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Moment quantization of inhomogeneous spin ensembles

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ABSTRACT

Robust excitation of a large spin ensemble is a long-standing problem in the field of quantum information science and engineering and presents a grand challenge in quantum control. A formal theoretical treatment of this task is to formulate it as an ensemble control problem defined on an infinite-dimensional space. In this paper, we present a distinct perspective to understand and control quantum ensemble systems. Instead of directly analyzing spin ensemble systems defined on a Hilbert space, we transform them to a space where the systems have reduced dimensions with distinctive network structures through the introduction of moment representations. In particular, we illustrate the idea of moment quantization for a spin ensemble and illuminate how this technique leads to a dynamically equivalent control system of moments. This equivalence enables the control of spin ensembles through the control of their moment systems, which in turn creates a new control analysis and design paradigm for quantum ensemble systems based on the use of truncated moment systems.

1. Introduction

Applications involving the control of a large ensemble of spin systems are prevalent in the domain of quantum science and technology. An essential step enabling these applications is to engineer a time-varying excitation that manipulates the dynamics and collective behavior of the spin ensemble as desired in an efficient or optimal manner. Prominent examples range from uniform or selective excitation of spins in nuclear magnetic resonance (NMR) spectroscopy and imaging (MRI) (Cory, Fahmy, & Havel, 1997; Glaser et al., 1998; Li, Ruths, Yu, Arthanari, & Wagner, 2011) to time-optimal control of spins for fast quantum transport in quantum optics and quantum information processing (Chen, Torrontegui, Stefanatos, Li, & Muga, 2011; Roos & Moelmer, 2004; Silver, Joseph, & Hoult, 1985; Stefanatos & Li, 2011, 2014).

The analysis and control of quantum ensembles are severely challenged by the complexity arising from the vast scale (typically infinite-dimension) and underactuated nature inherent in the system dynamics. This compels inventions of new methodologies of systems theory derived from a completely different angle beyond the reach of the modern paradigm. Extensive works have been conducted to overcome such bottlenecks in quantum control, including the developments of novel ensemble control (Belhadj, Salomon, & Turinici, 2015; Brockett & Khaneja, 2000; Li & Khaneja, 2009; Li, Ruths, & Glaser, 2017; Stefanatos & Li, 2011; Zhang & Li, 2021), robust control (Daems, Ruschhaupt, Sugny, & Guérin, 2013; Van Damme, Ansel, Glaser, & Sugny, 2017), computational optimal control (Khaneja, Li, Kehlet, Luy, & Glaser, 2004; Khaneja, Reiss, Kehlet, Herbruggen, & Glaser, 2005; Li, Ruths, & Stefanatos, 2009; Phelps, Royset, & Gong, 2016; Wang & Li, 2017), and learning-based methods (Chen, Dong, Long, Petersen, & Rabitz, 2014; Dong, 2020). A common theme of these emerging methods is to develop strategies to compensate for the variations in the dynamics of individual systems across the entire ensemble. This forms the basis of modern quantum pulse design and has motivated numerous novel design approaches, such as the perturbation-based robust optimal control method (Van Damme et al., 2017), iterative optimal control algorithms (Vu & Zeng, 2020; Wang & Li, 2017), and the non-harmonic Fourier synthesis approach (Zhang & Li, 2015).

Apart from these existing, successful and promising developments, in this paper, we present a distinct perspective to understand quantum ensemble systems. Instead of directly analyzing these large-scale systems defined on a Hilbert space, we transform them to a space where the systems have reduced dimensions with distinctive network structures through the introduction of *moment representations*. In particular, we present the idea of moment quantization for a continuum of spin systems and illustrate how this technique leads to a control system of moments, which is dynamically equivalent to the spin ensemble system. This equivalence enables the control of spin ensembles through the control of their moment systems, which in turn creates a new control

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design paradigm for quantum ensemble systems based on the use of the moment systems.

This paper is organized as follows. In Section 2, we represent the control of spin systems as an ensemble control problem, and introduce the notion of ensemble moments and moment quantization. We then derive moment representations of ensemble systems and reveal the nontrivial network structures induced by these representations. In Section 3, we establish a unified control paradigm for spin ensemble systems using their moment systems and demonstrate how this transition to the domain of moments facilitates the systems-theoretic analysis and control design using challenging pulse design problems in quantum control.

2. Quantization of spin ensembles via moment representations

2.1. Spin ensembles on Hilbert space

The time-evolution of a sample of nuclear spins immersed in an external magnetic field follows the Bloch equations, which forms a semiclassical model given by

$$\frac{d}{dt}M(t,\omega,\varepsilon) = \left[\omega\Omega_z + \varepsilon u(t)\Omega_y + \varepsilon v(t)\Omega_x\right]M(t,\omega,\varepsilon), \tag{1}$$

where $M(t,\omega,\varepsilon) \in \mathbb{R}^3$ denotes the magnetization vector of the spin characterized by the parameter vector $\beta = (\omega,\varepsilon)'$ at time t; $u(t)$ and $v(t)$ are the respective external control fields (i.e., radio-frequency (rf) pulses) applied in the x- and y-axis, respectively; and Ω_x, Ω_y, and Ω_z are the generators of rotation around the corresponding axis. The parameter ω denotes the Larmor frequency of the spin and ε depicts the intensity of the applied control field. In practice, variations in these parameters arise so that $\omega \in [\omega_1,\omega_2] \subset \mathbb{R}$ and $\varepsilon \in [1-\delta, 1+\delta]$ for $0<\delta<1$, referred to as Larmor dispersion and rf-inhomogeneity due to chemical shifts and inhomogeneity in the applied rf fields, respectively.

These inherent inhomogeneities make the Bloch model in (1) an infinite-dimensional system consisting of a continuum of spin systems and hinders the formal treatment of such an ensemble system with classical tools from systems theory. Consequently, establishing a new control-theoretic paradigm is compelled to enable proper analysis and design for ensemble systems. Analogous to the Fourier transform of a time-dependent signal or the Laplace transform of a time-invariant linear system to its frequency domain, in this work, we propose a kernel-based transformation that maps the ensemble system to a domain on which the analysis is eased and transparent.

2.2. Ensemble moments and moment quantization

In probability theory and statistics, the method of moments concerns with representing probability distributions of random variables in terms of infinite sequences. The function space setting of the spin ensemble in (1) renders the opportunity to adopt the *method of moments* for studying ensemble control systems. The building blocks are made of the time-dependent quantities which we refer to as *ensemble moments*. To introduce this concept in a general setting, we consider an ensemble of systems defined on a common manifold $M \subseteq \mathbb{R}^n$, indexed by the system parameter β, of the form

$$\frac{d}{dt}x(t,\beta) = F\left(t,\beta,x(t,\beta),u(t)\right), \tag{2}$$

where β takes values on a compact space $K \subset \mathbb{R}^d$, the state $x(t,\cdot)$ is an element in the space $\mathcal{F}(K,M)$ of M-valued functions defined on K, $F(t,\beta,\cdot,u(t))$ is a vector field on M for each $\beta \in K$, and $u(t) \in \mathbb{R}^m$ is a piecewise constant control input.

To place the focus on quantum systems, in this paper we assume that the state space $\mathcal{F}(K,M)$ of the ensemble system in (2) is a separable Hilbert space $\mathcal{H}$, and denote the state variable $x(t,\cdot)$ as $|x(t)\rangle$. The separability allows the expression of $|x(t)\rangle$ as a linear combination of basis elements of $\mathcal{H}$ (Folland, 2013), which inspires the idea of *moment quantization.*

Definition 1 (*Ensemble Moments*). Given an ensemble system defined on a separable Hilbert space $\mathcal{H}$ as in (2), the *kth ensemble-moment* of the system is defined by

$$m_k(t) = \langle\psi_k|x(t)\rangle, \tag{3}$$

where $k \in \mathbb{N}^d$ is a multi-index, and $\left\{|\psi_k\rangle : k \in \mathbb{N}^d\right\}$ is a basis of $\mathcal{H}$.

