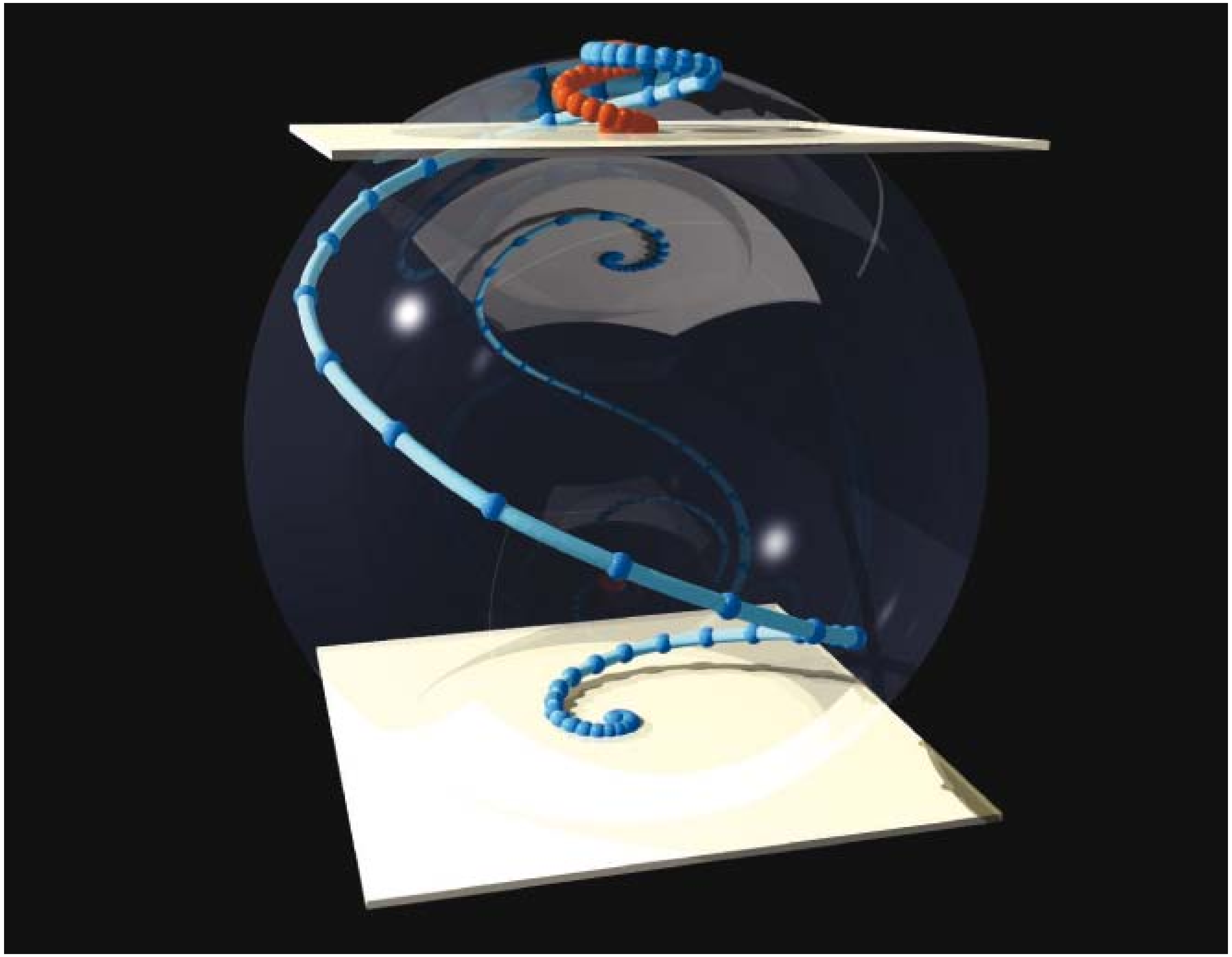
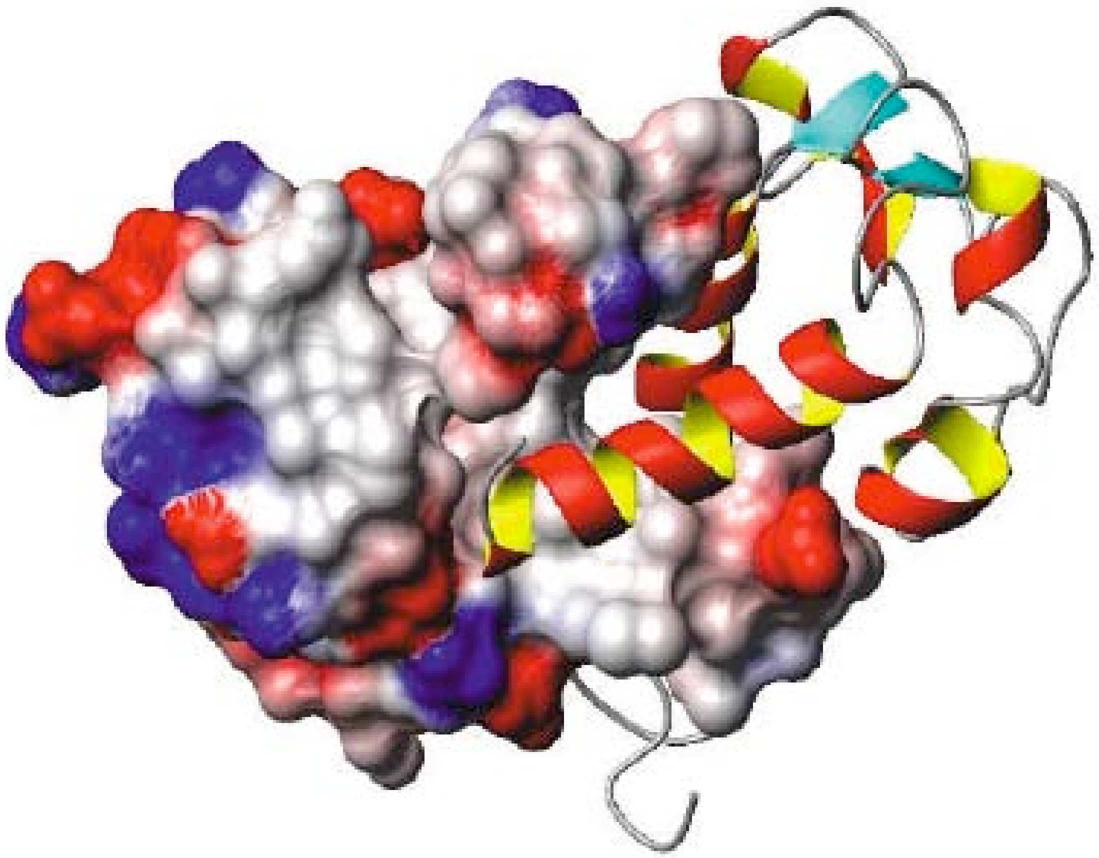
Optimal Control of Spin Dynamics

Steffen Glaser, TU München



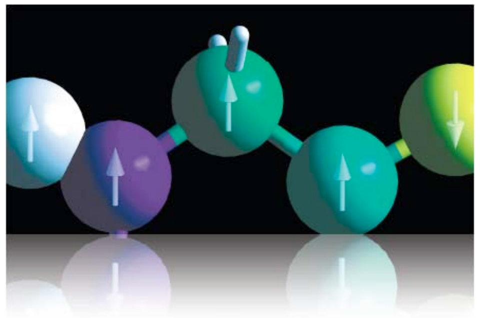


Nobel Prizes:

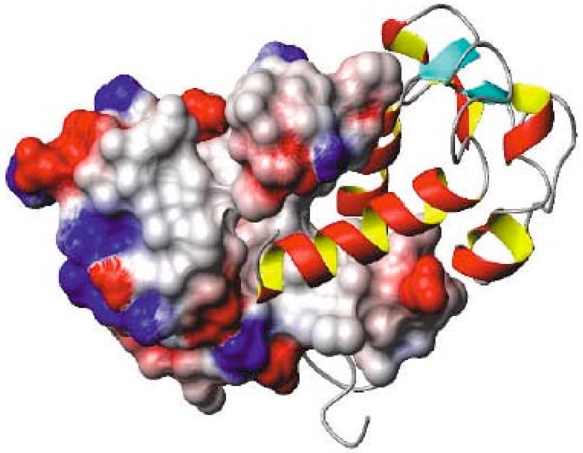
1952: Edward Purcell, Felix Bloch (Physics)
1991: Richard Ernst (Chemistry)
2002: Kurt Wüthrich (Chemistry)
2003: Paul Lauterbur, Peter Mansfield (Medicine)

Optimal Control in Spin Systems

physical limits of spin dynamics

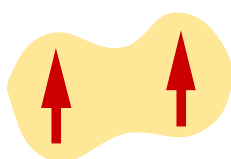


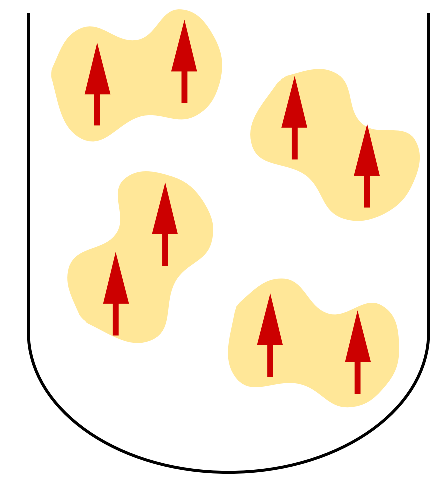
spectroscopy



quantum computing

local spin manipulation and imaging



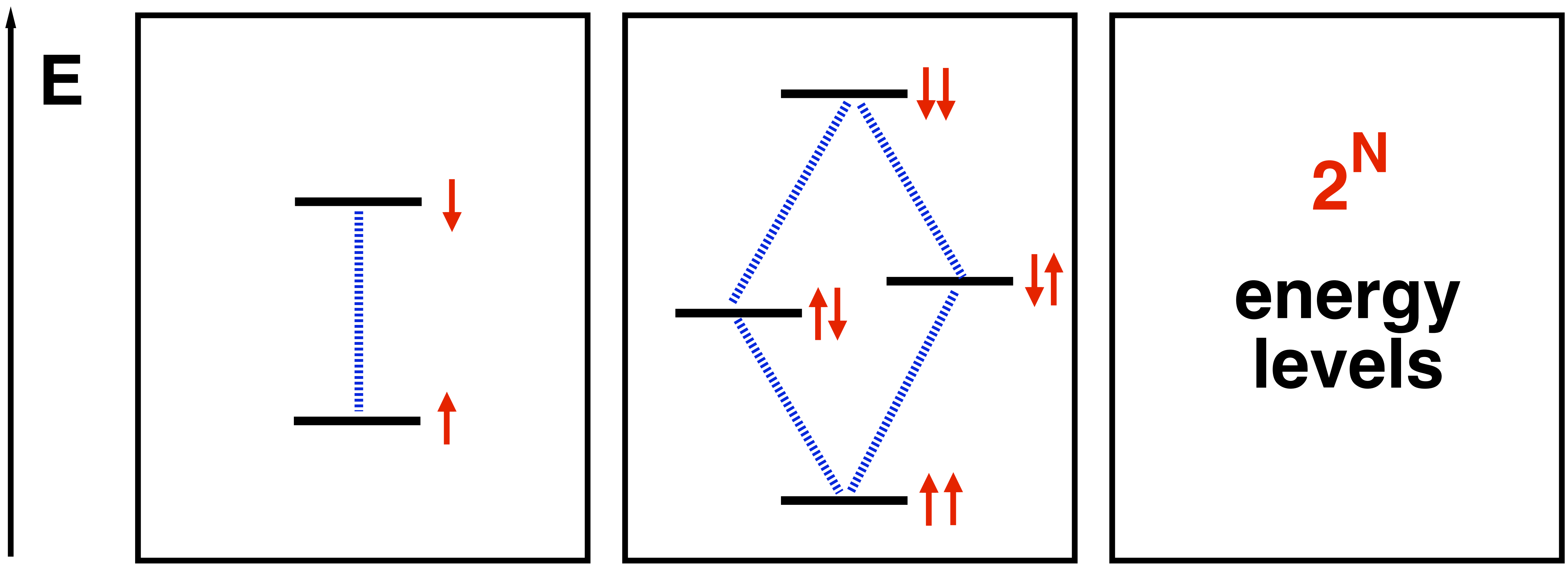


Pure state $|\Psi\rangle$

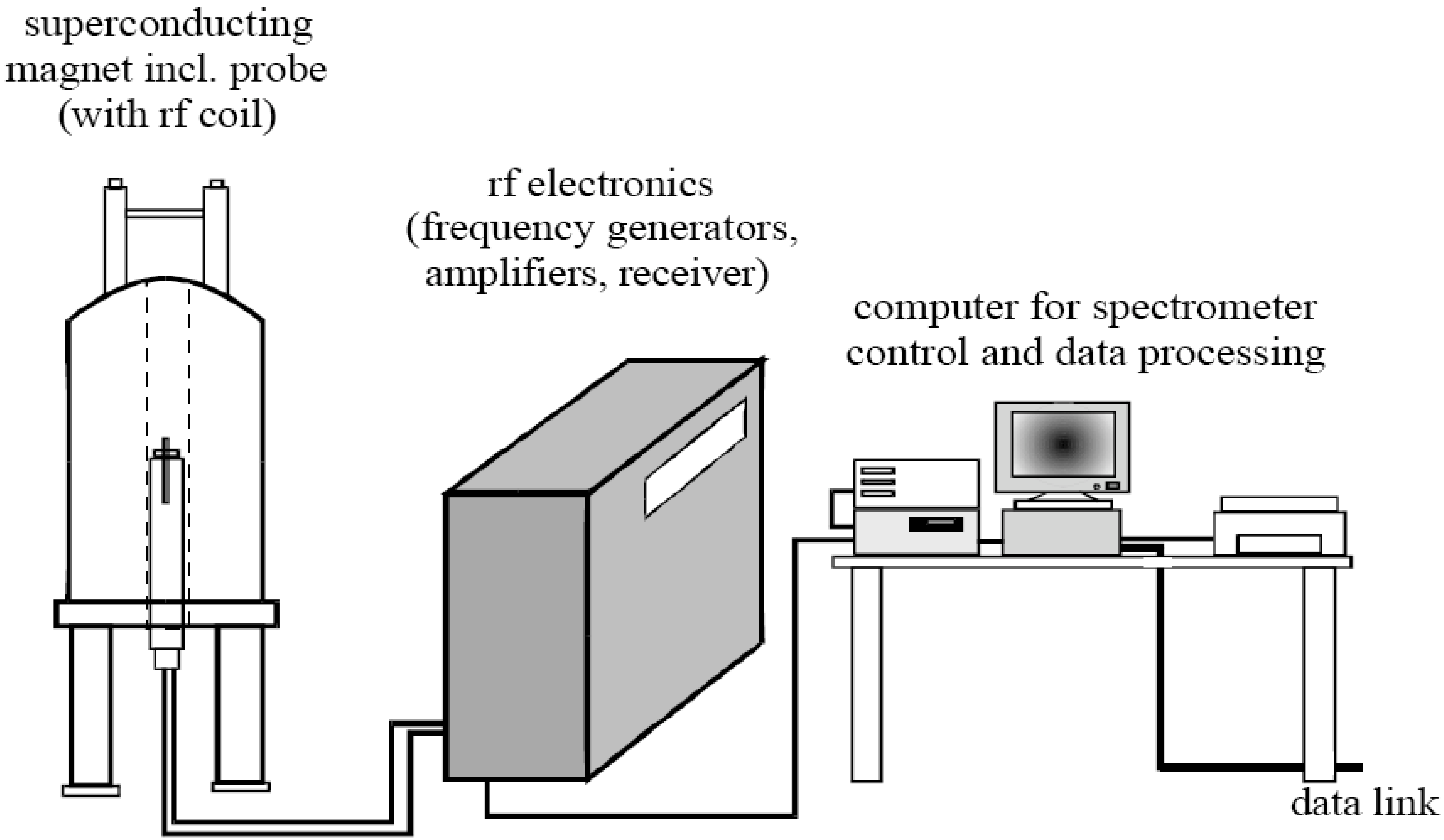
Density operator $\rho = \overline{|\Psi\rangle\langle\Psi|}$

Energy levels

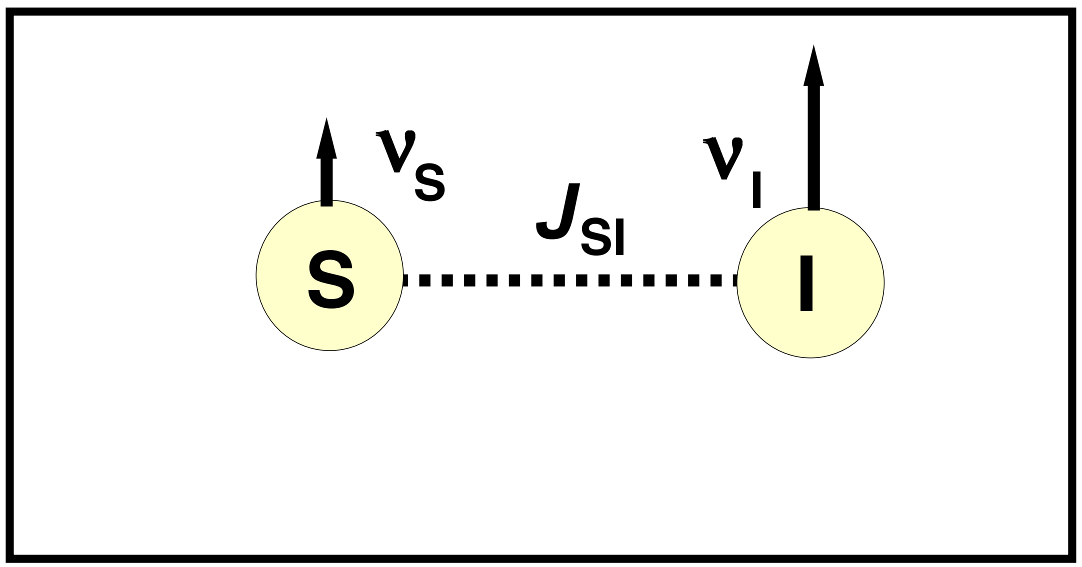
1 spin 1/2 2 spins 1/2 N spins 1/2



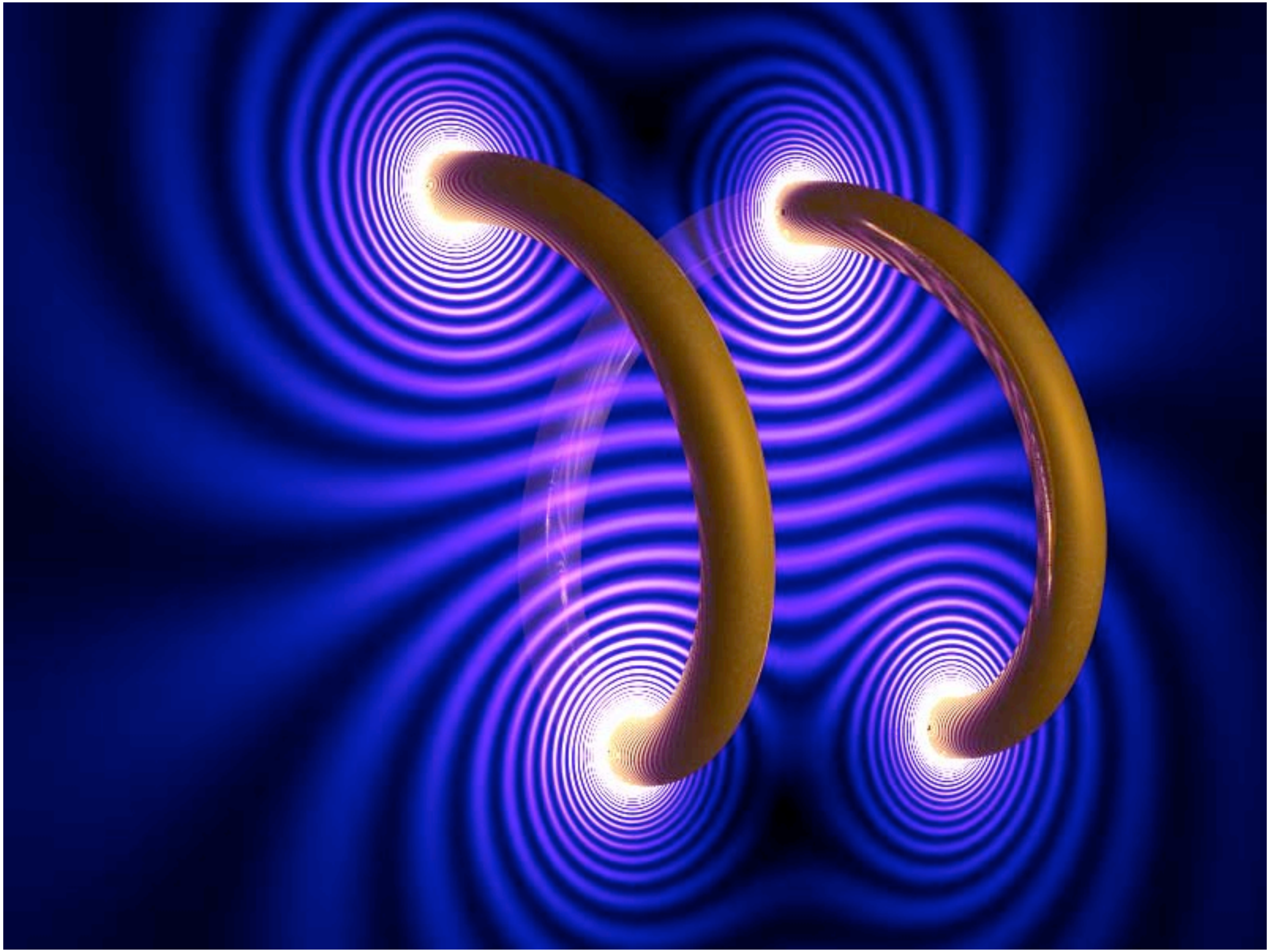
How do you measure an NMR signal?



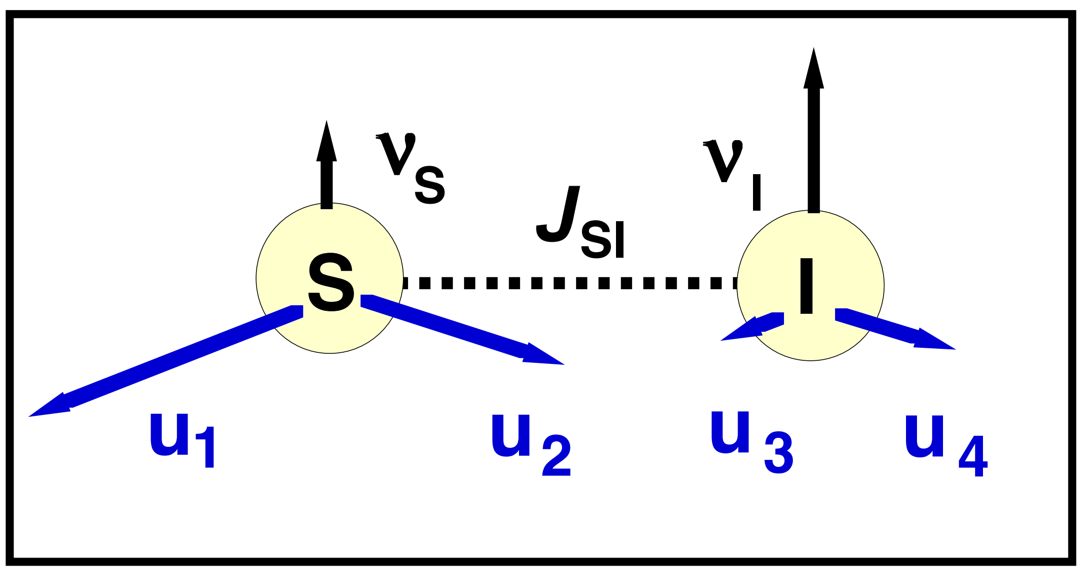

BRUKER
900
UltraStabilized™



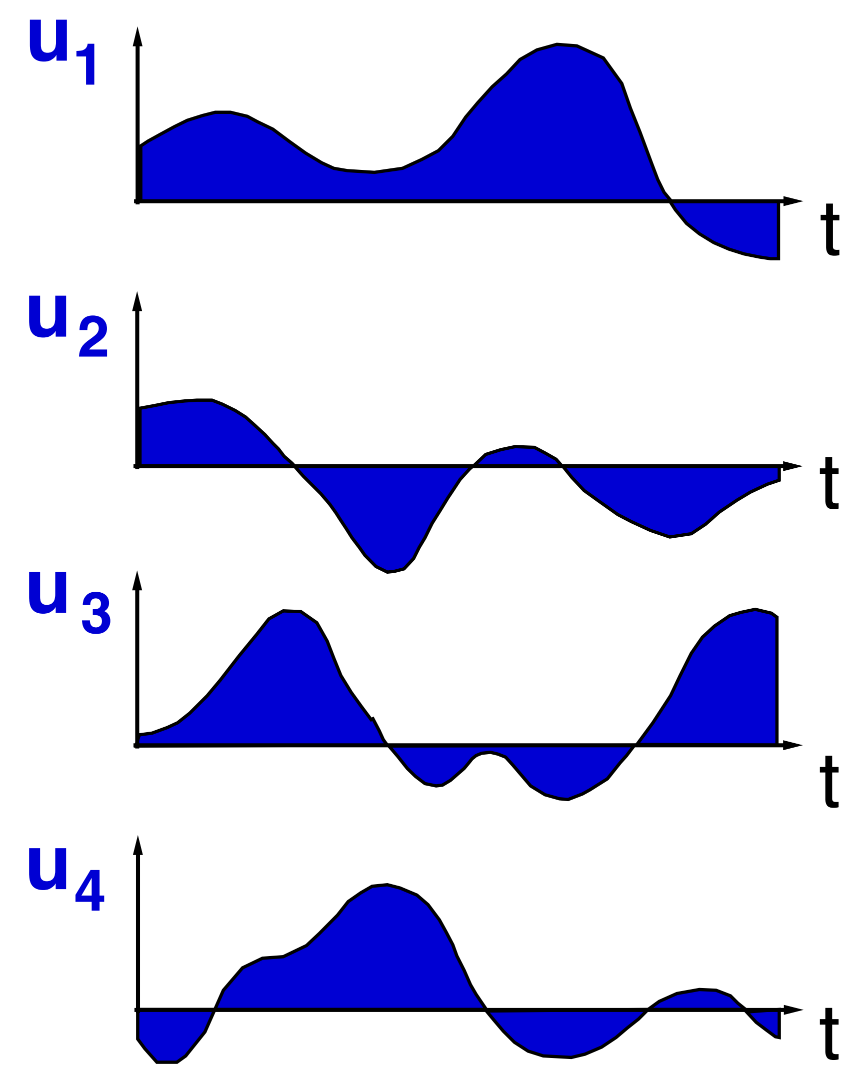
H_0



Control Parameters u_k (t)

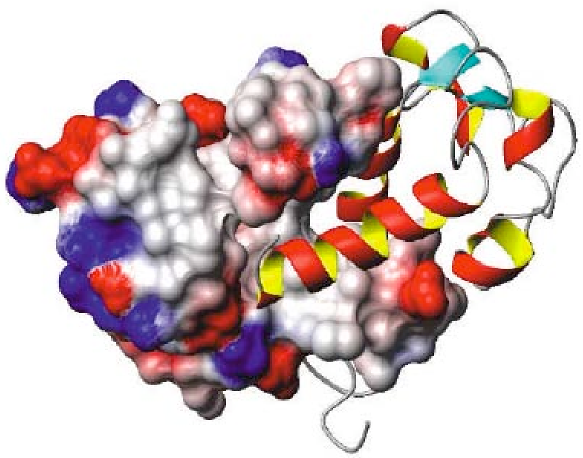


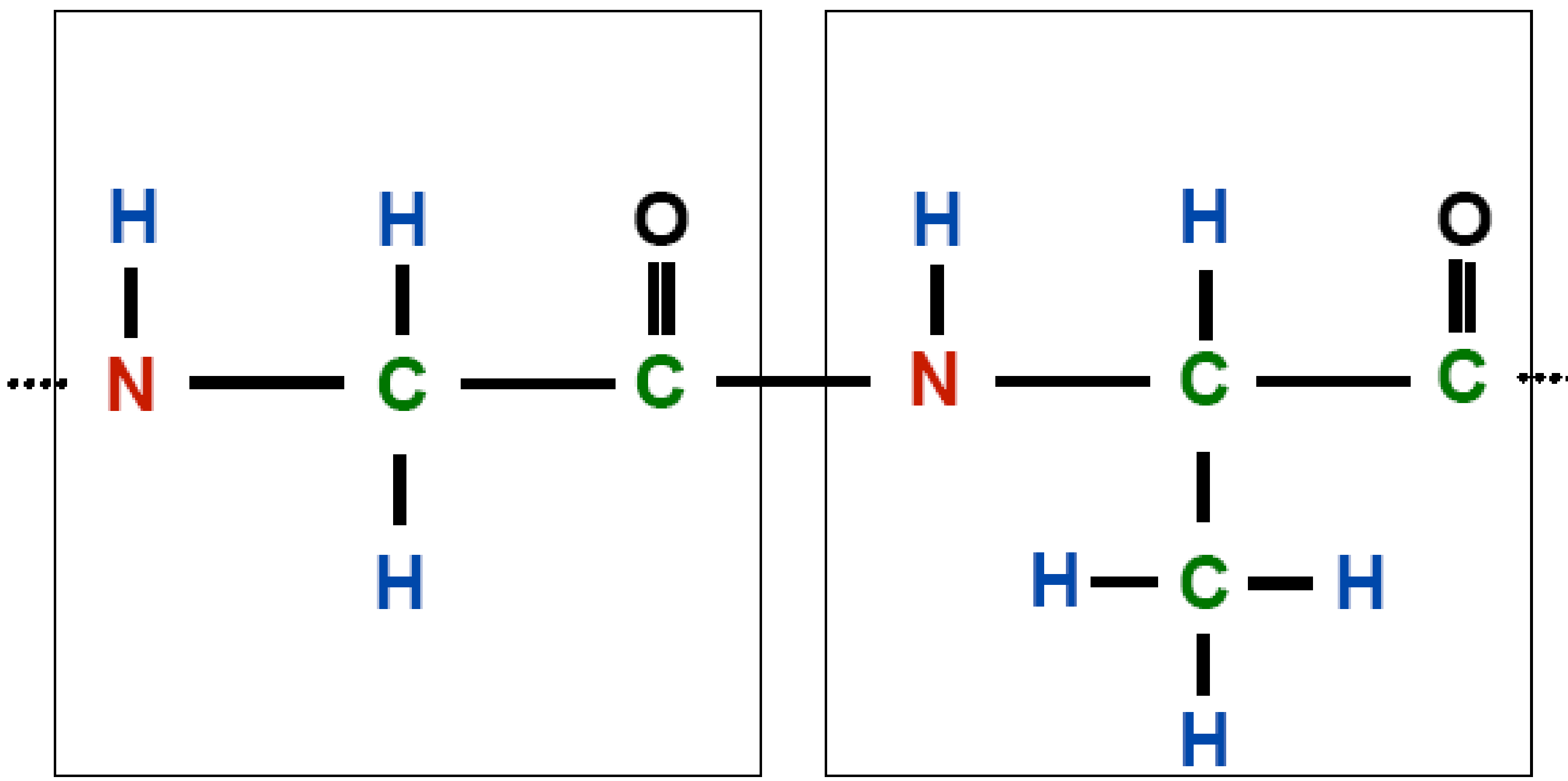
$$H_0 + \sum_k u_k(t)\, H_k$$

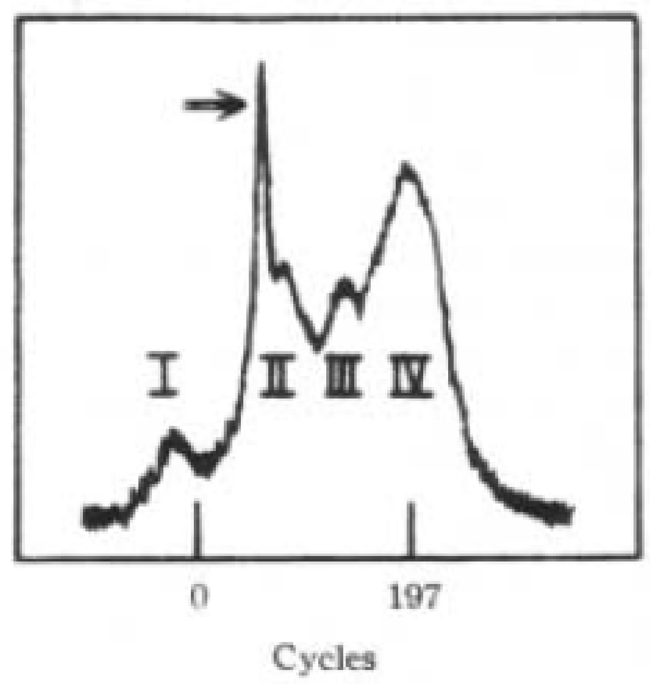


Resonance frequencies at 14 Tesla:

^{1}H	600 MHz
^{15}N	60 MHz
^{13}C	150 MHz



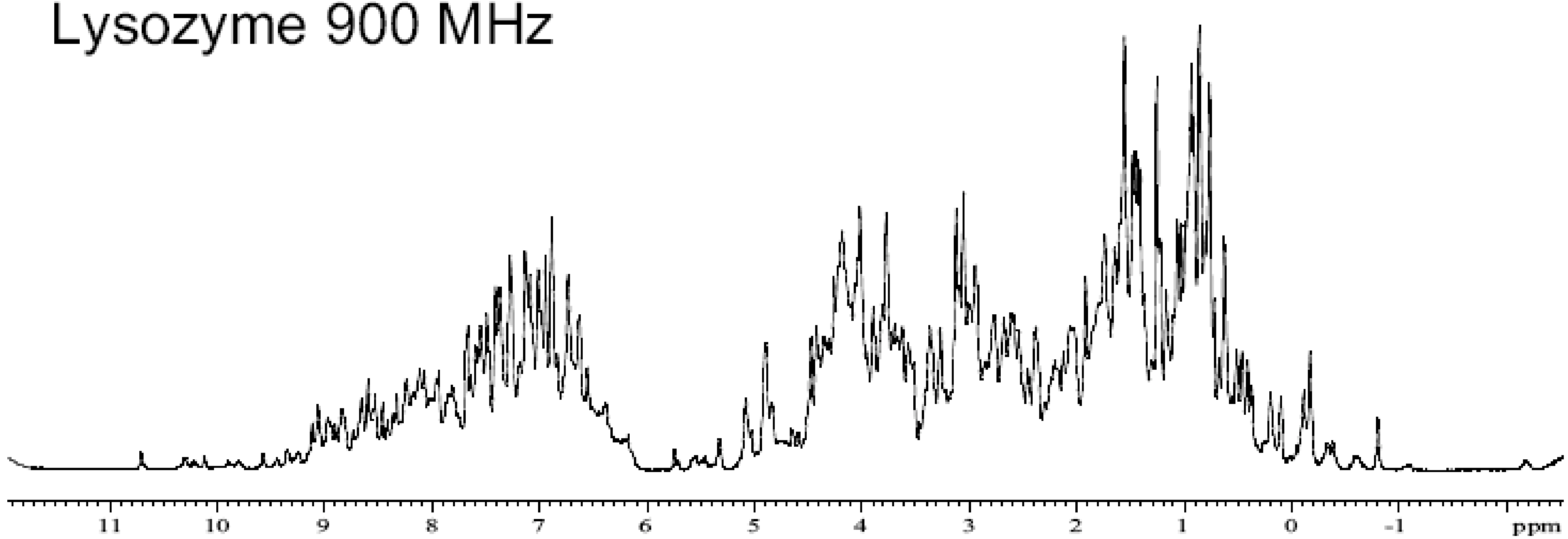




Ribonuclease
40 MHz

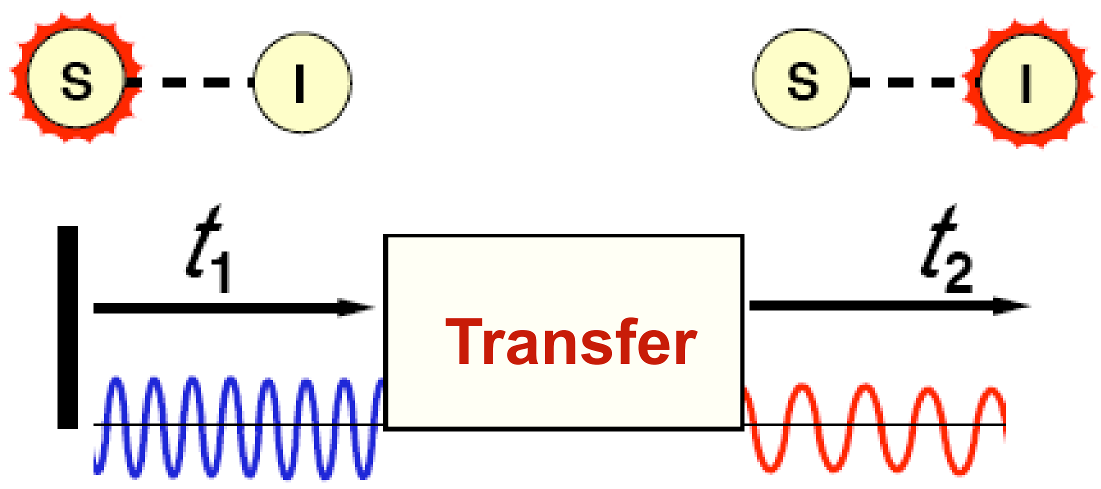
M. Saunders et al.
J.Amer.Chem.Soc. **1957**, *79*, 3289

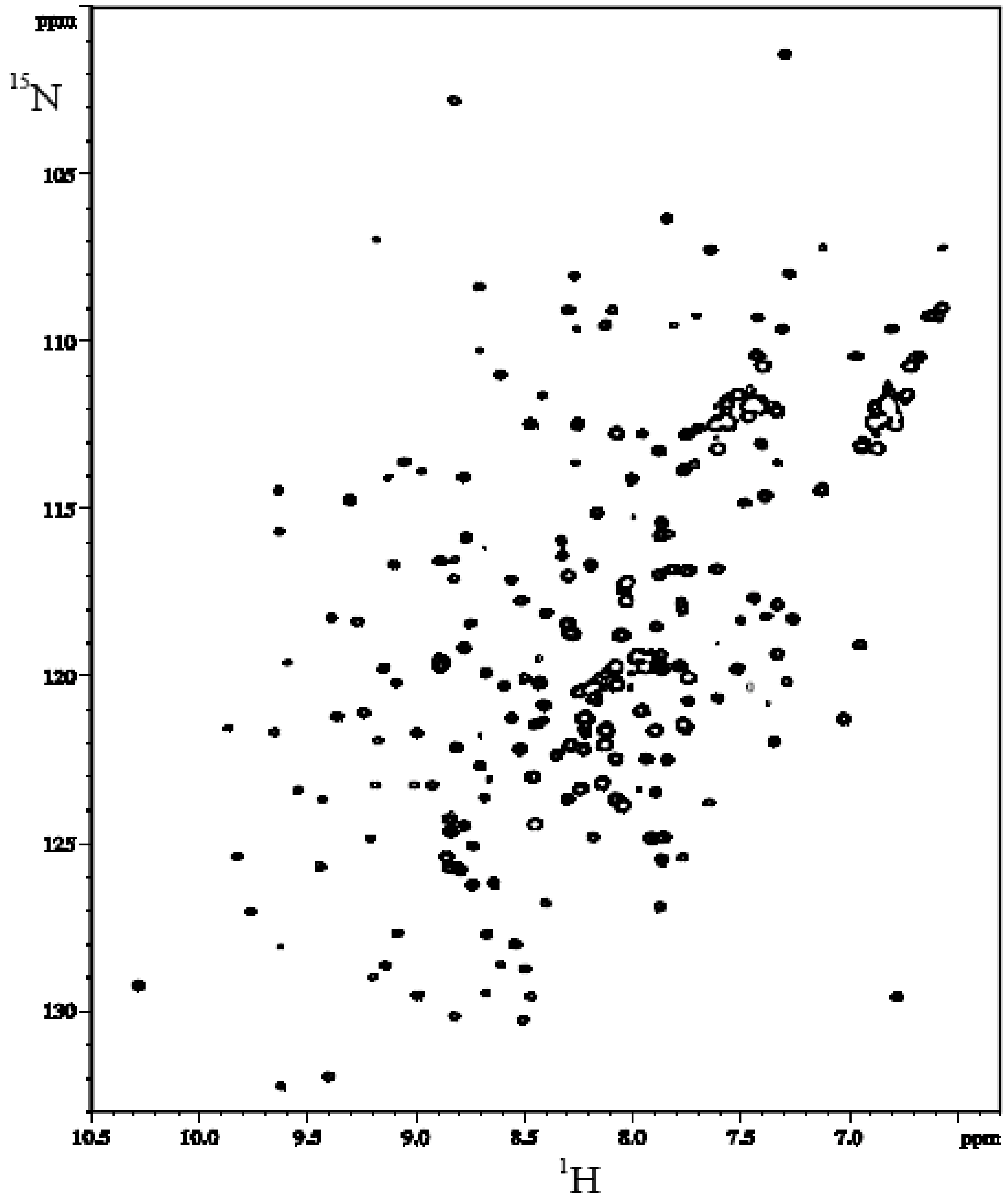
Lysozyme 900 MHz

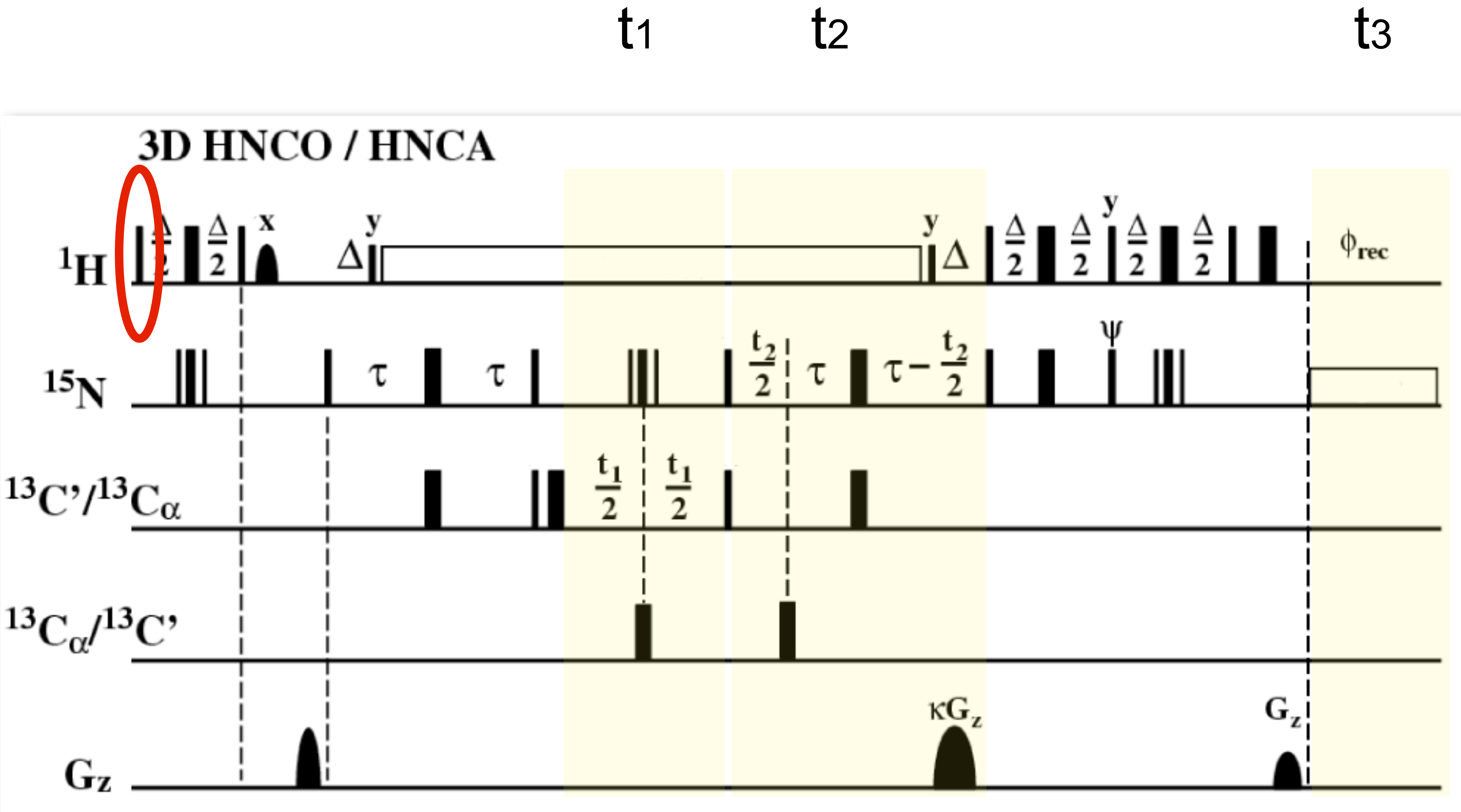


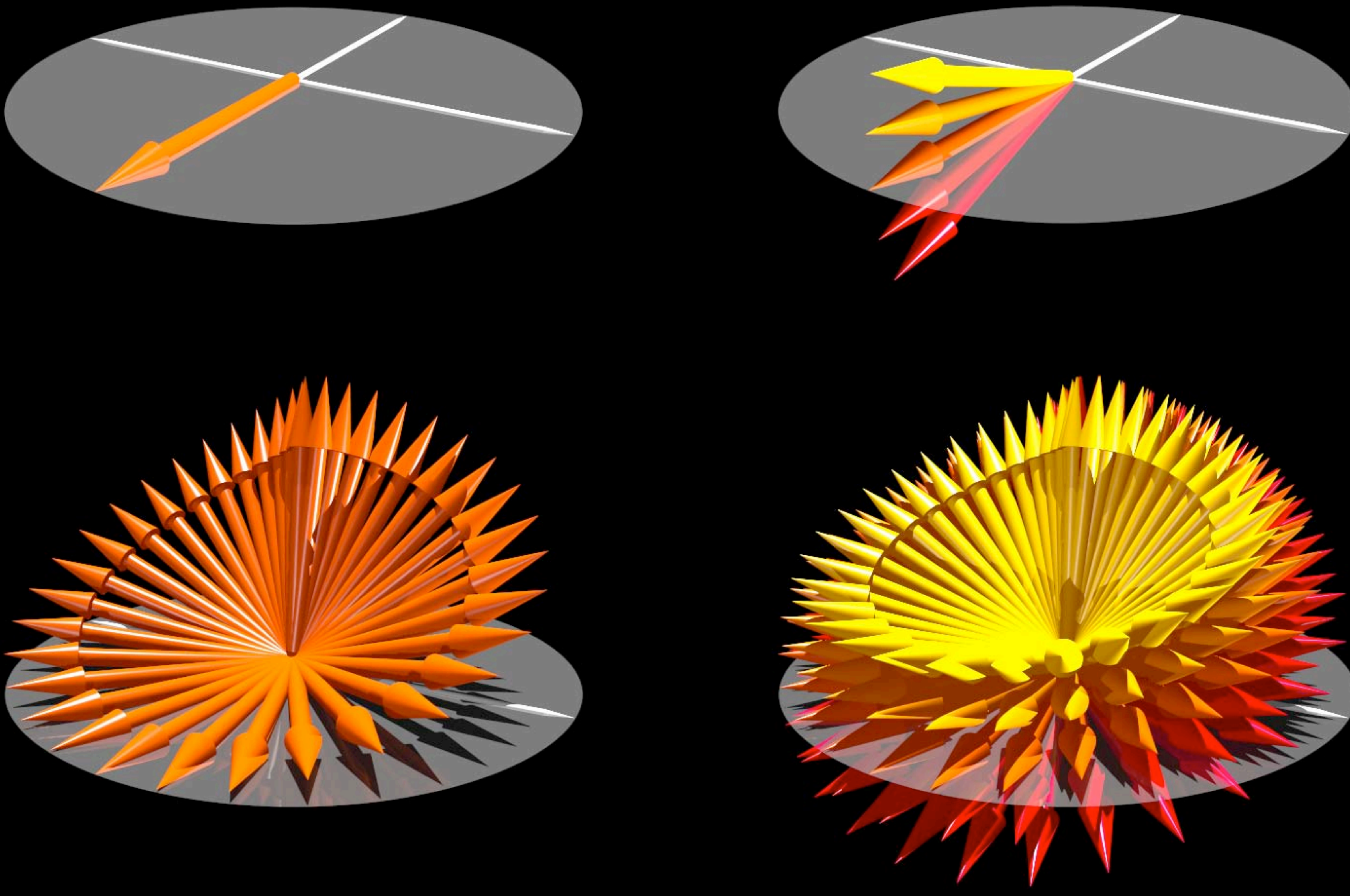
frequency dispersion: 10 kHz

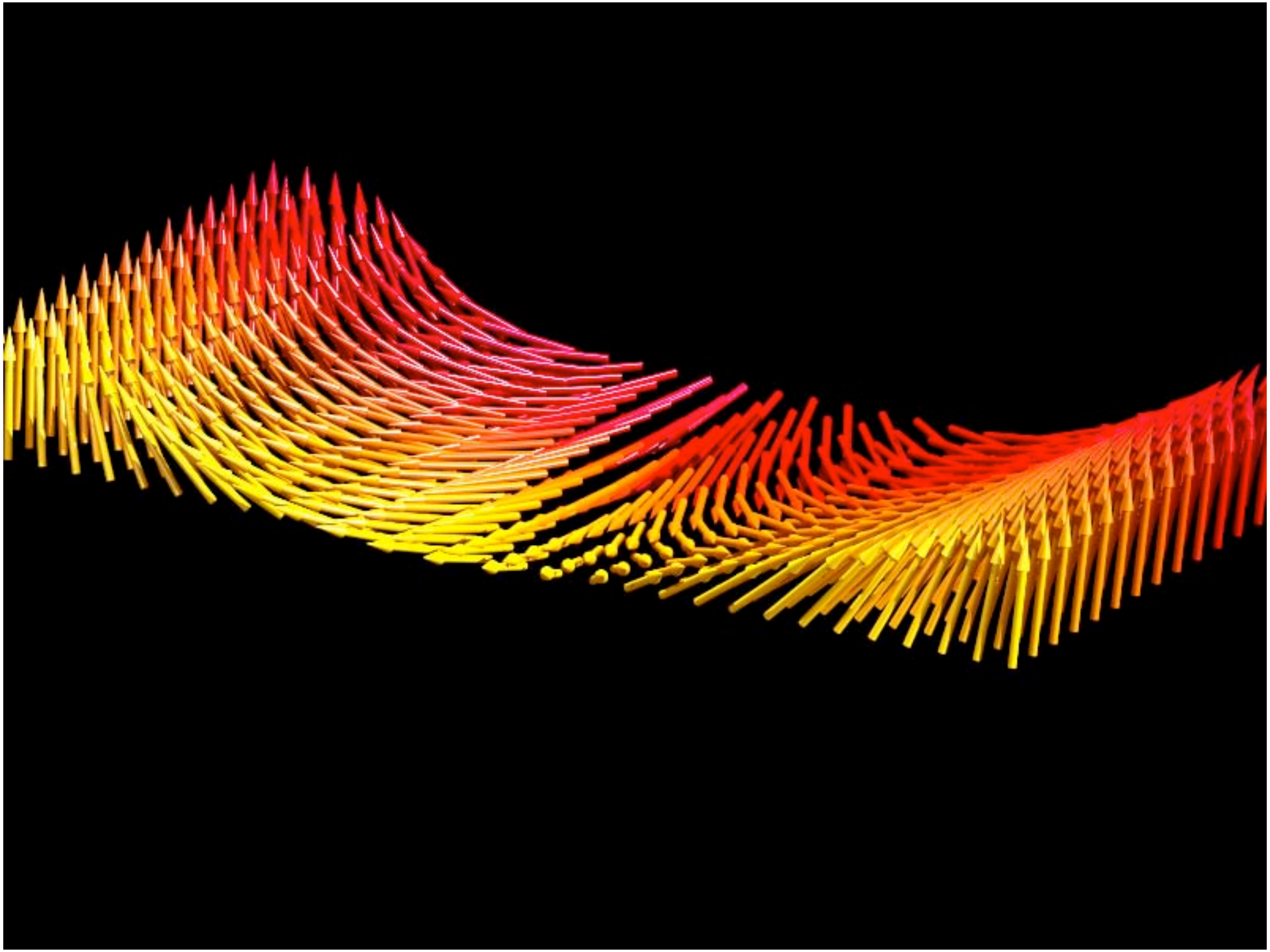
Two-dimensional NMR



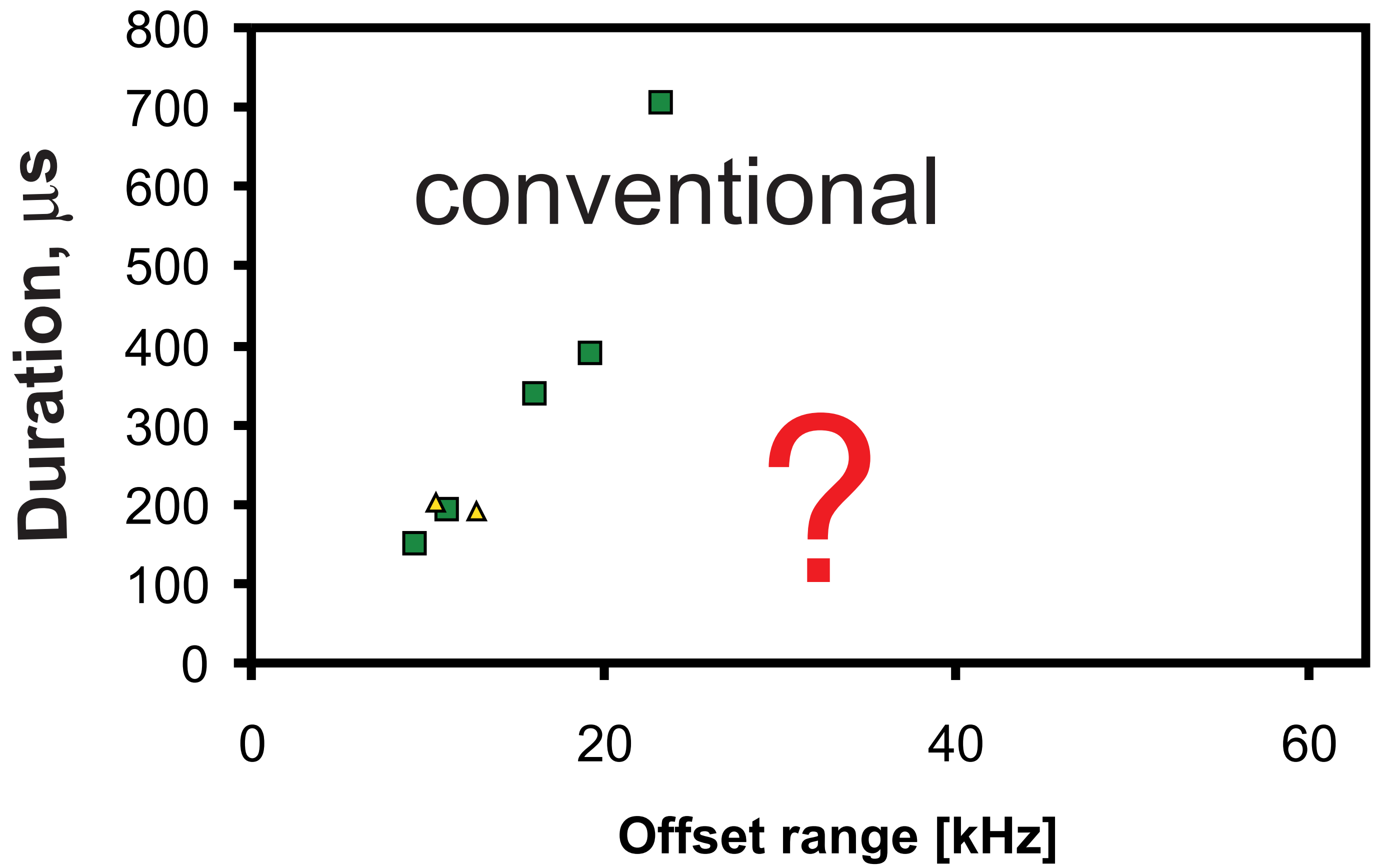



t1
t2
t3
3D HNCO / HNCA
1H
15N
13C'/13Cα
13Cα/13C'
Gz
κGz
Gz
φrec
ψ
τ
Δ
x
y

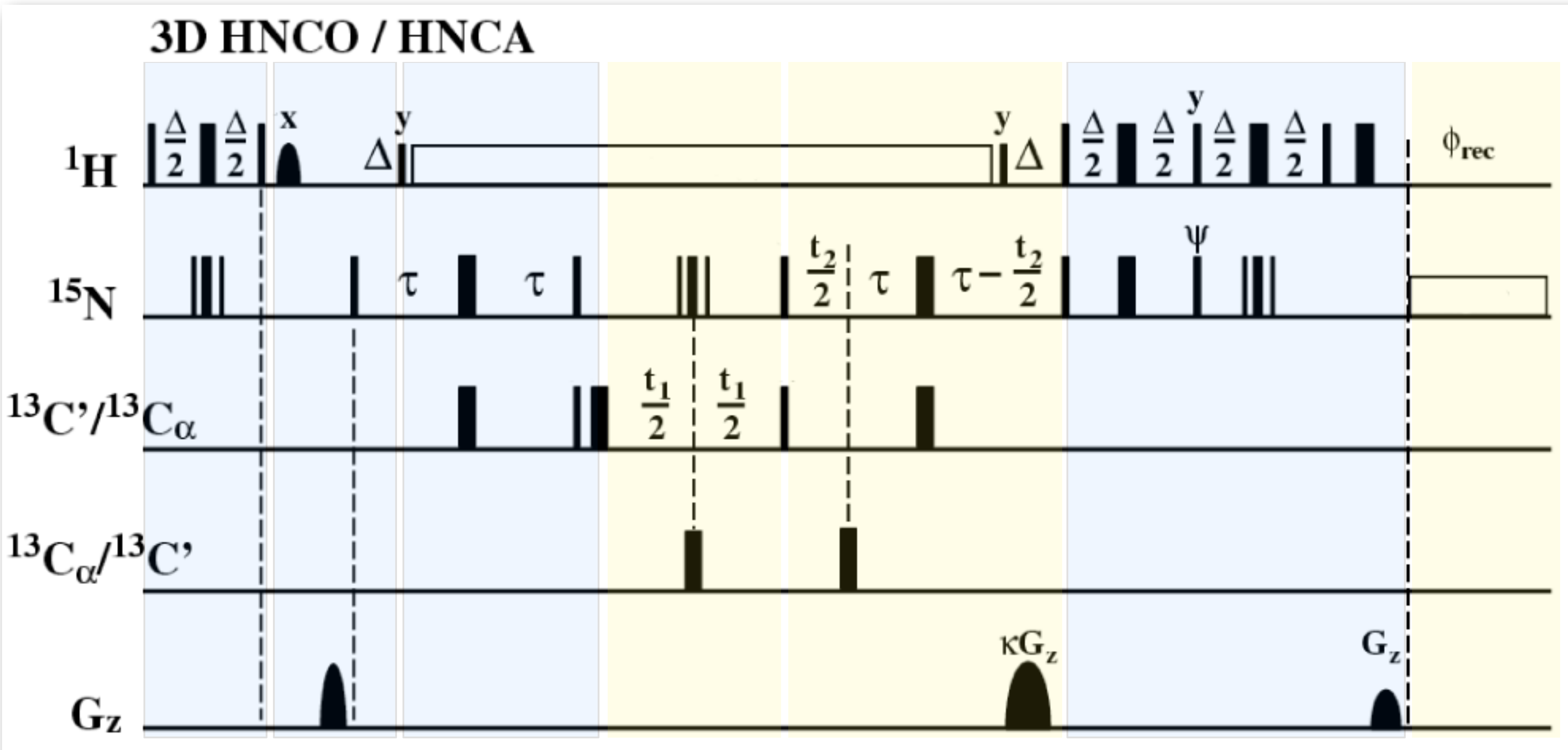




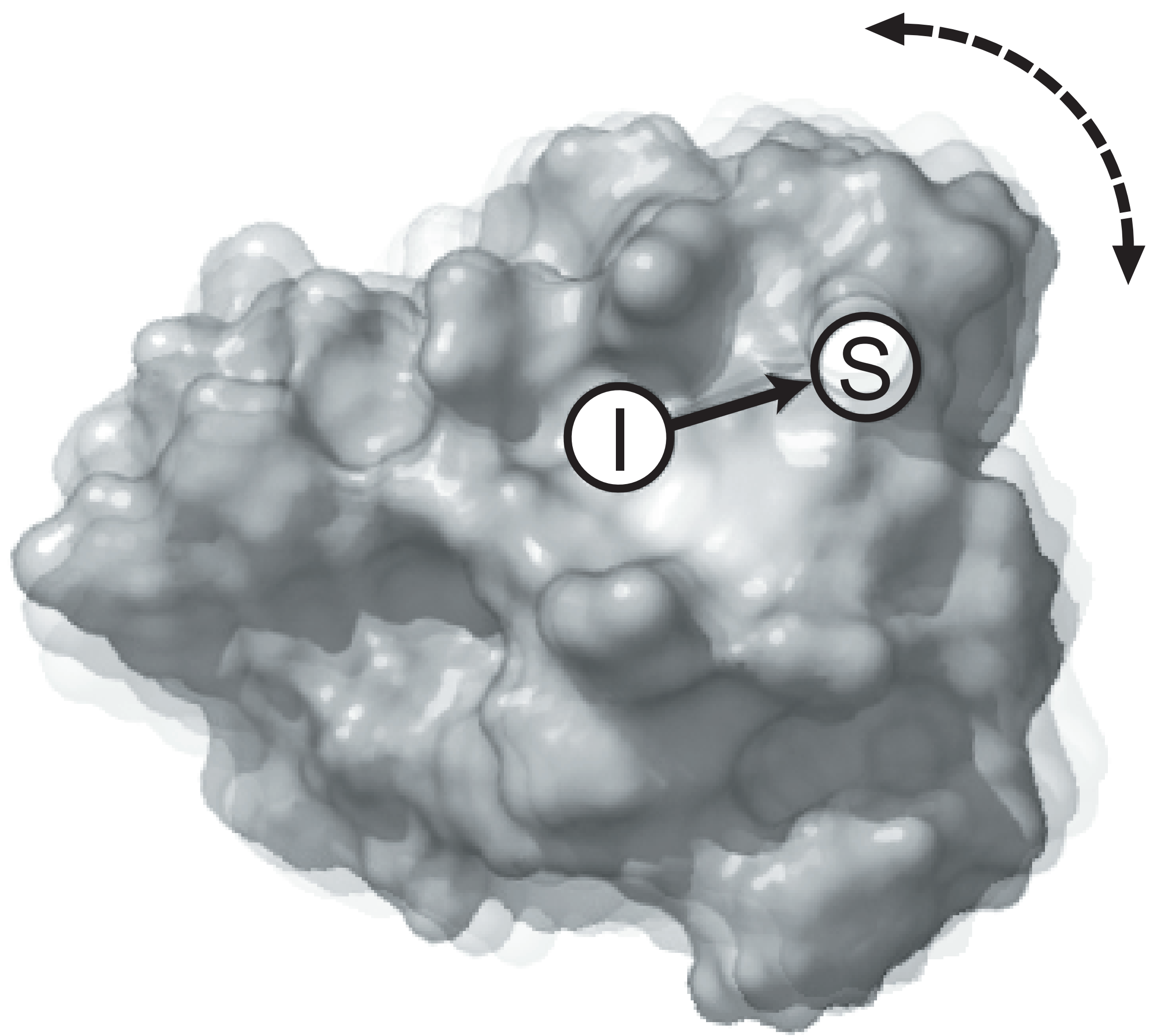
Performance of conventional composite pulses for broadband (robust) excitation



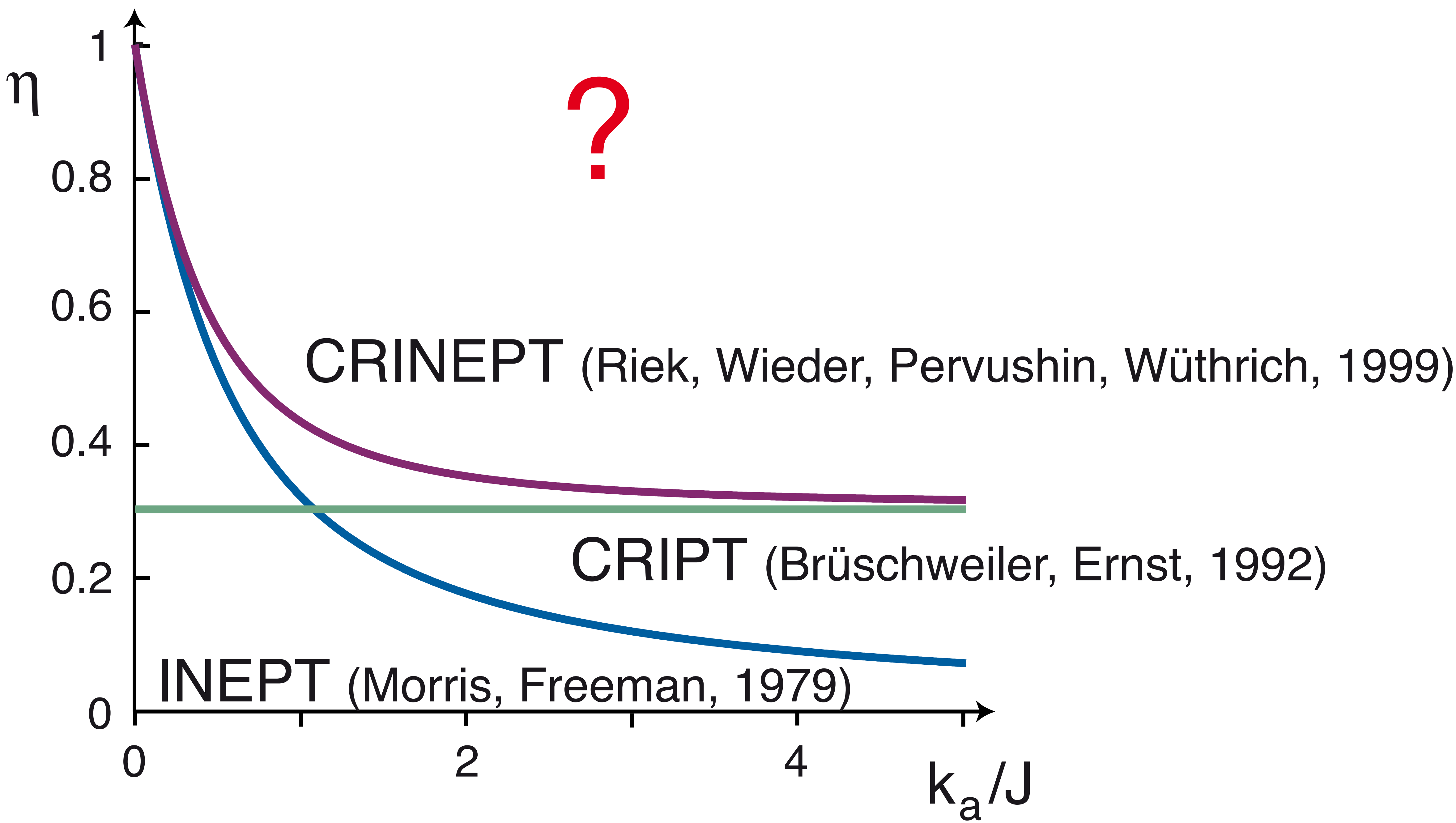
(excitation efficiency: 98%, max. rf amplitude: 10 kHz, no rf inhomogeneity) t1 t2 t3


3D HNCO / HNCA
1H
15N
13C'/13Cα
13Cα/13C'
Gz
x
y
Δ
Δ/2
τ
t1/2
t2/2
τ − t2/2
ψ
φrec
κGz
Gz

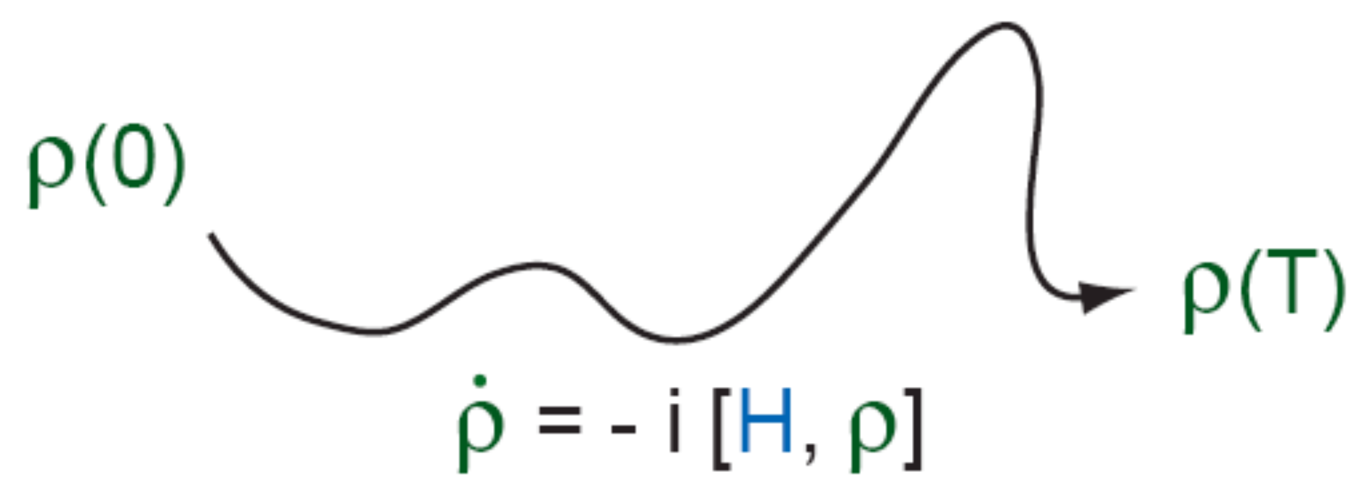
Relaxation rates k increase with molecular weight



