Hadamard Products of Product Operators and
the Design of Gradient-Diffusion Experiments
for Simulating Decoherence by NMR Spectroscopy

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Abstract

An extension of the product operator formalism of NMR is introduced, which
uses the Hadamard matrix product to describe many simple spin 1/2 re-
laxation processes. The utility of this formalism is illustrated by deriving
NMR gradient-diffusion experiments to simulate several decoher ence models
of interest in quantum information processing, along with their Lindblad and
Kraus representations.

1. Introduction

The product operator formalism is widely used for designing radio-frequency pulse se-
quences to control the coherent evolution of multi-spin-1/2 systems in liquid-state nuclear
magnetic resonance (NMR) spectroscopy [1–4]. This is a symbolic representation of the
$2^N \times 2^N$ Hermitian matrices of $N$-spin density operators, observables and Hamiltonians,
together with a collection of rules for evolving them under radio-frequency (RF) pulses and
free evolution delays (in the Schrödinger, Heisenberg, or interaction picture defined by a
rotating frame [4]). This representation is obtained by expanding the matrices versus the
“product operator” basis consisting of all possible $N$-fold Kronecker (or tensor) products of
the usual Pauli matrices $\sigma_x, \sigma_y, \sigma_z$ and the $2 \times 2$ identity $\sigma_I$. Together with the simpler and
more general rules provided by the underlying geometric algebra structure [5,6], the product
operator formalism has also proven invaluable in designing pulse sequences to implement a
wide variety of logic gates for quantum information processing (QIP) by NMR [7–10] (see
[11,12] for recent reviews).

In contrast, the general theory of NMR relaxation [4,13] is usually developed using the
“superoperator” representation of linear mappings on $N$-spin density matrices $\rho$ relative to
the matrix basis $|m\rangle\langle m'|$ obtained from the eigenvectors of the Zeeman Hamiltonian $|m\rangle$
$(m, m' \in \{0, 1\}^N)$. This representation consists of the $2^{2N} \times 2^{2N}$ matrices acting on the

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“columnized” density matrices versus the corresponding vector basis $|m⟩⟨m|$. The Lindblad differential operator $[14]$ and Kraus operator sum $[15]$ representations, which are becoming commonplace in quantum optics $[10]$, condensed matter physics $[17]$, and in foundational studies of decoherence $[18–20]$, are virtually unknown to NMR spectroscopists. The recent use of NMR gradient-diffusion methods to simulate theoretical decoherence processes in experimental studies of the control of decoherence for QIP $[21,22]$ has made it desirable to develop a new representation which: (i) blends naturally with the product operator formalism; (ii) avoids the unwieldy use of full $2^N \times 2^N$ superoperator matrices; (iii) is sufficient to analyze common NMR gradient-diffusion experiments; (iv) describes the theoretical decoherence models most often studied; (v) can easily be translated into the corresponding Lindblad, Kraus and superoperator representations.

This paper presents such a formalism, which is based on the well-known Hadamard product of matrices $[23,24]$. This formalism permits the representation of relaxation processes for which the $2^{2N} \times 2^{2N}$ superoperator matrix is diagonal relative to some operator (not superoperator) basis. As such it can handle both $T_1$ and $T_2$ relaxation (decoherence), but not cross-relaxation $[1,3]$. In terms of Kraus operator sums, this is equivalent to requiring that the Kraus operators can all be simultaneously diagonalized. The utility of the Hadamard product formalism will be demonstrated by giving streamlined derivations of closed-form, time-dependent density operators for a variety of NMR gradient-diffusion experiments. These include collective and independent decoherence about any axis, together with several more complex forms of decoherence including the collective isotropic model. When possible, the corresponding Lindblad and Kraus operators for these experiments will also be given. We have found, however, that the Hadamard product permits a concise analytic description of decoherence with arbitrary correlations among the fluctuating fields at the different spins involved, whereas the corresponding Lindblad and Kraus forms entail the solution of polynomial equations, and can be written down explicitly only in the simplest cases.