According to the Schwarz inequality (Folland, 2013), we have

$$|m_k(t)| = |\langle\psi_k|x(t)\rangle| = \sqrt{\langle\psi_k|\psi_k\rangle\langle x(t)|x(t)\rangle} < \infty,$$

because $|\psi_k\rangle \in \mathcal{H}$ and $|x(t)\rangle \in \mathcal{H}$. Therefore, all the ensemble moments are well-defined and finite. Geometrically, $m_k(t)$, or to be more rigorous,

$$m_k(t)\frac{|\psi_k\rangle}{\sqrt{\langle\psi_k|\psi_k\rangle}} = \frac{|\psi_k\rangle\langle\psi_k|}{\sqrt{\langle\psi_k|\psi_k\rangle}}|x(t)\rangle,$$

represents the projection of $|x(t)\rangle$ onto the subspace of $\mathcal{H}$ spanned by $|\psi_k\rangle$. We define $\hat{P}_{\psi_k}|x(t)\rangle$ by

$$\hat{P}_{\psi_k} = \frac{|\psi_k\rangle\langle\psi_k|}{\langle\psi_k|\psi_k\rangle} : \mathcal{H} \to \mathcal{H}$$

as the projection operator satisfying

$$\hat{P}^2_{\psi_k} = \frac{|\psi_k\rangle\langle\psi_k|\psi_k\rangle\langle\psi_k|}{\langle\psi_k|\psi_k\rangle^2} = \hat{P}_{\psi_k}.$$

In addition, because $\left\{|\psi_k\rangle : k \in \mathbb{N}^d\right\}$ is a basis of $\mathcal{H}$, the state $|x(t)\rangle$ can be represented in terms of the ensemble moments $m_k(t)$. To find this representation, we treat the inner product on $\mathcal{H}$ as a tensor field, equivalently a Riemannian metric, denoted by g, whose coordinate representation under this basis is given by $g_{ij} = \langle\psi_i|\psi_j\rangle$. Moreover, we denote $\left\{\langle\Psi_k| : k \in \mathbb{N}^d\right\}$ as the basis of $\mathcal{H}^*$, the dual space of $\mathcal{H}$, satisfying $\langle\Psi_i|\psi_j\rangle = \delta_{ij}$ with δ the Kronecker delta function satisfying

$$\delta_{ij} = \begin{cases} 1, & \text{if } i=j, \\ 0, & \text{otherwise.}\end{cases}$$

Correspondingly, the inner product on $\mathcal{H}^*$ under this dual basis has the coordinate representation of the form $g^{ij} = \langle\Psi_i|\Psi_j\rangle$. By viewing g as a matrix with g_{ij} being the (i,j)-entry, g^{ij} is simply the (i,j)-entry of the inverse matrix of g. Allowing a little bit abuse of notations, we also use g^{-1} to denote the metric tensor on the dual space $\mathcal{H}^*$.

Theorem 1 (*Moment Quantization*). *Given an ensemble system defined on a separable Hilbert space $\mathcal{H}$ as in (2), the state $|x(t)\rangle$ satisfies the decomposition,*

$$|x(t)\rangle = \sum_{i,j\in\mathbb{N}^d} g^{ij}m_i(t)|\psi_j\rangle, \tag{4}$$

where $\left\{|\psi_k\rangle : k \in \mathbb{N}^d\right\}$ is a basis of $\mathcal{H}$ and g^{ij} is the coordinate representation of the inner product g^{-1} on $\mathcal{H}^*$ under the dual basis $\left\{|\Psi_k\rangle : k \in \mathbb{N}^d\right\}$.

Proof. By the definitions of ensemble moments in (3) and the metric tensor g^{ij}, the right hand side of (4) yields

$$\sum_{i,j\in\mathbb{N}^d} g^{ij}m_i(t)|\psi_j\rangle = \sum_{i,j\in\mathbb{N}^d}\langle\Psi_i|\Psi_j\rangle\langle\psi_i|x(t)\rangle|\psi_j\rangle$$
$$= \sum_{i,j\in\mathbb{N}^d}\left(|\psi_j\rangle\langle\Psi_i|\right)\left(|\Psi_j\rangle\langle\psi_i|\right)|x(t)\rangle.$$

The duality between the bases $\{\langle\Psi_k|\}$ and $\{|\psi_k\rangle\}$ gives

$$|\psi_j\rangle\langle\Psi_i| = |\Psi_j\rangle\langle\psi_i| = \begin{cases}\hat{P}_{\psi_i}, & \text{if } i=j, \\ 0, & \text{otherwise,}\end{cases}$$

by using $|\Psi_i\rangle = \sum_{j\in\mathbb{N}^d} g^{ij}|\psi_j\rangle$ and $\langle\Psi_i| = \sum_{j\in\mathbb{N}^d} g_{ij}\langle\Psi_j|$. We then arrive at

$$\sum_{i,j\in\mathbb{N}^d} g^{ij}m_i(t)|\psi_j\rangle = \sum_{i\in\mathbb{N}^d}\hat{P}_{\psi_i}|x(t)\rangle = |x(t)\rangle$$

as desired. □

Theorem 1 reveals a decomposition of the state of an ensemble system into a linear combination of countably many "pure states" ($|\psi_j\rangle$) under the moment representation. This is fundamentally different from the classical quantum mechanical representation, in which pure states are eigenfunctions of the system Hamiltonian. Here, the ensemble moments m_j and states $|\psi_j\rangle$ are analogous to the energy levels and pure quantum states, respectively. Owing to this interpretation, we refer to (4) as the *moment quantization* of the ensemble system in (2).

Note that when $\{|\psi_k\rangle : k \in \mathbb{N}^d\}$ forms an orthonormal basis of the state-space $\mathcal{H}$ of the ensemble system in (2), then $g^{ij} = \delta_{ij}$ holds so that the moment quantization in (4) is reduced to a spectral representation. This further indicates explicit dependence of the moment quantization on the choices of the basis of $\mathcal{H}$. As a result, a generic question to ask is how the ensemble moments, and correspondingly the moment quantizations, change with respect to different bases of $\mathcal{H}$. This simply pertains to the question of change of coordinates in linear algebra.

Corollary 1 (*Change of Moment Coordinates*). *Consider an ensemble system defined on a separable Hilbert space $\mathcal{H}$ as in (2). Let $\{|\psi_k\rangle : k \in \mathbb{N}^d\}$ and $\{|\widetilde{\psi}_k\rangle : k \in \mathbb{N}^d\}$ be two bases of $\mathcal{H}$ satisfying $|\widetilde{\psi}_k\rangle = \sum_{i\in\mathbb{N}^d} a_{ik}|\psi_i\rangle$ for some $a_{ik} \in \mathbb{R}$ and all $k \in \mathbb{N}^d$, then the two respective ensemble moment sequences $m(t)$ and $\widetilde{m}(t)$ associated with this system under these two bases satisfy the relation*

$$\widetilde{m}_k(t) = \sum_{i\in\mathbb{N}^d} a_{ik} m_i(t). \tag{5}$$

Proof. Applying $\langle\widetilde{\psi}_k|$ to the moment quantization in (4) under the basis $\{|\psi_k\rangle : k \in \mathbb{N}^d\}$ gives

$$\begin{aligned}\widetilde{m}_k(t) = \langle\widetilde{\psi}_k|x(t)\rangle &= \sum_{i,j\in\mathbb{N}^d} g^{ij} m_i(t)\langle\widetilde{\psi}_l|\psi_j\rangle \\ &= \sum_{i,j\in\mathbb{N}^d} g^{ij} m_i(t) \sum_{l\in\mathbb{N}^d} a_{lk}\langle\psi_l|\psi_j\rangle \\ &= \sum_{i,j,l\in\mathbb{N}^d} a_{lk} g^{ij} g_{lj} m_i(t).\end{aligned}$$

Because g^{ij} is the metric tensor on the dual space $\mathcal{H}^*$ and is symmetric, i.e., $g_{lj} = g_{jl}$, we have $\sum_{j\in\mathbb{N}^d} g^{ij} g_{kj} = \sum_{j\in\mathbb{N}^d} g^{ij} g_{jk} = \delta_{ik}$, which leads to the desired conclusion, i.e., $\widetilde{m}_k(t) = \sum_{i\in\mathbb{N}^d} a_{ik} m_i(t)$. □

It is worth noting that although the moment quantization of the state $|x(t)\rangle$ depends on the metric tensor g^{-1} of $\mathcal{H}^*$, the change of moment coordinates formula in (5) is independent of both g and g^{-1}. This is due to the duality between $\mathcal{H}$ and $\mathcal{H}^*$, which eliminates the effect of g^{-1} by g on $\mathcal{H}$, as shown in the proof of Corollary 1.

