PHYSICAL REVIEW A **66**, 060301(R) (2002)

Efficient transfer of coherence through Ising spin chains

Navin Khaneja*
Division of Applied Sciences, Harvard University, Cambridge, Massachusetts 02138

Steffen J. Glaser
Institute of Organic Chemistry and Biochemistry II, Technische Universität München, 85747 Garching, Germany

(Received 11 April 2002; published 12 December 2002)

Experiments in coherent spectroscopy correspond to control of quantum mechanical ensembles guiding them from initial to final target states by unitary transformations. The control inputs (pulse sequences) that accomplish these unitary transformations should take as little time as possible so as to minimize the effects of relaxation and to optimize the sensitivity of the experiments. Here, we present a novel approach for efficient control of dynamics in spin chains of arbitrary length. The approach relies on creating certain three spin encoded states, which can be efficiently transferred through a spin chain. The methods presented are expected to find applications in control of spin dynamics in coherent spectroscopy and quantum information processing.

DOI: 10.1103/PhysRevA.66.060301 PACS number(s): 03.67.Lx, 05.50.+q

According to the postulates of quantum mechanics, the evolution of the state of a closed quantum system is unitary and is governed by the time-dependent Schrödinger equation. This evolution can be controlled by systematically changing the Hamiltonian of the system. The control of quantum systems has important applications in physics and chemistry [1–4]. In particular, the ability to steer the state of a quantum system (or of an ensemble of quantum systems) from a given initial state to a desired target state forms the basis of spectroscopic techniques such as nuclear magnetic resonance (NMR) and electron spin resonance (ESR) spectroscopy [1,3], laser coherent control [4] and quantum computing [5,6]. Achieving a desired unitary evolution in a quantum system in minimum time is an important practical problem in coherent spectroscopy. Developing short pulse sequences (control laws) which produce a desired unitary evolution has been a major thrust in NMR spectroscopy [1]. For example, in the NMR spectroscopy of proteins, the transfer of coherence along spin chains is an essential step in a large number of key experiments.

A spectroscopist has at his disposal a limited set of control Hamiltonians $\{\mathcal{H}_j\}$ (produced by external electromagnetic fields) that can be turned on and off to modify the net Hamiltonian of the system. There is a natural coupling (interaction) between the spins and in the absence of any external control Hamiltonians, the state of spin system evolves under this interaction or coupling Hamiltonian $\mathcal{H}_c$. The task of the pulse designer is to find the right sequence of external pulses interspersed with evolution of the system under the coupling Hamiltonian $\mathcal{H}_c$ for different time periods, in order to create a net evolution or unitary transformation that transforms the state of the system from some initial to a desired final state in minimum possible time.

Even for two coupled spins 1/2, the time-optimal transfer of polarization or of coherence is not trivial [7,8]. Numerous approaches have been proposed and are currently used [9] to transfer polarization or coherence through chains of coupled spins. Examples are the design of radio-frequency (RF) pulse trains that create an effective Hamiltonian [1,10], which makes it possible to propagate spin waves in such chains [11–15]. In order to achieve the maximum possible transfer amplitude, many other approaches, that rely either on a series of selective transfer steps between adjacent spins or on concatenations of two such selective transfer steps [9,10] have been developed.

Here, we consider an approach to control the transfer of coherence in spin chains of arbitrary length. The approach relies on the creation of a three spin encoded state and efficient propagation of this encoded state through the spin chain. Our method is based on variational ideas as captured by the theory of optimal control [8]. In the present context of control of nuclear spin ensembles, we are interested in finding a sequence of RF pulses that will efficiently transfer any state (known or unknown) of a given spin in a spin chain to a desired target spin. Compared to conventional experiments, this new approach makes it possible to speed up the transfer rate by up to a factor of three, which suggests applications in NMR spectroscopy and experimental quantum computation.

