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ON THE EXACT UNITARY INTEGRATION OF TIME-VARYING QUANTUM LIOUVILLE EQUATIONS

Abstract. In this paper, the Dyson series corresponding to the time-varying Hamiltonian of a finite dimensional quantum mechanical system is expanded in terms of products of exponentials of a complete basis of commutator superoperators in the corresponding Liouville space. The Cayley-Hamilton theorem and the Wei-Norman formula allow to express explicitly the functional relation between the Dyson series and the product of exponentials via a set of first order differential equations. Since the method is structure preserving, it can be used for the exact unitary integration of the driven Liouville-von Neumann equation.

1. Introduction

The general solution of a quantum Liouville equation for time-varying Hamiltonians is given by the Dyson series. Normal procedure for its practical use is to truncate this expansion and work with the corresponding approximation. Beside providing approximate solutions, the main drawback of such truncations is that the unitarity of the time evolution is not necessarily preserved [17]. The method we present here relies on the formalism of the canonical coordinates of the first and second kind of the adjoint representation of the unitary group, and on the relation between them. In fact, the differential operator governing the Liouville equation can be related to a product of exponentials of the noncommuting operators corresponding to a complete basis of the adjoint representation of the Lie algebra via a set of nonlinear differential equations known as Wei-Norman formulæ. Such formulæ allows one to express the unitary evolution of the density operator *exactly* in terms of the product of exponentials. For $N \times N$ density operators, this is best understood in Liouville space. Once the parameterization of the density matrix is given in terms of the $N^2 - 1$ basis elements of $\text{ad}_{\mathfrak{su}(N)}$ (i.e. the commutator superoperator of the Liouville space) plus the identity operator, then the method corresponds to solving a time-varying system of ODEs and is one of the most popular structure preserving algorithm used by the numerical algebra community [8, 14]. The algorithm preserves unitarity, as the real time-varying parameters are multiplied by skew-hermitian matrices (the corresponding infinitesimal generators in the basis) and then exponentiated. Particular cases of the formula we use have already appeared in the literature to treat $\mathfrak{su}(2)$ -systems like spin $\frac{1}{2}$ or two-level systems [15, 17], examples which we also discuss below. The method, however, is absolutely general for all finite dimensional unitary operator algebras and for all “generalized” Euler angles one can choose on such algebras. Furthermore, it can be used for both pure and mixed states. A couple of applications, other than exact numerical simulation, are as follows. It can be used to reconstruct the behavior of a driven Hamiltonian from sequences of pulses or, on the other direction, to decompose a time-varying Hamiltonian into pulse sequences. This last will be treated in Section 4.

2. Time-independent Hamiltonians

In quantum mechanics, Liouville equations are very common to describe the time evolution of density operators or of observables in the Heisenberg picture. If H is a constant finite dimensional Hamiltonian, the density operator differential equation

$$(1) \quad \dot{\rho}(t) = -i[H, \rho] = -i\text{ad}_H(\rho)$$

is solved by

$$(2) \quad \rho(t) = e^{-itH} \rho(0) e^{itH} = \text{Ad}_{e^{-itH}} \rho(0) = e^{-it\text{ad}_H} \rho(0)$$

If $-iH \in \mathfrak{su}(N)$, then in (2) $-i\text{ad}_H$ is a so-called commutator superoperator i.e. a linear operator in the N^2 dimensional Liouville space obtained by expanding the density operator in a complete set of basis operators like the one obtained by choosing the N -dimensional Pauli matrices $\lambda_1, \dots, \lambda_{N^2-1}$ (see [10] for an explicit expression for these matrices) plus the identity matrix $\lambda_0 = N^{-\frac{1}{2}}I$: $\rho = \sum_{j=0}^n \rho_j \lambda_j$. As is well-known for this parameterization, the coefficient ρ_0 along λ_0 is a constant fixed by the $\text{tr}(\rho) = 1$ condition to $\rho_0 = N^{-\frac{1}{2}}$. Thus the evolution represented by (1) occurs along an hyperplane of the Liouville space, see [6]. Call $n = N^2 - 1$ the dimension of such hyperplane (equal to $\dim \mathfrak{su}(N)$). On the vector of n real components ρ_j , call it $\vec{\rho}$, the action of $-i\text{ad}_H$ is linear:

