RIVIC
Research Institute of
Visual Computing

School of
Computer Science
and Informatics
Cardiff University

Optimal Control of Chemical Reactions with Data-Driven Models

FC Langbein, Cardiff University

The EU Quantum Control Network

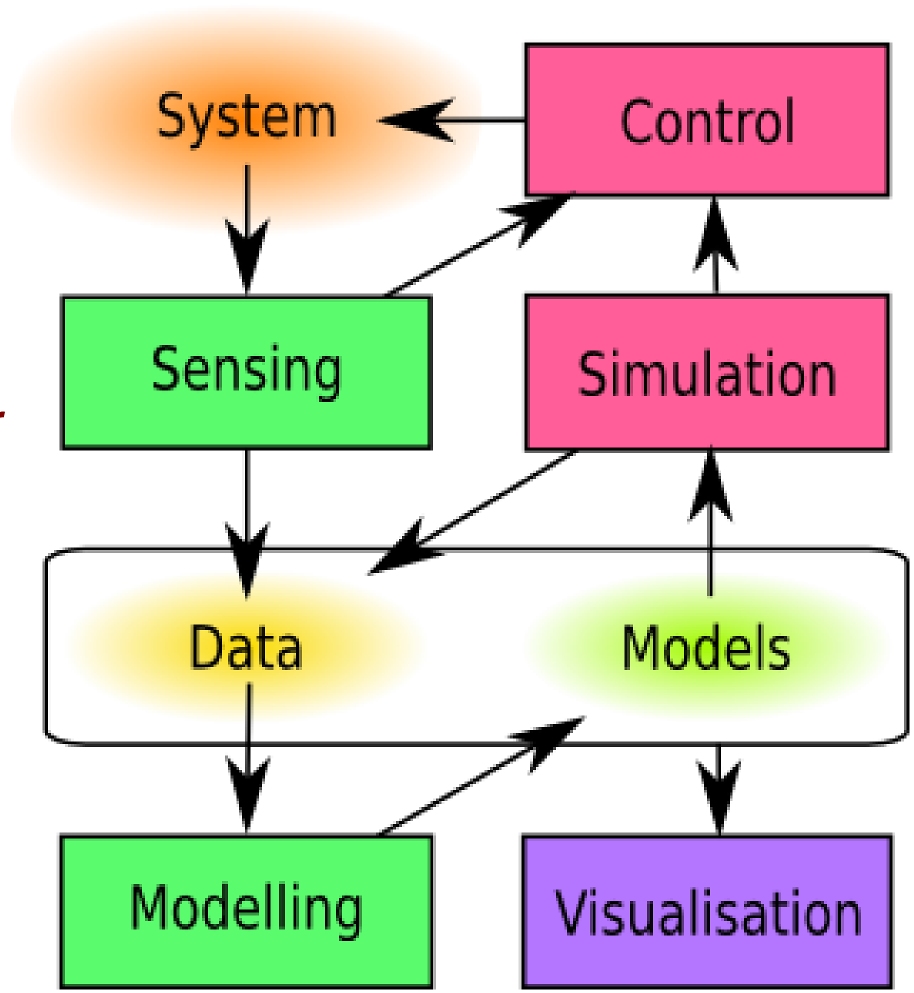
Optimal Control (of Quantum Systems)

Aim: *Design desired physical process* based on interference, entanglement, etc. with external controls such as magnetic, electric and laser fields

➔ Requires control with

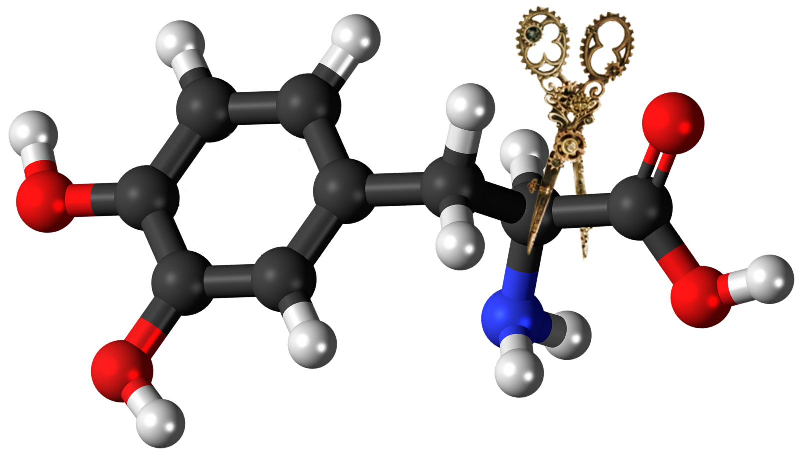
- high *precision* and *speed* to
- *accurately* and *deliberately steer* a quantum system
- while isolating it from *unwanted noise*

- Under *tough conditions* imposed by
  - instrumental sensitivity
  - power
  - timing
  - accuracy



Photonic Reagents

Idea: Laser selective photochemistry aimed to use narrow-bandwidth light in resonance with chemical bonds as *molecular scissors*



- *Not* so easy due to *intramolecular vibrational energy redistribution*

Exploit the quantum nature of light-matter interaction

Brumer, Shapiro

- Exploit *interference* between *different excitation pathways* leading to *same final state*
- Control laser fields to *enhance* one while *suppressing* others

P Brumer, M Shapiro, Annu Rev Phys Chem 43, 257, 1992.

