Chemistry

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Optimal Control of Quantum Systems

ss-NMR, Noise Resilience, and a Scalable Newton's Method

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Optimal Control at Southampton

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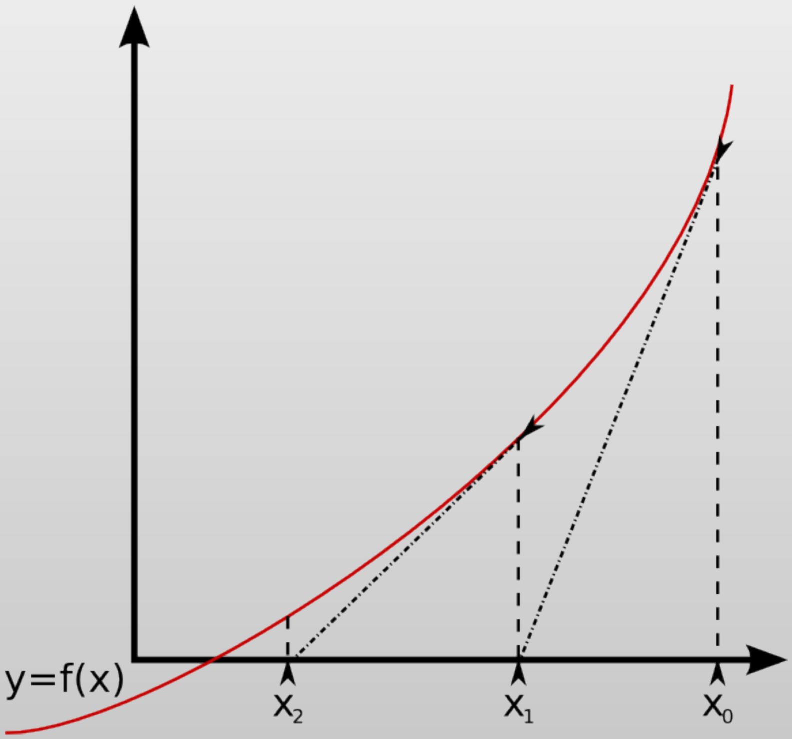
Introduction to Optimal Control

- Optimisation is the process of finding minimum or maximum (extrema) of functions
- Newton-Raphson method uses the gradient at a trial point to, incrementally, approach an extrema of an objective function
- $x_{k+1} = x_k - \frac{f(x)}{\nabla f(x)}$



- Basic method to use a **step length**, α, and a **step direction**, p, to iteratively find and extrema.
- We calculate a step direction, then decide how far to move in that direction:

$$x_{k+1} = x_k + \alpha_k p_k$$

- for minimisation, the step direction should be a descent direction.

$$p_k = -B_k^{-1} \nabla f_k$$

Gradient Descent B_k is the identity matrix

Newton's Method B_k is the exact Hessain matrix $\nabla^2 f(x_k)$

Quasi-Newton Methods B_k is an approximation to the Hessian.

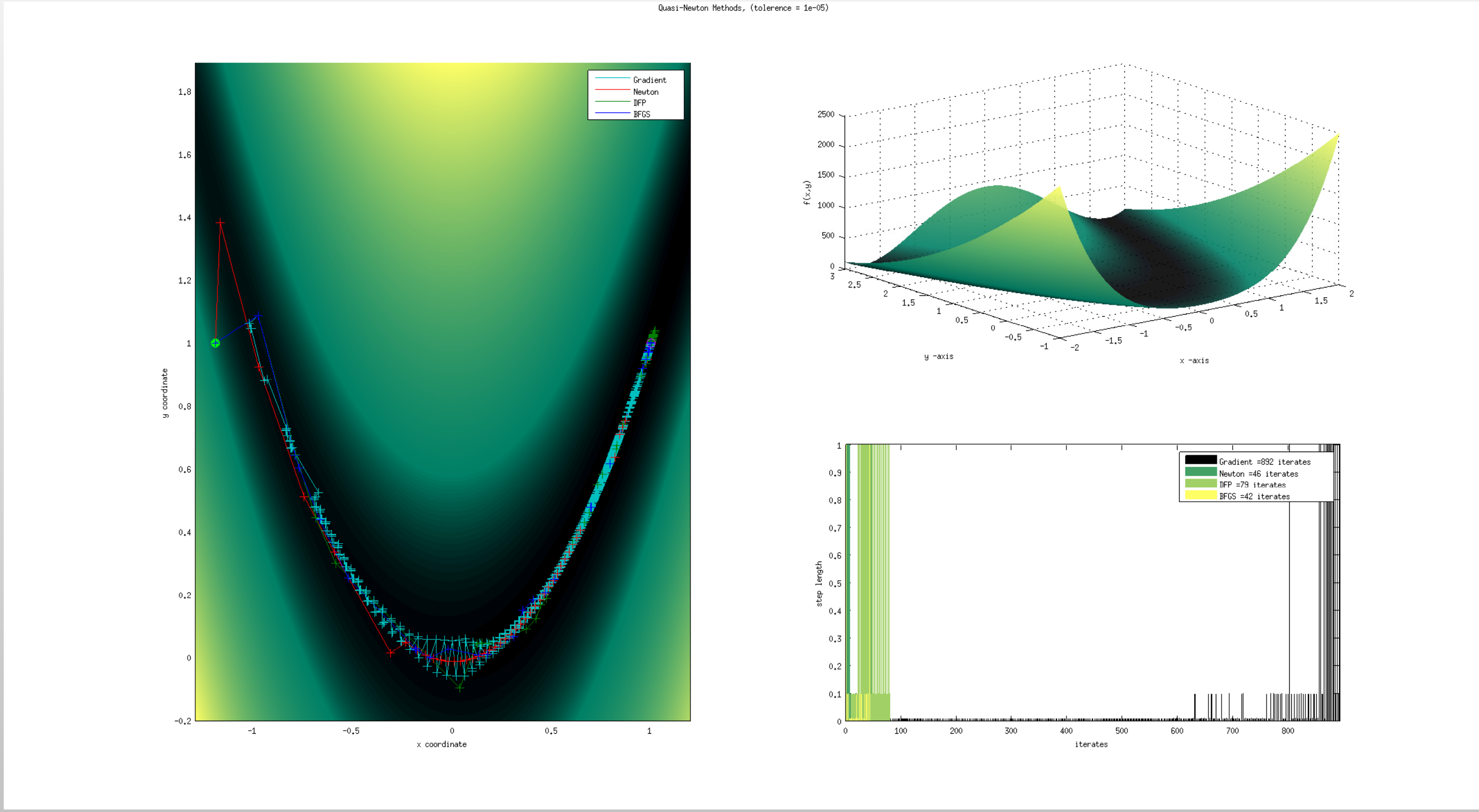
Gradient Descent Cheap, Scalable, Slow Convergence

