Control of Quantum Dynamics Phenomena: Analyses, Options and Opportunities

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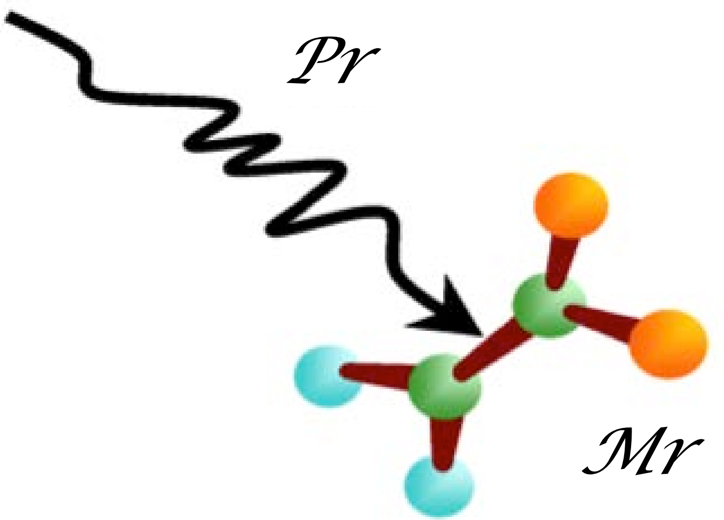
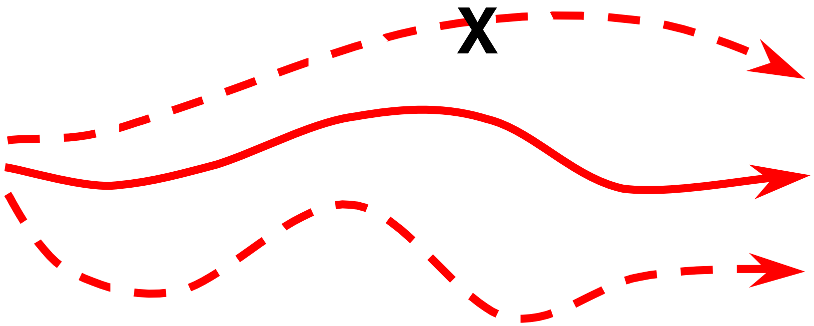


- $\mathcal{P}r$ Photonic Reagents as Controls
- $\mathcal{M}r$ Material Reagents as Controls

Control of Quantum Systems



ed Target

Control

Why explore this subject?

- Manage dynamical events at the atomic and molecular scale

- Reveal fundamental insights into dynamics

Applications and technology transfer

Quantum Control

- **Goal A:** Optimally achieve control objective
- **Goal B:** Understand control mechanism

Better Physical Understanding ⟷ **Better Control Performance**

How to identify optimal control solutions?

$$J_{\mathrm{opt}} = \max_{\varepsilon(t)} J[\varepsilon(t)]$$

Open-loop control

Find optimal controls for theoretical model

Apply theoretical optimal control designs to actual system in laboratory

Closed-loop control

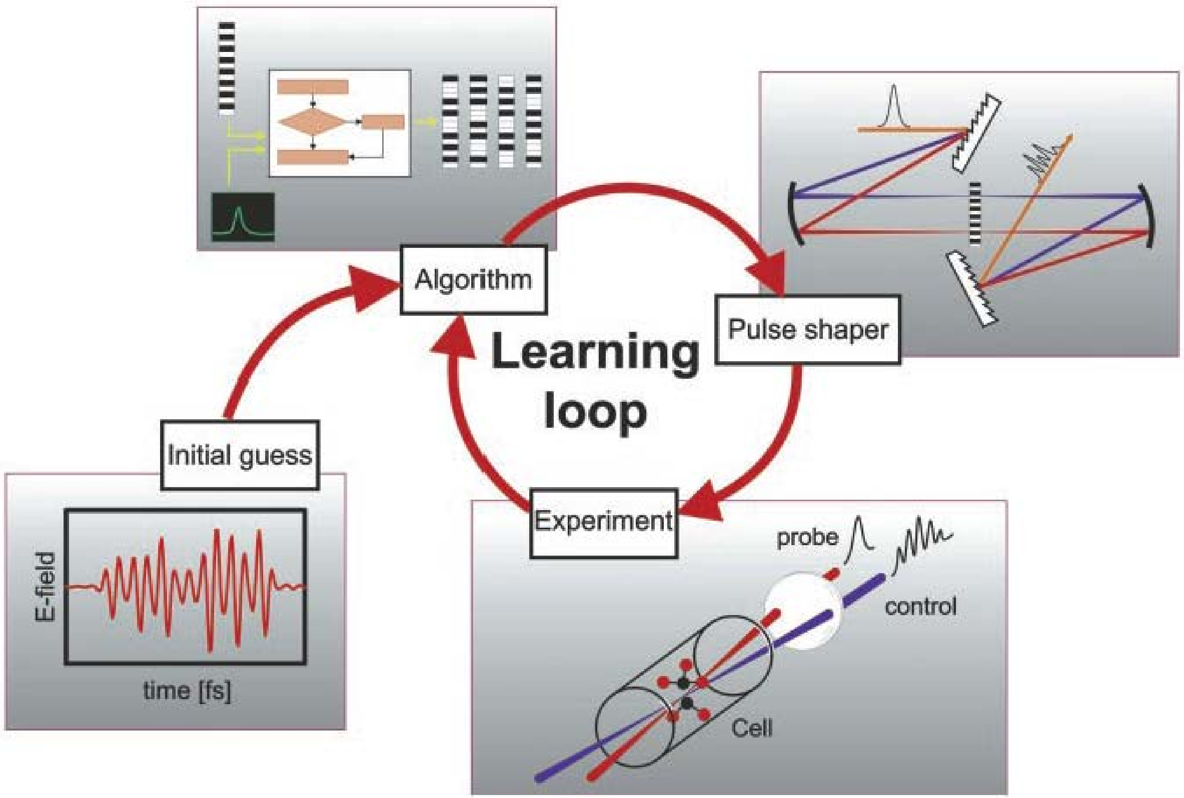
Find optimal controls directly in laboratory using feedback signal from controlled system

Adaptive feedback control
Measure control objective to guide optimization. Reset system after each measurement.

Real-time feedback control
Measure signal from system and use in real time to select next control action. Measurements affects system dynamics

