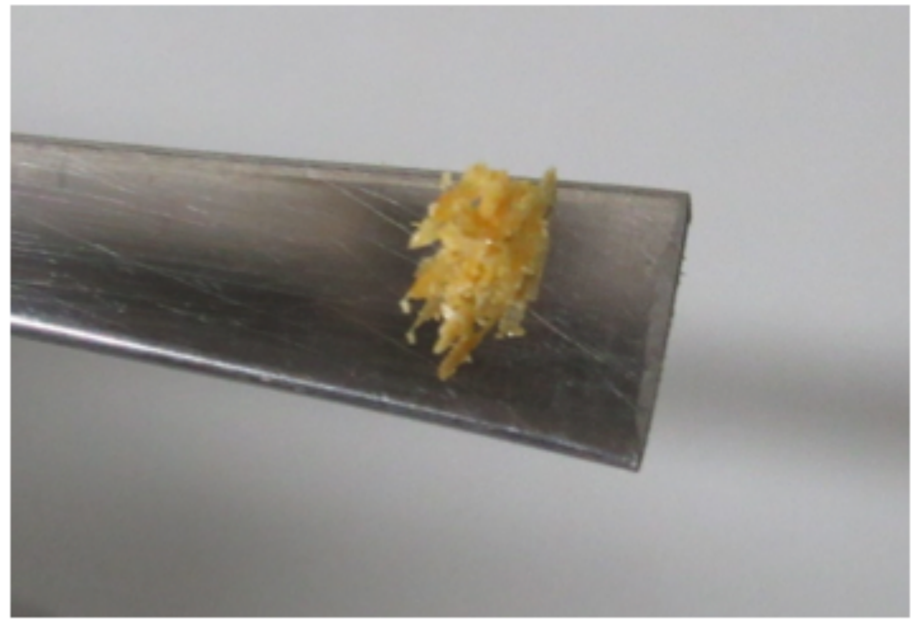
O=NN(Cc1ccccc1O)C



500nm

CC(=O)C(=O)CN(C)C





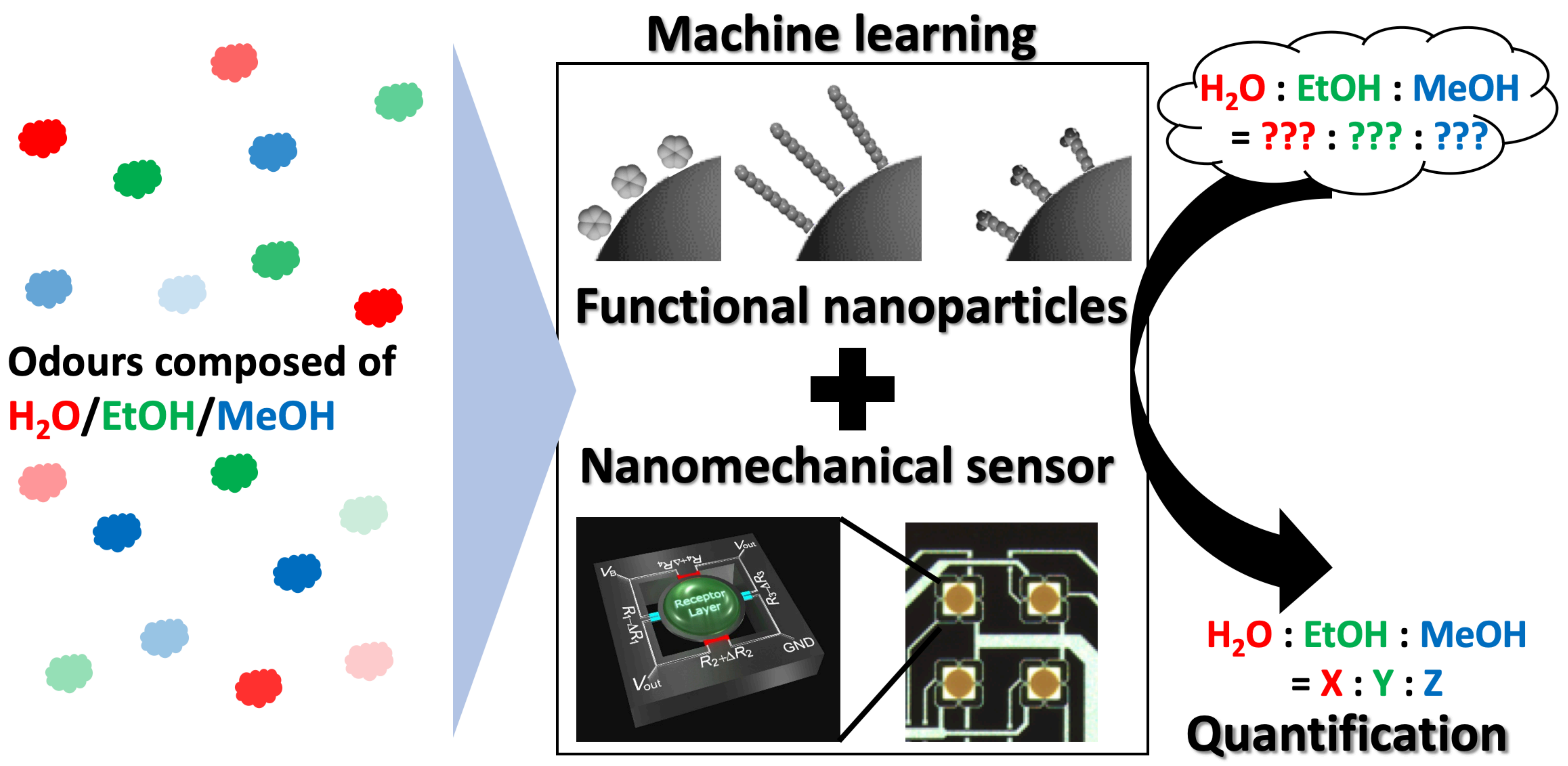
Target wavelength	200 nm	300 nm	400 nm	500 nm	600 nm
Simulator-Qualified	34	26	13	12	1
Synthesized	2	2	1	1	0
Functional	1	2	1	1	0

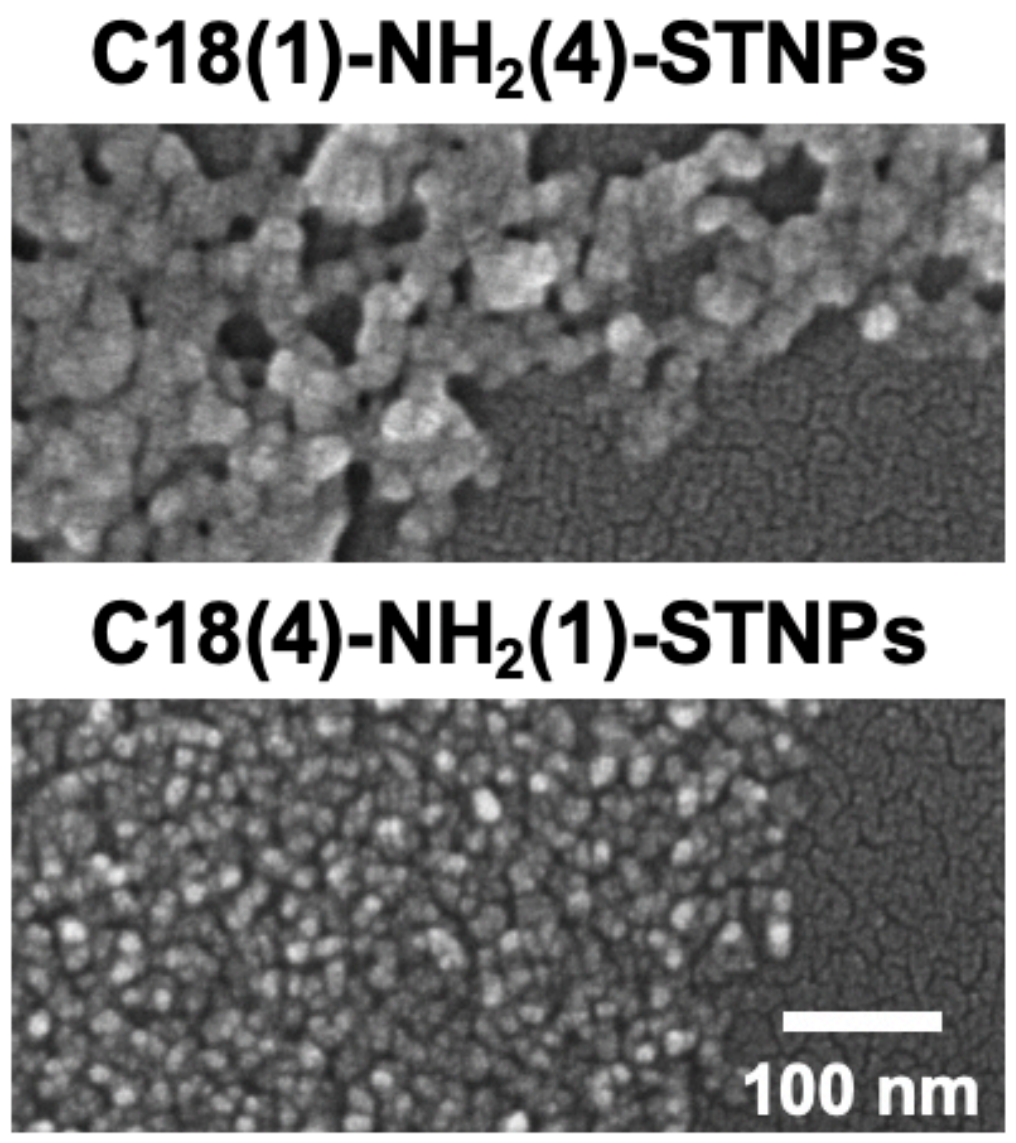
ACS Central Science 4, 1126 (2018).

Data driven nano mechanical sensing

By combination of MSS, ML and systematic material design, we can perform quantitative odor analysis.

