# Robust Population Transfer for Coupled Spin Ensembles

Wei Zhang, Vignesh Narayanan, and Jr-Shin Li

*Abstract*— **Finely manipulating a large population of interacting nuclear spins is an extremely challenging problem arising in wide-ranging applications in quantum science and technology. Prominent examples include the design of robust excitation and inversion pulses for nuclear magnetic resonance spectroscopy and imaging, coordination of spin networks for coherence transfer, and control of superposition and entanglement for quantum computation. In this paper, by integrating the technique of small angle approximation with non-harmonic Fourier analysis, we establish a systematic method to construct robust pulse sequences that neutralize the effect of coupling variations in a spin network. In addition, we explore an alternating optimization procedure for tailoring the constructed pulses to satisfy practical design criteria. We also provide numerical examples to demonstrate the efficacy of the proposed methodology.**

## I. Introduction

Theoretical and experimental research toward control of quantum systems has been gaining significant attention among broad scientific communities. Many applications involving control of large populations of quantum systems, e.g., spin ensembles, require sensorless manipulation of their dynamic behavior with limited control variables [1, 2]. In particular, the control fields are required to be robust to the variations in the system dynamics across the entire population. This forms a bottleneck in the control design in quantum control. For example, in magnetic resonance imaging (MRI), inhomogeneity in the applied radio frequency (rf) fields may result in imperfect excitation of spin ensembles, which compromises the fidelity of the resolution of images [3, 4, 5, 6]; and in multidimensional nuclear magnetic resonance (NMR) spectroscopy, variation in the relaxation rates and dispersion in the coupling coefficients of coupled spin pairs degrade sensitivity and significantly increases measurement times [7, 8].

In the past few decades, a variety of methods have been proposed to design control fields that are robust to parameter variations in quantum systems. Among these methods, the Shinnar-Le Roux (SLR) algorithm is renowned for its robustness to engineer frequency selective pulses in MRI [9]. Based on the spinor representation of spin systems, the SLR algorithm transforms such pulse design problems into the finite impulse response (FIR) filter design. In addition to the SLR algorithm, numerical optimal control methods have also been extensively employed to achieve more specific objectives in pulse design, such as gradient ascent, Krotov algorithms, and multivariate pseudospectral methods [10, 11, 12, 13, 6, 14, 3, 4]. In addition to these computational approaches, Fourier analysis based methods were also proposed for the identification and manipulation of spin systems in the presence of rf inhomogeneity [15, 16, 17]. It is worth noting that the majority of existing works addressing the pulse design problem was developed for exciting a single spin ensemble, and limited studies were undertaken for considering problems involving the control of quantum networks, for example, coherence transfer between two spin species [3, 4].

In this paper, we develop a control-theoretic approach to designing robust pulse sequences for manipulating coupled spin ensembles, which neutralize the effect of variations in the coupling strength within a two-spin network. The proposed approach is based on the technique of small angle approximation and non-harmonic Fourier analysis. Specifically, we decompose a parameter-dependent rotation into a sequence of small angle rotations, which leads to a non-harmonic Fourier series expansion of a desired propagation. It is worth emphasizing that due to the drift in the dynamics of a coupled spin ensemble, the pulse design problem for coherence transfer becomes complicated in contrast to the problem presented in [17] for an isolated spin ensemble system. To mitigate this challenge, we present a strategy to approximate each of the decomposed small angle rotations. Then, an effective alternating optimization algorithm is presented to obtain the non-harmonic Fourier expansions approximating the desired rotations, which allows for constructing pulse sequences satisfying the prescribed design criteria.

The paper is organized as follows. In the next section, we formulate the coherence transfer task for spin networks as an ensemble control problem. We then present our pulse design method, where we introduce the small angle approximation and the resulting non-harmonic Fourier series representation. In Section III, an alternating optimization procedure is presented to compute the design parameters that are required to synthesize the pulse sequences based on practical considerations. In Section IV, we present simulation examples wherein we use our proposed design for population transfer in coupled spin ensembles.

## II. Coherence Transfer in the Presence of Coupling Variations

In this section, we propose a control-theoretic method to design robust pulses for manipulating coupled two spin sys-

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tems without relaxation that neutralize the effect of coupling variations.

