PHYSICAL REVIEW A **86**, 022315 (2012)

Control of inhomogeneous ensembles on the Bloch sphere

Philip Owrutsky* and Navin Khaneja†
School of Engineering & Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA
(Received 30 March 2012; published 13 August 2012)

Developing control fields (pulse sequences) that can compensate for the dispersion in the parameters governing the evolution of a quantum system is an important problem in coherent spectroscopy and quantum information processing. The use of composite pulses for compensating for dispersion in system dynamics is widely known and applied. In this paper, we introduce pulse elements for correcting pulse errors. These design methods are analytical and can be used to prove arbitrarily good robust performance. Furthermore, the time to compensation is superior to existing Fourier synthesis methods, which is critical for minimizing errors due to relaxation effects.

DOI: 10.1103/PhysRevA.86.022315 PACS number(s): 03.67.Pp, 05.30.Ch, 02.30.Yy

I. INTRODUCTION

Many applications in control of quantum systems involve controlling a large ensemble by using the same control field. In practice, the elements of the ensemble could show variation in the parameters that govern the dynamics of the system. For example, in magnetic resonance experiments, the spins of an ensemble may have large dispersion in their natural frequencies (Larmor dispersion), strength of applied rf field (rf inhomogeneity), and the relaxation rates of the spins [1] to name a few. In solid state NMR spectroscopy of powders, the random distribution of orientations of internuclear vectors of coupled spins within an ensemble leads to a distribution of coupling strengths [2]. A canonical problem in control of quantum ensembles is to develop external excitations that can simultaneously steer the ensemble of systems with variation in their internal parameters from an initial state to a desired final state. These are called compensating pulse sequences as they can compensate for the dispersion in the system dynamics. From the standpoint of mathematical control theory, the challenge is to simultaneously steer a continuum of systems between points of interest with the same control signal. Typical applications are the design of excitation and inversion pulses in NMR spectroscopy in the presence of larmor dispersion and rf inhomogeneity [1,3–11] or the transfer of coherence or polarization in a coupled spin ensemble with variations in the coupling strengths [12]. In many cases of practical interest, one wants to find a control field that prepares the final state as some desired function of the parameter, for example, slice selective excitation and inversion pulses in magnetic resonance imaging [13–16]. The problem of designing excitations that can compensate for dispersion in the dynamics is a well studied subject in NMR spectroscopy and extensive literature exists on the subject of composite pulses that correct for dispersion in system dynamics [1,3–8]. Composite pulses have recently been used in quantum information processing to correct for systematic errors in single and two qubit operations [17–22].

The focus of this paper is to present pulse elements for compensating errors arising from uncertainties or imperfections in the pulse amplitude. The constructions presented here have the advantage that they are analytical and exhibit favorable performance compared to the existing analytical Fourier synthesis method [23]. Namely, the same level of rf robustness is obtained with reduced pulse length and power, which is critical for minimizing errors due to relaxation.

To fix ideas, consider an ensemble of noninteracting spin $\frac{1}{2}$ particles in a static field B_0 along the z axis and a transverse rf field, $(A(t)\cos(\phi(t)), A(t)\sin(\phi(t)))$, in the x-y plane. Let x,y,z represent the coordinates of the unit vector in the direction of the net magnetization vector of the ensemble. The dispersion in the amplitude of the rf field is given by a dispersion parameter ϵ such that $A(t) = \epsilon A_0(t)$ where $\epsilon \in [1-\delta, 1+\delta]$, for $\delta > 0$. The Bloch equations for the ensemble take the form

$$\frac{d}{dt}\begin{bmatrix} x \\ y \\ z \end{bmatrix} = \begin{bmatrix} 0 & -\omega & \epsilon u(t) \\ \omega & 0 & -\epsilon v(t) \\ -\epsilon u(t) & \epsilon v(t) & 0 \end{bmatrix}\begin{bmatrix} x \\ y \\ z \end{bmatrix}, \tag{1}$$

where

$$(u(t), v(t)) = \gamma(A_0(t)\cos(\phi(t)), A_0(t)\sin(\phi(t))).$$

Let $X(t)$ denote the unit vector $(x(t), y(t), z(t))$. Consider now the problem of designing controls $u(t)$ and $v(t)$ that simultaneously steer an ensemble of such systems with dispersion in the strength of their rf field from an initial state $X(0) = (0,0,1)$ to a final state $X_F = (1,0,0)$ [9]. This problem raises interesting questions about controllability, i.e., showing that in spite of bounds on the strength of the rf field, $\sqrt{u^2(t) + v^2(t)} \leqslant A_{\max}$, there exist excitations $(u(t), v(t))$, which simultaneously steer all the systems with dispersion in ϵ, to a ball of desired radius r around the final state $(1,0,0)$ in a finite time (which may depend on $A_{\max}$, B, δ, and r). Besides steering the ensemble between two points, we can ask for a control field that steers an initial distribution of the ensemble to a final distribution, i.e., different elements of the ensemble now have different initial and final states depending on the value of the their dispersion parameter ϵ. The initial and final state of the ensemble are described by functions $X_0(\epsilon)$ and $X_F(\epsilon)$, respectively. Consider the problem of steering an initial distribution $X_0(\epsilon)$ to within a desired distance r of a target function $X_F(\epsilon)$ by appropriate choice of controls in Eq. (1). We use the L2 norm as our error metric

$$E = \sqrt{\int_{1-\delta}^{1+\delta} ||X_F(\epsilon) - X_{\text{target}}(\epsilon)||^2 d\epsilon}. \tag{2}$$

*philip.owrutsky@gmail.com
†navin@eecs.harvard.edu; http://hrl.harvard.edu/∼navin

If a system with dispersion in its parameters can be steered between states that have dependency on the dispersion parameter arbitrarily well, then we say that the system is ensemble controllable with respect to those parameters.

We present pulse elements that compensate for inhomogeneity or uncertainty in the amplitude of the rf field. The presented method extends known techniques for pulse sequences that are robust to rf inhomogeneity. The methods presented may also find applications in anisotropy compensating pulse design or in solid state NMR experiments [24].