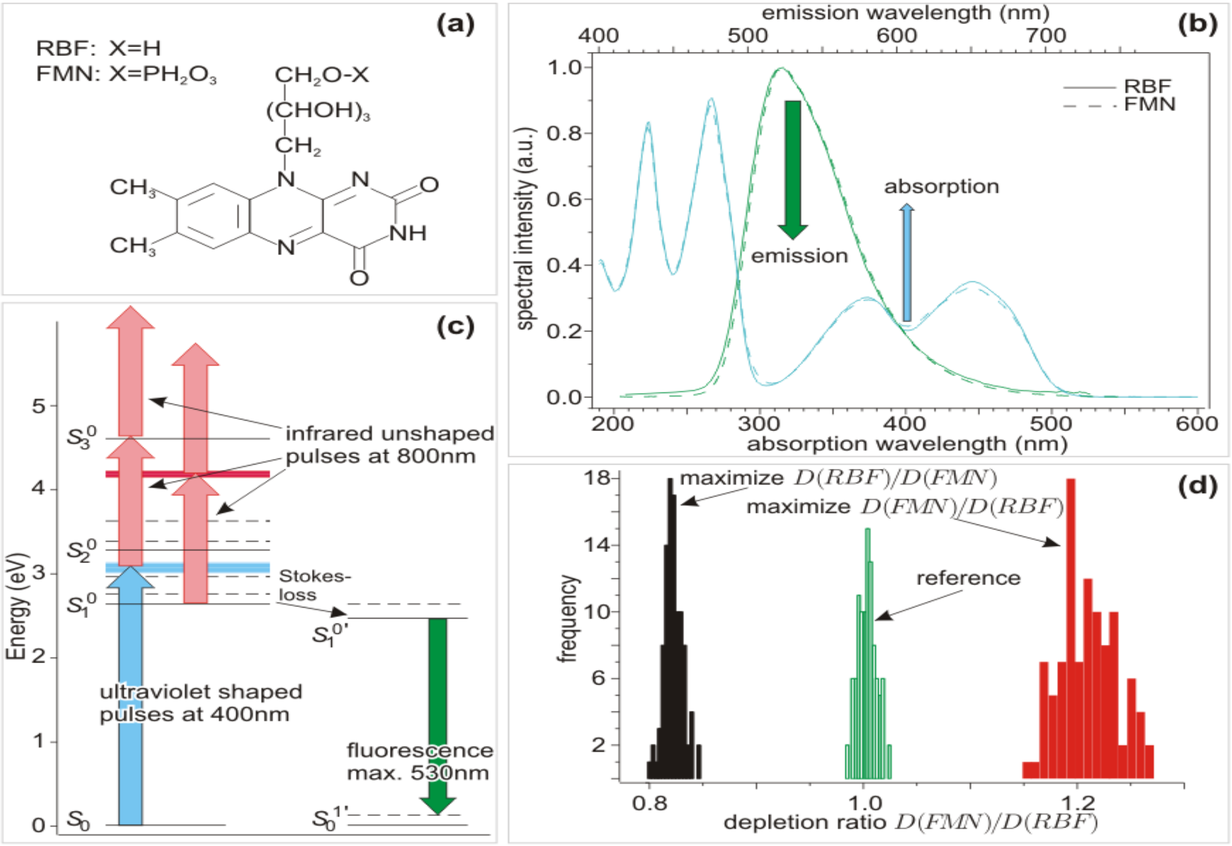
Coherent feedback control
Coherently process signal from controlled quantum system (“plant”) by an auxiliary quantum system (“controller”)

Automated Optimization of Quantum Phenomena



Pushing the Limits of Quantum Control

- *Static* spectroscopic methods fail to differentiate RBF and FMN



- Create distinct spectral signatures by optimal tailoring of the *dynamics*

Domains Subjected To Optimal Quantum Control

1. Atomic excitation
2. Vibrational excitation
3. Vibronic excitation
4. Selective isotope excitation
5. Discrimination of very similar molecules
6. Ionization, single to multiple
7. Molecular isomerization
8. Molecular fragmentation
9. Molecular alignment
10. Nonlinear spectroscopy
11. Ultrafast semi-conductor optical switching
12. Biomolecular energy transfer
13. Filimentation
14. Pulse propagation is nonlinear media
15. Decelerating and trapping of molecules
16. Surface ablation
17. Subsurface imaging
18. Decoherence mitigation

Comments

- 5~10 examples/domain
- Control mechanism often hard to discern
- Rules to be discovered
- How “quantum” is the dynamics?

Results from Seeking Optimal Fields

- Optimal control simulations typically attain excellent yields

- ~200 optimal control experiments with good-excellent yields (constrained controls)

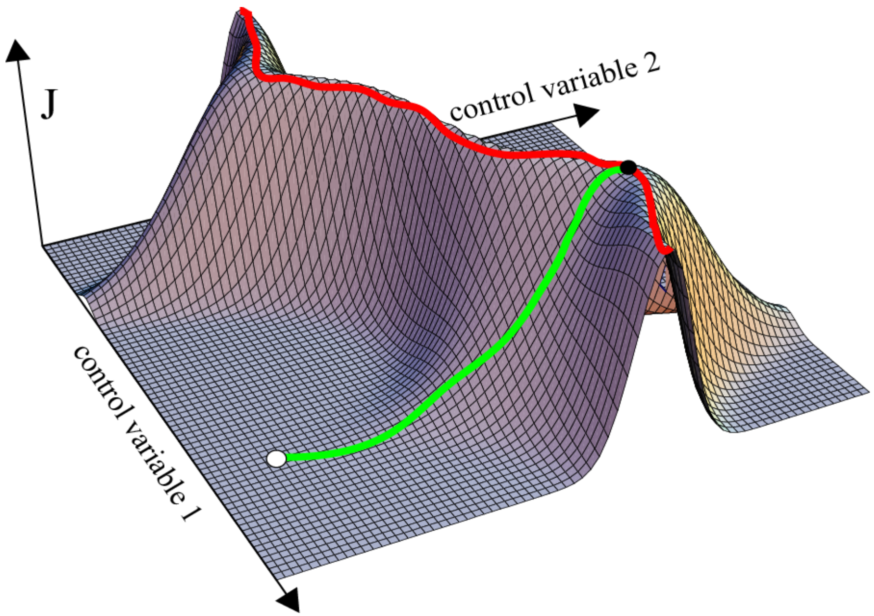
Common finding: ~10^3-10^4 iterations to optimize, despite the search space being of size ~10^{100}

Why the good fortune*?

* *Beyond overhead*

Why is the Search for Optimal Control Fields so Efficient?

- Physical Objective $J[\varepsilon(t)] = Tr\{\rho[\varepsilon(t)]O\}$



Landscape Features determine the ease of finding a successful optimal control $\varepsilon(t)$

- Topological Features: critical points, $\delta J / \delta\varepsilon(t) = 0, \forall t$
- Structural Features: non-topological “twists and turns”

Landscape Analysis

Assumptions

$$i\hbar \frac{\partial U(t)}{\partial t} = [H_0 - \mu\varepsilon(t)]U(t)$$

a) The system is controllable

b) No (significant) constraints are placed on the controls

c) The control $\rightarrow$ state map is full rank: $\delta U(T)/\delta\varepsilon(t) = \frac{-i}{\hbar}U(T)U^{\dagger}(t)\mu U(t)$

Conclusion

➢ No sub-optimal traps exist on the control landscape

- When are the assumptions satisfied?

Are Traps Lurking on Quantum Control Landscapes?

Numerical Simulations: employ a myopic algorithm

$P_{i\to f}$ ~20,000 runs

$Tr[\rho O]$ ~15,000 runs

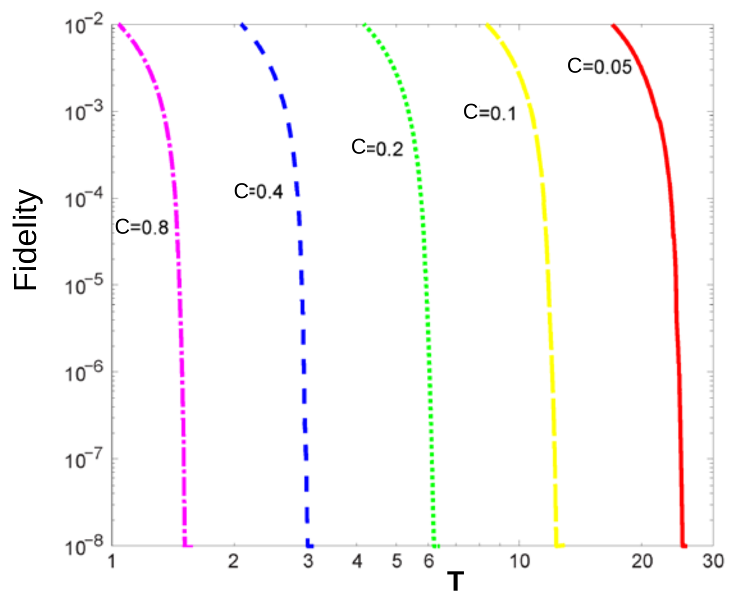
$\|W - U\|$ ~50,000 runs

All reached full fidelity (~0.999)!

