Feedback control of NMR systems: a control-theoretic perspective
– PART 2 –

Claudio Altafini

SISSA-ISAS

International School for Advanced Studies

Trieste, Italy

This lecture

- Model for a 2 spin $1/2$ system
- State feedback control for a weakly coupled system
- Tracking an Hamiltonian different from the real one: suppression of unwanted weak couplings
- design open-loop controls based on “feedback on the simulator”
- examples on 3 and 4 spins

Product operators basis

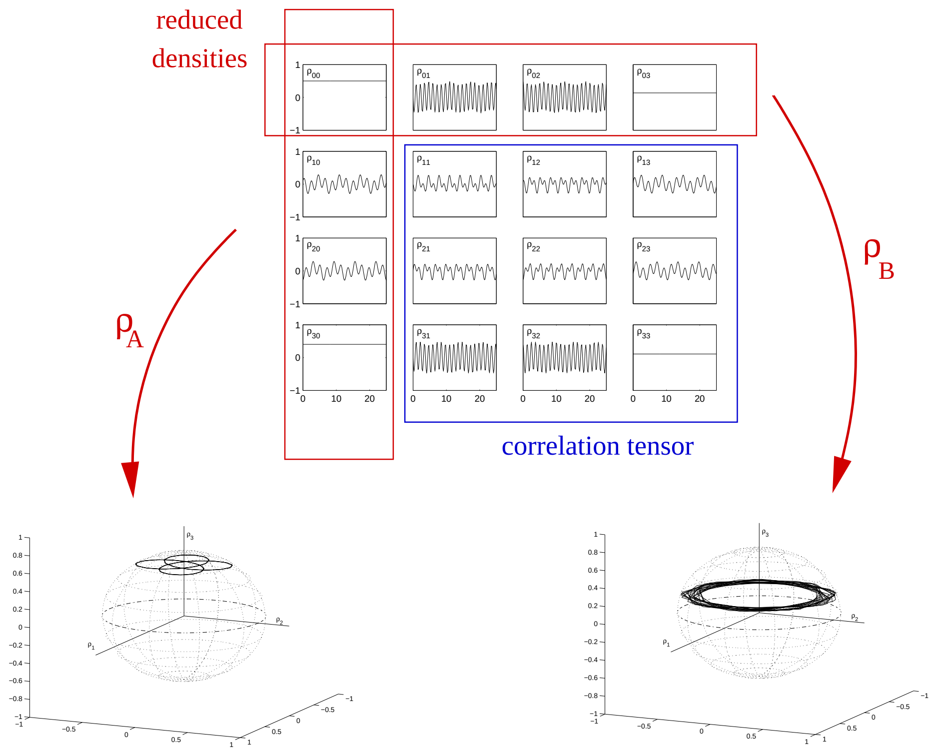
- (rescaled) identity + Pauli matrices

$$\lambda_0 = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}, \qquad \lambda_j = \frac{1}{\sqrt{2}} \sigma_j, \qquad j = 1, 2, 3$$

- *product operators* $\Lambda_{jk} = \lambda_j \otimes \lambda_k, \quad j, k = 0, \ldots, 3$:
  - 0 spin operators Λ_{00}
  - 1 spin operators $\Lambda_{01}, \Lambda_{02}, \Lambda_{03}, \Lambda_{10}, \Lambda_{20}, \Lambda_{30}$
  - 2 spin operators $\Lambda_{11}, \Lambda_{12}, \Lambda_{13}, \Lambda_{21}, \Lambda_{22}, \Lambda_{23}, \Lambda_{31}, \Lambda_{32}, \Lambda_{33}$
- basis for
  - density $\rho = \sum_{j,k=0}^{3} \varrho^{jk} \Lambda_{jk}$ where $\varrho^{jk} = \mathrm{tr}\left(\rho \Lambda_{jk}\right)$
  - Hamiltonian $H = h^{jl} \Lambda_{jk}$

Two spin $\frac{1}{2}$: density operator as a tensor

- state tensor $\boldsymbol{\varrho}$: 16 components
  - $\varrho^{00} = \mathrm{tr}\,(\rho\,\Lambda_{00})$ = const $\Longrightarrow$ trace component of ρ
  - $\{\varrho^{10},\ \varrho^{20},\ \varrho^{30}\}$ reduced density $\boldsymbol{\varrho}_A$
  - $\{\varrho^{01},\ \varrho^{02},\ \varrho^{03}\}$ reduced density $\boldsymbol{\varrho}_B$
  - $\{\varrho^{11},\ \varrho^{12}, \ldots, \varrho^{33}\}$ 2-body correlations



Two spin $\frac{1}{2}$: density operator as a tensor

- structure of the space of tensors ϱ^{jk}
  - ϱ^{jk} = elements of a "Liouville space"
  - $\boldsymbol{\varrho} = \{ \varrho^{jk} \}$ = Stokes tensor
  - $\varrho^{jk} \in \mathbb{R} \Longrightarrow \boldsymbol{\varrho} \in \mathcal{S} \subset \mathbb{S}^{14}$
  - 15 components
  - 6 independent degrees of freedom for pseudopure states
  - $\Longrightarrow$ structure of $\mathcal{S}$ includes several constraints (independent from the degree of purity)
  - $\Longrightarrow$ structure of $\mathcal{S}$ is complicated to "visualize"

Two spin $\frac{1}{2}$: density operator as a tensor

- $tr\Lambda_{jk}\Lambda_{lm} = \delta_{jk}\delta_{lm} \Longrightarrow \Lambda_{jk}$ form a complete orthonormal set
- trace norm $\Longrightarrow$ Euclidean norm in ϱ^{jk}-space

$$\mathrm{tr}\left(\rho^2\right) = \mathrm{tr}\left((\varrho^{jk}\Lambda_{jk})^2\right) = \sum_{j,k=0}^{3} (\varrho^{jk})^2 = \|\boldsymbol{\varrho}\|^2 \leqslant 1$$

- inner product

$$\mathrm{tr}\,(\rho_1\rho_2) = \langle\!\langle \boldsymbol{\varrho}_1, \boldsymbol{\varrho}_2 \rangle\!\rangle = \boldsymbol{\varrho}_1^T \boldsymbol{\varrho}_2$$

- distance function: assume $\|\boldsymbol{\varrho}_1\| = \|\boldsymbol{\varrho}_2\|$

$$d(\boldsymbol{\varrho}_1, \boldsymbol{\varrho}_2) = \|\boldsymbol{\varrho}_1\|^2 - \langle\!\langle \boldsymbol{\varrho}_1, \boldsymbol{\varrho}_2 \rangle\!\rangle = \|\boldsymbol{\varrho}_1\|^2 - \boldsymbol{\varrho}_1^T \boldsymbol{\varrho}_2.$$

Two spin $\frac{1}{2}$: Ising model

- free Hamiltonian
  - in the lab frame

$$H_f = \omega_{o,\alpha}\Lambda_{03} + \omega_{o,\beta}\Lambda_{30} + h^{33}\Lambda_{33}$$

  - $\omega_{o,\alpha},\ \omega_{o,\beta} =$ Larmor frequencies of the single spins ($\simeq$ MHz)
  - $h^{33} =$ scalar coupling ($\simeq$ hundreds of Hz)
  - if spins are homonuclear (gyromagnetic ratios $\gamma_\alpha = \gamma_\beta$) $\Longrightarrow$ $\omega_{o,\alpha}$ and $\omega_{o,\beta}$ differ only because of the chemical shift
  - if spins are heteronuclear: difference between $\omega_{o,\alpha}$ and $\omega_{o,\beta}$ can be of many MHz

Two spin $\frac{1}{2}$: Ising model

- control Hamiltonian
  1. when $\omega_{o,\alpha} \simeq \omega_{o,\beta} \Longrightarrow$ spins are not selectively excitable: r.f. field resonating with $\omega_{o,\alpha}$ will cross-talk with the other spin $\Longrightarrow$ one single control field
  2. when difference between $\omega_{o,\alpha}$ and $\omega_{o,\beta}$ is high $\Longrightarrow$ spins are selectively excitable $\Longrightarrow$ 2 distinct control fields tuned at $\omega_{o,\alpha}$ and $\omega_{o,\beta}$

  - in the lab frame

$$H_{\rm rf} = -B_1 \left(\cos(\omega_{\rm rf} t + \phi) \left(\gamma_\alpha \Lambda_{10} + \gamma_\beta \Lambda_{01}\right) + \sin(\omega_{\rm rf} t + \phi) \left(\gamma_\alpha \Lambda_{20} + \gamma_\beta \Lambda_{02}\right)\right.$$

  - in the rotating frame, with $\phi = 0$

$$H_{\rm c} = -B_1 \left(\gamma_\alpha \Lambda_{10} + \gamma_\beta \Lambda_{01}\right)$$

Two spin $\frac{1}{2}$: Ising model

Case 1: *nonselective control*

- in a "single" rotating frame

$$
\begin{aligned}
H_{\mathrm{f}} &= h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{33}\Lambda_{33} \\
H_{\mathrm{c}} &= u\left(\Lambda_{01} + \Lambda_{10}\right)
\end{aligned}
$$

- $h^{30} = -(\omega_{o,\alpha} - \omega_{\mathrm{rf}}), \quad h^{03} = -(\omega_{o,\beta} - \omega_{\mathrm{rf}})$
- $u = -\gamma_\alpha B_1 = -\gamma_\beta B_1$

Case 2: *selective control*

- in a "doubly rotating" frame

$$
\begin{aligned}
H_{\mathrm{f}} &= h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{33}\Lambda_{33} \\
H_{\mathrm{c}} &= u_{01}\Lambda_{01} + u_{10}\Lambda_{10}
\end{aligned}
$$

- $h^{30} = -(\omega_{o,\alpha} - \omega_{\mathrm{rf},\alpha}), \quad h^{03} = -(\omega_{o,\beta} - \omega_{\mathrm{rf},\beta})$
- $u_{10} = -\gamma_\alpha B_{1,\alpha}, \quad u_{01} = -\gamma_\beta B_{1,\beta}$

Two spin $\frac{1}{2}$: Lie algebra structure

- "local" Lie algebra:

$$\mathfrak{su}(2) \oplus \mathfrak{su}(2) = \mathrm{span}\{-i\Lambda_{j0},\ -i\Lambda_{0k}\}$$

- "nonlocal" Lie algebra:

$$\mathfrak{su}(2) \otimes \mathfrak{su}(2) = \mathrm{span}\{-i\Lambda_{jk},\ j,k \neq 0\}$$

- "total" Lie algebra:

$$\mathfrak{g}_{2s} = \mathrm{Lie}\{-i\Lambda_{jk},\ j,k = 0,\ldots,3\} = \mathfrak{su}(2) \oplus \mathfrak{su}(2) \cup \mathfrak{su}(2) \otimes \mathfrak{su}(2)$$

- $\dim(\mathfrak{g}_{2s}) = 3 + 3 + 9 = 15$

Two spin $\frac{1}{2}$: controllability

case 1: nonselective control

- intuitively:
  - nonselective case: control field is the same for both spins
  - coupling is only of the "z-z" type $\Longrightarrow$ symmetric
  - in order to have controllability I need to "break the symmetry" by means of the local Larmor precessions
  - $\Longrightarrow$ there must be some chemical shift $h^{03} \neq h^{30}$
- *Lie algebraic rank condition* (LARC): if $\mathrm{Lie}\{-iH_\mathrm{f},\,-iH_\mathrm{c}\} = \mathfrak{su}(2)\oplus\mathfrak{su}(2)\cup\mathfrak{su}(2)\otimes\mathfrak{su}(2) \;\Longrightarrow$ the system $\dot\rho = -i[H_\mathrm{f} + uH_\mathrm{c},\ \rho]$ is controllable
- controllability depends on the rotating frame chosen: consequence of the lack of small-time controllability
- check the LARC means compute exhaustively all the commutators $[-iH_f,\,-iH_c]$, $[-iH_f,\,[-iH_f,\,-iH_c]\,]$, $[-iH_c,\,[-iH_f,\,-iH_c]\,]$
- .... long procedure also for 2 spin systems
- last time: sufficient condition for controllability in terms of energy levels

Two spin $\frac{1}{2}$: controllability

- free Hamiltonian

$$H_f = \begin{bmatrix} h^{03}+h^{30}+h^{33} & & & \\ & -h^{03}+h^{30}-h^{33} & & \\ & & h^{03}-h^{30}-h^{33} & \\ & & & -h^{03}-h^{30}+h^{33} \end{bmatrix}$$

- control Hamiltonian

$$H_c = \begin{bmatrix} 0 & 1 & 1 & 0 \\ 1 & 0 & 0 & 1 \\ 1 & 0 & 0 & 1 \\ 0 & 1 & 1 & 0 \end{bmatrix}$$

- H_c is enabling the following transitions

$$1 \longleftrightarrow 2, \quad 1 \longleftrightarrow 3, \quad 2 \longleftrightarrow 4, \quad 3 \longleftrightarrow 4$$

- $\Longrightarrow \mathrm{Graph}(H_c)$ is connected

Two spin $\frac{1}{2}$: controllability

- sufficient condition for controllability: H_f is H_c strongly regular, meaning
  - energy levels of H_f are nondegenerate
  - energy levels of H_f are not equispaced along the transitions enabled by H_c

$$\text{i.e.,} \quad \begin{cases} h^{03} \neq h^{30} \\ h^{33} \neq \pm(h^{03} - h^{30})/2 \end{cases}$$

- when $h^{03} = h^{30}$, H_f has a degenerate energy level (of multiplicity 2)
- $\Longrightarrow$ sufficient conditions for controllability do not apply
- $\Longrightarrow$ system may be noncontrollable

Two spin $\frac{1}{2}$: controllability

case 2: selective control

- 2 control degrees of freedom u_{01} and u_{10}
- $\Longrightarrow$ LARC is always verified
- $\Longrightarrow$ the system

$$\dot{\rho} = -i[H_{\mathrm{f}} + u_{01}\Lambda_{01} + u_{10}\Lambda_{10},\ \rho]$$

  is always controllable

- you also have small time controllability $\Longrightarrow$ possibility to “kill” the drift

