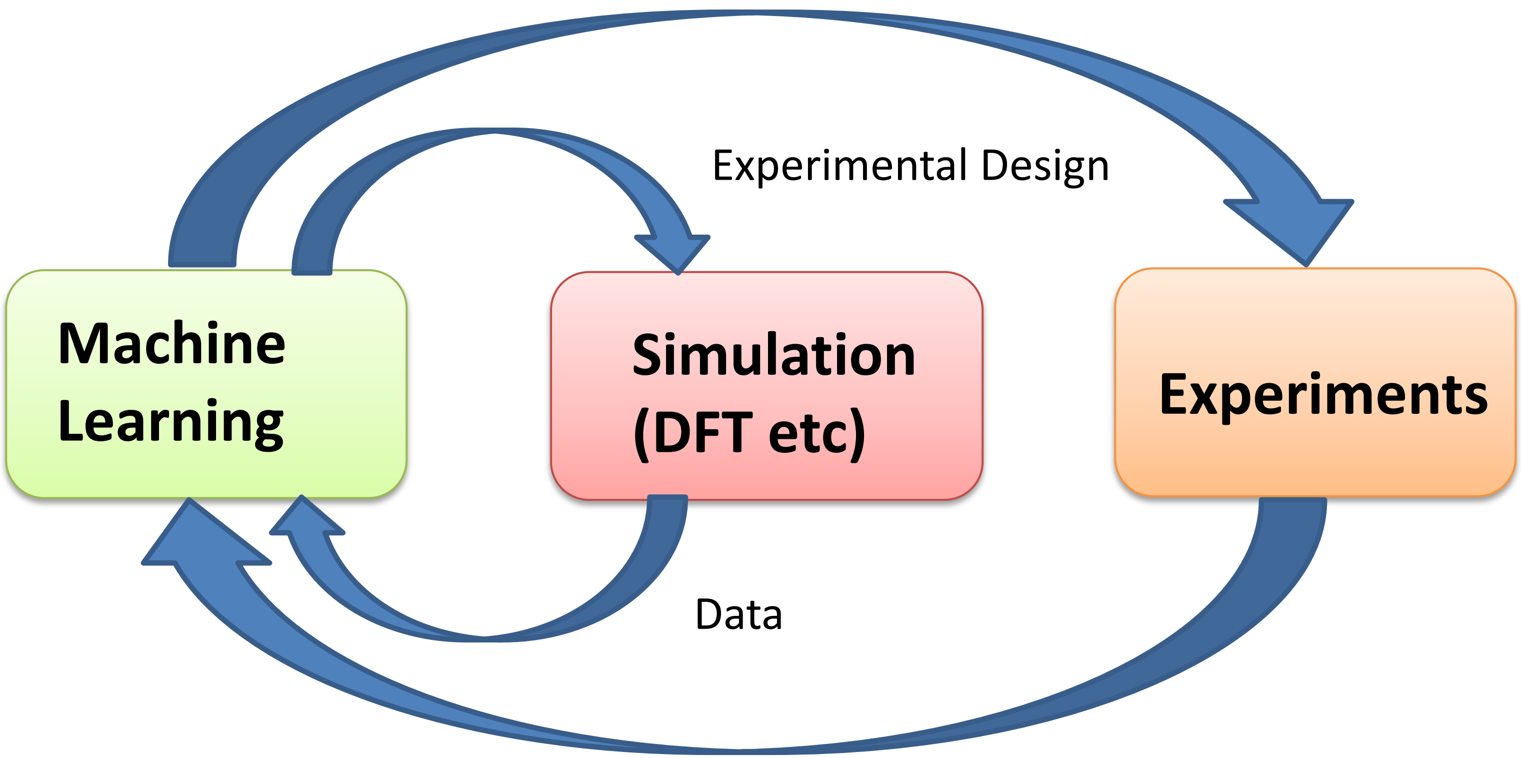
Designing materials with machine learning and quantum annealing

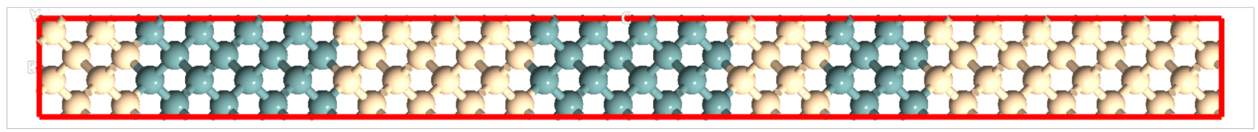
Koji Tsuda
University of Tokyo / NIMS / RIKEN

Automatic Materials Design

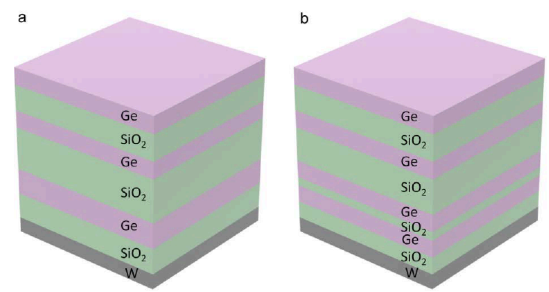


Agenda

- Bayesian Optimization
- Design of Si-Ge nanostructures (Ju+, PRX 2017)



- Wavelength selective thermal radiator (Sakurai+, ACS Cent Sci, 2019)



- D-wave quantum annealer (Kitai+, Arxiv, 2019)

Bayesian Optimization
(Jones et al., 1998)

- Find best data points with minimum number of observations

- Choose next point to observe to discover the best ones as early as possible

Screening by first principles calculations alone

Mat. 1	Mat. 2	Mat. 3	Mat. 4	Mat. 5	Mat. 6	Mat. 7	Mat. 8	Mat. 9	Mat. 10

First Principles Calc.

Score 1	Score 2	Score 3	Score 4	Score 5	Score 6	Score 7	Score 8	Score 9	Score 10

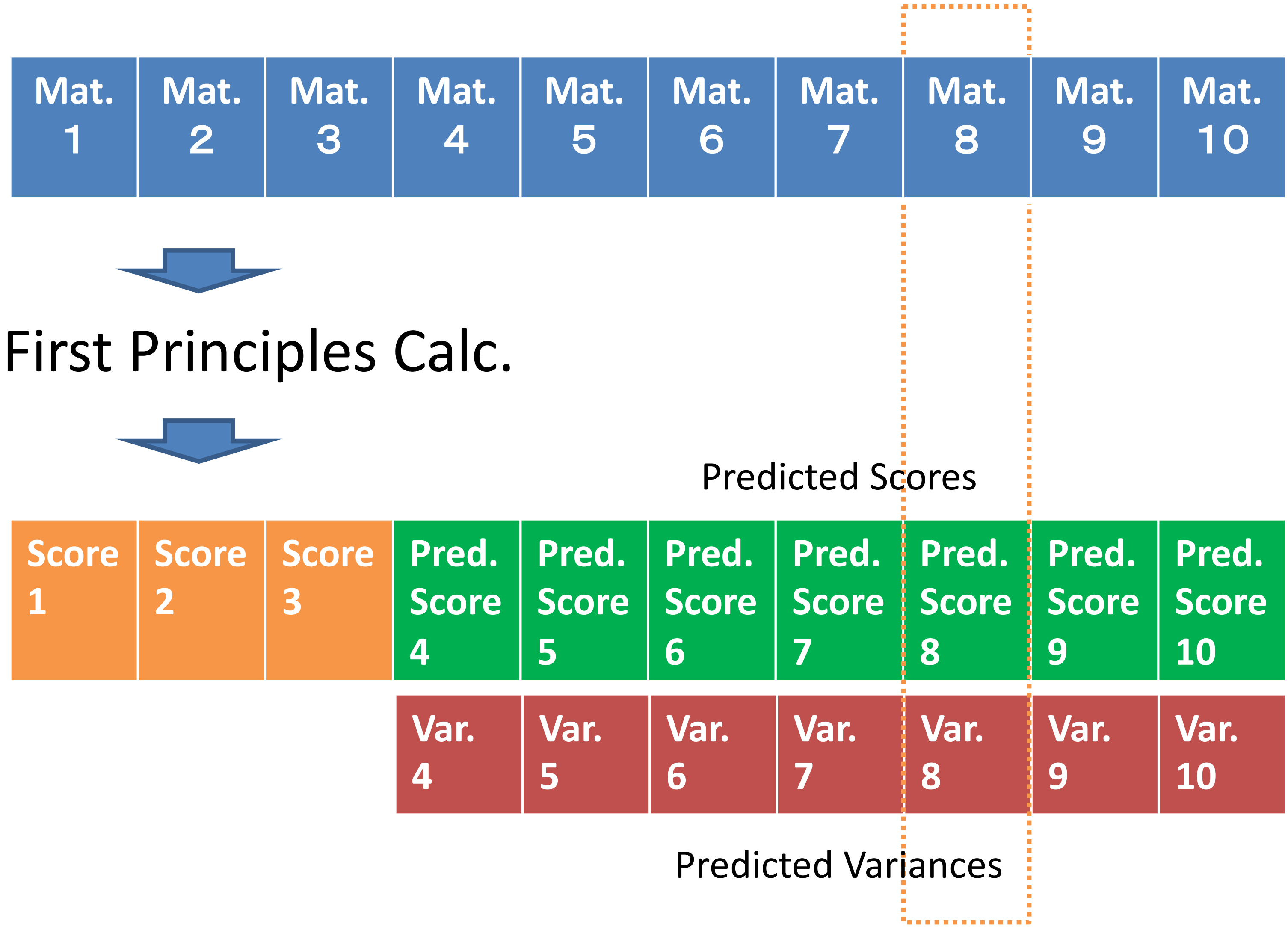
Bayesian Optimization (1)