Tannor, Kosloff, Rice

- *Pump* creates electronic wave package on excited-state energy surface
- *Dump* brings it down to ground state along desired channel

DJ Tannor, R Kosloff, SA Rice, J Chem Phys 85, 5805, 1986

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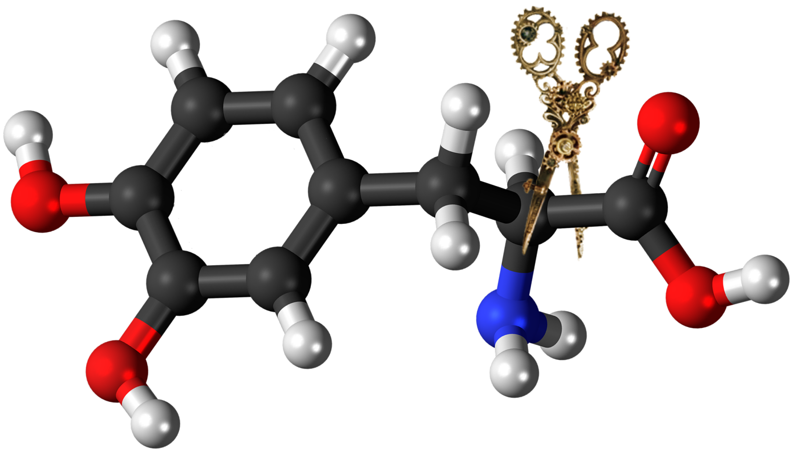

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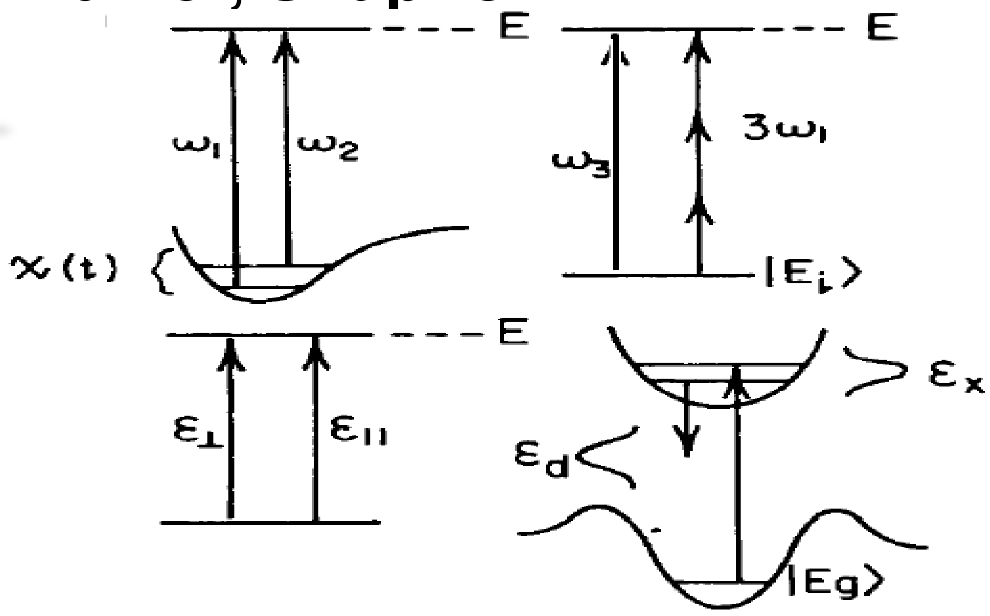