2. Hadamard products of product operators

Given two $M \times M'$ complex-valued matrices $A = [a_{mm'}]_{m,m'=0}^{M-1,M'-1}$ and $B = [b_{mm'}]_{m,m'=0}^{M-1,M'-1}$, their Hadamard product is the matrix consisting of the complex products of corresponding pairs of elements, $A \odot B = [a_{mm'}b_{mm'}]_{m,m'=0}^{M-1,M'-1}$. Like the usual matrix product, the Hadamard product satisfies the mixed product identity with the Kronecker product $[23,24]$.

$$(A \odot B) \odot (C \otimes D) = (A \odot C) \otimes (B \odot D), \quad (1)$$

but unlike the usual matrix product it is commutative.

In the following, all matrices will be expressed as linear combinations of product operators, as described above. Some further notations which prove useful include: $M = 2^N$; $I = \sigma_1 \otimes \cdots \otimes \sigma_1$ (the $M \times M$ identity); $\sigma_\mu$ in the $n$-th place, $n = 1, \ldots, N$; $E_0^n = (I + \sigma_\mu^n)/2 = \sigma_1 \otimes \cdots \otimes (|0⟩⟨0|) \otimes \cdots \otimes \sigma_1$, $E_1^n = (I - \sigma_\mu^n)/2 = \sigma_1 \otimes \cdots \otimes (|1⟩⟨1|) \otimes \cdots \otimes \sigma_1$ (so $(E_0^n)^2 = E_0^n$, $(E_1^n)^2 = E_1^n$, and $E_0 E_1^n = E_1^n E_0^n = O$, the $M \times M$ zero matrix); and
\[ E_m = E^1_m \cdots E^N_m, \quad \sigma^m_\mu = (\sigma^1_\mu)^{s^1_m} \cdots (\sigma^N_\mu)^{s^N_m} \]  

(2)

where \( \delta^m_n \) is the \( n \)-th bit in the binary representation \( m \) of the integer \( m \), and the matrix powers \( (\sigma^n_\mu)^0 = I \), \( (\sigma^n_\mu)^1 = \sigma^n_\mu \). On further abbreviating \( E_0 \) by \( E = \ket{00 \cdots 0} \bra{00 \cdots 0} \), a matrix \( A = [a_{mm'}]_{m,m'=0}^{M-1,M-1} = [a_{mm'}]_{m,m'=0}^{M-1} \) may be expressed using these operations as

\[ A = \sum_{m,m'=0}^{M-1} a_{mm'} \sigma^m_x E^m_x \sigma^{m'}_x , \]  

(3)

and in particular, a diagonal matrix \( D = \text{Diag}(d) \) \( (d = [d_{mm}]_{m=0}^{M-1}) \) may be written as

\[ D = \sum_{m=0}^{M-1} d_{mm} E^m = \sum_{m=0}^{M-1} d'_{mm} \sigma^m_z \]  

(4)

with \( d' = H d \), where \( H = H^1 \cdots H^N \) \( (H^n = (\sigma^n_x + \sigma^n_z)/\sqrt{2}) \) is the Hadamard transform \( \text{(not product)} \) of all the spins \([25]\).

The Hadamard product of any two such product operator expressions can be worked out from the mixed product formula (1) and the multiplication table below (in which \( \sigma_0 \) is the \( 2 \times 2 \) zero matrix).

**Table 1.**

Hadamard multiplication table for identity and Pauli matrices.

| ⊙ | \( \sigma_1 \) | \( \sigma_x \) | \( \sigma_y \) | \( \sigma_z \) |
|---|---|---|---|---|
| \( \sigma_1 \) | \( \sigma_1 \) | \( \sigma_0 \) | \( \sigma_0 \) | \( \sigma_2 \) |
| \( \sigma_x \) | \( \sigma_0 \) | \( \sigma_x \) | \( \sigma_y \) | \( \sigma_0 \) |
| \( \sigma_y \) | \( \sigma_0 \) | \( \sigma_y \) | \( -\sigma_x \) | \( \sigma_0 \) |
| \( \sigma_z \) | \( \sigma_0 \) | \( \sigma_z \) | \( \sigma_0 \) | \( \sigma_1 \) |

The essential property of Hadamard products to be used in this Letter will now be given. Let \( A, B \) and \( C \) be \( M \times M \) complex-valued matrices with \( A, C \) diagonal, and let \( \mathbf{a} = \text{diag}(A) \), \( \mathbf{c} = \text{diag}(C) \) be the column vectors formed from their diagonal elements. Then if \( \mathbf{c}^\dagger \) and \( \mathbf{C}^\dagger \) are the Hermitian conjugates of \( \mathbf{c} \) and \( \mathbf{C} \), respectively:

\[ A B C^\dagger = (\mathbf{a} \mathbf{c}^\dagger) \odot B \]  