ML results ⟶ Development of useful materials

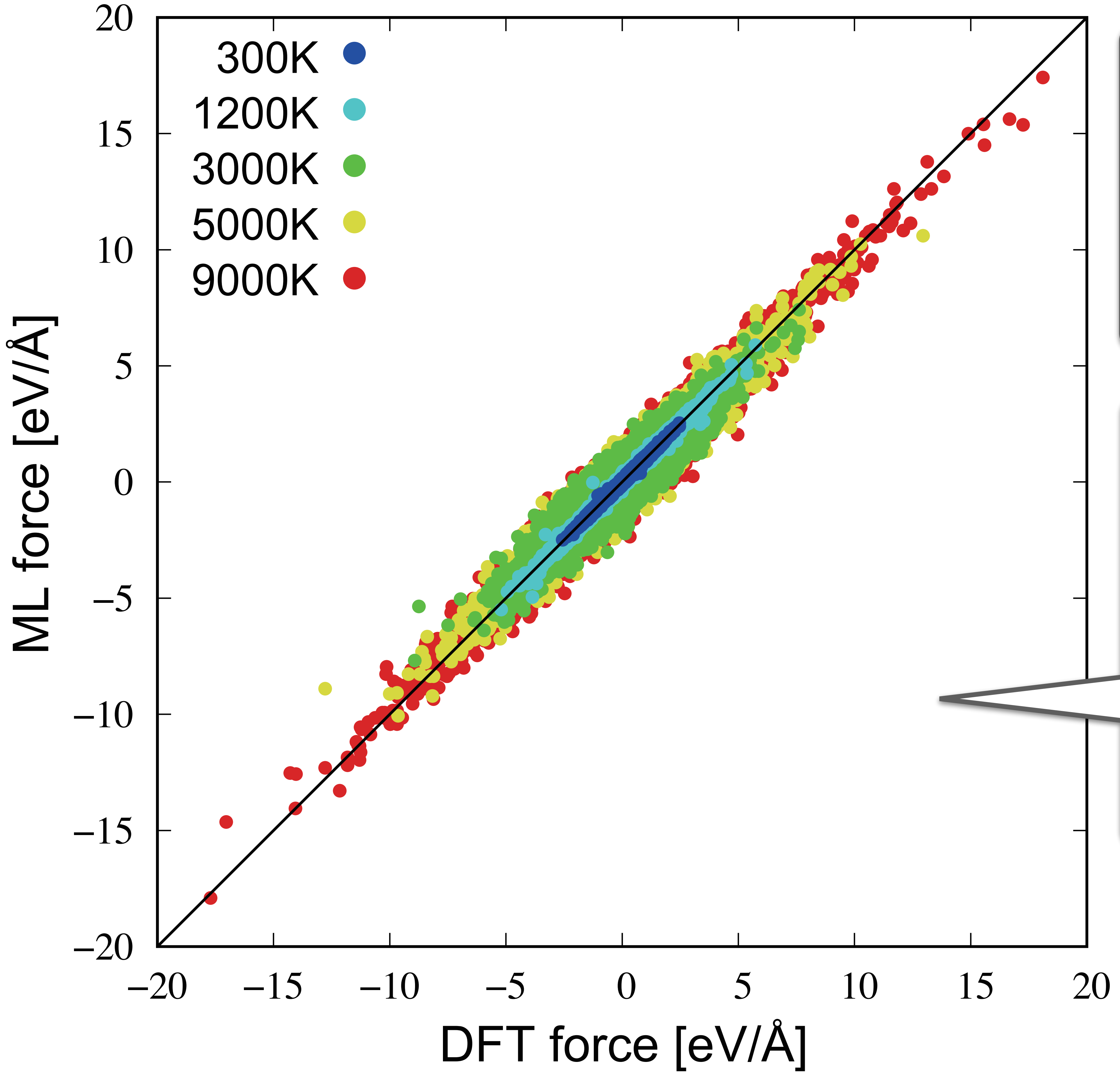




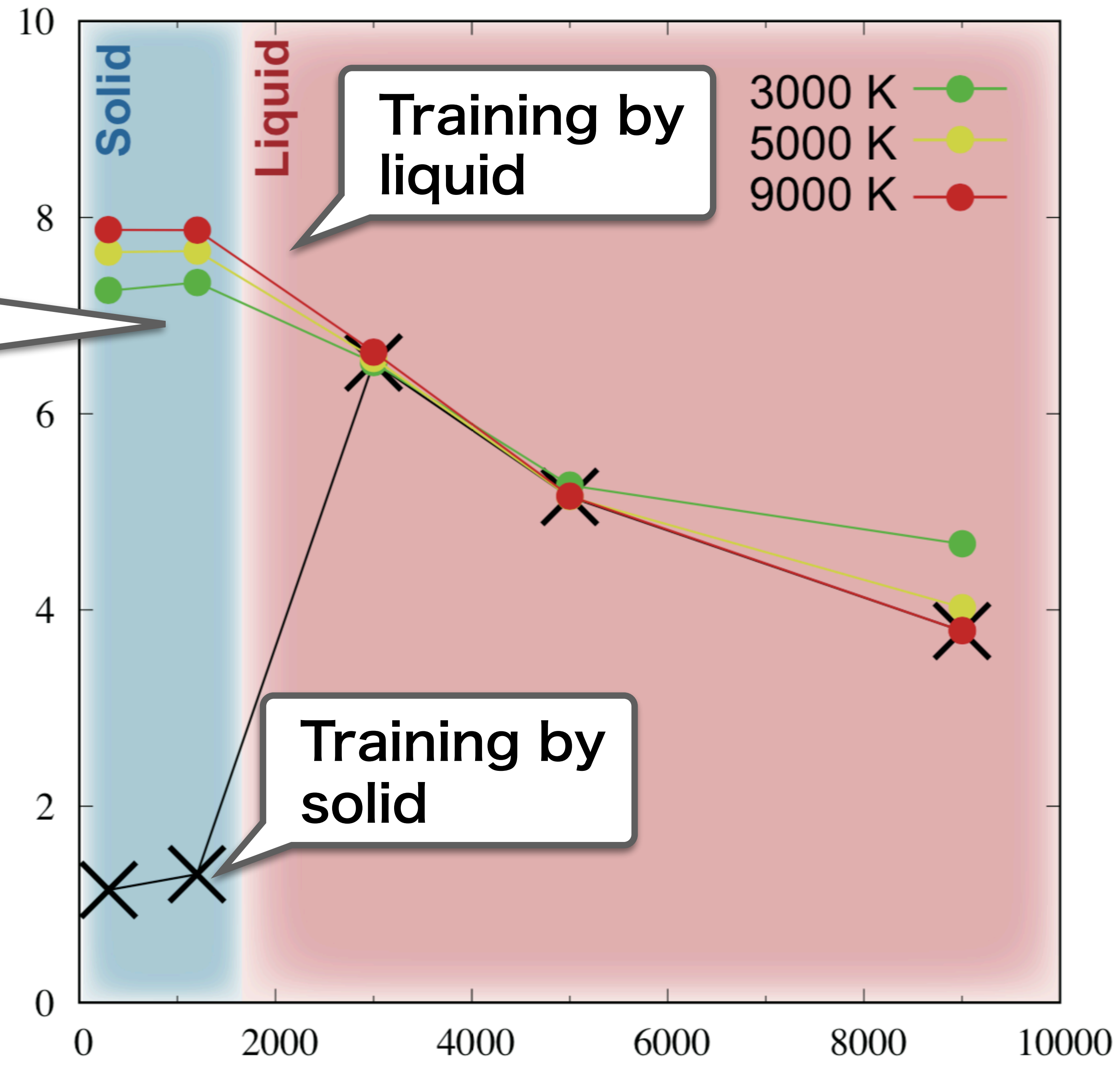
ACS Sensors 3, 1592 (2018).

ML force field for inorganic materials

By using supervised learning,
we can obtain high quality force field for solid and liquid states.



But,
transferability
is not good…



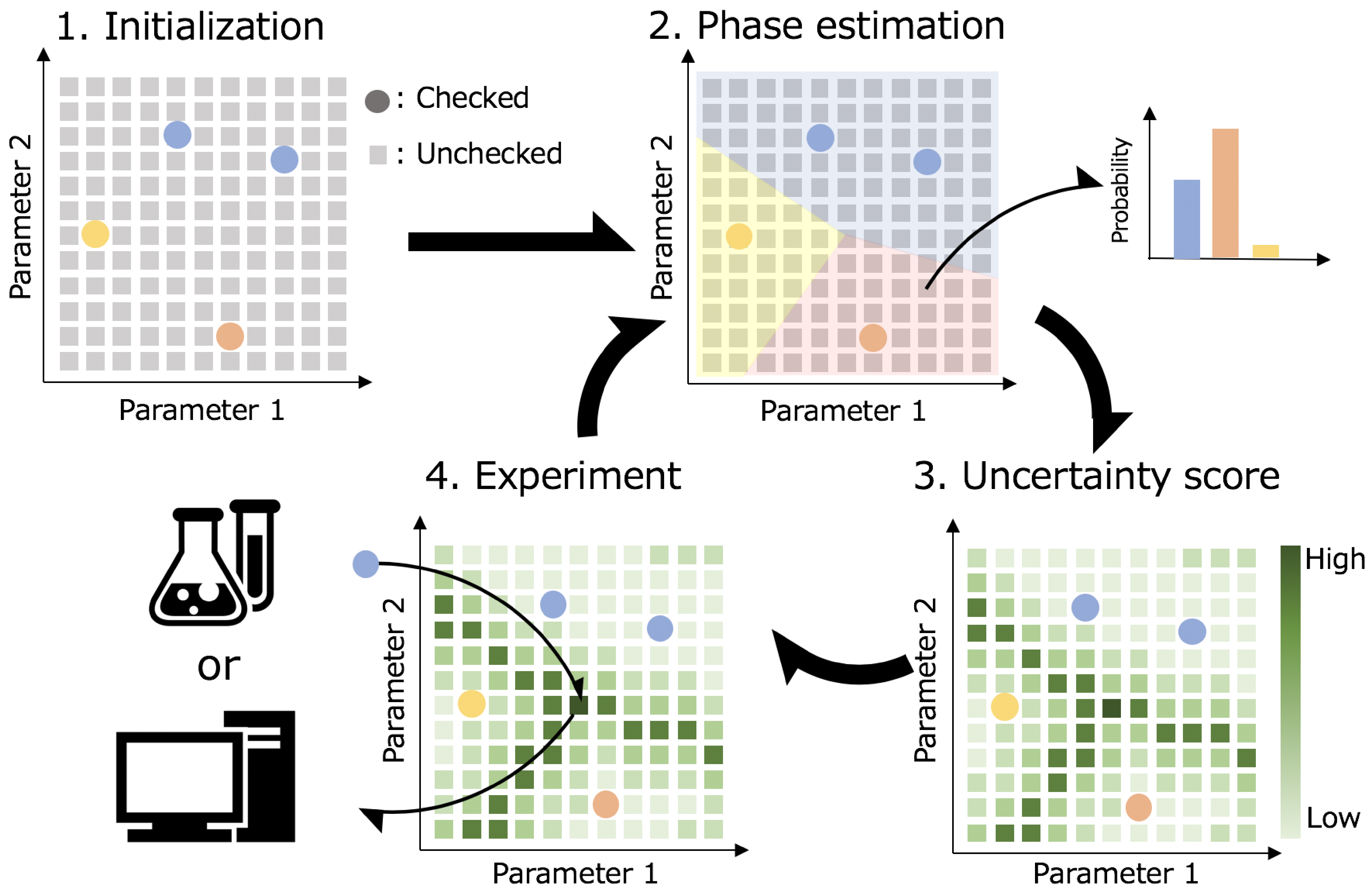
Gaussian process regression for silicon

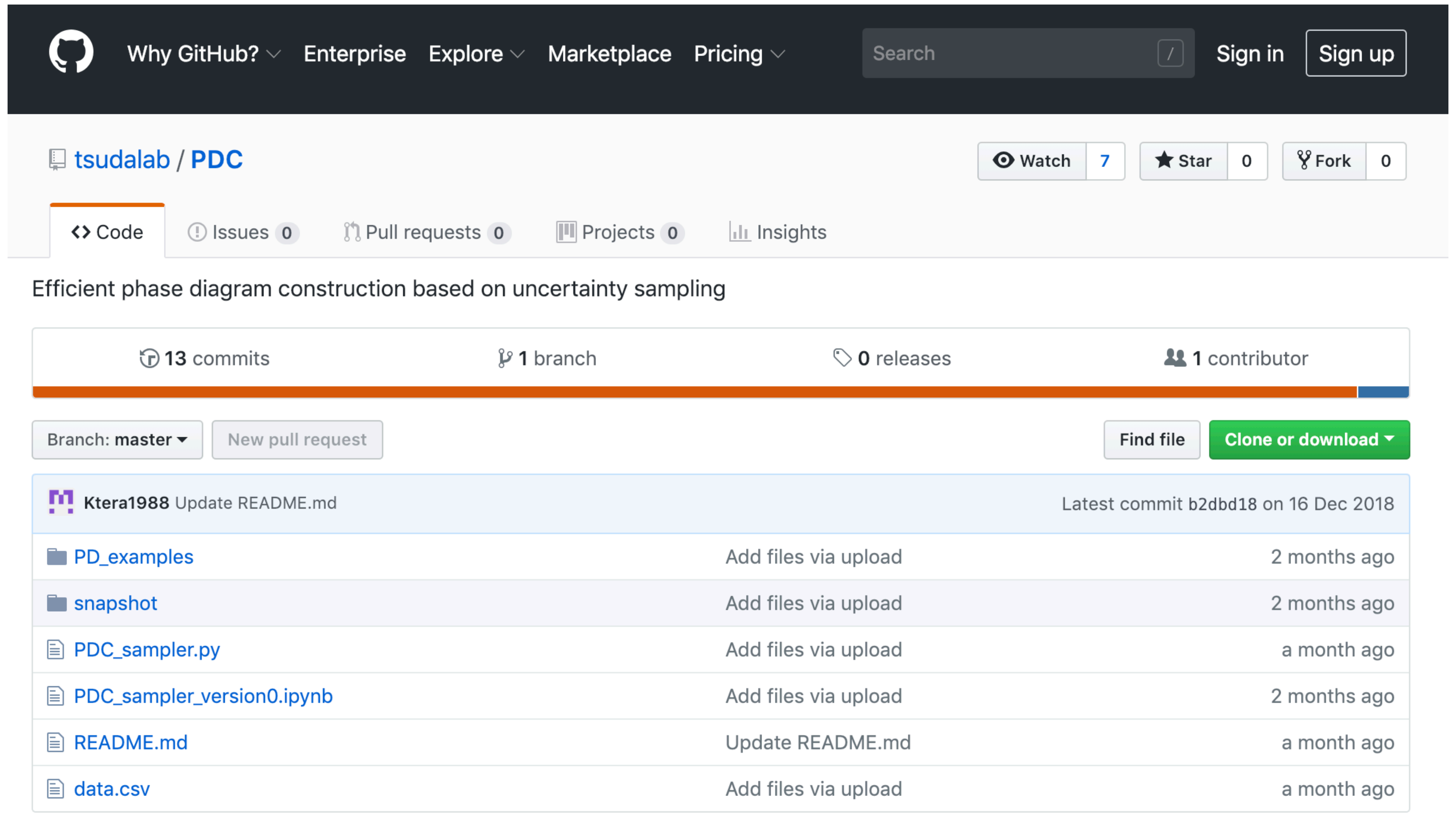
Int. J. Quant. Chem. 117, 33 (2017).

J. Phys. Soc. Jpn. 88, 044601 (2019).

Phase diagram construction by AI

By using active learning (uncertainty sampling), we can efficiently construct phase diagram.



