Proceedings of the
44th IEEE Conference on Decision and Control, and
the European Control Conference 2005
Seville, Spain, December 12-15, 2005

Feedback stabilization of spin systems

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***Abstract*— The feedback stabilization problem for ensembles of coupled spin $1/2$ systems is discussed. The noninvasive nature of the bulk measurement allows for a fully unitary and deterministic closed loop. The Lyapunov-based feedback design does not require that the spins are selectively addressable.**

I. INTRODUCTION

NMR spectroscopy deals with the manipulation of nuclear spins of quantum ensembles, see [1], [5]. These systems exhibit most of the essential features of quantum mechanical systems, like the state space of tensorial type (providing exponential growth of the degrees of freedom available) and natural coupling mechanisms between spins, which guarantee the nonclassical nonlocality characteristic of quantum evolutions. For the purposes of state manipulation, over the last 40 years the field of NMR has developed a bewildering set of open loop control tools, in the form of sequences of electromagnetic pulses. While these methods are extremely versatile and universally accepted, from a control perspective NMR systems constitute a remarkable opportunity to device feedback control methods at a quantum level for a number of reasons:

1) the model of the system is known in detail;
2) its control mechanism is also very accurate;
3) the measurement is classical, thus avoiding all complications due to the state collapse problem (weak or less) unavoidable in other quantum control contexts [12], [11];
4) the relaxation times are sufficiently long to make the real-time interface with a control device feasible.

For all these reasons, a completely classical, unitary and deterministic feedback in the context of NMR systems is *theoretically feasible*, although major technical problems remain to be addressed, like the very low signal-to-noise ratio and the on-line extraction of measurements from coil magnetization in real time and in presence of rf excitation.

The main purpose of this work is to investigate in detail this feedback synthesis from a theoretical viewpoint. For this purpose, the system is formulated as a bilinear control system living on a compact homogeneous space. For the task of tracking a given orbit, a class of control Lyapunov functions is naturally defined by the notion of distance induced by the real Euclidean structure with which the homogeneous space is endowed. This construction resembles closely the Jurdjevic-Quinn stabilization technique [8] (see also [6], [13], [7], [10] for related material dealing with pure states only), although the computation of the largest invariance set via LaSalle principle is more complicated. This is a consequence of the nontrivial topology of the state space (for a single spin $1/2$ it is a sphere $\mathbb{S}^2$), implying that no (smooth) feedback design can achieve global stabilization. At most one can achieve convergence out of a singular set of isolated, repulsive points. The emphasis on the exact knowledge of the singular locus is motivated by the fact that near a singularity the convergence can be very slow. Moreover, a local design is of limited practical interest in a quantum context. For multispin systems it is shown that the tensor product nature of the state space does not complicate exceedingly the feedback synthesis. On the contrary, the singular set of the control law can be computed explicitly thanks to this tensorial structure.

While the analysis is easier for the Ising Hamiltonian, all the results are valid for different types of interactions like Heisenberg or dipole-dipole. In particular, we show how it is possible to reject unwanted coupling terms, provided they are sufficiently slow compared to the residual nonlocal part of the Hamiltonian.

Throughout the paper we consider only the case of spins that are not selectively excitable. This is clearly the most difficult case, as an rf field affects all spins and interacts with all couplings. A similar feedback synthesis for selective controls is much simpler (especially for what concerns the convergence analysis) and can be deduced by similar means.

II. MODEL FORMULATION FOR SPIN ENSEMBLES

For a single spin $1/2$ system, assume the Hamiltonian is composed of a free part H_f (the drift) and a forcing part H_c (the control term). If λ_j, $j = 0, \dots, 3$, are the (normalized) Pauli matrices (see [4]), in a suitable reference frame,

$$H_f = -(\omega_o - \omega_{rf})\lambda_3 = h^3\lambda_3 \tag{1}$$
$$H_c = u\lambda_1 \tag{2}$$

where ω_o is the precession frequency and ω_{rf} the frequency of the rf field whose amplitude u is our real valued control parameter (the phase of the rf field is fixed and kept constant). If we have a two spin $1/2$ weakly coupled system and a single rf field, calling γ_α and γ_β the gyromagnetic ratios of the two nuclear species, the Hamiltonian in the rotating frame is given by

$$H_f = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{33}\Lambda_{33} \tag{3}$$
$$H_c = uH_{c,ns} = u\left(\Lambda_{01} + \Lambda_{10}\right) \tag{4}$$

where h^{30} and h^{03} are the differences between the Larmor frequencies of each spin, call them $\omega_{o,\alpha}$ and $\omega_{o,\beta}$, and the carrier frequency ω_{rf}, $h^{30} = -(\omega_{o,\alpha} - \omega_{rf})$, $h^{03} = -(\omega_{o,\beta} - \omega_{rf})$, h^{33} represents the so-called J (or scalar) coupling and $\Lambda_{jk} = \lambda_j \otimes \lambda_k$, $j, k = 0, \ldots, 3$, form a basis of frequent use in the NMR literature under the name of product operators basis. The control Hamiltonian (4) is nonselective. Such a model is suited for homonuclear species with $\omega_{o,\alpha}$ and $\omega_{o,\beta}$ not sufficiently separated by chemical shifts. We shall focus on this model, as from a mathematical (as well as practical) point of view, it is the most difficult case to control.