### A. Ensemble Control of Coupled Spin Systems

The dynamics of a network of coupled two spin systems is governed by the equation

$$\frac{d}{dt}X(t,J) = A(t,J)X(t,J), \qquad (1)$$

where

$$A(t,J) = \left[\begin{array}{ccc|ccc} 0 & -v(t) & u(t) & 0 & 0 & 0 \\ v(t) & 0 & 0 & -J & 0 & 0 \\ -u(t) & 0 & 0 & 0 & J & 0 \\ \hline 0 & J & 0 & 0 & 0 & -u(t) \\ 0 & 0 & -J & 0 & 0 & v(t) \\ 0 & 0 & 0 & u(t) & -v(t) & 0 \end{array}\right]$$
$$= J(\Omega_{24} - \Omega_{35}) + u(t)(\Omega_{46} - \Omega_{13}) + v(t)(\Omega_{12} - \Omega_{56}),$$

$\Omega_{ij} \in \mathbb{R}^{6\times 6}$ is the matrix with $-1$ in the $ij^{\text{th}}$ entry, 1 in the $ji^{th}$ entry, and 0 elsewhere; $J$ denotes the coupling between the two spin systems varying on the interval $[1-\delta, 1+\delta]$, $0 < \delta < 1$.

Due to the variation in the coupling $J$, the system in (1) becomes a bilinear *ensemble system*, i.e. a family of bilinear systems parameterized by the coupling parameter $J \in [1-\delta, 1+\delta]$. One of the fundamental goal of manipulating an ensemble system is to design parameter-independent control inputs to steer the ensemble systems between desired parameter-dependent states. Specifically, in the context of coherence transfer, a typical control task is to construct a piecewise constant control pair $(u(t), v(t))$, called a pulse sequence, that is immune to the coupling variation $J$, and steers the spin ensemble in (1) from the constant function $X_0(J) = X(0,J) = (1,\ldots,0)' = e_1$ to the constant function $X_F(J) = X(T,J) = (0,\ldots,1)' = e_6$ for some final time $T$.

Moreover, notice that the matrix $A(t,J)$ is a skew-symmetric matrix, and hence, solutions of the system in (1) are constrained on a sphere centered at the origin of $\mathbb{R}^6$. Therefore, state transitions of the system in (1) correspond to rotations on the sphere $\mathbb{S}^5 \subset \mathbb{R}^6$, where $\mathbb{S}^5 = \{x \in \mathbb{R}^6 : \|x\| = 1\}$ is the 5-dimensional unit sphere and $\|\cdot\|$ denotes the Euclidean norm. In particular, the coherence transfer from $e_1$ to $e_6$ corresponds to a $\pi/2$ rotation with respect to $\Omega_{16}$.

### B. Small Angle Approximation and Non-harmonic Fourier Series

For convenience, we introduce the following notations: $A_{12} = \Omega_{12} - \Omega_{56}$, $A_{13} = \Omega_{46} - \Omega_{13}$, and $A_{14} = \Omega_{24} - \Omega_{35}$. To illustrate the idea of our pulse design procedure, we aim to construct a pulse sequence that generates a $J$-dependent rotation $\phi(J)$ with respect to $A_{12}$, i.e., to produce the rotation $\exp(\phi(J)A_{12})$ by using the control inputs.

Specifically, a piecewise constant control input $v(t)$ will be applied to the ensemble system in (1) to produce a sequence of small angle rotations $U_1, \ldots, U_n$ so that the total evolution $U = \prod_{k=1}^{n} U_k$ approximates the desired rotation $\exp(\phi(J)A_{12})$. To this end, for each $k$, we define

$$U_{1k} = \exp(\lambda_k J A_{14}) \exp(\frac{\beta_k}{2} A_{12}) \exp(-\lambda_k J A_{14})$$
$$U_{2k} = \exp(-\lambda_k J A_{14}) \exp(\frac{\beta_k}{2} A_{12}) \exp(\lambda_k J A_{14}), \qquad (2)$$

where $\lambda_k, \beta_k$ are design parameters (to be determined) related to the magnitude and time-period of the control pulses. Then, the Baker-Campbell-Hausdorff formula yields

$$U_{1k} = \exp(\frac{\beta_k}{2}(\cos(\lambda_k J)A_{12} + \sin(\lambda_k J)A_{24})),$$
$$U_{2k} = \exp(\frac{\beta_k}{2}(\cos(\lambda_k J)A_{12} - \sin(\lambda_k J)A_{24})).$$

If $\beta_k$ is small enough such that $U_{1k}U_{2k} \approx U_{2k}U_{1k}$, these two rotations generates a total evolution

$$U_k = U_{1k}U_{2k} \approx \exp(\beta_k \cos(\lambda_k J)A_{12}). \qquad (3)$$

This approach is referred to as the *small angle approximation* [16]. Then, successively propagating the sequence of small angle rotations $U_1, \ldots, U_n$ results in

$$U = \prod_{k=1}^{n} U_k = \exp(\sum_{k=1}^{n} \beta_k \cos(\lambda_k J)A_{12}). \qquad (4)$$

