Supplementary Information

Argyrodite Configuration Determination for DFT and AIMD Calculations Using an Integrated Optimization Strategy†

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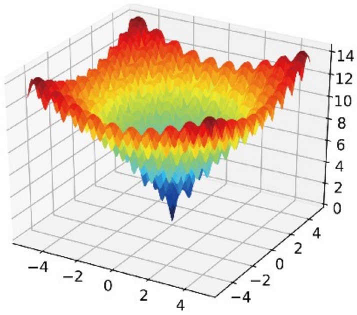
Ackley

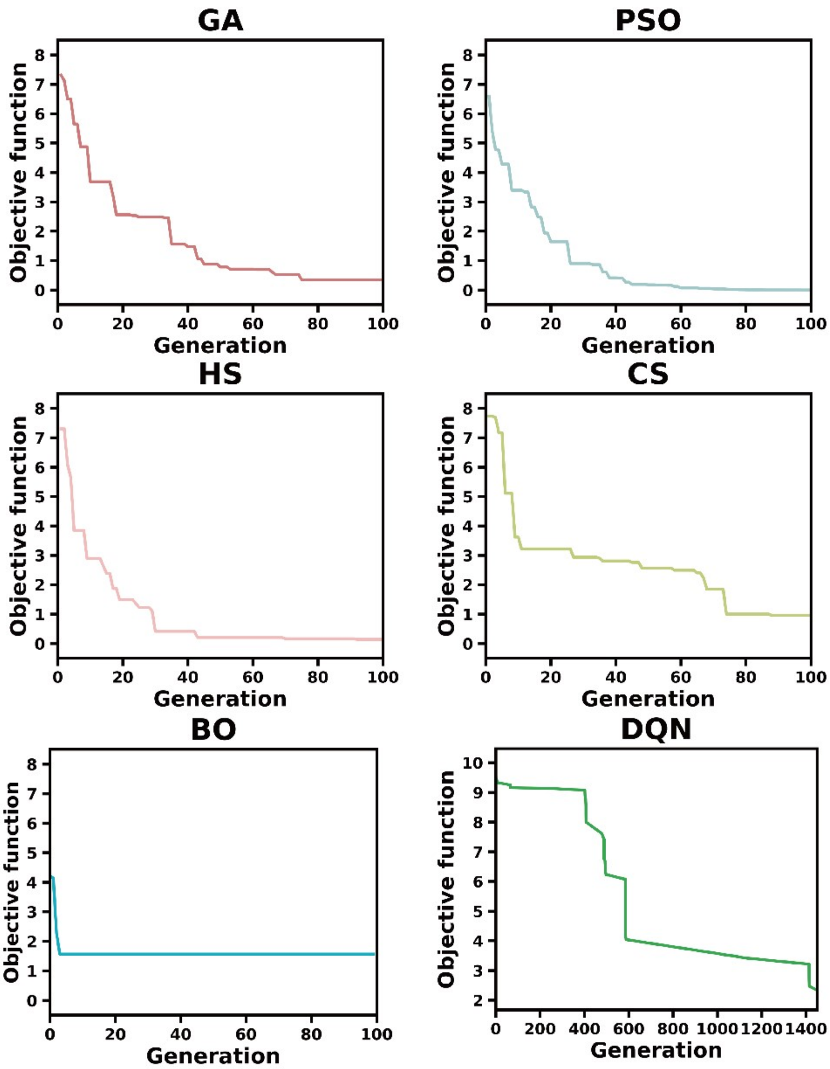
$$f(\mathrm{x}) = -a\exp\left(-b\sum_{i=1}^{d}x_i^2\right) - exp\left(\frac{1}{d}\sum_{i=1}^{d}\cos(cx_i)\right) + a + \exp(1)$$



Population Size = 25

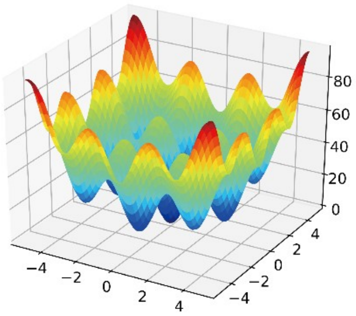
$-5 \leq (X_1, X_2 \cdot\cdot\cdot\cdot X_6) \leq 5$

$Y_{min} = 0$ at (0,0,0,0,0,0)

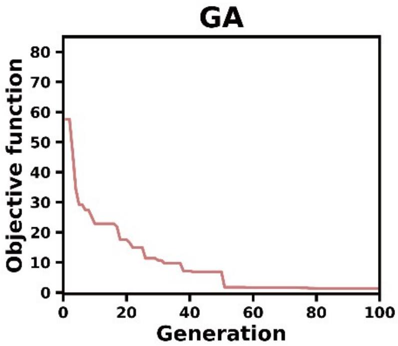


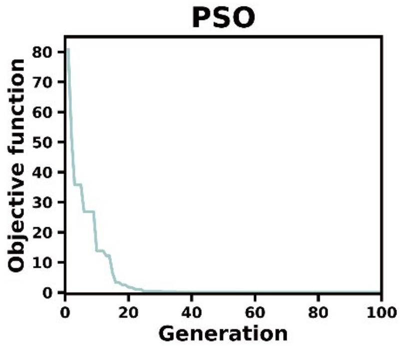
Egg Crate

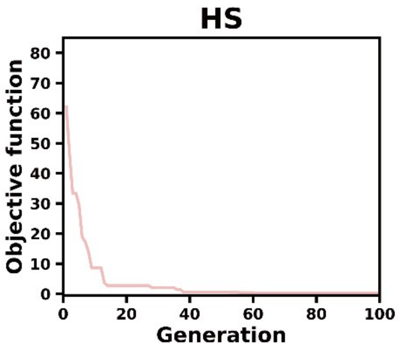
$$f(x) = \sum_{i=1}^{d} \left(x_i^2 + 25 \sin x_i^2 \right)$$

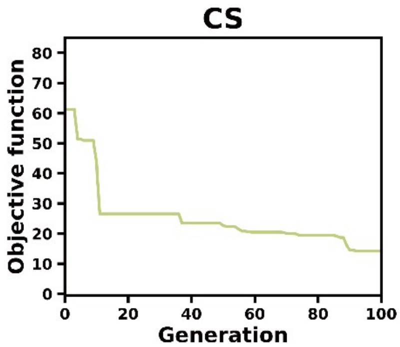


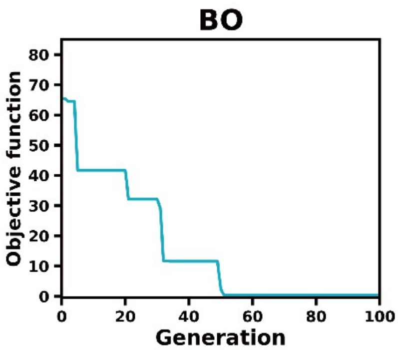
Population Size = 25
-5 ≤ (X_1, X_2 · · · · X_6) ≤ 5
Y_{min} = 0 at (0,0,0,0,0,0)