Hamiltonians like (3) with only a vertical coupling are often referred to as Ising Hamiltonians. In particular, when $h^{03} = h^{30}$ the unforced system has at least one degenerate eigenvalue of multiplicity 2, regardless of the value of h^{33}. This may result in a loss of controllability and complicates also the convergence in the closed-loop system. H_f is diagonal,

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

and its diagonal elements have the meaning of energy levels of the (unperturbed) system. From [3], since $\mathrm{Graph}(H_{c,ns})$ is connected, as soon as H_f is $H_{c,ns}$-strongly regular, i.e., has energy levels that are nondegenerate and transition frequencies all different in correspondence of the nonzero elements of $H_{c,ns}$, then the system is controllable, see Theorem 3 of [3].

Lemma 1 *Consider the system* (3)-(4). *H_f is $H_{c,ns}$-strongly regular if $h^{03} \neq h^{30}$, $h^{33} \neq 0$ and $h^{33} \neq \pm(h^{03} - h^{30})/2$.*

Proof: The graph of the control Hamiltonian $H_{c,ns}$ enables the following 4 (nonoriented) transitions: $1 \leftrightarrow 2$, $1 \leftrightarrow 3$, $2 \leftrightarrow 4$, $3 \leftrightarrow 4$. Computing the energy differences in terms of the h^{03}, h^{30} and h^{33}, lack of degenerate transitions corresponds to the three inequalities stated above. $\square$

For n spin $1/2$, in a rotating frame of frequency ω_{rf}, the Ising Hamiltonian of a linear spin chain is still composed of a drift part containing the Larmor precessions (relative to ω_{rf} as in (3)) plus the J couplings between adjacent spins

$$H_f = \left(h^{0\ldots03}\Lambda_{0\ldots03} + \ldots + h^{30\ldots0}\Lambda_{30\ldots0} \right. \\ \left. + h^{0\ldots033}\Lambda_{0\ldots033} + \ldots + h^{330\ldots0}\Lambda_{330\ldots0} \right),$$

and of a forcing term along the λ_1 axis of each spin which, in the nonselective case, is

$$H_c = u H_{c,ns} = u \left(\Lambda_{0\ldots01} + \ldots + \Lambda_{10\ldots0} \right).$$

Next we introduce a particular representation of density operators, the so-called Stokes tensor parametrization, which allows to rewrite the forced Liouville equation of motion as a real bilinear control system. All details are given in [4]. For a single spin $1/2$, it is well-known that one can write the density operator ρ as a real vector of expectation values along the Pauli matrices λ_j, $j = 0, \ldots, 3$: $\rho = \varrho^j \lambda_j = \boldsymbol{\varrho} \cdot \boldsymbol{\lambda}$. In terms of $\boldsymbol{\varrho}$, and for the Hamiltonian in (1) and (2) one obtains the Bloch equations:

$$\dot{\boldsymbol{\varrho}} = -i \left(h^3 \mathrm{ad}_{\lambda_3} + u \mathrm{ad}_{\lambda_1} \right) \boldsymbol{\varrho}. \tag{5}$$

The notation "ad" in (5) originates from the notion of adjoint representation, and the adjoint operators ad_{λ_j} stand for matrices of structure constants with respect to the $\mathfrak{su}(2)$ basis given by the $-i\lambda_j$: $\mathrm{ad}_{\lambda_j}\lambda_k = [\lambda_j, \lambda_k] = \sum_{l=0}^{3} c_{jk}^l \lambda_l$. In general, the adjoint representation of a semisimple Lie algebra is a real linear isomorphic representation of the algebra. This enables us to formulate the control problem in terms of standard real bilinear control systems also for multispin systems. In fact, for 2 or more spin $1/2$ densities, a parametrization similar to the Bloch vector yields a tensor, called the Stokes tensor and also denoted by $\boldsymbol{\varrho}$: if $\rho \in \mathcal{H}_2^{\otimes 2}$, $\rho = \varrho^{jk}\Lambda_{jk} = \boldsymbol{\varrho} \cdot \boldsymbol{\Lambda}$ where $\varrho^{jk} = \mathrm{tr}\left(\rho \Lambda_{jk}\right)$ are expectation values capturing all 15 degrees of freedom of ρ along the complete set of observables $\boldsymbol{\Lambda} = \{\Lambda_{jk},\ j, k = 0, \ldots, 3\}$. Calling

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

the real skew-symmetric operators obtained from the ad_{λ_j} above and the "antiadjoint" operators aad_{λ_j} (which have a similar meaning, only involving the "symmetric" structure constants $\mathrm{aad}_{\lambda_j}\lambda_k = \{\lambda_j, \lambda_k\} = \sum_{l=0}^{3} s_{jk}^l \lambda_l$), then we obtain the following adjoint representation of the Liouville equation

$$\dot{\boldsymbol{\varrho}} = -i \left(\mathrm{ad}_{H_f} + u \mathrm{ad}_{H_{c,ns}} \right) \boldsymbol{\varrho} \tag{6}$$

or, in components,

$$\begin{aligned} \dot{\rho}^{pq} = & -i \left(h^{03}\mathrm{ad}_{\Lambda_{03}} + h^{30}\mathrm{ad}_{\Lambda_{30}} + h^{33}\mathrm{ad}_{\Lambda_{33}} \right)_{lm}^{pq} \rho^{lm} \\ & - iu \left(\mathrm{ad}_{\Lambda_{01}} + \mathrm{ad}_{\Lambda_{10}} \right)_{lm}^{pq} \rho^{lm}. \end{aligned} \tag{7}$$

By writing ρ^{jk} as a 16-vector and expanding the tensor products, a bilinear control system with drift and control vector fields that are 16×16 matrices is obtained.