Mat. 1	Mat. 2	Mat. 3	Mat. 4	Mat. 5	Mat. 6	Mat. 7	Mat. 8	Mat. 9	Mat. 10

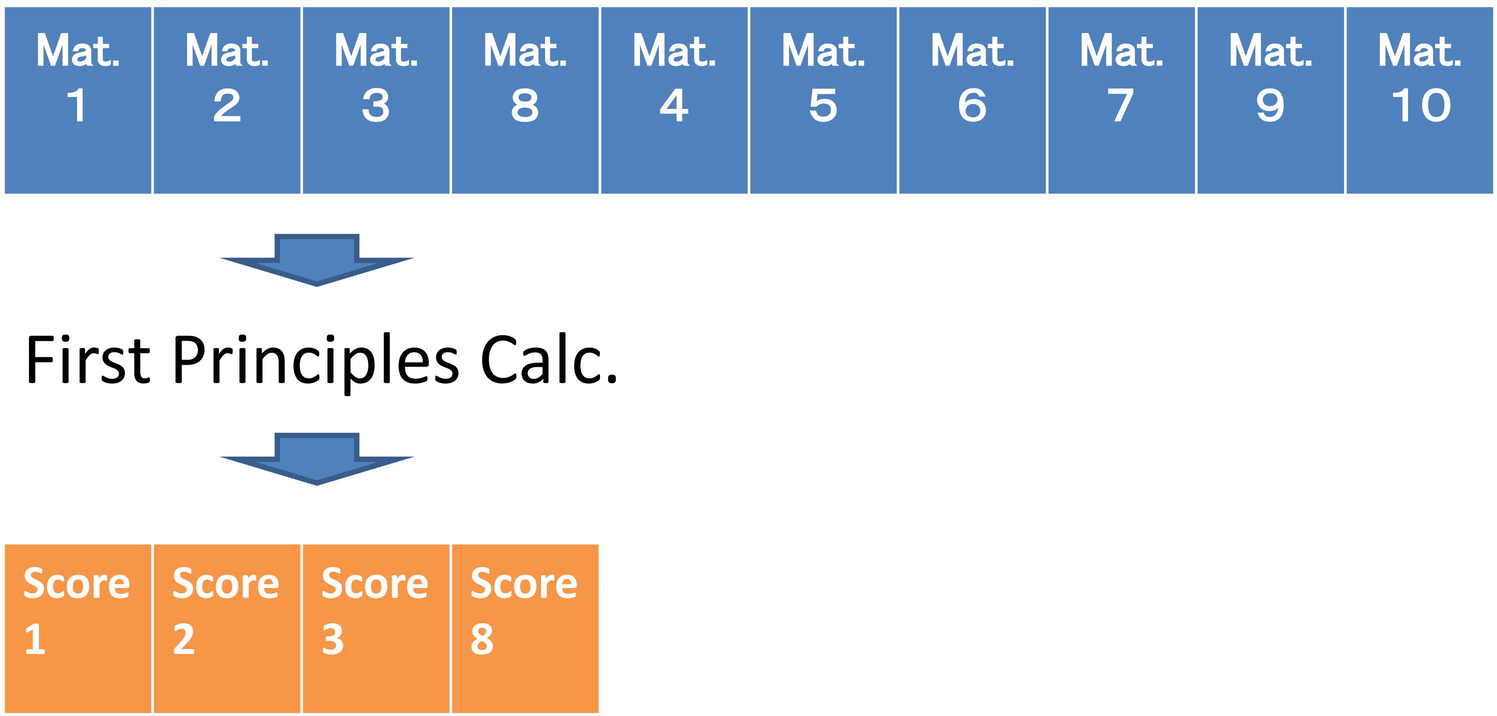
First Principles Calc.

Score 1	Score 2	Score 3

Bayesian Optimization (2)



Bayesian Optimization (3)



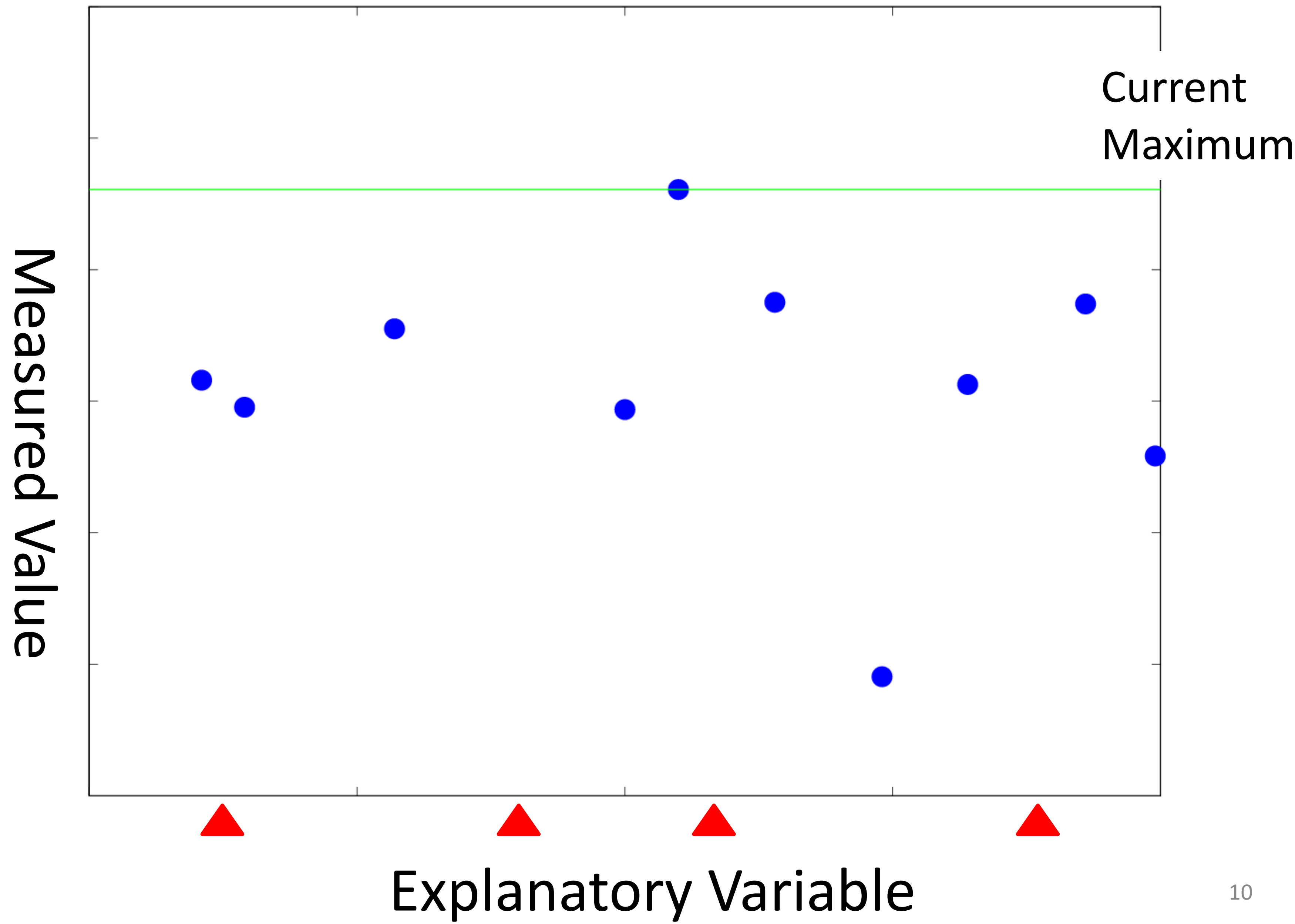
Bayesian Optimization (4)