Moreover, this change of moment coordinates is dramatically distinct from the regular change of coordinates. This can be best illustrated through the finite-dimensional analogy. For example, if the dimension of $\mathcal{H}$ is n, then the relation $|\widetilde{\psi}_k\rangle = \sum_{i=1}^n a_{ik}|\psi_i\rangle$ means that the linear transformation of $\mathcal{H}$ sending $|\psi_k\rangle$ to $|\widetilde{\psi}_k\rangle$, for all $k = 1, \ldots, n$, has the matrix representation A, whose (i,j)-entry is a_{ij}. This follows that for any state $|x(t)\rangle \in \mathcal{H}$, its coordinate representations $x(t) \in \mathbb{R}^n$ and $\widetilde{x}(t) \in \mathbb{R}^n$ under the two bases satisfy

$$\begin{aligned}|x(t)\rangle &= \sum_{i=1}^n x_i(t)|\psi_i\rangle = \sum_{i=1}^n \widetilde{x}_i(t)|\widetilde{\psi}_i\rangle \\ &= \sum_{i=1}^n\Big(\widetilde{x}_i(t)\sum_{j=1}^n a_{ji}|\psi_j\rangle\Big) = \sum_{j=1}^n\Big(\sum_{i=1}^n a_{ji}\widetilde{x}_i(t)\Big)|\psi_j\rangle\end{aligned}$$

so that

$$x_j(t) = \sum_{i=1}^n a_{ji}\widetilde{x}_i(t)$$

for all $j = 1, \ldots, n$, where $x_i(t)$ and $\widetilde{x}_i(t)$ are the i-entries of $x(t)$ and $\widetilde{x}(t)$, respectively. Consequently, in the matrix form, $x(t)$ and $\widetilde{x}(t)$ satisfy the *contravariant* relation, i.e., $\widetilde{x} = A^{-1}x$. However, Corollary 1 indicates that the moment sequences $m(t)$ and $\widetilde{m}(t)$ of $|x(t)\rangle$ under the two bases follow the *covariant* relation, i.e., $\widetilde{m}(t) = Am(t)$, which is exactly the rule for the change of coordinates under a dual basis of $\mathcal{H}^*$. This relation gives the interpretation of ensemble moments as elements in the dual space $\mathcal{H}^*$ and sheds light on duality between ensemble and moment systems to be discussed in the next section.

2.3. Moment representation of ensemble systems

The introduced notion of ensemble moments defined on a separable Hilbert space and the developed moment quantization in Section 2.2 allow us to transform an ensemble system, e.g., the spin ensemble in (1), to a system governed by the moment dynamics. To put this idea into a general setting, let us consider the control-affine ensemble system defined on a Hilbert space $\mathcal{H}$ of the form,

$$\frac{d}{dt}x(t,\beta) = f(\beta, x(t,\beta)) + \sum_{i=1}^r u_i(t) g_i(\beta, x(t,\beta)), \tag{6}$$

where the system parameter β takes values on a compact set $K \subset \mathbb{R}^d$, $|x(t)\rangle := x(t,\cdot)$ denotes the state, f and g_i are vector fields on $\mathcal{H}$. The spin ensemble in (1) is in this control-affine form with the drift, $f(\omega,\varepsilon,M(t,\omega,\varepsilon)) = \omega\Omega_z M(t,\omega,\varepsilon)$, and control vector fields, $g_1(\omega,\varepsilon,M(t,\omega,\varepsilon)) = \varepsilon\Omega_y M(t,\omega,\varepsilon)$ and $g_2(\omega,\varepsilon,M(t,\omega,\varepsilon)) = \varepsilon\Omega_x M(t,\omega,\varepsilon)$.

Now, let us consider a specific basis $\{|\psi_k\rangle : k \in \mathbb{N}^d\}$ of $\mathcal{H}$ and denote the space of ensemble moment sequences associated with elements in $\mathcal{H}$ by $\mathcal{M}$. Then, we define the *moment transformation* $\mathcal{L} : \mathcal{H} \to \mathcal{M}$, which assigns each function $|x\rangle \in \mathcal{H}$ a moment sequence $m \in \mathcal{M}$ by $m(t) = \mathcal{L}x(t,\cdot)$. Specifically, from Definition 1, the kth component of $m(t)$ is given by $m_k(t) = \langle\psi_k|x(t)\rangle$. Note that the map $\mathcal{L}$ is clearly well-defined as the zero state $|0\rangle \in \mathcal{H}$ is mapped to the zero sequence in $\mathcal{M}$.

Lemma 1. *The moment transformation $\mathcal{L} : \mathcal{H} \to \mathcal{M}$ is a vector space isomorphism.*

Proof. It is equivalent to proving that $\mathcal{L}$ is a bijective linear map. The linearity and subjectivity of $\mathcal{L}$ directly follow from its definition. To show the injectivity, we pick a state $|x\rangle \in \mathcal{H}$ such that $\mathcal{L}x = 0$, which gives $\langle\psi_k|x\rangle = 0$ for all $k \in \mathbb{N}^d$. Therefore, $|x\rangle$ is orthogonal to all the basis elements, and hence must be $|0\rangle$. This concludes that $\mathcal{L}$ has a trivial kernel, implying the injectivity. □

From Lemma 1, it is natural to equip $\mathcal{M}$ with the quotient topology generated by $\mathcal{L}$, under which $\mathcal{M}$ becomes a Banach space and $\mathcal{L}$ is a diffeomorphism (Lee, 2012). Under this setting, the pushforward map, equivalently the differential $\mathcal{L}_* : T\mathcal{H} \to T\mathcal{M}$ between the tangent bundles of $\mathcal{H}$ and $\mathcal{M}$ is a well-defined global diffeomorphism. This property allows us to derive the moment system associated with a control-affine ensemble system as in (6) by using the moment transformation, which yields

$$\begin{aligned}\frac{d}{dt}m(t) &= \frac{d}{dt}\mathcal{L}x(t,\beta) = \mathcal{L}_*\Big(\frac{d}{dt}x(t,\beta)\Big) \\ &= \mathcal{L}_*\Big(f(\beta,x(t,\beta)) + \sum_{i=1}^r u_i(t)g_i(\beta,x(t,\beta))\Big) \\ &= (\mathcal{L}_*f)(m(t)) + \sum_{i=1}^r u_i(t)(\mathcal{L}_*g_i)(m(t)).\end{aligned}$$

In this derivation, we used the definition and linearity of the pushforward map $\mathcal{L}_*$ (Lee, 2012). Because $\mathcal{L}_*$ is diffeomorphic, the induced vector fields $\mathcal{L}_*f$ and $\mathcal{L}_*g$ are well-defined on $\mathcal{M}$. Therefore, the derived moment system, governing the dynamics of the ensemble moment sequences, is well-defined on $\mathcal{M}$ and has an intimate relation to the original ensemble system in (6).

Proposition 1 (*Dynamic Moment Problem*). *Given the control-affine ensemble system defined on a Hilbert space $\mathcal{H}$ as in (6), then*

(1) *the associated moment system transformed by $\mathcal{L}$ is in the same control-affine form defined on $\mathcal{M}$, given by*

$$\frac{d}{dt}m(t) = \bar{f}(m(t)) + \sum_{i=1}^{r} u_i(t)\bar{g}_i(m(t)), \tag{7}$$

where $\bar{f} = \mathcal{L}_* f$ and $\bar{g}_i = \mathcal{L}_* g_i$ for all $i = 1, \dots, m$; and

(2) *there is a one-to-one correspondence between the two control-affine systems in* (6) and (7).

Proof. The proof follows directly by the fact that $\mathcal{L}_*$ is a diffeomorphism and the analysis presented above. □

This result reveals a duality between a control-affine ensemble system on $\mathcal{H}$ and its moment system represented with the moment coordinates. This equivalence paves the way for understanding and controlling ensemble systems through their moment systems.

