Iterative maps for broadband excitation of transverse coherence in two level systems

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An iterative scheme has been used to derive a pulse sequence, compensated for off-resonance and rf inhomogeneity pulse errors, which implements a $\pi/2$ rotation of the spin density operator around a well-defined axis in the transverse plane. A fixed point analysis is applied to this and other iterative schemes revealing the source and nature of the compensation. Contrasting features of the different schemes are uniquely revealed by this analysis. General considerations for the construction of iterative schemes with other stable fixed sets are considered.

I. INTRODUCTION

The bandwidth response of nuclear spins to applied radio-frequency (rf) radiation can be substantially broadened,[1–15] narrowed,[11,13,15–18] or shaped[17,18] in a nuclear magnetic resonance (NMR) or other coherent spectroscopic experiment[19] by using composite pulses in place of single pulses. Composite pulses are closely spaced trains of rf pulses with the amplitudes, phases, and frequencies selectively modulated so as to produce some desired effect, normally one which cannot be practically achieved with a single pulse. Such techniques for exciting nuclear spins are frequently necessitated both by the unusual bandwidth response required in some types of NMR experiments, e.g., selective multiple-quantum NMR[20] and spatially selective, *in vivo* NMR,[21] and by the practical limitations on rf instrumentation which inhibit uniform excitation of nuclear spins with different resonance frequencies and strong couplings to one another.

Much of the work on composite pulse sequences, with few exceptions,[9,12,14,22,23] has been concentrated on the problem of inverting the populations of nuclear spin energy levels. Such a response can, of course, be accomplished with a single pulse, but over relatively small bandwidths determined and limited by the size of the applied rf field. Several workers have demonstrated that the range of transition frequencies, rf amplitudes, and spin coupling constants over which the rf radiation is effective at inverting nuclear spin populations can be made dramatically broader, narrower, or "tailored" with relatively simple composite pulse sequences.[1–8,10–13,15–18] Comparatively less attention has been paid to the related problem of creating transverse magnetization from longitudinal magnetization, an obviously important component of pulsed coherence experiments. Pulse sequences which produce this effect are commonly known as composite $\pi/2$ pulses.

One main difficulty with such pulse sequences is that there are few readily evident rules for combining pulses to form a composite $\pi/2$ pulse. Not only are such rules difficult to determine, but they must also be compatible with the objective of finding sequences with the desired compensation of pulse errors.

The work presented in this paper demonstrates how such problems can be resolved with the use of iterative schemes. First, some general rules are advanced which simplify the construction of pulse sequences approximating a desired response, such as a $\pi/2$ rotation of the magnetization. Next, it is shown how such rules can be manipulated to determine sequences which compensate for individual pulse errors. These principles are utilized to derive practical sequences which convert longitudinal magnetization to transverse magnetization over wide ranges of both transition frequencies (ω_0) and rf intensities (ω_1). The effective rotation of the longitudinal magnetization is about an axis lying in the transverse plane. These properties distinguish these iteratively generated sequences from other, previously reported sequences,[9,12–15] and make them suitable for refocusing,[24] pulsed spin locking,[25] time reversal,[26] polarization transfer,[27] and multiple-quantum experiments,[20] for which a net rotation of the magnetization of angle $\pi/2$ about an axis in the transverse plane is necessary.

II. THEORY

A. Background

Irradiation of a quantum mechanical system with some pulse sequence S_i initiates evolution of the system from some starting state, usually a state of thermal equilibrium described by a density matrix proportional to the spin angular momentum operator I_z, to some desired final state. The propagator determining this time evolution will be written $U_i(\lambda,t)$ to signify the dependence of the time evolution of the system on both the time and on parameters of the spin system or incident radiation. Since λ may vary for different spins in the sample, it is possible for $U_i(\lambda,t)$ to assume a distribution of values after some time t. One of the basic objectives of broadband composite sequence design is to derive sequences for which $U_i(\lambda,t)$ assumes a specific form at some time τ and is insensitive to the value of λ.

A variety of methods have been developed for the treatment of this problem. The approach we adopt in this paper is to use an iterative scheme[28] to generate the broadband pulse sequence. This approach has several appealing conceptual

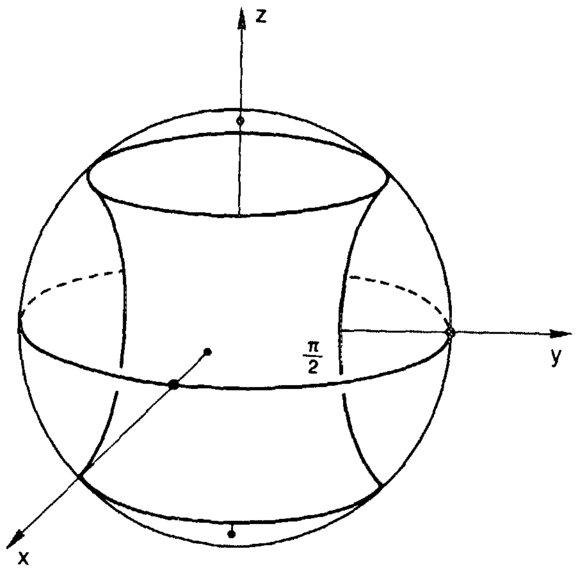

FIG. 1. Surface in SO(3) representing the set of rotations which rotate the z component of an arbitrary vector into the transverse plane. The three-dimensional rotation operators corresponding to such points are those which have the R_{zz} element identically equal to zero.

advantages, discussed elsewhere,[13] over other methods, particularly in the analysis of long sequences. An iterative scheme is a set of operations which can be applied to an arbitrary pulse sequence S_i to generate a new sequence S_{i+1}:

$$f(S_i) = S_{i+1} . \quad (1)$$

The propagator $U_i(\lambda,t)$ for S_i is accordingly transformed:

$$F[U_i(\lambda,t)] = U_{i+1}(\lambda,t) . \quad (2)$$

Propagator algorithms of this type have been analyzed by Tycko *et al.* using the formalism of iterative maps and their fixed points.[11,13,17,18] From this perspective, two conditions must be satisfied by f if it is to generate a broadband composite pulse sequence:

(a) If $\overline{U}$ is the propagator corresponding to the desired response, then $F(\overline{U}) = \overline{U}$. Such an operator is then said to be fixed or invariant.

(b) If, in addition, broadband behavior over λ is desired, $\overline{U}$ must be a stable fixed point of F with respect to variation of λ. Narrowband behavior results when $\overline{U}$ is unstable and fixed.

A more complete description of this terminology has been presented elsewhere.[13,18]

The theoretical analysis discussed below assumes an ensemble of isolated spin-1/2 nuclei. The space of operators applicable to the analysis of such an ensemble is the space of real rotations SO(3).[29] This space can be graphically portrayed as a solid sphere of radius π, as shown in Fig. 1. Elements of this space will be written $R(\boldsymbol{\alpha})$ to emphasize the fact that they represent rotations about an axis $\boldsymbol{\alpha}$, through an angle $|\boldsymbol{\alpha}|$. The locus of points of particular concern to us here are the rotations which take I_z into the xy plane, appearing within the sphere of Fig. 1 as a cylinder-like surface. This set represents the apparent $\pi/2$ rotations of I_z. True $\pi/2$ rotations of I_z into the transverse plane comprise the subset of this surface which intersects the xy plane of SO(3). These rotations are represented in SO(3) by vectors of the form

$$\boldsymbol{\alpha} = \pi/2[\cos\psi,\sin\psi,0] . \quad (3)$$

Using previously defined notation,[13,18] this set of rotations will be written $R_\psi(\pi/2)$, where ψ is an arbitrary phase shift measured from the x axis. While the rest of the surface will rotate a longitudinal vector into a transverse vector, the actual angle of the rotation will be greater than $\pi/2$ and will be about an axis not in the xy plane of SO(3).