Two spin $\frac{1}{2}$: adjoint representation

- commutator of $A_1 \otimes A_2$ and $B_1 \otimes B_2$

$$[A_1 \otimes A_2,\, B_1 \otimes B_2] = \frac{1}{2}\Big([A_1,\, B_1] \otimes \{A_2,\, B_2\} + \{A_1,\, B_1\} \otimes [A_2,\, B_2]\Big)$$

- commutator for basis elements

$$[\Lambda_{jk},\, \Lambda_{lm}] = \frac{1}{2}\left([\lambda_j,\, \lambda_l] \otimes \{\lambda_k,\, \lambda_m\} + \{\lambda_j,\, \lambda_l\} \otimes [\lambda_k,\, \lambda_m]\right)$$

- want to write it as “linear” operator

$$[\Lambda_{jk},\, \Lambda_{lm}] = \mathrm{ad}_{\Lambda_{jk}} \Lambda_{lm}$$

- need to compute the structure constants (both symmetric and skew-symmetric ones)

Two spin $\frac{1}{2}$: adjoint representation

- skew-symmetric structure constants

$$[\lambda_j,\, \lambda_k] = \mathrm{ad}_{\lambda_j}\lambda_k = \sum_{l=0}^{3} c_{jk}^{l}\lambda_l$$

$$\mathrm{ad}_{\lambda_0} = 0, \qquad \mathrm{ad}_{\lambda_1} = i\left[\begin{array}{c|ccc} 0 & 0 & 0 & 0 \\ \hline 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{array}\right]$$

$$\mathrm{ad}_{\lambda_2} = i\left[\begin{array}{c|ccc} 0 & 0 & 0 & 0 \\ \hline 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{array}\right], \qquad \mathrm{ad}_{\lambda_3} = i\left[\begin{array}{c|ccc} 0 & 0 & 0 & 0 \\ \hline 0 & 0 & -1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{array}\right]$$

Two spin $\frac{1}{2}$: adjoint representation

- symmetric structure constants

$$\{\lambda_j, \lambda_k\} = \mathrm{aad}_{\lambda_j} \lambda_k = \sum_{l=0}^{3} s_{jk}^{l} \lambda_l$$

$$\mathrm{aad}_{\lambda_0} = \left[\begin{array}{c|ccc} 1 & 0 & 0 & 0 \\ \hline 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{array}\right], \qquad \mathrm{aad}_{\lambda_1} = \left[\begin{array}{c|ccc} 0 & 1 & 0 & 0 \\ \hline 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{array}\right]$$

$$\mathrm{aad}_{\lambda_2} = \left[\begin{array}{c|ccc} 0 & 0 & 1 & 0 \\ \hline 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{array}\right], \qquad \mathrm{aad}_{\lambda_3} = \left[\begin{array}{c|ccc} 0 & 0 & 0 & 1 \\ \hline 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{array}\right]$$

Two spin $\frac{1}{2}$: adjoint representation

- Lie bracket

$$
\begin{aligned}
[\Lambda_{jk}, \Lambda_{lm}] &= \frac{1}{2}\big([\lambda_j, \lambda_l] \otimes \{\lambda_k, \lambda_m\} + \{\lambda_j, \lambda_l\} \otimes [\lambda_k, \lambda_m]\big) \\
&= \frac{1}{2}\big(\mathrm{ad}_{\lambda_j}\lambda_l \otimes \mathrm{aad}_{\lambda_k}\lambda_m + \mathrm{aad}_{\lambda_j}\lambda_l \otimes \mathrm{ad}_{\lambda_k}\lambda_m\big) \\
&= \frac{1}{2}\big(\mathrm{ad}_{\lambda_j} \otimes \mathrm{aad}_{\lambda_k} + \mathrm{aad}_{\lambda_j} \otimes \mathrm{ad}_{\lambda_k}\big)\, \lambda_l \otimes \lambda_m \\
&= \mathrm{ad}_{\Lambda_{jk}}\Lambda_{lm}
\end{aligned}
$$

- adjoint operators $\mathrm{ad}_{\Lambda_{jk}} \longrightarrow$ infinitesimal superoperators

$$
\mathrm{ad}_{\Lambda_{jk}} = \frac{1}{2}\big(\mathrm{ad}_{\lambda_j} \otimes \mathrm{aad}_{\lambda_k} + \mathrm{aad}_{\lambda_j} \otimes \mathrm{ad}_{\lambda_k}\big)
$$

- 16×16 skew-symmetric matrices

Two spin $\frac{1}{2}$: Lie algebra of "unitary" superoperators

- "local" adjoint Lie algebra

$$\mathrm{ad}_{\mathfrak{su}(2)} \oplus \mathrm{ad}_{\mathfrak{su}(2)} = \mathfrak{so}(3) \oplus \mathfrak{so}(3) = \mathrm{span}\{-i\mathrm{ad}_{\Lambda_{j0}},\ -i\mathrm{ad}_{\Lambda_{0k}}\}$$

- "nonlocal" adjoint Lie algebra

$$\mathrm{ad}_{\mathfrak{su}(2)} \otimes \mathrm{ad}_{\mathfrak{su}(2)} = \mathfrak{so}(3) \otimes \mathfrak{so}(3) = \mathrm{span}\{-i\mathrm{ad}_{\Lambda_{jk}},\ j,k \neq 0\}$$

- "total" Lie algebra:

$$\mathrm{ad}_{\mathfrak{g}_{2s}} = \mathrm{Lie}\{-i\mathrm{ad}_{\Lambda_{jk}},\ j,k = 0,\ldots,3\} = \mathfrak{so}(3) \oplus \mathfrak{so}(3) \cup \mathfrak{so}(3) \otimes \mathfrak{so}(3)$$

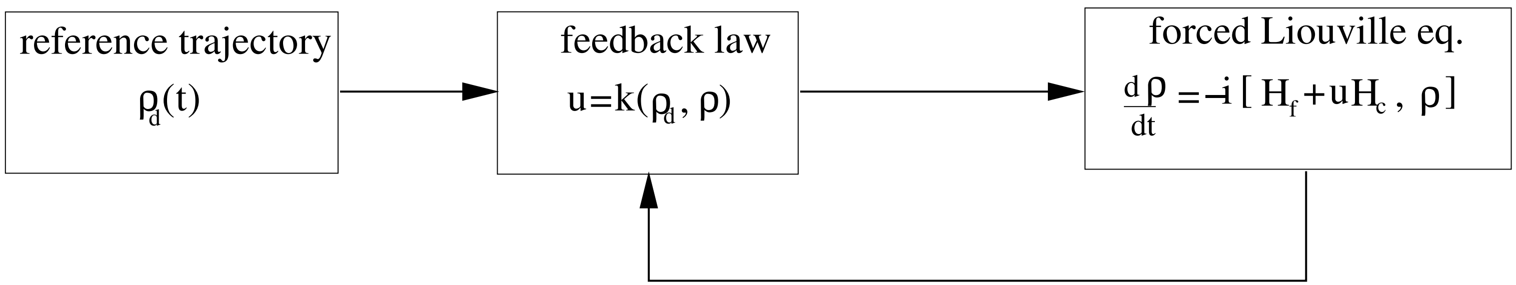
- $\dim(\mathrm{ad}_{\mathfrak{g}_{2s}}) = 3 + 3 + 9 = 15$

State feedback stabilization

Assumptions: state feedback stabilization problem:

- the entire state ϱ is available on-line
- nonselective case: only one control degree of freedom
- desired state to track ϱ_d is a pseudopure state
- ρ and ρ_d have the same eigenvalues

Scheme:



Feedback problem formulation

- given "reference density" $\varrho_d(t)$
- "reference evolution" given by H_{f_d}

$$\dot{\varrho}_d = -i \mathrm{ad}_{H_{f_d}} \varrho_d$$

- want that the "true" evolution

$$\dot{\varrho} = -i \left(\mathrm{ad}_{H_f} + u \, \mathrm{ad}_{H_c} \right) \varrho$$

tracks the reference state determined by $\varrho_d(t)$

$$\varrho \xrightarrow{t \to \infty} \varrho_d$$

  - full state stabilization

Jurdjevic-Quinn sufficient condition for stabilization

- given a bilinear control system, if the so-called “ad-brackets” generate the entire Lie algebra, then $\exists$ a Lyapunov based feedback design $\longrightarrow$ global stabilization
- $\Longrightarrow$ automatically answers the problem of convergence (LaSalle invariance principle)
- it is never the case for manifolds with nontrivial topology

$$\mathrm{span}\left\{-i\mathrm{ad}_{H_f},\, -i\mathrm{ad}_{H_c},\, [-i\mathrm{ad}_{H_f},\, -i\mathrm{ad}_{H_c}], [-i\mathrm{ad}_{H_f},\, [-i\mathrm{ad}_{H_f},\, -i\mathrm{ad}_{H_c}]], \ldots, \right\} \neq \mathrm{ad}_{\mathfrak{g}}$$

- to show it: compute the first commutators and verify that the basis directions $-i\Lambda_{11}$, $-i\Lambda_{12}$, $-i\Lambda_{21}$ and $-i\Lambda_{22}$ are never touched by such commutators
- stabilization design cannot be global!
- $\longrightarrow$ nontrivial singular locus
- $\longrightarrow$ in general: difficult to find what is the region of attraction

State tracking

Proposition *When* $h^{03} \neq h^{30}$ *and* $H_{f_d} = H_f$ *, the feedback law*

$$u = k\langle\langle \boldsymbol{\varrho}_d, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle, \qquad k > 0$$

asymptotically stabilizes the system

$$\dot{\boldsymbol{\varrho}} = -i\left(\mathrm{ad}_{H_f} + u\,\mathrm{ad}_{H_c}\right)\boldsymbol{\varrho}$$

to the reference state $\boldsymbol{\varrho}_d(t)$ *for all* $\boldsymbol{\varrho}(0)$ *except for the following initial conditions*

1. *antipodal point of the reduced densities* $(\boldsymbol{\varrho}_A(0), \boldsymbol{\varrho}_B(0)) = -(\boldsymbol{\varrho}_{A_\mathrm{d}}(0), \boldsymbol{\varrho}_{B_\mathrm{d}}(0))$
2. *horizontal great circles of the reduced densities* $(\varrho_A^3, \varrho_{A_d}^3) = (0, 0)$ *and* $(\varrho_B^3, \varrho_{B_d}^3) = (0, 0)$

- singular locus is the “replica” of the 1 spin 1/2 case
- k = feedback gain = parameter to tune

State tracking

Sketch of the proof

- take Lyapunov function as before

$$V(t) = \|\boldsymbol{\varrho}_d\|^2 - \langle\langle \boldsymbol{\varrho}_d(t),\, \boldsymbol{\varrho}(t)\rangle\rangle \; > 0$$

- differentiate V
- $H_{f_d} = H_f \implies$ drift disappears
- $\dot{V} = -u\langle\langle \boldsymbol{\varrho}_d,\, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle$
- with $u = k\langle\langle \boldsymbol{\varrho}_d,\, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle \implies \dot{V} = -k\langle\langle \boldsymbol{\varrho}_d,\, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle^2 \leqslant 0$
- LaSalle invariance principle: want to find closed-loop trajectories that are in $\mathcal{N} = \{\boldsymbol{\varrho}$ s.t. $\dot{V} = 0\}$
- $h^{03} \neq h^{30}$
    - $\implies$ local dynamics must be distinguishable
    - $\implies u = 0$ cannot belong to $\mathcal{N}$, except for the singular points

Jurdjevic-Quinn sufficient condition for stabilization

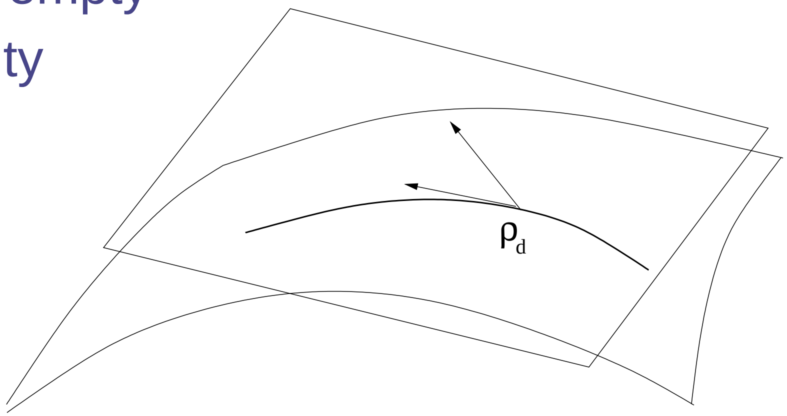
- meaning of the Jurdjevic-Quinn condition:
  - take the linearization around the reference trajectory ϱ_d

$$\dot{\varrho} = -i\mathrm{ad}_{H_f}\varrho + bu \qquad b = -i\mathrm{ad}_{H_c}\varrho_d$$

  - linearization lives on the tangent plane of ϱ_d
  - if linearization satisfies the Kalman controllability condition

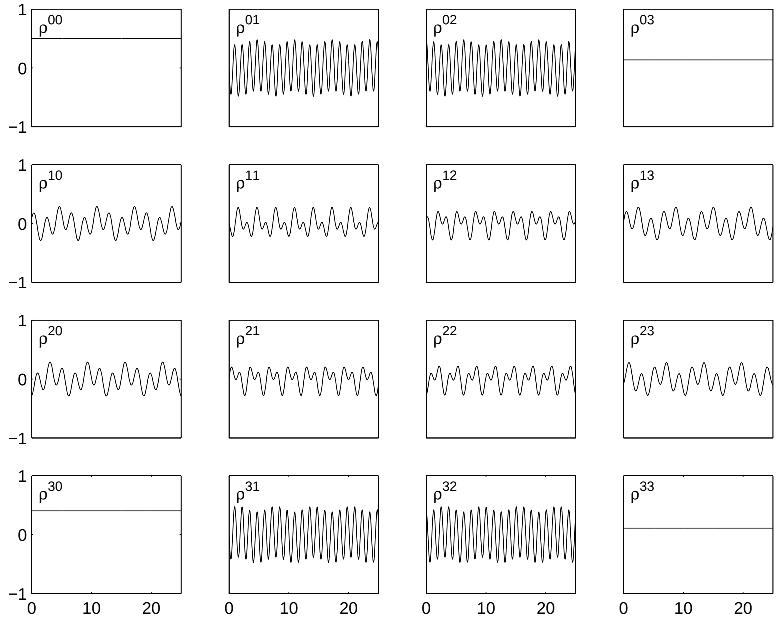
$$\mathrm{rank}\left[b \;\; -i\mathrm{ad}_{H_f}b \;\; (-i\mathrm{ad}_{H_f})^2 b \ldots\right] = \dim(\mathcal{S})$$

    then there is no "direction" in which you can move the closed loop system while staying in $\mathcal{N} \Longrightarrow \mathcal{N}$ is empty
  - ad-bracket $\Longleftrightarrow$ Kalman controllability
- when topology is nontrivial: linearization does not give global answers, only local

