

はじめに ベイズ最適化の紹介

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Discovery of new functional molecules and materials is of national importance

The screenshot shows the White House website's page for the Materials Genome Initiative. At the top, the navigation bar includes the White House logo, the text "the WHITE HOUSE PRESIDENT BARACK OBAMA", and links for "Get Email Updates" and "Contact Us". Below this is a secondary navigation bar with links for "BLOG", "PHOTOS & VIDEO", "BRIEFING ROOM", "ISSUES", "the ADMINISTRATION", "the WHITE HOUSE", and "our GOVERNMENT". A search bar is located on the right side of the page.

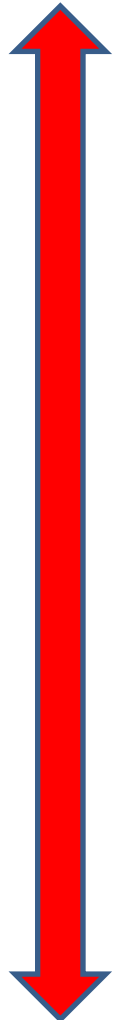
The main content area features the "Materials Genome Initiative" logo on the left and the title "Materials Genome Initiative" in the center. Below the title is a navigation menu with links for "About", "Goals", "Examples", "News & Announcements", "Federal Programs", "External Stakeholder Activities", and "Contact Us".

The main text on the page reads: "To help businesses discover, develop, and deploy new materials twice as fast, we're launching what we call the Materials Genome Initiative. The invention of silicon circuits and lithium-ion batteries made computers and iPods and iPads possible -- but it took years to get those technologies from the drawing board to the marketplace. We can do it faster."

Below the text is a quote attributed to President Obama: "– President Obama, June 2011 at Carnegie Mellon University". To the right of the text is a photograph of President Obama in a factory setting, wearing safety glasses and holding a long, glowing fluorescent light fixture. He is surrounded by other people, including a woman in a red jacket who is also looking at the light fixture.

First Principles Calculations

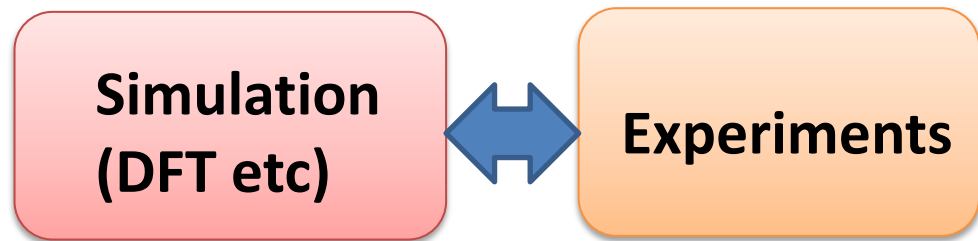
Accurate, Slow



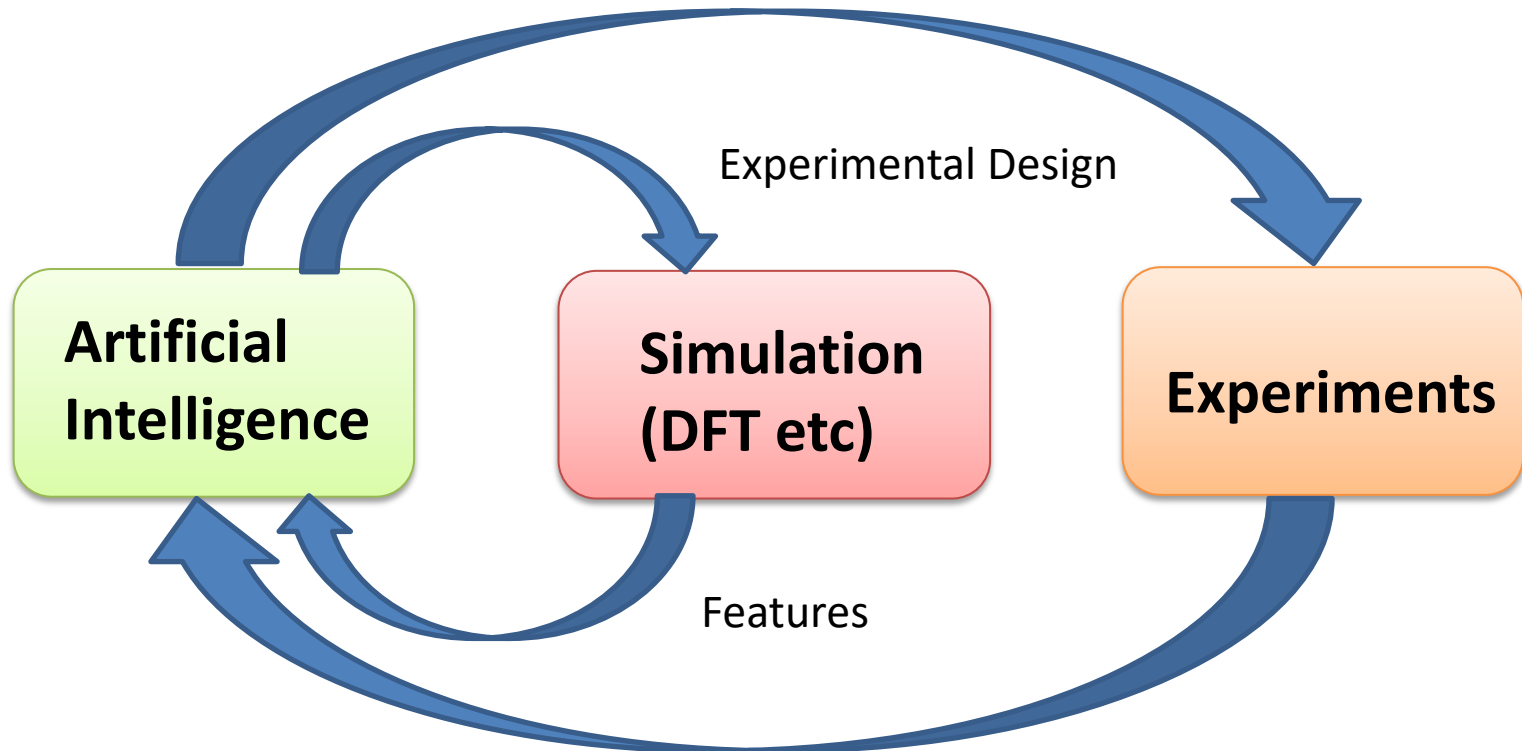
- Full configuration interaction
- Wave function based
- Density functional theory
- Semi-empirical
- Empirical potentials

Inaccurate, Fast

Old Picture



New Picture



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
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First Principles Calc.