In the next two subsections we examine first how to construct pulse sequence iteration schemes which result in maps on SO(3) with true $\pi/2$ rotations as a fixed set. After these operations have been identified, we consider next how to determine those operations which are stable at the desired fixed set.

B. Specification of invariance

The iterative schemes developed in this paper consist of two operations, namely, adding a constant phase shift to all the pulses of a sequence, followed by concatenation of phase shifted versions of the sequence. Adhering to past convention, a sequence S_i with all of its pulses phase shifted by some constant amount ϕ will be denoted $S_i(\phi)$.

Concatenation of N phase shifted versions of S_i constitutes the iterative scheme:

$$S_{i+1} = S_i(\phi_1)S_i(\phi_2)\cdots S_i(\phi_{N-1})S_i(\phi_N) \quad (4)$$

with the corresponding transformation of rotation operators:

$$R(\boldsymbol{\alpha}_{i+1}) = R(\boldsymbol{\alpha}_{i,N})R(\boldsymbol{\alpha}_{i,N-1})\cdots R(\boldsymbol{\alpha}_{i,2})R(\boldsymbol{\alpha}_{i,1}) , \quad (5)$$

where $R(\boldsymbol{\alpha}_{i,j}) = R_z(\phi_j)R(\boldsymbol{\alpha}_i)R_z^{-1}(\phi_j)$. The operator $R_z(\phi_j)$ denotes a positive rotation around the z axis by the angle ϕ_j. For high field NMR Hamiltonians, Eq. (5) generally holds true. This combination of operations will be summarized by the notation $[\phi_1,\phi_2,...,\phi_{N-1},\phi_N]$.

The problem we address in this section is the determination of possible iterative transformations of pulse sequences which result in maps on SO(3) with $R_\psi(\pi/2)$ as a fixed set. These are iterative operations which, when performed on a composite $\pi/2$ pulse S_i, guarantee that S_{i+1} will also be a composite $\pi/2$ pulse.

Several operations are conceivable. One possibility is to form a cycle, or several cycles, with S_i, concatenate the cycles, and then insert S_i at the end, beginning, or between cycles. We use the strict definition of cycle here to denote sequences which have the unit operator as a net propagator. If applied to a sequence producing a $R_\psi(\pi/2)$ rotation, the following phase shift-concatenation operations constitute cyclic sequences:

(i) $[\phi,\phi+\pi]$,

(ii) $[\phi,\phi,\phi,\phi]$,

(iii) $[\phi,\phi+\pi/2,\phi+3\pi/2,\phi+\pi]$.

The last sequence is a cycle of the WAHUHA type.[30] Insertion of a cycle anywhere within a composite $\pi/2$ pulse will leave the sequence a composite $\pi/2$ pulse.

A second operation is to phase shift S_i and concatenate it with itself, forming $S_i(\phi_j)S_i(\phi_j)$. If S_i is a true $\pi/2$ pulse sequence, with a corresponding rotation operator which can be written $R_\psi(\pi/2)$, $S_i(\phi_j)S_i(\phi_j)$ will be an inverting, or π sequence, with net rotation operator $R_{\psi+\phi_j}(\pi)$. Concatenation of N phase shifted versions of $S_i(\phi_j)S_i(\phi_j)$, N odd, produces a nominal inverting sequence with a net π rotation operator $R_\gamma(\pi)$, where γ can be computed from the relation[13]

$$\gamma = \psi + \phi_1 - \phi_2 + \cdots - \phi_{N-1} + \phi_N . \quad (6)$$

Inserting $S_i(\gamma - \psi)$ or $S_i(\gamma - \psi + \pi)$ at the beginning or end of this sequence will then result in a true $\pi/2$ pulse sequence.

The final operation we shall consider are rotations of the operator $R_\psi(\pi/2)$ around the z axis. This operation can be understood as follows. If S_i is a sequence with net rotation operator $R_\psi(\pi/2)$, then the sequence formed by the phase shift concatenation scheme $[\phi,\phi+\pi/2,\phi+\pi]$ has as its net rotation operator $R_z(\pi/2)$. A variation of this scheme is given by $[\phi,\phi+\pi/2,\phi+\pi/2,\phi+\pi]$ which results in the net rotation $R_z(\pi)$. Sandwiching S_i between two such z rotation sequences, e.g., the schemes $[\phi,\phi+\pi/2,\phi+\pi]$ and $[\theta,\theta+3\pi/2,\theta+\pi]$, results in the overall scheme

$$[\phi,\phi+\pi/2,\phi+\pi,0,\theta,\theta+3\pi/2,\theta+\pi] ,$$

with a net rotation operator which can be written $R_{\psi+\pi/2}(\pi/2)$.

The abovementioned schemes clearly do not exhaust the potential operations which lead to the desired invariance of $R_\psi(\pi/2)$. Nor are such schemes confined solely to the case of composite $\pi/2$ pulses; by straightforward extensions, they can be applied to conceive operations with other rotations or propagators as invariants as well. In addition, the various schemes can be combined with one another to produce more exotic sequences with the desired invariant propagators. The criteria governing the choice, ordering, phase, and number of these schemes is the topic of the next section.

C. Specification of stability

Stability is a general concept which refers to the behavior of points transformed by a map in the vicinity of the fixed points of the map. Denoting a fixed point of some mapping F in SO(3) as $R(\overline{\alpha})$, points in the neighborhood of $R(\overline{\alpha})$ can be written

$$R(\beta_i) = R(\overline{\alpha})R(\delta_i) , \quad (7)$$

where $|\delta_i|$ is small. This separation of an operator into a product of two operators, one an "ideal" operator, and the other arising from some perturbation, is formally identical to the separation of operators performed when transforming into an interaction representation in time-dependent quantum mechanical problems.

Applying the mapping F with fixed point $R(\overline{\alpha})$ to some point $R(\beta_i)$ results in a new point $R(\beta_{i+1})$ which can be expressed as

$$R(\beta_{i+1}) = R(\overline{\alpha})R(\delta_{i+1}) . \quad (8)$$

For a generalized phase shift-concatenation scheme $[\phi_1,\phi_2,...,\phi_{N-1},\phi_N]$, δ_{i+1}, in the linear approximation, is transformed to

TABLE I. Phase iteration schemes which generate broadband rotations of the form $R_\psi(\pi/2)$, as determined by the numerical procedure described in Sec. II. The phases are given in degrees. Symmetric schemes have been marked with an asterisk.