2.4. Moment-induced network structures for spin ensembles

The notion and technique of moment quantization developed in Sections 2.2 and 2.3 provide a new angle to represent and visualize spin ensembles. To illuminate this new prospect, we consider the case in the absence of Larmor dispersion, i.e., ω is a constant, and put the spin ensemble in (1) in a rotating frame with respect to ω, that is,

$$\frac{d}{dt}M(t,\epsilon) = \left[\epsilon u(t)\Omega_y + \epsilon v(t)\Omega_x\right]M(t,\epsilon), \tag{8}$$

where $M(t,\epsilon)$ denotes the bulk magnetization vector at time t with $M(0,\epsilon) = (0,0,1)'$ for all $\epsilon \in K = [1-\delta, 1+\delta]$, where $0 < \delta < 1$, and the matrices

$$\Omega_y = \begin{bmatrix} 0 & 0 & -1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix} \quad \text{and} \quad \Omega_x = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & -1 & 0 \end{bmatrix}.$$

Now, we consider the Hilbert space $\mathcal{H}_s = L^2(K,\mathbb{R}^3) = \{f : K \to \mathbb{R}^3 \mid \|f\|_2 < \infty\}$ consisting of $\mathbb{R}^3$-valued L^2-functions, in which $\|f\|_2 = \left(\int_{1-\delta}^{1+\delta} |f(\epsilon)|^2 d\epsilon\right)^{1/2}$ with $|\cdot| : \mathbb{R}^3 \to \mathbb{R}$ being the Euclidean norm on $\mathbb{R}^3$. Because the spin ensemble in (8) evolves on a subspace of $\mathcal{H}_s$, we may construct the corresponding moment system with respect to different choices of basis of $\mathcal{H}$. Before giving explicit illustrations, we define the inner product on $\mathcal{H}$ by $\langle f|g\rangle = \int_{1-\delta}^{1+\delta} f'(\epsilon)g(\epsilon)d\epsilon$ for any $f, g \in L^2(K,\mathbb{R}^3)$, where $f'(\epsilon)$ denotes the transpose of $f(\epsilon) \in \mathbb{R}^3$. In particular, we will present three moment representations formed by monomials, Legendre polynomials, and Chebyshev polynomials, which have significant theoretical and computational implications.

2.4.1. Monomial-moment system of spin ensemble

Our first attempt is to define the ensemble moments in (3) using monomials, which are consistent with classical "statistical moments". In this case, we have the basis $\left\{|\psi_k\rangle = \epsilon^k : k \in \mathbb{N}\right\}$ of $\mathcal{H} = L^2(K,\mathbb{R}^3)$, and then the k^{th}-ensemble moment of the spin system in (8) is given by

$$m_k(t) = \langle \psi_k | M(t)\rangle = \int_{1-\delta}^{1+\delta} \epsilon^k M(t,\epsilon)d\epsilon.$$

Taking the derivative of $m_k(t)$ with respect to t yields

$$\begin{aligned} \frac{d}{dt}m_k(t) &= \frac{d}{dt}\int_{1-\delta}^{1+\delta} \epsilon^k M(t,\epsilon)d\epsilon \\ &= \int_{1-\delta}^{1+\delta} \epsilon^k \frac{d}{dt}M(t,\epsilon)d\epsilon \\ &= \int_{1-\delta}^{1+\delta} \epsilon^k \left[\epsilon u(t)\Omega_y + \epsilon v(t)\Omega_x\right]M(t,\epsilon)d\epsilon \\ &= \left[u(t)\Omega_y + v(t)\Omega_x\right]\int_{1-\delta}^{1+\delta} \epsilon^{k+1} M(t,\epsilon)d\epsilon \\ &= \left[u(t)\Omega_y + v(t)\Omega_x\right]m_{k+1}(t); \end{aligned} \tag{9}$$

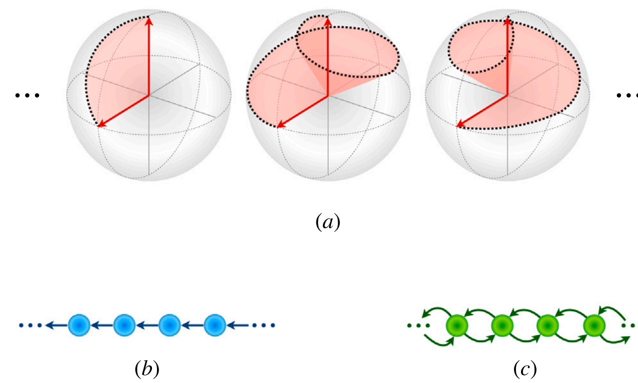

Fig. 1. Illustration of the network structure of the moment systems associated with the Bloch ensemble in (8). (a) Bloch ensemble consisting of uncoupled spin systems evolving on the unit sphere. (b) Unidirectional chain network structure of the monomial moment system in (9). (c) Bidirectional chain network structure of the Legendre and Chebyshev moment systems in (12) and (14), respectively.

or, equivalently, we can express this moment system in the tensorial form,

$$\frac{d}{dt}m(t) = \left(R \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)m(t), \tag{10}$$

where R denotes the right-shift operator and $\otimes$ is the tensor product of operators. Apparently and interestingly, under the standard basis of the moment space $\mathcal{M}$, that is, $\{e_i : i \in \mathbb{N}\}$ with e_i the sequence having 1 in the ith component and 0 elsewhere, the system in (10) also admits the matrix representation

$$\frac{d}{dt}\begin{bmatrix} m_0(t) \\ m_1(t) \\ m_2(t) \\ \vdots \end{bmatrix} = \left(U_R \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix} m_0(t) \\ m_1(t) \\ m_2(t) \\ \vdots \end{bmatrix},$$

where

$$U_R = \begin{bmatrix} 0 & 1 & & \\ & 0 & 1 & \\ & & 0 & \ddots \\ & & & \ddots \end{bmatrix}$$

is the matrix representation of the right shift operator R and $\otimes$ the Kronecker product of matrices.

As shown in Fig. 1(b), we observe that the involvement of the right shift operator R in the moment system in (10) gives rise to a chain-network structure, in which the dynamics of $m_k(t)$ depends on its successor $m_{k+1}(t)$. More interestingly, this newly unveiled structure reveals, on a conceptual level, the *moment-induced quantum entanglement* for an uncountable ensemble of uncoupled spins transformed to a moment space.

2.4.2. Legendre-moment system of spin ensemble

In addition to the canonical monomial basis, it is natural and advantageous to use orthogonal bases, such as Legendre or Chebyshev polynomials, in a Hilbert space.

Let $P_k(\epsilon)$ be the kth Legendre polynomial, defined by the recurrence relation,

$$\epsilon P_k(\epsilon) = c_k P_{k+1}(\epsilon) + c_{k-1}P_{k-1}(\epsilon) \tag{11}$$

with $P_0(\epsilon) = \sqrt{1/2}$, $P_1(\epsilon) = \sqrt{3/2}\,\epsilon$, and $c_k = (k+1)/\sqrt{(2k+1)(2k+3)}$, then $\left\{|P_k\rangle : k \in \mathbb{N}\right\}$ forms an orthogonal basis of $\mathcal{H}$. The system governing the dynamics of the Legendre-moments,

$$m_k(t) = \langle P_k | M(t)\rangle = \int_{1-\delta}^{1+\delta} P_k(\epsilon)M(t,\epsilon)d\epsilon,$$

then follows

$$\begin{aligned}\frac{d}{dt}m_k(t) &= \int_{1-\delta}^{1+\delta} P_k(\varepsilon)\frac{d}{dt}M(t,\varepsilon)d\varepsilon \\ &= \int_{1-\delta}^{1+\delta} \varepsilon P_k(\varepsilon)\left[u(t)\Omega_y + v(t)\Omega_x\right]M(t,\varepsilon)d\varepsilon \\ &= \left[u(t)\Omega_y + v(t)\Omega_x\right]\int_{1-\delta}^{1+\delta}\left[c_k P_{k+1}(\varepsilon) + c_{k-1}P_{k-1}(\varepsilon)\right]M(t,\varepsilon)d\varepsilon \\ &= \left[u(t)\Omega_y + v(t)\Omega_x\right]\left(c_k m_{k+1}(t) + c_{k-1}m_{k-1}(t)\right), \end{aligned} \tag{12}$$