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

Bayesian Optimization (1)

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



Score 1	Score 2	Score 3
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Bayesian Optimization (2)

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



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

Predicted Scores

Predicted Variances



Bayesian Optimization (3)

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



Score 1	Score 2	Score 3	Score 8
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Bayesian Optimization (4)

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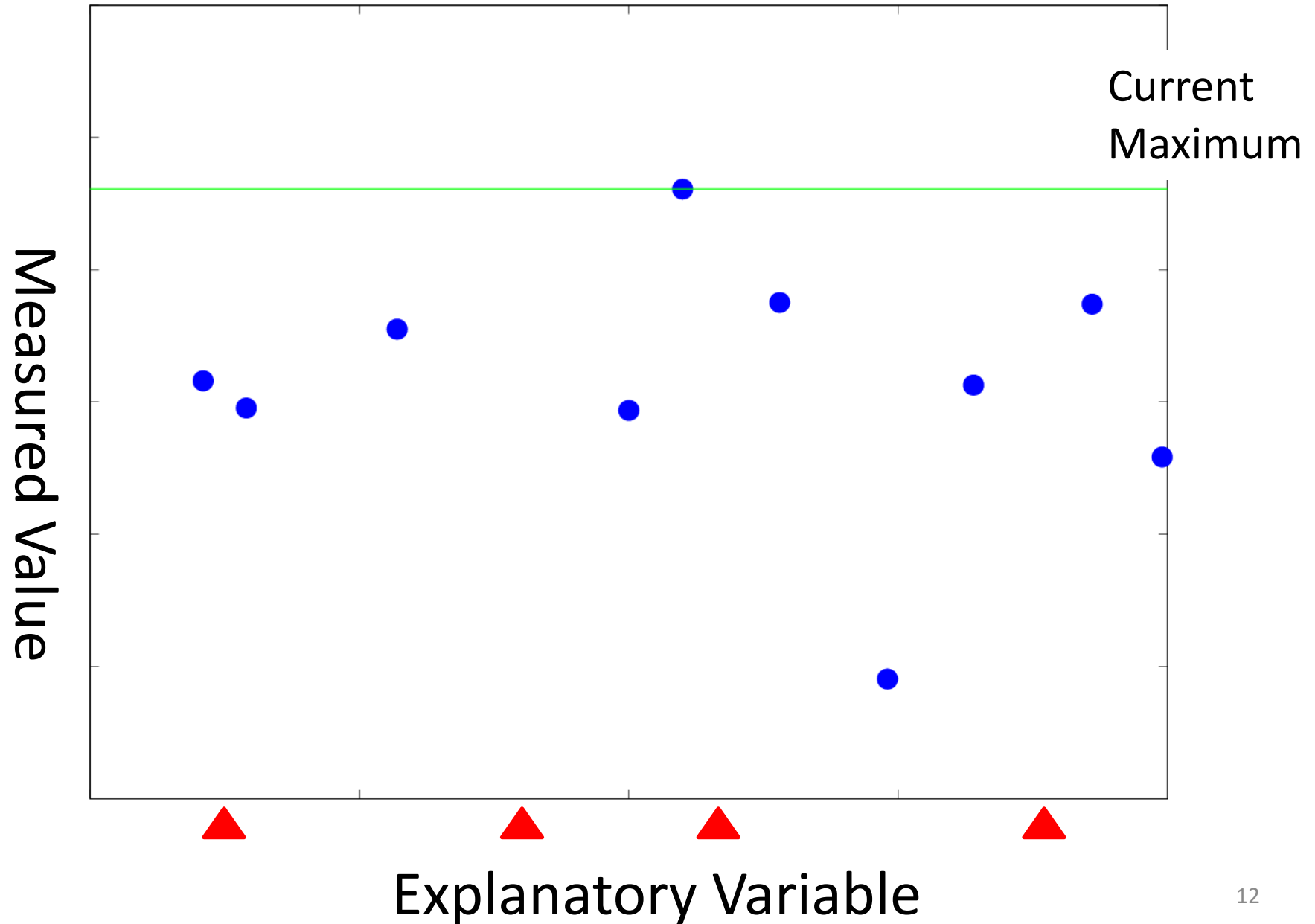


First Principles Calc.