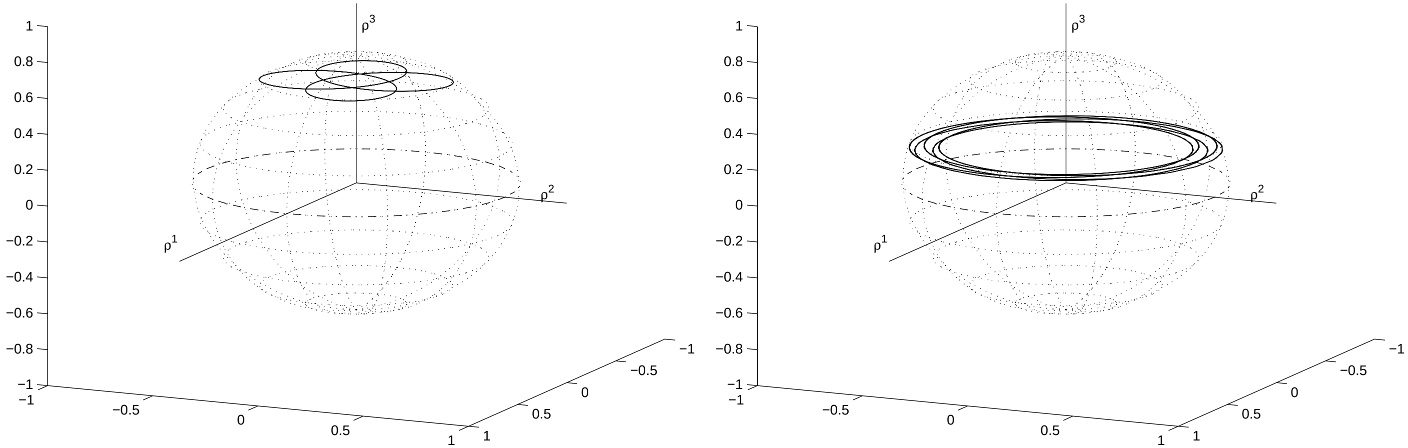


State tracking

- desirede state ϱ_d

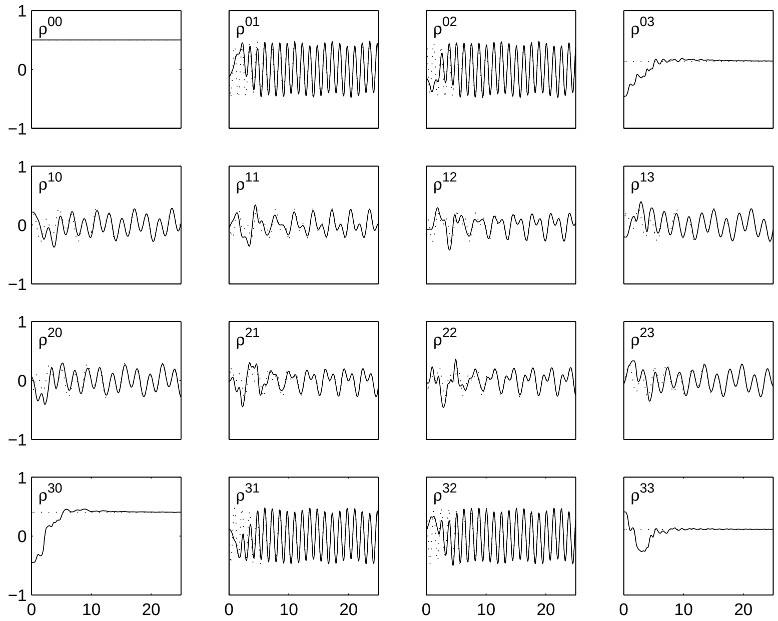


- its reduced densities

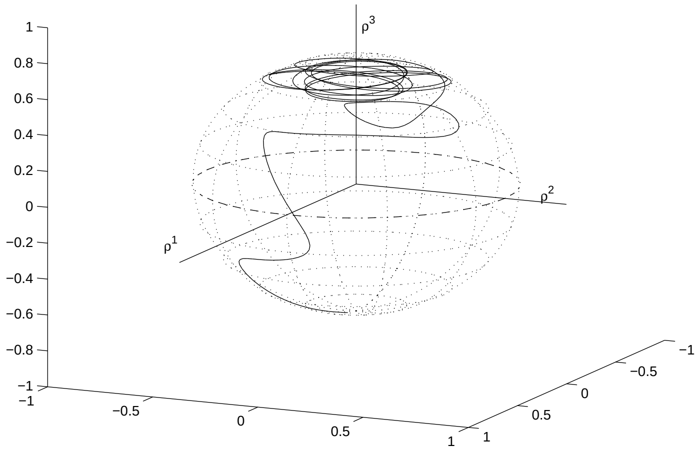


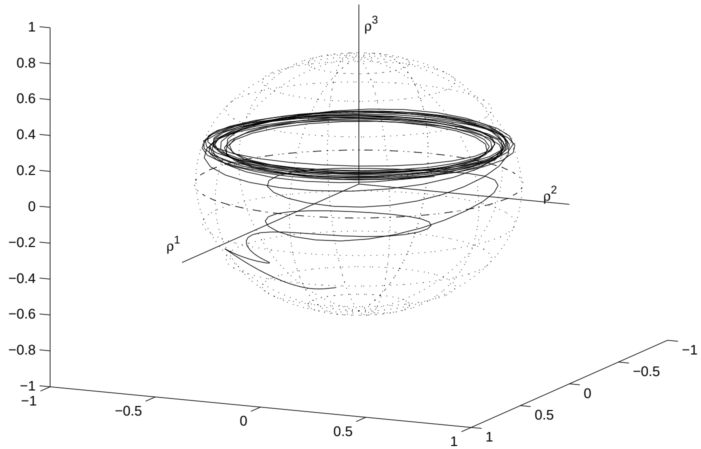
State tracking

- the 15 components



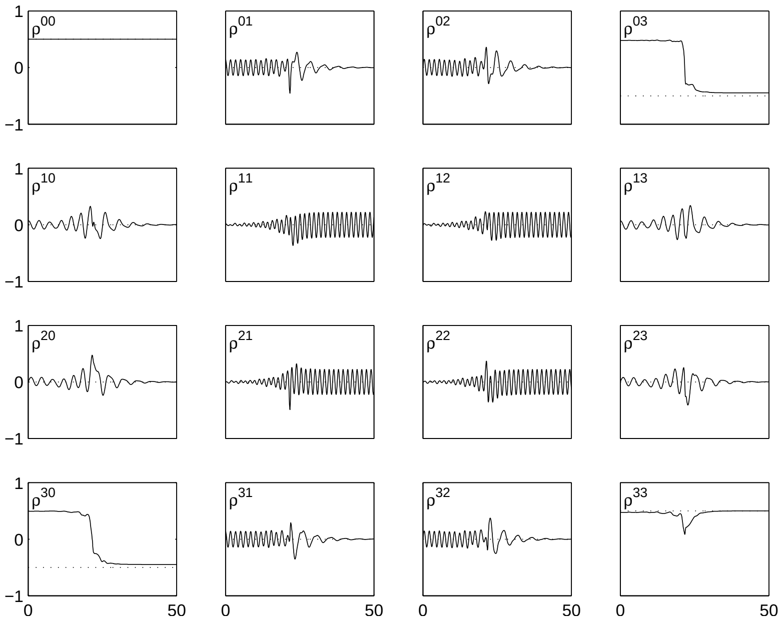
- its reduced densities





State tracking

- presence of singularities
  - “theoretically” state-to-state trasfer may fail
  - “practically” state-to-state transfer may be slow around the singular points (control action has to “build up” from 0)
- example $|00\rangle \rightarrow |11\rangle$
- it is better to apply an open-loop pulse to get approximately near the target and only then switch on the feedback



Generalizations

1. selective controls: tracking is easier since there are more control degrees of freedom
2. same theorem holds for n coupled spin 1/2
3. tracking Hamiltonians with any coupling

$$H_f \qquad \longrightarrow \qquad H_f = \sum_{jk} h^{jk} \Lambda_{jk}$$

- example: Heisenberg or dipole-dipole Hamiltonian

$$H_f = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{11}\Lambda_{11} + h^{22}\Lambda_{22} + h^{33}\Lambda_{33}$$

- any other transversal term can be added as well, also $h^{jk}\Lambda_{jk}$, $j \neq k$
- prerequisite: controllability

4. only caveat: need to use $H_{f_d} = H_f$ in the theorem. Next: want to relax this constraint

Suppression of unwanted couplings

- so far: $H_{f_d} = H_f$
- $\Longrightarrow$ the derivative of the Lyapunov function is homogeneous in u

$$\dot{V} = -u\langle\langle \boldsymbol{\varrho}_\mathrm{d}, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle$$

- $\Longrightarrow$ design of the feedback is "natural"

$$u = k\langle\langle \boldsymbol{\varrho}_\mathrm{d}, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle$$

  and guarantees at least $\dot{V} \leqslant 0$.
- if $H_{f_d} \neq H_f$ then feedback design is more difficult, since $\dot{V}$ is no longer homogeneous in u:

$$\dot{V} = \underbrace{\langle\langle \boldsymbol{\varrho}_\mathrm{d}, -i\mathrm{ad}_{(H_{f_d}-H_f)}\boldsymbol{\varrho}\rangle\rangle}_{\text{sign indefi nite term}} - u\langle\langle \boldsymbol{\varrho}_\mathrm{d}, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle$$

- want to see whether the algorithm is still converging

Suppression of unwanted couplings

- call $H_\delta = H_{f_d} - H_f$
  - H_δ = unwanted Hamiltonian
  - $\longrightarrow$ *disturbance to reject*
- example:
  - H_{f_d} Ising Hamiltonian

$$H_{f_d} = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{33}\Lambda_{33}$$

  - H_f Heisenberg Hamiltonian or dipole-dipole Hamiltonian

$$H_f = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{11}\Lambda_{11} + h^{22}\Lambda_{22} + h^{33}\Lambda_{33}$$

  - $\Longrightarrow H_\delta$ contains only transversal couplings

$$H_\delta = h^{11}\Lambda_{11} + h^{22}\Lambda_{22}$$

Suppression of unwanted couplings

Proposition *If H_δ contains only "slow" couplings (approximately of one order of magnitude smaller than those of H_{f_d}) then $\exists$ a sufficiently high feedback gain k and a ω_{rf} such that $\dot{\boldsymbol{\varrho}} = -i(\mathrm{ad}_{H_f} + u\mathrm{ad}_{H_c})\boldsymbol{\varrho}$ with the (nonselective) feedback controller $u = \langle\langle \boldsymbol{\varrho}_d, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho} \rangle\rangle$ tracks the reference trajectory $\dot{\boldsymbol{\varrho}}_d = -i\mathrm{ad}_{H_{f_d}}\boldsymbol{\varrho}_d$*

- H_δ is a "persistent" disturbance $\longrightarrow$ never vanish
  - $\Longrightarrow$ you never reach a steady state because of the persistent excitation
  - $\Longrightarrow$ stability is only up to a small error $\longrightarrow$ *practical stability*
- meaning of the Proposition: if H_δ is slow with respect to the feedback dynamics then it may not destroy convergence

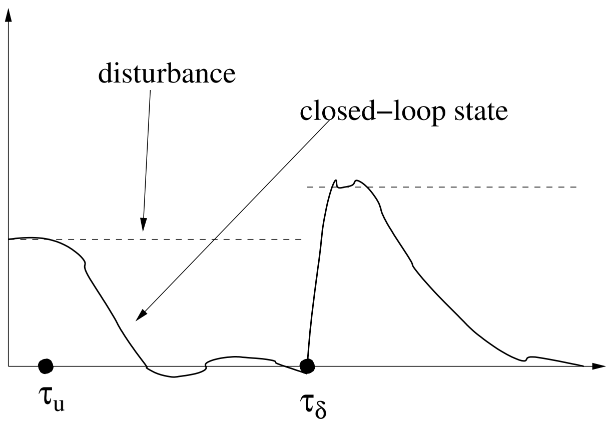
Suppression of unwanted couplings

Sketch of the proof

- derivative of the Lyapunov function

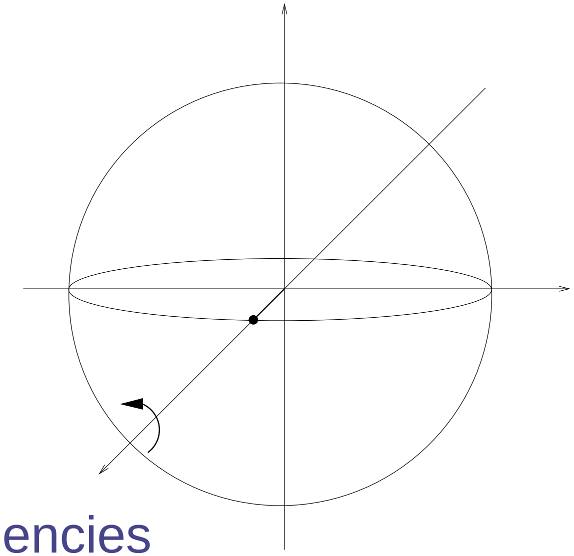
$$\dot{V} = \underbrace{\langle\langle \boldsymbol{\varrho}_{\mathrm{d}},\, -i\mathrm{ad}_{(H_{f_d}-H_f)}\boldsymbol{\varrho}\rangle\rangle}_{\text{slow time scale } \tau_\delta} - \underbrace{u\langle\langle \boldsymbol{\varrho}_{\mathrm{d}},\, -i\mathrm{ad}_{H_c}\boldsymbol{\varrho}\rangle\rangle}_{\text{fast time scale } \tau_u}$$