Mat. 1	Mat. 2	Mat. 3	Mat. 8	Mat. 4	Mat. 5	Mat. 6	Mat. 7	Mat. 9	Mat. 10

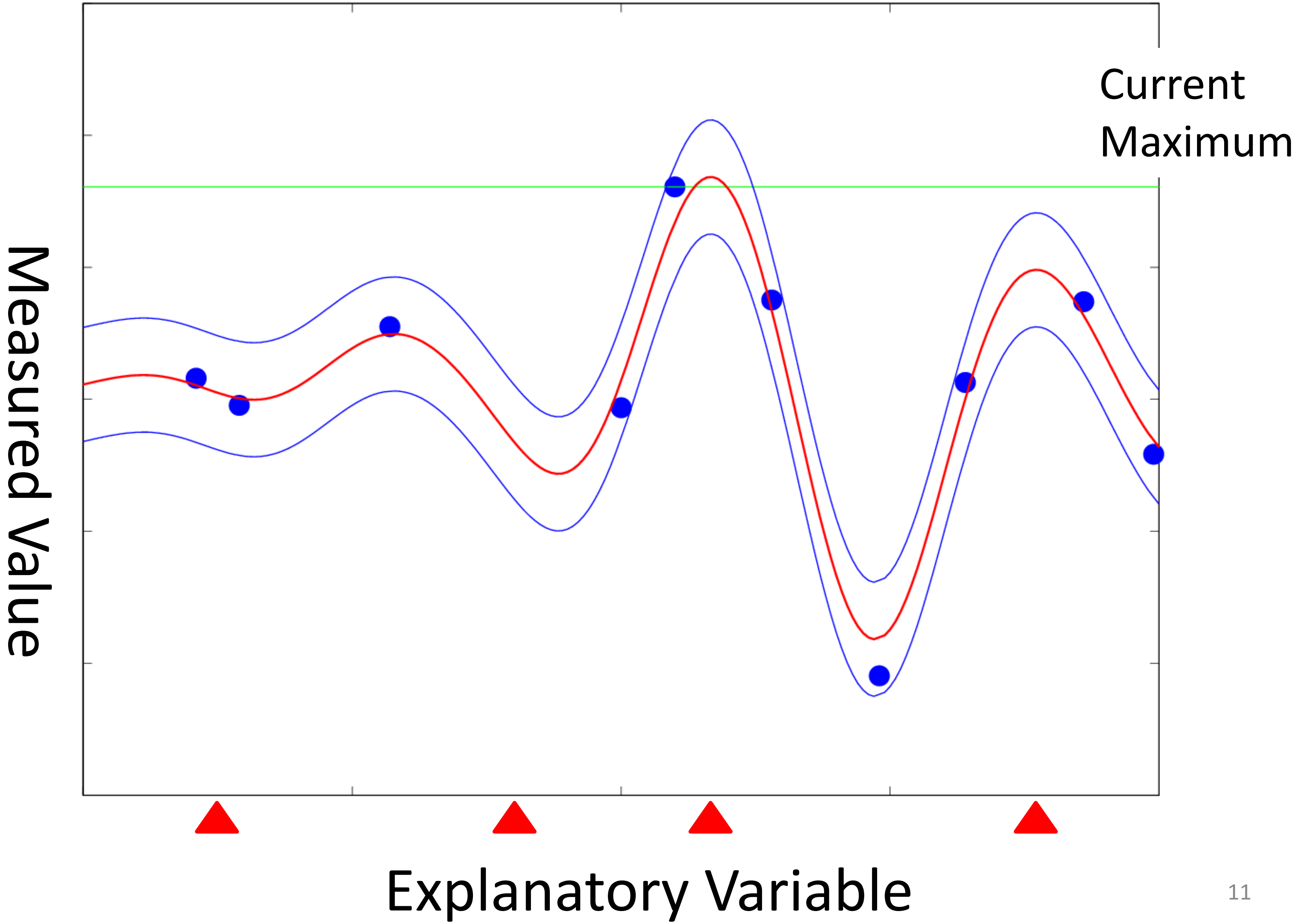
First Principles Calc.

Score 1	Score 2	Score 3	Score 8	Pred. Score 4	Pred. Score 5	Pred. Score 6	Pred. Score 7	Pred. Score 9	Pred. Score 10
				Var. 4	Var. 5	Var. 6	Var. 7	Var. 9	Var. 10

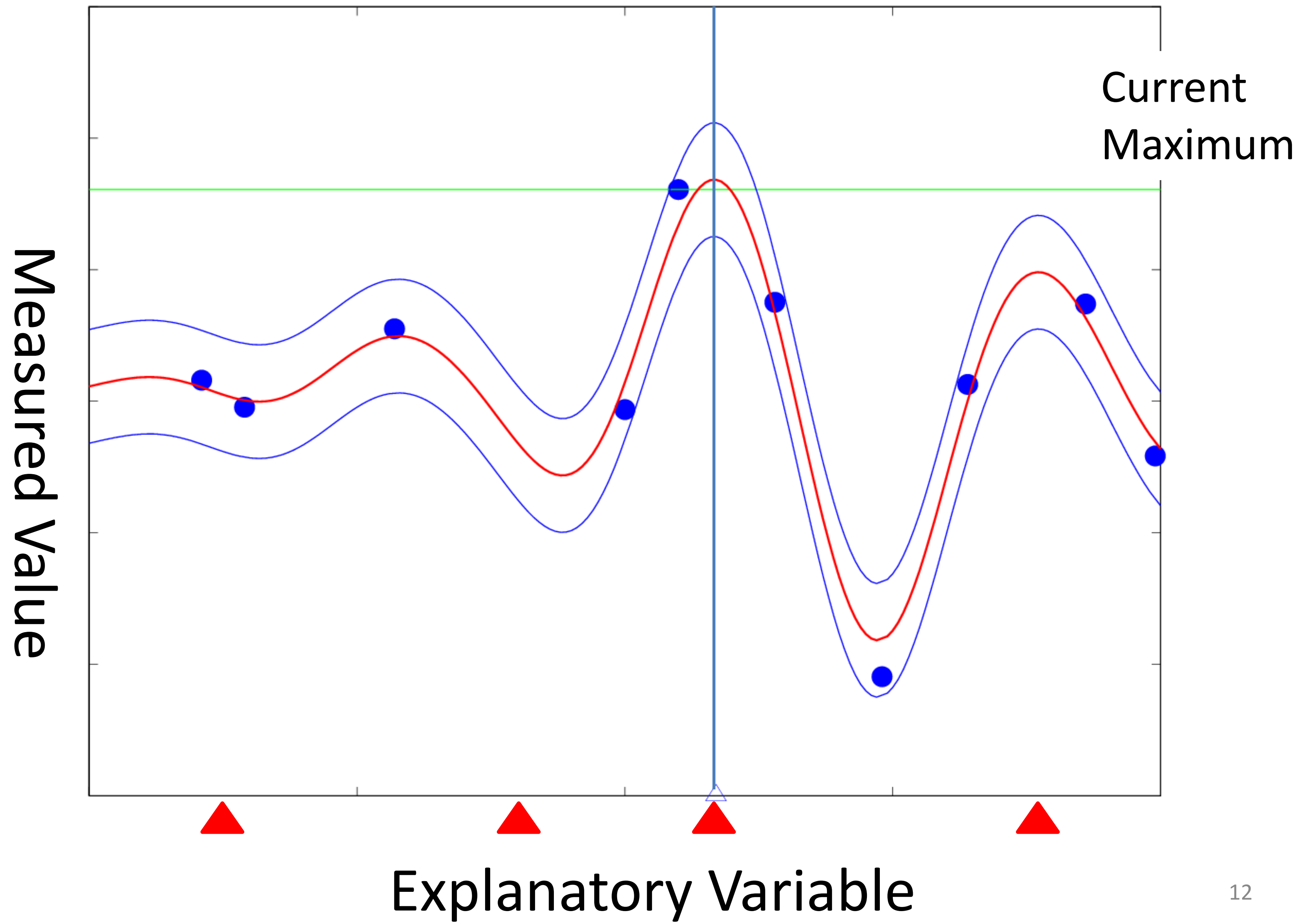
Where to observe next?



Gaussian Process

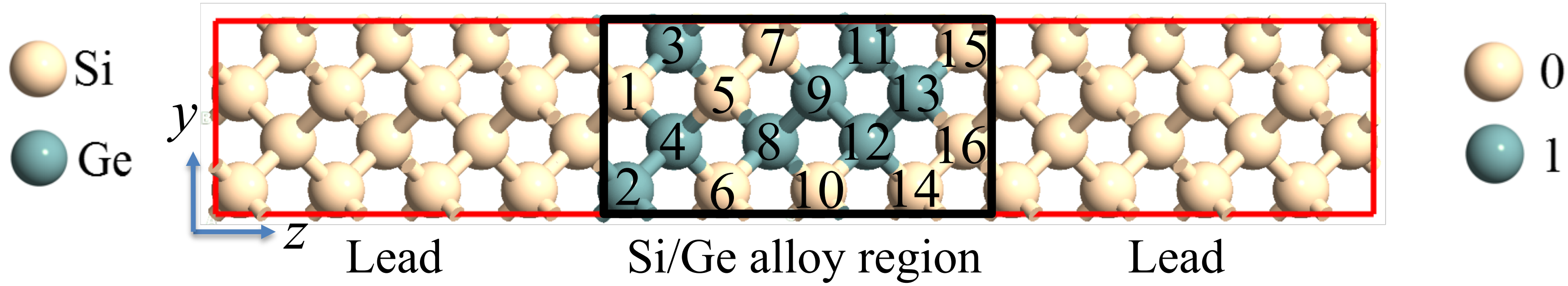


Maximum probability of improvement



Alloy Structure Optimization (Phys Rev X, 2017)

Question: How to organize 16 alloy atoms (Si: 8, Ge: 8) to obtain the largest and smallest interfacial thermal conductance?



Descriptors: $C_{16}^{8} = 12{,}870$

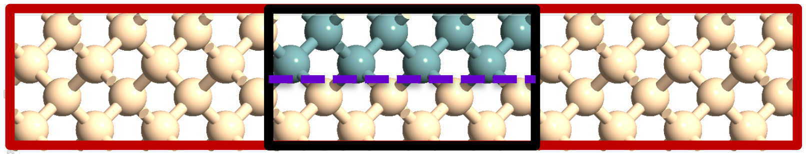
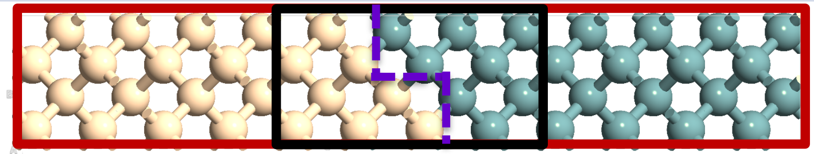
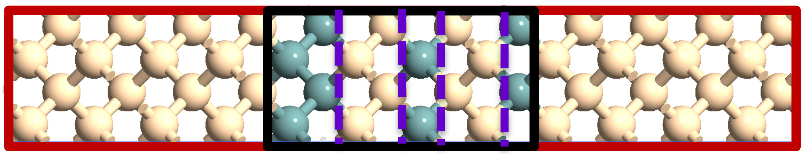
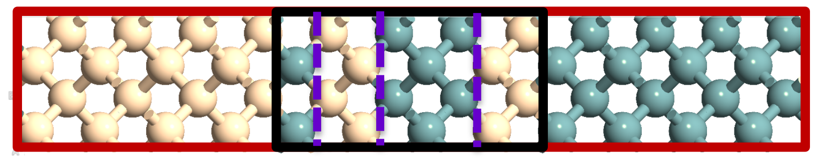
Case	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
1	1	1	1	1	1	1	1	1	0	0	0	0	0	0	0	0
2	1	1	1	1	1	1	1	0	1	0	0	0	0	0	0	0
3	1	1	1	1	1	1	1	0	0	1	0	0	0	0	0	0
…	…	…	…	…	…	…	…	…	…	…	…	…	…	…	…	…