Newton's Method Can be Expensive, Good Convergence.

BFGS Requires line search, No matrix inversion, Each update in $O(n^2)$.

l-BFGS Requires line search, No matrix inversion, Each update in $O(n)$.

Numerical Optimisation
Example - Rosenbruck Function



Optimal Control Theory

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- The idea of optimal control is to use all controllable parts of the full Hamiltonian to steer an initial quantum state into a target state.
- A measure of "success" for this control can be fidelity; the overlap between the controlled, final state and the target state (1 begin identical states, 0 being orthogonal states).
- The control parts of the Magnetic Resonance Hamiltonians are rf pulses for NMR, and mw pulses for ESR.
- The optimal solutions to the stated optimisation problem are a set of discrete-time control pulses.

Optimal Control Theory

- Separate Hamiltonian into the drift, $\hat{\mathcal{H}}_0$, and the parts we can control, $\hat{\hat{\mathcal{H}}}^{(k)}$.

$$\hat{\mathcal{H}}(t) = \hat{\mathcal{H}}_0 + \sum_k c^{(k)}(t)\hat{\hat{\mathcal{H}}}^{(k)}$$

- The control operators (e.g. Electromagnetic fields), each have a coefficient, $c^{(k)}(t)$, dictating the controllable time-evolution.
- Find $\{c^{(k)}(t)\}$ to take our system from an initial state, $|\psi_0\rangle$, to a target state, $|\sigma\rangle$

Optimal Control Theory

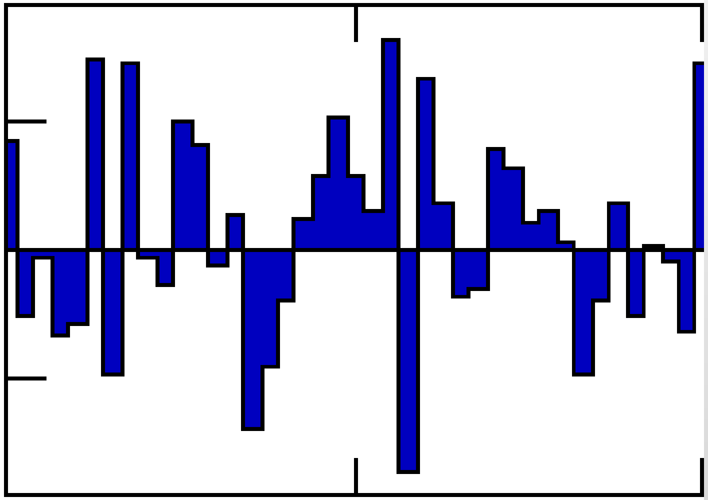
- Use fidelity as a metric to measure the overlap between target state and final state (1 = total overlap, 0 = no overlap)

$$J\{c^{(k)}(t)\} = \mathrm{Re}\,\langle\sigma|\,\exp_{(0)}\int_0^T -i\left(\hat{\mathcal{H}}_0 + \sum_k c^{(k)}(t)\hat{\hat{\mathcal{H}}}^{(k)}\right)dt\,|\psi_0\rangle$$

- The problem is to find the maximum of the fidelity (or the minimum of 1−fidelity).

