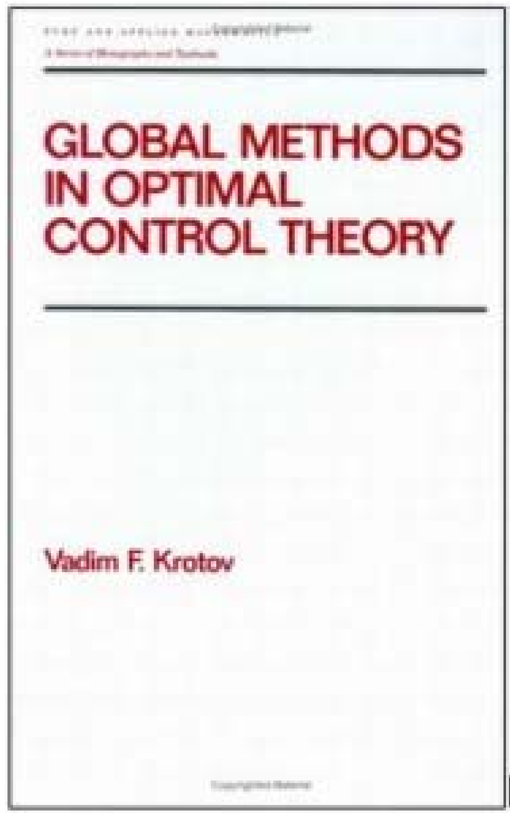
Which optimal control approach to use? – Reflections from Magnetic Resonance

G
R
A
P
E



Experiences by:

ZdenekTosner
Ivan Maximov

and NCN

C
l
o
s
e
c
o
l
l
a
b
o
r

Kavli Institute for Theoretical Physics, UCSB, Santa Barbara, June 17, 2009

Optimal control in relation to magnetic resonance: GRAPE

ELSEVIER

Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of Magnetic Resonance 172 (2005) 296–305

JMR Journal of Magnetic Resonance

www.elsevier.com/locate/jmr

Introduction of the method to NMR spectroscopy

Optimal control of coupled spin dynamics: design of NMR pulse sequences by gradient ascent algorithms

Navin Khaneja[a,*], Timo Reiss[b], Cindie Kehlet[b], Thomas Schulte-Herbrüggen[b], Steffen J. Glaser[b,*]

[a] *Division of Applied Sciences, Harvard University, Cambridge, MA 02138, USA*

[b] *Department of Chemistry, Technische Universität München, 85747 Garching, Germany*

Journal of Magnetic Resonance 197 (2009) 120–134

ELSEVIER

Contents lists available at ScienceDirect

Journal of Magnetic Resonance

journal homepage: www.elsevier.com/locate/jmr

JMR

Free open source Software distribution

Optimal control in NMR spectroscopy: Numerical implementation in SIMPSON

Zdeněk Tošner[a,b,*], Thomas Vosegaard[a], Cindie Kehlet[a], Navin Khaneja[c], Steffen J. Glaser[d], Niels Chr. Nielsen[a,*]

[a] *Center for Insoluble Protein Structures (inSPIN), Interdisciplinary Nanoscience Center (iNANO) and Department of Chemistry, University of Aarhus, Langelandsgade 140, DK-8000 Aarhus C, Denmark*

[b] *Department of Chemistry, Faculty of Science, Charles University in Prague, Hlavova 8, CZ-128 43, Czech Republic*

[c] *Division of Applied Sciences, Harvard University, Cambridge, MA 02138, USA*

[d] *Department of Chemistry, Technische Universität München, 85747 Garching, Germany*

Optimal control in relation to magnetic resonance: KROTOV

THE JOURNAL OF CHEMICAL PHYSICS **128**, 184505 (2008)

Optimal control design of NMR and dynamic nuclear polarization experiments using monotonically convergent algorithms

Ivan I. Maximov,[1] Zdeněk Tošner,[1,2] and Niels Chr. Nielsen[1,a)]
[1]*Center for Insoluble Protein Structures (inSPIN), Interdisciplinary Nanoscience Center (iNANO) and Department of Chemistry, University of Aarhus, Langelandsgade 140, DK-8000 Aarhus C, Denmark*
[2]*Faculty of Mathematics and Physics, Charles University in Prague, Ke Karlovu 3, 121 16 Praha 2, Czech Republic*

++ MORE IMPORTANTLY seminal papers by

Tannor
Rabitz
Maday and Turinici

D. J. Tannor, V. Kazakov, and V. Orlov, in *Time-Dependent Quantum Molecular Dynamics*, edited by J. Broeckhove and L. Lathouwers (Plenum, New York, 1992).
Y. Maday and G. Turinici, J. Chem. Phys. **118**, 8191 (2003).
W. Zhu and H. Rabitz, J. Chem. Phys. **109**, 385 (1998).

Issues in comparison:

- Convenience of use

- Computational time

- Robustness with respect to outcome

- Sensitivity towards starting guesses

- Sensitivity towards local minima

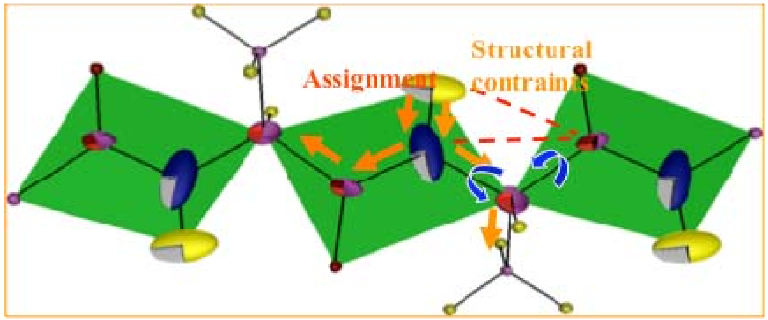
- Running cost issues

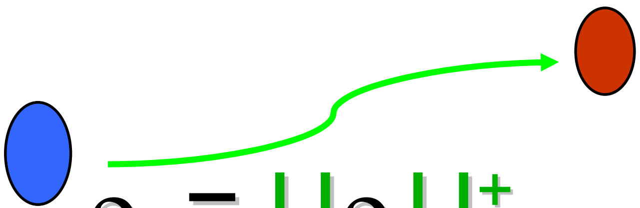
- Challenges with respect to system dimensionalities

Optimal control design of NMR experiments

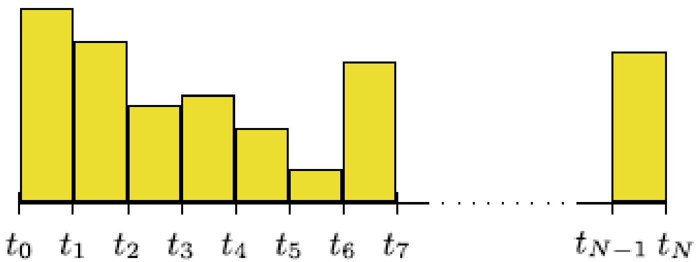
- improved sensitivity
- band selective operation
- less rf power consumption

Kehlet et al, *JACS, 2004*
Maximov et al, *J. Chem. Phys., 2008*
Tosner et al, *J. Magn. Reson. 2009*





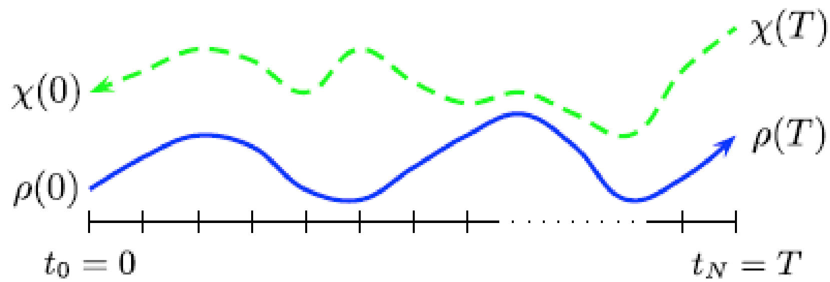
$$\rho_f = U \rho_i U^+$$



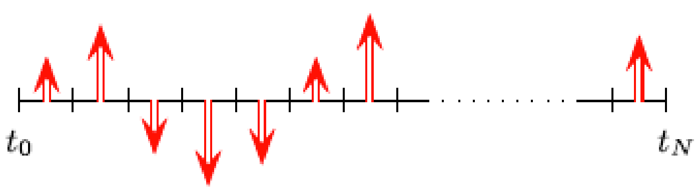
Optimal control => Design of Ū

Cost function:

$$J_i = \phi_i - \lambda \int_0^T \sum_k u_k^2(t) \mathrm{d}t$$



State to State or optimization of U or H_{eff}



Spin Hamiltonian

$$H(t) = H_{int}(t) + \sum_k \omega_k(t) H_k$$

Equation of motion

$$\frac{d}{dt}U(t) = -iH(t)U(t)$$

Target functional

$$J = \Phi_i - \lambda \int_0^T \sum_k \omega_k^2(t) dt$$

efficiency *penalty on rf power*

Transfer between Hermitian operators ρ_0 and C

$$\Phi_1 = \mathrm{Tr}\left\{C^\dagger U(T)\rho_0 U^\dagger(T)\right\}$$

Transfer between non-Hermitian operators

$$\Phi_2 = \mathrm{Re}\left\{\mathrm{Tr}\left\{C^\dagger U(T)\rho_0 U^\dagger(T)\right\}\right\}$$

$$\Phi_3 = \left|\mathrm{Tr}\left\{C^\dagger U(T)\rho_0 U^\dagger(T)\right\}\right|^2$$