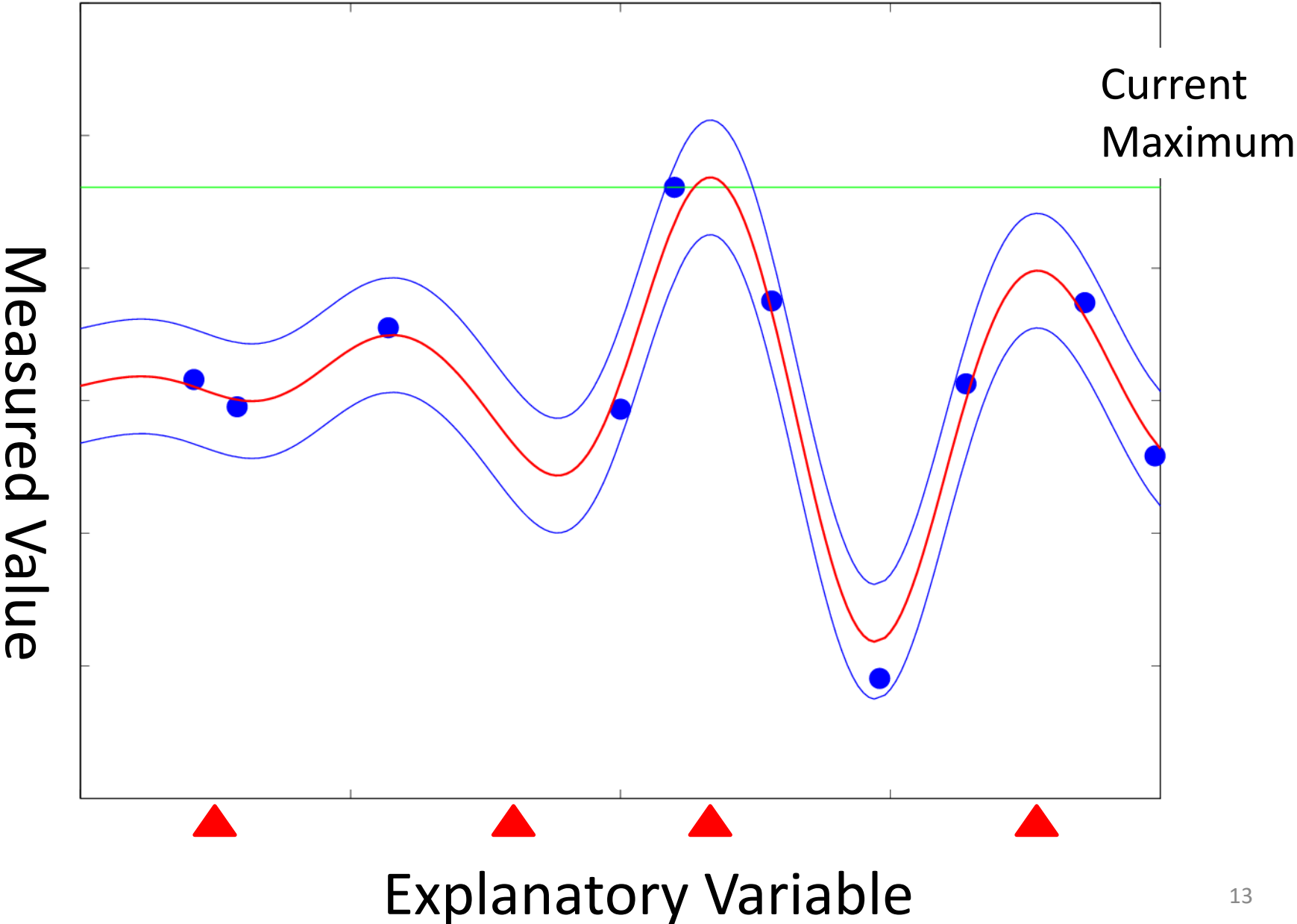


Score 1	Score 2	Score 3	Score 8	Pred. Score 4	Pred. Score 5	Pred. Score 6	Pred. Score 7	Pred. Score 9	Pred. Score 10
				Var. 4	Var. 5	Var. 6	Var. 7	Var. 9	Var. 10

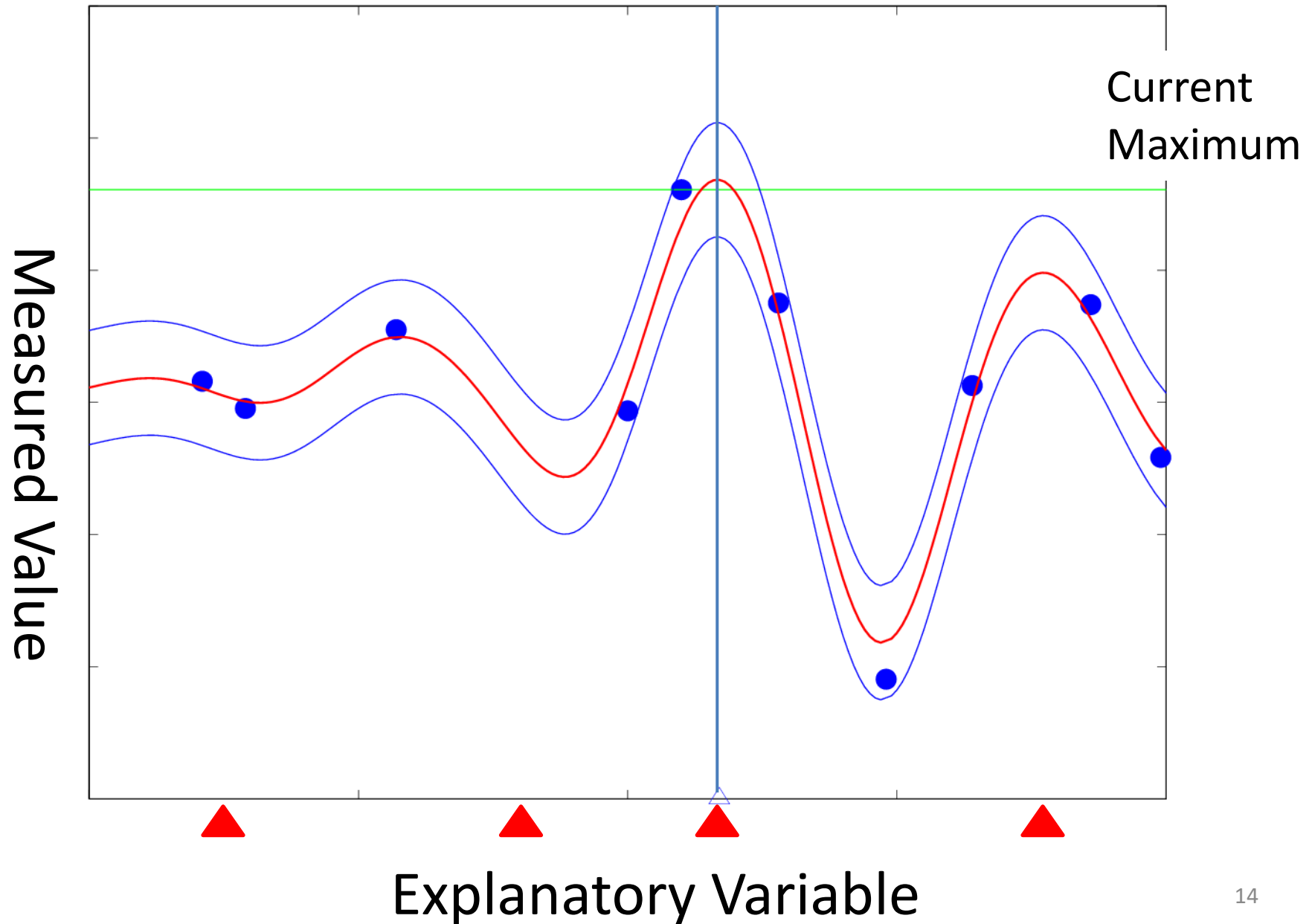
Where to observe next?