GRAPE

- Assumption: control pulse sequence is piecewise constant.
- Time-ordered exponential now a sequential multiplication of propagators.



$$J\{c^{(k)}(t)\} = \mathrm{Re}\left\langle \sigma \middle| \hat{\mathcal{U}}_N \hat{\mathcal{U}}_{N-1} \cdots \hat{\mathcal{U}}_2 \mathcal{U}_1 \middle| \psi_0 \right\rangle$$

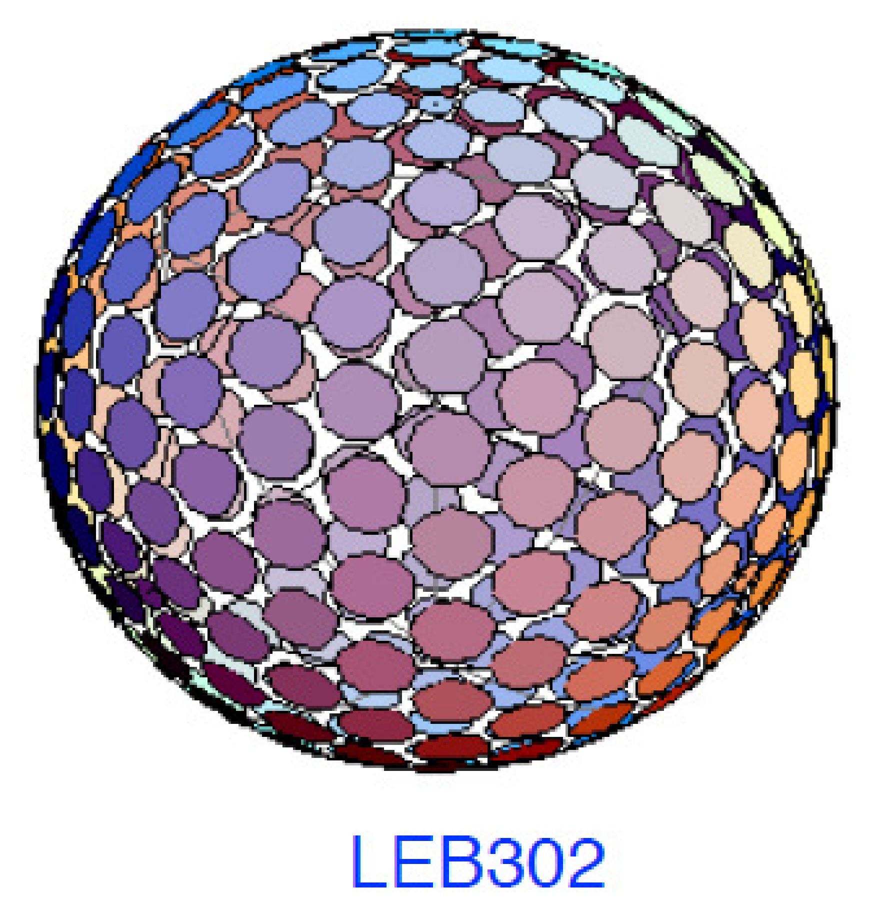
$$\hat{\mathcal{U}}_n = \exp_{(0)}\left[-i \left(\hat{\mathcal{H}}_0 + \sum_k c^{(k)}(t_n) \hat{\hat{\mathcal{H}}}^{(k)} \right) \Delta t \right]$$