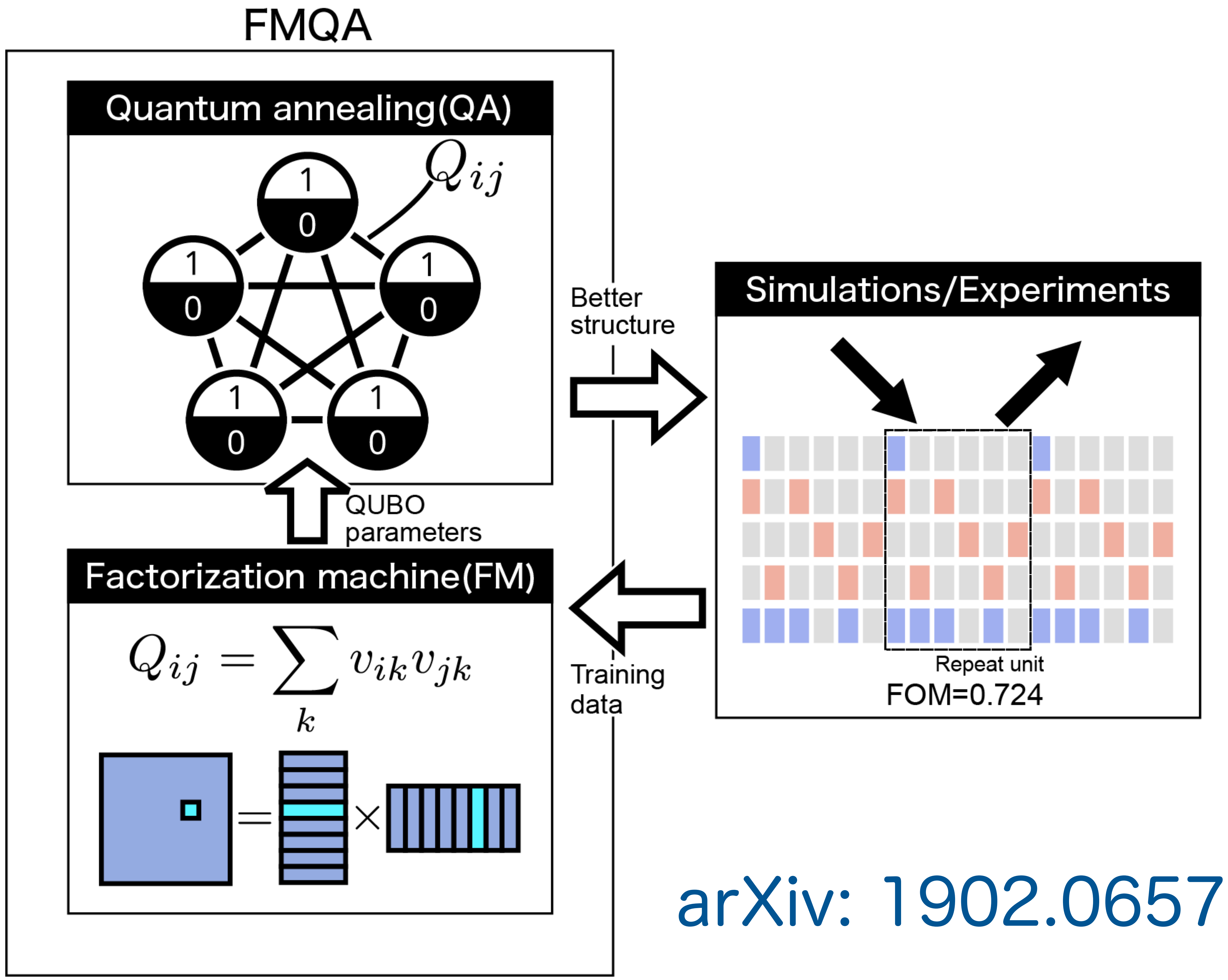


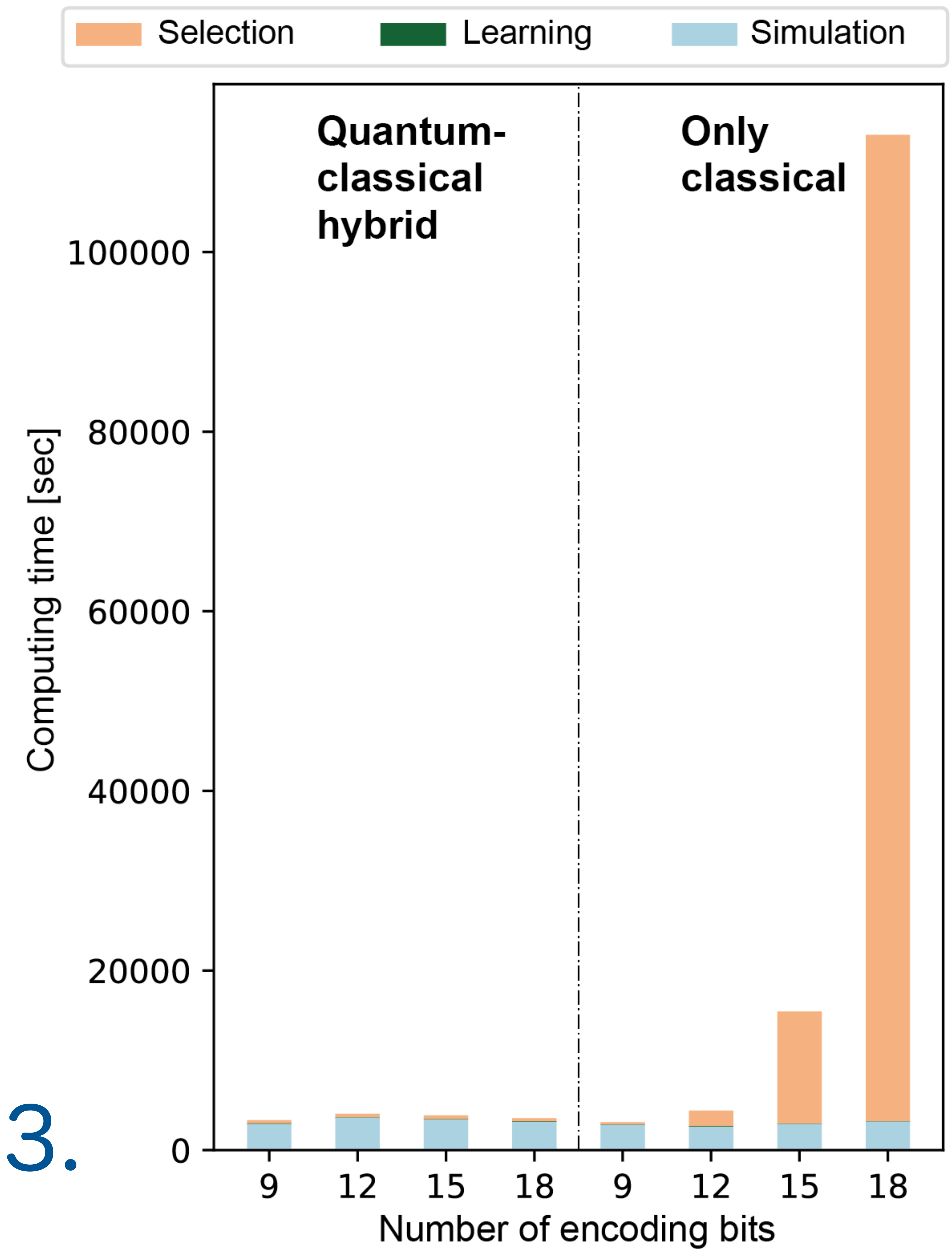
https://github.com/tsudalab/PDC

Physical Review Materials 3, 033802 (2019).

Metamaterial design by QA

By using quantum annealing and factorization machine, we can efficiently design metamaterial.





arXiv: 1902.06573.

Current my works

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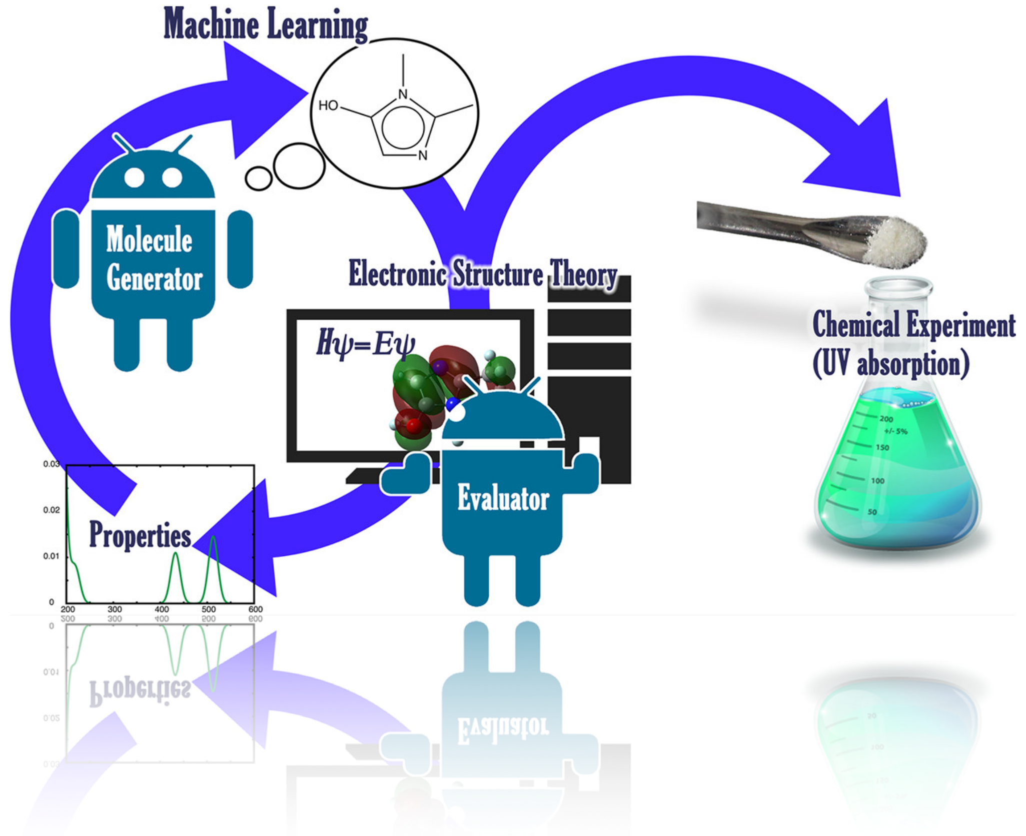
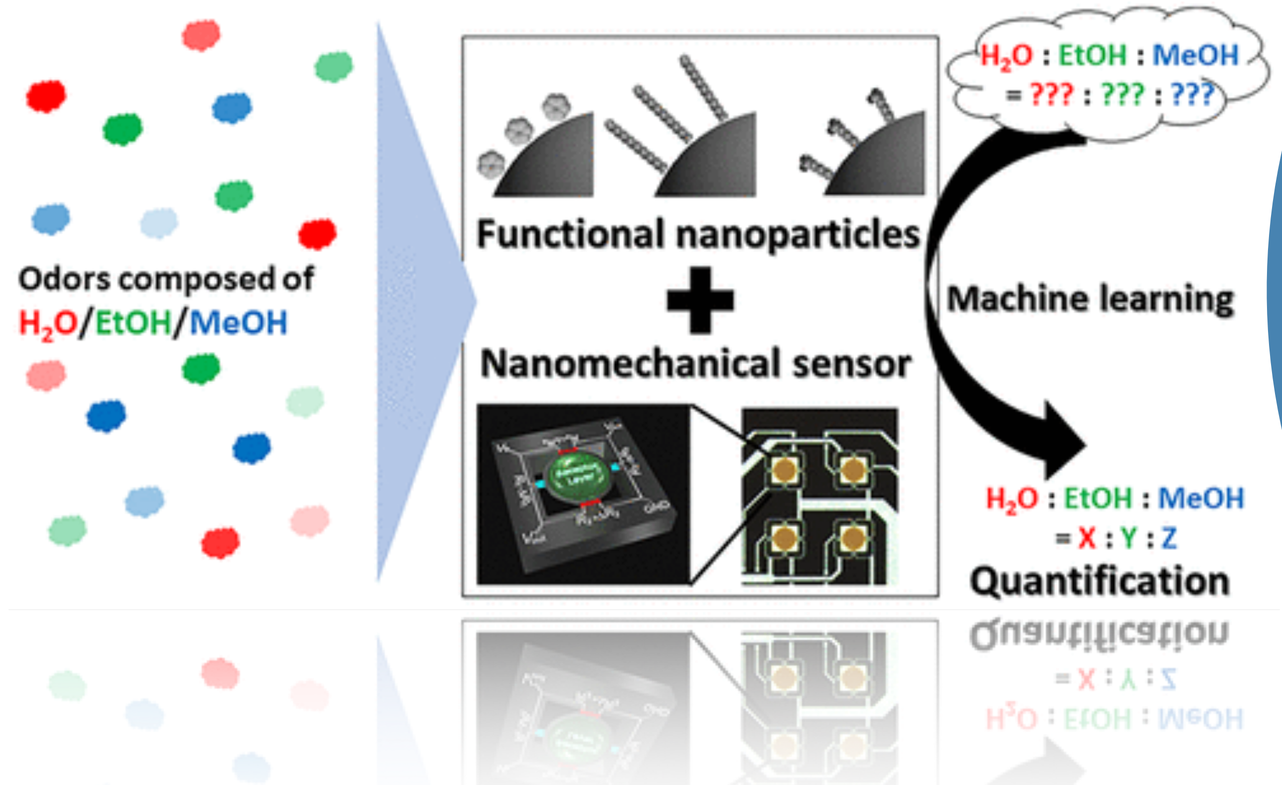
Organic



ACS Cent. Sci. 4, 1126 (2018)

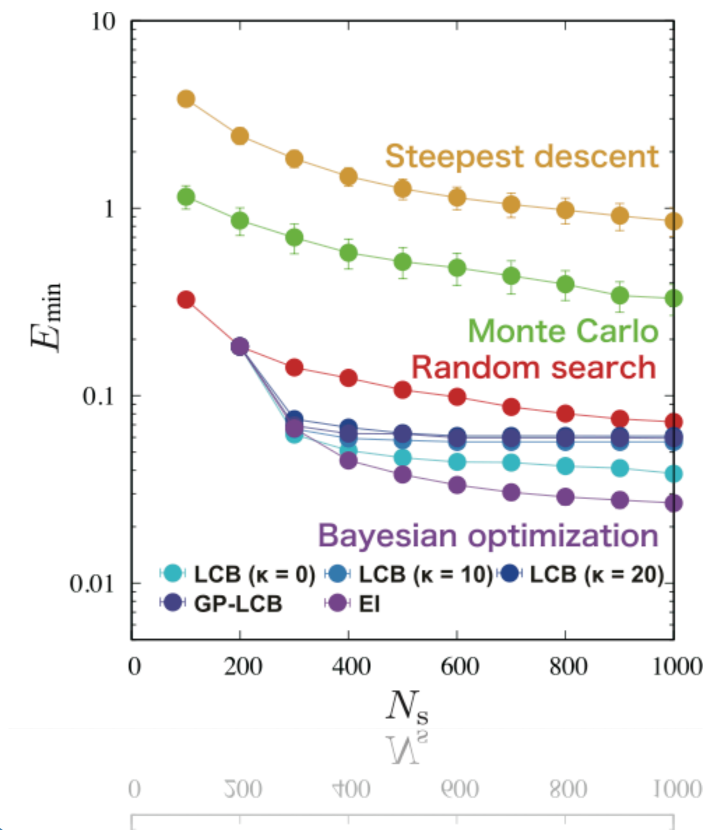
Smells Sensor

Sci. Rep. 7, 3661 (2017)



ACS Sensors 3, 1592 (2018)

Bayesian Opt.



PLoS ONE 13, e0193785 (2018)

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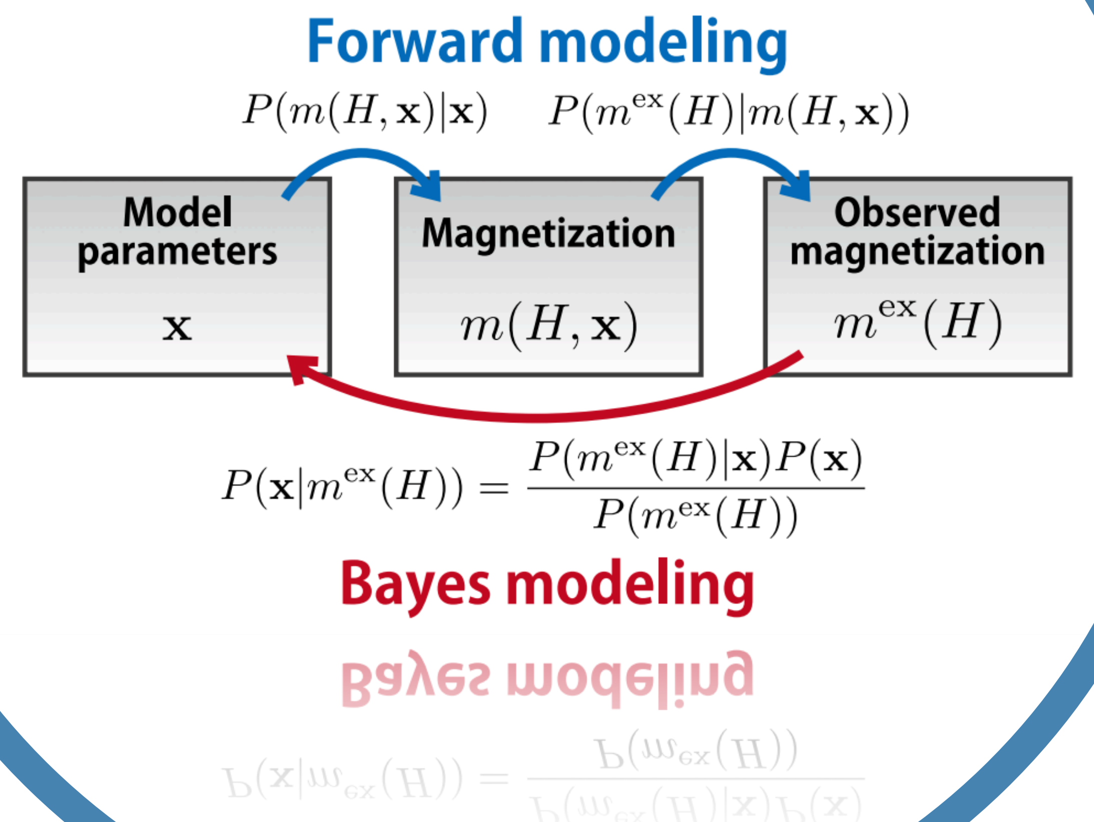
Magnet



Phys. Rev. B 95, 064407 (2017)