- in the fast time scale τ_u: disturbance can be thought as frozen
- $\Longrightarrow$ in the closed-loop dynamics it amounts to a constant “displacement”
- if the feedback can recover fast from such a displacement then convergence still holds



Suppression of unwanted couplings

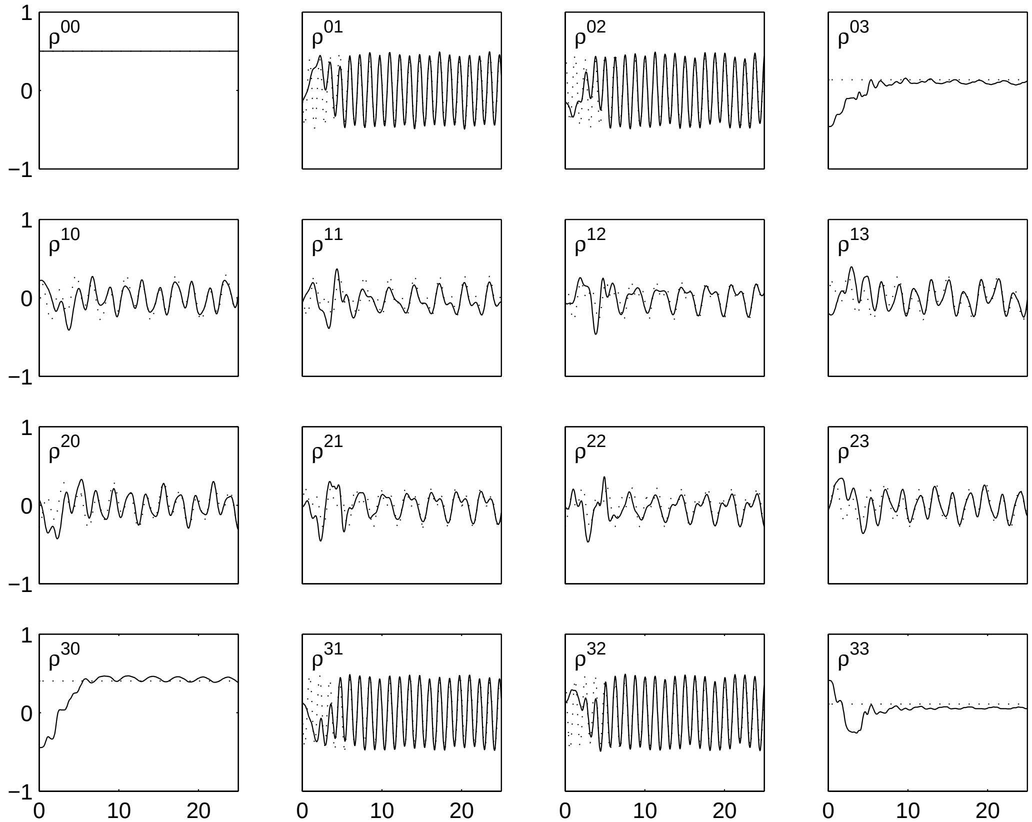
- Problem: control is along the λ_1 axis $\Longrightarrow$ when reduced density is aligned with λ_1 axis you have no control action
- $\Longrightarrow$ singularity of the control law



- you need to get away from this alignement
  - by means of the coupling
  - by means of the local precession
- to do it fast: choose ω_{rf} so that the Larmor frequencies $h^{30} = -(\omega_{0,\alpha} - \omega_{rf})$ and $h^{03} = -(\omega_{0,\beta} - \omega_{rf})$ are in the fast time scale
- $\Longrightarrow$ feedback loop
  - exits fast from the singularities
  - can recover the disturbance H_δ in the fast time scale
- feedback does not work for H_δ of the same order as H_{f_d}

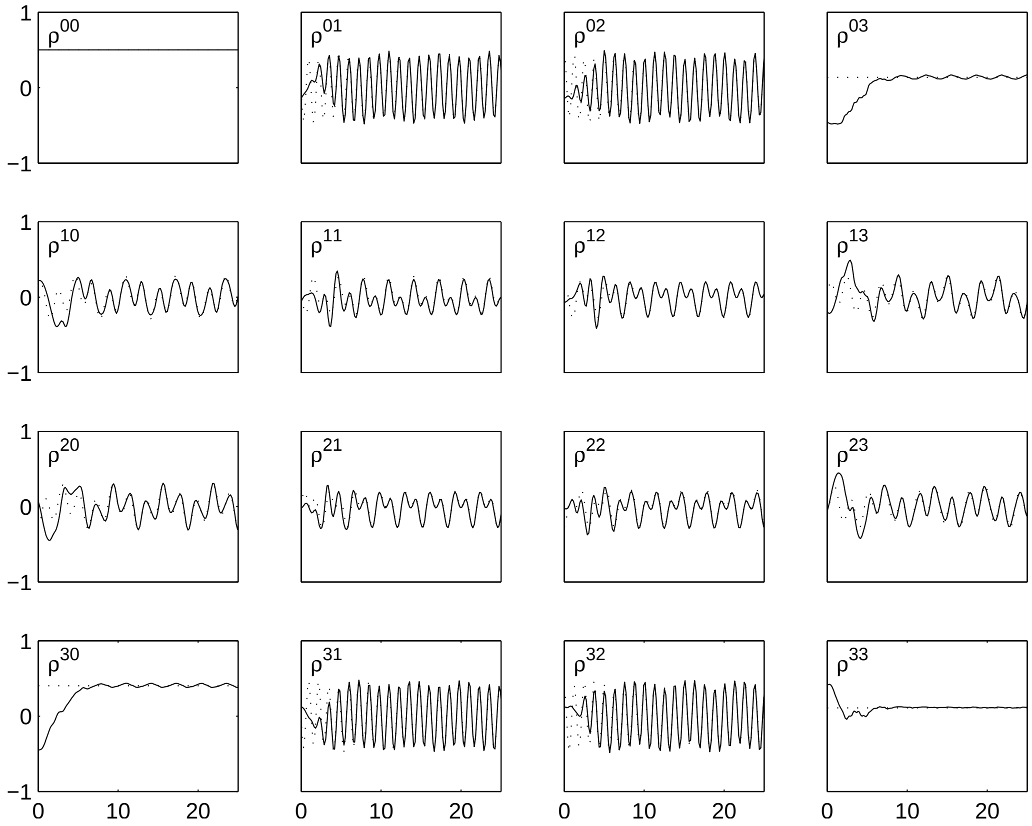
Suppression of unwanted couplings

- example mentioned above:
- H_{f_d} Ising, $\quad H_f$ Heisenbeg
- $H_\delta = h^{11}\Lambda_{11} + h^{22}\Lambda_{22}$



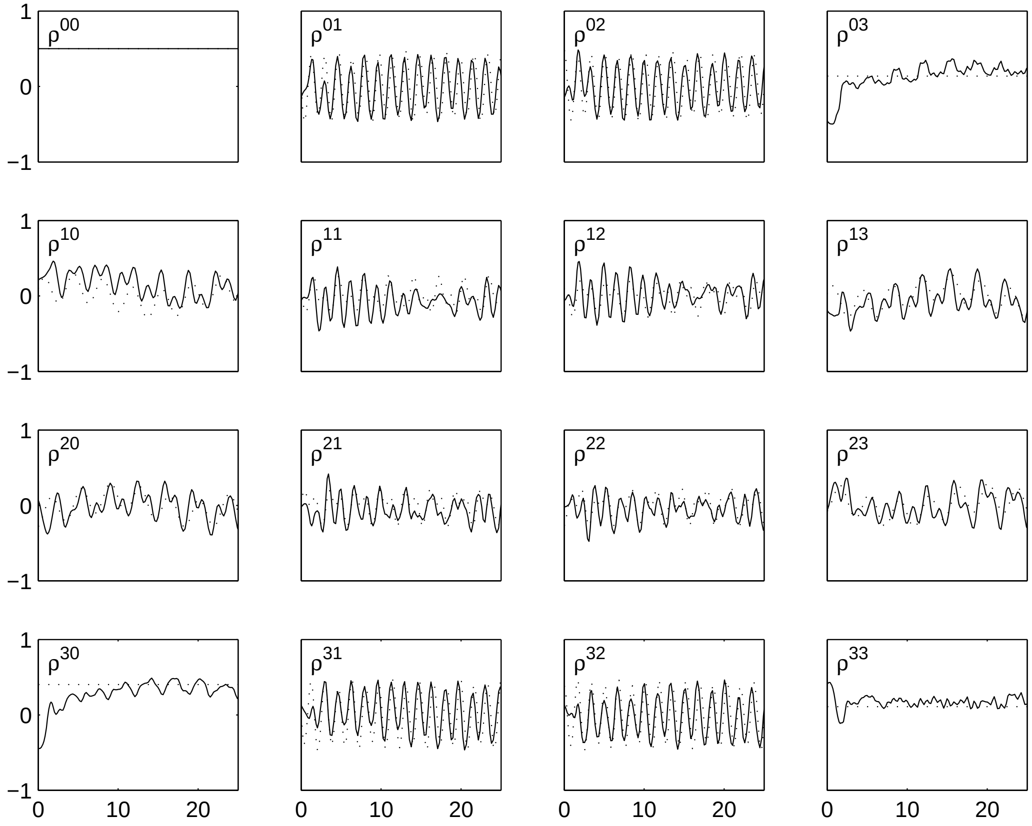
Suppression of unwanted couplings

- if the controls are selective, then any coupling can be suppressed
- previous example: slow coupling



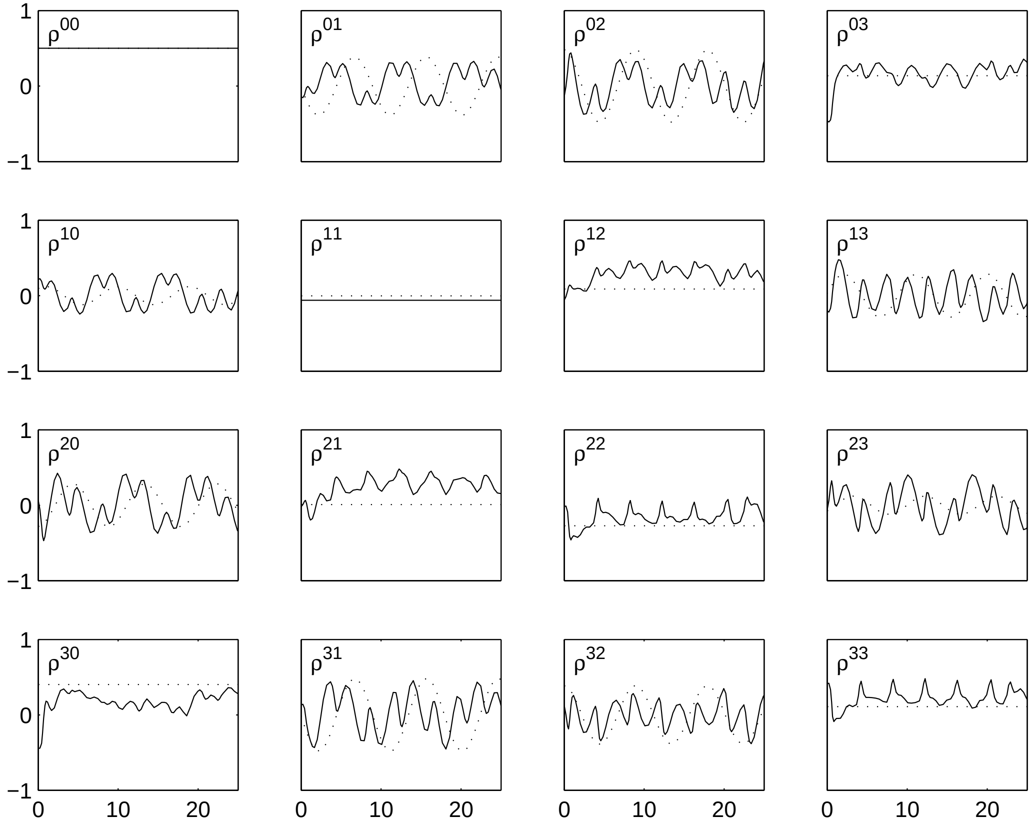
Suppression of unwanted couplings

- selective controls
- example: fast coupling to reject
  - $H_\delta = h^{11}\Lambda_{11} + h^{22}\Lambda_{22}$
  - $\tau_\delta \simeq \tau_u$



Suppression of unwanted couplings

- selective controls
- choosing two rf frequencies slightly off-resonance helps convergence
- same example:
  - $H_\delta = h^{11}\Lambda_{11} + h^{22}\Lambda_{22}$
  - $h^{03} = h^{30} = 0$