[0, 0,180,135,135,150,330,0]
[0, 0,180,135,135,160,340,0]
[0,280,100,135,135,100,280,0]*
[0,280,100,135,135,110,290,0]
[0,280,100,135,135,120,300,0]
[0,280,100,135,135,130,310,0]
[0,280,100,135,135,140,320,0]
[0,290,110,135,135, 90,270,0]
[0,290,110,135,135,100,280,0]
[0,290,110,135,135,110,290,0]*
[0,290,110,135,135,120,300,0]
[0,290,110,135,135,150,330,0]
[0,300,120,135,135, 90,270,0]
[0,300,120,135,135,100,280,0]
[0,300,120,135,135,110,290,0]
[0,300,120,135,135,160,340,0]
[0,310,130,135,135,100,280,0]
[0,310,130,135,135,170,350,0]
[0,320,140,135,135,100,280,0]
[0,320,140,135,135,170,350,0]
[0,330,150,135,135,110,290,0]
[0,330,150,135,135,160,340,0]
[0,330,150,135,135,170,350,0]
[0,340,160,135,135,120,300,0]
[0,340,160,135,135,150,330,0]
[0,340,160,135,135,160,340,0]*
[0,340,160,135,135,170,350,0]
[0,350,170,135,135,130,310,0]
[0,350,170,135,135,140,320,0]
[0,350,170,135,135,150,330,0]
[0,350,170,135,135,160,340,0]
[0,350,170,135,135,170,350,0]*

$$\begin{aligned}\delta_{i+1} &\approx \delta_i(\phi_1) + R^{-1}(\overline{\alpha}_1)\delta_i(\phi_2) \\ &\quad + R^{-1}(\overline{\alpha}_1)R^{-1}(\overline{\alpha}_2)\delta_i(\phi_3) \\ &\quad + R^{-1}(\overline{\alpha}_1)R^{-1}(\overline{\alpha}_2)R^{-1}(\overline{\alpha}_3)\delta_i(\phi_4) + \cdots \\ &= T_{\overline{\alpha}}\delta_i . \end{aligned} \quad (9)$$

The notation $\delta_i(\phi_j) = R_z(\phi_j)\delta_i$ and $R(\overline{\alpha}_j) = R_z(\phi_j)R(\overline{\alpha})R_z^{-1}(\phi_j)$ has been used here. The last equality in Eq. (9) reveals the fact that the expression above it is a well-defined linear transformation in three dimensions of δ_i.

The fixed point $R(\overline{\alpha})$ will be stable in all directions if the three complex eigenvalues λ_1, λ_2, and λ_3 of $T_{\overline{\alpha}}$ satisfy the inequality

$$|\lambda_j| < 1 , \quad (10)$$

and will be superstable in all directions if $|\lambda_j| = 0$ for all j. These conditions ensure that $|\delta_{i+1}| < |\delta_i|$ for $|\delta_i|$ small, and hence imply that $R(\beta_i)$ converges to $R(\overline{\alpha})$.

The set for which invariance and stability are sought consists of all rotations which can be written $R_\psi(\pi/2)$. Employing the principles outlined in the previous section, the iterative scheme [0,135,135,0], which has a map on SO(3) with $R_\psi(\pi/2)$ as an invariant set, was modified by inserting two cycles within the phase shift scheme to form the new eight shift scheme

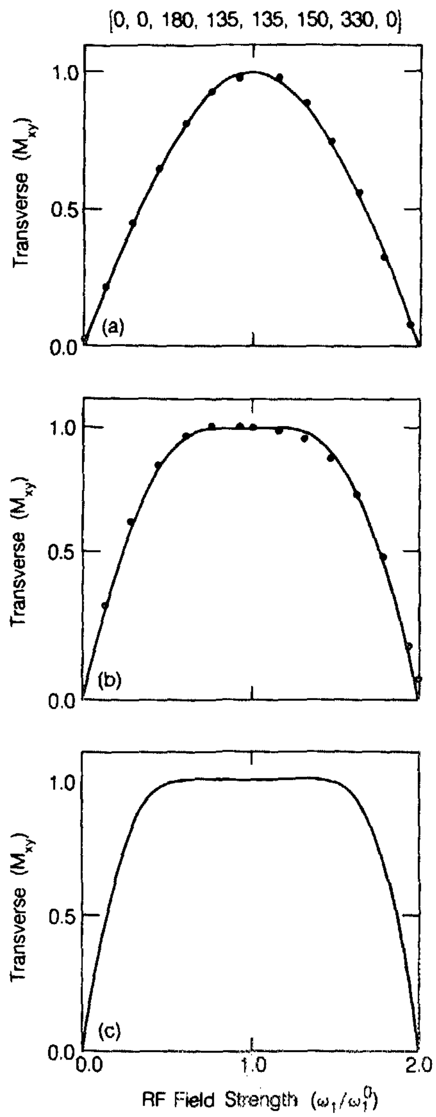

FIG. 2. Transverse magnetization created from the initial state I_z as a function of normalized rf field strength. Theoretical values appear as lines, experimental points as dots. Shown are the results for a single nominal $\pi/2$ pulse, a nominal $\pi/2$ pulse iterated once according to the scheme [0,0,180,135,135,150,330,0] (8 pulses), and the single pulse iterated twice (64 pulses).

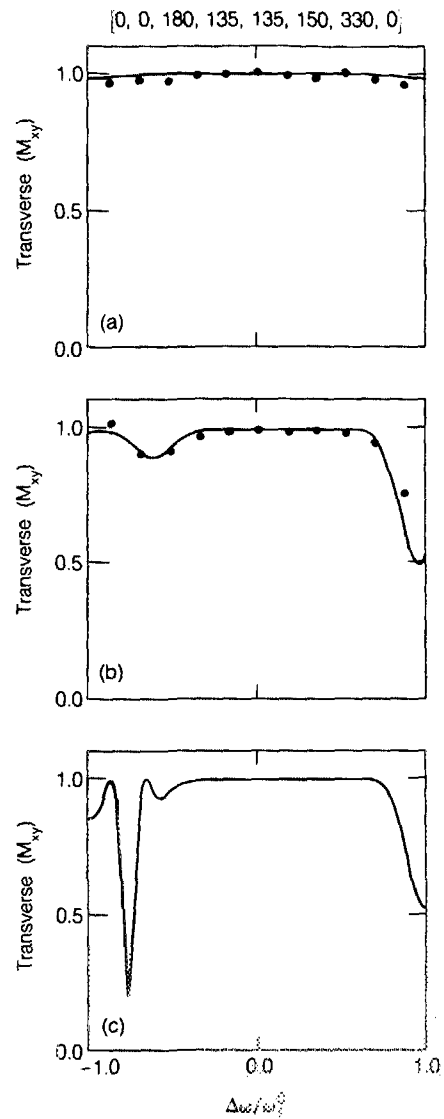

FIG. 3. Transverse magnetization created from the initial state I_z as a function of normalized resonance offset. The progression of pulse sequences for the three boxes follows that of Fig. 2. Experimental points appear as dots, and theoretical predictions as lines.

$$[0,\phi_1,\phi_1+180,135,135,\phi_2,\phi_2+180,0]\ .$$

The phases are indicated in degrees. The scheme [0,135,135,0] was chosen as a starting point since the SO(3) map of this scheme is already stable at $R_\psi(\pi/2)$ for displacements in the xy plane of SO(3). Insertion of the two additional cycles was necessary to obtain stability in all directions at $R_\psi(\pi/2)$. A general analytical expression for the eigenvalues of the linear operator $T_{\bar{\alpha}}$ can be derived for schemes of this form using Eq. (9), with ϕ_1 and ϕ_2 as independent variables. The variables ϕ_1 and ϕ_2 were then searched on a computer for values which satisfied the eigenvalue inequalities (10). Iterative schemes identified in this way appear in Table I. Based on the magnitude of the eigenvalues, and the number of phases in the scheme coincident with the four quadrature phases, the sequence [0,0,180,135,135,150,330,0] was selected from this table for closer examination.