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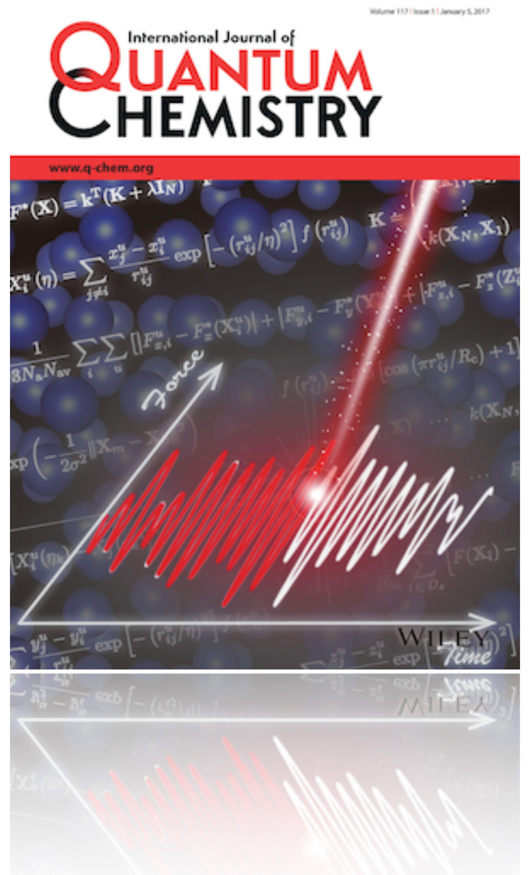
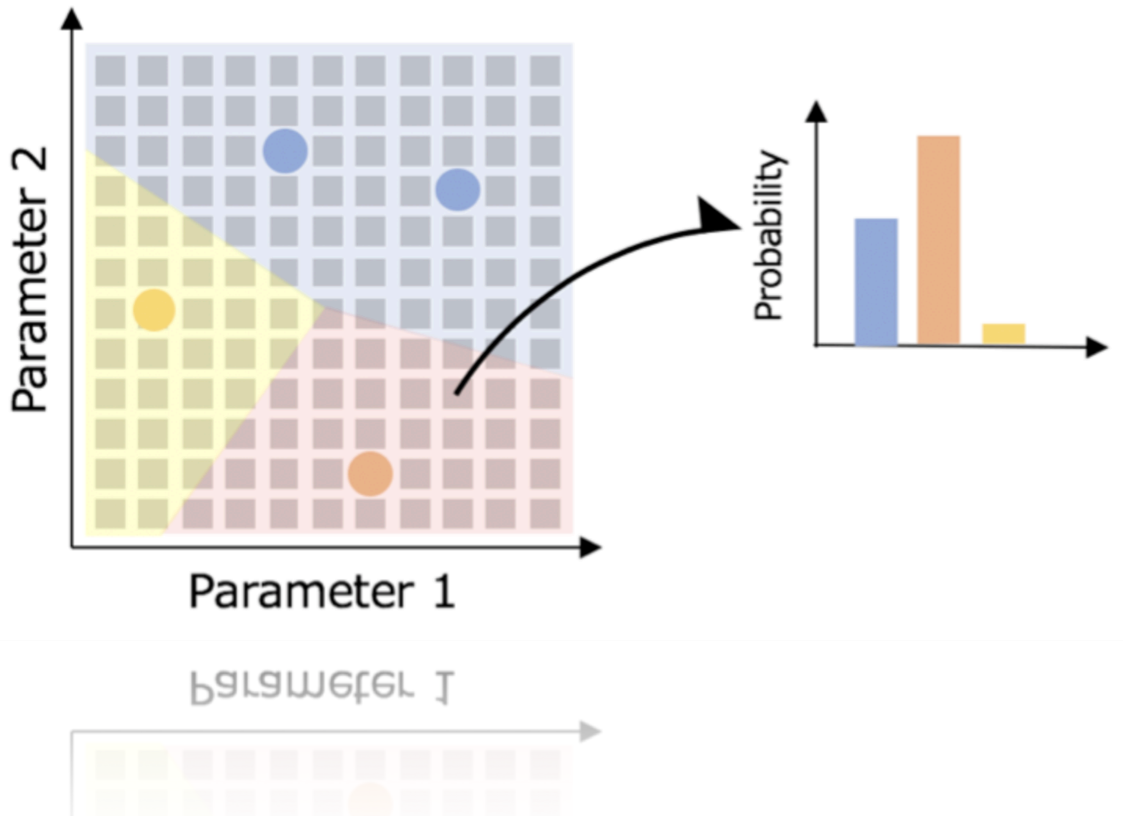


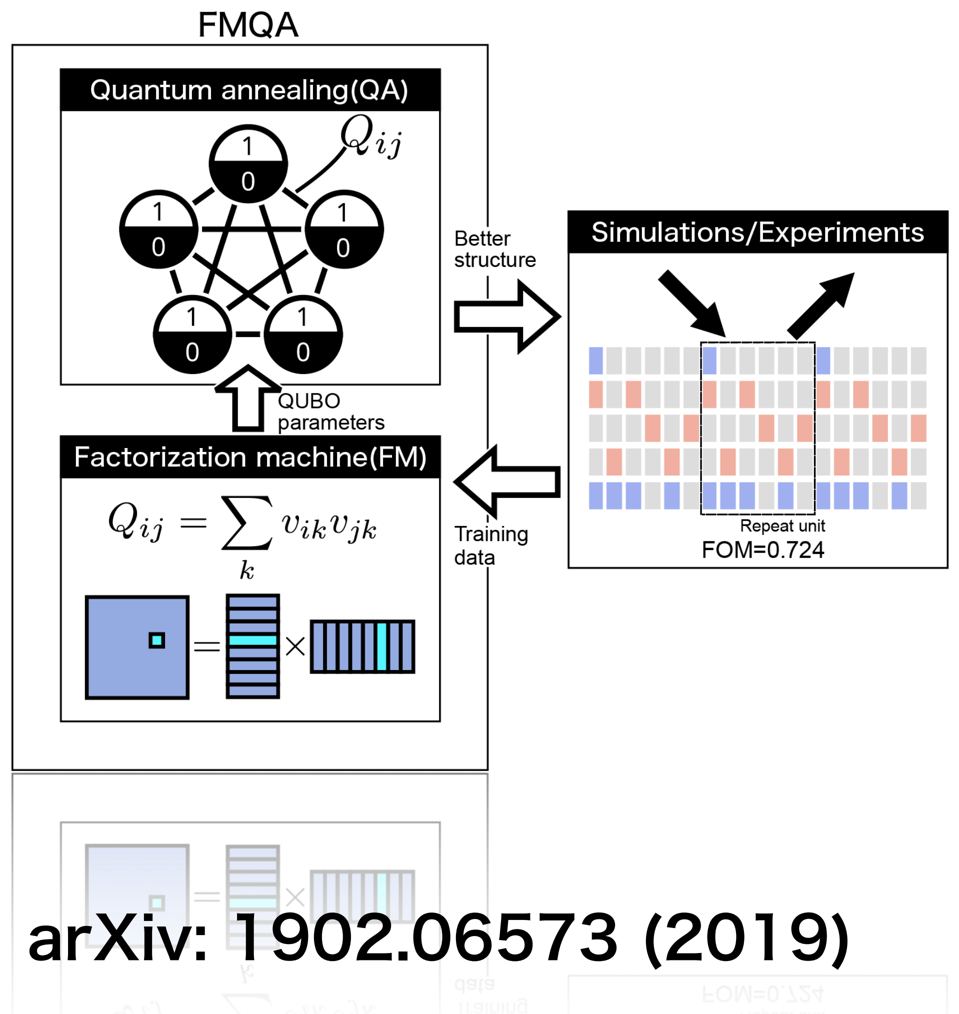
IJQC. 117, 33 (2017)
JPSJ. 88, 044601 (2019)

Phase Diagram



Phys. Rev. Mat. 3, 033802 (2019)

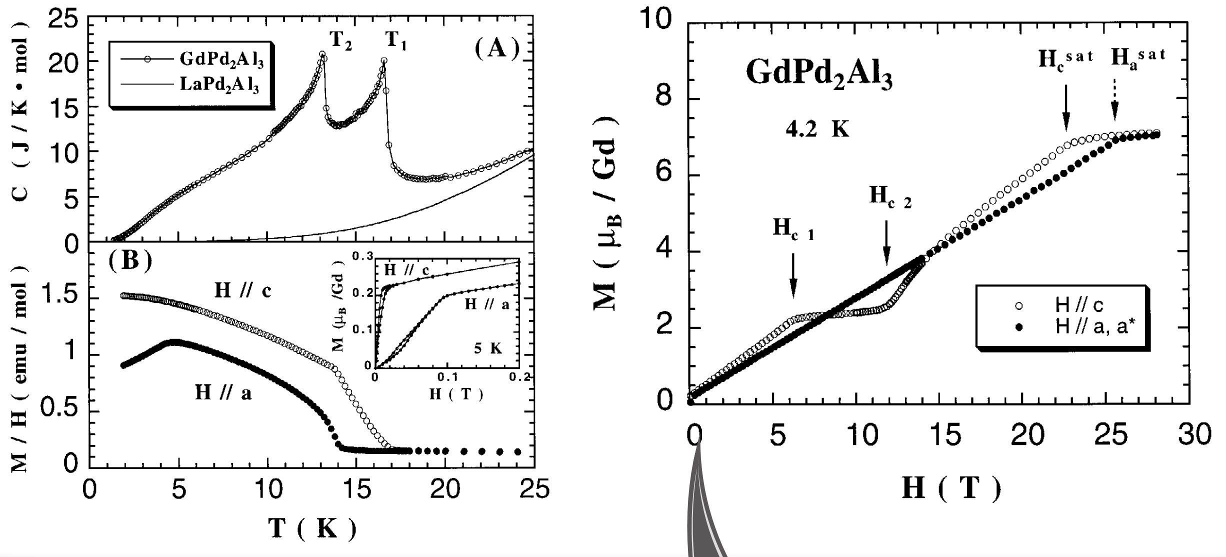
Metamaterial



arXiv: 1902.06573 (2019)

Effective model estimation method

Experimental results



Candidate models

$$b_{ij}(\mathbf{s}_i \cdot \mathbf{s}_j)^2$$

$$J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j \quad \mathbf{d}_{ij} \cdot [\mathbf{s}_i \times \mathbf{s}_j] \quad D_i(s_i^z)^2$$

$$\frac{\mathbf{s}_i \cdot \mathbf{s}_j}{r_{ij}^3} - 3\frac{(\mathbf{s}_i \cdot \mathbf{r}_{ij})(\mathbf{s}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5}$$

input

input

Automatic model estimation

Bayesian statistics

L1 regularization
L2 regularization
Full search
+
Cross validation
Elbow method

output

Plausible effective model for experimental results

Forward modeling and Bayes modeling

e.g. magnetization is target input property

Forward modeling

$P(m(H,\mathbf{x})|\mathbf{x})$ $\qquad$ $P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x}))$

$\mathbf{x}=$ {model parameters}

$$\mathcal{H}=-\sum_{i,j}J_{ij}\mathbf{s}_i\cdot\mathbf{s}_j+\cdots$$

magnetization

$$m(H,\mathbf{x})$$

observed magnetization

$$m^{\mathrm{ex}}(H)$$

$$P(\mathbf{x}|m^{\mathrm{ex}}(H))=\frac{P(m^{\mathrm{ex}}(H)|\mathbf{x})P(\mathbf{x})}{P(m^{\mathrm{ex}}(H))}$$

Bayes modeling

$P(B|A)$: Conditional probability of event B given event A (Posterior distribution)

Forward modeling and Bayes modeling

Forward modeling

$P(m(H,\mathbf{x})|\mathbf{x})$ $P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x}))$

$\mathbf{x} =$ {model parameters}

$$\mathcal{H} = -\sum_{i,j} J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j + \cdots$$

magnetization

$$m(H,\mathbf{x})$$

observed magnetization

$$m^{\mathrm{ex}}(H)$$

Definition of magnetization as thermal average of spins

$$\langle \mathbf{s}_i \rangle_{H,\mathbf{x}} = \frac{\mathrm{Tr}\mathbf{s}_i \mathrm{e}^{-\beta\mathcal{H}}}{\mathrm{Tr}\mathrm{e}^{-\beta\mathcal{H}}} \longrightarrow m(H,\mathbf{x}) = \left| \frac{1}{N|\mathbf{s}|} \sum_{i=1}^{N} \langle \mathbf{s}_i \rangle_{H,\mathbf{x}} \right|$$

Conditional probability of $m(H,\mathbf{x})$ given $\mathbf{x}$

$$P(m(H,\mathbf{x})|\mathbf{x}) = \delta\left(m(H,\mathbf{x}) - \left| \frac{1}{N|\mathbf{s}|} \sum_{i=1}^{N} \langle \mathbf{s}_i \rangle_{H,\mathbf{x}} \right| \right)$$

Magnetization is uniquely obtained when the model parameters are given.

Observation noise

Forward modeling

$P(m(H,\mathbf{x})|\mathbf{x})$ $\qquad$ $P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x}))$

$\mathbf{x} =$ {model parameters}

$\mathcal{H} = -\sum_{i,j} J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j + \cdots$

magnetization

$m(H,\mathbf{x})$

observed magnetization

$m^{\mathrm{ex}}(H)$

Existence of observation noise in $m^{\mathrm{ex}}(H)$

$$m^{\mathrm{ex}}(H) = m(H,\mathbf{x}) \underline{+\,\varepsilon}$$

observation noise

Assumption : $P(\varepsilon) \propto \exp\left(-\frac{\varepsilon^2}{2\sigma^2}\right)$

Conditional probability of $m^{\mathrm{ex}}(H)$ given $m(H,\mathbf{x})$

$$P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x})) \propto \exp\left(-\frac{1}{2\sigma^2}(m^{\mathrm{ex}}(H) - m(H,\mathbf{x}))^2\right)$$

Conditional probability

Forward modeling

$P(m(H,\mathbf{x})|\mathbf{x})$ $\quad$ $P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x}))$

$\mathbf{x} =$ {model parameters}

$\mathcal{H} = -\sum_{i,j} J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j + \cdots$

magnetization

$m(H,\mathbf{x})$

observed magnetization

$m^{\mathrm{ex}}(H)$

Conditional probability of $m^{\mathrm{ex}}(H)$ given $\mathbf{x}$

$$P(m^{\mathrm{ex}}(H)|\mathbf{x}) \propto \int dm(H,\mathbf{x}) P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x}))P(m(H,\mathbf{x})|\mathbf{x})$$

$$\propto \exp\left[-\frac{1}{2\sigma^2}\left(m^{\mathrm{ex}}(H) - \left|\frac{1}{N|\mathbf{s}|}\sum_{i=1}^{N}\langle \mathbf{s}_i\rangle_{H,\mathbf{x}}\right|\right)^2\right]$$

$m^{\mathrm{ex}}(H)$ where $P(m^{\mathrm{ex}}(H)|\mathbf{x})$ is maximize. → Observed magnetization

Bayes modeling

Forward modeling

$P(m(H,\mathbf{x})|\mathbf{x})$ $\qquad$ $P(m^{\mathrm{ex}}(H)|m(H,\mathbf{x}))$

$\mathbf{x}=$ {model parameters}

$$\mathcal{H}=-\sum_{i,j}J_{ij}\mathbf{s}_i\cdot\mathbf{s}_j+\cdots$$

magnetization

$$m(H,\mathbf{x})$$

observed
magnetization

$$m^{\mathrm{ex}}(H)$$

$$P(\mathbf{x}|m^{\mathrm{ex}}(H))=\frac{P(m^{\mathrm{ex}}(H)|\mathbf{x})P(\mathbf{x})}{P(m^{\mathrm{ex}}(H))}$$

: Posterior distribution

Bayes modeling

$\mathbf{x}$ where $P(\mathbf{x}|m^{\mathrm{ex}}(H))$ is maximize. → Plausible model parameters

Summary of effective model estimation

We search the maximizer of the posterior distribution when the measured physical quantities are inputted.