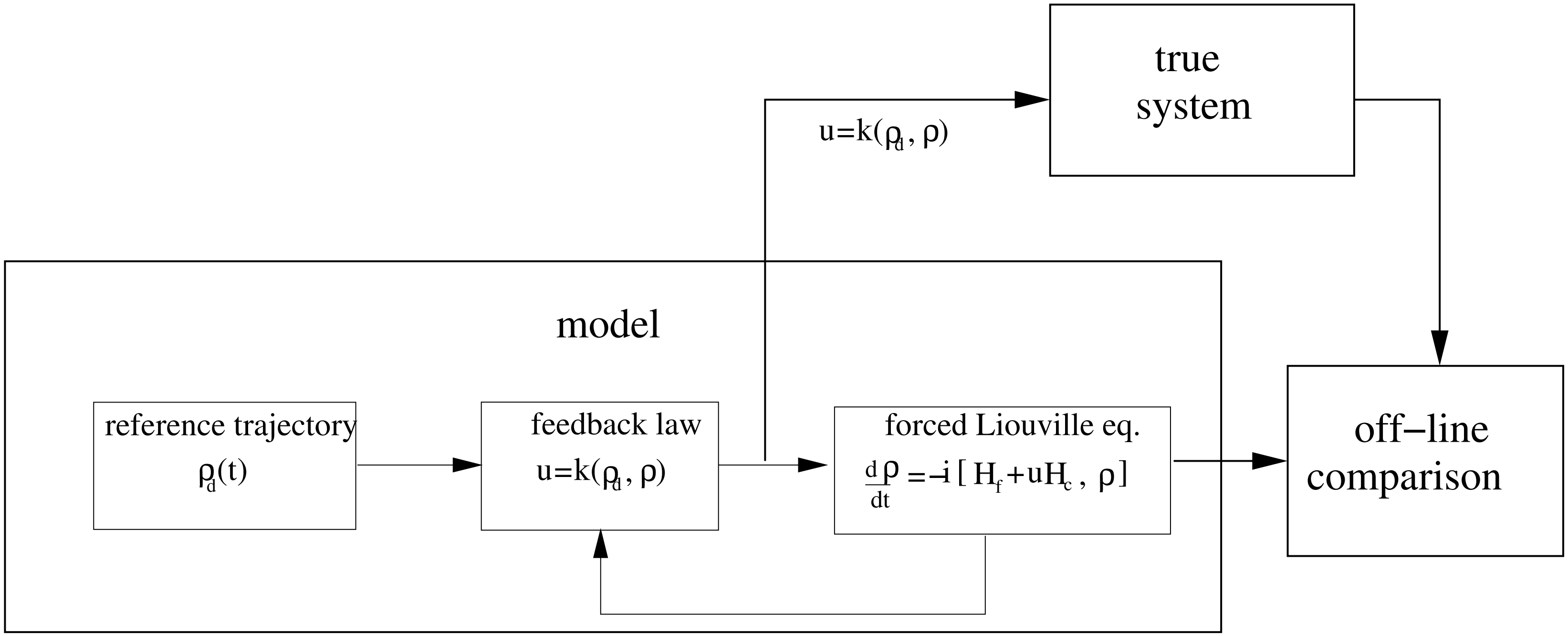


Using the algorithm for open loop control

- full state is *not* available $\Longrightarrow$ impossible to implement for real
- use scheme in an open-loop fashion, to generate time-dependent shaped pulses mapping $\varrho \longrightarrow \varrho_d$
- standard open-loop control methods
  1. hard pulses
     - high power $\Longrightarrow$ shorter times
  2. soft, shaped pulses
     - low power $\Longrightarrow$ long times
  - both need selectivity
  - simultaneous selective pulses $\Longrightarrow$ cross talk $\Longrightarrow$ need to precalculate the corrections
  - In presence of complicated couplings (solid state), both methods are difficult to use (remember: open-loop methods are “NP-hard”)
- Is it possible to let the simulator compute the pulses by means of the “feedback on the model”?

Using the algorithm for open loop control

- open-loop control based on *feedback on the model* can be used for
  1. “one-shot” gate (more properly: state transfer map)
  2. “learn” from the simulator the control inputs that decouple an unwanted Hamiltonian

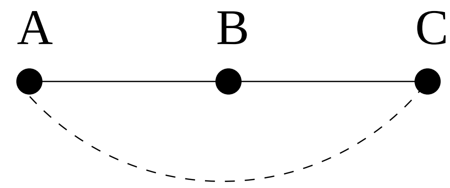


Using the algorithm for open loop control

- have the "true" state of the simulator track the desired reference, take the time-dependent control signal produced by the simulator and go to the lab.
- improvement w.r.t. the previous simulations: at $t = 0$ ϱ can start already on the desired ϱ_d
  - $\longrightarrow$ no need to "show asymptotic stability"
  - $\longrightarrow$ no "transient" behavior
- prerequisite: need to known the initial condition $\longrightarrow$ always the case in NMR
- any time you change the initial condition $u(t)$ is different (simulator and feedback algorithms remain the same)
- drawback: profile of $u(t)$ is normally not "nice"

Open loop control: a 3 spin example

aim: suppress unwanted couplings



- three identical spins, no chemical shift
- free Hamiltonian: dipole-dipole coupling
  1. two couplings between A-B and B-C

$$(-\Lambda_{110} - \Lambda_{220} + 2\Lambda_{330}) + (-\Lambda_{011} - \Lambda_{022} + 2\Lambda_{033})$$

  2. one coupling (of weak strength) that I want to suppress between A-C

$$\frac{1}{8}(-\Lambda_{101} - \Lambda_{202} + 2\Lambda_{303})$$

- control Hamiltonian: nonselective control field

$$u_1(\Lambda_{001} + \Lambda_{010} + \Lambda_{100})$$

Open loop control: a 3 spin example

- “disturbance” Hamiltonian

$$H_\delta = H_{f_d} - H_f = \frac{1}{8}\left(-\Lambda_{101} - \Lambda_{202} + 2\Lambda_{303}\right)$$

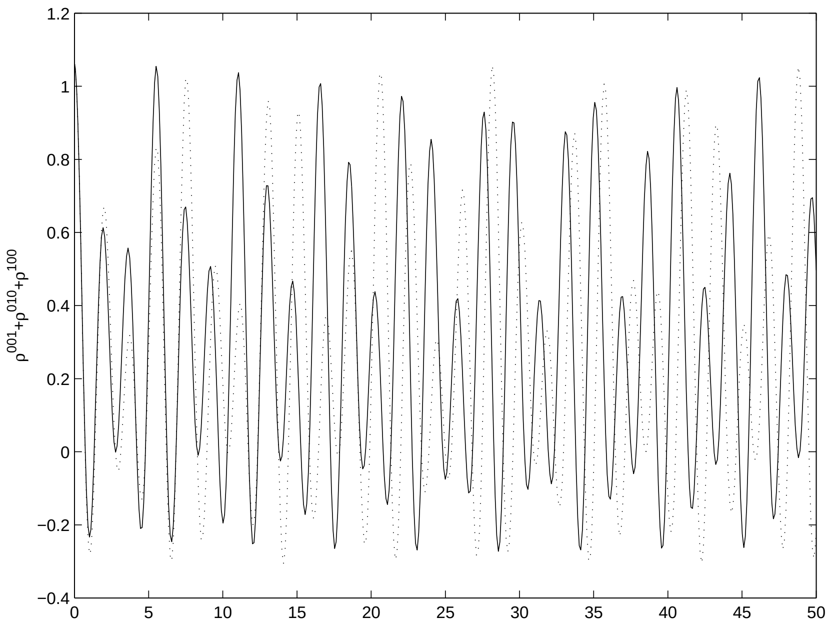
- initial state along the λ_1 axis

$$\boldsymbol{\varrho}(0) = \frac{1}{(\sqrt{2})^3}\begin{bmatrix}1\\1\\0\\0\end{bmatrix} \otimes \begin{bmatrix}1\\1\\0\\0\end{bmatrix} \otimes \begin{bmatrix}1\\1\\0\\0\end{bmatrix}$$

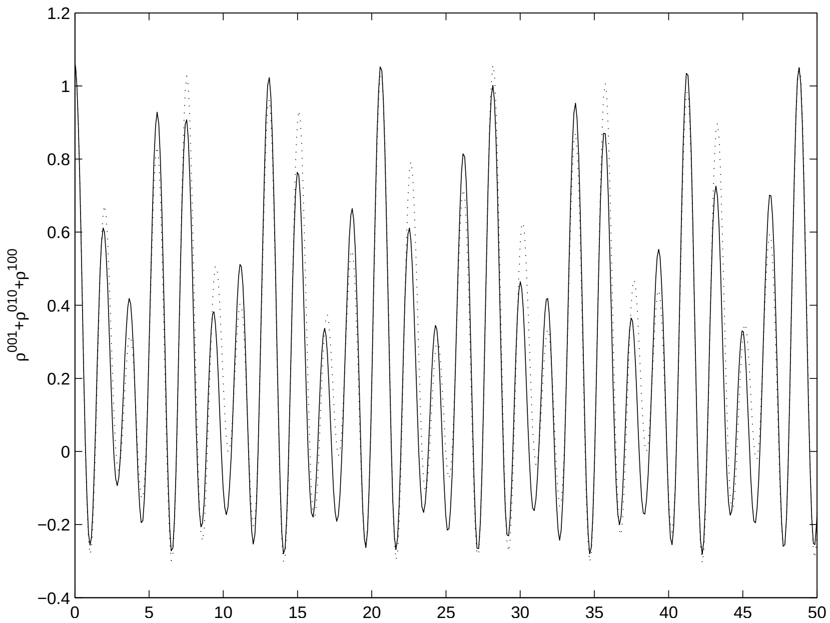
- look at the FID of the signal $\varrho^{001} + \varrho^{010} + \varrho^{100}$

