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Resonance Offset Tailored Composite Pulses

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We describe novel composite pulse sequences which act as general rotors and thus are particularly suitable for nuclear magnetic resonance quantum computation. The resonance offset tailoring to enhance nutations approach permits perfect compensation of off-resonance errors at two selected frequencies placed symmetrically around the frequency of the radiofrequency source. © 2001 Academic Press

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1. INTRODUCTION

Composite pulses (*1, 2*) play an important role in many nuclear magnetic resonance (NMR) experiments, as they allow the effects of experimental imperfections, such as pulse length errors and off-resonance effects, to be reduced. Such pulses can also prove useful in NMR implementations of quantum information processing devices, such as simple NMR quantum computers (*3–7*), where they act to reduce systematic errors in quantum logic gates (*8*). Unfortunately, many conventional composite pulse sequences are not appropriate for quantum computers as they only perform well for certain initial states, while pulse sequences designed for quantum information processing must act as *general rotors,* that is they must perform well for *any* initial state.

Composite pulses of the this kind, which are sometimes called Class A composite pulses (*1*), are rarely needed for conventional NMR experiments, and so relatively little is known about them. One important example is a composite 90° pulse developed by Tycko (*9*), which has recently been generalized to arbitrary rotation angles (*8*). These composite pulses give excellent compensation of off-resonance effects at small offset frequencies, such as those found for ^{1}H nuclei, but are of no use for the much larger off-resonance frequencies typically found for ^{13}C.

Fortunately, when composite pulses are used for NMR quantum computation one great simplification can be made: it is only necessary that the pulse sequence perform well over a small number of discrete frequency ranges, corresponding to the resonance frequencies of the nuclei used to implement qubits; it is *not* necessary to design pulses which work well over the whole frequency range. In particular, it is quite common in NMR quantum computation to use at most two spins of each nuclear species (see, for example, (*10*)), and it is convenient to place the radiofrequency (RF) frequency in the center of the spectrum, so that the two spins have equal and opposite resonance offsets (*11*). Thus it suffices to tailor the composite pulse sequence to work well at these two frequencies; the performance at all other frequencies can be completely ignored.

Here we explain how resonance offset tailoring to enhance nutations may be used to produce composite pulse sequences which give perfect compensation of off-resonance effects. These ROTTEN pulses act as perfect general rotors at two frequencies, offset from the RF frequency by $\pm\delta$ and are well suited to NMR quantum computation; in combination with periods of free precession they provide an adequate set of gates, permitting any operation to be performed (*11*). ROTTEN pulses are simple to implement and may be derived for any desired resonance offset as long as $\delta \le \sqrt{3}\nu_1$.

2. RESULTS

We choose to implement our composite rotation using a sequence of three radiofrequency pulses. In the absence of off-resonance effects any such pulse sequence can be described by stating two angles describing each pulse, θ_j, the nutation angle of the jth pulse, and ϕ_j, the phase angle of the nutation axis in the xy-plane. In the presence of off-resonance effects it is convenient to retain this description, except that θ_j is now a nominal nutation angle, and the nutation axis is no longer in the xy-plane (although ϕ_j remains a good description of the phase angle within the plane). It is also necessary to characterize the off-resonance behavior, which is conveniently parameterized using either the off-resonance fraction $f = \delta/\nu_1$ (where δ is the off-resonance frequency and ν_1 the nutation rate, both measured in hertz) or, equivalently, the RF tilt angle (the tilt angle

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TABLE 1

The ROTTEN Pulse Sequence: A Perfectly Compensated θ_ϕ Pulse Can Be Implemented Using a Sequence of Three Pulses with Flip Angles and Phases as Given Below

Pulse	Flip angle	Phase
1	$\frac{180°}{\sqrt{1+f^2}}$	$\phi + \arccos\left(\frac{\sqrt{1+f^2}}{2}\right)$
2	$\frac{\theta}{\sqrt{1+f^2}}$	$\phi + \pi - \arccos\left(\frac{\sqrt{1+f^2}}{2}\right)$
3	$\frac{180°}{\sqrt{1+f^2}}$	$\phi + \arccos\left(\frac{\sqrt{1+f^2}}{2}\right)$

of the nutation axis away from the z-axis), given by $\tan(\Delta) = \nu_1/\delta = 1/f$. The propagator describing a single RF pulse is then

$$U_j = e^{-i\theta_j(I_x\cos\phi_j + I_y\sin\phi_j + I_z f)} \quad [1]$$

(where I_x, I_y, and I_z are the conventional one-spin product operators (*12*)), while the overall propagator describing the three pulse sequence is $U = U_3U_2U_1$.