Solid State Nuclear Magnetic Resonance

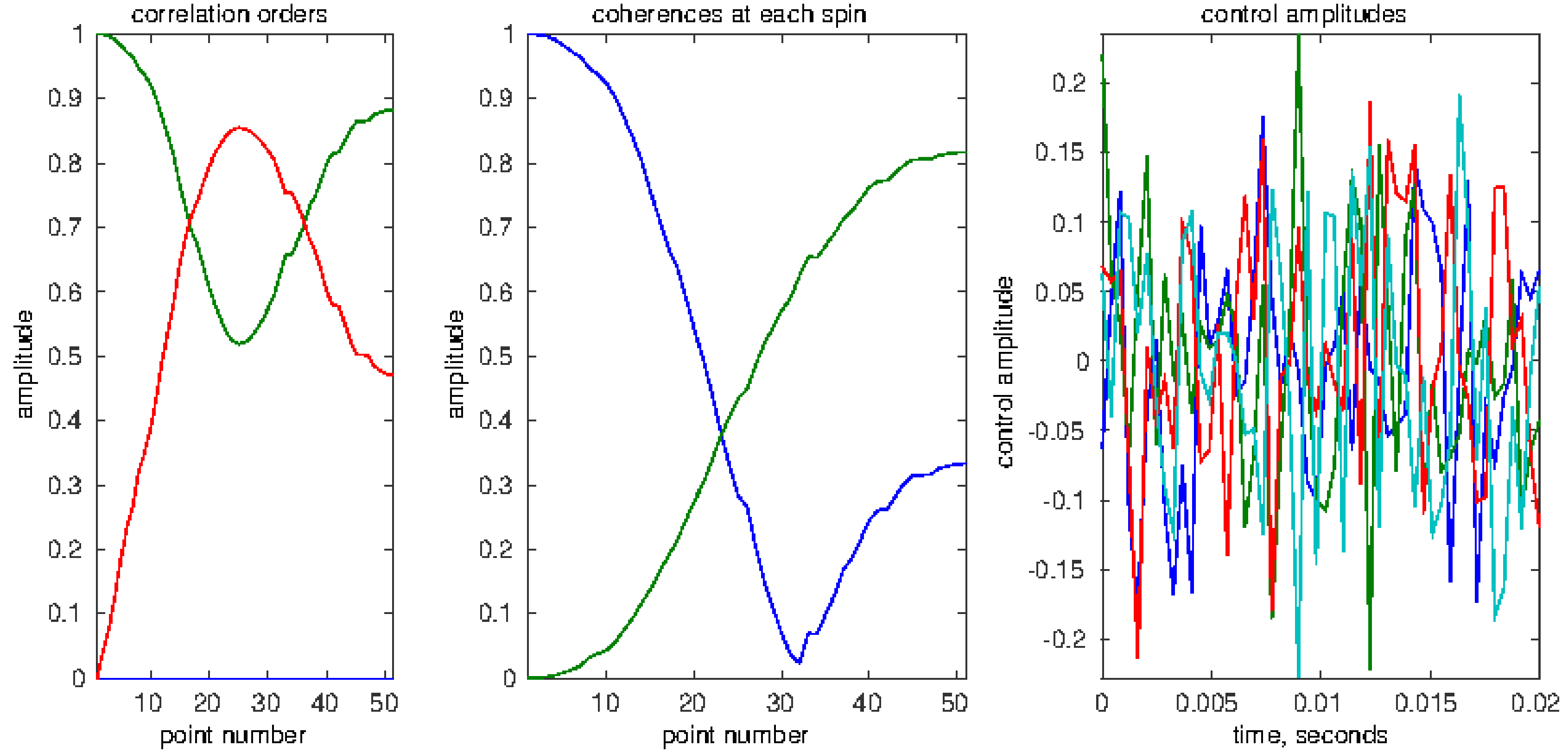
Ensemble Average

Crystallite orientation, Lebedev grid, Floquet space

- Take an ensemble average over the crystallite orientations.
- Average over a Lebedev grid.
- Initialise each of the ensemble to the same random guess.
- Calculate the Liouvillian in a Floquet basis at each of the grid weights.
- find the local cost and its gradient.
- Take the weighted average over the cost and gradient.
- Feed into a numerical optimisation through GRAPE.



Ensemble Average



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Noise Resilience of Optimal Control Pulses

HCF simulations

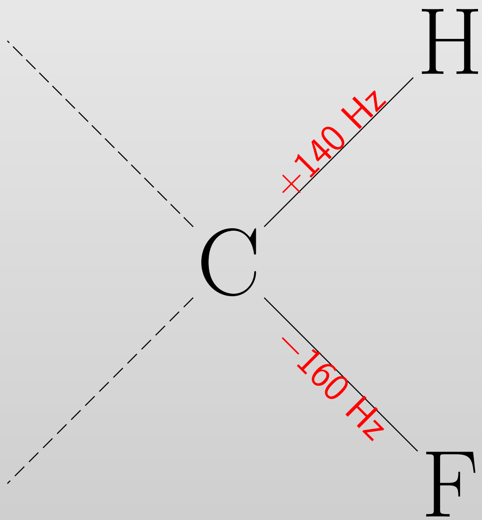

Figure: Interaction parameters of a molecular group used in HCF small system state transfer simulations, with a magnetic induction of 9.4 Tesla.

