A Universal Computational Method for Optimal Pulse Design in NMR & MRI

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Abstract

Many key aspects of control of quantum systems involve manipulating a large quantum ensemble exhibiting variation in the value of parameters characterizing the system dynamics. Developing electromagnetic pulses to produce a desired evolution in the presence of such variation is a fundamental and challenging problem in this research area. We present such robust pulse designs as an optimal control problem of a continuum of bilinear systems with a common control function. We then develop a unified computational method for optimal pulse design using ideas from pseudospectral approximations, by which a continuous-time optimal control problem of pulse design can be discretized to a constrained optimization problem with spectral accuracy. This is a highly flexible and efficient numerical method that requires low order of discretization and yields inherently smooth solutions.

Contribution

Challenge To design robust, practical pulses for quantum control.

Impacts Immediate applicable to structural biology, molecular imaging and quantum optics.

Universal The pseudospectral method delivers a highly general framework for pulse design in conjunction with optimal ensemble control theory.

New Formulation for Optimal Pulse Design

$$\min \int_\Omega \left[\varphi(T, x(T,s)) + \int_0^T \mathcal{L}(x(t,s), u(t))\,dt\right] ds$$

$$\text{s.t.}\ \frac{d}{dt}x(t,s) = f(x(t,s), u(t), s)$$

$$e(x(0,s), x(T,s)) = 0$$

$$g(x(t,s), u(t)) \le 0, \qquad \forall \quad s \in \Omega \subset \mathbb{R}^d,\ t \in [0,T]$$

Mutlidimensional Pseudospectral Discretization

$$\min\ \frac{b-a}{2}\sum_{r=0}^{N_s}\left[\varphi(T, \bar{x}_{Nr}) + \frac{T}{2}\sum_{i=0}^{N}\mathcal{L}(\bar{x}_{ir}, \bar{u}_i) w_i^N\right] w_r^{N_s}$$

$$\text{s.t.}\ \sum_{k=0}^{N} D_{jk}\bar{x}_{kr} = \frac{T}{2} f(\bar{x}_{jr}, \bar{u}_j)$$

$$e(\bar{x}_{0r}, \bar{x}_{Nr}) = 0$$

$$g(\bar{x}_{jr}, \bar{u}_j) \le 0, \qquad \forall \quad j \in \{0, 1, \dots, N\},\ r \in \{0, 1, \dots, N_s\}$$

References

- Li J-S, Khaneja N. *Ensemble Control of Bloch Equations.* IEEE Trans Autom Control 54: 528-536 (2009).
- J.-S. Li, J. Ruths, D. Stefanatos. *A pseudospectral method for optimal control of open quantum systems*, J. Chem. Phys. 131, 164110 (2009).
- C. Canuto, M. Y. Hussaini, A. Quarteroni and T. A. Zang, *Spectral Methods* (Springer, Berlin, 2006).
- J. Ruths, J.-S. Li. *Optimal ensemble control of open quantum systems with a pseudospectral method*, 49th IEEE Conference on Decision and Control, Atlanta, 2010.

Optimal Coherence Transfer (NMR)

$$\max\ \mathcal{J}_{\text{avg}} = \frac{1}{2\delta(\xi_2-\xi_1)}\int_{1-\delta}^{1+\delta}\int_{\xi_1}^{\xi_2} x_4(T,\xi,J)\, d\xi\, dJ$$

$$\text{s.t.}\ \frac{d}{dt}\begin{bmatrix}x_1\\x_2\\x_3\\x_4\end{bmatrix} = \begin{bmatrix}0 & -u_1 & 0 & 0\\ u_1 & -\xi & -J & 0\\ 0 & J & -\xi & -u_2\\ 0 & 0 & u_2 & 0\end{bmatrix}\begin{bmatrix}x_1\\x_2\\x_3\\x_4\end{bmatrix}$$