Without loss of generality, here we consider the problem of transferring the coherence of a spin at one end of a spin chain (label spin 1) to a spin at the opposite end of the chain (label spin n) in a spin ensemble. Consider an initial density operator ρ_0 representing coherence or polarization on the first spin, which has the general form $\rho_0 = \frac{1}{2}\mathbf{1} + a_1 I_{1x} + a_2 I_{1y} + a_3 I_{1z}$. The goal is to transfer this density operator to the operator $\frac{1}{2}\mathbf{1} + a_1 I_{nx} + a_2 I_{ny} + a_3 I_{nz}$. Note that it suffices to find a unitary transformation U that transfers $I_{1x} \rightarrow I_{nx}$ and $I_{1y} \rightarrow I_{ny}$. The same unitary transformation will also transfer $[I_{1x}, I_{1y}] \rightarrow [I_{nx}, I_{ny}]$, i.e., $I_{1z} \rightarrow I_{nz}$. Therefore, the transfer of the coherent state of spin 1 to spin n is equivalent to the transfer of the non-Hermitian operator $I_1^- = I_{1x} - iI_{1y}$ to $I_n^- = I_{nx} - iI_{ny}$. The transfer between such non-Hermitian operators arises naturally in coherent spectroscopy of ensembles and constitutes a fundamental step in multidimensional NMR spectroscopy of biological macromolecules [7]. We emphasize again that any unitary transformation that transfers I_1^- to I_n^-, implies that any state of the form $\rho_0 = \frac{1}{2}\mathbf{1} + a_1 I_{1x} + a_2 I_{1y} + a_3 I_{1z}$ is transferred to spin n.

---

*Email address: navin@hrl.harvard.edu.

Besides applications in spectroscopy, finding optimal methods to control the dynamics of coupled spin networks is of fundamental importance for the practical implementation of quantum information processing. In recent years, many innovative proposals have come out to harness the dynamics of spins in the liquid [5,6] and solid state [16,17] for the purpose of information processing. Like many coherence transfer experiments in multidimensional NMR spectroscopy, these NMR quantum computing architectures rely on elaborate sequences of radio frequency (RF) pulses for realizing desired effective Hamiltonians. Recent proposals by Yamamoto [17] use a chain of nuclear spins 1/2 in the solid state for purpose of computing. A major challenge in such architectures, which is also found in various other quantum information devices, is finding efficient ways of making qubits interact if they are not directly coupled. A prototype example of this problem is finding efficient ways to generate unitary transformations which exchange the states of spins on the two opposite ends of a spin chain. Pulse sequences presented in this paper can be used to accomplish such operations efficiently.

Consider a linear chain of n weakly interacting spin $\frac{1}{2}$ particles placed in a static external magnetic field in the z direction and with Ising type couplings between next neighbors [18,19]. In a suitably chosen (multiple) rotating frame which rotates with each spin at its resonant frequency, the Hamiltonian that governs the free evolution of the spin system is given by the coupling Hamiltonian

$$\mathcal{H}_c = 2\pi \sum_{k=2}^{n} J_{k-1,k} I_{(k-1)z} I_{kz},$$

where $J_{k-1,k}$ is the coupling constant between spin $k-1$ and k. If the resonance frequencies of the spins are well separated, spin k can be selectively excited (addressed) by an appropriate choice of the amplitude and phase of the RF field at its resonant frequency. The goal of the pulse designer is to make appropriate choice of the control variables comprising of the frequency, amplitude and phase of the external RF field to effect a net unitary evolution $U(t)$ which efficiently transfers the initial operator $A = I_1^-$ to $B = I_n^-$. For simplicity but without loss of generality (vide infra), we assume that all coupling constants in the spin chain are equal, i.e., $J_{k-1,k} = J$ for $1 < k \leqslant n$.

A straightforward way of transferring the operator I_1^- to I_n^- is to perform sequential transfers, whereby I_k^- is transferred to I_{k+1}^- [21,22]. Each of these sequential steps takes $3/2J$ units of time, resulting in a total time of $3(n-1)/2J$ (see Fig. 1). However this is far from optimal. It can be shown [8] that given any initial operator A_k that represents a state involving spin k and spins with label less than k, the minimum time required to advance this operator one step up the spin chain is $1/2J$. This suggests that an efficient approach to transferring the state I_1^- to I_n^- is to prepare an encoded state Λ_k^- such that it is possible to go from Λ_k^- to Λ_{k+1}^- in a time of $1/2J$. Furthermore, it is desirable that these encoded states be sufficiently localized so that Λ_1^- can be encoded and decoded in a short time. We will refer to such encoded states as effective soliton operators (see end of this section).