Synthesis of desired propagator U_D

$$\Phi_4 = \left|\mathrm{Tr}\left\{U_D^\dagger U(T)\right\}\right|^2$$

The problem is solved by Lagrange method using adjoint variable $B(t)$

$$\frac{d}{dt}B(t) = -iH(t)B(t) \qquad B(T) = \frac{\partial \Phi_i}{\partial U(T)}$$

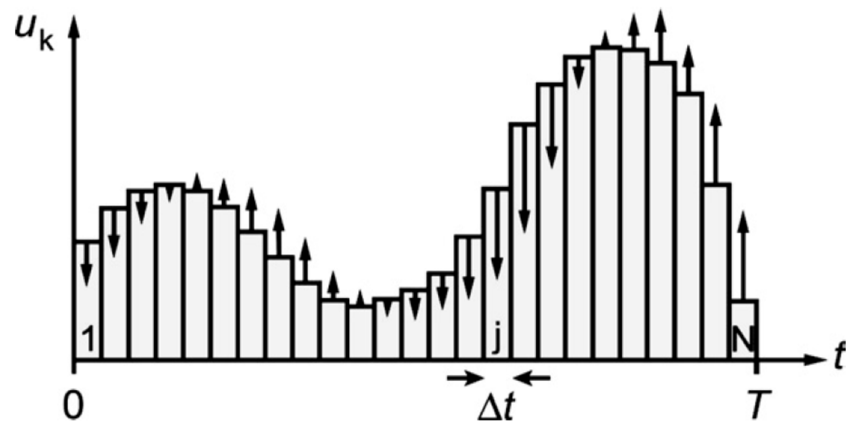
For Krotov algorithm, corrections in Φ_i (i=1,2,3) are necessary [1]

$$\Phi_i' = \Phi_i + \kappa \mathrm{Tr}\left\{U^\dagger(T)U(T)\right\}$$

A gradient based approach to optimal control:
The GRAPE algorithm

Hamiltonian:

$$H(t) = H_{int}(t) + H_{rf}(t)$$



Propagator:

$$\rho(t_N) = U_N ... U_2 U_1 \rho(0) U_1^\dagger U_2^\dagger ... U_N^\dagger$$

$$\lambda(t_j) = U_{j+1}^\dagger ... U_N^\dagger C U_N ... U_{j+1}$$

$$U_j = \exp\{-i \int_{t_j}^{t_{j+1}} H_{int}(t)dt + \omega_{rf,j}^x I_x + \omega_{rf,j}^y I_y\}$$

Change of rf: gradient:

$$\omega_{rf,j}^q \rightarrow \omega_{rf,j}^q + \epsilon Tr\{i\Delta t_j [I_q, \rho(t_j)]\lambda(t_j)\}$$

Iterative procedure changing all pulses at the same time!

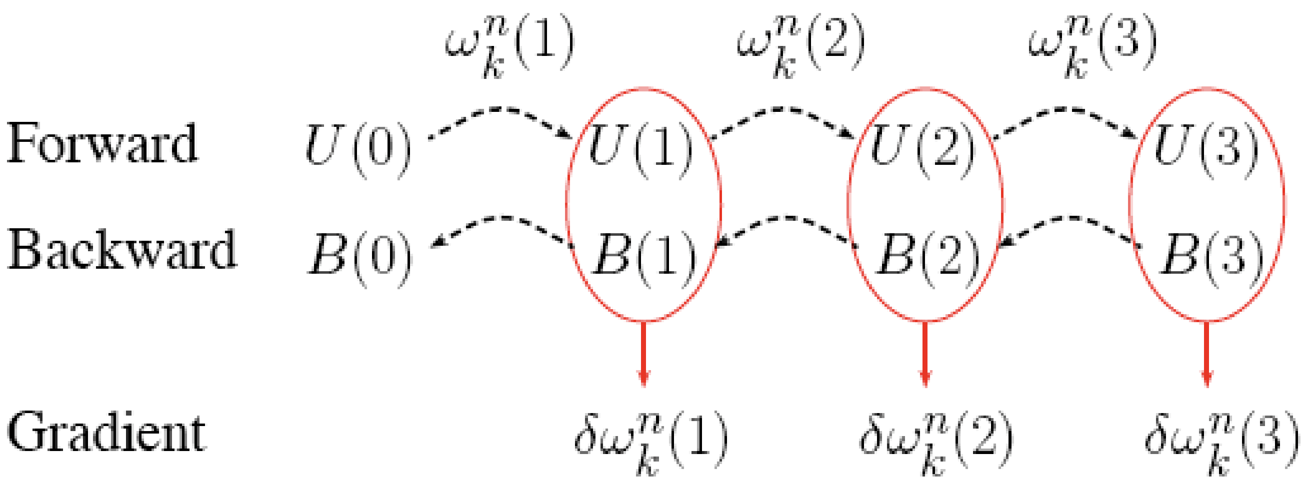
Khaneja, Glaser et al.

Flow of calculations: GRAPE

GRAPE

delayed feedback

Optimal pulse sequence parameters $\omega_k(t)$ are obtained in "steepest ascent" manner. It updates $\omega_k(t)$ along the direction determined by gradient of the functional J (details in Ref. [2]).



$$\omega_k^{n+1}(t) = \omega_k^n(t) + \alpha\delta\omega_k^n(t)$$

α determined by line-search

$\Rightarrow$ REPEATED EVALUATION OF THE FUNCTIONAL

Another monotonic convergence method:
The KROTOV algorithm

In each step we perform both Forward and Backward calculation.

$$\frac{d}{dt}U_n(t) = -i\left[H_0 + \sum_k \omega_{k,n}(t) I_k\right] U_n(t),$$

$$U_n(0) = \mathbf{1},$$

$$\omega_{k,n}(t) = (1-\delta)\tilde{\omega}_{k,n-1}(t) + \frac{\delta}{\lambda}\,\mathrm{Im}\{\mathrm{Tr}[B^{\dagger}_{n-1}(t) I_k U_n(t)]\},$$

δ and η is algorithm unifying parameters

$$\frac{d}{dt}B_n(t) = -i\left[H_0 + \sum_k \tilde{\omega}_{k,n}(t) I_k\right] B_n(t),$$

$$B_n(T) = \frac{\partial \phi_4}{\partial U(T)} = U_D\,\mathrm{Tr}[U_D^{\dagger} U_n(T)],$$

$$\tilde{\omega}_{k,n}(t) = (1-\eta)\omega_{k,n}(t) + \frac{\eta}{\lambda}\,\mathrm{Im}\{\mathrm{Tr}[B^{\dagger}_n(t) I_k U_n(t)]\},$$