Obviously, from $\mathfrak{g}_{2s} = \mathrm{Lie}\{-i\Lambda_{jk}, j, k = 0, \ldots, 3\} = \mathfrak{su}(2) \oplus \mathfrak{su}(2) \cup \mathfrak{su}(2) \otimes \mathfrak{su}(2)$, one gets for the adjoint representation $\mathrm{ad}_{\mathfrak{g}_{2s}} = \mathrm{Lie}\{-i\mathrm{ad}_{\Lambda_{jk}}, j, k = 0, \ldots, 3\} = \mathfrak{so}(3) \oplus \mathfrak{so}(3) \cup \mathfrak{so}(3) \otimes \mathfrak{so}(3)$.

The generalization to n spin $1/2$ is completely analogous: the $2^n \times 2^n$ density matrix ρ can be described by an n-index tensor $\rho = \varrho^{j_1 \ldots j_n}\Lambda_{j_1 \ldots j_n} = \boldsymbol{\varrho} \cdot \boldsymbol{\Lambda}$, each index ranging in $0, \ldots, 3$, $\Lambda_{j_1 \ldots j_n} = \lambda_{j_1} \otimes \ldots \otimes \lambda_{j_n}$. The corresponding ODE is still given by an equation like (6).

III. Feedback stabilization for spin-half systems

In this Section, we are only interested in full state feedback. Assume the entire state vector (or tensor) $\boldsymbol{\varrho}$ is available in real-time. The feedback scheme consists in choosing the amplitude profile of the rf field as a function of the desired (fixed or time-dependent) reference state $\boldsymbol{\varrho}_d$ and of the current value of the state $\boldsymbol{\varrho}$.

The topology of the manifolds discussed in this work (spheres and compact homogeneous spaces obtained by taking the "envelope" of tensor products of "affine" spheres) forbids to have globally converging smooth algorithms. For example, for the Bloch sphere $\mathbb{S}^2$ there does not exist smooth positive definite functions with less than two points having zero derivative. In control theory, the functions having such minimal number of zeros are sometimes referred to as Morse functions [9]. Hence in the simplest case of a single spin $1/2$, the Lyapunov-based design will always be characterized by the presence of at least a spurious equilibrium point, which can, however, be rendered repulsive.

Consider the system (5) from a given initial condition $\boldsymbol{\varrho}(0)$. Describe the desired orbit $\boldsymbol{\varrho}_d(t)$ (with the obvious prerequisite $\|\boldsymbol{\varrho}_d\| = \|\boldsymbol{\varrho}\|$) by means of a ODE like (5) but without forcing terms

$$\dot{\boldsymbol{\varrho}}_d = -ih_d^3 \mathrm{ad}_{\lambda_3} \boldsymbol{\varrho}_d. \tag{8}$$

This means that ϱ_d^1 and ϱ_d^2 evolve on a circle while $\varrho_d^3(t) = \varrho_d^3(0)$ is the fixed value that characterizes the orbit.

Proposition 1 *The system* (5) *with the time-varying feedback law*

$$u = k\langle\langle \boldsymbol{\varrho}_d, \, -i\mathrm{ad}_{\lambda_1} \boldsymbol{\varrho} \rangle\rangle, \tag{9}$$

where $k \in \mathbb{R}^+$, is tracking the reference orbit $\boldsymbol{\varrho}_d(t)$ given by (8) *with $h_d^3 = h^3$, in an asymptotically stable manner, for all $\boldsymbol{\varrho}(0)$ with the exception of $\boldsymbol{\varrho}(0) = -\boldsymbol{\varrho}_d(0)$ and of $\boldsymbol{\varrho}(0)$, $\boldsymbol{\varrho}_d(0)$ such that $\varrho^3(0) = \varrho_d^3 = 0$.*

Proof: In terms of Bloch vectors $\boldsymbol{\varrho}_d$, $\boldsymbol{\varrho}$, consider the following $\mathbb{S}^2$ distance between $\boldsymbol{\varrho}_d$ and $\boldsymbol{\varrho}$, see e.g. [14]:

$$d(\boldsymbol{\varrho}_d, \boldsymbol{\varrho}) = \|\boldsymbol{\varrho}_d\|^2 - \langle\langle \boldsymbol{\varrho}_d, \boldsymbol{\varrho} \rangle\rangle = \|\boldsymbol{\varrho}_d\|^2 - \boldsymbol{\varrho}_d^T \boldsymbol{\varrho}. \tag{10}$$

Take as candidate Lyapunov function the time-dependent distance (10): $V(t) = d(\boldsymbol{\varrho}_d, \boldsymbol{\varrho})$. Clearly $V \geqslant 0$ and $V = 0$ only when $\boldsymbol{\varrho}_d = \boldsymbol{\varrho}$. Since $\frac{d\|\boldsymbol{\varrho}_d\|^2}{dt} = 0$,

$$\begin{aligned} \dot{V} &= -\langle\langle \dot{\boldsymbol{\varrho}}_d, \boldsymbol{\varrho} \rangle\rangle - \langle\langle \boldsymbol{\varrho}_d, \dot{\boldsymbol{\varrho}} \rangle\rangle \\ &= -\langle\langle -ih^3 \mathrm{ad}_{\lambda_3} \boldsymbol{\varrho}_d, \boldsymbol{\varrho} \rangle\rangle - \langle\langle \boldsymbol{\varrho}_d, -i\left(h^3 \mathrm{ad}_{\lambda_3} + u\mathrm{ad}_{\lambda_1}\right) \boldsymbol{\varrho} \rangle\rangle \\ &= u\langle\langle \boldsymbol{\varrho}_d, -i\mathrm{ad}_{\lambda_1} \boldsymbol{\varrho} \rangle\rangle, \end{aligned} \tag{11}$$

because $i\mathrm{ad}_{\lambda_3}$ is skew-symmetric. Inserting (9):

$$\dot{V} = -(u)^2 = -k\left(i\boldsymbol{\varrho}_d^T \mathrm{ad}_{\lambda_1} \boldsymbol{\varrho}\right)^2 \leqslant 0.$$