In particular, $(\beta_1, \lambda_1), \ldots, (\beta_n, \lambda_n)$ can be chosen so that

$$\phi(J) \approx \sum_{k=1}^{n} \beta_k \cos(\lambda_k J), \qquad (5)$$

and consequently, we obtain an approximation of the desired rotation $U \approx \exp(\phi(J)A_{12})$. The expansion of $\phi(J)$ in terms of sinusoidal functions in (5) is called the *non-harmonic Fourier series* expansion of $\phi(J)$. The strategy for selecting $(\beta_k, \lambda_k)$ will be discussed in detail in the next section, where we will present an alternating optimization algorithm to find the optimal values of $(\beta_k, \lambda_k)$ satisfying (5). Instead, the remaining part of this section concerns with the generation of the small angle rotations $U_{1k}$ and $U_{2k}$, or equivalently, $\exp(-\lambda_k J A_{14})$, $\exp(\lambda_k J A_{14})$, and $\exp(\frac{\beta_k}{2} A_{12})$, by using the control input $v(t)$.

At first, to generate the rotation $\exp(-\lambda_k J A_{14})$, we apply a large amplitude constant control $v(t) = v_{k1}$ for a short time period $t_{k1}$ such that $v_{k1}t_{k1} = \pi$ and

$$\exp(t_{k1} J A_{14}) \exp(\pi A_{12}) \approx \exp(\pi A_{12}) \exp(t_{k1} J A_{14}). \qquad (6)$$

Then, the two controls $u(t)$ and $v(t)$ are turned off for time $\lambda_k - 2t_{k1}$. We then apply $v(t) = -v_{k1}$ for a period of time $t_{k1}$. This procedure results in a net rotation given by

$$U_{1k1} = \exp(t_{k1} J A_{14} - \pi A_{12}) \exp((\lambda_k - 2t_{k1}) J A_{14})$$
$$\exp(t_{k1} J A_{14} + \pi A_{12}).$$

Using (6) we get

$$U_{1k1} \approx \exp(-\pi A_{12}) \exp(\lambda_k J A_{14}) \exp(\pi A_{12})$$
$$= \exp(-\lambda_k J A_{14}). \qquad (7)$$

Next, to generate the rotations $\exp(\lambda_k J A_{14}) \exp(\frac{\beta_k}{2} A_{12})$, we apply a constant control $v(t) = v_{k2}$ for a time period $t_{k2}$ so that $2v_{k2}t_{k2} = \beta_k$. Since $\beta_k$ is required to be small enough to guarantee the small angle approximation in (3), a rotation $U_{1k2}$ is produced in this step satisfying

$$\begin{aligned} U_{1k2} &= \exp(t_{k_2}(JA_{14}) + v_{k2}A_{12}) \\ &\approx \exp(t_{k_2}JA_{14}) \exp(\frac{\beta_k}{2}A_{12}). \end{aligned} \tag{8}$$

At last, evolving the system for time $\lambda_k - t_{k2}$ without any control input (i.e., $v(t) = u(t) = 0$) results in

$$U_{1k3} = \exp((t_{k2} - \lambda_{k2})JA_{14}). \tag{9}$$

The composition of these three rotations ((7), (8), (9)) results in a total evolution $U_{1k3}U_{1k2}U_{1k1} \approx U_{1k}$.

A similar strategy can be employed to generate the small angle rotations corresponding to $U_{2k}$ in (2). Furthermore, note that by using $u(t)$ together with $v(t)$, the small angle approximation and non-harmonic Fourier series based pulse design approach presented here can also be employed for the generation of $J$-dependent rotations with respect to $A_{14}$ and $A_{13}$.

**Remark 1.** *Note that unlike the problem of pulse design addressed in [17] for an isolated spin ensemble, the rotations* $\exp(\frac{\beta_k}{2}A_{12})$ *and* $\exp(-\lambda_k JA_{14})$ *in* $U_{1k}$ *and* $U_{2k}$ *given in* (2) *cannot be directly generated by applying constant control inputs. This is due to the control-independence of the drift term* $JA_{14}$ *in the system* (1). *Specifically,* $\exp(-\lambda_k JA_{14})$ *cannot to be generated by turning off the two controls* $(u(t), v(t))$*, and* $\exp(\frac{\beta_k}{2}A_{12})$ *cannot be produced by applying a constant input* $v(t) = v_k$ *because of the drift term. This necessitated the elaborate approximation procedure derived in* (7)-(9).