Impact of control constraints

- Mild constraints can still permit a trap free climb of the landscape
- Significant constraints will fracture the landscape
- Even the target time T is a control resource

C=Coupling strength



Qubit System

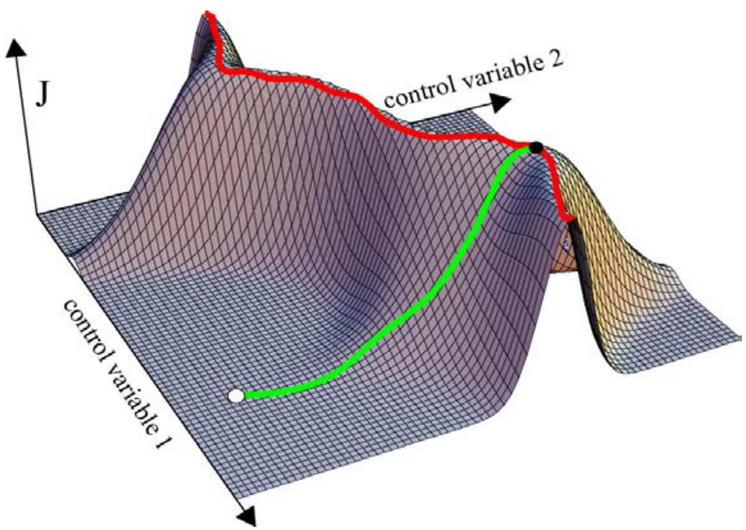
Does Discovery of an Optimal Field Follow a Tortured Path through Control Space?

- Under reasonable assumptions, there are no traps lurking on control landscapes

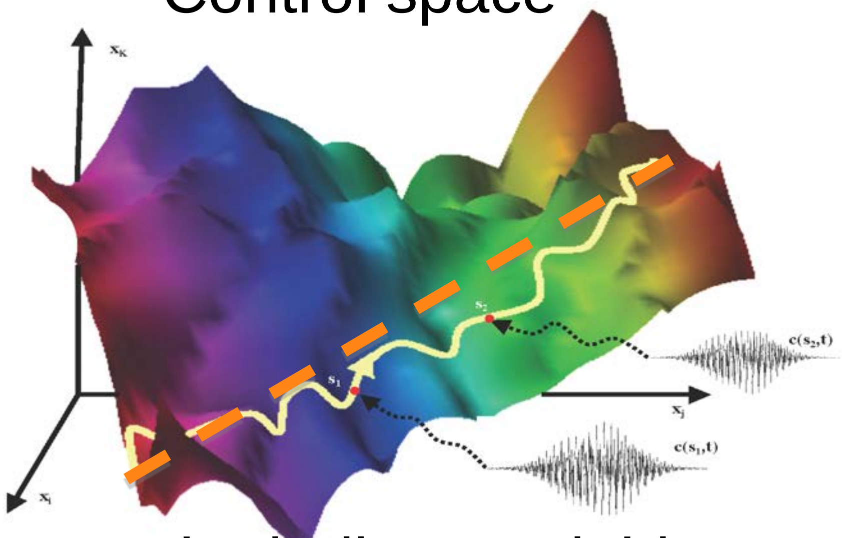
- But, is the path to the top gnarled?