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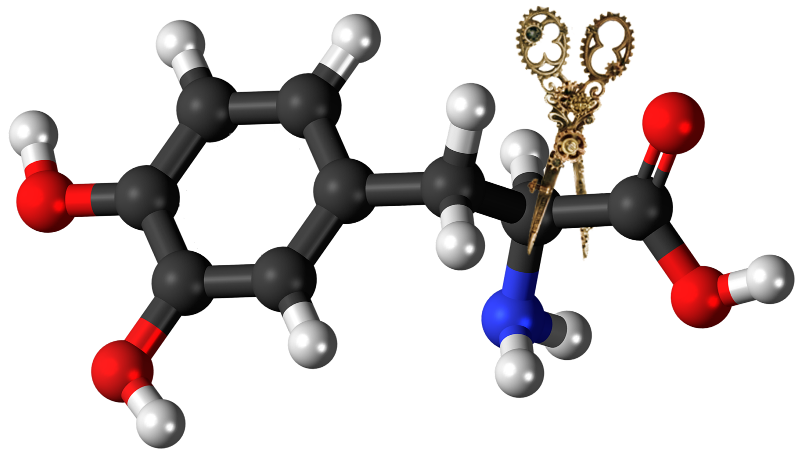

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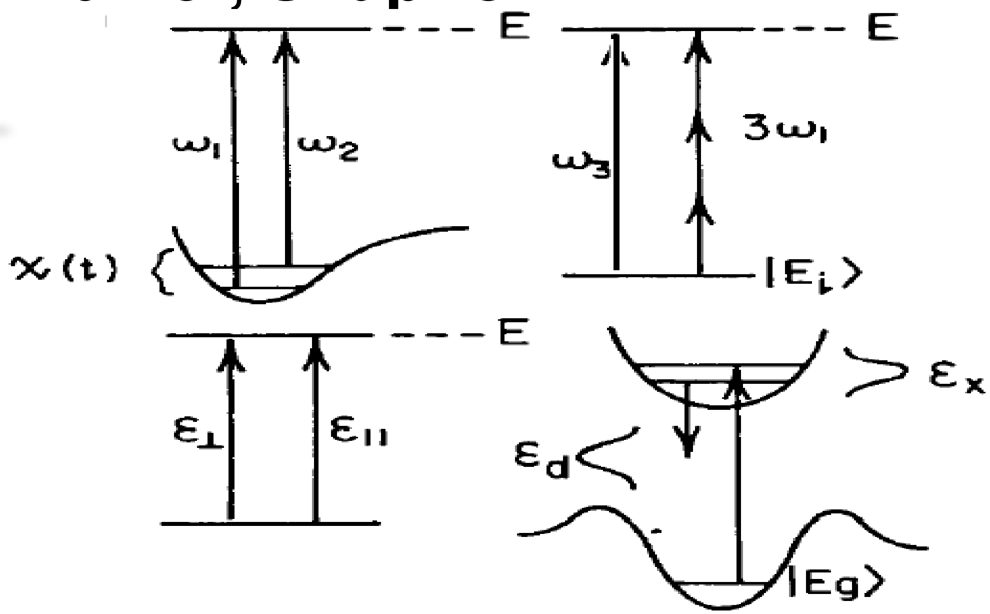