In order to produce a perfectly compensated pulse it is necessary to find values of the six angles (θ_1, ϕ_1, θ_2, ϕ_2, θ_3, and ϕ_3) such that U implements the desired rotation for the desired value of f. A general search over these six values would be a major task, but fortunately the problem can be substantially simplified. The requirement that the composite pulse has *identical* effects on resonances at frequencies $\pm\delta$, so that $U(f) = U(-f)$, imposes major restrictions on the allowed values. Furthermore, a family of solutions exists for which the first and last pulses are identical, thus reducing the underlying search space to four independent values. Examining the form of the asymmetric response term $U(f) - U(-f)$ suggests the choices

$$\theta_1 = \pi/\sqrt{1+f^2} \quad [2]$$

and

$$\cos(\phi_1 - \phi_2) = (1 - f^2)/2. \quad [3]$$

These values can then be inserted back into the expression for U, the result equated with the desired propagator (neglecting any irrelevant overall phase term), and the equations then solved for θ_2 and ϕ_1. It is simplest to begin by solving for a θ_x pulse, that is an ideal pulse with nutation angle θ and phase angle 0; pulses with any other phase angle can then be created by simply adding a phase shift to all three pulses. This gives the results

$$\theta_2 = \theta/\sqrt{1+f^2} \quad [4]$$

and

$$\phi_1 = \pm\arccos\left(\frac{\sqrt{1+f^2}}{2}\right). \quad [5]$$

(Throughout this paper we will use the positive solution of Eq. [5]). Finally, combining Eqs. [3] and [5] gives

$$\phi_2 = \pi - \phi_1. \quad [6]$$

These results are summarised in Table 1.

Examining Eqs. [3] and [5] reveals a limitation to this approach: the phase angles only have real solutions when $|f| \le \sqrt{3}$. It is, of course, possible that other composite pulse families exist with larger ranges of applicability, but clearly there is a limit beyond which any three pulse sequence will cease to function. However, even the limited range of f values described here is far greater than anything which can be achieved with conventional (nontailored) composite pulses and is likely to be adequate for most purposes. Furthermore, the family of pulses described above exhibits fairly good tolerance of RF inhomogeneity relative to other families which we considered.

It must, however, be remembered that ROTTEN composite pulses are only effective at the resonance offsets for which they have been tailored; at other resonance offsets these pulses may perform very poorly indeed. This is shown in Fig. 1, which plots the fidelity of a ROTTEN composite pulse sequence optimised for $f = \pm\sqrt{3}$ over a range of values of f. As ROTTEN pulses are designed to act as general rotors, it is necessary to use a fidelity measure which applies over all possible starting values; we have chosen to use the rotor

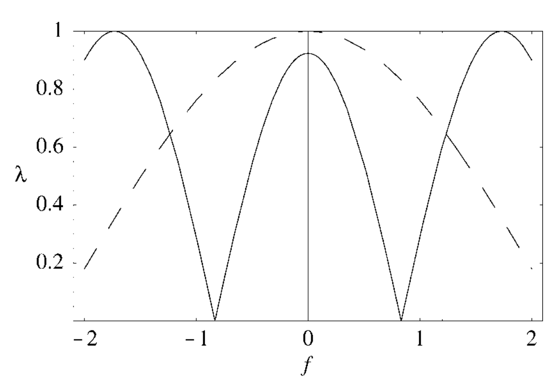

FIG. 1. The fidelity, λ, of a simple 90° pulse (dashed line) and a ROTTEN composite 90° pulse for a range of values of the off-resonance fraction, f; the ROTTEN pulse was tailored for the values $f = \pm\sqrt{3}$.