Tannor:
$\delta = 1$; $\eta = 0$

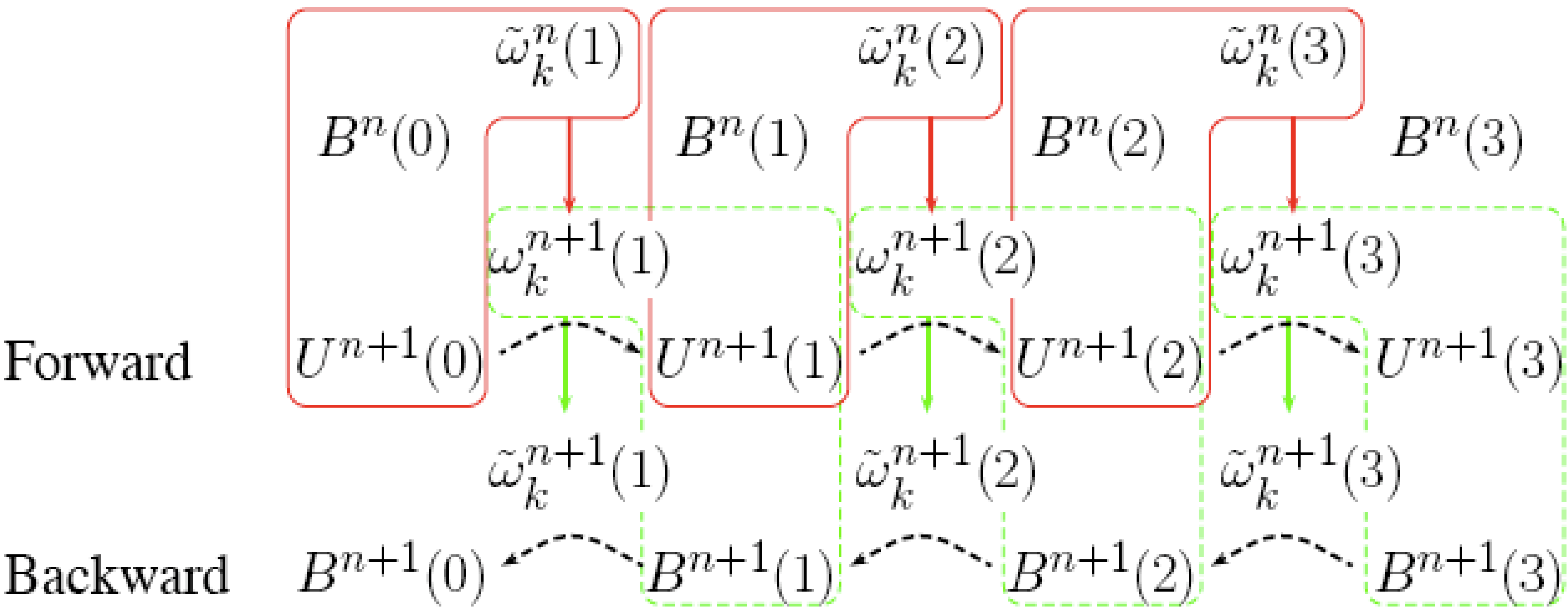
Zhu, Rabitz:
$\delta = 1$; $\eta = 1$

Flow of calcuations: KROTOV

Krotov algorithm

immediate feedback

Iteratively solves optimality conditions (given by Pontryagin maximum principle) in self-consistent manner. There are explicit formulas for optimal pulse sequence parameters $\omega_k(t)$ (details in Ref. [1]).



- forward and backward propagations done with separate rf parameters
- mixing controled by parameters δ and η

Advantage: NO EVALUATION OF THE FUNCTIONAL!!!

Evaluation criteria: Many sides of the coin

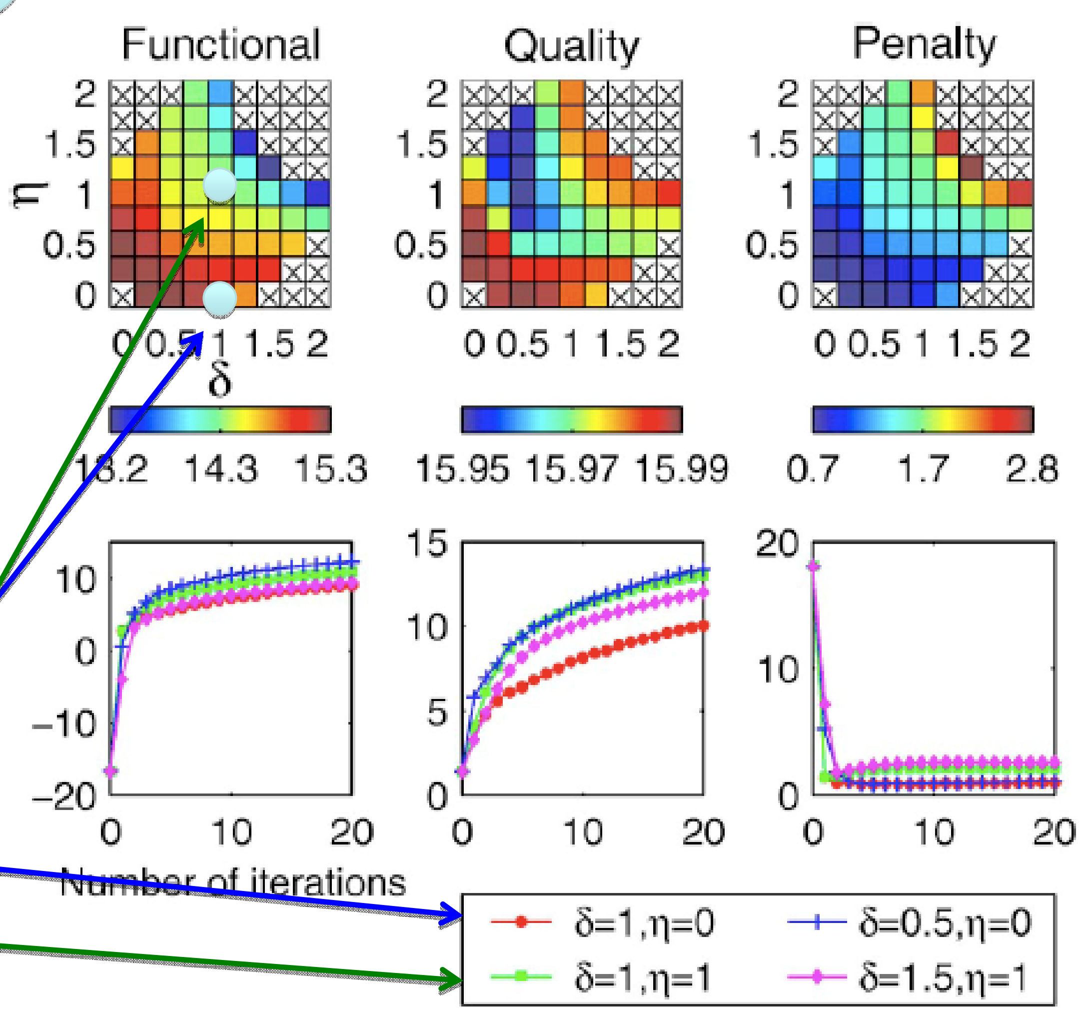
Functional

Penalty (running cost)

$$J = \Phi_i - \lambda \int_0^T \sum_k \omega_k^2(t)dt$$

Quality / Efficiency (final cost)

