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Symmetric Phase-Alternating Composite Pulses

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We derive sequences of new composite pulses that can provide constant rotations of arbitrary flip angle in the presence of large resonance offset effects. These symmetric sequences use only 180° phase shifts, and have the same symmetry properties as a single radiofrequency pulse. For two-level systems, these composite pulses behave like ideal single rf pulses, making them of potential use in a wide variety of experimental situations.

INTRODUCTION

Composite pulses (*1, 2*) have found use in many NMR experiments for which a single radiofrequency pulse, due to its restricted bandwidth, is inadequate. Examples of improvements in excitation or inversion bandwidths in isotropic liquids (*3–5*) and solids (*6–9*) are well-established. Typical composite pulses are constructed from constant-amplitude rectangular pulses using a piecewise constant phase function; in addition the frequency of the rf irradiation is almost always fixed. These restrictions have arisen both from the hardware limitations imposed by most high-resolution spectrometers and from the difficulty of analyzing more elaborate irradiation strategies. There are, nevertheless, examples of continuous modulation schemes in the recent literature (*10–13*).

Of the more conventional composite pulses, an important subset is the set of all phase-alternating composite pulses, where the phase function ψ_k for the kth pulse is restricted to the values of 0 or π. These sequences have proven effective for resonance offset compensation in broadband spin decoupling (*14–18*), broadband spin inversion (*5*), and spatial localization (*19*) experiments in liquids, and for broadband three-level excitation in solids (*7, 8*). Here we restrict ourselves to the family of phase-alternating composite pulses that are symmetric in time, and demonstrate analytically that even this small subset of all possible composite pulses can support sequences providing both controlled uniform rotations applicable to any initial state of the spins and specific uniform "point-to-point" transformations of one spin operator, for example, I_z, into another, for example, I_y. These latter composite pulses can be used to create transverse magnetization of constant phase over quite large bandwidths, eliminating the usual phase shift of the excitation as a function of resonance offset.

THEORY

We consider an ensemble of isolated spin-$\frac{1}{2}$ nuclei, and concentrate on the single imperfection that the irradiation frequency ω is mismatched to the resonance frequency

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ω_0, producing a resonance offset $\Delta\omega = \omega - \omega_0$. In the usual rotating frame, the Hamiltonian during the kth pulse, of duration τ_k, can be written

$$H_k = H_{k\mathrm{rf}} + V \quad [1]$$

where

$$H_{k\mathrm{rf}} = (-1)^k \omega_1 I_x \quad [2]$$

$$V = \Delta\omega I_z. \quad [3]$$

The symmetric phase-alternating composite pulse leads to an overall propagator at time $\tau = \sum_k \tau_k$ given by

$$U(\tau) = \prod_k \exp(-i\tau_k H_k) = \exp(i\alpha \mathbf{n} \cdot \mathbf{I}) \quad [4]$$

corresponding to a pure rotation of angle α about a unit axis n. In general, both α and n will depend on $\Delta\omega$. Our first goal is to show that, by the correct choice of the pulsewidths τ_k, this dependence can be eliminated over a range of $\Delta\omega$ about exact resonance for any desired flip angle α.

Our analysis begins with coherent averaging theory (*20*), in which the operator V, after transforming into an interaction representation, is treated as a perturbation. By examining the perturbation series in powers of $\Delta\omega/\omega_1$, we can estimate how strong the dependence on V will be. To this end, we decompose the propagator $U(\tau)$ in the manner suggested by Tycko *et al.* (*9*) and write

$$U(\tau) = U_{\mathrm{rf}}(\tau) U_V(\tau) \quad [5]$$

where $U_{\mathrm{rf}}(\tau)$ represents the ideal transformation and $U_V(\tau)$ the imperfection:

$$U_V(\tau) = T \exp\left\{-i \int \tilde{V}(t) dt\right\} \quad [6]$$

$$\tilde{V}(t) = U_{\mathrm{rf}}(t)^{-1} V U_{\mathrm{rf}}(t) \quad [7]$$

and T denotes time ordering. We then use the Magnus expansion (*21*) to write $U_V(\tau)$ as the complex exponential of an averaged operator $\bar{V}$, which is expressed as an infinite series:

$$\bar{V} = V^{(0)} + V^{(1)} + V^{(2)} + \cdots. \quad [8]$$

The terms in the series are well-known. For $V^{(0)}$ and $V^{(1)}$ we have

$$V^{(0)} = \frac{1}{\tau} \int_0^\tau dt \tilde{V}(t) \quad [9]$$

$$V^{(1)} = \frac{-i}{2\tau} \int_0^\tau dt_1 \int_0^{t_1} dt_2 [\tilde{V}(t_1), \tilde{V}(t_2)] \quad [10]$$

and higher-order terms are available (*22, 23*). By nulling successive terms $V^{(n)}$ we insure that $U_V(\tau)$ approaches the identity operator, so the propagator $U(\tau)$ approaches the ideal propagator $U_{\mathrm{rf}}(\tau)$. Since it is $\tau\bar{V}$ that enters into the calculation of $U_V(\tau)$, and hence the departure of $U(\tau)$ from $U_{\mathrm{rf}}(\tau)$, we shall always consider the quantities $\tau V^{(n)}$.

In the case of a single rf pulse, we find that $V^{(0)} = 0$ only if the flip angle α on resonance is a multiple of 2π. Geometrically, we can understand this result by realizing that the spins will least be able to detect a finite $\Delta\omega/\omega_1$ when they undergo a complete revolution about the effective field. In such a case, the net rotation axis $\mathbf{n}$ is of no importance, and only the dependence of α on $\Delta\omega$ need be considered. $V^{(1)}$ is always nonvanishing for a single rf pulse:

$$\tau V^{(1)} = \frac{\Delta\omega^2 I_x}{2\omega_1^2}\{\alpha - \sin\alpha\} \qquad [11]$$

and $\tau V^{(1)}$ is a rapidly increasing function of α. A 2π pulse is more nearly cyclic with respect to $\Delta\omega$ than a multiple of 2π. Roughly speaking, the term $V^{(0)}$ measures the deviation of the rotation axis $\mathbf{n}$, while $V^{(1)}$ monitors the increased rotation angle, as a function of $\Delta\omega$.

The next class we consider is all composite pulses of the form $\bar{\alpha}_1\alpha_2\bar{\alpha}_1$, where the overbars denote a phase shift of π and the flip angles α_i are understood to be the nominal rotation angles (when $\Delta\omega = 0$). The zeroth-order term in the Magnus expansion is

$$\tau V^{(0)} = \frac{\Delta\omega}{\omega_1}\{\bar{a}I_x + \bar{b}I_y + \bar{c}I_z\} \qquad [12]$$

where

$$\bar{a} \equiv 0 \qquad [13]$$

$$\bar{b} = 1 - 2\cos\alpha_1 + 2\cos(\alpha_1 - \alpha_2) - \cos(2\alpha_1 - \alpha_2) \qquad [14]$$

$$\bar{c} = 2\sin\alpha_1 - 2\sin(\alpha_1 - \alpha_2) + \sin(2\alpha_1 - \alpha_2). \qquad [15]$$

In the notation of Ref. (*9*), a zeroth-order composite pulse is obtained if positive angles α_1 and α_2 can be found so that both $\bar{b}$ and $\bar{c}$ are zero. In fact, Eqs. [14] and [15] are the real and imaginary parts of a single complex equation, and the relationship

$$\bar{b} = \bar{c}\tan[(2\alpha_1 - \alpha_2)/2] \qquad [16]$$

holds. There is a continuum of solutions as a function of the net flip angle on resonance, $\alpha = \alpha_2 - 2\alpha_1$. Given α, the solution is

$$\alpha_1 = \arg[1 - e^{-i\alpha} \pm [1 + 14e^{-i\alpha} + e^{-2i\alpha}]^{1/2}] \qquad [17]$$

$$\alpha_2 = 2\alpha_1 + \alpha \qquad [18]$$

where we recall $\arg\{re^{i\theta}\} \equiv \theta$ for real positive r. Equations [17] and [18] constitute an explicit prescription to construct composite pulses of *any* net flip angle α so that a constant rotation is obtained as a function of $\Delta\omega$, to zeroth-order in the Magnus expansion. In degrees we have, for example,