Using the LaSalle's invariance principle, we want to compute the largest invariance set for (5) confined to $\mathcal{N} = \{\boldsymbol{\varrho} \text{ such that } \dot{V} = 0\}$ (and corresponding to $u = 0$), call it $\mathcal{E}$. Following the same idea of the proof of Theorem 2 of [8], in $\mathcal{N}$ it must also be

$$\frac{du}{dt} = kh^3 \boldsymbol{\varrho}_d^T [-i\mathrm{ad}_{\lambda_1}, \, -i\mathrm{ad}_{\lambda_3}] \boldsymbol{\varrho} = 0 \tag{12}$$

Notice, however, that $u = \frac{du}{dt} = 0$ yields bilinear forms as opposed to the quadratic forms of the original proof of [8]; that in addition we have the constraint of $\|\boldsymbol{\varrho}(t)\| = \mathrm{const} \neq 0$ to deal with; and that the resulting Lie algebra is composed of only skew-symmetric matrices, which applied to a point yields (out of the singularities) the tangent plane to such sphere (not $\mathbb{R}^3$). This makes the condition of [8] nonglobal. For example, both bilinear forms $u = 0$ and (12) are identically zero on the great circles $\varrho_d^3 = \varrho^3 = 0$, regardless of the values of ϱ_d^j, ϱ^j, $j = 1, 2$. Using the isospectral constraint $\|\boldsymbol{\varrho}(t)\| = \mathrm{const}$, it is easy to check that when $\varrho_d^3 \neq 0$ in $\mathcal{N}$ we have $\mathcal{E} = \{\boldsymbol{\varrho}(t) = \pm \boldsymbol{\varrho}_d(t)\}$ and the closed loop system almost globally tracks the desired orbit in an asymptotically stable manner, since $-\boldsymbol{\varrho}$ is the antipodal point to the desired position, an isolated equilibrium point rendered repulsive by (9).

□

Notice that the exact cancellation of the drift in (11) is crucial for the proof of stability. If $h_d^3 \neq h^3$, in fact, the set of critical equilibria in $\mathcal{N}$ is larger and the stability of the reference orbit is not asymptotic. This implies for example that almost global asymptotic stabilization to a point (other than north and south poles) of $\mathbb{S}^2$ is not achievable, at least with this scheme. For a single spin in a rotating frame, something similar is however possible provided ω_{rf} is tuned exactly at the Larmour frequency so that both (6) and (8) are driftless: $h^3 = -(\omega_o - \omega_{rf}) = 0$. In this case, however, the singular set is larger.

In Figg. 1-3, simulations of the closed loop system with the controller (9) are shown [1]. In particular, Fig. 3 shows the instability of the antipodal point: while $\boldsymbol{\varrho}(0) = -\boldsymbol{\varrho}_d(0)$ implies the state (dashed line) is not converging to $\boldsymbol{\varrho}_d(t)$ (dotted line), a small perturbation is enough to make $\boldsymbol{\varrho}(t)$ (solid line) converging to $\boldsymbol{\varrho}_d(t)$.

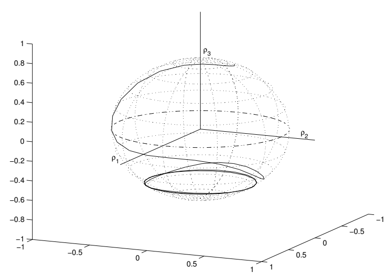

Fig. 1. Closed-loop trajectory on the Bloch sphere for the controller (9).

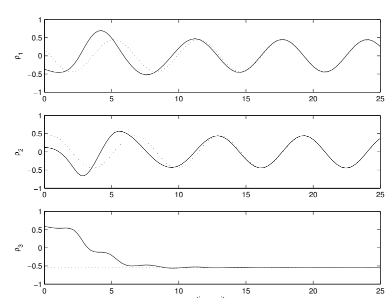

Fig. 2. The components of the Bloch vector of $\boldsymbol{\varrho}_d(t)$ (dotted line) and $\boldsymbol{\varrho}(t)$ (solid line) for the same data as in Fig. 1.

[1] All plotted signals, here and below, are suitably normalized.

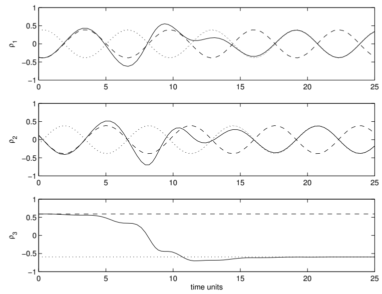

Fig. 3. The closed-loop trajectories of the system with initial state (dashed line) "antipodal" to the desired state (dotted line) $\boldsymbol{\varrho}_d(0) = -\boldsymbol{\varrho}(0)$ and from the same antipodal inital state plus a small perturbation (solid line).