protein backbone simulations

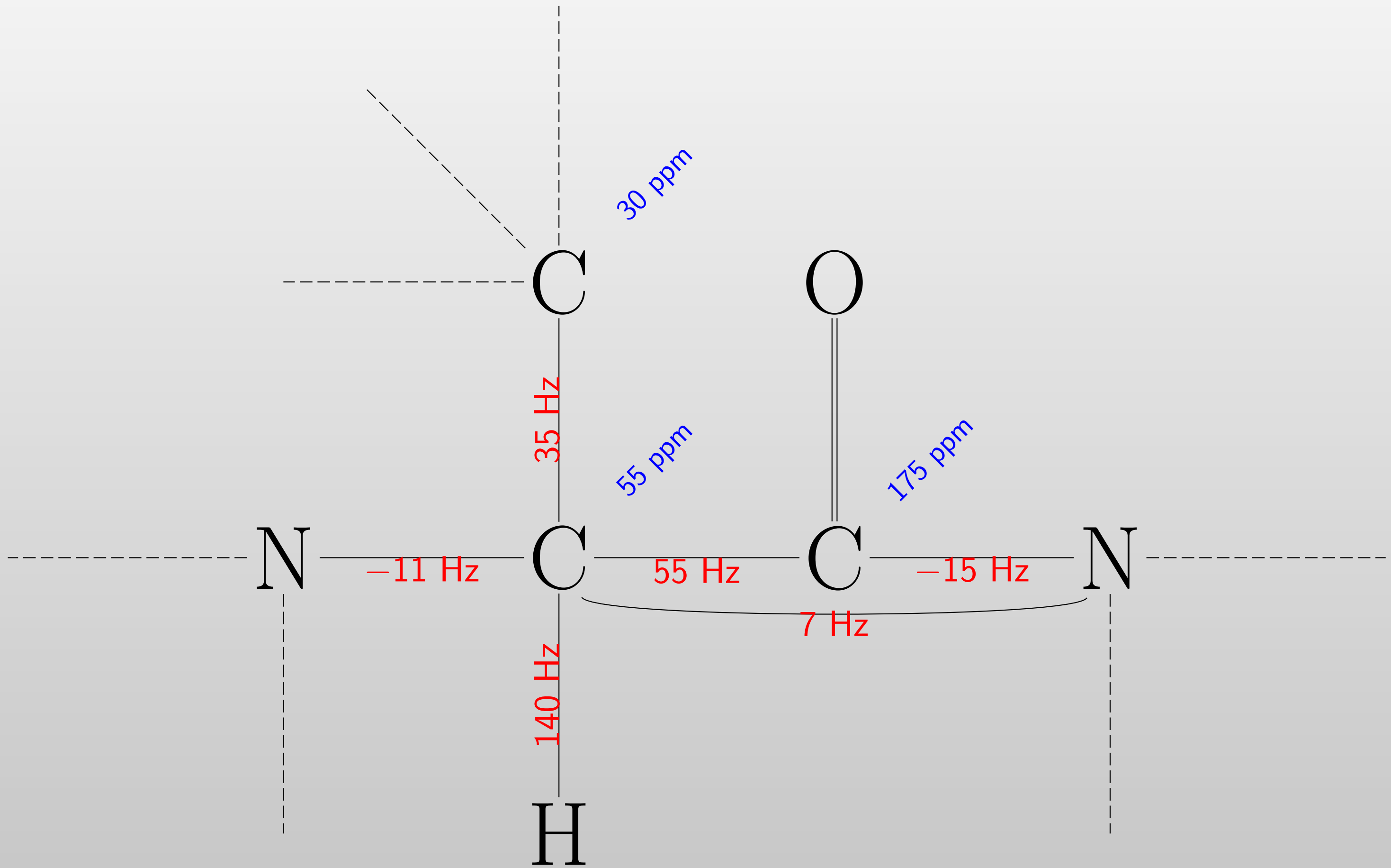

Figure: Interaction parameters of a typical protein backbone used in large system state transfer simulations, with a magnetic induction of 9.4 Tesla.

Ensemble Average

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- In real experimental apparatus, the control channels will include some level of noise as most electrical systems do
- Just as with the ssNMR optimisation, we average over an ensemble of systems.
- each system has the same initial random guess
- In simulating this noise, the solution is to create an ensemble of systems, each with their own instance of noise affecting the control channels.
- Noise is additive Gaussian, and is defined at the start of the simulation for each noise instance.
- We then optimise over the ensemble.

Ensemble Average

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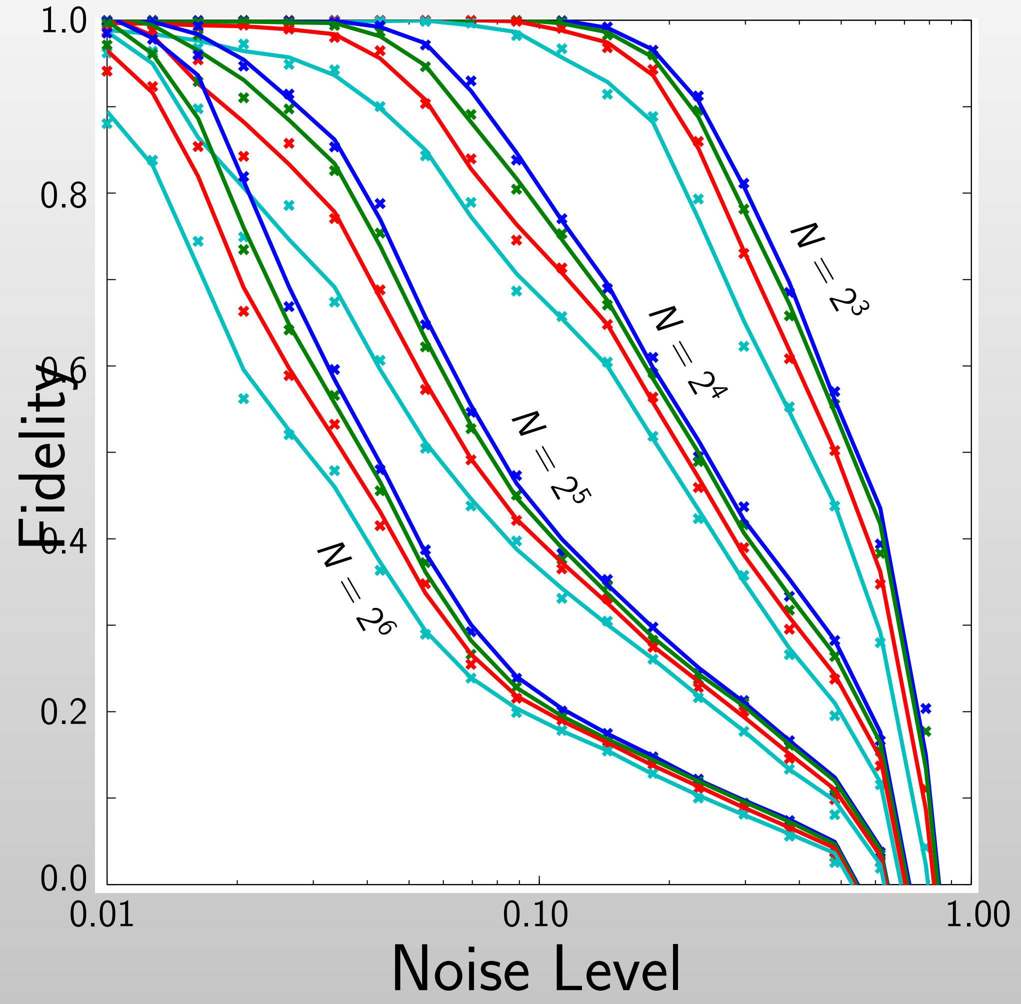