$$\alpha = 180: \quad U = \overline{60}\ 300\ \overline{60} \qquad [19]$$

$$\alpha = 135: \quad U = \overline{85}\ 305\ \overline{85} \qquad [20]$$

$$\alpha = 90: \quad U = \overline{114.3}\ 318.6\ \overline{114.3} \qquad [21]$$

$$\alpha = 45: \quad U = \overline{146.5}\ 338\ \overline{146.5}. \qquad [22]$$

To zeroth-order in the Magnus expansion, these composite pulses yield constant rotations of the form $U = \exp(i\alpha I_x)$. This is the first example we know of in which a continuum of exact solutions, as a function of the nominal flip angle of the composite pulse, has been found.

The calculation for $V^{(1)}$ can be carried through similarly, and we find

$$\tau V^{(1)} = \frac{\Delta\omega^2}{2\omega_1^2}\{\bar{a}_1 I_x + \bar{b}_1 I_y + \bar{c}_1 I_z\} \quad [23]$$

where $\bar{b}_1 = \bar{c}_1 = 0$ and

$$\bar{a}_1 = 2\alpha_1 - \alpha_2 - \sin(2\alpha_1 - \alpha_2) + 4\{\sin(\alpha_1 - \alpha_2) - [\sin\alpha_1 - \sin\alpha_2]\}. \quad [24]$$

Since α_1 and α_2 are already fixed by the requirement that $V^{(0)} = 0$, the value of $V^{(1)}$ is fixed. For the sequences of Eqs. [19]–[22] we find

$$\alpha = 180: \quad \tau V^{(1)} = -3.303 I_x \Delta\omega^2/\omega_1^2 \quad [25]$$

$$\alpha = 135: \quad \tau V^{(1)} = -3.169 I_x \Delta\omega^2/\omega_1^2 \quad [26]$$

$$\alpha = 90: \quad \tau V^{(1)} = -2.608 I_x \Delta\omega^2/\omega_1^2 \quad [27]$$

$$\alpha = 45: \quad \tau V^{(1)} = -1.493 I_x \Delta\omega^2/\omega_1^2 \quad [28]$$

while the ($\alpha = 0$) sequence $\overline{180}\ 360\ \overline{180}$ has $V^{(0)} = V^{(1)} = 0$. We note that since $V^{(1)}$ commutes with U_{rf} the first sign of poor performance should be a deviation in α from the prescribed value, rather than any deviation in **n**. Since the value of $\tau V^{(1)}$ is quite large for these sequences, we expect only a modest operating bandwidth. It is a general property of all phase alternating sequences, whether symmetric or not, that the even-order terms in the Magnus expansion will be a linear combination of I_y and I_z whereas the odd-order terms will be proportional to I_x.

For 5-pulse sequences of the form $\bar{\alpha}_1\alpha_2\bar{\alpha}_3\alpha_2\bar{\alpha}_1$ the corresponding equations for $V^{(0)}$ and $V^{(1)}$ reduce to

$$\bar{b} = 1 - 2\cos\alpha_1 + 2\cos(\alpha_1 - \alpha_2) - 2\cos(\alpha_1 - \alpha_2 + \alpha_3) + 2\cos(\alpha_1 - 2\alpha_2 + \alpha_3) - \cos(2\alpha_1 - 2\alpha_2 + \alpha_3) \quad [29]$$

$$\bar{c} = 2\sin\alpha_1 - 2\sin(\alpha_1 - \alpha_2) + 2\sin(\alpha_1 - \alpha_2 + \alpha_3) - 2\sin(\alpha_1 - 2\alpha_2 + \alpha_3) + \sin(2\alpha_1 - 2\alpha_2 + \alpha_3) \quad [30]$$

and

$$\bar{a}_1 = 2\alpha_1 - 2\alpha_2 + \alpha_3 - \sin(2\alpha_1 - 2\alpha_2 + \alpha_3) + 4\{\sin(\alpha_1 - \alpha_2) - \sin\alpha_1 + 2\sin\alpha_2 - \sin\alpha_3 - 2\sin(\alpha_2 - \alpha_3) - \sin(\alpha_1 - \alpha_2 + \alpha_3) + \sin(2\alpha_2 - \alpha_3) + \sin(\alpha_1 - 2\alpha_2 + \alpha_3)\}. \quad [31]$$