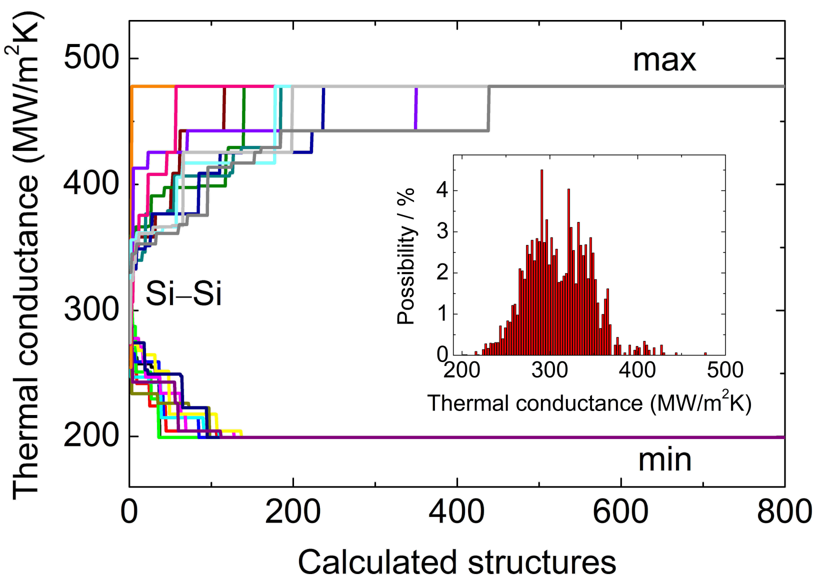
Calculator: Atomistic Green's Function (AGF): Phonon transmission

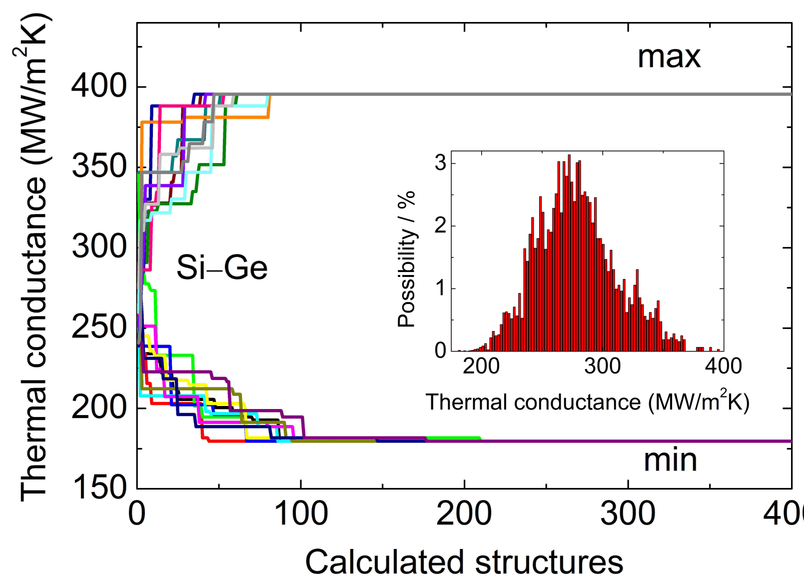
Evaluator: Interfacial Thermal Conductance (ITC)

Optimization method: Thompson Sampling (Bayesian Optimization)

Alloy Structure Optimization

ITC	Si-Si	Si-Ge
Max		
Min		

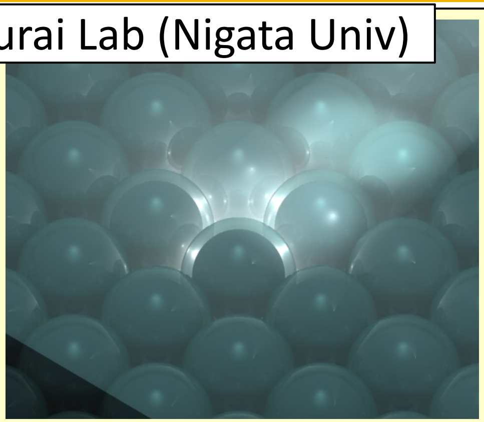




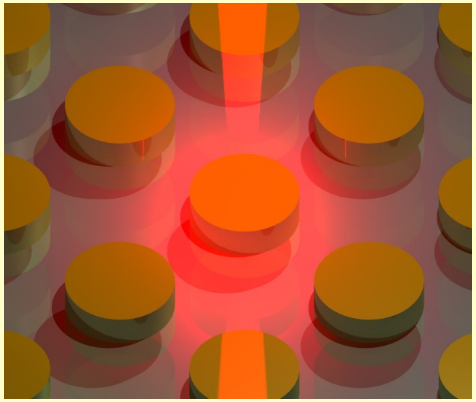
Optimal structures were obtained by calculating only 3.4% of all candidates.

Wavelength selective thermal radiator

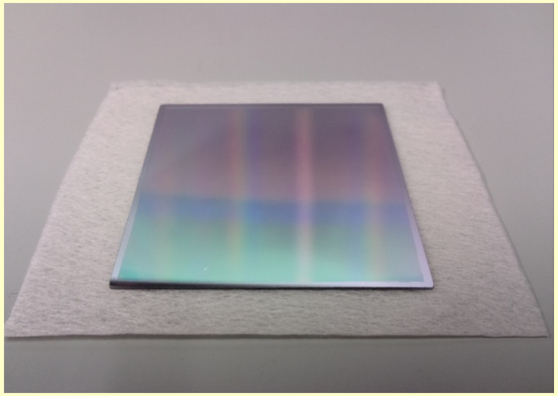
Sakurai Lab (Nigata Univ)