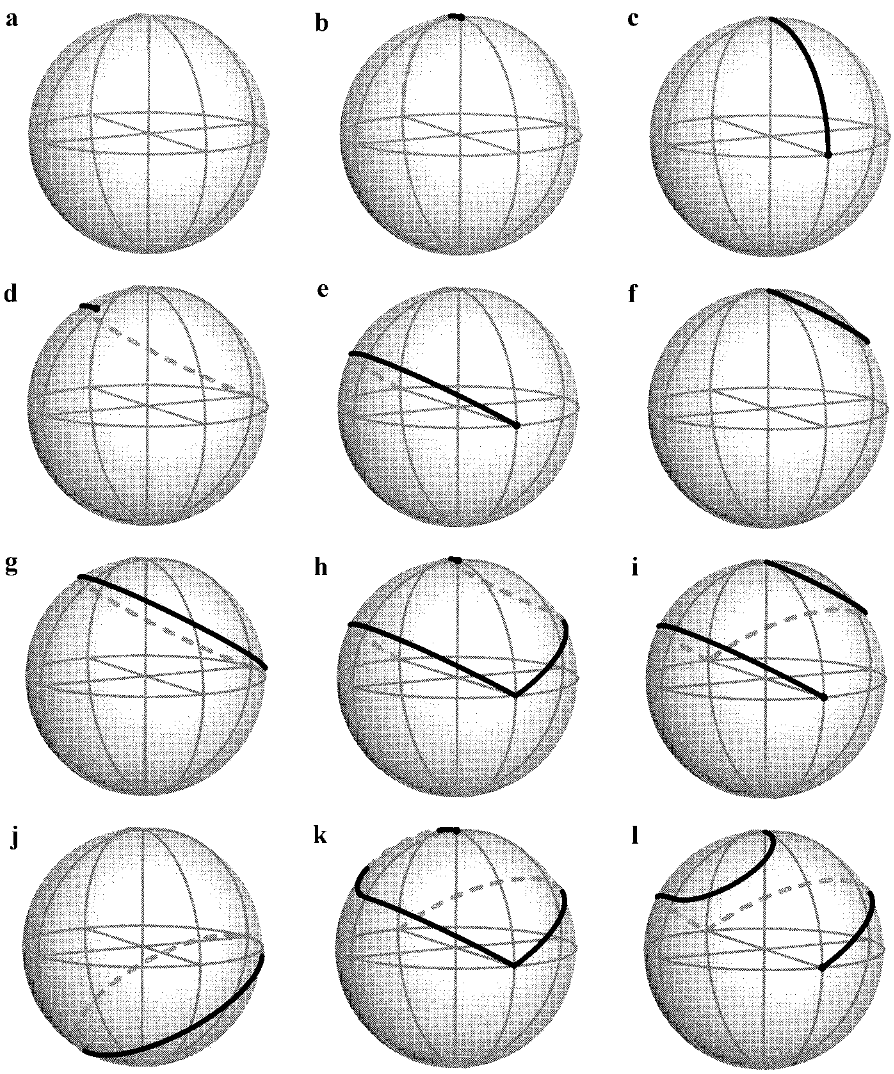

FIG. 2. Grapefruit plots showing magnetization trajectories for 90°_x pulses using simple pulses and ROTTEN composite pulses ($90^\circ_x 45^\circ_{-x} 90^\circ_x$) optimized for an off-resonance fraction $f = \sqrt{3}$. Simple pulses (a, b, c) with no off-resonance effects ($f = 0$); simple pulses (d, e, f) in the presence of large positive off-resonance effects ($f = \sqrt{3}$); ROTTEN pulses (g, h, i) in the presence of large positive off-resonance effects ($f = \sqrt{3}$); ROTTEN pulses (j, k, l) in the presence of large negative off-resonance effects ($f = -\sqrt{3}$). Initial states are I_x (a, d, g, j), I_y (b, e, h, k), and I_z (c, f, i, l). Note that for a perfect 90°_x pulse applied to I_x (a) the magnetization does not leave the *x*-axis.

fidelity, λ, defined by Levitt (*1*). The rotor fidelity of the simple pulse is low except at small values of f, while that for the composite pulse is high in two regions around $\pm\sqrt{3}$. This is ideal for some implementations of NMR quantum computation, if unlikely to prove useful in most conventional NMR experiments.

For the remainder of this paper we will consider composite pulses tailored for the case $f = \pm\sqrt{3}$; this is not only the limit of our approach (and so the case where ROTTEN pulses give the greatest improvement in comparison with conventional pulses), but also a choice which results in a particularly simple sequence. To achieve an ideal θ_ϕ rotation the values required are $\theta_1 = \theta_3 = \pi/2$, $\theta_2 = \theta/2$, $\phi_1 = \phi_3 = \phi$, and $\phi_2 = \phi + \pi$. The operation of simple and ROTTEN composite 90°_x pulses is shown by magnetization trajectories in Fig. 2 for initial states of I_x, I_y, and I_z. The magnetization trajectories are complicated, but those for ROTTEN composite pulses terminate in the correct locations, while simple 90°_x pulses give extremely poor results at such large resonance offsets.