Gaussian Process



Maximum probability of improvement



Gaussian Process

Multivariate Gaussian Distribution

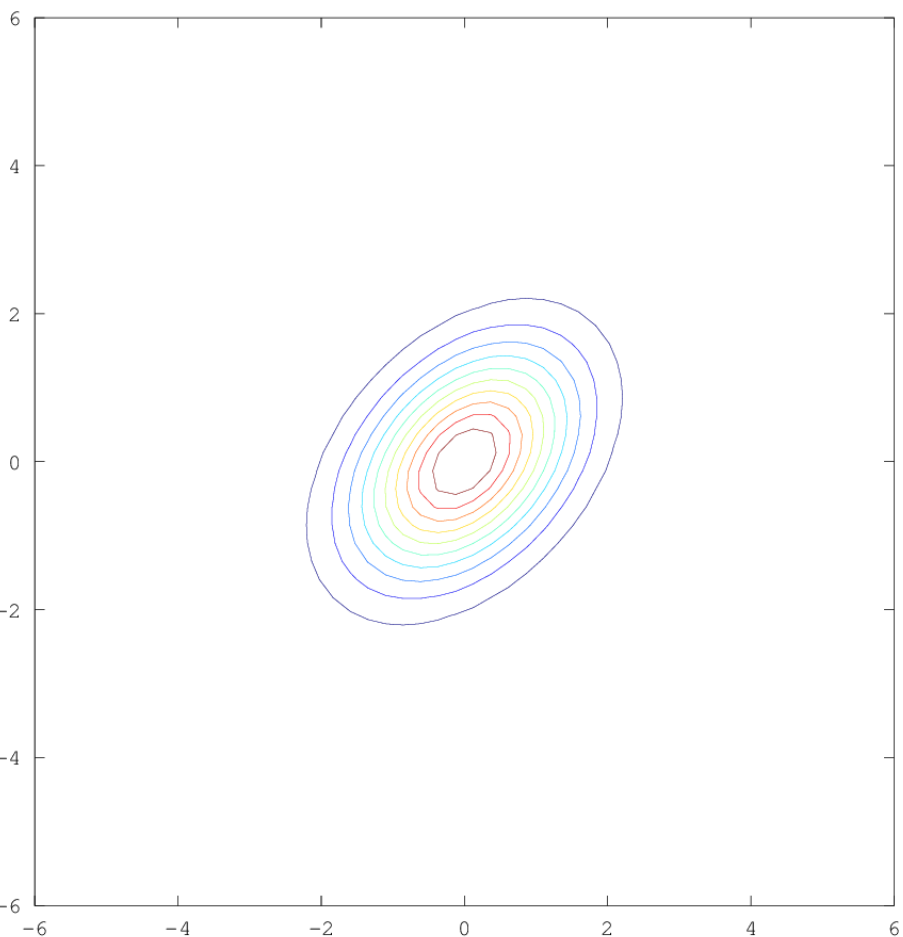
- Probability density function

$$p(\mathbf{x} \mid \boldsymbol{\mu}, \boldsymbol{\Sigma}) = \frac{1}{(2\pi)^{d/2} |\boldsymbol{\Sigma}|^{1/2}} \exp\left(-\frac{1}{2} (\mathbf{x} - \boldsymbol{\mu})^\top \boldsymbol{\Sigma}^{-1} (\mathbf{x} - \boldsymbol{\mu})\right)$$

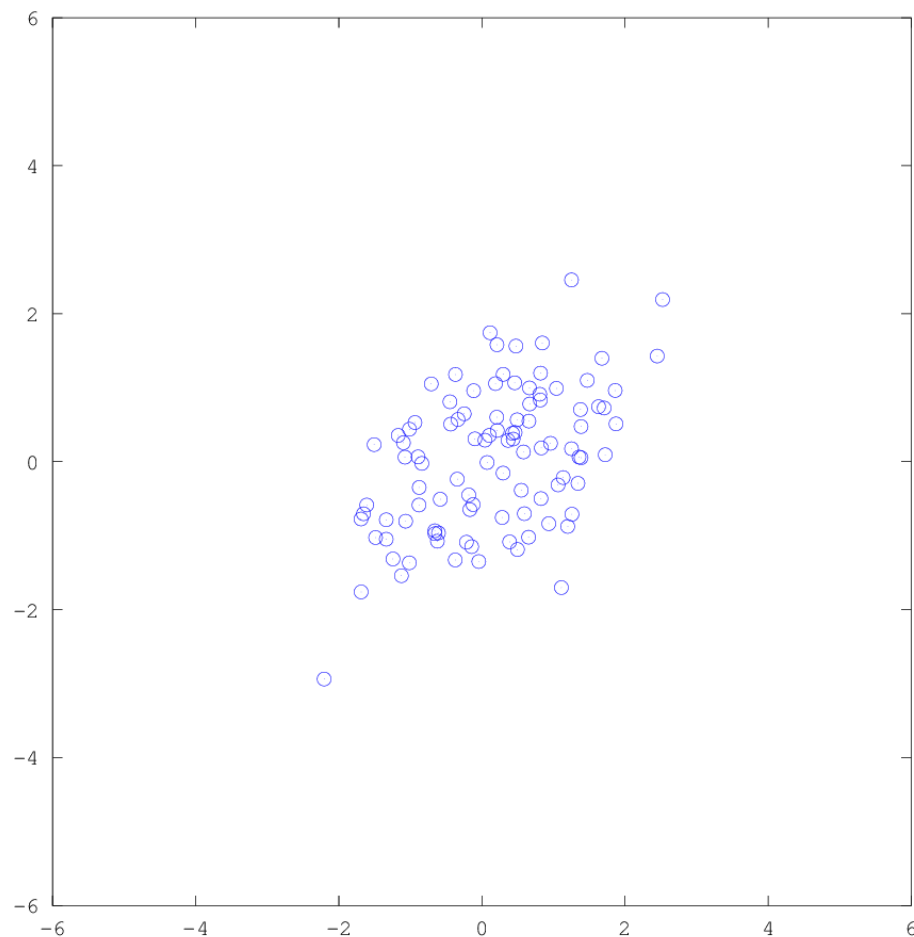
$\boldsymbol{\mu}$ Mean

$\boldsymbol{\Sigma}$ Covariance Matrix

Probability Density



100 Samples



$$\boldsymbol{\mu} = (0, 0)^\top$$

$$\boldsymbol{\Sigma} = \begin{pmatrix} 0.4 & 1 \\ 1 & 0.4 \end{pmatrix}$$

Conditional Distribution

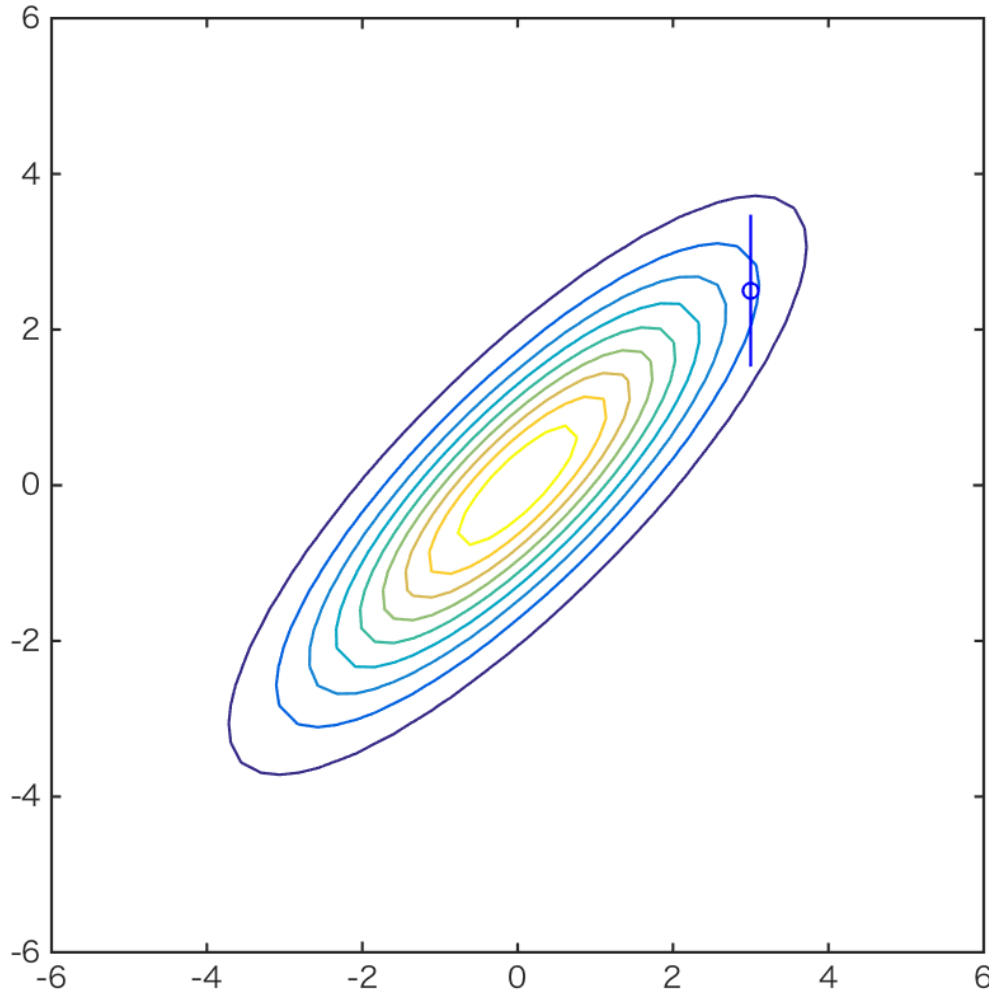
$$\begin{array}{ccc} & \text{Mean} & \text{Covariance} \\ x = \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} & \mu = \begin{pmatrix} \mu_1 \\ \mu_2 \end{pmatrix} & \Sigma = \begin{pmatrix} \Sigma_{11} & \Sigma_{12} \\ \Sigma_{21} & \Sigma_{22} \end{pmatrix} \end{array}$$