$$(3) \quad \dot{\vec{\rho}} = -i\text{ad}_H \vec{\rho}$$

The $\{\lambda_j\}$ basis of $\mathfrak{su}(N)$ corresponds to purely imaginary structure constants. For the scope of this paper, it is convenient to choose a skew-hermitian basis for $\mathfrak{su}(N)$, call it A_1, \dots, A_n for which we have all real structure constants $[A_i, A_j] = \sum_{k=1}^n c_{ij}^k A_k$, $c_{ij}^k \in \mathbb{R}$. Then $-iH = \sum_{j=1}^n u_j A_j$ with $u_j \in \mathbb{R}$. The corresponding basis in the adjoint representation is given by the $n \times n$ matrices $\text{ad}_{A_1}, \dots, \text{ad}_{A_n}$ and $-i\text{ad}_H = \sum_{j=1}^n u_j \text{ad}_{A_j}$, where the ad_{A_i} have matrix elements $(\text{ad}_{A_i})_{jk} = c_{ij}^k$. The $n \times n$ matrices $\text{ad}_{A_1}, \dots, \text{ad}_{A_n}$ are real and skew-symmetric and as such they are part of a basis of $\mathfrak{so}(n)$. Since $\dim \mathfrak{so}(n) = \frac{n(n-1)}{2} = \frac{N^4 - 3N^2 + 2}{2}$, for $N > 2$ the n matrices $\text{ad}_{A_1}, \dots, \text{ad}_{A_n}$ span only a proper subalgebra of $\mathfrak{so}(n)$. For example for $N = 3$ $n = \dim \mathfrak{su}(3) = 8$ while $\dim \mathfrak{so}(8) = 28$! Just like the time evolutor of the Schrödinger equation is unitary, $|\psi\rangle = U(t)|\psi(0)\rangle$, $U(t) \in SU(N)$, for the Liouville equation (3) the adjoint representation giving matrices on $\mathfrak{so}(n)$, the propagator for $\vec{\rho}$ is an orthogonal matrix:

$$\vec{\rho}(t) = O(t)\vec{\rho}(0), \quad O(t) \in SO(n).$$

For a generic (i.e. not necessarily diagonal) H , there exist many ways to compute the exponential $e^{-it\text{ad}_H}$ other than its infinite series expansion:

$$(4) \quad e^{-it\text{ad}_H} = \sum_{k=0}^{\infty} \frac{(-it)^k}{k!} \text{ad}_H^k$$

see the classical survey [12] and the recent “classroom notes” of SIAM Review [7, 9] and references therein. Here we use a method based on the Cayley-Hamilton theorem. The method consists in expressing the series expansion of $e^{-i\text{ad}_H}$ in terms of the first $n - 1$ powers of ad_H with suitable coefficients depending on the coefficients of the characteristic polynomial of $-i\text{ad}_H$ and on t . It is most suited for the adjoint representation, as the powers of the basis elements ad_{A_i} are immediately expressed in terms of the structure constants of the Lie algebra. The r -th power of ad_{A_i} is in fact given by

$$(5) \quad \text{ad}_{A_i}^r = (\text{ad}_{A_i}^r)_{kj} = \sum_{l_1, \dots, l_{r-1}=1}^n c_{1l_1}^k c_{1l_2}^{l_1} \cdots c_{1l_{r-1}}^{l_{r-2}} c_{1j}^{l_{r-1}}$$