Open loop control: a 3 spin example

FID without control

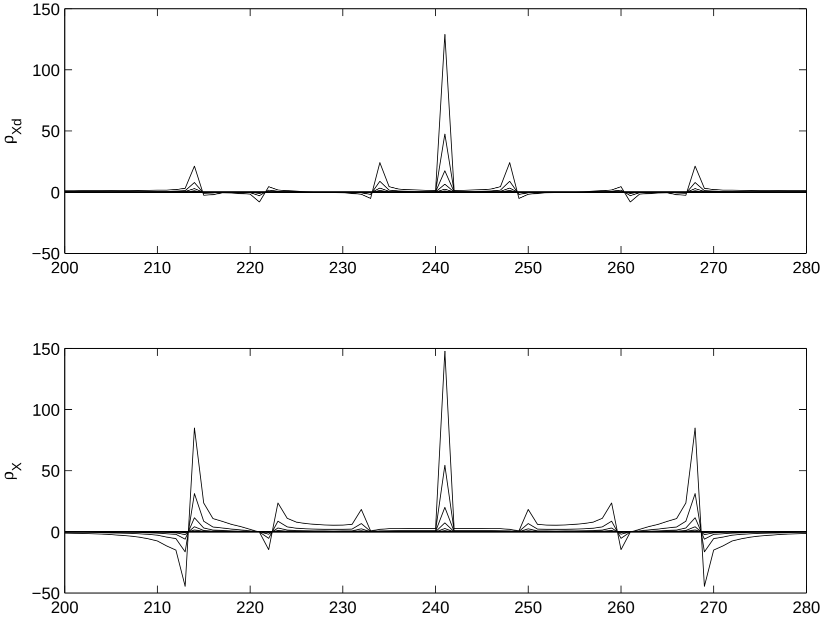


FID with feedback

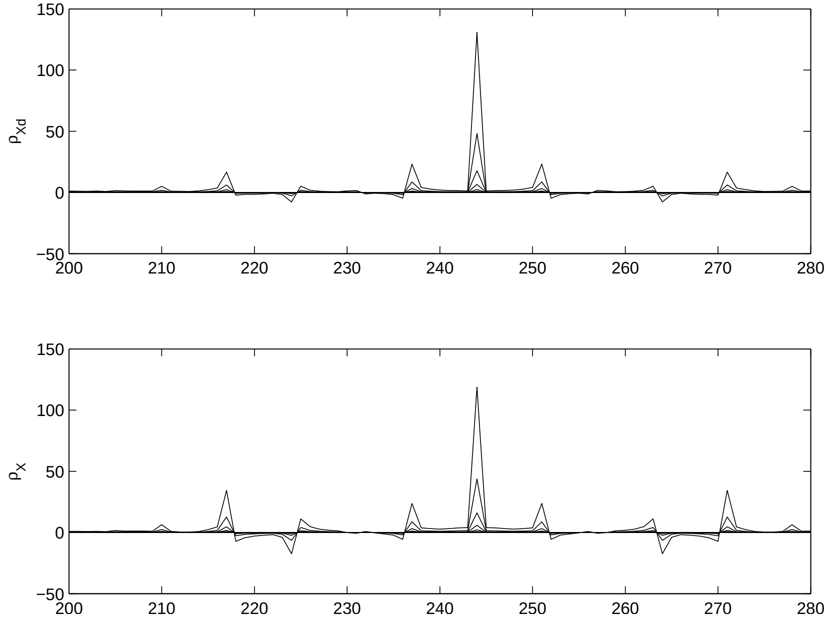


Open loop control: a 3 spin example

FID without control

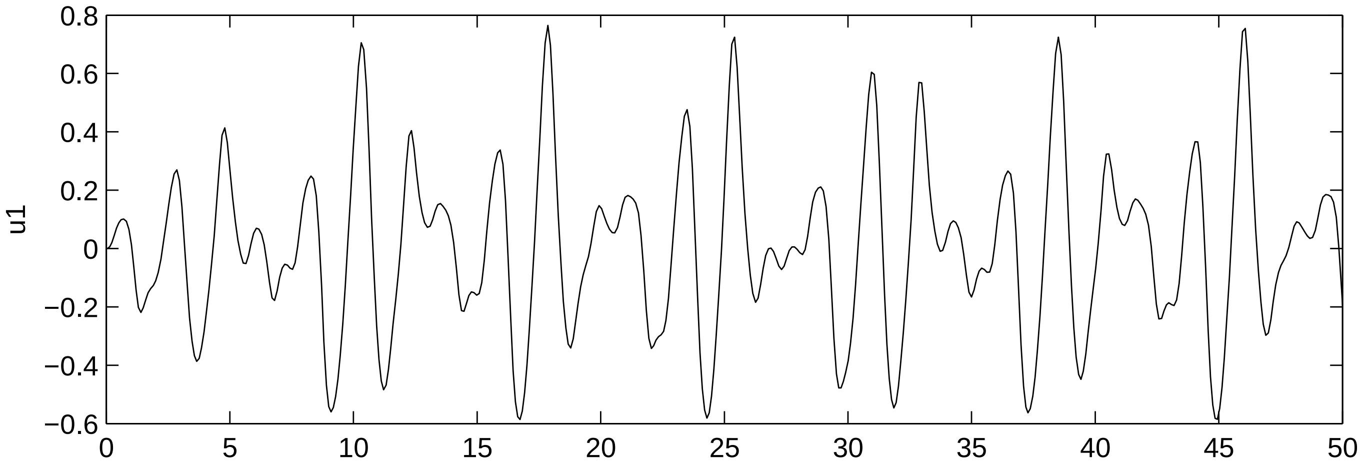


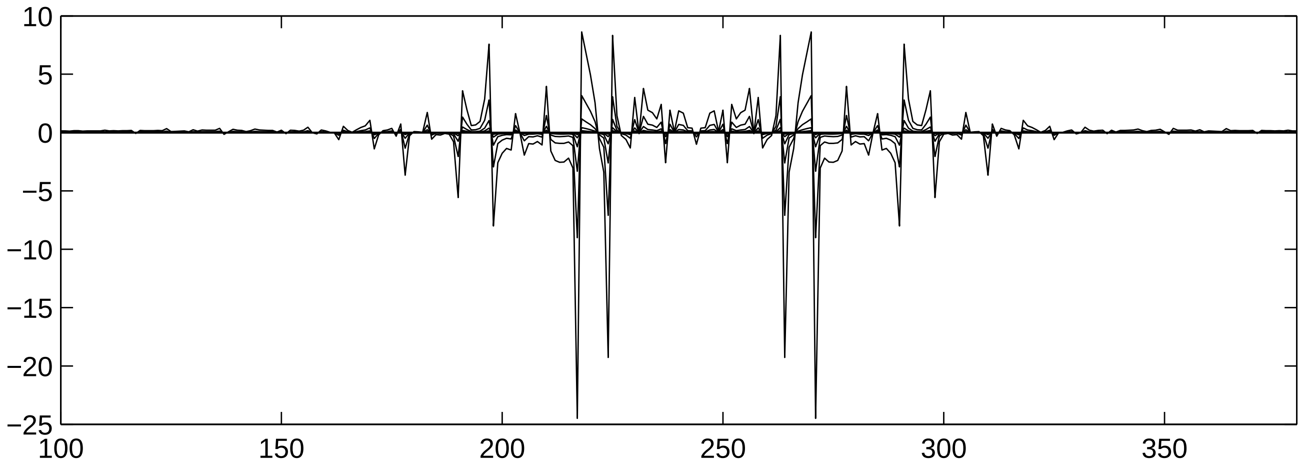
FID with feedback



Open loop control: a 3 spin example

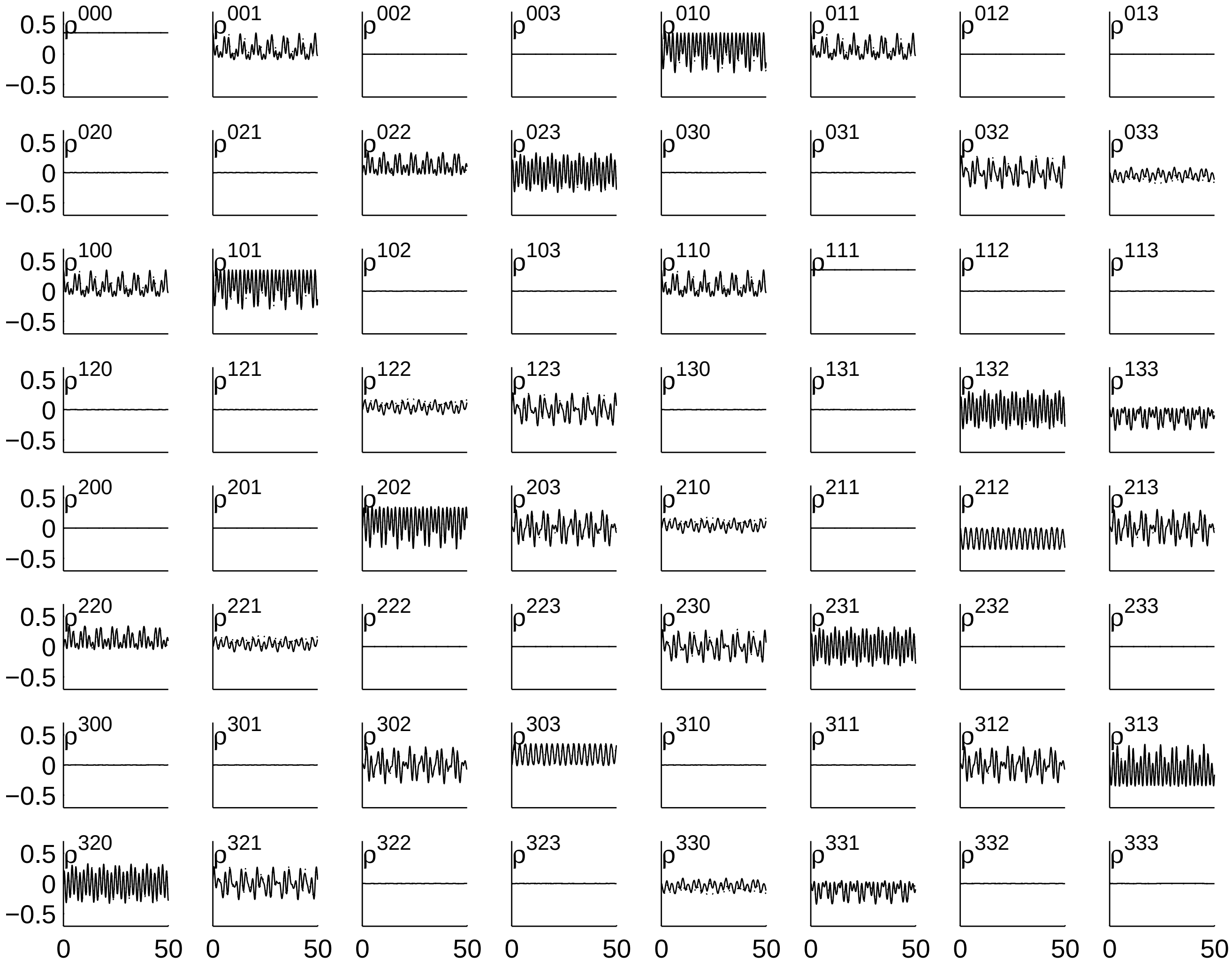
- control signal that does it





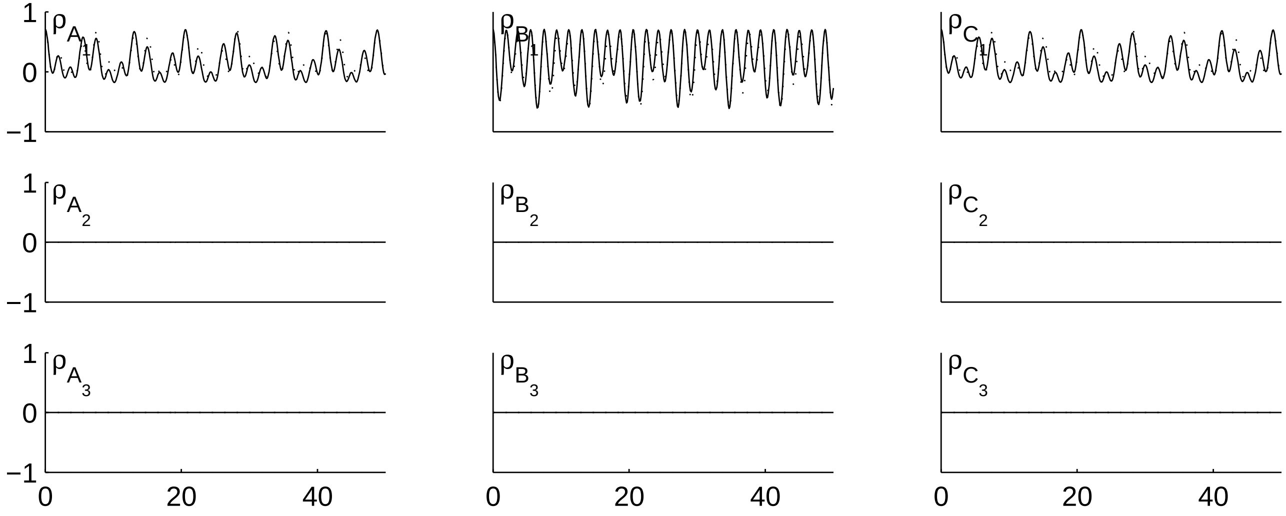
Open loop control: a 3 spin example

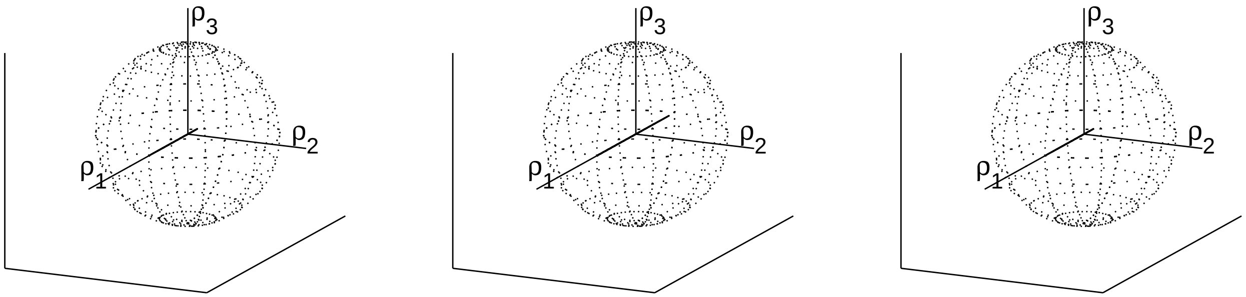
- how about the rest of the state? The feedback is decoupling the entire state space!



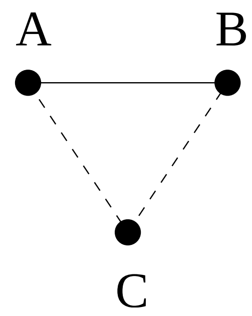
Open loop control: a 3 spin example

- for the 3 reduced densities





Open loop control: another 3 spin example



- three identical spins, no chemical shift
- free Hamiltonian: dipole-dipole coupling
  1. a couplings between A-B

$$(-\Lambda_{110} - \Lambda_{220} + 2\Lambda_{330})$$

  2. two couplings (of weak strength) that I want to suppress between A-C and B-C

$$+\frac{1}{8}(-\Lambda_{011} - \Lambda_{022} + 2\Lambda_{033}) + \frac{1}{8}(-\Lambda_{101} - \Lambda_{202} + 2\Lambda_{303})$$

$\Longrightarrow$ I want to decouple C from A-B

- control Hamiltonian: nonselective control field

$$u_1(\Lambda_{001} + \Lambda_{010} + \Lambda_{100})$$

Open loop control: another 3 spin example

- “disturbance” Hamiltonian

$$H_\delta = H_{f_d} - H_f = \frac{1}{8}\left(-\Lambda_{101} - \Lambda_{202} + 2\Lambda_{303}\right) + \frac{1}{8}\left(-\Lambda_{011} - \Lambda_{022} + 2\Lambda_{033}\right)$$

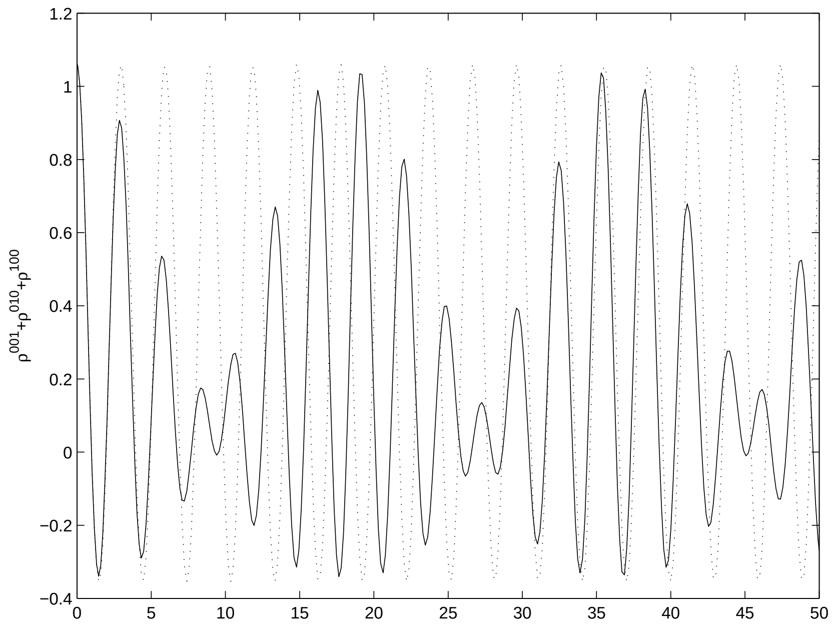
- initial state along the λ_1 axis

$$\boldsymbol{\varrho}(0) = \frac{1}{(\sqrt{2})^3}\begin{bmatrix}1\\1\\0\\0\end{bmatrix} \otimes \begin{bmatrix}1\\1\\0\\0\end{bmatrix} \otimes \begin{bmatrix}1\\1\\0\\0\end{bmatrix}$$

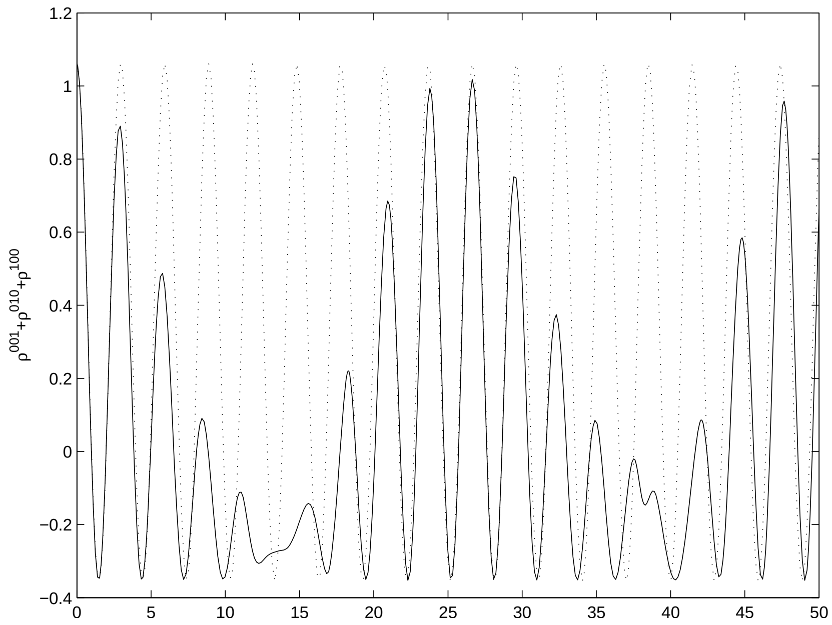
- look at the FID of the signal $\varrho^{001} + \varrho^{010} + \varrho^{100}$

Open loop control: another 3 spin example

FID without control



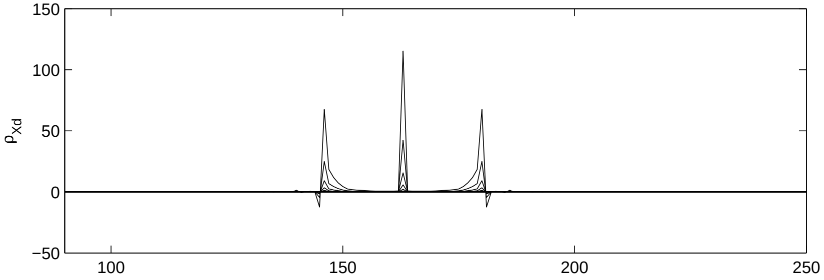
FID with feedback

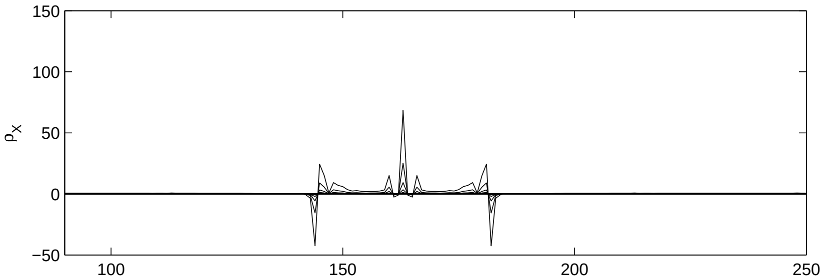


decoupling is not very good....