Krotov also needs to worry about δ and η



Tannor

Rabitz

Niels Chr. Nielsen

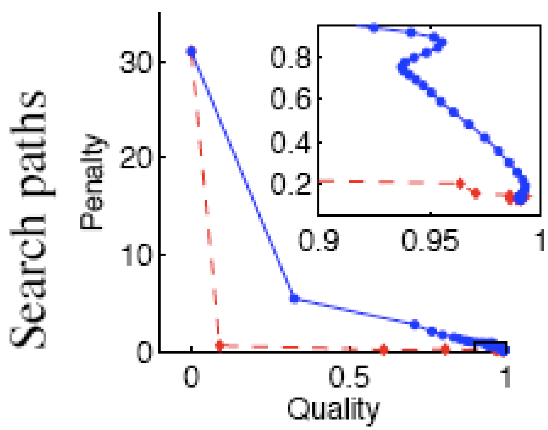
Transfer between operators

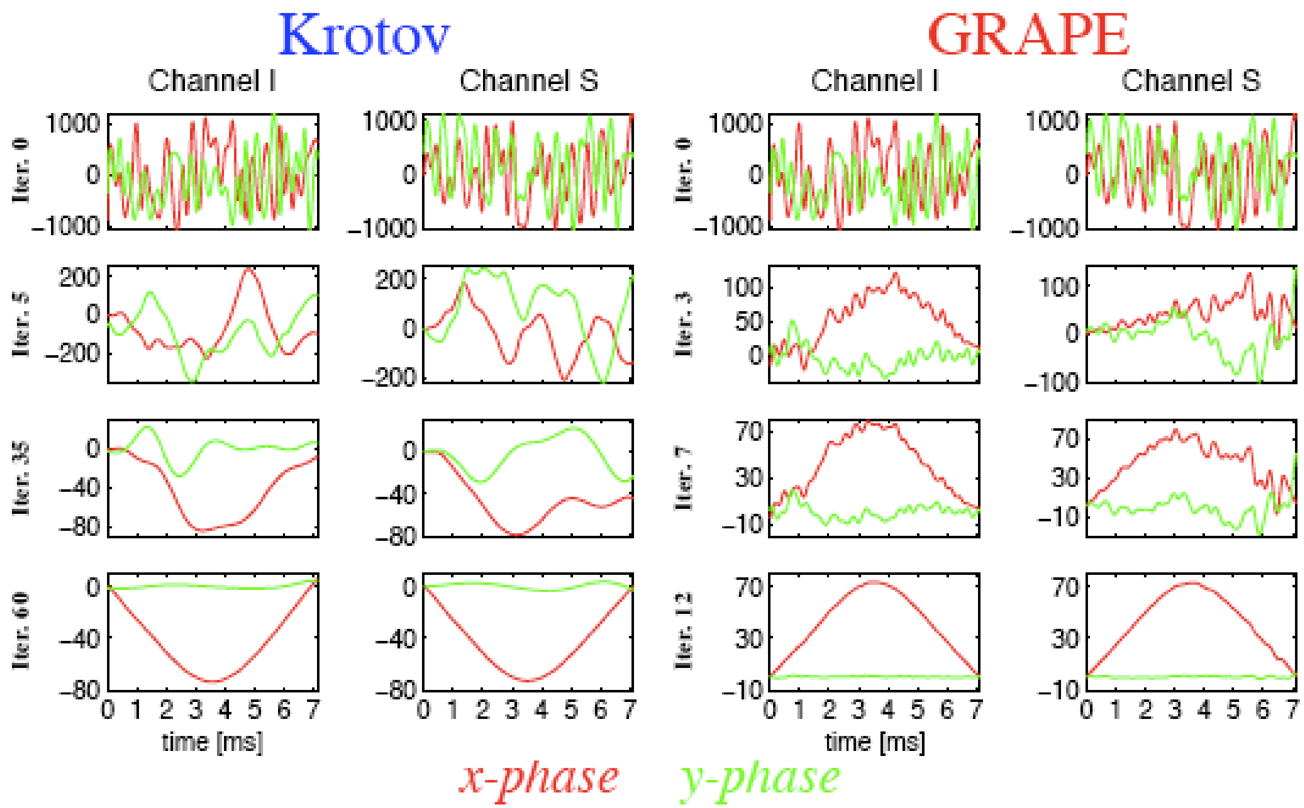
<u>Goal:</u> pulse sequence with minimal rf power for efficient transfer of polarization $I_x \rightarrow S_x$ in two spin-1/2 system with weak J-coupling.

$$H_{int} = \pi J 2 I_z S_z$$

$$J\text{=140 Hz}, T = \frac{1}{J}$$

$$\Phi_1', \text{100\% efficiency}$$



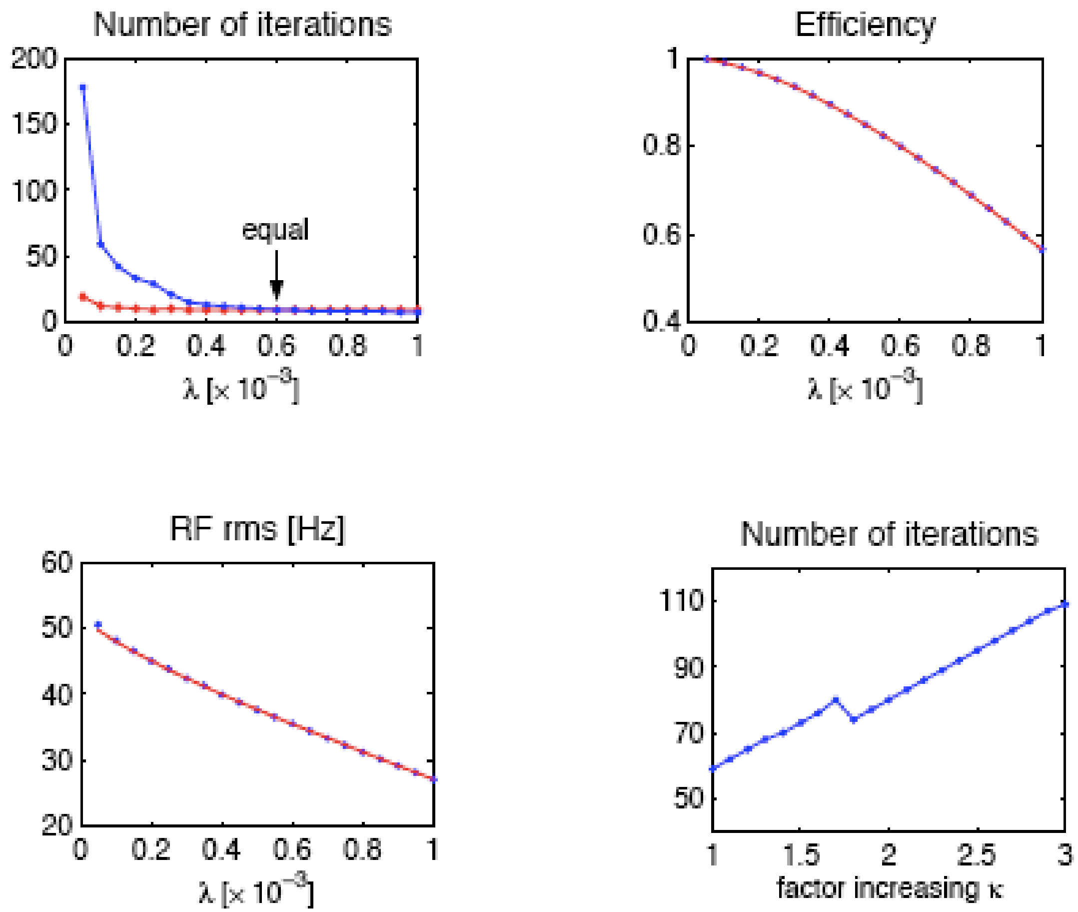


GRAPE and Krotov algorithms take different paths to essentially the same result. Effective Hamiltonian of the pulse sequence is $\overline{H} = \pi J \left(I_z S_z + I_y S_y \right) + \beta \left(I_x + S_x \right)$

inSPIN Center for Insoluble Protein Structures

Niels Chr. Nielsen

Transfer between operators



When rf penalty parameter λ is increased Krotov algorithm converges faster and is eventually faster than GRAPE. Increasing κ slows down the algorithm.