(5)

If the matrices are sums of product operators as above, the dyadic product \( \mathbf{a} \mathbf{c}^\dagger \) may also be expressed in product operator form as

\[ \mathbf{a} \mathbf{c}^\dagger = A H E H C^\dagger . \]  

(6)
3. Lindblad operators for gradient-diffusion

Pulsed magnetic field gradients have many uses in NMR, for example to estimate conditional displacement probabilities in diverse transport phenomena [27]. The underlying theory is also important in relating NMR relaxation rates to the internal Brownian dynamics of molecules [27]. The experiments relevant to this Letter take advantage of the spatial extent of the ensemble of spin systems (molecules) in a liquid NMR sample. A magnetic field gradient \( \nabla B_z \) parallel to the static field \( B_z \) along the \( z \)-axis causes the Zeeman precession rate of the spins to vary linearly with their spatial \( z \)-coordinates, winding the transverse \((xy)\) magnetization into a spiral about \( z \) for which the net transverse magnetization vanishes. The phase coherence thus rendered unobservable can be refocussed by either a second gradient pulse of the opposite polarity, or by an RF \( \pi \)-pulse followed by a gradient of the same polarity. Hence to obtain true irreversible decoherence, it is necessary to wait for diffusion to randomize the positions of the molecules so that the correlation between their spins’ phases and \( z \)-coordinates is lost. A more detailed account of this process is outside the scope of this Letter, and may be found in Ref. [28].

Let \( \rho \) be the \( M \times M \) density matrix of an ensemble of spin systems each consisting of \( N \) spin 1/2 particles, assumed for simplicity to be noninteracting and to have the same gyromagnetic ratio \( \gamma \), which has been polarized by a static magnetic field \( B_z \) along the \( z \)-axis [1]. A uniform magnetic field gradient \( \nabla B_z \) correlates the phases of the spins with their spatial \( z \)-coordinates via the semiclassical propagator

\[
G(z) = \sum_{m=0}^{M-1} g_{mm}(z) E_m = e^{-i z k (\sigma_1^z + \cdots + \sigma_N^z)/2}.
\]  

(7)

In this expression, \( k = \gamma \int_0^t dt' \partial B_z / \partial z \) is the wave number of the phase ramp along the \( z \) axis, \( g_{mm}(z) = \exp(-i z k (N - 2h(m))/2) \) where \( h(m) = \sum_{n=1}^N \delta_{mn} \) is the Hamming weight of \( m \), and \( i^2 = -1 \). Since \( G(z) \) is diagonal, its action on a density operator \( \rho \) can be written as the Hadamard product \( G(z) \rho G^\dagger(z) = (g(z) g^\dagger(z)) \odot \rho \), and it is easily seen that

\[
g(z) g^\dagger(z) = \left[ e^{i z k (h(m) - h(m'))} \right]_{m,m'=0}^{M-1}.
\]  

(8)

The effect of the molecular diffusion period \( t \) on the elements of \( g(z) g^\dagger(z) \) is to convolute them with a Gaussian in \( z \) whose variance is \( D t \), where \( D \) is the diffusion constant [22,28]. These may be evaluated via Fourier transform methods to yield the corresponding phase damping matrix

\[
D(t) = \left[ e^{-(k(h(m) - h(m')))^2 D t} \right]_{m,m'=0}^{M-1}.
\]  

(9)

Following a refocusing gradient pulse of equal magnitude and opposite polarity, the loss of coherence due to diffusion is now given (assuming no coherent evolution) by the time-dependent density matrix \( \rho(t) = D(t) \odot \rho \) (so \( \rho(0) = \rho \)). Differentiation yields \( \dot{\rho}(t) = -R \odot \rho(t) \), where \( R = [r_{mm'}]_{m,m'=0}^{M-1} \) is the rate matrix

\[
R = \left[ (k(h(m) - h(m')) D)_{m,m'=0}^{M-1}.
\]  

(10)
Conversely, integration yields $D(t) = \exp(-Rt) = \left[\exp(-r_{m,n} t)\right]_{m,n=1}^{M-1}$. As $t \to \infty$ all elements of $D(t)$ and hence $\rho(t)$ vanish save those with $h(m) = h(m')$. Those with $m \neq m'$ represent coherences between states with equal angular momentum about the $z$-axis, which are called zero-quantum coherences in NMR \[1\].