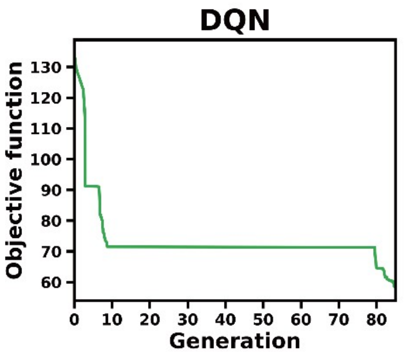






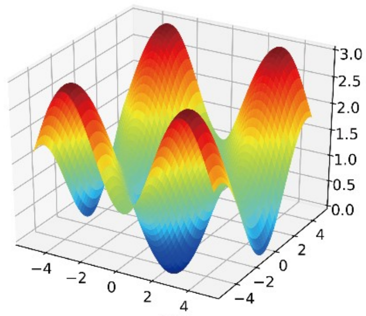






Griewank

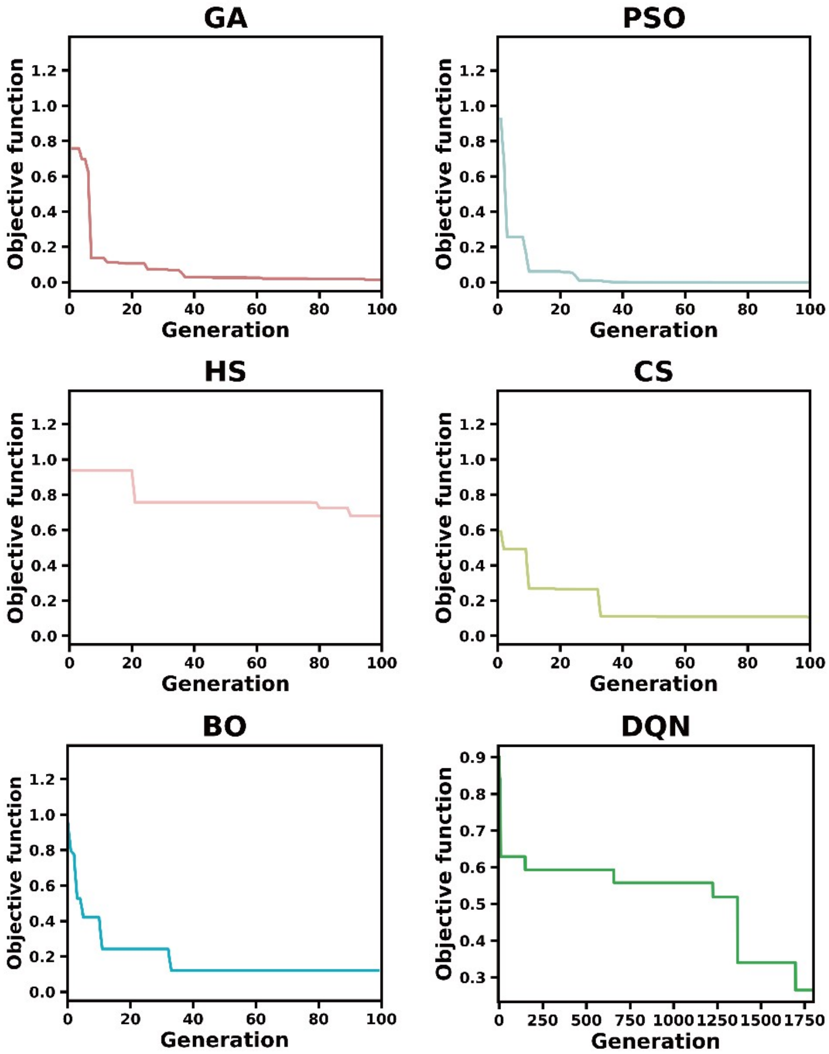
$$f(x) = \sum_{i=1}^{d} \frac{x_i^2}{4000} - \prod_{i=1}^{d} \left(\frac{x_i}{\sqrt{i}}\right) + 1$$



Population Size = 25

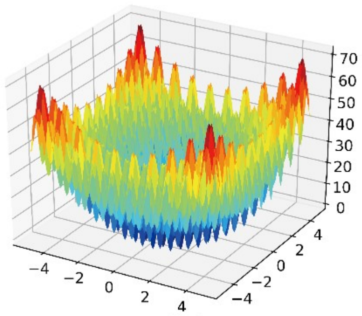
$-5 \le (X_1, X_2 \cdot \cdot \cdot \cdot X_6) \le 5$

$Y_{min} = 0$ at (0,0,0,0,0,0)



Rastrigin

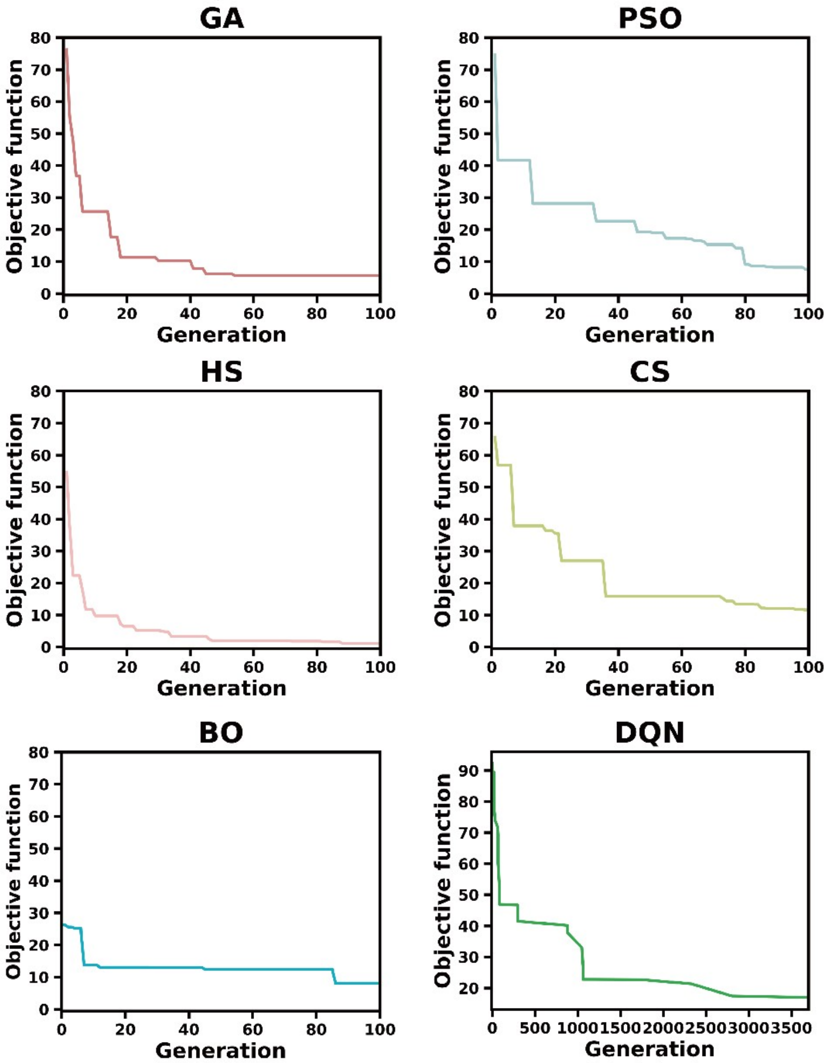
$$f(\mathrm{x}) = 10d + \sum_{i=1}^{d}\left[x_i^2 - 10\cos(2\pi x_i)\right]$$



Population Size = 25

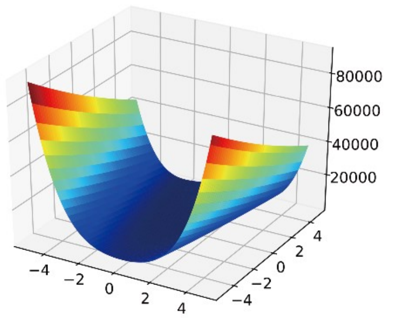
$-5 \leq (X_1, X_2 \cdot \cdot \cdot \cdot X_6) \leq 5$

$Y_{min} = 0$ at (0,0,0,0,0,0)



Rosenbrock

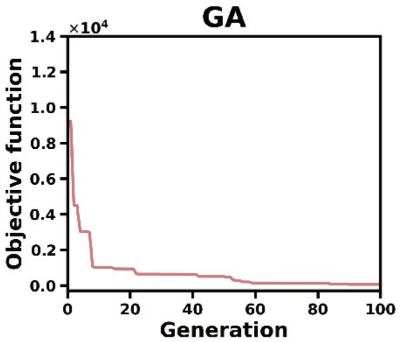
$$f(x) = \sum_{i=1}^{d-1}\left[100\left(x_{i+1} - x_i^2\right)^2 + (x_i - 1)^2\right]$$

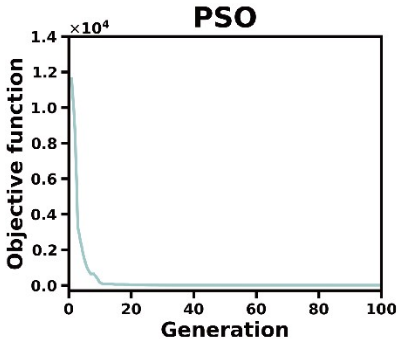


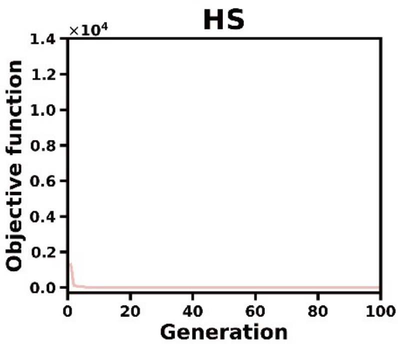
Population Size = 25

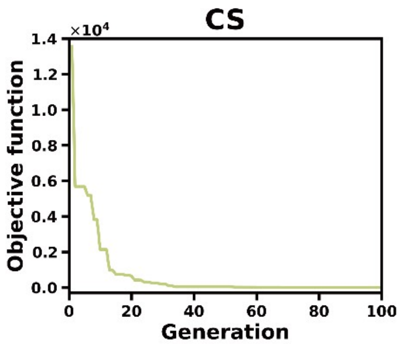
$-5 \leq (X_1, X_2 \cdot\cdot\cdot\cdot X_6) \leq 5$

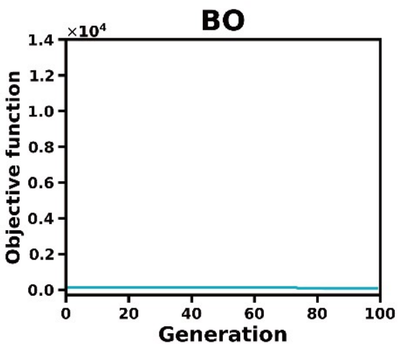
$Y_{min} = 0$ at (1,1,1,1,1,1)

