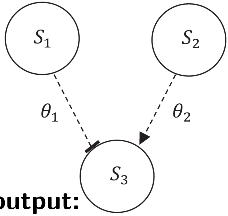
MOCU: motivating example



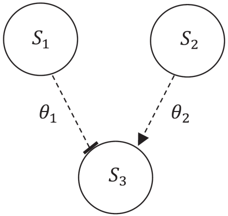
- **System dynamics and output:**

$$R(S,\psi) = \psi_1\theta_1 S_1 + \psi_2\theta_2 S_2 \quad , \quad (S_1, S_2) \sim \rho(S_1, S_2)$$

$$S_3 = \begin{cases} 1 & \text{if } R(S,\psi) > 0 \\ 0 & \text{if } R(S,\psi) < 0 \\ 1(\text{prob } 0.5) & \text{if } R(S,\psi) = 0 \end{cases}$$

- **Uncertainty:** $\theta_i \in \{-1, 1\}$, $\theta_i \sim \rho(\theta_i)$
- **Intervention:** $\Psi = \{(1,0),(0,1)\}$
- **Cost:** $C(\theta,\psi) = S_3$ (we don't want S_3 activated)
- **Experiment:** "probe" x_i measures y_j probability $\rho(y_j|x_i,\theta_k)$

MOCU: motivating example, informal discussion



- **Given:**
  - Uncertain system dynamics
  - User-specified control/intervention
  - Cost of intervention under uncertainty
  - Experimental measurement (assumed to be expensive)
- **Question:** what experiment should be made next that (1) reduces uncertainty and (2) is optimal across all uncertainty w.r.t. the cost?

MOCU: Algorithm and Pseudocode

inputs : set of $\Theta = \{\theta_1 \ldots \theta_k\}$ with $\rho(\Theta)$, computer model $\hat{y} = f_m(x, \theta)$, set of experimental choices $X = \{x_1 \ldots x_n\}$ with possible outcomes $Y = \{y_1 \ldots y_m\}$

Monte Carlo sample the computer model over $X \times Y \times \Theta$ and approximate $\rho(Y|X, \Theta)$

for all x_i in X:

  for all y_j in Y:

    Compute $\rho(\theta|x_i = y_j)$ via Bayes with priors $\rho(\theta)$, $\rho(y_j|x_i, \theta)$

    Compute $\psi(\Theta|x_i = y_j) = \text{argmin}_\psi \; \mathbb{E}_{\theta|x_i=y_j}[C(\theta, \psi)]$

    Compute $\omega(x_i = y_j) = \mathbb{E}_{\theta|x_i=y_j}[C(\theta, \psi(\Theta|x_i = y_j))]$

  Compute $\mathbb{E}_{y|x_i}[\omega(x_i)]$ (i.e., averaged over the m outcomes in Y)

Compute $x^* = \text{argmin}_X \; \mathbb{E}_{y|x}[\omega(x)]$ (i.e., minimization over the n experiments in X)

outputs : x^*

Application: BCPs

How does this map to BCPs and materials discovery?

- **Uncertain system dynamics:** Cahn-Hilliard with uncertain RHS terms added (“model inadequacies”, closures)

$$\delta_t c = f_{ch}\left(c; \epsilon^2(T,\theta), \sigma(T,\theta), m\right)$$

  - Example: $\epsilon^2(T,\theta) = \epsilon^2_{scft}(T) + \theta\epsilon^2(T^2) + \theta^2\epsilon^2(T^3) + \dots$
  - Example: $\epsilon^2(T,\theta) = \epsilon^2_{scft}(T) + \theta g(x_{micro})$, x_{micro} are microstate variables, perhaps computed locally with MD
- **Intervention:** $\Psi = \{T, m, c(\boldsymbol{x}, t_0), c(\boldsymbol{x} \in d\Omega, t)\}$
- **Cost:** $C(\theta,\psi) = d\left(c(\boldsymbol{x}, t_f; \theta, \psi) - c^*(\boldsymbol{x})\right)$ (distance from target structure)
- **Experiment:** $X = \{c(\boldsymbol{x}, t_0), c(\boldsymbol{x} \in d\Omega, t)\}$,
  $Y = \{$close-packed spheres , cylinders , lamellae , gyroids$\}$, $\psi = \{T^*, m^*\}$ (fixed operating conditions)

Test problem: Toy SDE

Let's start simple...