A major problem of the Lyapunov design for spheres and other homogeneous spaces, is that its extension to systems with more that 2 levels makes the characterization of the region of convergence of the controller difficult to describe. See for example the N-level construction of [10]. The class of systems considered in this work, coupled spins $1/2$, makes a pleasant exception: for them the singular locus of the multispin case is a replica of the single spin case. Consider for example the two weakly coupled spin $1/2$ case. Assuming the entire state tensor $\boldsymbol{\varrho}$ is available on-line, we want to obtain feedback laws for the Hamiltonian (3)-(4). Let $\boldsymbol{\varrho}_\alpha$, $\boldsymbol{\varrho}_\beta$ and $\boldsymbol{\varrho}_{\alpha_d}$, $\boldsymbol{\varrho}_{\beta_d}$ be the reduced densities respectively of $\boldsymbol{\varrho}$ and $\boldsymbol{\varrho}_d$. We shall assume that the initial condition is a product state $\boldsymbol{\varrho}(0) = \boldsymbol{\varrho}_\alpha(0) \otimes \boldsymbol{\varrho}_\beta(0)$. The aim is to achieve asymptotically stable tracking of the following periodic orbit:

$$\begin{aligned}\dot{\boldsymbol{\varrho}}_d &= -\, i\mathrm{ad}_{H_{f_d}} \boldsymbol{\varrho}_d \\ &= -\, i \left(h_{\mathrm{d}}^{03} \mathrm{ad}_{\Lambda_{03}} + h_{\mathrm{d}}^{30} \mathrm{ad}_{\Lambda_{30}} + h_{\mathrm{d}}^{33} \mathrm{ad}_{\Lambda_{33}} \right) \boldsymbol{\varrho}_{\mathrm{d}}\end{aligned} \tag{13}$$

by means of a single control input.

Proposition 2 *Whenever H_f is $H_{c,ns}$-strongly regular, the feedback*

$$u = k \langle\!\langle \boldsymbol{\varrho}_d, \, -i\mathrm{ad}_{H_{c,ns}} \boldsymbol{\varrho} \rangle\!\rangle \tag{14}$$

with $k \in \mathbb{R}^+$, asymptotically stabilizes the system (6) *to the time-varying reference state $\boldsymbol{\varrho}_d(t)$ given by* (13) *with $H_{f_d} = H_f$, for all $\boldsymbol{\varrho}(0)$ with the exception of $\boldsymbol{\varrho}(0)$ such that $(\boldsymbol{\varrho}_\alpha(0), \, \boldsymbol{\varrho}_\beta(0)) = -(\boldsymbol{\varrho}_{\alpha_d}(0), \, \boldsymbol{\varrho}_{\beta_d}(0))$ and of all pairs $(\boldsymbol{\varrho}(0), \, \boldsymbol{\varrho}_d(0))$ having $(\varrho_\alpha^3, \, \varrho_{\alpha_d}^3) = (0, \, 0)$ and $(\varrho_\beta^3, \, \varrho_{\beta_d}^3) = (0, \, 0)$.*

Proof: We shall only sketch the arguments of the proof. As in the proof of Proposition 1, take as Lyapunov function the analogous of the distance (10), $V(t) = \|\boldsymbol{\varrho}_d\|^2 - \langle\!\langle \boldsymbol{\varrho}_d(t), \, \boldsymbol{\varrho}(t) \rangle\!\rangle$. Again, when differentiating the drift disappears,

$$\dot{V} = -u \langle\!\langle \boldsymbol{\varrho}_d, \, -i\mathrm{ad}_{H_{c,ns}} \boldsymbol{\varrho} \rangle\!\rangle, \tag{15}$$

and $\dot{V}$ is made negative semidefinite by the choice of feedback (14). Complications arise when using LaSalle invariance principle. In fact, the Jurdjevic-Quinn condition never applies to tensor product systems. Furthermore, since the reduced densities $\boldsymbol{\varrho}_\alpha(t)$ and $\boldsymbol{\varrho}_\beta(t)$ in $\mathcal{N}$ have time-varying norm due to the J coupling, neither the method used in the proof of Proposition 1 is directly applicable. However, the strong regularity of $H_{c,ns}$ guarantees that the two reduced closed-loop evolutions (each obtained tracing out a subsystem) restricted to $\mathcal{N}$ are nonidentical and hence they cannot simultaneously satisfy the constraints $u = du/dt = d^2u/dt^2 = 0$ for all times. This allows to compute explicitly the singular set, which is essentially a replica of the one in Proposition 1.

□

A typical simulation is shown in Figg. 4-5. The entire 16-state reference tensor (dotted) and the tensor of the closed-loop system (solid) are shown in Fig. 4; the tracking on the two reduced densities in Fig. 5.

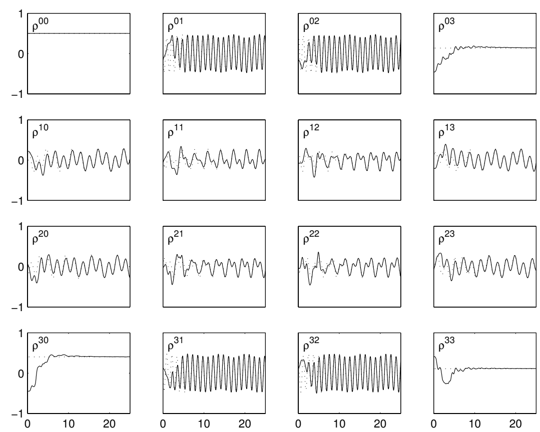

Fig. 4. The 16-components of the tensor $\boldsymbol{\varrho}_d$ (dotted lines) and $\boldsymbol{\varrho}$ (solid line) for the orbit tracking problem.

