

Cyclicity of interaction-frame transformations

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We study the structure of rotation sequences composed of piecewise displacements by an angle β about arbitrary axes in three dimensions. We find that when $\beta = 2\pi/m$, repeated toggling-frame transformations return the sequence to its original form after m applications. We develop this cyclicity property to unify several families of error-tuned control sequences used across quantum technologies, including NMR, MRI, atomic clocks, and atom-scale computing. For the $m = 2$ case, we show a duality between narrowband and broadband π -element sequences. For $m > 2$, we relate the cyclic structure to polyhedral symmetry, leading to original sequence designs based on m -fold rotational axes. The result may guide the construction of robust control protocols grounded in geometric and group-theoretic principles.

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I. INTRODUCTION

Rotational periodicity arises in a wide variety of systems whose configuration spaces are discrete rather than continuous. In one dimension, examples include gear trains, Möbius strips, and spinor behavior such as the double cover of $SO(3)$ by $SU(2)$. In higher dimensions, notable cases include twisty puzzles like the Rubik's cube [1] and quantum systems exhibiting geometric phase [2], where repeated sequences of noncommuting operations restore the system to its initial state. Recognizing the cyclic structure of these systems provides a perspective on their underlying symmetries and has implications for algorithm design, especially in quantum control [3].

In this article, we introduce and examine a rotational periodicity property that is intrinsic to the so-called *toggling frame*—the interaction frame defined as corotating with a piecewise-time-dependent torque or Hamiltonian applied to a system—that is bread and butter to dynamics analysis, qubit dynamical decoupling [4–6], noise filtering [7,8], and optimal control [9]. A context where this periodicity can be applied is *composite* rotation sequences with tuned resilience to control field imperfections and inhomogeneities [10–14]. Composite rotations are used widely for high-fidelity manipulation of spin qubits, for instance, in nuclear magnetic resonance (NMR) [14–16], where this field began, in molecular spectroscopy [17,18], and more recently in photonic quantum

memories [19], quantum computing [13,20] in excited nuclear states [21], trapped ions [7,22–24], trapped neutral atoms [25,26], color centers [27], and superconducting transmons [28–31]. Analogous sequences may apply to classical systems with limited rotational-control degrees of freedom, such as spherical wheels and rolling robots [32,33].

II. THEORY: CYCLIC TOGGLING FRAMES

The main result is as follows: for a sequence of n rotations through uniform angle steps, $\beta \bmod 2\pi = 2\pi/m$, about arbitrary axes in three dimensions, denoted $\mathbf{e}_i^{(0)}$ ($i \in \{0 : n-1\}$), it turns out that transformation to the toggling frame [$\hat{M}$, defined by Eq. (1)] is cyclic; when m is an integer, m successive axis transformations returns the original axis set. This is illustrated graphically in Fig. 1 for the arbitrary set $(\mathbf{e}_0^{(0)}, \mathbf{e}_1^{(0)}, \mathbf{e}_2^{(0)}, \mathbf{e}_3^{(0)}) = (\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_x, \mathbf{e}_y)$ [with $\mathbf{e}_x = (1, 0, 0)$ and $\mathbf{e}_y = (0, 1, 0)$ in Cartesian coordinates], where we observe $\mathbf{e}_i^{(0)} \equiv \mathbf{e}_i^{(m)}$ upon applying $\hat{M}$ to $\{\mathbf{e}_i^{(0)}\}$ m times.

A proof of the result for arbitrary 3D rotations is given as follows. Let the rotation sequence be $\mathcal{S}^{(0)} \equiv (\beta)_{\mathbf{e}_0^{(0)}}(\beta)_{\mathbf{e}_1^{(0)}} \dots (\beta)_{\mathbf{e}_{n-1}^{(0)}}$, using left-to-right chronology and (angle)_{axis} notation. If a single rotation is also denoted by the matrix $R(\beta, \mathbf{e}_i^{(0)})$, the overall sequence is described by the propagator

$$U_n(\mathcal{S}^{(0)}) = \prod_{j=0}^{n-1} R(\beta, \mathbf{e}_j^{(0)}) \equiv R(\beta, \mathbf{e}_{(n-1)}^{(0)}) \dots R(\beta, \mathbf{e}_1^{(0)}) R(\beta, \mathbf{e}_0^{(0)}), \quad (1)$$

which by closure of the rotation group is also a rotation. $\hat{M}$ is then defined as a map on $\{\mathbf{e}_i^{(0)}\}$ using the shorthand

$$\{\mathbf{e}_i^{(1)}\} = \hat{M}\{\mathbf{e}_i^{(0)}\} \quad (2)$$

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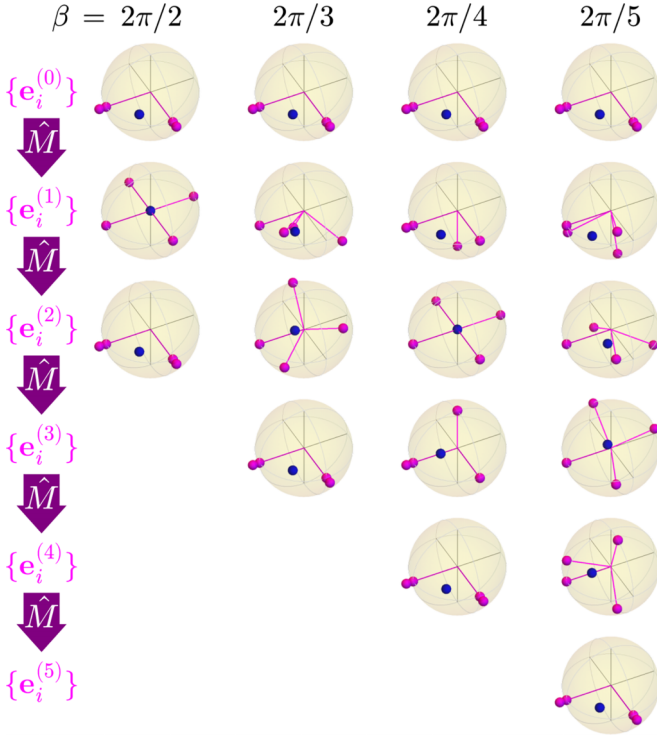


FIG. 1. Cyclicity of the transformation $\hat{M}$ defined by Eqs. (2) and (3) for a sequence of rotations through angle $\beta = 2\pi/m$, for integer m , about unit-vector axes (in magenta) given by $\{\mathbf{e}_i^{(0)}\} = (\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_x, \mathbf{e}_y)$; $\mathbf{e}_x = (1, 0, 0)$ and $\mathbf{e}_y = (0, 1, 0)$. The cycle property is $\mathbf{e}_i^{(m)} = \hat{M}^m \mathbf{e}_i^{(0)} \equiv \mathbf{e}_i^{(0)}$. Blue spheres show the centroid of the axes sets, defined by Eq. (4).

to describe the accumulative rotation:

$$\begin{aligned} \mathbf{e}_i^{(1)} &= U_i^{-1}(\mathcal{S}^{(0)})\mathbf{e}_i^{(0)}; \quad i \geq 1, \\ \mathbf{e}_0^{(1)} &= \mathbf{e}_0^{(0)}. \end{aligned} \quad (3)$$

A sequence of rotations about the axis set $\{\mathbf{e}_i^{(1)}\}$ then defines $\mathcal{S}^{(0)}$ in the toggling frame.

As shown in Appendix A, $\hat{M}$ provides a transformation into the toggling frame for rotations with a systematic angle error $\beta \rightarrow \beta' \equiv \beta + \epsilon$, and the effective propagator in this frame is a sequence of rotations about axes $\mathbf{e}_i^{(1)}$ ($i \in \{0 : n-1\}$), not through angles β but ϵ : $(\epsilon)_{\mathbf{e}_0^{(1)}}(\epsilon)_{\mathbf{e}_1^{(1)}} \dots (\epsilon)_{\mathbf{e}_{n-1}^{(1)}}$. Coherent averaging theory states that the first-order effect of ϵ is zero if the centroid

$$\mathbf{C}^{(1)} = (1/n) \sum_{i=0}^{n-1} \mathbf{e}_i^{(1)} \quad (4)$$

or arithmetic mean of $\{\mathbf{e}_i^{(1)}\}$ is zero; this is a target for broadband pulse sequence design, which we review briefly in Appendix B. In Fig. 1 we see $\mathbf{C}^{(1)} = \mathbf{0}$ for $\{\mathbf{e}_i^{(0)}\} = (\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_x, \mathbf{e}_y)$, a key result for dynamical decoupling.

For present purposes, however, the error angle is unimportant, because we focus on the abstract scenario where $\hat{M}$ is applied iteratively. For instance, applying $\hat{M}$ to the sequence $\mathcal{S}^{(1)} \equiv (\beta)_{\mathbf{e}_0^{(1)}}(\beta)_{\mathbf{e}_1^{(1)}} \dots (\beta)_{\mathbf{e}_{n-1}^{(1)}}$ with angles β , not ϵ , to generate a next set of axes $\{\mathbf{e}_i^{(2)}\}$, and so on. Subsequent sets of vectors $\{\mathbf{e}_i^{(m)}\}$ are generated by concatenating rota-

tions by β around the axes from the previous transformations $\{\mathbf{e}_j^{(m-1)}\}$, ($j \leq i$):

$$(\{\mathbf{e}_0^{(m)}, \mathbf{e}_1^{(m)}, \dots, \mathbf{e}_{(n-1)}^{(m)}\}) = \hat{M}^m(\{\mathbf{e}_0^{(0)}, \mathbf{e}_1^{(0)}, \dots, \mathbf{e}_{(n-1)}^{(0)}\}), \quad (5)$$

where we stress it is β , not ϵ , used each time for the rotation angle.