By expanding the square in Eq. (10), it is easily seen that $R$ can be written as

$$R = \frac{1}{2} (1 (\ell \odot \ell)^T + (\ell \odot \ell) 1^T) - \ell \ell^T,$$

where $1$ is a length $M$ vector of 1’s, $\ell = [k \sqrt{2D} h(m)]_{m=0}^{M-1}$, and $1^T, \ell^T$ are their transposes. The application of Eq. (5) thus yields

$$\dot{\rho}(t) = L \rho(t) L - \frac{1}{2} L^2 \rho(t) - \frac{1}{2} \rho(t) L^2,$$

which is the desired Lindblad master equation with a single real diagonal Lindblad operator $L = \text{Diag}(\ell) = L^\dagger$. Because $\text{diag}(\sigma_1^1 + \cdots + \sigma_z^N) = [N - 2h(m)]_{m=0}^{M-1}$, this can also be expressed as $L = k \sqrt{2D/2} (N 1 - \sigma_1^z - \cdots - \sigma_z^N)$, and because the action of $L$ on $\rho(t)$ is unchanged by its overall sign or by adding on a multiple of the identity $I$, it can be further simplified to

$$L = k \sqrt{D/2} (\sigma_1^1 + \cdots + \sigma_z^N).$$

This result is readily generalized to cases in which the transverse magnetization from each type of spin has its own wave number $k^n$. Then the propagator in (6) becomes $G(z) = \exp(-iz(k^1 \sigma_1^1 + \cdots + k^N \sigma_z^N))$, and an essentially identical derivation leads to Eq. (12) with

$$L = \sqrt{D/2} (k^1 \sigma_1^1 + \cdots + k^N \sigma_z^N).$$

Decoherence processes of this kind occur naturally in homonuclear gradient-diffusion experiments, and can be obtained in homonuclear by a sequence of gradient pulses interspersed with $\pi$-pulses. If these refocus certain spins so that $k^n = 0$, the sequence applies the decoherence selectively to the remaining spins. This analysis can be further generalized to the conditional gradient operations introduced in \[31\], where the gradient pulses are interspersed with more complex RF pulse sequences and delays which implement conditional quantum logic gates such as the controlled-NOT \[31\].

To illustrate such “conditional decoherence”, consider a two-spin system subjected to a pair of gradient pulses selective for the first spin and interspersed by controlled-NOT’s $S^{1|2} = \sigma_x^1 E_x^1 + E_0^2$ to spin 1 conditional on 2, giving a net propagator \[31\]

$$G(z) = e^{-izk_2 \sigma_z^2/2} S^{1|2} e^{-izk_1 \sigma_z^1/2} S^{1|2}$$

$$= e^{-iz \sigma_z^1 (k_1 + k_2 \sigma_z^2)/2} ,$$

where the subscripts on the $k$’s now specify the temporal order of the corresponding gradient pulses. A similar derivation then gives the Lindblad operator

$$L = \sqrt{D/2} \sigma_z^1 (k_1 I + k_2 \sigma_z^2)$$
which for \( k_1 = k_2 \) selectively decoheres all off-diagonal elements of \( \rho \) except \( \sigma^1_x E^2_i \leftrightarrow \rho_{13} = \rho_{31}^* \). As another example, take a three-spin system and substitute the controlled-NOT’s in Eq. (15) by Toffoli gates \( T^{1|23} = I - E^2_i E^3_i + \sigma^1_x E^2_i E^3_i \). Then the Lindblad operator
\[
L = \sqrt{D/2} \sigma^1_x \left( k_1 I + k_2 (I + \sigma^2 + \sigma^3_z - \sigma^2_y \sigma^3_z) \right)
\]
decohers the off-diagonal elements \( \sigma^1_x E^2_i E^3_i \leftrightarrow \rho_{37} = \rho_{73}^* \) only if \( k_1 \neq k_2 \), and otherwise creates this pseudo-pure state directly from \( \rho = \sigma^1_x \). In general, the Lindblad operator will be proportional to the effective Hamiltonian of the gradient propagator preceding the diffusion period.