Posterior distribution

$$P(\underline{\mathbf{x}}|\{y^{\mathrm{ex}}(g_l)\}_{l=1,\cdots,L}) = \exp[-E(\mathbf{x})]$$

Model parameters

Energy function

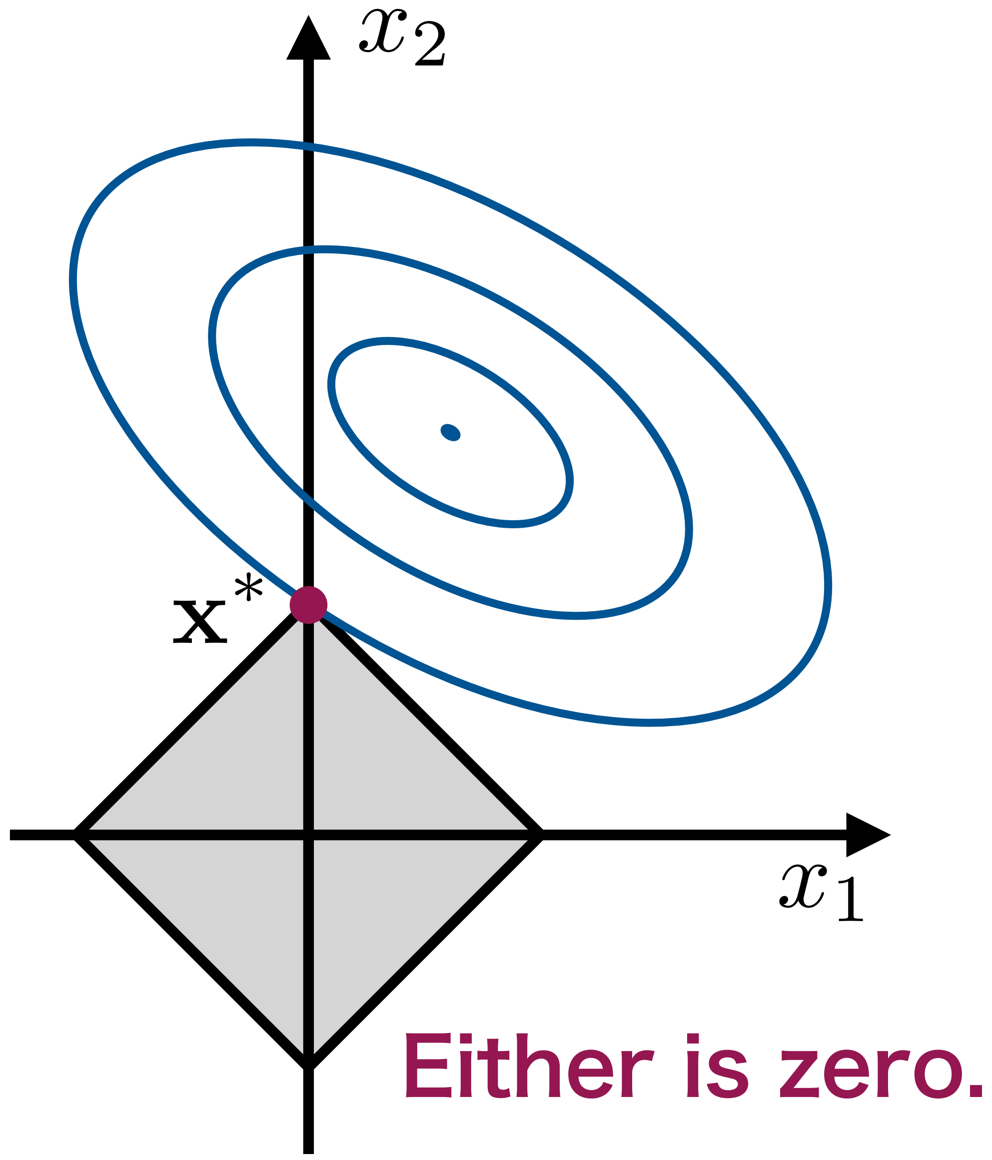
$$E(\mathbf{x}) = \frac{1}{2\sigma^2}\sum_{l=1}^{L}\Big[\underbrace{y^{\mathrm{ex}}(g_l)}_{\text{Input physical quantities}} - \underbrace{y^{\mathrm{cal}}(g_l,\mathbf{x})}_{\text{Calculated physical quantities by effective model}}\Big]^2 - \log \underbrace{P(\mathbf{x})}_{\text{Prior distribution}}$$

R. Tamura and K. Hukushima, Phys. Rev. B **95**, 064407 (2017).

Prior distribution

L1 reguralization

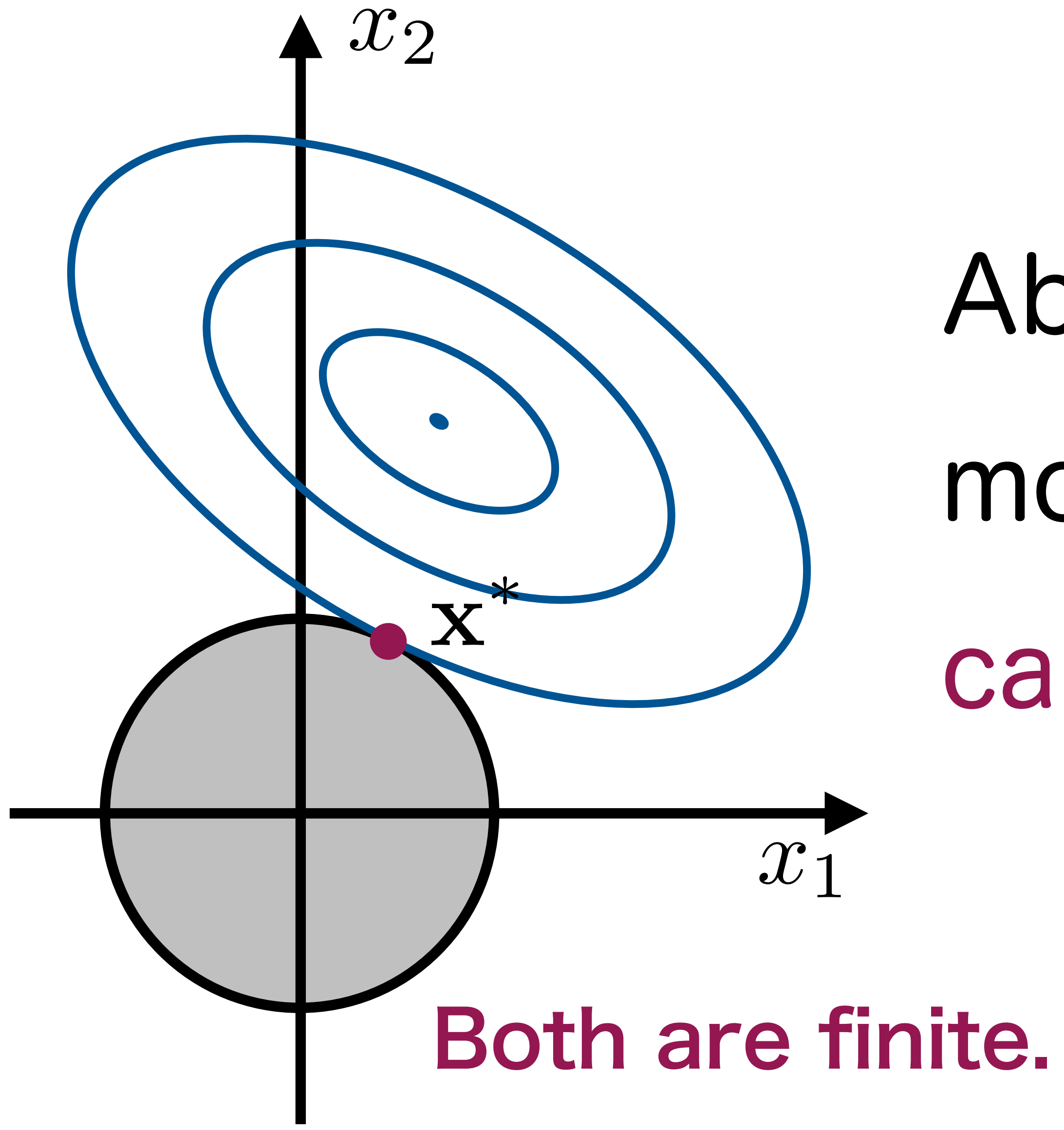
$$P(\mathbf{x}) \propto \exp\left(-\lambda|\mathbf{x}|\right)$$



Model parameters with large contributions can be selected based on the feature selection.

L2 reguralization

$$P(\mathbf{x}) \propto \exp\left(-\lambda||\mathbf{x}||^2\right)$$



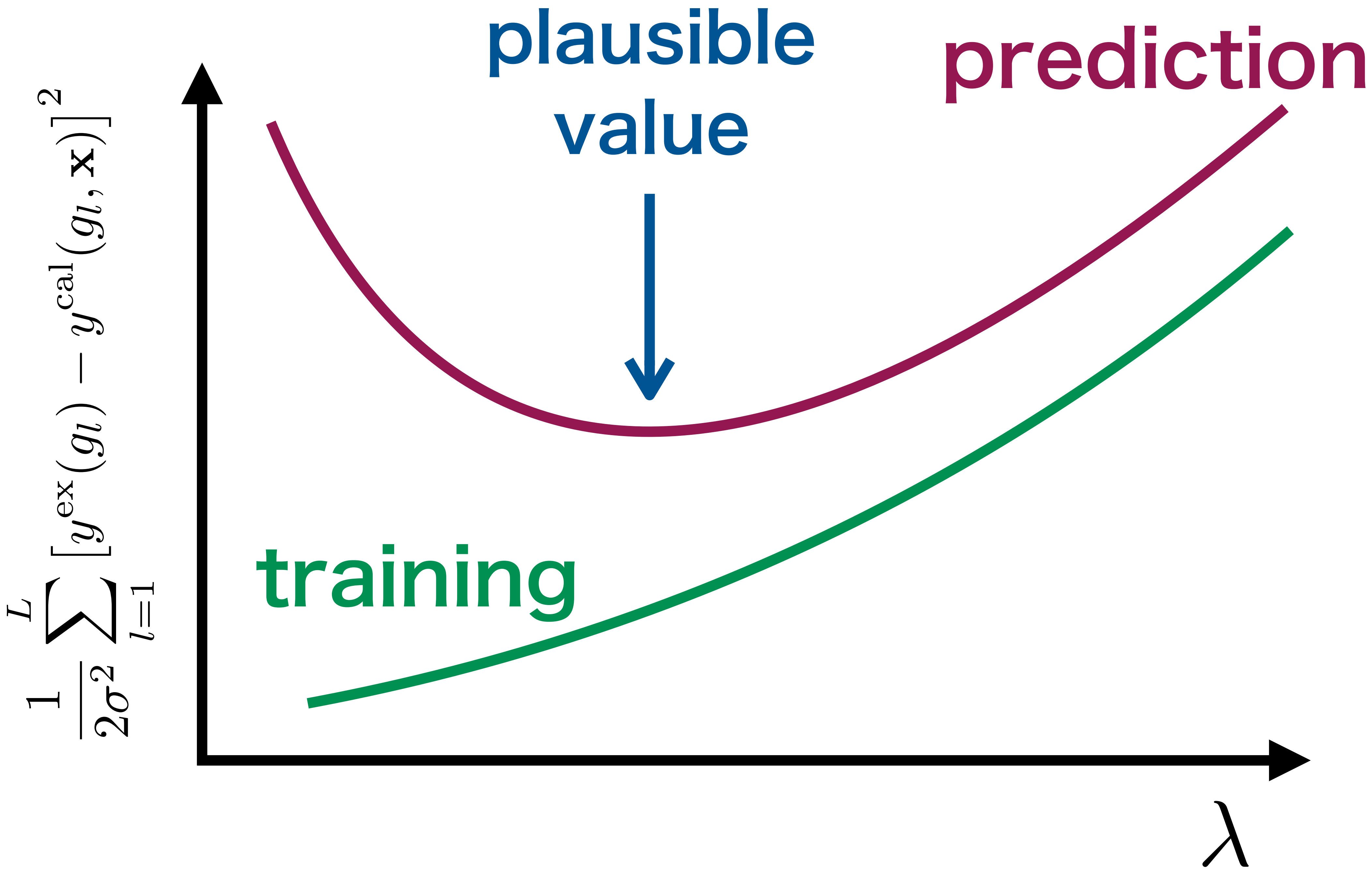
Absolute values of model parameters can be suppressed.

Depending on the situation, it is necessary to select a proper prior distribution.