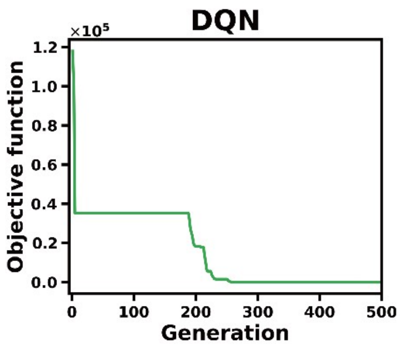












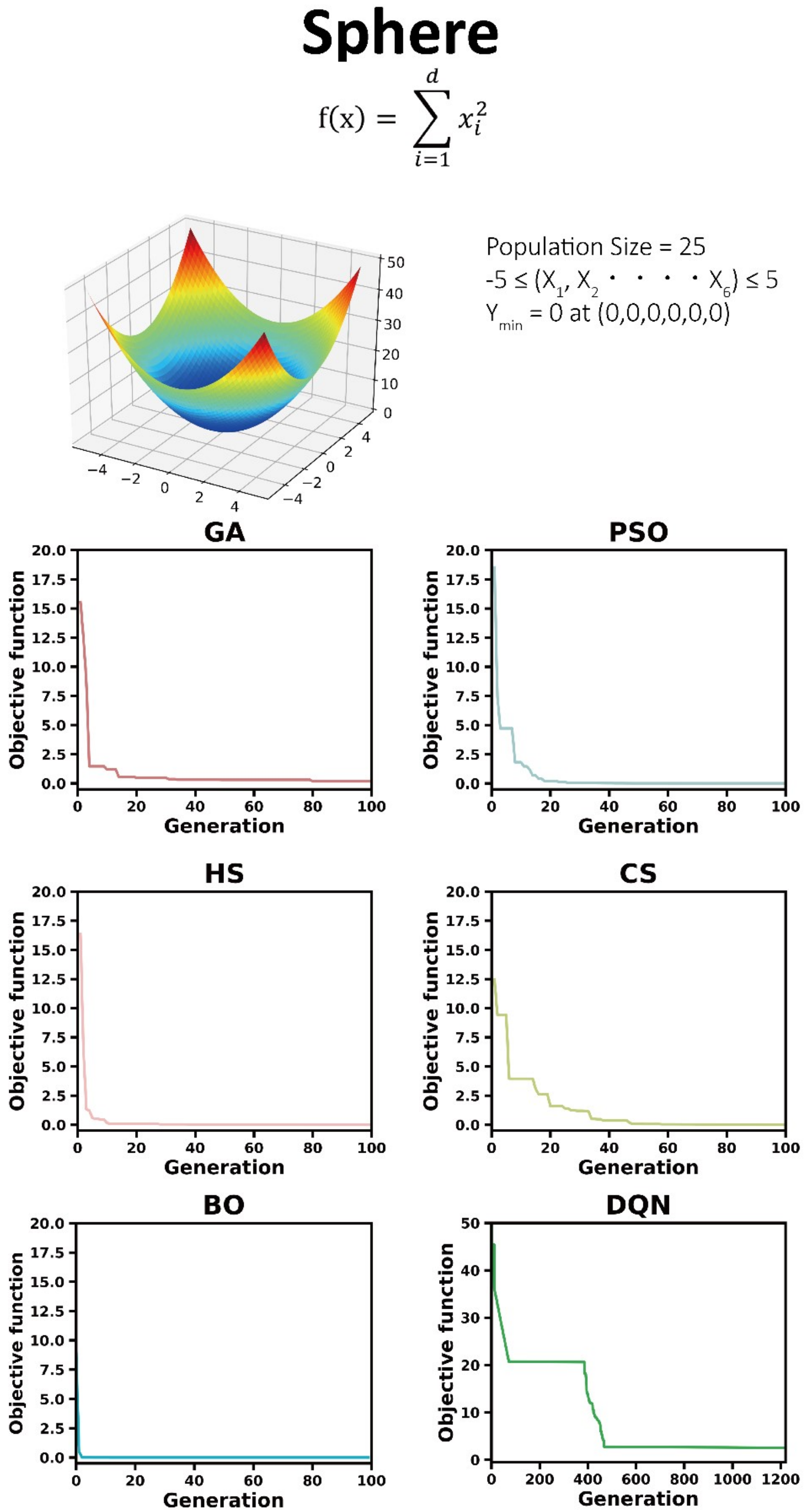
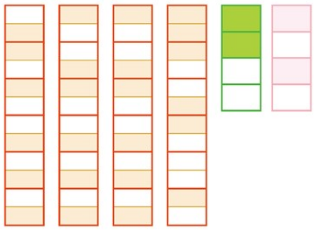


Fig. S1 Test result for all the optimization algorithms using several well-known benchmark test functions such as Ackley, Egg Crate, Griewank, Rastrigin, Rosenbrock, and Sphere functions, each of which is schematically described in the 2D scheme. The instaneous objective function value versus the generation number for each benchmark function.

Genetic Algorithm

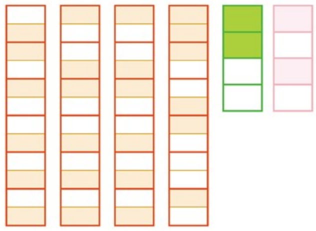
10th Gen.

E_c = -248.57 eV / f. u.



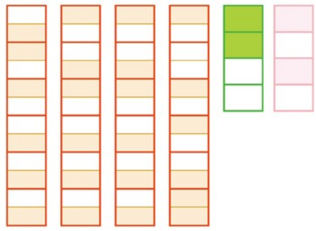
20th Gen.

E_c = -248.57 eV / f. u.



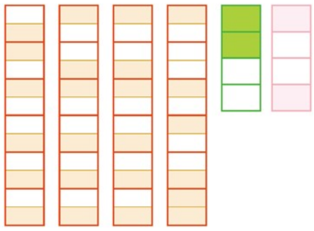
30th Gen.

E_c = -248.65 eV / f. u.



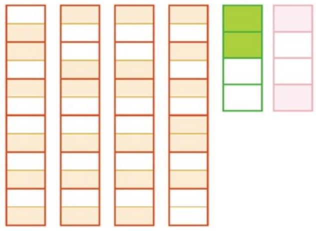
40th Gen.

E_c = -248.75 eV / f. u.



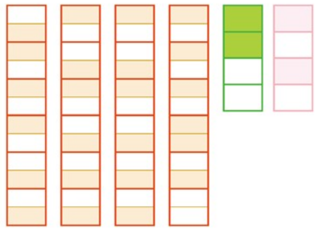
50th Gen.

E_c = -248.87 eV / f. u.



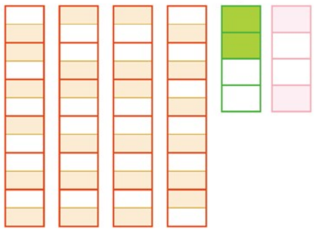
60th Gen.

E_c = -249.12 eV / f. u.



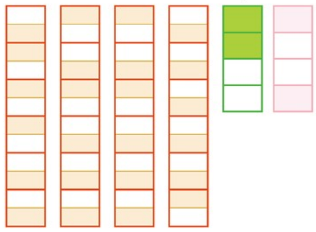
70th Gen.

E_c = -249.15 eV / f. u.



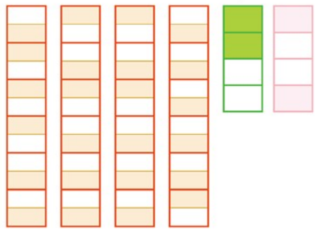
80th Gen.

E_c = -249.15 eV / f. u.



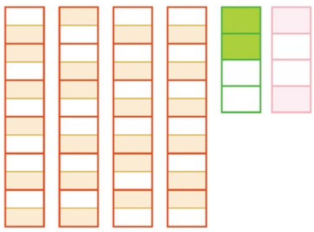
90th Gen.

E_c = -249.15 eV / f. u.



100th Gen.

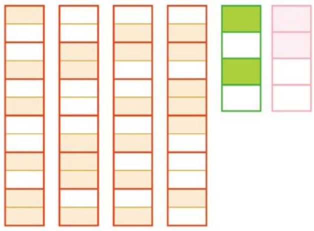
E_c = -249.27 eV / f. u.



Particle Swarm Optimization

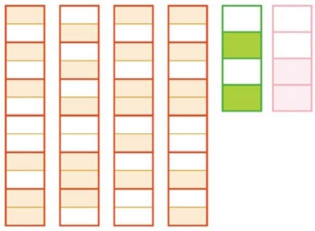
10th Gen.