Remarkably, despite the folded nature of $\hat{M}$, each transformed vector $\mathbf{e}_i^{(m)}$ can also be expressed simply in terms of the initial set $\{\mathbf{e}_i^{(0)}\}$. The formula

$$\mathbf{e}_i^{(m)} = \left(\prod_{j=0}^{i-1} R(m\beta, \mathbf{e}_j^{(0)}) \right)^{-1} \mathbf{e}_i^{(0)} \quad (6)$$

follows naturally from the cumulative definition of $\hat{M}$. For example, $\mathbf{e}_3^{(m)} = U_3^{-1}(\mathcal{S}^{(m-1)}) \dots U_3^{-1}(\mathcal{S}^{(0)})\mathbf{e}_3^{(0)} = R(-m\beta, \mathbf{e}_0^{(0)})R(-m\beta, \mathbf{e}_1^{(0)})R(-m\beta, \mathbf{e}_2^{(0)})\mathbf{e}_3^{(0)}$. The result appears upon expansion of the rotation products from Eq. (1), with later U transformations “unwinding” the effects of the previous ones; see Appendix C for an explicit evaluation. From a physical perspective, however, such a simple equation is unexpected and nonintuitive due to the inherent noncommutativity of 3D rotations.

Equation (6) now reveals the periodic property. For rotation angles that are unit fractions of 2π , ($\beta = 2\pi/m$), the net effect of $\hat{M}^m$ is a unity operation (or a unity super-superoperator), returning the original vectors regardless of orientation: $\{\mathbf{e}_i^{(m)}\} \equiv \hat{M}^m\{\mathbf{e}_i^{(0)}\} = \{\mathbf{e}_i^{(0)}\}$, as shown in Fig. 1. In these cases we say $\hat{M}$ has a cycle of length m . The inverse transformation is simply $\hat{M}^{-1} \equiv \hat{M}^{m-1}$, because $\hat{M}^{m-1}\hat{M} = 1$.

In terms of the relation to spin dynamics, Eq. (6) implies that for $\beta = 2\pi/m$ there are families of m iterated rotation sequences $\mathcal{S}^{(k)} = (\beta)_{\mathbf{e}_0^{(k)}}(\beta)_{\mathbf{e}_1^{(k)}} \dots (\beta)_{\mathbf{e}_{n-1}^{(k)}}$ ($0 \leq k < m$) with respective toggling-frame propagators determined by $\{\mathbf{e}_0^{(k+1) \bmod m}\}$. In the next sections we apply this concept to (1) reinterpret known composite pulses and (2) analyze dynamical decoupling sequences.

III. RESULTS: APPLICATION TO COMPOSITE PULSES

Despite wide application of the toggling frame to dynamical systems, Eq. (6) as a general property appears unrecognized. A very limited use of the formula for $m = 2$ (an involution) and equatorial-plane vectors $\{\mathbf{e}_i^{(0)}\} = \{(\cos \phi_i^{(0)}, \sin \phi_i^{(0)}, 0)\}$ appears in the NMR literature [34–36], in relation to the rf and toggling-frame phases of $\beta = \pi$ -composite pulses. Phases of the rf pulses are related by the formulas

$$\phi_i^{(1)} = \phi_0^{(0)} + \sum_{j=1}^i (-1)^j (\phi_j^{(0)} - \phi_{j-1}^{(0)}), \quad (7)$$

$$\phi_i^{(0)} = \phi_0^{(1)} + \sum_{j=1}^i (-1)^j (\phi_j^{(1)} - \phi_{j-1}^{(1)}), \quad (8)$$

where vectors $\{\mathbf{e}_i^{(1)}\} = \{(\cos \phi_i^{(1)}, \sin \phi_i^{(1)}, 0)\}$ result directly from Eq. (6) because any exact π rotation involving two rf-frame axes, $R(\pi, \mathbf{e}_i^{(0)})\mathbf{e}_k^{(0)}$, keeps the vector $\mathbf{e}_i^{(1)}$ confined to

the xy plane. The phase relation for $m = 2$ was also mentioned by Pines *et al.* in relation to the fixed-point theory of iterative dynamical decoupling [37–39].

Aside from these works, Eq. (6) is unexplored, especially for values $m > 2$ and axes outside the equatorial plane [40]. An immediate conclusion is that there are m sequences of rotations by $2\pi/m$, $\mathcal{S}^{(k)}$; $0 \leq k < m$, that cyclically interconvert with one another under $\hat{M}$.

For $m = 2$, this means a “duality” property of π rotation sequences, where the toggling-frame vectors of one sequence equal the nontoggling (e.g., rf-frame) vectors of the dual sequence [41]. As a general rule, a broadband compensated sequence $\mathcal{S}^{(0)}$ involving vectors $\{\mathbf{e}_i^{(0)}\}$ maps to a narrowband sequence $\mathcal{S}^{(1)}$, involving rotations about vectors $\{\mathbf{e}_i^{(1)}\}$, and vice versa. One interpretation is through the centroid of the vector sets. Sequences for broadband angle-error compensation have balanced vectors $\mathbf{C}^{(1)} = \mathbf{0}$ and a far-from-zero $\mathbf{C}^{(0)}$, while for narrowband sequences it is the opposite: $\mathbf{C}^{(0)} = \mathbf{0}$ and a far-from-zero $\mathbf{C}^{(1)}$.

The interchange of $\mathbf{e}_i^{(0)}$ and $\mathbf{e}_i^{(1)}$ between dual sequences also equates to a glide-reflection relationship in the profile of vector inversion vs rotation angle. If $q_{\mathcal{S}^{(0)}}(\mathbf{e}_\xi, \beta')$ is a function describing the transformation amplitude of a unit vector $\mathbf{e}_\xi$ (such as spin orientation) under $\mathcal{S}^{(0)}$ with $\beta = \beta'$, where all rotation axes lie in the xy plane,

$$q_{\mathcal{S}^{(0)}}(\mathbf{e}_\xi, \beta') = \text{Tr}\{(\mathbf{e}_\xi U_n(\mathcal{S}^{(0)})^\dagger U_n(\mathcal{S}^{(0)}) \mathbf{e}_\xi\}, \quad (9)$$

then provided that constraints $q_{\mathcal{S}^{(0)}}(\mathbf{e}_z, 0) = 1$ and $q_{\mathcal{S}^{(0)}}(\mathbf{e}_z, \pi) = -1$ are satisfied (which describe nominal inversion), we find

$$q_{\mathcal{S}^{(1)}}(\mathbf{e}_\xi, \beta') = -q_{\mathcal{S}^{(0)}}(\mathbf{e}_\xi, \pi \pm \beta'). \quad (10)$$

For $\mathbf{e}_\xi = \mathbf{e}_z \equiv (0, 0, 1)$, Eq. (10) means the profiles of vector inversion vs β' for two dual sequences are related by a vertical flip and a translation by π along the β' axis, modulo 2π . The horizontal shift of π is the period of β for $m = 2$, and the vertical flip, for one half of the dual, is supplied by the minus sign in $U_n(\mathcal{S}^{(k)})\mathbf{e}_z = -\mathbf{e}_z$ on the right-hand side of Eq. (3).

Another general property of duals is parity reversal, where if the phase list $\{\phi_i^{(0)}\}$ is order-reversal antisymmetric then the list $\{\phi_i^{(1)}\}$ is order-reversal symmetric, and vice versa. The property arises from the alternating sign structure of Eq. (7), which causes phase differences in the second half of the list to be the opposite of those in the first half. This may be seen more clearly by stating Eq. (7) in a recursive form

$$\phi_i^{(1)} + (-1)^{i-1} \phi_i^{(0)} = \phi_{i-1}^{(1)} + (-1)^{i-1} \phi_{i-1}^{(0)} \quad (11)$$

or

$$\Delta_{-1} \phi_i^{(1)} = (-1)^i \Delta_{-1} \phi_i^{(0)} \quad (12)$$

by introducing the finite difference operator $\Delta_a f(x) = f(x+a) - f(x)$.