Determination of hyperparameter

- **Cross validation**

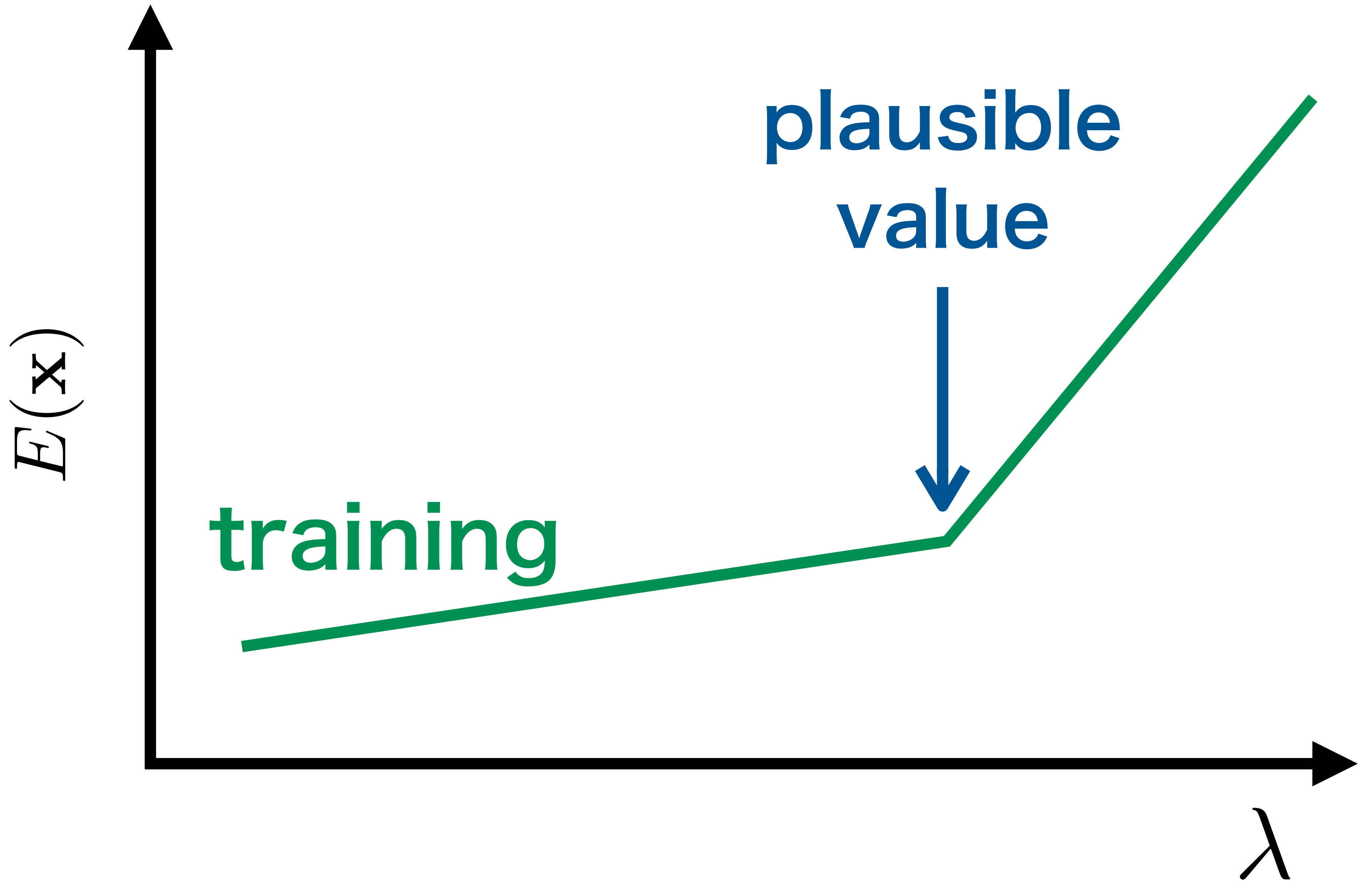
  Plausible value is determined at the point where prediction error is minimized.



useful for L1 regularization
(overfitting occurs)

- **Elbow method**

  Plausible value is determined by the large change point in energy function.



useful for L2 regularization
(overfitting does not occur)

Demonstration: Theoretical model

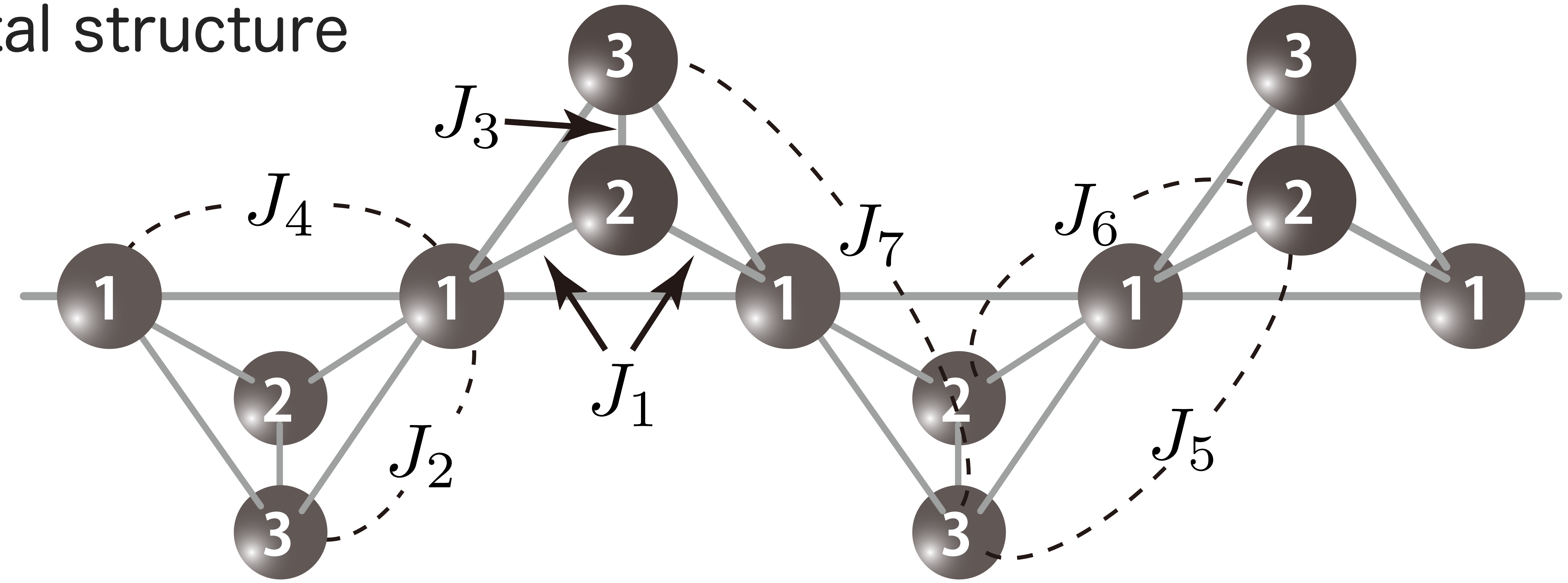
Target classical Heisenberg model with biquadratic interactions

(magnetization plateau is appeared)

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} \left[\mathbf{s}_i \cdot \mathbf{s}_j - b_{ij} (\mathbf{s}_i \cdot \mathbf{s}_j)^2 \right] - H \sum_i s_i^z \qquad b_{ij} = bJ_{ij}$$

$\mathbf{s}_i$: Classical Heisenberg spin (S=1/2)

Crystal structure



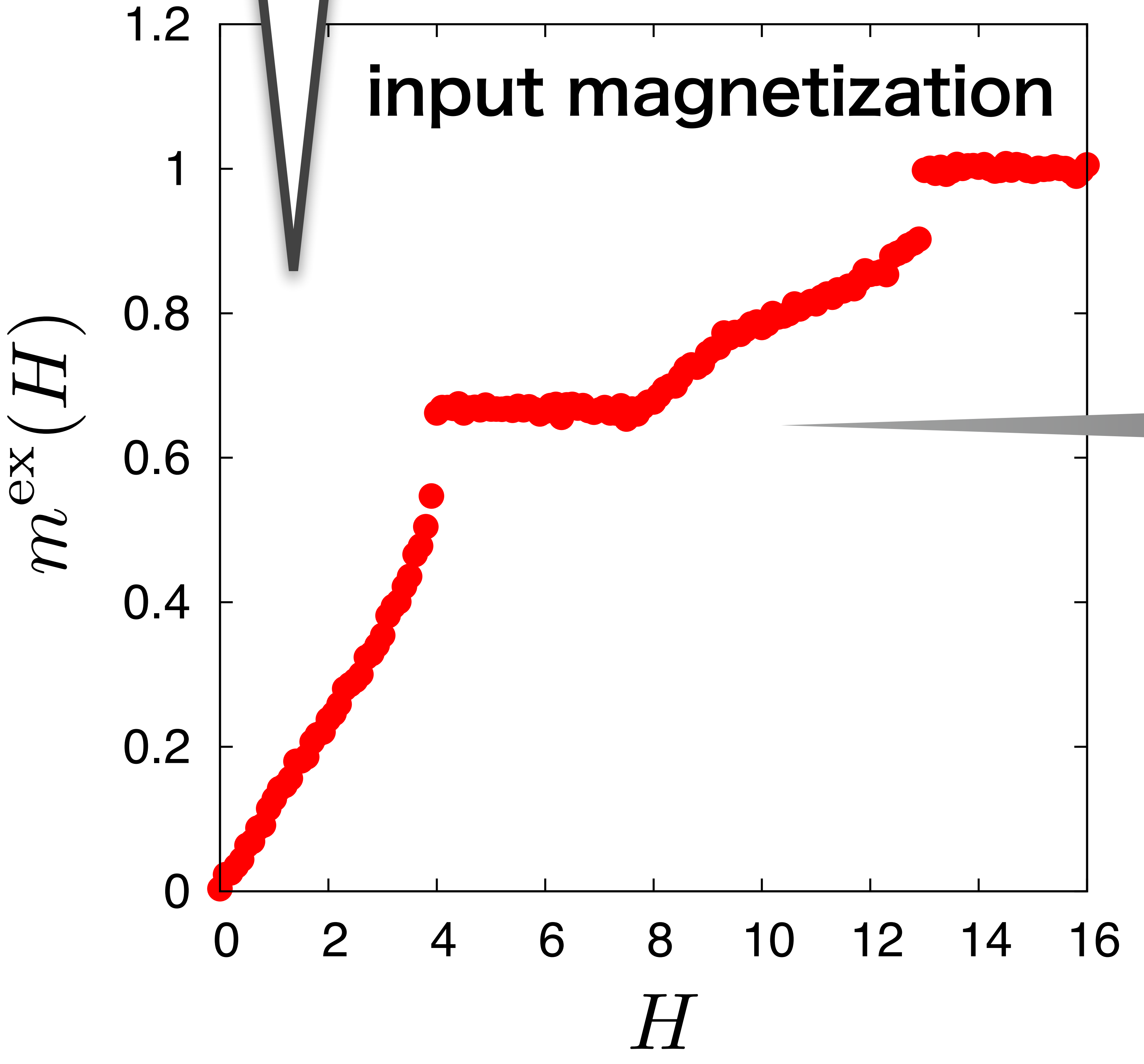
We tried:
L1 regularization
&
cross validation

model parameters : $\mathbf{x} = \{J_1, J_2, J_3, J_4, J_5, J_6, J_7, b\}$

Estimation results

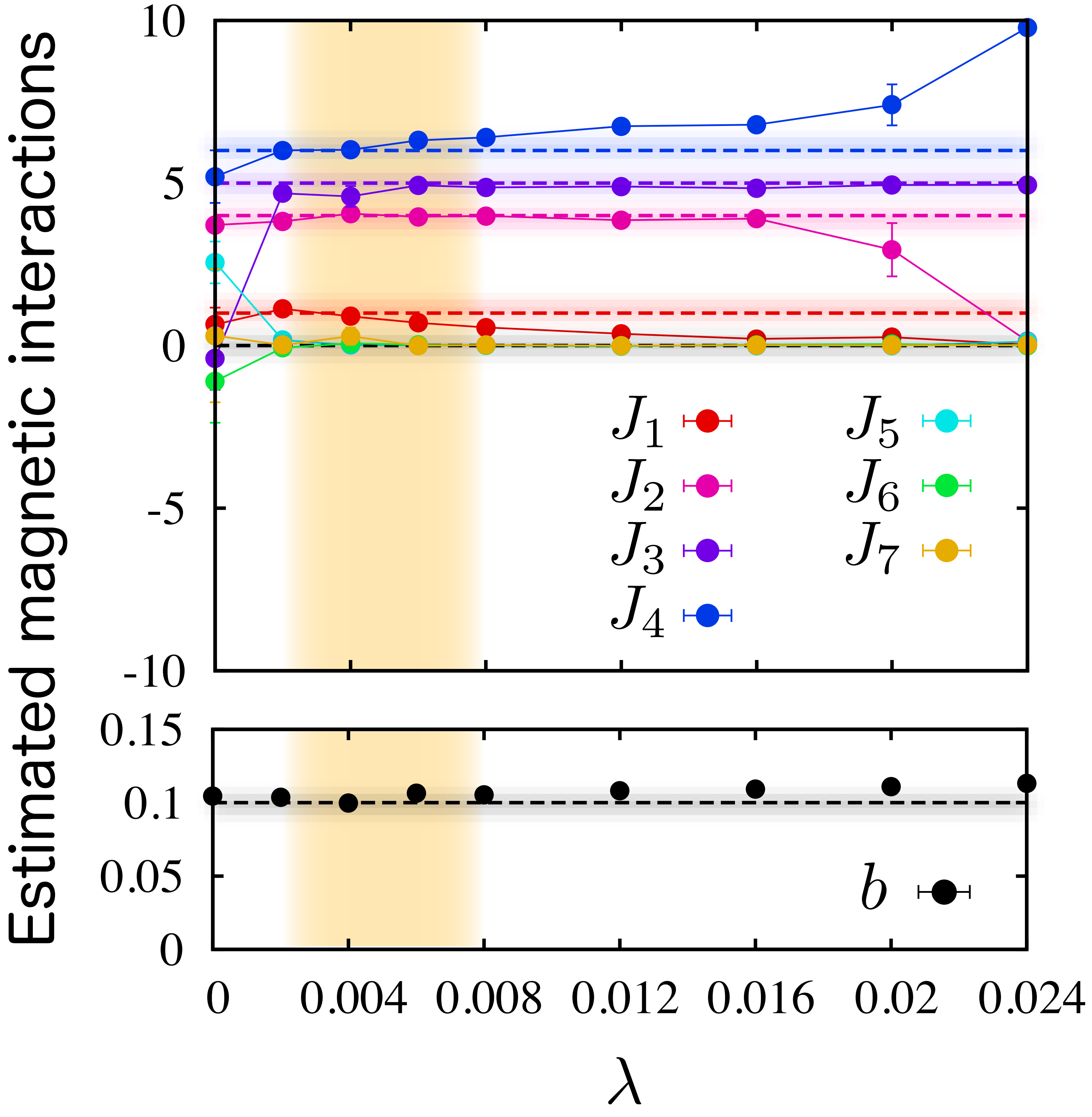
Zero temperature magnetization curve for $J_1 = 1, J_2 = 4, J_3 = 5, J_4 = 6, b = 0.1$ $\boxed{J_5 = J_6 = J_7 = 0}$ + Gaussian noise

Magnetization is calculated by the steepest descent method.