$$R = \frac{Path\ Length}{Euclidean\ Distance} \geq 1$$

Observable landscape



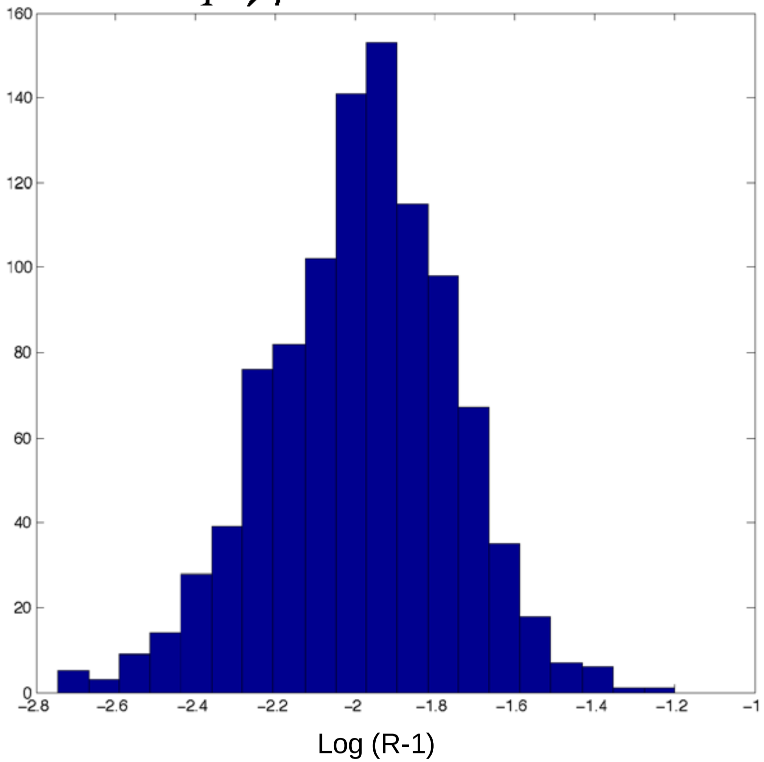
Control space



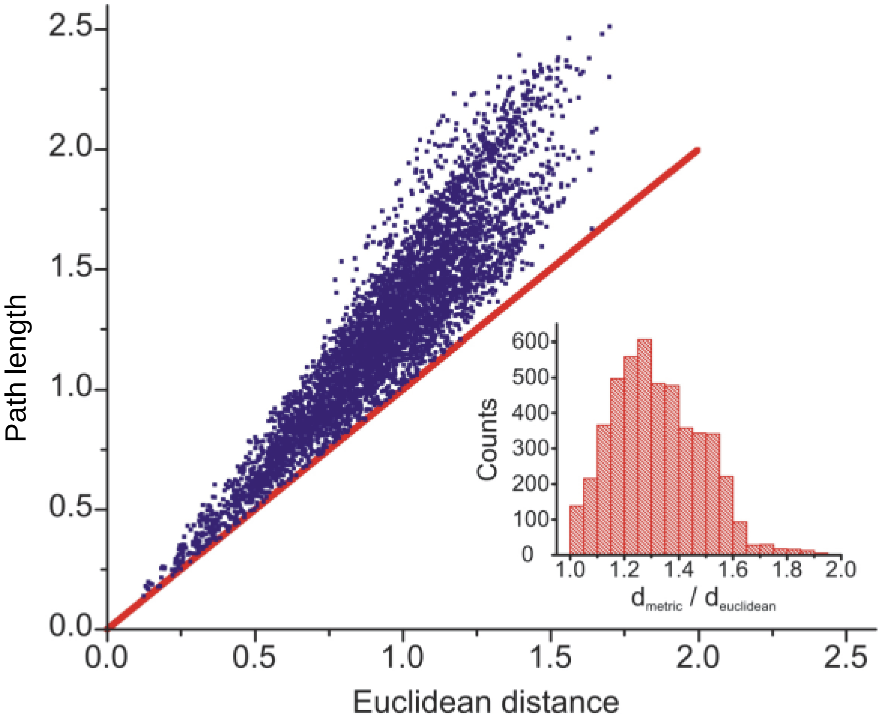
Color indicates yield

Control Space is Dense with Nearly Straight Shots to the Top of the Landscape

$P_{1\to 4}$ Simulation



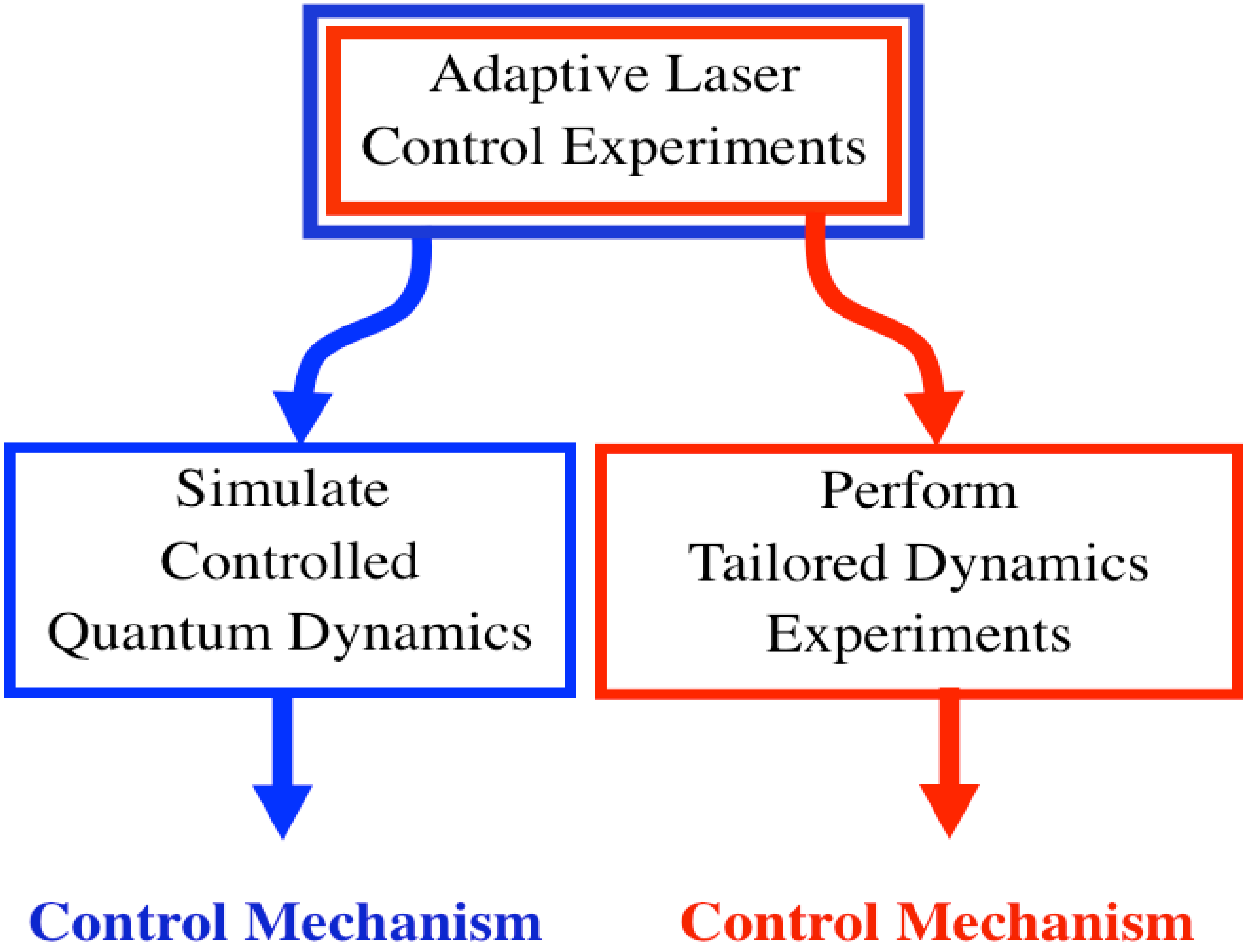
Frequency Doubling Experiment



Both cases with random initial fields

- **Control space looks like "Quantum Swiss Cheese" with easy passages to traverse**

Revealing Mechanisms of Laser-Controlled Dynamics



Revealing Quantum Control Mechanisms

Perspective: The quantum system + shaped laser pulse acts as a complex functioning machine

Problem: Deduce the machine operating mechanism without numerically solving the system Schrödinger equation

Solution: Modulate ε(t) with a pseudo time-like variable s

Approach:

- Encode a "secret" message m(s) in the Hamiltonian
- Decode the nonlinear distortion of the message in observations

$$H_0 \xrightarrow[\text{Control}]{\text{Optimal}} H \xrightarrow[\text{Hamiltonian}]{\text{Encode}} H\left[m(s)\right] \xrightarrow[\text{Dynamics}]{\text{Controlled}} O\left[m(s)\right] \xrightarrow[\text{Observations}]{\text{Decode}} \left\{U_{if}^{\ell}\right\}$$

Mechanism revealed by identification of the dynamical amplitudes $\left\{U_{if}^{\ell}\right\}$

ℓ =

Implementation of Hamiltonian Encoding - Observable Decoding for Control Mechanism Analysis

Formulation

$$\langle O \rangle = Tr[U\rho U^{\dagger} O]$$

Dyson expansion

$$U = \sum_{n=0}^{\infty} U^{n}$$

$$U^{n} = \left(\frac{-i}{\hbar}\right)^{n} \int_0^T \int_0^{t_{n-1}} \dots \int_0^{t_1} \mu(t_n)\varepsilon(t_n)\dots\mu(t_1)\varepsilon(t_1)dt_1\dots dt_n$$

$$\varepsilon(t) = \sum_p a_p \varepsilon_p(t)$$

Encoding $\varepsilon(t) \rightarrow \varepsilon(t,s) = \sum_p a_p h_p(s)\varepsilon_p(t)$

$h_p(s)$-*encoding function*

$$U \Rightarrow U(s) = \sum_{n=0}^{\infty} \sum_{\{p_l\}} h_{p_n}(s) h_{p_{n-1}}(s) \dots h_{p_1}(s) U^{n(p_1 \dots p_n)}$$

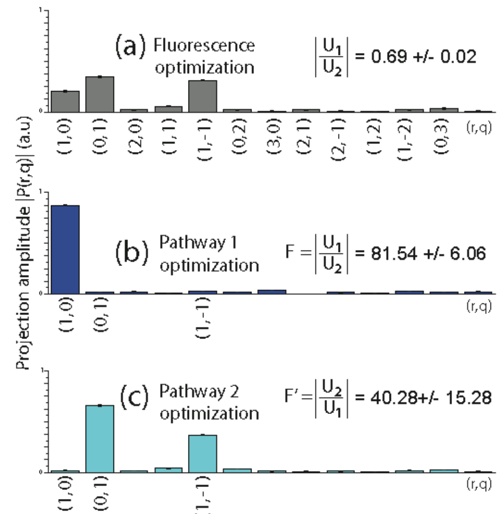
$$\langle O \rangle \Rightarrow \langle O(s) \rangle$$

Decoding: Measure $\langle O(s) \rangle$ to extract the amplitudes $U^{n(p_1 \dots p_n)}$

Identifying and Manipulating Quantum Control Mechanisms

- Mechanism identification by Hamiltonian encoding and observable decoding

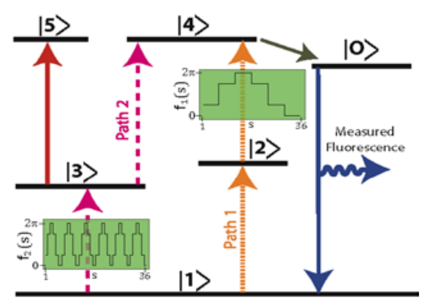
- Fast operation enables on-the-fly steering of the mechanism