If the characteristic polynomial is $\det(sI - (-i\text{ad}_H)) = s^n - a_{n-1}s^{n-1} - \dots - a_1s - a_0$, with coefficients $a_{n-1} = \text{tr}(-i\text{ad}_H), \dots, a_0 = (-1)^n \det(-i\text{ad}_H)$, the Cayley-Hamilton theorem affirms that $-i\text{ad}_H$ satisfies its own characteristic equation, i.e.

$$(6) \quad -i\text{ad}_H^n = a_0I + a_1(-i\text{ad}_H) + a_2(-i\text{ad}_H)^2 + \dots + a_{n-1}(-i\text{ad}_H)^{n-1}$$

and the infinite sum (4) can always be written as

$$(7) \quad e^{-it\text{ad}_H} = \sum_{k=0}^{n-1} \beta_k (-i\text{ad}_H)^k$$

for suitable $\beta_k = \beta_k(a_0, \dots, a_{n-1}, t)$, computed in detail in [3].

The procedure is valid also for the the skew-hermitian basis elements $A_i \in \mathfrak{su}(N)$, with the powers of ad_{A_i} expressed in terms of the structure constants as in (5). Using the notation $\beta_0^{[i]}, \beta_1^{[i]}, \dots, \beta_{n-1}^{[i]}$ for the coefficients corresponding to (7), we have

$$(8) \quad \begin{aligned} e^{\gamma_i \text{ad}_{A_i}} &= \beta_0^{[i]} \delta_j^k + \beta_1^{[i]} c_{ij}^k + \sum_{l_1=1}^n \beta_2^{[i]} c_{il_1}^k c_{ij}^{l_1} + \dots + \sum_{l_1, \dots, l_{n-2}=1}^n \beta_{n-1}^{[i]} c_{il_1}^k c_{il_2}^{l_1} \cdots c_{ij}^{l_{n-2}} \\ &= \sum_{r=0}^{n-1} \sum_{l_1, \dots, l_{r-1}=1}^n \beta_r^{[i]} c_{il_1}^k c_{il_2}^{l_1} \cdots c_{ij}^{l_{r-1}} \end{aligned}$$

where it is intended that

$$\sum_{l_1, \dots, l_{r-1}=1}^n c_{il_1}^k c_{il_2}^{l_1} \cdots c_{ij}^{l_{r-1}} = \delta_j^k \quad \text{for } r = 0$$

and

$$\sum_{l_1, \dots, l_{r-1}=1}^n c_{il_1}^k c_{il_2}^{l_1} \cdots c_{ij}^{l_{r-1}} = c_{ij}^k \quad \text{for } r = 1$$

(the lower index in $\beta_k^{[i]}$ gives the number of times the structure constants c_{i*}^* appear in the corresponding term).

3. Time-varying Hamiltonians

For time varying Hamiltonians, the linearity of (3) as a differential equation in the $\vec{\rho}$ coordinates implies that there are two standard ways to express its local solution, similarly to what happens for all linearly time-varying systems of differential equations [19]. The situation is obviously specular to the case of the time-varying Schrödinger equation, which has already been studied via similar techniques in [3].

When $H = H(t)$ (i.e. $u_j = u_j(t)$ in the $\mathfrak{su}(N)$ basis), the local solution of (1) can be expressed in terms of infinite formal series in the style of chronological calculus [1] or of the Dyson series, as it is commonly referred to in quantum physics [4]. Using the Dyson time-ordering operator T for $-i\text{ad}_{H(t)} = -i\text{ad}_{(u_1(t)A_1 + \dots + u_n(t)A_n)} = -i(\text{ad}_{u_1(t)A_1} + \dots + \text{ad}_{u_n(t)A_n})$:

$$(9) \quad \begin{aligned} \vec{\rho}(t) &= T \exp \left(\int_0^t -i\text{ad}_{H(\tau)} d\tau \right) \vec{\rho}(0) \\ &= \left(I + \sum_{k=1}^{\infty} \int \int \dots \int_{0 \leq \tau_1 < \dots < \tau_k \leq t} (-i\text{ad}_{H(\tau_1)}) \dots (-i\text{ad}_{H(\tau_k)}) d\tau_k \dots d\tau_1 \right) \vec{\rho}(0) \end{aligned}$$