Now we consider the three specific effective soliton operators $\Lambda_{kx} = 2I_{(k-2)x} I_{(k-1)z}$, $\Lambda_{ky} = 2I_{(k-1)x} I_{kz}$, and $\Lambda_{kz} = 4I_{(k-2)x} I_{(k-1)y} I_{kz}$, which obey the commutation relations $[\Lambda_\alpha, \Lambda_\beta] = i\epsilon_{\alpha\beta\gamma}\Lambda_\gamma$, where $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita symbol which is 1 (or -1) if $\{\alpha\beta\gamma\}$ is an even (or odd) permutation of $\{x,y,z\}$ and 0 if two or more of the indices α, β, γ are identical. Each individual soliton operator $\Lambda_{k\alpha}$ is advanced along the spin chain by one unit if the propagator $U_\Lambda = \exp\{-i\Delta\mathcal{H}_c\}\exp\{-i(\pi/2)F_y\}$ with $F_y = I_{1y} + I_{2y} + \cdots I_{ny}$ is applied:

$$\Lambda_{k\alpha} \xrightarrow{U_\Lambda} \Lambda_{(k+1)\alpha}.$$

The propagator U_Λ can be realized by applying a nonselective $90°_y$ pulse (with negligible duration) to all spins, followed by the evolution of the spin system under the coupling Hamiltonian $\mathcal{H}_c$ for a duration $\Delta = (2J)^{-1}$.

With the help of the soliton operators $\Lambda_k^- = \Lambda_{kx} - i\Lambda_{ky}$, it is possible to transfer $I_1^- = I_{1x} - iI_{1y}$ efficiently to $I_n^- = I_{nx} - iI_{ny}$:

$$I_1^- \xrightarrow{U_1} \xrightarrow{U_2} \Lambda_3^- \underbrace{\xrightarrow{U_\Lambda} \ldots\ldots \xrightarrow{U_\Lambda}}_{(n-3)\text{times}} \Lambda_n^- \xrightarrow{U_\Lambda} \xrightarrow{\quad U_{n+1} \quad} I_n^- .$$

Here, the encoding of I_1^- as the soliton operator Λ_3^- is effected by the propagators

$$U_1 = \exp\{-i\Delta\mathcal{H}_c\}\exp\left\{i\frac{\pi}{2}I_{1y}\right\}\exp\left\{-i\frac{\pi}{2}I_{1x}\right\},$$

and

$$U_2 = \exp\{-i\Delta\mathcal{H}_c\}\exp\{-i(\pi/2)(I_{1x} + I_{2y})\}.$$

Finally, the decoding of the soliton operator Λ_n^- into I_n^- is achieved by the propagators U_Λ and

$$U_{n+1} = \exp\{i(\pi/2)I_{nx}\}\exp\{-i\Delta\mathcal{H}_c\}\exp\{i(\pi/2)(I_{nx} - I_{(n-1)y})\}.$$

U_1, U_2, U_Λ, and U_{n+1} require a period $\Delta = (2J)^{-1}$ each, resulting in the time $\tau_{\text{soliton}} = (n+1)/2J$ for the complete transfer from I_1^- to I_n^-.

The flow of soliton operators is summarized in Fig. 1(b). The panel schematically traces the evolution of the initial operators I_{1x}, I_{1y}, and I_{1z} via the soliton operators Λ_{kx}, Λ_{ky}, and Λ_{kz} in the spin chain as a function of time. The operators Λ_{kx}, Λ_{ky} and Λ_{kz} represent local correlations of spin $k-1$ with its neighbors. Under the proposed pulse sequences, these correlations advance one step in the spin chain, every $\tau_{\text{step}} = 1/2J$ units of time. Although these operators evolve to other operators under the proposed pulse sequences, if the spin system is observed stroboscopically, every τ_{step} units of time, the correlations maintain their shape