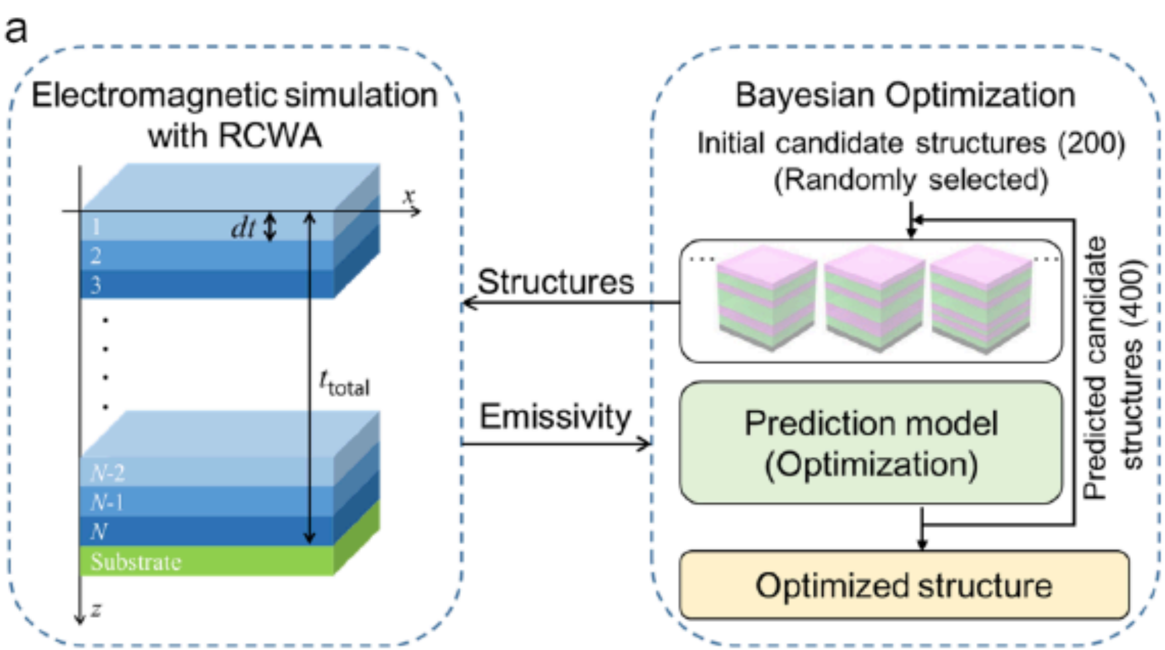
Solar absorber

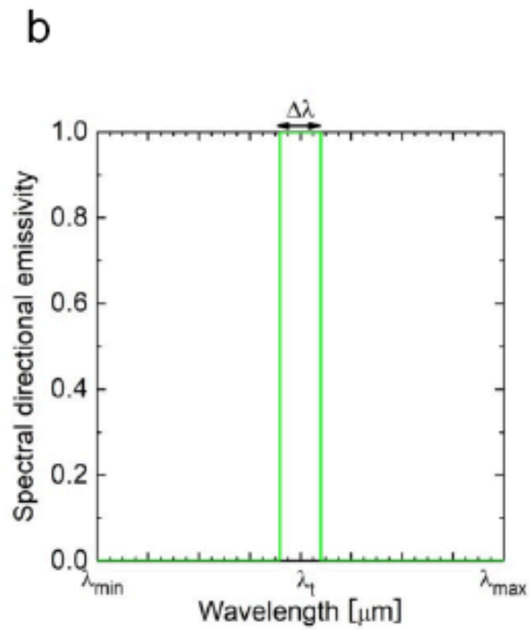


Sky radiator



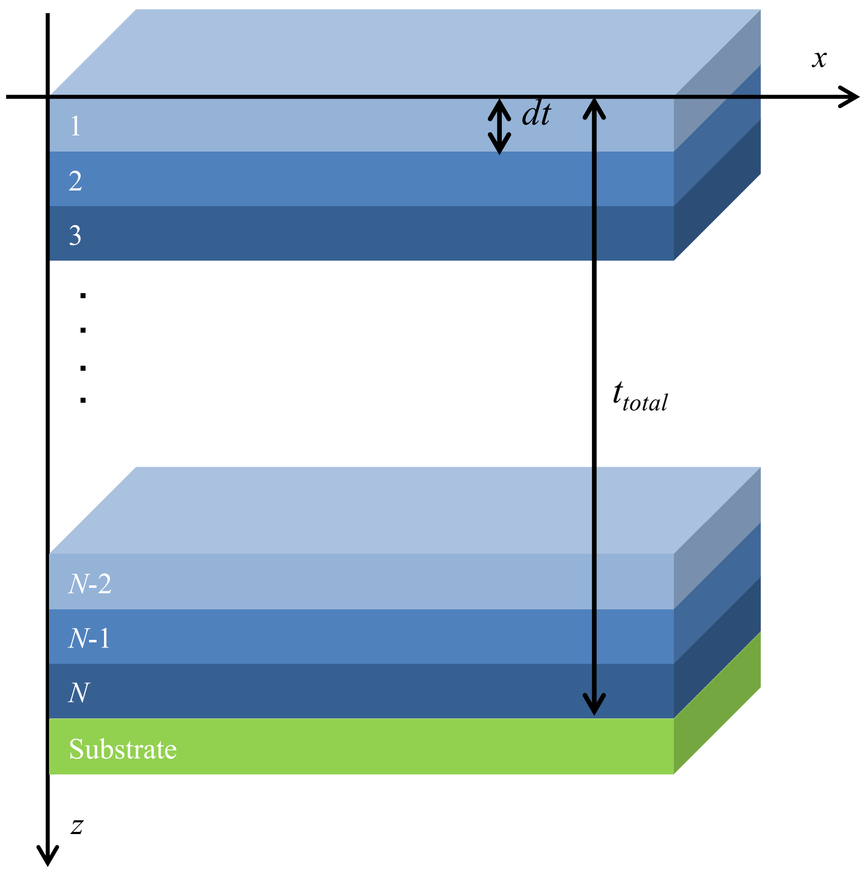
Heater for drying





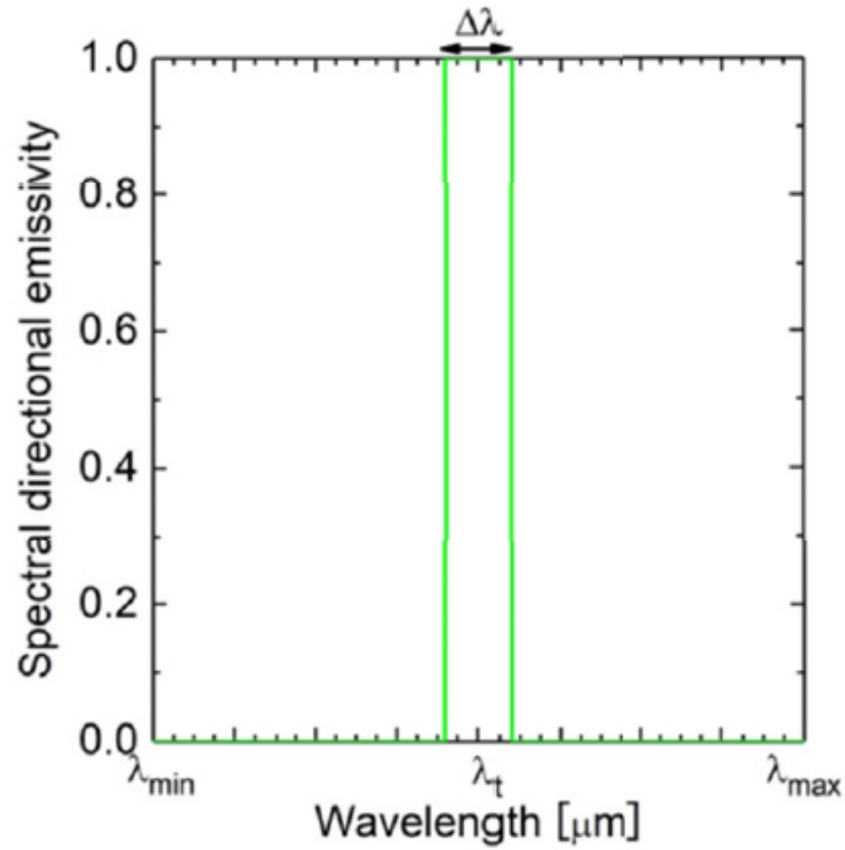
Designing layered material

- 18 layers: Ge, Si or SiO_2
- Total thickness: 21 grid points between 3.6 µm and 4.0 µm
- Number of candidate structures: $3^{18} \times 21 =$ 8,135,830,269

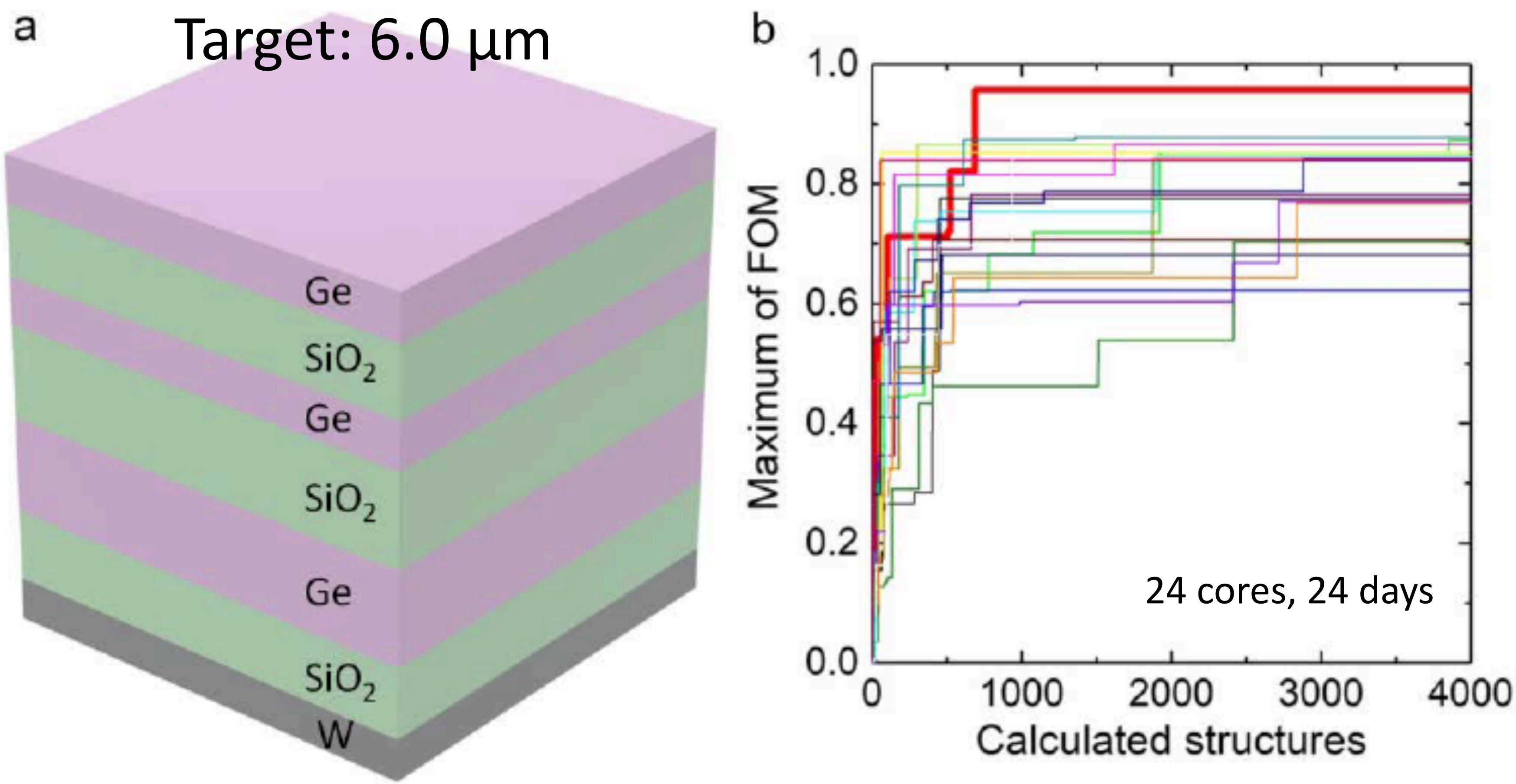


What to optimize

- Figure of Merit
  - Appreciates peaks near target, penalizes peaks outside
- Calculation of emissivity spectra
  - Electromagnetic simulation via transfer matrix method