where we use the recurrence relation in (11) in the third equality. Putting this system into a tensorial form gives

$$\frac{d}{dt}m(t) = \left(R \otimes \left[u(t)\Omega_y + v(t)\Omega_z\right]\right)[\begin{pmatrix}0\\c\end{pmatrix}\circ m(t)] + \left(L \otimes \left[u(t)\Omega_y + v(t)\Omega_z\right]\right)[c\circ m(t)], \tag{13}$$

where $c = (c_0, c_1, \dots)'$, L is the left-shift operator, and '∘' denotes the Hadamard product, i.e., component-wise product, of matrices. In addition, under the standard basis $\{e_i : i \in \mathbb{N}\}$ for $\mathcal{M}$, we have the matrix representation of the Legendre-moment system, given by

$$\begin{aligned}\frac{d}{dt}\begin{bmatrix}m_0(t)\\m_1(t)\\m_2(t)\\m_3(t)\\\vdots\end{bmatrix} &= \left(\begin{bmatrix}0 & c_0 & 0 & 0 & \\ c_0 & 0 & c_1 & 0 & \\ 0 & c_1 & 0 & c_2 & \\ 0 & 0 & c_2 & 0 & \\ & & & & \ddots\end{bmatrix} \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}m_0(t)\\m_1(t)\\m_2(t)\\m_3(t)\\\vdots\end{bmatrix} \\ &= \left(\begin{bmatrix}0 & 1 & 0 & 0 & \\ 0 & 0 & 1 & 0 & \\ 0 & 0 & 0 & 1 & \\ 0 & 0 & 0 & 0 & \\ & & & & \ddots\end{bmatrix} \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}0\\c_0m_1(t)\\c_1m_2(t)\\c_2m_3(t)\\\vdots\end{bmatrix} \\ &\quad + \left(\begin{bmatrix}0 & 0 & 0 & 0 & \\ 1 & 0 & 0 & 0 & \\ 0 & 1 & 0 & 0 & \\ 0 & 0 & 1 & 0 & \\ & & & & \ddots\end{bmatrix} \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}c_0m_0(t)\\c_1m_1(t)\\c_2m_2(t)\\c_3m_3(t)\\\vdots\end{bmatrix} \\ &= \left(U_R \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}0\\c_0m_1(t)\\c_1m_2(t)\\c_2m_3(t)\\\vdots\end{bmatrix} + \left(U_L \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}c_0m_0(t)\\c_1m_1(t)\\c_2m_2(t)\\c_3m_3(t)\\\vdots\end{bmatrix},\end{aligned}$$

Similar to the monomial-moment system in (9), the Legendre-moment system also preserves a chain-network structure. In particular, it is a bidirectional chain-network, illustrated in Fig. 1(c), because the Legendre-moment system in (12) or (13) is governed by both right- and left-shift operators, so that the dynamics of each ensemble-moment is determined by both of its predecessor and successor.

2.4.3. Chebyshev-moment system of spin ensemble

Chebyshev polynomials are another class of widely-used orthogonal bases for Hilbert space. Here, we choose the set of Chebyshev polynomials of the first kind, denoted $T_k(\theta)$, $k \in \mathbb{N}$, as the basis of $\mathcal{H}$, and they satisfy the recurrence relation,

$$T_{k+1}(\theta) + T_{k-1}(\theta) = 2\theta T_k(\theta)$$

with $T_0(\theta) = 1$ and $T_1(\theta) = \theta$. This is in fact equivalent to the recurrence relation in (11) for the Legendre polynomials with $c_K = 1/2$ for all $k \in \mathbb{N}$. Similar to the derivations for the monomial- and Legendre-moment systems, we immediately obtain the coordinate, tensorial, and matrix representations of the Chebyshev-moment system, given by

$$\frac{d}{dt}m_k(t) = \frac{1}{2}\left[u(t)\Omega_y + v(t)\Omega_z\right](m_{k-1}(t) + m_{k+1}(t)), \tag{14}$$

$$\frac{d}{dt}m(t) = \left((R+L) \otimes \frac{1}{2}\left[u(t)\Omega_y + v(t)\Omega_x\right]\right)m(t), \tag{15}$$

and

$$\begin{aligned}\frac{d}{dt}\begin{bmatrix}m_0(t)\\m_1(t)\\m_2(t)\\m_3(t)\\\vdots\end{bmatrix} &= \left(\begin{bmatrix}0 & \frac{1}{2} & 0 & 0 & \\ \frac{1}{2} & 0 & \frac{1}{2} & 0 & \\ 0 & \frac{1}{2} & 0 & \frac{1}{2} & \\ 0 & 0 & \frac{1}{2} & 0 & \\ & & & & \ddots\end{bmatrix} \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}m_0(t)\\m_1(t)\\m_2(t)\\m_3(t)\\\vdots\end{bmatrix} \\ &= \frac{1}{2}\left((U_R + U_L) \otimes \left[u(t)\Omega_y + v(t)\Omega_x\right]\right)\begin{bmatrix}m_0(t)\\m_1(t)\\m_2(t)\\m_3(t)\\\vdots\end{bmatrix},\end{aligned}$$

respectively.

Remark 1 (*Moment Space Induced by Hilbert Space Basis*). It is also crucial to note that a different choice of the basis of $\mathcal{H}_s$ may result in a dramatically different moment space $\mathcal{M}$. For example, the moment space under the monomial basis consists of infinite sequences $(m_0, m_1, \dots)$ satisfying the Carleman's condition,

$$(n+1)\sum_{k=0}^{n}\left[\binom{n}{k}\Delta^{n-k}m_k\right]^2 \le K,$$

for some K independent of n and for all $n \in \mathbb{N}$, where $\Delta^{n-k}m_k = \sum_{i=0}^{n-k}\binom{n-k}{i}(-1)^i m_{k+i}$. This is a direct consequence of the Hausdorff moment problem (Hausdorff, 1923). On the other hand, because the sets of Legendre and Chebyshev polynomials are orthonormal with respective to the L^2-inner product, by the Pythagorean theorem, the moment spaces induced by these bases are both ℓ^2, the space of square-summable sequences.

3. Moment control of spin ensembles

The introduced notion of moment quantization and developed moment transformations offer a formal channel to control an ensemble system through controlling its moment system. In this section, we will leverage this equivalence to build a new moment-based ensemble control paradigm and to draw the parallel of the fundamental properties between this pair of dual systems.

3.1. Ensemble and moment controllability

Since the ensemble system as in (2) is infinite-dimensional defined on the function space $\mathcal{F}(K, M)$, it is proper to define the notion of controllability, which we refer to as *ensemble controllability*, in the approximate sense.

Definition 2 (*Ensemble Controllability*). The system in (2) is said to be *ensemble controllable* on $\mathcal{F}(K, M)$, if for any $\varepsilon > 0$ and starting with any initial profile $x_0 \in \mathcal{F}(K, M)$, there exists a piecewise-constant control law $u(t)$ that steers the system into an ε-neighborhood of a desired target profile $x_F \in \mathcal{F}(K, M)$ in a finite time $T > 0$, i.e., $d(x(T, \cdot), x_F(\cdot)) < \varepsilon$, where $d : \mathcal{F}(\Omega, M) \times \mathcal{F}(K, M) \to \mathbb{R}$ is a metric on $\mathcal{F}(\Omega, M)$ (Li, Zhang, & Tie, 2020).

When $\mathcal{F}(K, M)$ is a Hilbert space, denoted $\mathcal{H}$, which is the case of particular interest in this paper, the norm $\|\cdot\| : \mathcal{H} \to \mathbb{R}$ induced by the inner product $\langle \cdot|\cdot \rangle : \mathcal{H} \times \mathcal{H} \to \mathbb{R}$ gives a metric on $\mathcal{H}$ as $\|f-g\| = \sqrt{\langle f-g|f-g \rangle}$. In the following, we will restrict our attention to ensemble controllability defined in this way.

Definition 2 refers to the notion of "approximate controllability" for any control system defined on an infinite-dimensional function space. Specifically, for the moment system defined on $\mathcal{M}$ derived in Sections 2.3 and 2.4, we may regard the moment sequences as real-valued functions defined on $\mathbb{N}^d$. This, together with the isomorphic property of the moment transformation described in Lemma 1, leads to a nice equivalence of the controllability property between the two systems.