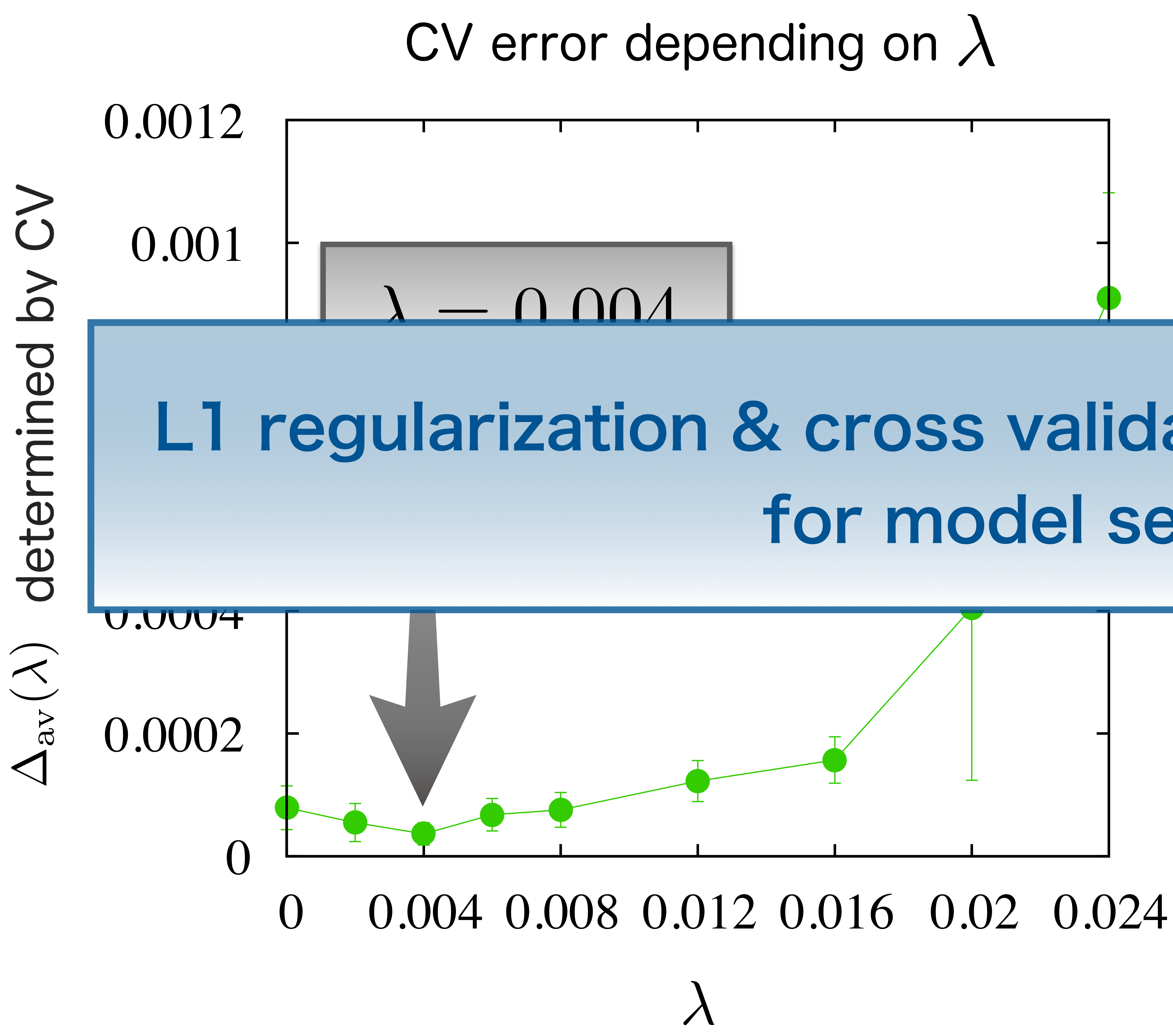


Estimation results

Prior distribution: L1 regularization



Estimated results and prediction



	Estimated	Correct
J_1	1.074	1.000
J_2	3.850	4.000
J_5	0.011	0.000
J_6	-0.051	0.000
J_7	0.002	0.000
b	0.102	0.100

L1 regularization & cross validation is effectively worked for model selection.

Time consuming problem

Energy function in effective model estimation

$$E(\mathbf{x}) = \frac{1}{2\sigma^2}\sum_{l=1}^{L}\left[y^{\mathrm{ex}}(g_l) - y^{\mathrm{cal}}(g_l, \mathbf{x})\right]^2 - \log P(\mathbf{x})$$

We should calculate the thermal average of physical quantity by Monte Carlo, exact diagonalization, DMRG, mean-field, etc.

Searching procedure of maximizer of posterior distribution

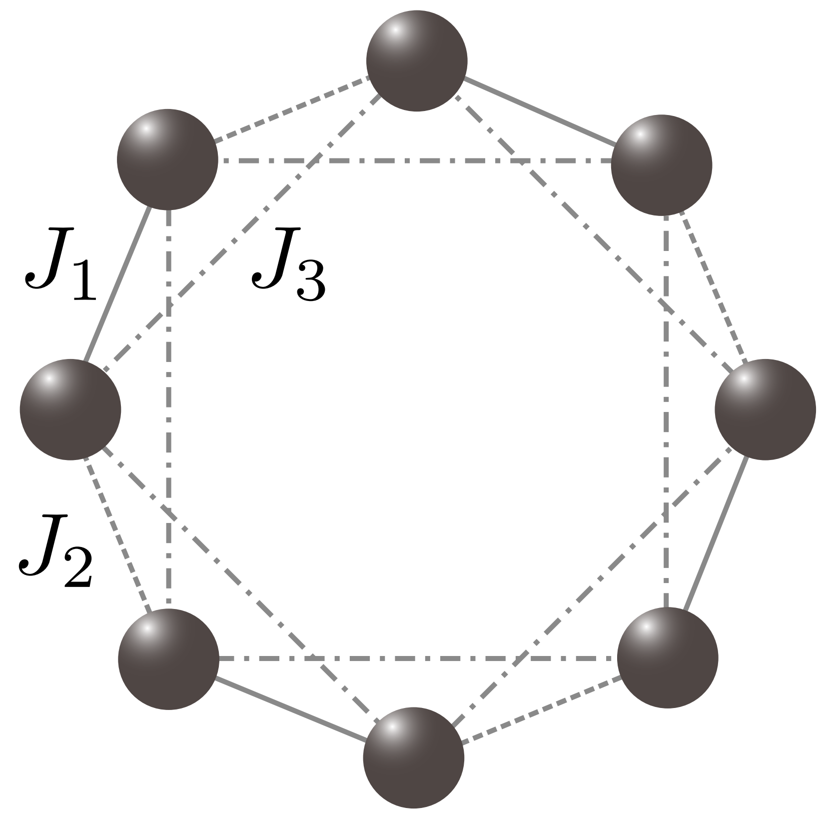
Values of model parameters are changed. → Thermal average is calculated. → One point value on posterior distribution is obtained.

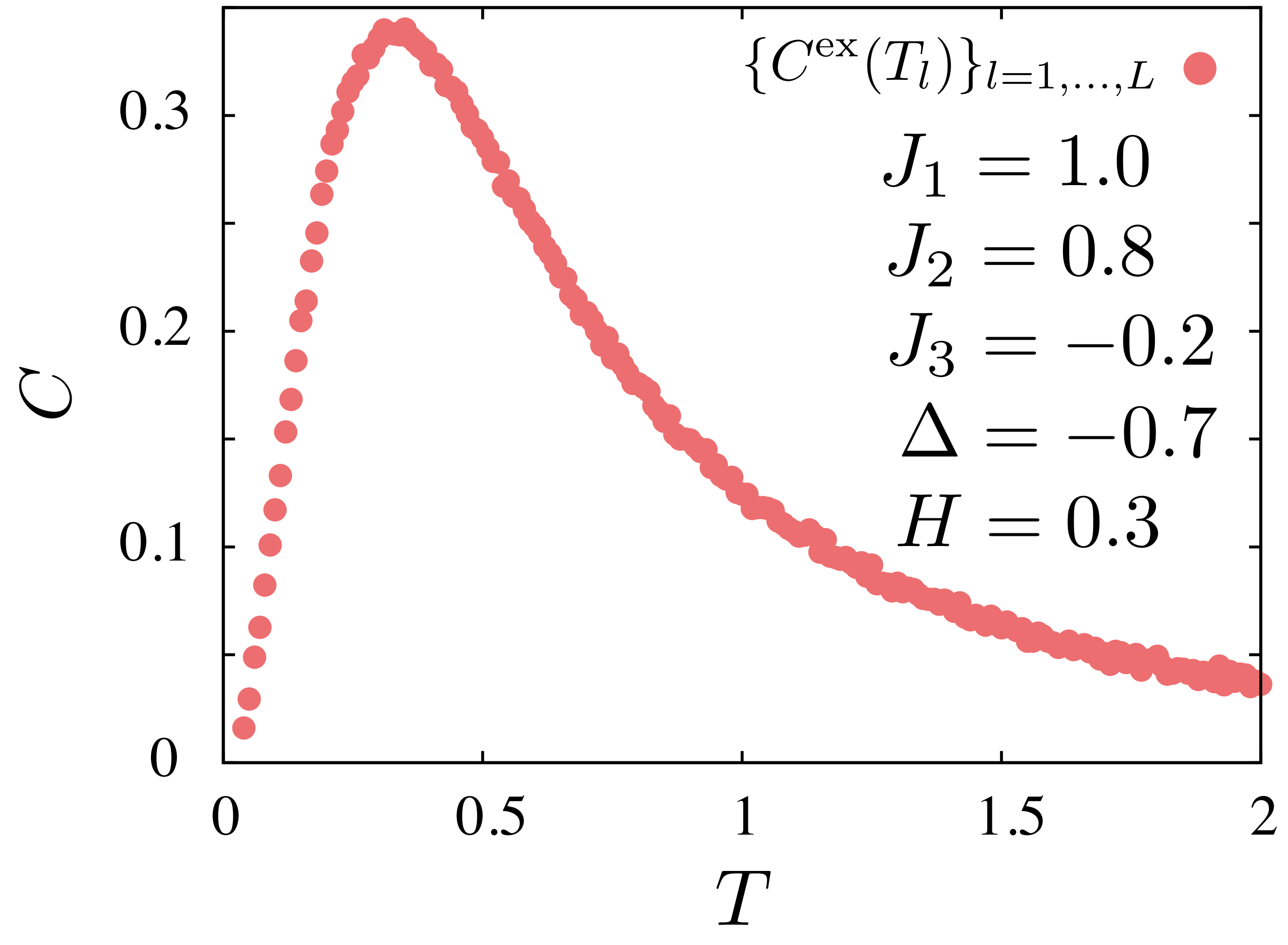
Time consuming !

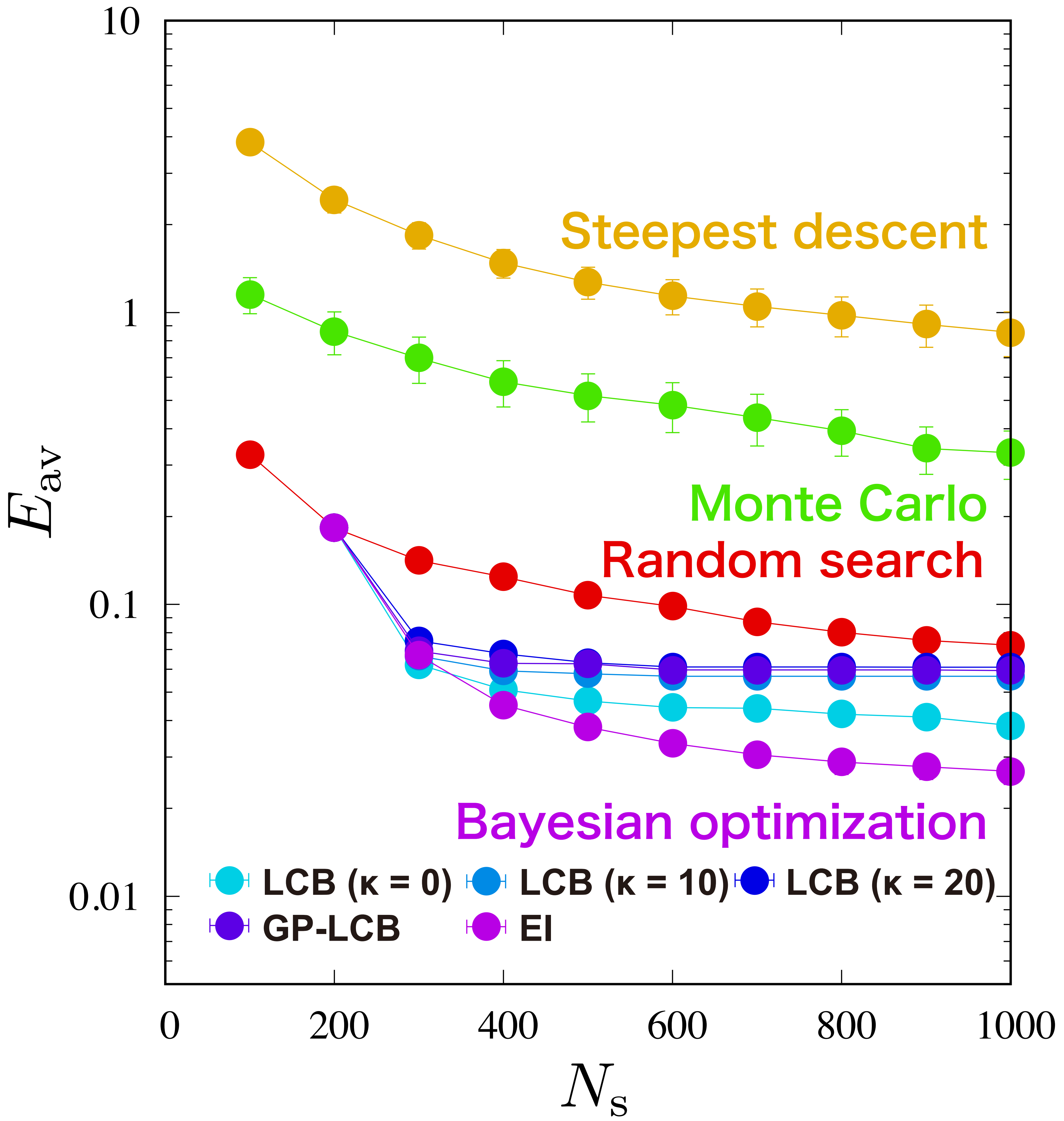
We want to reduce the number of calculations of thermal averages. → **Bayesian optimization**

Bayesian optimization

Quantum Heisenberg model : $\mathcal{H} = -\sum_{i,j} J_{ij}\left[s_i^x s_j^x + s_i^y s_j^y + \Delta s_i^z s_j^z\right] - H\sum_i s_i^z$





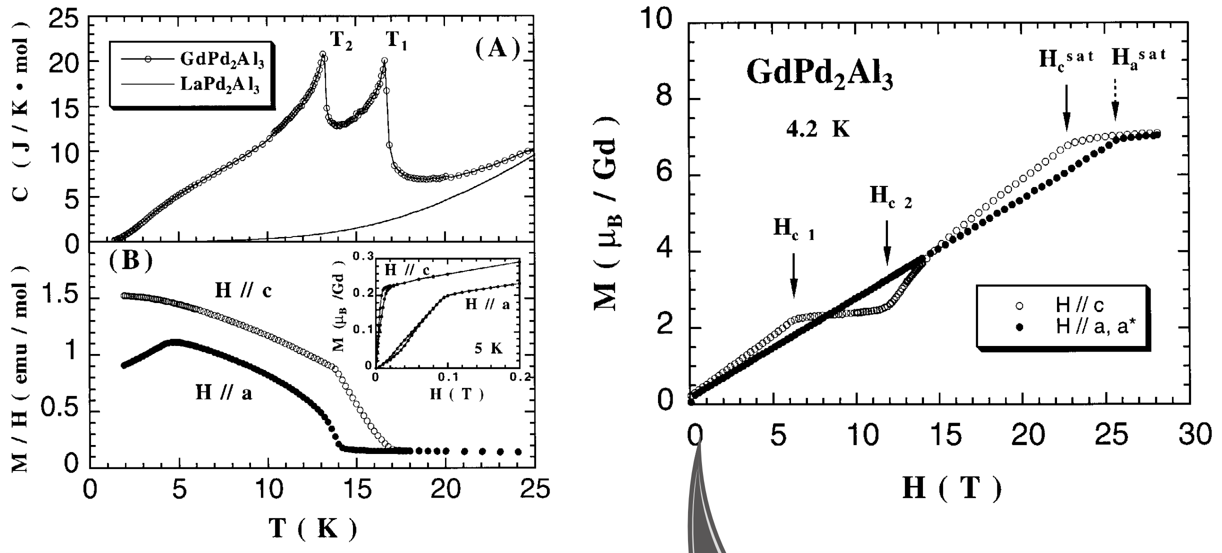


R. Tamura and K. Hukushima, PLoS ONE 13, e0193785 (2018).

Time consuming problem will be overcome by using Bayesian optimization.