III. RESULTS AND DISCUSSION

A. Bandwidth properties

The simplest composite pulse sequence which can be generated from the scheme [0,0,180,135,135,150,330,0] is an eight pulse sequence, each a nominal $\pi/2$ pulse ordered consecutively with the eight prescribed phases. The efficacy of this eight pulse sequence at creating transverse magnetization from the initial state I_z as a function of rf field strength is presented in Fig. 2 for zero, one, and two iterations of the scheme. The y axis in this plot represents the projection of the density operator onto the transverse plane, and is defined by the relation

$$\langle M_{xy}\rangle = [(\mathrm{Tr}\{I_x U(t) I_z U^\dagger(t)\})^2 + (\mathrm{Tr}\{I_y U(t) I_z U^\dagger(t)\})^2]^{1/2}/\mathrm{Tr}\{I_z^2\}\ . \quad (11)$$

The increase in the effective bandwidth of the sequence is plain.

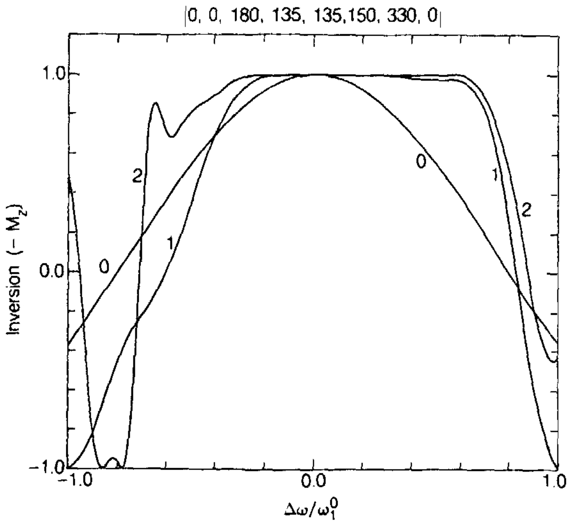

FIG. 4. Simulations of the normalized population inversion ($-M_z$) as a function of resonance offset for a single nominal π pulse (0), the 16 pulse sequence generated by concatenating $[90_0,90_0,90_{180},90_{135},90_{135},90_{150},90_{330},90_0]$ with itself (1), and the 128 pulse sequence formed by concatenating the second iterate of the 8 shift scheme with itself (2). The greater bandwidth range of the concatenated composite $\pi/2$ pulse sequences is a sensitive demonstration of the quality of the rotations which they produce.

A similar plot is shown in Fig. 3 for the excitation of transverse coherence, this time as a function of resonance offset. A single pulse is more effective at creating transverse magnetization from the initial condition I_z than a composite pulse. This phenomenon is well understood as arising from the fact that the net rotation produced by a single, off-resonance pulse $\pi/2$ pulse, while neither about an axis in the xy plane nor about an angle $\pi/2$, nevertheless does rotate I_z into the transverse plane quite effectively.[12,31]

This point can be made clearer by comparing the off-resonance population inversion produced by a pulse sequence consisting of two concatenated $\pi/2$ pulses vs that of two concatenated composite $\pi/2$ pulse sequences. Theoretical simulations of the population inversion produced by these concatenated sequences as a function of resonance offset are shown in Fig. 4. It is apparent from these simulations that the concatenated composite pulses produce a better π rotation than do the concatenated single pulses, even though the composite pulse sequence appears less effective at converting longitudinal polarization to transverse polarization than the single pulse. Moreover, the phase of the signal from a single $\pi/2$ pulse varies linearly as a function of the rf offset; this phase distortion is diminished in the composite pulse sequence over a small range of offsets. Following Levitt's classification scheme,[15] the eight pulse sequence can be considered an A type of sequence within this range. Outside this range, the sequence becomes of the B 1 type.

All experiments reported in this section were performed on an $H_2O(1)$ sample sealed in a 1.5 mm capillary tube at a proton Larmor frequency of 360 MHz. The rf strength for these experiments varied from 7 to 11 kHz. The radio frequencies for the probe and the receiver mixing were generated in separate, but locked, sources, permitting off-resonance excitation with on-resonance detection.

B. Previous work

Levitt and Ernst have proposed designing composite pulse sequences for converting longitudinal magnetization to transverse magnetization by use of an iterative procedure.[22] One version of their procedure, which they called a recursive expansion, consists of concatenating a starting sequence S_i with its inverse sequence S_i^{-1} phase shifted by $\pi/2$ rad. These two operations are summarized by the notation $S_{i+1} = S_i(S_i^{-1})_{90}$. If we denote the propagator describing the time evolution of the spin system during the sequence S_i as $U_i(\lambda,t)$, the inverse sequence S_i^{-1} can be defined as the sequence which has as its propagator the unitary operator $U_i^\dagger(\lambda),t)$.

Two features distinguish the recursive expansion approach from the phase shift-concatenation schemes developed in this paper. The first is that the fixed set of the map on SO(3) of the recursive expansion does not include rotations of the form $R_\psi(\pi/2)$. The procedure instead generates rotations which take place about unit vectors of the form $[(2/3)^{1/2}\cos\psi,(2/3)^{1/2}\sin\psi,-3^{-1/2}]$ through an angle of $2\pi/3$, leading to the effective propagator

$$\bar{U} = \exp\left\{-\frac{2\pi i}{3}\left[(2/3)^{1/2}\cos\psi I_x + (2/3)^{1/2}\sin\psi I_y - (3)^{1/2} I_z\right]\right\}. \quad (12)$$

One consequence of this fact is that concatenating two identical such sequences will not result in an inverting (π) pulse sequence.

The second difference of the recursive expansion is that it requires, as one step of the procedure, the formation of the inverse sequence S_i^{-1}. If such an inverse sequence could be constructed, the recursive expansion would produce sequences effective at virtually all resonance offsets and ω_1 values. However, when the resonance offset is not zero, there is no known method for creating the inverse sequence S_i^{-1}. To minimize this problem, methods for constructing approximate inverses have been suggested, and starting with an initial sequence which is itself compensated for resonance offset errors has been proposed. Even with these measures, it is difficult to accurately implement the recursive expansion procedure off resonance. A large phase distortion in the spectrum as a function of offset results.[12]

A second iterative procedure for generating $\pi/2$ rotations is the phase shift-concatenation scheme [0,135,135,0].[13] Iteration of this scheme transforms rotation operators of the form $R_\psi(\pi/2)$ to operators of the form $R_{\psi+\pi/2}(\pi/2)$. Sequences generated by this scheme have been shown to be compensated for rf inhomogeneity effects, and hence are broadband over ω_1. However, such sequences are not compensated for resonance offset errors, and the performance of these sequences is accordingly degraded when the rf is moved away from the spin resonance frequency.