II. PULSE ELEMENTS FOR COMPENSATING RF INHOMOGENEITY

In this section we present two methods for producing ϵ robust rotations. The first is the previously known Fourier synthesis methods [23] and the second is a modified method using δ modulation that we call modified Fourier synthesis. The latter will be shown to have favorable pulse duration to compensation properties.

A. Fourier synthesis method

To fix ideas, we begin by considering the Bloch equations in a rotating frame with only rf inhomogeneity and no Larmor dispersion:

$$\dot{X} = \epsilon(u(t)\Omega_y + v(t)\Omega_x)X, \tag{3}$$

where

$$\Omega_x = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & 1 & 0 \end{bmatrix}, \quad \Omega_y = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix},$$

$$\Omega_z = \begin{bmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

are the generators of rotation around x, y, and z axis, respectively.

We define the pulse elements [23]

$$U_1(\beta_k,\gamma_k) = \exp(-\gamma_k\epsilon\Omega_x)\exp\left(\frac{\beta_k}{2}\epsilon\Omega_y\right)\exp(\gamma_k\epsilon\Omega_x), \tag{4}$$

$$U_2(\beta_k,\gamma_k) = \exp(\gamma_k\epsilon\Omega_x)\exp\left(\frac{\beta_k}{2}\epsilon\Omega_y\right)\exp(-\gamma_k\epsilon\Omega_x), \tag{5}$$

which correspond to directly accessible evolutions.

For suitably small β_k, we have

$$V_k = U_2U_1 \sim \exp(\epsilon\beta_k\cos(\gamma_k\epsilon)\Omega_y). \tag{6}$$

To effect a larger amplitude rotation, we consider the sequence of transformations,

$$\Phi_1 \equiv \prod_k (V_k)^{n_k} \sim \exp\left(\epsilon\sum_k \alpha_k\cos(\gamma_k\epsilon)\Omega_y\right), \tag{7}$$

where $\alpha_k = n\beta_k$. In practice, $\beta_k < \frac{\pi}{10}$ is suitably small and results in an error that is less than 1 in the L2 sense in Eq. (6). Now, the coefficients α_k can be chosen so that

$$\epsilon\sum_k \alpha_k\cos(\gamma_k\epsilon) \approx \theta \tag{8}$$

for $1-\delta \leqslant \epsilon \leqslant 1+\delta$, with $0<\delta<1$.

Therefore,

$$\Phi_1 \sim \exp(\theta\Omega_y) \tag{9}$$

approximately independent of ϵ. The dependence on ϵ can be made arbitrarily small by increasing the pulse length and extending the number of terms in the summation, leading to pulses that are immune to dispersions in the rf amplitude as claimed.

B. Modified Fourier synthesis using δ modulation

In this section we develop a modified Fourier synthesis method that will be shown to have favorable time-robustness properties to the original Fourier synthesis technique. To this end we consider the following system:

$$\dot{Y} = (\epsilon u(t)\Omega_x + v(t)\Omega_z)Y, \tag{10}$$

which corresponds to a system with one pure control by way of $v(t)$ and one control with dispersion, $\epsilon u(t)$. We will apply a similar Fourier synthesis method (FSM) analysis on the system and show that this results in a modified Hamiltonian to that of the previous section, the advantages of which will be discussed in subsequent sections. We then show how the previous system with both controls exhibiting dispersion (3) can be transformed into (10) by an appropriate change of coordinates.

Consider the modified transformations,

$$\tilde{U}_1 = \exp(-\gamma_k\epsilon\Omega_x)\exp\left(\frac{\beta_k}{2}\Omega_z\right)\exp(\gamma_k\epsilon\Omega_x), \tag{11}$$

$$\tilde{U}_2 = \exp(\gamma_k\epsilon\Omega_x)\exp\left(-\frac{\beta_k}{2}\Omega_z\right)\exp(-\gamma_k\epsilon\Omega_x). \tag{12}$$

By again choosing β_k sufficiently small, we have

$$\tilde{V}_k = \tilde{U}_2\tilde{U}_1 \sim \exp(\beta_k\sin(\gamma_k\epsilon)\Omega_y). \tag{13}$$

Applying a sequence of such transformations

$$\Phi_2 \equiv \prod_k (\tilde{V}_k)^{n_k} \sim \exp\left(\sum_k \alpha_k\sin(\gamma_k\epsilon)\Omega_y\right), \tag{14}$$

where again $\alpha_k = n\beta_k$ is used to control the error from the approximation in (13). Now, the coefficients α_k and γ_k can be chosen so that

$$\sum_k \alpha_k\sin(\gamma_k\epsilon) \approx \theta, \tag{15}$$

over the range of ϵ of interest $1-\delta \leqslant \epsilon \leqslant 1+\delta$ resulting in a robust rotation. We point out that (15) resembles (8), but no longer contains an ϵ factor external to the trigonometric argument and that cos has been replaced with sin.

To see how (10) can be generated from (3), we return to (3) and let $A = \gamma A_0$,

$$\dot{X} = \epsilon A(\cos\underbrace{(\phi_1(t)+\phi_2(t))}_{\phi(t)}\Omega_x + \sin\underbrace{(\phi_1(t)+\phi_2(t))}_{\phi(t)}\Omega_y)X$$

and move into the frame,

$$Y = \exp(-\phi_2(t)\Omega_z)X, \tag{16}$$

$$\dot{Y} = [\epsilon A(t)(\cos\phi_1(t)\Omega_x + \sin\phi_1(t)\Omega_y) - \dot{\phi}_2(t)\Omega_z]Y, \tag{17}$$