Finally, we show the performance of these pulse sequences in an actual NMR experiment. First, Fig. 3 shows ^{13}C spectra of ^{13}C-labeled glycine acquired using simple and ROTTEN composite 90° pulses. (The glycine framework formed the basic structure of the first five-qubit NMR quantum computer (*10*), and so glycine provides an excellent model system; as described previously (*11*) quantum computations can be performed in systems such as this by combining hard pulses with delays.) In order to emphasize the performance of our com-

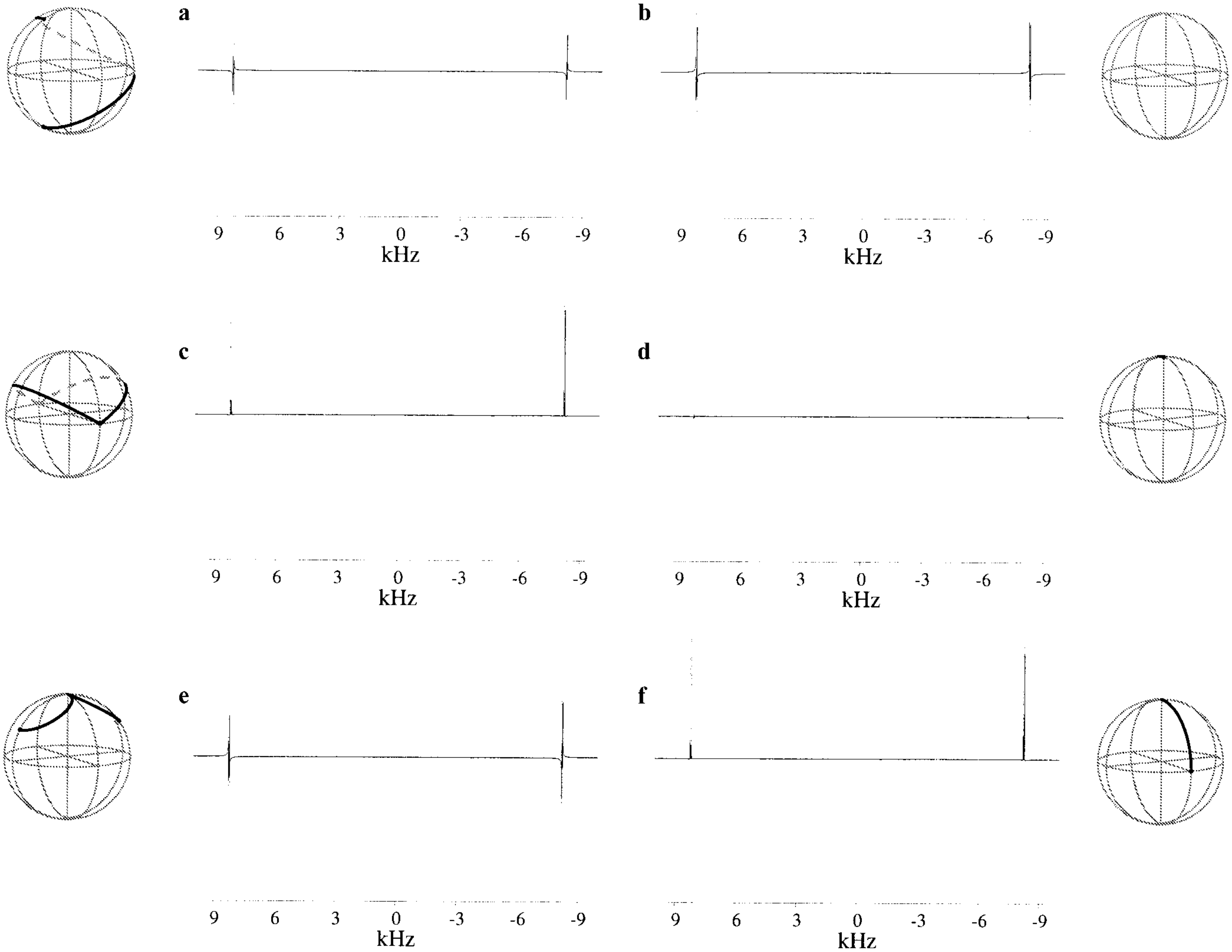

FIG. 3. Experimental ^{13}C spectra of ^{13}C-labeled glycine in a homebuilt 500-MHz (^{1}H frequency) NMR spectrometer at the Oxford Centre for Molecular Sciences: (a, c, e) using a simple 90°_x pulse; (b, d, f) using a ROTTEN composite pulse ($90^\circ_x 45^\circ_{-x} 90^\circ_x$). The frequency separation between the C_α and C' multiplets was 16480 Hz and the RF pulse power was reduced to about 44% of its maximum value (around 10.9 kHz) so that the off-resonance fraction was $f \approx \pm\sqrt{3}$. Low-power CW ^{1}H decoupling was applied to the CH_2 protons. Initial states were (a, b) I_x; (c, d) I_y; (e, f) I_z. (Initial states other than I_z were themselves prepared from I_z by using appropriate ROTTEN pulses.) All spectra were phased using the same phasing parameters so that the state $-I_y$ appears in positive absorption mode. Marginal grapefruit plots indicate the operation of conventional pulses on the left-hand side and *ideal* pulses (rather than ROTTEN pulses, for which the trajectories are much more complex) on the right-hand side.