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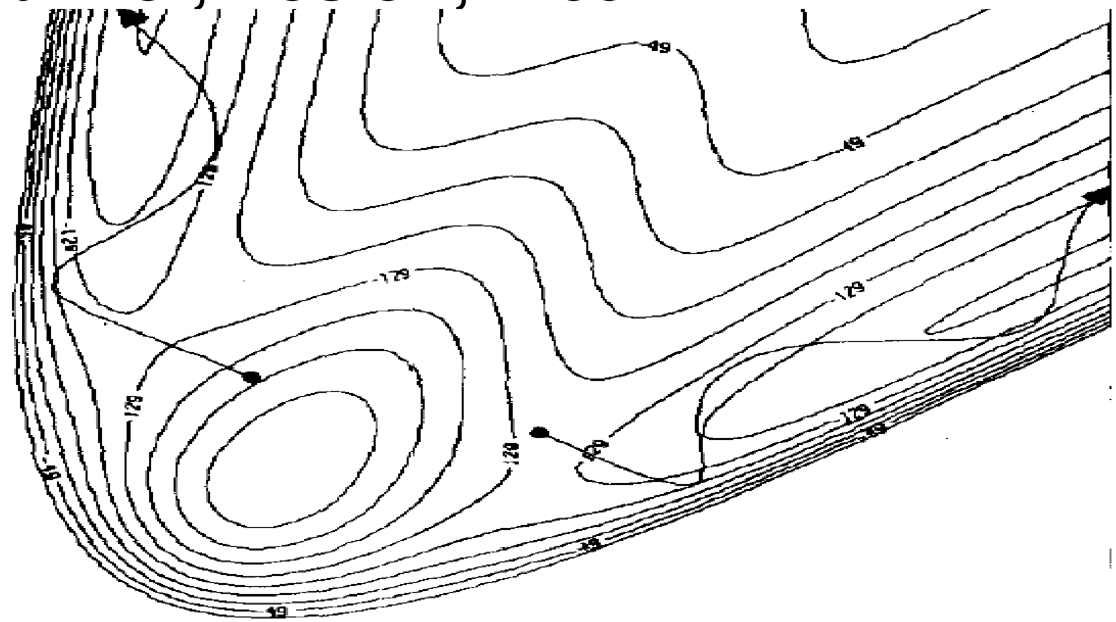
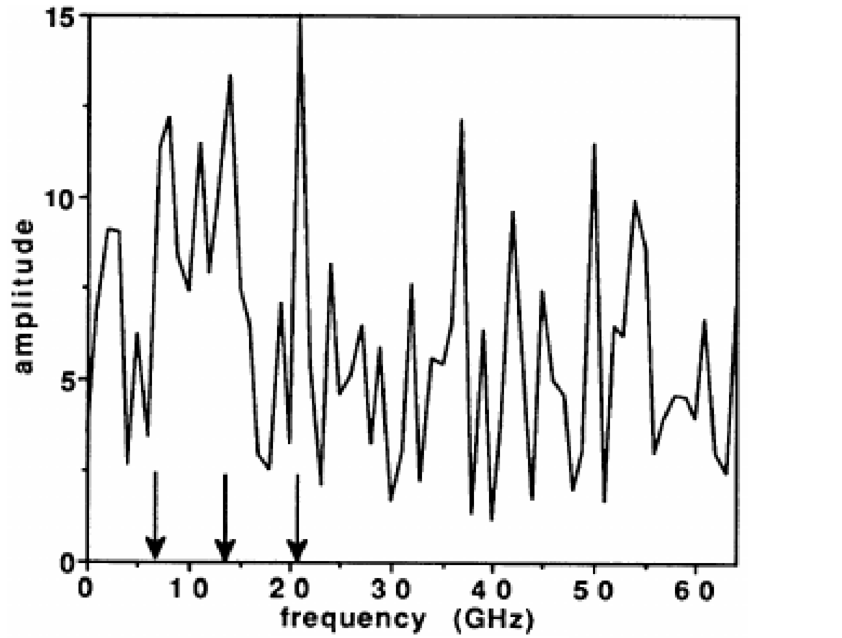


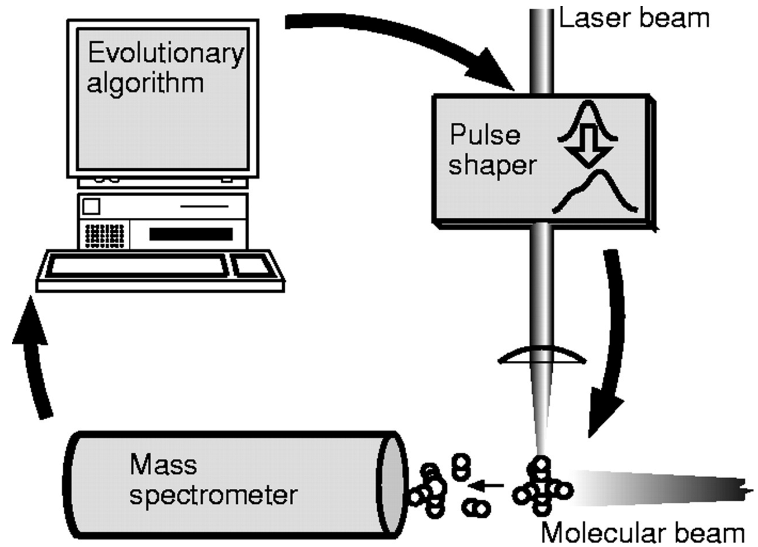
DJ Tannor, R Kosloff, SA Rice, J Chem Phys 85, 5805, 1986

Optimal Control of Reactions

- *Optimise* laser pulse spectrum to enhance reaction channel while suppressing others
  - Optimal pulse very complex, but easy to realise with pulse shaping technologies
  - However, potential energy surface of molecule is not known *with sufficient accuracy* to allow *reliable* design of optimal control pulse
- *Closed-Loop* method
  - Feed results of actual experiments into a *learning algorithm* to inform the control algorithm
  - Often use the experiment *directly* as “simulator”