Corollary 2. *A control-affine ensemble system defined on a separable Hilbert space $\mathcal{H}$ as in (6) is ensemble controllable if and only if the associated moment system in (7) is approximately controllable on $\mathcal{M}$.*

Proof. The proof follows directly from the isomorphic property of the moment transformation, which guarantees $\|m_F - m(t)\| = \|\mathcal{L}x_F(\cdot) - \mathcal{L}x(t, \cdot)\| = \|x_F(\cdot) - x(t, \cdot)\|$ for any desired final state $x_F \in \mathcal{H}$ of the ensemble system in (6). □

Corollary 2 then provides the theoretical guarantee of controlling ensemble systems through controlling the associated moment systems.

Remark 2 (*Controllability-preserving Moment Quantization*). Recall that a moment system always consists of countably many dynamic components, although the corresponding ensemble system contains a continuum of (uncountably many) individual systems. Therefore, the moment transformation gives rise to a controllability-preserving quantization (or dimensionality reduction) of ensemble systems.

Computationally, the countable nature of moment systems defined on $\mathcal{H}^*$ will greatly benefit and facilitate the ensemble control design through the use of truncated moment systems. In particular, the design based on such reduced finite-dimensional approximation systems will result in a desired uniform performance across the entire ensemble. This can also be understood by observing the kth moment representing the "averaged ensemble dynamics" over the subspace spanned by $|\psi_k\rangle$, where $\{|\psi_i\rangle : i \in \mathbb{N}^d\}$ is the basis of the state space of the corresponding ensemble system. This formal finite-dimensional approximation by the truncated moment representations is of particular importance to improve the design paradigm typically based on taking dense samples of the systems in the ensemble, for which a uniform performance is often not guaranteed (Zlotnik & Li, 2012). Therefore, the ensemble control inputs designed based on the moment systems will exhibit inherent persistence to the dispersion of system parameters. We will illustrate this feature below using challenging pulse design problems in nuclear magnetic resonance (NMR).

Remark 3. Conventionally, quantum ensemble systems are studied by using the state-space model defined on an infinite-dimensional function space as presented in (1). Extensive theoretical and numerical methods, integrating ideas and tools from Lie algebras, functional analysis, and algebraic and differential geometry, have been developed to overcome the challenges in analysis and control design due to the infinite-dimension and underactuated nature inherent in the system dynamics. The moment quantization method proposed in this work maps the quantum ensemble system to a dynamically-equivalent moment system, so that the quantum ensemble can be understood through the well-defined finite-dimensional truncations of the moment system. This approach shifts ensemble control to a new paradigm and opens the door for quantum pulse design by utilizing any state-of-the-art methods for finite-dimensional systems.

3.2. Pulse design for spin ensembles

Designing robust electromagnetic pulses to excite a large quantum ensemble is an essential step to enabling numerous applications in the domain of quantum control, including NMR spectroscopy and imaging, quantum optics, and quantum information processing (Cory et al., 1997; Dong & Petersen, 2010; Glaser et al., 1998). This long-standing problem is challenged by typically imperfect excitation caused by inhomogeneities inherent in the system dynamics across the ensemble, such as rf inhomogeneity and Larmor dispersion in NMR, so that sensitivity in the experiments is significantly degraded (Li et al., 2011). In this section, we will illuminate the design of uniform excitation pulses essential to many aforementioned applications by using the moment quantization technique.

3.2.1. Approximation of inverse moment quantization

In Section 2.3, we have illustrated that the moment quantization transforms the Bloch ensemble in (8) to the moment systems in the form of (10), (13), or (15) under different basis representations. This transformation opens the door for conducting systems-theoretic analysis and control design based on the truncated moment systems. To evaluate how the analysis and design perform on the original ensemble system, it is essential to construct and apply the inverse moment quantization.

To fix ideas, we define $P_k : \mathcal{M} \to \mathbb{R}^k$ as the projection operator projecting an element on $\mathcal{M}$ onto its first k components by $P_k m(t) = \big(m_0(t), \ldots, m_{k-1}(t)\big) \doteq \hat{m}(t)$, where $\mathbb{R}^k$ is considered as a vector subspace of $\mathcal{M}$ by identifying $\big(m_0(t), \ldots, m_{k-1}(t)\big)$ with the moment sequence $\big(m_0(t), \ldots, m_{k-1}(t), 0, 0, \ldots\big)$. A truncated moment system with the truncated moment sequence $\hat{m}(t)$ as the state variable can then be expressed as

$$\frac{d}{dt}\hat{m}(t) = \hat{R} \otimes \big[u(t)\Omega_y + v(t)\Omega_z\big]\, \hat{m}(t), \tag{16}$$

$$\frac{d}{dt}\hat{m}(t) = \hat{R} \otimes \big[u(t)\Omega_y + v(t)\Omega_z\big][\begin{pmatrix} 0 \\ \hat{c} \end{pmatrix} \circ \hat{m}(t)] + \hat{L} \otimes \big[u(t)\Omega_y + v(t)\Omega_z\big][\begin{pmatrix} \hat{c} \\ 0 \end{pmatrix} \circ \hat{m}(t)], \tag{17}$$

$$\frac{d}{dt}\hat{m}(t) = \frac{1}{2}(\hat{R} + \hat{L}) \otimes \big[u(t)\Omega_y + v(t)\Omega_z\big]\, \hat{m}(t), \tag{18}$$

with respect to the monomial, Legendre, and Chebyshev polynomials, respectively, where $\hat{c} = (c_0, \ldots, c_{k-1})' \in \mathbb{R}^k$ with c defined in (13) and $\hat{R}, \hat{L} \in \mathbb{R}^{k\times k}$ denote the upper-left $k \times k$ blocks of R and L, respectively. In addition, we have $\hat{m}(t) \to m(t)$ as $k \to \infty$, and hence, by Lemma 1, $\mathcal{L}^{-1}\hat{m}(t) \to \mathcal{L}^{-1}m(t) = M(t, \cdot)$. This implies that given a desired final state $M_F \in \mathcal{H}_s$ of the Bloch ensemble with the associated moment sequence $m_F \in \mathcal{M}$, a pulse $(u(t), v(t))'$ steering the truncated moment system to $\hat{m}(t)$ will drive the Bloch ensemble to a neighborhood of M_F, denoted $\mathcal{B}_r(M_F)$, where the radius r of the neighborhood depends on the truncation order.

To construct the inverse moment quantization, we partition the parameter space $K = [1-\delta, 1+\delta]$ by $1 - \delta = \varepsilon_0 < \varepsilon_2 < \cdots < \varepsilon_N = 1 + \delta$,

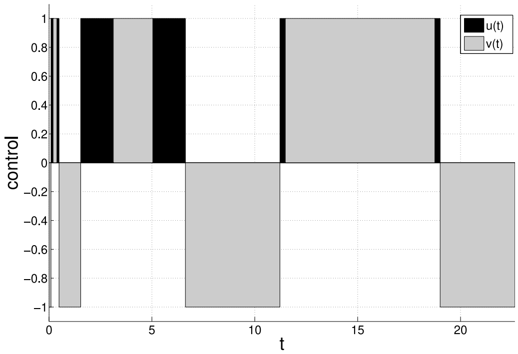

Fig. 2. The uniform $\pi/2$-pulse sequence $(u(t), v(t))'$ designed in our previous work (Zhang & Li, 2015) by using the techniques of small angle approximation and non-harmonic Fourier series for the Bloch ensemble in (8) with 40% rf-inhomogeneity ($\delta = 0.4$).