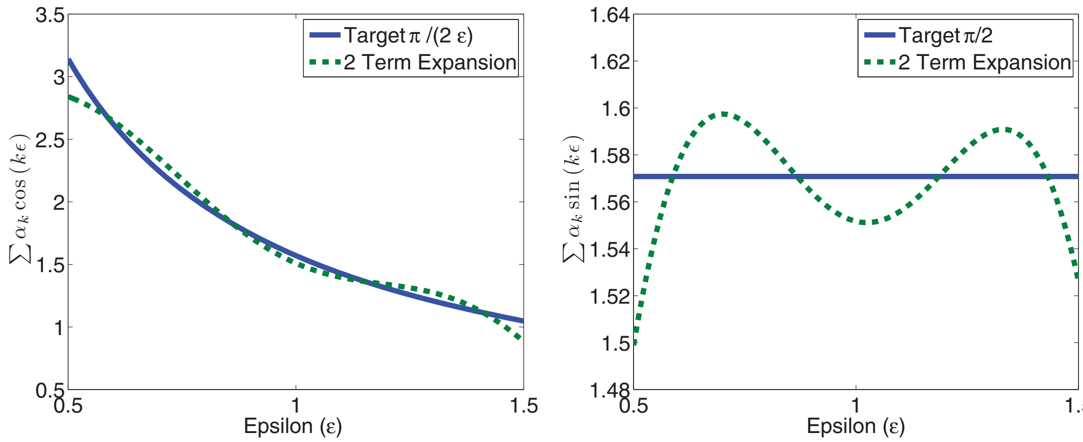

FIG. 1. (Color online) Left: two term Fourier synthesis method approximation for $\frac{\pi}{2\epsilon}$ using gradient descent for frequency selection. Right: two term δ modulation approximation for $\frac{\pi}{2}$ using gradient descent for frequency selection.

which corresponds to (10) once the appropriate identifications are made.

This means that $\tilde{V}_k$ can be directly produced by implementing $\phi_1(t)$ as 0, π, π, and 0 over Δt time intervals such that $A\Delta t = \gamma_k$, and with $-\dot{\phi}_2$ a delta pulse with area $\frac{\alpha_k}{2}$, and $-\frac{\alpha_k}{2}$ at time Δt and $3\Delta t$ respectively in (3). As $\phi_2(4\Delta t) = 0$, $X(4\Delta t) = Y(4\Delta t)$ and the laboratory frame and Y frame coincide after each pulse sequence, completing the δ modulated pulse design method.

C. Remarks

The previous sections presented a constructive means to produce the following rotations:

$$\Phi_1(\vec{\alpha},\vec{\gamma}) = \prod_k V(\beta_k,\gamma_k)^{n_k} = \exp\left(\overbrace{\sum_k \epsilon\alpha_k \cos(\epsilon\gamma_k)\,\Omega_y}^{H_1(\epsilon,\vec{\alpha},\vec{\gamma})}\right), \tag{18}$$

$$\Phi_2(\vec{\alpha},\vec{\gamma}) = \prod_k \tilde{V}(\beta_k,\gamma_k)^{n_k} = \exp\left(\underbrace{\sum_k \alpha_k \sin(\epsilon\gamma_k)\,\Omega_y}_{H_2(\epsilon,\vec{\alpha},\vec{\gamma})}\right), \tag{19}$$

$$\beta_k = \frac{\alpha_k}{n_k}, \tag{20}$$

using corresponding pulse elements consisting of directly accessible rotations:

$$V(\beta_k,\gamma_k) = \exp(\gamma_k\epsilon\Omega_x)\exp\left(\frac{\beta_k}{2}\epsilon\Omega_y\right)\exp(-2\gamma_k\epsilon\Omega_x)$$
$$\times \exp\left(\frac{\beta_k}{2}\epsilon\Omega_y\right)\exp(\gamma_k\epsilon\Omega_x), \tag{21}$$

$$\tilde{V}(\beta_k,\gamma_k) = \exp(\gamma_k\epsilon\Omega_x)\exp(-2\gamma_k\epsilon\Omega_\phi)\exp(\gamma_k\epsilon\Omega_x), \tag{22}$$

$$\Omega_\phi = \cos\left(\frac{\beta_k}{2}\right)\Omega_x - \sin\left(\frac{\beta_k}{2}\right)\Omega_y. \tag{23}$$

By completeness of a Fourier sine series, H_2 can be made to approximate any odd function with arbitrary accuracy. Similarly, H_1/ϵ can approximate any even function. As we are only interested in positive values of ϵ, H_1, and H_2 can be made to approximate any $f(\epsilon)\Omega_y$ rotation, where $f(\epsilon)$ is a continuous function of ϵ. We will show in the next section that keeping only the first few terms in the series is often sufficient in practice. Moreover, interchanging Ω_x and Ω_y will produce analogous rotations about the x axis so that any rotation,

$$\begin{aligned}\exp(\theta(\epsilon)(\cos\beta(\epsilon)\Omega_y &+ \sin\beta(\epsilon)\cos\phi(\epsilon)\Omega_z \\ &+ \sin\beta(\epsilon)\sin\phi(\epsilon)\Omega_x) \\ &= \exp(\phi(\epsilon)\Omega_y)\exp(\beta(\epsilon)\Omega_x)\exp(\theta(\epsilon)\Omega_y) \\ &\quad \exp(-\beta(\epsilon)\Omega_x)\exp(-\phi(\epsilon)\Omega_y),\end{aligned} \tag{24}$$

where θ, ϕ, and β are continuous functions of ϵ, can be produced.

This effectively reduces the problem of rf compensation to one of function fitting through selection of $\vec{\alpha}$ and $\vec{\gamma}$. Selecting $f(\epsilon) = \theta$, a constant, corresponds to robust rotations, which are the primary focus of this paper.

An important consideration in robust rf pulse design is the total required flip angle to achieve a level of compensation as this is a measure of the time required to implement a pulse. Long duration pulses are undesirable as relaxation effects can become non-negligible.

While we defer an in depth comparison of the methods to the following section, it is clear that Eq. (18) and Eq. (19) represent different possible bases for expansion. Consequently, series truncation is expected to result in differing levels of error for a given target function. In the case of a robust rotation, $f(\epsilon) = \theta$, the basis from δ modulation will be found to be preferable, requiring fewer terms in the expansion for a given level of error. As a result, δ modulated pulses will be shorter for a given level of robustness. Figures 1 and 2 show this graphically for the case of a two term expansion.