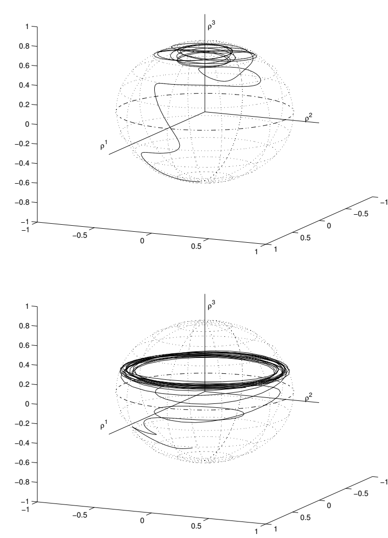

Fig. 5. Closed-loop reduced densities $\boldsymbol{\varrho}_\alpha(t)$ (left) and $\boldsymbol{\varrho}_\beta(t)$ (right) corresponding to Fig. 4.

Remark 1 Since $-i\mathrm{ad}_{H_{c,ns}}$ is local, in the feedback action (14) only the reduced densities matter.

In spite of this, the closed loop retains a nonlocality due to the J coupling. The changes in the norms $\|\boldsymbol{\varrho}_\alpha\|$ and $\|\boldsymbol{\varrho}_\beta\|$ visible in Fig. 5 are a consequence of this coupling.

A consequence of the "distinguishability" argument, mentioned in the proof of Proposition 2, is that the same tracking scheme can be used for more complicated free Hamiltonians than (3). In particular, any

$$H_f = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{jk}\Lambda_{jk}, \qquad j,\, k \neq 0 \tag{16}$$

can be used in Proposition 2, provided that $H_{f_d} = H_f$. We state it as a Corollary.

Corollary 1 *If H_f given by* (16) *is $H_{c,ns}$-strongly regular, the feedback law* (14) *asymptotically stabilizes the system $\dot{\boldsymbol{\varrho}} = -i\mathrm{ad}_{(H_f+uH_{c,ns})}\boldsymbol{\varrho}$ to the time-varying reference state $\boldsymbol{\varrho}_d(t)$ given by $\dot{\boldsymbol{\varrho}}_d = -i\mathrm{ad}_{H_f}\boldsymbol{\varrho}_d$ for all $\boldsymbol{\varrho}(0)$, except for the same singular set described in Proposition 2.*

For the Ising Hamiltonian, the generalization of the feedback stabilization algorithm to n spin $1/2$ can be done along the same lines. We only sketch the equivalent of Proposition 2 without proof, as it makes use of the same techniques used above, only the notation is more cumbersome. If we start from a product state, label the spins as α, …, ν and call ρ_α , …, ρ_ν the corresponding reduced densities, then we can arrive at the same conclusion as Proposition 2, provided all energy levels are neither equal nor equispaced.

Proposition 3 *If H_f is $H_{c,ns}$-strongly regular, the feedback*

$$u = k\langle\!\langle \boldsymbol{\varrho}_d,\, -i\mathrm{ad}_{H_{c,ns}}\boldsymbol{\varrho}\rangle\!\rangle$$

with $k \in \mathbb{R}^+$, asymptotically stabilizes the system $\dot{\boldsymbol{\varrho}} = -i(\mathrm{ad}_{H_f} + u\mathrm{ad}_{H_c})\boldsymbol{\varrho}$ to the time-varying reference orbit $\boldsymbol{\varrho}_d(t)$ given by $\dot{\boldsymbol{\varrho}}_d = -i\mathrm{ad}_{H_f}\boldsymbol{\varrho}_d$, for all $\boldsymbol{\varrho}(0)$, with the exception of the antipodal point $(\boldsymbol{\varrho}_\alpha(0), \ldots, \boldsymbol{\varrho}_\eta(0)) = -(\boldsymbol{\varrho}_{\alpha_d}(0), \ldots, \boldsymbol{\varrho}_{\eta_d}(0))$ and of all pairs $(\boldsymbol{\varrho}(0),\, \boldsymbol{\varrho}_d(0))$ having $(\varrho^3_\alpha,\, \varrho^3_{\alpha_d}) = (0,\, 0)$, …, $(\varrho^3_\eta,\, \varrho^3_{\eta_d}) = (0,\, 0)$.

IV. Suppressing unwanted weak couplings

The J-coupling used in the previous Sections is an indirect coupling mechanics, physically due to the electrons shared in the chemical bonds between the atoms. Apart from this coupling, there are other interaction mechanisms, due to the direct or electron-mediated interactions between the spins.

Consider the nonselective two spin $1/2$ Hamiltonian (16) and

$$H_{f_d} = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{jk}_d\Lambda_{jk}, \qquad j,k \neq 0. \tag{17}$$

Call $H_\delta = H_{f_d} - H_f$ the difference between the desired and the true Hamiltonian. While Corollary 1 affirms that the tracking design is still possible, we are interested here in treating the extra terms H_δ as disturbances and suppressing them by means of feedback.

Assume the frequencies h^{jk}_d are of the same order of magnitude, call it $1/\tau_d$. When the frequency of the disturbance H_δ, call it $1/\tau_\delta$ ($\simeq h^{jk}_\delta$), is about one order of magnitude smaller than that of the desired drift H_{f_d} then it can be suppressed by the control action.