An archetype of toggling-frame duality is the pair of composite pulse sequences F1 and NB1_{TPG}. F1 [35,42] is a cyclically permuted version of Wimperis’s broadband population-inversion pulse BB1 [43], while NB1_{TPG} is a reversal-symmetric narrowband inversion pulse devised by Tycko, Pines, and Guckenheimer in the 1980s [39]. The

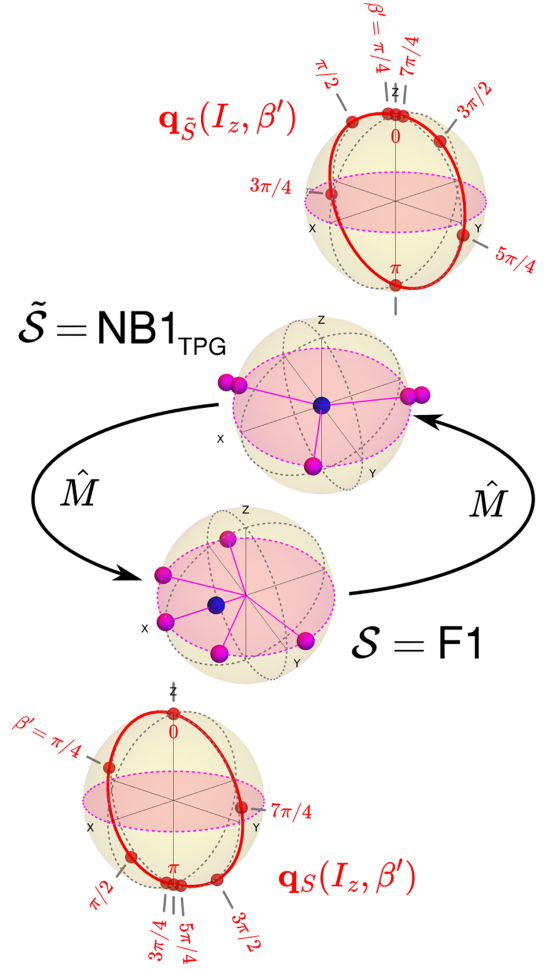


FIG. 2. A representation of the universal duality ($m = 2$ cycle) between sequences $\mathcal{S}^{(0)} = \text{F1}$ [Eq. (13)] and $\mathcal{S}^{(1)} \equiv \text{NB1}_{\text{TPG}}$ [Eq. (14)]. Magenta lines and spheres show the rotation axes of the piecewise elements of the sequence, while the blue spheres represent the centroids. At the top and bottom of the figure, the z -inversion performance is represented by continuous paths showing the vectors $\mathcal{S}^{(0)} \cdot (0, 0, 1)$ and $\mathcal{S}^{(1)} \cdot (0, 0, 1)$ as a function of $0 \leq \beta' < 2\pi$. The dual paths are related by a π rotation about the $x = (1, 0, 0)$ axis.

sequences are given in the form $\mathcal{S}^{(k)} = (\beta)_{\phi_0^{(k)}} (\beta)_{\phi_1^{(k)}} \dots (\beta)_{\phi_{n-1}^{(k)}}$ by

$$\text{F1} = (\pi)_{-3\varphi} (\pi)_{-\varphi} \pi_0 (\pi)_{\varphi} (\pi)_{3\varphi}, \quad (13)$$

$$\text{NB1}_{\text{TPG}} = (\pi)_{\varphi} (\pi)_{-\varphi} (\pi)_0 (\pi)_{-\varphi} (\pi)_{\varphi}, \quad (14)$$

with $\varphi = \arccos(-1/4)$, and the common values of φ and n already hint at some level of connection between the two. Applying Eqs. (7) and (8), we observe

$$\phi_i^{(1)}[\text{NB1}_{\text{TPG}}] = \phi_i^{(0)}[\text{F1}] + 4\varphi, \quad (15)$$

$$\phi_i^{(1)}[\text{F1}] = \phi_i^{(0)}[\text{NB1}_{\text{TPG}}] - 4\varphi. \quad (16)$$

The phase shift of 4φ arises because in the original definition of NB1_{TPG}, the net rotation is about the 4φ axis, i.e., $(\cos \varphi, \sin \varphi, 0)$, rather than the x axis. The dual properties including the glide-reflection relation of Eq. (10) are illustrated in Fig. 2 and the upper panel of Fig. 3.

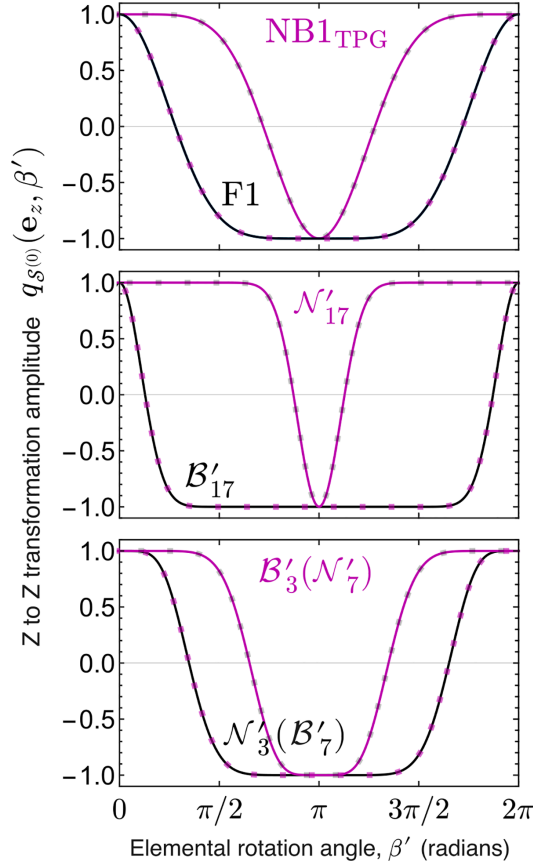


FIG. 3. Vector transformation profiles for pairs of selected $\beta = \pi$ rotation sequences related by duality, where each dual offers narrowband or broadband tolerance to amplitude variation in β' about $\beta' = \pi$. Solid lines represent curves $q_{S(0)}(\mathbf{e}_z, \beta')$ defined by Eq. (9), while sparse-dotted lines of the same color represent the same profile after applying a horizontal translation by π (modulo 2π) and then multiplication by -1 ; exact overlap of these with the counterpart solid curve illustrates the glide-reflection rule of Eq. (10).

We now identify several other duals, including between (1) antisymmetric narrow-band [44] and (2) symmetric broadband [45–47] π -rotation sequences both involving $\beta = \pi$ and phase digitizations $2\pi/n$, introduced by Vitanov and colleagues. We use modified versions where a global phase shift is added to ensure the phase list is order-reversal antisymmetric. These modified sequences are denoted with a prime ($\mathcal{N}'_n$ narrowband, $\mathcal{B}'_n$ broadband),

$$\phi_i[\mathcal{N}'_n] = \frac{2\pi}{n} \left[(-1)^{i+1} \left\lfloor \frac{i+1}{2} \right\rfloor + (-1)^{(n-1)/2} \left\lfloor \frac{n+1}{4} \right\rfloor \right], \quad (17)$$

$$\begin{aligned} &= -\phi_{n-1-i}[\mathcal{N}'_n], \\ \phi_i[\mathcal{B}'_n] &= -\frac{2\pi}{n} \times \frac{2i(i+1) - n^2 + 1}{4}, \\ &= +\phi_{n-1-i}[\mathcal{B}'_n], \end{aligned} \quad (18)$$

and produce nominal π rotations about the x axis. Proof that $\mathcal{N}'_n$ and $\mathcal{B}'_n$ are dual is via Eq. (12), to show that the differences between adjacent phases $\phi_i[\mathcal{N}'_n]$ and $\phi_{i-1}[\mathcal{N}'_n]$ in the toggling

frame equate to those for $\mathcal{B}'_n$. The glide-reflection property is illustrated for $n = 17$ in the center panel of Fig. 3.

Other implementations of Vitanov's sequences include scaling the phases by an integer k , such that $\phi_i[\mathcal{N}'_{n,k}] = k \times \phi_i[\mathcal{N}'_n]$ and $\phi_i[\mathcal{B}'_{n,k}] = k \times \phi_i[\mathcal{B}'_n]$. As long as k is not a divisor of n , these sequences perform identically to $\mathcal{N}'_{n,1}$ and $\mathcal{B}'_{n,1}$, respectively, when considering errors $|\epsilon| > 0$, but differently under detuning errors [12,48]. Generally, $\mathcal{B}'_{n,k}$ and $\mathcal{N}'_{n,k}$ are also duals. Additionally, Kyoseva and Vitanov [49] constructed a recipe for generating band-pass inversion sequences via a nested composition [50] of broadband and narrowband sequences; these are called $\mathcal{B}_v(\mathcal{N}_n)$ and $\mathcal{N}_v(\mathcal{B}_n)$ and are defined through

$$\begin{aligned} \phi[\mathcal{N}_v(\mathcal{B}_n)] &= \{\phi_0[\mathcal{N}_v] + \phi[\mathcal{B}_n], \\ &\quad \phi_1[\mathcal{N}_v] + \phi[\widetilde{\mathcal{B}}_n], \\ &\quad \phi_2[\mathcal{N}_v] + \phi[\mathcal{B}_n], \\ &\quad \dots, \\ &\quad \phi_{v-1}[\mathcal{N}_v] + \phi[\mathcal{B}_n]\}, \end{aligned} \quad (19)$$

$$\begin{aligned} \phi[\mathcal{B}_v(\mathcal{N}_n)] &= \{\phi_0[\mathcal{B}_v] + \phi[\mathcal{N}_n], \\ &\quad \phi_1[\mathcal{B}_v] + \phi[\widetilde{\mathcal{N}}_n], \\ &\quad \phi_2[\mathcal{B}_v] + \phi[\mathcal{N}_n], \\ &\quad \dots, \\ &\quad \phi_{v-1}[\mathcal{B}_v] + \phi[\mathcal{N}_n]\}, \end{aligned} \quad (20)$$

where $\widetilde{\mathcal{N}}$ and $\widetilde{\mathcal{B}}$ indicate sequences applied in reverse order for odd-integer values of v and n . The linear property $\hat{M}\mathcal{B}(\mathcal{N}) = (\hat{M}\mathcal{B})(\hat{M}\mathcal{N})$ implies that nested dual pulses are also duals, namely, that $\mathcal{B}'_v(\mathcal{N}'_n)$ is the dual of $\mathcal{N}'_v(\mathcal{B}'_n)$; see lower panel of Fig. 3 for $\mathcal{N}'_3(\mathcal{B}'_7)/\mathcal{B}'_3(\mathcal{N}'_7)$.