Alternatively, it can be written as a product of exponentials over all basis elements on the adjoint representation with arbitrarily fixed order, here the cardinal order:

$$(10) \quad \vec{\rho}(t) = e^{\gamma_1(t)\text{ad}_{A_1}} \dots e^{\gamma_n(t)\text{ad}_{A_n}} \vec{\rho}(0)$$

with $\gamma_j(t)$ real valued parameters expressing the time-dependence of the solution. Notice that, as said above, $\text{ad}_{\mathfrak{su}(N)} \subseteq \mathfrak{so}(n)$ (\subsetneq for $N > 2$), but $\text{ad}_{\mathfrak{su}(N)}$ is a subalgebra and therefore it is closed under commutation. Hence all integral curves of (9) will belong to $\exp(\text{ad}_{\mathfrak{su}(N)})$ and the canonical coordinates of the second kind i.e. the product of exponentials (10) can be restricted to this subalgebra of $\mathfrak{so}(n)$ only.

The relation between (9) and (10) is given by the so-called Wei-Norman formula, [18], which is the Jacobian of the (locally invertible) transformation from (9) to (10) and is obtained by comparing (3) and the derivative of (10)

$$\begin{aligned} \left(\sum_{j=1}^n u_j \text{ad}_{A_j} \right) \vec{\rho} &= \frac{d}{dt} \left(e^{\gamma_1(t)\text{ad}_{A_1}} \dots e^{\gamma_n(t)\text{ad}_{A_n}} \right) \vec{\rho}(0) \\ &= \sum_{j=1}^n \left(\prod_{k=1}^j e^{\gamma_k \text{ad}_{A_k}} \right) \dot{\gamma}_j \text{ad}_{A_j} \vec{\rho}(t) \end{aligned}$$

along each of the $\mathfrak{su}(N)$ basis directions ad_{A_j} . Equation (8) can be used to compute in a closed form the $e^{\gamma_k \text{ad}_{A_k}}$. The result is a set of n differential equations nonlinear in the

$\gamma_j(t)$ but linear in the $u_j(t)$ that relate the two sets of parameters:

$$(11) \quad \begin{bmatrix} u_1 \\ \vdots \\ u_n \end{bmatrix} = \Xi(\gamma_1, \dots, \gamma_n) \begin{bmatrix} \dot{\gamma}_1 \\ \vdots \\ \dot{\gamma}_n \end{bmatrix}$$

which can be (locally) inverted to give:

$$(12) \quad \begin{bmatrix} \dot{\gamma}_1 \\ \vdots \\ \dot{\gamma}_n \end{bmatrix} = \Xi(\gamma_1, \dots, \gamma_n)^{-1} \begin{bmatrix} u_1 \\ \vdots \\ u_n \end{bmatrix}$$

See [3, 2] for issues related to the non-globality of the used parameterization. The two sets of parameters live on the Lie algebra $\mathfrak{ad}_{\mathfrak{su}(N)}$. Its skew-hermitian structure plus the exponentiation operation guarantee that unitarity is preserved in both the single exponential (9) and the product of exponentials (10).

While the matrix Ξ can always be obtained explicitly, see [3], except for a few simple cases like two-level systems, see [13, 15, 16], the analytic solution of (12) becomes quickly prohibitive with the dimension N . However, the systems of ODEs (11) or (12) can be numerically integrated in a structure preserving fashion using ordinary simulation tools.

4. Application: time-varying Hamiltonian and pulse sequences for a spin $\frac{1}{2}$ system

In NMR spectroscopy, a nuclear spin is manipulated by the application of suitable pulses along different axes, see [5, 11]. One of the main issues is then the reconstruction of the time-varying Hamiltonian which would correspond to a sequence of pulses and viceversa.