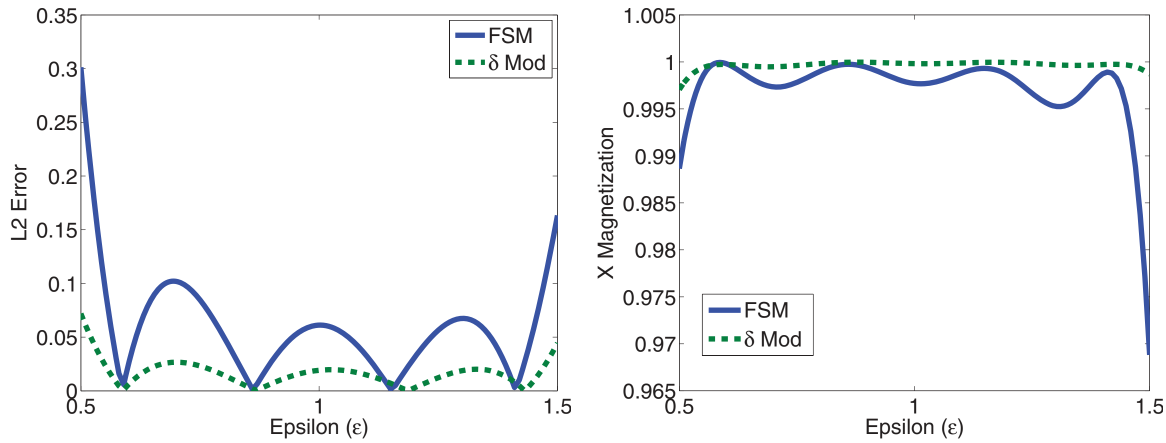

FIG. 2. (Color online) Left: L2 error with respect to the desired final magnetization $[1,0,0]'$ for two term expansions in Φ_1 and Φ_2 as a function of the dispersion parameter ϵ. Frequencies were selected using gradient descent. Right: corresponding final X magnetization for the two term sequences for both Fourier synthesis and δ modulation. δ modulation has a favorable magnetization profile while requiring a shorter pulse duration.

III. SIMULATIONS AND ERROR PERFORMANCE OF FOURIER SYNTHESIS AND δ MODULATION

The previous section reduced the problem of RF dispersion compensation to parameter selection, γ_k and α_k. Given the inhomogeneity parameter, $\epsilon \in [1-\delta_0, 1+\delta_0]$, we compute the error performance of synthesizing the effective rotations,

$$\Phi_1 = \exp\left(\epsilon \sum_k \alpha_k^1 \cos\left(\epsilon \gamma_k^1\right)\Omega_y\right), \tag{25}$$

$$\Phi_2 = \exp\left(\sum_k \alpha_k^2 \sin\left(\epsilon \gamma_k^2\right)\Omega_y\right), \tag{26}$$

to approximate

$$\Phi = \exp(\theta \Omega_y).$$

We note that the $\{\alpha_k^1\}$ can be directly calculated given the $\{\gamma_k^1\}$ as

$$\vec{\alpha}^1 = M^{-1}V, \quad \langle f,g\rangle = \int_{1-\delta}^{1+\delta} f(\epsilon)g(\epsilon)d\epsilon, \tag{27}$$

$$M_{ij} = \left\langle \cos\left(\gamma_i^1\epsilon\right), \cos\left(\gamma_j^1\epsilon\right)\right\rangle, \quad V_i = \left\langle \cos\left(\gamma_i^1\epsilon\right), \frac{\theta}{\epsilon}\right\rangle \tag{28}$$

and similarly the $\{\alpha_k^2\}$ can be calculated as

$$\vec{\alpha}^2 = M^{-1}V, \quad \langle f,g\rangle = \int_{1-\delta}^{1+\delta} f(\epsilon)g(\epsilon)d\epsilon, \tag{29}$$

$$M_{ij} = \left\langle \sin\left(\gamma_i^2\epsilon\right), \sin\left(\gamma_j^2\epsilon\right)\right\rangle, \quad V_i = \left\langle \sin\left(\gamma_i^2\epsilon\right), \theta\right\rangle, \tag{30}$$

so that the problem reduces to selecting the optimal frequencies.

We report performance for three frequency selection methods, heuristically, greedy selection and gradient descent. δ modulation outperforms Fourier synthesis methods for all

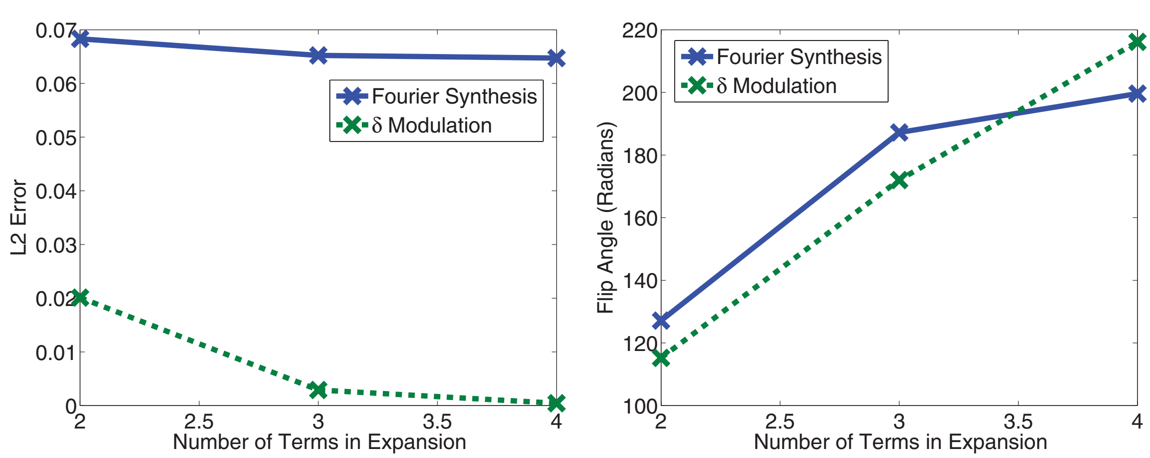

FIG. 3. (Color online) L2 error (left) and total flip angle (right) for heuristic frequency selection for Fourier synthesis and δ modulation. Error for a fixed pulse duration is smaller with δ modulation.