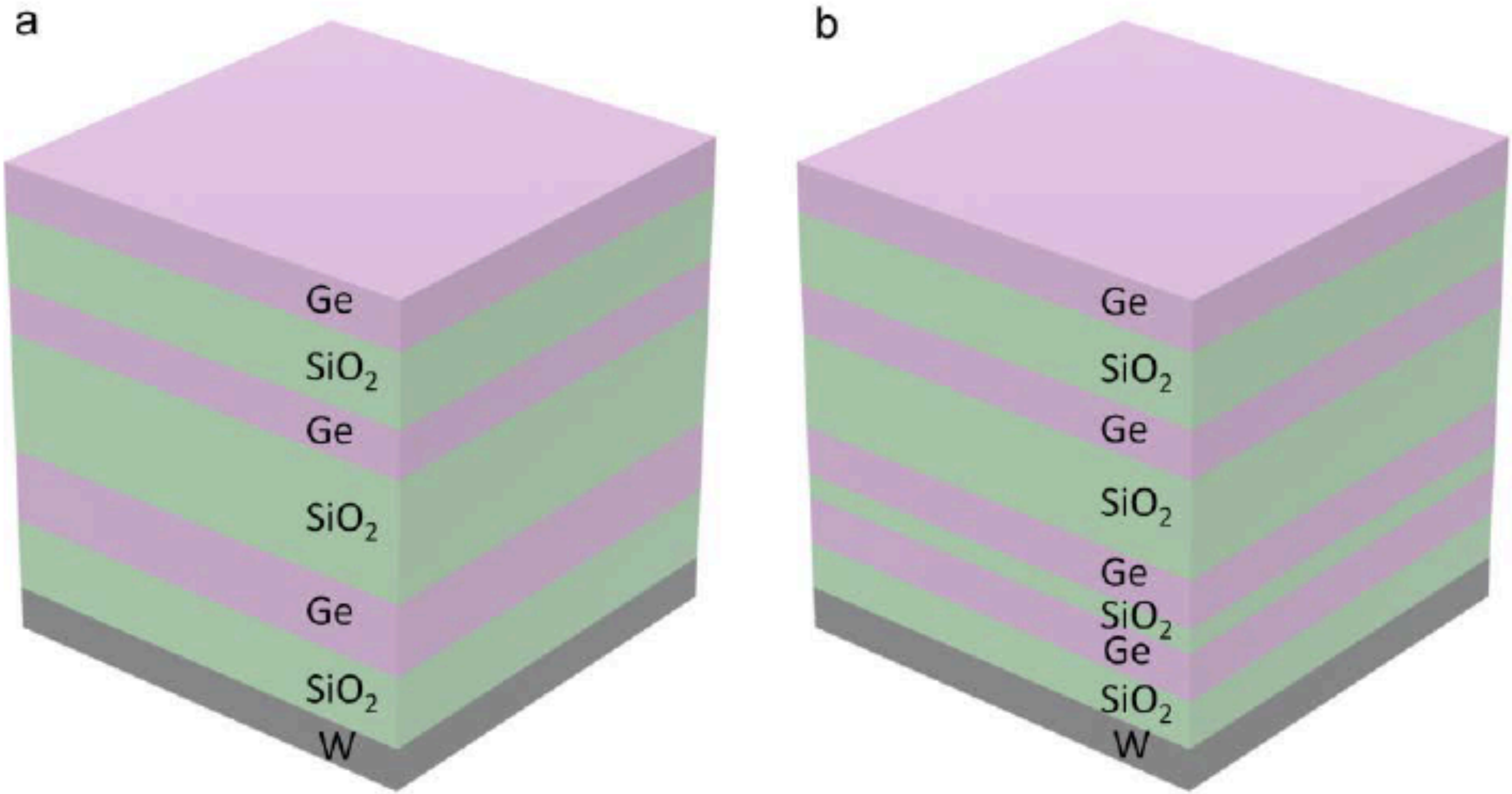


Optimal solution found with 168 million calculations on average (2.06% of all possibilities)



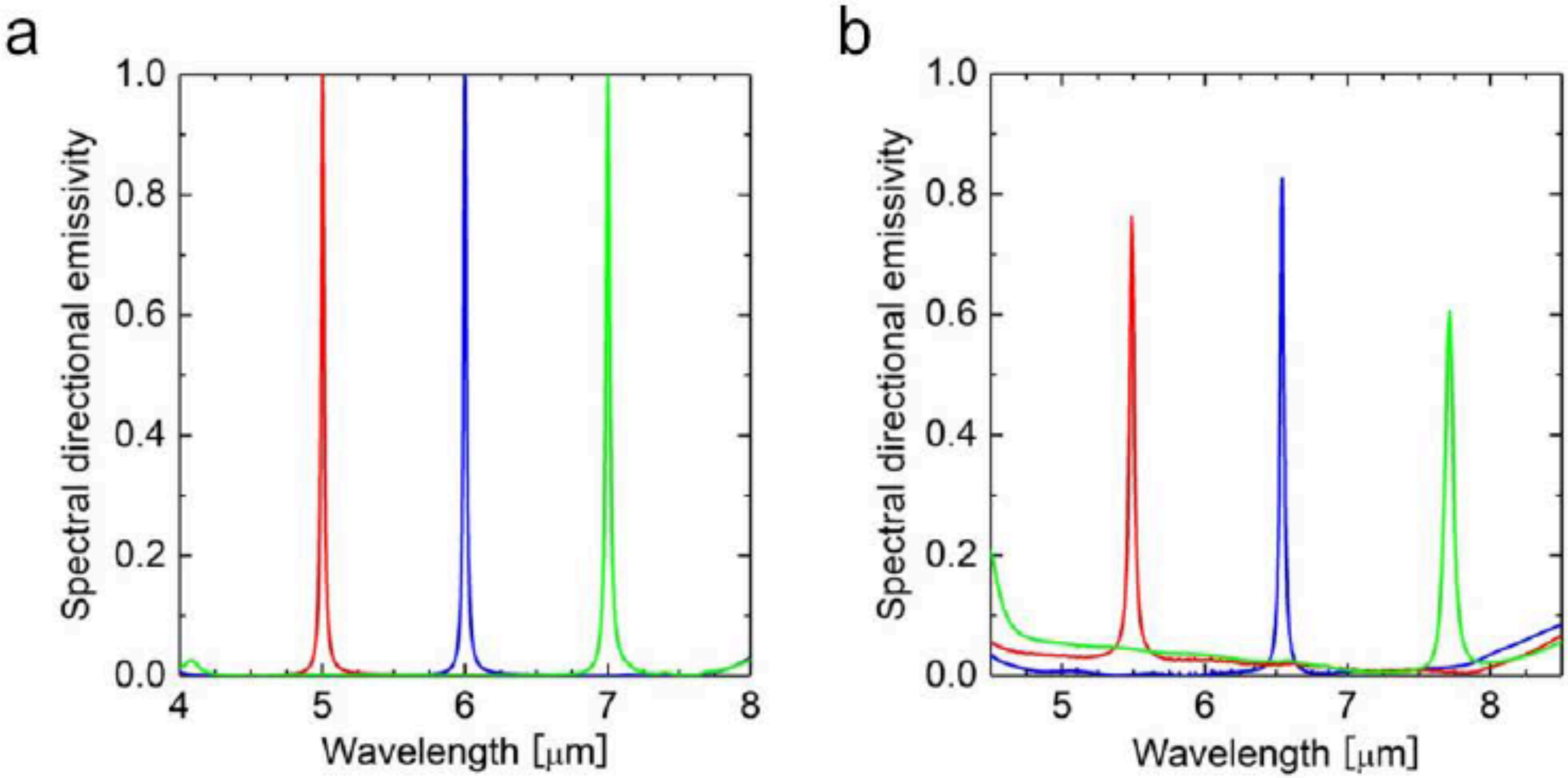
Target: 5.0 μm

Target: 7.0 μm



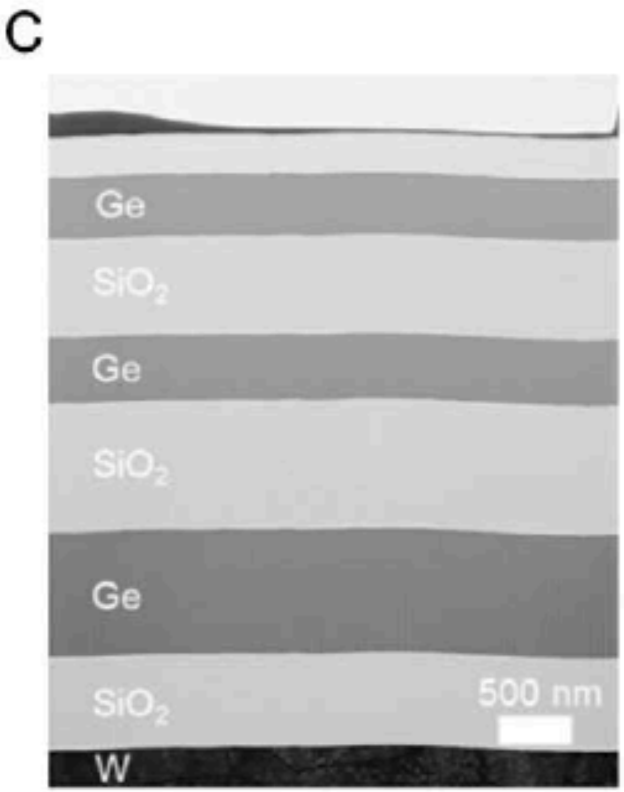
Calculated

Experimental Validation



Experimental Validation

TEM image



Layer Thickness

Target wavelengths, λ_t	5.0		6.0		7.0	
Layer No.	Sim.	Exp.	Sim.	Exp.	Sim.	Exp.
1	0.42	0.42	0.42	0.43	0.44	0.44
2	0.63	0.61	0.63	0.69	0.66	0.62
3	0.42	0.43	0.42	0.45	0.44	0.44
4	1.05	0.97	0.85	0.91	0.88	0.84
5	0.63	0.63	0.85	0.87	0.44	0.45
6	0.63	0.58	0.63	0.65	0.22	0.22
7	-	-	-	-	0.44	0.44
8	-	-	-	-	0.44	0.41