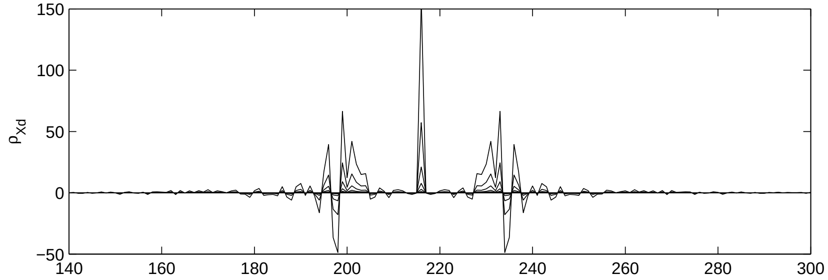
Open loop control: another 3 spin example

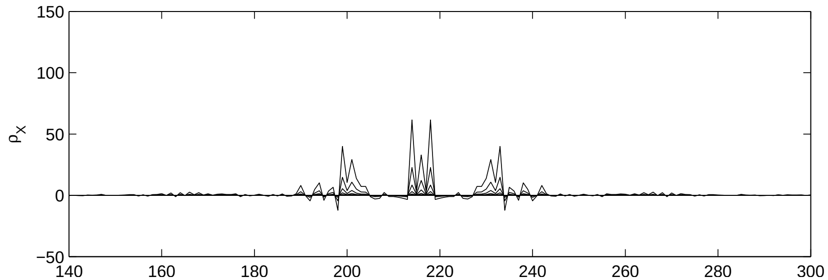
FID without control





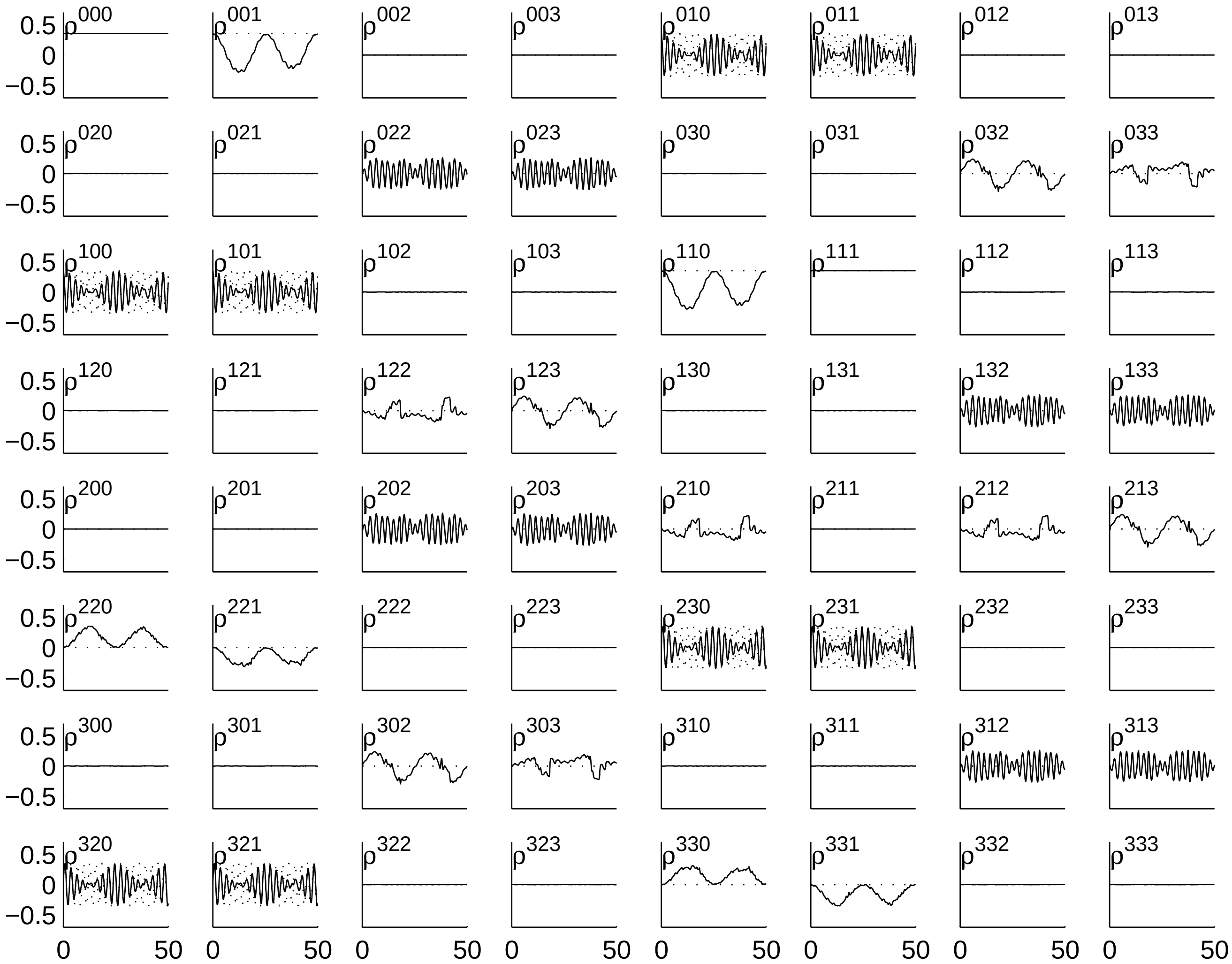
FID with feedback





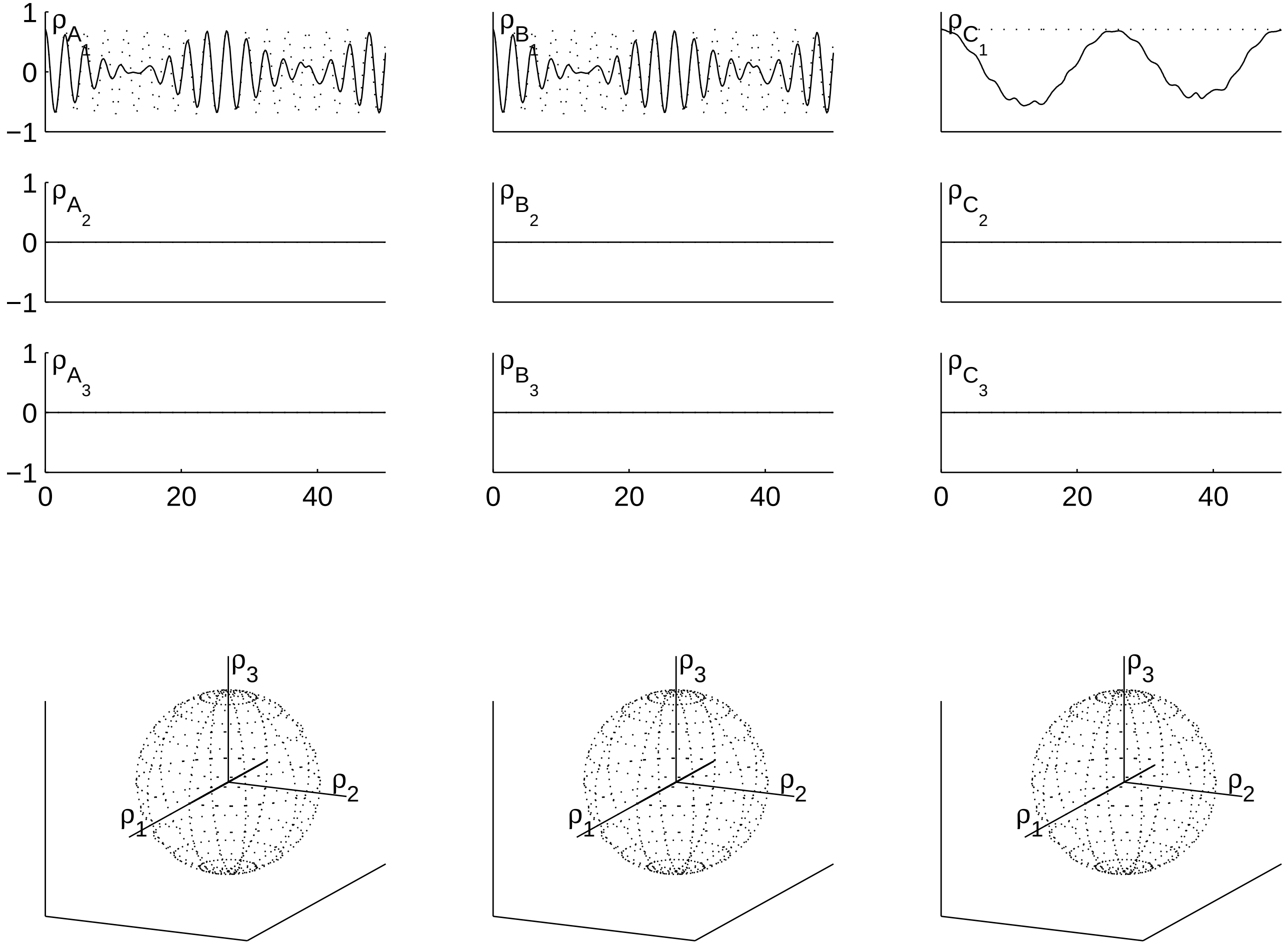
Open loop control: another 3 spin example

- rest of the state

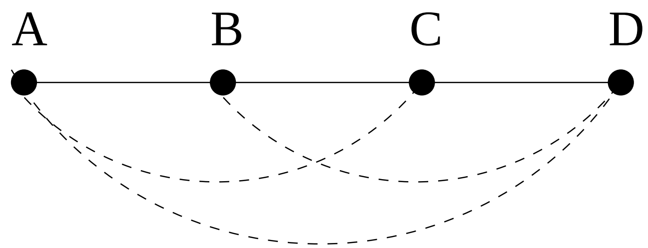


Open loop control: another 3 spin example

- for the 3 reduced densities



Open loop control: a 4 spin example

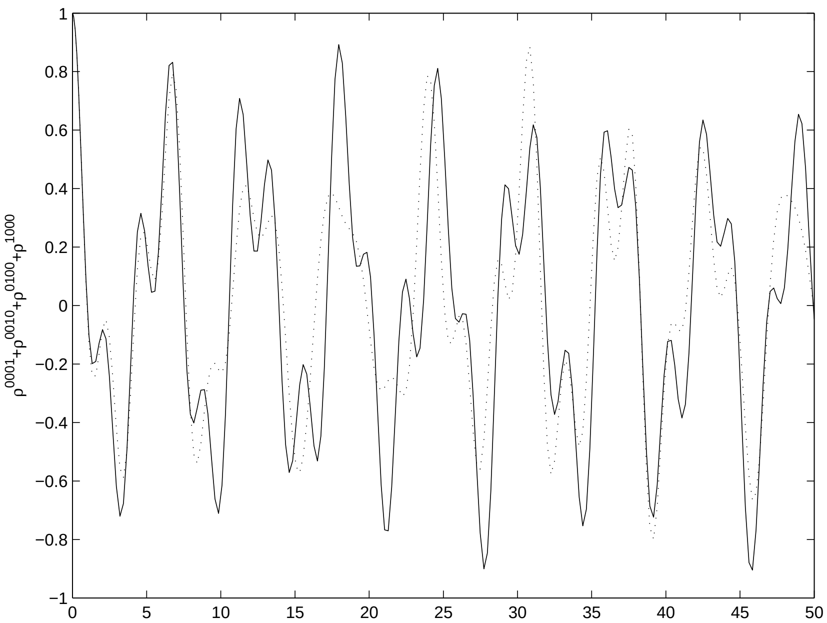


- 4 identical spins, no chemical shift
- free Hamiltonian: dipole-dipole coupling
  1. a couplings between A-B, B-C, C-D
  2. couplings to reject: A-C, B-D and A-D
  3. $\Longrightarrow$ want to make a linear spin chain
- control Hamiltonian: nonselective control field