Due to the presence of both α_k and $\sin\alpha_k$, we have found no general solution to Eqs. [29]–[31]. However, it is once again true that $\bar{b}$ and $\bar{c}$ are related, and only the former need be considered when looking for numerical solutions. Furthermore, it is unclear whether any solutions exist. For an overall rotation $\alpha = \pi$ a solution exists

$$\alpha = 180: \quad U = 343\ \overline{280}\ 54\ \overline{280}\ 343 \quad [32]$$

but for other flip angles the requirements $\bar{b} = 0$ and $\bar{a}_1 = 0$ appear incompatible. Nevertheless, sequences with $\bar{b} \sim 0$ and small $\bar{a}_1$ can be found.

Rather than follow through a case by case analysis of 7-, 9-, and longer $(2m + 1)$-pulse sequences, we summarize the relevant equations for sequences of arbitrary length. For an arbitrary sequence of the form $\bar{\alpha}_1\alpha_2 \cdots \alpha_{2m}\bar{\alpha}_{(2m+1)}$ where we understand $\alpha_k = \alpha_{(2m+2-k)}$ and defining Δ_n by

$$\Delta_n = \sum_{k=0}^{n} (-1)^{k+1}\alpha_k \qquad [33]$$

with $\alpha_0 \equiv 0$ we find

$$\bar{b} = 1 - \cos\Delta_{(2m+1)} + 2\sum_{j=1}^{2m}(-1)^j \cos\Delta_j \qquad [34]$$

$$\bar{c} = \sin\Delta_{(2m+1)} - 2\sum_{j=1}^{2m}(-1)^j \sin\Delta_j \qquad [35]$$

$$\bar{a}_1 = \sum_{j=1}^{2m+1} (-1)^{j+1}V_j + \sum_{k=2}^{2m+1}\sum_{j=1}^{k-1}(-1)^{k+j}V_{kj} \qquad [36]$$

where V_j and V_{kj} are defined by

$$V_j = \alpha_j - \sin\alpha_j \qquad [37]$$

$$V_{kj} = \sin(\Delta_{k-1} - \Delta_{j-1}) - \sin(\Delta_{k-1} - \Delta_{j-1} + (-1)^{k+1}\alpha_k) - \sin(\Delta_{k-1} - \Delta_{j-1} - (-1)^{j+1}\alpha_j) + \sin(\Delta_{k-1} - \Delta_{j-1} + (-1)^{k+1}\alpha_k - (-1)^{j+1}\alpha_j). \qquad [38]$$

The symmetry properties

$$V_j = V_{2m+2-j} \qquad [39]$$

$$V_{kj} = V_{(2m+2-j)(2m+2-k)} \qquad [40]$$

greatly reduce the computational work involved in evaluating Eq. [36].

Any phase-alternating composite pulse $\bar{\alpha}_1\alpha_2\bar{\alpha}_3 \cdots$ satisfying Eqs. [34]–[36] produces a constant rotation as a funtion of $\Delta\omega$ to first order in the Magnus expansion; Eqs. [39] and [40] are only necessarily true for symmetric sequences, however.

SYMMETRY PROPERTIES

It is perfectly reasonable to derive closed expressions for $V^{(2)}$ and even $V^{(3)}$ for the specific composite pulses under consideration. While the solution of the resulting equations would guarantee very uniform performance around $\Delta\omega = 0$, it does not necessarily guarantee a large bandwidth. As the offset increases, $\Delta\omega/\omega_1$ approaches unity, and ever more terms in the Magnus expansion become important. When $\Delta\omega/\omega_1$ exceeds unity, the whole perturbation approach breaks down. In addition, the numerical calculation of the terms $V^{(n)}$ quickly becomes more costly than the exact calculation of $U(\tau)$ itself. This is to be recognized as a fundamental limitation of the perturbation approach. To achieve large bandwidths, it is $U(\tau)$ we must consider.

