

Recursive Evaluation of Interaction Pictures

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The interaction picture is a useful tool in quantum mechanics, allowing one to investigate slow and possibly complicated processes in the presence of fast motions whose behavior is well understood. Important examples in NMR are the rotating-frame transformation (1) and the "toggling-frame" picture used in multiple-pulse NMR (2, 3) and composite pulses (4). The interaction picture is related to the laboratory frame by a time-dependent unitary transformation operator. If this operator commutes with itself at different times, as in the rotating-frame transformation, the evaluation of the transformation is usually quite simple. In multiple-pulse NMR, however, the pulses are usually applied with different phases, leading to noncommuting transformation operators. In this case the transformation is conventionally performed in a straightforward fashion which requires a number of transformations proportional to the square of the number of pulses in the sequence. In this paper we show that this method is not inherent to the transformation and demonstrate a simplification which is recursive, thereby making the number of necessary transformations linear in the number of pulses in the sequence.

The Hamiltonian describing a multiple-pulse sequence can be written in the laboratory frame as

$$\mathcal{H}(t) = \mathcal{H}^0 + \mathcal{H}^{\text{RF}}(t), \quad [1]$$

where $\mathcal{H}^{\text{RF}}(t)$ describes the effect of the radiofrequency pulses, while \mathcal{H}^0 contains all the other interactions. The general solution of the Liouville-von-Neumann equation

$$\rho(t) = T \exp \left[-i \int_0^t \tilde{\mathcal{H}}(t') dt' \right] \rho(0) T \exp \left[i \int_0^t \mathcal{H}(t') dt' \right], \quad [2]$$

where T represents the Dyson time-ordering operator, is usually transformed into an interaction picture where

$$\tilde{\rho}(t) = T \exp \left[-i \int_0^t \tilde{\mathcal{H}}(t') dt' \right] \rho(0) T \exp \left[i \int_0^t \tilde{\mathcal{H}}(t') dt' \right]. \quad [3]$$

Here

$$\tilde{\mathcal{H}}(t) = U^{-1}(t) \mathcal{H}^0 U(t) \quad [4]$$

represents the Hamiltonian in the so-called "toggling frame" and

$$U(t) = T \exp \left[-i \int_0^t \mathcal{H}^{\text{RF}}(t') dt' \right] \quad [5]$$

is the transformation operator. The motivation for this separation is that we are not interested in the fast common motion of the spins due to the RF pulses, but in the evolution of the system under the internal Hamiltonian \mathcal{H}^0 which is modified by the effect of the pulses. This goal can be achieved by stroboscopic observation at times when $U(t) = \mathbb{1}$. As can be seen immediately from Eq. [3], the evolution of the system is then determined entirely by the interaction representation Hamiltonian $\tilde{\mathcal{H}}(t)$.

The evaluation of the interaction picture defined by [4] is often performed as follows: The transformation operator $U(t)$ is divided into discrete parts, corresponding to individual RF pulses. The toggling-frame Hamiltonian after the n th pulse can then be written as

$$\tilde{\mathcal{H}}_n(t) = P_1^{-1} P_2^{-1} \cdots P_n^{-1} \mathcal{H}^0 P_n \cdots P_2 P_1, \quad [6]$$

where

$$P_i = T \exp \left[-i \int_{t_i}^{t'_i} \mathcal{H}^{\text{RF}}(t') dt' \right] \quad [7]$$

and t_i and t'_i mark the beginning and end of the i th pulse, respectively. This procedure corresponds to first applying the inverse of the n th pulse to the laboratory-frame Hamiltonian, followed by the inverse of the $(n-1)$ th pulse, etc., until the transformation by the inverse of the first pulse leads to the final form of the interaction Hamiltonian. Because this approach proceeds backward in time, it does not permit the calculation of $\tilde{\mathcal{H}}_{n+1}$ from $\tilde{\mathcal{H}}_n$ and the $(n+1)$ th pulse; it is necessary to repeat the whole calculation for the next step, starting with P_{n+1} . The number of transformations required therefore grows quadratically with the number of pulses in the sequence. In addition, the apparent time reversal in [6], together with the lack of a recursion formula, makes the values of $\tilde{\mathcal{H}}(t)$ in the different windows appear to be unrelated to each other, thereby obscuring the relation between $\tilde{\mathcal{H}}_n$ and $\tilde{\mathcal{H}}_{n+1}$.