Figure: Convergence of the noise simulations to a number of noise instances

Ensemble Average

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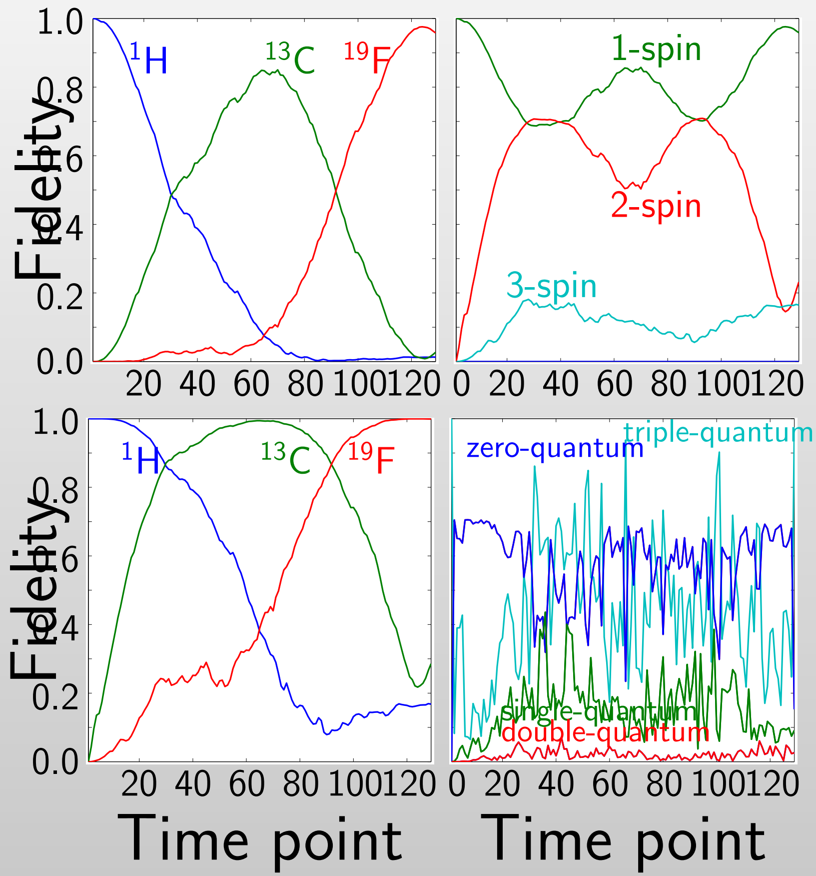

Figure: Population of states local at each spin of HCF (upper-left), and total population of each state (lower-left).

Total populations of correlations in the system (upper-right), and coherence orders (lower-right).

a direct product basis set the full state space, $\mathfrak{L}$, is a direct sum of correlation order subspaces:

$$\mathfrak{L} = \mathfrak{L}_0 \oplus \mathfrak{L}_1 \oplus \mathfrak{L}_2 \oplus \mathfrak{L}_3$$

where the population of a correlation order, p_k, is given by the projection onto its subspace

$$p_k = \left\| \hat{\hat{P}}_{\mathfrak{L}_k} \left| \hat{\rho} \right\rangle \right\|$$

Ensemble Average

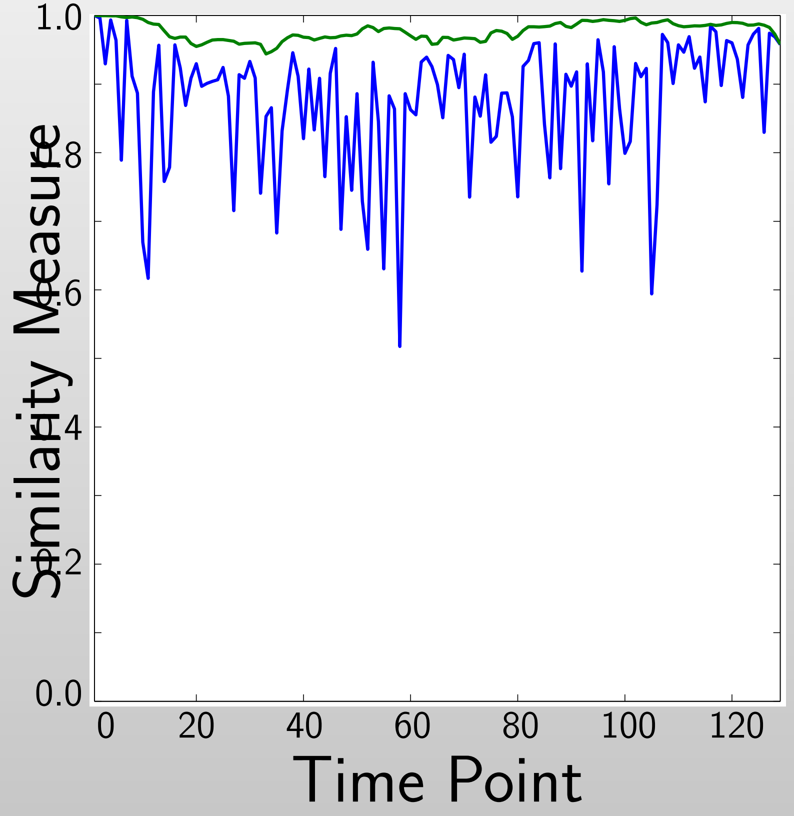

Figure: Measures of similarity between the trajectory of a noisy system compared to that of a similar one without noise

The Newton-Raphson Method

Objective function and Gradient Calculation

- The problem of second order methods, that require a gradient calculation, is reduced to:
  1. Propagate forwards from the source
  2. Propagate backwards from the target
  3. Compute the expectation of the derivative

$$\left\langle \sigma \middle| \hat{\mathcal{U}}_N \hat{\mathcal{U}}_{N-1} \cdots \frac{\partial}{\partial c_{n=t}^{(k)}} \hat{\mathcal{U}}_{n=t} \cdots \hat{\mathcal{U}}_2 \mathcal{U}_1 \middle| \psi_0 \right\rangle$$