$$P(x_1 \mid x_2 = a) = \mathcal{N}(\mu_c, \Sigma_c)$$

$$\mu_c = \mu_1 + \Sigma_{12} \Sigma_{22}^{-1} (a - \mu_2)$$

$$\Sigma_c = \Sigma_{11} - \Sigma_{12} \Sigma_{22}^{-1} \Sigma_{21}$$

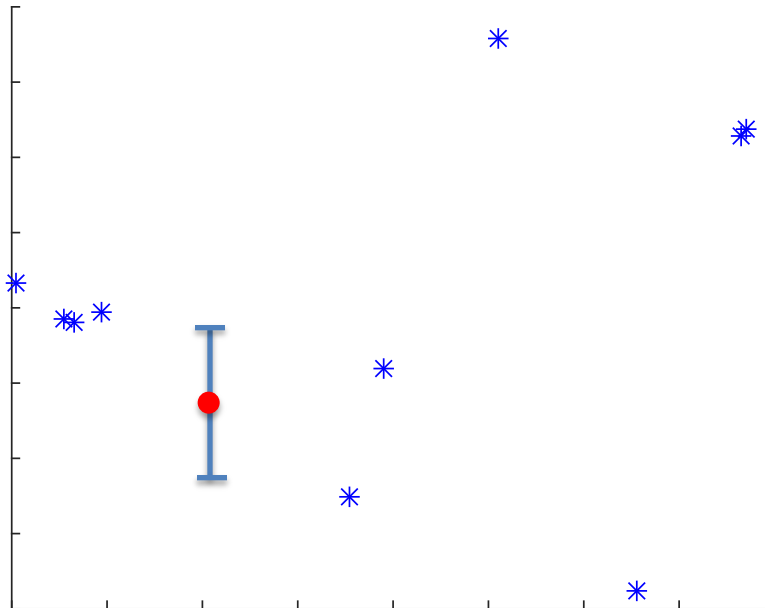
Conditional distribution at $X=3$



$$\Sigma = \begin{pmatrix} 3 & 2.5 \\ 2.5 & 3 \end{pmatrix}$$

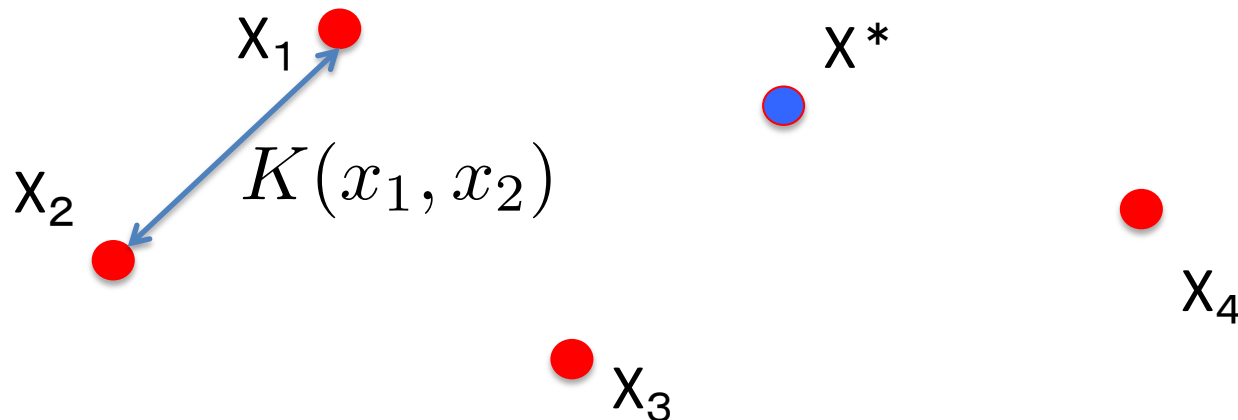
Gaussian Process

- Kernel method for regression
- Provides predictive variance in addition to regression function



Gaussian Process (No noise)

- Training points $\{x_i\}_{i=1,\dots,n}$ 、 Test point x^*
- Observed outcomes y_i, y^* are subject to $n+1$ dim Gaussian
- Mean of y_i is 0.
- Covariances are given as $K(x_i, x_j)$



Covariance matrix by Gaussian kernel

$$\begin{pmatrix} k(\mathbf{x}^*, \mathbf{x}^*) & \mathbf{k}^{*\top} \\ \mathbf{k} & K \end{pmatrix}$$

$$K(x, x') = \exp(-\|x - x'\|^2 / \eta)$$

Gaussian Process (No noise)

- K : Kernel matrix for training points
- \mathbf{y} : Observed outcomes for training points
- Predicted outcome at \mathbf{x}^*

$$E[y^*] = \mathbf{k}^{*\top} K^{-1} \mathbf{y}$$

- Predicted variance

$$V[y^*] = k(\mathbf{x}^*, \mathbf{x}^*) - \mathbf{k}^{*\top} K^{-1} \mathbf{k}^*$$

Gaussian process with noise

- Observed outcome include mean 0, variance σ^2 noise
- Covariance matrix

$$\begin{pmatrix} k(\mathbf{x}^*, \mathbf{x}^*) + \sigma^2 & \mathbf{k}^{*\top} \\ \mathbf{k} & K + \sigma^2 I \end{pmatrix}$$

Gaussian Process (with noise)

- K : Kernel matrix for training points
- \mathbf{y} : Observed outcomes for training points
- Predicted outcome at \mathbf{x}^*