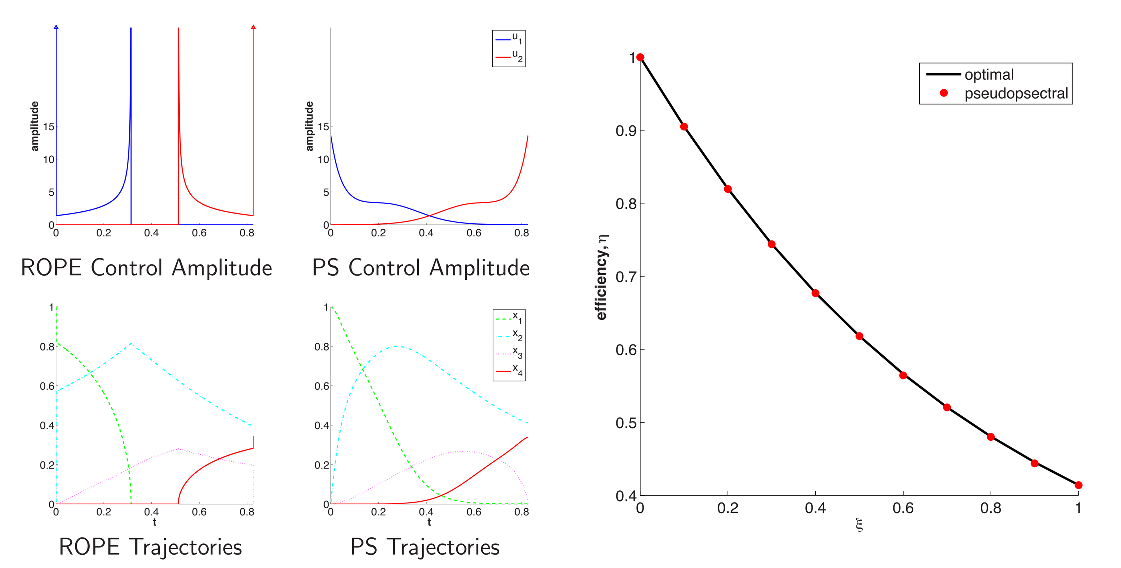
$$x(0,\xi,J) = [1\ 0\ 0\ 0]'$$

$$\sqrt{u_1^2(t)+u_2^2(t)} \le A,\ \forall t \in [0,T]$$

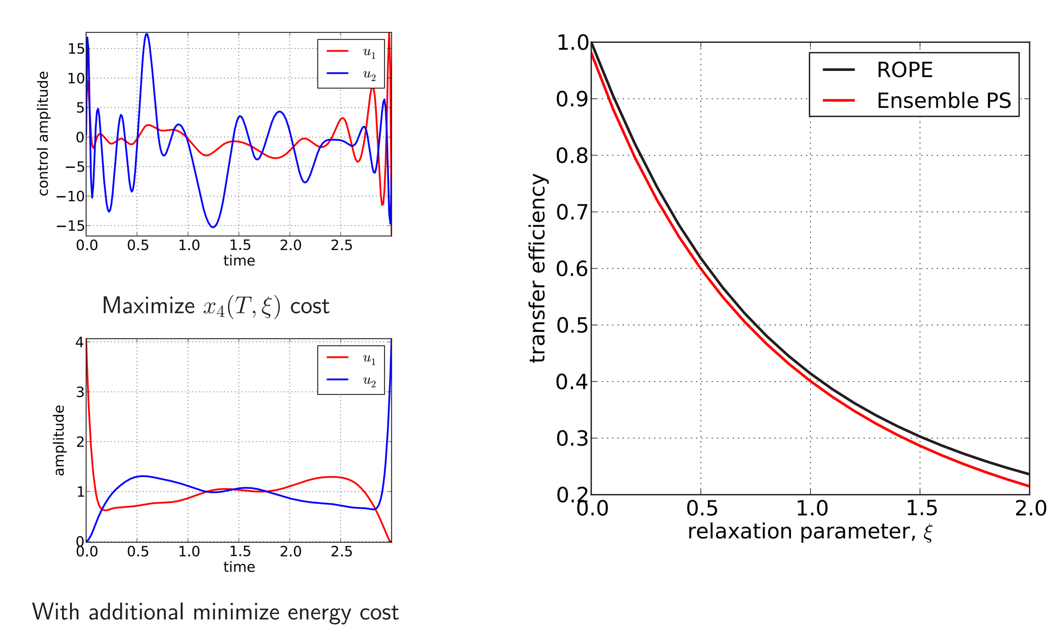
$$\xi \in [\xi_1, \xi_2], \quad J \in [1-\delta, 1+\delta]$$