E_c = -245.67 eV / f. u.



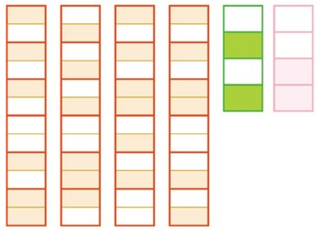
20th Gen.

E_c = -246.62 eV / f. u.



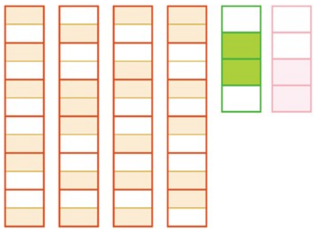
30th Gen.

E_c = -246.62 eV / f. u.



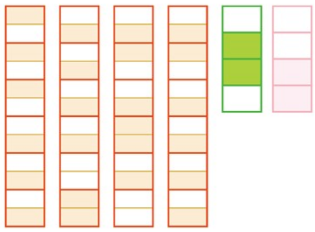
40th Gen.

E_c = -247.25 eV / f. u.



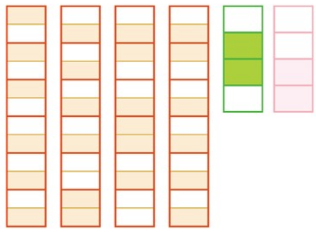
50th Gen.

E_c = -247.76 eV / f. u.



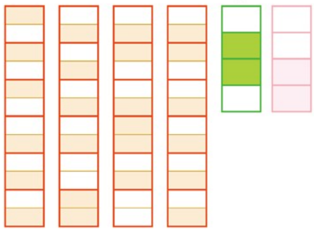
60th Gen.

E_c = -247.76 eV / f. u.



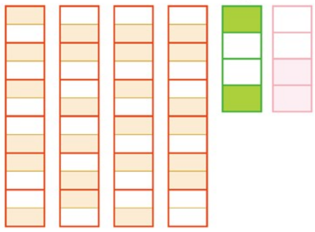
70th Gen.

E_c = -247.76 eV / f. u.



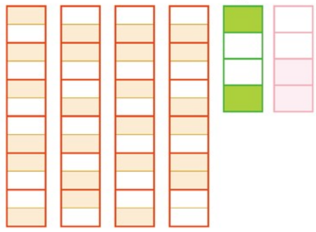
80th Gen.

E_c = -248.10 eV / f. u.



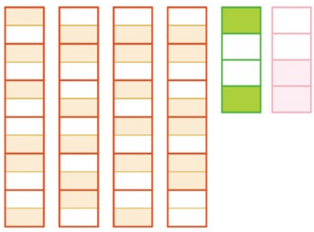
90th Gen.

E_c = -248.10 eV / f. u.

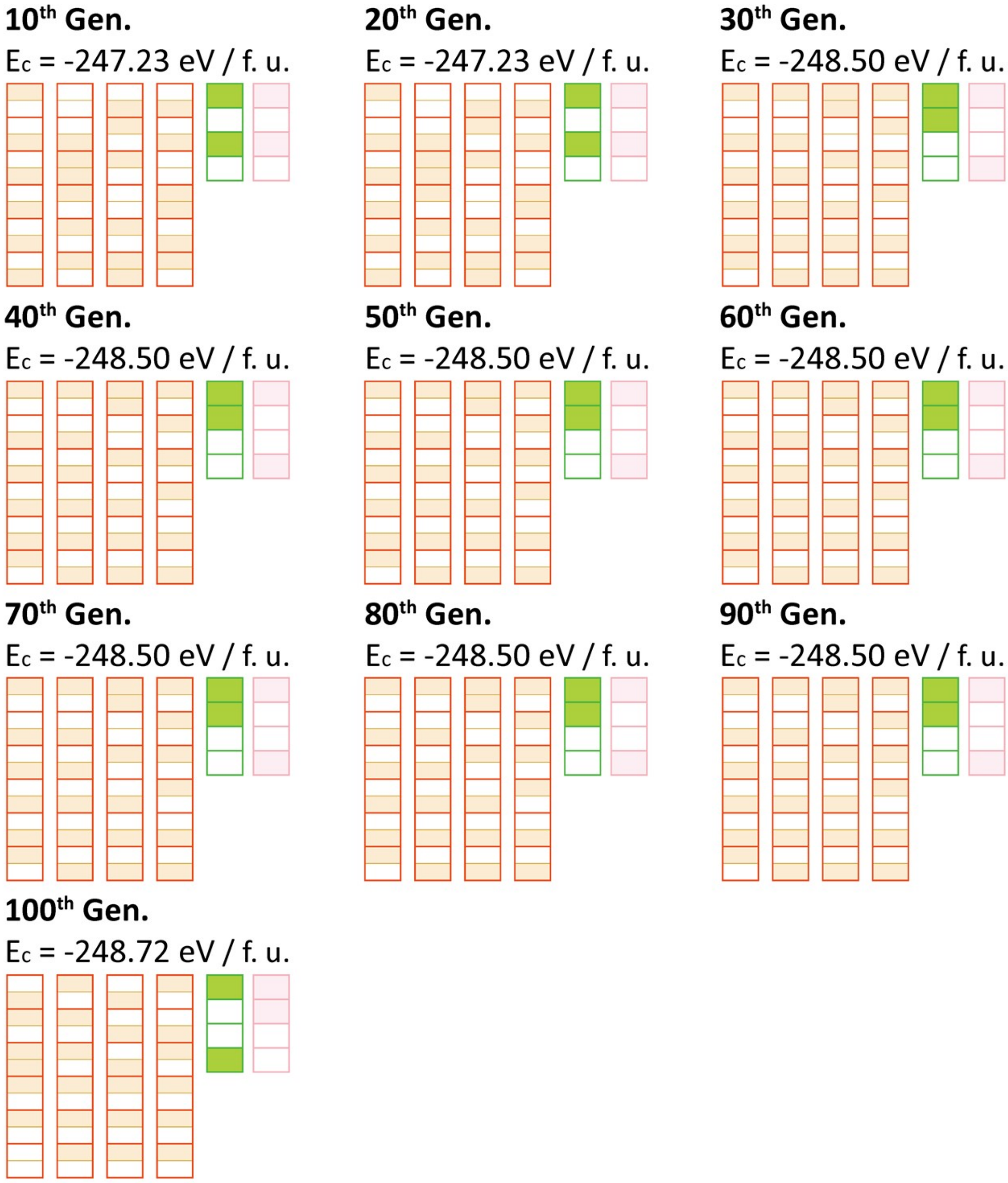


100th Gen.

E_c = -248.10 eV / f. u.



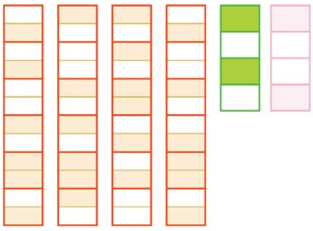
Harmony Search



Cuckoo Search

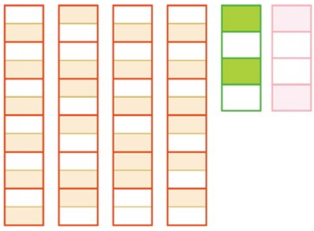
10th Gen.

E_c = -247.05 eV / f. u.



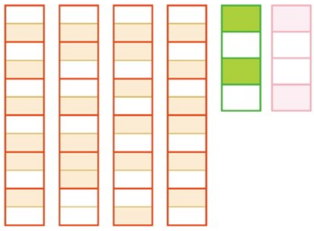
20th Gen.

E_c = -248.59 eV / f. u.



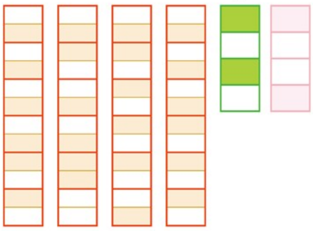
30th Gen.

E_c = -247.67 eV / f. u.



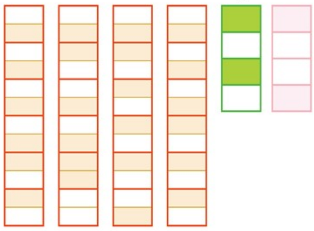
40th Gen.

E_c = -247.67 eV / f. u.



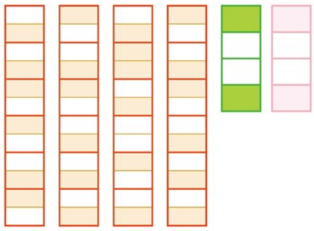
50th Gen.

E_c = -247.67 eV / f. u.



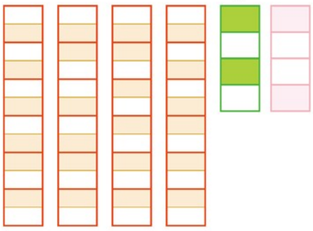
60th Gen.