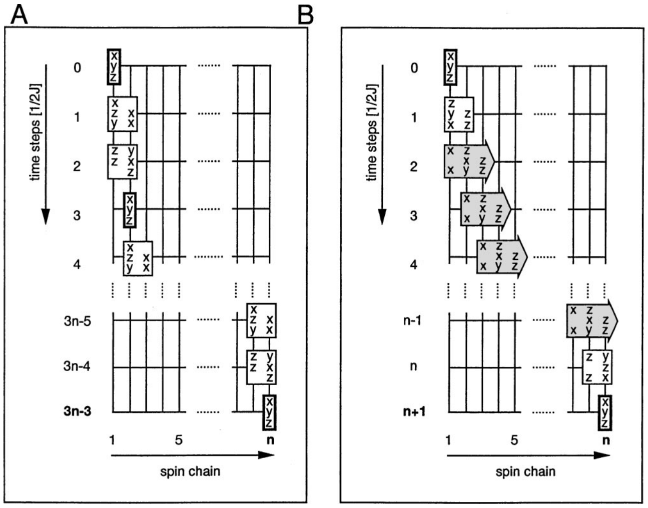

FIG. 1. Panel (a) shows the flow of coherence and polarization in a spin chain under a sequence of selective isotropic mixing periods [10,21,22], (sequential swap operations) each of which can be decomposed in three steps of duration $\Delta=(2J)^{-1}$ with effective Hamiltonians $2\pi JI_{(k-1)x}I_{kx}$, $2\pi JI_{(k-1)y}I_{ky}$, and $2\pi JI_{(k-1)z}I_{kz}$, respectively. After the first step the initial operators I_{1x}, I_{1y}, I_{1z} are transferred to I_{1x}, $2I_{1z}I_{2x}$, $2I_{1y}I_{2x}$ respectively. Coherence is transferred in a sequential manner where the state of the spin k is transferred to spin $k+1$ in $3J/2$ units of time. The total transfer takes $3(n-1)/2J$ units of time. Panel (b) shows the flow of coherence and polarization under the proposed pulse sequence based on effective soliton operators (indicated by gray arrows). Here, a localized spin wave is created which moves one step in the spin chain in every $1/2J$ seconds. The total transfer time for the proposed pulse sequence is $(n+1)/2J$. For clarity, operators such as I_{kx} are indicated by the letter x at position k within a box; each box displays, in a vertical array, what the initial operators x,y,z have evolved to at the time indicated on the vertical axis. Similarly, bilinear (or trilinear) product operators such as Λ_{kx} (or Λ_{kz}) are indicated only by the axis labels x, y, or z at the corresponding spin position, omitting prefactors of 2 (or of 4) and possible algebraic signs.

and are just translated one step up in the spin chain. Hence the name effective soliton operators.

The time τ_{soliton} taken by the proposed pulse sequence should be compared with the transfer time for conventional pulse sequences which transfer I_1^- to I_n^- [10,21,22]. These pulse sequences require $n-1$ steps of selective isotropic transfers in which the jth step transfers the operator I_j^- to I_{j+1}^-. In the jth step, only spins j and $j+1$ are active and the remaining spins in the chain are decoupled. This mode of transfer is depicted in panel (a) of Fig. 1. Each such isotropic transfer step requires $3/2J$ units of time and therefore the total time is $3(n-1)/2J$. In the limit of large n, the proposed soliton sequences only take 1/3 amount of time as compared to state of the art pulse sequences. A comparison of the time taken for the coherence transfer by the conventional sequence of selective isotropic pulse sequences τ_{conv} and the proposed pulse sequences τ_{soliton} is shown in Fig. 2 for $n \leqslant 10$.

As the implementation of spin-selective pulses is often difficult, it is of interest to note that in the new sequence, spin-selective pulses are only required in the encoding and decoding steps but not in the propagation steps. The propagators U_Λ that advance the effective soliton operators by one step in the chain can be implemented entirely without spin-selective pulses. Hence, it is only required to selectively address the first and last spin in the chain but it is not necessary to implement selective rotations for the remaining spins. In contrast, a sequence of sequential swap operations requires spin-selective rotations for all spins in the chain, because during a swap operation between two directly coupled spins spins k and $k+1$, the spins $k-1$ and $k+2$ must be selectively decoupled. A straightforward implementation of such a swap operation between spins k and $k+1$ consists of three time periods of duration $\Delta=(2J)^{-1}$ with effective propagators $\exp\{-i\pi I_{kx}I_{(k+1)x}\}$, $\exp\{-i\pi I_{ky}I_{(k+1)y}\}$, and $\exp\{-i\pi I_{kz}I_{(k+1)z}\}$. The creation of these effective propagators requires the application of several (spin-selective) radio frequency pulses (for a detailed discussion of various experimental pulse sequences for the implementations of swap operations, see [20]). For some applications, the significantly reduced selectivity requirements of the new transfer scheme may be yet another advantage compared to a sequence of selective swap operations. A further issue to be considered is the relative sensitivity of the sequences to errors in timing, strength of the radio-frequency pulses and due to their finite duration. For each time period Δ and each radio-frequency pulse, there are finite errors in both sequences. However, in the new sequence, not only the number of transfer steps of duration Δ (see Fig. 1), but also the number of pulses is reduced by about a factor of three, which helps to increase the robustness of the new sequence to experimental imperfections. As quantum information is encoded differently in the new transfer scheme (spread over three spins or qubits instead of one or two), it will in general have a different degree of robustness with respect to relaxation or external noise. A detailed study of the transfer efficiency for various types of relaxation mechanisms or environmental noise is of interest but beyond the scope of the present paper.