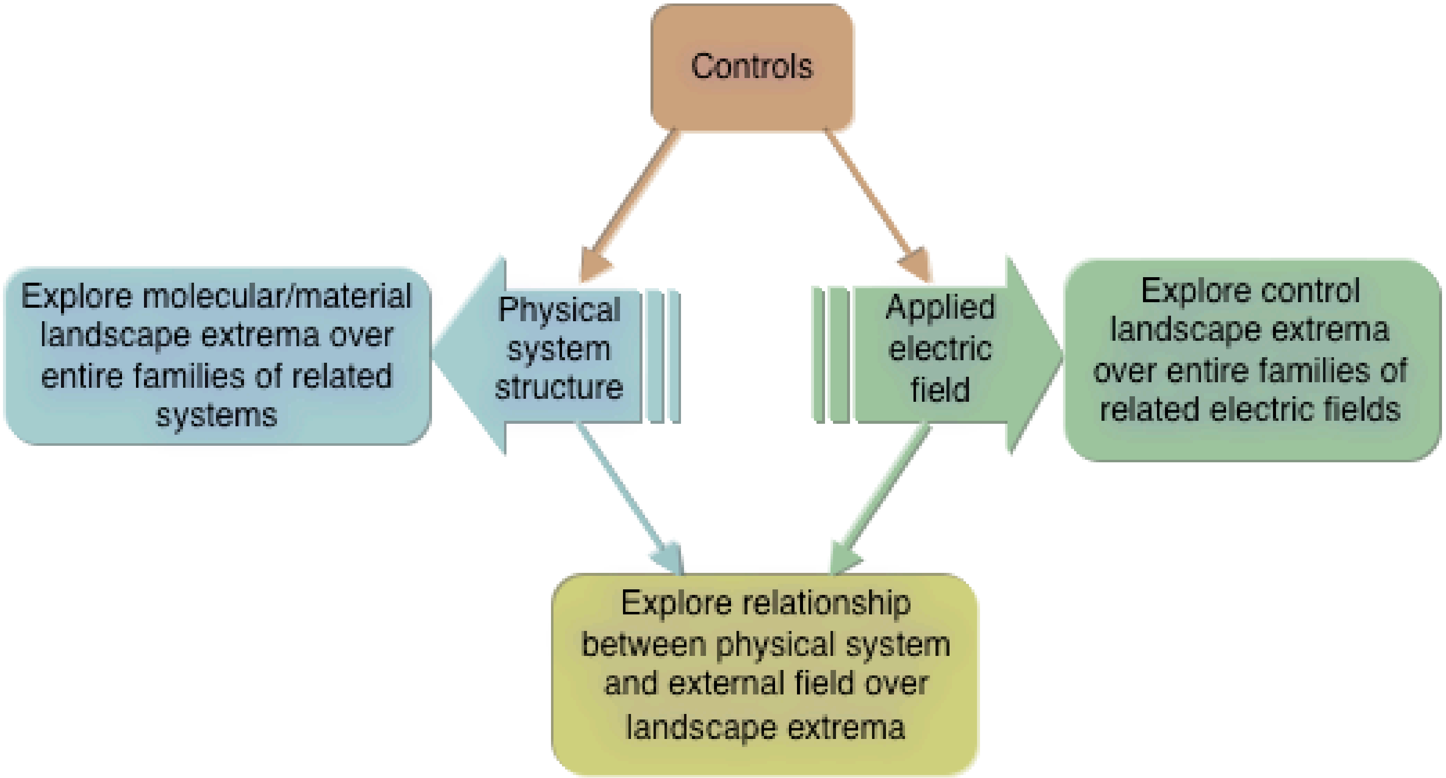
(i,j):

Atomic Rb

i-th order in control pathway 1
j-th order in control pathway 2



The Full Prospects for Control Resources



Mix and Match Control Resources from the Photonic ($\mathcal{P}$r) and Material ($\mathcal{M}$r) Reagent Stockrooms

Where is Optimization Encountered in the Sciences?

Everywhere!

Glimpse at Optimization:

- Quantum dynamics phenomena
- Chemical & material science
- Natural evolution
- Directed evolution
- ……

The Limit of Field Free Control: Chemistry

"I'm on the verge of a major breakthrough, but I'm also at that point where physics leaves off and chemistry begins, so I'll have to drop the whole thing."

"Chemistry is all about getting lucky" -Robert Curl

What is the truth about the nature of Chemistry?

Optimization in Chemistry & Material Science

Objectives

a. Optimize synthesis yield
b. Optimize property

Controls

Chemicals, solvents, catalysts, processing conditions

Experimental findings

Optimization in chemistry & material science are widely successful

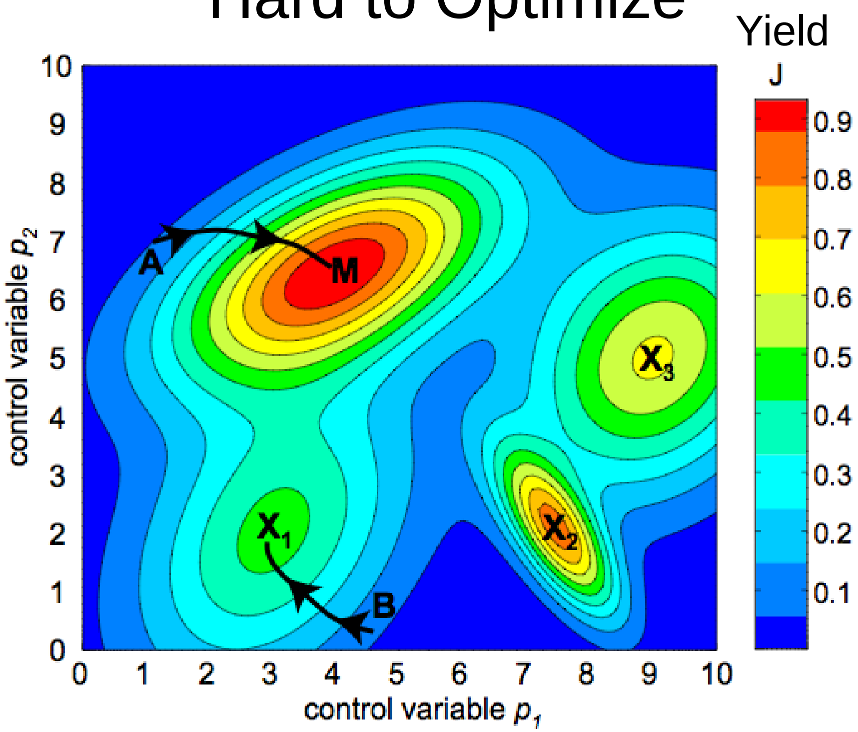
Automated Optimization of Chemical & Material Synthesis and Properties

Ref.	Var.	Objective	Expts.	Space
9	6	Binding to stromelysin	300	6.4×10^7
16	8	Propane → propene	328	NA
17	4	Inhibition of thrombin	400	1.6×10^5
18	8	Propane → CO	150	NA
19, 20	8	Propane → propene	280	NA
21	13	Propane → propene	60	NA
22	23	$NH_3 + CH_4 \rightarrow HCN$	644	NA
23	9	$CO \rightarrow CO_2$	189	NA
24	4	$CO + CO_2 + H_2 \rightarrow CH_3OH$	115	2.7×10^9
25	5	$3CO + 3H_2 \rightarrow C_2H_6O + CO_2$	160	2.4×10^{11}
26	6	$CO + CO_2 + H_2 \rightarrow CH_3OH$	235	4.7×10^9
27	10	*n*-Pentane isomerization	72	1.44×10^4
28	7	Propane → aldehydes	80	NA
29	8	Isobutane → methacrolein	90	10^9
30	8	Membrane permeability	192	9×10^{21}
31	4	Cyclohexene epoxidation	114	NA
32	3	Protein inhibition	160	10^{16}
33	6	Red luminescence	216	NA
34	7	Green luminescence	540	10^{14}
35	6	Color chromaticity	168	NA
36	8	Red luminescence	270	NA
37, 38	7	Red luminescence	1080	NA