E_c = -248.18 eV / f. u.



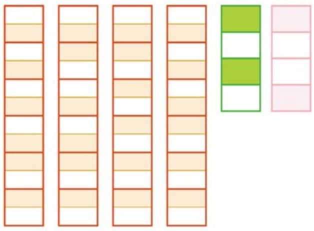
70th Gen.

E_c = -248.80 eV / f. u.



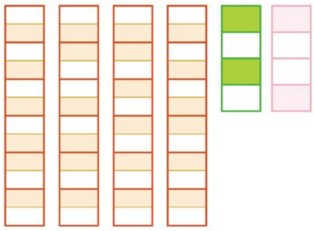
80th Gen.

E_c = -248.80 eV / f. u.



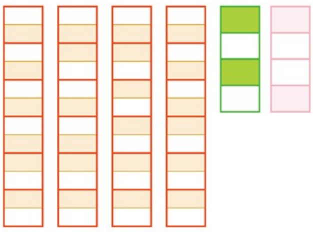
90th Gen.

E_c = -248.80 eV / f. u.

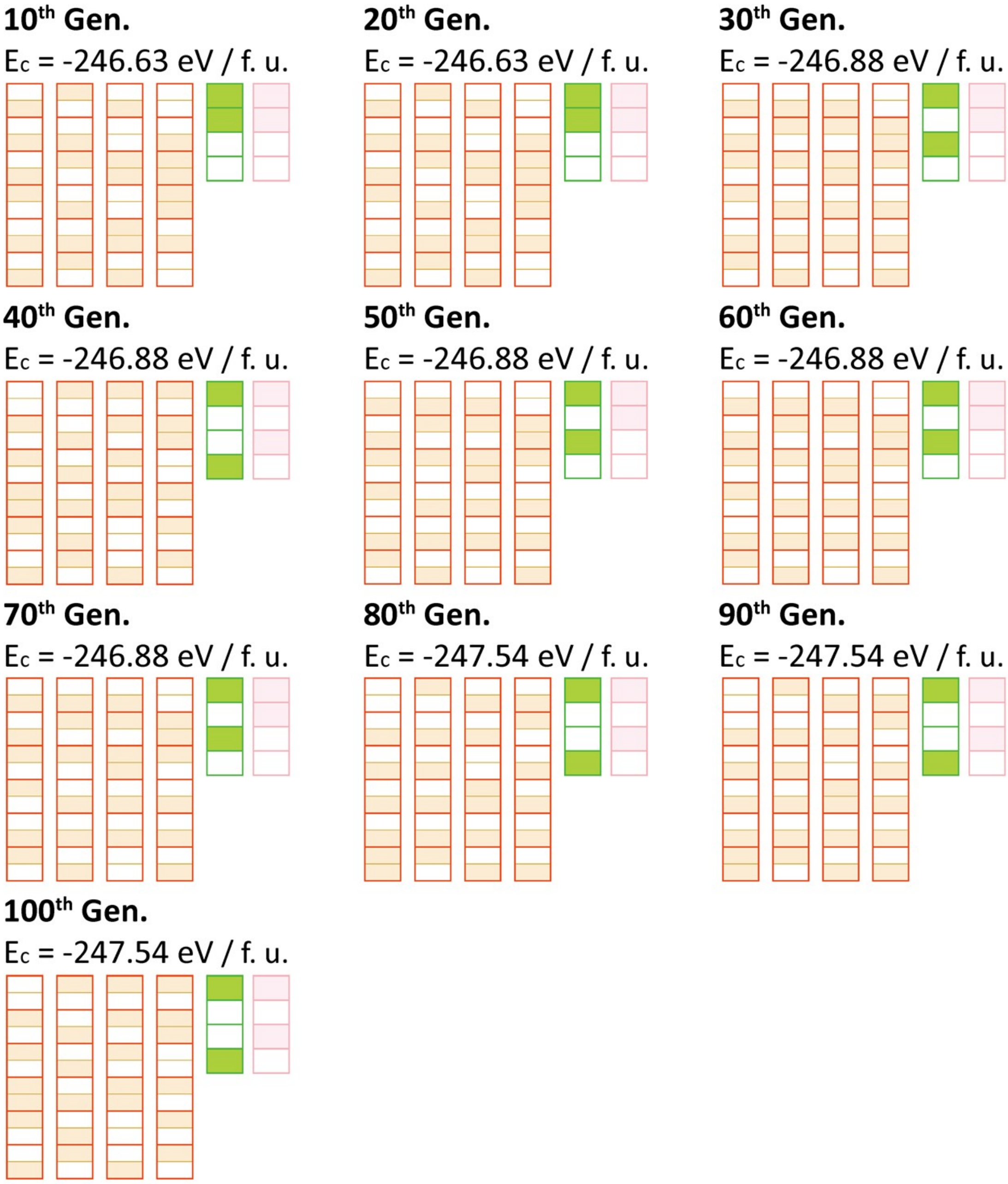


100th Gen.

E_c = -248.80 eV / f. u.