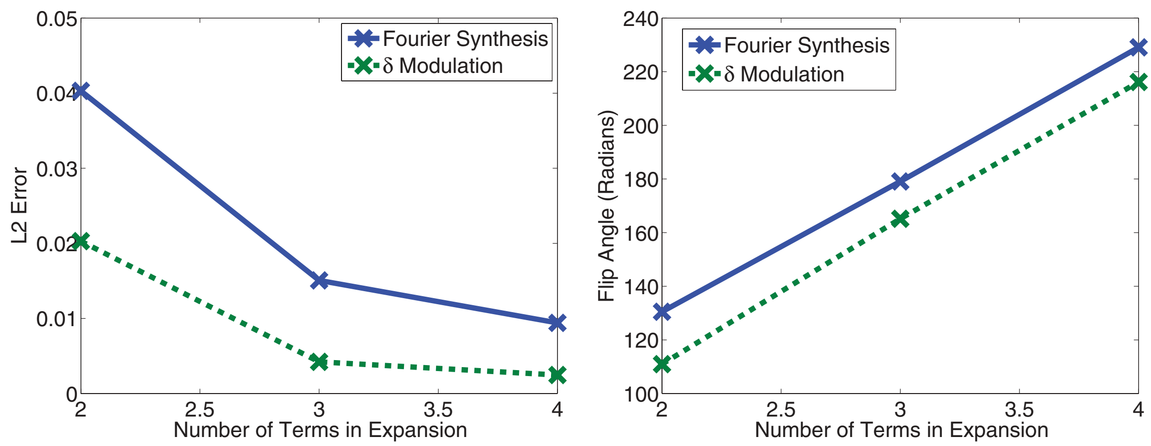

FIG. 4. (Color online) L2 error (left) and total flip angle (right) for greedy frequency selection for Fourier synthesis and δ modulation. Both duration and error are smaller for δ modulation.

frequency selection methods as shown in Figs. 3–5. Unless stated otherwise, the notion of optimal is with respect to L2 error for a given pulse duration. L2 error is calculated with respect to the desired final magnetization [1,0,0] and pulse duration is reported in total flip angle.

As a starting point, we consider the problem of selecting the optimal amplitudes given known frequencies which we will choose heuristically. As sine obtains its maximum at $\pi/2$ and is relatively horizontal about this point, a natural selection for the frequencies in (15) is $\gamma_k = \frac{(2k-1)\pi}{2}$. Similarly, selecting γ_k in (8) to maximize flatness about $\epsilon = 1$ corresponds to

$$\left[\frac{d}{d\epsilon}\epsilon\cos(\gamma_k\epsilon)\right]\Bigg|_{\epsilon=1} = \cos(\gamma_k) - \gamma_k\sin(\gamma_k) = 0. \quad (31)$$

Numerically solving gives the first several $\gamma_k = [0.860, 3.426, 6.437]$.

The amplitude coefficients $\vec{\alpha}$ were then calculated according to (27)–(30). The comparative performance of standard FSM, Φ_1, to δ modulation, Φ_2, is tabulated in Table I and a complete description of the pulses is given in the Appendix.

An alternative algorithm is to sequentially select the frequencies employing a greedy algorithm, in which already determined frequencies are held fixed, and only the newest frequency is optimized over. Explicitly, we sequentially minimize the cost functions with respect to $\gamma_k^{1/2}$,

$$F_1(\gamma_k^1,\ldots,\gamma_1^1) = \int_{1-\delta}^{1+\delta}\left\|\sum\alpha_k\cos(\epsilon\gamma_k^1) - \frac{\theta}{\epsilon}\right\| d\epsilon, \quad (32)$$

$$F_2(\gamma_k^2,\ldots,\gamma_1^2) = \int_{1-\delta}^{1+\delta}\left\|\sum\alpha_k\sin(\epsilon\gamma_k^2) - \theta\right\| d\epsilon, \quad (33)$$

again using (27)–(30) to calculate the $\vec{\alpha}$. This was done using gradient descent and numerically calculating the necessary derivatives. Table I shows that the δ modulation outperforms standard FSM.

The most general method we applied (and best performing) was simultaneously optimizing F_1 and F_2 with respect to all frequencies using gradient descent, where derivatives were again calculated numerically. As with all descent schemes, there is concern that one merely obtains a local minima. Moreover, the problem of unspecified frequencies is how to project onto an over-represented subspace which is known to have local minima. To combat such issues we chose the optimal result after numerous starting points and note that the performance exceeds the other methods and the results are tabulated in Table I.

As an example we consider the resulting parameters from optimizing a two term δ modulated pulse using gradient descent:

$$\gamma^2 = [88.6^\circ, 265.1^\circ], \quad \alpha^2 = [105.5^\circ, 16.6^\circ].$$

TABLE I. Performance of Fourier synthesis and δ modulation for heuristic, greedy, and gradient descent based frequency selection. In all cases, δ modulation outperforms the Fourier synthesis method in terms of L2 error for a given pulse duration, which is important for minimizing relaxation effects. L2 error is calculated with respect to the desired final magnetization [1,0,0]′.

Heuristic				**Greedy**				**Gradient**			
	$n=2$	$n=3$	$n=4$		$n=2$	$n=3$	$n=4$		$n=2$	$n=3$	$n=4$
Error FSM	0.06831	0.06523	0.06473	Error FSM	0.04031	0.01506	0.00941	Error FSM	0.07339	0.01874	0.00423
Error δ mod	0.02012	0.00290	0.00044	Error δ mod	0.02029	0.00422	0.00247	Error δ mod	0.01940	0.00280	0.00044
Flip $\angle$ FSM	127.120	187.200	199.600	Flip $\angle$ FSM	130.497	179.062	229.101	Flip $\angle$ FSM	120.519	225.780	347.413
Flip $\angle$ δ mod	115.230	172.001	216.138	Flip $\angle$ δ mod	110.952	165.178	216.177	Flip $\angle$ δ mod	113.341	170.134	216.137