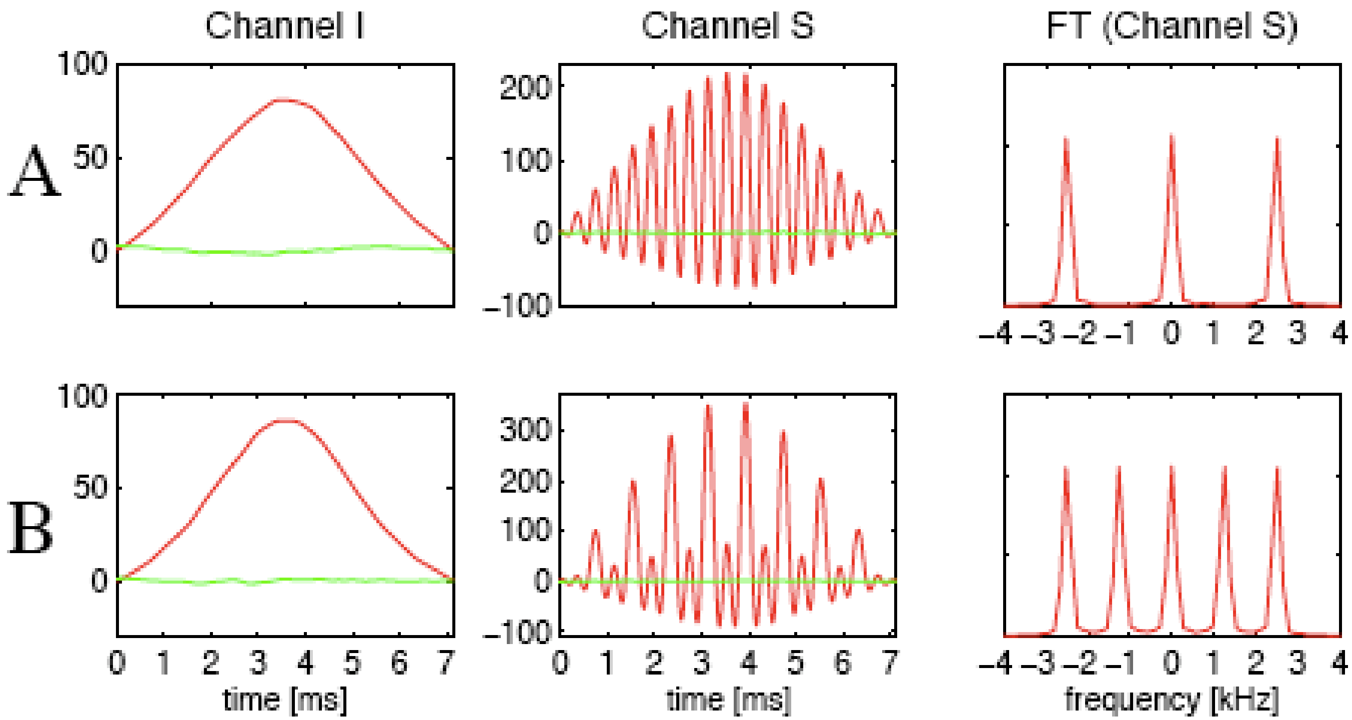
BroadbandOptimization

Goal: single pulse sequence optimal for a set of spin system parameters

$$H_{int} = \omega_S S_x + \pi J 2 I_z S_z$$

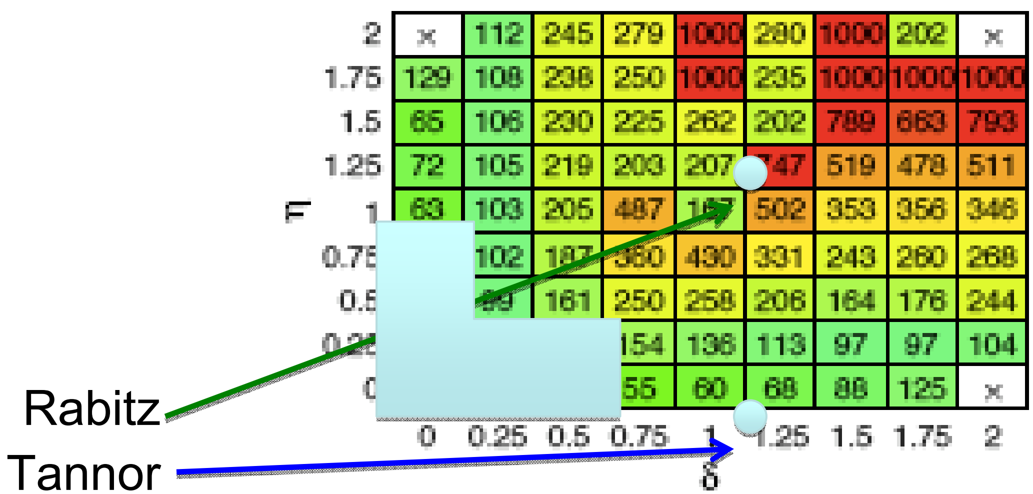
A: $\omega_S = -2.5, 0, 2.5\,\mathrm{kHz}$

B: $\omega_S = -2.5, -1.25, 0, 1.25, 2.5\,\mathrm{kHz}$



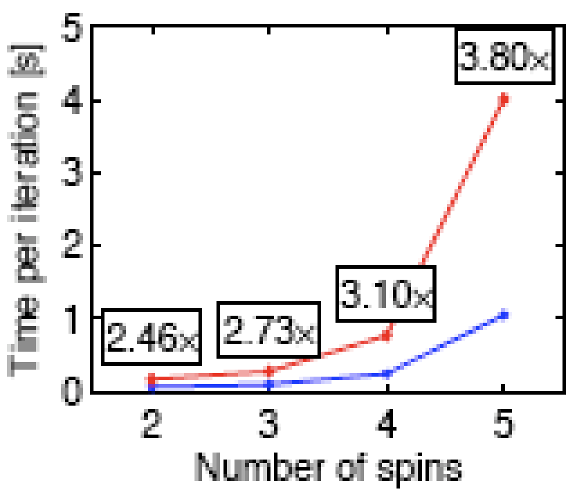
BroadbandOptimization: Comparing Speed

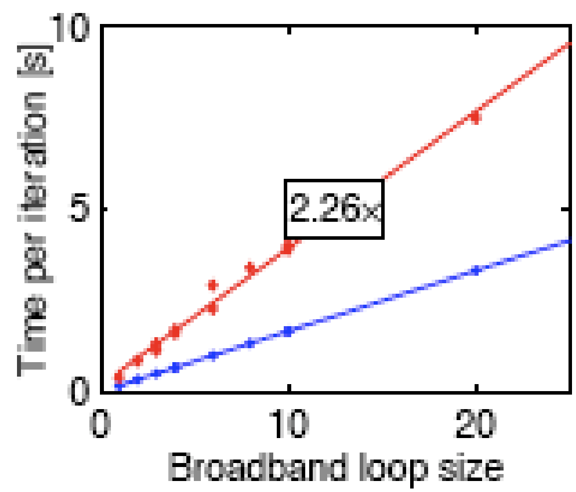
For Krotov algorithm, number of iterations also depends on parameters δ and η. Small values are recommendable.



BroadbandOptimization: Comparing Speed

Krotov algorithm is much faster per iteration. This advantage grows with complexity of the problem - when increasing size of a spin system or increasing loops over broadband parameters.





Synthesis of a desiredpropagator: IsotropicMixing

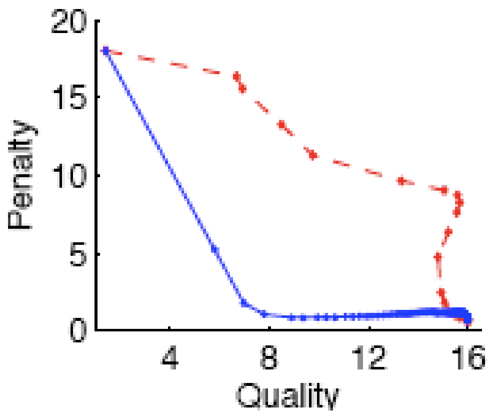
Goal: pulse sequence with minimal rf power for creation of isotropic mixing Hamiltonian in two spin-1/2 system with weak J-coupling.