There are many other π -composite rotation sequences with distinguishing properties, for example, the PB1, PB2 [43] and T1 [34] universal inversion pulses,

$$\text{T1} = (\pi)_0(\pi)_{\varphi_{\text{T1}}}(\pi)_{\varphi_{\text{T1}}}(\pi)_{-\varphi_{\text{T1}}}(\pi)_{-\varphi_{\text{T1}}}, \quad (21)$$

$$\begin{aligned} \text{PB1} &= (\pi)_{-\varphi_{\text{PB1}}}(\pi)_{-\varphi_{\text{PB1}}}(\pi)_{\varphi_{\text{PB1}}}(\pi)_{\varphi_{\text{PB1}}} \\ &\quad (\pi)_{\varphi_{\text{PB1}}}(\pi)_{\varphi_{\text{PB1}}}(\pi)_{-\varphi_{\text{PB1}}}(\pi)_{-\varphi_{\text{PB1}}}(\pi)_0, \end{aligned} \quad (22)$$

with $\varphi_{\text{T1}} = \arccos(-1/4)$, $\varphi_{\text{PB1}} = \arccos(-1/8)$. These have the general form $(\pi)_{\phi_a}(\pi)_{\phi_a}(\pi)_{\phi_b}(\pi)_{\phi_b} \dots (\pi)_0$; because axes or phases are repeated twice, $\hat{M}$ automatically maps the vector set onto itself, $\mathbf{e}_i^{(1)} \equiv \pm \mathbf{e}_i^{(0)}$ (or $\phi_i^{(1)} \equiv \pm \phi_i^{(0)}$), and we call these “half band” composite rotations, because each k th-order average propagator has the exact same magnitude about both $\beta' = 0$ and $\beta' = \pi$.

Returning to the general scenario $\beta = 2\pi/m$, where m is an integer, natural axes $\{\mathbf{e}_i^{(0)}\}$ for composing error-tuned rotation sequences are the vertices of regular shapes with m -fold rotational symmetry. For $m = 2$ these are vertices of regular n -gons (polygons) [44,50–52], and for higher m the vertices of regular polyhedra, namely, for $m = 3$, a tetrahedron, cube, or dodecahedron, and for $m = 4$ and $m = 5$, the vertices of an octahedron and icosahedron, respectively. Many

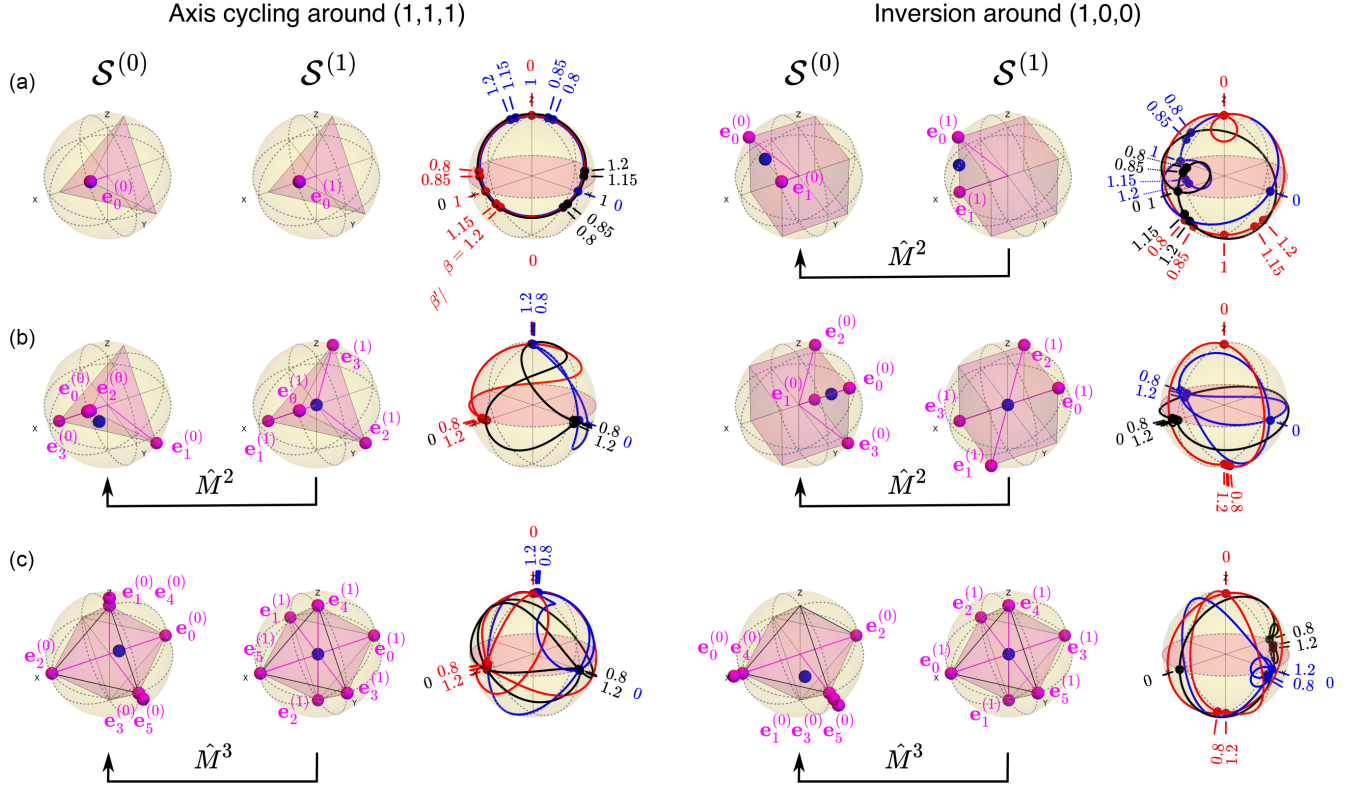


FIG. 4. Platonic sequences of composite rotations yielding $(1,1,1)$ -axis cycling or $(\pi)_x$ -inversion gates: (a) uncompensated, $\beta = 2\pi/3$; (b) compensated, $\beta = 2\pi/3$ [see Eqs. (25) and (26)]; (c) compensated, $\beta = 2\pi/4$ [see [57], Eqs. (27) and (28)]. Left and center plots show in 3D the orientations of $\{e_i^{(0)}\}$ and $\{e_i^{(1)}\}$, using the color convention given in Fig. (2). Right plots show orientation after the sequence as a function of β' , where the black, blue, and red curves denote an initial condition along the x , y , and z axis, respectively, and labels indicate positions for selected values of β'/β .

composite-rotation pulses, for example, $90_x 180_y 90_x$ (written equivalently as $90_x 90_y 90_y 90_x$), $\text{CORP}_2\text{SE}-\pi_0$, and phase-alternating sequences [53–56] involve $\pi/2$ rf-phase steps and octahedral symmetry. We say "natural" for the following reasons: (1) all vertices $e_i^{(k)}$ belong to the same finite set and (2) the vertices contain balanced subsets, meaning that n vectors $\{e_i^{(1)}\}$ can be chosen to satisfy $C^{(1)} = \mathbf{0}$ and then mapped via Eq. (6) to sequences $S^{(0)}$.

For $m = 3$ and $n = 4$, we applied this approach exhaustively and found two original sequences. First, choosing $C^{(1)} = \mathbf{0}$ for 4 vectors located in a diagonal plane,

$$\begin{aligned} \{e_i^{(1)}[\text{I}_3 4(1, 0, 0)]\} = \{ & (-1, 1, 1), (1, 1, -1), \\ & (1, -1, -1), (-1, -1, 1)\}/\sqrt{3}, \end{aligned} \quad (23)$$

we obtained a compensated $(\pi)_0$ rotation:

$$\begin{aligned} \{e_i^{(0)}[\text{I}_3 4(1, 0, 0)]\} = \{ & (-1, 1, 1), (-1, -1, -1), \\ & (-1, -1, 1), (-1, 1, -1)\}/\sqrt{3}, \end{aligned} \quad (24)$$

which can be compared with two $2\pi/3$ rotations applied sequentially about $(1,1,1)$ and $(1, -1, 1)$ [Figs. 4(a) and 4(b)].