RS Judson, H Rabitz, Phys Rev Lett, 68, 1500, 1992



A Assion, T Baumert, et al, Science, 282, 919, 1998

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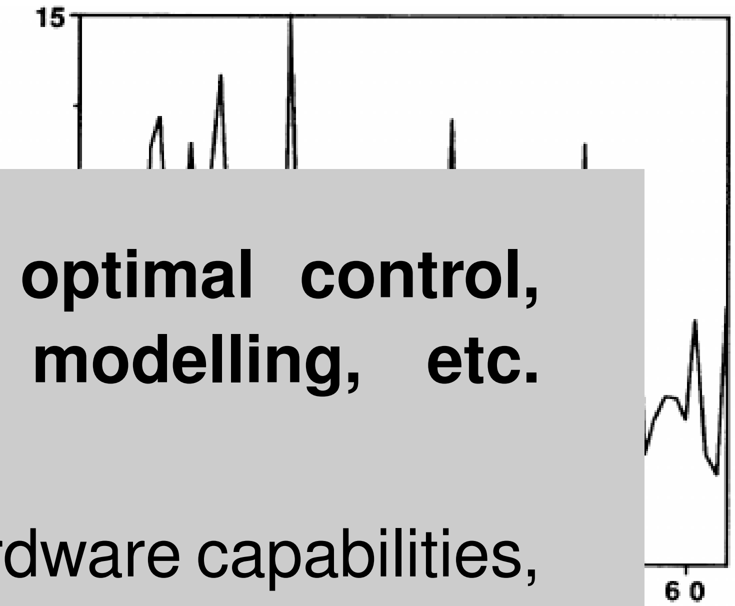

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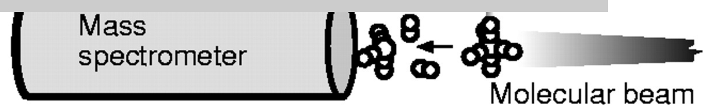
Careful experimental setups with optimal control, machine learning, computational modelling, etc. enabled significant progress

- Progress mainly due to improved hardware capabilities, refined optimisation algorithms and improved machine learning techniques
- Core issues on *efficient optimisation*, *accurate models*, *robust controls* remain
- General application limited by the *complexity of chemical systems*

- Often use the experiment *directly* as "simulator"



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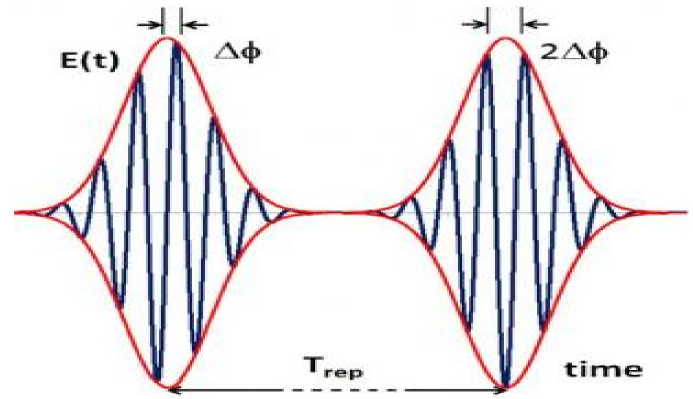
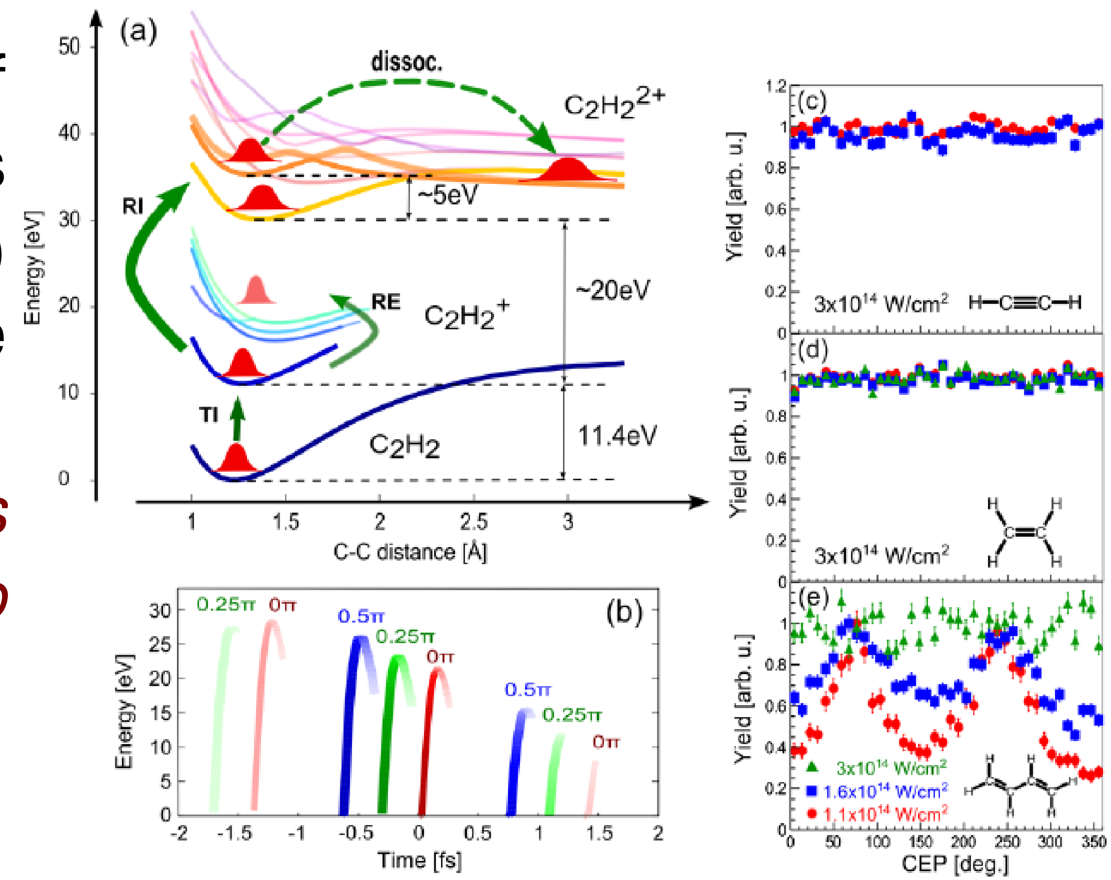
Carrier-Envelope Phase