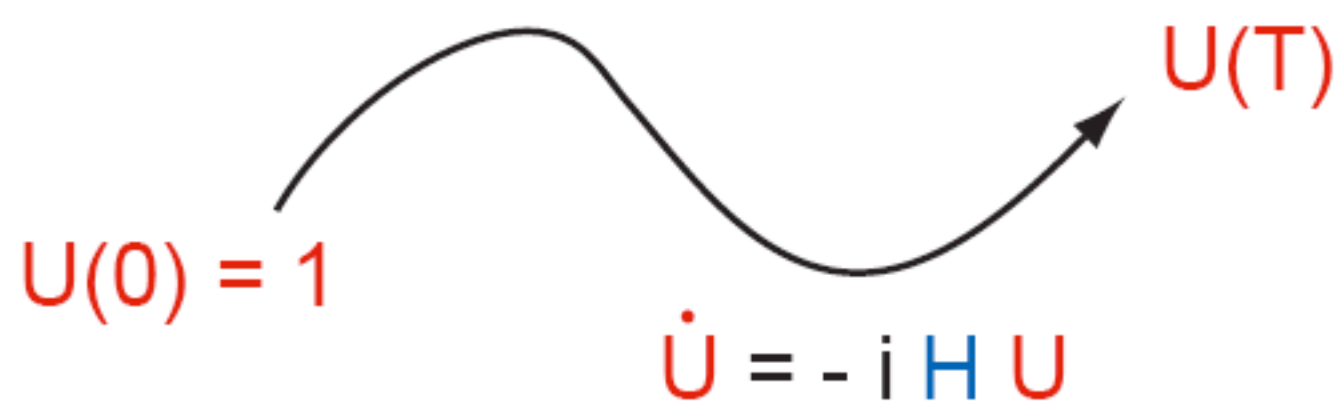
Transfer Efficiency $I_x \rightarrow 2I_z S_y$



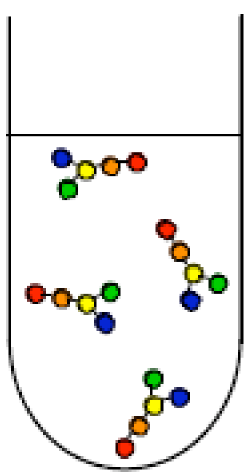
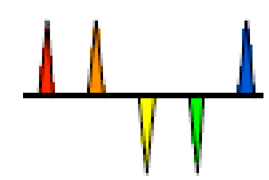
Transformation of the Density Operator



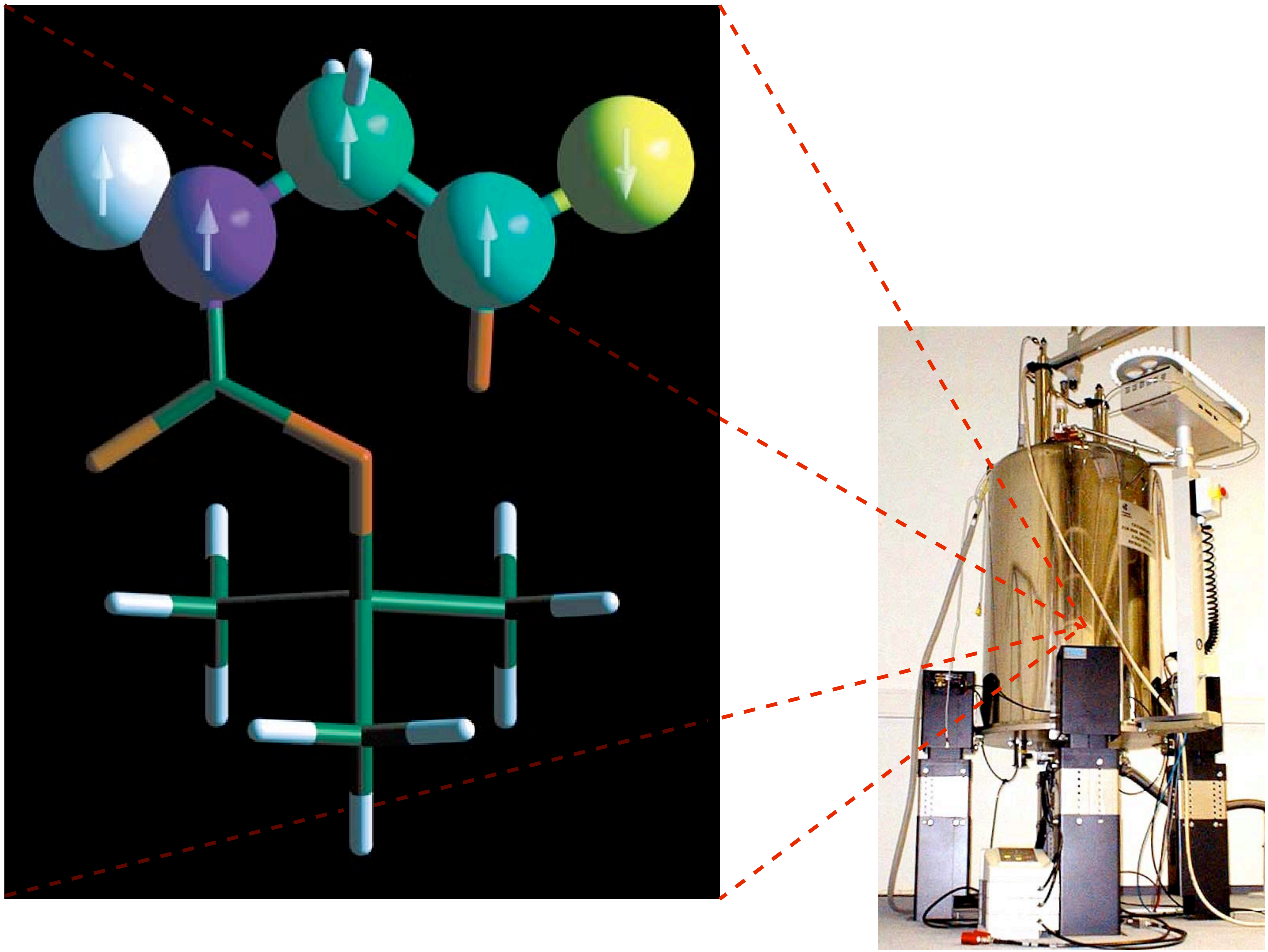
Generation of Unitary Operators



NMR Quantum Computing

INPUT	CALCULATION	OUTPUT
prepare initial state	**apply quantum-algorithm**	**detect NMR spectrum**
ρ_o	$\xrightarrow{U}$	ρ_f
		

Cory, Fahmy, Havel (1996)
Gershenfeld, Chuang (1997)



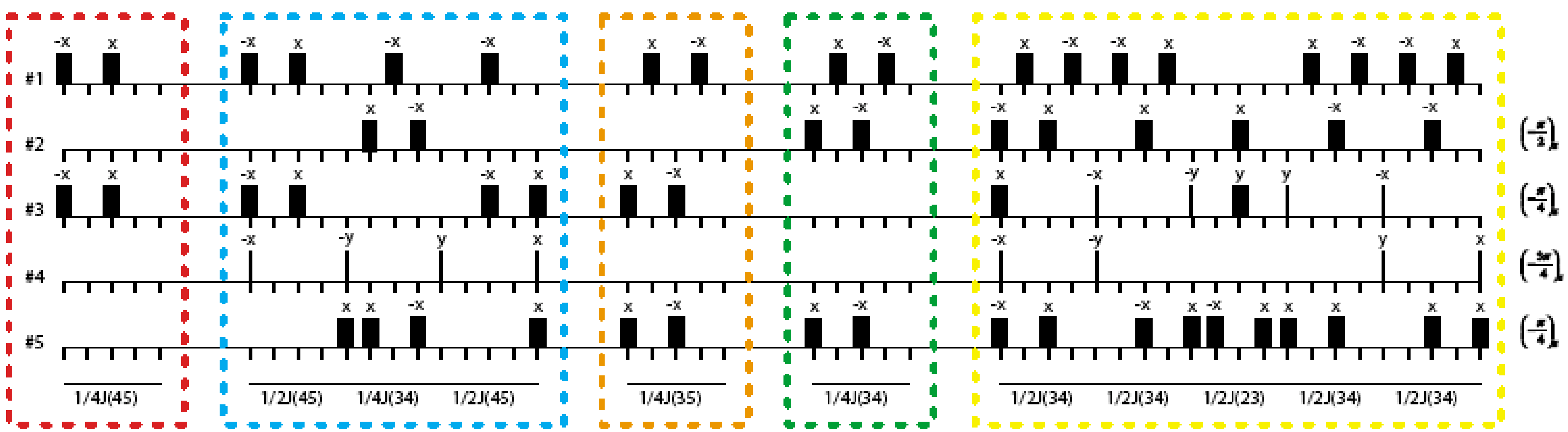
Implemented Test Functions for Thermal Deutsch Jozsa Algorithm

constant function: $f(\vec{x}) = 0$

$$f'(\vec{x}) = f(\vec{x}) \cdot \overline{x_4} = 0$$

balanced function: $f(\vec{x}) = x_2 \oplus x_3 \cdot x_5$

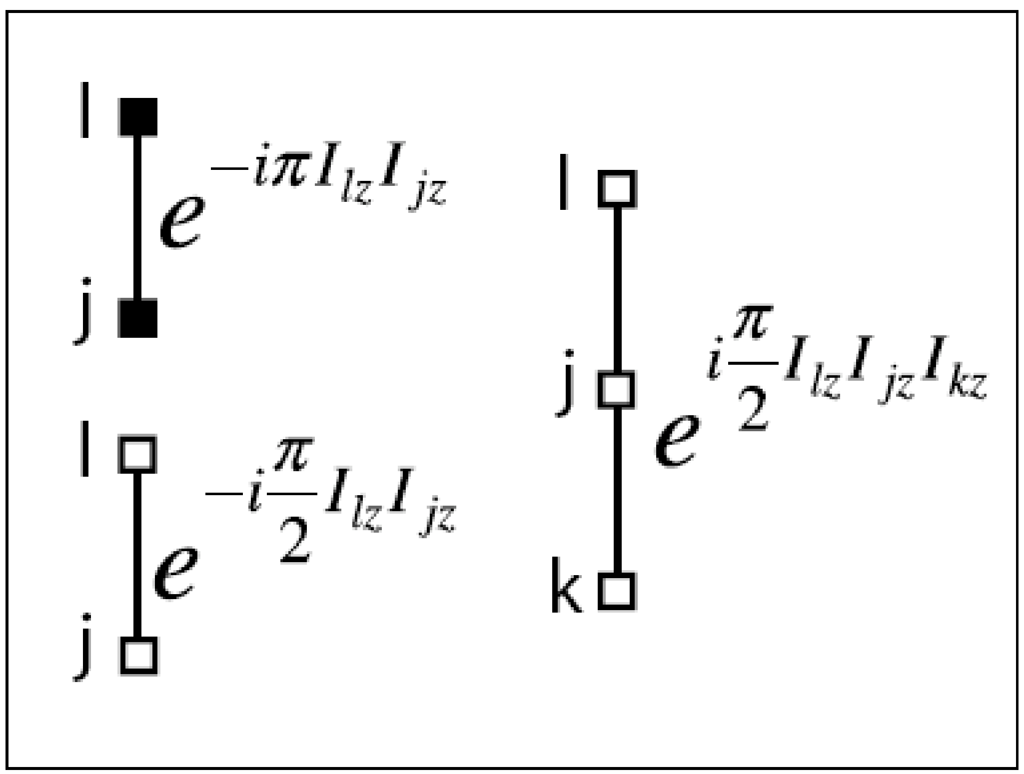
$$f'(\vec{x}) = f(\vec{x}) \cdot \overline{x_4} = x_2 \cdot \overline{x_4} \oplus x_3 \cdot \overline{x_4} \cdot x_5$$

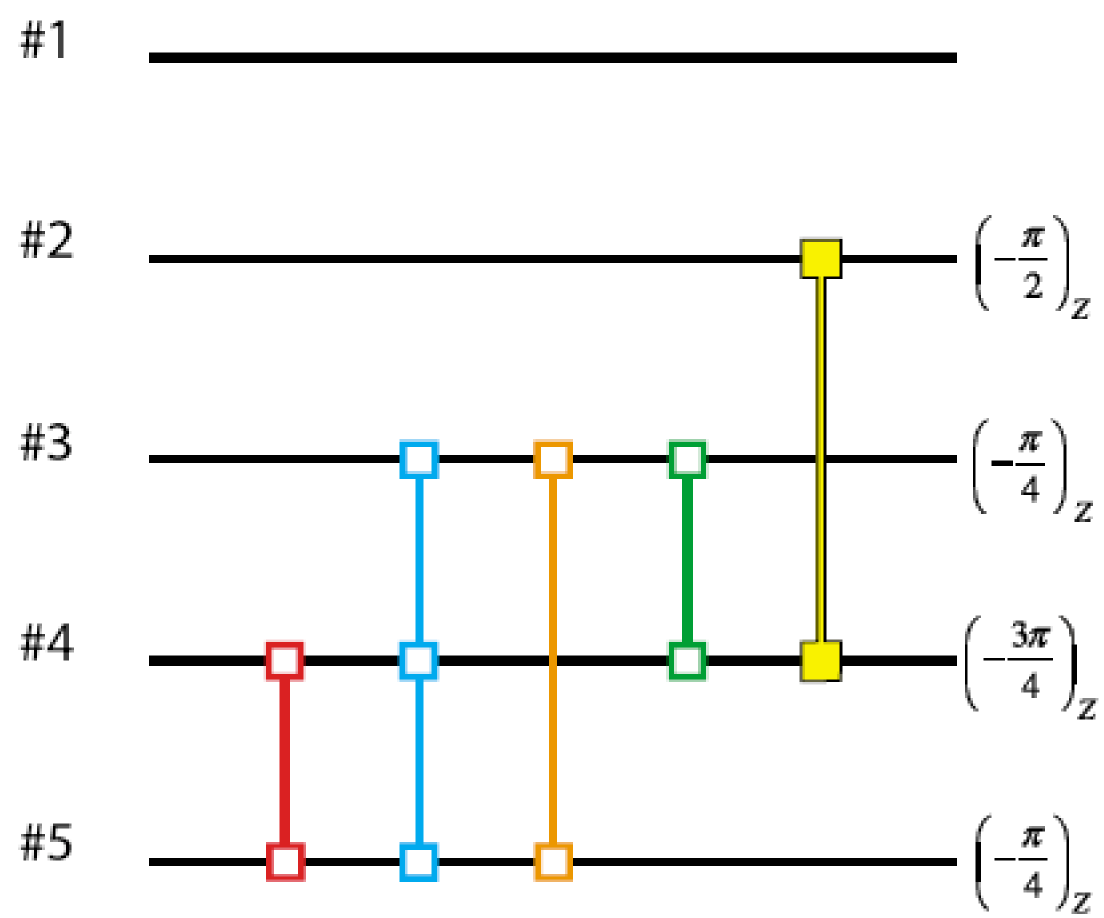


A. F. Fahmy, R. Marx, W. Bermel, S.J.G., Phys. Rev. A 78, 022317 (2008)

Scheme for the implementation of the function

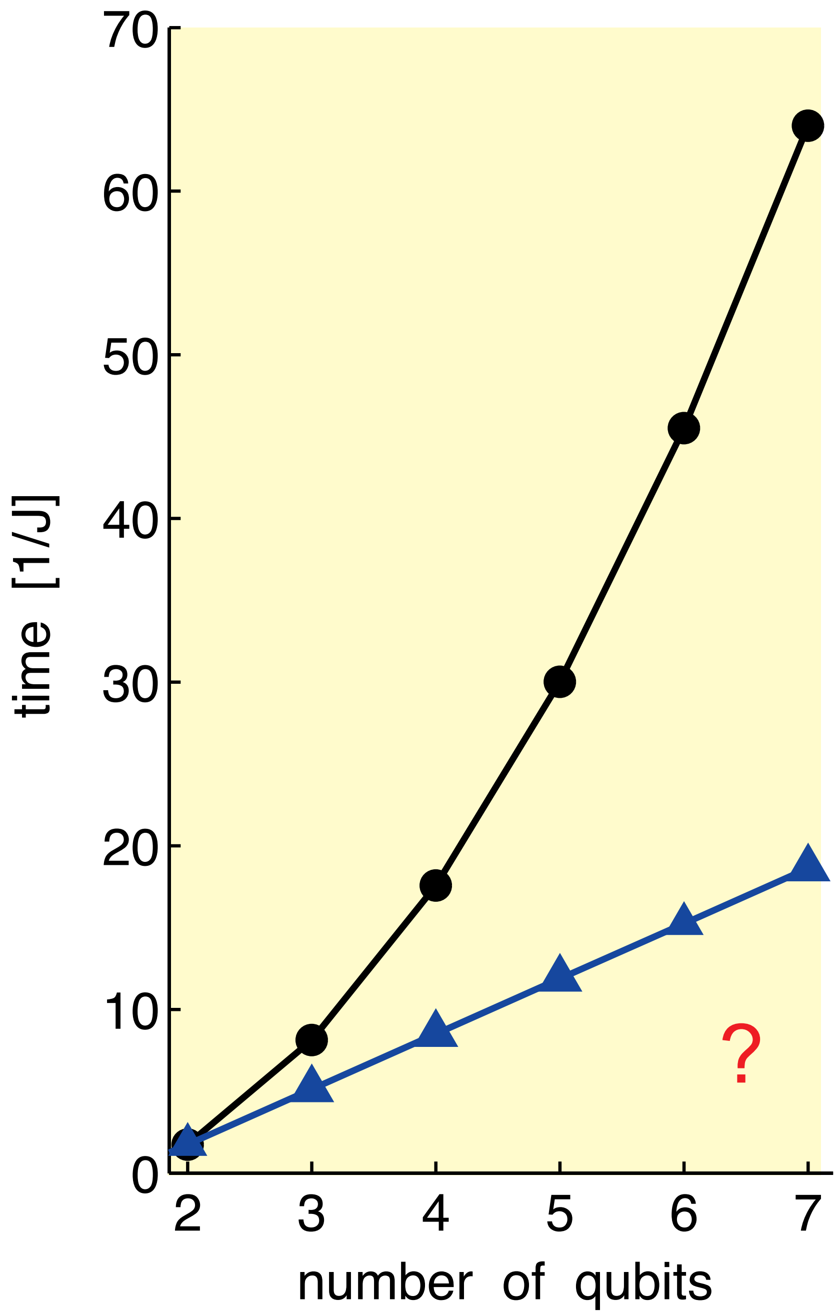
$$f'(\vec{x}) = f(\vec{x}) \cdot \overline{x_4} = x_2 \cdot \overline{x_4} \oplus x_3 \cdot \overline{x_4} \cdot x_5$$





A. F. Fahmy, R. Marx, W. Bermel, S.J.G., Phys. Rev. A 78, 022317 (2008)

Time-optimal implementation of the quantum Fourier transform ?



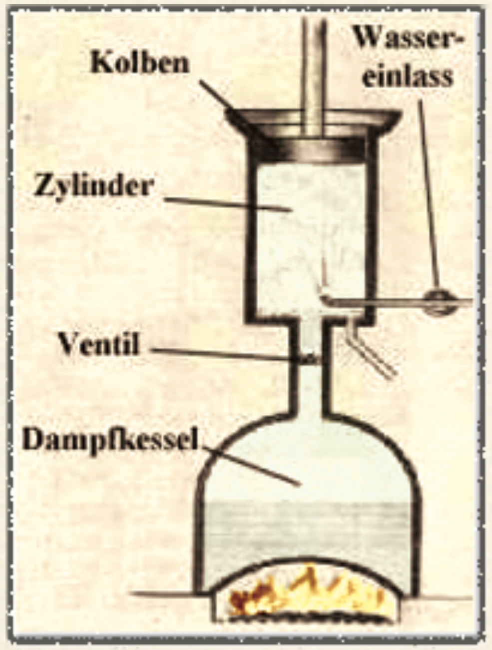
Saito et al. (2000)
quant-ph/0001113

Blais (2001)
PRA 64, 022312

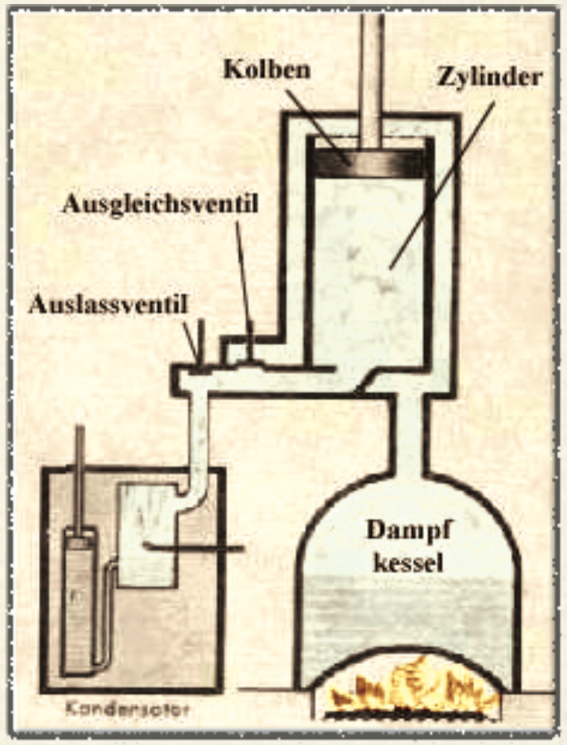
T. Schulte-Herbrüggen, A. Spörl, N. Khaneja, S.J.G., Phys. Rev. A 72, 042331 (2005)

Steam Engine

1697
D. Papin



1712
T. Newcomen



1765
J. Watt

?

Steam Engine

„The theory of its operation is rudimentary and attempts to improve its performance are still made in an almost haphazard way.“

1824

RÉFLEXIONS

SUR LA

PUISSANCE MOTRICE

DU FEU

ET

SUR LES MACHINES

PROPRES A DÉVELOPPER CETTE PUISSANCE.

PAR S. CARNOT,

ANCIEN ÉLÈVE DE L'ÉCOLE POLYTECHNIQUE.

A PARIS,

CHEZ BACHELIER, LIBRAIRE,

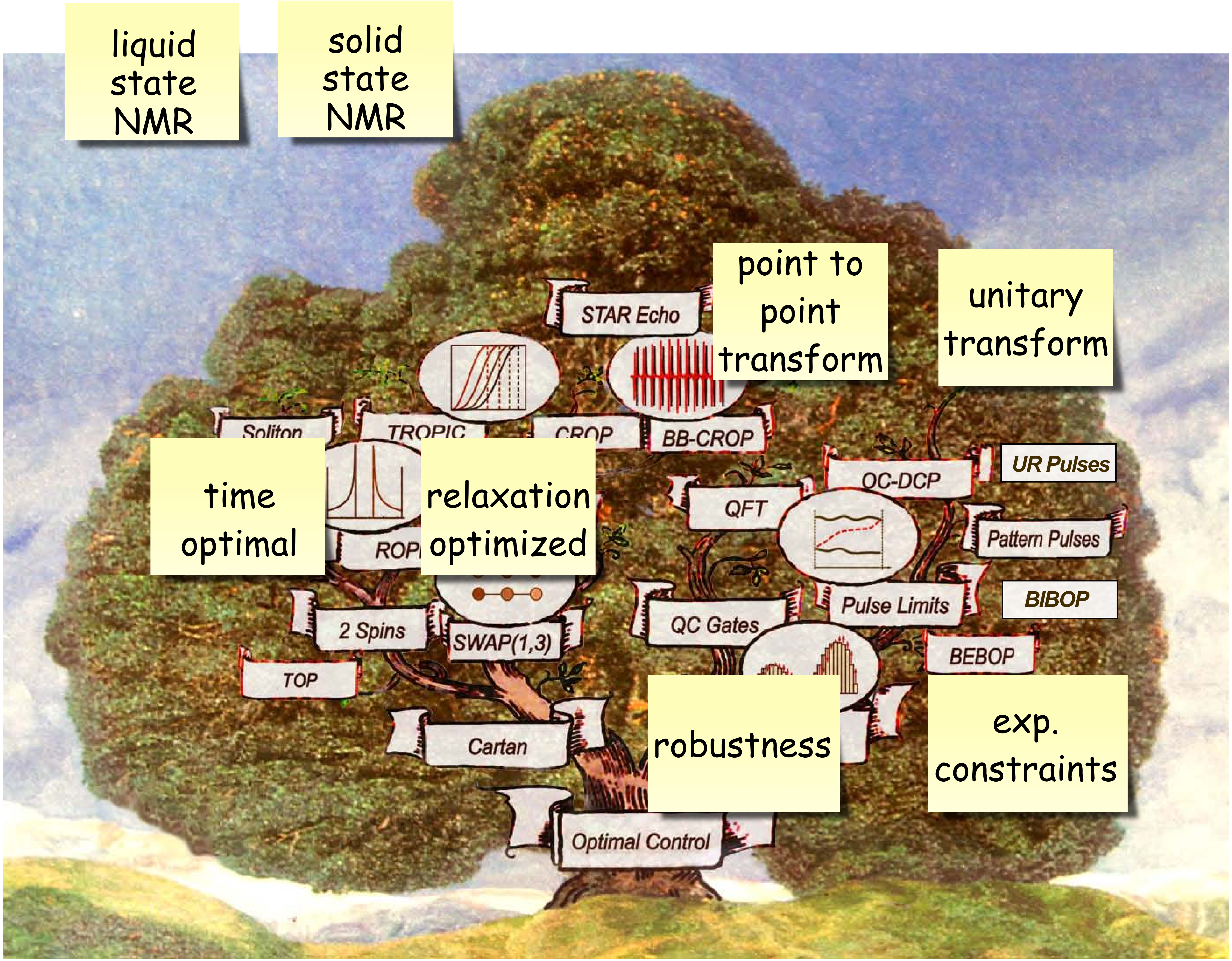
QUAI DES AUGUSTINS, N°. 55.

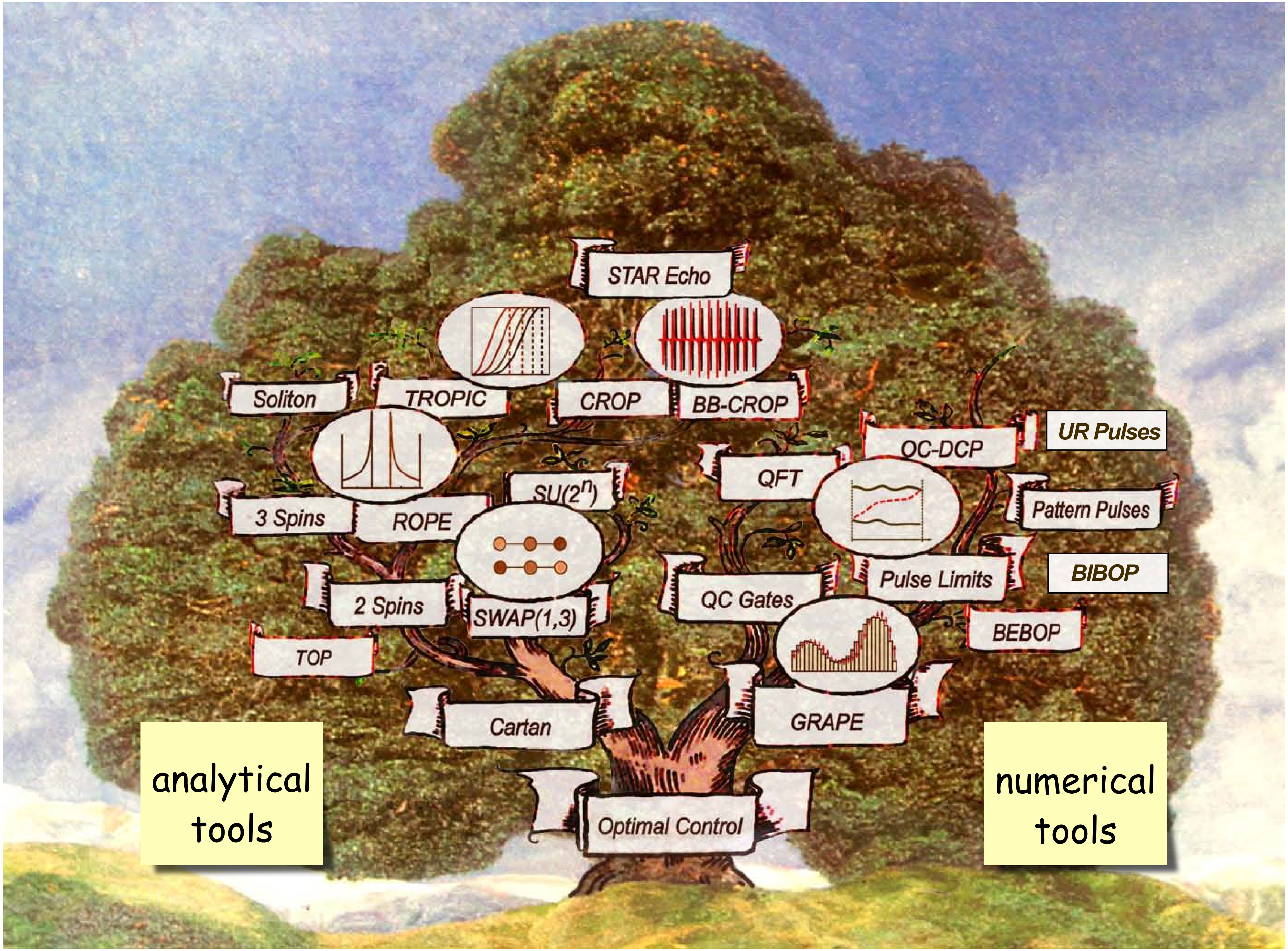
1824.

Optimal Control of Spin Systems

Optimal Control Theory

Spin Physics


liquid state NMR
solid state NMR
point to point transform
unitary transform
STAR Echo
Soliton
TROPIC
CROP
BB-CROP
time optimal
relaxation optimized
QFT
OC-DCP
UR Pulses
Pattern Pulses
2 Spins
SWAP(1,3)
QC Gates
Pulse Limits
BIBOP
BEBOP
TOP
Cartan
robustness
exp. constraints
Optimal Control


STAR Echo
Soliton
TROPIC
CROP
BB-CROP
UR Pulses
OC-DCP
QFT
SU(2^n)
3 Spins
ROPE
Pattern Pulses
Pulse Limits
BIBOP
2 Spins
SWAP(1,3)
QC Gates
BEBOP
TOP
Cartan
GRAPE
analytical tools
numerical tools
Optimal Control

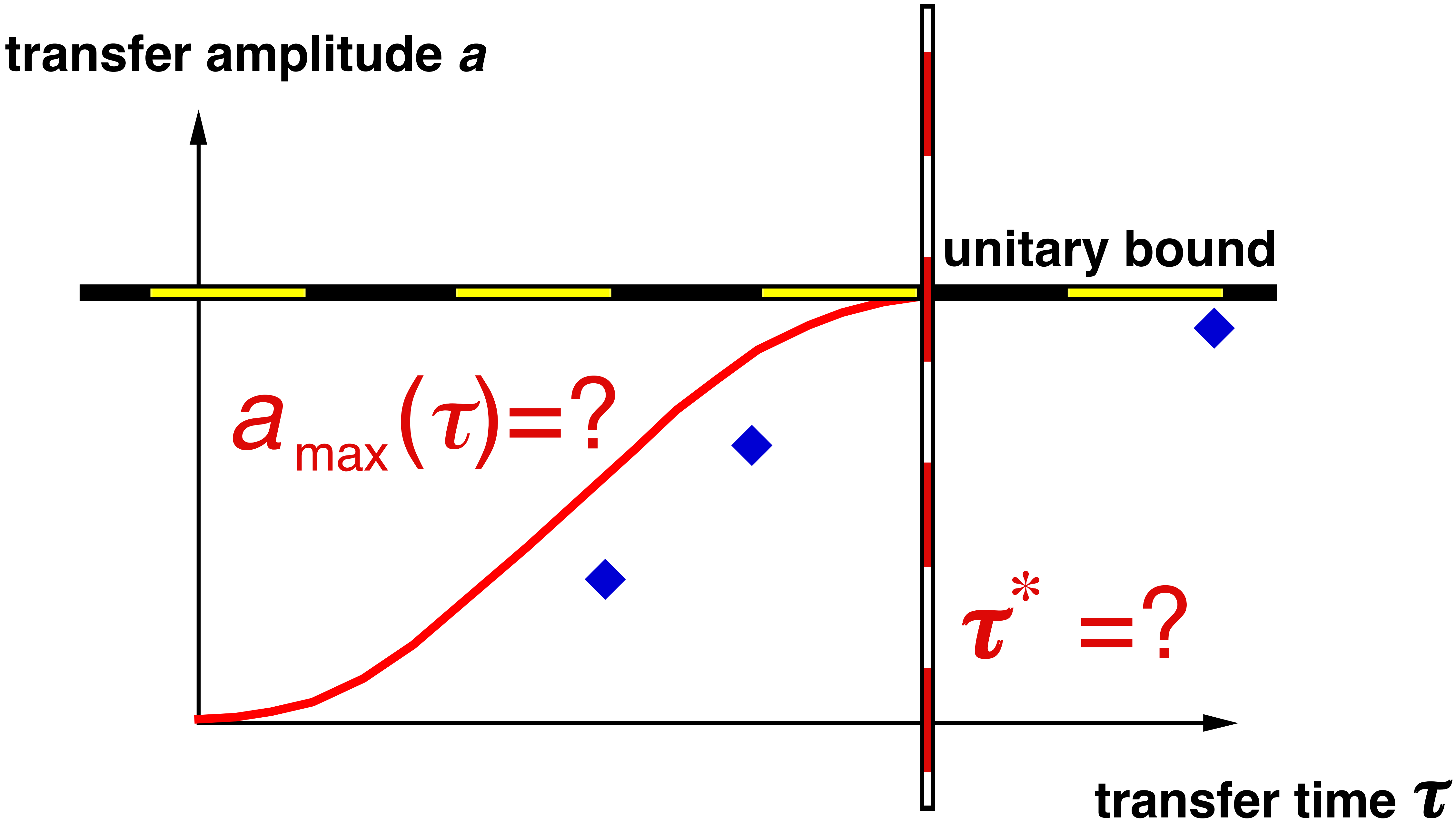
Optimal control in NMR:
band-selective excitation and inversion

S. Conolly, D. Nishimura, A. Macovski, Optimal control solutions to the magnetic resonance selective excitation problem, IEEE Trans. Med. Imaging MI-5 (1986) 106–115.