*Moreover, in practice, it is preferable to either turn on or turn off the control with the maximum possible amplitude for the purpose steering the ensemble from a given initial state to the desired final state with minimum energy. In this case, the magnitudes* $v_{k_1}$ *and* $v_{k_2}$ *can be made equal by appropriately adjusting their pulse timing.*

## III. Alternating Minimization for Synthesizing Parameter-Dependent Rotations

The approximation presented in (5) can be optimized to get the design parameters $(\beta_k, \lambda_k)$ required to construct the control pulses that generate the desired rotation. In this section, we present an alternating minimization approach for computing these parameters.

Consider the optimization problem with the objective function

$$\min_{\beta,\lambda} \; F(\beta, \lambda) = \Big\| \phi(J) - \sum_{k=1}^{n} \beta_k \cos(\lambda_k J) \Big\|_2^2, \tag{10}$$

where $\beta = (\beta_1, \ldots, \beta_n) \in \mathbb{R}^n$, $\lambda = (\lambda_1, \ldots, \lambda_n) \in \mathbb{R}^n$, and $\|\cdot\|_2$ denotes the $L^2$-norm on the Hilbert space $\mathcal{H} = L^2([1-\delta, 1+\delta])$, i.e., $\|f\|_2 = \sqrt{\int_{1-\delta}^{1+\delta} |f(J)|^2 dJ}$.

The first order optimality condition applied to the nonlinear program in (10) yields that

$$\begin{aligned} \frac{\partial F}{\partial \beta_i} &= \int_{1-\delta}^{1+\delta} \frac{\partial}{\partial \beta_i} \Big[ \phi(J) - \sum_{k=1}^{n} \beta_k \cos(\lambda_k J) \Big]^2 dJ \\ &= -2 \int_{1-\delta}^{1+\delta} \cos(\lambda_i J) \Big[ \phi(J) - \sum_{k=1}^{n} \beta_k \cos(\lambda_k J) \Big] dJ \end{aligned} \tag{11}$$

and

$$\begin{aligned} \frac{\partial F}{\partial \lambda_i} &= \int_{1-\delta}^{1+\delta} \frac{\partial}{\partial \lambda_i} \Big[ \phi(J) - \sum_{k=1}^{n} \beta_k \cos(\lambda_k J) \Big]^2 dJ \\ &= 2 \int_{1-\delta}^{1+\delta} \beta_i J \sin(\lambda_i J) \Big[ \phi(J) - \sum_{k=1}^{n} \beta_k \cos(\lambda_k J) \Big] dJ \end{aligned} \tag{12}$$

are vanishing for all $i = 1, \ldots, n$. We can further reduce (11) and (12) to get

$$G(\lambda)\beta = c(\lambda) \tag{13}$$

and

$$H(\lambda)\beta = d(\lambda), \tag{14}$$

respectively, where $G(\lambda), H(\lambda) \in \mathbb{R}^{n \times n}$ with their $ij^{\text{th}}$ entries defined by $G_{ij}(\lambda) = \langle \cos(\lambda_i J), \cos(\lambda_j J) \rangle$ and $H_{ij}(\lambda) = \langle J \sin(\lambda_i J), \cos(\lambda_j J) \rangle$, respectively, and $c(\lambda), d(\lambda) \in \mathbb{R}^n$ with their $i^{\text{th}}$ entries defined by $c_i(\lambda) = \langle \phi(J), \cos(\lambda_i J) \rangle$ and $d_i(\lambda) = \langle \phi(J), J \sin(\lambda_i J) \rangle$, respectively. In addition, the inner product is given by $\langle f, g \rangle = \int_{1-\delta}^{1+\delta} f(J)g(J)dJ$ for any $f, g \in \mathcal{H}$.

To solve the nonlinear program in (10), we present an alternating optimization procedure by using (13) and (14). Staring from an initial condition $(\beta^0, \lambda^0)$, we first update $\beta^0$ to $\beta^1$ by solving (13) with respect to $\beta$ for the fixed $\lambda = \lambda^0$, i.e., $\beta^1$ satisfies $G(\lambda^0)\beta^1 = c(\lambda^0)$. Then, we fix $\beta = \beta^1$ in (14), and denote the solution of this equation by $\lambda^1$, i.e., $\lambda^1$ satisfies $H(\lambda^1)\beta^1 = d(\lambda^1)$. Repeating this procedure results in a sequence $\{(\beta^k, \lambda^k) : k = 0, 1 \ldots\}$. The following theorem shows that this sequence converges to a local minimizer of the objective function $F$ of the nonlinear program in (10).