The symmetry of the composite pulse can best be examined by considering the symmetry of the underlying piecewise-constant phase function $\psi(t)$ defined by

$$H_{rf}(t) = \omega_1 \exp(-i\psi(t)I_z)I_x \exp(i\psi(t)I_z). \quad [41]$$

The symmetry of ψ imparts important symmetry properties to U so that, in two important ways, symmetric phase alternating composite pulses behave exactly like a single rf pulse. Firstly, since the phase shift scheme is symmetric in time, $\psi(t) = \psi(\tau - t)$, the performance is independent of the sign of $\Delta\omega$ (24). Second, since $0 = -0$ and $\pi = -\pi$ for rf phase shifts, the scheme is also *antisymmetric* in time, $\psi(t) = -\psi(\tau - t)$. This property constrains U, at all resonance offsets, to be a rotation about some axis in the xz plane (*25*). A single rf pulse automatically possesses these properties since $\psi(t)$ is identically zero.

The propagator U can be written in the form $U = P^t P$, where P denotes the transformation resulting from the first half of the composite pulse, and P^t that from the last half, in which the rf pulses are simply reversed in time. If $U(\tau, +)$ is the propagator at some positive offset $\Delta\omega$ and $U(\tau, -)$ that for the corresponding negative offset, then the symmetry property means that

$$U(\tau, +) = \exp(i\pi I_z)U(\tau, -)^{-1}\exp(-i\pi I_z) \quad [42]$$

$$P^t(+) = \exp(i\pi I_z)P(-)^{-1}\exp(-i\pi I_z) \quad [43]$$

whereas the antisymmetry property yields

$$U(\tau, +) = \exp(i\pi I_y)U(\tau, +)^{-1}\exp(-i\pi I_y) \quad [44]$$

$$P^t(+) = \exp(i\pi I_y)P(+)^{-1}\exp(-i\pi I_y). \quad [45]$$

Aside from the obvious saving in computation by halving both the offset range and the number of pulses, these properties show that, under some circumstances, P itself may be of special interest.

Suppose that, over a given offset range, U approximates a perfect inversion pulse, $U = \exp(i\pi I_x)$. Then multiplying both sides of Eq. [45] on the right by P we have

$$\exp(i\pi I_x) = \exp(i\pi I_y)P^{-1}\exp(-i\pi I_y)P \quad [46]$$

which can be rearranged to give

$$P\exp(-i\pi I_z)P^{-1} = \exp(-i\pi I_y). \quad [47]$$

It follows that

$$PI_zP^{-1} = I_y \quad [48]$$

that is, P converts longitudinal magnetization into transverse magnetization with *no phase gradient as a function of resonance offset.* Since P is not necessarily a constant rotation over the same range, yet P maps a particular point of the state space onto another particular point, we call P a point-to-point transformation. Such composite pulses are ideal substitutes for the initial 90° pulse in a wide range of experiments, and should simplify the required phase correction in, for example, phase-sensitive two-dimensional NMR experiments.

COMPUTATIONAL

Composite pulses offering bandwidths up to about $\Delta\omega/\omega_1 = \pm 0.5$ result from approximate solutions of Eqs. [34]–[36]. There are many such composite pulses, varying in both duration and complexity. Shorter and less complicated sequences result in smaller bandwidths, but may prove useful in situations where the overall duration of the composite pulse is a consideration. To explore bandwidths up to $\Delta\omega/\omega_1 = \pm 1.0$, or beyond, it is necessary to optimize U directly. This can be accomplished by accepting an approximate solution in the perturbation series as an initial iterate for a numerical nonlinear optimization. The optimization is then performed by minimizing the norm $\| U_{rf} - U \|$, measuring the deviation of U from the ideal propagator U_{rf}. Details of the optimization procedure have been described elsewhere (*5*).

We have found that, for any given sequence consisting of up to 15 pulses, there seems to be a "natural" bandwidth over which the composite pulse performs extremely well, but that any attempt to increase this bandwidth results in unacceptable deterioration in the performance over the intermediate offset range. In Table 1 we summarize representative results for flip angles of 180, 135, and 90°. The stated bandwidths for each composite pulse reflect the offset range over which the composite pulse delivers the specified rotation $\exp(i\alpha I_x)$ with acceptable accuracy. In this case, the z component, n_z, of the rotation axis is kept below a few percent, and the flip angle α never deviates by more than a few degrees over the indicated bandwidths. In more concrete terms, these composite pulses perform, within the stated limits, about as well as the equivalent single pulse performs over the range $\Delta\omega/\omega_1 = \pm 0.08$. The corresponding point-to-point 90° pulses can be obtained from the first half of the composite 180° pulses listed in the table, for example, $\overline{59}$ 149, 58 $\overline{140}$ 172, etc.