Thus far the discussion has been restricted to a single diffusion period, so that the decoherence, although possibly selective or conditional, acts collectively on all the affected spins. The use of multiple diffusion periods permits implementation of arbitrary correlations including the independent case [23]. The phase damping matrix in this latter case is the sum of those for each spin individually, leading to the Lindblad master equation
\[
\dot{\rho}(t) = \sum_{n=1}^{N} \left( L_n \rho(t) L_n - \frac{1}{2} L_n^2 \rho(t) - \frac{1}{2} \rho(t) L_n^2 \right)
\]
with \( L_n = \sqrt{D/2} k^n \sigma^a \) for \( n = 1, \ldots, N \). A general formula for arbitrary correlations is likewise expected to involve \( N \) Lindblads, but appears quite complicated. The result for a two-spin system, however, can be given as
\[
L_1 = a_+ \sigma^1_x + b_- \sigma^2_z, \quad L_2 = a_- \sigma^1_x + b_+ \sigma^2_z,
\]
with \( a_{\pm}^2 = \frac{1}{2} R^1 \pm S \Delta \), \( b_{\pm}^2 = \frac{1}{2} R^2 \pm S \Delta \), and
\[
\Delta = \sqrt{\frac{R^1 R^2 - (S)^2}{4(S)^2 + (R^1 - R^2)^2}}.
\]
Here, \( R^n = (k^n)^2 D/2 \) are the selective decoherence rates for the two spins, and \( S \) with \(-\sqrt{R^1 R^2} \leq S \leq \sqrt{R^1 R^2} \) is a measure of the correlation in their mutual decoherence. These equations are indeterminate if \( R^1 - R^2 = 0 = S \), and care must be taken in the choice of signs for the square roots of \( a_{\pm}^2 \) and \( b_{\pm}^2 \). Specifically, if \( R^1 \geq R^2 \) and \( S^2 > R^2 (R^1 - R^2)/2 \) all four roots are positive, whereas \( b_- \) is negative if \( S^2 < R^2 (R^1 - R^2)/2 \), and if \( R^1 \leq R^2 \) then all four roots are positive unless \( S^2 < R^1 (R^2 - R^1)/2 \), in which case the negative square root is taken for \( a_- \); if \( S = 0 \) then Eq. (18) is used instead.

In closing this section, we note that standard results in the theory of Kronecker products [24] show that the \( M^2 \times M^2 \) matrix \( \mathcal{L} \) of a Lindbladian superoperator of the form given in Eq. (18) (with any Hermitian \( L_n \)) is
\[
\mathcal{L} = \frac{1}{2} \sum_{n=0}^{N} \left( L_n^\dagger \otimes I - I \otimes L_n \right)^2,
\]
and that this is diagonal whenever the \( L_n \) are.
4. Kraus operator sums and correlated decoherence

Performing an eigenvalue decomposition of the phase damping matrix \([3]\) gives the integrated evolution equation in the standard Kraus operator sum form,

\[
\rho(t) = \rho \odot D(t) = \rho \odot \sum_{m=0}^{M-1} k_m(t) \kappa_m(t) k_m^\dagger(t) \\
= \sum_{m=0}^{M-1} K_m(t) \rho K_m(t) ,
\]

(22)

where the real diagonal matrices \(K_m(t) = \sqrt{\kappa_m(t)} \text{Diag}(k_m(t))\) are in general complicated functions of time \(t\). The only assumption made here is that \(D(t)\) is positive-semidefinite for all \(t\), as it must if \(\rho(t)\) is to be positive-semidefinite for all \(t\) and initial states \(\rho = \rho(0)\).

Due to its algebraic complexity, in general the Kraus form can be obtained only by numerically diagonalizing \(D(t)\) at each time point. For independent decoherence at the different spins, however, the rates \(r_{nm}\) are easily seen to be proportional to the squared Hamming distances \(\sum_{n=1}^{N} (\delta_m^n - \delta_{m'}^n)^2\), rather than to \((h(m) - h(m'))^2\). This leads to the Kraus form \([23]\)

\[
\rho(t) = 2^{-N} \sum_{m=0}^{M-1} \kappa_m(t) \sigma_z^m \rho \sigma_z^m ,
\]