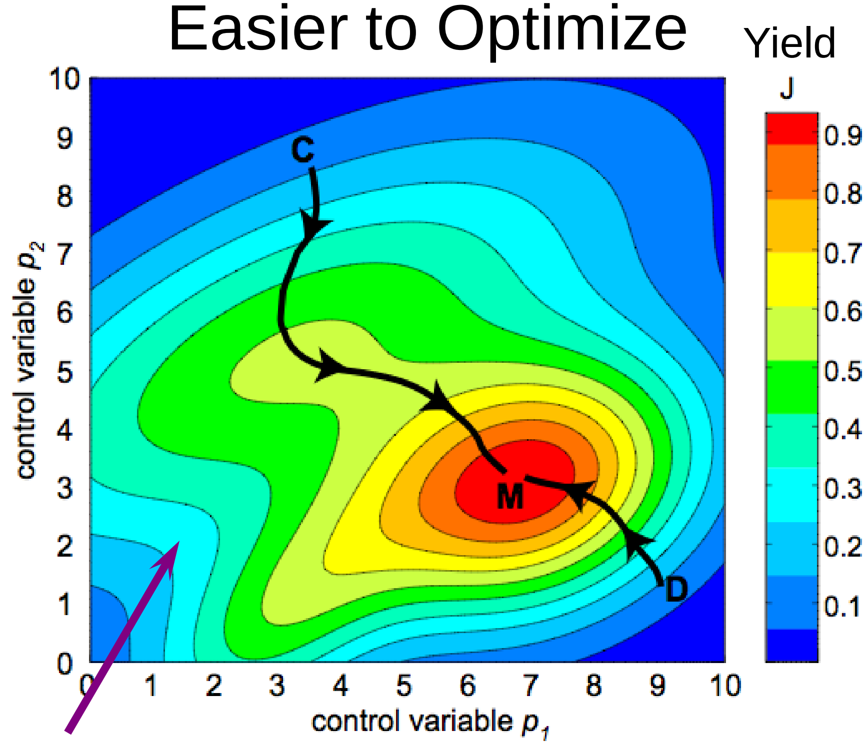
"The studied parameter space had $9x10^{21}$ possible combinations. A total of 192 polymeric solutions were synthesized…. producing a substantially improved membrane performance." **Jacobs (2006).**

What does the Chemistry Control Landscape Look Like?

Contains “Traps”
Hard to Optimize



Trap-Free
Easier to Optimize

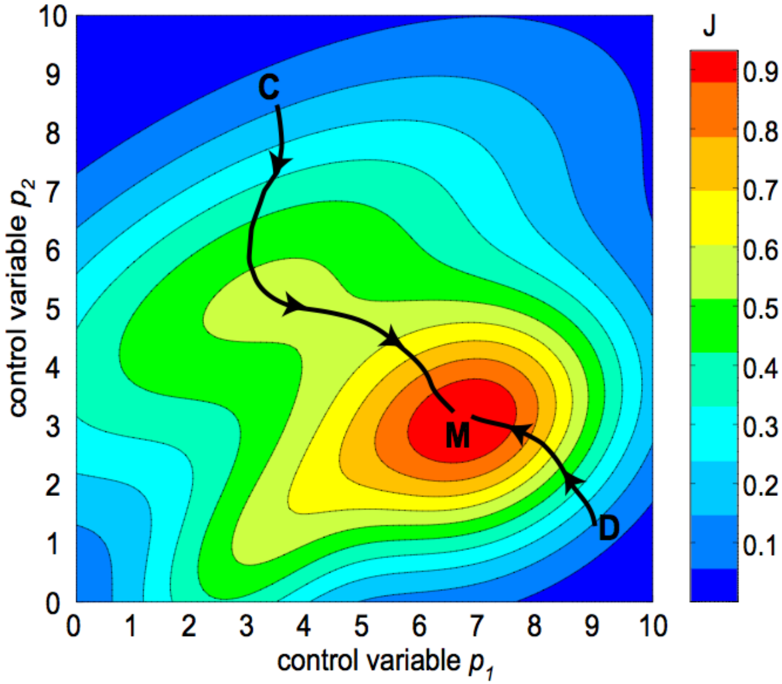


Chemical optimization looks like this!

OptiChem* : Chemistry has Trap-Free Landscapes!

- Convex analysis leads to the conclusion that the landscape should be trap-free

- Assumptions
  - Objective is well-posed
  - No significant constraints are placed on the controls
  - Freedom to move over the landscape