Another balanced set comprises the four vertices of a tetrahedron,

$$\begin{aligned} \{e_i^{(1)}[\text{P}_3 4(1, 1, 1)]\} = \{ & (1, 1, 1), (1, -1, -1), \\ & (-1, 1, -1), (-1, -1, 1)\}/\sqrt{3}, \end{aligned} \quad (25)$$

and the net transformation is a compensated $(2\pi/3)$ rotation about $(1,1,1)$,

$$\begin{aligned} \{e_i^{(0)}[\text{P}_3 4(1, 1, 1)]\} = \{ & (1, 1, 1), (-1, 1, -1), \\ & (1, 1, 1), (1, -1, -1)\}/\sqrt{3}, \end{aligned} \quad (26)$$

or an axis-cycling Clifford gate [11,58], and is accurate within 5 degrees for up to $\pm 20\%$ error in β [Figs. 4(a) and 4(b)].

For $m = 4$ and $n = 6$, axes can be chosen as $\{e_i^{(1)}\} = \{\mathbf{e}_x, \mathbf{e}_{-x}, \mathbf{e}_y, \mathbf{e}_{-y}, \mathbf{e}_z, \mathbf{e}_{-z}\}$ (no specific order) to ensure balance. To within a global z rotation, we find the only sequence providing a compensated π_ϕ rotation is

$$(\pi/2)_0(\pi/2)_{\pi/2}(\pi/2)_\pi(\pi/2)_{\pi/2}(\pi/2)_0(\pi/2)_{\pi/2}, \quad (27)$$

which is Derome's $(\pi)_y$ sequence [57,59]. Relaxing the balance constraint to $C_z^{(1)} = 0$ we also find point-to-point compensated sequences, such as Tycko's amplitude-error-compensated sequence $(\pi/2)_0(\pi/2)_0(\pi/2)_{2\pi/3}(\pi/2)_{2\pi/3}(\pi/2)_0(\pi/2)_0 \equiv (\pi)_0(\pi)_{2\pi/3}(\pi)_0$ [59,60] for $n = 6$, and Levitt's offset-error-compensated sequence $(\pi/2)_0(\pi/2)_{\pi/2}$

$(\pi/2)_{\pi/2}(\pi/2)_{\pi/2}(\pi/2)_0 \equiv (\pi/2)_0(3\pi/2)_{\pi/2}(\pi/2)_0$ [61,62] for $n = 5$.

If vectors outside the equatorial plane are permitted, we also find undiscovered solutions for compensated axis cycling rotations such as

$$\{\mathbf{e}_i^{(0)}[P_46(1, 1, 1)]\} = \{-\mathbf{e}_x, \mathbf{e}_z, \mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z, \mathbf{e}_y\}, \quad (28)$$

as shown in Fig. 4(c), or

$$\{\mathbf{e}_i^{(0)}[P_46'(1, 1, 1)]\} = \{-\mathbf{e}_z, \mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z, \mathbf{e}_x, \mathbf{e}_y\}, \quad (29)$$

both for $n = 6$.

In qubit systems, sequences involving non-equatorial axes such as the one given in Eq. (26) can be implemented using near-resonant ac fields with the specific detuning frequencies to set the latitude of the rotation axes $\mathbf{e}_i^{(0)}$. This approach evidently befits higher-symmetry sets, for example, $m = 5$ cyclic sequences involving axis subsets of the vertices of the regular icosahedron. A couple of sequences employing this structure have been used with angles $\beta = 2\pi/5$ and $n = 10$ or $n = 12$ for dipolar-decoupling of diamond nitrogen-vacancy centers [63].

Polyhedral symmetry of the axis sets is, however, not always necessary. The design of compensated rotation sequences may capitalize upon relationships between interaction frame cycles for different β values, such as between $\beta = \pi/2$ ($m = 4$) and $\beta = \pi$ ($m = 2$). According to Eq. (6), for $\beta = \pi/2$, $\mathbf{e}_i^{(2)}$ must relate to $\mathbf{e}_i^{(0)}$ in the same way that $\mathbf{e}_i^{(1)}$ relates to $\mathbf{e}_i^{(0)}$ for $\beta = \pi$. Known principles follow from this result include the doubling theorems, where (1) bisection of any $\beta = \pi$ inversion sequence that is symmetric under phase order reversal generates a narrowband $\pi/2$ sequence [64] and (2) concatenation of any $\beta = \pi/2$ sequence with its phase-reversed inverse generates a narrowband π sequence [42].

We have used Eq. (6) to identify a third connection, that appears to be hitherto undiscovered, between narrowband universal $(\pi)_x$ rotations composed of units $\beta = \pi/2$ and broadband universal $(\pi)_x$ rotations composed of units $\beta = \pi/2$. Vectors $\{\mathbf{e}_i^{(1)}\}$ for both sequences are related by a global rotation of $\pi/2$ about the x axis plus a global rotation about z . The global rotation of toggling frame axes does not alter the centroid, since it is already zero. The overall recipe is then (1) replace every π element in a narrowband $(\pi)_x$ sequence with two $\pi/2$ elements of the same phase, (2) rotate each vector $\mathbf{e}_i^{(0)}$ about x by $\pi/2$, and cyclically permuting the sequence order by half its length, (3) apply the $m = 4$ inverse-toggling-frame transformation $\hat{M}^3$, and finally (4) rotate each vector $\mathbf{e}_i^{(0)}$ about the z axis by a constant angle to become reflection symmetric in the xz plane. This method relates several of the $\beta = \pi/2$ sequences discovered by brute force in Ref. [59] with the analytical solutions of Eq. (17). Figure 5 illustrates the relationship for $n = 15$, a value for which the $\pi/2$ counterpart would be intractable by brute-force computation.

IV. RESULTS: APPLICATION TO DYNAMICAL DECOUPLING (DD)

Let us now relate these findings to qubit control protocols aimed at noise-suppression and decoherence mitigation, which involve sequences of n β kicks that intersperse free-system evolution delays τ_j ($j \in [0, n]$) [6,7,39]. In the limit

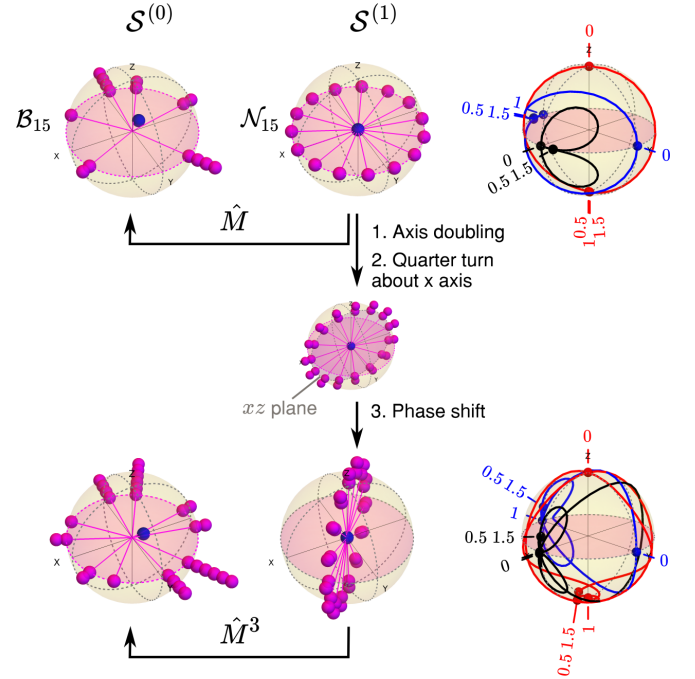


FIG. 5. Compensated rotation sequences with cycles $m = 2$ and $m = 4$ are interrelated by the general principle where any order-reversal-symmetric, broadband inversion sequence composed of n $\beta = \pi$ rotations can be converted to an order-reversal-antisymmetric broadband sequence composed of $2n$ $\beta = \pi/2$ rotations. The example shown is for $n = 15$.

$\tau_j \rightarrow 0$ these can equate to composite pulses. In the delta-pulse limit, DD sequences are of the form

$$S^{(0)} = \tau_n \prod_{j=0}^{n-1} R(\beta, \mathbf{e}_j^{(0)}) \tau_j, \quad (30)$$

where axes and delays are tailored for robustness to pulse errors, plus averaging of oscillatory or static external fields. Equation (6) has implications in the following scenarios:

(1) An oscillating field along the z axis, of time-dependent amplitude $a \cos(\omega t)$. The propagator of $S^{(0)}$ can be expressed as a product of rotations about axes that are phase-shifted by angles θ_j depending on ω and the kick time $t_j = \sum_{l=0}^j \tau_l$:

$$U_n(S^{(0)}(\omega)) = R(\theta_n(\omega), \mathbf{e}_z) \prod_{j=0}^{n-1} R(\beta, \hat{R}(\theta_j(\omega), \mathbf{e}_z) \cdot \mathbf{e}_j^{(0)}), \quad (31)$$

$$\theta_j(\omega) = (a/\omega) \sin(\omega t_j); \quad j \in [0, n-1], \quad (32)$$

$$\theta_n(\omega) = (a/\omega) [\sin(\omega \tau_n) - \sin(\omega \tau_{n-1})]. \quad (33)$$

Toggling-frame phases are determined using Eq. (7) and $\phi_j^{(0)}(\omega) = \phi_j^{(0)} + \theta_j(\omega)$.