- Compare against analytic optimal solution: ROPE
- ROPE is only valid for a single choice of (ξ, J)
- Applications in protein NMR spectroscopy

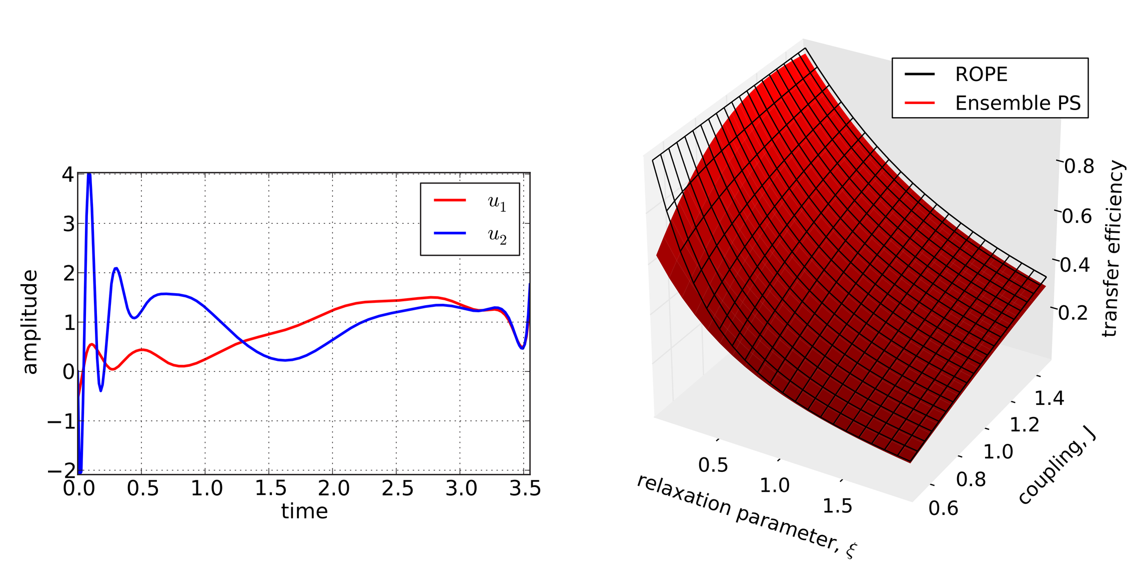
Single Parameter Optimization



Ensemble Optimization, $\xi \in [0,2]$, $J = 1$



Ensemble Optimization, $\xi \in [0,2]$, $J \in [0.5, 1.5]$



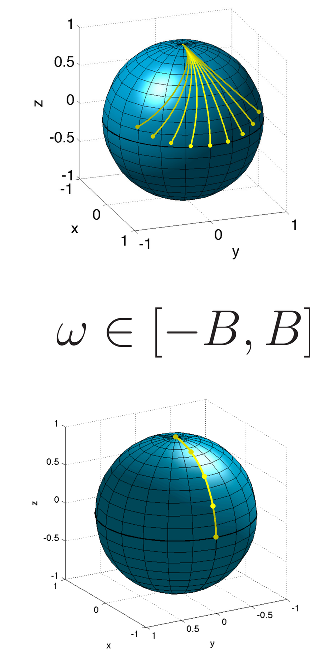
Optimal Pulse Design (MRI & NMR)

$$\max\ \frac{1}{4\delta B}\int_{1-\delta}^{1+\delta}\int_{-B}^{B} M_x(T,\omega,\epsilon)\, d\omega\, d\epsilon$$

$$\text{s.t.}\ \frac{d}{dt}\begin{bmatrix}M_x\\M_y\\M_z\end{bmatrix} = \begin{bmatrix}0 & -\omega & \epsilon u\\ \omega & 0 & -\epsilon v\\ -\epsilon u & \epsilon v & 0\end{bmatrix}\begin{bmatrix}M_x\\M_y\\M_z\end{bmatrix}$$