Augmented Exponential
Gradient Calculation

- We can use a the work of C. Van Loan (1970's) and Sophie Schirmer to find the derivative of the control pulse at a specific time point

$$\exp\begin{pmatrix} -i\hat{\hat{L}}\Delta t & -i\hat{\hat{H}}_k^{(j)}\Delta t \\ \mathbf{0} & -i\hat{\hat{L}}\Delta t \end{pmatrix} = \begin{pmatrix} e^{-i\hat{\hat{L}}\Delta t} & \frac{\partial}{\partial c_k^{(j)}} e^{-i\hat{\hat{L}}\Delta t} \\ \mathbf{0} & e^{-i\hat{\hat{L}}\Delta t} \end{pmatrix}$$

Hessian Matrix
Avoiding Singularities

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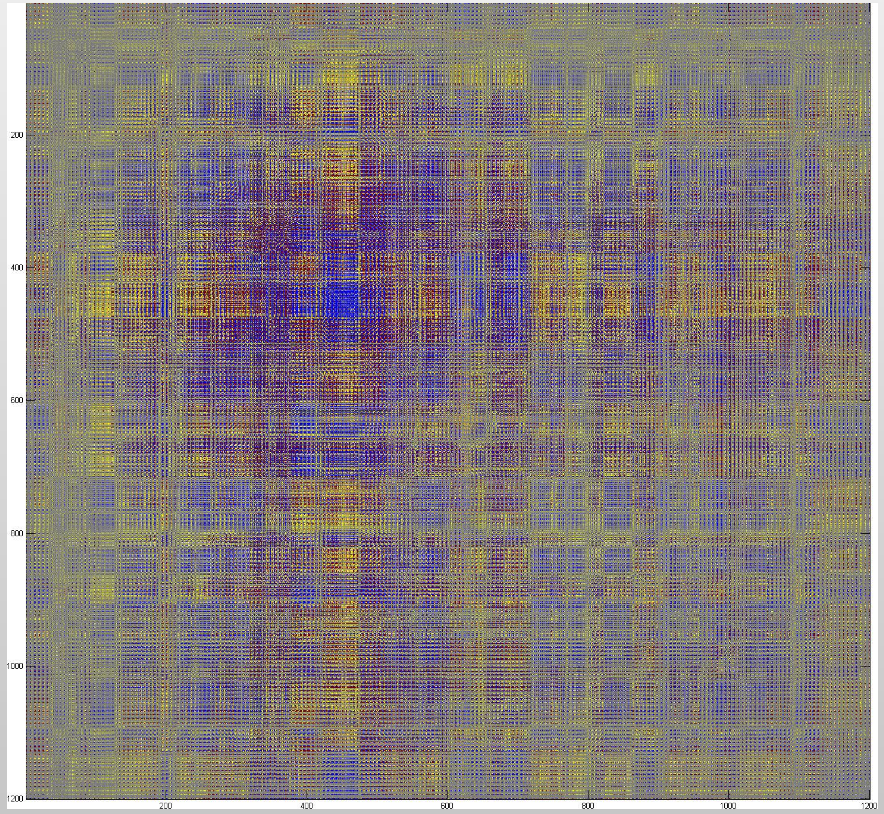
- Symmetric
- Non-singular
- Size is time-points multiplied by control channels.
- Diagonally dominant.



Hessian Matrix
Calculating the Hessian elements

- Calculation of the Hessian elements requires the second order derivatives
- Scales to $O(n^2)$ calculations (compared with $O(n)$ for a gradient calculation).
- second order derivatives can be calculated with a 3×3 augmented exponential.

$$\exp\begin{pmatrix} -i\hat{\hat{L}}\Delta t & -i\hat{\hat{H}}_{n_1}^{(k_1)}\Delta t & 0 \\ 0 & -i\hat{\hat{L}}\Delta t & -i\hat{\hat{H}}_{n_2}^{(k_2)}\Delta t \\ 0 & 0 & -i\hat{\hat{L}}\Delta t \end{pmatrix} =$$

$$\begin{pmatrix} e^{-i\hat{\hat{L}}\Delta t} & \frac{\partial}{\partial c_{n_1}^{(k_1)}} e^{-i\hat{\hat{L}}\Delta t} & \frac{\partial^2}{\partial c_{n_1}^{(k_1)} \partial c_{n_2}^{(k_2)}} e^{-i\hat{\hat{L}}\Delta t} \\ 0 & e^{-i\hat{\hat{L}}\Delta t} & \frac{\partial}{\partial c_{n_2}^{(k_2)}} e^{-i\hat{\hat{L}}\Delta t} \\ 0 & 0 & e^{-i\hat{\hat{L}}\Delta t} \end{pmatrix}$$

Hessian Calculation
Expectation of second order derivatives

- We can reduce the computation to scale with $O(n)$ by realising that we can store propagators from gradient calculation, and then perform an extra set of 3×3 augmented exponentials for the (block) diagonal elements.