A nonrobust DD sequence that has been used in both liquid-state [65] and solid-state [66] qubit control is Uhrig decoupling (UDD), a n $\beta = \pi$ -pulse scheme that is optimal for highpass noise filtering (giving a filter of cutoff order n in ω , due to design where kick times correspond to the Chebyshev nodes of the second kind on $[0, n]$: $t_j = n \sin^2[\pi j/(2n+2)]$,

$\mathbf{e}_j^{(0)} \equiv \mathbf{e}_x$) but is highly sensitive to inaccuracy and inhomogeneity in β . The source of the nonrobustness is the constant pulse phase $\phi_j^{(0)} = 0$, where $\mathbf{C}^{(1)} \approx \mathbf{e}_x$ for small ω . Other DD sequences based on dual-compensated rotations, e.g., URn [5], on the other hand, can be robust to pulse errors but not necessarily noise [67].

Fully robust DD schemes—with simultaneous tolerance to β , rf-detuning, and ω errors—generally involve concatenation of noise-tolerant and dual-compensated rotation sequences [68,69]. One of the most experimentally successful to date is the $n = 20$ -pulse “KDD” scheme [4,10,70,71], which is a concatenation (or *supercycle* [72]) of the UR4 sequence $\text{XY4} = \tau/2 - (\pi)_0 - \tau - (\pi)_{\pi/2} - \tau - (\pi)_0 - \tau - (\pi)_{\pi/2} - \tau/2$ [5] and the element $\text{KDD} = \tau/2 - (\pi)_{\pi/6} - \tau - (\pi)_0 - \tau - (\pi)_{\pi/2} - \tau - (\pi)_0 - \tau - (\pi)_{\pi/6} - \tau/2$, which for $\tau = 0$ is the Tycko-Pines-Guckenheimer U_5 composite pulse [37,73,74]. The phases are $\{\phi^{(0)}[\text{KDD}]\} \equiv \{\phi^{(0)}[\text{XY4}(\text{U}_5)]\}$ following the notation of Eq. (19). Other concatenations of URn and dual-compensated rotations, e.g., $\text{UR6}(\text{U}_5) \equiv \text{BB1}(2\pi)(\text{U}_5)$, $\text{UR8}(\text{U}_5)$ [75], $\text{UR10}(\text{U}_5)$, $\text{XY4}(\text{U}_7)$, and so on, extend KDD to arbitrarily improved DD against pulse errors β . We note, historically, that KDD was first reported by the Pines group well before the advent of quantum information processing. Fujiwara and Nagayama, around that time, also presented schemes involving U_n concatenated with $\text{MLEV4} = \tau/2 - (\pi)_0 - \tau - (\pi)_{\pi/2} - \tau - (\pi)_{\pi/2} - \tau - (\pi)_0 - \tau/2$, which were compensated against rf-pulse error but not noise [76,77].

It is because of the cyclic map for $m = 2$ Eqs. (7) and (8) that the duals of U_n or ASBOn , moreover, can be nested in URn to produce “anti-DD” sequences that are robust in ω and narrowband in β ; see Fig. 6. We also find that U_n can be substituted by other n -odd dual-compensated sequences, such as $\text{ASBOn}\Omega$ and ASBOnB_1 [36], giving $\text{XY4}(\text{ASBOn}\Omega)$ and $\text{XY4}(\text{ASBOnB}_1)$ as robust sequences.

The axis-doubling principle used for mapping $m = 2 \rightarrow m = 4$ in Fig. 5 provides further solutions, such as $\text{XY4}(\text{U}_5 \vee \text{U}_5)$ as a robust DD sequence for $\beta = \pi/2$. Here $\vee$ indicates a self-riffling operation to duplicate the phases: e.g., $\{\phi_a, \phi_b, \dots\} \vee \{\phi_a, \phi_b, \dots\} = \{\phi_a, \phi_a, \phi_b, \phi_b, \dots\}$.

(2) A time-independent field of irreducible tensor rank λ . Consider a field $a_{\lambda,-\mu} T_{\lambda,\mu}$, where the rank λ is invariant under global rotation. The operator part transforms as $R(\beta, \mathbf{e}_j^{(i)})^{-1} T_{\lambda,\mu} R(\beta, \mathbf{e}_j^{(i)}) = \sum_{\mu'=-\lambda}^{\lambda} T_{\lambda,\mu'} D_{\mu,\mu'}^{\lambda} [R(\beta, \mathbf{e}_j^{(i)})]$, where $\mathbf{D}^{\lambda}$ is the $(2\lambda + 1)$ -by- $(2\lambda + 1)$ Wigner matrix, and substitution into Eq. (30) leads to a first-order-average propagator given by

$$(\tilde{\mathbf{V}}_n)^{[1]}(a_{\lambda,-\mu}) = \exp \left(-ia_{\lambda,-\mu} \sum_{\mu'=-\lambda}^{\lambda} T_{\lambda,\mu'} \kappa_{\lambda,\mu,\mu'} \right), \quad (34)$$

$$\kappa_{\lambda,\mu,\mu'} = \frac{1}{\sum_j \tau_j} \sum_{j=0}^{n-1} \tau_j D_{\mu,\mu'}^{\lambda} (\mathbf{U}_j^{-1}). \quad (35)$$

For $\lambda = 1$, this reduces to the familiar vector average in Eq. (B1).

Sequences that cancel $\mathbf{D}^{\lambda}$ components by enforcing $\kappa_{\lambda,\mu,\mu'} = 0$ are well established, and for $\lambda \geq 1$ almost invariably involve polyhedral symmetries. Solid-state NMR, for

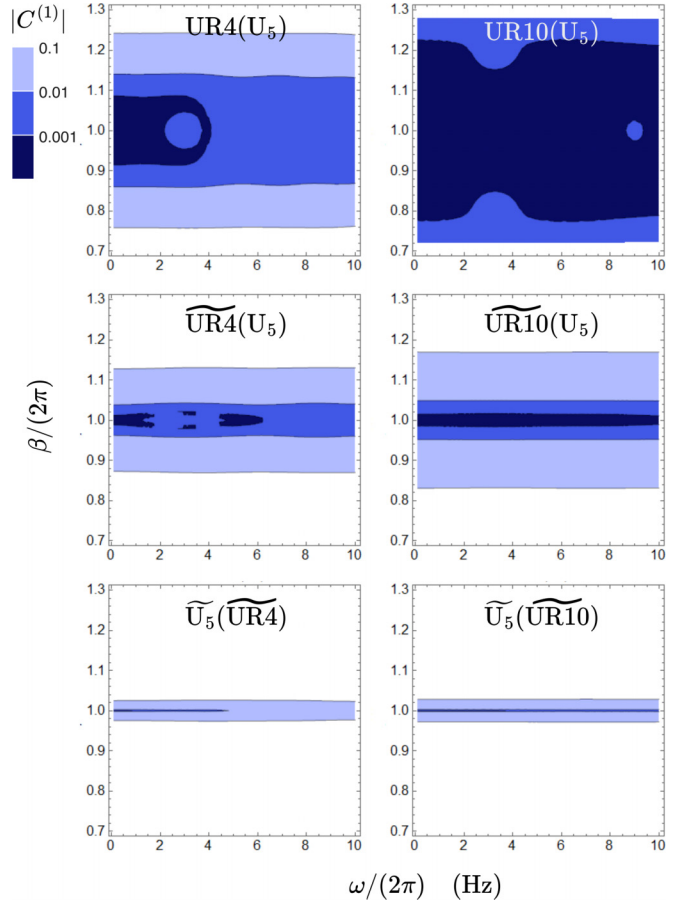


FIG. 6. Contour-plot representations of the magnitude of the centroid ($|\mathbf{C}^{(1)}| \propto |\langle \tilde{\mathbf{V}}_n \rangle^{[1]}|$) in the pulse toggling frame for single cycles of DD sequences composed of a concatenation between two compensated rotation sequences with $m = 2$. Down each column, we explore the concatenation of duals (indicated by the tilde), which produces sequences tolerant to a wide range of ω , but tunable performance in β' . $\text{UR4}(\text{U}_5)$ is the well-known KDD sequence [4,10,70,71].

example, since the 1980s has used sequences with $\kappa_{2,0,\mu'} = 0$ to decouple the secular part of g-tensor anisotropies or pair dipole-dipole couplings—both rank-2 interactions [78]. An example involving a tetrahedral geometry is virtual magic-angle spinning (MAS) by $\beta = 2\pi/3$ about $(1, 1, 1)/\sqrt{3}$ axes [79–81]. Other sequences, such as $\text{WHH-4} = (\pi/2)_0 - \tau - (\pi/2)_{\pi/2} - 2\tau - (\pi/2)_{-\pi/2} - \tau - (\pi/2)_{\pi} - 2\tau$, MREV [82,83], echo-WAHUHA [6], BR24 [84], etc. [85] involve $\beta = 2\pi/4$ and are octahedral. Zero-field analogs developed by Lee, Suter, and Pines achieve the same type of decoupling by suppressing $\kappa_{2,\mu,\mu'}$ without the secular approximation [86] and belong to the same respective symmetries. Other sequences using icosahedral vertices and $\beta = 2\pi/5$ are known to suppress higher-rank terms such as $\kappa_{3,0,\mu'} = 0$ [87] or to decouple rank-1 or rank-2 components selectively [63]. Pulse sequence construction using polyhedral symmetry may be viewed as a special case of constructing pulse sequences using the vertices of polytopes [88,89] that may have an arbitrary number of dimensions in general.