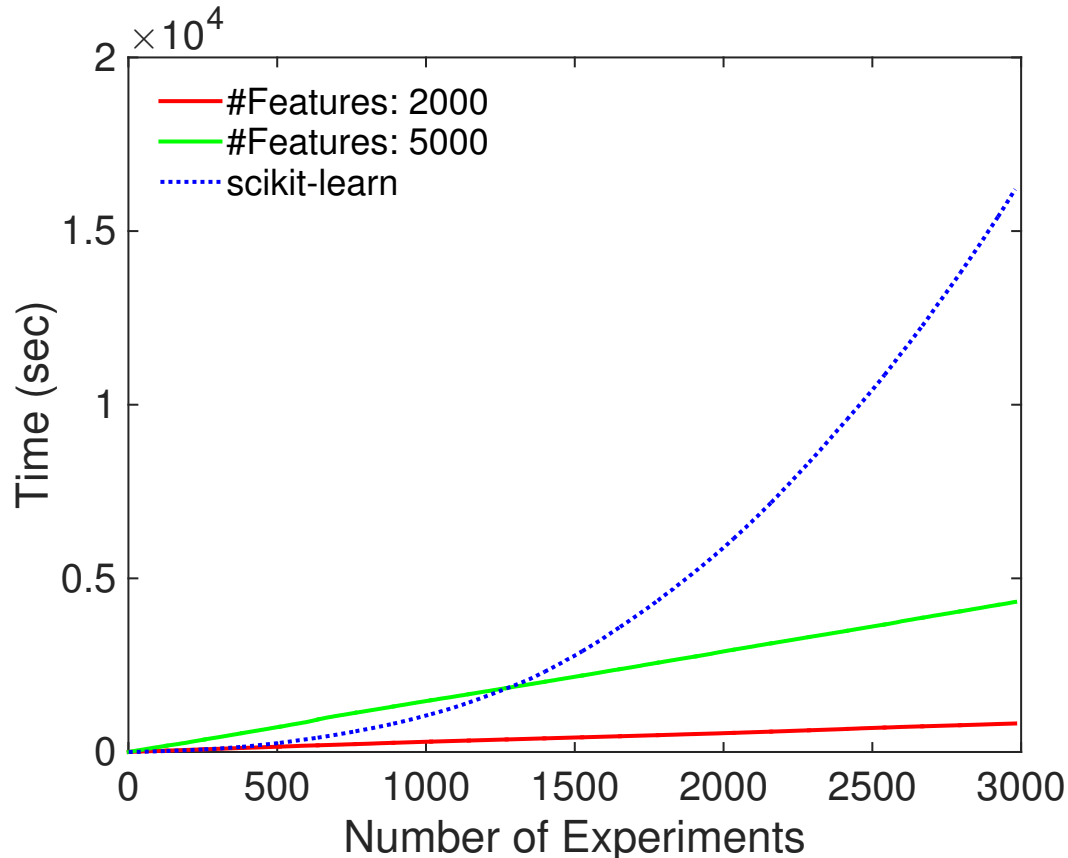
$$E[y^*] = \mathbf{k}^{*\top} (K + \sigma^2 I)^{-1} \mathbf{y}$$

- Predicted variance

$$V[y^*] = k(\mathbf{x}^*, \mathbf{x}^*) + \sigma^2 - \mathbf{k}^{*\top} (K + \sigma^2 I)^{-1} \mathbf{k}^*$$

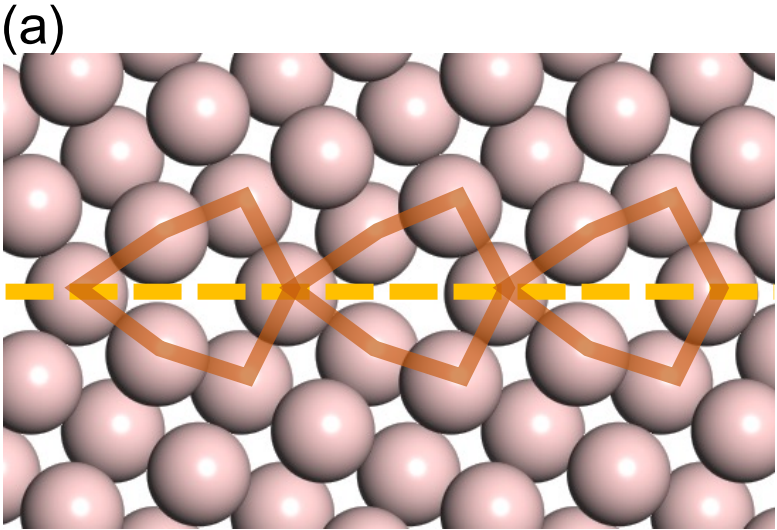
PHYSBO (COMBO)

- Fast learning by random feature maps
- Automatic hyperparameter initialization & update