$$\left\langle \sigma \left| \hat{\mathcal{U}}_N \hat{\mathcal{U}}_{N-1} \cdots \frac{\partial^2}{\partial {c_{n=t}^{(k)}}^2} \hat{\mathcal{U}}_{n=t} \cdots \hat{\mathcal{U}}_2 \mathcal{U}_1 \right| \psi_0 \right\rangle$$

$$\left\langle \sigma \left| \hat{\mathcal{U}}_N \hat{\mathcal{U}}_{N-1} \cdots \frac{\partial}{\partial c_{n=t}^{(k)}} \hat{\mathcal{U}}_{n=t+1} \frac{\partial}{\partial c_{n=t}^{(k)}} \hat{\mathcal{U}}_{n=t} \cdots \hat{\mathcal{U}}_2 \mathcal{U}_1 \right| \psi_0 \right\rangle$$

$$\left\langle \sigma \left| \hat{\mathcal{U}}_N \hat{\mathcal{U}}_{N-1} \cdots \frac{\partial}{\partial c_{n=t}^{(k)}} \hat{\mathcal{U}}_{n=t_2} \cdots \frac{\partial}{\partial c_{n=t}^{(k)}} \hat{\mathcal{U}}_{n=t_1} \cdots \hat{\mathcal{U}}_2 \mathcal{U}_1 \right| \psi_0 \right\rangle$$

Hessian Regularisation
Avoiding Singularities

- A singular matrix is one that is not invertable.
- Common when we have negative eigenvalues.
- We can regularise the Hessian matrix:

TRM - We can add a multiple of the identity to the Hessian

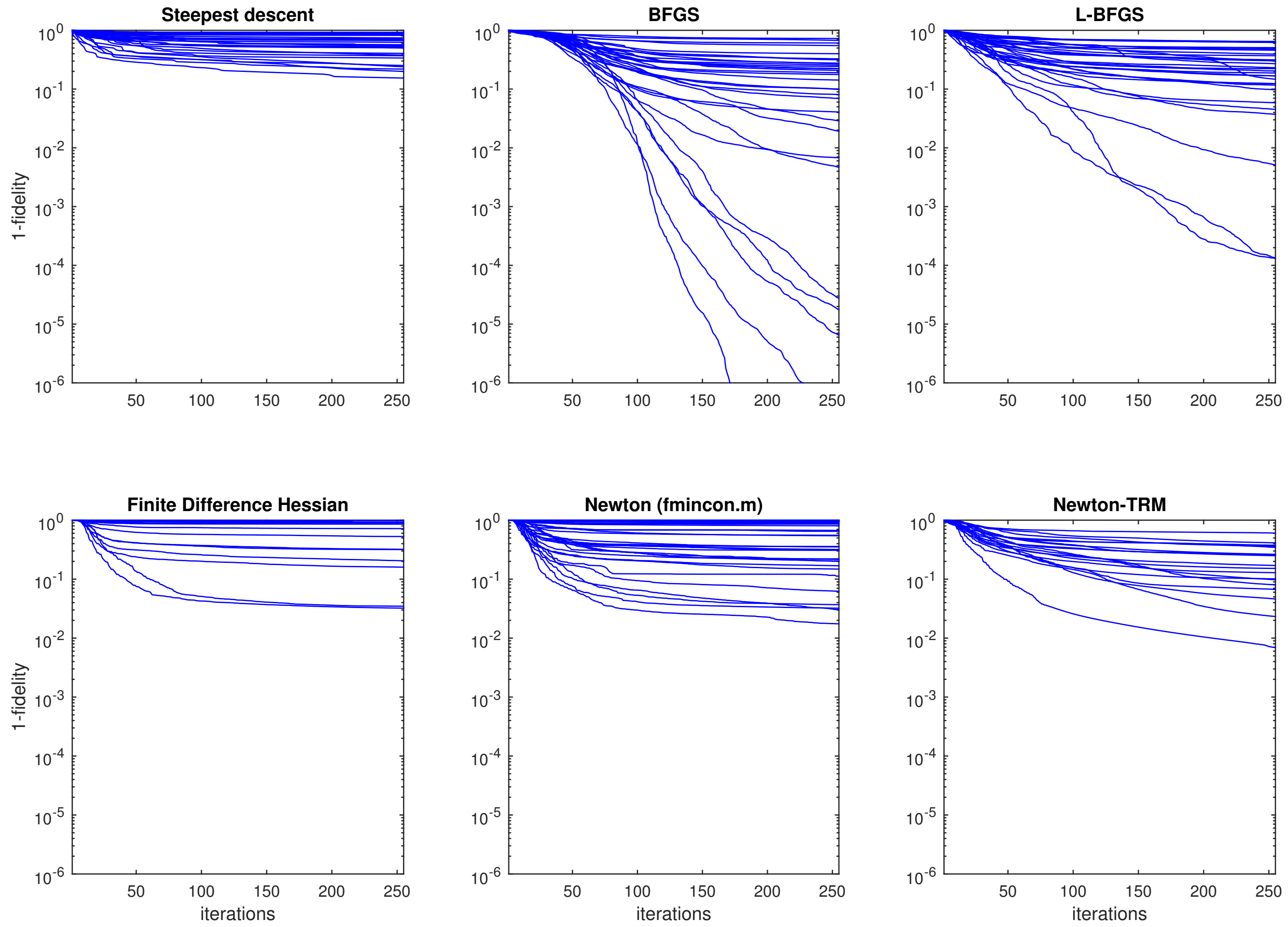
$$H_{reg} = H + \lambda I$$

where $\lambda > max\{0, -min[eig(H)]\}$

RFO - Prevents the algorithm taking large steps....

Results

Comparing the Newton method



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