Comparison with Existing Materials

- Q-factor: Peak sharpness
- Our material: Q=273 (Simulation), Q= 188 (Realized)
- Highest known Q-factor: 200 (2D grating coupled surface phonon polaritons, 2008)
  - Large unwanted peaks: Poor FOM = 0.02
  - High cost for nanofabrication

Quantum annealing

- Solves quadratic unconstrained binary optimization (QUBO)

$$\min_{z \in \{-1,1\}^m} \sum_i h_i z_i + \sum_{i \leq j} g_{ij} z_i z_j .$$

- D-wave 2000Q
  - Implementation of quantum annealing with superconducting semiconductor
  - Annealing time 170µs, up to 64 bits
  - Machine in Canada, accessed via API from Japan

Principle of quantum annealing

- QUBO + transverse field term
- Qubit has distribution of up and down
- When measured, up or down appears
- First, strong transverse field is applied
  - [up,down] = [0.5,0.5] is the ground state
- Then transverse field is weakened slowly
  - Ground state slides to global optimum of QUBO

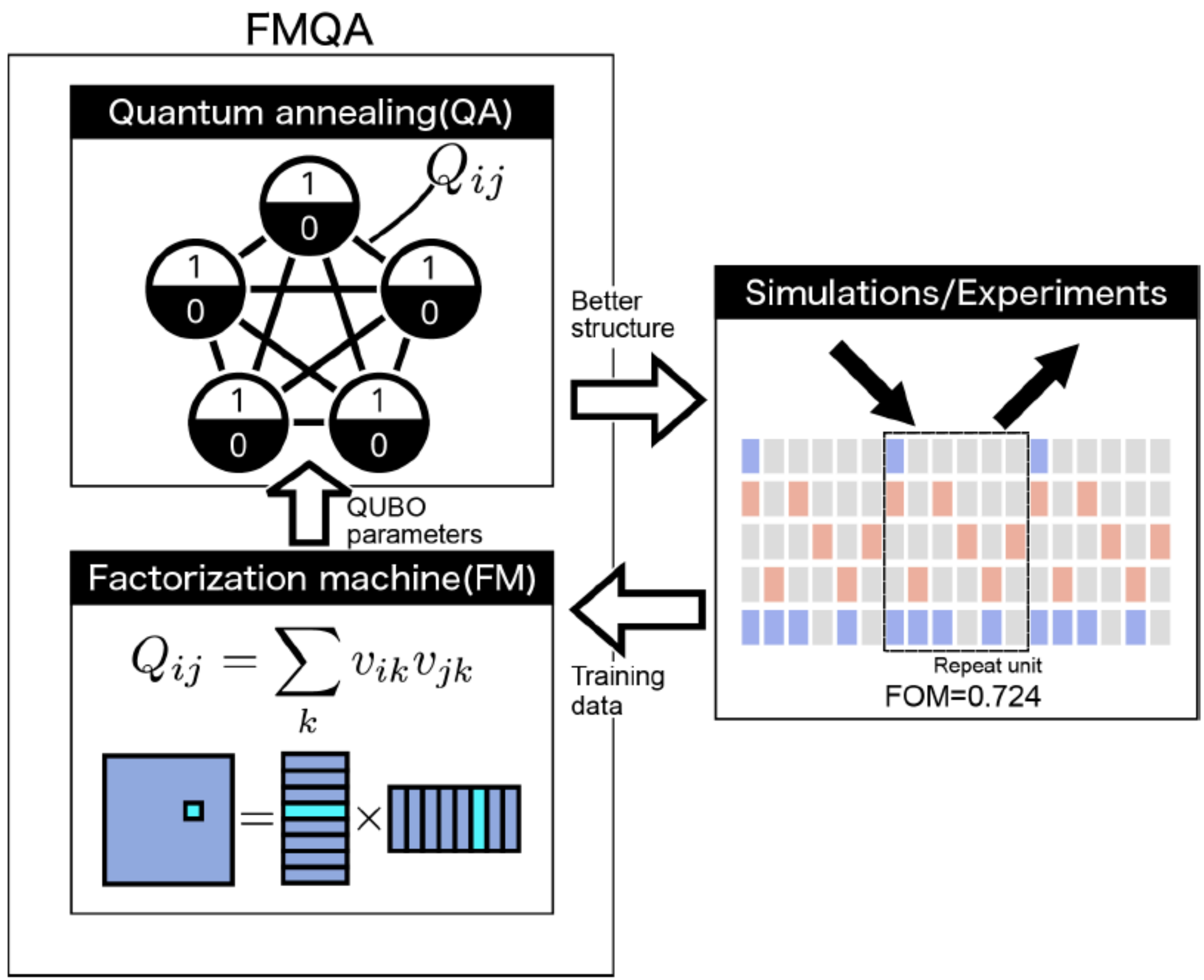
- *Conceptually similar to regularization path following (?)*

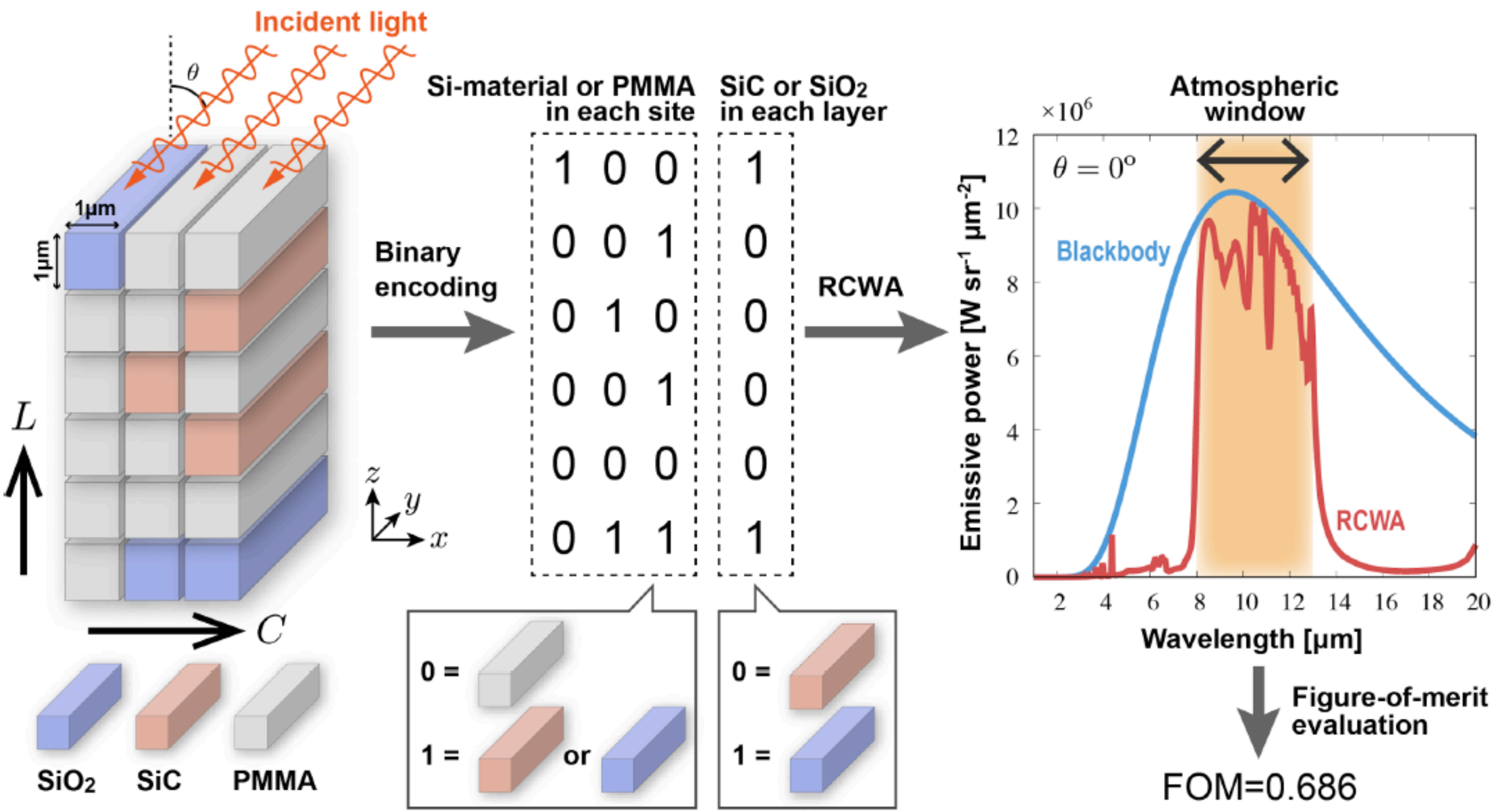
Using QA for black-box optimization

- GP's acquisition function is not QUBO (BAD!)
- Use factorization machine instead