For sake of simplicity, we work in the case:

1. there is no constant magnetic field applied, i.e. the free Hamiltonian is zero;
2. the pulses are gaussian in shape;

In particular the first assumption means that only the interaction part of the Hamiltonian is considered and that we can work in the laboratory frame. Choosing Pauli-like skew-Hermitian matrices

$$(13) \quad A_1 = \frac{1}{2} \begin{bmatrix} 0 & i \\ i & 0 \end{bmatrix} \quad A_2 = \frac{1}{2} \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix} \quad A_3 = \frac{1}{2} \begin{bmatrix} i & 0 \\ 0 & -i \end{bmatrix}$$

we get the adjoint basis for $\mathfrak{su}(2)$

$$\text{ad}_{A_1} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & 1 & 0 \end{bmatrix} \quad \text{ad}_{A_2} = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \quad \text{ad}_{A_3} = \begin{bmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

Eq. (12) will look like:

$$(14) \quad \Xi^{-1} = \begin{bmatrix} 1 & \sin \gamma_1 \tan \gamma_2 & -\cos \gamma_1 \tan \gamma_2 \\ 0 & \cos \gamma_1 & \sin \gamma_1 \\ 0 & -\sec \gamma_2 \sin \gamma_1 & \cos \gamma_1 \sec \gamma_2 \end{bmatrix}$$

which corresponds to the inverse of equation (6) of [15]. Notice that, while on (10) the cardinal order is followed, changing the ordering (and also using repeated generators along the same direction) will lead to still admissible formulæ, see [2] for details.

Pulses of known shape are applied along the X and Y directions in different ways. The shape of the k -th gaussian pulse of amplitude $\frac{A_k}{\sigma_k \sqrt{2\pi}}$ and centered at $\tau_k = t_{k-1} + \frac{\Delta t_k}{2}$, where Δt_k is the time support of the k -th pulse and σ^2 its “variance”, is

$$(15) \quad u_k(t) = \frac{A_k}{\sigma_k \sqrt{2\pi}} e^{-\frac{1}{2} \left(\frac{t-\tau_k}{\sigma_k} \right)^2}$$

Under the assumptions above, the Magnus expansion

$$(16) \quad \vec{\rho}(t) = T \exp \left(\int_0^t (\text{ad}_{u_1(\tau)A_1} + \text{ad}_{u_2(\tau)A_2}) d\tau \right) \vec{\rho}(0)$$

maps the (pure or mixed) density operator $\vec{\rho}_0$ to

$$(17) \quad \vec{\rho}(t_3) = e^{\gamma_1 \text{ad}_{A_1}} e^{\gamma_2 \text{ad}_{A_2}} e^{\gamma_3 \text{ad}_{A_3}} \vec{\rho}_0$$

with the $\gamma_j = \gamma_j(t)$ obtained numerically from the ODEs (12). In all the simulations below, the model is adimensional: \hbar and the gyromagnetic ratio are set to 1 and the time scales and amplitudes refer to arbitrary units.

4.1. Case I: simultaneous pulses

Along X and Y apply simultaneously identical gaussian pulses with $\sigma_1 = \sigma_2 = 1$, $A_1 = A_2 = 1$ and centered at $\tau_1 = \tau_2 = 3$. The u_1 and u_2 coordinates of the Magnus expansion (16) and the corresponding time-varying coordinates γ_j , $j = 1, 2, 3$ in the product of exponentials representation (17) are shown respectively Fig. 1 and Fig. 2. While $u_3 \equiv 0$, $\gamma_3 \neq 0$ because of noncommutativity.