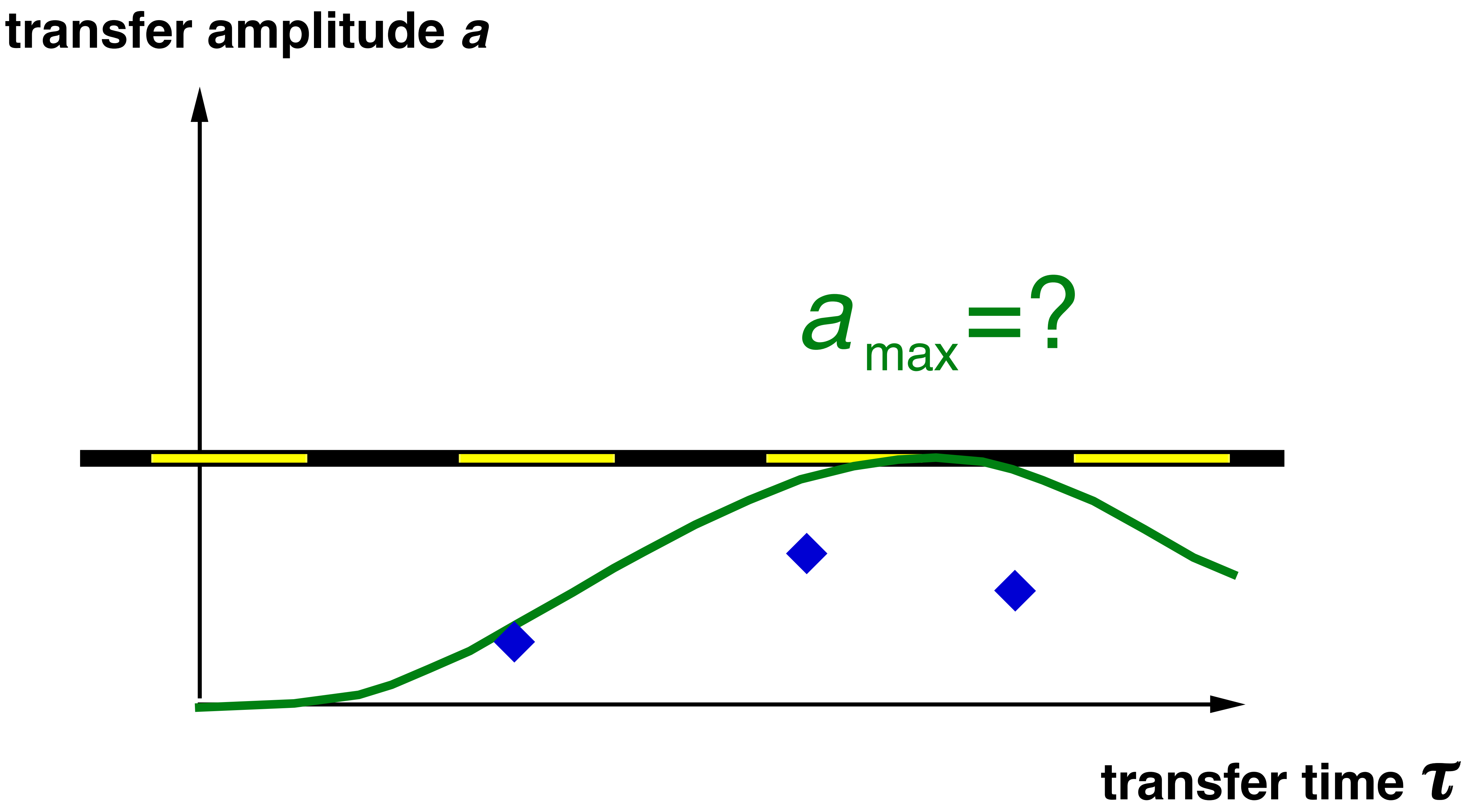
J. Mao, T.H. Mareci, K.N. Scott, E.R. Andrew, Selective inversion radiofrequency pulses by optimal control, J. Magn. Reson. 70 (1986) 310–318.

D. Rosenfeld, Y. Zur, Design of adiabatic selective pulses using optimal control theory, Magn. Reson. Med. 36 (1996) 401–409.

Unitary Quantum Evolution (no Relaxation)



Quantum Evolution in Presence of Relaxation



Time-Optimal Control of Two-Spin Systems

Strong-Pulse Limit: $H_{rf} >> H_c$ (2 time scales)

Cartan Decomposition

Characterization of ALL unitary operators that can be created in time T

Derivation of
- time-optimal transfer function (TOP curve)
- minimum time for maximum transfer
- pulse sequence

Khaneja, Brockett, Glaser (2001)

Khaneja, Kramer, Glaser (2005)

Maximum transfer efficiency $\eta^*(t)$ and minimum time $t_{\min}$ for complete transfer

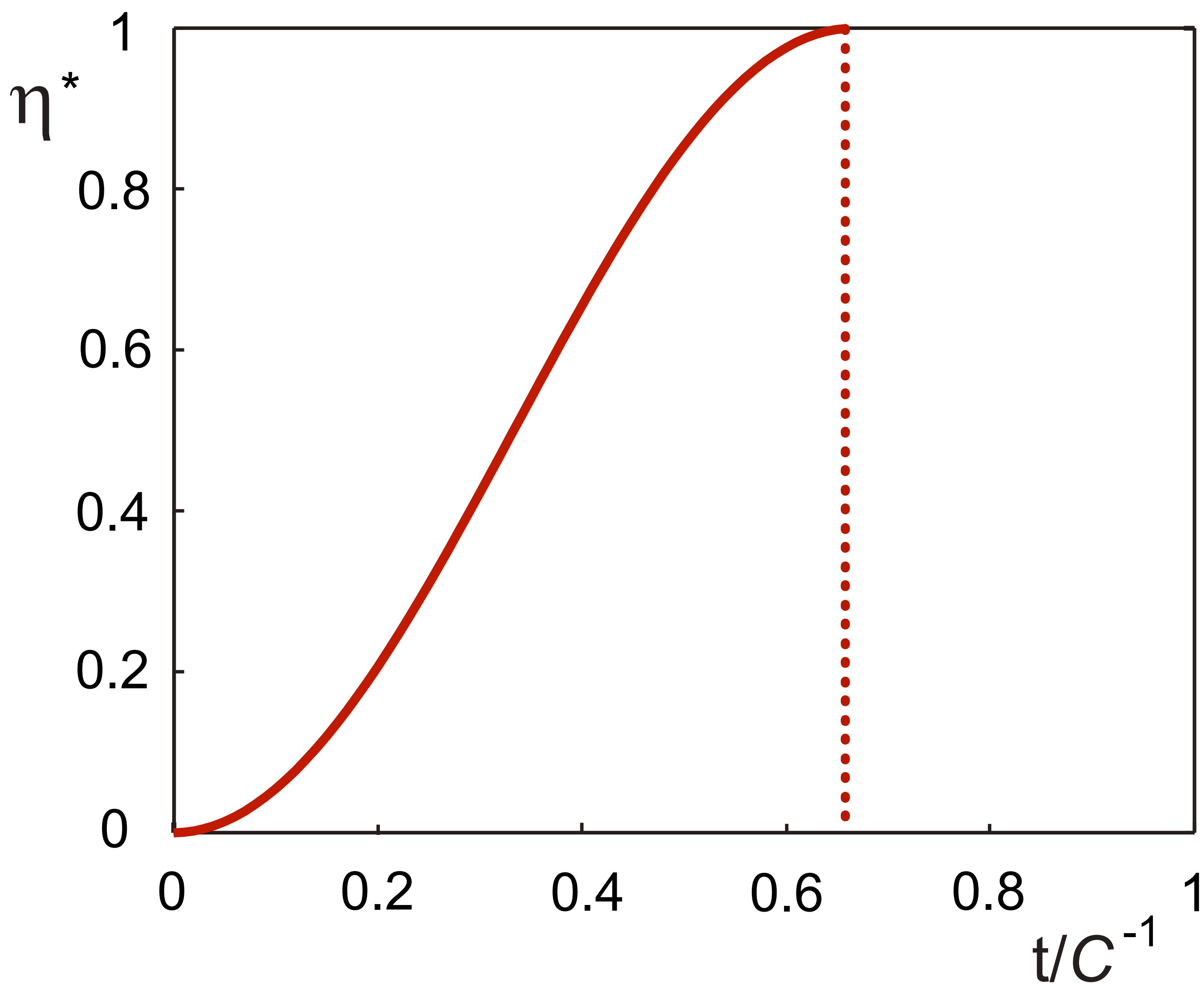
Transfer	$\eta^*(t)$	$t_{\min}^{-1}$
$I_x \to S_x$	$\sin^2(\frac{\pi}{2}C(\|\mu_3\| + \|\mu_2\|)t)$	$C(\|\mu_3\| + \|\mu_2\|)$
$I^- \to S^-$	$\sin(\pi C a)\sin(\pi C b)$	$\frac{2}{3}C(\|\mu_3\| + \|\mu_2\| + \|\mu_1\|)$
$I_x \to 2I_zS_x$	$\sin(\pi C\|\mu_3\|t)$	$2C\|\mu_3\|$
$I^- \to 2I_zS^-$	$\max_x \ \sin(\frac{\pi}{2}C\{\|\mu_3\| + \|\mu_2\| - \|\mu_1\| + x\}t)\cos(\pi Ctx)$	$C(\|\mu_3\| + \|\mu_2\| - \|\mu_1\|)$
$I_xS_\beta \to I_\beta S_x$	$\sin(\frac{\pi}{2}C(\|\mu_3\| + \|\mu_2\|)t)$	$C(\|\mu_3\| + \|\mu_2\|)$
$I^-S_\beta \to I_\beta S^-$	$\sin(\frac{\pi}{2}C(\|\mu_3\| + \|\mu_2\|)t)$	$C(\|\mu_3\| + \|\mu_2\|)$

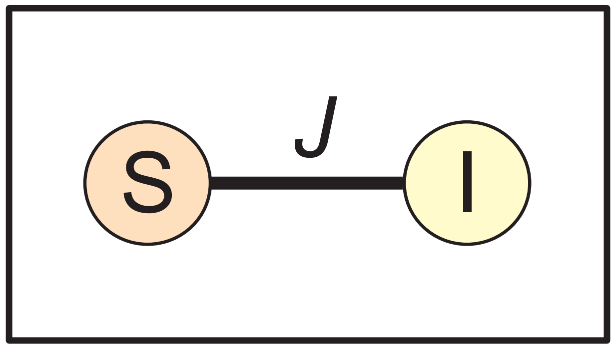
Note: $I^- = I_x - \mathrm{i}I_y$ and $I_\beta = \frac{\mathbf{1}}{2} - I_z$. For the transfer $I^- \to S^-$, the optimal values of a and b are completely characterized by the two conditions $a + 2b = (|\mu_3| + |\mu_2| + |\mu_1|)\ t$ and $\tan(\pi C a) = 2\ \tan(\pi C b)$.

Khaneja, Kramer, Glaser (2005)

TOP (time-optimal pulse) curves for dipolar coupling

$$(\mu_1, \mu_2, \mu_3) = (-1/2, -1/2, 1)$$

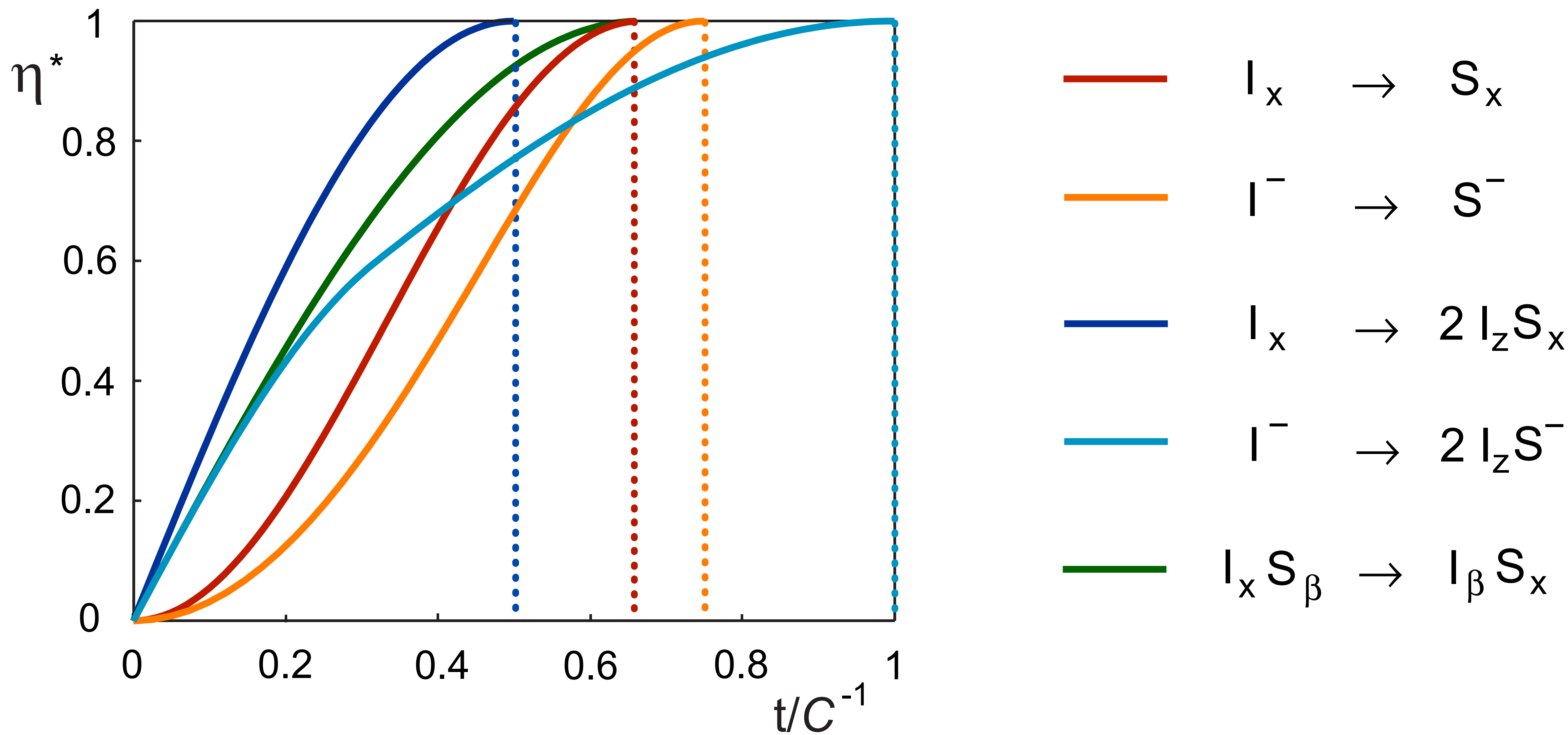




Khaneja, Kramer, Glaser (2004)

TOP (time-optimal pulse) curves for dipolar coupling

$$(\mu_1, \mu_2, \mu_3) = (-1/2, -1/2, 1)$$



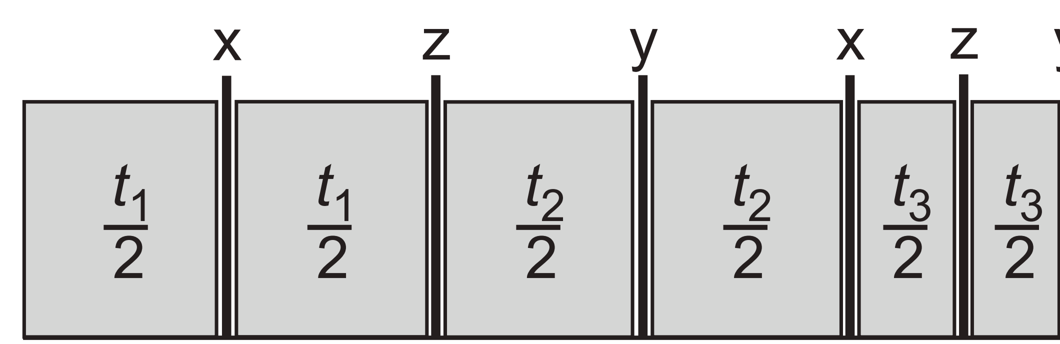
Khaneja, Kramer, Glaser (2004)

Optimal sequence of effective Hamiltonians

$$I^- \quad \to \quad S^-$$

$\lvert\mu_1\rvert$	$\lvert\mu_1\rvert$	$\lvert\mu_3\rvert$	$\lvert\mu_2\rvert$	$\lvert\mu_2\rvert$	$\lvert\mu_3\rvert$
$\lvert\mu_2\rvert$	$\lvert\mu_3\rvert$	$\lvert\mu_1\rvert$	$\lvert\mu_1\rvert$	$\lvert\mu_3\rvert$	$\lvert\mu_2\rvert$
$\lvert\mu_3\rvert$	$\lvert\mu_2\rvert$	$\lvert\mu_2\rvert$	$\lvert\mu_3\rvert$	$\lvert\mu_1\rvert$	$\lvert\mu_1\rvert$

Pulse sequences



Khaneja, Kramer, Glaser (2004)

Time-Optimal Simulation of Trilinear Coupling Terms

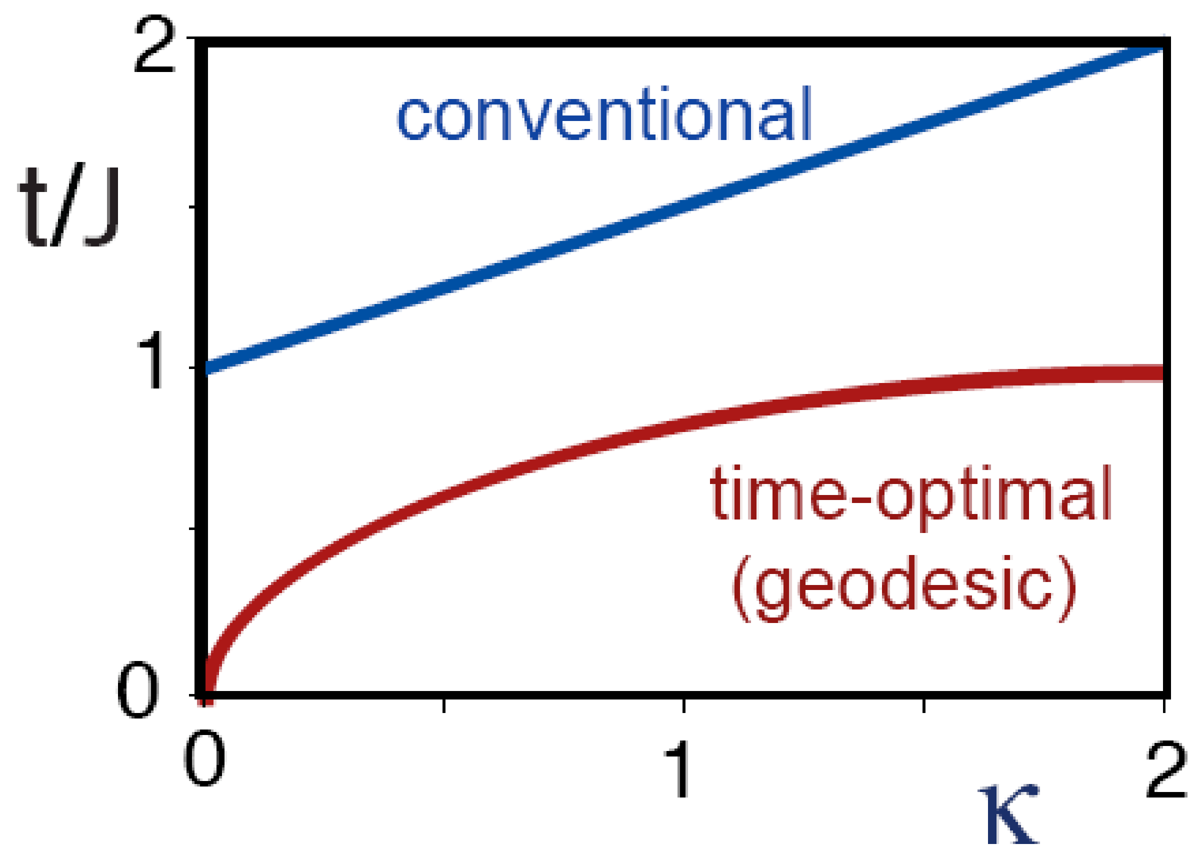
given:

$$H = 2\pi J\,(I_{1z} I_{2z} + I_{2z} I_{3z})$$

desired:

$$U = \exp\{-i\,\kappa\, 2\pi\, I_{1z} I_{2z} I_{3z}\}$$

$$H_{eff} = 2\pi J_{eff}\,(I_{1z} I_{2z} I_{3z})$$

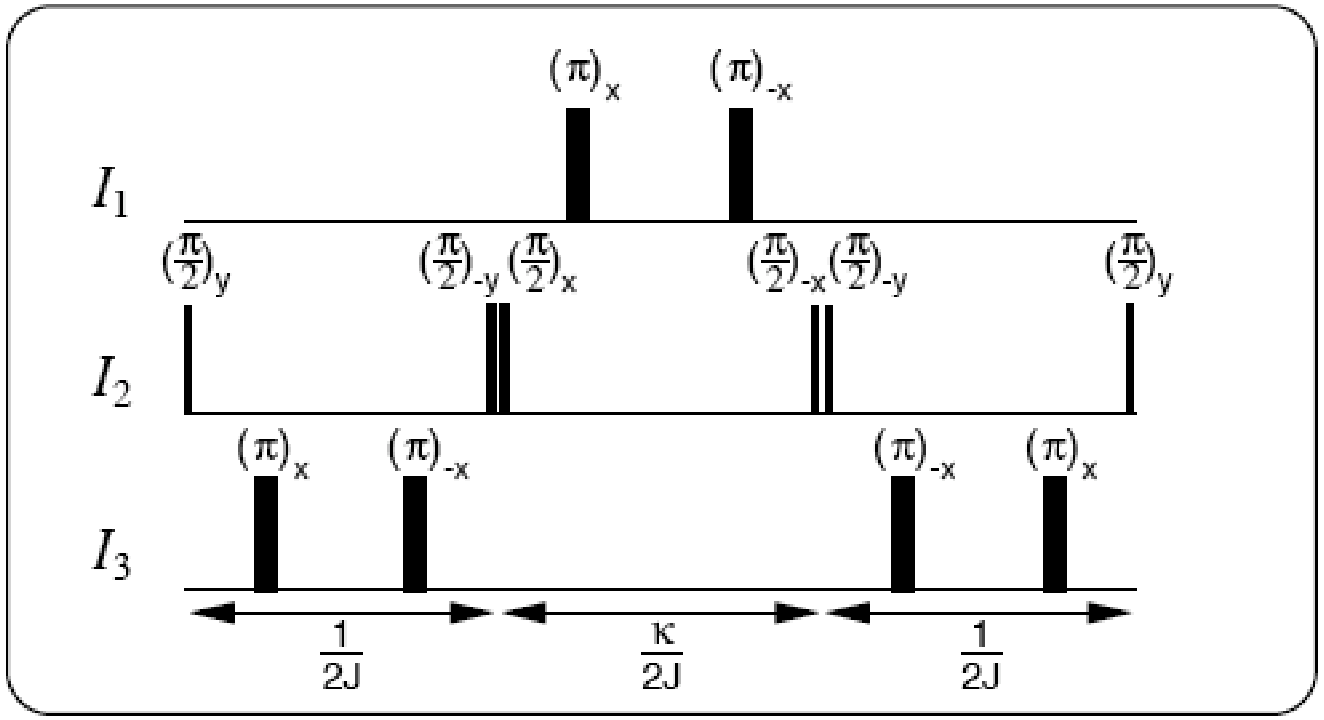


Tseng, Somaroo, Sharf, Knill, Laflamme, Havel, Cory, Phys. Rev. A 61, 012302 (2000)

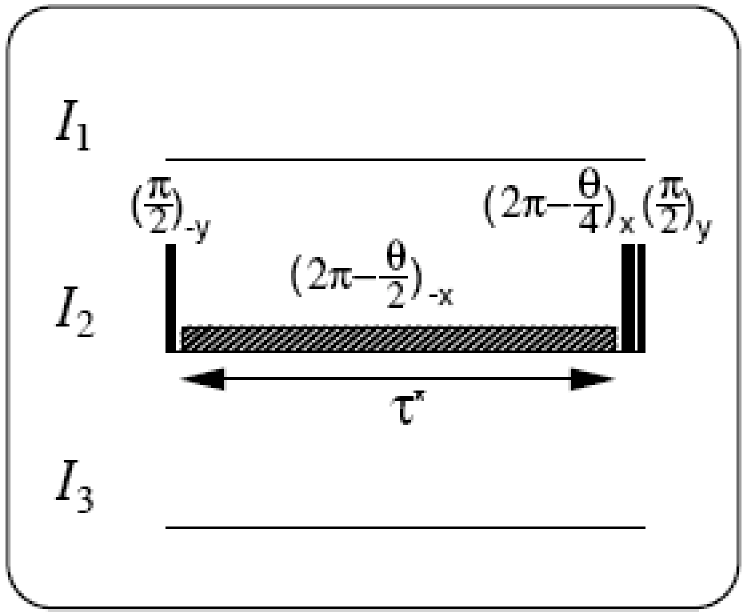
Khaneja, Glaser, Brockett, Phys. Rev. A 65, 032301 (2002)

Pulse Sequences ("zzz")

conventional

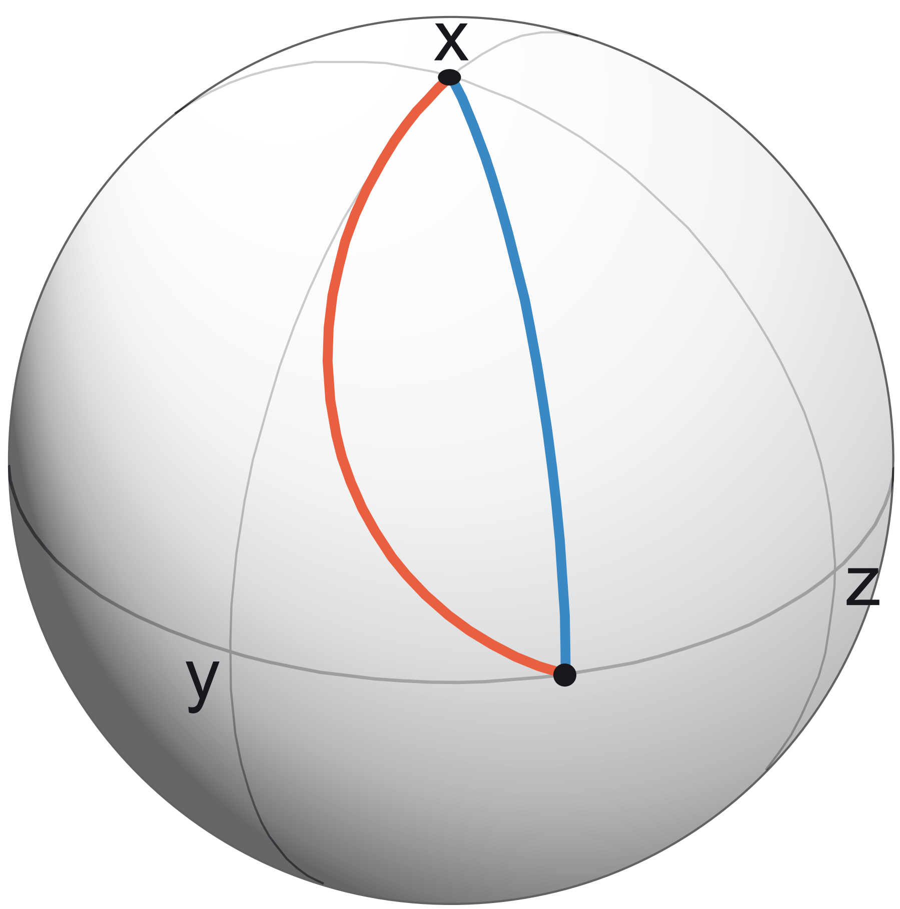


geodesic



Khaneja, Glaser, Brockett, Phys. Rev. A 65, 032301 (2002)

Geodesics on a sphere



Euklidian metric

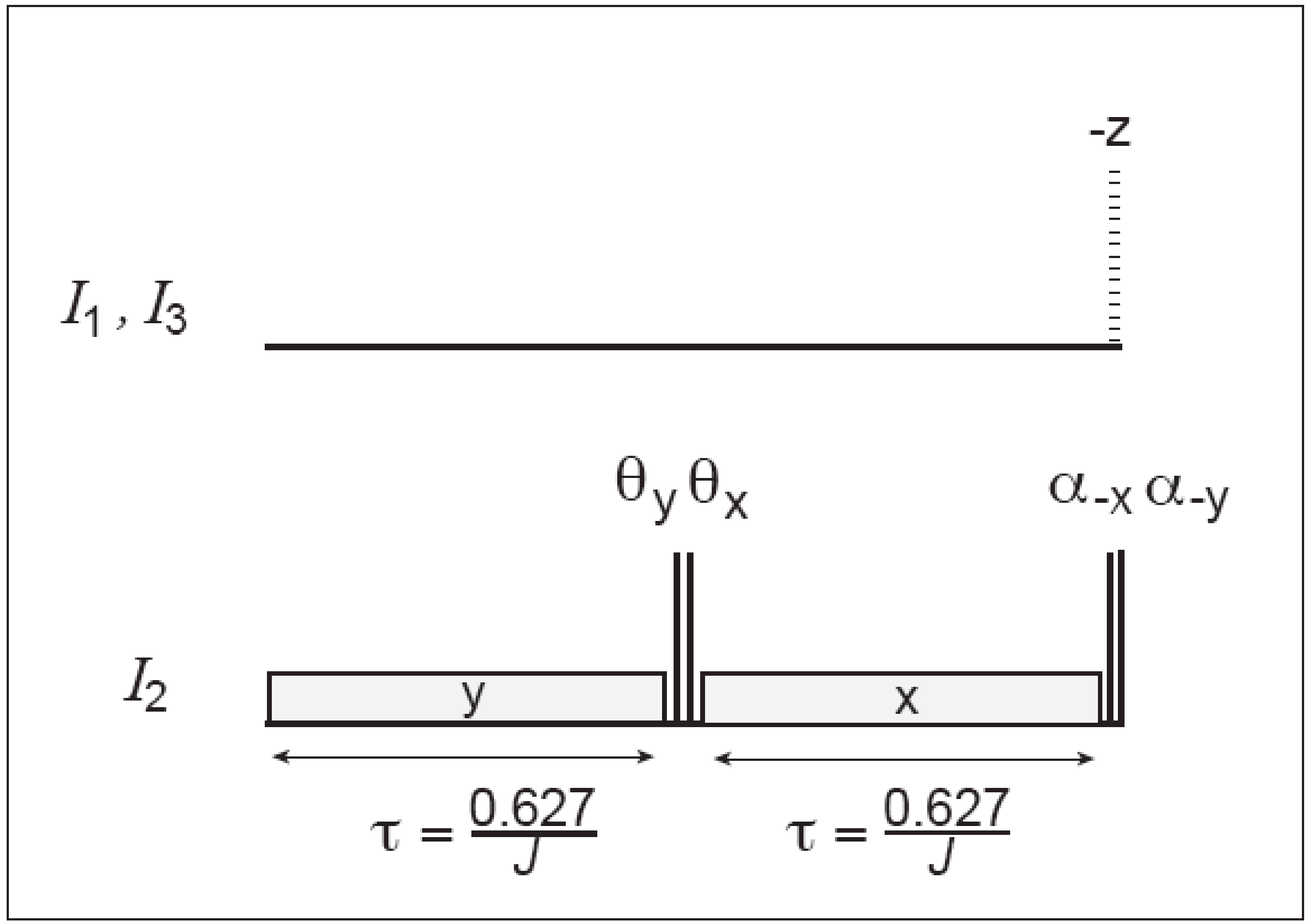
$$(dx)^2 + (dy)^2 + (dz)^2$$

“quantum gate design metric”

$$\frac{(dx)^2 + (dz)^2}{y^2}$$

Khaneja et al., Phys. Rev. A 75, 012322 (2007).

Pulse sequence for creating $U_{13}=\exp\{-i\,\pi\, I_{1z}I_{3z}\}$



$\theta=180^\circ-\alpha=31.4^\circ$, weak pulse amplitude: 0.52 J

Khaneja et al., Phys. Rev. A 75, 012322 (2007)

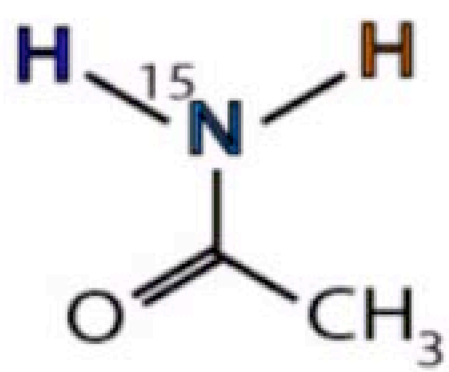
TABLE I. Duration τ_C of various implementations of the CNOT(1,3) gate.

Pulse sequence	τ_C (units of J^{-1})	Relative duration (%)
Sequence 1 (C1)	3.5	100
Sequence 2 (C2)	2.5	71.4
Sequence 3 (C3)	2.0	57.1
Sequence 4 (C4)	1.866	53.3
Sequence 5 (C5)	1.253	38.8

(C1, C2) D. Collins, K. W. Kim, W. C. Holton, H. Sierzputowska-Gracz, and E. O. Stejskal, Phys. Rev. A **62**, 022304 (2000).