The proposed pulse sequences are also compared with the widely used concatenated INEPT pulse sequences [23,24], which transfer only one component of magnetization along a spin chain, i.e., $I_{1x}\rightarrow I_{nx}$. If all couplings are equal to J, the time required for transferring $I_{1x}\rightarrow I_{nx}$ by the concatenated INEPT pulse sequences is $n/2J$. For large n, this is approximately the same as the time required for the new soliton-based pulse sequences. However, the soliton sequences transfer the complete state of spin 1 to spin n, which may result in appreciable gain of signal to noise ratio in spectroscopic applications [9].

The proposed pulse sequences can be used to efficiently exchange the (arbitrary) states of spins at the two ends of a spin chain with possible applications to proposed quantum computing architectures [17]. This exchange operation between spin 1 and n, in general will not preserve the state of other spins on the spin chain and hence donot represent a swap gate between spin 1 and n in the usual sense. The proposed soliton sequences can be used to transfer the state of spin 1 to spin n and spin n to spin 1 simultaneously in $(n+1)/2J$ units of time. This should be compared to the approach where states of spin 1 and n are exchanged through

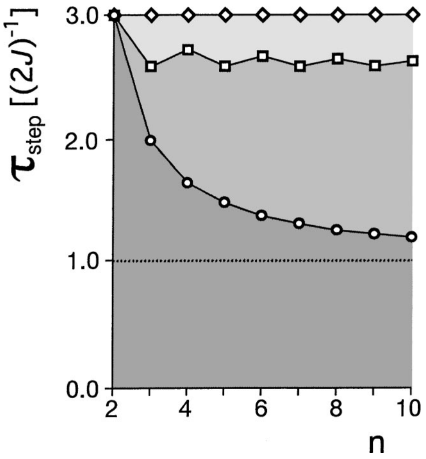

FIG. 2. A comparison of $\tau_{\text{step}}=\tau/(n-1)$, the average time required to advance by one step in a chain of n coupled spins for pulse sequences which effect full transfer from I_1^- to I_n^-. Diamonds: conventional sequence of selective isotropic mixing steps between neighboring spins [21,22]. Squares: sequence of optimal indirect SWAP$(k,k+2)$ operations [25] (of duration $3\sqrt{3}/2J$) which are followed by a selective isotropic mixing step between spins $(n-1)$ and n if n is even. Circles: soliton pulse sequence.

a sequence of neighboring swap operations [20]. Each swap operation requires $3/2J$ units of time. Therefore the total time required to exchange states of spin 1 and n by sequential swapping is at least $3(n-1)/2J$, which in the limit of large n is three times longer than the proposed approach based on soliton operators.

Finally, we analyze the time taken by the proposed coherence transfer methodology when couplings are not equal. To produce the effect of the propagator U_Λ acting on Λ_{kx}, Λ_{ky}, Λ_{kz} in Eq. (1), the only terms in the coupling Hamiltonian $\mathcal{H}_c$ that are instrumental are $2\pi(J_{k-2,k-1}I_{(k-2)z}I_{(k-1)z}+J_{k-1,k}I_{(k-1)z}I_{kz}+J_{k,k+1}I_{kz}I_{(k+1)z})$. If $J_{k,k+1}$ is the smallest of the coupling constants $J_{k-2,k-1}$, $J_{k-1,k}$, $J_{k,k+1}$, i.e., if $J_{k,k+1}=\min(J_{k-2,k-1},J_{k-1,k},J_{k,k+1})$, then it will take $\tau_k=1/2J_{k,k+1}$ units of time to produce the propagator $\exp\{-i\pi(I_{(k-2)z}I_{(k-1)z}+I_{(k-1)z}I_{kz}+I_{kz}I_{(k+1)z})\}$. This is achieved by letting the coupling $J_{k,k+1}$ evolve during τ_k while letting $J_{k-2,k-1}$ and $J_{k-1,k}$ couplings evolve only during $(J_{k,k+1}/J_{k-2,k-1})\tau_k$ and $(J_{k,k+1}/J_{k-1,k})\tau_k$ respectively and decoupling these couplings for the remaining time. This can be achieved by standard refocusing techniques [1]. Therefore the total time required for propagation of the soliton Λ_3 to Λ_n is