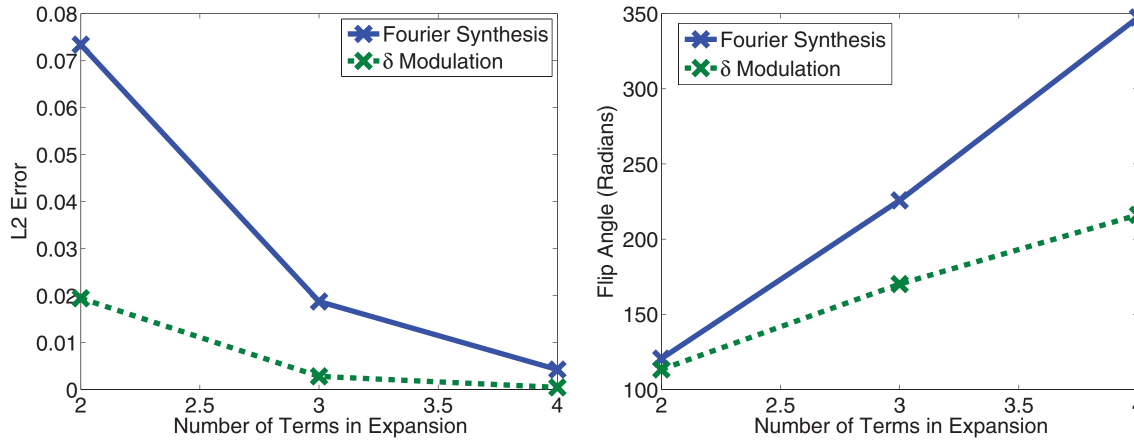

FIG. 5. (Color online) L2 error (left) and total flip angle (right) for gradient descent frequency selection for Fourier synthesis and δ modulation. Both duration and error are smaller for δ modulation.

These are converted into a pulse sequence by first dividing large amplitudes of α_k^2 into repeated sequences with smaller amplitudes according to (14) using a threshold value of 9°, which yields the modified parameters

$$\gamma^{2'} = [\underbrace{88.6^\circ,\ldots,88.6^\circ}_{12\text{ times}},\underbrace{265.1^\circ,\ldots,265.1^\circ}_{2\text{ times}}]$$
$$\beta^2 = [\underbrace{8.8^\circ,\ldots,8.8^\circ}_{12\text{ times}},\underbrace{8.4^\circ,\ldots,8.4^\circ}_{2\text{ times}}].$$

Pulses are calculated as described in Sec. II B where pulse elements are

$$[(\gamma_k)_0(2\gamma_k)_{180^\circ-\beta_k/2}(\gamma_k)_0]$$

with numbers inside the parentheses representing the flip angle and the subscripts, the phase. Applying to the parameters above yields the pulse sequence

$$[(88.6)_0(177.1)_{175.6}(88.6)_0]^{\times 12}$$
$$\times [(265.1)_0(530.1)_{175.9}(265.1)_0]^{\times 2}.$$

The performance is displayed in Fig. 2. The more terms kept in the series, the longer the sequence and overall pulse, but the more ϵ robust.

IV. GENERAL MODULATION SCHEMES

In many ways δ modulation is the most natural choice as it has a nice correspondence with existing FSMs. However, other modulation schemes are possible and their analysis is warranted for the sake of completeness or in the event that abrupt phase adjustments in the RF fields are not available. We begin by considering linear modulation.

A. Linear modulation

Returning to Eq. (10) with $\Delta t = \frac{\pi}{A}$ and

$$\phi_1 = \begin{cases} 0, & 0 < t \leqslant \Delta t, \\ \pi, & \Delta t < t \leqslant 3\Delta t, \\ 0, & 3\Delta t < t \leqslant 4\Delta t, \end{cases} \tag{34}$$

$$\dot{Y} = (A\epsilon \cos\phi_1(t)\Omega_x - \dot{\phi}_2\Omega_z)Y. \tag{35}$$

Moving into the interaction frame

$$Z \equiv \exp\left(-\int_0^t \epsilon A\cos\phi_1(\tau)d\tau\ \Omega_x\right)Y, \tag{36}$$

$$\dot{Z} = -\dot{\phi}_2\left(\cos\epsilon\int_0^t A\cos\phi_1(\tau)d\tau\ \Omega_z + \sin\epsilon\int_0^t A\cos\phi_1(\tau)d\tau\ \Omega_y\right)Z. \tag{37}$$

We note that under the assumptions

$$\int_0^T \cos\phi_1(t)dt = \int_0^T \dot{\phi}_2(t)dt = 0, \tag{38}$$

the Z frame will agree with the Y frame which will agree with the laboratory frame at time T, so that it is sufficient to analyze the system in the interaction frame. Letting $\phi_2(t)$ be a linear modulation of the form

$$\dot{\phi}_2 = \begin{cases} -B, & 0 \leqslant t < 2\Delta t, \\ B, & 2\Delta t \leqslant t \leqslant 4\Delta t, \end{cases} \tag{39}$$

we can analyze the resulting rotation with the Peano-Baker series,

$$\begin{aligned}\Phi &= I + \int_0^{\frac{4\pi}{A}} H(t)dt + \int_0^{\frac{4\pi}{A}} H(t)\int_0^t H(\sigma_1)d\sigma_1 dt + \cdots \\ &= I + \int_0^{\frac{4\pi}{A}} H(t)dt + O\left(\frac{B}{A}\right)^2 \\ &= I + 4B\int_0^{\frac{\pi}{A}} \sin(A\epsilon t)dt\Omega_y + O\left(\frac{B}{A}\right)^2 \\ &= I + \frac{4B}{A\epsilon}(1-\cos(\pi\epsilon)) + O\left(\frac{B}{A}\right)^2 \\ &= I + \frac{8B}{A}(1-\delta) + o(\delta)^2 + O\left(\frac{B}{A}\right)^2,\end{aligned} \tag{40}$$

which, to first order, has resulted in an evolution with the dispersion term reversed. Combining with a directly accessible

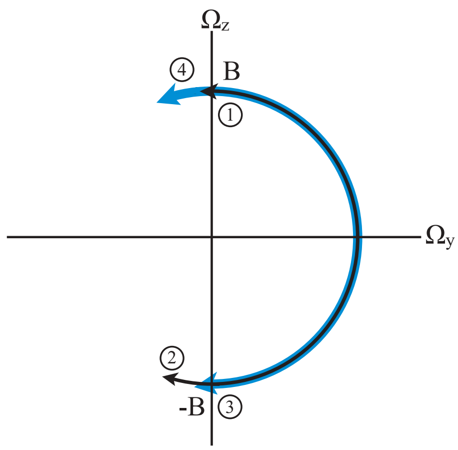

FIG. 6. (Color online) Trajectory in the interaction frame for linearly modulated controls. The trajectory is the black path from 1-2 and back to 1, followed by the blue path from 3-4 and back to 3.

evolution of $\frac{8B(1+\delta)}{A}\Omega_y$ will produce a pulse that is robust to first order in δ. Figure 6 displays the trajectory in the interaction frame for the linearly modulated pulse with $\epsilon > 1$ and provides the intuition for why the dispersion term is negated.