By a simple change in picture the same calculation can be performed in a simpler fashion. The idea is to express the effect of the pulses themselves in the toggling frame. We thus define

$$\tilde{P}_n = P_1^{-1} P_2^{-1} \cdots P_{n-1}^{-1} P_n P_{n-1} \cdots P_2 P_1. \quad [8]$$

By inverting this equation to solve for P_n and inserting the result in Eq. [6], we can express $\tilde{\mathcal{H}}(t)$ as

$$\tilde{\mathcal{H}}_n(t) = \tilde{P}_n^{-1} \cdots \tilde{P}_2^{-1} \tilde{P}_1^{-1} \mathcal{H}^0 \tilde{P}_1 \tilde{P}_2 \cdots \tilde{P}_n. \quad [9]$$

Thus, if the effect of the pulses, and therefore of the transformation operator $U(t)$, is itself described in the toggling frame, the transformation can be accomplished in the forward sense. Successive values of the interaction-frame Hamiltonian are now related via the recursion relation

$$\tilde{\mathcal{H}}_{n+1}(t) = \tilde{P}_{n+1}^{-1} \tilde{\mathcal{H}}_n(t) \tilde{P}_{n+1}. \quad [10]$$

Accordingly, the number of necessary transformations becomes linear in the number of pulses. The reversal of order observed here is analogous to the coordinate transformation by Euler angles, where the sequence of rotations in the modified frame occurs in the opposite sense compared to their application in the original frame.

The actual evaluation of the \tilde{P}_i does not revert to Eq. [8], but proceeds via direct transformation of $\mathcal{H}^{\text{RF}}(t)$ into the toggling frame and subsequent integration

$$\tilde{P}_i = T \exp \left[-i \int_{t_i}^{t_i'} \tilde{\mathcal{H}}^{\text{RF}}(t') dt' \right]. \quad [11]$$

In general, all the angular-momentum operators that occur in the coupling Hamiltonian $\mathcal{H}^{\text{RF}}(t)$ must be transformed into the toggling frame. In most cases this means that \tilde{I}_x and \tilde{I}_y must be evaluated for the whole sequence, even though only \tilde{I}_z usually occurs in \mathcal{H}^0 for high-field NMR. The evaluation of all three components is normally required anyway for multiple-pulse sequences at zero field (5) or for iterative expansions (5, 6), where all three terms occur in \mathcal{H}^0 .

As a simple example we calculate the interaction Hamiltonian for the WHH-4 sequence (7). The basic pulse cycle can be written as $-\tau/2-\bar{x}-\tau-y-2\tau-\bar{y}-\tau-x-\tau/2-$. We use here the usual notation, abbreviating 90° pulses with lower case letters designating the relative phase. Figure 1 illustrates the procedure: the relative phase of the pulses in the laboratory frame is written on top of the pulse while the corresponding phase in the toggling frame appears at the bottom. The first pulse is always the same in the laboratory frame and in the toggling frame, in this case P_{-x} . It transforms the laboratory-frame operators (I_x, I_y, I_z) into ($I_x, -I_z, I_y$), using the same sense of rotation as Haebleren (3). Thus, I_y , the coupling operator for the second pulse, has been turned into $-I_z$ and the effect of the second pulse can be written as P_y or \tilde{P}_{-z} . Using the latter notation, it is obvious that the three operators are transformed into $(-I_y, -I_z, I_x)$. The last two pulses simply reverse the effect of the first two, making the calculation straightforward in either notation.