$$\sum_{k=1}^{n-3}\frac{1}{2\min(J_{k,k+1},J_{k+1,k+2},J_{k+2,k+3})}.$$

Similar arguments yield that the time required for preparation of soliton state from the initial state is $1/2J_{12}+1/2\min(J_{12},J_{23})$ and finally the time required to reduce the soliton state to the final state is $1/2\min(J_{n-2,n-1},J_{n-1,n})+1/2J_{n-1,n}$.

The proposed methods for control of spin chain dynamics may have the potential to improve the sensitivity of multidimensional heteronuclear triple resonance experiments, used for example for sequential resonance assignments in protein NMR spectroscopy [9]. The proposed method of manipulating dynamics of spin chains could also reduce decoherence effects in experimental realizations of quantum information devices [17]. Although minimizing the time required to produce desired unitary evolutions in a quantum system is expected to reduce dissipation and relaxation effects, optimizing pulse sequences by incorporating a realistic relaxation model may further improve the sensitivity of experiments in coherent spectroscopy.

This work was funded by DARPA-Stanford Grant No. F49620-01-1-0556. This work was funded by the Fonds der Chemischen Industrie and the Deutsche Forschungsgemeinschaft under Grant No. G1 203/4-1.

[1] R. R. Ernst *et al.*, *Principles of Nuclear Magnetic Resonance in One and Two Dimensions* (Clarendon, Oxford, 1987).
[2] H. M. Wiseman and G. J. Milburn, Phys. Rev. Lett. **70**, 548 (1993).
[3] A. Schweiger, in *Modern Pulsed and Continuous Wave Electron Spin Resonance*, edited by M. K. Bowman (Wiley, London, 1990), p. 43.
[4] W. S. Warren *et al.*, Science **259**, 1581 (1993).
[5] D. G. Cory *et al.*, Proc. Natl. Acad. Sci. U.S.A. **94**, 1634 (1997).
[6] N. A. Gershenfeld and I. L. Chuang, Science **275**, 350 (1997).
[7] S. J. Glaser *et al.*, Science **208**, 421 (1998).
[8] N. Khaneja *et al.*, Phys. Rev. A **63**, 032308 (2001).
[9] J. Cavanagh *et al.*, *Protein NMR Spectroscopy: Principles and Practice* (Academic, San Diego, 1996).
[10] S. J. Glaser and J. J. Quant, in *Advances in Magnetic and Optical Resonance*, edited by W. S. Warren (Academic, San Diego, 1996), Vol. 19, p. 59.
[11] E. H. Lieb *et al.*, Ann. Phys. (Paris) **16**, 407 (1961).
[12] H. M. Pastawski *et al.*, Chem. Phys. Lett. **261**, 329 (1996).
[13] R. M. White, *Quantum Theory of Magnetism* (Springer, Berlin, 1983).
[14] D. C. Mattis, *The Theory of Magnetism I, Statistics and Dynamics* (Springer, Berlin, 1988).
[15] Z. L. Mádi *et al.*, Chem. Phys. Lett. **268**, 300 (1997).
[16] B. E. Kane, Nature (London) **393**, 133 (1998).
[17] F. Yamaguchi and Y. Yamamoto, Appl. Phys. A: Mater. Sci. Process. **68**, 1 (1999).
[18] E. Ising, Z. Phys. **31**, 253 (1925).
[19] W. J. Caspers, *Spin Systems* (World Scientific, London, 1989).
[20] T. Schulte-Herbrüggen and O. W. Sørensen, Concepts Magn. Reson. **12**, 389 (2000).
[21] D. P. Weitekamp *et al.*, J. Chem. Phys. **77**, 2870 (1982).
[22] P. Caravatti *et al.*, Chem. Phys. Lett. **100**, 305 (1983).
[23] L. E. Kay *et al.*, J. Magn. Reson. (1969-1992) **91**, 84 (1991).
[24] A. Majumdar and E. P. Zuiderweg, J. Magn. Reson., Ser. A **113**, 19 (1995).
[25] N. Khaneja *et al.*, Phys. Rev. A **65**, 032301 (2002).