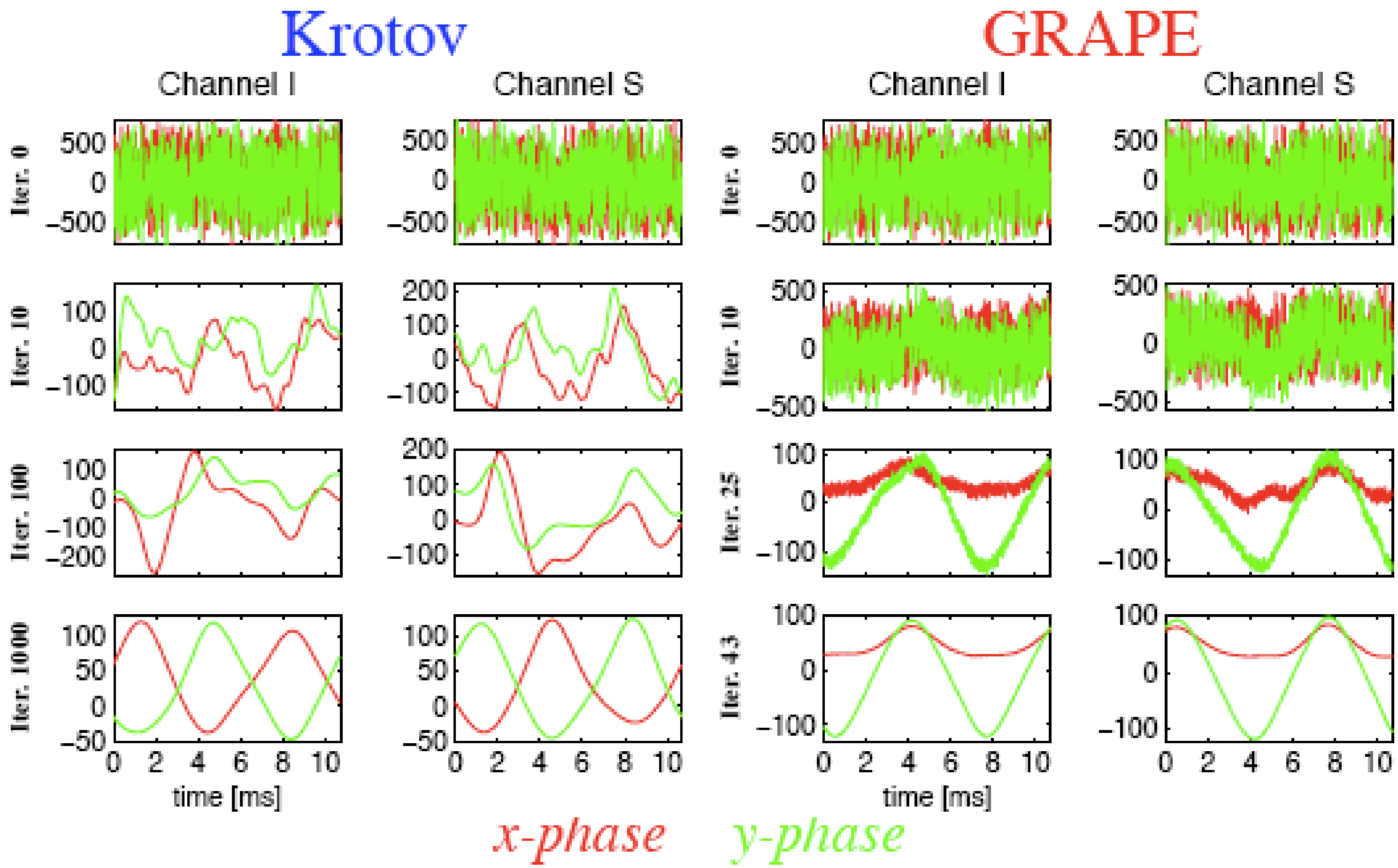
$$H_{int} = \pi J 2 I_z S_z$$

$$U_D = \exp\{-i\pi \mathbf{I}\cdot\mathbf{S}\}$$

J=140 Hz, $T = \frac{3}{2J}$

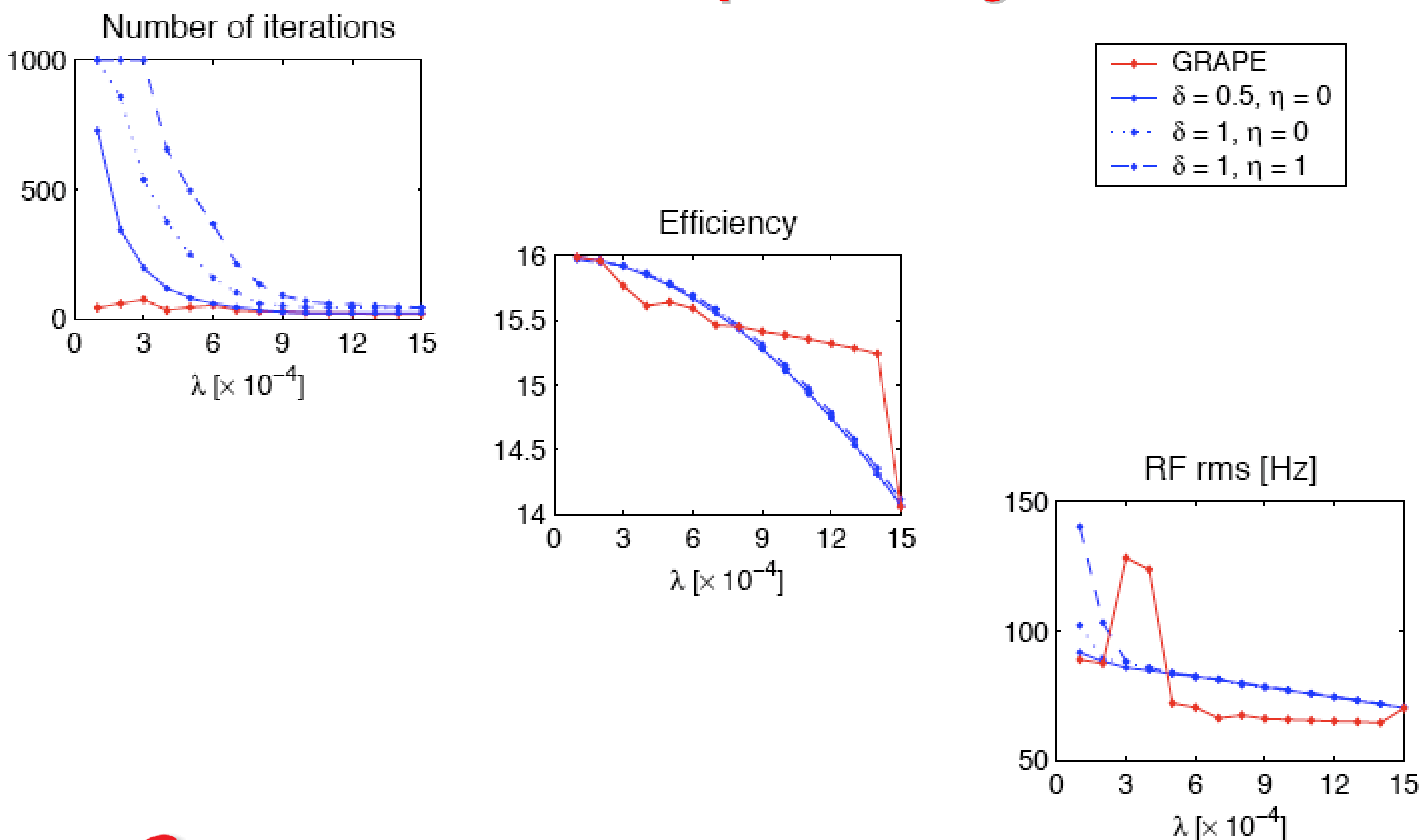
Φ_4, 100% efficiency





inSPIN Center for Insoluble Protein Structures
Niels Chr. Nielsen

Synthesis of a desiredpropagator: IsotropicMixing



Optimizationstatistics: INEPT

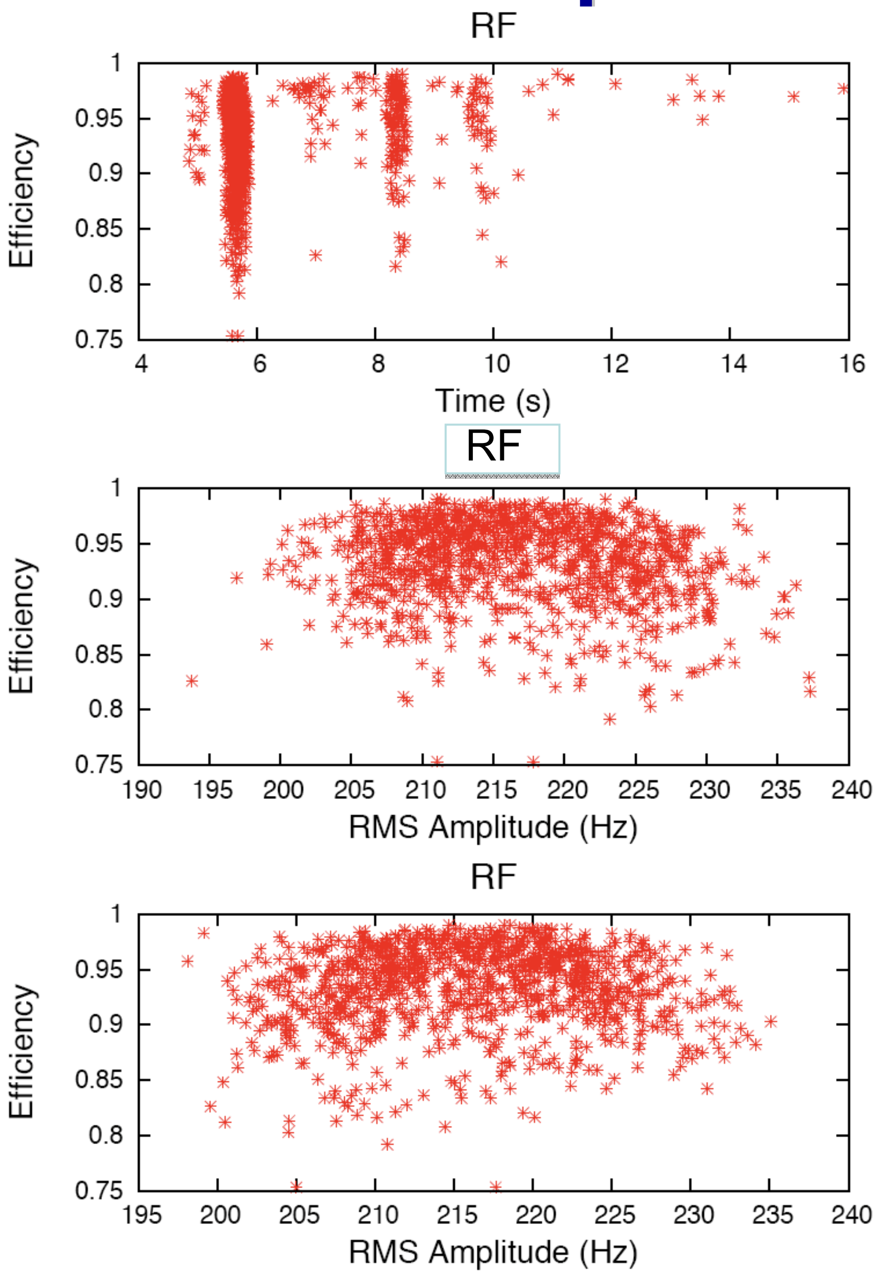


FIG. 15: GRAPE: statistics in INEPT case, N=200.