The method is equally applicable to composite pulse schemes or windowless multiple-pulse sequences. As an example consider the BLEW-12 sequence (8). The toggling-frame values of \tilde{P}_i can be obtained by replacing x or y with the corresponding table entry from the line starting with I_x or I_y , respectively. The procedure is the same as

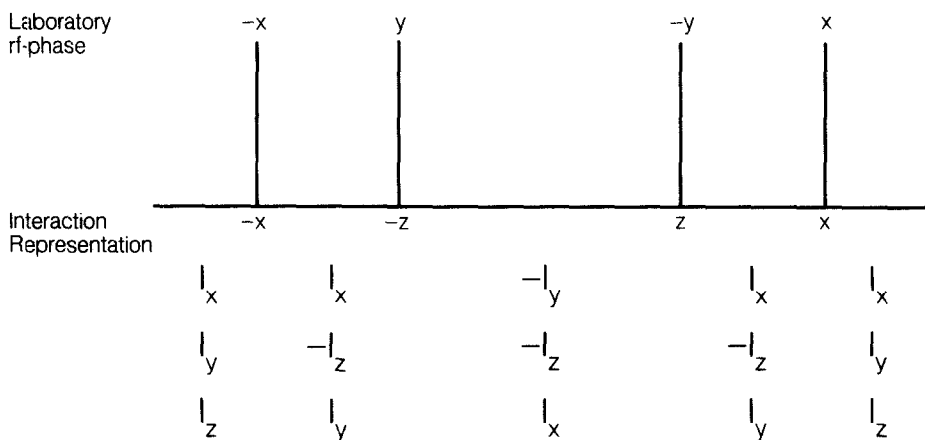


FIG. 1. Schematic representation of the procedure for the WHH-4 sequence. The phases of the pulses in the laboratory frame are indicated on top of the drawing. The letters under the pulses indicate the direction of the pulses in the toggling frame. Note that in this frame pulses in the $\pm z$ direction occur naturally. Each successive step of the interaction picture can be evaluated from the preceding one by applying a rotation around the axis indicated under the pulse.

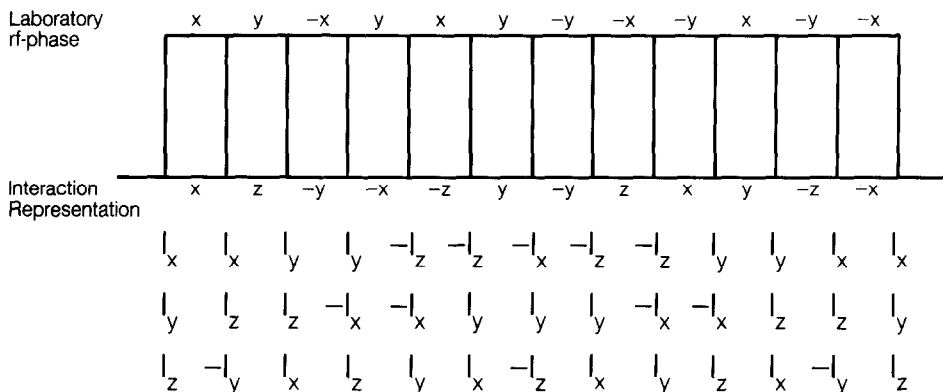


FIG. 2. Same as Fig. 1, for the windowless BLEW-12 sequence. The interaction picture values of the angular momentum operators refer to the instantaneous values between two pulses.

that for the WHH-4 sequence and is illustrated in Fig. 2. The resulting values of the angular-momentum operators at the times between the pulses are shown in the figure. The Hamiltonian during the n th pulse can be calculated as

$$\tilde{\mathcal{H}}_n(t) = \exp\left[i \int_{t_n}^t \tilde{\mathcal{H}}^{\text{RF}}(t') dt'\right] \tilde{\mathcal{H}}_{n-1}(t_n) \exp\left[-i \int_{t_n}^t \tilde{\mathcal{H}}^{\text{RF}}(t') dt'\right]. \quad [12]$$

In conclusion, we have shown that it is possible to evaluate interaction pictures starting with the first transformation and proceeding forward in time by expressing the transformation operator itself in the interaction frame. This allows one to evaluate successive steps recursively, thereby reducing the number of coordinate transformations needed from a quadratic function in the number of pulses to a linear function. This recursive transformation is also easier to grasp conceptually. The method is of course not limited to NMR but can be used for the evaluation of any interaction representation in time-dependent quantum mechanical problems.

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