posite pulses these spectra were acquired with the RF pulse power reduced so that the off-resonance fraction was $f \approx \pm\sqrt{3}$. Under these circumstances a simple 90° excitation pulse results in phase errors of ±90°, while a ROTTEN pulse should give perfect compensation. Similar improvements should occur for other initial states such as I_x and I_y. In practice, small phase errors are observed in the ROTTEN spectra; calculations suggest that these arise principally from pulse length errors, although *J*-coupling also makes a minor contribution.

Second, Fig. 4 shows the performance of ROTTEN pulses as excitation sequences away from their optimal off-resonance frequencies. The experimental setup was the same as that described in Fig. 3, except that the RF frequency was shifted so that the off-resonance frequency of the C_α carbon varied from $f = -3\sqrt{3}/2$ to $f = 3\sqrt{3}/2$. Only the signal from the C_α carbon is shown, and the frequency axis of each spectrum has been shifted to move all the lines toward the center of the spectrum. The actual off-resonance fraction for each spectrum is shown on the *y*-axis. As expected, the sequence performs almost perfectly at $f = \pm\sqrt{3}$ but relatively poorly at other frequencies. The secondary maximum at $f = 0$ is also clearly visible; in this case the ROTTEN pulse acts as a 45°_x excitation pulse.

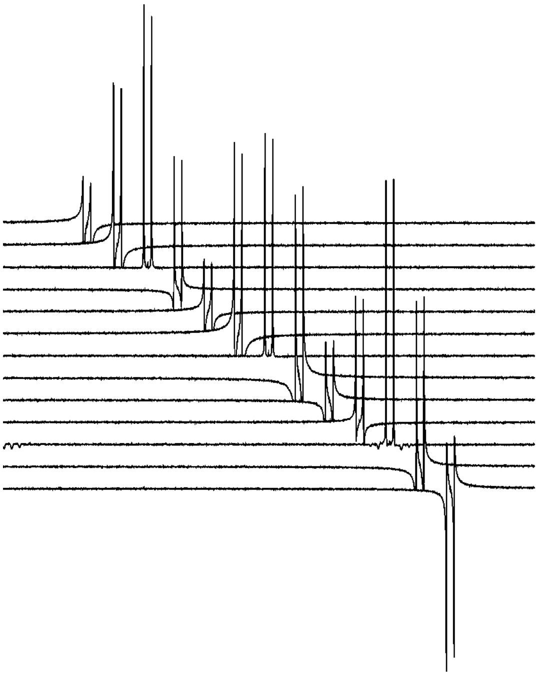

FIG. 4. Experimental spectra showing the performance of ROTTEN excitation pulses away from resonance. Experimental parameters were the same as in Fig. 3 except that the RF frequency was varied as described in the text.

3. CONCLUSIONS

Resonance offset tailoring provides a simple and effective approach for removing large off-resonance effects in systems where NMR resonances occur at two well-separated frequencies. Unlike most conventional composite pulses, ROTTEN pulses provide theoretically perfect compensation and act as perfect rotors. These properties make them well suited to NMR quantum computation; it is not yet clear whether there are any applications in more conventional NMR experiments.

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