Bayesian Optimization



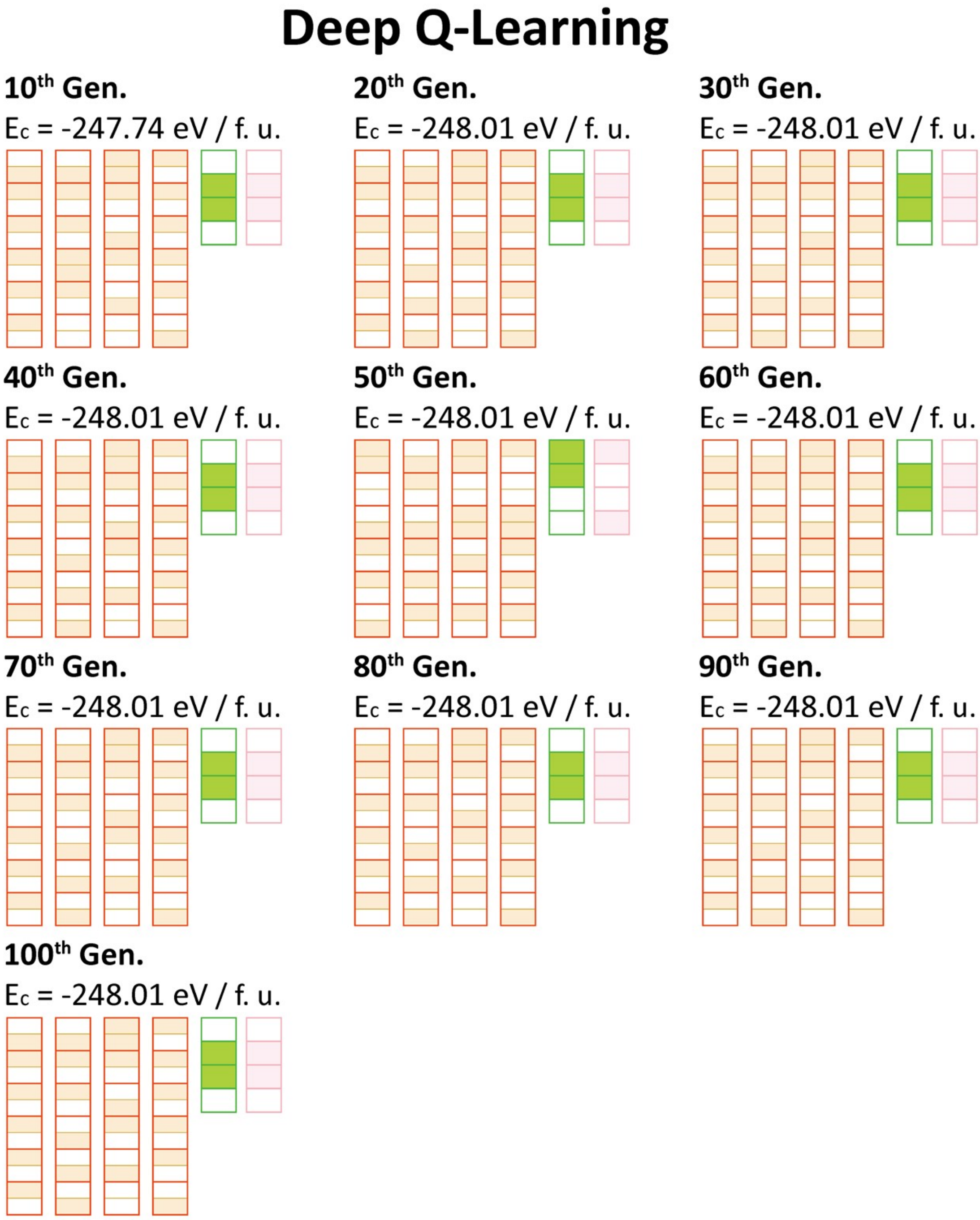
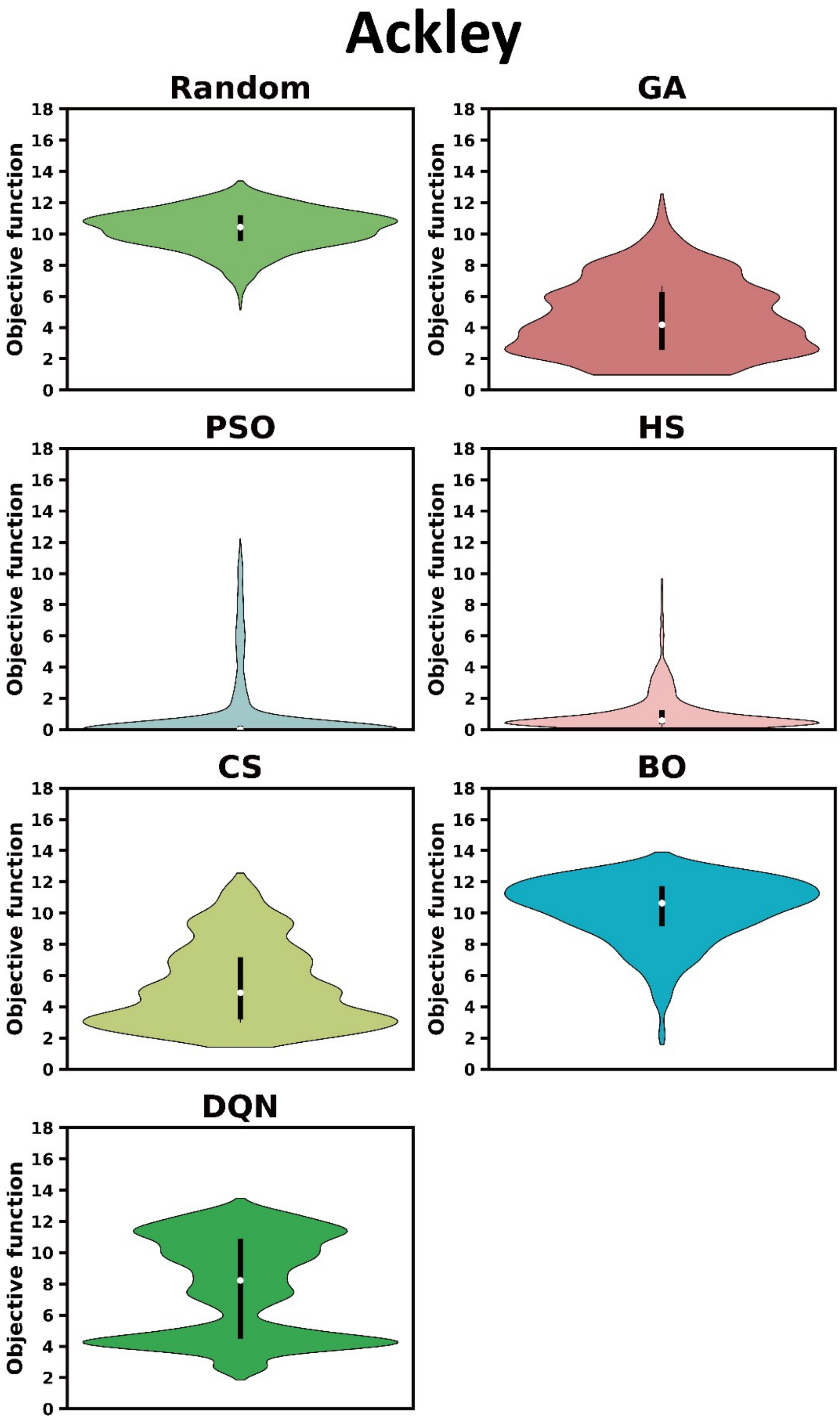