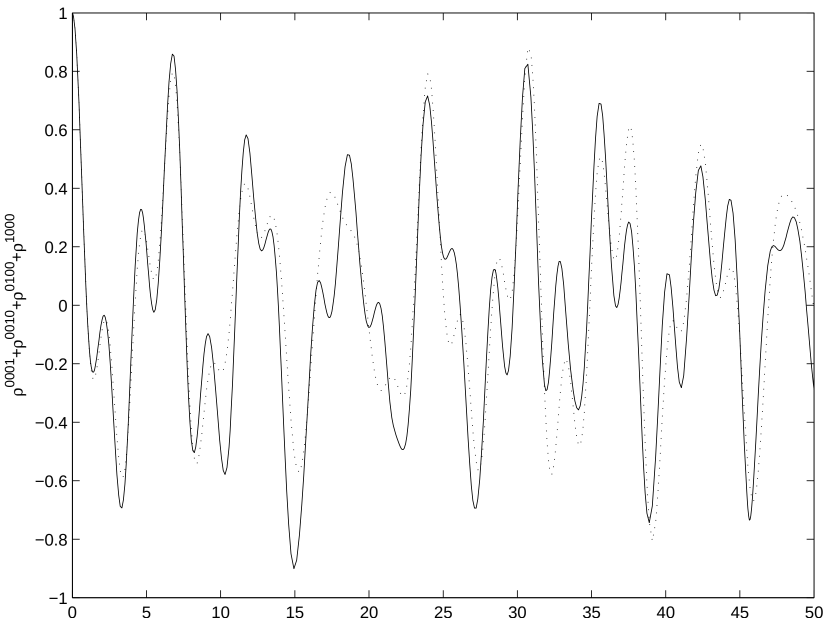
$$u_1(\Lambda_{0001} + \Lambda_{0010} + \Lambda_{0100} + \Lambda_{1000})$$

Open loop control: a 4 spin example

FID without control



FID with feedback

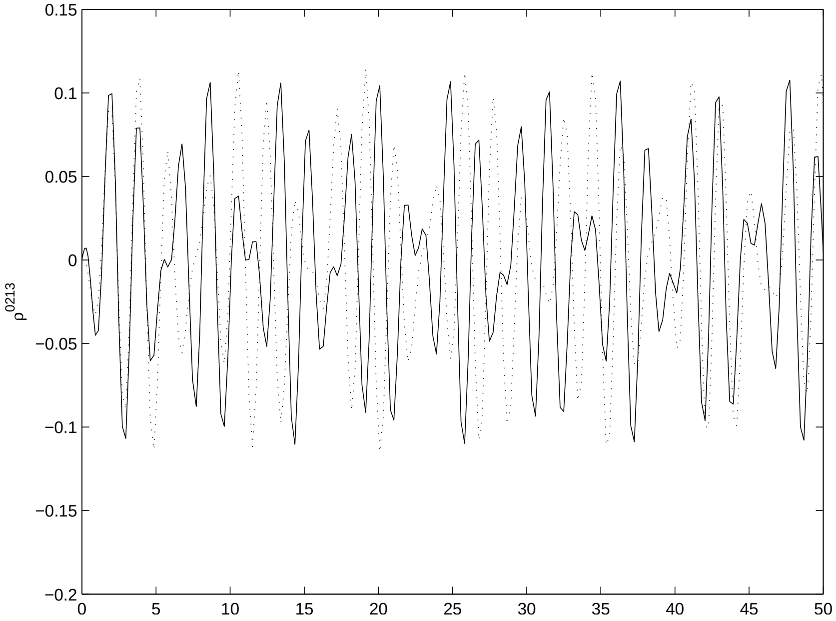


entire state space is already decoupled? Not really....

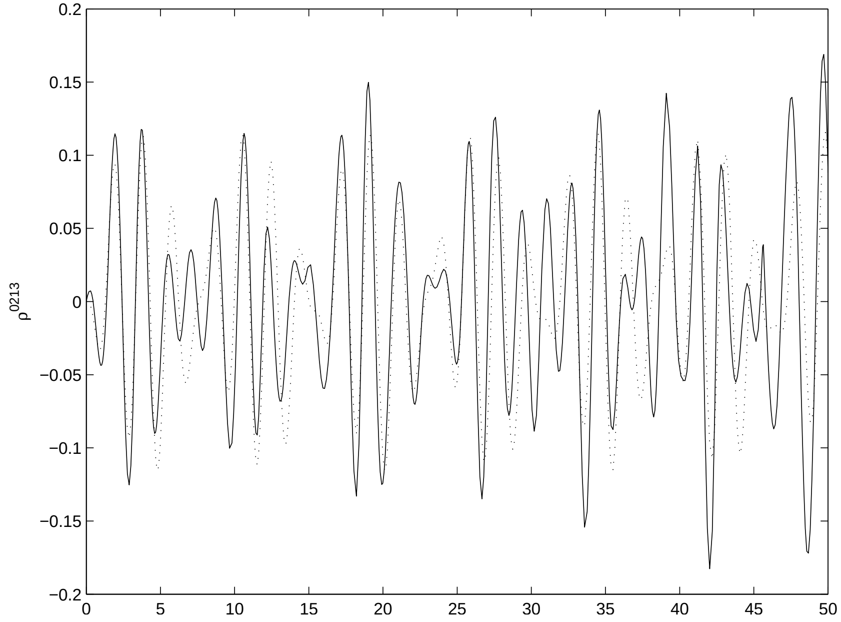
Open loop control: a 4 spin example

- take e.g. a 3-body correlation: ϱ^{0213}

ϱ^{0213} without control



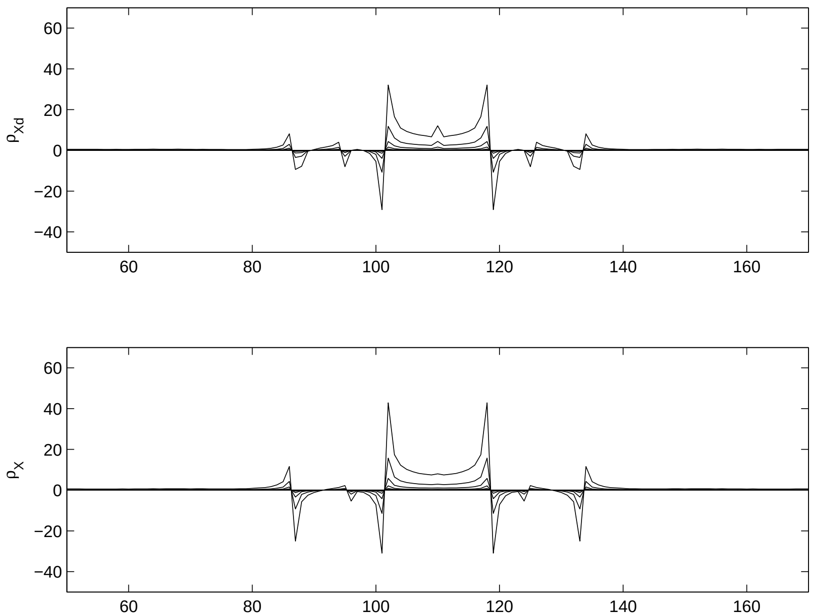
ϱ^{0213} with feedback



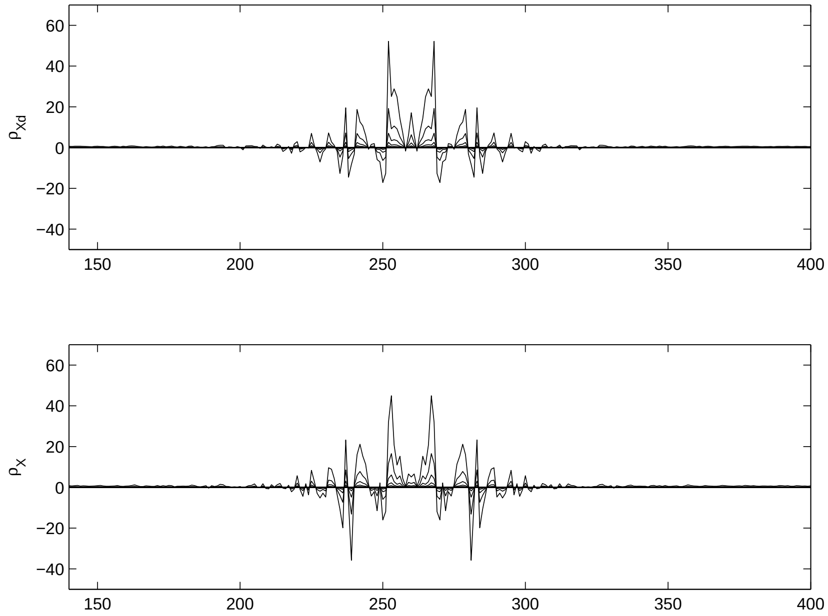
$\Longrightarrow$ solution is improved

Open loop control: a 4 spin example

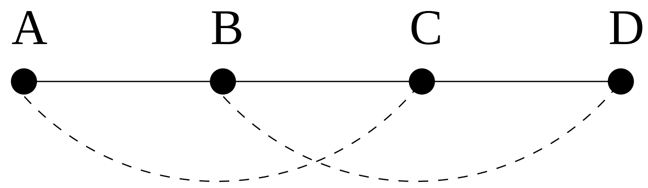
FID without control



FID with feedback



Open loop control: another 4 spin example

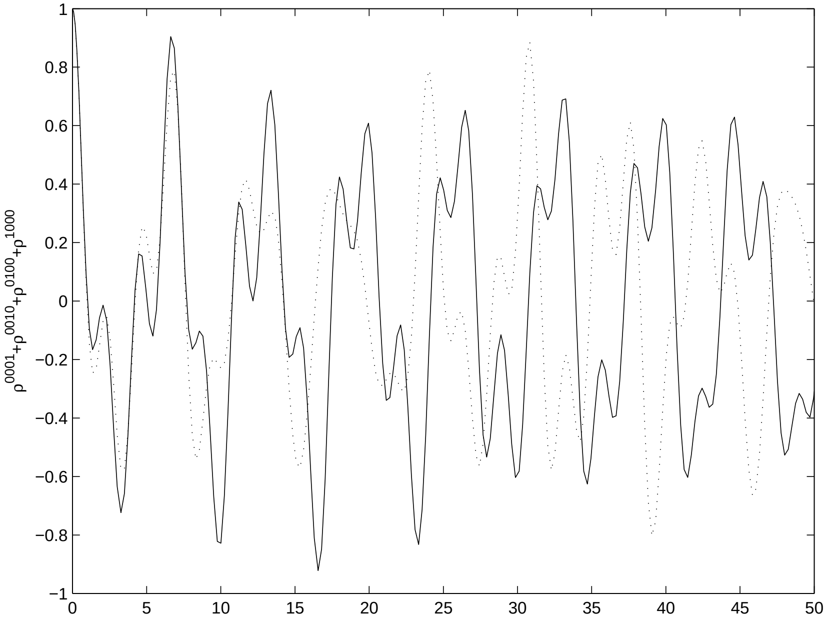


- 4 identical spins, no chemical shift
- free Hamiltonian: dipole-dipole coupling
  1. a couplings between A-B, B-C, C-D
  2. couplings to reject: A-C and B-D
  3. $\Longrightarrow$ want to make a linear spin chain
- control Hamiltonian: nonselective control field

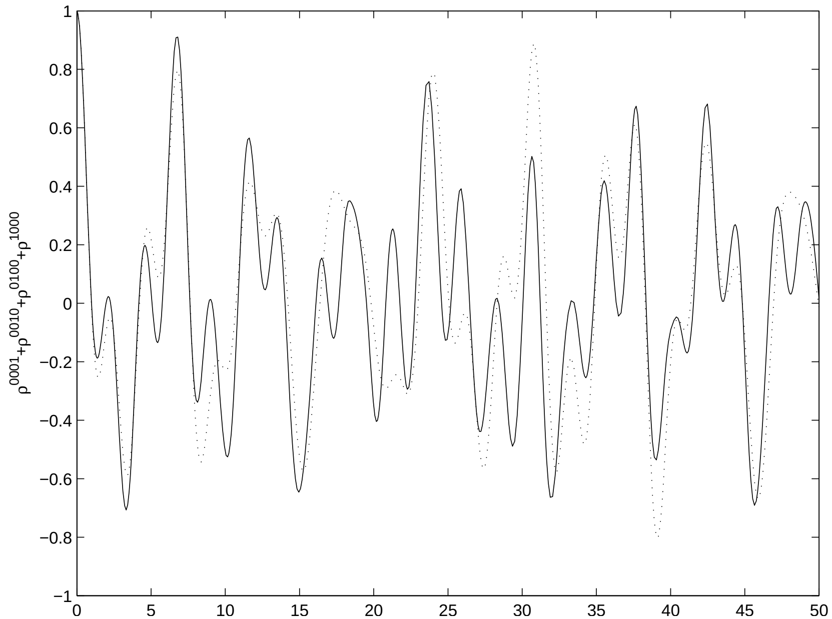
$$u_1(\Lambda_{0001} + \Lambda_{0010} + \Lambda_{0100} + \Lambda_{1000})$$

Open loop control: another 4 spin example

FID without control

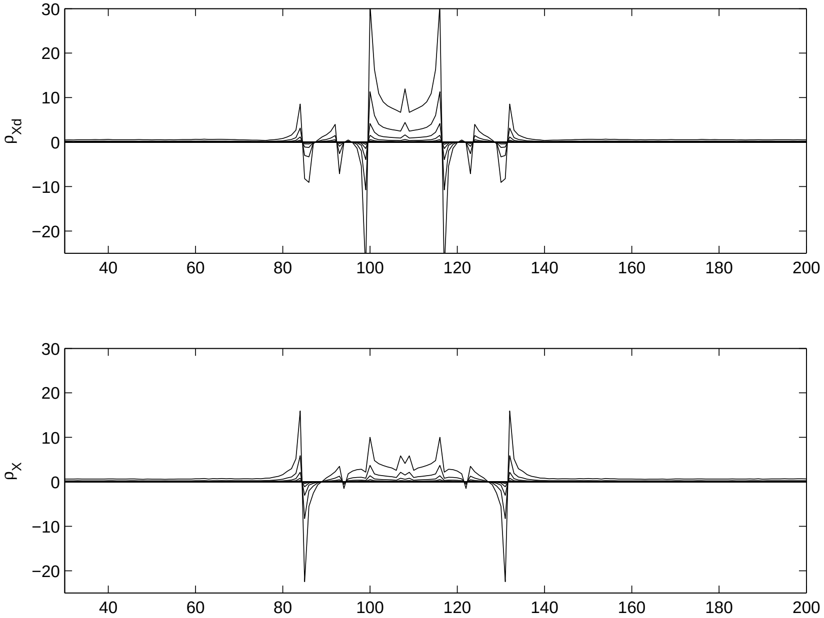


FID with feedback



Open loop control: another 4 spin example

FID without control



FID with feedback