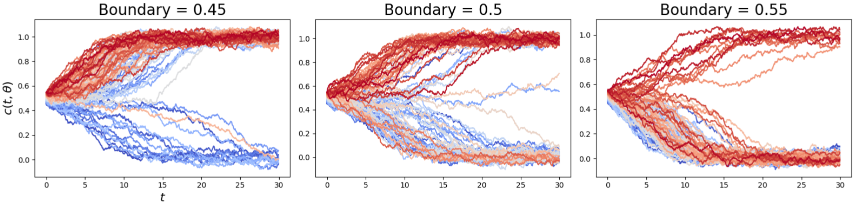
- **Uncertain system dynamics:**

$$dc = \lambda\, c(c-1)\left(c - (a|\psi - \theta|^2 + b)\right)\; dt + \sigma\, dW$$

$$dW\ ,\ dW \sim \mathcal{N}(0, 1^2)\ ,\ \theta \in \Theta = \{0, 0.5, 1\}\ ,\ \rho(\Theta)$$

- **Intervention**
  - $\psi \in \Psi = \{0, 0.5, 1\}$
  - Material design option that affects the final "phase" of a given IC
- **Cost:** $C(\theta, \psi) = \|c(t_f; \theta, \psi) - 1\|$ (distance from desired final "phase")
- **Experiment**
  - $c(t_0) \in X = \{a, 0.5(a+b), a+b\}$ (initial condition)
  - $c(t_f) \in Y = \{0\ ,\ 1\}$ (final phase)
  - $\psi^* = 0$ (fixed operating conditions)
  - $\theta^* = 0.5$ (true, "unknown" value of θ in the "real" system)

Test problem: Toy SDE



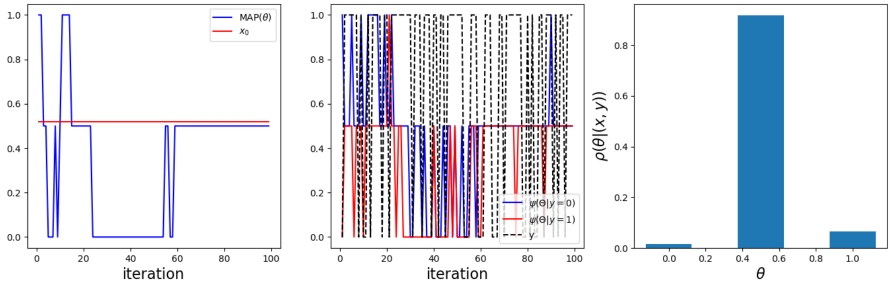
- Small $|\psi - \theta| \mapsto$ low boundary, best for getting phase 1
- Large $|\psi - \theta| \mapsto$ high boundary, best for getting phase 0

Test problem: Toy SDE

Use some intuition...

- Our goal is to design materials that get to phase 1
- Optimal experiment
  - The bigger the quantity $(c(t_0) - [\text{boundary}])$, the more certain we can be that $c(t_f) = 1$, regardless of the value of θ
  - Pick the highest IC value $c(t_0)$
- Optimal intervention
  - Small $|\psi - \theta| \mapsto$ low boundary, best for getting phase 1
  - Pick ψ to minimize $|\psi - \theta|$ over average weighted by $\rho(\theta)$

Test problem: Toy SDE



- Plot 1
  - MAP estimate correctly converges to true value $\theta^* = 0.5$
  - Optimal experiment always correct (highest possible IC value)
- Plot 2
  - Dash-black: experimental observations
  - Blue/Red: optimal interventions conditioned on an observation
- Plot 3: Posterior distribution for θ (centered around correct value)