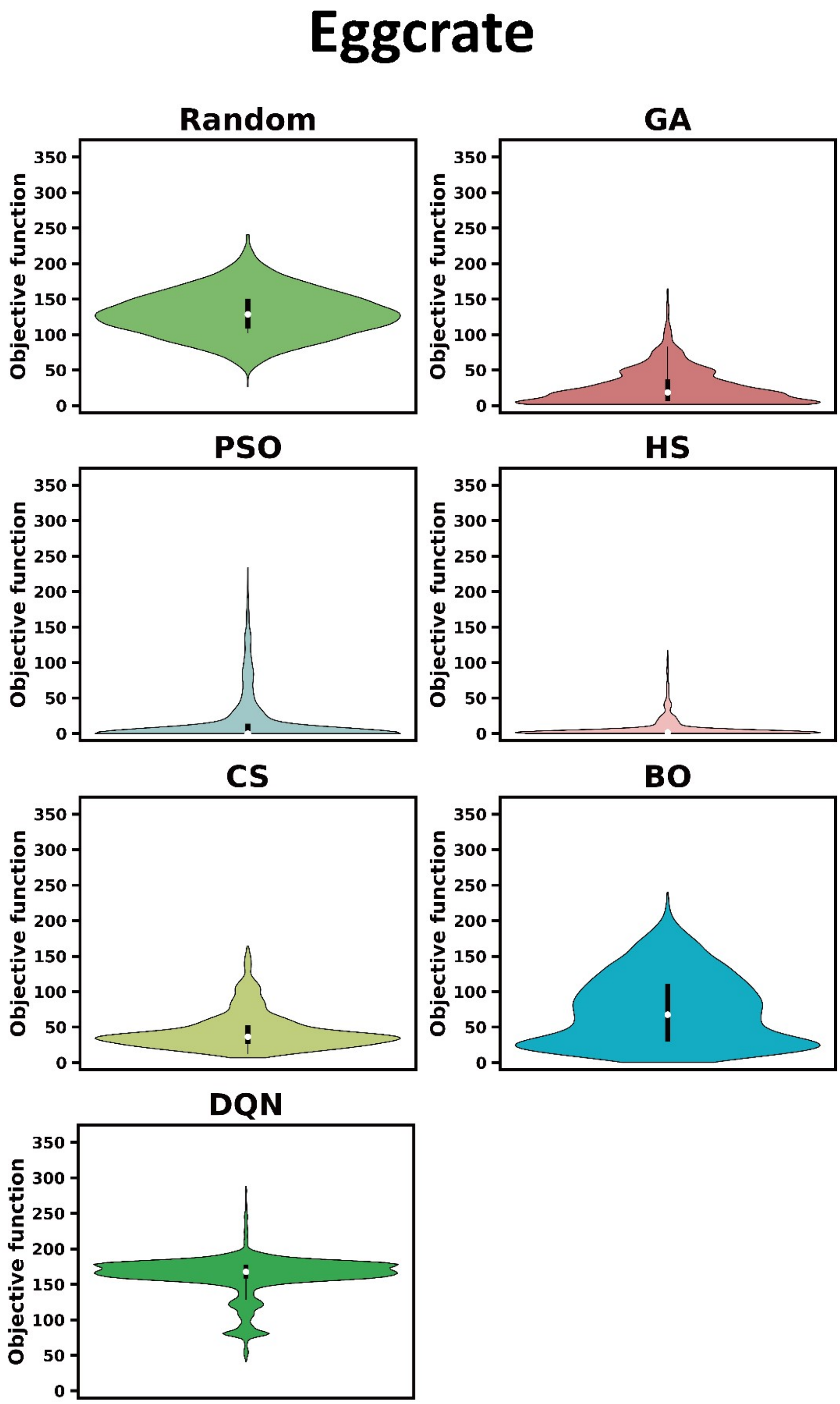
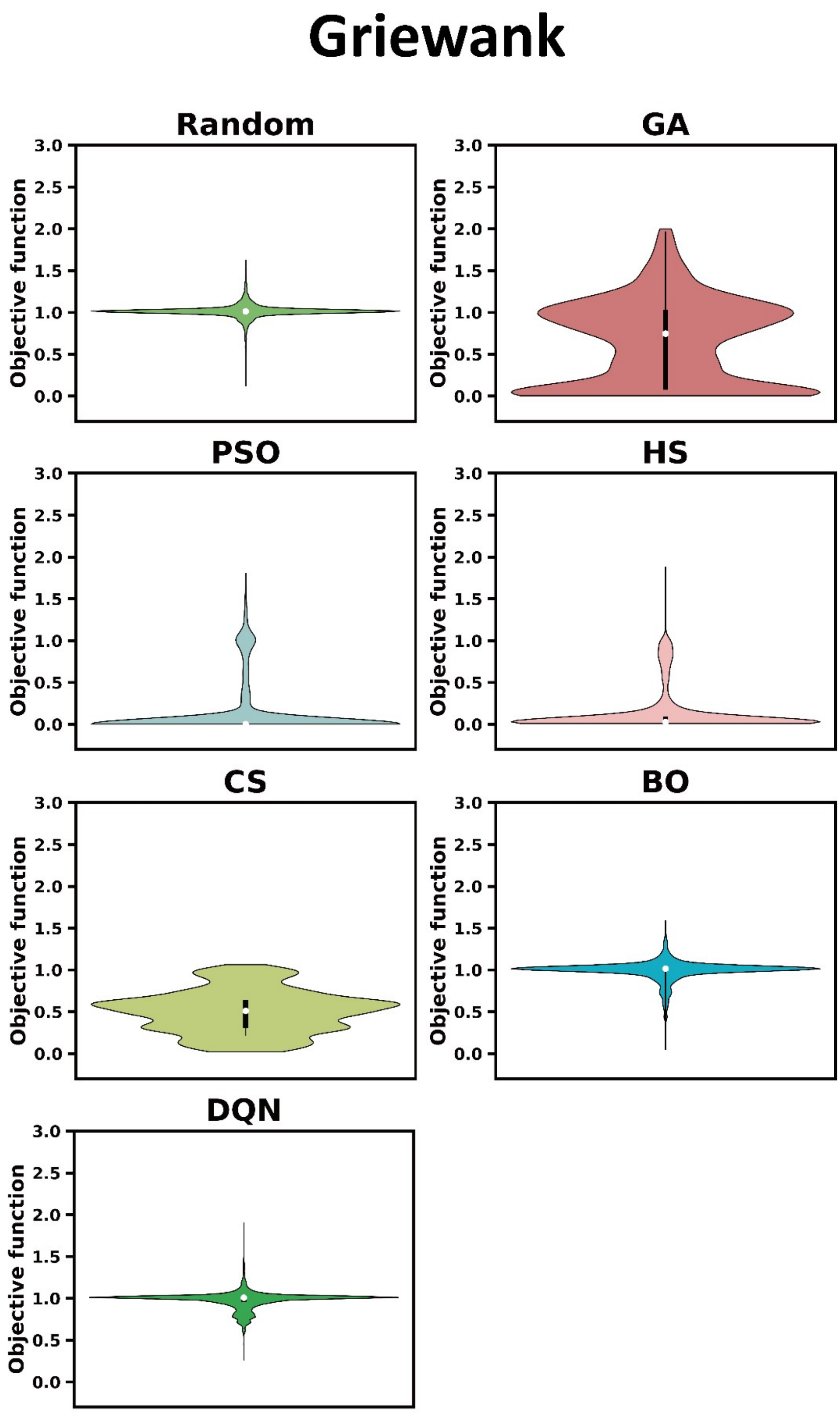
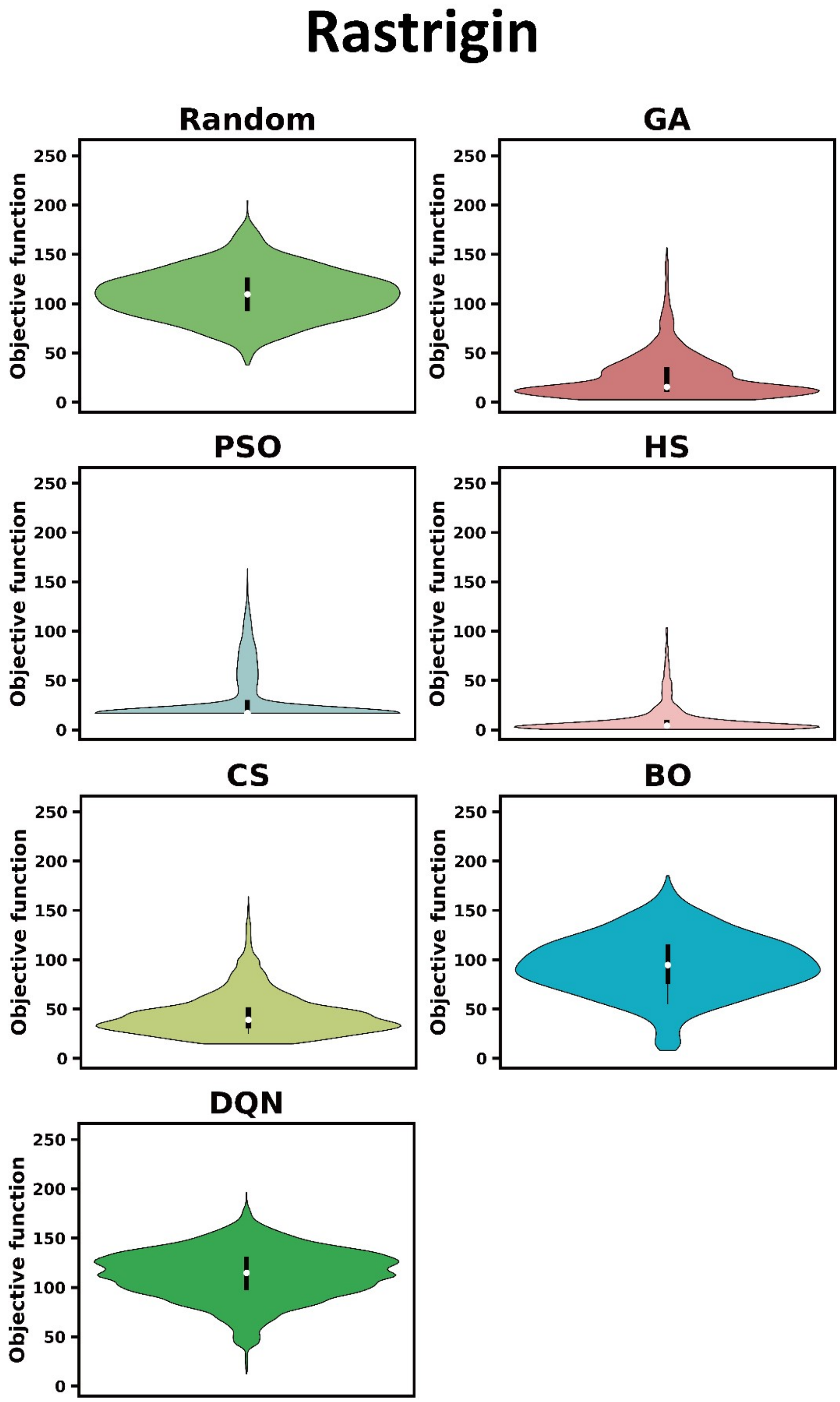