(C3, C4, C5) *Khaneja et al., Phys. Rev. A 75, 012322 (2007)*

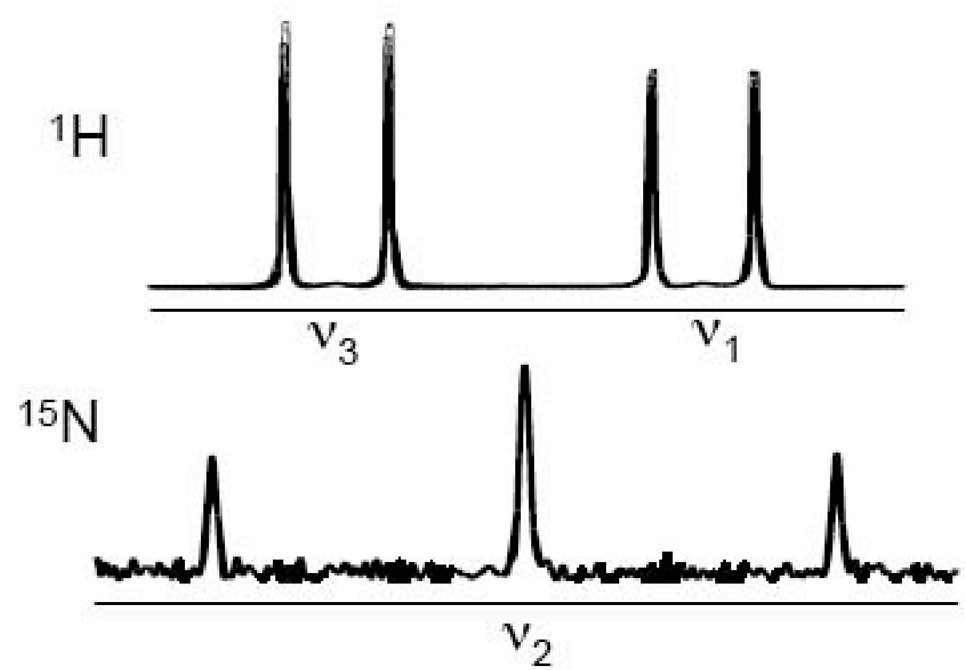
Experimental model system



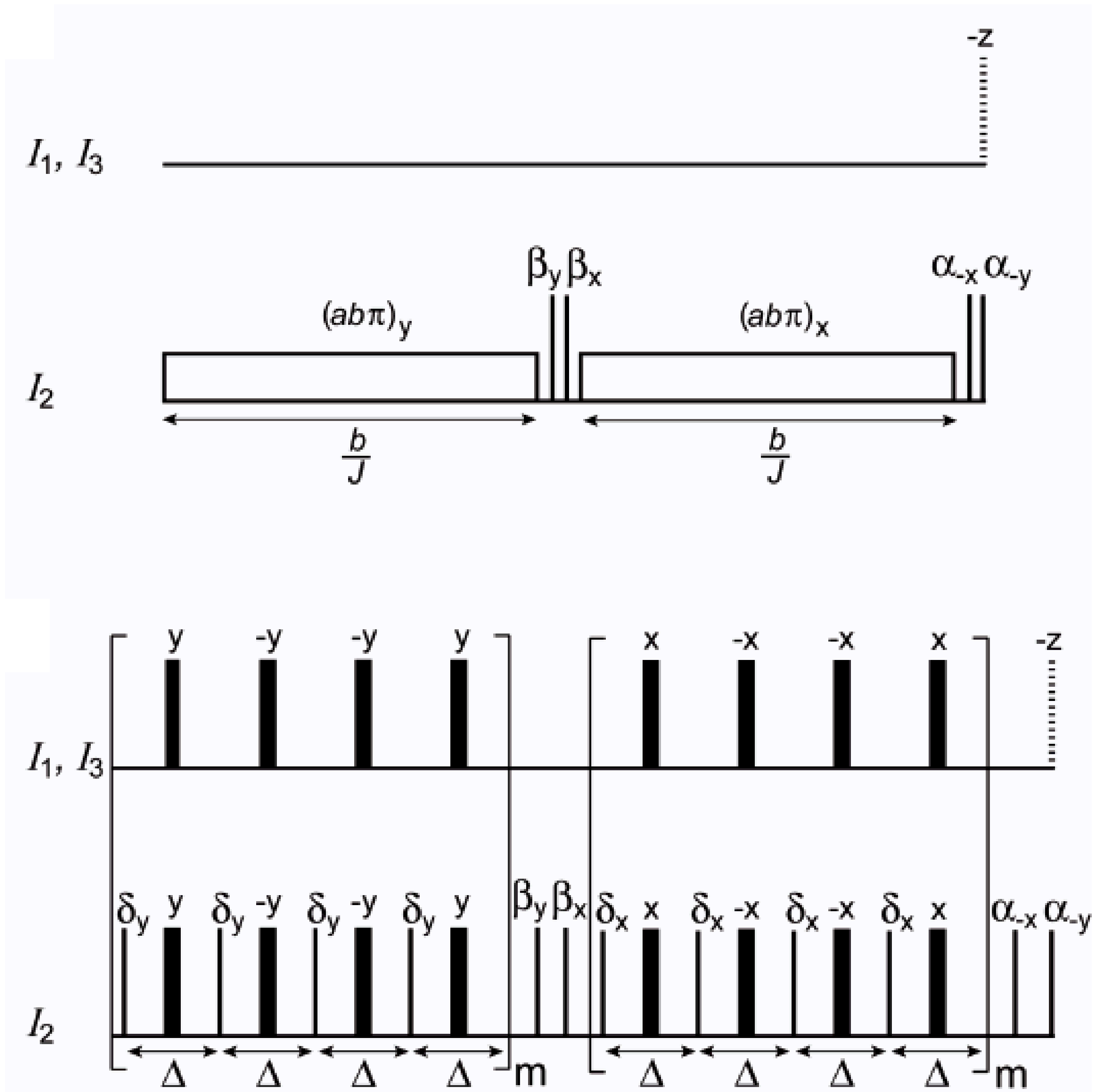
^{15}N - acetamide

Solvent: DMSO-d_6
Temp.: 295 K
Bruker 500 Avance Spectrometer

$J_{12} = -87.3$ Hz $\approx J_{23} = -88.8$ Hz » $J_{13} = 2.9$ Hz
$\Delta\nu_{13} = 310$ Hz



Experimental Demonstration U_{13}

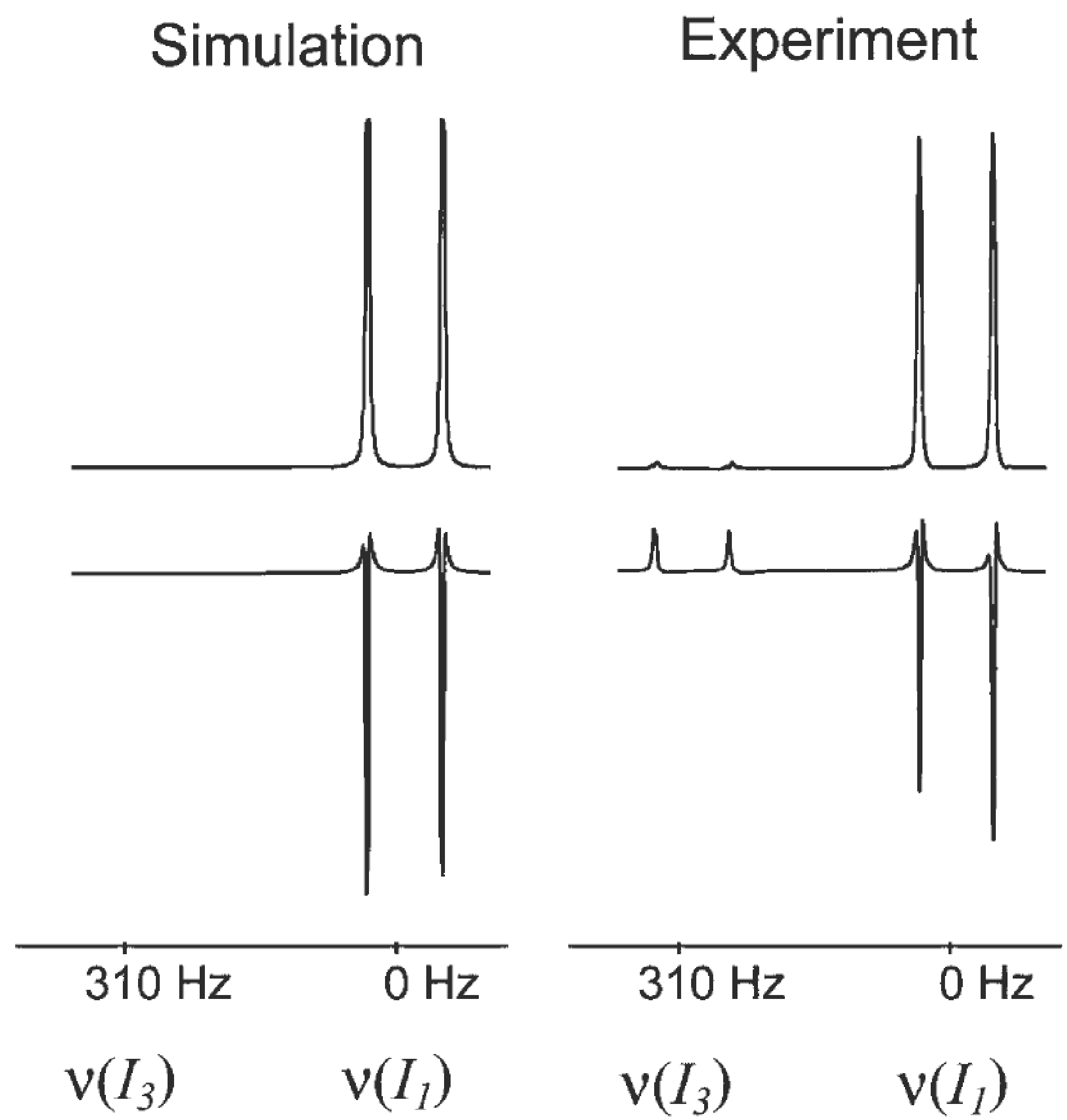


Khaneja et al., Phys. Rev. A 75, 012322 (2007)

$$\boxed{\mathcal{U}_{13}=\exp\{-i\tfrac{\pi}{2}2I_{1z}I_{3z}\}}$$

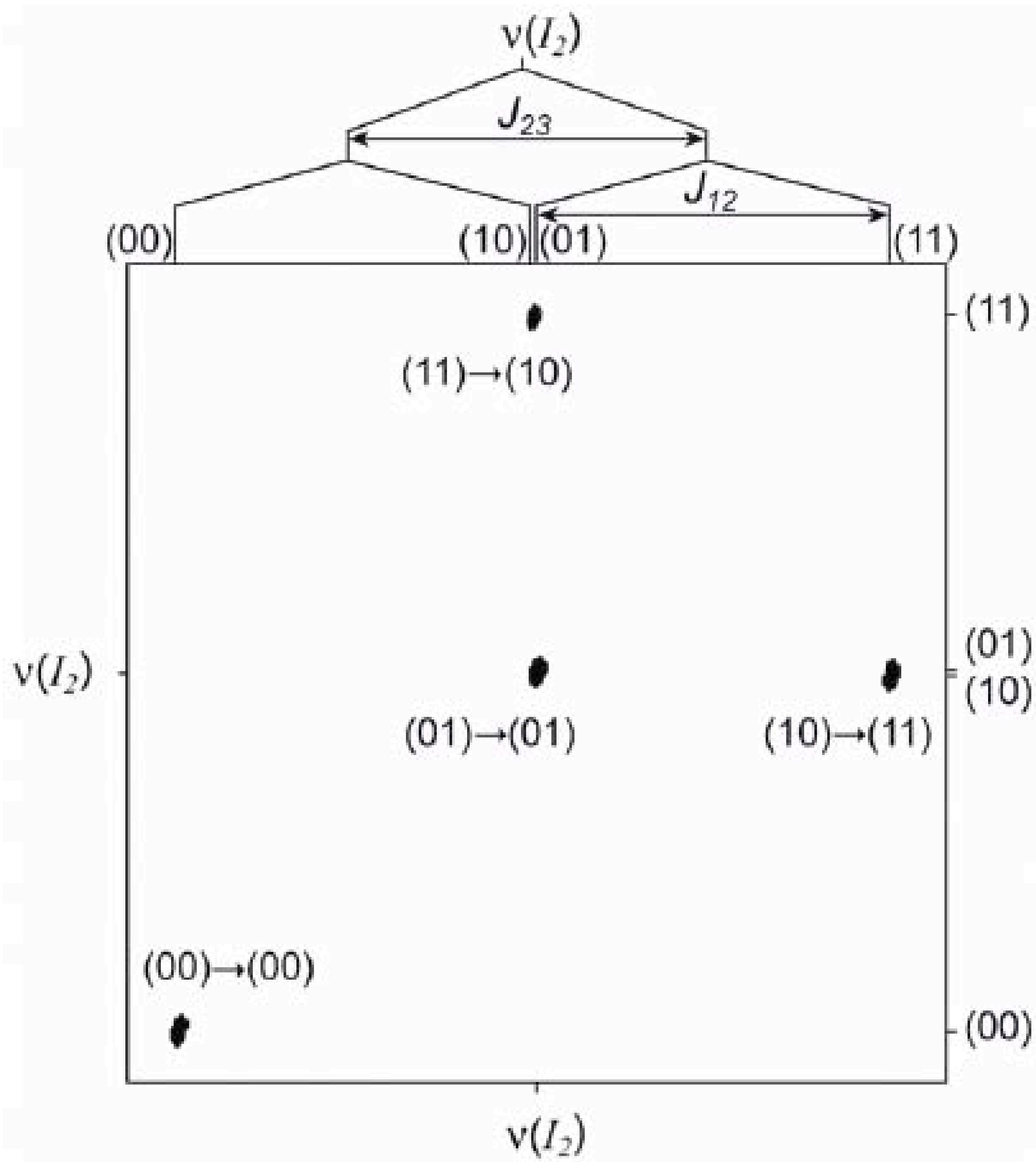
$\rho_A = I_{1x}$

$\rho_B = 2I_{1y}I_{3z}$

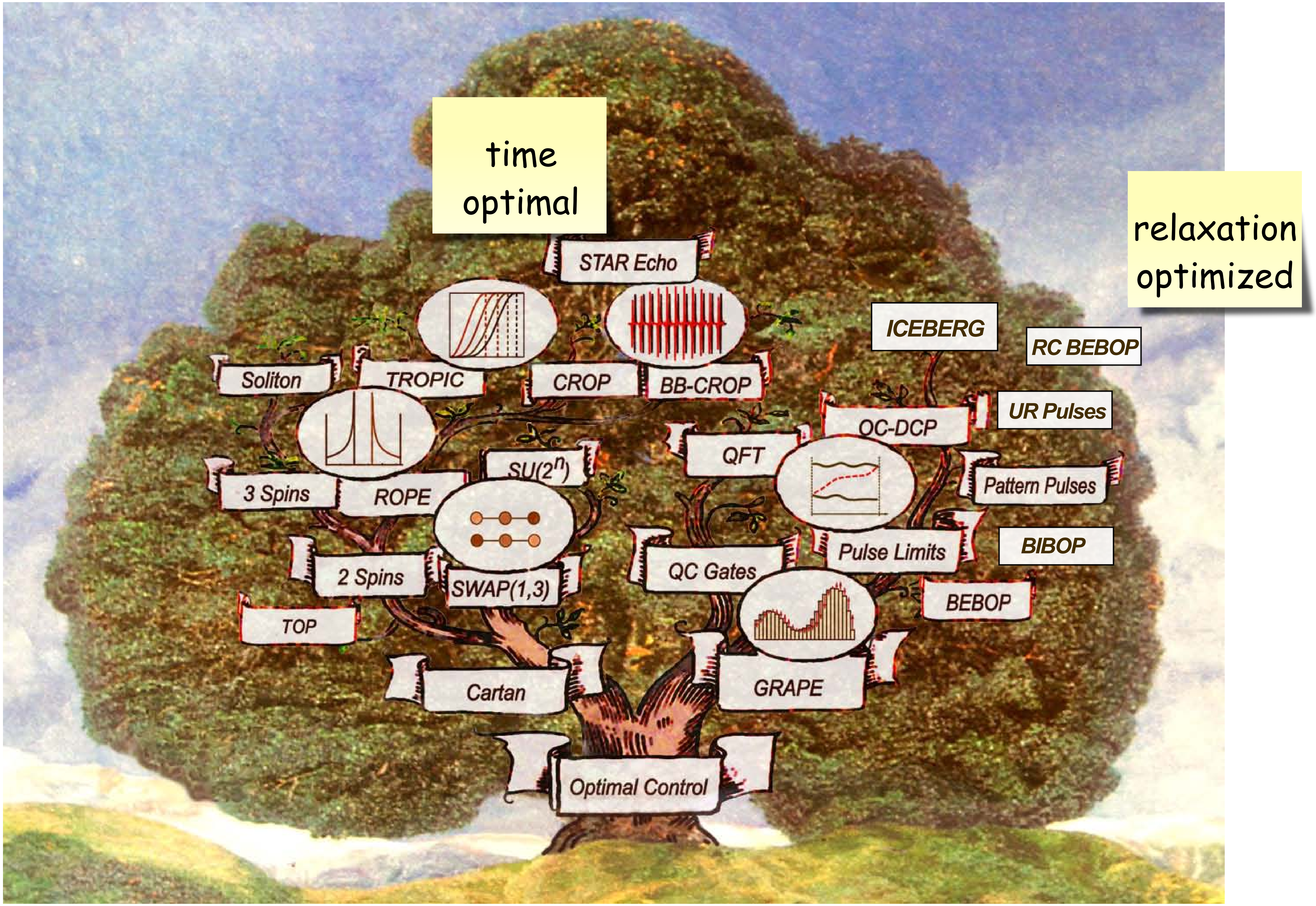


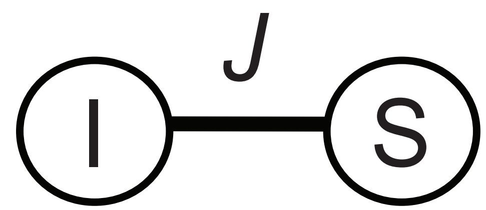
Khaneja et al., Phys. Rev. A 75, 012322 (2007)

Experimental demonstration of CNOT(1,3)



Khaneja et al., Phys. Rev. A 75, 012322 (2007)


time optimal
relaxation optimized
STAR Echo
ICEBERG
RC BEBOP
Soliton
TROPIC
CROP
BB-CROP
UR Pulses
OC-DCP
QFT
SU(2^n)
3 Spins
ROPE
Pattern Pulses
Pulse Limits
BIBOP
2 Spins
SWAP(1,3)
QC Gates
BEBOP
TOP
Cartan
GRAPE
Optimal Control



Dipol-Dipol Relaxation in the Spin-Diffusion Limit

$$\dot{\rho} = \pi J\,[-i\,2I_z S_z, \rho] + \pi k\,[2I_z S_z, [2I_z S_z, \rho]]$$

$$\xi = k/J$$

$$I_x \xrightarrow{?} 2I_y S_z$$

Conventional transfer (INEPT)

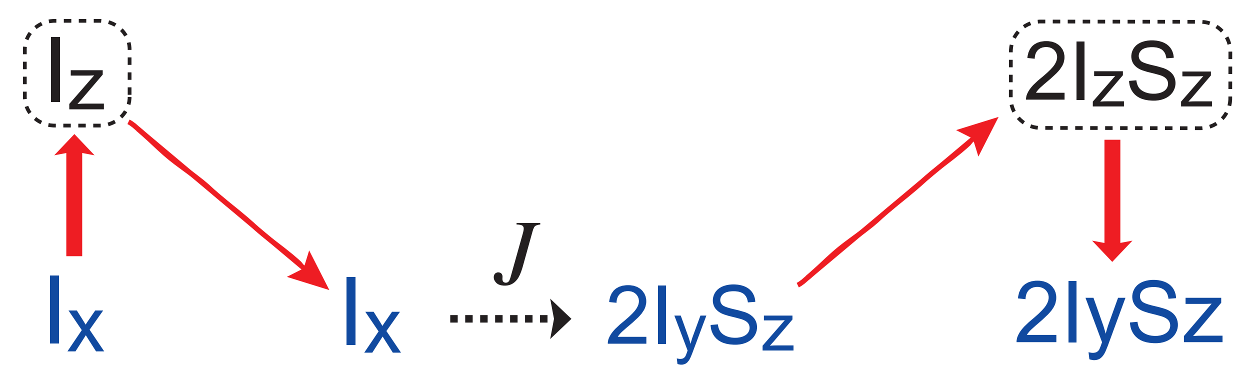
$$I_x \xrightarrow{J} 2I_yS_z$$

Morris, Freeman, J. Am. Chem. Soc. 101,760 (1979)

Conventional transfer (INEPT)

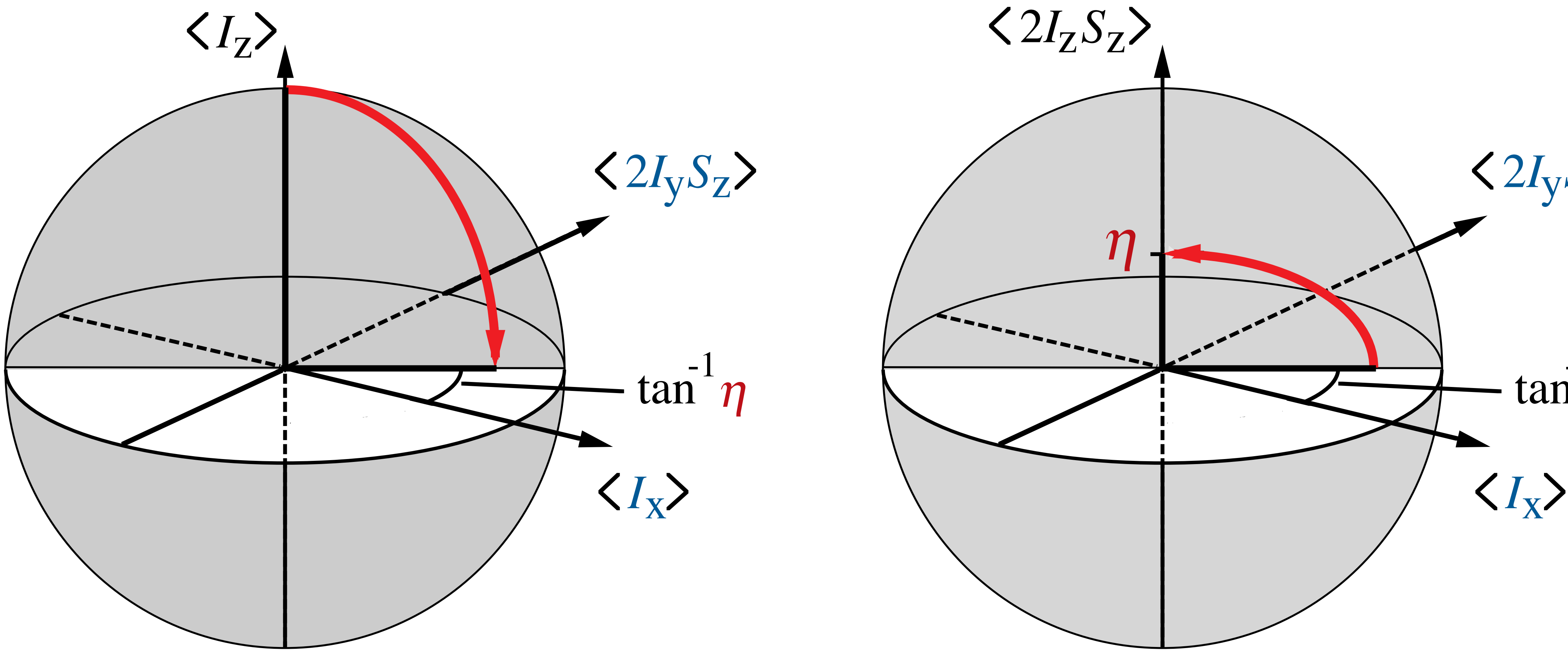
$$I_x \xrightarrow{J} 2I_yS_z$$

Relaxation-optimized transfer (ROPE)



Khaneja, Reiss, Luy, Glaser, J. Magn. Reson.162, 311 (2003)

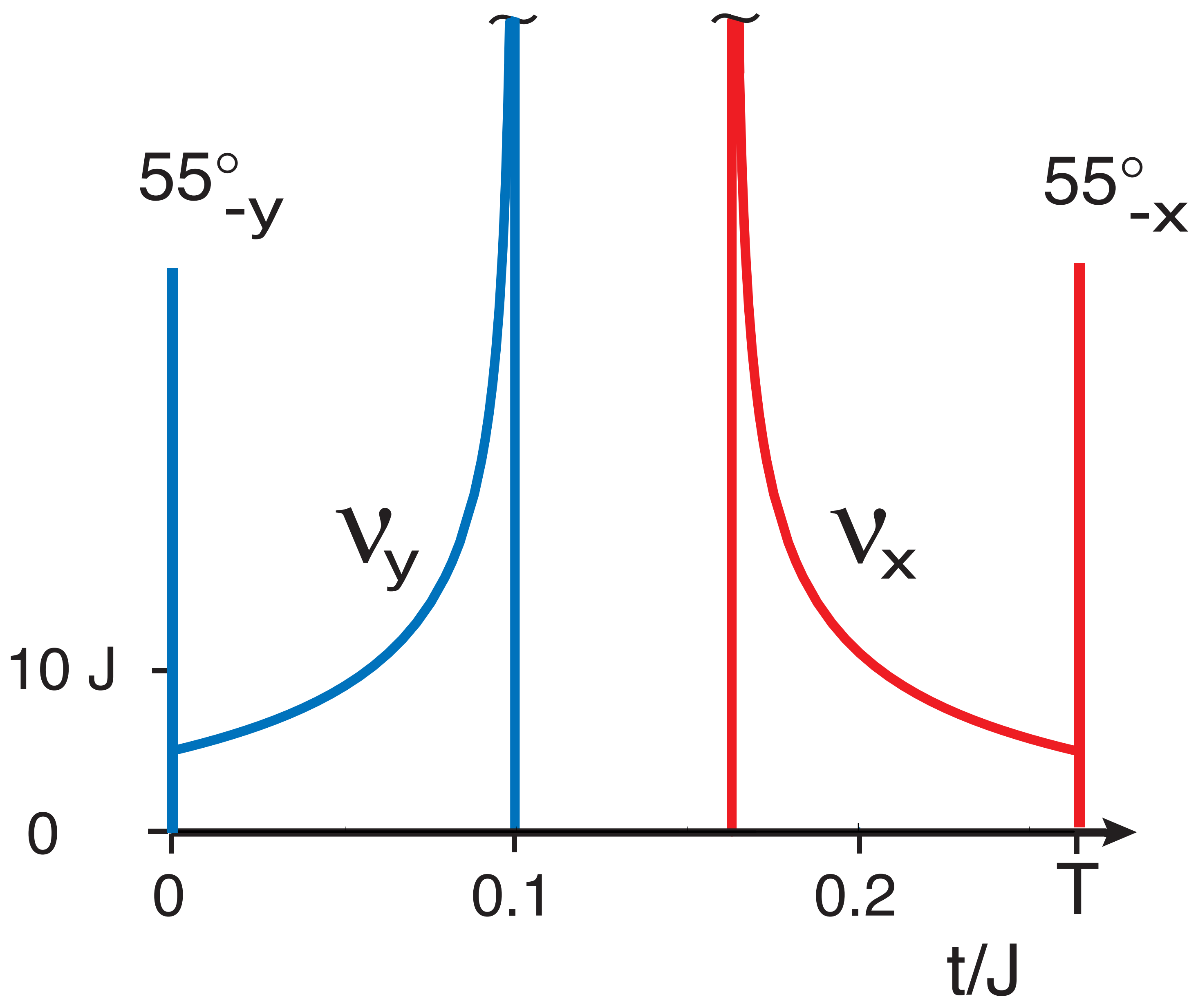
ROPE Trajectory



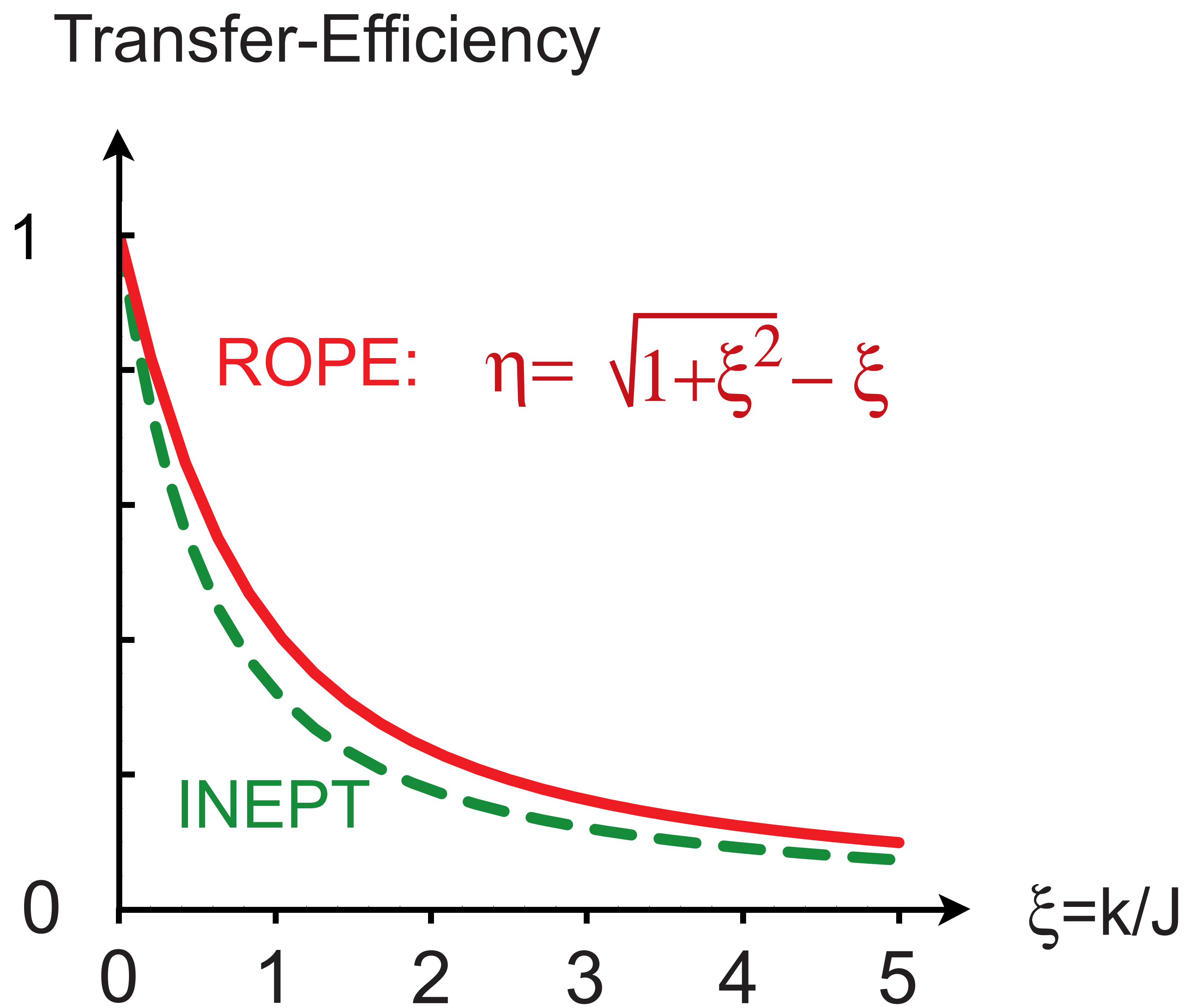
Optimal transfer efficiency $\eta = \sqrt{1+\xi^2} - \xi$ $\xi = k/J$

Khaneja et al., J. Magn. Reson.162, 311 (2003)