**Theorem 1.** *The sequence* $\{(\beta^k, \lambda^k) : k = 0, 1, \ldots, \}$ *generated by alternately solving the systems of equations in* (13) *and* (14) *for* $\beta$ *and* $\lambda$*, respectively, i.e., the sequence satisfying*

$$G(\lambda^k)\beta^{k+1} = c(\lambda^k)$$

*and*

$$H(\lambda^{k+1})\beta^{k+1} = d(\lambda^{k+1})$$

*for all* $k = 0, 1, \ldots,$ *converges to a local minimizer of the nonlinear program in* (10).

*Proof.* At first, notice that for any $\lambda^k$ fixed, the nonlinear program in (10) reduces to a least squares best approximation problem on the Hilbert space $\mathcal{H}$. Consequently, the projection theorem implies that the solution, denoted by $\beta^{k+1}$, of the normal equation $G(\lambda^k)\beta = c(\lambda^k)$ in (13) is a global minimizer of the objective function $F(\beta, \lambda^k)$ [18]. In particular, this gives

$$F(\beta^{k+1}, \lambda^k) \leq F(\beta^k, \lambda^k).$$

Alternatively, we fix $\beta = \beta^{k+1}$, and then solve $H(\lambda)\beta^{k+1} = d(\lambda)$ for $\lambda$. Let $\lambda^{k+1}$ be one of the solutions such that the Hessian of $F(\beta^{k+1}, \lambda)$ evaluated at $\lambda = \lambda^{k+1}$ is positive definite, then $\lambda^{k+1}$ is a local minimizer of $F(\beta^{k+1}, \lambda)$, which leads to

$$F(\beta^{k+1}, \lambda^{k+1}) \leq F(\beta^{k+1}, \lambda^k) \leq F(\beta^k, \lambda^k).$$

As a result, the sequence $\{F(\beta^k, \lambda^k) : k = 0, 1, \dots\}$ is monotonically decreasing. Moreover, because the objective function $F$ is bounded below by 0, the sequence $\{F(\beta^k, \lambda^k) : k = 0, 1, \dots\}$ converges to some nonnegative real number, i.e.,

$$\lim_{k\to\infty} F(\beta^k, \lambda^k) \to F^*$$

for some $F^* \geq 0$.

Let $\beta^* = \lim_{k\to\infty} \beta^k$ and $\lambda^* = \lim_{k\to\infty} \lambda^k$, where the convergence is guaranteed by replacing the sequence $\{(\beta^k, \lambda^k) : k = 0, 1, \dots\}$ by a subsequence if necessary, then the continuity of $F$ with respect to both $\beta$ and $\lambda$ implies $F^* = F(\beta^*, \lambda^*)$. Moreover, because $\beta^k$ and $\lambda^k$ are the solutions of the two systems of equations $G(\lambda^{k-1})\beta = c(\lambda^{k-1})$ and $H(\lambda)\beta^k = d(\lambda)$, respectively, we obtain

$$\left.\frac{\partial F}{\partial \beta}\right|_{(\beta^k,\lambda^{k-1})} = 0 \quad \text{and} \quad \left.\frac{\partial F}{\partial \lambda}\right|_{(\beta^k,\lambda^k)} = 0.$$

Taking limit from both sides of the above two equalities, as $k$ approaches to infinity, yields

$$\left.\frac{\partial F}{\partial \beta}\right|_{(\beta^*,\lambda^*)} = 0 \quad \text{and} \quad \left.\frac{\partial F}{\partial \lambda}\right|_{(\beta^*,\lambda^*)} = 0.$$

This then implies that $(\beta^*, \lambda^*)$ is a local minimizer of $F(\beta, \lambda)$. □

**Remark 2.** *When $\beta_k$ obtained by solving the nonlinear program in* (10) *is too large for* (3) *to represent a good approximation, one can divide the rotation angle $\beta_k$ into a sequence of identical smaller angles, $\beta_0$, such that $\beta_k = m\beta_0$ for some positive integer $m$. In this way, the total propagator $U$ in* (4) *becomes*

$$U = \exp\left[\beta_k \cos(\lambda_k J) A_{12}\right] \approx \Big(\exp\left[\beta_0 \cos(\lambda_k J) A_{12}\right]\Big)^m,$$
$$= \Bigg(\exp\left[\frac{\beta_0}{2}\left(A_{12}\cos(\lambda_k J) + A_{24}\sin(\lambda_k J)\right)\right]\cdot$$
$$\exp\left[\frac{\beta_0}{2}\left(A_{12}\cos(\lambda_k J) - A_{24}\sin(\lambda_k J)\right)\right]\Bigg)^m.$$

## IV. Pulse Synthesis for Coupled Spin Ensembles

In this section, we adopt the small angle approximation and non-harmonic Fourier series based method described in Section II and Section III to design control pulses that compensate for the coupling variation in networked spin pairs.