The last class of composite sequences we shall consider are those developed using coherent averaging theory.[6,12] Several sequences of phase and time-modulated pulses were designed which produce a constant $\pi/2$ net rotation of the spin density operator about a fixed axis in the xy plane of SO(3). As a result, these sequences show little phase distor-

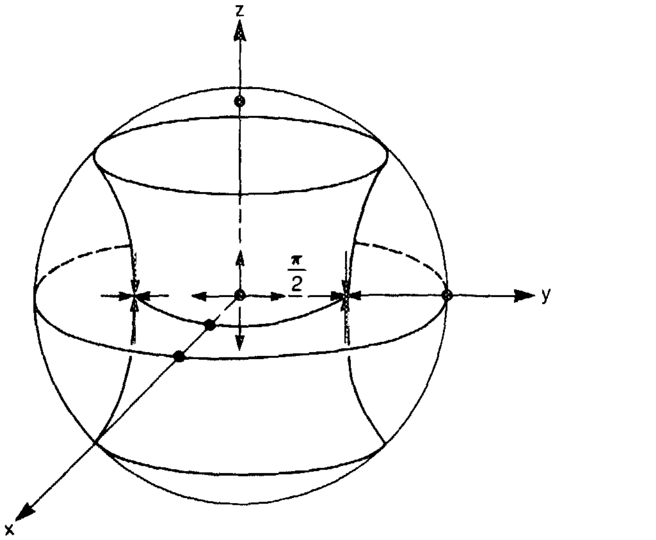

FIG. 5. The movement of points in SO(3) resulting from the iteration of a map with the set $R_\psi(\pi/2)$ as a stable fixed set and the origin as an unstable fixed set. $R_\psi(\pi/2)$ is shown here as being stable in all directions. The map of the scheme [0,0,180,135,135,150,330,0] possesses this type of stability.

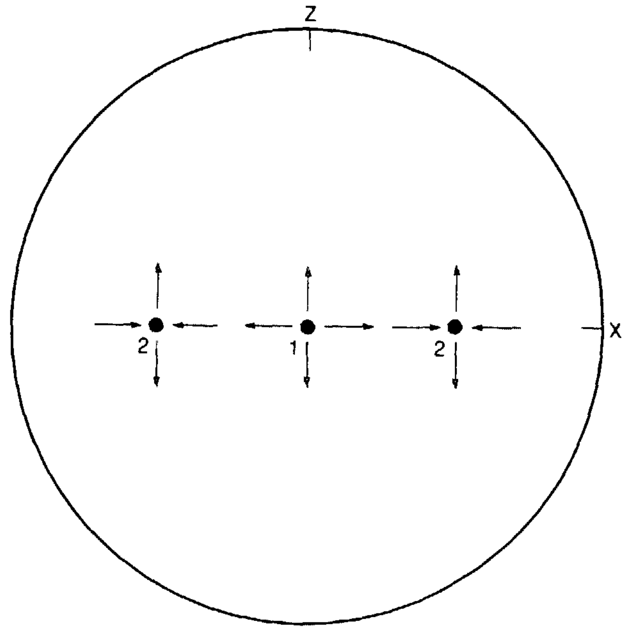

FIG. 7. Flow for a map which has $R_\psi(\pi/2)$ as a fixed set unstable in the z direction but stable otherwise. The map of the scheme [0,135,135,0] exhibits flow of this type. The instability off the transverse plane means that sequences generated by this scheme will not be compensated for imperfections caused by off-resonance effects. The origin is an incidental, unstable fixed point of this map.

tion of the NMR signal even for large pulse errors. These sequences were formulated to compensate for errors of a specific type, e.g., resonance offsets or rf missets. By way of contrast, the sequences presented here are designed to compensate for more than one type of error, and can be successively refined by iteration. Similar refinement is much more difficult to achieve within the context of coherent averaging theory.

C. Fixed point analysis of iterative schemes

The displacement of points in SO(3) by some map is termed the flow.[13,18] The direction of the flow for the map of the scheme [0,0,180,135,135,150,330,0] is schematically shown in Fig. 5 by the arrows pointing in towards the $R_\psi(\pi/2)$ circle, indicative of the stability of the map at this fixed point. The origin is an incidental, unstable fixed point of the map, which is reflected in the outward directed arrows emanating from this point. A two-dimensional version of this image appears in Fig. 6. This two-dimensional image can be contrasted with the two-dimensional flow for the map of the scheme [0,135,135,0] shown in Fig. 7. Points displaced from the fixed set $R_\psi(\pi/2)$ in the xy plane converge towards the fixed set for this map, but move away from this fixed set if there is a displacement in the z direction. The fixed set is therefore stable only in certain preferred directions.

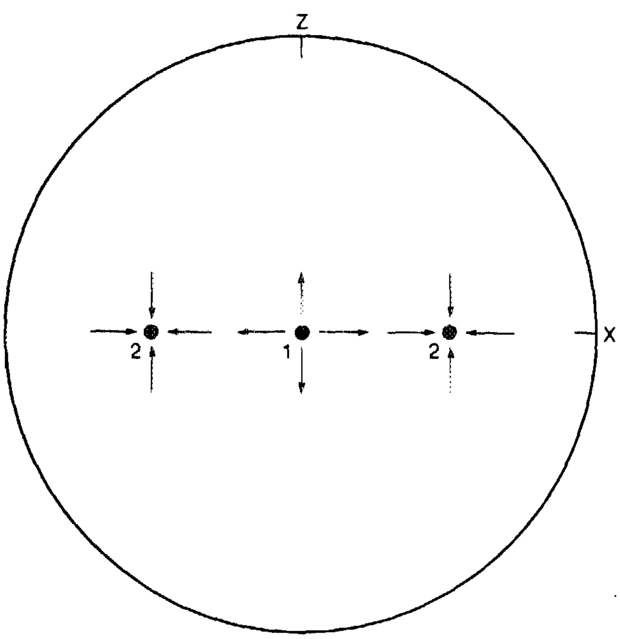

FIG. 6. Two-dimensional projection of Fig. 5. For simple phase shift-concatenation schemes, cylindrical symmetry permits such two-dimensional representations. The distance between points 1 and 2 is $\pi/2$.

The recursive expansion procedure has a map on SO(3) producing flow of the type diagrammed in Figs. 8 and 9. This flow assumes formation of a perfect inverse rotation in the scheme $S_i(S_i^{-1})_{90}$. The whole of SO(3) excluding the z axis and the equator converges smoothly to rotations of the type described by Eq. (12). It has been shown previously that this fixed set is superstable.[13] The superstability of this map was demonstrated for small displacements from the fixed set; however, the assumption of small displacements is not necessary, and it can be shown that for the idealized recursive expansion, the whole of SO(3) excluding the z axis and the equator must converge to the stable fixed set indicated. The remainder of the space converges to the origin, which is superstable only in the z direction.

The flow of these maps can be further studied by following the individual trajectories of an initial distribution of points in SO(3). One possible configuration appears in

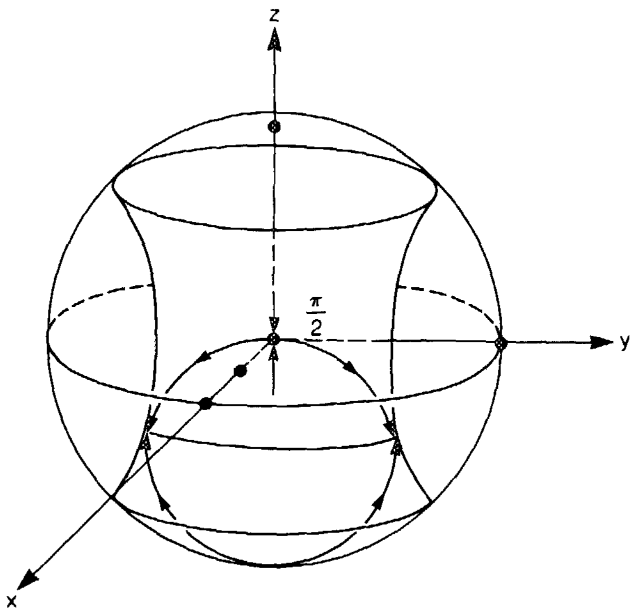

FIG. 8. The fixed sets and flow for a map on SO(3) of the recursive expansion scheme $S_i(S_i^{-1})_{90}$. The stable fixed set is a circle on the set of "apparent $\pi/2$ rotations" shown in Fig. 1. This flow assumes the formation of a perfect inverse rotation in the recursive expansion, and hence represents an idealization of its stability properties.