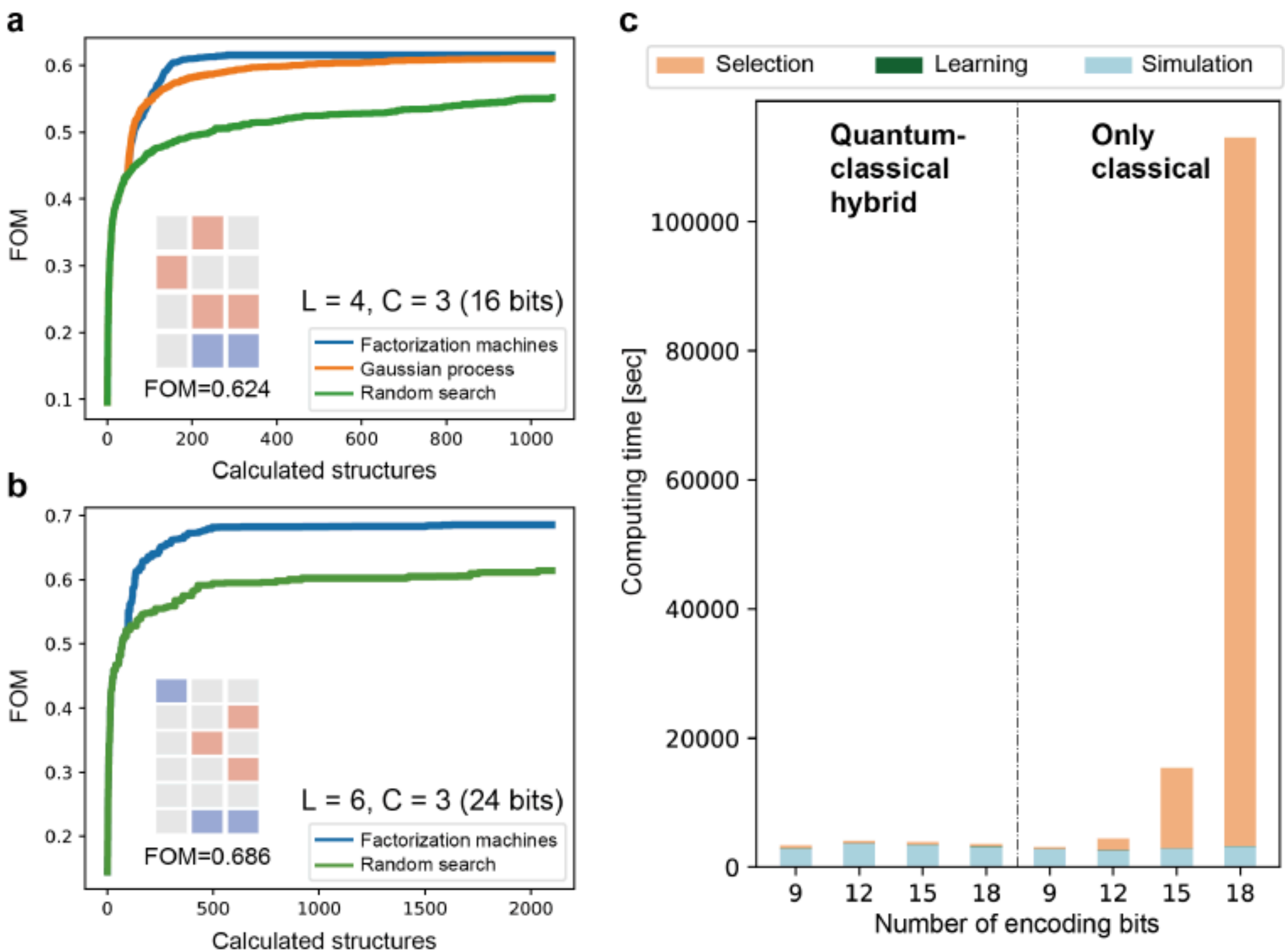
$$y(\mathbf{x}) = \sum_{i=1}^{N} w_i x_i + \sum_{i=1}^{N} \sum_{j=1}^{N} \sum_{k=1}^{K} v_{ik} v_{jk} x_i x_j,$$

- A learned model becomes QUBO
- 50 annealing at a time, select the best unseen solution

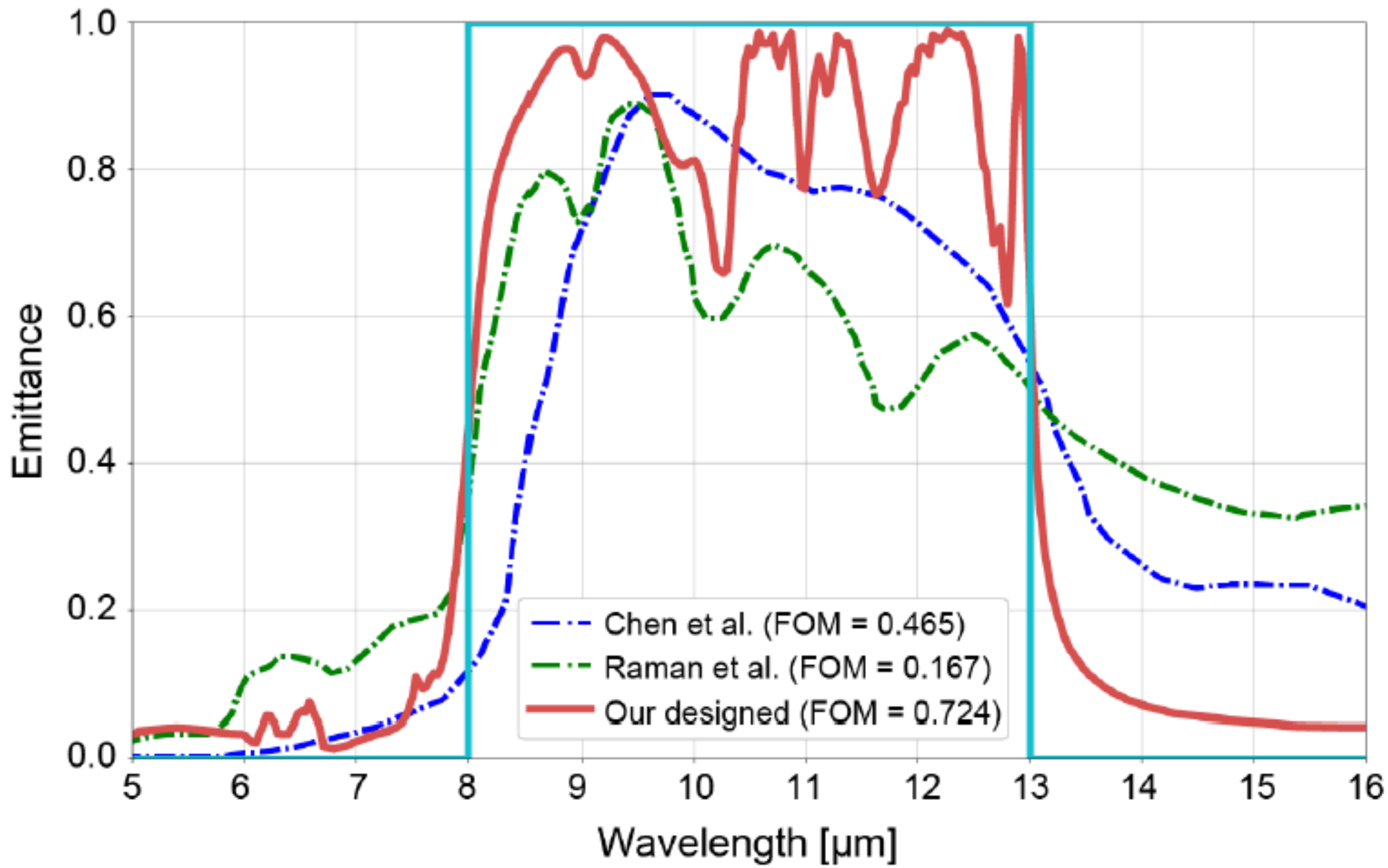

FMQA
Quantum annealing(QA)
Q_{ij}
1
0
QUBO parameters
Factorization machine(FM)
$Q_{ij} = \sum_k v_{ik} v_{jk}$
Better structure
Simulations/Experiments
Repeat unit
FOM=0.724
Training data



Incident light
θ
1μm
1μm
L
C
SiO2
SiC
PMMA
Binary encoding
Si-material or PMMA in each site
SiC or SiO2 in each layer
1 0 0
0 0 1
0 1 0
0 0 1
0 0 0
0 1 1
1
0
0
0
0
1
0 =
1 = or
0 =
1 =
z y x
RCWA
Atmospheric window
×10^6
θ = 0°
Blackbody
RCWA
Emissive power [W sr-1 μm-2]
Wavelength [μm]
Figure-of-merit evaluation
FOM=0.686


a
FOM
Calculated structures
L = 4, C = 3 (16 bits)
FOM=0.624
Factorization machines
Gaussian process
Random search
b
FOM
Calculated structures
L = 6, C = 3 (24 bits)
FOM=0.686
Factorization machines
Random search
c
Selection
Learning
Simulation
Quantum-classical hybrid
Only classical
Computing time [sec]
Number of encoding bits

Comparison to existing materials



Conclusion

- Designing complex materials is beyond ability of human intuition
- New “class” of materials enabled by ML & QA

- Tsuda Lab, UTokyo
  - Koki Kitai
  - Ryo Tamura
- Dept of Mech Eng, UTokyo
  - Junichiro Shiomi
  - Takuma Shiga
  - Shenghong Ju
  - Lei Fang
  - Jiang Guo
  - Makoto Kashiwagi
- Niigata Univ
  - Atsushi Sakurai
  - Kyohei Yada
  - Hideyuki Okada
  - Tetsushi Shimomura
- NIMS
  - Zhufeng Hou
  - Tadaaki Nagao
- Waseda Univ
  - Shu Tanaka