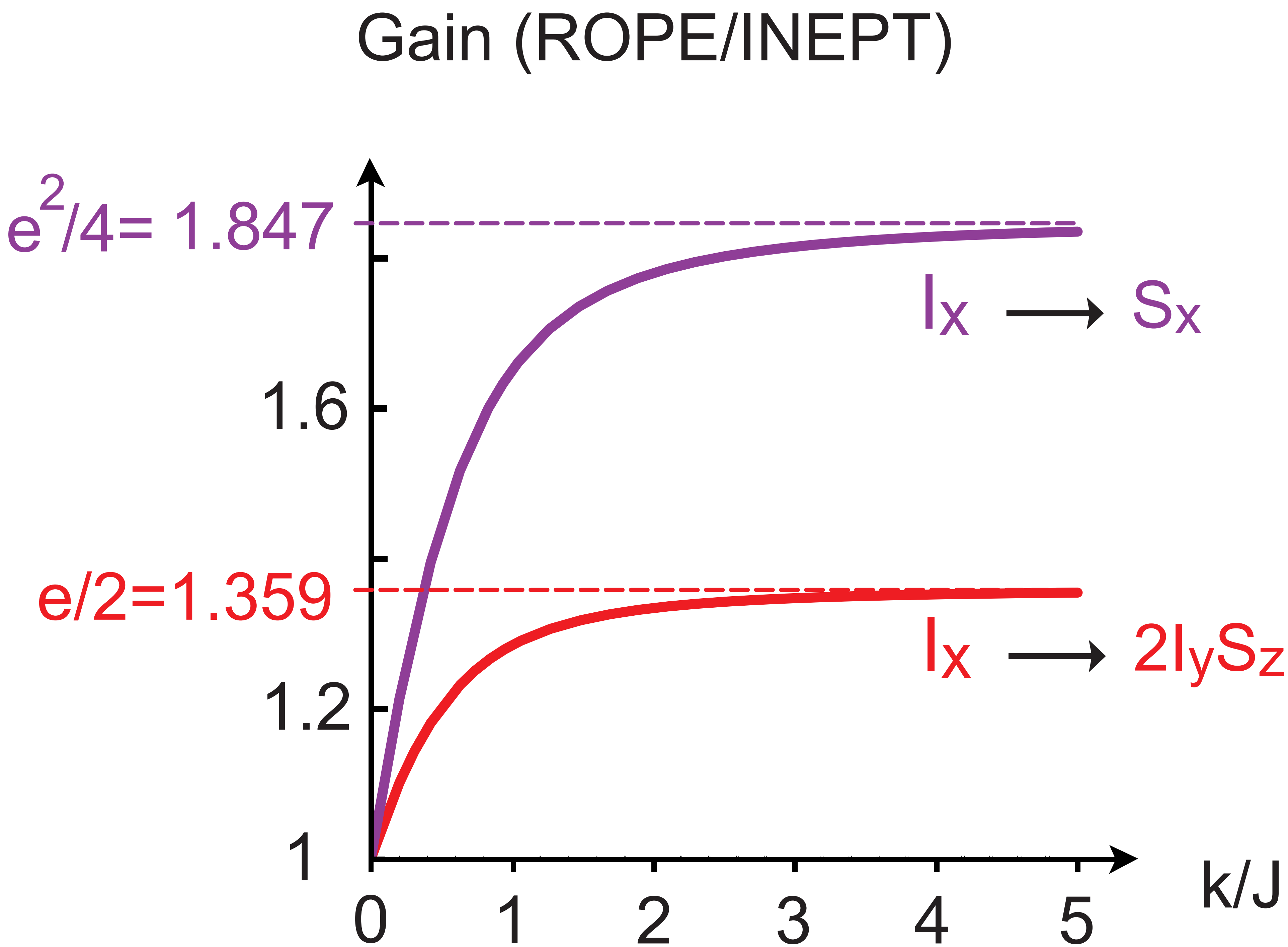
ROPE Pulse Sequence



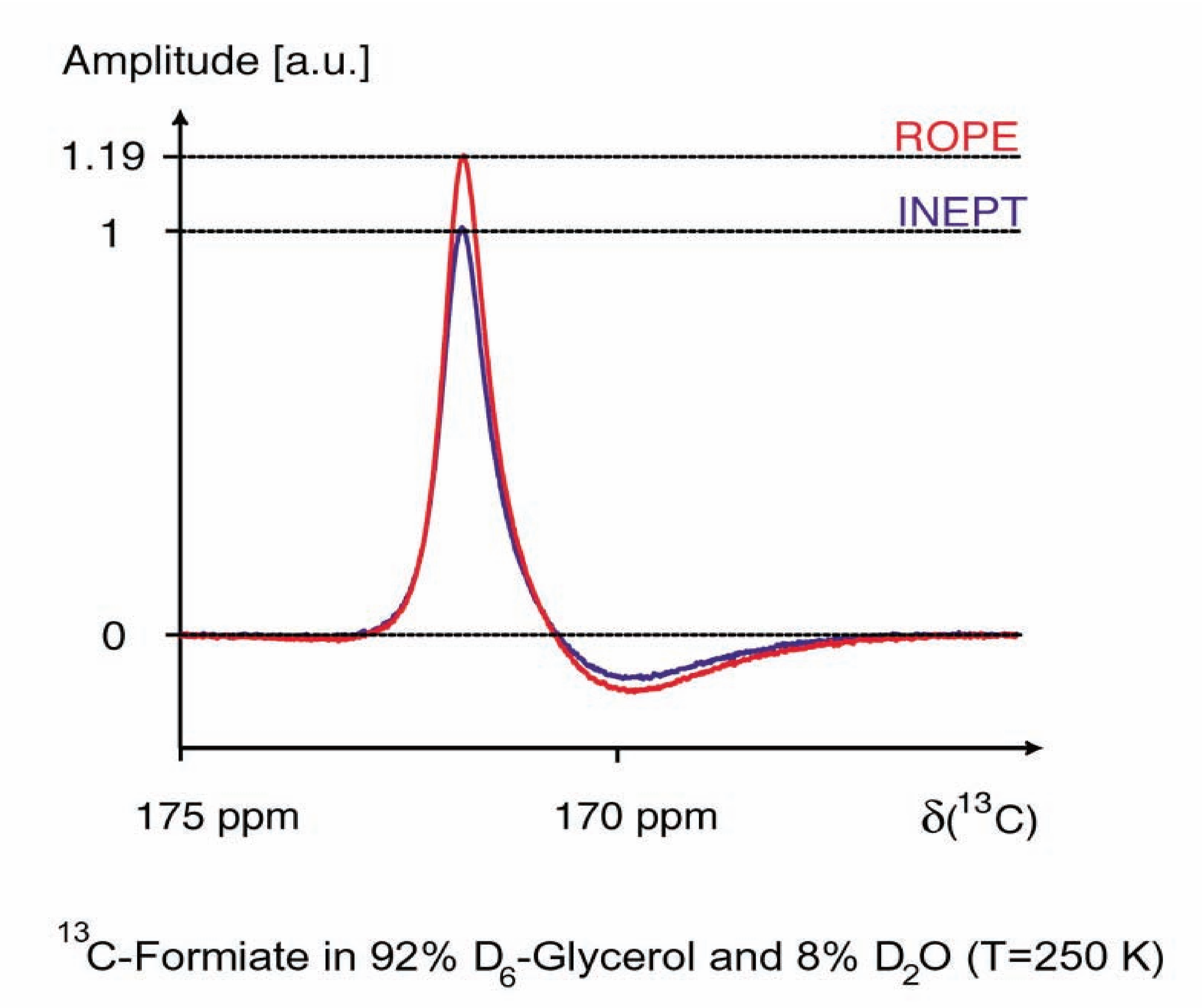
Khaneja et al., J. Magn. Reson.162, 311 (2003)



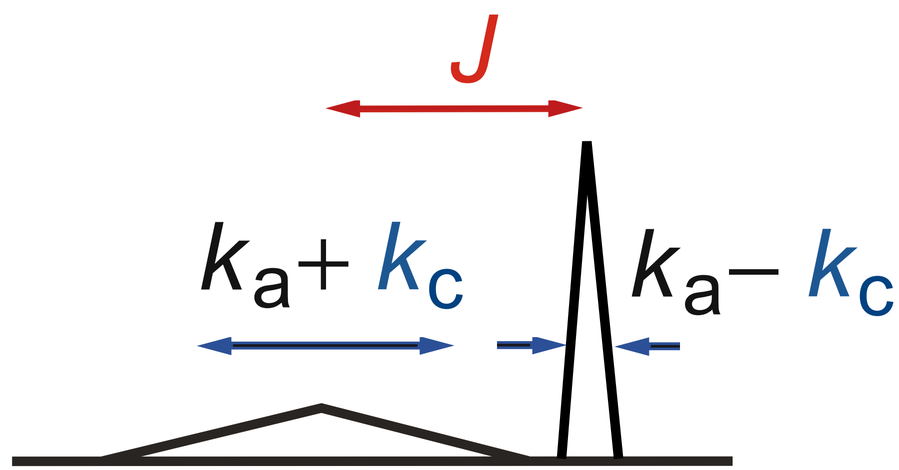
Khaneja et al., J. Magn. Reson.162, 311 (2003)



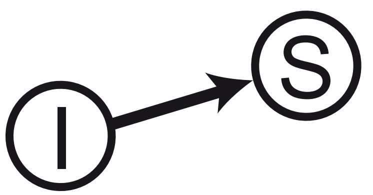
Khaneja et al., J. Magn. Reson.162, 311 (2003)



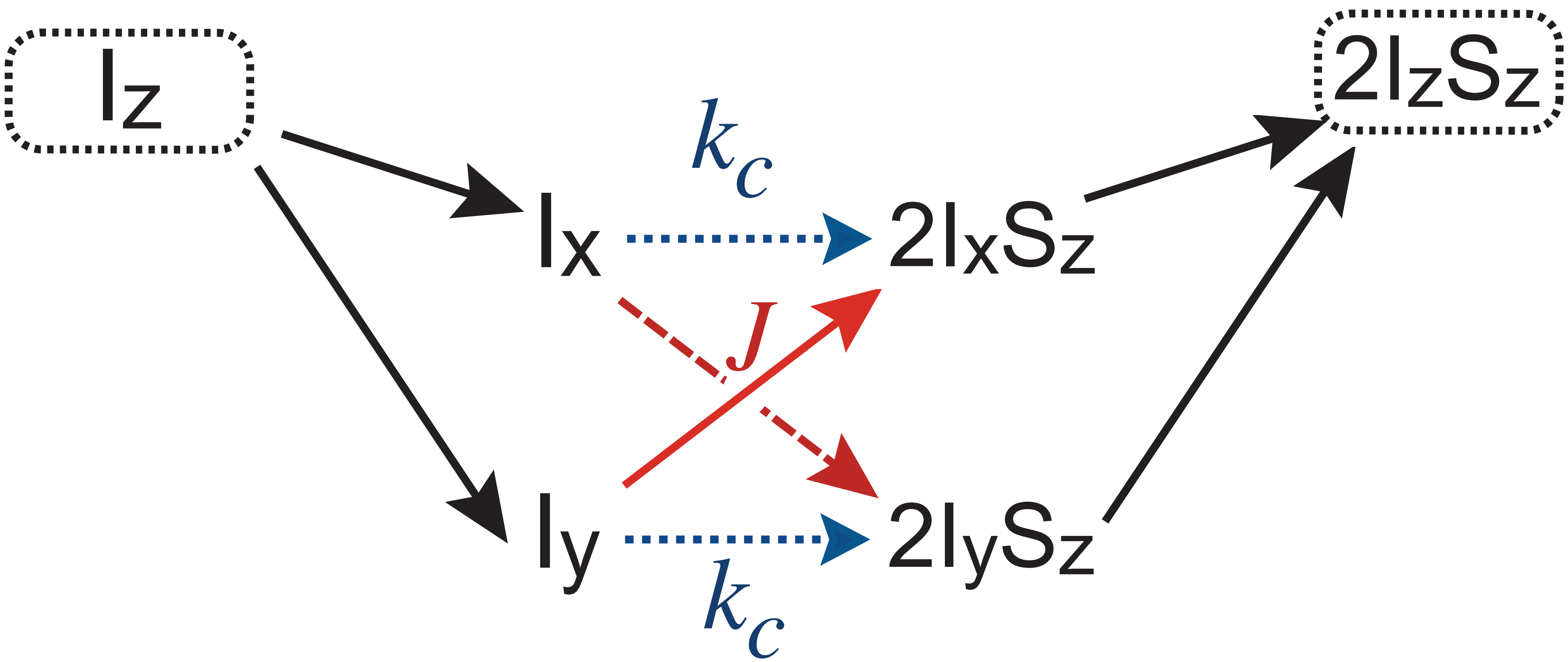
Khaneja et al., J. Magn. Reson.162, 311 (2003)



Multiplet of Spin I



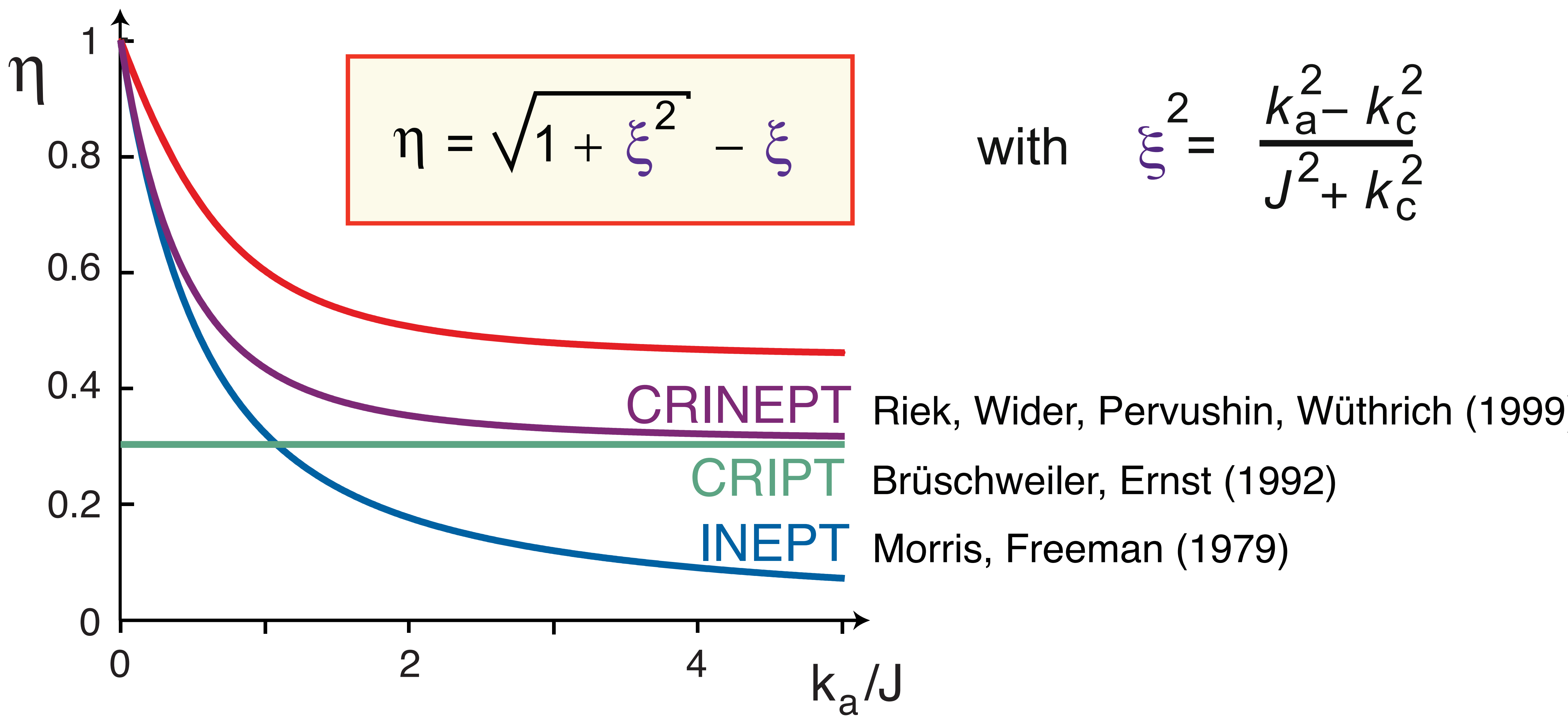
Polarization Transfer in the Presence of Cross-Correlated Relaxation



Transfer Efficiency η for $k_c / k_a = 0.75$

$$\eta = \sqrt{1 + \xi^2} - \xi$$

with $\xi^2 = \dfrac{k_a^2 - k_c^2}{J^2 + k_c^2}$



maximum transfer efficiency:

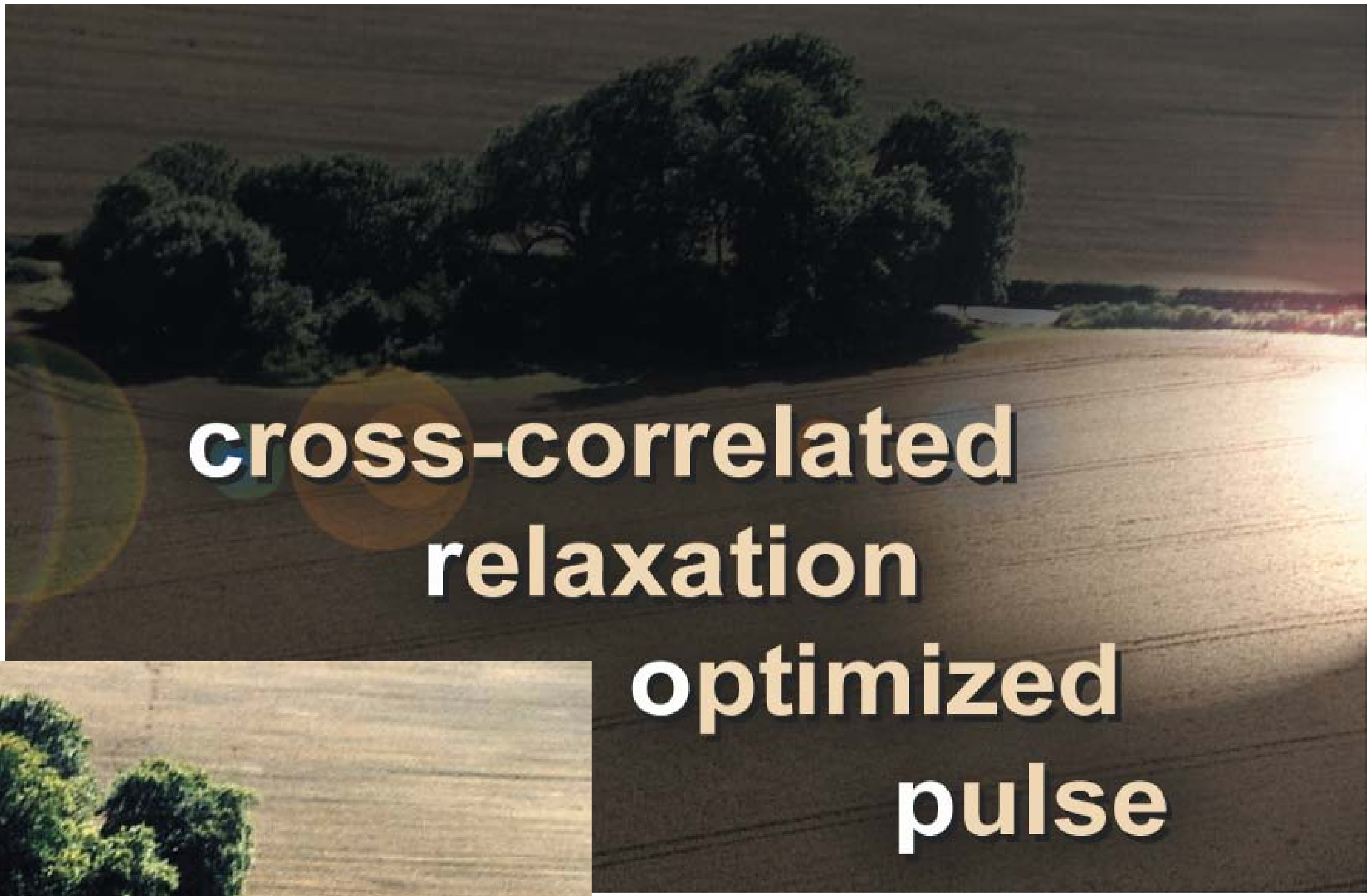
$$\boxed{\eta = \sqrt{1+\xi^2} - \xi}$$

formal proof (based on principles of optimum control theory):

optimal return function $V(r_1, r_2)$

Hamilton-Jacobi-Bellman equation

$$\max_{u_1, u_2} \left[\frac{\partial V}{\partial r_1} \delta r_1 + \frac{\partial V}{\partial r_2} \delta r_2 \right] = 0$$

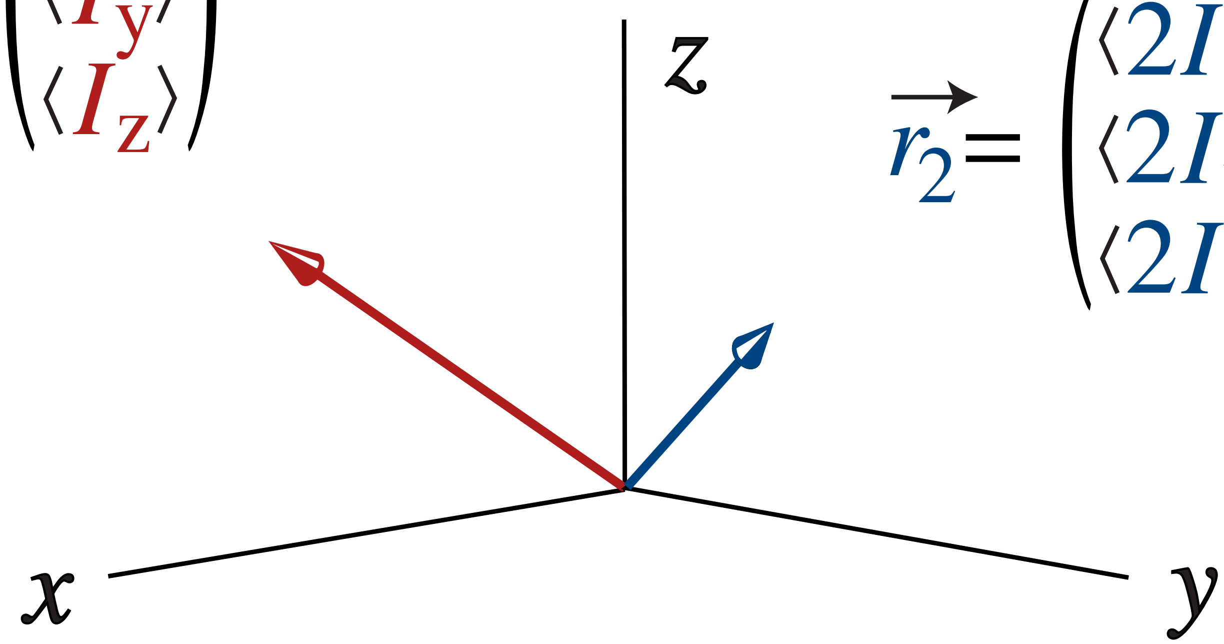


Khaneja, Luy, Glaser,
Proc. Natl. Acad. Sci (2003)

Representing in-phase and antiphase magnetization vectors $\vec{r}_1$ and $\vec{r}_2$ in a common frame of reference

$$\vec{r}_1 = \begin{pmatrix} \langle I_x \rangle \\ \langle I_y \rangle \\ \langle I_z \rangle \end{pmatrix}$$

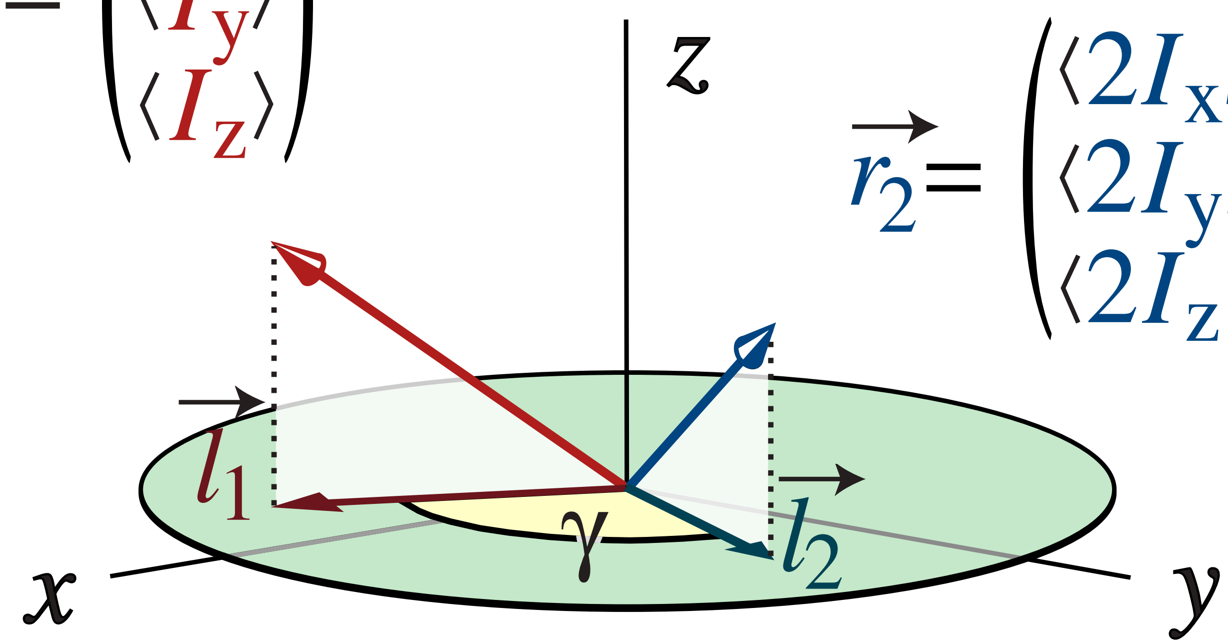
$$\vec{r}_2 = \begin{pmatrix} \langle 2I_xS_z \rangle \\ \langle 2I_yS_z \rangle \\ \langle 2I_zS_z \rangle \end{pmatrix}$$



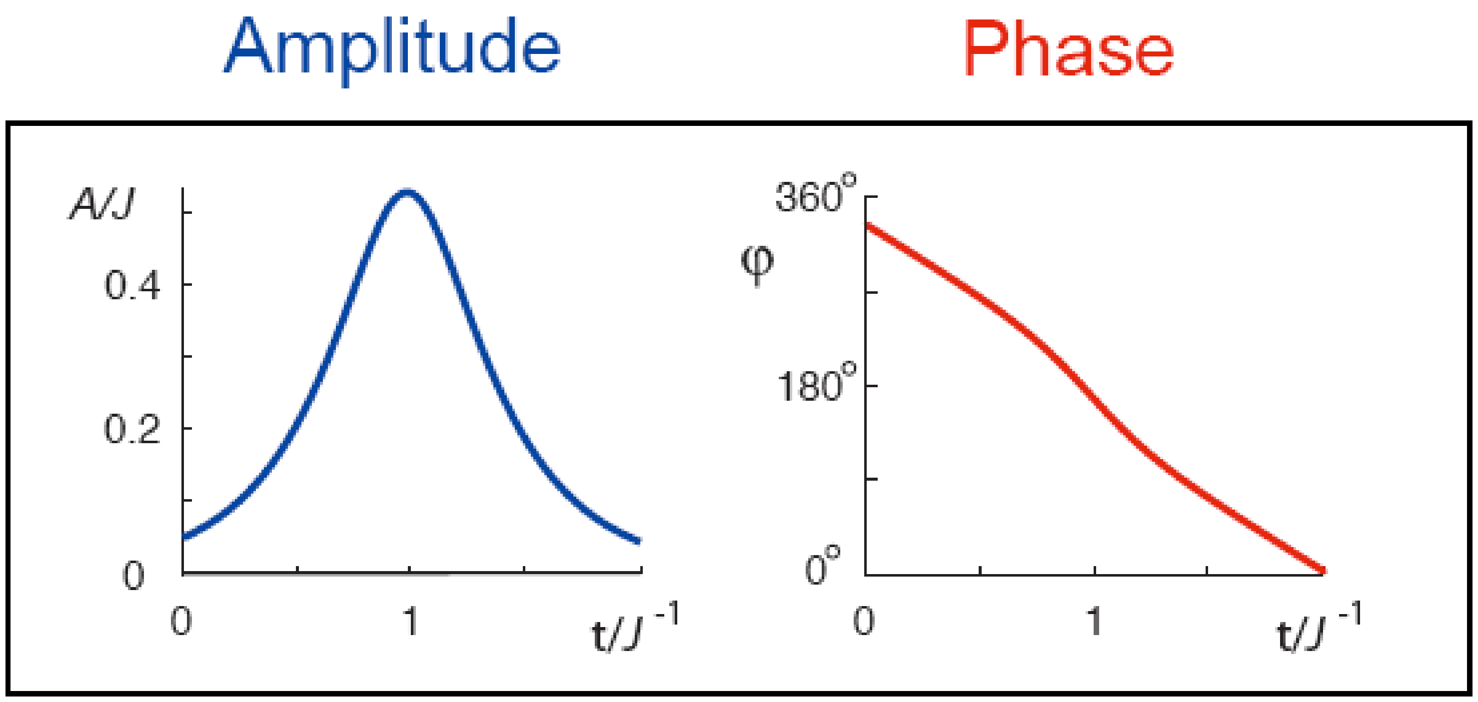
Optimal trajectory preserves ratio $\dfrac{l_2}{l_1} = \eta$ and angle γ

$$\vec{r}_1 = \begin{pmatrix} \langle I_x \rangle \\ \langle I_y \rangle \\ \langle I_z \rangle \end{pmatrix}$$

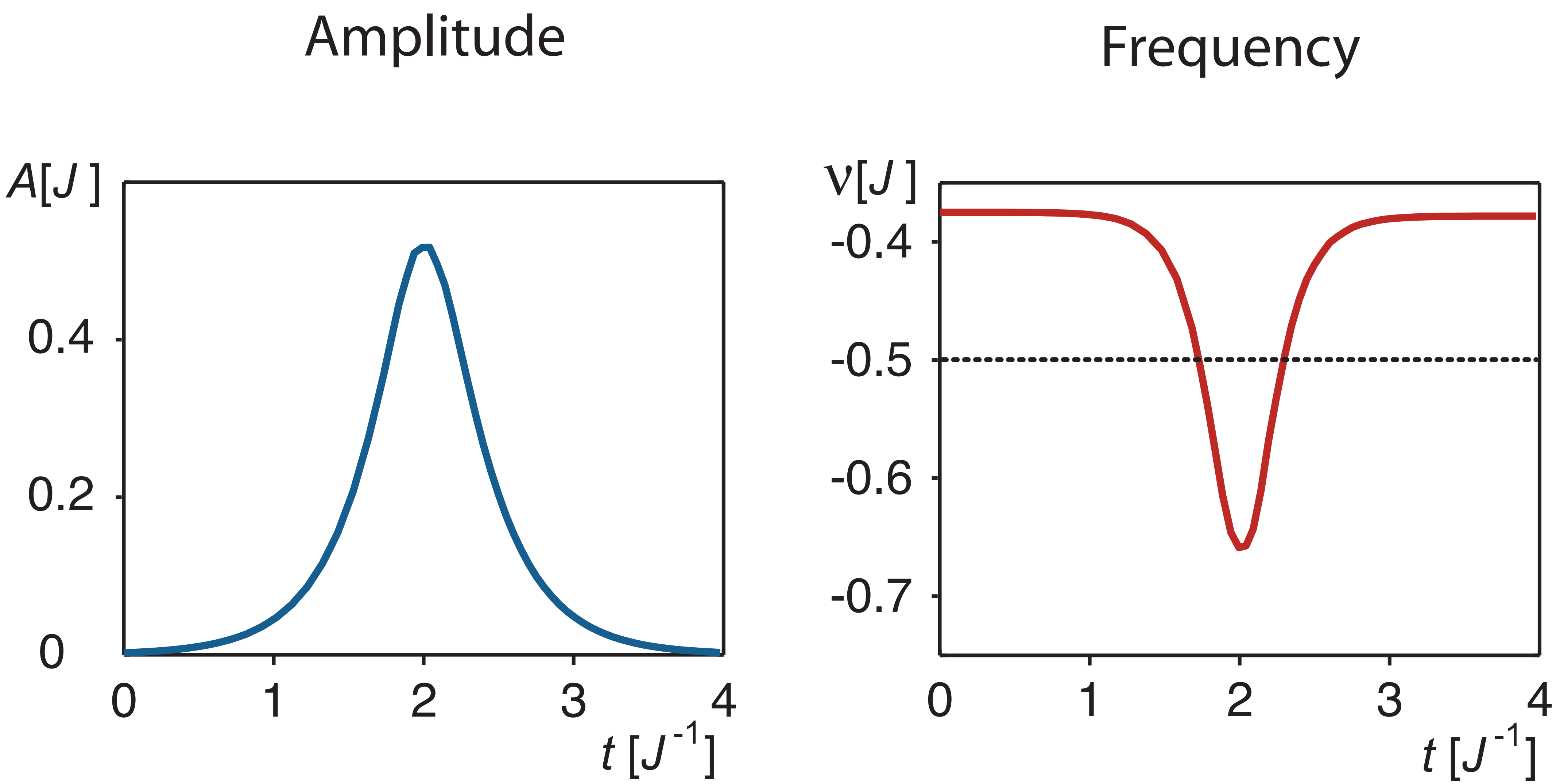
$$\vec{r}_2 = \begin{pmatrix} \langle 2I_x S_z \rangle \\ \langle 2I_y S_z \rangle \\ \langle 2I_z S_z \rangle \end{pmatrix}$$



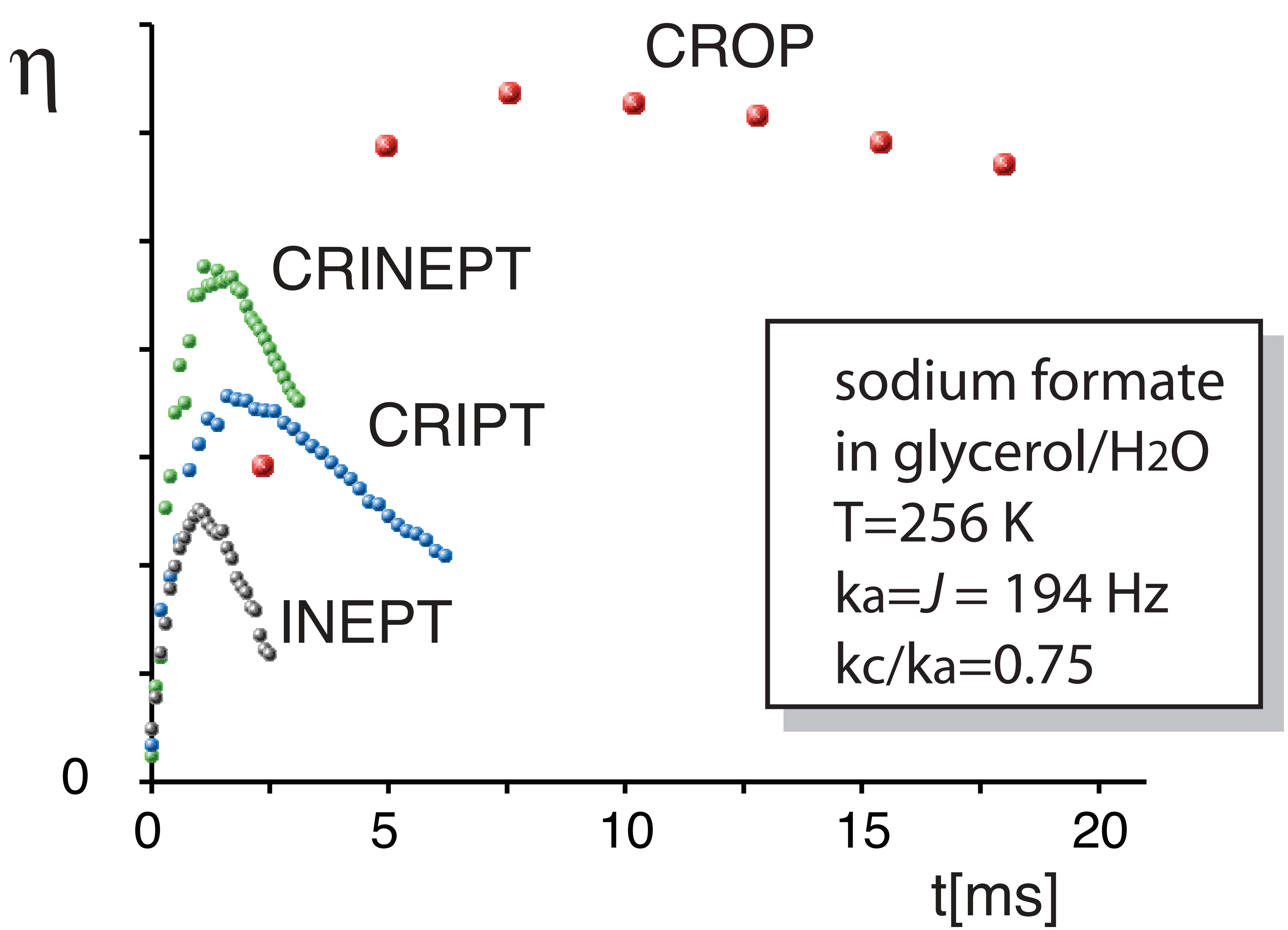
Optimal pulse shape for $k_c / k_a = 0.75$



CROP (**c**ross-correlated **r**elaxation **o**ptimized **p**ulse)



Experimental Transfer Functions



Relaxation-optimized heteronuclear transfer of polarization and spin order

- in two-spin systems

  without cross-correlated relaxation (ROPE)

  with cross-correlated relaxation (CROP)