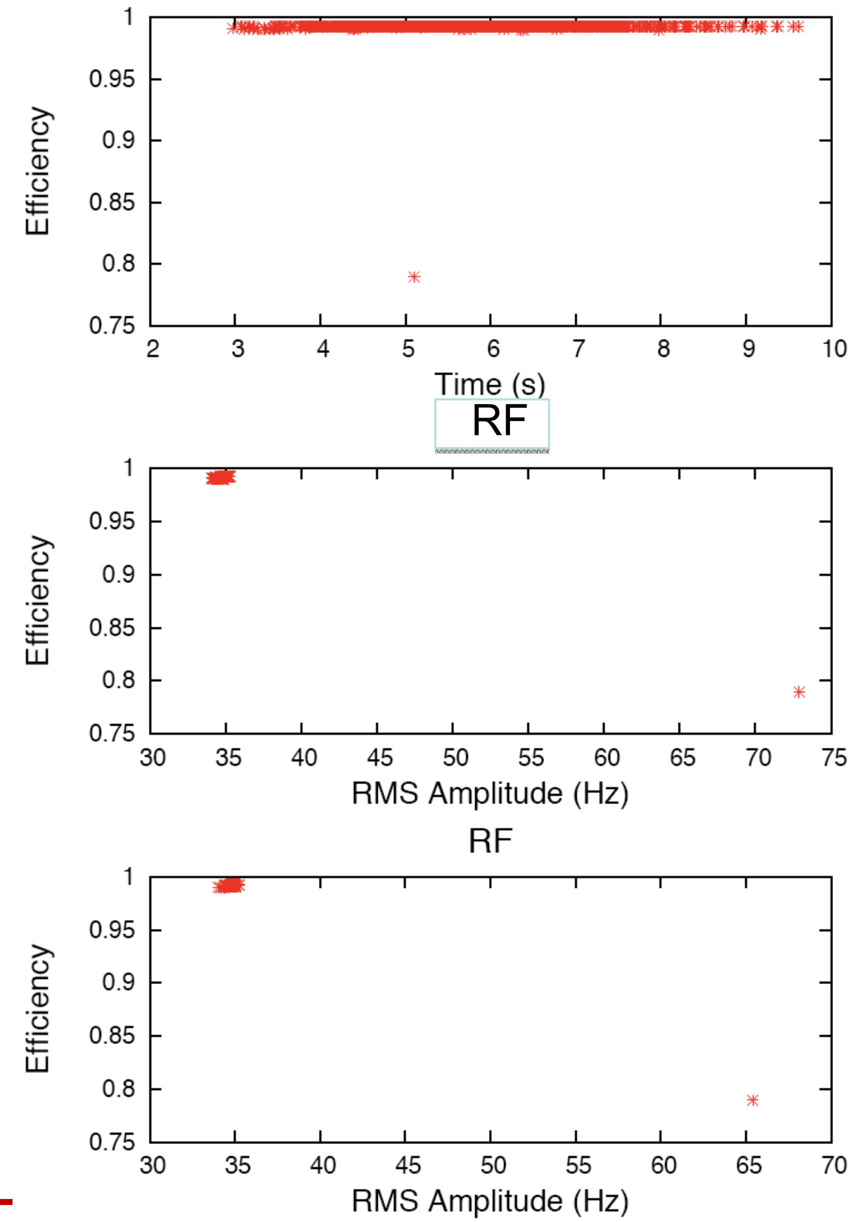


FIG. 13: Krotov: statistics in INEPT case, N=200.

Optimizationstatistics: DNP

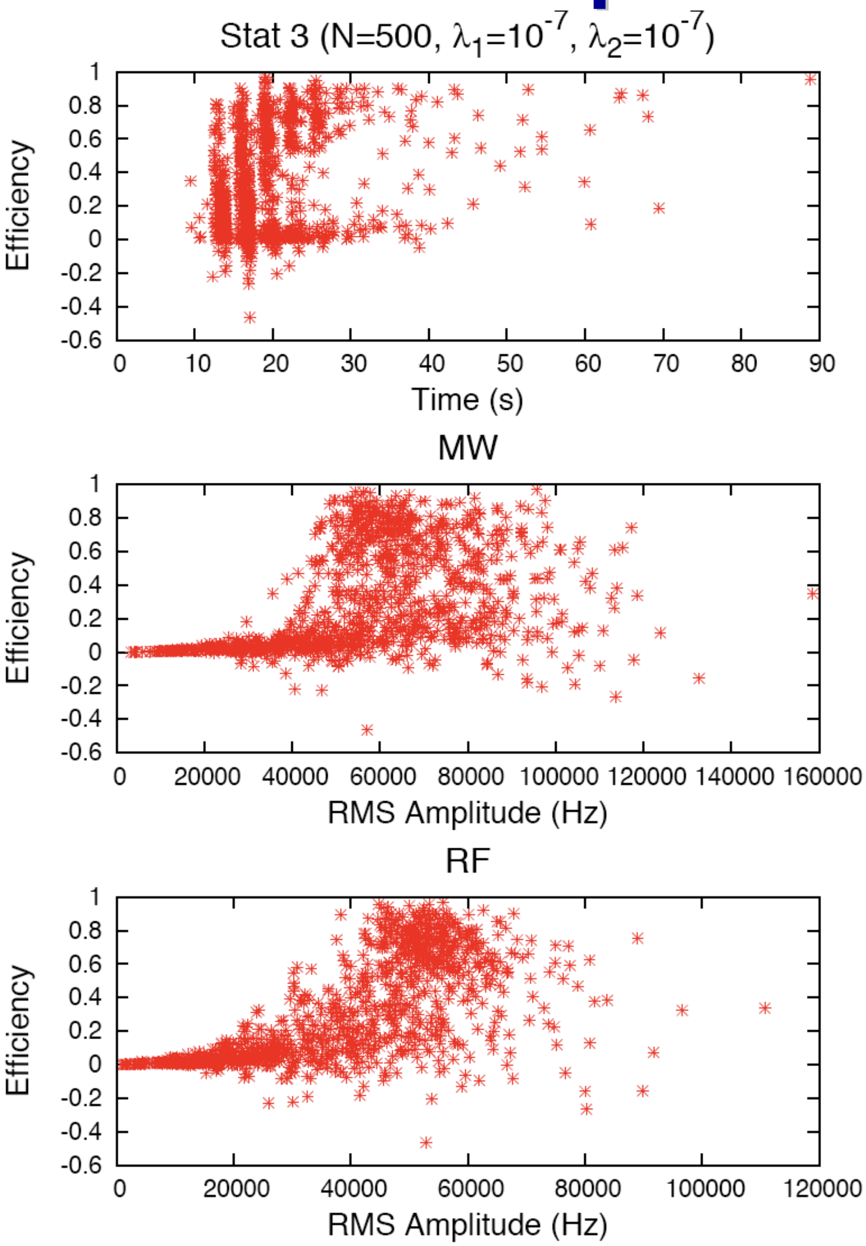


FIG. 10: GRAPE statistics

parameter	range
δ	$[1, 10^{-10}]$
η	$\eta = \delta$
λ_1	$[10^{-3}, 10^{-13}]$
λ_2	$[\lambda_1, 10^{-15}]$
N	300
$A/2\pi$	6 MHz
$B/2\pi$	3 MHz
$\omega_n/2\pi$	10MHz