4.2. Case II: pulses with disjoint support

In (15), if $A_k = 1$ and we consider the “classical” 3σ case, i.e. choose $\frac{\Delta t_k}{2} = 3\sigma$, then 99.7% of the pulse area is contained in the interval $[t_{k-1}, t_k]$. This is the case normally considered for example in NMR, as the approximation of (16) with a product of exponentials is acceptable if the pulses are disjoint in time. In particular, a sequence XY of pulses (i.e. first a pulse along Y, then along X) with $\sigma_1 = \sigma_2 = 1$, $A_1 = A_2 = 1$

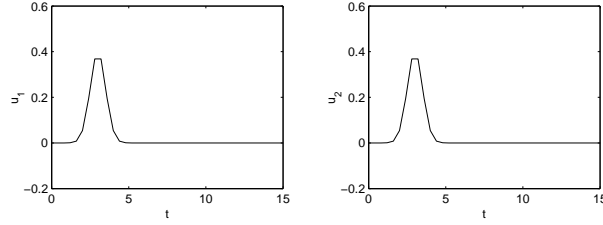


Figure 1: Case I: u_1 and u_2 coordinates

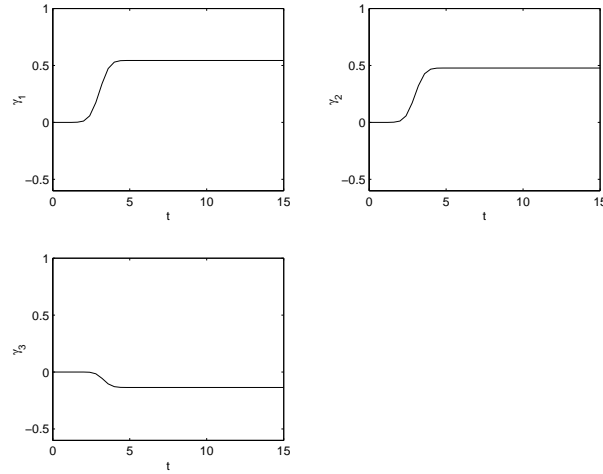


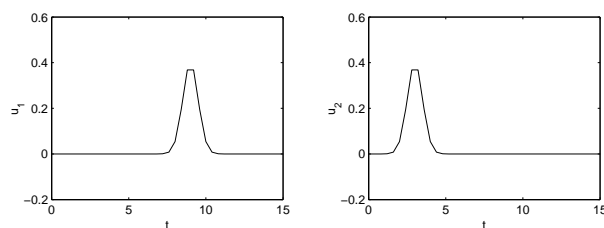
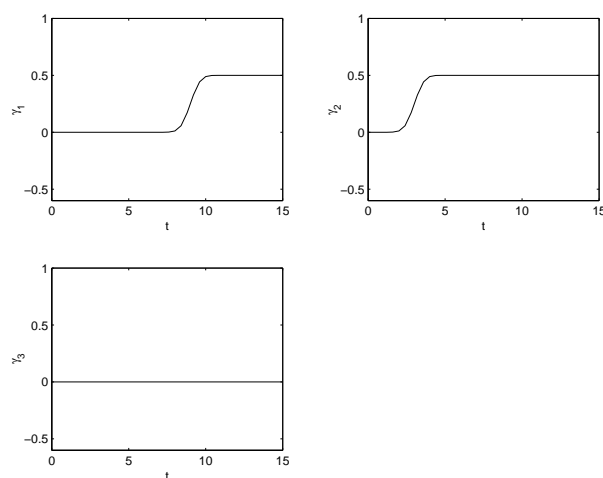
Figure 2: Case I: γ_1 , γ_2 and γ_3 coordinates

and respectively centered at $\tau_1 = 9$ and $\tau_2 = 3$ implies that

$$\begin{aligned} \vec{\rho}(t) &= T \exp \left(\int_0^t (\text{ad}_{u_1(\tau)A_1} + \text{ad}_{u_2(\tau)A_2}) d\tau \right) \vec{\rho}(0) \\ &\simeq \exp \left(\int_0^t \text{ad}_{u_1(\tau)A_1} d\tau \right) \exp \left(\int_0^t \text{ad}_{u_2(\tau)A_2} d\tau \right) \vec{\rho}(0) = e^{\gamma_1 \text{ad}_{A_1}} e^{\gamma_2 \text{ad}_{A_2}} \vec{\rho}_0 \end{aligned}$$

In this case, the coordinate along the Z direction remains constantly zero also in the product of exponentials coordinates. See Fig. 3 and Fig. 4 for the values of the time-varying parameters u_j and γ_j .