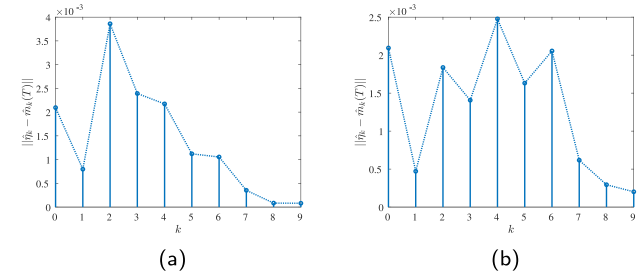

Fig. 3. Validation of the uniform $\pi/2$-pulse shown in Fig. 2 by using the truncated Legendre- and Chebyshev-moment systems in (17) and (18), respectively, with truncation order $N = 9$. (a) Component-wise ℓ^2-error, $\|\hat{m}_k(T) - \hat{\eta}_k\|$ for $k = 0, \ldots, N$, between the final and target states of the truncated Legendre-moment system. (b) Component-wise ℓ^2-error, $\|\hat{m}_k(T) - \hat{\eta}_k\|$ for $k = 0, \ldots, N$, between the final and target states of the truncated Chebyshev-moment system.

and approximate the moment sequence $m(t)$ corresponding to the state $M(t,\cdot)$ of the Bloch ensemble in (8) by using Riemann sum, i.e.,

$$\begin{aligned} m_k(t) &= \langle \psi_k | M(t) \rangle = \int_\Omega \psi_k(\varepsilon) M(t,\varepsilon) d\varepsilon \\ &= \sum_{i=1}^{N} \int_{\varepsilon_{i-1}}^{\varepsilon_i} \psi_k(\varepsilon) M(t,\varepsilon) d\varepsilon \\ &\approx \sum_{i=1}^{N} (\varepsilon_i - \varepsilon_{i-1}) \psi_k(\varepsilon_i) M(t,\varepsilon_i). \end{aligned} \tag{19}$$

Putting this approximation into a matrix form yields $m_k(t) \approx \hat{M}(t)\Psi_k$, where $\Psi_k = [(\varepsilon_1 - \varepsilon_0)\psi_k(\beta_1) \mid \cdots \mid (\varepsilon_N - \varepsilon_{N-1})\psi_k(\varepsilon_N)]' \in \mathbb{R}^N$ and $\hat{M}(t) = [\, M(t,\varepsilon_1) \mid \cdots \mid M(t,\varepsilon_N)\,] \in \mathbb{R}^{3\times N}$. Consequently, the truncated moment sequence $\hat{m}(t) = P_k m(t)$ can be expressed as

$$\begin{aligned} \hat{m}(t) &= \left[m_0(t) \mid m_1(t) \mid \cdots \mid m_{k-1}(t) \right] \\ &\approx \left[\hat{M}(t)\Psi_0 \mid \hat{M}(t)\Psi_1 \mid \cdots \mid \hat{M}(t)\Psi_{k-1} \right] \\ &= \hat{M}(t)\Psi, \end{aligned} \tag{20}$$

where $\Psi = \left[\Psi_0 \mid \Psi_1 \mid \cdots \mid \Psi_{k-1} \right] \in \mathbb{R}^{N\times k}$. The construction of the inverse moment transformation then boils down to computing the pseudoinverse $\Psi^\dagger$ of Ψ.

We will use the truncated singular value decomposition (TSVD) to compute $\Psi^\dagger$, which regularizes the ill-posedness resulting from those near-zero singular values of Ψ. Specifically, we have $\Psi = U\Sigma V'$, where $\Sigma \in \mathbb{R}^{r\times r}$ with $r \le \min\{N,k\}$ is the diagonal matrix consisting of the non-zero singular values $s_1 \ge s_2 \ge \cdots \ge s_r$ of Ψ and $U \in \mathbb{R}^{N\times r}$ and $V \in \mathbb{R}^{k\times r}$ are the semi-orthogonal matrices satisfying $U'U = V'V = I_r$, the $r \times r$ identity matrix. Then, from (20) we obtain the approximation of the inverse moment quantization, given by

$$\hat{M}(t) \approx \sum_{i=1}^{N(\eta)} \frac{\hat{m}(t) v_i}{s_i} u_i', \tag{21}$$

where $N(\eta) = \min\{i : s_1/s_i < \eta\}$ is the number of singular values determined by the chosen threshold η, and u_i and v_i denote the left- and right- singular vectors corresponding to s_i for $i = 1, \ldots, N(\eta)$.

In the next section, we will illustrate the theoretical development of the moment quantization from two alternative angels. On the one hand, we will show that a uniform excitation pulse $(u(t), v(t))'$ designed with the Bloch model in (8) will be driving the associated moment systems between the moment sequences corresponding to the ground and excited states. On the other hand, we will reverse engineering to design uniform pulses using the moment systems as in (10), (13), and (15), and then verify the resulting control designs with the Bloch ensemble in (8).

3.2.2. Uniform excitation pulses

Here, we showcase the design of uniform excitation pulses in the presence of rf-inhomogeneity. Specifically, we consider the spin ensemble following the Bloch system as in (8) with 40% variation, i.e., $\varepsilon \in K = [1-\delta, 1+\delta] = [0.6, 1.4]$. The goal is to design open-loop control fields, $(u(t), v(t))'$ such that the ensemble is steered from the equilibrium state $M_0(\varepsilon) = M(0,\varepsilon) = (0,0,1)'$ to the excited state $M_F(\varepsilon) = (1,0,0)'$ at a prescribed time $T > 0$. Methods for broadband and uniform pulse design have been extensively proposed (see Li et al., 2011 and the references therein). Here, we specifically illustrate how such a challenging ensemble control design task can be eased by translating it to the domain of moment space.

In our previous work (Zhang & Li, 2015), we presented a constructive approach based on systematic synthesis of non-harmonic Fourier series that generate the desired evolution (i.e., a sequence of rotations) from $M_0(\varepsilon)$ to $M_F(\varepsilon)$. As a validation of moment quantization, we apply a uniform $\pi/2$-pulse designed using this method, shown in Fig. 2, to the truncated moment systems induced by the Legendre polynomial and Chebyshev polynomial bases in (13) and (15), respectively, and evaluate whether this pulse will steer these moment systems between the truncated moment sequences, $\hat{m}(0)$ and $\hat{\eta}$, corresponding to $M_0(\varepsilon)$ and $M_F(\varepsilon)$, respectively. Fig. 3 shows the resulting ℓ^2-error, i.e., $\|\hat{\eta}_k - \hat{m}_k(T)\|$, $k = 0, \ldots, N$, between the desired and final moments following the designed pulse, where $\hat{\eta} = (\hat{\eta}_0, \ldots, \hat{\eta}_N)'$ denotes the truncated moment sequence of order N corresponding to $M_F(\varepsilon)$. These results numerically validate the equivalence between controlling the Bloch ensemble and its moment systems.

Alternatively, as a two-fold validation, we design uniform excitation pulses using these truncated moment systems. In particular, we will design controls to steer these systems between the moment sequences ξ and η corresponding to the equilibrium $M_0(\varepsilon)$ and the excited state $M_F(\varepsilon)$, respectively. To illustrate the main idea, we use the Chebyshev moment system in (15). The control design task can be formulated as the following optimal control problem,

$$\begin{aligned} &\min_{u(t),v(t)} \ \|\eta - m(T)\| \\ &\text{s.t.} \ \ \dot{m}(t) = \left((R+L) \otimes \frac{1}{2}\left[u(t)\Omega_y + v(t)\Omega_x \right] \right) m(t). \end{aligned} \tag{22}$$

Various computational methods can be utilized to solve for this optimal control problem, such as gradient-based methods (Khaneja et al., 2004, 2005), pseudospectral methods (Li et al., 2009, 2011; Phelps et al., 2016), and iterative algorithms (Vu & Zeng, 2020; Wang & Li, 2017, 2018). Here, we synthesize a direct method by discretizing this continuous-time problem in the time domain with the partition $0 = t_0 < t_1 < \cdots < t_n = T$ and approximate the dynamics of the truncated Chebyshev moment system of order N as in (18) by Euler discretization, that is,

$$\hat{m}(t_k) = \hat{m}(t_{k-1}) + (t_k - t_{k-1})$$

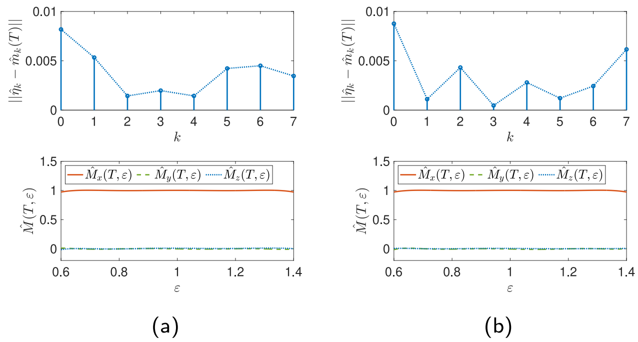

Fig. 4. Optimal control of the truncated Legendre- and Chebyshev-moment systems in (17) and (18), respectively, with the truncation order $N = 7$. (a) Component-wise ℓ^2-error, $\|\hat{\eta}_k - \hat{m}_k(T)\|$ for $k = 0, \ldots, N$, between the final and target states (top), and the final ensemble state $(\hat{M}_x(T,\varepsilon), \hat{M}_y(T,\varepsilon), \hat{M}_z(T,\varepsilon))'$ (bottom) obtained by the application of the inverse moment transformation to the final state $\hat{m}(T)$ of the truncated Legendre-moment system. (b) Component-wise ℓ^2-error, $\|\hat{\eta}_k - \hat{m}_k(T)\|$ for $k = 0, \ldots, N$, between the final and target states (top), and the final ensemble state $(\hat{M}_x(T,\varepsilon), \hat{M}_y(T,\varepsilon), \hat{M}_z(T,\varepsilon))'$ (bottom) obtained by the application of the inverse moment transformation to the final state $\hat{m}(T)$ of the truncated Chebyshev-moment system.