$$H = -\omega_n I^z + A I^z S^z + B I^x S^z .$$

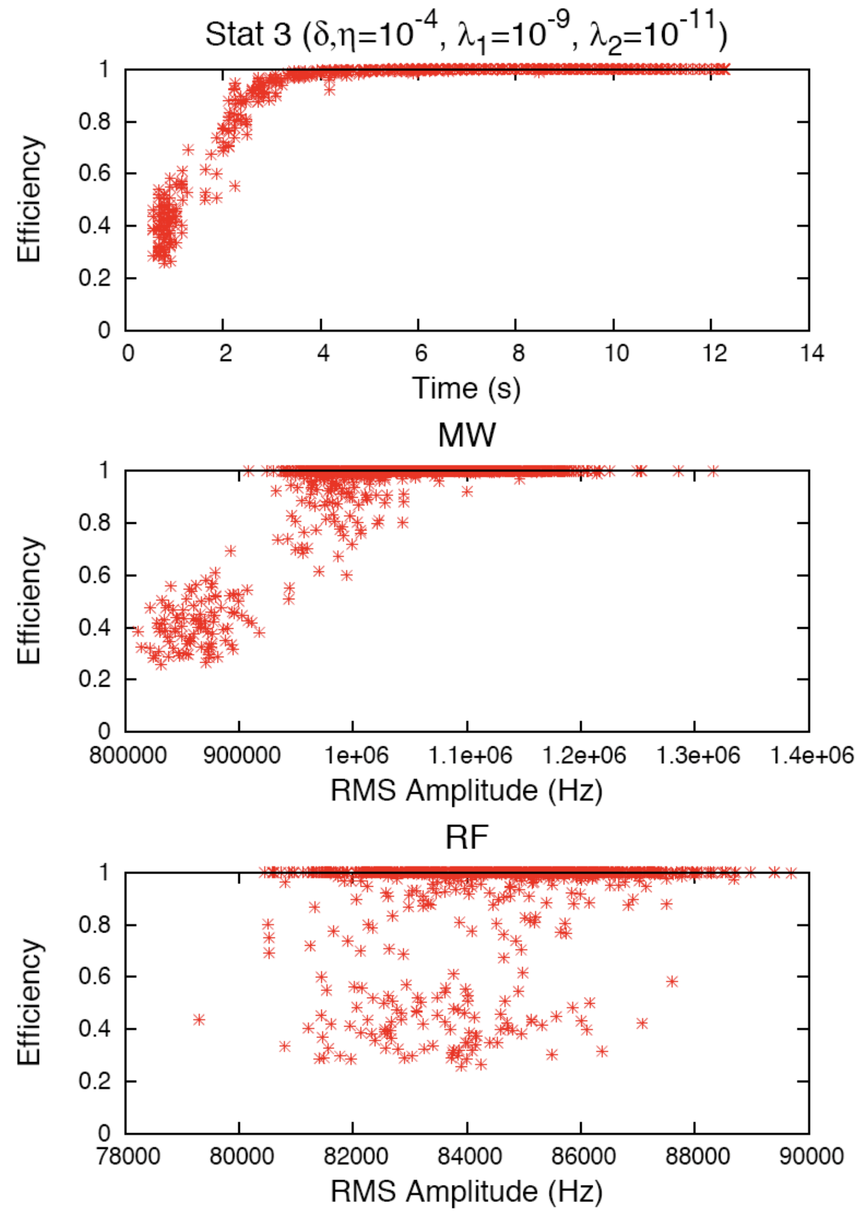


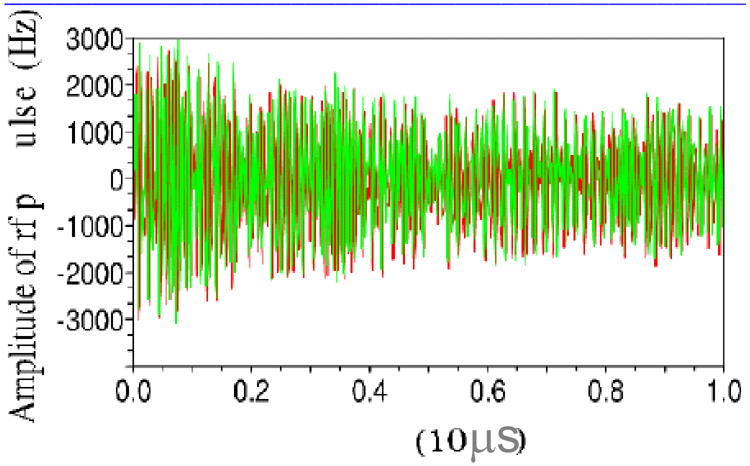
FIG. 5: Krotov statistics set III.

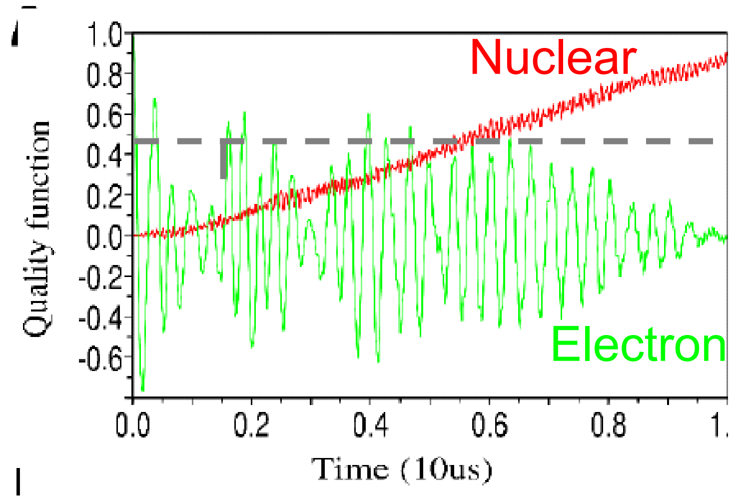
Optimal control and DNP

S(Electron)-I(Nucleus)
two-spin system

$$H_0 = \Omega_n I_z + A S_z I_z + B S_z I_x$$

$\Omega_n / 2\pi = -14$ MHz [1]H Zeeman
$A/2\pi = 6$ MHz
$B/2\pi = 3$ MHz

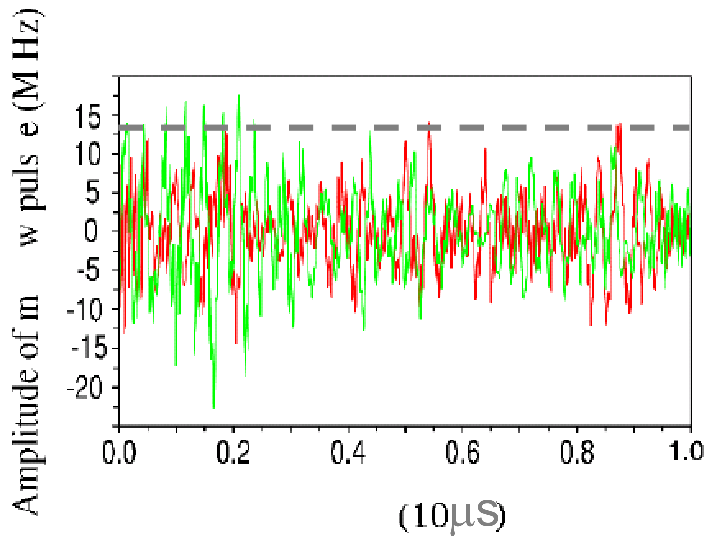




$S_x \rightarrow I_z$

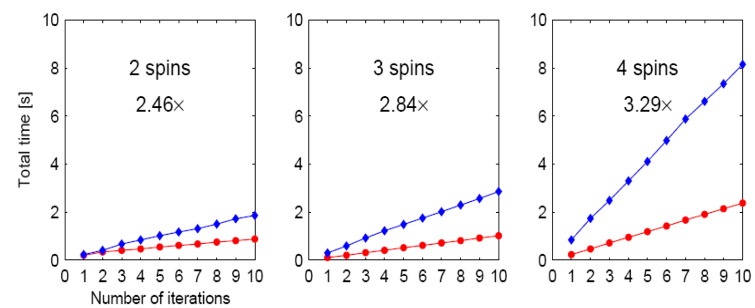
A: secular part of hyperfine interaction

B: Pseudo secular part of hyperfine interaction



Computational time

Krotov: Red
GRAPE: Blue



MW @ 9 GHz (X-band) –
ca. 0.3 T (14 MHz) NMR

Jeschke, Schweiger, Mol Phys 1996

Maximov, Tosner, Nielsen, JCP (2008)

inSPIN Center for Insoluble Protein Structures

Niels Chr. Nielsen

Midwayconclusions

- Both GRAPE and Krotov algorithms are capable to find global maxima.
- Including rf power penalty to optimization process simplifies resulting pulse sequences. These sequences are more feasible for analytical analysis.
- Krotov algorithm is significantly faster per iteration compared to GRAPE and thus is superior to GRAPE for large spin systems or broadband optimizations.
- Krotov algorithm may need more iterations when low rf penalty is used but it converges faster when using stronger rf penalties.
- Combined optimization approach when the problem is first tackled by Krotov algorithm with high rf penalty and then refined by GRAPE (with lower rf penalty) may lead to quick results.
- Krotov appears significantly less sensitive to appropriate choice of initial guesses.
- Krotov needs optimization of δ and η to obtain stability – low values are relatively safe, in particular very low values in which may slow down optimimizations relative to high values
- Krotov works also in cases where you have VERY FEW CONTROLS (e.g., 2-3 pulses)

Remember

Everything is based on “local experience” and all conclusions may be different (?) for other systems/cases etc – and all depends a lot on the objective of you optimization (including costs, robustness and size of system)

Optimal control is a great tool independent on whether you are proGRAPE or proKROTOV – coexistence through complementarity may be the optimum