(23)

where \(\kappa_m(t) = \prod_{n=1}^{N} (1 + (-1)^{\delta_m^n} p^n(t))\) for the one-spin survival probabilities \(p^n(t) = \exp(-tR^n)\). The case of collective decoherence appears substantially more difficult, and the eigenvalues \(\kappa_m(t)\) involve radicals even for just two spins. Assuming as in Eq. \([4]\) that the one-spin survival probabilities are both equal to \(p = p(t)\), a simple algebraic form, not based on diagonalization, exists and is given by

\[
\rho(t) = \frac{1}{4} \left( (1 + p) I - p \sigma_z^1 \sigma_z^2 \rho ((1 + p) I - p \sigma_z^1 \sigma_z^2) \\
+ \frac{1}{2} (1 - (p)^2) (I + \sigma_z^1 \sigma_z^2) \rho (I + \sigma_z^1 \sigma_z^2) \\
+ \frac{1}{2} (1 - (p)^2) (\sigma_z^1 + \sigma_z^2) \rho (\sigma_z^1 + \sigma_z^2) \right) .
\]

(24)

This does not seem to extend to larger numbers of spins.

The extended Kraus (diagonal) operator sum form,

\[
\rho(t) = \sum_{m,m'=0}^{M-1} c_{mm'}(t) \sigma_z^m \rho \sigma_z^{m'} ,
\]

(25)

turns out to be algebraically simpler, and (given that \([c_{mm'}(t)]\) is positive-semidefinite) is readily converted into the above standard form by diagonalization. To derive the extended form with arbitrary correlations between two spins, Eq. \([4]\) is used to express the microscopic effect of an arbitrary decoherence process as

\[
\left( e^{-i \frac{\pi}{2} (k^1 \sigma_z^1 + k^2 \sigma_z^2)/2} H E H e^{i \frac{\pi}{2} (k^1 \sigma_z^1 + k^2 \sigma_z^2)/2} \right) \odot \rho \\
= \frac{1}{4} \left( I + e^{-i \frac{\pi}{2} k^1 \sigma_z^1} \sigma_z^1 + e^{-i \frac{\pi}{2} k^2 \sigma_z^2} \sigma_z^2 + e^{-i \frac{\pi}{2} (k^1 \sigma_z^1 + k^2 \sigma_z^2)} \sigma_z^1 \sigma_z^2 \right) \odot \rho ,
\]

(26)

where the exponentials on the right-hand side have been commuted past each term of \(H E H = (I + \sigma_z^1 + \sigma_z^2 + \sigma_z^1 \sigma_z^2)/4\), changing the sign of their arguments as appropriate. On combining the pulse sequence which implements this propagator with one or more diffusion periods, the resulting macroscopic average is given by \([22]\).
\[
\rho(t) = \frac{1}{4} \left( I + e^{-R^1 t} \sigma_x^1 + e^{-R^2 t} \sigma_x^2 + e^{-(R^1 + R^2 + S \sigma_z^1) t} \sigma_x^1 \sigma_x^2 \right) \odot \rho ,
\]

(27)

where the rates \( R^1, R^2 \) and \( S \) are as given previously.

The Hadamard multiplication table (Table 1) shows the identity term in (27) annihilates all but the \( I, \sigma_x \), \( \sigma_y \) and \( \sigma_z \) components of \( \rho \), while the \( \sigma_x^1 \) term annihilates all but the \( \sigma_x \), \( \sigma_y^1 \) and \( \sigma_z^1 \) components, etc. These components are isolated by the projections parallel (+) and perpendicular (−) to the \( z \)-axis,

\[
\rho_{\epsilon_1 \epsilon_2} = \frac{1}{4} (\rho + \epsilon_1 \sigma_z^1 \rho \sigma_z^1 + \epsilon_2 \sigma_z^2 \rho \sigma_z^2 + \epsilon_1 \epsilon_2 \sigma_z^1 \rho \sigma_z^1 \sigma_z^2)
\]

(ε₁, ε₂ ∈ \{±1\}), which enables the evolution equation to be written without using the Hadamard product as

\[
\rho(t) = \rho_{++} + e^{-R^1 t} \rho_{+-} + e^{-R^2 t} \rho_{-+} + e^{-R^1 t} e^{-R^2 t} e^{-S \sigma_z^1 \sigma_z^2 t} \rho_{--}.
\]

(29)