Fig. 10. The movement of points from this initial state for the three iterative schemes [0,135,135,0], [0,0,180,135,135,150,330,0], and $S_i(S_i^{-1})_{90}$ to their respective fixed points is clearly evidenced in Figs. 11, 12, and 13 by the congregation of points around the fixed points. Again, the difference in the fixed sets of the recursive expansion scheme and the phase shift-concatenation schemes is obvious.

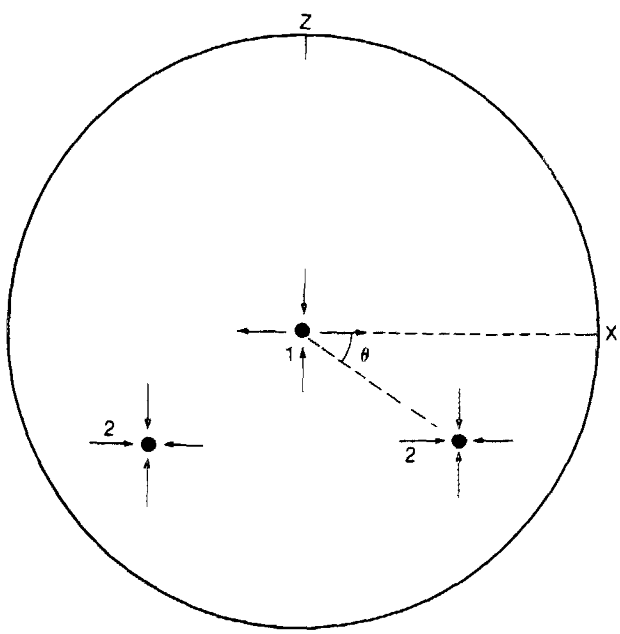

FIG. 9. Two-dimensional version of Fig. 8 for the recursive expansion procedure. The idealized recursive expansion procedure produces a mapping on SO(3) with cylindrical symmetry. The distance in SO(3) between points 1 and 2 is $2\pi/3$, and the angle θ is $\cos^{-1}(3^{-1/2})$.

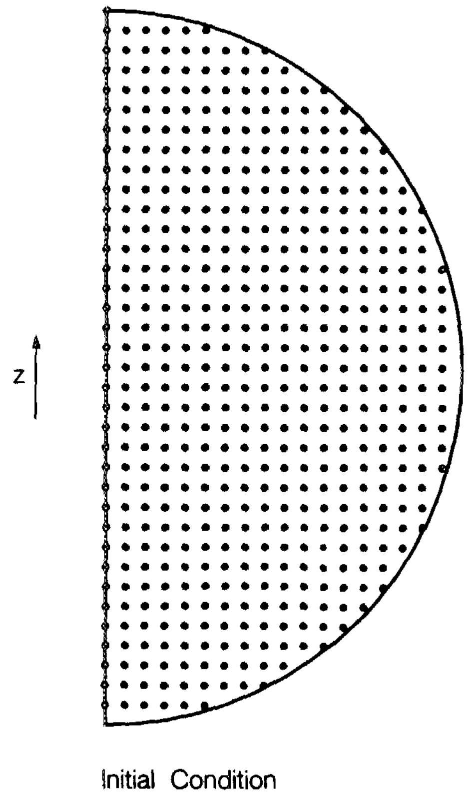

FIG. 10. An initial distribution of points in SO(3). The general movement of points in SO(3) when transformed by some map can be inferred by following how points progress away from this initial condition as the map is iterated. Stable fixed points can be identified by the clustering of points around certain areas. The trajectory of points for various maps with this configuration of points as the initial condition are displayed in the next three figures.

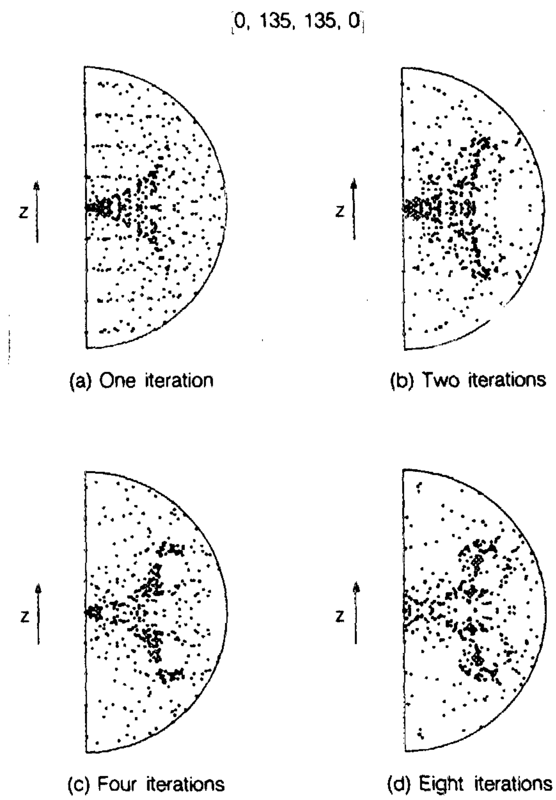

FIG. 11. The distribution of points in SO(3) following iteration of the scheme [0,135,135,0], assuming the initial configuration in Fig. 10. The results of one, two, four, and eight iterations are shown.

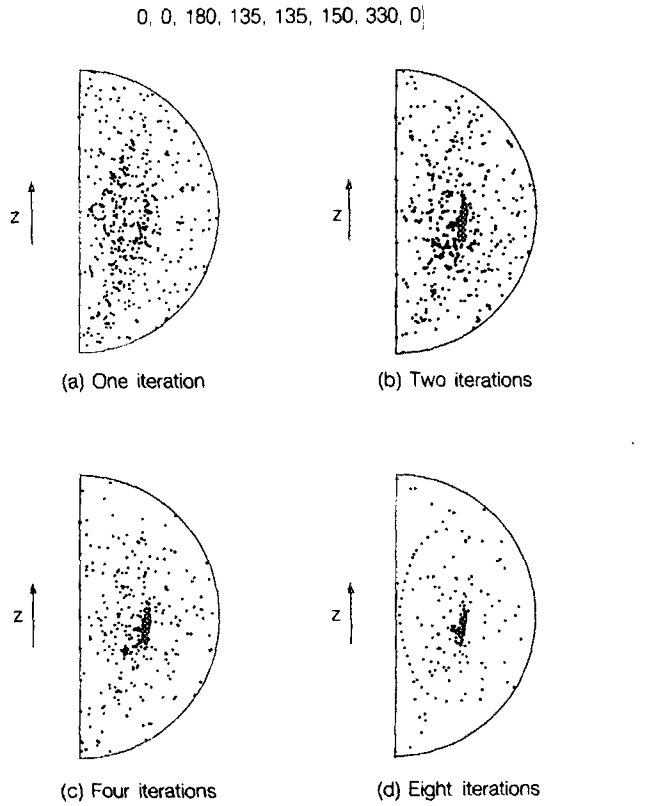

FIG. 12. Configuration of points following iteration of the scheme [0,0,180,135,135,150,330,0], assuming the initial condition in Fig. 10. Again, one, two, four, and eight iterations are shown.