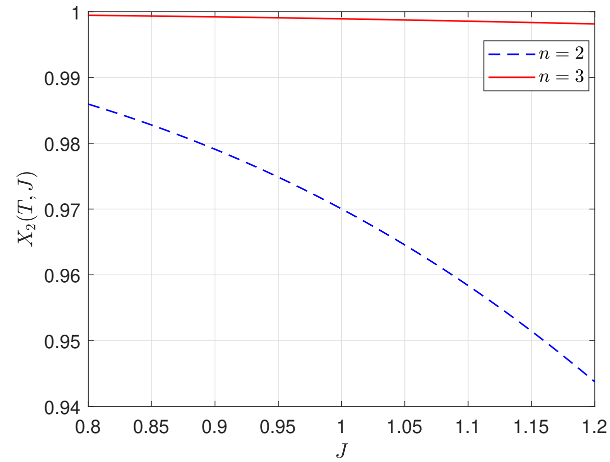


Fig. 1. The final states $X_2(T, J)$ of the coupled spin ensembles in (1) for $J \in [0.8, 1.2]$ with control pulses constructed using two and three non-harmonic Fourier terms (with magnitude of the pulses fixed) to achieve $e_1$ to $e_2$ uniformly.

As mentioned in Section II-A, one typical control task of coherence transfer is to steer the coupled spin systems in (1) from $X_0(J) = X(0, J) = (1, 0, \dots, 0)' = e_1$ to $X_F(J) = (0, \dots, 0, 1)' = e_6$ uniformly regardless of the variation in $J$. In particular, we set a 20% variation in the coupling constant, i.e., $\delta = 0.2$, and so $J \in [0.8, 1.2]$. By using the established pulse design method, we will complete this task in two steps. In the first step, we design a pulse sequence to steer the ensemble system in (1) from $X_0(J) = (1, 0, 0, 0, 0, 0)' = e_1$ to $\hat{X}_F(J) = (0, 1, 0, 0, 0, 0)' = e_2$. Then, the same design strategy can be carried over to construct pulses which realize the uniform transfer of the ensemble from $\hat{X}_F(J) = (0, 1, 0, 0, 0, 0)' = e_2$ to $X_F(J) = (0, 0, 0, 0, 0, 1)' = e_6$. In the following, we first focus on the design of the pulse sequence steering the ensemble system in (1) from $e_1$ to $e_2$.

To this end, the control sequence must induce a net $J$-dependent rotation of $\frac{\pi}{2}$ degree with respect to $A_{12}$, i.e., $\exp(\phi(J)A_{12})$ with $\phi(J) = \frac{\pi}{2}$ for all $J$. At first, to obtain the non-harmonic Fourier expansion, as shown in (5), of this constant function, we solve the nonlinear program in (10) using the alternating algorithm presented in Section III. Then, we generate the pulse sequence by using the 3 non-harmonic Fourier series of $\phi(J) = \frac{\pi}{2}$ for constructing the desired pulse sequence, respectively, with the magnitude of the pulses fixed. Fig. 1 shows the plot of the final states $X_2(T, J)$ as a function of $J$ and Fig. 2 shows the control pulses corresponding to three non-harmonic Fourier series terms.

As mentioned in Section II, to approximately generate the rotations $\exp(-\lambda_k J A_{14})$, it is necessary to apply a control with a large amplitude for a short duration. Intuitively, with a larger amplitude pulse, the rotation $\exp(-\lambda_k J A_{14})$ can be approximated more accurately. Fig. 3 shows the plot of