Proposition 4 *Assume $\tau_\delta \simeq 10\tau_d$ and that H_f is $H_{c,ns}$-strongly regular. Then there exists a ω_{rf} and a sufficiently high gain k such that the system $\dot{\boldsymbol{\varrho}} = -i\mathrm{ad}_{(H_f+uH_{c,ns})}\boldsymbol{\varrho}$ with the feedback* (14) *can track the reference trajectory $\boldsymbol{\varrho}_d(t)$ given by $\dot{\boldsymbol{\varrho}}_d = -i\mathrm{ad}_{H_{f_d}}\boldsymbol{\varrho}_d$ and reject the disturbance H_δ.*

Proof: Since $H_{f_d} \neq H_f$, in the proof of Proposition 2 the derivative of the Lyapunov function is no longer homogeneous in the control:

$$\dot{V} = \langle\!\langle \boldsymbol{\varrho}_d,\, -i\mathrm{ad}_{H_\delta}\boldsymbol{\varrho}\rangle\!\rangle - k\langle\!\langle \boldsymbol{\varrho}_d,\, -i\mathrm{ad}_{H_{c,ns}}\boldsymbol{\varrho}\rangle\!\rangle^2. \tag{18}$$

Provided k is sufficiently high, in (18) the last term has a fast dynamics with respect to the first one. Hence, in the time scale τ_d, $\langle\!\langle \boldsymbol{\varrho}_d,\, i\mathrm{ad}_{H_\delta}\boldsymbol{\varrho}\rangle\!\rangle$ can be thought of as frozen. The drift term in (18) implies negative semidefiniteness of $\dot{V}$ is not a priori guaranteed. In particular this may happen when the controllable term of (18) vanishes, i.e., when any of the 4 reduced densities $\boldsymbol{\varrho}_{\alpha_d}$, $\boldsymbol{\varrho}_{\beta_d}$, $\boldsymbol{\varrho}_\alpha$, and $\boldsymbol{\varrho}_\beta$ approaches the λ_1 axis. Recall (Remark 1) that the amplitude of the feedback action only depends from the reduced densities. In absence of a local precession motion, the system leaves this non-convergence region only due to the coupling terms and it is not possible to guarantee a recovery from the disturbance-induced instability for all H_{f_d}. However, since $h^{03} = -(\omega_{o,\alpha} - \omega_{rf})$ and $h^{30} = -(\omega_{o,\beta} - \omega_{rf})$, it is always possible to choose ω_{rf} so that $1/\tau_\delta$ is small compared to h^{03} and h^{30}. The effect of the local precessions is to steer the corresponding reduced dynamics, both the desired and the real ones, out of the uncontrollable alignment with the λ_1 axis. Therefore, since both local closed-loop dynamics evolve fast compared to H_δ and so does H_{f_d}, the displacement due to the drift term in (18) can be rejected in the fast time scale. □

Two common coupling models often used in the literature are the Heisenberg interaction and the dipole-dipole interaction [1], [5]. For example, the Heisenberg Hamiltonian is given by

$$H_{he} = -\omega_{he}(\Lambda_{11} + \Lambda_{22} + \Lambda_{33}) \tag{19}$$

Up to (mathematically irrelevant) coefficients in front of the Λ_{jj}, H_{he} may also model the direct coupling due to the magnetic dipole-dipole interaction between the magnetic moments in solid state NMR. As an example, assume that the Hamiltonian of our system includes also the coupling (19) but that its strength is about one order of magnitude smaller that H_{f_d} and the local precession frequencies. With reference to Proposition 4, we shall assume that $10\omega_{he} \simeq (\omega_{o,\alpha} - \omega_{rf}) \simeq (\omega_{o,\beta} - \omega_{rf})$. In the rotating frame ω_{rf}, the free Hamiltonian is then

$$H_f = h^{03}\Lambda_{03} + h^{30}\Lambda_{30} + h^{33}\Lambda_{33} + h^{11}\Lambda_{11} + h^{22}\Lambda_{22} \tag{20}$$

where h^{33} includes both the J-coupling of (3) and Λ_{33} component of (19), $H_\delta = h^{11}\Lambda_{11} + h^{22}\Lambda_{22}$, and $h^{11} =$

$h^{22} = -\omega_{he}$. The control Hamiltonian is still nonselective and given by (4). A typical closed loop behavior for this choice is shown in Fig. 6. Even after the offset due to the initial condition is recovered, the system does not reach an unperturbed steady state due to the persistent excitation given be H_δ. Notice that Corollary 1 implies that the coupling

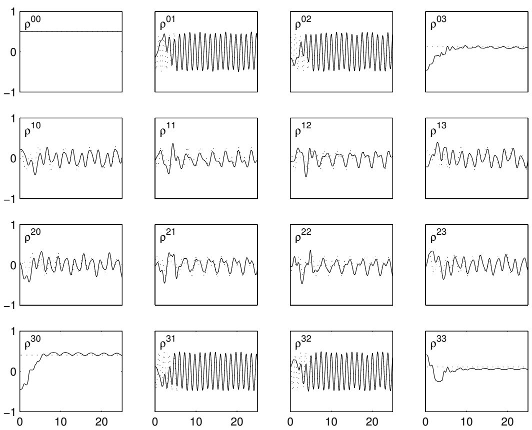

Fig. 6. The 16-components of the tensor $\boldsymbol{\varrho}_d$ (dotted lines) and $\boldsymbol{\varrho}$ (solid line) for the weak coupling suppression problem.

included in H_{f_d} needs not be restricted to the vertical Λ_{33} direction. Only the weakness of the unwanted coupling $H_f - H_{f_d}$ with respect to H_{f_d} and with respect to the local precession frequencies matters for the disturbance rejection.