- in spin chains (SPORTS ROPE)

  simple case: no cross-correlated relaxation

  same transverse relaxation rates,
  no cross-corr. relaxation
  - analytical upper limit for spin order transfer
  - only one smoth pulse, approx. Gaussian
  - more efficient than concatenated INEPT

  solutions for general chains

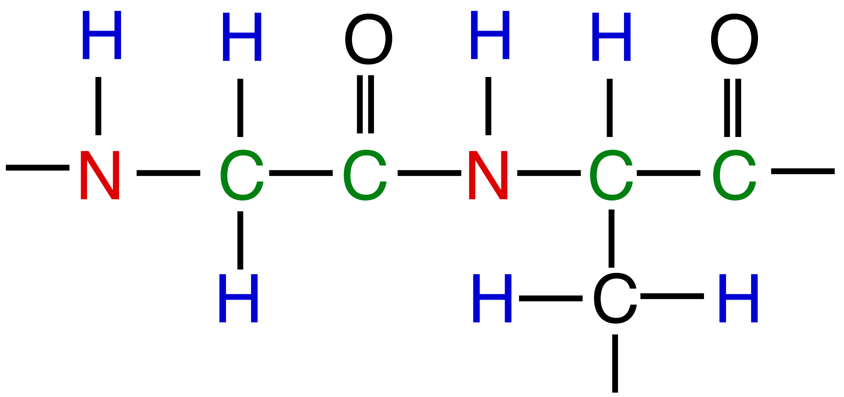
Challenges for biological applications

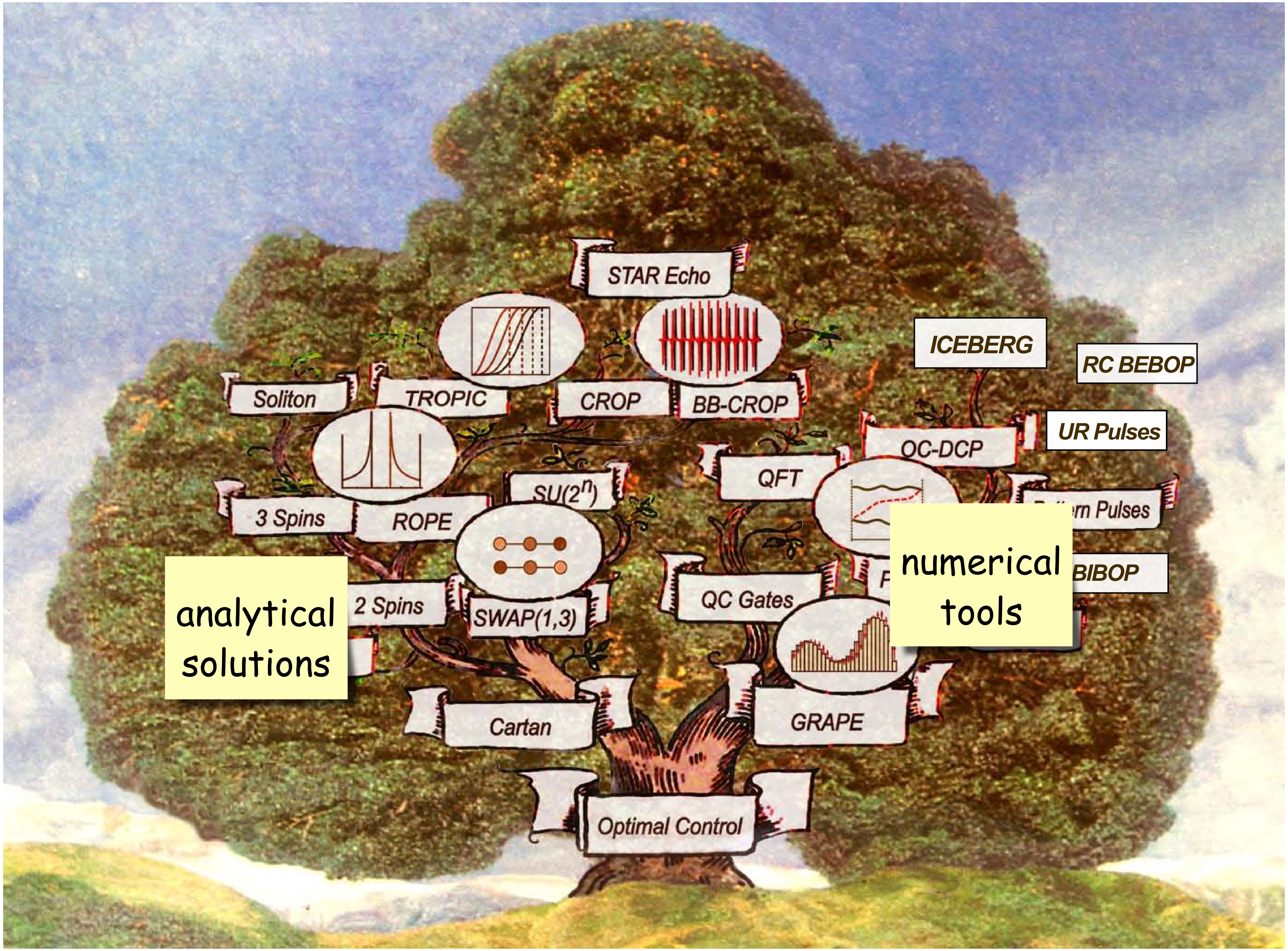
- offset compensation

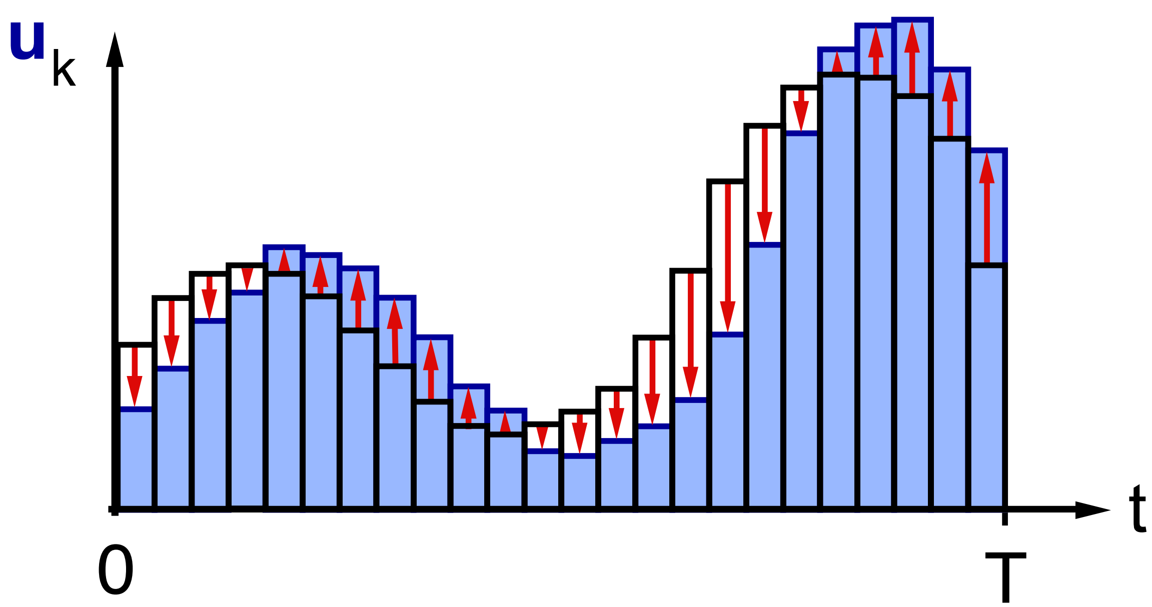
- experimental imperfections

- homonuclear couplings

- branched coupling networks




STAR Echo
ICEBERG
RC BEBOP
Soliton
TROPIC
CROP
BB-CROP
UR Pulses
OC-DCP
QFT
3 Spins
ROPE
$SU(2^n)$
BIBOP
analytical solutions
numerical tools
2 Spins
SWAP(1,3)
QC Gates
Cartan
GRAPE
Optimal Control



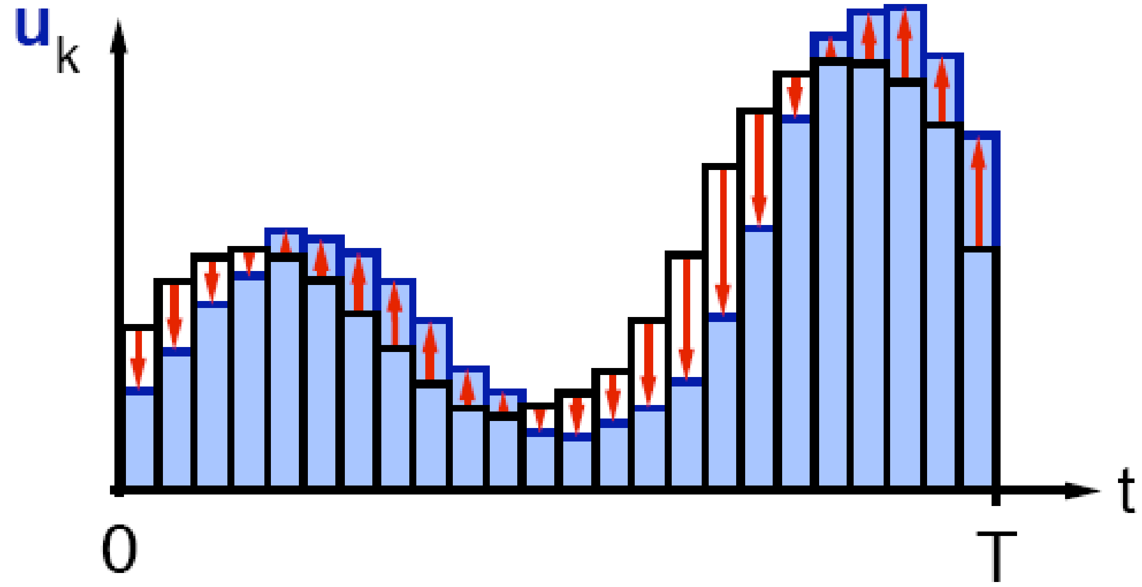
desired transfer: $A \longrightarrow C$

performance: $\langle C \mid \rho(T) \rangle$

$\rho(0) = A$

$\lambda(T) = C$

$$\boxed{u_k(t) \longrightarrow u_k(t) + \varepsilon \langle \lambda(t) \,|\, [-i H_k, \rho(t)] \rangle}$$



desired propagator: U_F

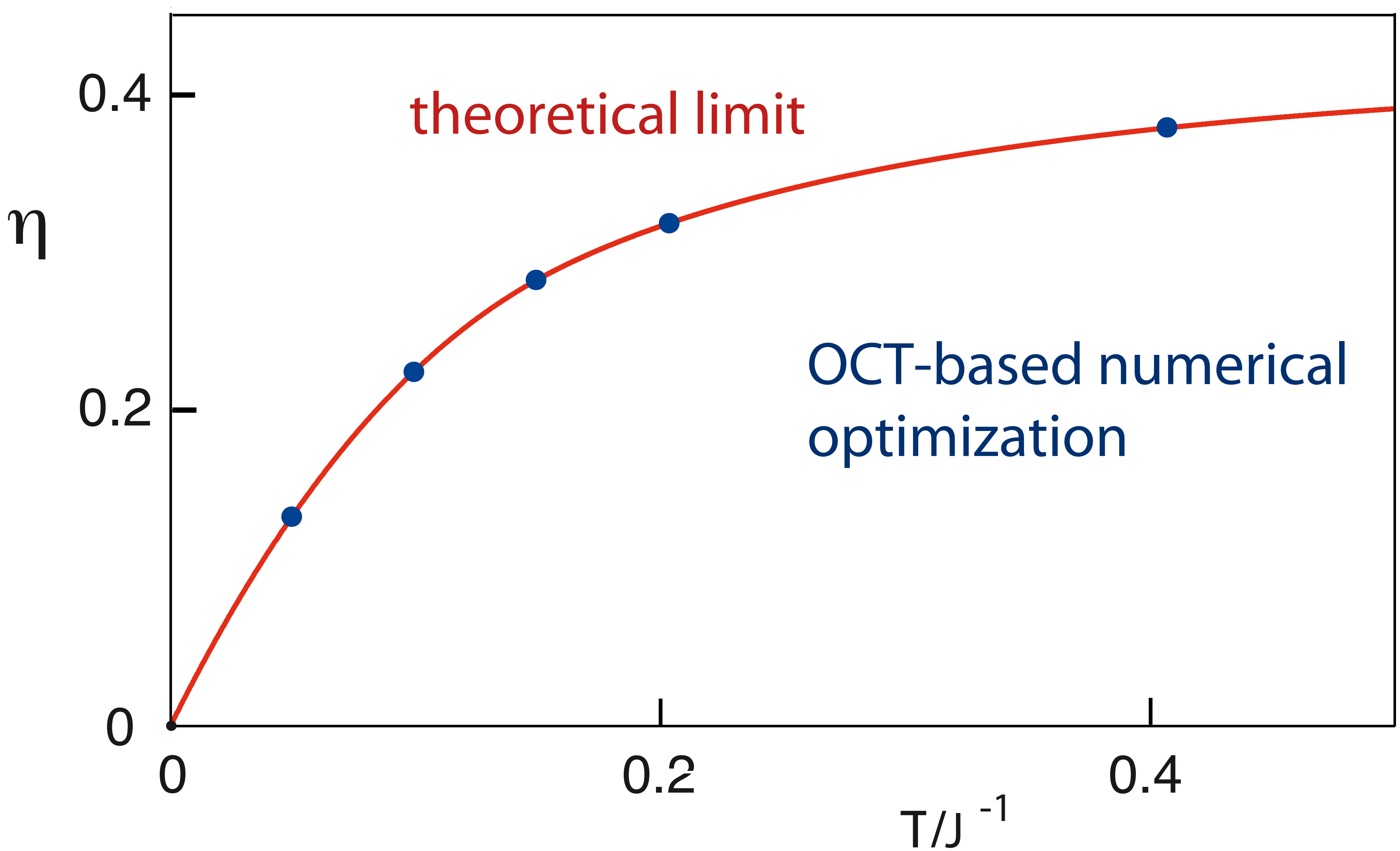
performance: $\left|\left\langle U_F \middle| U(T) \right\rangle\right|^2$

$U(0) = \mathbf{1}$

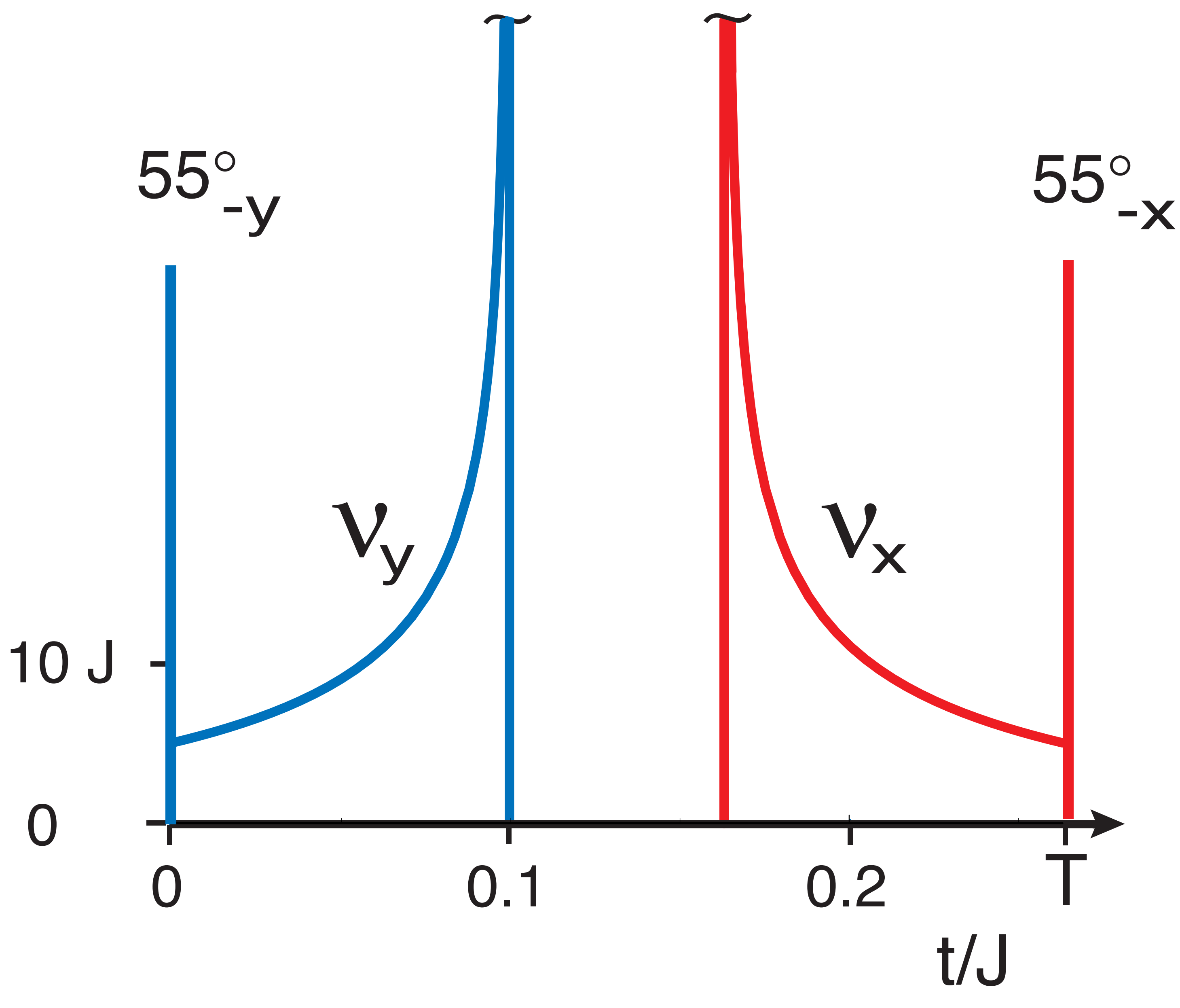
$P(T) = U_F$

$$\boxed{\mathbf{u}_k(t) \longrightarrow \mathbf{u}_k(t) + \varepsilon\, Re \left\{ \left\langle P(t) \middle| -i\, H_k U(t) \right\rangle \left\langle U(t) \middle| P(t) \right\rangle \right\}}$$

relaxation-optimized coherence transfer

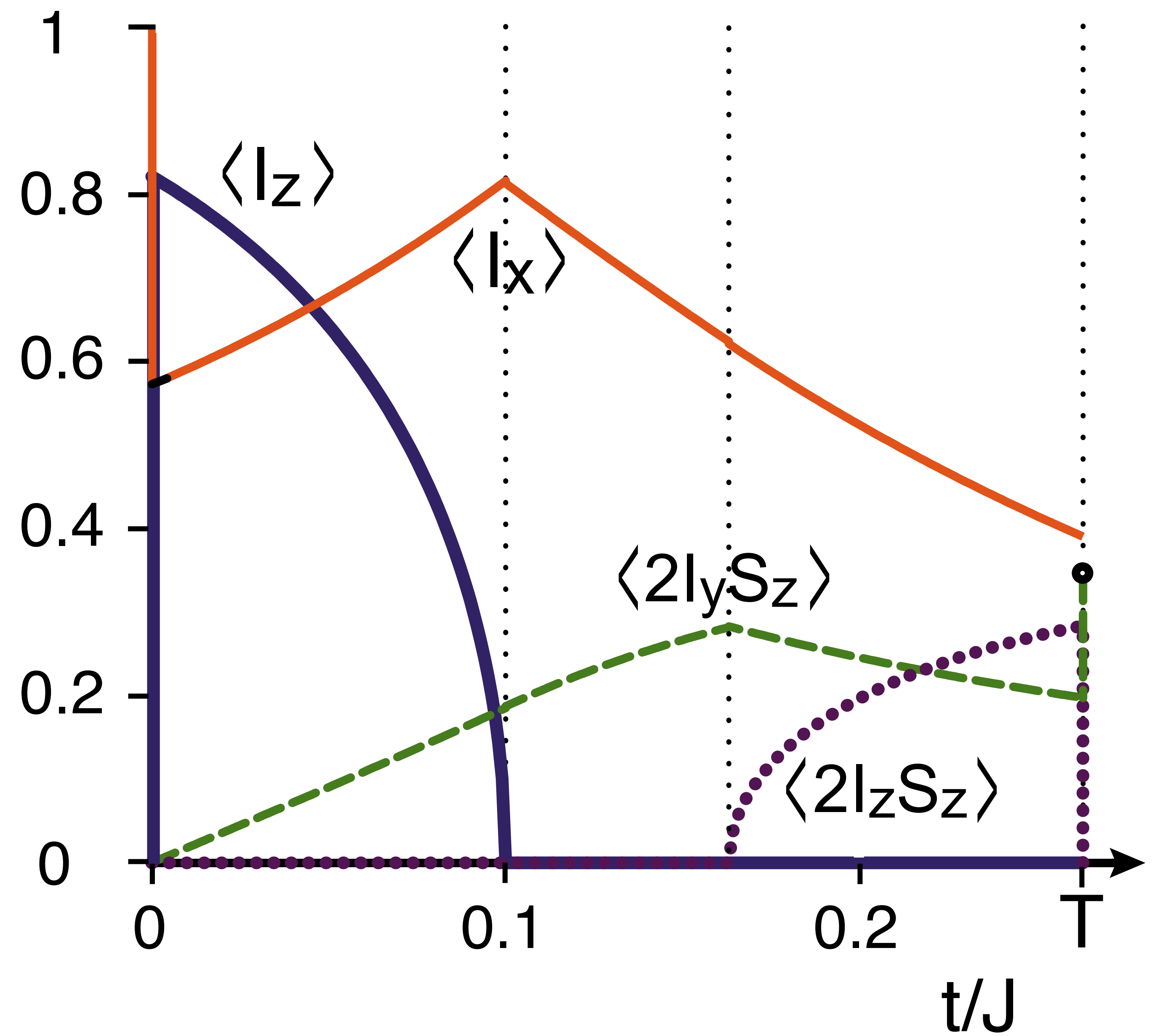


ROPE Pulse Sequence (finite time T)



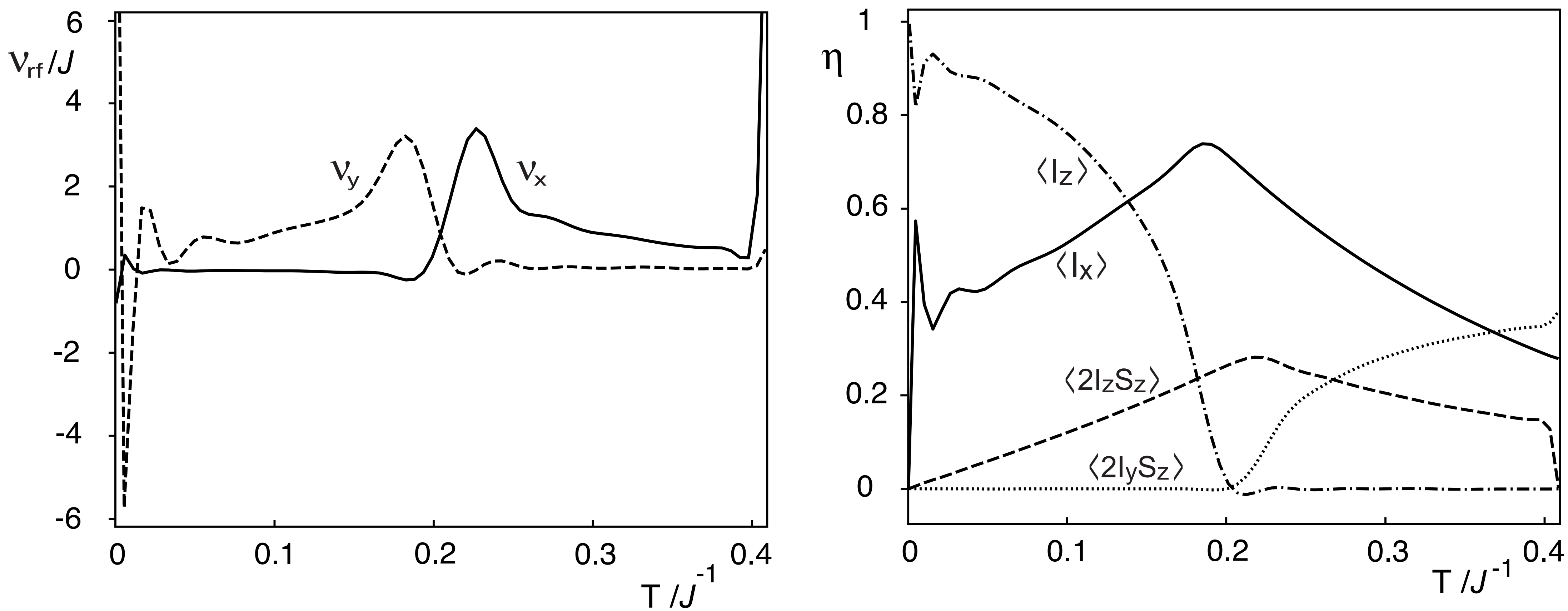
Khaneja et al., J. Magn. Reson.162, 311 (2003)

ROPE Trajectory (finite time T)

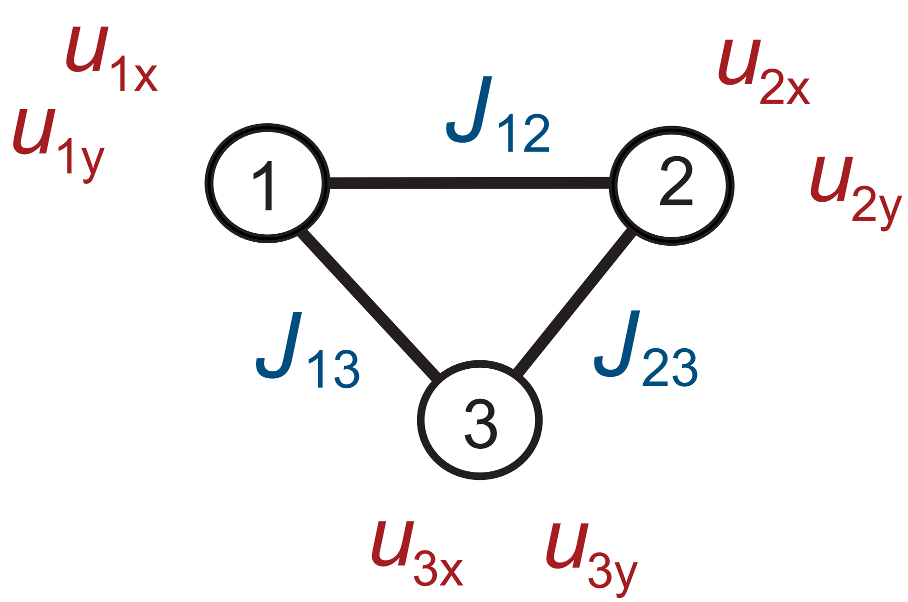


Khaneja, Reiss, Luy, Glaser (2003)

Numerically optimized ROPE sequences



Polarization transfer in homonuclear three spin systems



$$I_{1z} \longrightarrow I_{2z}$$

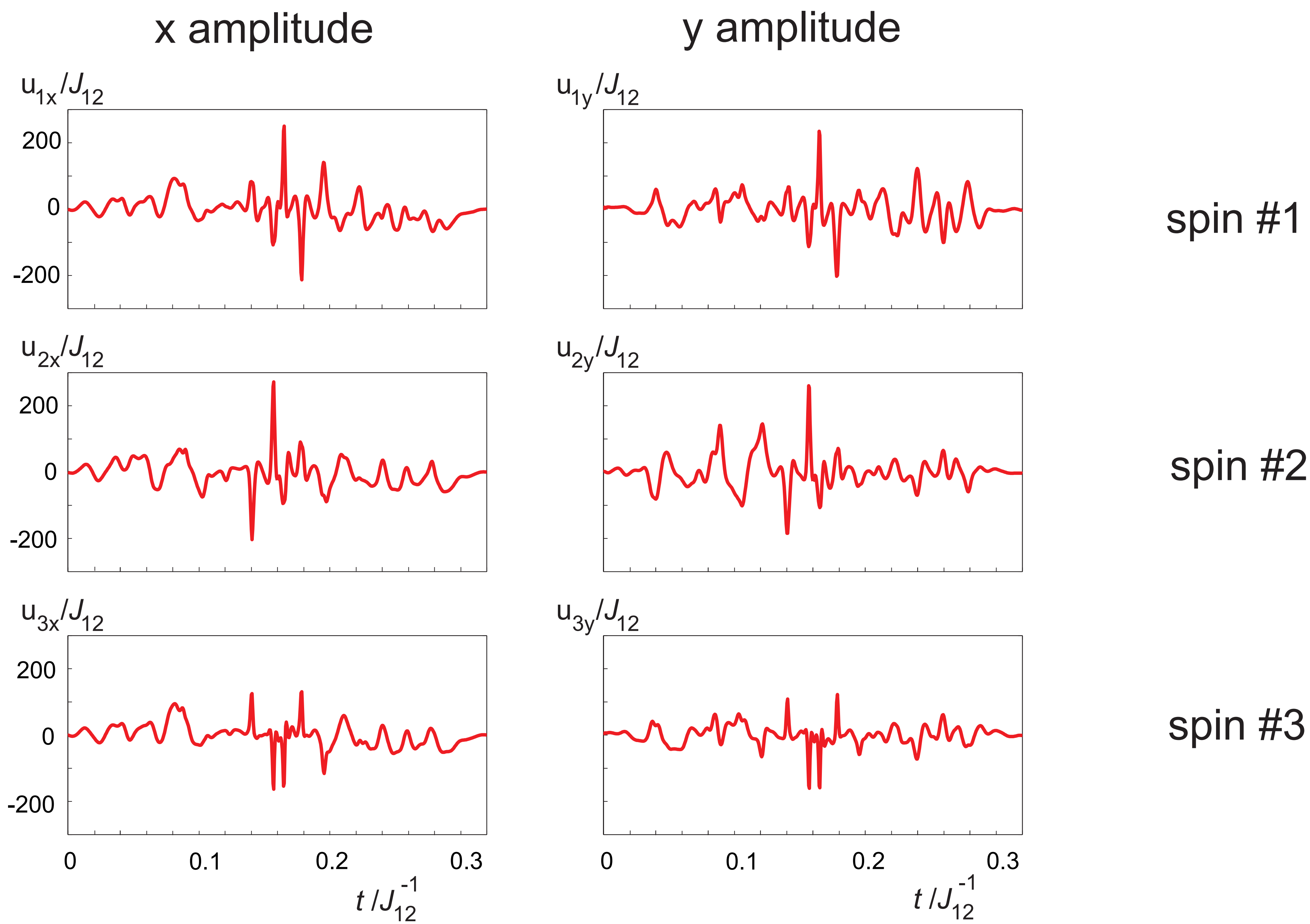
isotropic (Heisenberg) couplings

$$\sum_{m<n} 2\pi J_{mn}(I_{mx}I_{nx} + I_{my}I_{ny} + I_{mz}I_{nz})$$

idealized setting: fast, selective pulses (six control amplitudes)

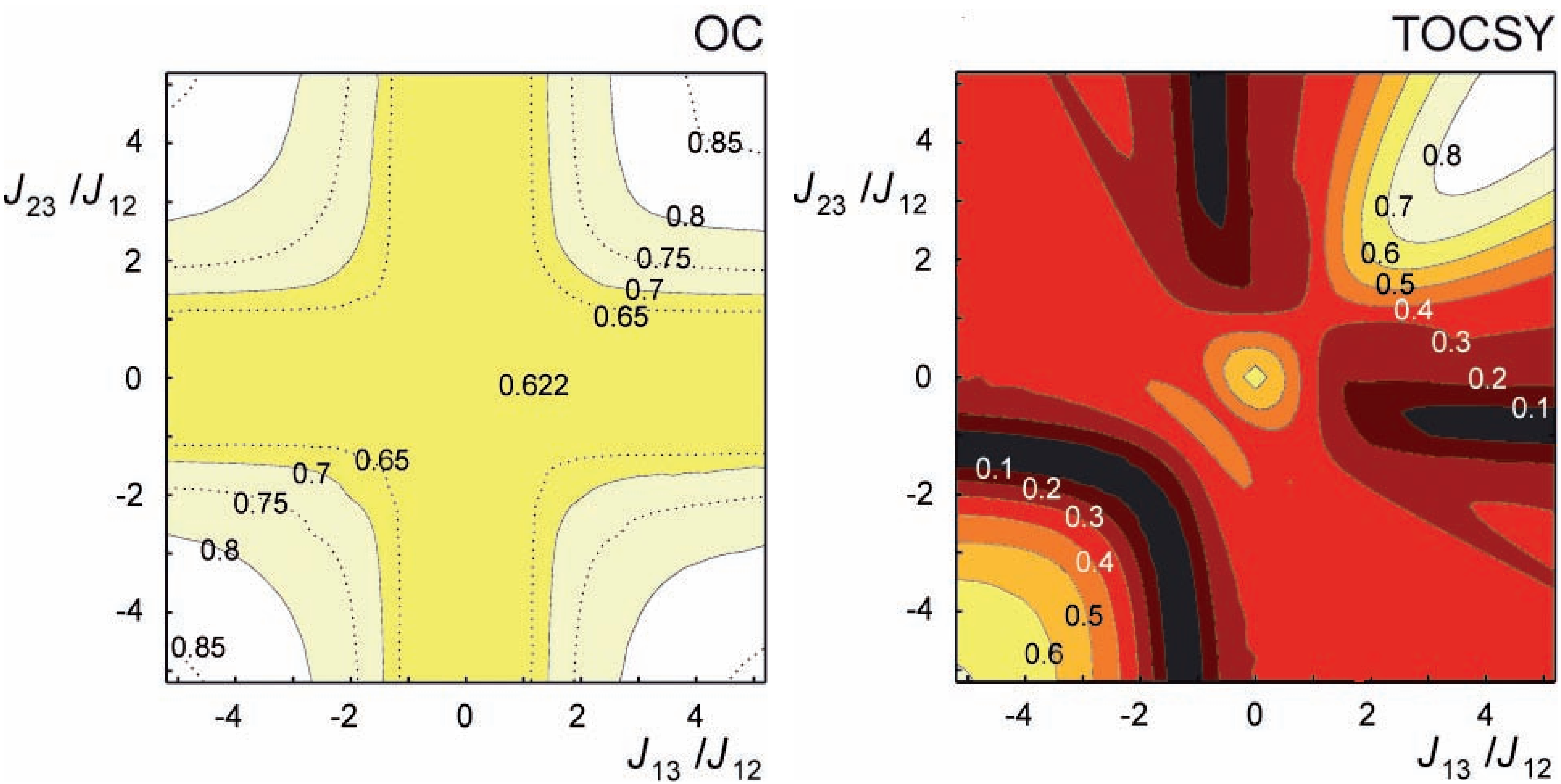
$$2\pi \sum_{m=1}^{3} \{u_{mx}(t)\; I_{mx} + u_{my}(t)\; I_{my}\}$$

Optimized controls (radio frequency amplitudes)

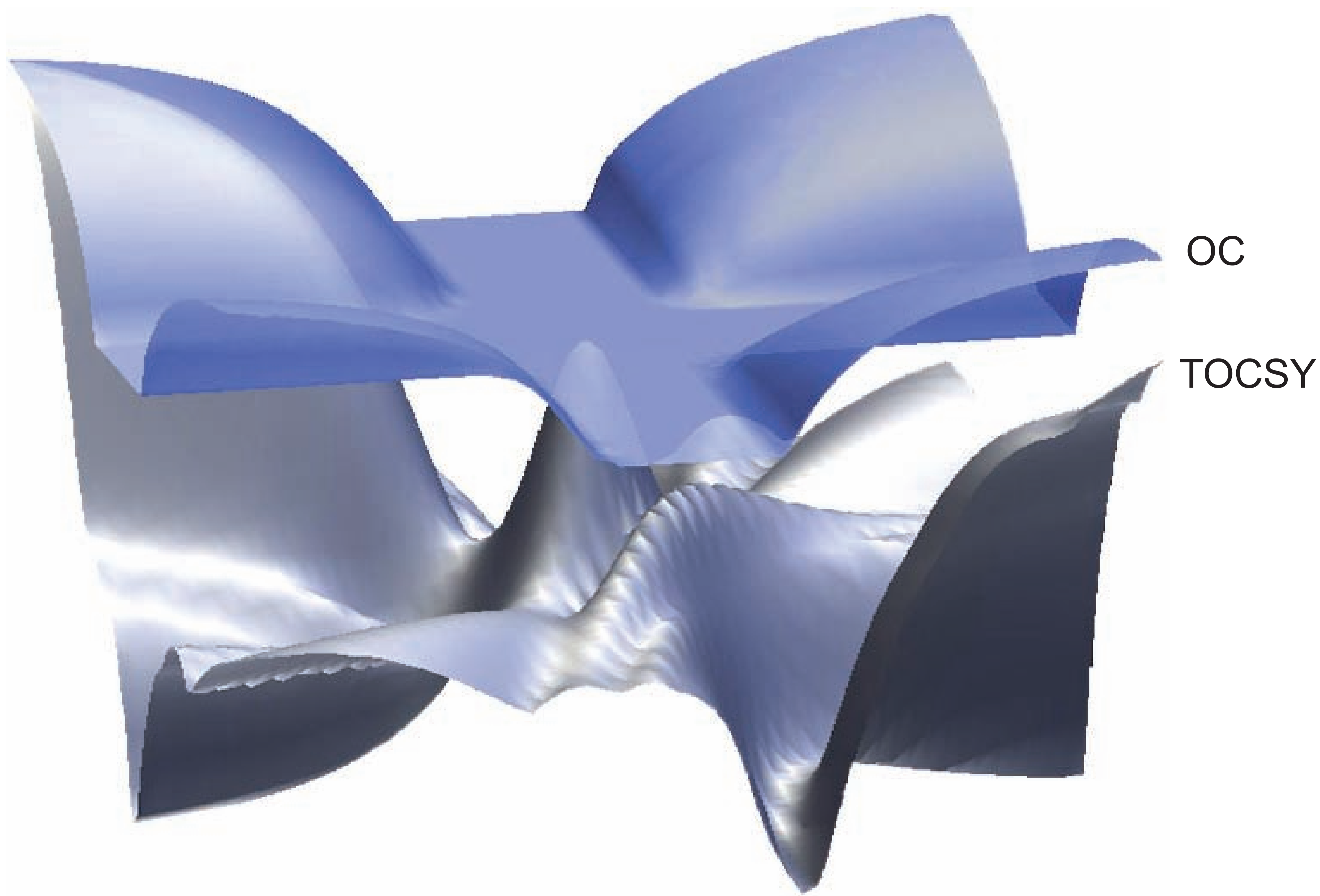


Neves et al. (2006)

Transfer efficiency as a function of relative coupling constants J_{13}/J_{12} and J_{23}/J_{12}



Transfer efficiency as a function of relative coupling constants J_{13}/J_{12} and J_{23}/J_{12}



Remark: The conventional (so-called TOCSY) experiment is equivalent to the transfer of energy between three coupled pendulums,

see

R. Marx, S. J. Glaser,
"Spins Swing Like Pendulums Do: An Exact Classical Model for TOCSY Transfer in Systems of Three Isotropically Coupled Spins 1/2",
J. Magn. Reson. 164, 338-342 (2003).