Remarkably, Eqs. (27, 29) extend easily to any number of spins. The last term in (27), previously derived in (22), can be cast in a more symmetric form by expanding \( \exp(-S \sigma_z^1 \sigma_z^2 t) = \cosh(St) - \sigma_z^1 \sigma_z^2 \sinh(St) \) and applying it to \( \rho_{--} \) in Eq. (28), leading eventually to the two-spin extended Kraus operator sum

\[
8 \rho(t) = a (\sigma_z^1 \rho \sigma_z^2 + \sigma_z^2 \rho \sigma_z^1 - \sigma_z^1 \sigma_z^2 \rho - \rho \sigma_z^1 \sigma_z^2) + b_{++}^1 + \rho
\]

+ \( b_{--}^1 \rho \sigma_z^1 + b_{-+} \sigma_z^1 \rho \sigma_z^1 + b_{++} \sigma_z^1 \rho \sigma_z^1 \sigma_z^2 \)

(30)

where \( a = p^1 p^2 (1 - (q)^2)/q \) and \( b_{\epsilon_1 \epsilon_2} = 2(1 + \epsilon_1 p^1)(1 + \epsilon_2 p^2) + \epsilon_3 p^3 p^2 (q - 1)^2/q \) (ε₁, ε₂, ε₃ ∈ \{±1\}), with \( p^1 = \exp(-R^1 t), p^2 = \exp(-R^2 t), q = \exp(-St) \).

Finally, by sandwiching a gradient between RF pulses for a rotation and its inverse, the spins can be decohered about any axis. For example, a Hadamard transform may be used to obtain collective decoherence about the \( x \)-axis as

\[
\rho(t) = H(D(t) \odot (H \rho H)) H ,
\]

(31)

where \( D(t) \) is as given in Eq. (3). Isotropic collective decoherence about all three axes is obtained by applying three identical gradients, two of which are sandwiched between RF pulses for \( (\pi/2) \)-rotations about \( x \) and \( y \), and each followed by equal diffusion periods. The Lindblad operators in this case are that in Eq. (13) together with the two obtained by replacing \( \sigma_z \) by \( \sigma_x \) and \( \sigma_y \) throughout. The integrated form may be written compactly as

\[
\rho(t) = Z H Z \left( D(t) \odot (Z H D(t) \odot (H (D(t) \odot \rho) H) H Z H)^I \right) Z H \dagger ,
\]

(32)

where \( Z = \exp(-i \pi (\sigma_z^1 + \cdots + \sigma_z^N)/4) \).

5. Conclusions

It has been shown that NMR gradient-diffusion methods enable precise implementations of the adiabatic decoherence processes most often studied in QIP (24), and that the Hadamard product formalism is a simple and efficient means of analyzing such processes (regardless of their underlying physical mechanism). Nonadiabatic relaxation of \( \rho \) towards
the equilibrium density matrix $\rho_{\text{eq}}$ can also be described using the Hadamard product, simply by decohering the diagonal part of $\rho - \rho_{\text{eq}}$ about the x-axis. Using the fact that $I \otimes X = \text{Diag}(\text{diag}(X))$ for any $X$, this leads to
\begin{equation}
\rho(t) = H(D(t) \otimes (H(I \otimes (\rho - \rho_{\text{eq}}))H))H + \rho_{\text{eq}},
\end{equation}
where $D(t) = \prod_{n=1}^{N} \exp(\otimes(-t(I + \sigma_{n}^{x})/T_{1}^{n}))$ for independent decoherence (although any of the foregoing decoherence models could be used). Nonadiabatic $T_{2}$ processes can similarly be described using complex-valued phase damping matrices $D(t)$.

The ability to simulate complicated decoherence processes by gradient-diffusion, together with modulation of the natural decoherence processes operative in NMR [31], should significantly enhance the utility of NMR as a testbed for the development of more powerful quantum information processors [12]. As a means of designing such experiments, the Hadamard formalism is effectively limited to operations for which the propagator $G(z)$ can be diagonalized by a unitary matrix that is independent of $z$, since otherwise the algebra and integrations will not usually be analytically tractable. Some decoherence processes, e.g. those involving cross-relaxation [12], cannot be efficiently described by Hadamard products, and it is not known if they can be obtained by gradient-diffusion. Further work on the universality [32,33], and complexity, of these simulations is needed.

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