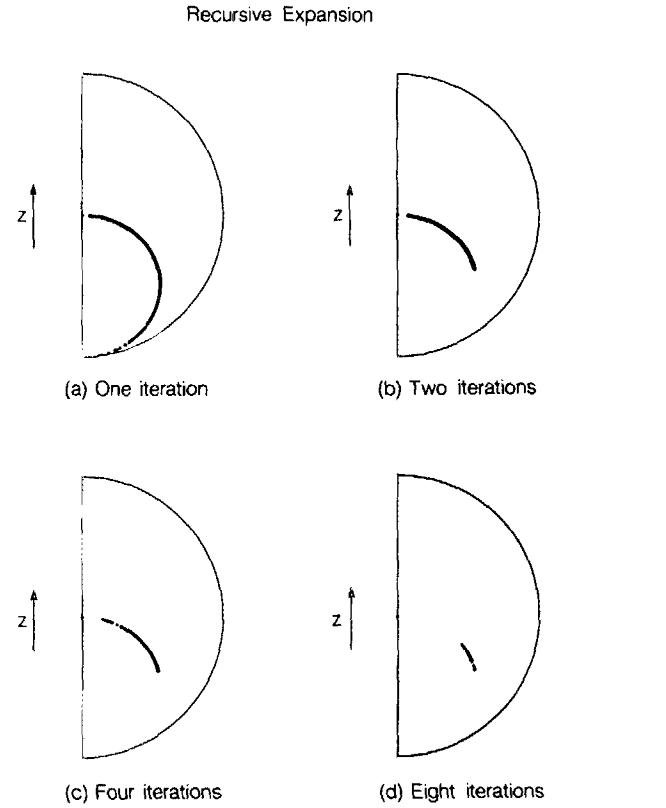

FIG. 13. Distribution of points following iteration of the idealized recursive expansion scheme $S_i(S_i^{-1})_{90}$, assuming the initial condition in Fig. 10. The results of one, two, four, and eight iterations are shown.

A second notable feature to observe in these figures is the orderly flow resulting from the recursive expansion, as contrasted with the two phase iteration schemes. This orderly flow is a consequence of the use of an exact inverse operation as part of the iterative scheme. Any practically realizable operation for forming an inverse sequence, however, results in chaotic flow similar to that exhibited by the two other schemes.

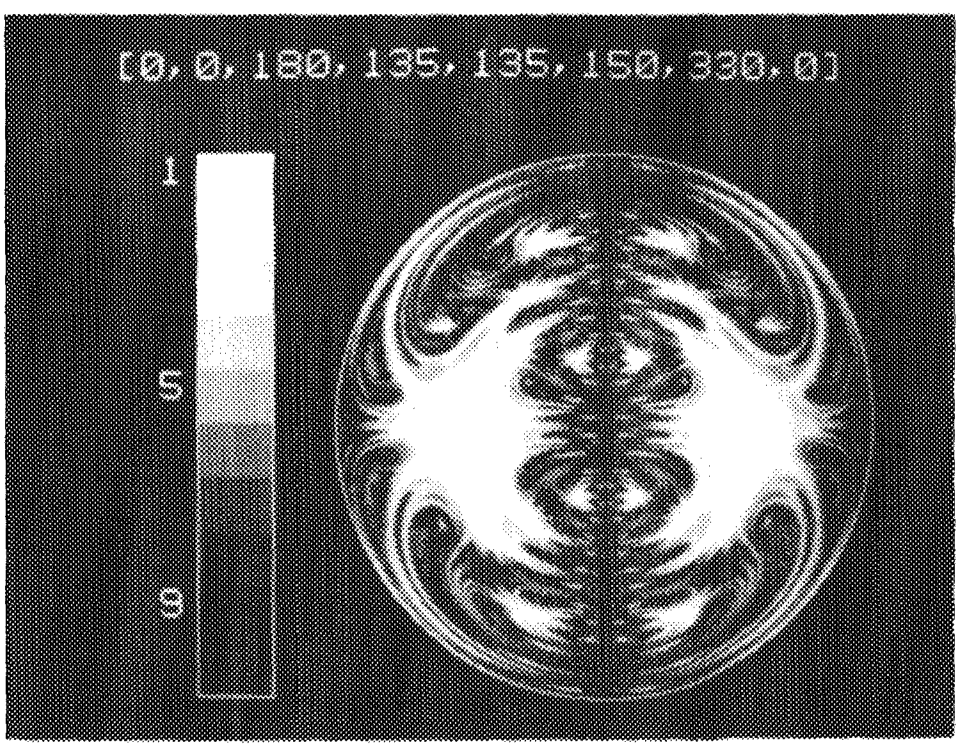

FIG. 14. Two-dimensioinal basin image of SO(3) containing the z axis for the map of the scheme [0,135,135,0]. The cylindrical symmetry of the map ensures that all such slices will be identical. Light colored regions represent rotations which converge to the form $R_\phi(\pi/2)$, the stable fixed point of Fig. 5, as the map is iterated. The density scale to the left indicates the number of iterations required for convergence.

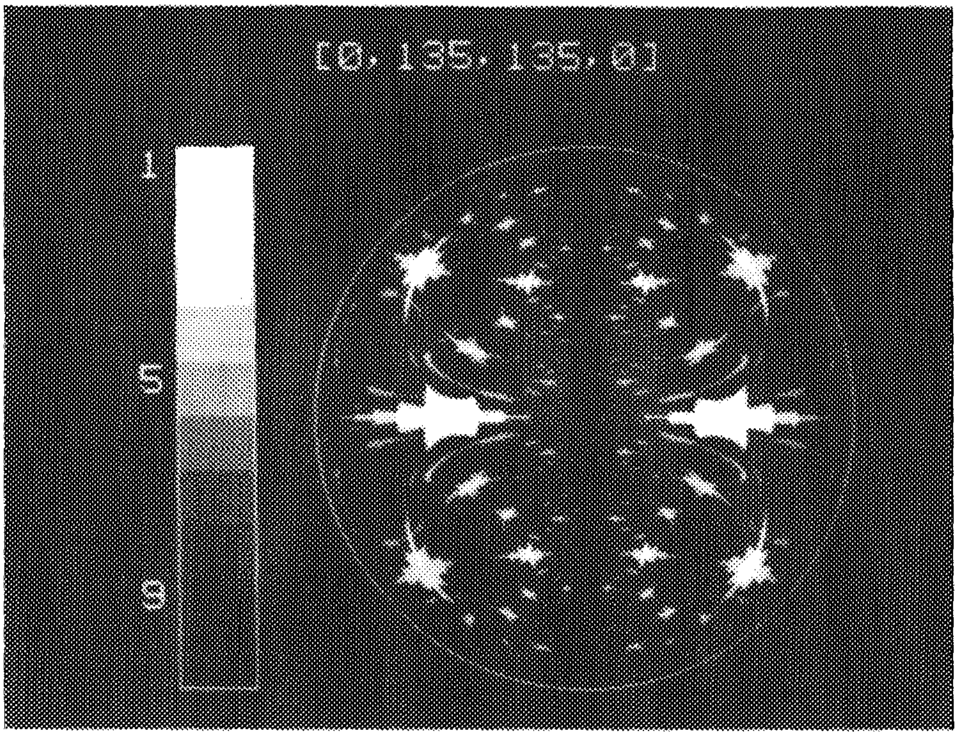

FIG. 15. Two-dimensional basin image of SO(3) containing the z axis for the map of the scheme [0,0,180,135,135,150,330,0]. Light colored regions show rotations which converge to the stable fixed set, namely, rotations of the form $R_\psi(\pi/2)$.