R. Wu et al., *J. Math. Phys.* **49** 022108 (2008)

K. Moore, A. Pechen, X. Feng, J. Dominy, V. Beltrani, and H. Rabitz, *Chemical Science* (2011)

***Opti**mization of syntheses and properties in **Chem**istry

Evidence for Trap-Free Landscapes in Chemistry

Synthesis Optimization Landscapes

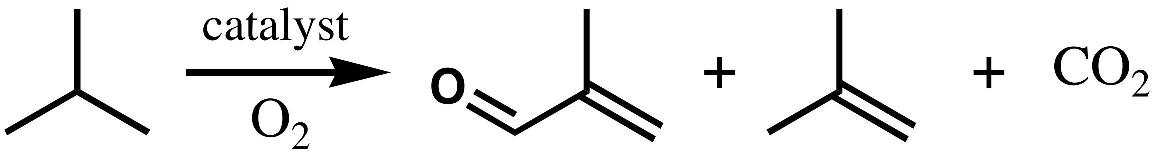
- Chemical products
- Material products

Property Optimization Landscapes

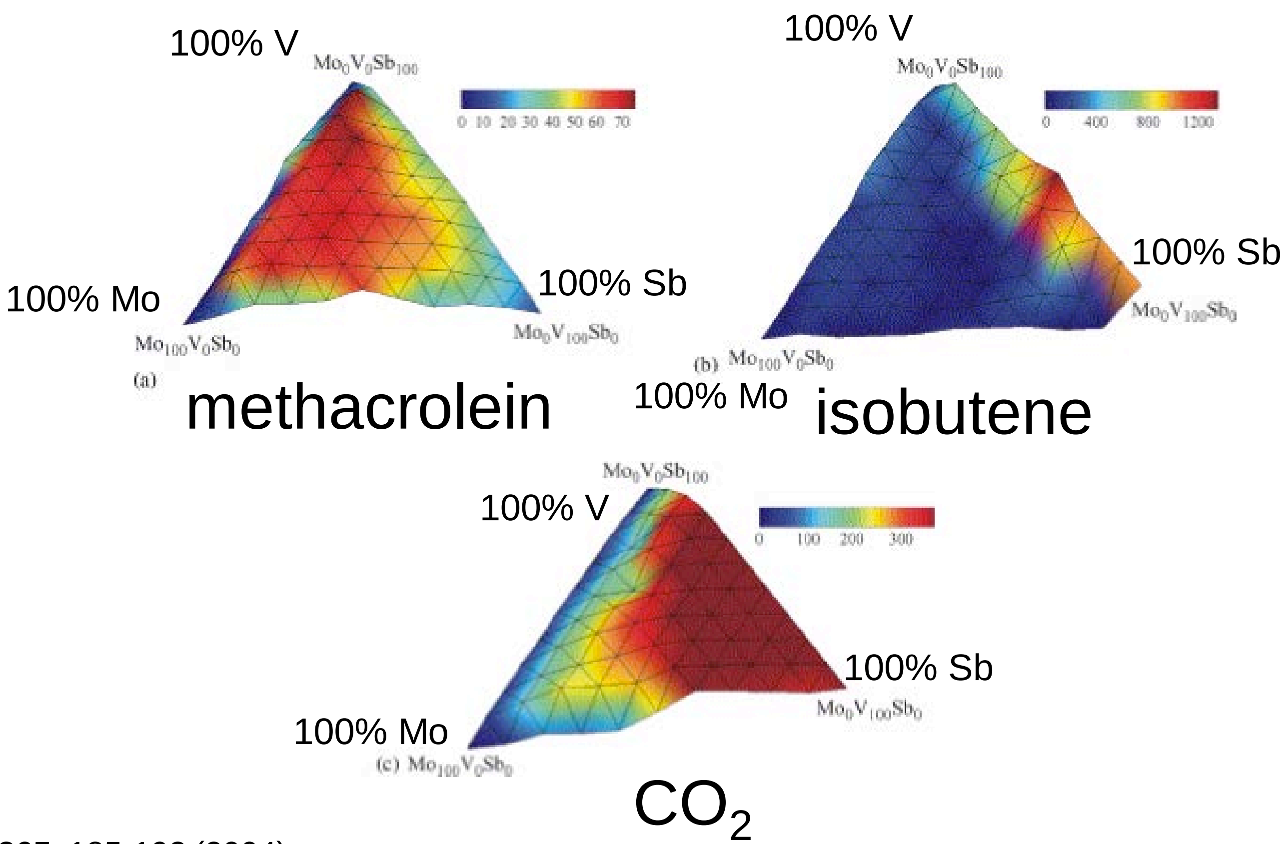
- Light emission and spectral properties
- Electrical properties of materials
- Mechanical properties of materials
- Substituent selection for protein binding

Hundreds of landscapes in the literature, ~90% are trap-free

Optimization of Catalyst Composition



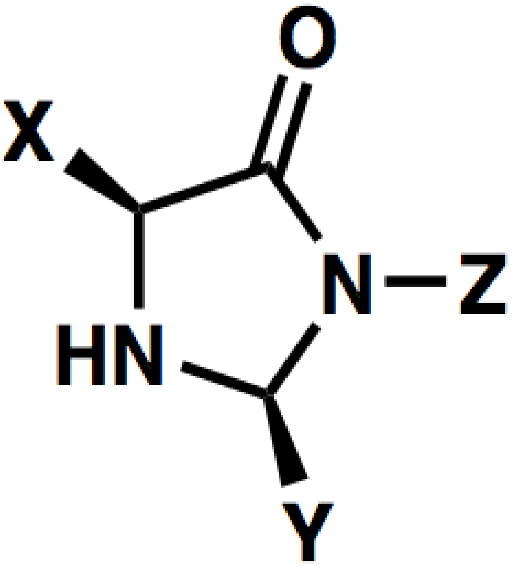
- Objective: maximize catalytic oxidation of isobutane
- Controls: mole fraction of Mo, V, Sb in catalyst



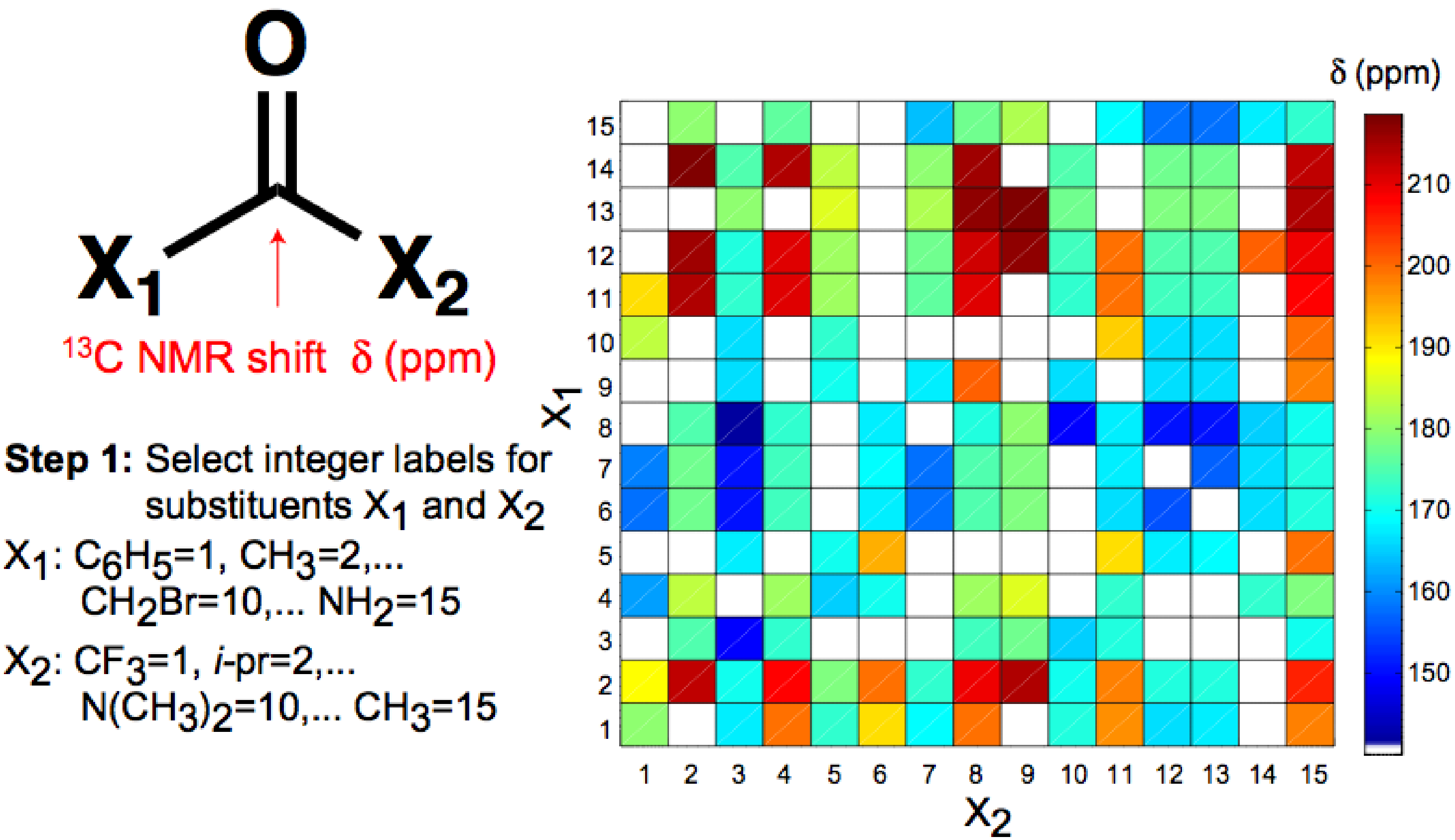
J. S. Paul, et al., *Applied Catalysis A* **265**, 185-193 (2004)

Molecular Discovery Landscapes

- **Goal**: Assess the landscape from a minimal set of property data
- **Means**: Optimally reorder the substituents on a scaffold
- **Outcome**: Estimate property of as yet un-synthesized molecules