Polarization transfer in homonuclear three spin systems

isotropic (Heisenberg) couplings

$$\sum_{m<n} 2\pi J_{mn}(I_{mx} I_{nx} + I_{my} I_{ny} + I_{mz} I_{nz})$$

idealized control: fast, selective pulses (six control amplitudes)

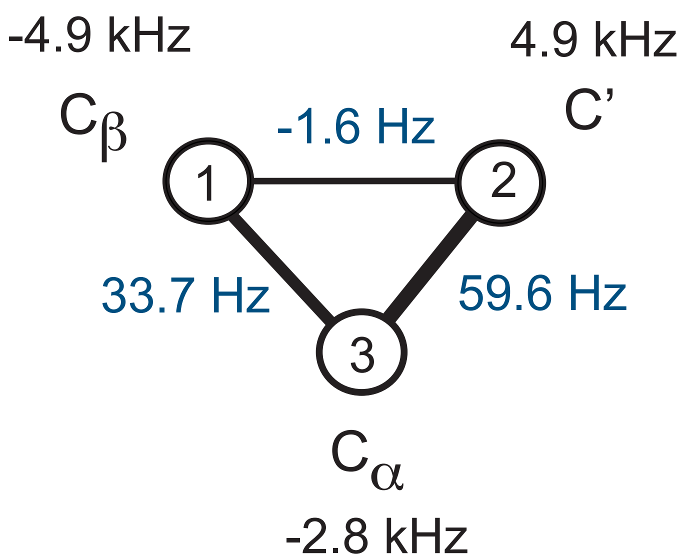
$$2\pi \sum_{m=1}^{3} \{u_{mx}(t)\, I_{mx} + u_{my}(t)\, I_{my}\}$$

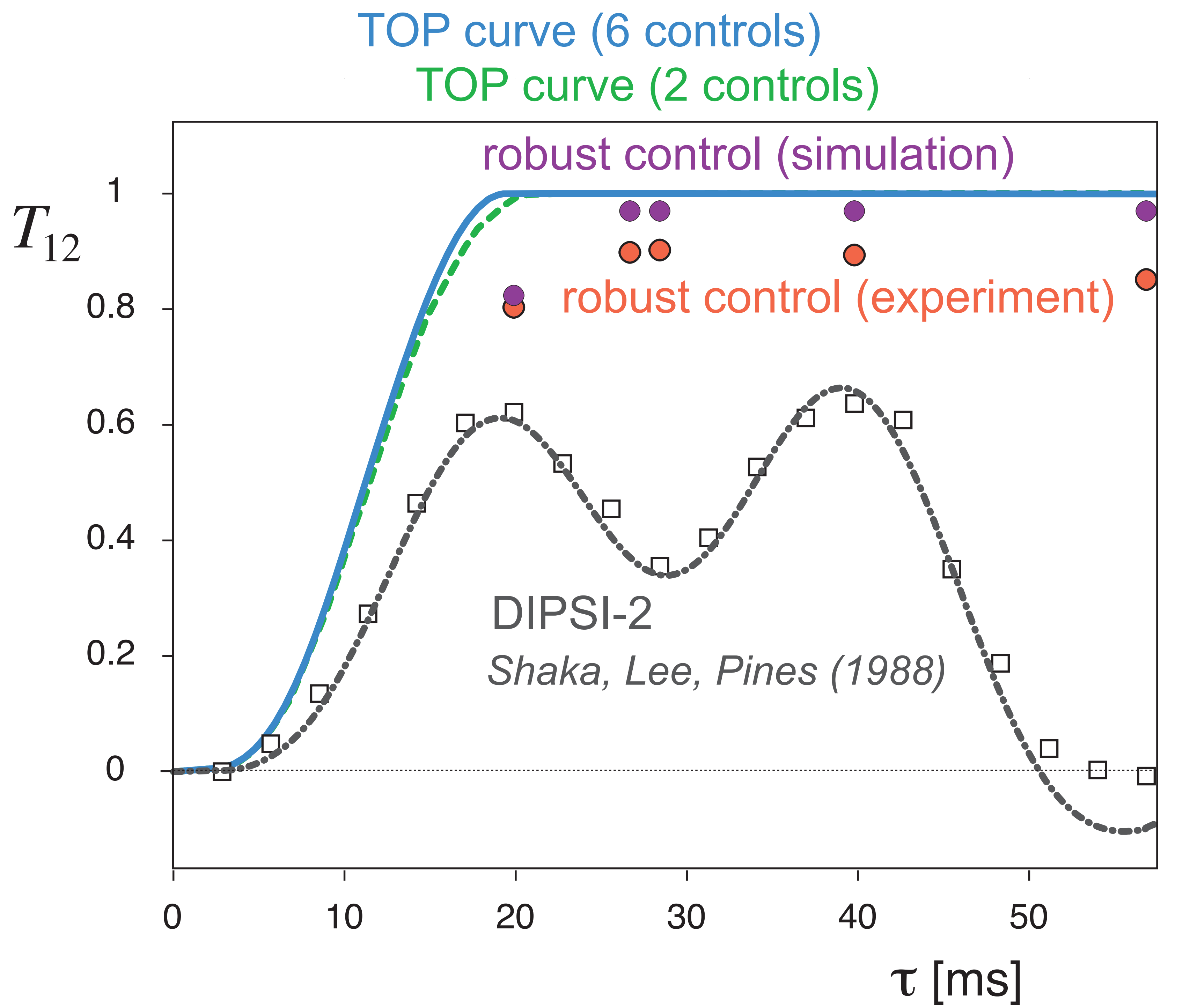
realistic control: non-selective pulses (two control amplitudes)

$$2\pi\, u_x(t) \sum_{m=1}^{3} I_{mx} + 2\pi\, u_y(t) \sum_{m=1}^{3} I_{my}$$

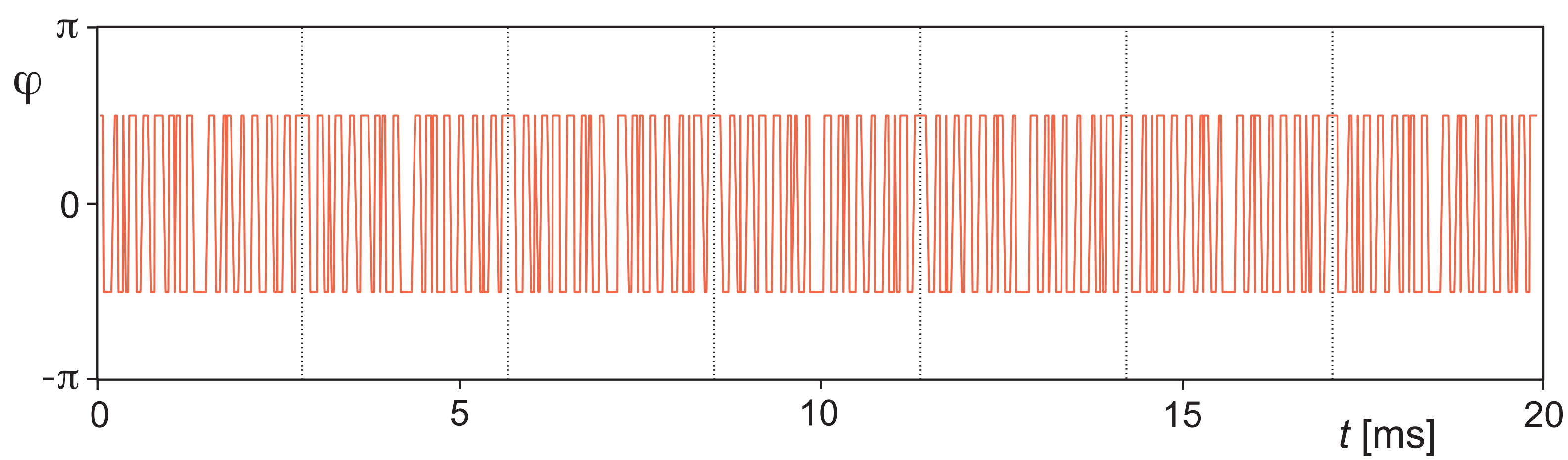
and (constant) offset terms $2\pi \sum_{m=1}^{3} \nu_{mz} I_{mz}$

Homonuclear three spin model system: ^{13}C labelled alanine



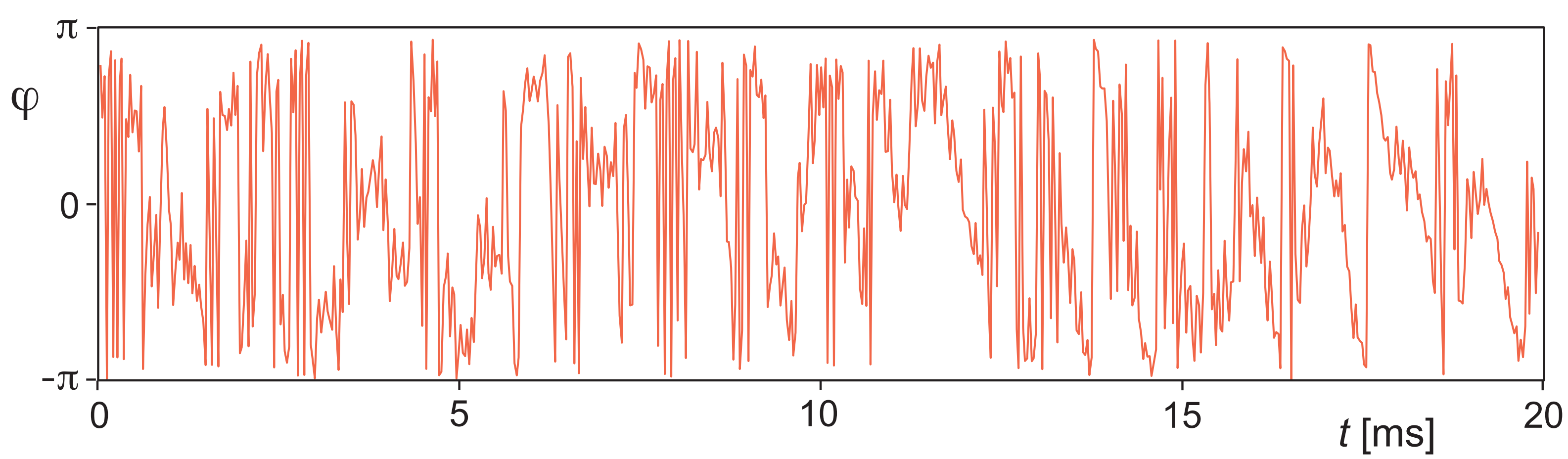


Conventional TOCSY sequence, e.g. DIPSI-2

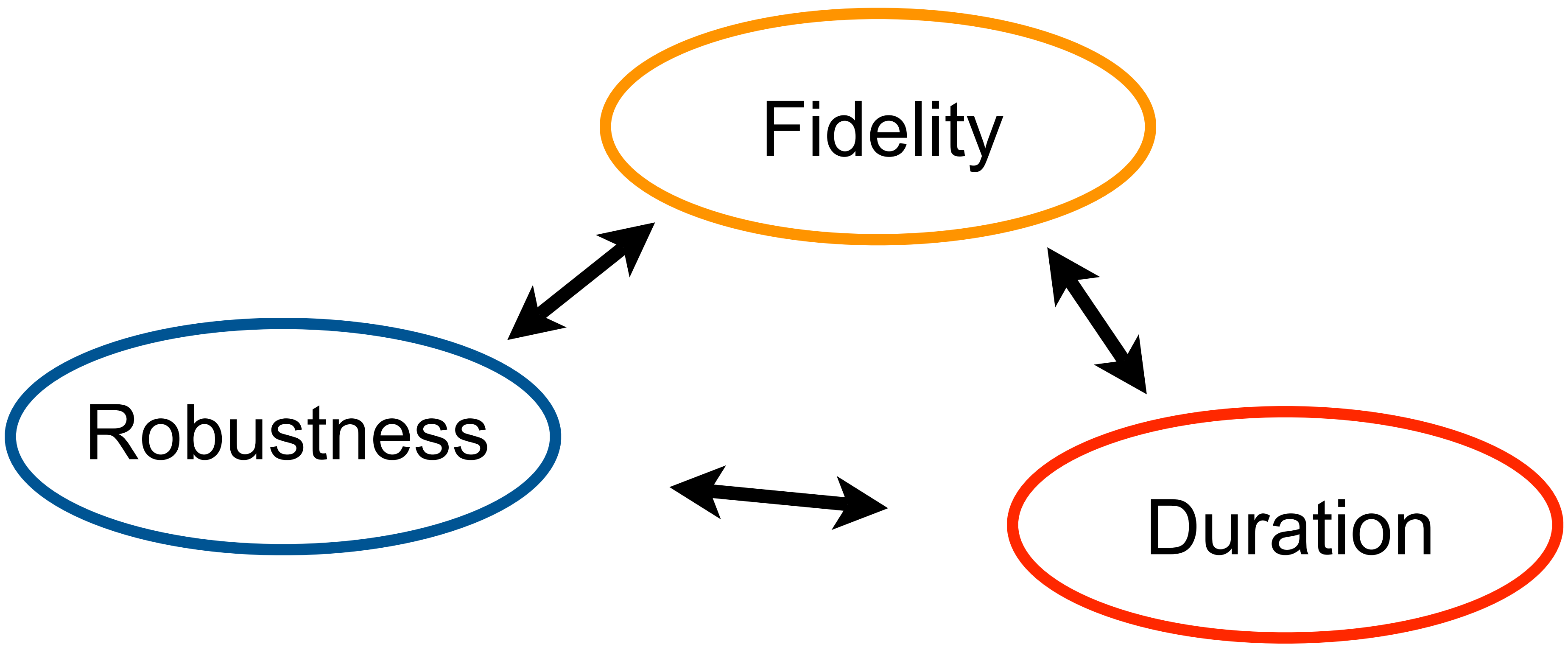


Shaka et al. (1988)

Robust phase-modulated polarization transfer sequences:



Neves et al. (2006)


Fidelity
Robustness
Duration

Low power heteronuclear decoupling

$$\mathcal{H}(t) = \mathcal{H}^{I}_{off} + \mathcal{H}^{S}_{off} + \mathcal{H}^{IS}_{J} + \mathcal{H}^{S}_{rf}(t)$$

$$\mathcal{H}^{I}_{off} = 2\pi\nu_I I_z$$

$$\mathcal{H}^{S}_{off} = 2\pi\nu_S S_z$$

$$\mathcal{H}^{IS}_{J} = 2\pi J S_z I_z$$

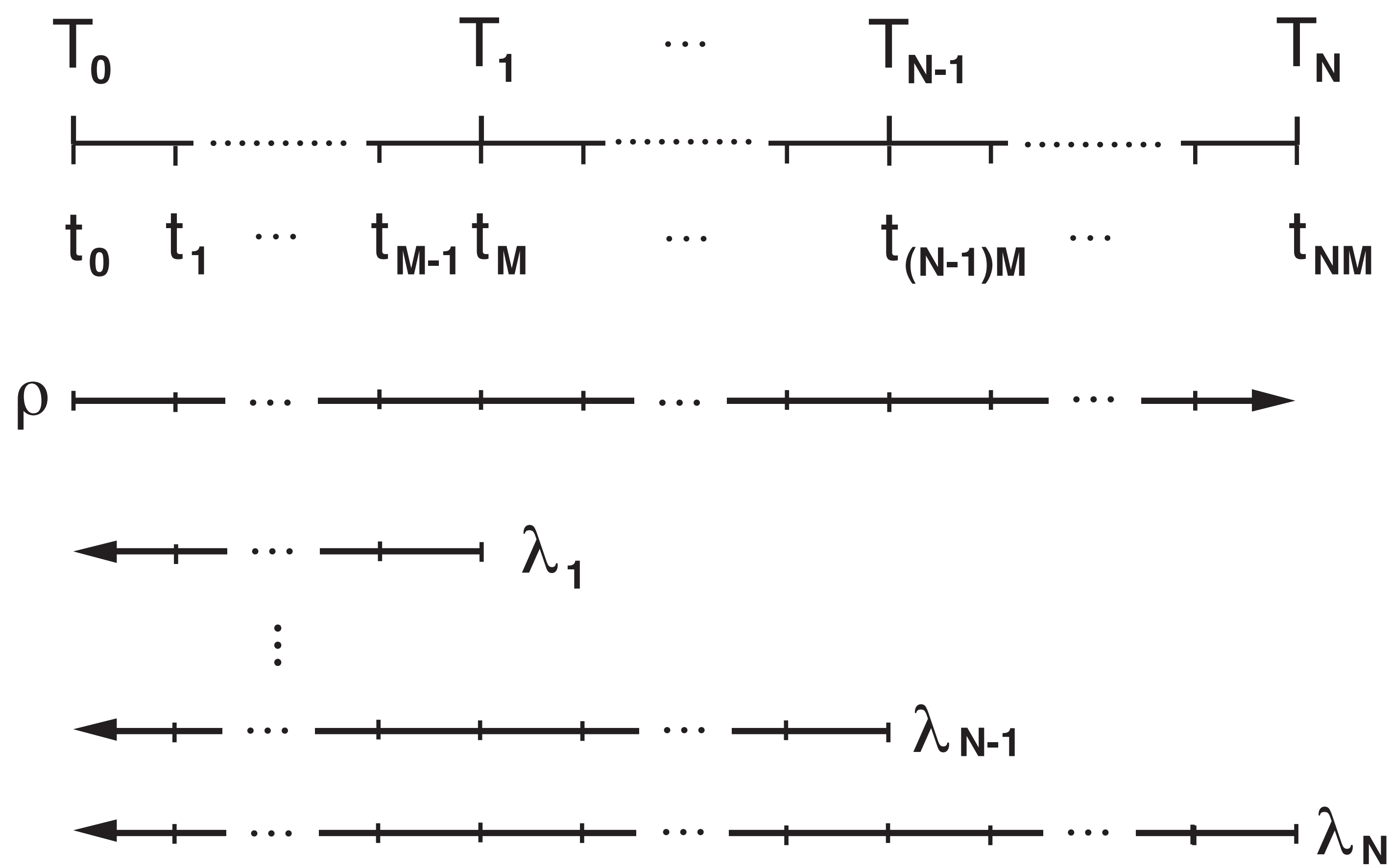
$$\mathcal{H}^{S}_{rf}(t) = 2\pi\ \epsilon\ \{u_x(t)S_x + u_y(t)S_y\}$$

J. L. Neves, B. Heitmann, N. Khaneja, S. J. Glaser, submitted (2009)

Low power heteronuclear decoupling

$$s_k = \langle I_x \rangle (T_k)$$

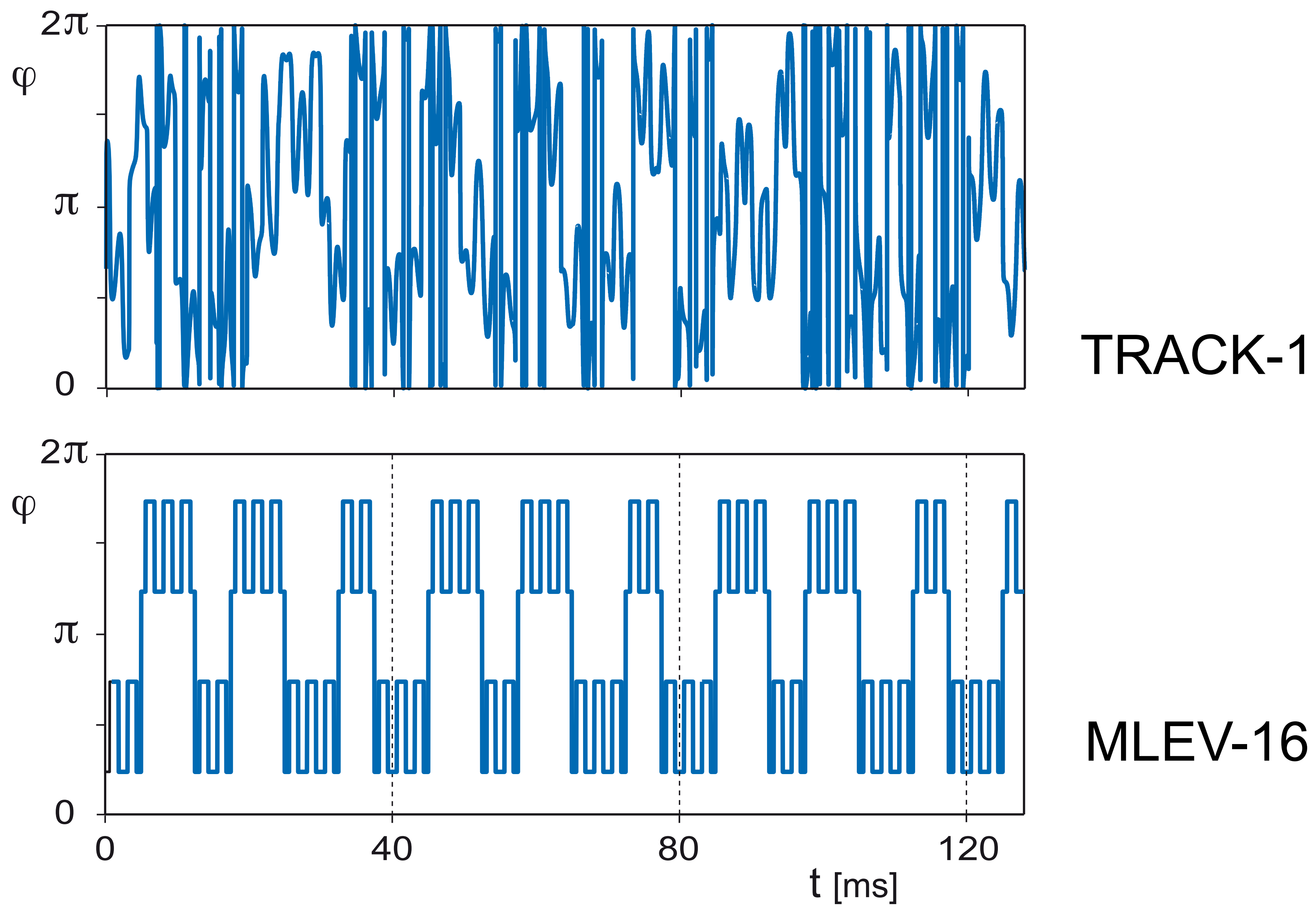
$$S_0(\epsilon, \nu_S) = \frac{1}{N+1} \sum_{k=0}^{N} s_k(\epsilon, \nu_S) \qquad \Phi = \frac{1}{N_\epsilon N_\nu} \sum_{p=1}^{N_\epsilon} \sum_{q=1}^{N_\nu} \phi(\epsilon^{(p)}, \nu_S^{(q)})$$



J. L. Neves, B. Heitmann, N. Khaneja, S. J. Glaser, submitted (2009)

Low power heteronuclear decoupling

nominal rf amplitude: 400 Hz



J. L. Neves, B. Heitmann, N. Khaneja, S. J. Glaser, submitted (2009)

Low power heteronuclear decoupling

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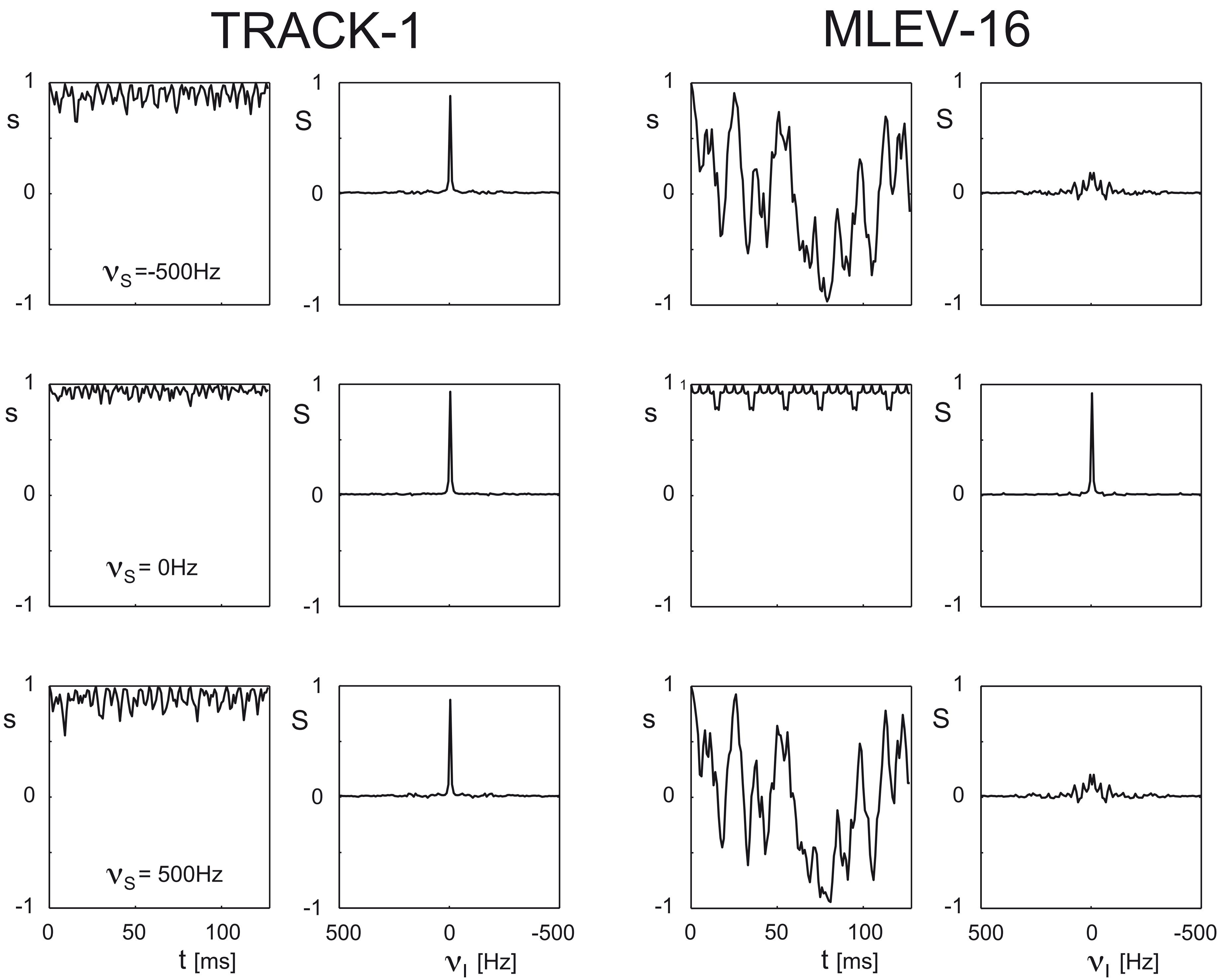
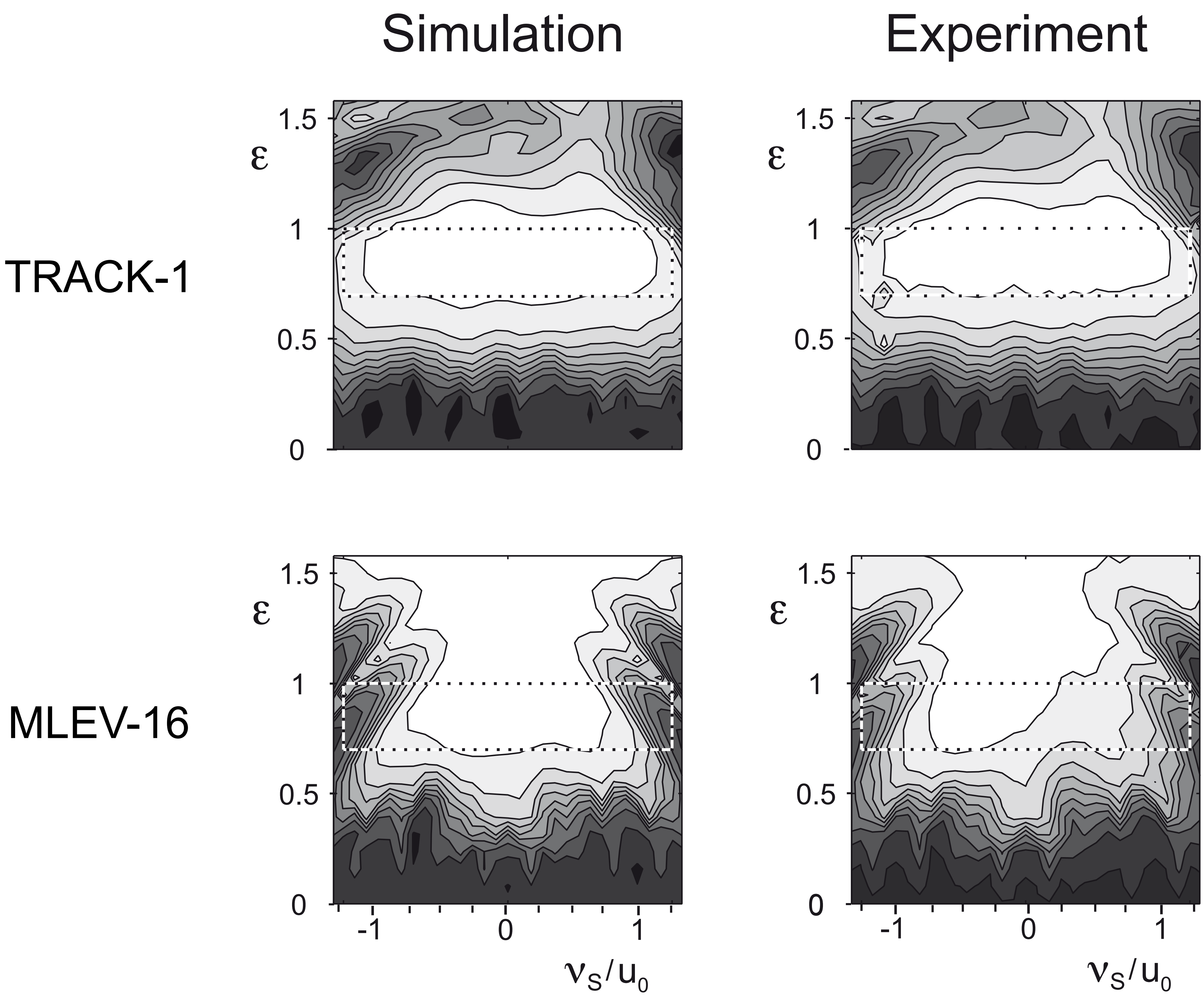


Low power heteronuclear decoupling



Low power heteronuclear decoupling

TRACK-1