B. Arbitrary modulation schemes

Other modulation functions are also possible. Let $|f(t)| < B$ be such a candidate modulation, then choosing

$$\dot{\phi}_2 = \begin{cases} f(t), & 0 < t \leqslant \Delta t, \\ f(2\Delta t - t), & \Delta t < t \leqslant 2\Delta t, \\ -f(t - 2\Delta t), & 2\Delta t < t \leqslant 3\Delta t, \\ -f(4\Delta t - t), & 3\Delta t < t \leqslant 4\Delta t \end{cases} \tag{41}$$

will produce a rotation

$$I - 4\int_0^{\frac{\pi}{A}} f(t)\sin(A\epsilon t)dt\Omega_y + O\left(\frac{B}{A}\right)^2, \tag{42}$$

which can be used to produce new dispersion dependencies and thereby robust pulses as was done in the linear case.

V. CONCLUSION

We have presented a method for pulse design in the presence of RF inhomogeniety that extends existing Fourier synthesis methods. The method displays superior time-compensation properties to conventional Fourier synthesis methods. These methods are analytical and can be used to produce arbitrarily robust performance.

ACKNOWLEDGMENTS

The work was supported by ONR 38A-1077404, AFOSR FA9550-05-1-0443, and AFOSR FA9550-04-1-0427.

APPENDIX: PULSE PARAMETER

Heuristic FSM, $n = 2$:

$$\begin{aligned} \alpha &= [187.3, 33.8], \\ \gamma &= [49.3, 196.5], \\ Pulse &: [(49.3)_0(4.5)_{90}(98.5)_{180}(4.5)_{90}(49.3)_0]^{\times 21}[(196.5)_0(4.2)_{90}(393.0)_{180}(4.2)_{90}(196.5)_0]^{\times 4}. \end{aligned}$$

Heuristic FSM, $n = 3$:

$$\begin{aligned} \alpha &= [201.1, 49.2, 7.3], \\ \gamma &= [49.3, 196.5, 369.0], \\ Pulse &: [(49.3)_0(4.4)_{90}(98.5)_{180}(4.4)_{90}(49.3)_0]^{\times 23} \\ &\quad \times [(196.5)_0(4.1)_{90}(393.0)_{180}(4.1)_{90}(196.5)_0]^{\times 6}[(369.0)_0(3.6)_{90}(738.0)_{180}(3.6)_{90}(369.0)_0]^{\times 1}. \end{aligned}$$

Heuristic FSM, $n = 4$:

$$\begin{aligned} \alpha &= [175.2903, 18.3977, -10.8059, -5.67454], \\ \gamma &= [49.3, 196.5, 369.0, 546.0], \\ Pulse &: [(49.3)_0(4.4)_{90}(98.5)_{180}(4.4)_{90}(49.3)_0]^{\times 20}[(196.5)_0(3.1)_{90}(393.0)_{180}(3.1)_{90}(196.5)_0]^{\times 3} \\ &\quad \times [(369.0)_0(-2.7)_{90}(738.0)_{180}(-2.7)_{90}(369.0)_0]^{\times 2}[(546.0)_0(-2.8)_{90}(1092.1)_{180}(-2.8)_{90}(546.0)_0]^{\times 1}. \end{aligned}$$

Heuristic δ mod, $n = 2$:

$$\begin{aligned} \alpha &= [105.5, 16.7], \\ \gamma &= [90, 270], \\ Pulse &: [(90.0)_0(180.0)_{175.6}(90.0)_0]^{\times 12}[(270.0)_0(540.0)_{175.8}(270.0)_0]^{\times 2}. \end{aligned}$$

Heuristic δ mod, $n = 3$:

$$\begin{aligned}\alpha &= [108.3, 22.4, 4.3],\\ \gamma &= [90, 270, 450],\\ Pulse &: [(90.0)_0(180.0)_{175.8}(90.0)_0]^{\times 13}[(270.0)_0(540.0)_{176.3}(270.0)_0]^{\times 3}[(450.0)_0(900.0)_{177.9}(450.0)_0]^{\times 1}.\end{aligned}$$

Heuristic δ mod, $n = 4$:

$$\begin{aligned}\alpha &= [109.8, 25.7, 7.1, 1.2],\\ \gamma &= [90, 270, 450, 630],\\ Pulse &: [(90.0)_0(180.0)_{175.8}(90.0)_0]^{\times 13}[(270.0)_0(540.0)_{175.7}(270.0)_0]^{\times 3}\\ &\quad \times [(450.0)_0(900.0)_{176.4}(450.0)_0]^{\times 1}[(630.0)_0(1260.0)_{179.4}(630.0)_0]^{\times 1}.\end{aligned}$$

Greedy FSM, $n = 2$:

$$\begin{aligned}\alpha &= [191.9, 35.9],\\ \gamma &= [49.9, 192.7],\\ Pulse &: [(49.9)_0(4.4)_{90}(99.9)_{180}(4.4)_{90}(49.9)_0]^{\times 22}[(192.7)_0(4.5)_{90}(385.4)_{180}(4.5)_{90}(192.7)_0]^{\times 4}.\end{aligned}$$