Uncovering
Landscapes for Molecular Properties

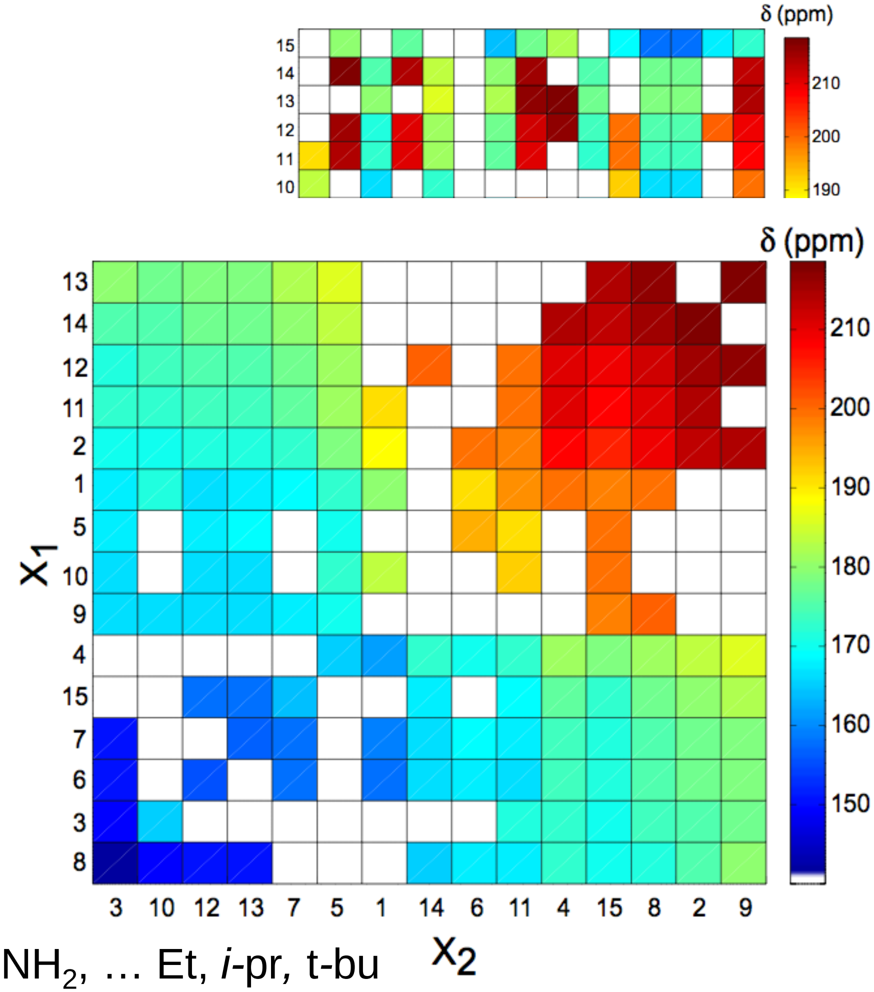


- Property: NMR shift
- Controls: Chemical substituents x_1 and x_2

Optimal Substituent Reordering

- **Goal**: Find ordering of integer labels for substituents that gives smoothest property landscape

- **Allowed movements**: reorder rows and columns of property landscape



Optimal order for x_1: Cl, $N(CH_3)_2$, OEt, OMe, NH_2, … Et, *i*-pr, t-bu

The “rules” of NMR are discovered by substituent reordering

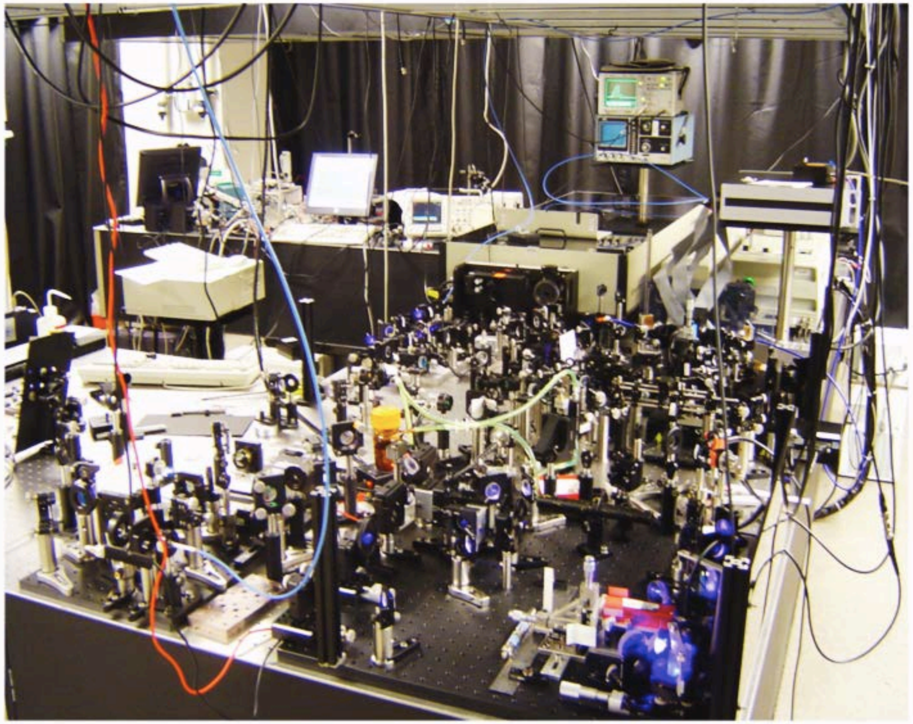
Features of Optimization

Domain	Control Variables	What Is Optimized	Outcome	Mechanism Understanding	Systematics	Laboratory Implementation
Quantum	Photonic	Yield, Fidelity	good	Initial stages	Rules emerging	Automated
Chemical & Material	Chemicals, Processing Conditions	Yield, Property	Good - Excellent	Good	Rule driven	Automated

- Number of experiments to reach optimality is generally small
- Yet, libraries of controls are enormous

Laboratories Controlling Events in Nano/Micro-World

Control Laboratory Employing Photonic Reagents



Control Laboratory Employing Molecular/Material Reagents

What is common here?

Optimal Control over Vast Length and Time Scales

- Is common high search efficiency a coincidence?
- Under optimization, is a unified picture operative?
- What are practical control consequences?

Control of Quantum Phenomena has a Special Role

The Control Landscape Principles Encompass Quantum and Classical Phenomena

Quantum Mechanics

$$\langle O_{qm} \rangle = Tr(\rho_{qm} O)$$

Classical Mechanics

$$\langle O_{cl} \rangle = \int dw \; \rho_{cl}(w) O(w)$$

- Control in both cases is a convex optimization problem, upon satisfaction of the key assumptions

➢ The associated control landscapes should be trap free

Implications of Common Control Landscape Behavior over Vast Length and Time Scales

- Systematic discovery of operational “rules” in the sciences

- Early identification of flawed experimental designs

- Curse of dimensionality may be overcome by *increasing* the number of variables

Unification

Assumptions

- Objective is well posed
- No significant constraints on resources (controls)
- Free movement on objective landscape
  (i.e., control $\rightarrow$ state map is full rank)

Satisfaction of the assumptions implies that the objective landscape should be trap-free

Optimal control of phenomena in the sciences share common landscape topology

Common Features of Optimization in Science

Science	Control Variables	What Is Optimized	Outcome	Mechanism Understanding	Systematics	Laboratory Implementation
Quantum	Photonic	Yield, Fidelity	good	Initial stages	Rules emerging	Automated
Chemical & Material	Chemicals, Processing Conditions	Yield, Property	Good - Excellent	Good	Rule driven	Automated
Natural Evolution	Genomic	Fitness	Excellent	Poor	Not clear	Micro-organisms (laboratory)
Directed Evolution	Genomic	Enzymatic activity	Good	Fair	Vague rules	Semi-automated

- Libraries of controls are enormous
- Yet, number of experiments to reach optimality is generally small

Why this good fortune?