Fig. S2 The configurations selected every 10th generation for GA, PSO, HS, CS, BO, and DQN algorithms.

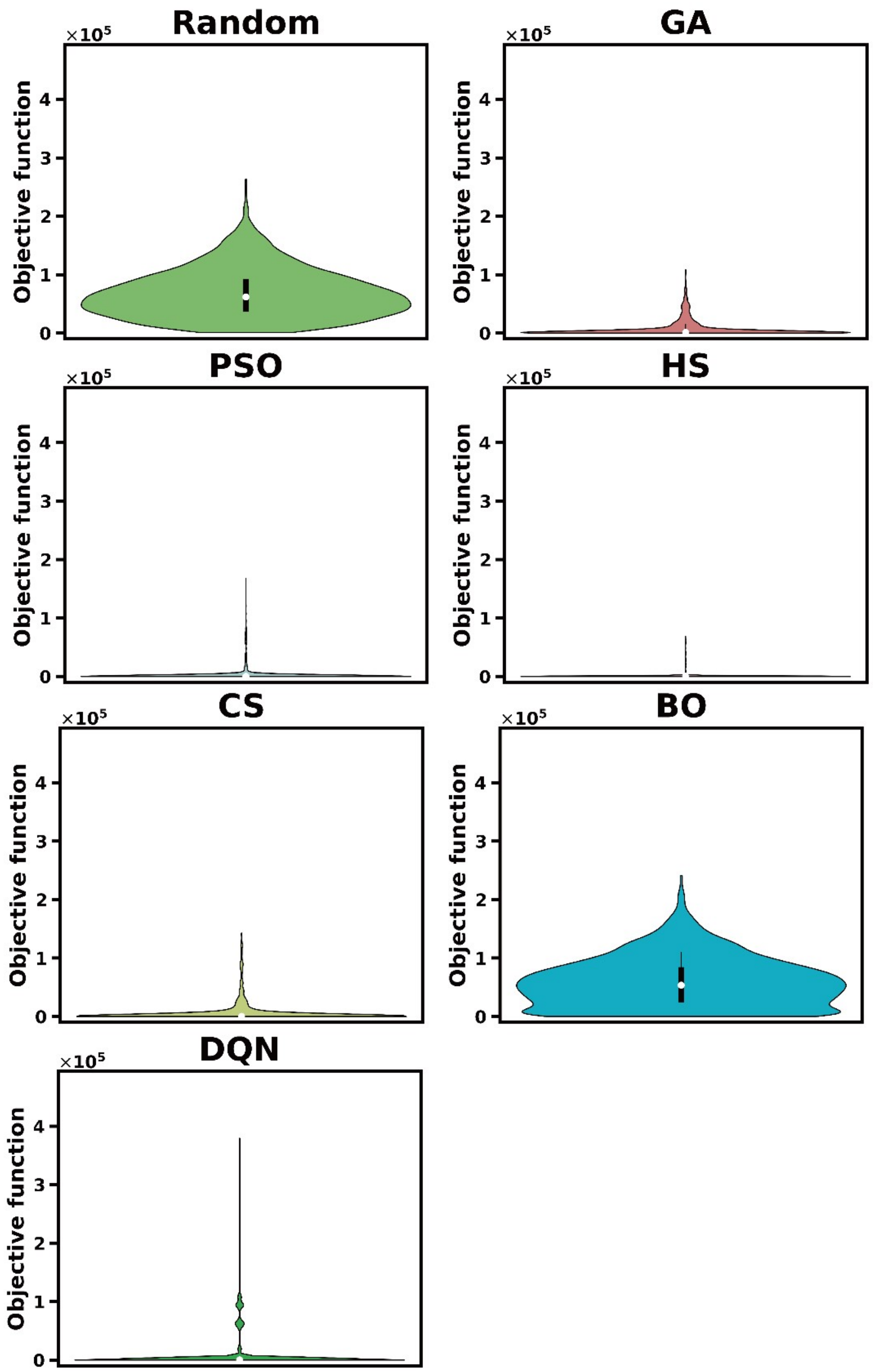

Ackley
Random
GA
PSO
HS
CS
BO
DQN
Objective function
0
2
4
6
8
10
12
14
16
18


Eggcrate
Random
GA
PSO
HS
CS
BO
DQN
Objective function
0
50
100
150
200
250
300
350


Griewank
Random
GA
PSO
HS
CS
BO
DQN
Objective function
0.0
0.5
1.0
1.5
2.0
2.5
3.0


Rastrigin
Random
GA
PSO
HS
CS
BO
DQN
Objective function
0
50
100
150
200
250

Rosenbrock



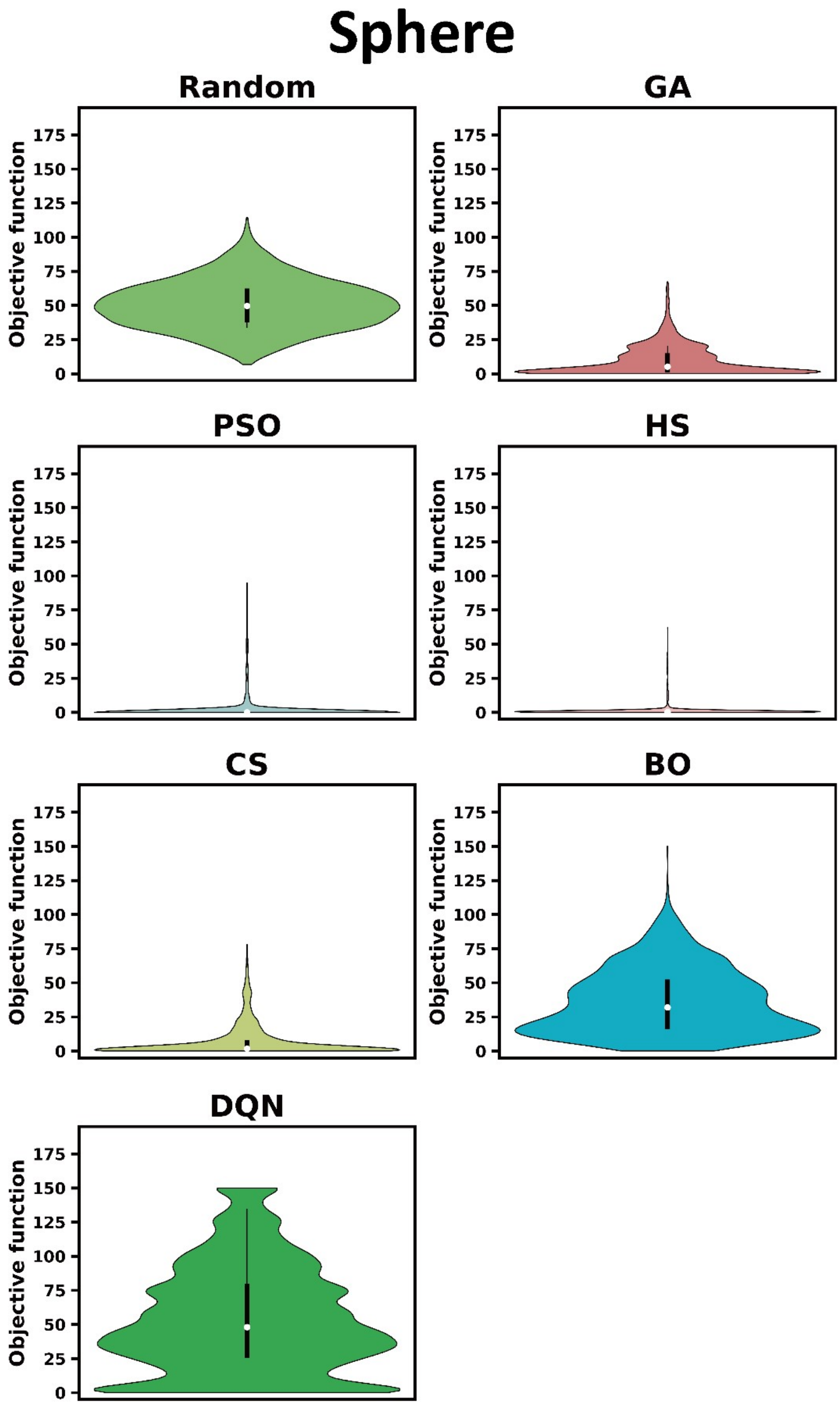


Fig. S3 Violin plots for every benchmark function data

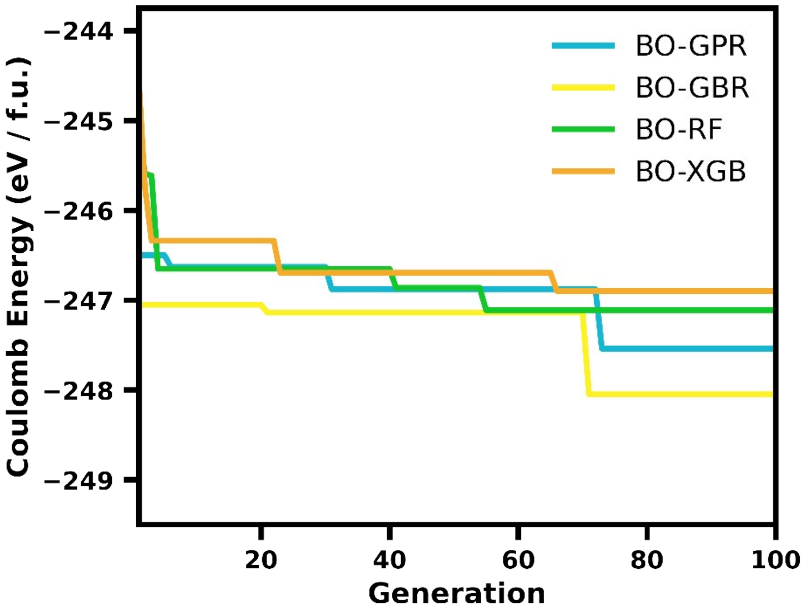


Fig. S4 The instantaneous Coulomb energy value versus generation for BO with four different surrogate functions. GPR, GBR, RF and XGB are refered to as the gaussian process regression, gradient boost regression, randum forest and extreme gradient(XG) boost regression, respectively.

Table S1 Finally optimized objective function (benchmark function) values that was reached by each of the metaheuristics (GA, PSO, HS and CS), BO and DQN algorithms

Method	Ackley	Egg Crate	Griewank	Rastrigin	Rosenbrock	Sphere
Random	5.134	26.708	0.124	37.574	973.423	6.472
GA	0.346	1.341	0.013	5.606	66.077	0.168
PSO	0.003	0.628×10^{-9}	0.246×10^{-2}	7.583	3.821	0.390×10^{-11}
HS	0.138	0.193	0.631	1.099	1.957	0.001
CS	0.953	14.253	0.102	11.577	6.954	0.003
BO	0.853	9.566	0.343	27.283	35.055	0.002
DQN	2.072	57.792	0.265	16.82	0.185	0.051

Table S2 Hyperparameter for benchmark function

Method	Hyperparameter
Random	-
GA	Mutation probability : 0.1 , elite size : 0.01 crossover probability
PSO	C_1 : 1, C_2 : 2, w : 0.5
HS	HMCR : 0.7 PAR : 0.3 bw : 0.01
CS	P_a : 0.25 β : 1.5
BO	α : 10^{-5} , β : 10^{-7}
DQN	Episode length : 2500, Learning rate : 0.001