ベイズ最適化による粒界構造決定の高速化

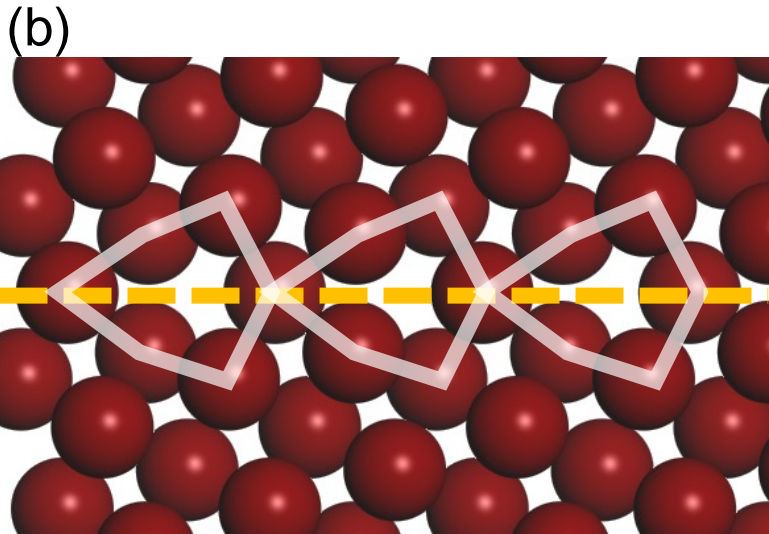
Cu [001] (210) $\Sigma 5$ 粒界



網羅的計算により決定

GB energy=0.96J/m²

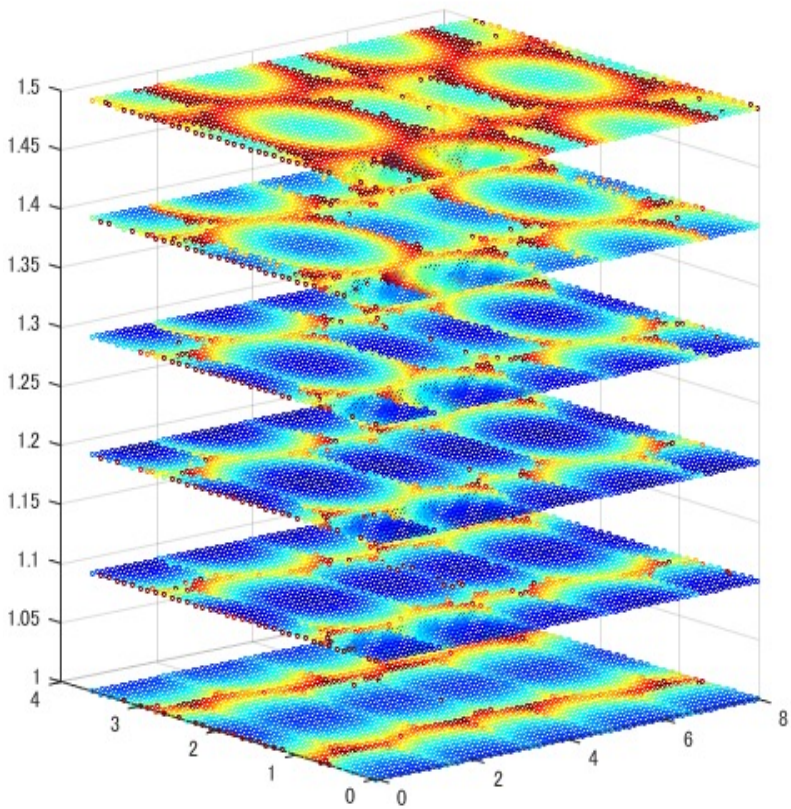
計算回数=16,983回



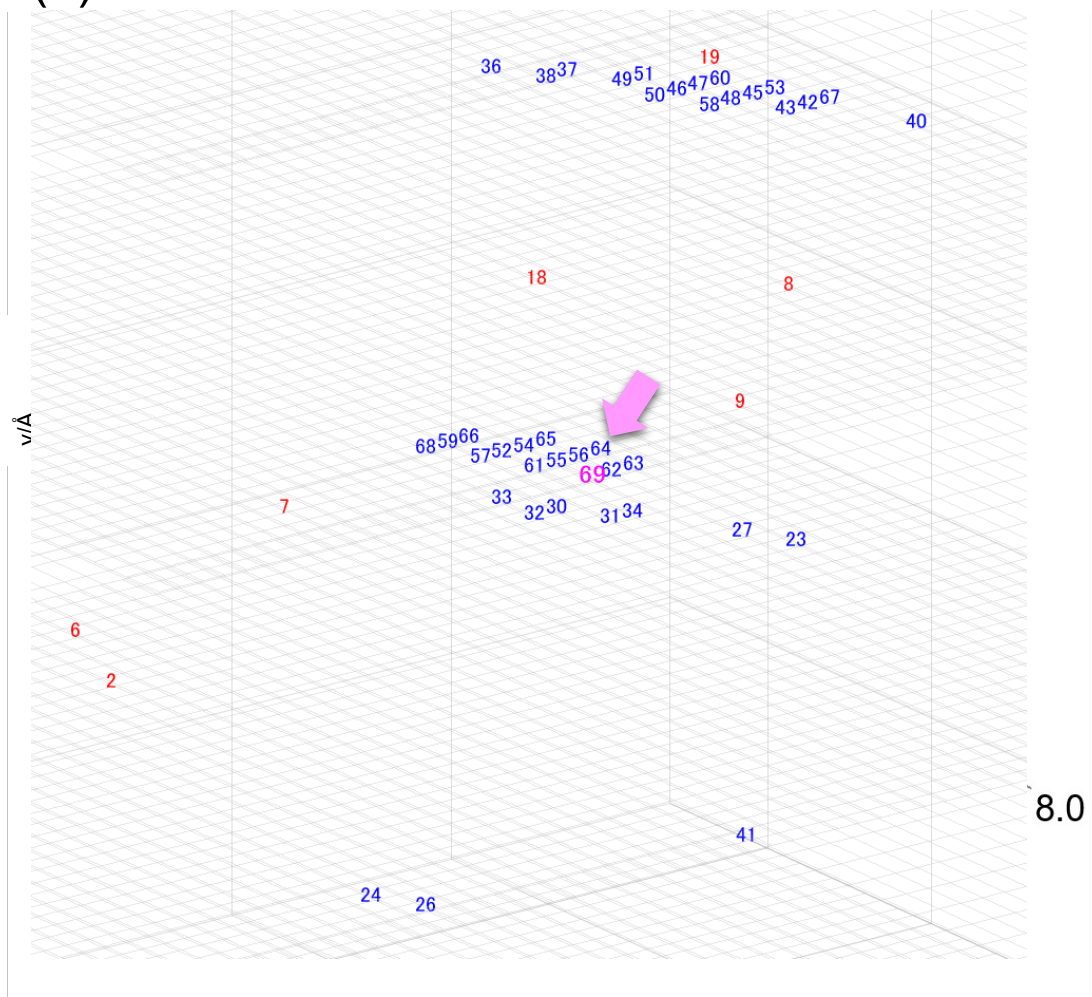
ベイズ最適化により決定

GB energy=0.96J/m²

計算回数=69回
(20回のinitial trial含む)



(b)



二元化合物の融点データ を用いた計算実験

- 226個ある材料のなかから、融点が最高のも
のを発見する
- 5%をランダムに選んで融点を観測する
- その後、ベイズ最適化を用いて、観測順を自
動的に決定していく

17個の説明変数

#Ecoh:一原子あたりの凝集エネルギー(計算値)

#bm:体積弾性率(計算値)

#V:一原子あたりの格子体積(計算値)

#NN:最近接原子間距離(計算値)