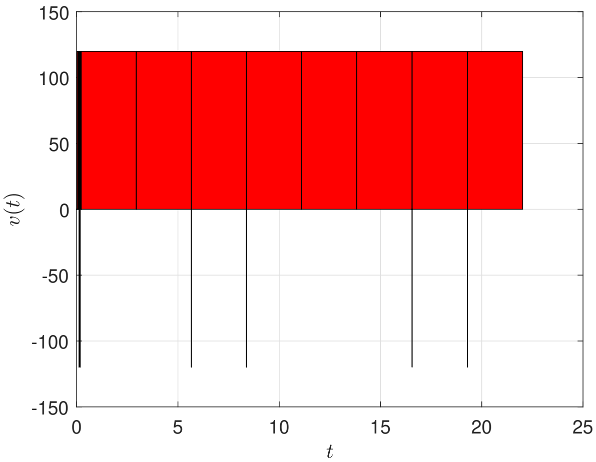


Fig. 2. The pulse sequence ($v(t)$) for the coupled spin ensemble in (1) constructed using three non-harmonic Fourier terms to achieve $e_1$ to $e_2$ rotation uniformly.

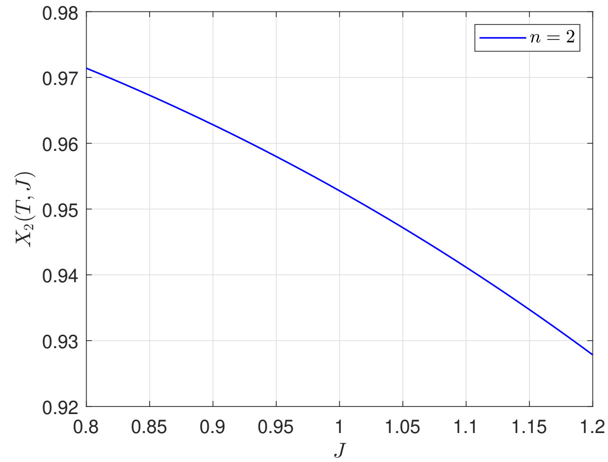


Fig. 4. The final states $X_2(T, J)$ of the coupled spin ensemble in (1) for $J \in [0.8, 1.2]$ with the control pulse constructed using two non-harmonic Fourier terms to achieve $e_1$ to $e_2$ uniformly.

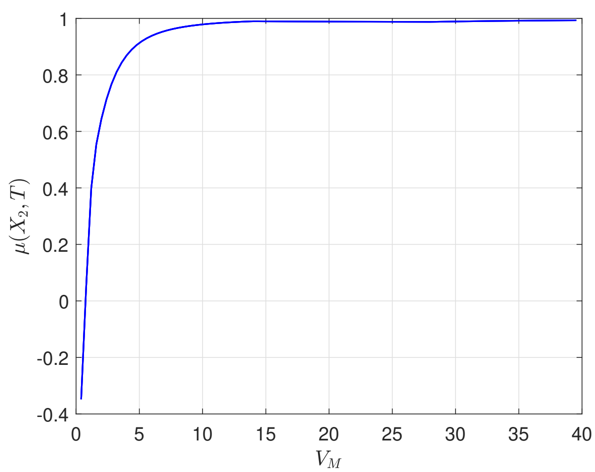


Fig. 3. The performance $\mu(X_2, T)$ of the control pulse with respect to different amplitude of the pulses $V_M$, where the control pulse is constructed using three non-harmonic Fourier terms to achieve $e_1$ to $e_2$ uniformly.

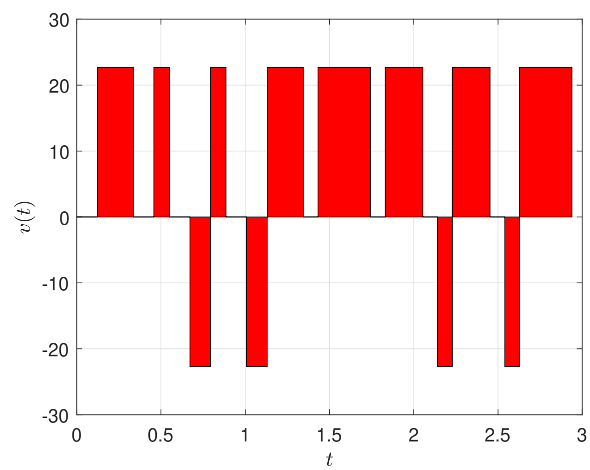


Fig. 5. The pulse sequence $v(t)$ designed for the coupled spin ensemble in (1) constructed using two non-harmonic Fourier terms to achieve $e_1$ to $e_2$ uniformly.

the performance (defined as the average value of the final state over the variation interval) versus the amplitude of the pulse sequence, in which the performance is monotonically increasing with respect to the amplitude. This indeed coincides with our intuition.

