General comments

The author provides an exhaustive collection of analytical expressions for cases, in which the spin density operator evolves in a low-dimensional subspace of the Liouville space. Although the theory behind the procedure is not new and some of the examples are known from literature, the "completeness" of the work presented in the manuscript makes it a very useful source of information for researchers who want to go beyond numerical "black box" calculations and want to get a better understanding of the evolution of spin systems. I think the manuscript is very interesting both for experts and for newcomers in the field of magnetic resonance and I would like to see this contribution published.

Specific comments

To make the manuscript easier to understand for newcomers I suggest to include a bit more background information. Some essential background knowledge is taken for granted, making the understandability sometimes a bit difficult. In my opinion, it should be explained explicitly what is behind the "arrow notation" of the propagation rules introduced in equation (1) and (2), for example, by providing an equation like

$$\hat{\rho}(t) = \exp(-i\hat{H}t)\hat{\rho}(0)\exp(i\hat{H}t).$$

By expanding the exponentials in this equation the occurence of the multiple commutators can be readily explained. Without this, the mentioning of repeated calculations of commutators comes "out of the blue", at least for less experienced readers.

I also have a problem understanding section 1.1 of the SI: I find it difficult to bring the equation and the text above it together — perhaps the text can be rephrased more clearly. I guess the meaning of the arrow $\mapsto$ is *maps to*. It may also help to explicitly explain the different symbols used for abstract operators (such as $\hat{\rho}$) and their matrix representations (such as $\boldsymbol{\rho}$).

Technical issues

Overall, the text is diligently written but it is no surprise that such a comprehensive document comes with some errors. In the following, both the errors I found (not having read every sentence or equation!), some suggestions and minor questions are listed.

1. page 1, line 22ff: The introductory example mentioned here is propagation of transversal magnetization of spin $I = 1/2$ (which is a good choice) but the following equation (1) and the text on page 2, line 25, contain I_z instead of I_x. In eq. (1), both occurrences of I_z should be replaced.

2. page 2, line 28: I suggest to replace *in this case* by *in this example.*

3. page 2, line 31: Typo in *dipol-dipol*

4. same line: I suggest to add *or* before *cross polarization.*

5. page 3, lines 50–51: I suggest to replace *independent of the dimension of the latter* by *although the Liouville space has a much larger dimension.*

6. page 3, line 52: I wonder if the statement *"However, condition (3) cannot be fulfilled if more than one interaction has to be considered"* is always true.

7. page 3, line 54: *... an initial state* $\rho_0 = \hat{I}_z$. (Shouldn't the density operator carry a ˆ?). This is one of the few instances, where the equal sign (=) is used for assigning the initial state. In most of the manuscript (including SI), assignments of special values are indicated by arrows (→). I prefer the equal sign because the arrow can be misinterpreted as indication of a limiting value or, in the context of this manuscript, a time evolution.

8. page 3, line 63: I suggest to rephrase the sentence (for better understandability) and write: *... note the* 2×4 *matrix in Eq. (5) is the exponential of the* 2×2 *matrix in Eq. (6) multiplied by* $-it$ *....*

9. page 3, line 67: change *formed* to *formulated*

10. page 3, line 70: change *was possible* to *is possible*

11. page 3, line 74: change *for the further work here* to *this work*

12. page 4, line 85: I think *estimating* should be changed to *calculating.* (There are more instances, where *estimate* is used instead of *calculate.* Please check.)

13. page 4, lines 86–87. I suggest to rephrase the sentence: *... but it depends on the relevant space, which is different for different numbers of spins.*

14. page 5, line 118: change estimation to *calculation*

15. page 5, line 122: The operator $\hat{A}$ is missing its hat.

16. page 6, lines 152–153: *see Example 1D-1 in the SI.* In the SI, there is no such example. A 1D subspace is mentioned in section 4.1.

17. page 7, line 172: extra *all*

18. page 7, line 177: parenthesis not closed

19. page 7, line 181: extra *above*

20. page 7, lines 181–182: Shouldn't all N be replaced by n?

21. page 8, Eq. 22: Isn't the matrix U multiplied from the left, resulting in $\hat{A}_1 \cdot U_{11} + \hat{A}_2 \cdot U_{21} + \ldots$ (inverted indices of U_{kl})?

22. page 9, line 219: I suggest *... appearing in Eq. (5) and (6).*

23. page 10, line 243: replace *estimate* by *calculate*

24. page 12, line 286: typo, it should probably read: *... with the amplitude* $\frac{D_{IS}^2}{\omega_{IS}^2+D_{IS}^2}$.

25. page 13, line 319: replace DRKS by *doubly rotating frame* (I think it should be "doubly rotating" instead of "double rotating" everywhere.)

26. page 17, line 367: I think *... larger prefactor, which reflects the roof effect* is correct.

27. page 18, line 376: *a limited power*

28. page 20, line 406: *an I spin*

29. page 21, lines 432–433: N and n not clear. I think N is the total number of spins, and n the number of factors in the product. For clarity one should write $2^{(N/2)-n}$—if I understood it correctly.

30. page 22, line 461: What is $\omega_{1I;S}$?

31. page 23, lines 469–470: Perhaps better *... developed the method, used it to derive the examples given here and ...*?

32. page 23, line 472: no plural for *advice*

33. SI, page 4, line after (S2): instead of *Similarly* the use of *Similar to the dipolar Hamiltonian* might be more informative.

34. SI, page 4, line 4 from bottom: How about *... is parallel magnetization of spins* I_1 *and* I_2*, aligned transversal to* $\mathbf{B_0}$?

35. SI, page 5, line after 3.2.2.3: The extra punctuation mark after *Hamiltonian:* should be deleted.

36. SI, page 6, sentence before 3.2.3.2: *... not* $-(3/2)D_{II}$ *... (example 2D-1)* (minus-sign for completeness, wrong example number)

37. SI, page 7, line 7 (including eq.): *... can be detected ...*

38. SI, page 7, change of sentence: *The cases where the relevant magnetic field strengths are not large with respect to the coupling frequency and where deviations from Hartmann-Hahn condition occur are problems ...*

39. SI, page 7, eq. (S15) and (S16): What is q?

40. SI, page 8, line 1 after 3.3.2.1 Here and elsewhere: replace all *Equ.* by *Eq.*

41. SI, page 8, line 2 after (S17): replace *what* by *which*

42. SI, page 9, (S21): typo, change to *crossing*

43. SI, page 10, line 7 after (S23): Do you mean *approaches* instead of *approximates*?

44. SI, page 10, line 7 after (S23): Avoid starting the sentence with I.e., one could write *In other words, it describes ...*

45. SI, page 10, line 3 before 3.3.3.2: Missing word: *The constant component is subject ...*

46. SI, page 12, line 1: *approach* instead of *approximate*

Sorry, I had to stop here because I ran out of time.