- CEP provides additional DoF for control
  - Enables controlling intensity profile and *electric field* of light wave form
  - Attosecond pulse generation/measurement
  - Control of electron emission from atoms



- *Photofragmentation* of polyatomic molecules (acetylene, ethylene, . . .) via CEP control with few-cycle pulses
  - *Experimental observations linked to theoretical model to identify mechanism*
  - Recollision ionisation



X Xie, et al, Phys Rev Lett, 109, 243001, 2012

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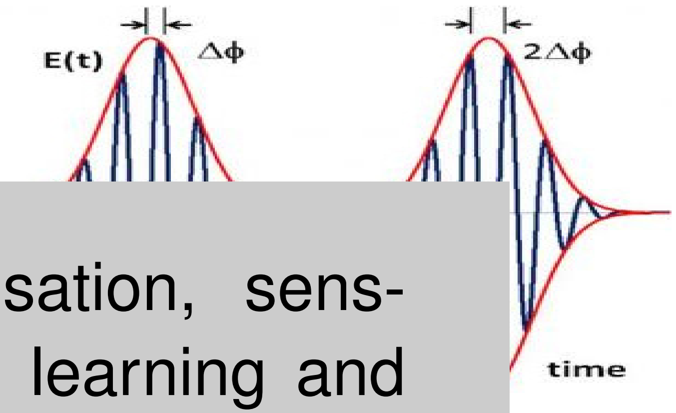
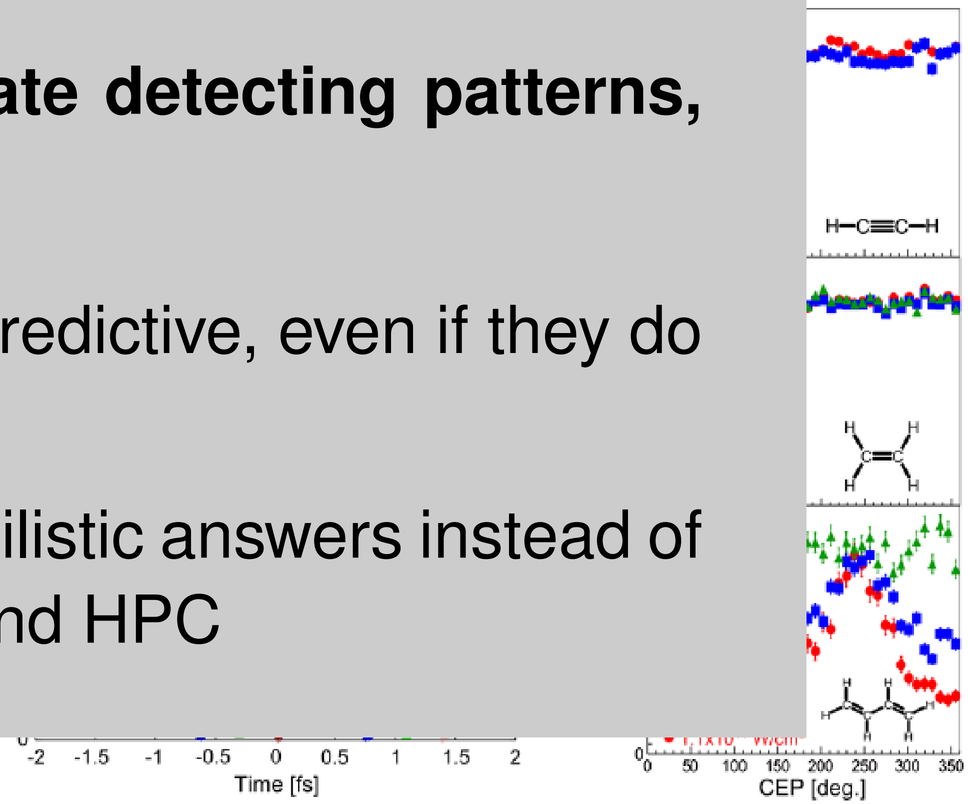


We have a wide range of control/optimisation, sensing/signal processing, simulation, machine learning and modelling techniques.