In addition, we also construct a pulse sequence by only keeping the first 2 terms of the non-harmonic Fourier series of $\phi(J) = \frac{\pi}{2}$. Fig. 4 and Fig. 5 show the second state $X_2(T, J)$ at the final time $T$ as a function of the parameter $J$, and the pulse sequence $v(t)$, respectively. The performance of this pulse is 0.9517. Moreover, by comparing Fig. 5 with Fig. 2, we notice that the amplitude of this pulse is much lower than the amplitude of the pulse obtained by using 3 non-harmonic Fourier terms, but the trade-off is in the performance which is reduced only by about 2%.

To further steer the ensemble from $e_2$ to $e_6$, a net rotation of $-\frac{\pi}{2}$ degree with respect to $A_{34} = \Omega_{26} - \Omega_{15}$ is required. For the construction of this pulse sequence, a slight modification to the small angle rotation $U_{2k}$ in (2) is required as follows

$$U_{2k} = \exp(-\lambda_k J A_{14}) \exp(-\frac{\beta_k}{2} A_{13}) \exp(\lambda_k J A_{14}). \quad (15)$$

After that, following the same design procedure, we are able to construct a pulse sequence $u(t)$ to steer the spin pair from $e_2$ to $e_6$ uniformly. Fig. 6 and 7 show the pulse sequence $u(t)$ and the sixth state $X_6(T, J)$ at the final time as a function of $J$. In particular, this pulse is constructed by using 2 non-harmonic Fourier terms, and its performance is 0.9593.

## V. CONCLUSIONS

In this paper, we develop a systematic pulse design method to accomplish coherence transfer tasks for networked spin pairs in the presence of coupling variations. This method employs the technique of small angle approximation

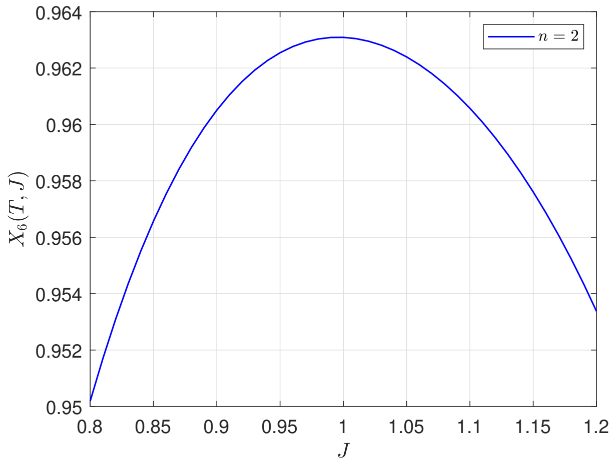


Fig. 6. The final states $X_6(T, J)$ of the coupled spin ensemble in (1) for $J \in [0.8, 1.2]$ with the control pulse constructed using two non-harmonic Fourier terms to achieve $e_2$ to $e_6$ uniformly.

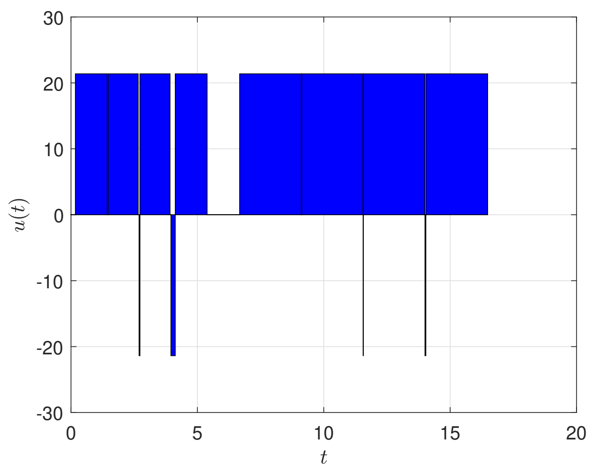


Fig. 7. The pulse sequence $u(t)$ designed for the coupled spin ensemble in (1) constructed using two non-harmonic Fourier terms to achieve $e_2$ to $e_6$ uniformly.

to expand desired parameter-dependent rotations in terms of non-harmonic Fourier series. Moreover, an alternating optimization procedure is presented to obtain the design parameters in the non-harmonic Fourier expansion that leads to a pulse sequence achieving the best performance. We also provide the convergence proof of this alternating algorithm, in which its computational efficiency is revealed. In addition to coherence transfer, the proposed method further sheds light on constructing broadband pulses in the context of (1-dimensional) NMR spectroscopy and imaging, which may provide an alternative to the well-known SLR algorithm, and also has great potential for designing piecewise constant control inputs for general bilinear ensemble systems.

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