The selection rules determining which components $\kappa_{\kappa,\mu,\mu'}$ are zero are also well established from group theory [90,91]

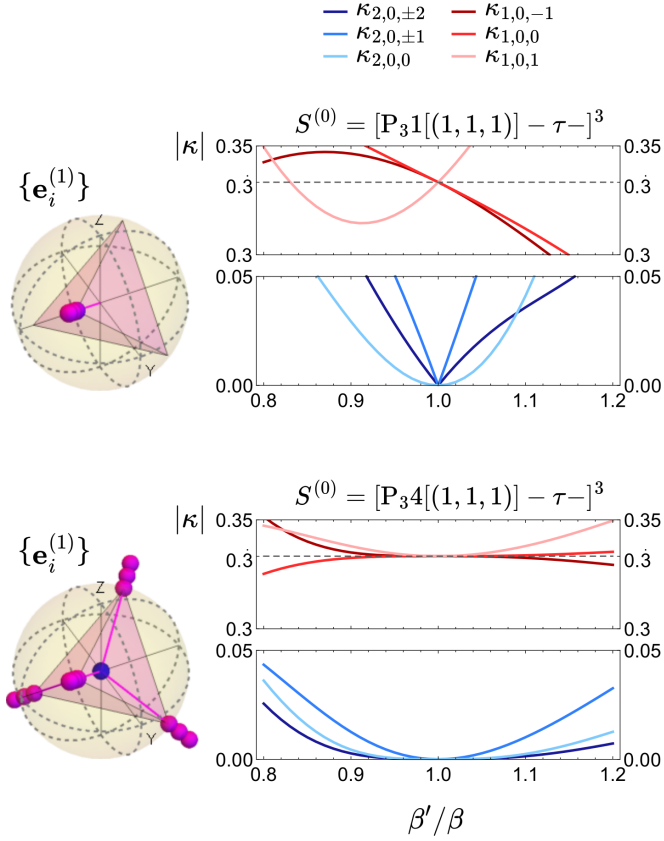


FIG. 7. First-order average Hamiltonian suppression of a time-independent background field with rank $\lambda = 2$, using (top) single $\beta = 2\pi/3$ pulses about the $(1, 1, 1)/\sqrt{3}$ axis as a form of “virtual” magic-angle spinning (MAS); (bottom) virtual MAS with $(1, 1, 1)/\sqrt{3}$ pulses compensated for angle error $\epsilon = \beta' - \beta$, defined in Eq. (26). $\kappa_{\lambda,\mu,\mu'}$ are as defined in Eq. (35).

and are a central feature of symmetry-based sequence design [72]. To guarantee $\mathbf{D}^\lambda(R) = \mathbf{0}$, the weighted sum in Eq. (35) can be viewed as a finite-sampling implementation of the Peter-Weyl theorem. This may be solved using the orbit-stabilizer theorem, leading to vector sets $\{\mathbf{e}_i^{(1)}\}$ comprising vertices of a regular polyhedron and its dual polyhedron, weighted by rotation number: e.g., for OEDD ($n = 48$) [87], $\{\mathbf{e}_i^{(1)}\}$ comprises cube vertices $\{(\pm 1, \pm 1, \pm 1)/\sqrt{3}\}$ with weight 3 and rotation angle $\beta_i = 2\pi/3$, plus octahedron vertices $\pm \mathbf{e}_x, \pm \mathbf{e}_y, \pm \mathbf{e}_z$ with weight 4 and $\beta_i = \pi/2$. Alternatively, for IEDD ($n = 120$) [87], $\{\mathbf{e}_i^{(1)}\}$ comprises the 20 dodecahedron vertices with weight 3 and rotation angle $\beta_i = 2\pi/3$, plus 12 icosahedron vertices with weight 5 and $\beta_i = 2\pi/5$. For the tetrahedron, which is a self-dual polyhedron, this geometric construction leads to a cube (TEDD, $n = 24, m = 3$ [87]). We note that solutions like OEDD and IEDD obey a modified form of Eq. (6) where β is replaced by β_i , and the cyclic relationship still holds, with m given by a lowest common multiplier: for OEDD, $m = 12 = \text{lcm}(3, 4)$, and for IEDD $m = 15 = \text{lcm}(3, 5)$.

The sequences above are not inherently robust to errors in β . However, replacing each axis-cycling pulse with a compensated rotation can restore the desired averaging even under angle miscalibration. In Fig. 7 we propose a mini-

mal, tetrahedral-symmetry composite pulse for virtual-MAS, where the single axis-cycling rotations are replaced by a compensated rotation of the form given in Eq. (26). Whether such constructions are robust to other error sources still remains an open question; however, they may offer a practical path forward.

V. CONCLUSION

Normally in coherent averaging theory, only one toggling-frame transformation is used to collect error terms. Here we considered: what happens if we apply the same transformation repeatedly? Surprisingly, the axes cycle back to themselves after a fixed number of steps, depending only on the rotation angle: a succession of m iterative transformations always returns the original axis set. A general formulation of this result appears to be previously unrecognized, and the cyclicity reveals hidden structure in many known composite pulses.

First, the implication of self-mapping vector sets $\{\mathbf{e}_i^{(k)}\}$ $k \in \{0, \dots, m\}$ is a global property that explains why most error-tolerant composite-rotation and DD sequences—other than optimal control—involve angles $2\pi/m$. For $m = 2$, it leads to the duality of π -pulse sequences in pulse-length error. For $m = 3$ and 4, this led us to derive several original rotation sequences. More shall be published in future work. We also established direct connections between cycles that relate existing sequence families, particularly between $m = 2$ and $m = 4$.

All of these results arguably constitute a unified theory of compensated rotations, because for given values of n and m , there is only a finite number of sequences with zero centroid. For now, however, a more exhaustive analysis of that sort is future work and we close by repeating the appropriate words of Ray Freeman from almost 40 years ago [92]: “We have certainly not heard the last word on composite pulses.”

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are openly available [93].

APPENDIX A: TOGGLING FRAMES FOR ROTATION ERRORS

The error-tolerance properties of $S^{(0)}$ can be analyzed by transforming into alternative reference frames. We review the case where errors in rotation angle (but not axis) take the general form $\beta \rightarrow \beta' \equiv \beta + \epsilon$ for fixed nominal rotation angle β and a constant or proportional error ϵ , and arise when a spin is addressed by on-resonance ac pulses with a miset Rabi frequency ($\omega_{R,i}$) or length ($\tau_i = \beta' / \omega_{R,i}$). To make $S^{(0)}$ tolerant to these, one must satisfy the condition $U_n(S^{(0)}; \beta + \epsilon) \approx U_n(S^{(0)}; \beta)$ for $|\epsilon|$ below some target threshold. For this, the toggling frame defined by

$$U_n(S^{(0)}; \beta + \epsilon) = U_n(S^{(0)}; \beta) \tilde{V}_n(\epsilon) \quad (\text{A1})$$

is convenient because the overall propagator is split into parts that depend separately on ϵ and β .

The ϵ -dependent part, denoted in Eq. (A1) using a tilde ($\sim$), is a sequence of rotations by ϵ , $\tilde{S}^{(0)} \equiv (\epsilon)_{\tilde{e}_0^{(0)}}(\epsilon)_{\tilde{e}_1^{(0)}} \dots (\epsilon)_{\tilde{e}_{n-1}^{(0)}}$, about n axes defined by

$$\tilde{V}_n = \prod_{i=0}^{n-1} R(\epsilon, \tilde{e}_i^{(0)}), \quad (\text{A2})$$

$$\tilde{e}_i^{(0)} = [U_i(S, \beta)]^{-1} e_i^{(0)}; \quad i \geq 1, \quad (\text{A3})$$

$$\tilde{e}_0^{(0)} = e_0^{(0)}. \quad (\text{A4})$$

This is the same transformation, $\hat{M}$, defined by Eq. (2) and Eq. (3) and used in the main text.

Throughout the main text we have used the alternative notation where, in lieu of the tilde, we add 1 to the superscript index, so $\tilde{e}_i^{(0)} \equiv e_i^{(1)}$. This is a more natural way to indicate the cyclic property, and recognizes the fact that $\tilde{V}_n$ is the propagator of a rotation sequence, and if—within that—we used angles β rather than ϵ , then we would equate $\tilde{e}_i^{(1)} \equiv e_i^{(2)}$, and so on. We also note that the axes sets are coincident, $e_i^{(m)} = e_i^{(0)}$, always, for $\beta' = 0$ and 2π .