We emphasize that these predictions suppose that no other pulse imperfections are present: in particular, the flip angles of the constituent pulses in each composite pulse

TABLE 1

Composite Pulses For Broadband Constant Rotations

Angle	Bandwidth[a]	Length[b]	Sequence
90	±0.20	542	$\overline{113}$ 316 $\overline{113}$
90	±0.35	698	24 $\overline{152}$ 346 $\overline{152}$ 24
90	±0.60	1218	$\overline{16}$ 300 $\overline{266}$ 54 $\overline{266}$ 300 $\overline{16}$
90	±0.80	1410	119 $\overline{183}$ 211 $\overline{384}$ 211 $\overline{183}$ 119
90	±1.0	2538	160 $\overline{324}$ 141 $\overline{204}$ 320 $\overline{84}$ 72 $\overline{84}$ 320 $\overline{204}$ 141 $\overline{324}$ 160
135	±0.15	471	$\overline{84}$ 303 $\overline{84}$
135	±0.35	713	39 $\overline{144}$ 347 $\overline{144}$ 39
135	±0.60	1251	$\overline{13}$ 320 $\overline{266}$ 53 $\overline{266}$ 320 $\overline{13}$
135	±0.80	1411	10 $\overline{105}$ 182 $\overline{214}$ 389 $\overline{214}$ 182 $\overline{105}$ 10
135	±1.1	2399	158 $\overline{308}$ 137 $\overline{178}$ 304 $\overline{80}$ 69 $\overline{80}$ 304 $\overline{178}$ 137 $\overline{308}$ 158
180	±0.15	416	$\overline{59}$ 298 $\overline{59}$
180	±0.35	740	58 $\overline{140}$ 344 $\overline{140}$ 58
180	±0.65	1232	325 $\overline{263}$ 56 $\overline{263}$ 325
180	±0.75	1352	$\overline{66}$ 180 $\overline{227}$ 406 $\overline{227}$ 180 $\overline{66}$
180	±0.85	1420	27 $\overline{99}$ 180 $\overline{211}$ 386 $\overline{211}$ 180 $\overline{99}$ 27
180	±1.2	2320	158 $\overline{294}$ 144 $\overline{152}$ 291 $\overline{89}$ 64 $\overline{89}$ 291 $\overline{152}$ 144 $\overline{294}$ 158

[a] In terms of the dimensionless offset parameter $\Delta\omega/\omega_1$.

[b] Total rotation in degrees.

should be accurately set, and no compensation for rf inhomogeneity is offered. Near to exact resonance, these phase-alternating sequences all show the same sensitivity to rf inhomogeneity as a single pulse; at other resonance offsets they can be either more or less sensitive.

CONCLUSION

We have demonstrated that symmetric phase-alternating composite pulses can provide constant rotations of arbitrary flip angles over large bandwidths. For two-level systems, these composite pulses behave like ideal single rf pulses, and can be substituted for conventional pulses in complicated multiple-pulse sequences or two-dimensional NMR experiments whenever resonance offset effects represent the principal problem. When networks of coupled spins are involved, the composite pulses must be of short duration compared with the inverse of a representative coupling constant if the compensation is to be effective.

Our sequences are based on a perturbation treatment which is known to result in composite pulses providing constant rotations (*4, 6, 9*). Our improvement has been to extend the operational bandwidth far beyond the regime over which the first few terms of the perturbation series provide an adequate approximation to the exact solution. The success of this approach has relied on the symmetry properties of the composite pulse and the great simplifications that result by using only 180° phase shifts.

Of the class of constant amplitude composite pulses, only the symmetric phase-alternating sequences retain the symmetry and antisymmetry properties of a single rf pulse. In the more general case where the pulse amplitude is allowed to vary, the corresponding sequences must have a symmetric amplitude function. A number of such sequences, based on a Gaussian amplitude function, have appeared in the high-resolution literature (*26–28*).

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