$$M(0,\omega,\epsilon) = [0\ 0\ 1]'$$

$$\sqrt{u^2(t)+v^2(t)} \le A,\ \forall t \in [0,T]$$

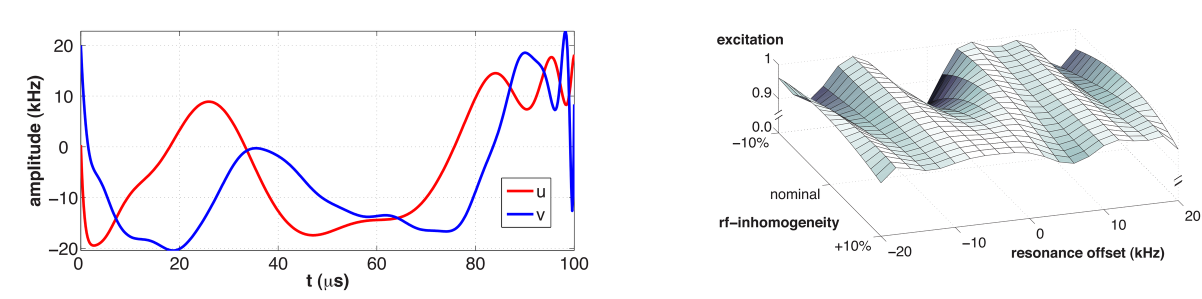


$\omega \in [-B, B]$

$\epsilon \in [1-\delta, 1+\delta]$

Ensemble Optimization

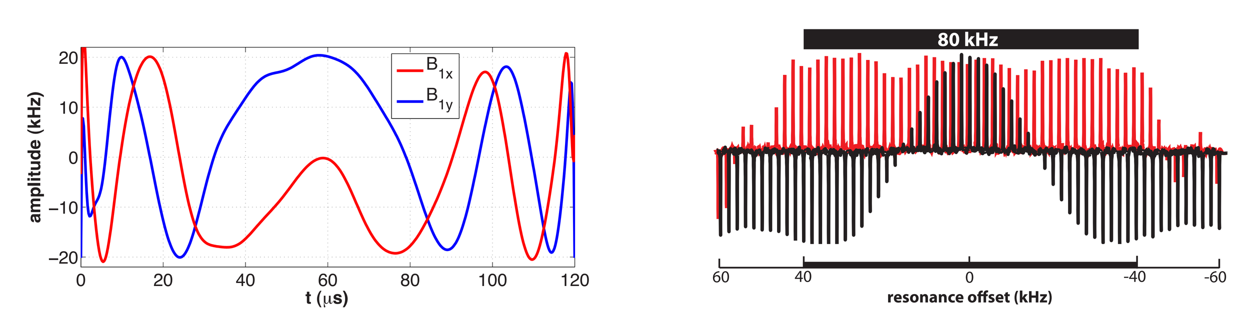
- Maximum Amplitude, $A = 20$ kHz
- Larmor Dispersion, $\omega \in [-20, 20]$ kHz
- RF inhomogeneity, $\delta = 10\%$



Ensemble Optimization & Experimental Verification

$A = 20$ kHz, $\omega \in [-40, 40]$ kHz

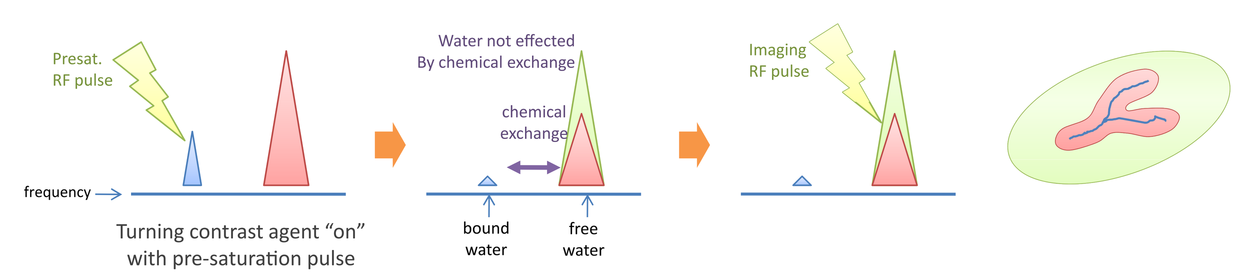
In collaboration with G. Wagner's Lab, Harvard Medical School