Frequency-detuning error ($\delta\omega$) is another source of imperfection for ac pulses. This scenario corresponds to changing the latitude, θ_i , of the vectors $e_i^{(0)}$ in the pulse rotating frame to $(\theta_i + \delta\theta_i) = \arctan(\Delta\omega_i + \delta\omega, \omega_R)$, where $\Delta\omega_i$ is the nominal resonance frequency offset and we assume field oscillates in the xy plane, perpendicular to the main static field axis along z . Generally this case presents no simple analytical form for the toggling frame vectors. However, for $\beta = \pi$ and $\Delta\omega_i = 0$, the vectors are simply related to those of the tilde frame, $\{\tilde{e}_i\}$, by alternating $\pi/2$ rotations about the z axis [94,95]. We denote this toggling frame with a tilde plus a prime ($\tilde{\prime}$):

$$\tilde{e}'_i \equiv R((-1)^i \pi/2, e_z) \tilde{e}_i. \quad (\text{A5})$$

APPENDIX B: AVERAGE LIOUVILLIAN THEORY

Provided the error is small, ($|\epsilon/\beta| < 1$), the Baker-Campbell-Hausdorff-Dynkin formula can expand $\tilde{V}_n$ as a convergent power series in ϵ . The index k indicates a mean or "average" propagator in ϵ^k , denoted as the k th-order average Liouvillian (according to the Nielsen-Levitt convention [96] for which k is numbered 1 higher than in earlier literature). The lowest few terms, where we indicate k as a superscript

in square brackets $(\tilde{V}_n)^{[k]}(\epsilon)$, have the following form with implicit β' dependence in $\{\tilde{e}_i\}$:

$$(\tilde{V}_n)^{[1]}(\epsilon) = R\left(\epsilon, \frac{1}{n} \sum_{i=0}^{n-1} \tilde{e}_i\right), \quad (\text{B1})$$

$$(\tilde{V}_n)^{[2]}(\epsilon) = R\left(\epsilon^2, \frac{1}{2n} \sum_{j=0}^{n-1} \sum_{i=0}^{j-1} \tilde{e}_j \wedge \tilde{e}_i\right), \quad (\text{B2})$$

$$\begin{aligned} (\tilde{V}_n)^{[3]}(\epsilon) = R\left(\epsilon^3, \frac{1}{6n} \sum_{k=0}^{n-1} \sum_{j=0}^{k-1} \sum_{i=0}^{j-1} \right. \\ \left. \times \{[\tilde{e}_i \wedge \tilde{e}_j] \wedge \tilde{e}_k + [\tilde{e}_k \wedge \tilde{e}_j] \wedge \tilde{e}_i\} \right. \\ \left. - \frac{1}{2} \{[\tilde{e}_k \wedge \tilde{e}_j] \wedge \tilde{e}_j + [\tilde{e}_j \wedge \tilde{e}_k] \wedge \tilde{e}_k\} \right). \end{aligned} \quad (\text{B3})$$

and ensemble average is indicated by the overbar ($\overline{}$). The superscript is formatted as $[k]$ to clearly distinguish the index from that of the toggling-frame axis sets, (j) .

Expressions for higher k orders in terms of vector-product sums are generally not needed, but may be obtained by explicit evaluation or by recursion [97,98].

The leading error-compensation properties of $S^{(0)}$ can thus be linked to *balance* of the vectors sets $\{e_i\}$, $\{\tilde{e}_i\}$, and $\{\tilde{e}'_i\}$. A vector set $\{v_i\}$ is balanced if its arithmetic mean or *centroid*

$$\begin{aligned} C[\{v_i\}] &= (1/n) \sum_{i=0}^{n-1} v_i \\ &\equiv (C_x[\{v_i\}], C_y[\{v_i\}], C_z[\{v_i\}]) \end{aligned} \quad (\text{B4})$$

is zero, and if the set is not balanced, we say it is *unbalanced*.

Given Eq. (B1), $(\tilde{V}_n)^{[1]} = R(\epsilon, C[\tilde{e}_i])$, we begin to form a recipe to formulate $S^{(0)}$: pick a set of balanced vectors, and reverse transform these to the nontoggling frame of rotations. Generally, a sequence that is broadband to errors ϵ about $\beta' = \beta$ will have balance $C[\{\tilde{e}_i^{(0)}\}] = \mathbf{0}$. A sequence that is narrowband will be unbalanced about $\beta' = \beta$, $C[\{\tilde{e}_i^{(0)}\}] \neq \mathbf{0}$, but balanced about $\beta' = 0$ $C[\{e_i^{(0)}\}] = \mathbf{0}$. The second result follows because Eqs. (B1)–(B3) can be applied using $\tilde{e}_i \equiv e_i$ to describe the average propagators around $\beta' = 0$, by equivalence to $\beta' = 2\pi$. For detuning errors where Eq. (A5) is valid, balance is required separately for both odd- and even-indexed vectors $\tilde{e}'_i$ (see, for example, the U_5 or “Knill” pulse described in [37] and [73]).

The second- and higher-order error terms in Eqs. (B2) and (B3) can be made zero for certain chronological-order symmetries of $S^{(0)}$. Known relations comprise (1) order-reversal symmetry [99,100] of the sequence, leading to vanishing even order propagators $(\tilde{V}_n)^{[2k]} = 0$ about $\beta' = \beta$ for $\tilde{e}_i = \tilde{e}_{(n-i-1)}$ and similarly $(\tilde{V}_n)^{[2k]} = 0$ about $\beta' = 0$ for $e_i = e_{(n-i-1)}$; (2) order-reversal antisymmetry, leading to vanishing of *all* order propagators $(\tilde{V}_n)^{[k]} = 0$ for $\tilde{e}_i = -\tilde{e}_{(n-i-1)}$; and (3) nesting, where if $S^{(0)}$ consists of two or more subunits all individually satisfying $(\tilde{V}_n)^{[1]} = 0$, then the second- and third-order average propagators of $S^{(0)}$ are simply the product of the second-

and third-order average propagators over the individual sub-cycles, respectively [84].

Combining these properties, widest error tolerance is given when $C[\tilde{e}_i] = \mathbf{0}$ and $S^{(0)}$ is order-reversal symmetric.

APPENDIX C: SUCCESSIVE $\hat{M}$ TRANSFORMATIONS

Here we show how Eq. (6) is generated by expanding and collecting rotation operators. To avoid notational clutter, we use the abbreviation $R_j = R(-\beta, \mathbf{e}_j^{(0)})$, thus rewriting Eq. (3) as

$$\mathbf{e}_i^{(1)} = U_i^{-1} \mathbf{e}_i^{(0)} = R_0 R_1 \dots R_{i-1} \mathbf{e}_i^{(0)}. \quad (\text{C1})$$

The first few terms are

$$\begin{aligned} \mathbf{e}_1^{(1)} &= R_0 \mathbf{e}_1^{(0)}, \\ \mathbf{e}_2^{(1)} &= R_0 R_1 \mathbf{e}_2^{(0)}, \\ \mathbf{e}_3^{(1)} &= R_0 R_1 R_2 \mathbf{e}_3^{(0)}, \end{aligned} \quad (\text{C2})$$

and so on. If we then want to apply $\hat{M}$ to $\{\mathbf{e}_i^{(1)}\}$, the rotations through β must be performed about these new axes. To keep the original axes $\{\mathbf{e}_i^{(0)}\}$ we use the identity for a rotation R of an object about a rotated vector, $A\mathbf{e}$, where A is unitary ($A^\dagger A = 1$):

$$R(\beta, A\mathbf{e}) = AR(\beta, \mathbf{e})A^{-1}. \quad (\text{C3})$$

The resulting expressions for $\mathbf{e}_i^{(2)}$ are

$$\begin{aligned} \mathbf{e}_1^{(2)} &= R_0 \cdot \underbrace{R_0 \mathbf{e}_1^{(0)}}_{\mathbf{e}_1^{(1)}} = R_0^2 \mathbf{e}_1^{(0)}, \\ \mathbf{e}_2^{(2)} &= R_0 \cdot \underbrace{R_0 R_1 R_0^{-1}}_{R(\beta, \mathbf{e}_1^{(1)})} \cdot \underbrace{R_0 R_1 \mathbf{e}_2^{(0)}}_{\mathbf{e}_2^{(1)}} = R_0^2 R_1^2 \mathbf{e}_2^{(0)}, \\ \mathbf{e}_3^{(2)} &= R_0 \cdot \underbrace{R_0 R_1 R_0^{-1}}_{R(\beta, \mathbf{e}_1^{(1)})} \cdot \underbrace{R_0 R_1 R_2 R_1^{-1} R_0^{-1}}_{R(\beta, \mathbf{e}_2^{(1)})} \\ &\quad \cdot \underbrace{R_0 R_1 R_2 \mathbf{e}_3^{(0)}}_{\mathbf{e}_3^{(1)}} = R_0^2 R_1^2 R_2^2 \mathbf{e}_3^{(0)}. \end{aligned} \quad (\text{C4})$$

By induction from the next few sets of terms, such as

$$\begin{aligned} \mathbf{e}_3^{(3)} &= R_0 \cdot \underbrace{(R_0 R_0) R_1 (R_0^{-1} R_0^{-1})}_{R(\beta, \mathbf{e}_1^{(2)})} \\ &\quad \cdot \underbrace{(R_0 R_0 R_1 R_1) R_2 (R_1^{-1} R_1^{-1} R_0^{-1} R_0^{-1})}_{R(\beta, \mathbf{e}_2^{(2)})} \\ &\quad \cdot \underbrace{R_0^2 R_1^2 R_2^2 \mathbf{e}_3^{(0)}}_{\mathbf{e}_3^{(2)}} = R_0^3 R_1^3 R_2^3 \mathbf{e}_3^{(0)}, \end{aligned} \quad (\text{C5})$$

we see the general formula is

$$\mathbf{e}_i^{(k)} = \prod_{j=0}^{i-1} R_j^k \mathbf{e}_i^{(0)}, \quad (\text{C6})$$

which is identical to Eq. (6).

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