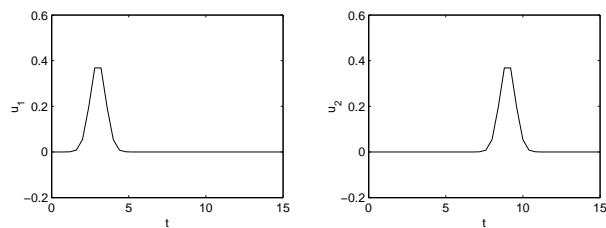
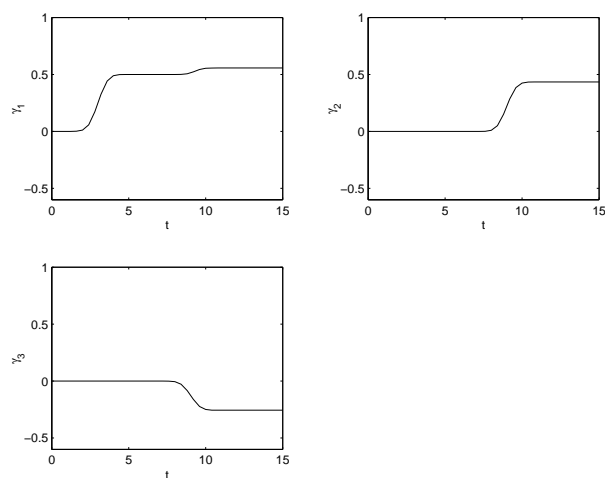
However, the Wei-Norman formula is a coordinate dependent expression and also the order in which the basis elements are taken in (10) matters. For example, if instead of the XY sequence of Fig. 3 and Fig. 4 we apply the same pulses but in the opposite order (YX: first along X then along Y) then the result changes when represented in the basis ordering given by the cardinality, as in (10) and (17). The

Figure 3: Case II: u_1 and u_2 coordinatesFigure 4: Case II: γ_1 , γ_2 and γ_3 coordinates

time-evolutions of the γ_j in this case are represented in Fig. 6. As can be seen, also the γ_3 component becomes nonnull. Obviously, when a constant magnetic field is applied along the Z direction, H splits into constant and time-varying parts: $-iH = \bar{u}_3 A_3 + (u_1(t)A_1 + u_2(t)A_2)$, $\bar{u}_3 = \text{const}$, and an interaction representation has to be used to recover the results.

4.3. Hamiltonians from sequences of pulses: an outlook

The common way to reconstruct a time-varying Hamiltonian in NMR is through some form of averaging directly on the truncation of expressions like our product of exponentials [5]. When the method of this Section is applied to known sequences of pulses, i.e. to smooth time-varying coordinate functions $\gamma_j(t)$ in the product of exponentials (10), then it provides the time-depending functional expression of the parameters $u_j(t)$, from which an averaged expression for the Hamiltonian could be easily attained, with-

Figure 5: Case II bis: u_1 and u_2 coordinatesFigure 6: Case II bis: γ_1 , γ_2 and γ_3 coordinates

out resorting to truncations.

5. Conclusion

If two-level systems like those of Section 4 are simple enough that explicit solutions are available [13, 15], the methodology presented here is general enough to describe realistic cases of laser-driven molecules or tensor products of nuclear spins in rf fields. It could be used to simulate the exact response of the systems to pulse shaping and for their coherent control.

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