Greedy FSM, $n = 3$:

$$\begin{aligned}\alpha &= [197.4, 40.9, -3.8],\\ \gamma &= [49.9, 192.7, 502.9],\\ Pulse &: [(49.9)_0(4.5)_{90}(99.9)_{180}(4.5)_{90}(49.9)_0]^{\times 22}[(192.7)_0(4.1)_{90}(385.4)_{180}(4.1)_{90}(192.7)_0]^{\times 5}\\ &\quad \times [(502.9)_0(-1.9)_{90}(1005.8)_{180}(-1.9)_{90}(502.9)_0]^{\times 1}.\end{aligned}$$

Greedy FSM, $n = 4$:

$$\begin{aligned}\alpha &= [200.7, 43.7, -5.9, -1.9],\\ \gamma &= [49.9, 192.7, 502.9, 666.8],\\ Pulse &: [(49.9)_0(4.4)_{90}(99.9)_{180}(4.4)_{90}(49.9)_0]^{\times 23}[(192.7)_0(4.4)_{90}(385.4)_{180}(4.4)_{90}(192.7)_0]^{\times 5}\\ &\quad \times [(502.9)_0(-3.0)_{90}(1005.8)_{180}(-3.0)_{90}(502.9)_0]^{\times 1}[(666.8)_0(-0.9)_{90}(1333.7)_{180}(-0.9)_{90}(666.8)_0]^{\times 1}.\end{aligned}$$

Greedy δ mod, $n = 2$:

$$\begin{aligned}\alpha &= [105.5, 16.6],\\ \gamma &= [86.7, 259.1],\\ Pulse &: [(86.7)_0(173.4)_{175.6}(86.7)_0]^{\times 12}[(259.1)_0(518.1)_{175.8}(259.1)_0]^{\times 2}.\end{aligned}$$

Greedy δ mod, $n = 3$:

$$\begin{aligned}\alpha &= [108.2, 22.2, 4.1],\\ \gamma &= [86.7, 259.1, 427.8],\\ Pulse &: [(86.7)_0(173.4)_{175.8}(86.7)_0]^{\times 13}[(259.1)_0(518.1)_{176.3}(259.1)_0]^{\times 3}[(427.8)_0(855.7)_{177.9}(427.8)_0]^{\times 1}.\end{aligned}$$

Greedy δ mod, $n = 4$:

$$\begin{aligned}\alpha &= [108.5, 22.9, 4.6, -0.3],\\ \gamma &= [86.7, 259.1, 427.8, 730.2],\\ Pulse &: [(86.7)_0(173.4)_{175.8}(86.7)_0]^{\times 13}[(259.1)_0(518.1)_{176.2}(259.1)_0]^{\times 3}\\ &\quad \times [(427.8)_0(855.7)_{177.7}(427.8)_0]^{\times 1}[(730.2)_0(1460.5)_{180.2}(730.2)_0]^{\times 1}.\end{aligned}$$

Gradient descent FSM, $n = 2$:

$$\begin{aligned}
\alpha &= [163.4, -15.7],\\
\gamma &= [51.5, 373.7],\\
Pulse &: [(51.5)_0(4.3)_{90}(103.0)_{180}(4.3)_{90}(51.5)_0]^{\times 19}[(373.7)_0(-3.9)_{90}(747.4)_{180}(-3.9)_{90}(373.7)_0]^{\times 2}.
\end{aligned}$$

Gradient descent FSM, $n = 3$:

$$\begin{aligned}
\alpha &= [169.6, -23.9, -10.3],\\
\gamma &= [52.4, 379.1, 550.3],\\
Pulse &: [(52.4)_0(4.5)_{90}(104.9)_{180}(4.5)_{90}(52.4)_0]^{\times 19}[(379.1)_0(-4.0)_{90}(758.2)_{180}(-4.0)_{90}(379.1)_0]^{\times 3}\\
&\quad \times [(550.3)_0(-2.6)_{90}(1100.6)_{180}(-2.6)_{90}(550.3)_0]^{\times 2}.
\end{aligned}$$

Gradient descent FSM, $n = 4$:

$$\begin{aligned}
\alpha &= [174.4, -30.6, -19.0, -5.1],\\
\gamma &= [53.1, 381.4, 554.0, 727.9],\\
Pulse &: [(53.1)_0(4.4)_{90}(106.1)_{180}(4.4)_{90}(53.1)_0]^{\times 20}[(381.4)_0(-3.8)_{90}(762.7)_{180}(-3.8)_{90}(381.4)_0]^{\times 4}\\
&\quad \times [(554.0)_0(-3.2)_{90}(1108.0)_{180}(-3.2)_{90}(554.0)_0]^{\times 3}[(727.9)_0(-2.6)_{90}(1455.8)_{180}(-2.6)_{90}(727.9)_0]^{\times 1}.
\end{aligned}$$

Gradient descent δ mod, $n = 2$:

$$\begin{aligned}
\alpha &= [105.5, 16.6],\\
\gamma &= [88.6, 265.1],\\
Pulse &: [(88.6)_0(177.1)_{175.6}(88.6)_0]^{\times 12}[(265.1)_0(530.1)_{175.9}(265.1)_0]^{\times 2}.
\end{aligned}$$

Gradient descent δ mod, $n = 3$:

$$\begin{aligned}
\alpha &= [108.3, 22.4, 4.3],\\
\gamma &= [89.1, 267.0, 444.5],\\
Pulse &: [(89.1)_0(178.1)_{175.8}(89.1)_0]^{\times 13}[(267.0)_0(534.1)_{176.3}(267.0)_0]^{\times 3}[(444.5)_0(889.0)_{177.9}(444.5)_0]^{\times 1}.
\end{aligned}$$

Gradient descent δ mod, $n = 4$:

$$\begin{aligned}
\alpha &= [109.8, 25.7, 7.1, 1.2],\\
\gamma &= [90.0, 270.0, 450.0, 630],\\
Pulse &: [(90.0)_0(180.0)_{175.8}(90.0)_0]^{\times 13}[(270.0)_0(540.0)_{175.7}(270.0)_0]^{\times 3}\\
&\quad \times [(450.0)_0(900.0)_{176.4}(450.0)_0]^{\times 1}[(630.0)_0(1260.0)_{179.4}(630.0)_0]^{\times 1}.
\end{aligned}$$

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