V. Feedback from measurable quantities

Recall that for ensembles the measurement is a *classical* process and that its result is an expectation value. In principle, a typical NMR measurement apparatus can provide a *continuous* collective magnetization measurement in the (λ_1, λ_2) plane. While for a single spin ensemble this, together with $\|\boldsymbol{\varrho}\| = \text{const}$, allows to easily recover the entire Bloch vector, for two spin systems what is measurable depends on the nuclear species we are considering. In the nonselective case, the measurement corresponds to the output vector $y = [y_1\, y_2]^T$, where $y_j(t) = \varrho^{0j}(t) + \varrho^{j0}(t)$. Since also the controls are nonselective, a feedback from y cannot stabilize the system. However, if we can, by means of a state observer [2], access the entire 3-vectors ϱ^{0j} and ϱ^{j0} rather than just the sums of their (λ_1, λ_2) components, then several control designs are possible. We will not investigate the state estimation issue in detail here. We only notice that the system is easily seen to be observable as soon as it is controllable [2] but that due to the complicated structure of the state space, linear state observers are inadequate (they do not preserve $\|\boldsymbol{\varrho}\|$). In the heteronuclear case, the estimation of $\boldsymbol{\varrho}_\alpha$ and $\boldsymbol{\varrho}_\beta$ is an easier task than in the homonuclear one. In fact, for different spin species, different coils (tuned at the different, well-separated, precession frequencies $\omega_{o,\alpha}$, $\omega_{o,\beta}$) can be used so that both pairs $(\varrho^{01}, \varrho^{02})$ and $(\varrho^{10}, \varrho^{20})$ can be available simultaneously from direct measurements. Since in NMR experiments the initial condition $\boldsymbol{\varrho}(0)$ is always known, a naive way to recover the ϱ^{03} and ϱ^{30} components in this case is obviously to numerically integrate the control Hamiltonian H_c (H_f is not contributing to the evolution of the two vertical coordinates). Assuming $\boldsymbol{\varrho}_\alpha(t)$ and $\boldsymbol{\varrho}_\beta(t)$ are available (by this or by more sophisticated estimation procedures yet to be devised), then one could use for example the product state $\boldsymbol{\varrho}_p(t) = \boldsymbol{\varrho}_\alpha(t) \otimes \boldsymbol{\varrho}_\beta(t)$ in place of $\boldsymbol{\varrho}(t)$ in the (nonselective) feedback controller of Proposition 2

$$u = k\langle\langle \boldsymbol{\varrho}_d, \, -i\mathrm{ad}_{H_{c,ns}} \boldsymbol{\varrho}_\alpha \otimes \boldsymbol{\varrho}_\beta \rangle\rangle. \tag{21}$$

The approximation of the true state $\boldsymbol{\varrho}(t)$ with the product state $\boldsymbol{\varrho}_\alpha(t) \otimes \boldsymbol{\varrho}_\beta(t)$ corresponds to disregarding at each time the correlation that is being built by the J-coupling h^{33}. From Remark 1, this difference is to a large extent negligible.

[2]The word "observer" is used in the system-theoretic sense of state estimator.

References

[1] A. Abragam. *Principles of Nuclear Magnetism*. Oxford University Press, Oxford, UK, 1983.

[2] F. Albertini and D. D'Alessandro. Observability, measurement and parameter identification of quantum mechanical systems; application to spin networks. In *Proc. 42nd Conference on Decision and Control*, pages 439–444, Maui, Hawaii, 2003.

[3] C. Altafini. Controllability of quantum mechanical systems by root space decomposition of su(N). *Journal of Mathematical Physics*, 43(5):2051–2062, 2002.

[4] C. Altafini. Representing multiqubit unitary evolutions via Stokes tensors. *Physical Review A*, 70:032331, 2004.

[5] R. R. Ernst, G. Bodenhausen, and A. Wokaun. *Principles of Magnetic Resonance in One and Two Dimensions*. Clarendon Press, Oxford, UK, 1987.

[6] A. Ferrante, M. Pavon, and G. Raccanelli. Driving the propagator of a spin system: a feedback approach. In *Proc. of the 41st Conference on Decision and Control*, pages 46–50, Las Vegas, NV, December 2002.

[7] S. Grivopoulos and B. Bamieh. Lyapunov-based control of quantum systems. In *Proc. of the 41st Conference on Decision and Control*, pages 434–438, Maui, Hawaii, December 2004.

[8] V. Jurdjevic and J. P. Quinn. Controllability and stability. *Journal of Differential Equations*, 28:381–389, 1978.

[9] D. E. Koditschek. The application of total energy as a Lyapunov function for mechanical control systems. In J. E. Marsden, P. S. Krishnaprasad, and J. C. Simo, editors, *Dynamics and control of multibody systems*, Contemp. Math., 97. American Mathematical Society, Providence, 1989.

[10] M. Mirrahimi and P. Rouchon. Trajectory tracking for quantum systems: a Lyapunov approach. In *MTNS Mathematical Theory of Networks and Systems*, Leuven, Belgium, 2004.

[11] H. Rabitz, R. de Vivie-Riedle, M. Motzkus, and K. Kompa. Whither the future of controlling quantum phenomena? *Science*, 288:824–828, 2000.

[12] R. van Handel, J. K. Stockton, and H. Mabuchi. Feedback control of quantum state reduction. *Preprint arXiv:quant-ph/0402136*, 2004.

[13] P. Vettori. On the convergence of a feedback control strategy for multilevel quantum systems. In *MTNS Mathematical Theory of Networks and Systems*, Perpignan, France, 2002.

[14] K. Zyczkowski and W. Slomczyński. Monge metric on the sphere and geometry of quantum states. *J. Phys. A*, 34:6689, 2000.