

1. The basis sets for the matrix representations should be given.

The matrix form of the propagator is expressed in the Cartesian basis. This has been clarified in the text.

2. Some of the equations, like Eq. (24), are not obvious, pointers at derivation may be given.

A term proportional to  $y_1(t)$  is absent from Eq. 24. Thus, the homogeneous solution can be found by two integrations and the inhomogeneous solution found by variation of parameters. We have added this information and a reference to a standard text on differential equations. The same is true for Eq. 27.

3. It is written that HAM works well with an appropriate choice of a linear operator and starting functions. Are these given in the manuscript for the examples illustrated? How easy is to fix these for an arbitrary pulse? What are the guiding parameters in this regard?

The choices of linear operator and starting approximation are given in the text for each application of HAM. For Method 1, the linear operator is given in Eq. 19 and the initial starting function given in the following text. In the present case we chose, mostly by trial-and-error, the linear operator and starting function so that the differential equation for  $y_1(t)$  had a very simple form and the equations for  $y_n(t)$  were “special” second-order differential equations with known analytical solutions. The monograph by Liao discusses choices of these parameters in a number of applications. Certainly, the choice of these parameters is the difference between success and failure.

4. Any comments on the quaternions and Euler angles calculation for the pulse methods illustrated here?

We are not sure what specifically is being asked by the reviewer. The quaternion and Euler angle methods are highly accurate in calculating the performance of shaped pulses and the purpose of the paper is not to criticize these techniques. Indeed, the ‘exact’ calculations provided in the paper for comparison to approximations using HAM were calculated using the Euler angle approach.

5. When the authors say that the HAM-Riccati approach may work well for other cases in NMR, what do they have in mind?

The Riccati equation can be modified to include radiation damping, but not relaxation (Rourke, D.E. (2002), Solutions and linearization of the nonlinear dynamics of radiation damping. *Concepts Magn. Reson.*, 14: 112-129. <https://doi.org/10.1002/cmr.10005>). HAM however is generally applicable to solution of single or systems of differential equations, provided suitable linear operators, initial approximations  $y_0(t)$ , and auxiliary functions  $H(t)$  can be found.

6. In the current manuscript, although elegant examples are given, I am not sure how the approach can be used to make the schemes better. Perhaps this can be explained in the text.

As noted in the text, the efficiency of the Method 1 calculations may be useful in optimization of new pulse shapes, in which many iterative steps must be taken.

7. On a semantic level, I am not sure what is actually meant by theoretical magnetic resonance. It is essentially an experimental field with solid inputs from theory. We may not want to just keep calculating some parameters from sophisticated equations which may have no practical relevance.

We have removed the word “theoretical” from the Introduction. As noted above, the HAM approach may be useful for optimization of new pulse shapes. However, in some cases, the HAM results (as in other analytic approximations) may provide new insights. For example, Method 1 gives  $f(t)$  essentially as the ratio of sums of iterated integrals of  $w(t)$ . The iterated integrals are related to the Fourier integrals used by Warren in a perturbation expansion using average Hamiltonian theory. This correspondence suggests a connection between the two approaches that might prove useful, although beyond the scope of the present paper. Finally, one purpose of the paper is to introduce to NMR spectroscopists a powerful approach for approximating the solutions to systems of differential equations. We anticipate that other spectroscopists will find interesting applications of the approach in their own work.