A final point to note about these diagrams is the reflection symmetry present in the diagram for the scheme [0,135,135,0]. This symmetry is a natural result of the symmetry of the scheme itself.[13] Since [0,0,180,135,135,150,330,0] is not a symmetric scheme, the reflection symmetry is absent in the diagram of the map for this scheme.

Computer generated basin images showing the regions of SO(3) which converge to the fixed set $R_\psi(\pi/2)$, and the number of iterations required for convergence, for maps of the schemes [0,135,135,0] and [0,0,180,135,135,150,330,0] are shown in Figs. 14 and 15. The program written to generate these images employed two criteria for determining convergence to the fixed set: the first is that the net angle α_i of the rotation operator must meet the inequality $|90° - \alpha_i| \leqslant 5°$; the second is that the square of the R_{zz} element of the three-dimensional rotation matrix $R(\alpha_i)$ must be less than 0.0076. These two criteria in combination test for convergence to only those rotations of the general form $R_\psi(\pi/2)$. As expected, the basin image for [0,135,135,0] is mostly dark, indicating the instability of the fixed set for displacements in the z direction. This is to be contrasted with

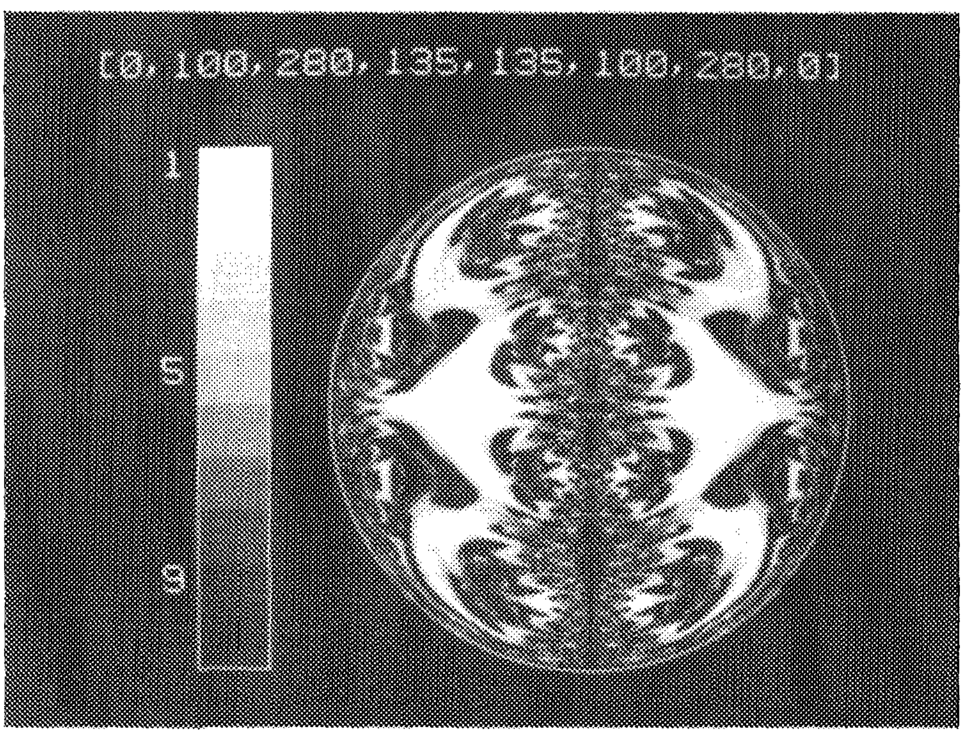

FIG. 16. Two-dimensional basin image in SO(3) containing the z axis for the map of the scheme [0,280,100,135,135,100,280,0]. The main feature distinguishing this image from that in Fig. 15 is the additional horizontal plane of symmetry resulting from the phase symmetry of the scheme.

the lighter basin image for the scheme [0,0,180,135,135,150,330,0], which is stable in all directions around the fixed set. Again, the xy reflection symmetry in the image for [0,135,135,0] is absent in the basin image for [0,0,180,135,135,150,330,0]. The additional symmetry of basin images for symmetry schemes is more readily apprehended for broadband schemes, which have bigger basins. An example of this appears in Fig. 16, which displays the basin image for the broadband symmetric scheme [0,280,100,135,135,100,280,0] taken from Table I.

A z slice basin image for the recursive expansion procedure has been presented in Ref. 13. The basin image appearing there was generated by testing only for the second of the two criteria enumerated above. The contours of this basin are smooth and well defined, and suggest in shape the object shown in Fig. 1 containing the set of apparent $\pi/2$ rotations. Adding the first circulation from above results in an image which is completely dark, while adding the criterion $|120° - \alpha_i| \leqslant 5°$ results in a basin image indistinguishable from the one in Ref. 13.

IV. CONCLUSION

We have demonstrated an eight pulse iterative scheme, consisting only of conveniently calibrated $\pi/2$ pulses, which provides substantial compensation of off-resonance and timing misset pulse errors. This sequence produces a net rotation of the spin density operator about an axis in the xy plane of angle $\pi/2$. Although the sequence requires pulses with phases other than the four standard quadrature phases, the eight pulse sequence should still be relatively easy to generate by placing a programmable, variable phase shifter in series with the quadrature circuit. This phase shifter need only be able to produce three distinct phase shifts of 0°, 135°, and 150° for the sequence we have examined here. This capability is becoming increasingly standard in most modern commercial NMR spectrometers. Furthermore, simulations indicate that systematic phase errors of at least $\pm 5°$ can be tolerated without significant degradation in performance.

Improvement and analytical development of these sequences might proceed along several lines. The first is to develop similar phase shift-concatenation schemes which are either symmetric or antisymmetric. Several of the sequences appearing in Table I are, indeed, symmetric, though none are antisymmetric. Antisymmetric schemes are, in principle, more desirable, since they offer the possibility of generating sequences which excite phase coherent NMR signals. The role and utility of the symmetry of a composite pulse sequence has been thoroughly considered elsewhere.[13]

A second improvement would be to determine schemes with maps on SO(3) which are superstable at the fixed set. It is a remarkable feature of the schemes in this paper that none of them results in a vanishing first order error term, i.e., none of them are superstable, and yet the sequences they generate are still well compensated for pulse errors. Nevertheless, improved sequences might be discovered with the use of superstable maps.

Another route to an improved sequence would be to investigate other initial sequences on which to begin iterating. Initial sequences represented by rotations in SO(3) preferentially concentrated within the basin of the map might provide a better starting point than a single $\pi/2$ pulse.

Deriving schemes with other fixed sets in addition to $R_\psi(\pi/2)$ is a fourth possible area of development. Maps with more than one stable fixed set provide the means for a shaped or tailored bandwidth response, as has been demonstrated elsewhere.[17,18] The methods proposed in this paper can serve as the guidelines for specifying one or more other arbitrary fixed sets.

The formalism presented here has distinct advantages in the design and analysis of highly compensated NMR pulse sequences. Though the focus of this paper has been on the special case of $\pi/2$ pulse sequences, the general principles used here can be applied to find iterative schemes for other types of responses as well.

ACKNOWLEDGMENTS

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