#c:組成

#Z1:構成元素の原子番号の二乗和

#Z2:構成元素の原子番号の積

#Z3:構成元素の原子番号の和

#M1:構成元素の原子量の二乗和

#M2:構成元素の原子量の積

#M3:構成元素の原子量の和

#n1:構成元素の価電子数の二乗和

#n2:構成元素の価電子数の積

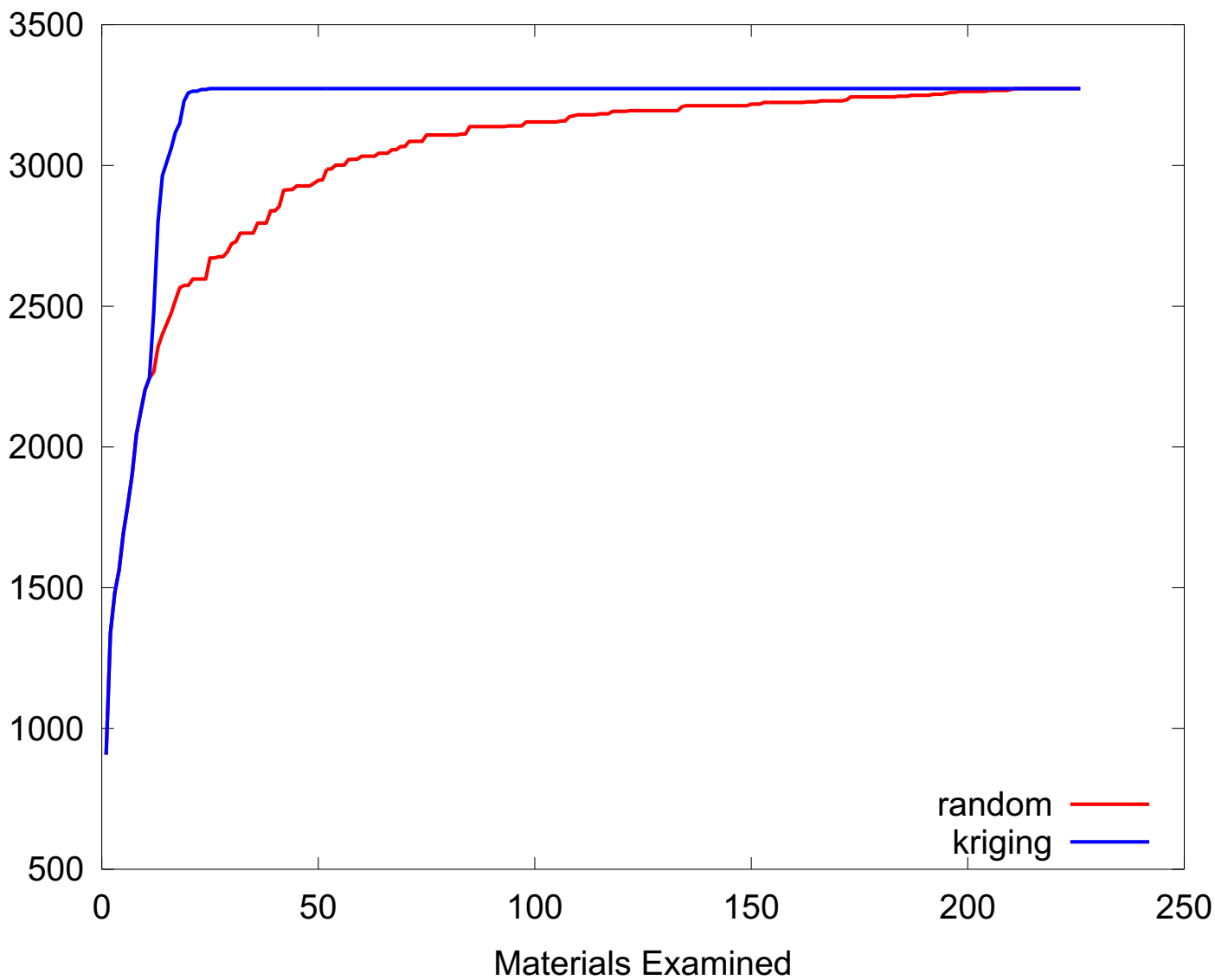
#n3:構成元素の価電子数の和

#p1:構成元素の周期の二乗和

#p2:構成元素の周期の積

#p3:構成元素の周期の和

融点の最高値



観測数

最高融点の材料を見つけ出すまでの 平均観測数

ベイズ最適化
16.1回

ランダム
133.4回

ベイズ最適化による実験順

- AlBr₃ - As₄S₄ - GeSe - Se - BaSe - SnO₂ - Sb₂S₃ - Sb₂Te₃ - Pb - SnF₂ - GeBr₂ - SnSe₂
- BaO - BaS - SrSe - SiC - BeO - **[AlN]** - Be₃N₂ - Al₂O₃ - Si₃N₄ - Al₄C₃ - MgO - CaO
- CaC₂ - LiH - Cs - Be - BaH₂ - Bi₂O₄ - K - BeF₂ - Tl - RbN₃ - LiF - PbTe - CsI - Li - P₂O₅
- Ti₂O₃ - BaF₂ - Bi - Ba - CaS - SrO - CaSi - PbO - CaF₂ - Rb - MgH₂ - Si - BaSi₂ - IBr -
Bi₂O₃ - SrS - NaF - Ga₂O₃ - Al - TlI - CsO₂ - KCl - In - I₂ - BiF₃ - SrF₂ - LiCl - InN - CsBr
- ICl - SrH₂ - Pb₃O₄ - Na - Na₂O₂ - In₂O₃ - RbI - S - PbF₂ - Bi₂Te₃ - Sn - CaH₂ - KF -
InSb - Ca - BiI₃ - CsCl - K₂O₂ - MgF₂ - Ge - PbS - SrSi₂ - TeO₂ - TlSe - Sr - BaI₂ - AlP -
Li₂O - RbO₂ - CsF - P₄S₃ - BiF₅ - Mg - GeO₂ - NaCl - CaSi₂ - BaCl₂ - Te - PbSe - TeF₄ -
PbI₂ - TlF - KI - P - MgS - SnTe - NaO₂ - GaAs - RbCl - Ti₂O - SiS₂ - KO₂ - InAs - BaBr₂
- P₂S₃ - Sb - KBr - TeI₄ - Li₃N - TeO₃ - RbBr - SiI₄ - LiBr - GaSb - TlCl - SeO₃ - GaP -
RbF - SnI₄ - Cs₂O - As₂O₃ - SrCl₂ - Mg₂Si - TlBr - AlAs - LiI - P₄S₇ - Bi₂S₃ - Mg₂Sn -
CaCl₂ - AlI₃ - As₂O₅ - SnSe - Ca₃N₂ - Li₂S - NaBr - InI₃ - BeCl₂ - Sb₂O₃ - NaI -
Mg₂Ge - InI - BiBr₃ - GeS - BeI₂ - SeBr₄ - Ti₂S - InP - GaTe - P₂S₅ - SbF₃ - K₂S - BiCl₃
- SrBr₂ - InF₃ - GeTe - SbI₃ - AlSb - In₂Te₃ - GeF₂ - Mg₃Sb₂ - SrI₂ - PbCl₂ - GaS - PI₃
- Na₂S - SnS - Al₂S₃ - GaI₃ - Rb₂S - GaSe - MgCl₂ - TeCl₄ - Rb₂Se - PbBr₂ - GeI₄ -
K₂Se - CaI₂ - BeBr₂ - P₂I₄ - Sb₂Se₃ - CaBr₂ - As₂Te₃ - In₂Se₃ - AlCl₃ - InS - GeBr₄ -
As₂S₃ - Ga₂Se₃ - SnBr₄ - InCl - As₂Se₃ - AsBr₃ - AsI₃ - GaBr₃ - Al₂Te₃ - In₂S₃ -
SbBr₃ - MgI₂ - InBr₃ - GeS₂ - MgBr₂ - Ga₂S₃ - GaCl₃ - SbCl₃ - SnBr₂ - GaCl₂ - SnCl₂
-