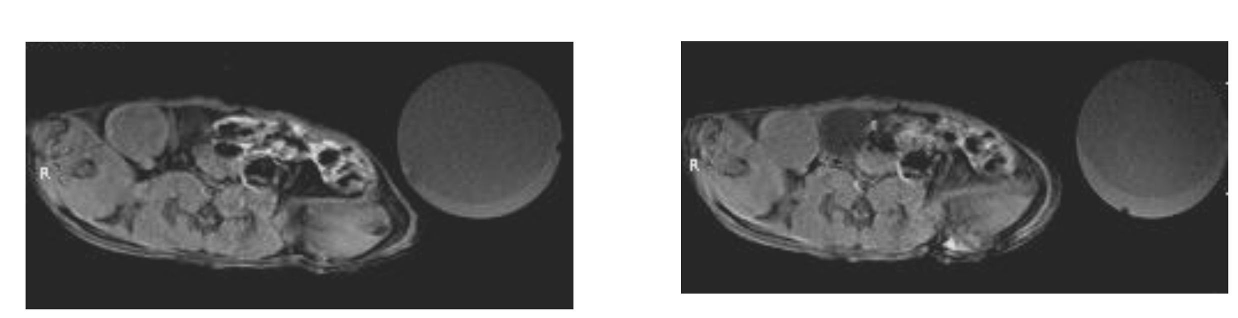


PARACEST Contrast Optimization (MRI)

$$\frac{d}{dt}\begin{bmatrix}z_a\\x_a\\y_a\\y_b\\x_b\\z_b\end{bmatrix} = \left[\begin{array}{ccc|ccc}-k_{1a} & -v & u & 0 & 0 & C_b\\ v & -k_{2a} & -\omega_a & 0 & C_b & 0\\ -u & \omega_a & -k_{2a} & C_b & 0 & 0\\ \hline 0 & 0 & C_a & -k_{2b} & \omega_b & -u\\ 0 & C_a & 0 & -\omega_b & -k_{2b} & v\\ C_a & 0 & 0 & u & -v & -k_{1b}\end{array}\right]\begin{bmatrix}z_a\\x_a\\y_a\\y_b\\x_b\\z_b\end{bmatrix} + \begin{bmatrix}\frac{M_0^a}{T_{1a}}\\0\\0\\0\\0\\\frac{M_0^b}{T_{1b}}\end{bmatrix}$$

- Motivates designing optimal pulses robust to rf inhomogeneity including relaxation effects





Pseudospectral Features

Arbitrary Cost Function & Constraints
The direct transformation of the continuous-time optimal control problem into a finite-dimensional constrained optimization enables any type or combination of cost functions and constraints.

Smoothness
The pseudospectral method is based on polynomial approximations, which guarantees that the solutions will be smooth. Certain choices of cost functions can yield pulses with different properties, such as slowly varying pulses for practical experimental implementation.

Convergence
Discretizing with the pseudospectral method empirically exhibits exponential convergence of the solution as the number of discretizations increases.

Ease of Implementation
The pseudospectral method can be implemented in virtually any programming language and can be expressed very concisely within the AMPL optimization framework.

Computational Complexity
The low order of approximation characteristic of orthogonal polynomial approximation greatly reduces the size of the optimization problem necessary to be solved.

Future Directions

Experimental verification
We are beginning verification of the optimal pulses for coherence transfer for systems with J coupling variation in collaboration with G. Wagner at Harvard.

Extension to PARACEST
Modify and run the pseudospectral method to derive presaturation pulses for PARACEST imaging based on our accomplishments in relaxation optimized pulses and pulses robust to inhomogeneity.

Convergence
Pseudospectral convergence rates remain largely at the forefront of research. Quantification and proven results are only available for certain classes of systems.

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