Can we extend these to facilitate detecting patterns, physical laws, mechanisms?

At least models should become predictive, even if they do not help human understanding.

Bayesian approaches and probabilistic answers instead of exact ones enabled by big data and HPC



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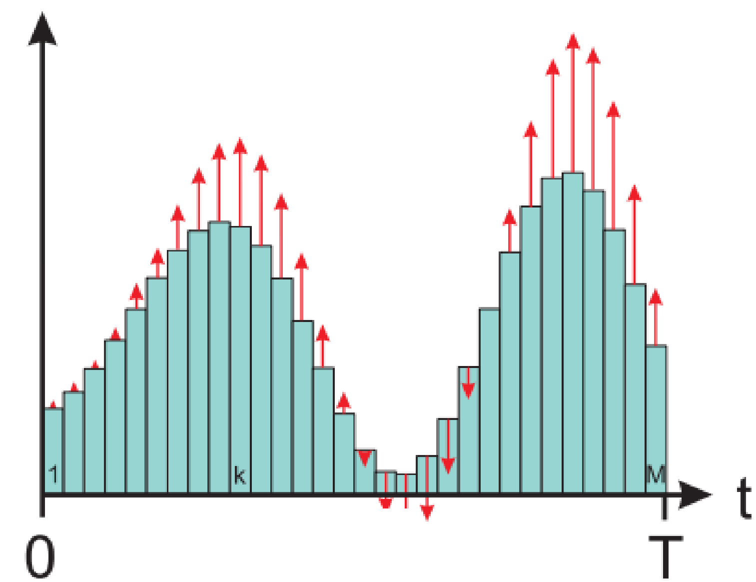
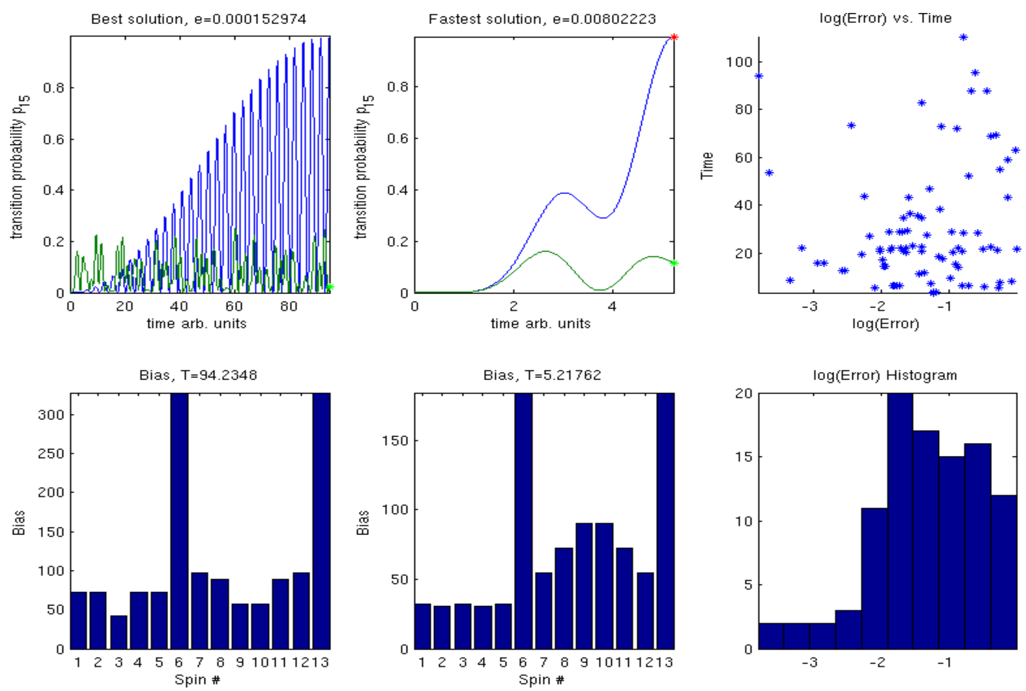
Optimal Control Toolbox