Summary

Experimental results



Candidate models

$$b_{ij}(\mathbf{s}_i\cdot\mathbf{s}_j)^2$$

$$J_{ij}\mathbf{s}_i\cdot\mathbf{s}_j \qquad \mathbf{d}_{ij}\cdot[\mathbf{s}_i\times\mathbf{s}_j] \qquad D_i(s_i^z)^2$$

$$\frac{\mathbf{s}_i\cdot\mathbf{s}_j}{r_{ij}^3}-3\frac{(\mathbf{s}_i\cdot\mathbf{r}_{ij})(\mathbf{s}_j\cdot\mathbf{r}_{ij})}{r_{ij}^5}$$

input

input

Automatic model estimation

Bayesian statistics

Bayesian optimization

Machine learning based optimization

L1 regularization
L2 regularization
Full search
+
Cross validation
Elbow method

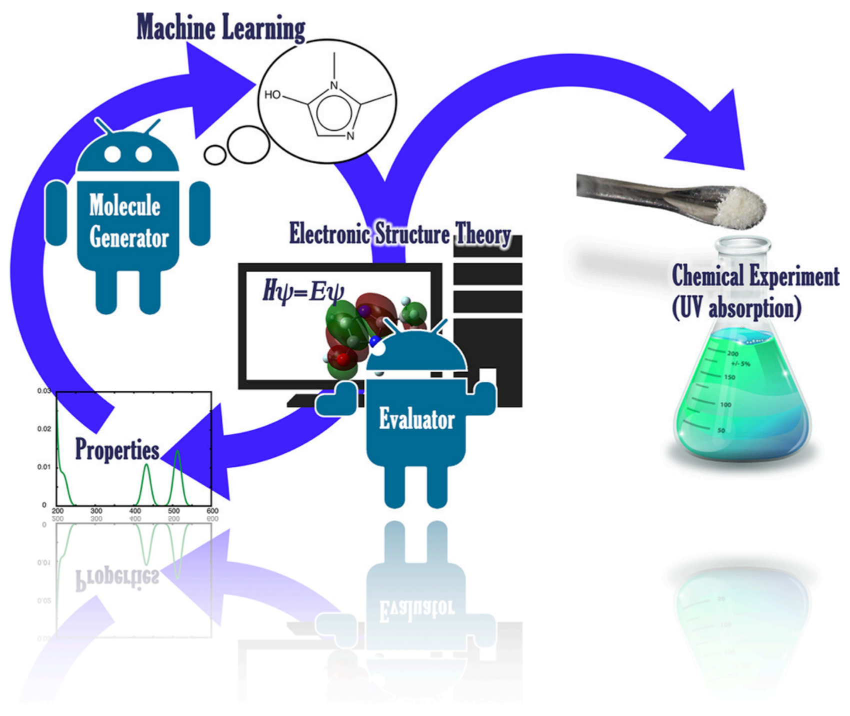
output

Plausible effective model for experimental results

Summary

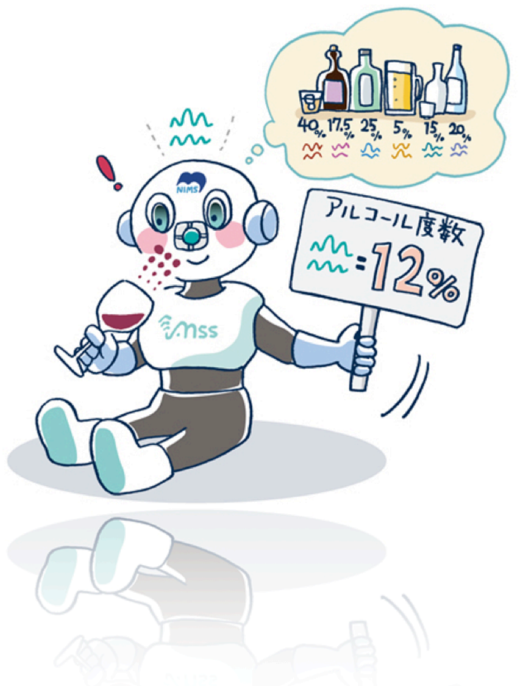
Machine learning is very useful as support tool in materials science.

Organic

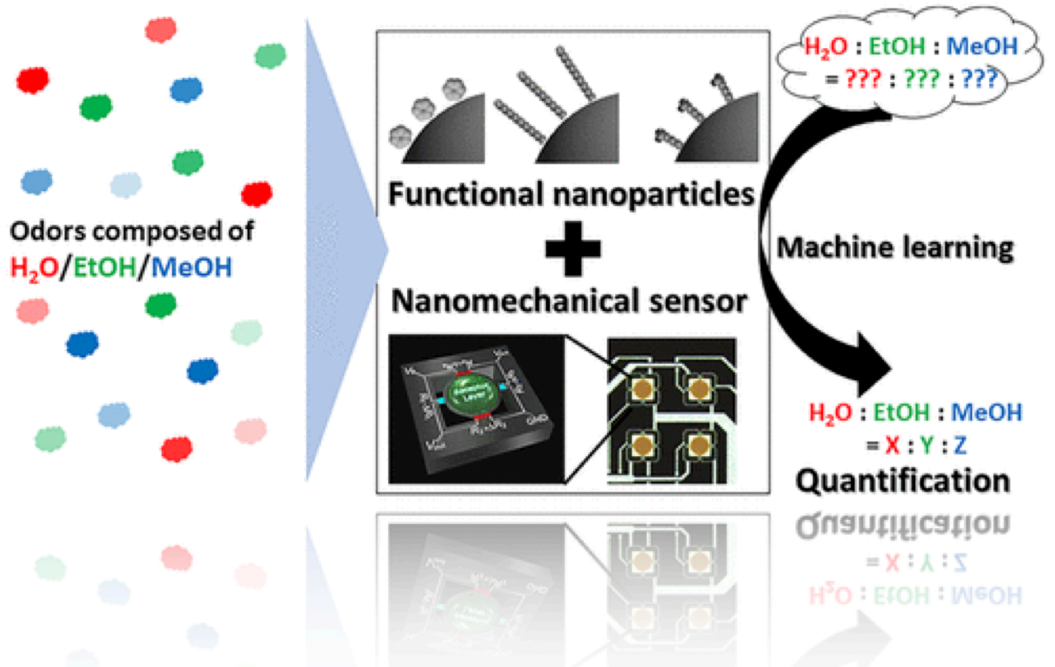


ACS Cent. Sci. 4, 1126 (2018)

Smells Sensor

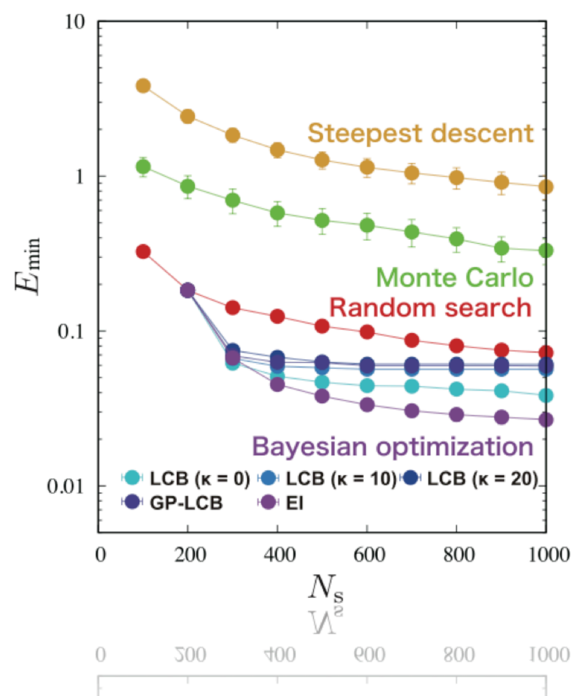


Sci. Rep. 7, 3661 (2017)



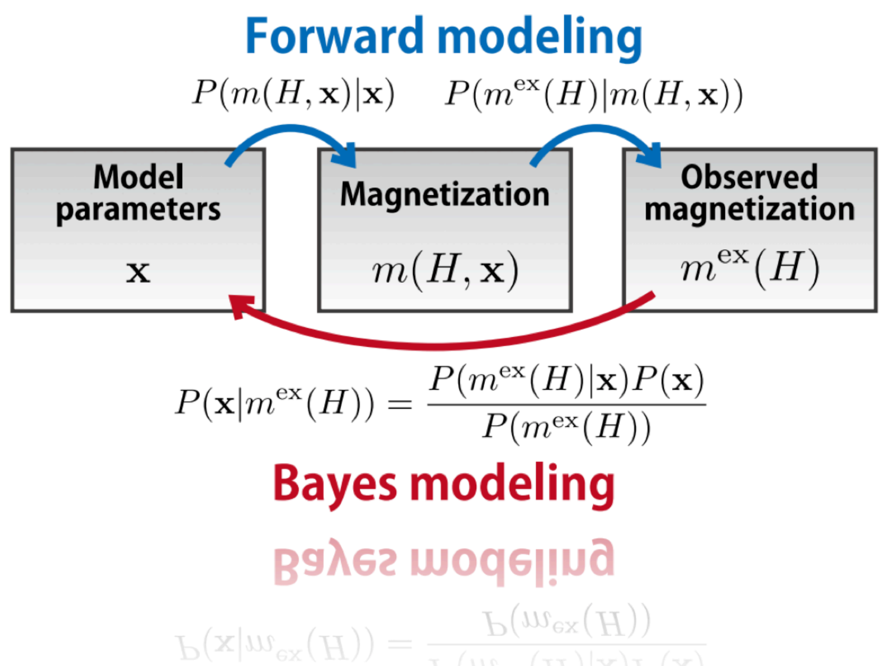
ACS Sensors 3, 1592 (2018)

Bayesian Opt.



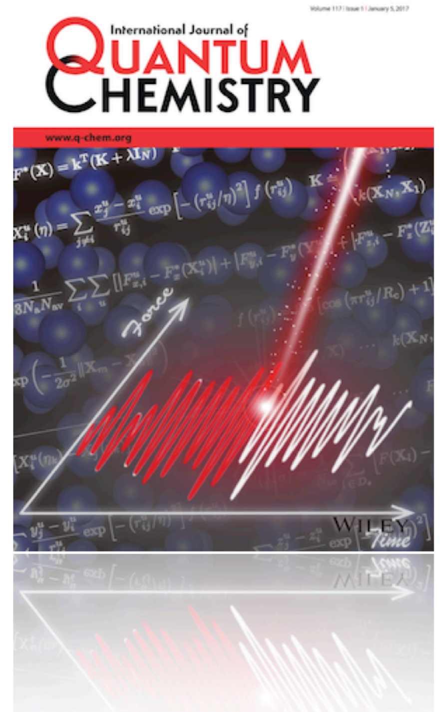
PLoS ONE 13, e0193785 (2018)

Magnet



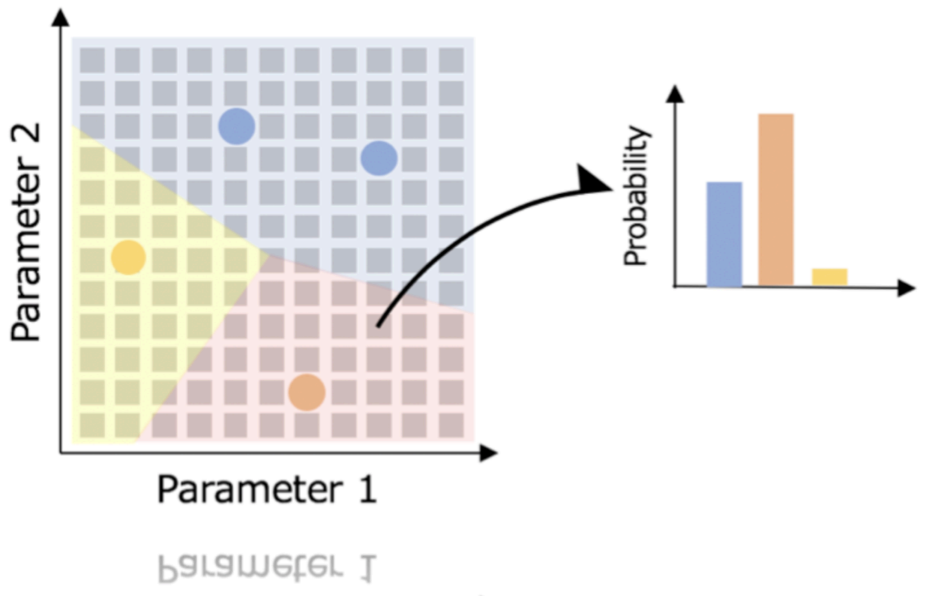
Phys. Rev. B 95, 064407 (2017)

Atomic Force



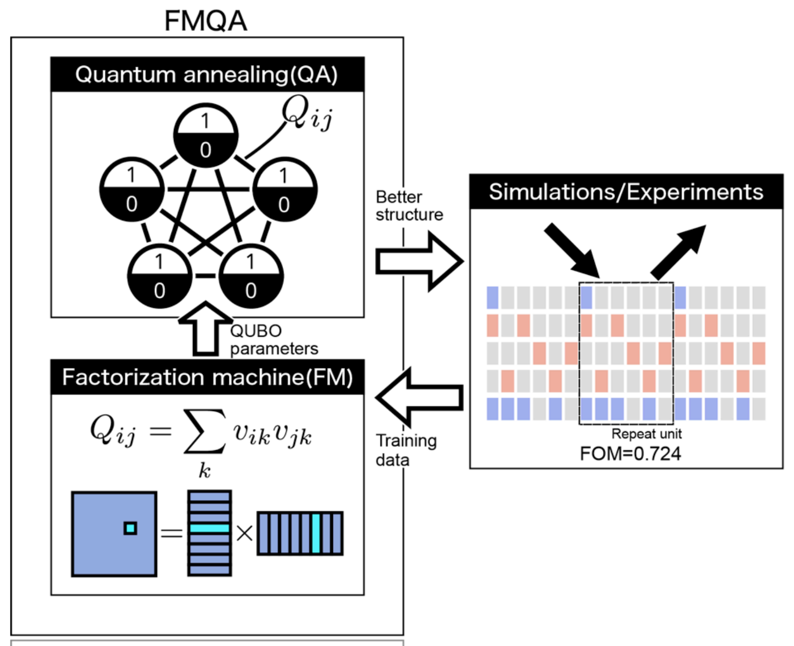
IJQC. 117, 33 (2017)
JPSJ. 88, 044601 (2019)

Phase Diagram



Phys. Rev. Mat. 3, 033802 (2019)

Metamaterial



arXiv: 1902.06573 (2019)