$$\cdot \left((\hat{R} + \hat{L}) \otimes \frac{1}{2}\left[u_k \Omega_y + v_k \Omega_x\right]\right) m(t_{k-1})$$

$$= \Big\{ I + (t_k - t_{k-1}) \cdot \left((\hat{R} + \hat{L}) \otimes \frac{1}{2}\left[u_k \Omega_y + v_k \Omega_x\right]\right) \Big\} m(t_{k-1}), \quad (23)$$

where $u_k = u(t_k)$, $v_k = v(t_k)$, and $k = 1, \ldots, n$. Note that the difference equation in (23) represents a discrete-time linear system with the system matrix $\Omega(u_k, v_k) = \left\{ I + (t_k - t_{k-1})\left((\hat{R}+\hat{L}) \otimes \frac{1}{2}\left[u(t_k)\Omega_y + v(t_k)\Omega_x\right]\right)\right\}$, and hence the final state can be concretely calculated as

$$\hat{m}(t_n) = \Omega(u_n, v_n) \cdots \Omega(u_1, v_1)\hat{m}(t_0) = \prod_{k=n}^{1} \Omega(u_k, v_k)\hat{\xi},$$

where $\hat{\xi} = (\hat{\xi}_0, \ldots, \hat{\xi}_N)'$ is the order N truncation of the initial moment sequence ξ. This yields an approximation of the continuous-time optimal control problem in (22) by a discrete unconstrained nonlinear program,

$$\min_{u_k, v_k} \left\| \hat{\eta} - \prod_{k=n}^{1} \Omega(u_k, v_k)\hat{\xi} \right\|. \quad (24)$$

We designed a uniform $\pi/2$-pulse using the optimization formulation presented in (24). Fig. 4 shows the ℓ^2-error, $\|\hat{\eta}_k - \hat{m}_k(T)\|$, for each $k = 0, \ldots, N$, and the corresponding final states $\hat{M}(T)$ of the Bloch ensemble obtained by applying the inverse moment transformation in (21) to the truncated moment sequence at time T, i.e., $\hat{m}(T) = (\hat{m}_0(T), \ldots, \hat{m}_N(T))$. In these cases, we took the truncation order $N = 7$ and the pulse duration $T = 15$ with uniform discretization of size $n = 1500$, i.e., $t_i - t_{i-1} = 0.01$ for all $i = 1, \ldots, n$, for both the Legendre- and Chebyshev-moment systems. Fig. 5 shows the designed $\pi/2$ pulses using both the truncated Legendre and Chebyshev moment systems of order $N = 7$, as well as the trajectories and final states of the spin ensemble excited by these pulses. Specifically, the performance of both pulses, which is defined as the averaged x-component, $M_x(T,\varepsilon)$, at time T over $\varepsilon \in [0.6, 1.4]$, is 0.9999.

4. Conclusion

In this paper, we propose a moment-based framework for quantization of quantum ensemble systems defined on a separable Hilbert space. The underlying idea is to project the dynamics of a quantum ensemble onto the subspace spanned by each basis element of the state-space of the ensemble system. The moment quantization induces a moment system, which is equipped with a distinctive network structure and

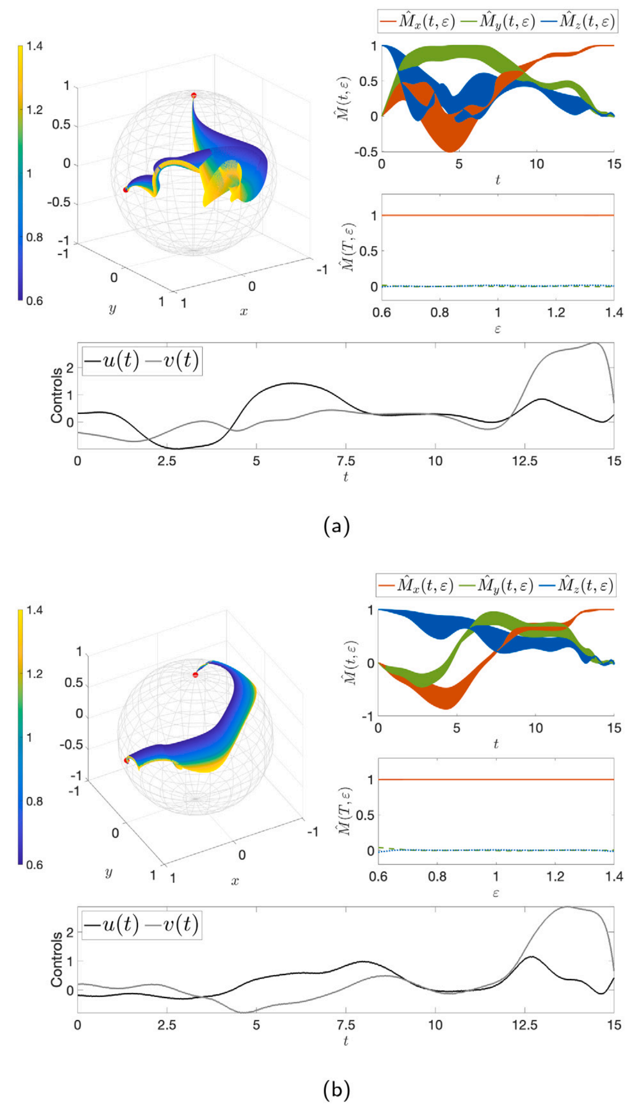

Fig. 5. Uniform $\pi/2$-excitation pulse designed by using the Legendre- and the Chebyshev-moment system. (a) Spin ensemble trajectories (top-left) and the final states $(\hat{M}_x(T,\varepsilon), \hat{M}_y(T,\varepsilon), \hat{M}_z(T,\varepsilon))'$ (top-right) following the uniform $\pi/2$-pulse $(u(t), v(t))'$ (bottom) designed by using the Legendre-moment system in (17). (b) Spin ensemble trajectories (top-left) and the final states $(\hat{M}_x(T,\varepsilon), \hat{M}_y(T,\varepsilon), \hat{M}_z(T,\varepsilon))'$ (top-right) following the uniform $\pi/2$-pulse $(u(t), v(t))'$ (bottom) designed by using the Chebyshev-moment system in (18).

dynamically equivalent to the original quantum ensemble system. We show that the control of a quantum ensemble system can be achieved by the control of its moment system and demonstrate this through pulse design in NMR spectroscopy and imaging. In particular, we design $\pi/2$-pulses robust to rf-inhomogeneity based on different representations of moment systems under the bases of Legendre and Chebyshev polynomials, which yield high fidelity across the entire ensemble. The presented notion and method of moment quantization promises a new perspective and direction towards a holistic understanding of quantum ensemble systems and enriches the repertoire of analytical and design tools in advancing quantum control theory. To date, open-loop control strategies have been the focus in ensemble control. The moment-based framework enables the application of aggregated feedback, which can be synthesized using population-level measurements, to close the loop in ensemble systems. Ideas and tools from robust control theory will play an important role in filling this gap and taking the next step towards advancing a complete theory of ensemble control.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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