- Advanced *optimisation algorithms* that can deal well with optimal control problems
  - CRAB, GRAPE, Krotov variants of algorithms based on Nelder-Mead, genetic & evolutionary approaches, gradient-based, Newton, truncated and quasi Newton
  - For *piecewise-constant* dynamic controls



S Machnes, et al, PRA, 84, 022305, 2011



FC Langbein, SG Schirmer, E Jonckheere, submitted, CDC 2015

- *Semi-smooth Newton* and evolutionary approaches, *exploiting specific problem structures*, for very *complex landscapes*
- Handle *non piecewise-constant* controls, *static/spatial* controls, *experimental constraints*, *multiple targets* (e.g. time)

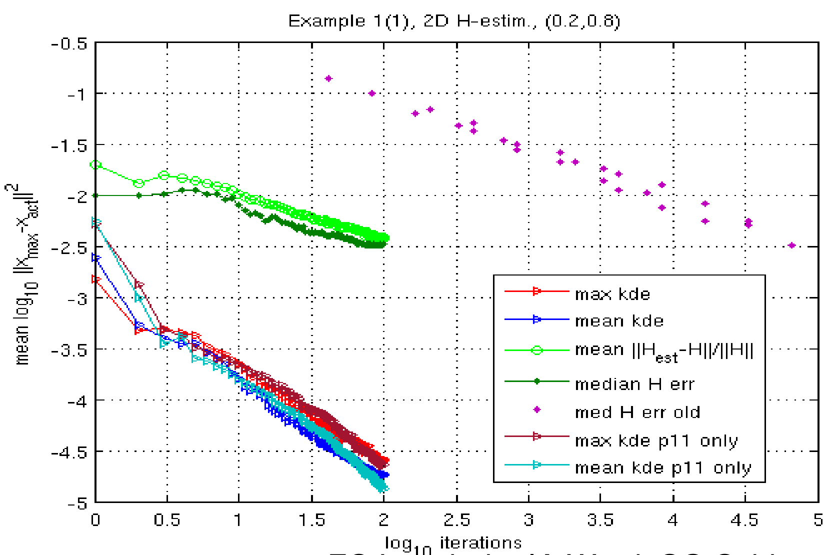
Modelling

Main problem: *Insufficiently accurate models* to describe the *evolution of the physical system* that are *predictive* and *feasible* to compute

- Time to move from *exact* models and answers to *probabilistic* models

➤ Mainly *model parameter estimation* using *maximum likelihood*

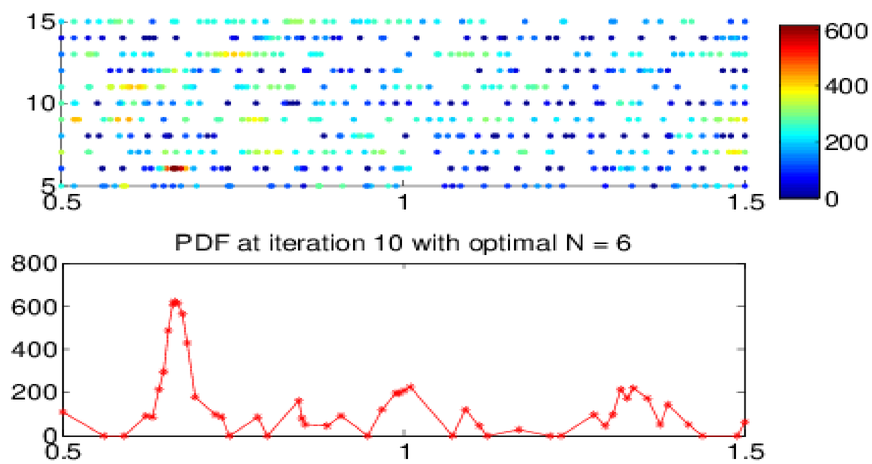
- PDF represented as KDE



FC Langbein, IA Ward, SG Schirmer, QCC 2015

– tracking *multiple* peaks
– *fast* convergence with *few* measurements

- Hilbert curve particle filter



SG Schirmer, FC Langbein, ISCCSP 2014
IA Ward, FC Langbein, SG Schirmer, 2015

– *more* parameters
– some *model discrimination*

Conclusions

- Optimal control of quantum systems, including reactions, an active topic for some time
  - Success *limited* mainly by *computational* power and suitable *models*
- Recent progress in quantum control created a range of *efficient optimisation* algorithms
  - More powerful techniques for *very complex landscapes*, *experimental constraints*, *more complex temporal and static/spatial controls* and *multiple targets* on the horizon
- Exact simulations start to be replaced by *probabilistic* simulations
  - Still mainly *parameter estimation*, but *model discrimination* techniques and *larger, discrete search spaces* become feasible